

ORAU TEAM Dose Reconstruction Project for NIOSH

Oak Ridge Associated Universities | NV5|Dade Moeller | MJW Technical Services

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DOE Review Release 04/08/2019

| | | ORAUT-RPRT-0060 | Rev. 00 | |
|-----------------------------|---|------------------------------------|-----------|-------------|
| Neutron Dose fron | n Highly Enriched Uranium | Effective Date: | 03/28/2 | 019 |
| | | Supersedes: | None | |
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Page Change

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PUBLICATION RECORD

| EFFECTIVE | REVISION | |
|------------|----------|---|
| DATE | NUMBER | DESCRIPTION |
| 03/28/2019 | 00 | New document initiated for evaluation of neutron dose from highly enriched uranium at the Oak Ridge Gaseous Diffusion Plant (K-25), Paducah Gaseous Diffusion Plant, Portsmouth Gaseous Diffusion Plant, and Y-12 Plant. Incorporates formal internal and NIOSH review comments. Training required: As determined by the Objective Manager. Initiated by Matthew H. Smith. |

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ACRONYMS AND ABBREVIATIONS

| cm | centimeter |
|-------------|--|
| DOE DU | U.S. Department of Energy depleted uranium |
| ETTP | East Tennessee Technology Park |
| EU | enriched uranium |
| ft | feet |
| g | gram |
| GDP | gaseous diffusion plant |
| HEU | highly enriched uranium |
| hr | hour |
| ICPP | Idaho Chemical Processing Plant |
| ICRP | International Commission on Radiological Protection |
| in. | inch |
| IREP | Interactive RadioEpidemiological Program |
| LEU | low-enriched uranium |
| LOD | limit of detection |
| m | meter |
| MeV | megaelectron-volt, 1 million electron-volts |
| mR | milliroentgen |
| mrem | millirem |
| n | neutron |
| NFS | Nuclear Fuel Services |
| NIOSH | National Institute for Occupational Safety and Health |
| N:P | neutron-to-photon ratio |
| NU | natural uranium |
| ORAU | Oak Ridge Associated Universities |
| ORNL | Oak Ridge National Laboratory |
| ppb | parts per billion |
| PW | product withdrawal |
| RU | recycled uranium |
| s | second |
| SRDB Ref ID | Site Research Database reference identification (number) |
| SRS | Savannah River Site |
| t | ton |
| TLD | thermoluminescent dosimeter |
| USEC | United States Enrichment Corporation |

- VHEU very highly enriched uranium
- WGU weapons-grade uranium wt % weight percent
- α alpha particle

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1.0 INTRODUCTION

Neutron dose to workers at gaseous diffusion and uranium metals plants has been an issue of concern because some workers were unmonitored or monitored for exposure to photon and beta radiation only. Information is provided on this topic in the Guide of Good Practices for Occupational Radiological Protection in Uranium Facilities (DOE 2009), but the basis for that information is limited. Therefore, this report focuses on measurements and calculations of the neutron dose and neutron-to-photon (N:P) ratio from various uranium compounds with an emphasis on exposures to highly enriched uranium (HEU) compounds at gaseous diffusion plants and at uranium metal processing facilities, such as the Y-12 Plant in Oak Ridge, Tennessee.

2.0 BACKGROUND

From 1944 to 1952, all uranium used by the Manhattan Engineer District and the U.S. Atomic Energy Commission [(AEC) U.S. Department of Energy (DOE) predecessor agencies] was derived from natural uranium (NU) sources (DOE 2003, p. 29). NU contains the following three isotopes: ²³⁸U, ²³⁵U, and ²³⁴U. Uranium that has been processed to increase the weight concentration of ²³⁵U is referred to as enriched uranium (EU). The extent of the enrichment depends on the intended use of the uranium (DOE 2009). Commercial light-water reactors are designed for use with the ²³⁵U enriched from 3% to 5% by weight, or so-called low-enriched uranium (LEU). HEU is required for use in high-temperature gas-cooled reactors, naval nuclear propulsion reactors, most research reactors, and weapons (DOE 2009, p. 19). Weapons-grade uranium (WGU) is produced using separation techniques that increase the weight concentration of ²³⁵U to more than 90% by weight (NAS 2005). For example, the concentrations by weight of various uranium isotopes in materials from NU sources and from recycled very highly enriched uranium (VHEU) sources such as the Savannah River Site (SRS) and the Idaho Chemical Processing Plant (ICPP) are provided in Table 2-1.

| Material | U-233 (wt %) | U-234 (wt %) | U-235 (wt %) | U-236 (wt %) | U-238 (wt %) | Non-U:U α emission rate (%) |
|--|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------------------------|
| NU ^a | <0.001 | 0.0057 | 0.7193 | <0.001 | 99.275 | ≈0 |
| LEU ^a | <0.001 | 0.025 | 3.5 | <0.001 | 96.475 | ≈0 |
| HEU ^a | <0.01 | 0.12 | 94.0 | <0.01 | 5.88 | ≈0 |
| High burn-up recycled VHEU from SRS ^b | 0.01 | 1.28 | 52.2 | 29.2 | 17.3 | 0.04 |
| Lower burn-up recycled VHEU from ICPP ^b | 0.0003 | 1.0 | 78.0 | 20.0 | 1.7 | 0.08 |
| Recycled VHEU (WGU) ^{b,c} | <0.01 | 1.1 | 93 | 0.75 | 6 | 0.21 |
| Recycled NU ^b | <0.0001 | 0.0054 | 0.72 | <0.0001 | 99.27 | 0.008 |
| Recycled LEU ^b | <0.01 | 0.02 | 2.0 | <0.01 | 97.98 | 0.04 |
| Recycled DU ^b | 0.001 | 0.2 | < 0.0001 | 0.0003 | 99.8 | 0.018 |

Table 2-1. Composition of various uranium materials and two specific VHEU materials from the SRS and ICPP.^a

a. Uranium composition from NAS (2005, Table A-2).

b. Recycled uranium (RU) composition from ORAUT (2012, Table 5-7).

c. WGU is produced using separation techniques to increase the concentration of U-235 to more than 90 wt % (NAS 2005).

In 1952, the AEC started reprocessing uranium from weapon stockpiles and reactors (DOE 2003). This reprocessed uranium is commonly referred to as recycled uranium (RU). Uranium that has been irradiated in reactors contains transuranic elements (such as plutonium and ²³⁷Np), fission products (such as ⁹⁹Tc), and reactor-generated products (such as ²³⁶U). After chemical processing to separate and extract plutonium, as well as to recover uranium for reuse, trace quantities of plutonium, neptunium, technetium, and ²³⁶U remain in the RU. These constituents make the RU more radioactive than NU. Thus, the handling, reprocessing, and reenrichment of RU may present a greater potential for personnel exposure than that normally associated with the processing of

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unirradiated uranium (BWXT 2000). In the early days of RU processing, the Y-12 Plant set an acceptance limit of 10 ppb for the plutonium concentration in RU being sent to the site for subsequent processing. This limit has continued in use for the duration of the RU production (DOE 2003). Shipments of RU with plutonium concentrations above this limit required site permission before shipment. Due to these requirements, nearly all RU coming from these sites contained less than 10 ppb of plutonium (DOE 2003).

The RU process streams involved the processing of uranium metals, uranium alloys, chemical compounds (such as UF₄ and UF₆), uranium oxides (UO₂, UO₃, U₃O₈), uranium chloride (UCl₄), and sodium diuranate (Na₂U₂O₇) (ORAUT 2005, p. 3). Comparisons of neutron production rates due to spontaneous fission and (α ,n) reactions in various isotopes and two chemical compounds of uranium are provided in Table 2-2. The interaction of alpha particles from uranium with nuclei of fluorine, oxygen, and other low-atomic weight atoms generates neutrons with energies of approximately 2 MeV (DOE 2009). The magnitude of the neutron flux varies based on total activity of the uranium (a function of enrichment) and the chemical compound (combining of uranium with fluorine or oxygen). In the case of UF₆, the typically measured neutron dose equivalent rates for storage containers are as follows (DOE 2009):

 NU to 5% EU:
 0.01 to 0.2 mrem/hr

 VHEU (97%+):
 2 to 4 mrem/hr (contact)

 1 to 2 mrem/hr (3 ft or 1 m).

| Uranium isotope or | Spontaneous fission vield | Alpha particle vield | (α,n) yield in oxides | (α,n) yield in fluorides |
|---------------------------------|------------------------------|-------------------------|--------------------------|-----------------------------|
| material | (n/s-g) | (α/s-g) | (n/s-g) | (n/s-g) |
| U-232ª | 1.3 | 8.0 × 10 ¹¹ | 1.49 × 10 ⁴ | 2.6 × 10 ⁶ |
| U-233ª | 8.6 × 10 ⁻⁴ | 3.5 × 10 ⁸ | 4.8 | 7.0 × 10 ² |
| U-234 ^a | 5.02 × 10 ⁻³ | 2.3 × 10 ⁸ | 3.0 | 5.8 × 10 ² |
| U-235ª | 2.99 × 10 ⁻⁴ | 7.9 × 10 ⁴ | 7.1 × 10 ⁻⁴ | 8 × 10 ⁻² |
| U-236ª | 5.49 × 10 ⁻³ | 2.3 × 10 ⁶ | 2.4 × 10 ⁻² | 2.9 |
| U-238ª | 1.36 × 10 ⁻² | 1.2 × 10 ⁴ | 8.3 × 10 ⁻⁵ | 2.8 × 10 ⁻² |
| NU ^b | 5.45 × 10 ⁻³ | 3.07 × 10 ⁴ | 3.04 × 10 ⁻⁴ | 6.66 × 10 ⁻² |
| LEU ^b | 5.31 × 10 ⁻³ | 7.18 × 10 ⁴ | 8.55 × 10 ⁻⁴ | 1.75 × 10⁻¹ |
| HEU ^b | 7.69 × 10 ⁻² | 3.51 × 10⁵ | 4.27 × 10 ⁻³ | 7.73 × 10 ⁻¹ |
| High burn-up VHEU ^c | 4.18 × 10 ⁻³ | 3.69 × 10 ⁶ | 4.63 × 10 ⁻² | 7.43 |
| Lower burn-up VHEU ^c | 1.61 × 10 ⁻³ | 2.83 × 10 ⁶ | 3.52 × 10 ⁻² | 6.44 |
| WGU recycled VHEU ^c | 1.19 × 10 ⁻³ | 2.62 × 10 ⁶ | 3.38 × 10 ⁻² | 6.48 |
| Recycled NU ^c | 1.35 × 10 ⁻² | 2.44 × 10 ⁴ | 2.38 × 10 ⁻⁴ | 5.74 × 10 ⁻² |
| Recycled LEU ^c | 1.33 × 10 ⁻² | 5.97 × 10 ⁴ | 6.96 × 10 ⁻⁴ | 1.45 × 10 ⁻¹ |
| Recycled DU ^c | 1.36 × 10 ⁻² | 1.48 × 104 | 1.14 × 10 ⁻⁴ | 3.39 × 10 ⁻² |

Table 2-2. Neutron production in various uranium isotopes and materials.

a. Spontaneous fission neutron yields and (α, n) reactions in oxides and fluorides from Tables 6-4 and 6-5 of DOE (2009).

b. See Table 2-1 of this report and uranium compositions (wt %) from Table A-2 of NAS (2005).

c. See Table 2-1 of this report and RU compositions (wt %) from Table 5-7 of ORAUT (2012).

The potential for significant worker exposures to neutrons generated by (α, n) reactions in uranium compounds is not very high unless a worker spends a large amount of time near uranium fluoride or uranium oxide compounds storage containers, or in processing areas for large quantities of those materials (DOE 2009). At very high ²³⁵U enrichments, the N:P dose ratio can be as much as 2:1 and neutrons can be the limiting radiation source for whole-body exposure (DOE 2009). As stated previously, there is a significantly limited basis for this information from DOE (2009). The N:P ratio of 2:1 should be considered an upper bound of the potential range of ratios seen in facilities handling

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HEU. The neutron doses from low-enriched ²³⁵U compounds or from uranium metals are not limiting doses (DOE 2009).

With respect to neutron energy spectra for HEU, Y-12 National Security Complex – Occupational External Dosimetry provides data for HEU storage areas at the Y-12 Plant that is summarized below in Table 2-3.

| Neutron energy group | Dose fraction | | | |
|-----------------------|---------------|--|--|--|
| <10 keV | 0.012 | | | |
| 10–100 keV | 0.003 | | | |
| 0.1–2 MeV | 0.970 | | | |
| 2–20 MeV | 0.015 | | | |
| Sources ODALIT (2000) | | | | |

Table 2-3. Neutron dose fractions for HEU storage areas.

Source: ORAUT (2009).

3.0 **NEUTRON DOSE DATA FROM FACILITIES**

3.1 **GASEOUS DIFFUSION PLANTS**

Studies have been published about measured neutron doses and N:P ratios near storage cylinders containing any DU, NU, LEU, or HEU in the form of UF₆ at the Paducah, Portsmouth, and K-25 gaseous diffusion plants. In some cases, these studies focused on large (14-t) cylinders in outside storage yards, while others were measurements of smaller HEU storage cylinders inside the facilities.

3.1.1 **Paducah Gaseous Diffusion Plant**

At the Paducah plant, measurements during a UF_6 cylinder painting project in the cylinder storage yard gave values for neutron-to-photon dose ratios ranging from 0.14 to 0.42 with an approximate average of 0.2 (BJC 1999). These cylinders, however, contained either DU or NU.

3.1.2 **Portsmouth Gaseous Diffusion Plant**

Neutron Dose Rate Data

Cardarelli (1997) quotes values for measurements of the neutron dose near small (5-in.-diameter) HEU storage containers that ranged from 3 mrem/hr (contact) to <0.5 mrem/hr (at 1 m). These values are in reasonably good agreement with the values in DOE (2009). This report also cites the neutron dose component of total external dose as 12.5% based on a monitoring study of uranium material handlers in 1995. It is important to note that this study included workers exposed to a variety of uranium enrichments.

In 1992, measurements taken in the PW (product withdrawal) vault (Building X-326) at Portsmouth indicated a neutron dose rate of 0.8 mrem/hr at a distance of 0.6 to 1.3 m facing a group of HEU cylinders (Soldat and Tanner 1992). Another survey was conducted in 1995 of a single 5A cylinder (97% HEU) in the X-345 Building. The recorded neutron dose rates at that time were 2.1 mrem/hr and 0.77 mrem/hr at distances of 15.2 cm and 30.5 cm, respectively (Scherpelz and Murphy 1995). Unfortunately, photon measurements were not made in either of these two studies.

Neutron and photon rate measurements were documented in a 1986 memo for four empty and six full 5-inch product cylinders stored in the X-326 PW vaults. Surface neutron and photon dose rates for the empty cylinders were ≤0.5 mrem/hr and ≤1.0 mR/hr respectively (N:P ratio = 0.5). Neutron and photon dose rates for the full cylinders were ≤3.0 mrem/hr and ≤2.4 mR/hr respectively (N:P ratio = 1.25). At a distance of 1 meter, the combined neutron and photon dose rate was 0.2 mrem/hr for empty cylinders and 0.5 mrem/hr for full cylinders (Bassett 1986).

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N:P Ratio Data from Area Monitoring Measurements

A summary memorandum from 2008 (McGuire 2008) provides annual Portsmouth area dosimetry data that was collected to meet the requirements of *Dosimetry Program Standards* (Madder and Turner 1997; Dodd, Duncan, and Teeters 2001, 2002). Outdoor annual neutron and photon dose measurements for the period from 2004 to 2008 were available for the following radiological facilities: X-326, X-330, X-333, X-342, X-343 X-344, X-530, X-705, X-710, and X-745. Data were also provided for nonradiological facility areas as well as Perimeter Road near the cylinder yards. Paired positive photon and neutron measurements were recorded at the following facilities or areas: X-330, X-344, X-343, X-530, X-745, and Perimeter Road near the cylinder yards. The average N:P ratio derived from this data was 0.147 ±0.09 with a minimum value of 0.01 and a maximum value of 0.398.

N:P Ratio Data from Employee Dosimetry Measurements, 1992 to 2013

Four databases containing historical dosimetry records for Portsmouth employees were reviewed in order to calculate a favorable to claimant N:P ratio. The databases reviewed are as follows:

- 3_Individual_Dose_Ports (overlaps with REMS_Portsmouth 1992-2013 Exp Recs)
- Extra_REIRS_Ports (overlaps with 3_Individual_Dose_Ports)
- REIRS_Portsmouth 1992-2013 Exp Recs
- REMS_Portsmouth 1992-2013 Exp Recs

The Portsmouth REIRS database was not useful in calculating an N:P ratio because no neutron dose records were included. Also, as indicated above, two of the databases were actually subsets of other databases. Thus, only the REMS_Portsmouth 1992-2013 database was used. Positive paired neutron and photon doses from this database were used to calculate a favorable to claimant N:P ratio that can be applied to certain workers in facilities where enriched uranium was handled. It should be noted that positive neutron results appeared in the REMS database beginning in 1999 (Bechtel Jacobs era).

Evaluation of REMS_Portsmouth 1992-2013 Exp Recs Database

A total of 38,964 records are contained in the Portsmouth REMS database. These encompass the period 1992 to 2013. A review of these records revealed the following information:

- 10,721 unique individuals were monitored by external dosimetry during this period.
- Of the 10,721 individuals monitored, 722 were also monitored for neutrons (7%) at one time or another.
- This database indicates that neutron monitoring began at Portsmouth in 1996, although the hardcopy records in the NIOSH Division of Compensation Analysis and Support Claimant Tracking System indicate an earlier date:
 - 1987 sporadic monitoring for a few individuals, and
 - Fall 2005 widespread monitoring.
- It should be noted that United States Enrichment Corporation (USEC) dosimetry records were included in a separate database beginning in 1997 (REIRS).
- Of the 722 individuals monitored for neutrons, 56 had at least one neutron monitoring result that exceeded the neutron limit of detection (LOD) of 10 mrem.

- Positive neutron results do not appear in the REMS database until 1999. HPRS printouts in NOCTS, however, indicate positive neutron dose as early as 1987 for one individual (see [Redacted]).
- USEC personnel wore a combined thermoluminescent dosimeter (TLD).
- DOE personnel wore an albedo neutron dosimeter separate from their regular issued TLD.

Table 3-1 shows a breakdown of monitored individuals by year, based on a review of the REMS database:

| Table 3- | 1. Portsmouth employee dosimetry | uala, 1992 lu 2013. |
|----------|----------------------------------|---------------------------------|
| | Number of individuals monitored | Number of individuals |
| Year | for total external dose | monitored for neutron dose (ND) |
| 1992 | 3,628 | 0 |
| 1993 | 3,589 | 0 |
| 1994 | 4,404 | 0 |
| 1995 | 4,485 | 0 |
| 1996 | 3,957 | 444 (all ND) |
| 1997ª | 144 | 0 |
| 1998 | 176 | 4 (all ND) |
| 1999 | 204 | 36 |
| 2000 | 421 | 30 |
| 2001 | 639 | 32 |
| 2002 | 640 | 39 |
| 2003 | 541 | 26 |
| 2004 | 650 | 25 |
| 2005 | 643 | 64 |
| 2006 | 762 | 40 |
| 2007 | 731 | 31 |
| 2008 | 803 | 33 |
| 2009 | 1,219 | 95 |
| 2010 | 1,544 | 144 |
| 2011 | 2,718 | 142 |
| 2012 | 2,737 | 184 |
| 2013 | 2,662 | 147 |

Table 3-1. Portsmouth employee dosimetry data, 1992 to 2013.

Source: Portsmouth REMS Database – 1992-2013.

a. Databases split in 1997; REIRS database beginning in 1997 contains all USEC employees but no neutron monitoring results.

Development of N:P Ratio Based on Portsmouth Dosimetry Data

A total of 161 records in the REMS database indicate neutron dose values greater than the LOD of 10 mrem. These values were divided by their paired photon dose values to calculate an N:P ratio for each line of data. These ratios were then averaged to obtain an overall N:P ratio. If a positive neutron dose had a corresponding photon dose of less than its LOD of 10 mrem, the photon dose was set equal to 10 mrem. This occurred with 10 records. In addition, a doubling of the radiation weighting factor from 10 to 20 was accounted for beginning in 2010. Based on this approach, the average N:P ratio was calculated to be 0.369 ± 0.2 .

3.1.3 K-25 Gaseous Diffusion Plant

Neutron Dose Rate Data

Discussions of N:P ratio values were found in a variety of documents written to address radiological safety issues associated with the K-25 cylinder storage yards. In a May 2000 report, an N:P ratio of 0.25 was cited with the notation that many of the neutron dose rate measurements were zero (BJC

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2000). A Final Safety Analysis Report for the K-25 cylinder yards, written in 1997, indicated contact dose rate values between 1 to 6 mrem/hr and general area dose rates between 1.5 to 2 mrem/hr. The report further indicated that neutron dose was one-sixth of the dose received from photon radiation (N:P = 0.166) (LMES 1997). Data from an area characterization study done along the outside boundary of the K1066E cylinder yard in 2000 indicated an N:P ratio of 0.274. This value was based on nine locations that had positive neutron values out of a total of 86 sampling points (DOE 2000).

N:P Ratio Data from Modeling Study

A modeling study for the neutron dose and N:P ratio from a typical array of large storage containers (14-t cylinders) of DU, NU, and LEU (5% enrichment) at the K-25 Plant can be found in *Determination of Neutron to Gamma Dose Ratios for the UF₆ Cylinder Yards* (NISYS 2000). The array of tanks consisted of 2 rows of cylinders stacked 2 cylinders high and 25 cylinders long on a 6-in. concrete pad. Calculations of the neutron dose and N:P ratio were made at distances ranging from 1 m to 300 m at the mid-length of the array of tanks. These calculations considered the effects of radiation scattering by the 6-in. thick concrete pad and by air with a relative atmospheric humidity of 30%, 60%, and 90% (NISYS 2000). For tanks containing DU, NU, and LEU (5% enrichment), at 1 m the N:P dose ratios were approximately 0.10, 0.22, and 0.88, respectively (NISYS 2000). The calculations at larger distances from the tanks containing DU, NU, and LEU (5% enrichment) were used to estimate bounding N:P ratios for radiation exposures at the tank yard of approximately 0.2, 0.4, and 1.5, respectively (NISYS 2000).

N:P Ratio from Equipment and Area Monitoring Surveys

A summary of K-25 survey data was provided in 2015 by East Tennessee Technology Park [(ETTP) previously known as the K-25 Site] personnel. The data cover the period January 1999 through November 2006 (at which time the last of the UF₆ cylinders were removed). Over 6,000 paired neutron and photon survey data points were available that corresponded with equipment and general area measurements. Analysis of the equipment survey data yielded N:P ratio values of 0.099, 0.085, and 0.122 for distances on contact, 1 foot, and 1 meter respectively. An N:P ratio of 0.605 ±0.408 was calculated from 445 neutron/photon data points associated with general area measurements (Pope 2015).

N:P Ratio Data from Employee Dosimetry Measurements, 1989 to 2012

In the past, dosimetry services for K-25 were provided by either the Oak Ridge National Laboratory (ORNL) or the Y-12 Plant. Therefore, to establish an N:P ratio, dosimetry records data for Oak Ridge Gaseous Diffusion Plant (K-25), also known as the East Tennessee Technology Park (ETTP), employees was extracted based on employee information and collated into a merged copy of the Y-12 Plant Dosimetry and ORNL Safety and Health Databases (dated February 6, 2013). Although integration of the two databases resulted in a version containing a variety of different tables, only records extracted from Table 3_Individual_Dose_ETTP (hereafter referred to as Table 3 ETTP) were used because the others: (1) did not have neutron dosimetry information, (2) contained duplicate data, (3) were limited to extremity dose, or (4) contained only Portsmouth Gaseous Diffusion Plant records.

Evaluation of Table 3 ETTP

Table 3 ETTP initially contained a total of 187,295 records; dates ranged from January 1, 1989, through December 31, 2012. Duplicate data were removed, leaving only unique dosimetry records for K-25 employees during the period of interest. Also of note:

- Personnel wore the Harshaw four-element beta/photon dosimeter and a separate four-element neutron dosimeter.
- 129,132 unique beta/photon dosimeters remained after duplicates were removed.

- 16,701 unique neutron dosimeters remained after duplicates were removed.
- 11,599 individuals were monitored for beta/photon radiation from 1989 to 2012.
- 404 individuals were monitored for neutrons from 1989 to 2012.
- Approximately 3.5% of the individuals were monitored for beta/photon and neutron radiation.
- The first positive neutron result occurred in 1993.
- Results from 369 neutron dosimeters were greater than or equal to the limit of detection (LOD).
- Neutron monitoring seldom occurred from 1990 to 1996, but seems to have been more frequent from 1999 to 2012.

Table 3-2 below tabulates the number of photon/beta and neutron dosimeters exchanged for K-25 employees by year.

| | Photon/beta dosimeter | Neutron dosimeter | Neutron dosimeters (results |
|------|-----------------------|-------------------|-----------------------------------|
| Year | exchanges | exchanges | > or = to the limit of detection) |
| 1989 | 232 | 0 | 0 |
| 1990 | 401 | 7 | 0 |
| 1991 | 359 | 5 | 0 |
| 1992 | 550 | 2 | 0 |
| 1993 | 461 | 4 | 1 |
| 1994 | 327 | 3 | 0 |
| 1995 | 169 | 6 | 0 |
| 1996 | 253 | 12 | 0 |
| 1997 | 6,005 | 79 | 1 |
| 1998 | 6,304 | 59 | 0 |
| 1999 | 5,920 | 183 | 9 |
| 2000 | 5,980 | 773 | 20 |
| 2001 | 7,107 | 1,138 | 35 |
| 2002 | 7,002 | 1,415 | 11 |
| 2003 | 8,096 | 1,769 | 21 |
| 2004 | 10,239 | 1,923 | 26 |
| 2005 | 12,515 | 2,283 | 20 |
| 2006 | 10,637 | 2,030 | 25 |
| 2007 | 9,040 | 877 | 60 |
| 2008 | 8,137 | 959 | 44 |
| 2009 | 8,419 | 904 | 19 |
| 2010 | 8,618 | 857 | 54 |
| 2011 | 7,518 | 852 | 20 |
| 2012 | 4,843 | 561 | 3 |

Table 3-2. K-25 annual photon/beta and neutron badge exchanges from 1989 to 2012.

Source: K-25_PORTS_Exposure Database.

A total of 369 records in Table 3 ETTP had individual neutron doses greater than or equal to the LOD of 10 mrem. An analysis of dosimeters with paired photon and neutron doses greater than the LOD was performed. The N:P ratio was calculated by dividing the neutron dose by the deep dose for each badge exchange. A ratio of 0.490 was calculated based on the average of the paired N:P ratios for the period January 1, 1989 to December 31, 2012. Starting January 1, 2010, ORNL adopted the 1990 International Commission on Radiological Protection (ICRP) recommendations on neutron

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weighting factors (O'Connell 2011). Based on this information, an adjusted N:P ratio of 0.420 was calculated for the period January 1, 1989 to December 31, 2012.

3.2 Y-12 PLANT

The Y-12 Plant site profile discusses historical dosimetry information that can be used to determine neutron and photon exposure near HEU operations (ORAUT 2009). For this report, individual worker quarterly data for pairs of neutron and photon doses from workers in Y-12 Plant Departments 2301 (Development Operations) and 2160 (Material Engineering) during the period from 1955 to 1959 were combined and used as a basis for obtaining an average N:P ratio. These data were selected because unique work activities involving EU were carried out in these departments during this period as indicated in *Recycled Uranium Mass Balance Project Y-12 National Security Complex* (BWXT 2000). Excerpts from this report stated (BWXT 2000):

- In the late 1950s, continuous solvent extraction equipment was installed in the B-1 wing (of Building 9212). This period covered the transition from small-scale batch operations to the existing continuous recovery operation equipment in use today.
- The 9206 building has been used extensively over its lifetime for the chemical processing of uranium.
- Enriched uranium processes, activities, and/or missions of the 9206 Facility have included: conversion of UF₆ to UF₄ to uranium metal for weapons (1954 to 1964) and casting and machining of HEU metal for weapons (1955 to 1965).

Film-badge data for Departments 2160 (Material Engineering) and 2301 (Project Design Development and Analytical Development) appear to be somewhat typical of the N:P ratio for workers who were exposed to neutrons from HEU sources in Buildings 9202, 9206, and 9212. The data analysis shown in Figure 4-3 is based on 90 paired N:P results for 47 different workers in Departments 2160 and 2301.

A study was conducted in 1989 of neutron and photon emissions from small storage containers of HEU in Building 9212 at the Y-12 Plant in Oak Ridge, Tennessee (Soldat et al. 1990). At the time of the measurements, the storage area in Building 9212 held small HEU containers of both UF₄ and UO_3 . These small HEU containers were placed on a rack of shelves and arranged in a matrix that was criticality safe (Soldat et al. 1990). The exact enrichment value was not given, but the nature of the work in Building 9212 (as well as Buildings 9202 and 9206, discussed below) would indicate an enrichment value of 90% or greater. The measurements were made at a height of 39 in. (1 m) above the floor and 27 in. (0.7 m) from the nearest container. The location of the measurements was also selected so that it was only near containers of UF₄ (Soldat et al. 1990). The characterization of the neutron field consisted of measurements of the neutron energy spectrum and personal dose equivalent from both neutrons and photons. The measured values for the neutron dose rate ranged from 0.7 to 2.08 mrem/hr. An N:P ratio was derived from this data by comparing the integrated neutron dose of 76.2 mrem for a period of 40.1 hours as determined from measurements made with a tissue equivalent proportional counter and the integrated photon dose of 68.0 mrem as measured by phantom-mounted thermoluminescent dosimeters over the same time period. The resulting N:P ratio for these measurements was 1.12.

Table 3-3 summarizes N:P ratio data based on survey information from the Y-12 Plant for the period 1992 to 2004. The readings were taken either in general work areas or at a distance of 30 cm.

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| Date | Building | Room | N:P Range | N:P Average | Reference |
|------------|-----------|---------|-----------|-------------|--------------------------------|
| 06/02/1992 | 9981/9998 | G-2/G-5 | 0.13–1.0 | 0.37 | Y-12 2013; SRDB Ref ID: 129045 |
| 02/28/2001 | 9212 | D-107 | 0.06-0.50 | 0.2 | Y-12 2013; SRDB Ref ID: 126485 |
| 07/11/2002 | 9212 | 1004 | 0.1–0.5 | 0.3 | Y-12 2013; SRDB Ref ID: 128511 |
| 12/05/2003 | 9212 | 1004 | 0.25–1.0 | 0.6 | Y-12 2013; SRDB Ref ID: 128511 |
| 12/08/2003 | 9212 | 1004 | 0.25–1.0 | 0.6 | Y-12 2013; SRDB Ref ID: 128511 |
| 05/25/2004 | 9212 | 1014 | 0.22-0.43 | 0.31 | Y-12 2013; SRDB Ref ID: 126498 |

Table 3-3. Summary of Y-12 Plant N:P ratio data, 1992 to 2004.

4.0 <u>ADDITIONAL DATA ANALYSIS OF EMPLOYEE DATA MEASUREMENTS FOR K-25,</u> <u>PORTSMOUTH, AND Y-12 PLANT</u>

An additional method of analysis was applied to the employee dosimetry datasets for K-25 and Portsmouth. This method – quantile regression – is discussed in ORAUT-RPRT-0087, *Applications of Regression in External Dose Reconstruction* (ORAUT 2018). In summary, this method uses quantile regression to fit a linear model to the neutron and photon doses (not an N:P ratio), giving the 50th-and 95th-percentile regression fits directly – with the added benefit of requiring no special treatment of data that might yield a traditional N:P ratio of 1. This approach also allows for exploratory data analysis (i.e., deriving fits for the 50th- and 95th-percentile exposed populations).

Table 4-1 provides the results of the quantile regression fits for K-25, Portsmouth, and Y-12 Plant – and data for all three sites combined. In the case of K-25, the entire dataset from 1989 to 2012 was used since the site was in a single (post-production) mode during that era. The 50th- and 95th-percentile regression fits for K-25 can be seen in Figure 4-1. At Portsmouth, enrichment activities ended in 2001, so the fits seen in Figure 4-2 are based on data from 1992 to 2001. Figure 4-3 illustrates the fits for Y-12 Plant from 1955 to 1959. Finally, Figure 4-4 illustrates the fits for combining the datasets for all three sites.

Table 4-1. Summary of quantile regression fit parameters for K-25, Portsmouth, and Y-12 Plant employee data measurements.

| | | 50th- percentile | 50th-percentile intercept | 95th- percentile | 95th-percentile intercept |
|----------------------------|-------|---------------------|------------------------------|---------------------|------------------------------|
| Site | N | slope | (rem) | slope | (rem) |
| K-25 (1989–2012) | 375 | 0.139 | 0.01 | 0.112 | 0.03 |
| Portsmouth, Production Era | 3,727 | 0.133 | 0.002 | 0.231 | 0.004 |
| (1992–2001) | | | | | |
| Y-12 Plant (1955–1959) | 89 | 0.14 | 0.03 | 0.7 | 0.1 |
| K-25/Portsmouth/Y-12 Plant | 4,191 | 0.195 | 0.002 | 0.846 | 0.002 |
| Combined | | | | | |

The slope data for K-25, Portsmouth during the production era, and the Y-12 Plant show good agreement – especially at the 50th-percentile. Please note that production of HEU product at Portsmouth ended in 1991, therefore, this production dataset represents work with mostly LEU product.







Figure 4-2. Quantile regression results for Portsmouth employee dosimetry data, production era (1992–2001).







Figure 4-4. Quantile regression results for the combined K-25, Y-12, and Portsmouth employee dosimetry data sets. (Data for K-25 are in red, data for Portsmouth are in blue, and data for Y-12 are in black).

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5.0 PHOTON SUMMARY AND CONCLUSIONS

Table 5-1 provides a summary of the neutron dose rates, photon dose rates (where available), and N:P ratios (where available) associated with the measurements and calculations described above. Neutron dose rate measurements were in good agreement with the values in DOE (2009).

At locations where NU and DU were present, an N:P ratio of 0.2 was observed (Paducah and K-25).

At facilities where material with higher uranium enrichment was present, an increase in the N:P ratio was observed. A close-proximity (1 m) N:P ratio of 0.88 was calculated for LEU (5%) at the K-25 storage yard. Survey and area monitoring N:P values at the K-25 and Portsmouth gaseous diffusion plants ranged from 0.1 to 0.605. N:P ratios ranging from 0.41 to 1.12 (surveys and stationary measurement) were found for HEU material at the Y-12 Plant.

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Table 5-1. Summary of neutron dose rates, photon dose rates, and N:P ratios associated with natural, depleted, and enriched uranium materials at Paducah, Portsmouth, and K-25 Gaseous Diffusion Plants and Y-12 Plant.

| | | Measurement | Neutron dose rate | Photon dose rate | |
|--|---------------------|--------------|----------------------|---------------------|-----------------------|
| Location/reference | Material | distance | (mrem/hr) | (mrem/hr) | N:P ratio |
| K-25 GDP, Cylinder Storage Yard (NISYS 2000) | DU | 1 m | 0.07 | 0.7 | 0.1 |
| Paducah GDP, Cylinder Storage Yard (BJC 1999) | DU, NU | Not given | NAª | NA | 0.2 (0.14-0.42) |
| K-25 GDP, Cylinder Storage Yard (NISYS 2000) | NU | 1 m | 0.15 | 0.68 | 0.22 |
| DOE Good Practices Guide (DOE 2009) | NU to 5% LEU | Not given | 0.01-0.2 | NA | 2.0 |
| K-25 GDP, Cylinder Storage Yard (NISYS 2000) | 5% LEU | 1 m | 0.6 | 0.68 | 0.88 |
| Y-12 Plant, Bldg. 9212 (Soldat et al., 1990) | HEU | Not given | 1.9 | 1.7 | 1.12 |
| Y-12 Plant, Bldg. 9981/9998 (Y-12 2013) | HEU | 1 ft | 2-32 | 1-4 | 0.37 (0.13-1.0) |
| Y-12 Plant, Bldg. 9212, Rm D-107 (Y-12 2013) | HEU | 1 ft | 1-1.8 | 0.1-0.5 | 0.2 (0.06-0.50) |
| Y-12 Plant, Bldg. 9212, Rm 1004 (Y-12 2013) | HEU (SRS/UF4) | 1 ft | 0.4-8 | 0.2-0.4 | 0.3 (0.1-0.5) |
| Y-12 Plant, Bldg. 9212, Rm 1004 (Y-12 2013) | HEU | 1 ft | 0.1-0.4 | 0.1 | 0.6 (0.25-1.0) |
| Y-12 Plant, Bldg. 9212, Rm 1004 (Y-12 2013) | HEU | 1 ft | 0.1-0.5 | 0.1 | 0.6 (0.25-1.0) |
| Y-12 Plant, Bldg. 9212, Rm 1014 (Y-12 2013) | HEU | 1 ft | 1.4-1.8 | 0.4-0.6 | 0.31 (0.22-0.43) |
| Portsmouth GDP (Cardarelli 1997) | HEU | Contact | 3 | NA | 0.125 |
| DOE Good Practices Guide (DOE 2009) | 97% HEU | Contact | 2-4 | NA | 2 |
| Portsmouth GDP, X-326, Full 5" Cylinders (Bassett 1986) | 97% HEU | Contact | <3.0 | <2.4 | 1.25 |
| DOE Good Practices Guide (DOE 2009) | 97% HEU | 1 m | 1-2 | NA | 2 |
| K-25, FSAR (LMES 1997) | DU, NU, LEU, HEU | Contact | NA | NA | 0.166 |
| K-25 Equipment and Area Monitoring Surveys (Pope 2015) | DU, NU, LEU, HEU | Contact | NA | NA | 0.099 |
| K-25 Equipment and Area Monitoring Surveys (Pope 2015) | DU, NU, LEU, HEU | 1 ft | NA | NA | 0.085 |
| K-25 Equipment and Area Monitoring Surveys (Pope 2015) | DU, NU, LEU, HEU | 1 m | NA | NA | 0.122 |
| Portsmouth GDP (Cardarelli 1997) | HEU | 1 m | <0.5 | NA | 0.125 |
| K-25 Equipment and Area Monitoring Surveys (Pope 2015) | DU, NU, LEU, HEU | General area | NA | NA | 0.605 |
| K-25, FSAR (LMES 1997) | DU, NU, LEU, HEU | General area | NA | NA | 0.166 |
| Portsmouth Area Monitoring Results (McGuire 2008) | DU, NU, LEU, HEU | Not given | NA | NA | 0.147 (0.01-0.398) |

a. NA = not applicable.

Data based on analyses of personnel dosimeter measurements at these facilities (see Table 5-2) yielded N:P values between 0.369 and 0.420 and, for comparison, quantile regression 50th- and 95th-percentile relationships yielding slope values ranging from 0.133 to 0.231 (where the y-intercepts are close to zero) respectively during eras after active HEU production had ceased. Finally, quantile regression 50th- and 95th-percentile relationships yielding slope values ranging from 0.14 to 0.7

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respectively (again, where the y-intercept was close to zero) were found for workers actively handling HEU product at the Y-12 Plant.

Although a wide range of survey, modeling, and static dosimetry data is available for all three sites, the quantile regression results from each site's personnel dosimetry measurements would yield the most accurate N:P ratio result. In addition, good agreement was seen – especially when comparing the slope values at the 50th-percentile level – between the personnel measurement results from all three sites.

Table 5-2. Summary of N:P ratios and quantile regression relationships associated with natural, depleted, and enriched uranium materials at Portsmouth, K-25, and Y-12 Plants, based on personnel dosimetry.

| | | N:P | |
|----------------------|------------------|-------|---|
| Location/reference | Material | ratio | Quantile regression relationship |
| K-25 Personnel | DU, NU, LEU, HEU | 0.420 | 50th neutron = 0.139 [photon (rem)] + 0.01 (rem) |
| Dosimetry, 1989–2012 | | | 95th neutron = 0.112 [photon (rem)] + 0.03 (rem) |
| Portsmouth Personnel | DU, NU, LEU, HEU | 0.369 | 50th neutron = 0.133 [photon (rem)] + 0.002 (rem) |
| Dosimetry, 1992–2013 | | | 95th neutron = 0.231 [photon (rem)] + 0.004 (rem) |
| Y-12 Plant Personnel | HEU | NAa | 50th neutron = 0.14 [photon (rem)] + 0.025 (rem) |
| Dosimetry, 1955–1959 | | | 95th neutron = 0.7 [photon (rem)] + 0.098 (rem) |
| K-25/Portsmouth/Y-12 | DU, NU, LEU, HEU | NAa | 50th neutron = 0.195 [photon (rem)] + 0.002 (rem) |
| Plant Combined | | | 95th neutron = 0.846 [photon (rem)] + 0.002 (rem) |

a. NA = not applicable.

The personnel data take into account the actual distances and residence time personnel would have had near potential source terms, whereas the survey data – by their nature – are static measurements taken during a snapshot of time. Survey data are typically taken at maximum dose rate locations to set bounds for radiological control purposes, thus, the survey data can be considered an upper bound. Most of the surveys are indicating N:P ratios less than 0.4:1 with only three slightly greater than 1.2:1. This further confirms that the 2:1 upper bound N:P ratio discussed in DOE (2009) is indeed a conservative upper bound. Given multiple work locations and personnel movement between areas of neutron exposure with N:P ratios less than 0.4 – and areas of no neutron exposure – the data resulting from the quantile regression analysis of personnel dosimetry provides the best estimate for an overall N:P ratio for unmonitored workers.

An N:P relationship based on the quantile regression analysis of dosimetry data for all three sites combined – as summarized in Tables 4-1 and 5-2 – is recommended for assigning neutron dose for unmonitored workers in eras as described above. Application of the guidance in Attachment A yields a lognormal distribution that reflects the 50th- and 95th-percentile fits for the quantile regression analysis of the personnel dosimetry data from K-25, Portsmouth, and Y-12 Plant combined. This distribution is to be combined with photon dose (defined as a constant or distribution) – using Monte Carlo methods as needed – to yield a distribution of neutron dose.

6.0 ATTRIBUTIONS AND ANNOTATIONS

All information requiring identification was addressed via references integrated into the reference section of this document.

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ATTACHMENT A QUANTILE REGRESSION ANALYSIS IMPLEMENTATION WITHIN IREP

Using a quantile regression analysis of paired photon / neutron data, an N:P ratio can be derived for calculating neutron dose based on photon dose using the following inputs (note that this N:P ratio derivation is needed to implement the quantile regression results in the calculation tools used by the ORAU Team):

- 1. 50th-percentile equation,
- 2. 95th-percentile equation, and
- 3. Known photon dose (normal or lognormal distribution).

The neutron dose at any photon dose is assumed to have a lognormal distribution, the parameters of which are described by the 50th- and 95th-percentile equations.

In this discussion, $N_{50}(p)$ denotes the 50th-percentile neutron dose for a given photon dose *p* and $N_{95}(p)$ denotes the 95th-percentile neutron dose for a given photon dose (*p*).

The lognormal parameters for the N:P ratio of a given photon dose are:

$$GM(p) = \frac{N_{50}(p)}{(p)}$$
 (A-1)

where

 $z_{95} = 1.64485$ (normal standard deviation for the 95th-percentile)

and

$$GSD(p) = e^{\left(\frac{\ln\frac{N_{95}(p)}{N_{50}(p)}}{Z_{95}}\right)}$$
(A-2)

To obtain the neutron dose, the derived N:P ratio is multiplied by the photon dose distribution. (See example below).

Example Calculation:

N:P ratio from quantile regression relationship from K-25/Portsmouth/Y-12 Plant combined: $N_{95}(p) = 0.846p + 0.002$ $N_{50}(p) = 0.195p + 0.002$ Photon dose p = 0.05 rem

Calculation of N:P Ratio from Photon Dose and Quantile Regression Neutron Dose

$$GM(p) = \frac{0.011366}{0.05} = 0.22732 \tag{A-3}$$

and

ATTACHMENT A QUANTILE REGRESSION ANALYSIS IMPLEMENTATION WITHIN IREP (continued)

$$GSD(p) = e^{\left(\frac{\ln \frac{0.0444575}{0.011366}}{1.64485}\right)} = 2.291481$$
(A-4)

Monte Carlo Simulation to Assign Neutron Dose

Photon dose (p) = 0.05 rem ±0.015 rem (normal distribution) N:P ratio: GM = 0.22732, GSD = 2.291481 (lognormal distribution)

A sample from the normal photon dose distribution is multiplied by a sample from the derived N:P ratio. This process is repeated for 10,000 iterations and a distribution is fit to the resulting data set (see results in Figure A-1).



Figure A-1. Lognormal distribution fit of calculated neutron data based on photon sample and N:P ratio sample. Neutron dose lognormal fit from photon sample: 0.05 ± 0.015 rem. (The parameters for this fitted lognormal are: Mean = 0.016 rem, Standard Deviation = 0.015 and, for use with IREP: GM = 0.0135 rem, GSD = 2.090, and Shift = -0.00198)