



# ORAU TEAM Dose Reconstruction Project for NIOSH

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07/12/2011	00	New report to consolidate the information pertinent to the Feed Materials Production Center (FMPC) internal dosimetry that has been developed since the SEC Evaluation Report was presented. This information includes topic discussed in the Advisory Board FMPC Work Group meetings. Several distinct topics are included in this report. Incorporates formal NIOSH review comments. Training required: As determined by the Objective Manager. Initiated by Karin Jessen.

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**ACRONYMS AND ABBREVIATIONS**

AEC	U.S. Atomic Energy Commission
AMC	African Metals Company
AWE	Atomic Weapons Employer
Bq	becquerel
cfm	cubic feet per minute
CFR	Code of Federal Regulations
Ci	curie
d	day
DAC	derived air concentration
DCAS	Department of Compensation Analysis and Support
DOE	U.S. Department of Energy
dpm	disintegrations per minute
dps	disintegrations per second
DWE	daily weighted exposure
EEOICPA	Energy Employees Occupational Illness Compensation Program Act of 2000
FMPC	Feed Materials Production Center
ft	feet
g	gram
gal	gallon
GDP	gaseous diffusion plant
HASL	Health and Safety Laboratory
hr	hour
ICRP	International Commission on Radiological Protection
IMBA	Integrated Modules for Bioassay Analysis
in.	inch
K	Kelvin
kg	kilogram
L	liter
lb	pound
LEU	low-enriched uranium
LOOW	Lake Ontario Ordnance Works
LPF	leak path factor
m	meter
MAC	maximum allowable concentration
MCW	Mallinckrodt Chemical Works
MDA	minimum detectable amount
MIVRML	Mobile In Vivo Radioactivity Measurements Laboratory
mL	milliliter
MPC	maximum permissible concentrations
mph	miles per hour

MPLB	maximum permissible lung burden
MT	metric ton
MTU	metric ton of uranium
nCi	nanoCurie
NIOSH	National Institute for Occupational Safety and Health
NLO	National Lead of Ohio
NU	natural uranium
ORAU	Oak Ridge Associated Universities
pCi	picocurie
PGDP	Paducah Gaseous Diffusion Plant
POOS	plutonium out of specification
PORTS	Portsmouth Gaseous Diffusion Plant
ppb	parts per billion
ppm	parts per million
PUREX	plutonium-uranium extraction
RAC	Radiological Assessments Corporation
RFP	Rocky Flats Plant
RGM	radon gas monitor
RMI	Reactive Metals, Inc.
RU	recycled uranium
s	second
SC&A	S. Cohen & Associates
SEC	Special Exposure Cohort
SRDB Ref ID	Site Research Database Reference Identification (number)
SRS	Savannah River Site
t	ton (2,000 lb)
TASL	Track Analysis Systems, Ltd.
TRU	transuranic
TWA	time-weighted average
UNH	Uranyl nitrate hexahydrate
W	watt
wk	week
yr	year
°F	degrees Fahrenheit
μCi	microCurie
μg	microgram

## 1.0 INTRODUCTION

The purpose of this report is to consolidate the information on internal dosimetry at the Feed Materials Production Center (FMPC) that has been developed since the Special Exposure Cohort (SEC) Evaluation Report was presented (NIOSH 2006). This information includes topics discussed in the Advisory Board FMPC Work Group meetings. Several topics are included in this report; each stands alone in its own section. The following topics are included in this report:

- Radon releases from K-65 silos (Section 2.0),
- Exposure to Q-11, K-65, domestic uranium, and raffinates (Section 3.0),
- Recycled uranium (RU) (Section 4.0), and
- Use of daily weighted exposure (DWE) data to determine thorium intake rates (Section 5.0).

In addition, the National Institute for Occupational Safety and Health (NIOSH) has proposed a coworker model to describe the intake rates of thorium based on chest count data. That model, which is applicable from 1968 through 1988, is not included in this report but is expected to be published at a future date.

Attributions and annotations, indicated by bracketed callouts and used to identify the source, justification, or clarification of the associated information, are presented in Section 6.0.

## 2.0 RADON RELEASES FROM K-65 SILOS

### 2.1 BACKGROUND

Original approach defining the amount of radon released from the K-65 silos is defined in the site profile (ORAUT 2006b). In that document NIOSH estimated a  $^{222}\text{Rn}$  release rate of 5,000 to 6,000 Ci/year.

In November 2008, Sanford Cohen & Associates (SC&A) prepared a white paper, *An Alternative Assessment of Radon Releases from K-65 Silos* (Alternative Assessment; SC&A 2008) that provided assumptions and quantitative calculations to demonstrate its claim of the potential for  $^{222}\text{Rn}$  releases between 64,500 to 92,000 Ci/yr from Silo 1 at FMPC. In response to the Alternative Assessment, NIOSH prepared *Response to SC&A White Paper, An Alternative Assessment of Radon Releases from K-65 Silos* in November 2008 (NIOSH 2008), which stated:

*NIOSH disagrees with the draft findings reported by SC&A. The RAC [Radiological Assessments Corporation] model was supported by a National Academy of Sciences review; SC&A's was not. NIOSH will rely upon the individual exposure estimates produced using the Pinney/Hornung model, which utilized radon exposure levels from the RAC study, plus an additional radon source term at Fernald.*

Dr. John Mauro made the following statement in the Work Group Meeting on January 29, 2010 (ABRWH 2010, pp. 280-281):

*Please do one other thing for us. We certainly will look at those papers very carefully, and let's say we walk away from this. You read those papers and it looks pretty good, and then you take a look at our papers, at the arguments based on ... this deficit of lead-210, and you say, then where did the radon go? I mean, we've got ourselves quite a dilemma here because the radon had to go somewhere because of that deficit. Now, if we could somehow reconcile whatever you have here and our analysis, ... it would make me very happy that we could somehow reconcile how one analysis, which*

*is based on the radon deficit (our work), is coming up with some number, and I will look at the other work that was done and how they came up with theirs.*

Dr. Mauro reiterated this desire in his Division of Compensation Analysis and Support (DCAS)/SC&A action items status summary listing (Mauro 2010):

*Issue 5: Review of radon emissions from the K-65 silos*

*...during the meeting, the work group requested that NIOSH explicitly address the Pb-210 deficit arguments, and the external dose measurement arguments made by SC&A regarding this matter....*

In April 2010, SC&A issued *A Second White Paper Addressing Enhanced Radon Releases from the K-65 Silos at the Fernald Site* (Second White Paper; SC&A 2010) in response to NIOSH's rejection of the Alternative Assessment.

In May 2010, NIOSH prepared a response (NIOSH 2010) to the Second White Paper in which it responded to the stated issues on the same subject.

The manner by which K-65 radon release issues were raised as indicated above has resulted in the need to consolidate information in one document to provide a more cohesive and comprehensive response. To this end, this section focuses on responding to (1) the calculation methodology by which the enhanced radon releases were derived in the Alternative Assessment (SC&A 2008), (2) calculation modeling based on physical phenomena, and (3) the salient points made in the second White Paper (SC&A 2010) in relation to NIOSH's dose reconstruction position to "rely upon the individual exposure estimates produced using the Pinney/Hornung model, which utilized radon exposure levels from the RAC study, plus an additional radon source term at Fernald" (NIOSH 2008).

## **2.2 THE ALTERNATIVE ASSESSMENT OF RADON RELEASES FROM K-65 SILOS**

The Alternative Assessment (SC&A 2008) is based on the premise that the degree of disequilibrium between  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  in the K-65 waste medium determines the amount of radon released in the silo headspace. The  $^{210}\text{Pb}$  and  $^{226}\text{Ra}$  contents in the waste package were based on results from the radionuclide analyses of samples taken from the silos in 1991. The amount of radon released to the atmosphere before 1979 was based on the relationship of external dose rate on the top of the dome taken a minimum of 14 years apart.

The cumulative uncertainties of the bases for the quantities of radon release in the Alternative Assessment (SC&A 2008) are sufficiently large that they invalidate use of the derived radon releases. The following list identifies some of the prominent uncertainties that challenge the derivation of the release quantities in the Alternative Assessment:

1. The range of  $^{226}\text{Ra}$  concentrations in the 1991 sampling of the silos had shown that the K-65 material is not homogeneous. "The color of the material also varied greatly," stated RAC (1995). The samples were taken through the four former influent manholes on top of each silo, which are denoted by their direction from the center of the Silo (NE, SE, SW, and NW). At best, samples from each manhole would cover one-fourth of the K-65 materials. Table 2, "Results of Radionuclide Analyses on K-65 Material Obtained by ASI/IT from 1991 Sampling" in SC&A (2008) presented results of the samples from three of the four manholes (SE, NE, and NW). Implicitly, the radionuclide contents of a quarter of the K-65 materials were not measured. Confirmed by the fact that the K-65 material is not homogeneous, this introduces significant uncertainty to the radionuclide dataset for all K-65 materials.

2. The determination of radon release based on the argument of  $^{210}\text{Pb}$  deficit did not take into account the removal of lead during the chemical processing of the ore and handling of the raffinates. It is certain that the  $^{210}\text{Pb}$  could have plated out on the surface of the K-65 materials, as well as on the foam layer on top of the K-65 materials and on the inner concrete dome surfaces. On further examination of Table 2 in SC&A (2008), the mean  $^{210}\text{Po}$  content was found to be 242,000 pCi/g. This amount exceeded the mean  $^{210}\text{Pb}$  activity of 194,000 pCi/g, and must mean that  $^{210}\text{Pb}$  is still in the silos somewhere, because there is an excess of  $^{210}\text{Po}$ .
3. There is no evidence that the 1991 measured radionuclide contents can be extrapolated to the early years of the silos. As a point of reference, Table J-4 of RAC (1995) shows that significant variations existed in measurements from different periods. The activity concentration of radionuclides that have different solubility would be likely to change because of evaporation over time. It is predictable that greater differences would occur in the early years when the silo contents were in the process of settling down from previous loading operations.
4. The approach to relate measured external dose rates at the top of the dome to demonstrate buildup differences in the headspace is so uncertain as to be unusable. The Alternative Assessment (SC&A 2008) failed to ensure that the same equipment was used to measure the external dose rates on the silo domes. It is unlikely that the same dose rate meters were used from 1964 through 1987. The paper did not substantiate that the measurements were conducted in a highly proceduralized manner such that they were taken in the exact same geometry and in the exact same location on the silo dome. At least eight or nine different individuals took the dose rate measurements over the 23-year period, and it is highly unlikely that the measurements were taken in identical locations on the silo dome with the same geometries. In addition, the geometry of the silo dome varied over time. In fact, the concrete domes on the silos are thinnest in the center and thickest around the perimeter. By the 1970s, the degradation of the dome had advanced to a point where the inside of the concrete dome was spalling and likely resulted in a much thinner dome. The thinning of the concrete dome would dramatically decrease the gamma shielding and account for the observed increase in dose rates.
5. Section 3.4 of the report provides the basis for an estimate of  $^{226}\text{Ra}$  activity in K-65 Silos 1 and 2. The exact activity of  $^{226}\text{Ra}$  in the two silos is not known for certain, but the amount of activity is in the range from 1,650 to 4,600 Ci. The  $^{226}\text{Ra}$  activity in Silo 3 is expected to be less since the concentration in the Silo 3 material was lower, bringing the total to nominally 5000 Ci or less.  $^{226}\text{Ra}$  has a relatively long half-life of 1600 years so the radium activity is essentially unchanging during the period of concern. The activity of  $^{222}\text{Rn}$  is controlled by, and cannot exceed, the activity of  $^{226}\text{Ra}$ . Therefore the activity of  $^{222}\text{Rn}$  that is available to release is nominally 5000 Ci or less, which is in the range of values accepted by NIOSH, and much less than the range of values suggested in SC&A's alternate assessment.

Section 2.3 presents a simplified radon release rate calculation using a conventional modeling approach to illustrate the conservative nature of the RAC (1995) analytical method, for which the National Academy of Sciences review did not reveal technical issues that could lead to the prohibition on the use of its results.

## 2.3 RADON RELEASE CALCULATION USING CONVENTIONAL MODELING APPROACH

### 2.3.1 Calculation for Radon Release Rate from K-65 Material to the Silo Air

To reach the headspace of the silos, the radon from disintegration of  $^{226}\text{Ra}$  nuclides in the waste medium must (1) escape from the  $^{226}\text{Ra}$  particle of original residence to the pores of the medium through release of the kinetic energy of formation (recoil energy) and (2) migrate up through the medium materials. At the same time, the radioactive decay process removes quantities of radon depending on the time for the radon to migrate through about 20 ft of soil-like materials to the headspace.

The following is a description of radon transport through soil: Emanation is a several step process – first, the radon must escape from the particle in which the parent radium is contained through expending the kinetic energy of formation (recoil energy). Alpha recoil is defined as the process by which an atom (radon) recoils in the opposite direction from the path of particle ejection following the radioactive decay of its parent atom. Most of the radon atoms remain in the particle matrix without ever entering the pores in the medium.

Second, the process continues as radon is transported from a solid to a gas or liquid medium within the interstitial pores of the medium, i.e. after radon is produced at the soil particulate level from the radioactive decay of radium, it is released into small air or water containing pores between soil and rock particles. This transportation of radon through soil is primarily accomplished by the mechanical flow of air and/or water through the soil.

Third, after radon is released into the pore spaces, the efficiency of its eventual release into ambient air [in the case of the K-65 silos, the silo headspace], termed exhalation, is a function of:

The soil porosity

The concentration of radon in the soil/gas pore

a. Meteorological factors, including, precipitation and atmospheric pressure.

The K-65 silo radon production rate is generally constrained by existing radon in the air spaces. However, to ensure favorability to the claimant, an unconstrained production rate (no radon to inhibit release from the K-65 material) is calculated in the next section.

#### 2.3.1.1 Radon Exhalation Rate from K-65 Material in the Silos

The exhalation rate for radon permeated from the soil can be estimated based on the conservation of mass; an idealized, one-dimensional, steady-state model for the transportation and distribution of radon in the soil can be expressed as the following differential equation (Jasaitis and Girgzdys 2007):

$$D_e \frac{d^2 C}{dx^2} - \lambda C + \frac{A}{p_{ef}} = 0 \quad (1)$$

where

$D_e$  = the effective radon diffusion coefficient ( $\text{m}^2/\text{s}$ )

$C$  = the radon concentration in pore air of the soil ( $\text{Bq}/\text{m}^3$ )

$x$  = a distance from the ground surface with its positive direction downward (m)

$\lambda$  = the radon decay constant (1/s)

$p_{ef}$  = the effective porosity of the soil

$A$  = the production rate of radon gas in the pore space; defined as Equation 2:

$$A = \lambda p R E \quad (2)$$

where

- $\rho$  = the dry bulk density of the soil (kg/m<sup>3</sup>)
- $R$  = <sup>226</sup>Ra activity concentration in the soil particles (Bq/kg)
- $E$  = the radon emanation coefficient

Given the boundary conditions when  $x = 0$ ,  $C = C_0$ , and  $x \rightarrow \infty$ ,  $C$  is finite, the solution for Equation 1.

$$C = \frac{A}{\lambda p_{ef}} \left[ 1 - \exp\left(-\frac{\sqrt{\lambda D_e}}{D_e} x\right) \right] \quad (3)$$

On the other hand, as radon flux is continuous at the ground surface between the soil and atmosphere, the exhalation rate  $F$  of radon from the ground surface can be expressed as follows:

$$F = p_{ef} D_e \frac{dC}{dx} \quad (4)$$

Combined with Equation 3, Equation 4 can be rewritten as:

$$F = \sqrt{\lambda D_e} \rho E R \quad (5)$$

The effective radon diffusion coefficient  $D_e$  in the soil has been experimentally studied, which can be expressed as the following equation:

$$D_e = p D_0 \exp(-6mp - 6m^{14}p) \quad (6)$$

where

- $p$  = the total soil porosity
- $D_0$  = the radon diffusion coefficient in open air with a constant of  $1.1 \times 10^{-5}$  m<sup>2</sup>/s
- $m$  = the volume fraction of water saturation which can be calculated from water mass content  $\omega$  by using Equation 7:

$$m = \omega \rho / 1000 \rho \quad (7)$$

The above equations indicate that the radon flux rate can be calculated when the parameters of  $R$ ,  $\omega$ ,  $\rho$ ,  $p$ , and  $E$  in the soil are known.

### Parameter Values Used in the Calculations

The relevant characteristics of the K-65 raffinate can be found in Appendix J of RAC (1995) and are repeated in the following along with the values selected to be used for this calculation.

*No specific values were reported for the porosity of the K-65 material. However, the report of the 1989 sampling (DOE 1990) reports specific gravity for eight samples (of which two are composites) to be between 2.58 and 3.37, with mean 2.98 and standard deviation 0.29 (about 10%). Porosity can be calculated from bulk density and specific gravity.....results in relatively high (compared to typical uranium mill tailings) nominal estimates of porosity from about 0.6 to 0.8 (RAC 1995, p. J-9).*

Porosity  $p$  of 0.8 is used for the K-65 material.

*The range of moisture contents reported for the 1989 sampling is a summary of eight measurements (DOE 1990). Of the eight, five were 20-35%, one was roughly 50%, and two were between 70% and 75%. It seems likely that the two highest values were for saturated material (RAC 1995, p. J-9).*

Moisture content  $\omega$  of 15% is used for the K-65 material.

*Table J-4 have been converted to dry densities, using the moisture content (65% dry weight) calculated from information on the related laboratory analytical data sheet (NLCO 1972). The calculated dry bulk densities of about  $0.53$  to  $0.72$   $\text{g cm}^{-3}$  and the value of  $1.179$   $\text{g cm}^{-3}$  (basis unknown) seem quite low, relative to a more typical value of  $1.5$   $\text{g cm}^{-3}$  for uranium mill tailings or soils (RAC 1995, p. J-9).*

Bulk density  $\rho$  of  $1.5$   $\text{g/cm}^3$  is used for the K-65 material.

The decay constant  $\lambda$  of  $^{222}\text{Rn}$  is  $2.1 \times 10^{-6}/\text{s}$

The radon emanation coefficient  $E$  is 0.28 (for oven-dried uranium mill tailings (Yu et al. 1993).

The  $^{226}\text{Ra}$  activity concentration  $R$  is 525 nCi/g in Silo 1 and 299 nCi/g in Silo 2 (RAC 1995, Table 8.1).

The results of this method for the silos, substituting the values for  $R$ ,  $E$ ,  $\lambda$ ,  $\omega$ , and  $\rho$  in the above section into Equations 5, 6, and 7, yields a radon exhalation flux from the K-65 material of  $7.10 \times 10^{-7}$  Ci/m<sup>2</sup>/s and  $2.75 \times 10^{-7}$  Ci/m<sup>2</sup>/s in Silos 1 and 2, respectively. The total rate of radon emanating from the K-65 raffinate in the silo airspace is then the product of the exhalation flux and the surface area of the raffinate. It was assumed that the dome approximates a circle with a 40-ft radius, and thus the surface area of the raffinate is 467 m<sup>2</sup>. This resulted in 7,100 Ci/yr and 4,050 Ci/yr of radon entering into the headspace from the raffinate for Silos 1 and 2, respectively.

### **2.3.2 Calculation for the Radon Rate of Release to the Atmosphere**

The calculation continues by providing analytical treatments for determining (1) the amount of radon released to the environment that was driven by the diurnal temperature differential and the Venturi effect of the prevailing wind speed, (2) the retention time of radon gas in the headspace affected by the containment structure of the silo, and (3) the depletion of  $^{222}\text{Rn}$  in the headspace due to the radioactive decay process before release to the environment.

Once created in the K-65 materials, the radon would migrate to the headspace of the silo as described and quantified in Section 2.3.1. Being heavier than air, the radon would tend to stay close to the bottom, and would be moved toward the direction of its decreasing concentration in the dome airspace by atomic diffusion, unless it were mechanically stirred or pulled up by the upward air movements that were being driven to the openings at the top of the silo by the diurnal changes in temperature and the Venturi effects of the wind flow. These openings consist of a 6-in.-diameter gooseneck vent pipe and the gaps between the manholes and the manhole covers. Although RAC (1995) describes significant cracking in the walls that could provide flow paths for the migration of air to the outside, the extremely large resistance in the cracks would direct nearly all the airflow toward the unimpeded 6-in.-diameter vent pipe and the manhole cover gaps at the top of the dome. This conclusion is supported by the abrupt decrease in the environmental  $^{222}\text{Rn}$  concentrations after the sealing of the manhole covers and the removal of the gooseneck vent pipe in 1979.

The leak path factor (LPF) is conventionally used to predict the release of radioactive materials from their containment structures such as, in this case, the silos. The LPF is defined as the fraction of airborne radioactive materials that escape from their confinement structure via available pathways (i.e., the ratio of the radon released to the environment to the net radon source term in the headspace of the silos). LPF calculations were made by developing and applying physically realistic modeling using the Nuclear Regulatory Commission's generalized mass transport and thermal hydraulics computer program CONTAIN 2.0 (SNL 1997). Although this program was developed to predict the thermal hydraulic response inside containments and the release of radionuclides to the environment in the event of containment failure, the modeling capabilities of CONTAIN are sufficiently flexible that it can be applied to the analysis of containment structures in general.

The radon exhalation rate in Section 2.3.1.1 was used as the initial radon source term in the headspace. This amount is reduced by radioactive decay for the duration of the radon residence time in the headspace. The residence time is the period in which radon resides in the silo before exiting the silo. The residence time of the radon can be determined by dividing the flow rate of the radon-air mixture leaving the silo by the volume of the headspace. Subsequently, the amount of radon that had decayed during that period can be calculated. Only the surviving radon would reach the environment. As such, the net radon source term in the headspace of the K-65 silos ( $Q_{Rn}$ ) is represented by the following equations:

$$Q_{Rn} = q_{Rn}(1 - f_d) \quad (8)$$

where

$$\begin{aligned} Q_{Rn} &= \text{the net amount of } ^{222}\text{Rn in the K-65 silo headspace.} \\ q_{Rn} &= \text{the exhalation rate of } ^{222}\text{Rn from the K-65 material} \\ f_d &= \text{the fraction of } ^{222}\text{Rn depleted due to radioactive decay in the headspace of the silo.} \end{aligned}$$

The fraction of radon decayed  $f_d$  is calculated by:

$$f_d = 1 - e^{-\lambda_{Rn} t} \quad (9)$$

where

$$\begin{aligned} \lambda_{Rn} &= \text{the decay constant of } ^{222}\text{Rn} = 0.00755/\text{hr} \\ t &= \text{the residence time of } ^{222}\text{Rn in the headspace} \end{aligned}$$

The residence time  $t$  is obtained by:

$$t = \frac{V_{hs}}{q_{ex}} \quad (10)$$

where

$$\begin{aligned} V_{hs} &= \text{the volume of the silo headspace} \\ q_{ex} &= \text{the exiting radon-air mixture flow rate from the silo due to diurnal and wind effects} \end{aligned}$$

Substituting Equations 9 and 10 into Equation 8 yields Equation 11 showing the net  $^{222}\text{Rn}$  source term in the K-65 silo  $Q_{Rn}$  as a function of the radon-air mixture exiting flow rate  $q_{ex}$  due to the diurnal temperature difference and the Venturi effect from the wind:

$$Q_{Rn} = q_{Rn} \left\{ 1 - \left\{ 1 - \exp \left[ -\lambda_{Rn} \left( \frac{V_{HS}}{q_{ex}} \right) \right] \right\} \right\}$$

(11)

### 2.3.2.1 CONTAIN 2.0 Leak Path Factor Calculations

The CONTAIN 2.0 computer program (SNL 1997) is an integrated analysis tool used for predicting the physical conditions, chemical compositions, and distributions of radiological materials inside the containment and connected buildings of a nuclear reactor after the release of material from the primary system in a light-water reactor accident. It is designed to predict the thermal-hydraulic response inside containments and the release of radionuclides to the environment. CONTAIN analyzes the thermal-hydraulic phenomena, aerosol behavior, and fission product processes, and the interactions among these phenomena.

The code treats a containment system as a network of interconnected control volumes or "cells." A compartment can also be partitioned to model phenomena such as natural convection and stratification within the compartment. The cells communicate with each other by means of mass flow of material between cells and/or heat conduction between cells through heat transfer structures. The flow directions through the flow paths are determined by the code calculation. Because there is considerable flexibility in specifying the properties of each cell and the connections between cells, the code is able to handle a wide variety of containment types.

Figure 1 defines the physical configurations of the silos for the CONTAIN code modeling.

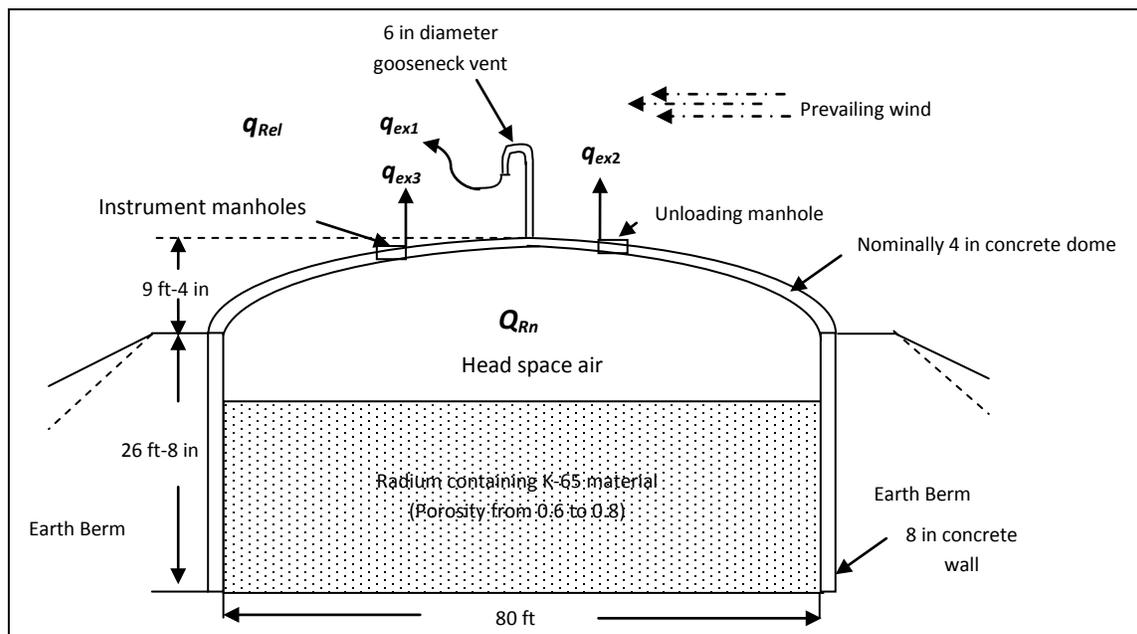


Figure 1. K-65 silos at the FMPC.

The LPF is defined as:

$$LPF = \frac{q_{rel}}{Q_{Rn}} \quad (12)$$

where

$q_{rel}$  = the radon released to the environment

$Q_{Rn}$  = the net amount of  $^{222}\text{Rn}$  in the K-65 silo headspace [Equation 11]

and:

$$q_{ex} = q_{ex1} + q_{ex2} + q_{ex3} \quad (13)$$

where

$q_{ex1}$  = the radon-air mixture exiting flow rate from the gooseneck vent pipe  
 $q_{ex2}$  = the radon-air mixture exiting flow rate from the unloading manhole cover gap  
 $q_{ex3}$  = the radon-air mixture exiting flow rate from the four instrument manhole cover gaps

The following assumptions were applied to the development of the CONTAIN input parameters:

- Flow through wall cracks are small in comparison with flow through the 6-in. vent and manhole cover gaps
- Total volume of a silo is about 160,000 ft<sup>3</sup> (4,500 m<sup>3</sup>) (RAC 1995)
- The silo is two-thirds full of K-65 materials with a porosity of 0.7 (RAC 1995)
- The diurnal temperature difference is 19.46°F (RAC 1995)
- The prevailing wind speed is 6.5 mph (ORAUT 2006b)
- Each instrument manhole has a diameter of 6 in. with 4-in.-thick concrete. The gap around the circumference of the manhole and manhole cover is 0.125 in.
- The unloading manhole has a diameter of 24 in. with 4-in.-thick concrete. The gap around the circumference of the manhole and manhole cover is 0.125 in.
- The total length of the 6-in. gooseneck vent is 5 ft.
- CO<sub>2</sub> gas is used as a surrogate for radon.

The K-65 silo is modeled as a series of nine cells and the environment is represented by the 10th cell. Cell 1 contains the radium-bearing K-65 materials having 0.7 porosity. Its volume counts only the portion available to the gas. Cell 9 is the domed headspace and is simulated by a cylinder. The connections of Cell 9 to the outside environment (Cell 10) are by three engineered vents: one for the unloading manhole, one for the gooseneck vent pipe, and one for the four instrument manholes combined. Cells 2 through 8 represent the remainder of the silo and are for observing gradients in the gas. All internal cells contain air with various molar fractions of CO<sub>2</sub>. The CONTAIN program has the ability to track materials with thermodynamic properties if they are in the code library, such as CO<sub>2</sub>. At 1 atmosphere and 300 K, CO<sub>2</sub> is about 53% denser than air. Carbon dioxide gas works well as a surrogate for radon and is claimant-favorable because radon is significantly denser than CO<sub>2</sub>, which leads to an overestimation of releases to the environment. Cell 10, the environmental cell, initially contains only air and the arriving mass of CO<sub>2</sub> over time is tracked by CONTAIN.

Attachment A presents the details of the CONTAIN calculations.

### 2.3.2.2 CONTAIN Code Results

Figure 2 shows the flow rates for the three vents. The total flow to the environment promptly reached a value of  $q_{ex} = 17.88$  cfm. Figure 3 shows the CO<sub>2</sub> mass in Cell 10 (environment) versus time for a 5% CO<sub>2</sub> molar fraction in the silo. The resulting CO<sub>2</sub> mass in Cell 10 was divided by the total initial CO<sub>2</sub> mass in the silo to obtain the values for the LPF. Figure 4 shows the LPF versus time for the 5% CO<sub>2</sub> and air mixture in the silo. The LPF value reached a plateau of 0.873% after 4 hours of release. The LPF calculation was repeated with 10%, 15%, and 20% CO<sub>2</sub> molar fractions and the corresponding LPFs were 0.864%, 0.855%, and 0.847%. The LPFs changed only slightly, decreasing as the molar fraction increased. The results indicate that the fraction vented is not sensitive to the molar fraction of CO<sub>2</sub> used. Therefore, it can be concluded that a value of 0.873% is an appropriate LPF (the fraction of radon vented to the environment) regardless of the amount of radon in the headspace for this problem.

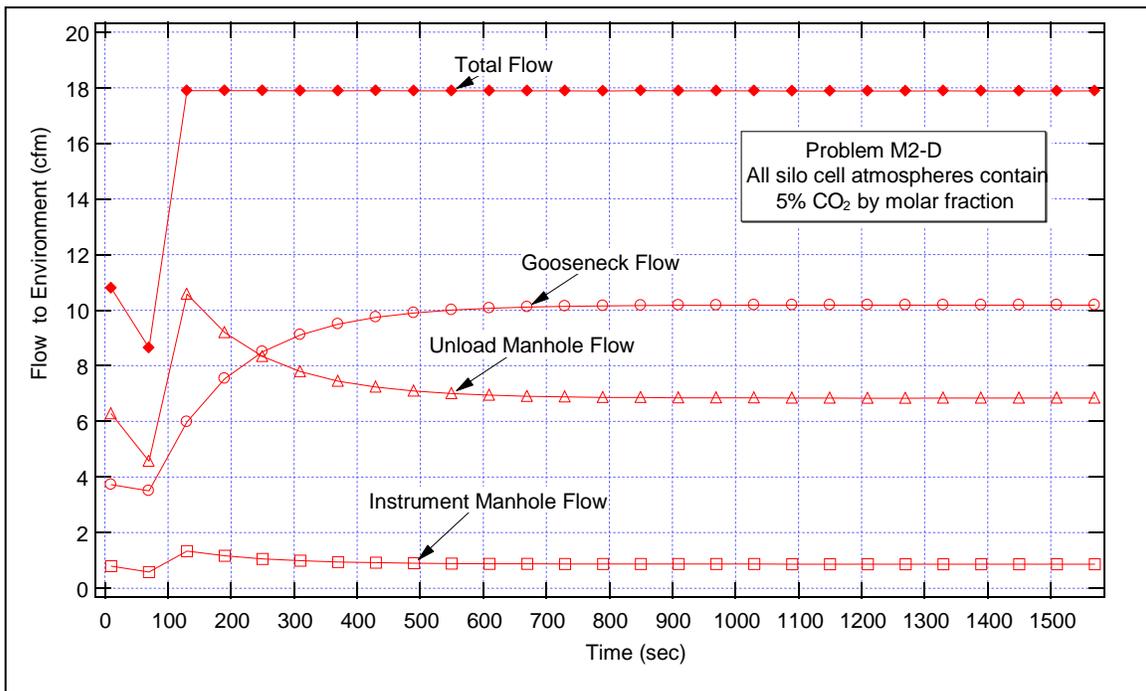


Figure 2. Flows to the environment from the engineered vents.

### 2.3.3 Environmental Radon Release Rate

The radon release from the K-65 silos can be obtained by solving Equation 14:

$$q_{rel} = LPF \times Q_{Rn} \tag{14}$$

where

- LPF = the leak path factor, 0.00873
- $Q_{Rn}$  = the net source term in the headspace

=

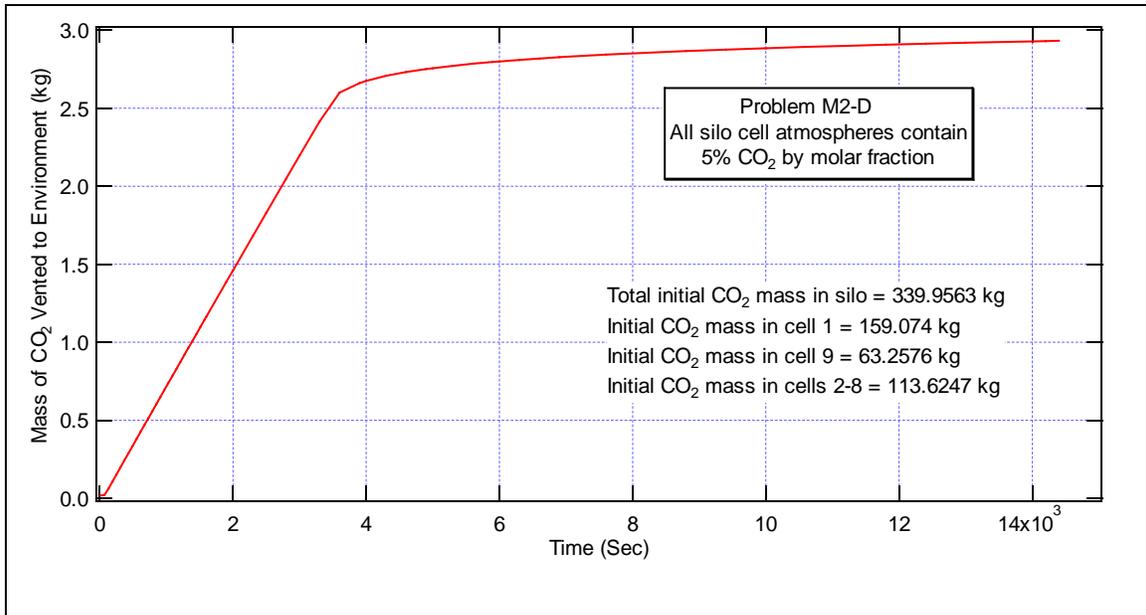
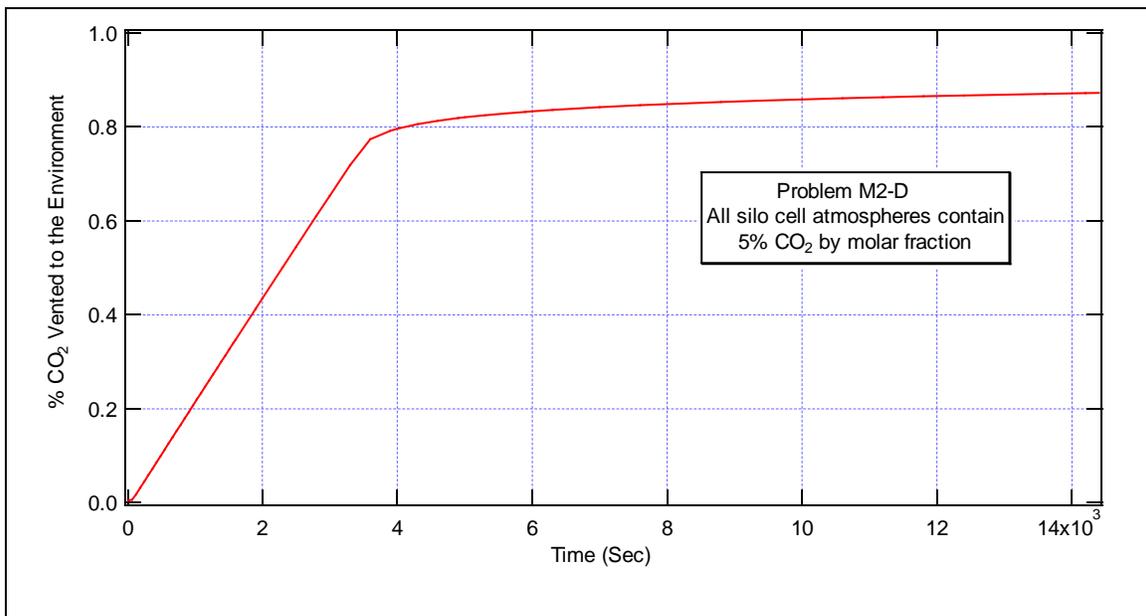


Figure 3. Flows to the environment from the engineered vents.

Figure 4. Percent of the CO<sub>2</sub> vented to the environment versus time.

where:

$$q_{Rn} = 7,100 \text{ Ci/yr Silo 1 and } 4,050 \text{ Ci/yr Silo 2}$$

$$\lambda_{Rn} = 0.00755/\text{hr}$$

$$V_{hs} = 4.00 \times 10^4 \text{ ft}^3 \text{ (RAC 1995, p. J-30)}$$

$$q_{ex} = 17.88 \times 60 = 1,073 \text{ ft}^3/\text{hr (From Figure 2)}$$

Solving Equation 14 yields the combined (Silos 1 and 2) annual environmental releases before 1980 of 70.4 Ci/yr.

### 2.3.4 Summary Conclusion

The alternative calculation of the radon release from the K-65 silos indicates a release rate of 70.4 Ci/yr, which is slightly higher than the 60-Ci/yr diffusion release calculations performed by Borak (1985). The incremental increase in release rate primarily represents the amount of radon that escaped through the openings on the top of the dome driven by the daily temperature difference and the Venturi effect from the wind. All previous calculations assumed a constant radon concentration in the silo air mixture. Being a mass transport and thermal hydraulics computer code, the air/radon air mixture flow rates through the dome openings to the outside environment predicted by CONTAIN based on physical configurations and thermal-hydraulic parameters are particularly accurate. Figure 2 shows a total flow to the environment from the engineered vents of approximately 18 cfm. When using the actual radon concentration in the headspace as shown in RAC (1995, Table J-12) ( $2.5 \times 10^7$  pCi/L for Silo 1), the alternative calculation would have resulted in a radon release rate to the atmosphere of 6,698 Ci/yr:

$$2.5 \times 10^7 \text{ pCi/L} \times 2.83 \times 10^{-11} \text{ Ci/ft}^3 \times 18 \text{ ft}^3/\text{min} \times 5.26 \times 10^5 \text{ min/yr} = 6,698 \text{ Ci/yr}$$

The alternative result of 6,700 Ci/yr is practically identical to the RAC (1995) results of 5,000 to 6,000 Ci/yr for the same period, and both calculations are in line with each other. On the other hand, perhaps the RAC report could be proven to have overestimated the radon release by CONTAIN.

Because of the very large uncertainties associated with the SC&A  $^{210}\text{Pb}$  deficit radon release calculation and because its validity has not been verified or reviewed by the National Academy of Sciences, NIOSH will continue to adopt the claimant-favorable model for radon releases, as given in Section 5.2.4 of ORAUT-TKBS-0017-5 (ORAUT 2004), that 5,000 to 6,000 Ci/year of  $^{222}\text{Rn}$  were released from the silos.

## 2.4 **RESPONSE TO APRIL 2010 SC&A WHITE PAPER**

The purpose of this section is to respond to the salient points in the Second White Paper (SC&A 2010) in relation to the NIOSH dose reconstruction position to:

*....rely upon the individual exposure estimates produced using the Pinney/Hornung model which utilized radon exposure levels from the RAC study, plus an additional radon source term at Fernald. ...*

The primary negative conclusions drawn by SC&A regarding the Pinney Report (Hornung 2008) are:

- (1) *The Pinney Report simply accepted the source term releases of radon from the K-65 Silos, as described by the following two RAC reports issued in 1995 and 1998:...*
- (2) *Using the modeled RAC (1995)/RAC (1998) radon release source terms, the "Pinney Report" investigators applied a modified Gaussian dispersion model originally ...*

These conclusions failed to recognize the extensive efforts to validate the RAC model by the Hornung/Pinney team. The description of the validation of model estimates in Hornung (2008) is repeated here to ensure their efforts are not overlooked.

From Pages 3 and 4 of the Pinney Report (Hornung et al. 2008):

### *Validation of model estimates*

*The original Gaussian plume model was calibrated to outdoor measurements of radon made at various locations on the site during the years 1981 through 1987 (Killough and Schmidt, 2000), with estimated emission rates from the K-65 silos as input. Before 1979, when vent pipes were removed and the silo domes were capped, very few radon measurements were available. The RAC dose-reconstruction team made a thorough search for any historical measurements that could have been used to calibrate their model, but the era of highest exposures (1952–1979) was difficult to validate. We sought a way of validating our exposure estimates, particularly before 1979. Several case-control studies of radon related lung cancer from exposures received in homes (Field, 2001; Steck et al., 2002) employed a method for estimating cumulative exposure. This method involved the use of CR-39 plastic film applied to glass in mirrors or framed pictures that were kept in each home that a case or control inhabited over their adult lifetime. The CR-39 film records tracks from  $^{210}\text{Po}$   $\alpha$ -particles that decay from  $^{210}\text{Pb}$  which is a long-lived (22 years half-life) decay product of  $^{222}\text{Rn}$  imbedded in the glass. Counts of tracks from the CR-39 plastics can be used to estimate integrated radon exposure over long periods of time (Lively and Steck, 1993; Mahaffey et al., 1993).*

*Since CR-39 had been primarily used in laboratories and homes, a test of its feasibility in a uranium processing facility was necessary. Eight CR-39 films were obtained from Track Analysis Systems Ltd. (TASL) in Bristol, England. They were placed on the inside and outside of glass window panes in a building near the center of the production area and in a building near the eastern edge of the plant farther from the K-65 silos. Four replicates were made by placing films side-by-side on the same pane of glass. These films were exposed for approximately 2 weeks in November 1999 and then sealed and sent to TASL for analysis with no indication of where they had been placed. Upon receipt of the exposed CR-39 films, TASL etched the films in a solution of NaOH, calibrated them with a  $^{252}\text{Cf}$  fission source, and scanned them with an automated image analysis system. Surface activity of  $^{210}\text{Po}$  is measured with corrections for lower energy particles from  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$  and similar lower energy  $\alpha$  particles (Fews and Henshaw, 1982).*

*Higher energy contamination ( $^{214}\text{Po}$ ,  $^{218}\text{Po}$ , and  $^{212}\text{Po}$ ) was eliminated from track counts and did not cross contaminate  $^{210}\text{Po}$  readings. The actual measurements were made by etching and scanning TASTRAK  $\alpha$ -particle sensitive CR-39 plastic, followed by spectroscopic analysis of associated  $\alpha$ -tracks (Fews, 1992). When track density was extremely high, tracks were counted by eye using averages of 10 microscope image frames randomly distributed over the sample. Results for all samples were reported as total  $^{210}\text{Po}$  activity per unit area in  $\text{Bq}/\text{m}^2$ .*

*In addition to the use of CR-39 film for validation of the air dispersion model, a set of radon measurements were discovered that were not used to validate the model when it was developed to estimate off-site radon levels for residents near Fernald. These data were collected for 7 months from March to September 1991 by a student for a Master's degree thesis (Cardarelli, 1992). Samples were taken around the K-65 silos and in the vicinity of five buildings on the Fernald site. The protocol for the Master's thesis called for a survey using three monitors simultaneously: Pylon monitors, Radon Gas monitors (RGMII), and Femto-tech monitors, all three located at each measurement site. The Pylon monitor utilizes passive diffusion of radon gas into a scintillation chamber where  $\alpha$ -particles are counted. The RGM II utilizes a pump to draw air through a particulate filter into a scintillation chamber. The Femto-tech monitor is*

*similar to the Pylon monitor in using passive diffusion, but it employs a pulsed ion-sensing chamber instead of a scintillation chamber. The data available for our validation consisted of the arithmetic mean (AM) of the three monitor results for each hour. The hourly means were then used to calculate geometric means (GMs) for each 8-h shift across the 7 months when sampling was done since the distribution of radon levels over time was right-skewed."*

The Pinney Report (Hornung et al. 2008) was not the first time the validation of the RAC model was described. The validation was also described in Pinney et al. (2004) by the Hornung/Pinney team on their work to assess the radon and cigarette smoking exposure of Fernald workers. Again the radon dispersion model developed by RAC for emissions from the K-65 silos was used and validated in the assessment.

From page 5 of Pinney et al. (2004):

*Since the RAC model is a conceptual mathematical model developed from physical principles and is not based directly upon measured data, we attempted two ways to investigate the accuracy of its estimates. First, we discovered radon measurement data that had not been used by the RAC group to calibrate their model. These were data collected by John Cardarelli for his Master's thesis during the period of March through September 1991. Data were collected both in the immediate vicinity of the K-65 silos and at various points around the production area. We then compared Cardarelli's measurements by shift and location to the closest year of our estimates, 1988. Since no remediation was done between 1988 and 1991, we assumed that results should be comparable. Results are shown in the attached Tables 2 and 3.*

*Table 2. Comparison of model predictions and 1991 radon measurements.*

Location	Shift	Model	Measurement	Accuracy	P-value
14A	1	0.273	0.233	0.285	0.057
	2	0.338	0.293	0.356	0.049
	3	0.293	0.494	0.437	<0.001
2A	1	0.458	0.079	0.458	<0.001
	2	0.453	0.107	0.425	<0.001
	3	0.354	0.335	0.449	NS
53A	1	0.240	0.072	0.328	<0.001
	2	0.299	0.134	0.745	<0.001
	3	0.259	0.231	0.361	<0.001
54A	1	0.388	0.228	0.269	<0.001
	2	0.467	0.365	0.338	<0.001
	3	0.396	0.666	0.662	<0.001

*Table 3. Comparison of model predictions to 1991 silo measurements.*

Location	Shift	Model	Measurement	Accuracy	P-value
K-65 NE	1	12.37	2.75	10.67	<0.001
	2	13.55	7.68	11.94	<0.001
	3	7.72	11.68	18.45	0.012
K-65 NW	1	3.39	1.30	3.29	<0.001
	2	4.27	3.10	4.96	<0.001
	3	1.35	3.54	4.97	<0.001
K-65 SE	1	8.92	1.75	8.10	<0.001
	2	9.99	5.19	9.57	<0.001
	3	8.20	8.41	13.06	NS
K-65 SW	1	8.70	1.29	9.53	<0.001
	2	6.46	4.24	8.77	<0.001
	3	2.99	5.50	9.61	<0.001

*Our second attempt to validate the model was the use of CR-39 film attached to windows in selected buildings around the site. The CR-39 film measures alpha activity from the decay of Pb-210, which was embedded in the glass over several decades. This assay, therefore, serves as a cumulative measure of radon and its decay products since each building was constructed. .... With NIOSH support, we expanded our sample size to 110 samples placed throughout the site on buildings selected for their proximity to both the K-65 silos and the plant 2/3 area where we found the initial high measurements. Results of the larger survey indicated that glass sampled near the Q-11 silos in the plant 2/3 area were substantially higher than glass closer to the K-65 silos. The Q-11 silos had been used in the period 1953-58 to store highly radioactive ore before it was processed in the plant 2/3 complex. .... Once we became aware of these CR-39 findings, we conducted additional document searches and met with workers several times to develop a better understanding of this local source of radon.*

## 2.5 CONCLUSION

The Hornung/Pinney team validated the RAC model estimates with an independent set of radon measurement data that had not been used by the RAC group to calibrate their model. These data were collected by John Cardarelli for his Master's thesis from March through September 1991. Data were collected in the immediate vicinity of the K-65 silos and at various points around the production area. Tables 2 and 3 of Pinney et al. (2004) show that the comparisons were in good agreement with the Killough model predictions for the first and second shifts. The model predicted higher exposures that were claimant-favorable. It also predicted lower exposures for the third shift and the differences, in general, are within a factor of 2. In the scientific community, a factor-of-2 difference is a reasonable validation of the model predictions for this type of analysis.

NRC (1994, p. 14) states:

*Measurements were used for comparison with the estimates derived from the model, and the overall results are verified to be within a factor of 2. ... Overall, the results of the comparison are a reasonable validation of the model predictions.*

The Pinney/Hornung model on which NIOSH will rely for individual exposure estimates is entirely credible and appropriate because the analytical model had initially been calibrated with measurement data and later validated with independent sets of measurement data.

### 3.0 Q-11, K-65, DOMESTIC URANIUM, AND RAFFINATES

#### 3.1 PROCESSING HISTORY OF PITCHBLEND AND DOMESTIC URANIUM FEEDSTOCK

During the Manhattan Project, extremely high-grade (50% to 80%) uranium ore was imported from African Metal Company (AMC) in the Belgian Congo and was designated with the code "Q-11." At this level of uranium content, the ore could be processed directly in the modified plutonium-uranium extraction (PUREX) process at FMPC. From 1946 until 1953, Mallinckrodt Chemical Works (MCW) in St. Louis, Missouri, produced uranium from the Q-11 ore. The Q-11 ore contained all of the uranium daughter products, including  $^{226}\text{Ra}$ . When the ore was refined,  $^{226}\text{Ra}$  and other daughter products were concentrated in the waste stream, which was identified with the code "K-65." The term *raffinate* was used to describe the waste stream. The K-65 residues were returned to Belgium until March 1949 (NLO 1979; Heatherton 1949). In 1949, an agreement was reached wherein the U.S. Atomic Energy Commission (AEC) would store the K-65 residues for later return to AMC. Beginning on March 22, 1949, the K-65 residues from MCW were sent to Lake Ontario Ordnance Works (LOOW) to be stored (Heatherton 1949).

Construction of FMPC began in 1951 and MCW began shipping K-65 material to FMPC rather than LOOW (Walden 1952; Vogel 1989). Between September 25, 1951 and April 30, 1952, 11,253 drums were sent from MCW to FMPC; a total of 25,000 drums were shipped in all (Vogel 1989). The shipments of K-65 residue from MCW continued until January 1956 (Vogel 1989).

When construction was completed, storage of all K-65 material was assigned to FMPC and two large ground-level silos were built for this purpose. In addition to the residues shipped from MCW and LOOW, FMPC began processing Q-11 ore and generating its own raffinate streams that needed to be stored. Pitchblende ore processing resulted in "hot" raffinates, indicating that relatively high concentrations of  $^{226}\text{Ra}$  plus daughter products were in the waste stream with the associated large increase in external radiation fields. The transfer of K-65 material to the silos began in late 1952 (Wunder 1953a). The silos were designed to hold about 24,000 drums each (Vogel 1989). Silo 1 contained K-65 material from LOOW and MCW. Silo 2 held K-65 material generated at FMPC and from off site, including MCW (Vogel 1989).

The transfer of the drummed K-65 (the hot raffinate) material to the silos took place at a drum handling station, where the material was dumped from the drum to a slurry tank. The slurried material was pumped to the storage silo and the excess water returned to the slurry tank. This operation did not run continuously; for example, freezing weather caused the dumping to be stopped on January 9, 1953, for about 2 months. At that time, Silo 1 contained 20,259 drums, with 2,257 drums from LOOW remaining (Wunder 1953a). In June 1953, the K-65 dumping station was being operated on the day shift only, the dumping was current on MCW material, and there were 671 drums remaining from LOOW. The south tank (Silo 1) was reported to be full and the dumping was moving to the north tank (Silo 2) (Wunder 1953b). The last of the K-65 material from MCW was dumped in April 1956 (Vogel 1989).

K-65 material from the processing of Q-11 ores at FMPC was initially transferred in 1954 and periodically through 1957 (Vogel 1989, Attachment 8). This processing occurred in Plants 2 and 3. See Section 3.4 for specific dates of these campaigns and a listing of other K-65 materials that were transferred to Silo 2. The total K-65 material in the two silos was 19,385,126 lb, including about 380,000 lb from the processing of "rum jungle" ore from Australia in 1957 and 1958 (Vogel 1989, Attachment 18; Shaw 1968; Robinson 1973). About 25% of the material in the two silos was generated at FMPC (Robinson 1973). In summary, the following events or campaigns contributed to the history of the K-65 silos:

1946–1953 All uranium ore processing occurred at MCW.

- 1946–1949 K-65 residues were sent from MCW to Belgium.
- 3/1949–1951 K-65 residues were no longer sent to Belgium, but were sent from MCW to LOOW.
- 1951–1/1956 MCW shipped K-65 residues directly to FMPC.
- 9-11/1952 Drums of MCW residue that were not dumped into the LOOW water tower were shipped to FMPC under the supervision of FMPC, but using workers from the Niagara Falls area.
- Late 1952 FMPC began dumping drums of K-65 from MCW and LOOW to Silo 1.
- June 1953 The drum dumping operation was current on MCW drums, with 671 drums left from LOOW; the south tank was full (Wunder 1953b).
- 4/54–4/57 Processing of Q-11 ores at FMPC occurred periodically during this period with the associated transfers of K-65 materials from the local process.
- 4/1956 The last K-65 material from MCW was transferred to Silo 2.
- 1957–1958 Australian pitchblende was processed with the raffinates being transferred to Silo 2.

Most U.S. and Canadian ores were low grade (1% or less uranium content) and required processing near the mine sites. The product from these mills was a “yellowcake” (U<sub>3</sub>O<sub>8</sub>) of approximately 60% uranium content. After 1958, when processing of the Q-11 and other pitchblende ores at FMPC had been completed, the source of the uranium for processing at FMPC was this yellowcake. The yellowcake had most of the radium and other impurities removed before being shipped to FMPC. This raffinate was called “cold raffinate” because the waste stream had much lower concentrations of <sup>226</sup>Ra plus daughters, compared with the K-65 raffinate. The cold raffinate was a calcined dry, dispersible powder that was transferred to Silo 3 using an enclosed air lift. (The fourth silo was never used and remained empty.)

The isotopic contents of the three silos are shown in Table 1.

Table 1. Isotopic composition in FMPC Silos 1, 2, and 3.

Isotope	Silo 1 activity concentration (nCi/g raffinate)	Silo 1 percent activity	Silo 2 activity concentration (nCi/g raffinate)	Silo 2 percent activity	Silo 3 activity concentration (nCi/g raffinate)	Silo 3 percent activity
Uranium	1.68	0.2	2.37	0.3	3.63	4.8
Ac-227	7.67	0.8	6.64	0.8	0.93	1.2
Pa-231	N/A	0.0	4.04	0.5	0.63	0.8
Pb-210	202	20.0	190	24.3	3.48	4.6
Po-210	281	27.8	231	29.6	N/A	0.0
Ra-224	N/A	0.0	N/A	0.0	0.37	0.5
Ra-226	447	44.2	263	33.6	3.87	5.2
Ra-228	2.28	0.2	N/A	0.0	0.41	0.5
Th-228	N/A	0.0	7.36	0.9	0.75	1.0
Th-230	68.9	6.8	76.2	9.7	60.2	80.1
Th-232	1.11	0.1	0.99	0.1	0.84	1.1
Total	1011.64	N/A <sup>a</sup>	781.6	N/A	75.11	N/A

a. N/A = not applicable.

Several observations are made from these data. Silos 1 and 2 contain the K-65 (hot) raffinates and include relatively high concentrations of  $^{226}\text{Ra}$ . Silo 3 contains cold raffinate with very little  $^{226}\text{Ra}$ . Because uranium is the extraction product, the raffinates in the silos contain low levels of uranium. Thorium-230 is present in each silo in similar concentrations, indicating that the uranium mills effectively removed radium but were not effective in removing thorium from the yellowcake. The total activity concentration in Silo 3 is approximately 10% of the activity concentration in Silos 1 and 2.

These observations indicate potential problems for dose reconstruction techniques that are based on uranium bioassay data because the uranium intake when handling these materials might not have been the dominant isotopes. As a consequence, alternative methods of dose reconstruction are defined.

### 3.2 AVAILABLE MONITORING DATA

An extensive air sampling program was conducted at FMPC. The program was designed for worker exposure control by monitoring radioactive air dust to identify process areas that needed better engineering controls or designs, and to identify operations that required respiratory protection. These studies involved air samples in all the production plants, and included general area samples and breathing zone samples for workers. The studies were documented in annual exposure studies from 1955 to 1968 (AEC 1955; Stefenac 1955a,b; Stefenac and Schumann 1956a,b; Halcomb and Huesing 1957; NLO 1957; Wing and Halcomb 1958a,b, 1959a,b; Wing and Ruhe 1960a,b; Wing et al. 1961; Wing, Ross, and Cline 1961; Wing, Ross, and Leininger 1962; Wing and Leininger 1965; Wing, Leininger, and Ruhe 1966; Ross, Leininger, and Lawrence 1968; Ross, Leininger, and Zimber 1969).

Tables 2, 3, and 4 are summaries of the results from these exposure studies in Plants 2 and 3, where feed materials were charged to the processes and raffinate processing occurred, for each job category and each year. The units are in multiples of the maximum allowable concentration (MAC). See Section 2 for more information about DWE reports.

Several observations can be made on plant operations and personnel exposures from these data.

- Data were collected separately for Plants 2 and 3 from 1955 to 1962. The two plants were shut down for the next 2 years. After restart, data were collected as combined Plant 2/3 data.
- The DWE for each job category did not take into consideration that respirators were used by the workers for jobs that created high dust levels. As a consequence, actual exposures to the workers would have been lower than depicted in these tables.
- The data for 1955 through 1957 covered the end of the processing of the pitchblende ores from the Belgian Congo and Australian sources in 1958 with high radium content. The raffinate stream from these ores was stored in Silo 2. The processing then switched to yellowcake from mills in the United States and Canada, which continued until Plants 2 and 3 were shut down in 1962. It was during this period that the raffinate stream was sent to Silo 3 for storage.
- The level of exposures to air activity as measured by the DWE sampling program gradually decreased over the 14 years of operations. The exposure studies attribute the decreases in part to improvements in process controls and procedures.
- The studies for Plants 2 and 3 indicate that the highest airborne dust concentrations were measured in the head end (ore and concentrate) dumping operations. The ore dumping operations were dusty and less well contained. The high values for the ore dumping operators were actually the results of very high values experienced while dumping drums containing the

Table 2. DWE data for Plant 2, 1955 to 1962, when Silos 2 and 3 were being filled. The # field represents the number of workers potentially exposed at that daily weighted value at the time the data were collected and analyzed by the FMPC staff.

Job category	1955 <sup>a</sup>		1956		1957		1958		1959		1960		1961		1962	
	#	DWE	#	DWE	#	DWE	#	DWE	#	DWE	#	DWE	#	DWE	#	DWE
Ore dumping and metal dissolver operators							3	2.81	3	0.76	6	4.1	3	8.3	3	24
Ore dumping	3	0.9	4	6.2	4	2.16	6	4.26	6	5.9	3	1.4				
Extraction operators and helpers	24	0.21								15	0.53					
Digestion operators and helpers	24	0.64														
Laborer	3	1	12	0.87	12	0.76										
Boildown and denitration operators	24	0.74														
Gulping helper			6	0.85	4	0.76										
Gulping operator	6	0.05	12	0.82	8	0.15										
Leadman			1	0.49	1	0.16										
Control room	6	0.06	4	0.73	4	0.16										
Pot man			8	0.31	8	0.15	6	0.96			3	0.71			5	0.1
Floor man			3	0.37	8	0.19	3	0.81	6	0.21	3	0.56	3	0.1		
Utility operator and helpers	3	0.06	12	0.32	12	0.19	3	0.81	3	0.25	3	0.54	3	0.1	3	0.2
Forklift operator							3	0.46	3	0.15	3	0.56	3	0.1	3	0.2
Shift clerk			4	0.35	4	0.11	2	0.46	2	0.13	2	0.49	2	0.1	1	0.2
Leadman, operators, and helpers	27	0.65	30	0.21	20	0.11										
Evaporator operator			4	0.31	4	0.13	3	0.63	3	1.24	3	0.9	3	0.2	2	0.2
Area foreman	1	0.06	1	0.3	1	0.11	1	0.57	1	0.26	1	0.42	1	0.1	1	0.2
Shift foreman	3	0.06	4	0.26	4	0.12	3	0.79	6	0.34	3	0.39	4	0.1	4	0.2
Pump man							3	0.57	3	0.31	2	0.23	2	0.1		
Production records clerk	5	0.12					3	0.16	3	0.12	2	0.23	2	0.1	2	0.2
General foreman	1	0.12	1	0.2	1	0.12	1	0.53	1	0.35	1	0.3	1	0.1	1	0.1
Analyst	3	0.25	4	0.73	4	0.08	3	0.1	3	0.24	3	0.22	3	0.1	3	0.2
Extraction operator	24	0.21					18	0.37			15	0.2	9	0.1		
Superintendent	1	0.11	1	0.2	1	0.06	1	0.37	1	0.22	1	0.17	1	0.1	1	0.2
Office assistant; admin. assistant	1	0.11	1	0.21	1	0.12	2	0.14	2	0.11	2	0.14	1	0.1	1	0.2
Sump operator	6	0.09	8	0.18	8	0.05	6	0.09	6	0.2	3	0.05	3	0.1	3	0.1
Solvent reclaimer													6	0.1		

Compiled from two exposure studies (Stefanec 1955a, Stefanec 1955b) performed in 1955.

Table 3. DWE data for Plant 3, 1955 to 1962, when Silos 2 and 3 were being filled. The # field represents the number of workers potentially exposed at that daily weighted value at the time the data were collected and analyzed by the FMPC staff.

Job category	1955		1956		1957		1958		1959		1960		1961		1962	
	#	DWE	#	DWE	#	DWE	#	DWE								
Pot man							6	0.63	6	0.55	6	0.8	4	0.2		
Leadman	6	0.44	6	0.23	6	0.09										
Laborers	3	1	7	0.21	7	0.08										
Gulping operator	6	0.09					6	0.73	6	0.46	6	0.6	5	0.2		
Utility operator											3	0.5	1	0.2		
Area foreman			1	0.25	1	0.07	1	0.18		0.18	1	0.2	1	0.1	1	0.2
Shift foreman	4	0.53	4	0.23	4	0.09	3	0.37	3	0.33	3	0.5	8	0.1	1	0.1
Systems operator									3	3.77	3	0.4				
Recovery operators and helpers	24	0.51	29	0.22	27	0.36	12	0.14	12	0.08	12	0.1	5	0.1	5	0.1

Table 4. DWE data for Plant 2 and 3, 1965 to 1968, during low acid processing period. The # field represents the number of workers potentially exposed at that daily weighted value at the time the data were collected and analyzed by the FMPC staff.<sup>a</sup>

Job category	1965		1966		1967		1968	
	#	DWE	#	DWE	#	DWE	#	DWE
Digestion operator	4	1.7	6	1.6	12	4.2	3	2.9
Denitration operators	6	0.3	9	2.2	6	0.5	4	1.7
Foreman	3	0.2	3	0.2	3	0.3	2	0.7
Clerk	1	0.2	1	0.1	1	0.1	1	0.1
Superintendent	1	0.2	1	0.2	1	0.2	1	0.2
Hot raffinate	3	0.1	3	0.1	3	0.2	3	0.2
Combined raffinate	4	0.1	5	0.1	3	0.1	3	0.2
Forklift operator	1	0.1	1	0.2	3	0.5	3	0.5
Shift foreman							6	0.4
Prod. records clerk							2	0.1
Extraction operator	6	0.1	6	0.1	3	0.1	3	0.1
Sump operator					3	0.1	3	0.1

a. The low acid period refers to a change in process from concentrated nitric acid (which for economic considerations required recovery by condensation from a calciner) to a more dilute acid process, which could be neutralized and pumped directly to waste disposal Pit 5.

uranium feed material into the head end of the process that were weighted or averaged with the time that was spent in other activities performed in lower air dust concentrations.

- The lowest concentrations were experienced by the workers in raffinate operations. The solvent extraction process was a liquid process completely contained in a closed tank/piping system. The workers in the raffinate areas experienced airborne concentrations that were consistently at the same levels as those assigned to records clerks and administrative assistants. These levels should be considered ambient levels very near the detection limit of the monitoring equipment and techniques. The value of 0.1 MAC is the same value consistently reported for other nonproduction plant areas.

The most significant exposure potential from the "hot" raffinates was the relatively high external exposure rates due to high concentrations of <sup>226</sup>Ra and daughter products. Shielding was provided for the equipment that was a part of the Q-11 processing to minimize external exposures to the workers. The handling of K-65 drums from MCW and LOOW was recognized as having high external exposures; workers were rotated through this work activity to meet the external exposure guidelines (Heatherton 1952a).

- The DWE data for Plant 2/3 during the period from 1965 through 1968 clearly show the significant differences in the front end processes in Plant 2 compared to the raffinate processes in Plant 3. The raffinate operators experienced DWE levels that were 10 to 20 times lower than the operators on the digestion/denitration side of the refinery.

These data demonstrate that exposures from the raffinate processes in Plant 3 were extremely low, essentially at background levels. The primary source of exposure for workers in Plants 2 and 3 was the beginning of the processing operation, the ore and concentrate charging operations. The source of the exposure is significant in establishing the ratio of the trace contaminants to which workers would have been exposed. The high thorium ratio observed in Silo 3 materials is associated with the liquid stream contained in a closed piping system and then to a closed calcination system, which provides little exposure potential to the workers. The process charging operation, which is the primary

source of exposure, has ratios of thorium to uranium that are typical of ore materials. At most the  $^{230}\text{Th}$  would be in effective equilibrium with the  $^{238}\text{U}/^{234}\text{U}$  activities in the feed material.

The  $^{230}\text{Th}$  concentration in Silo 3 is roughly the same (possibly 10% less) as the  $^{230}\text{Th}$  concentration in Silos 1 and 2. The  $^{230}\text{Th}$  in Silo 3 constitutes a high fraction of the total activity, compared with the fractions in Silos 1 and 2, due not to high  $^{230}\text{Th}$  levels, but solely to the absence of radium and its daughter products. If the  $^{226}\text{Ra}$  and its daughters were removed from Silos 1 and 2, the  $^{230}\text{Th}$  would constitute about 80% of the remaining relative hazard, as it does in the Silo 3 material. The concentration of  $^{230}\text{Th}$  is essentially the same in all three silos, indicating that the uranium milling processes removed little of the thorium from the ores.

A review of the job categories in Tables 2, 3, and 4 indicates that the workers with the highest exposures in Plants 2 and 3 were the operators in Plant 2. Job categories that might have been considered transient, such as helpers, laborers, forklift operator, etc., are shown to have low exposures. In fact, helpers are included in the operator category in Plant 3, which experienced very low exposures. Any special maintenance activities that might have required work on the raffinate process systems would have been performed with appropriate protective equipment (respirators and ventilation), but primarily would have constituted a relatively short exposure time compared with the full-time exposure considered in the DWE exposure studies. As a consequence, such exposures would have been small in comparison. Therefore, the default recommendation to add an amount of  $^{230}\text{Th}$  activity equal to the uranium intake activity is appropriate and conservative.

### Radon Breath Analyses for Offsite Raffinate Dumping Operations to FMPC Sites

During 1952 through 1954, to infer radium intakes, 609 breath samples were taken for radon at the site. The samples were sent for analysis to the AEC Health and Safety Laboratory (HASL; now the Environmental Measurements Laboratory) in New York City. At this time, no specific information is available about the measurement method; the assumption is that the ambient radon concentration present during the sampling was included in the result. Therefore, any dose reconstruction based on these uncorrected results would include a claimant-favorable bias.

A typical radon breath analysis result identifies the individual and provides the radon breath concentration measurement results in curies per liter. Annual and composite parametric values for the data are shown in Table 5. In all years, the minimum reported value is  $1 \times 10^{-13}$  Ci/L, which is assumed to be the reporting limit for the analysis. When the reported result is " $<1 \times 10^{-13}$  Ci/L" a value half of that ( $5 \times 10^{-14}$  Ci/L) was used in calculations. This method is endorsed in NIOSH (1993), which is included by reference in the implementing rules for the Energy Employees Occupational Illness Compensation Program Act of 2000 (EEOICPA) [42 CFR Part 82.16(a)]. Approximately 25% of the values were imputed by this method. In addition, approximately 26% of the submitted samples were not successfully analyzed due to processing errors or equipment failures. Lacking data, these were removed from consideration, resulting in a total of 449 valid samples over the period from 1952 through 1954.

Table 5. Radon breath analysis statistical parameters.

Parameter	1952	1953	1954	Composite 1952 through 1954
Number of attempted measurements	140	238	231	609
N, number of valid measurements	84	183	182	449
Number of valid measurements less than the reporting limit	55	54	46	155
Minimum, Ci/L (assumed to be the reporting limit)	5.00E-14	5.00E-14	5.00E-14	5.00E-14
Maximum, Ci/L	9.00E-13	3.30E-12	1.20E-12	3.30E-12

Parameter	1952	1953	1954	Composite 1952 through 1954
Mean, Ci/L	2.66E-13	2.84E-13	2.47E-13	2.66E-13
Std deviation	2.01E-13	3.26E-13	1.79E-13	2.53E-13
Coefficient of variation (std. dev./mean)	0.76	1.14	0.72	0.95
Geometric mean, Ci/L	2.01E-13	2.07E-13	1.91E-13	1.99E-13
GSD	2.18	2.13	2.11	2.13
Median, Ci/L	2.00E-13	2.00E-13	2.00E-13	2.00E-13
84th percentile, Ci/L	4.00E-13	4.00E-13	4.00E-13	4.00E-13
95th percentile, Ci/L	6.85E-13	6.90E-13	6.00E-13	6.00E-13

ORAUT-OTIB-0025, *Estimation of Radium-226 Activity in the Body from Breath Radon-222 Measurements*, provides information on partitioning of the whole-body radium activity among bone, lung, and soft tissue compartments (ORAUT 2005). The radiation dose to various organs from <sup>226</sup>Ra in the body can be derived by selecting an appropriate intake scenario and applying appropriate dose factors to the derived <sup>226</sup>Ra activity level. ORAUT-OTIB-0025 defines a whole-body <sup>226</sup>Ra activity conversion factor for radon breath data as  $2.52 \times 10^5$  pCi of <sup>226</sup>Ra per pCi/L of exhaled <sup>222</sup>Rn.

Table 6 shows the ratio of three thorium isotopes found in the Silo 1 and 2 materials to <sup>226</sup>Ra. These ratios are based on data in ORAUT-TKBS-0017-5, *Technical Basis Document for the Fernald Environmental Management Project (FEMP) – Occupational Internal Dose* (ORAUT 2004, Table 5-16). The ratios from both silos are similar but the more claimant-favorable ratio (the higher value) from Silo 2 data is recommended for dose reconstruction use, because it is not possible to know which material was included with the radium intake.

Table 6. Ratios of Silo 1 and Silo 2 thorium isotopes to <sup>226</sup>Ra.

Isotope	Activity ratio of isotope to Ra-226 in Silo 1 material	Activity ratio of isotope to Ra-226 in Silo 2 material
Th-228	0.0048	0.028
Th-230	0.14	0.29
Th-232	0.0023	0.0038

A comparison was made between the bounding scenario described in Fernald site profile (ORAUT 2004, Section 5.2.4) and the bounding estimate used for this report. The bounding estimate for this report considers the 95th percentile estimate for <sup>226</sup>Ra in the whole body of workers participating in the Silo 1 and 2 transfers. This bounding scenario was based upon air sampling taken during the drum transfer operations and the following assumptions:

- Occupancy time limited by external dose (6 wk/yr),
- Alpha airborne activity at 100 MAC ( $4.5 \times 10^{-9}$  μCi/mL) – maximum air activities measured,
- Breathing rate of  $9.6 \times 10^6$  cm<sup>3</sup>/d,
- No use of respiratory protection equipment, and
- 46% of activity in the air sample being <sup>226</sup>Ra (ORAUT 2004, Table 5-16, Silo 1).

The 95th-percentile whole-body estimate developed from the radon breath analysis is 0.15 μCi of <sup>226</sup>Ra. The whole-body estimate developed in ORAUT (2004) is 0.7 μCi of <sup>226</sup>Ra. These estimates are reasonably close considering the uncertainty and in the ORAUT (2004) air sample-based bounding assumptions. The bounding estimate in ORAUT (2004) is favorable to the claimant. Based on the data in this report, it a lower the bounding estimate in the next revision of ORAUT (2004) could be justified.

### 3.3 RECOMMENDATIONS

Use radon breath analysis data to estimate the  $^{226}\text{Ra}$  whole-body burden for workers associated with K-65 raffinate handling or processing of offsite raffinate transfer to the K-65 silos. The 95th-percentile value observed in K-65 raffinate workers was  $6 \times 10^{-13}$  Ci/L of exhaled  $^{222}\text{Rn}$ , which according to ORAUT-OTIB-0025 is equivalent to 0.15  $\mu\text{Ci}$  of  $^{226}\text{Ra}$  (ORAUT 2005). This value should be assigned to K-65 raffinate workers.

Air monitoring results show that no significant exposure to radon from cold raffinates occurred.

Unmonitored exposure to the impurities in uranium feed materials such as pitchblende and yellowcake might have occurred to workers who sampled or handled these materials. Workers in Plants 1, 2, 3, and 8 might have been exposed. The intake rates from these exposures can be bounded by adding an intake in the ore or concentrate in proportion to the uranium intake as measured by bioassay. Table 7 shows the recommended values for unmonitored isotopes that might be present in uranium mill concentrates. For dose reconstruction, the amount of uranium intake should be calculated from urine bioassay data. The results in mass quantities should be multiplied by the conversion factors in the table to derive the maximum activity intake of each unmonitored isotope.

Table 7. Factors for unmonitored isotopes for the period 1953 through 1962 only.

Isotope	nCi/mg U in urine
Th-230	0.3
Th-228	0.002
Th-232	0.005
Ra-226	0.02
Pb-210	0.02
Ac-227	0.005
Pa-231	0.003

### 3.4 DETAILED TIMELINE AND HISTORY OF K-65 MATERIAL

Beginning March 22, 1949, the K-65 residues from MCW were sent to Lake Ontario Ordnance Works (LOOW) to be stored for future shipment (Heatherton 1949). The initial shipment was stored under canvas in the open; later shipments were placed in an igloo, but some drums remained stored beside the road (Heatherton 1950). Starting in late 1950 and continuing into 1952, much of the drummed material was transferred to a modified water tower that was 165 ft tall. This operation was performed by National Lead Company of Ohio (NLO) personnel from FMPC. In December 1950, it was reported that there was a backlog of 11,000 drums in the igloo and new drums coming from MCW were going directly into the tower (NLO 1977; LOOW 1951). The estimates of the amount of material in the tower included values of 3,874 t, 3791 t, or 1,757 t (TAC 1982, p. 31). The K-65 material in the tower was ultimately moved to underground concrete water tanks in Building 411 on the LOOW site during the decontamination and demolition project in 1984 and 1985 and listed as 3,891 t (Glenn 1987).

A number of drums were not transferred to the water tower; the number has been reported to be between 5,000 and 6,000 drums (Boback 1978; TAC 1982; Hershman 1952; Heatherton 1952b; Vogel 1989). These drums were of varied sizes, from 30 to 55 gal. Between September 22 and November 14, 1952, 4,481 drums were transported from LOOW to FMPC (Schuman 1952; Strattman 1952; Boback 1978; Karl 1952). The lower number of drums actually transported is likely due to the consolidation of drums at LOOW due to drum conditions.

In 1951, MCW began shipping K-65 material to FMPC rather than LOOW (Walden 1952, Vogel 1989). Between September 26, 1951 and April 30, 1952, 11,253 drums were sent from MCW to FMPC. A

total of 25,000 drums were shipped (Vogel 1989). As an example of the rate of these shipments, in late 1952, FMPC was receiving five railcars per week from LOOW and three railcars per week from MCW (Heatherton 1952a). Each rail car had the capacity to hold 96 drums. The shipments from MCW continued until January 1956 (Vogel 1989). This K-65 material was transferred to Silos 1 and 2.

From 1954 to 1957, K-65 material was transferred to Silo 2 from Q-11 processing at FMPC. The dates of the specific campaigns are as follows:

A campaign	April 1954–June 7, 1954
Pilot Plant	September 24, 1954–October 14, 1954
B Campaign	October 23, 1955–February 3, 1956
C Campaign	July 31, 1956–October 30, 1956
D Campaign	March 7, 1957–April 2, 1957 (the date of the memo, Vogel 1989)

Processing of the Q-11 ores would have preceded these dates by 1-2 weeks (Vogel 1989).

Miscellaneous other materials were placed in these silos (most likely Silo 2) as described in Table 8 (Vogel 1989).

Table 8. Materials placed in Silo 2 after Q-11/K-65 processing was complete.

Amount	Period	Source	Reference
5 drums	4/11/1959	Clean out residue	Attachment 10 to Vogel (1989)
150 drums	6/10/59	Wastes and residues from K-65 activities	Attachment 12 & 13 to Vogel (1989)
Some ionium	6/20/60		Attachment 14 to Vogel (1989)
75 lbs	4/16/65	K-65 and Q-11 from Lucius Pitkin–New York City	Attachment 15 to Vogel (1989)
11 drums	8/15/66	7 drums from Middlesex Sampling Plant and 4 drums of cleanup material from hot raffinate building	Attachment 16 to Vogel (1989)

The total K-65 material in the two silos was 19,385,126 lb, including about 380,000 pounds from the processing of “rum jungle” ore from Australia in 1957 and 1958 (Vogel 1989, Attachment 18; Shaw 1968; Robinson 1973). About 25% of the material was generated at FMPC (Robinson 1973). In a memo from W. E. Shaw to J. E. Carvitti and J. R. Lynch dated May 24, 1968 (Shaw 1968), an attachment dated February 18, 1959, identifies the source of the material in the two silos. The page is entitled “K-65 Residue in 1 & 2 Storage Tanks” with a note, “check on this date 2/18/59 from 101 records and avg analyses.” The data in Table 9 is contained on the rest of the page.

Table 9. Inventory of K-65 material in Silos 1 and 2.

Source	Weight	Drums	Ra-226 (mg/t)	Ra-226 activity (Ci)
NLO	5,363,712	12,130	300	804
TOA	2,463,363	6,460	750	924
MCW	11,558,051	25,260	500	2,890

This attachment is handwritten and signed by J. E. Carvitti and T. C. Feist. This document is one of the sources for the total weight of residues in the two silos. A memo that S. L. Hinnefeld wrote in 1982 discusses estimates of the amount of material and the <sup>226</sup>Ra content of the two K-65 Silos (Hinnefeld 1982). The following values were discussed in this memo:

- 1,650 Ci – The memo indicates that this is the most commonly reported value (Hinnefeld 1982). It is also quoted in “History of the Operation of the FMPC by NLO, Inc,” by Mead et al. (1985).

- 2,960 Ci – The memo derives this from a reported <sup>226</sup>Ra content in a Draft SAR for the FMPC Waste Storage area (Hinnefeld 1982).
- 1,720 Ci – This number was derived from a 1957 memo that assumed the Belgian Congo ore was 40% U (Hinnefeld 1982). [A more common range was 50-80% (see Section 3.1); 60% U which would yield 2,580 Ci.]
- 4,600 Ci – The memo indicated this value was found in a Technical Division memo and indicated it might be old and therefore suspect, but noted that there was no reference to support the value. The data that is discussed above from Shaw (1968) appear to be the source of this value. Several other pages in that document reference this value for the <sup>226</sup>Ra content of the K-65 silos.

Vogel (1989, attachment 17, p. 21) has a value for the alpha activity for K-65 material of  $1.19 \times 10^4$  dps/g. This document is titled "The Origin of K-65 Material" and was written by a Westinghouse employee (Vogel) in 1989. This value yields a total of 2,770 Ci in the 19,385,126 lb of K-65 material in Silos 1 and 2.

As can be seen from the information above and data in this timeline presentation, the exact activity of <sup>226</sup>Ra in the two silos is not known for certain, but the amount of activity is certainly in the range from 1,650 to 4,600 Ci.

#### **4.0 RECYCLED URANIUM AT FMPC**

##### **4.1 HISTORICAL BACKGROUND AND PROCESS LIMITS**

The term Recycled uranium (RU) has been used in the nuclear weapons complex in a number of contexts; e.g., recycled scrap materials from uranium manufacturing operations, etc. as well as uranium that has been recovered from reactor fuel and targets that have been irradiated in production and/or research reactors. Throughout much of the history of nuclear weapons production there was no separate and distinct accountability category for RU, which contributed to confusion in developing the mass balance reports referenced in this report and discussed later. The term RU in this report is used only for reactor-irradiated and recovered uranium.

RU was recovered in the chemical processing plants at the Hanford, Savannah River, West Valley and Idaho Chemical Processing Plant sites. RU was known to contain trace quantities of transuranic elements (referred to as TRU in the rest of this report) and trace quantities of fission product impurities. The primary contaminants were Pu-238, Pu-239, Np-237, and a fission product, Tc-99. The Pu-239, Np-237, and Tc-99 were the radionuclides of greatest concern and were tracked and documented for control purposes. The levels of contaminants were recorded in parts per billion (ppb) on a uranium mass basis.

The first uranium feedstock introduced to the Fernald plant in 1953 was high grade ores (50-80% uranium), i.e. pitchblende ores from the Belgian Congo and other locations, which could be processed in a modified PUREX process directly to high purity uranium. Other uranium feed stock came from low grade ores, which was processed near the mines by mills that produced natural uranium in the form of yellow cake (U<sub>3</sub>O<sub>8</sub>) in the range of 60% uranium. The yellow cake required further processing by PUREX to a high purity state. When RU from the chemical processing plants was introduced to the process feed streams at Fernald it was a high purity oxide and was normally blended with natural uranium or with other existing feed stocks.

Relatively small quantities of RU (a recorded mass of 45 MTRU) from Hanford were received at FMPC as early as 1955 (2000a,b) but did not constitute a feed stream component of significance or at

all. The first significant quantity of RU was received by FMPC for processing on February 13, 1961, in a shipment of UF<sub>6</sub> that was to be converted to UF<sub>4</sub> (DOE 2000a) and introduced to the feed streams of the plant.

#### 4.1.1 Major Contaminants in FMPC RU

Table 10 details DOE's estimate of quantity and characteristics of RU at FMPC. Approximately 218 g of Pu-239, 25.7 kg of Np-237, and 332 kg of Tc-99 were received at FMPC in 246,683 metric tons (MT) of RU during the history of the plant. (DOE 2000a). The historical average of contaminants in RU was approximately 1 ppb Pu-239, 104 ppb Np-237, and 1,346 ppb Tc-99.

A limit of 10 ppb total plutonium in RU was set by the nuclear weapons complex (primarily Oak Ridge) for use at Hanford (ORAUT 2010; and DOE 2000b). Initially no numeric limits were specified for Np-237 or Tc-99. The plutonium limit of 10 ppb was set such that the radiological impurities did not exceed 0.1% of the alpha activity from natural uranium. Table 11 lists the activity percentages and illustrates that the activity ratios of the impurities vary as a function of the activity of uranium that changes with enrichment. Note that the percentage is higher for depleted uranium, but in each case

Table 10. Average characteristics of RU processed at FMPC.

Uranium enrichment	MTU <sup>a</sup> receipts	Percent uranium receipts	Total Pu-239 (g)	Pu-239 (ppb)	Total Np-237 (g)	Np-237 (ppb)	Total Tc-99 (g)	Tc-99 (ppb)
Enriched	60,181	24.4	207.9	3.5	19,048	316	328,740	5,382
Normal	89,649	36.3	4.1	<0.1	3,026	34	1,197	13
Depleted	96,853	39.3	5.7	<0.1	3,669	38	2,061	21
Total or average	246,683		217.7	0.9	25,742	104	331,998	1,346

a. MTU = metric tons of uranium.

the radioactivity from uranium alpha emissions far exceeded the radioactivity from plutonium alpha emissions.

Table 11. Activity in RU containing 10 ppb and 100 ppb <sup>239</sup>Pu for various uranium enrichments.

Uranium enrichment	Uranium activity concentration (pCi/μg U)	Plutonium activity concentration in 10-ppb RU (pCi/μgU)	Plutonium activity concentration in 100-ppb RU (pCi/μgU)	Fraction of total alpha activity from Pu-239 in 10-ppb RU (%)	Fraction of total alpha activity from Pu-239 in 100-ppb RU (%)
Depleted	0.422	6.175E-04	6.175E-03	0.154	1.54
Natural	0.685	6.175E-04	6.175E-03	0.09	0.90
1% enriched	0.976	6.175E-04	6.175E-03	0.06	0.63
2% enriched	1.616	6.175E-04	6.175E-03	0.04	0.38

#### 4.1.2 Mass Balance, Material Flow, and Pu-239 Levels in RU within the Weapons Complex

As will be explained later, from the beginning of operations urinalyses for uranium was conducted extensively for purposes of controlling biological heavy metal concerns. The presence of RU contaminants was recognized but was not specifically analyzed due to the anticipated low levels and related low hazard contribution. The approach to bound the maximum credible dose from these contaminants is to add a default concentration of each contaminant to the extensive uranium-in-urine data. For this reason, it is not necessary to precisely quantify the mass balance of RU and material flow of RU in the weapons complex to establish a bounding estimate of the contaminant concentration in RU at FMPC. It is necessary to have an understanding of the materials flow into the processes but basically to know the relative concentration in each of the major process streams, such that bounding level concentrations can be established. Figure 5 illustrates the flow of RU into and out of FMPC.

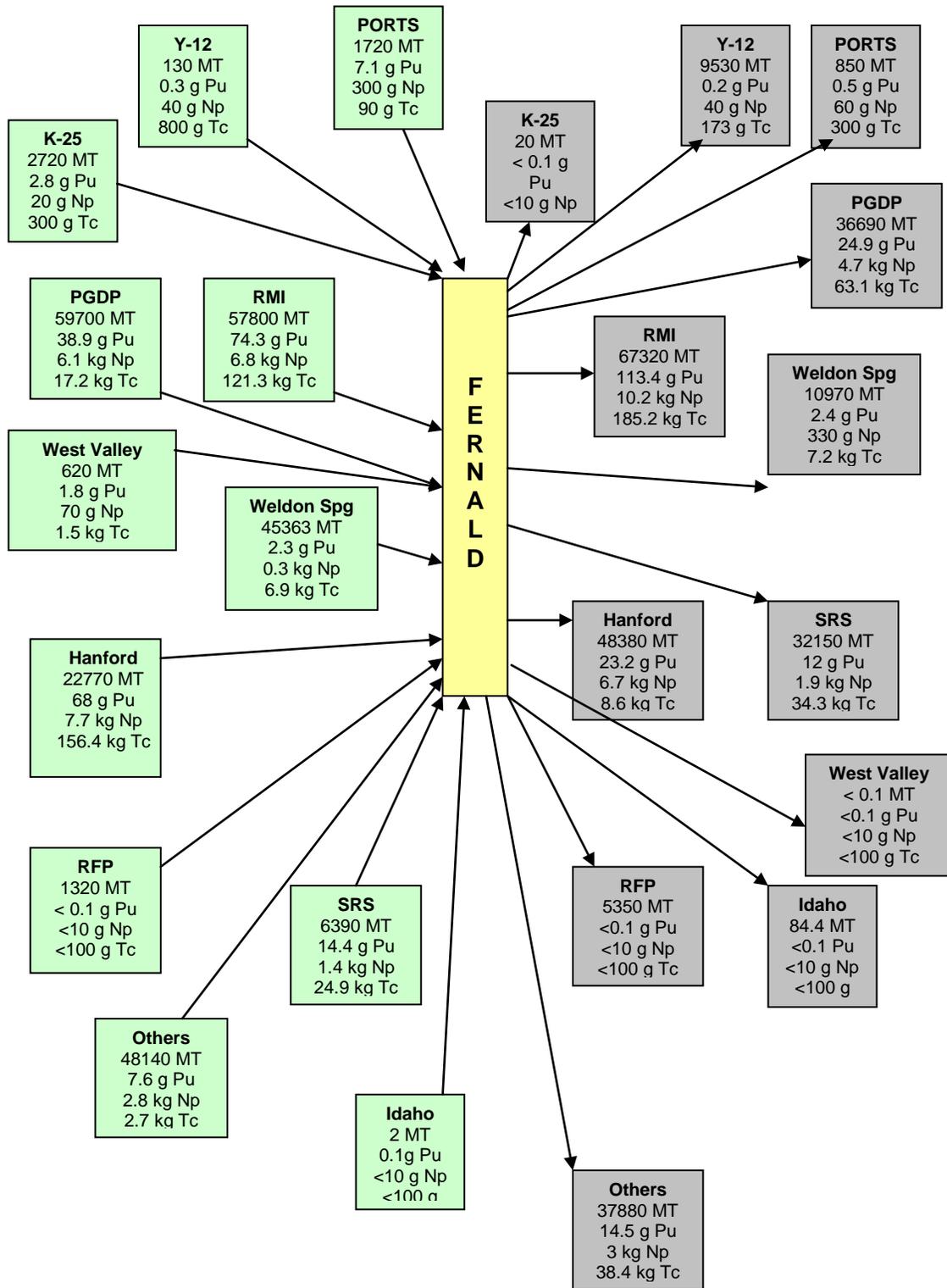


Figure 5. RU mass flow in and out of FMPC.

It is true that the DOE documents pertaining to mass flows of RU in the weapons complex include some discrepancies, but these discrepancies in mass flow do not preclude use of the data to define bounding assumptions. The inconsistencies in the amounts of RU in various reports can be attributed primarily to definition for protection purposes, i.e. many plants simply classified and treated as such all the uranium in the plant as RU once the RU was introduced in the processes - due to mixing, blending, etc. In addition, the availability of data at the time that each report was prepared, the scope

of each report, and the degree to which conservative assumptions were used contributed to the apparent discrepancies in mass flow. However, the DOE (2003a) mass balance report documented the quantities of contaminants leaving the RU generating sites (primarily Hanford). These numbers, though of interest in terms of understanding the flow and quantities of contaminants were not used in determining the recommended defaults. The defaults were determined by analyses of concentrations in the various processes and reported in Appendices F and F.1 in the Ohio Mass Balance report (DOE 2000a). The defaults were chosen such that they bound the highest values of all the processes, with the exception of the concentrations in the GDP tails and wastes.

The concentration of the contaminants also varies slightly with the source of the data. For example, the mass concentration of plutonium in DOE (2000a) is 0.9 ppb; for the complex-wide rollup, the mass concentration is 3.7 ppb; in DOE (2003), the mass concentration from the primary sites is 4.2 ppb. These values are essentially the same and all well below the 10 ppb control guideline specified for Hanford.

Further discussion of this issue and the DOE studies pertaining to uranium mass flow to and from FMPC are described in section 4.4.

#### 4.1.3 Other Contaminants in RU

The chemical separation of fission products and activation products from RU in the uranium processing plants was excellent in order to meet the uranium purity requirements, but did allow ppb levels of contaminants. Routine process chemical analysis at Hanford included gross beta and gamma measurements. These results were compared to the baseline measurements from aged natural uranium (NU) (DOE 2000b). Limits on these other contaminants were set so that the beta activity in RU was less than 100% of the beta activity in comparison with aged NU and gamma activity in RU was less than 100% (average of a ten lot sample with no sample > 300%) of the gamma activity in aged NU. The numeric value for these limits was 0.685 pCi/μg NU which is equal to 310 μCi/lb NU. With the advent of gamma spectrometry in 1967, the limits included isotopic levels as listed in Table 12.

Table 12. Limits for other contaminants in RU.

Isotope(s)	Limit (μCi/lb U)	Limit (pCi/μg U)
Gross beta count	<100% beta activity of NU	<0.685
Gross gamma count	<100%–300% gamma activity NU	<0.685
Zr/Nb-95	15	0.033
Ru-103/Ru-106	50	0.11
All other isotopes	2	0.0044

Gamma spectrometry performed at the Hanford Site did not identify <sup>137</sup>Cs in RU, although trace quantities were identified in the analyses of dust collector data taken at FMPC. Strontium-90 and 89 could have been among the other contaminants, but would be just 0.6% of the gross beta activity from NU and 1% of the activity from DU. Detectable levels of Zr/Nb-95 and the ruthenium isotopes (Ru/Rh-106) have been reported in RU samples from the chemical process plants. It was reported that additional reprocessing was necessary, primarily due to ruthenium contamination, until the product met the specifications. (DOE 2000b).

Some analyses have indicated that thorium isotopes were present in trace quantities. Insufficient data exist to establish a general default value for all sites, but modeling using the ORIGEN2 code has indicated the possible levels. For the major primary source (the Hanford Site), the analysis has indicated a default value for thorium of 10 ppm U. The values in Table 12 represent the acceptance limits used by Hanford. Specific analytical information for trace concentrations of other fission products as documented in Hanford references (DOE 2000b; ORAUT 2006a) are listed in Table 13..

Table 13. Other radioactive contaminants in RU at Hanford.

Element/isotope	Observed range
Plutonium (primarily Pu-239)	<1–2 ppb U
Neptunium (primarily Np-237)	0.04–0.16 ppm U
Thorium	8–10 ppm U
Technetium	3–4 ppm U
Ru-103/106	<6 $\mu$ Ci/lb U
Zr/Nb-95	<4 $\mu$ Ci/lb U
Other gamma emitters excluding Tc-99	0.09–0.75 $\mu$ Ci/lb U

#### 4.1.4 Tower Ash and Plutonium Out of Specification

The gaseous diffusion process concentrated the RU contaminants (primarily due to lack of volatility of the fluoride compounds as compared to those of uranium) in tower ash and other relatively low volume residues in the UF<sub>6</sub> conversion and handling processes. These ash and residues also contained significant amounts of uranium, which required that they be recovered according to economic discard criteria. In 1972 the decision was made by the AEC to process these materials from the Paducah Gaseous Diffusion Plant (PGDP) and the other GDP sites. Fernald was directed to process most of these materials. This decision introduced RU contaminants to Fernald with Plutonium-Out-of-Specification (POOS), in other words above the established specification of 10 ppb. POOS feed stock from PGDP was sent to FMPC for reprocessing with the prior knowledge that the material was above the established contamination limit. The 1970s and increased amounts in the 1980s GDP shipments accounted for a major portion of the total TRU inventory at FMPC, essentially doubling the plant inventory over this time period (Bassett et al. 1989 and DOE 2000a).

Consequently, exposures to RU contaminants greater than the 10 ppbU could have increased significantly starting in 1973.

According to the Fernald Closure Project (FCP) – Internal Dosimetry TBD (Tomes 2001), most of the POOS uranium on the site in 2001 contained less than 80 ppb plutonium. However, an examination of the statistical data sheets in the F and C Appendices of DOE (2000a) indicates that maximum values in some subgroups (as determined by the 95th percentile of a log normal distribution) ranged up to approximately 400 ppb Pu. These values are summarized in Table 14. A notable exception is the tower ash from PGDP, which had a mean value of 1732 ppb Pu (range of 0.6 to 3,505). This maximum-level POOS material was handled as a recognized short-term risk during feed preparations and blending operations. Additional protective precautions were applied during these short term tasks. These precautions included airline respiratory protection, protective clothing over normal plant clothing which was comprised of head covering, gloves, shoe covers. Tape was applied at ankles and sleeves. Weekly urine bioassay was required of those involved. (Author unknown) All other process subgroups as a result of the POOS insertion had 95th percentile values less than 400 ppb Pu. Thus, the short duration of the injection of the subgroup 10A materials into plant inventory materials with the immediate drop to subgroup 10B levels of 35 ppb Pu (range 2.5 to 53.1 ppb Pu) make the default level of 400 ppb recommended in this report adequate to ensure claimant favorability.

Table 14. RU contaminant levels by process subgroups.<sup>a</sup>

Subgroup	Description	Pu ppbU @95%	Np ppbU @95%	Tc ppbU @95%
1A	Miscellaneous	133	3,692	3,060
1B	Miscellaneous – minor offsite	1.6	179	1
2	UF <sub>6</sub> source UF <sub>4</sub> (GDP tails)	1.2	84.	171
3	UF <sub>6</sub> source metal and scrap	0.01	4.2	15
4	Normal U product, residues, and scrap	0.12	139	33
5	Enriched UF <sub>6</sub> source products/res.	3	109	15,410

Subgroup	Description	Pu ppbU @95%	Np ppbU@95%	Tc ppbU @95%
6A	UO <sub>3</sub> PUREX source (A508)(unblended)	4.6	655	14,119
6B	LEU products A508 UO <sub>3</sub> /UF <sub>4</sub> (low cross)	4.1	643	19,732
6C	LEU products A508 UO <sub>3</sub> /UF <sub>4</sub> (high cross)	34	1,198	4,823
6D	A500 coded enriched residues	7.6	246	2,701
6E	SRS UNH	37.8	nm <sup>b</sup>	nm
6F	SRS UO <sub>3</sub> – not shipped to FMPC	4.6	nm	nm
7A	A508 based derbies	17	513	2,870
7B	A508 based ingots and metal	1.7	433	133
8	Enriched MgF <sub>2</sub>	342	4,417	4,619
9	Incinerator ash and scrap residues–GDPs	283	5,116	474
10A	Tower ash and decontamination residues	1732	17,277	4,445
10B	UO <sub>3</sub> from tower ash	34.9	842	4,154
11	Waste residues	152	10,666	6,991

- a. \*Though the results in the table are all reported in ppb U, this measure may appear less meaningful in subgroups in which there is a reduced amount of uranium (in single digit percentage levels) such as subgroups 8 and 11, in which the MgF<sub>2</sub> and chemical plant raffinates accumulated some isotopes, but were relatively low in uranium by design, but still continued to be a process stream to recover uranium above the economic discharge levels. Note that even with decreased U levels the contamination default can still be a ratio of U.
- b. nm – not measured.

Over time, much of the POOS material was blended with the natural uranium feed stock and with RU of typical concentration. This was done to maintain the plant-wide limits that had been determined to provide an adequate level of safety, i.e., staying near the average complex-wide plutonium specification of 10 ppb). Workers handling the POOS materials directly prior to its being blended with other materials (Plant 1 and other locations) were provided with special protection. As a background comment, a small amount of this POOS material from the GDP was inadvertently stored for a period of years. The discovery of this material in 1985 was categorized as an unusual event with the material being subsequently processed and disposed.

After processing the PGDP POOS, the blended uranium oxide resulted in temporary storage of uranium with a maximum concentration of Pu of 43 ppb U. In addition, several of the uranium purification processes resulted in concentrating the TRU contaminants in waste streams. Formal investigations were conducted that traced the TRU contaminants through the various plant processes; one such study in 1975 defined contaminant affinity for the metal production furnace mold materials (some TRU studies are referenced in DOE 2000a). These analyses demonstrated that the extraction and metal conversion processes concentrated plutonium, neptunium, and uranium daughter product contaminants in the raffinate waste of the liquid extraction columns and in the MgF<sub>2</sub> from the thermite metal conversion furnaces. MgF<sub>2</sub> slag from the thermite metal reduction process was also processed through a slag leach process and then through the chemical extraction plant. These concentration points or areas were identified by analytical methods in early years. The discussions in this report make use of studies of the data from those early reports (DOE 2000a). The recommended defaults account for all of the possible concentration points in the processes.

## 4.2 WORKPLACE AND PERSONNEL MONITORING ASSOCIATED WITH RU

The FMPC Health and Safety staff was aware that TRU and fission product contaminants were present in the recycled uranium. The biological concerns were thought to represent an increase of less than 10% in hazard level compared to uranium that had never been irradiated. Radiological controls were determined and adjusted based on routine air sample analysis and urine bioassay for uranium.

Before 1986, routine TRU analyses was not performed for either airborne or urine activity, although a few examples of specific analyses of Pu on air samples were recorded and indicated the uranium to

be controlling. Exposure controls were set at levels that prevented chemical toxicity of uranium assuming that these controls were sufficient for all the radiological hazards (Bassett et al. 1989). Although the alpha activity from the TRU alpha emitters would have been collected and detected on the air samples, the reported results were all considered to be primarily uranium and compared to the Maximum Air Concentration (MAC) for uranium. Consequently bioassay measurements were not routinely performed on workers for the contaminants associated with RU until 1986. Note SRDB 4613 referenced in Section 4.2.1 below.

#### **4.2.1 In Vivo Analysis Data**

A review of the Oak Ridge Y-12 radiation protection program and internal dosimetry technology (Cofield 1959a; Scott 1964; Steckel and West 1966; West 1979) indicates that the internal dose technology, techniques, procedures, and philosophy used at FMPC were similar to those at Y-12. The fact that the Oak Ridge Y-12 Mobile In Vivo Radioactivity Measurements Laboratory (MIVRML) provided routine service to the FMPC in vivo internal dosimetry program is an indicator of the working relationship the FMPC staff had with the Y-12 program. However, prior to 1988 this technology did not provide detection for TRU at the very low levels that existed in the plant from RU contaminants. Typical results of the MIVRML were lung burdens for uranium and U-235, and data to determine thorium uptakes. Though TRU analyses were attempted, the limitations of the MIVRML, which included limited detection sensitivities for TRU isotopes and infrequent counts, restricted the ability to detect the anticipated levels at FMPC and in fact could not detect levels that met regulatory limits for chronic exposure to TRU isotopes. This lack of capability was a technological limitation of in vivo counting within the nuclear industry in general during this period of time. Early in 1989 an in vivo counting facility was constructed on the FMPC site to replace the mobile in vivo counting facility from Y-12.

Before 1989 when DOE Order 5480.11 was issued, bioassay data at FMPC was not routinely used to estimate intakes and internal organ doses. Instead chest (lung) counting was used to estimate the fraction of a Maximum Permissible Lung Burden (MPLB) for uranium primarily and infer annual dose by multiplying by 15 rem/y per MPLB. Lung burdens for thorium were also determined and recorded. Workers were removed from the higher exposure work or placed on radiological work restrictions when in vivo results exceeded 75% MPLB. The in vivo counting frequency was set at once per year for the high exposure potential work groups (chemical operators), with the exception that workers exceeding 50% MPBB were counted more frequently. Annual exposure reports listed uranium lung burdens in percent of MPLB only and did not address the systemic radiological burdens. See SRDB 4613, R. C. Heatherton paper, Occupational Health Experience With A Contractor Uranium Refinery, April 1975 for a discussion of the results of this program during the early time periods.

In summary, direct in vivo lung monitoring began in 1968 with the MIVRML and continued in 1989 with the FMPC IVEC counting facility until 2001. No RU contaminant analyses were reported in the in vivo records before 1989. Although there would have been some ability to detect TRU materials in the range of 0.1 to 10's of nCi depending on the radionuclide, the in vivo counts were not performed with a consistency or frequency to be of significant value in TRU dose reconstruction.

#### **4.2.2 Urine Analysis Data**

Measurements of uranium in urine were compared to limits based upon preventing toxic effects from heavy metals. A more formal program of internal dosimetry was introduced in 1986 at the change of contractors for the FMPC site. An immediate emphasis was placed on a more formal evaluation of the transuranic materials and fission products. From April 1986 to 1989, 675 bioassay samples from 441 workers were collected for plutonium analysis. "Those samples were collected primarily from workers who were expected to be and, in some cases, were actually involved in POOS processing and cleanup campaigns in Plants 4 and 8." Only 10 individuals exhibited quantities of plutonium in the

urine above 0.02 dpm/sample. (This contractual urine MDA was determined to be unrealistically low and later adjusted to 0.13 dpm/sample for Pu urine analyses). Further investigation (follow up samples) revealed that 3 of the 10 were below the specified detection limit of 0.02 dpm per sample. All of the 10 individuals were flown to Hanford, where in vivo counts were conducted for plutonium. All results were below the detection limit of the Hanford in vivo analyses. The conservative assumption was made by the site contractor that seven of the ten were technically at the urine MDA. Directions were given to "regard those results as valid indicators of exposure and to calculate the plutonium dose commitments to those workers based on these worst-case assumptions. The dose commitments are to be reported in each employee's permanent exposure record." (Bassett 1989). The FMPC radiation protection practices were adjusted to more rigorously account for those trace isotopes within the DOE radiation protection standards in effect at the time..

The information above is taken from the report of a DOE team of investigators and documents an organized effort by an incoming contractor to verify the previous assumptions that TRU exposures were not a major exposure source. The bioassay and in vivo results will be found in the claimant files and might be used by NIOSH dose reconstructors in some cases to assure bounding of doses. However, this data is insufficient to assign intakes/internal dose from contaminants in RU for all operational periods. The default intake analyses proposed in this paper are based upon recorded and maximized ratios of RU contaminants in uranium from which contaminant intake analyses can be performed.

#### 4.2.3 Air Sampling Data

Before February 1986, smears or air sampling filters were not specifically nor routinely analyzed for plutonium, neptunium, or thorium isotopes (Basset 1989), although these radionuclides would have been detected by the routine gross alpha counting. In 1989, several sets of air and surface smear samples from Plants 4 and 8 were analyzed for total uranium,  $^{239}\text{Pu}/^{240}\text{Pu}$ ,  $^{238}\text{Pu}$ ,  $^{237}\text{Np}$ ,  $^{228}\text{Th}$ ,  $^{230}\text{Th}$ , and  $^{232}\text{Th}$ . As stated above, a few examples of specific analyses of Pu on air samples were recorded at earlier times and indicated the uranium to be controlling. Table 15 lists the results of the air samples in derived air concentrations (DAC) from these analyses. The DAC ratios are provided not for dose reconstruction purposes, but only to indicate the relative measured levels of TRU contaminants in the plant in the 1989 time period. However, the ratios lend credence to the default assumptions proposed in this report, which are intended to account for the unmeasured TRU in the plants.

Table 15. Derived air concentrations from 1989 RU isotopic analysis.

Isotopes	Derived air concentrations
Pu-238, Pu-239, Pu-240	0.1-1% of Type S
Np-237	<0.03% to 0.1% of Type S
Th-228, Th-232	Ranged up to 3% of the Type S
Th-230	6 to 10% of Type S

Note: These initial results indicated that for those samples where the activity fraction of each radionuclide could be compared (generally those with a total alpha activity exceeding  $1 \times 10^{-12} \mu\text{Ci}/\text{cm}^3$ ), uranium was the controlling activity in air samples. In addition the results were reported in percentage of the Derived Air Concentration limits in effect at the time, which were also in the range of  $1 \times 10^{-12} \mu\text{Ci}/\text{cm}^3$ .

During the next several years, a major sampling and analytical program examined hundreds of process samples, air samples from the process areas, and biological samples for plutonium, neptunium, thorium, technetium, and uranium. The air monitoring program was used to establish work controls, such as respiratory protection requirements for workers, and was not routinely used to establish internal intake or exposure estimates although DAC-hr exposure evaluations were placed in the personnel dosimetry files for some specific operations, i.e. thorium materials.

### 4.3 RU IN WASTE STREAMS

#### 4.3.1 Process Raffinates and Other Wastes Below the Economic Disposal Limit (EDL)

This section deals only with raffinates and waste products during the period (post 1961) associated with processing RU. (Raffinate waste products associated with the processing of high-grade pitchblende ores and processed yellowcake from the uranium mines and mills are covered in Section 2.0 above.) The primary defaults recommended in this report for trace-level contaminants that accompanied RU rely upon routine bioassay analyses for uranium. There are processes in which concentrating mechanisms existed, and in which the uranium in those process streams will be low by design. Table 16 lists the processes that occurred at FMPC (DOE 2000a). These are process streams, determined to be waste on the basis that the remaining uranium is “below the economic disposal limit” (EDL) – but not zero.

Table 16. Processes that concentrated RU contaminants.

Process step/activity	Explanation/comments
Chemical extraction	87.2% initial fed Pu and 41.6% initial fed Np reports to the UO <sub>3</sub> product stream
UF <sub>6</sub> to UF <sub>4</sub>	Concentration in heel of constituents and remains with container
Hydrofluorination	Potential Tc vaporization
Reduction	46% Pu and 63% Np report to MgF <sub>2</sub> slag
Vacuum casting	5% Pu and 64% Np report to crucible and mold residues
Metal pickling	Surface residues (higher in Pu and Np) dissolve in pickling acid
Machining	Top Crops greater than metal product in feed

The predominant process streams of concern on the basis of low uranium content are enriched MgF<sub>2</sub> from the metal thermite process, a variety of raffinates, normal and depleted MgF<sub>2</sub>, sump cakes, etc. from liquid extraction processes. Enriched MgF<sub>2</sub> was processed 1) for uranium recovery, 2) reuse, and 3) disposal as waste, while the raffinates were treated or processed and disposed in silos, drums and/or disposal lagoons. On the surface the default approach of adding RU contaminants to the uranium intake determined from uranium bioassays for workers involved with these processes can appear to be less reliable. The following discussion presents a description which explains the choice of the default ratios as claimant-favorable.

#### 4.3.2 Daily Weighted Exposure (DWE) Studies

During the early period from 1953 to 1968 (including the RU period) air sampling time weighted exposure studies were performed throughout the plants. These studies illustrated the relationship between high concentrations and short exposure times to other more chronic exposures. The studies performed in the work locations defined as raffinate areas were generally lower in concentrations than those areas with higher concentrations of uranium by a factor of 10 or higher. DWE levels were generally in the 0.1 MAC levels or at the MDL. These data is shown in Section 3.2.

Even though the raffinates may have contained a relatively high ratio of RU contaminants to the uranium content compared to other plant locations, the air concentrations in the raffinate area were quite low. The lower concentrations in the Plant 3 Raffinate areas are the result of these processes being liquid based and totally contained within closed piping systems. These enclosed systems would have provided very little exposure to the workers, which the Daily Weighted Exposure studies demonstrate. The primary source of exposure occurred in the Plant 2 area where ore dumping operations were conducted. From the information above, exposure in the raffinate area was a factor of 20 less than in the Digestion areas, yet the MPC, based upon the measured isotopic composite, was only a factor of 15 smaller. For a specific example, if the exposure in the Digestion area were assumed to be at the U-Nat MPC of 220 dpm/100 m<sup>3</sup>, the corresponding exposure in the Raffinate area would be 11 dpm/100m<sup>3</sup>, less than the MPC calculated for the raffinate mixture.

Since the personnel of Plant 3 were rotated in the various jobs it was assumed that over a period of time each operator within a group would receive a similar exposure, i.e., no class of operators were assigned exclusively to the raffinate areas. This would tend to result in exposures to uranium that combined Pu contamination levels in the <1 ppbU with those in the range approaching 400 ppbU. If the Claimant were assumed to work in the Digestion areas full time, disregarding any work in the Raffinate area, the dose reconstruction recommendations listed in Section 4.3 below are sufficiently favorable to claimants to bound the potential exposures from the raffinate area based upon uranium intake analyses.

The DWE Exposure Study reports from 1968 and 1969 (Ross, Leininger, and Lawrence 1968; Ross, Leininger, and Zimber 1969) also included data from the urine sampling program. The following paragraph taken from the 1968 report documents the NLO policy and basis for their reporting levels.

*AEC Manual Chapter 0502 requires that all employees who are shown by bioassay to have one-half of one permissible body burden to be reported. National Lead Company of Ohio has arbitrarily chosen to report all employees whose urine results average 0.050 mg uranium per liter over a six-month period. This is believed to represent a much smaller uranium burden than the one referred to in the AEC Manual.*

Only rarely did any worker exceed the value representing one-half of the permissible body burden and most of the results were less than the value used for internal reporting, which was 0.025 mgU/L. These bioassay results are relatively low considering the results of the Exposure Studies, but reflect the conservative approach that was used in the study results to disregard the use of respirators, which in the higher exposure jobs was required. The uranium urinalyses results demonstrate that reportable uranium exposures occurred in all areas, including those identified as "raffinate" areas. Thus, RU contaminant intakes can be calculated by default ratios to positive uranium urine sampling results.

#### **4.3.3 Other Factors Associated with Recycled Uranium in Waste Streams**

The following mitigating considerations also support the default recommendations:

1. The primary source of RU from the reactors was generally not initially submitted to chemical extraction due to the high purity of this product. Thus, the levels of RU contaminants in the process areas will be higher prior to the concentration mechanisms.
2. Only a small portion (approximately 10%) of the process streams (scraps and process rejects, etc.) are processed by dissolution and chemical purification, and are diluted then with non-RU sources. Hence, the total quantities of RU contaminants in the processes that generate the raffinate streams are lower than in the other process areas.
3. The raffinate process has two aspects that minimize worker exposures: (1) being enclosed in process vessels and piping, which provide containment, and (2) the wet or moist material forms.
4. The raffinate process is a waste stream and is directly disposed. Generally, there is no continuing or further processing of the materials, which results in minimization of the exposure potential.

#### **4.4 RU MASS FLOWS AND DISCREPANCIES IN MASS FLOW REPORTS**

In the late 1990s, DOE recognized the need to better understand the internal dose potential of contaminants from the historical mass flow of RU in the weapons complex. Reports were generated

by each of the nine groupings of DOE sites or areas that were involved with RU. DOE (2000a) covers the Ohio sites, including the FMPC, and it is the primary source cited in this report.

Table 10 shows the average contaminant concentrations of the major contaminants in the RU that was received at FMPC during the period when RU was processed (1961-1999).

Process subgroup evaluations are summarized in Table 14. In many instances, the data from the process subgroups did not conform well to normal or lognormal distribution patterns so “bootstrap” analysis was employed. The results of these statistical analyses are presented in the DOE 2000a report. This statistical technique uses a sampling approach to approximate representative values for the data set (DOE 2000a). For purposes of this report and default analyses, the lognormal distribution assumption and defaulting at the 95th percentile level was used to assure a more conservative default for all plant workers for the time period of the GDP wastes processing (1973-1986).

An additional study was published by the DOE Office of Security in May, 2003 (DOE 2003), with the specific purpose of resolving the recognized inconsistencies in the mass flows among the nine site reports published in 2000. Table 3-4 of DOE (2000a) contains the data related to receipts and shipments at FMPC taken, in part, from WMCO (1986). However, DOE (2000a) defines “recycled uranium” at Fernald as all uranium received or shipped after 1962, while the receipts and shipments listed in WMCO (1986) were total uranium. The DOE (2000a) report acknowledges that the mass for shipments of RU exceeded the mass for receipts of RU “due to limiting the envelope for data to after recycled uranium started, i.e., not including the years prior to 1962 at the FEMP.” Due to blending during chemical processing, any uranium received prior to 1962 would likely have become RU if it were shipped after 1962 and therefore the apparent mismatch in RU receipts and shipments is a logical outcome.

The DOE Ohio Field Office report (2000a) discusses the difficulty of determining exact amounts of recycled uranium moving between the site and introduced the idea of “potentially recycled uranium for Fernald.” The report discusses the differences between the complex-wide rollup report and the site specific report, pointing out that although the amount of RU dropped from 246,000 MTU from the Site report to 55,419 MTU in the DOE complex-wide rollup, the amount of plutonium only dropped from 217 g to 201 g. The observation was made that “the majority of the constituents are contained in a relatively small mass of uranium.” Examples of these mass balance inconsistencies are listed in Table 17. However, it should be remembered that the mass flow discrepancies do not influence the default assumptions based on TRU to uranium ratios, which were determined by specific analyses of process streams.

Table 17. Examples of uranium mass balance inconsistencies in FMPC receipts.

Source	RU quantity (MTU)	Pu-239 (g)	Np-237 (g)	Tc-99 (g)	Comments
WMCO 1986	606,932	NR	NR	NR	1953–1986
DOE 2000a	246,683.1	217.7	25,742.1	331,998.1	1962–1999 <sup>a</sup> values used in this report
DOE 2000a	55,419	201	18,378	328,981	1962–1999 complex-wide rollup <sup>b</sup>
DOE 2003	17,966	74.3	5,735	71,000	Revision by DOE Office of Security <sup>c</sup>

NR = not reported.

- Includes receipts from both primary and secondary sites as well as an assumption of contamination of existing inventories.
- This complex-wide rollup summary also includes primary and secondary shipments, but does not include contamination of existing inventories from uranium ores processed on site and at uranium mills.
- Includes receipts from primary sites only.

It is true that the DOE documents pertaining to RU in the weapons complex include discrepancies, but these do not preclude the use of process data to define bounding assumptions. The inconsistencies in the amounts of RU in various reports can be attributed to the availability of data at the time that

each report was prepared, the scope of each report, and the degree to which conservative assumptions were used. See Section 4.1.2 for further discussion. The assumption that, beginning in 1961, all uranium at FMPC should be treated as RU is a major contributor to the RU inventory discrepancies. This assumption is favorable to the claimant.

Just as the mass inventories are different in the various reports, the concentration of the RU contaminants is somewhat, but less, different in the various reports. For example, the mass concentration of plutonium in DOE (2000a) is 0.9 ppb. But the complex-wide rollup DOE (2003) defines this value as 3.7 ppb. The mass concentration in a specific batch of uranium could vary from near-zero to values slightly in excess the Hanford guideline value of 10 ppb. However, these shipments from the primary sites in excess of the 10 ppb limit were rare and exception approved.

#### 4.5 SUMMARY AND RECOMMENDED DEFAULT VALUES FOR RU AT FMPC

Significant quantities of RU appeared at FMPC beginning in 1961, but there were no workplace indicators or bioassay monitoring for the contaminants in RU prior to 1986. Consequently, dose reconstruction for exposure to RU at FMPC is based on uranium bioassay results and should include default factors to account for exposure to the RU contaminants. The chemical forms of the RU contaminants are not known. The dose reconstructor should assume the solubility for the RU contaminants that matches the claimant-favorable selection made for the uranium, as recommended in ORAUT-OTIB-0060, *Internal Dose Reconstruction* (ORAUT 2007a). Table 18a lists the recommended mass concentration additions and activity conversions for the period after 1961 and to 1973. The actual contaminant levels were much lower than these values from 1961 to 1973, since their basis is process stream analyses in the time period of the highest activities following 1973. However, these are the defaults used in the dose reconstruction program for several years and will continue to be used for simplicity and conservatism. The defaults for the 1973 time period forward were adjusted, using a lognormal distribution at the 95th percentile.

Table 18a. Recommended RU default values beginning in 1961 and to 1973.

RU contaminant	Mass concentration addition	Activity concentration (Bq/g U)
Pu-239	100 ppb U	232.7
Np-237	3,500 ppb U	92.5
Tc-99	9,000 ppb U	5698
Th (232) <sup>a</sup>	10,000 ppb U	0.04
Ru-103/106	50 $\mu$ Ci/lb	4,075
Zr/Nb-95	15 $\mu$ Ci/lb	1,222
Others (Sr-90)	2 $\mu$ Ci/lb	163

a. Th-228 should be assumed to be at 70% equilibrium with Th-232.

Table 18b. Recommended RU default values beginning in 1973 and to 1986.

RU contaminant	Mass concentration addition	Activity concentration (Bq/g U)
Pu-239	400 ppb U	931
Np-237	11 ppm U	291
Tc-pp	20 ppm U	20,000

The indicated addition for each RU contaminant should be added for each gram of uranium in the urine.

The defaults for the time period of 1973 to 1986 should be those presented in Table 18b above, which represent the time period of processing the waste materials from the Gaseous Diffusion Plants (and

specifically the PGDP), which created a major increase in contaminant levels in all processes at Fernald. The other contaminant defaults should remain the same as in Table 18a.

## 5.0 USE OF DAILY WEIGHTED EXPOSURE REPORTS FOR ESTIMATION OF CHRONIC DAILY THORIUM INTAKE RATE

This report describes the average DWE hazard assessment process used at FMPC and explains how the DWE data can be interpreted and used to calculate inhalation and ingestion intake rates of thorium for use in dose reconstruction.

DWE data are especially important before 1968 when *in vivo* chest counting was introduced to the FMPC site. In addition, the concepts in this report can be used for dose reconstructions associated with raffinate or other waste-stream processing.

Information on where and when various materials were processed is in ORAUT (2004). Thorium was handled in several FMPC plants before 1968 (Morris 2008). Thorium-bearing materials were received, sampled, and occasionally repackaged in Plant 1. Thorium fluoride was produced in Plant 4 and, during the same period, reduced to metal and rolled into ingots in Plant 9. From 1959 through 1963, thorium residues were processed in a sludge furnace in Plant 6. Occasional solvent extraction and metal casting occurred in the Pilot Plant beginning in 1964 and continuing into the 1970s. Table 19 illustrates where and when thorium is known to have been processed before 1968 (Morris 2008). Plants that are not included in Table 19 are not known to have been involved in thorium processing before 1968.

Table 19. Where and when thorium was processed at FMPC before 1968.

Plant	54	55	56	57	58	59	60	61	62	63	64	65	66	67
1	X	X	X	X	X	X	X	X	X	X	X	X	X	X
4	X	X	X	N/A										
6	N/A	N/A	N/A	N/A	N/A	X	X	X	X	X	N/A	N/A	N/A	N/A
8	N/A	X	N/A											
9	X	X	X	N/A										
Pilot	X	X	X	N/A	X	X	X	X						

X = thorium was processed; N/A = thorium is not known to have been processed in that facility at that time.

## 5.1 DAILY WEIGHTED EXPOSURE REPORTS

An average DWE is an assessment of the exposure potential of an individual with a specific job title or job description at a specific facility. The concept of the average DWE was introduced to the AEC complex by the HASL staff (Adams and Strom 2008). The earliest DWE report at FMPC was prepared by the HASL staff (Breslin, Loysen, and Glauberman 1953). The monitoring concept was transferred to the FMPC staff, proceduralized (Wing 1960), and used at FMPC since the beginning of site operations. The procedure required that an annual analysis be prepared for all production plants including the Pilot Plant, Development Machine Shop, Technical Laboratory, and Laundry.

Nonproduction areas were also to be evaluated based on air dust samples from the Boiler and Water Purification Plant, Chemical Warehouse and Factory Stores, Decontamination Building, Mechanical Areas (Mechanical Building, Garage and Firehouse, and Heavy Equipment Building), and

Nonproduction Areas (Service Building, Administration Building, Personnel-Security Building, Health and Safety – Production Building, and Guard House).

In modern practice, the average DWE concept is similar to the time-weighted average (TWA) exposure that is in current use (ACGIH 1996), except the TWA assumes an 8-hour workday while the average DWE uses the actual workday duration. The peer-reviewed paper by Methner, Feng, and Utterback (2001) provides a description of the DWE method and data from FMPC.

DWE reports are summaries of data prepared by the FMPC Industrial Hygiene and Safety staff for use by management. The data in each report are in units of multiples of the MAC, which at FMPC was 70 alpha dpm/m<sup>3</sup> of air until it was changed to 100 alpha dpm/m<sup>3</sup> of air in 1963. The analytical technique applied to air dust samples was gross alpha counting. No isotopic information is available with gross alpha counting, and there are relatively few isotope-specific analyses in the record; therefore, the assumption about the radionuclide being measured was based on the material in process at that time. The reports are remarkably similar from year to year. For example, each report is typically between 30 and 70 pages long and includes the following information:

- Introduction containing a brief summary of the processes at the facility
- Description of the sampling and analysis method
- Summary of Data – Table I (see Figure 6 for an example), containing the average DWE for each job description in the facility, the number of workers employed in that job description, and an average DWE for the entire facility
- Summary of Data – Table II (see Figure 7 for an example), containing the average of the air dust sample concentrations for a specific operation or area
- Discussion, containing a detailed description of processes and controls
- Recommendations, containing general and specific recommendations, often in a form amenable to tracking
- Conclusions
- Appendix, containing Job Evaluation Reports.

DWE reports typically include a statement to the effect that respirators (filtering or air-supplied) were worn by some of the operations, but this was not taken into consideration in calculating DWE data, and it can be assumed that exposures of persons making proper use of prescribed respiratory protection were materially reduced from the calculated values. Another observation in some of the reports is that workers were rotated through jobs with the highest exposure potentials. Other data sources including interviews with former employees (ORAUT 2007b) confirm that use of respiratory protection was typical during tasks with high exposure.

DWE reports are available for some of the FMPC plants for some of the years 1953 to 1971. A useful summary of the FMPC DWE data is available in the SRDB (Author unknown ca. 1968). The available data from the DWE reports for plants and years during which thorium was processed have been transcribed to spreadsheets.<sup>1</sup> In the Pilot Plant during 1967 and 1969, no time-weighting information was recorded, possibly because the processes being characterized were in development and had not yet become routine. The transcribed DWE data represents six plants (1, 2/3, 4, 6, 8, 9), 14 different

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<sup>1</sup> The spreadsheets containing these transcribed data are available in the "AB Document Review" folder.

TABLE I  
WEIGHTED EXPOSURE OF PLANT 9 PERSONNEL

<u>Job Description</u>	<u>No. of Employees</u>	<u>Daily Weighted Exposure X MAC</u>
<u>CHEMICAL</u>		
Wet Area operator - up	3	34.8
Wet Area operator - down	3	215.1
Wet Area helper	3	46.9
Bump operator and helper	3	7.3
<u>REDUCTION</u>		
Reduction charging operator	3	4.11
Reduction charging helper	3	233.71
Jolter operator	3	4.10
Reduction furnace helper	3	4.11
Breakout operator	3	11.9
Derby cleaning operator	3	6.21
De-zincing operator and helper	6	10.60
<u>FURNACE</u>		
Primary welder	6	25.06
Secondary welder	3	8.83
Secondary welder helper	3	685.64
Primary arc furnace operator and helper	6	473.03
Secondary arc furnace operator and helper	6	75.71
<u>MACHINE</u>		
Derby saw operator	3	12.87
Ingot saw operator	3	17.67
Engine lathe operator	3	20.60
Centerless grinder operator and helper	6	10.46
Turret lathe operator #1	3	8.21
Turret lathe operator #2	3	25.41
<u>MISCELLANEOUS</u>		
Residue operator	3	9.66
Chip processing operator	3	4.39
Slug stamper	3	4.23
Quality Control operator	3	4.91
Final inspection	3	4.91
Fork lift operator	3	9.66
Shift foreman	3	34.46
Area foreman	3	9.44
Clerk	3	1.36
Superintendent	1	1.59

Figure 6. Excerpt from a 1955 Plant 9 report (Stefanec 1955b) showing DWE for 119 employees by job description. The average DWE is in units of multiples of the MAC where the MAC is 70 alpha dpm/m<sup>3</sup> of air.

years, 142 job descriptions, 361 operations, and 16,748 air samples. Air sampling duration ranged from 1 minute to 455 minutes, with the average being 63 minutes.

Figure 6 is an excerpt from a 1955 DWE report about FMPC Plant 9 (Stefanec 1955b) during a period when thorium processing was occurring. Figure 7 is an example of a Job Exposure Evaluation card for a Chemical Process Area Wet Area Helper from the same report. The card includes the time of the evaluated work shift. In almost all cases, the time on a card is 510 minutes or 8.5 hours. The

**NATIONAL LEAD OF OHIO**

Job Exposure Evaluation Survey Period \_\_\_\_\_

BUILDING 9 PROCESS AREA Chemical

OPERATOR Wet Area Helper 1 men/shift 3 shifts/day 3 men/day

OPERATION OR OPERATING AREA	TIME PER OPER.	OPER. PER SHIFT	TIME PER SHIFT (T)	NO. OF SAMPLES	CONCENTRATION $\mu\text{g}/\text{m}^3$			T x C
					High	Low	Aver. (C)	
EZ Dumping TWT into dissolving tank			60	3	1088	395	776	46440
EZ Cleaning predryer vents			16	3	1876	1040	1532	24512
EZ Dumping recycle oxide into predryer			15	8	125061	52395	4960	824400
GA Chemical Area, upper deck			300	13	5057	85	2518	778062
GA Smoking area			20	6	190	7	79	1580
GA Washroom			10	4	68	22	54	540
GA Process locker room			20	15	51	5	15	300
GA Street locker room			10	20	25	1	6	60
GA Cafeteria			30	20	33	0	4	120
GA Travel time			20	4	3	0	1.2	24

NLS-NRP-106 (4-1-54) IN LIEU OF NO. 12-N-2

$\Sigma T = 510$   $\Sigma(T \times C) = 1,676,038$

$\frac{\Sigma(T \times C)}{\Sigma(T)} = \frac{3286}{70} \mu\text{g}/\text{m}^3 = 46.9$  MAC

Figure 7. Excerpt from the appendix of a 1955 Plant 9 report (Stefanec 1955b) showing operations associated with a job description and the data used to assess each operation. Collectively this comprises a DWE for the job description.

Plant 9 Job Exposure Evaluations during 1955 were based on data from approximately 490 different air dust samples.

Figure 6 also provides insight into the impact of tasks involving very high airborne dust concentrations on worker exposure. Air dust samples reflecting very high concentrations are often observed and the suspicion is raised that they reflect an out-of-control situation that precludes dose reconstruction. On the contrary, Figure 7 shows that as early as 1955 high air sample data were routinely assessed and preserved in the record. For example, one 15-minute task called "Dumping recycle oxide into pre-dryer" was sampled on eight different occasions using a breathing zone air sampler. The highest

observed value was 125,061 dpm/m<sup>3</sup> and the average was 54,960 dpm/m<sup>3</sup>. This task accounts for 49% of the exposure for this job description. Despite the task's short duration and extremely high airborne dust concentration, its contribution to dose was assessed. A site-wide Dust and Fume Control Committee kept track of, routinely reviewed, and made recommendations about operations with the highest exposure potential (Starkey 1960–1962). The DWE reports also include an arithmetic mean value for the entire facility. For FMPC Plant 9 in 1955 the "average weighted exposure for the plant" is reported as 67.34 MAC.

## 5.2 CORRELATION OF DATA AND URINARY URANIUM EXCRETION RATE

At the 1958 Symposium on Occupational Health Experience and Practices in the Uranium Industry, Heatherton and Huesing (1958) presented a paper describing the correlation of weighted uranium air dust concentration with the urinary excretion rate of uranium at FMPC. This paper includes

information beginning in 1952 and summarized below that provides a validation of the usefulness of DWE data as a predictor of excretion rates and, with the aid of biokinetic modeling, the intake rate. The following excerpt from the paper establishes the operational approach to exposure controls at the beginning of plant operation:

*We have always used urine sample results as an indication of the air contamination problem within the plant or on a job. A high frequency of urine sample results above 0.050 mg/L within a particular job or plant has always required industrial hygiene investigation of the work to determine the cause. Field investigations have often led us to a source of exposure of which we were not aware at the time.*

Thus, frequent urine sampling was combined with comprehensive air monitoring to provide the means of detection and control of uranium intakes.

Figure 8, taken from the Heatherton and Huesing (1958) paper, illustrates the correlation of uranium-in-urine concentration with DWE, which was an analytical tool devised at the start of the programs for increased understanding of the effectiveness of the control mechanics. The Acme-Gridley operator job description is highlighted because of the abundant data available for this job – 120 urine samples were averaged to obtain the data point. Additional data associated with weighted air concentration levels ranging from 25 to 75 µg/m<sup>3</sup> were used to develop a linear model. Data was constrained to this range because fewer air samples were collected for jobs and locations with lower air concentrations and intakes for jobs with highly elevated concentrations were likely to be biased by the mandatory use of respirators. The resulting model predicts an intake rate which would yield a urine concentration of 0.030 mg U/L of urine when the worker is exposed to a DWE of 70 dpm/m<sup>3</sup> (1 MAC).

Beginning in 1958, urine sample reports were computerized and routinely sorted, listing the results by job number. This enabled comparison of urine samples with the DWE data for specific jobs. A urine sample concentration in excess of 0.050 mg/L was seldom found unless the DWE exceeded 1 MAC. This was based on an analysis of urine samples representing workers with negligible exposure who occupied administrative areas, workers who occupied production areas where the DWE was less than 1 MAC, and workers who occupied production areas where the DWE was greater than 1 MAC. As expected, workers occupying areas where the DWE was greater than 1 MAC had the highest proportion of elevated uranium-in-urine concentrations. Conversely, workers in administrative areas where DWE values approach zero had very low uranium-in-urine concentrations. Workers occupying production areas where the DWE was elevated, but less than 1 MAC had urine concentration values tending between the two extremes. Table 20 presents the results.

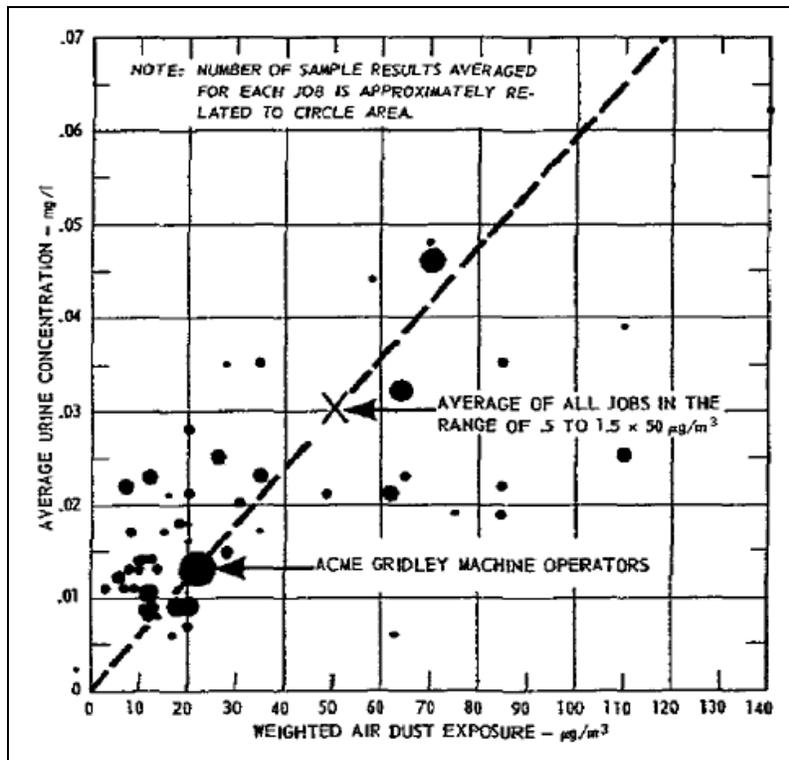


Figure 8. Uranium-in-urine excretion as a function of DWE (Heatherton and Huesing 1958).

Table 20. Comparison of uranium-in-urine concentration with nominal DWE.

Area	Number of urine samples	Nominal DWE	Urine concentration, mg/L (%)			
			<0.025	0.026 to 0.050	0.051 to 0.10	>0.10
Admin	267	<< 1 MAC	94	4.9	0.7	0.0
Production	762	< 1 MAC	84	14	1.6	0.0
Production	318	>1 MAC	31	51	16	2.5

From this information the authors deduced a rule of thumb: If a worker's uranium-in-urine concentration is usually less than 0.025 mg/L and never in excess of 0.050 mg/L, the DWE does not exceed 1 MAC.

The effect of respiratory protection on uranium-in-urine concentration was also reported. A specially planned job of "desludging" a salt bath furnace resulted in a DWE of 235 MAC on day 1, 5 MAC on day 2, and 2 MAC on day 3. Urine results for the 50 men wearing air-fed hoods were averaged. Before the job, the average uranium-in-urine concentration was 0.018 mg/L. At the end of day 3 the average concentration had increased only marginally to 0.038 mg/L. Fourteen of the men participating in this job wore dust-type respirators. For this group, the DWE during day 1 was 28 MAC. Days 2 and 3 were the same as for the air-fed hood group. The workers wearing dust-type respirators began the job with an average concentration of 0.018 mg/L. At the end of day 3 the average concentration was 0.035 mg/L. Collectively, these data show the respiratory protection program provided adequate protection while performing a job with high exposure potential.

This early study by the FMPC Health and Safety program (Heatherton and Huesing 1958) demonstrates the usefulness and reliability of using DWE data to bound intakes of contaminants in the workplace, when used as an integral element in the intake control program.

### 5.3 UNCERTAINTY

In a peer-reviewed publication, Davis and Strom (2008) evaluated DWE data and the supporting air sample data collected by the HASL staff at five Atomic Weapon Employer (AWE) sites: Electrometallurgical Co.; American Machine and Foundry Co.; Middlesex Sampling Plant; Horizons, Inc.; and Lake Ontario Ordinance Works. The data represented uranium, radium, and thorium operations performed from 1948 through 1954. The data from Horizons, Inc. was specific to thorium operations. The sampling methods and data analyses used by the HASL staff were the same as those used at FMPC. The engineered controls in these AWE facilities were similar to, but likely inferior to, those employed at FMPC, because FMPC was purpose-built in light of experience at earlier operating facilities.

Davis and Strom (2008) determined that the DWE values were usable as inputs in dose reconstructions conducted in accordance with EEOICPA, but that the DWE values were limited by their lack of numerical uncertainties. The uncertainty associated with DWE values is a function of blunders (in other words, data entry or math errors), variability, and measurement uncertainty. Measurement uncertainty was judged to be trivial in comparison with the other factors. Variability in air sampling data can be the result of many factors, including representativeness of the sample, ventilation changes, sampler placement, process variability, and aerosol particle size. Using a Monte Carlo analysis, Davis and Strom retrospectively assessed the uncertainty and variability in the DWE values for 63 job titles, thereby eliminating the limitation of usefulness of the DWE data.

Based on their analysis, Davis and Strom (2008) state it is reasonable to conclude that the upper 95th-percentile value of the geometric standard deviation (GSD) distribution is about 4 and the upper 99th-percentile value is between 7 and 8. This lends support to their generalization of assuming a GSD of 5 when a DWE concentration value is available but there is no information on uncertainty.

For instances in which the TWA of the air sample data are not available, Davis and Strom (2008) determined that the upper 95th percentile of the site-wide air concentration data will almost always be favorable to the claimant when making compensation decisions.

### 5.4 SELECTING A DAILY WEIGHTED EXPOSURE OR AIR SAMPLE CONCENTRATION VALUE FOR INTAKE RATE CALCULATION

Due to variations in job assignments, it is unlikely that unambiguous exposure scenarios can be defined for most workers. In addition, fugitive dust emissions have not been quantified except as the general air sample indicator. To ensure thorium intake potential is not underestimated, the highest DWE value associated with a specific job title or job description in the FMPC plant where thorium was handled for a specific year should be assigned to every worker in that facility. A GSD of 5 should be assumed.

If TWA data are not available, the upper 95th percentile of the air sampling data for that facility can be used instead.

If neither DWE data nor air sampling data are available for a specific year, or if they are judged to be inadequate or incomplete, DWE from the same facility for an adjacent period can be assigned. In these cases, a high value from an adjacent year should be selected. A GSD of 5 should be applied.

Table 21 summarizes the recommended DWE values for years and facilities in which thorium work is known to have occurred. These values represent the job description in each facility with the highest DWE value for that year.

Table 21. DWE values, in multiples of the MAC, recommended for use in estimation of thorium intake rates.

Plant	1954	1955	1956	1957	1958	1959	1960	1961	1962	1963	1964	1965	1966	1967
1	(a)	16.4	4.3	1.5	1.0	2.1	0.5	0.6	1.1	0.6	0.7	0.6	1.0	1.2
4	4.5	3.2	0.9	N/A										
6	N/A	N/A	N/A	N/A	N/A	0.8	4.3	1.6	2.8	17	N/A	N/A	N/A	N/A
8	N/A	7.1	N/A											
9	(b)	686	(b)	N/A										
Pilot Plant	5.9	(c)	2	N/A	(d)	(d)	(d)	77						

N/A = not applicable.

- Data are not available. Use the value for Plant 1, 1955.
- Data are not available. Use the value for Plant 9, 1955.
- Data are not available. Use the value for Pilot Plant, 1954.
- DWE data are not available. The value for Pilot Plant, 1967, is based on the 95th-percentile value of 18 air samples taken during thorium operations. Use this value for Pilot Plant, 1964 through 1966.

## 5.5 CALCULATION OF CHRONIC DAILY INTAKE RATES

The approach for intake rate calculations is based on the method defined in Battelle (2006).

Two modes of intake are considered for routine dose reconstruction: inhalation and ingestion. The methods presented here require an appropriate DWE parameter to be selected by the dose reconstructor. That value might be the average DWE for a specific job description. As an alternative, the DWE value might be selected from a lognormal distribution fitted to average DWE data for the facility or site.

For facilities in which thorium intake might have occurred, all activity should be assumed to be thorium in equilibrium between  $^{232}\text{Th}$  and  $^{228}\text{Th}$ . For example, if the thorium intake rate is 2 Bq/d, the  $^{232}\text{Th}$  intake rate would be 1 Bq/d and the  $^{228}\text{Th}$  intake rate would be 1 Bq/d.

### 5.5.1 Calculation of Inhalation Rate

The chronic daily inhalation intake rate *Chronic INH* for a worker can be calculated based on the appropriate DWE parameter, an assumed breathing rate *BR* and the hours worked in the job description per day  $T_d$  as shown in Equation 15.

$$\text{Chronic INH} \left( \frac{\text{Bq}}{\text{d}} \right) = \text{DWE} \left( \frac{\sum (T_d * C)}{\sum (T_d)} \right) \times \text{MAC}_{Op} \left( \frac{\text{dpm}}{\text{m}^3} \right) \times \text{BR} \left( \frac{\text{m}^3}{\text{h}} \right) \times T_d \left( \frac{\text{hr}}{\text{d}} \right) \times \frac{1}{60} \left( \frac{\text{Bq}}{\text{dpm}} \right) \times \frac{5}{7} \left( \frac{\text{d}}{\text{d}} \right) \quad (15)$$

where

- DWE* = DWE in multiples of the operational  $\text{MAC}_{Op}$ . A GSD of 5 is carried along with this value.
- $\text{MAC}_{Op}$  = MAC in  $\text{dpm}/\text{m}^3$ , either 70 alpha  $\text{dpm}/\text{m}^3$  of air before 1963 or 100 alpha  $\text{dpm}/\text{m}^3$  of air beginning in 1963.
- $T_d$  = hours worked per day, typically 8.5 at FMPC
- BR* = assumed breathing rate per hour
- $1/60$  = conversion to Bq from dpm.

This daily intake rate would be usable for dose reconstruction as a chronic inhalation intake rate if the worker were exposed to this concentration every day of the calendar year, but no employee was exposed every day of the year. To correct for the time away from the job due to weekends, the daily intake rate is multiplied by a factor of 5/7. This correction normalizes the calculated value and results

in an average chronic daily inhalation intake rate which is usable as input into the Integrated Modules for Bioassay Analysis (IMBA) program.

Assuming a typical breathing rate used in dose reconstruction of 1.2 m<sup>3</sup>/hr and a typical 8.5-hour workday, Equation 15 simplifies to Equation 16.

$$\text{ChronicINH} \left( \frac{\text{Bq}}{\text{d}} \right) = \text{DWE} \times \text{MAC}_{Op} \times (0.121) \quad (16)$$

### 5.5.2 Calculation of Daily Ingestion Intake Rate

The chronic daily ingestion rate (*Chronic ING*) for a worker can be calculated based on the appropriate DWE parameter, the hours worked in the job description per day ( $T_d$ ), and factors defined by NIOSH. NIOSH (2004) describes three modes of ingestion intake. Mode 1 is associated with swallowed particles that were initially inhaled and deposited in the respiratory tract. This mode is incorporated in the International Commission on Radiological Protections (ICRP) biokinetic models and is not addressed in this paper. Mode 2 is associated with consumption of radioactive material that might fall out in a drinking cup where airborne radioactive material is present. It assumes the cup is receiving fallout 8 hr/d, but that value can be adjusted to reflect a workday of any length [1]. Mode 3 is associated with pica, in which transfer of contamination from hand to mouth occurs. In addition, Mode 3 is based on the airborne radioactive material concentration, but it is unaffected by the length of the workday because airborne dust is assumed to be settling on surfaces 24 hr/d. Because Modes 2 and 3 are based on airborne radioactive material concentration, they can be combined.

Mode 2 assumes that the airborne radioactivity settles in a cup of a specified diameter with an assumed settling velocity over the course of an 8-hour day. In that case, the ingested activity is 0.0985 m<sup>3</sup>/d times the activity concentration per cubic meter. By dividing by 8, the workday length can become a variable,  $T_d$ , in hours per workday. This makes the equation versatile to accommodate the typical 8.5-hour workday at FMPC. One other change is to adjust for time away from the job due to weekends, creating a chronic ingestion intake rate suitable as input to IMBA. This is done by including a factor of 5/7 in the equation. The resulting formulation for chronic daily ingestion rate from Mode 2 sources is Equation 17.

$$\text{Mode2} \left( \frac{\text{Bq}}{\text{d}} \right) = \frac{\text{DWE} \left( \frac{\sum (T_d * C)}{\sum (T_d)} \right) \times \text{MAC}_{Op} \left( \frac{\text{dpm}}{\text{m}^3} \right) \times T_d \left( \frac{\text{hr}}{\text{d}} \right) \times \frac{1}{60} \left( \frac{\text{Bq}}{\text{dpm}} \right) \times 0.0985 \left( \frac{\text{m}^3}{\text{d}} \right) \times \frac{5}{7} \left( \frac{\text{d}}{\text{d}} \right)}{8 \left( \frac{\text{hr}}{\text{d}} \right)} \quad (17)$$

which simplifies to Equation 18.

$$\text{Mode2} \left( \frac{\text{Bq}}{\text{d}} \right) = \text{DWE} \times \text{MAC}_{Op} \times T_d \times (1.47 \times 10^{-4}) \quad (18)$$

where

- $\text{DWE}$  = DWE in multiples of the operational  $\text{MAC}_{Op}$ .
- $\text{MAC}_{Op}$  = MAC in dpm/m<sup>3</sup>, either 70 alpha dpm/m<sup>3</sup> of air before 1963 or 100 alpha dpm/m<sup>3</sup> of air beginning in 1963.
- $T_d$  = hours worked per day, typically 8.5 at FMPC

The Mode 3 daily ingestion intake rate resulting from 1 workday is defined in NIOSH (2004) as the airborne activity concentration in activity units per cubic meter times 0.1 m<sup>3</sup>/d. Just as for Mode 2, this is adjusted to accommodate a 5-day workweek as shown in Equation 19

$$\begin{aligned}
 Mode3\left(\frac{Bq}{d}\right) &= DWE\left(\frac{\sum(T_d * C)}{\sum(T_d)}\right) \times MAC_{Op}\left(\frac{dpm}{m^3}\right) \times \frac{1}{60}\left(\frac{Bq}{dpm}\right) \times \frac{5}{7}\left(\frac{d}{d}\right) \times 0.1\left(\frac{m^3}{d}\right) \\
 &= DWE \times MAC_{Op} \times (1.19 \times 10^{-3})
 \end{aligned}
 \tag{19}$$

Equations 18 and 19 can be combined to represent the chronic ingestion intake rate as shown in Equation 20.

$$Chronic\ ING = DWE \times MAC_{Op} \times \left[ \left( T_d \times (1.47 \times 10^{-4}) \right) + (1.19 \times 10^{-3}) \right]
 \tag{20}$$

Assuming an 8.5-hour workday, which is typical at FMPC, this simplifies to Equation 21.

$$Chronic\ ING\ \left(\frac{Bq}{d}\right) = DWE \times MAC_{Op} \times (2.44 \times 10^{-3})
 \tag{21}$$

where

$DWE$  = DWE in multiples of the operational  $MAC_{Op}$ .  
 $MAC_{Op}$  = MAC in dpm/m<sup>3</sup>, either 70 alpha dpm/m<sup>3</sup> of air before 1963 or 100 alpha dpm/m<sup>3</sup> of air beginning in 1963.

## 6.0 ATTRIBUTIONS AND ANNOTATIONS

Where appropriate in this document, bracketed callouts have been inserted to indicate information, conclusions, and recommendations provided to assist in the process of worker dose reconstruction. These callouts are listed here in the Attributions and Annotations section, with information to identify the source and justification for each associated item. Conventional references, which are provided in the next section of this document, link data, quotations, and other information to documents available for review on the Project's Site Research Database (SRDB).

- [1] Morris, Robert L. MH Chew & Associates, Inc. Principal Health Physicist. February 2008. This equation implicitly assumes the drinking cup is collocated with the worker at all times. It is reasonable to expect that a drinking cup would seldom be in areas where highest airborne concentrations occurred and would more likely be in a break area where exposures were lower. This assumption is, therefore, favorable to the claimant.

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## ATTACHMENT A CONTAIN CODE CALCULATIONS AND RESULTS

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Modeling the venting of radon gas from the K-65 Silos was performed by using carbon dioxide as a surrogate gas, because it is heavier than air and is in the CONTAIN computer program material library, while radon is not. A problem was set up with all cells containing the same atmosphere. The pressure reduction for lateral flow was calculated for a wind speed of 6.5 mph. The environment was assigned the corresponding reduced initial pressure of 99,420.30 Pa (see Attachment E). The wind had little influence on the problem. The internal cells had an initial pressure of 99,422.05 Pa. The pressure difference between the silo cells and the environment was eliminated rapidly as little air movement was required to achieve equilibrium. All of the internal cells quickly reached the outside environmental pressure. Three engineered vents were used to connect the unloading manhole, the gooseneck, and the instrument manhole to the environment. The turbulence coefficient used by the code was not optimized for the problem. The flow area connecting all internal cells was equal to the cross-section of the silo. The flow velocity for the engineered vents was too small for turbulence to be important. The principal driver for the problem was heat input to the headspace.

A heat source of 2,980.77 W was applied to cell 9 for 3,600 seconds (1 hour). The temperature in the headspace rose linearly with time from 293 K to 305.42 K. After the heat was turned off at 3,600 seconds the higher temperature persisted, but the flow rates to the environment fluctuated with outflow from the gooseneck, which was partially offset by inflow at the manholes. The calculation was run to a problem time of 4 hours.

For the first problem considered, all the silo cells had an initial atmosphere with 5% CO<sub>2</sub> by molar fraction. Initially, the flow from the unloading manhole was the largest because of the short path to the exterior and the instrument manhole had less area for the flow. The gooseneck path was much longer (1.524 m) and had noticeable flow inertia.

After slightly more than 4 minutes, the inertia was overcome and the gooseneck flow became the dominant contributor. Figure A-1 shows the vent flow velocities versus time. After a short time the velocities for all three vents are essentially the same. Table A-1 shows the vent velocities versus time.

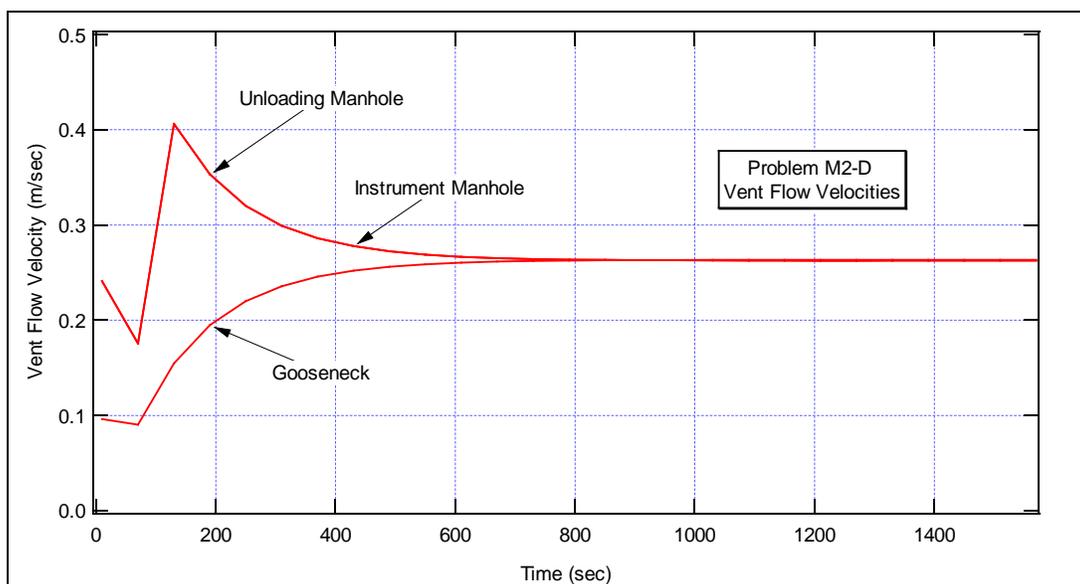


Figure A-1. Vent flow velocities versus time.

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Table A-1. Vent flow velocities versus time.  $V_1$  = unloading manhole,  $V_2$  = gooseneck,  $V_3$  = instrument manhole.

Time (s)	$V_1$ (m/s)	$V_2$ (m/s)	$V_3$ (m/s)
10	0.241058	0.096293	0.241061
70	0.175535	0.0904041	0.175538
130	0.406227	0.154696	0.406227
190	0.353121	0.194984	0.353121
250	0.320091	0.21999	0.320091
310	0.299227	0.235722	0.299227
370	0.286029	0.24571	0.286029
430	0.277905	0.252105	0.277905
490	0.272303	0.256172	0.272303
550	0.268871	0.258792	0.268871
610	0.266613	0.26047	0.266613
670	0.265165	0.261547	0.265165
730	0.264231	0.262239	0.264231
790	0.263559	0.262676	0.263559
850	0.263435	0.262977	0.263435
910	0.263014	0.263145	0.263014
970	0.262914	0.263261	0.262914
1,030	0.262823	0.263333	0.262823
1,090	0.262539	0.26336	0.262539
1,150	0.26258	0.263391	0.26258
1,210	0.262457	0.263398	0.262457
1,270	0.26248	0.263407	0.26248
1,330	0.262598	0.263417	0.262598
1,390	0.262578	0.263414	0.262578
1,450	0.26257	0.26341	0.26257
1,510	0.262575	0.263406	0.262575
1,570	0.262631	0.263404	0.262631

Figure A-2 shows the flow rates for the three vents. The total flow to the environment promptly reached a value of 17.88 cfm and remained steady thereafter. Table A-2 shows the volumetric flows from the engineered vents versus time.

The CO<sub>2</sub> mass in cell 10 (environment) was recorded versus time. Figure A-3 shows and Table A-3 lists the mass of CO<sub>2</sub> vented to the environment for this problem.

Flow rates were influenced somewhat by the presence of the CO<sub>2</sub> compared with plain air atmospheres.

**ATTACHMENT A**  
**CONTAIN CODE CALCULATIONS AND RESULTS**  
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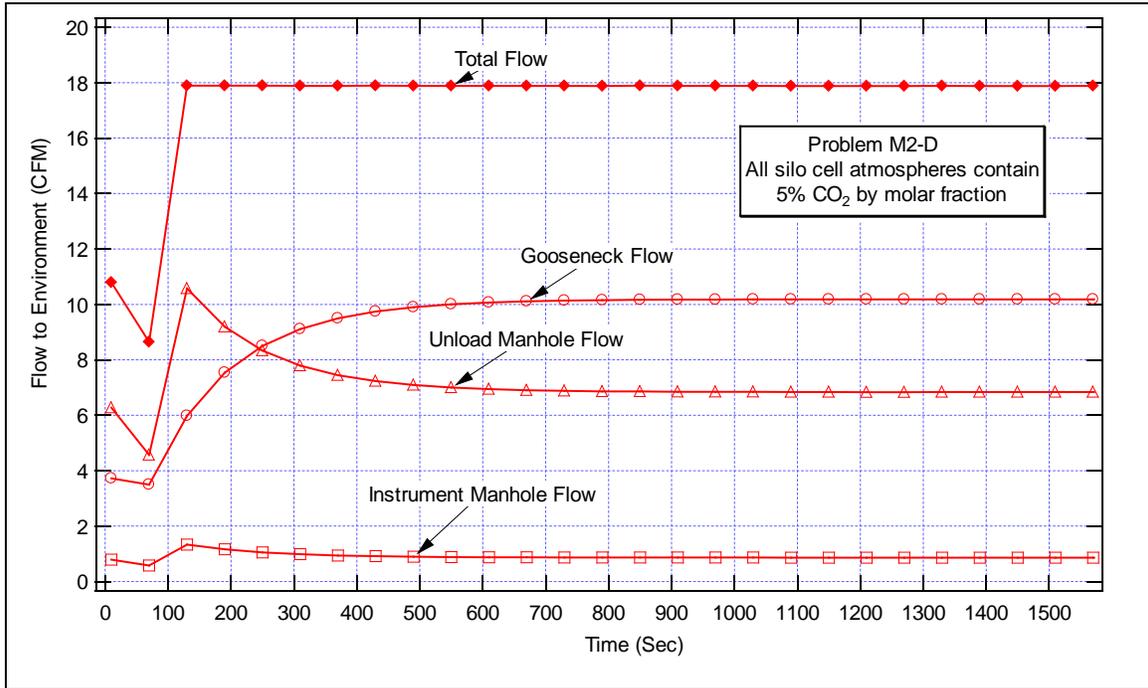


Figure A-2. Flows to the environment from the engineered vents.

Table A-2. Flow from the engineered vents versus time for Problem M2-D. Flow<sub>1</sub> = unloading manhole, Flow<sub>2</sub> = gooseneck, Flow<sub>3</sub> = instrument manhole.

Time (s)	Flow <sub>1</sub> (cfm)	Flow <sub>2</sub> (cfm)	Flow <sub>3</sub> (cfm)	Total (cfm)
10	6.2764	3.7218	0.79263	10.79083
70	4.5704	3.4942	0.57718	8.64178
130	10.5769	5.9791	1.3357	17.8917
190	9.1941	7.5362	1.1611	17.8914
250	8.3341	8.5027	1.0525	17.8893
310	7.7909	9.1108	0.98388	17.88558
370	7.4473	9.4968	0.94049	17.88459
430	7.2358	9.744	0.91377	17.89357
490	7.0899	9.9012	0.89535	17.88645
550	7.0005	10.0024	0.88407	17.88697
610	6.9417	10.0673	0.87664	17.88564
670	6.904	10.1089	0.87188	17.88478
730	6.8797	10.1357	0.86881	17.88421
790	6.8622	10.1526	0.8667	17.8815
850	6.859	10.1642	0.86619	17.88939
910	6.848	10.1707	0.86481	17.88351
970	6.8454	10.1752	0.86448	17.88508
1,030	6.8431	10.1779	0.86418	17.88515
1,090	6.8357	10.179	0.86325	17.87795
1,150	6.8367	10.1802	0.86338	17.88028
1,210	6.8335	10.1805	0.86298	17.87698

**ATTACHMENT A**  
**CONTAIN CODE CALCULATIONS AND RESULTS**  
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Time (s)	Flow <sub>1</sub> (cfm)	Flow <sub>2</sub> (cfm)	Flow <sub>3</sub> (cfm)	Total (cfm)
1,270	6.8341	10.1808	0.86305	17.87795
1,330	6.8372	10.1812	0.86344	17.88184
1,390	6.8367	10.1811	0.86338	17.88118
1,450	6.8365	10.1809	0.86335	17.88075
1,510	6.8366	10.1808	0.86337	17.88077
1,570	6.8381	10.1807	0.86355	17.88235

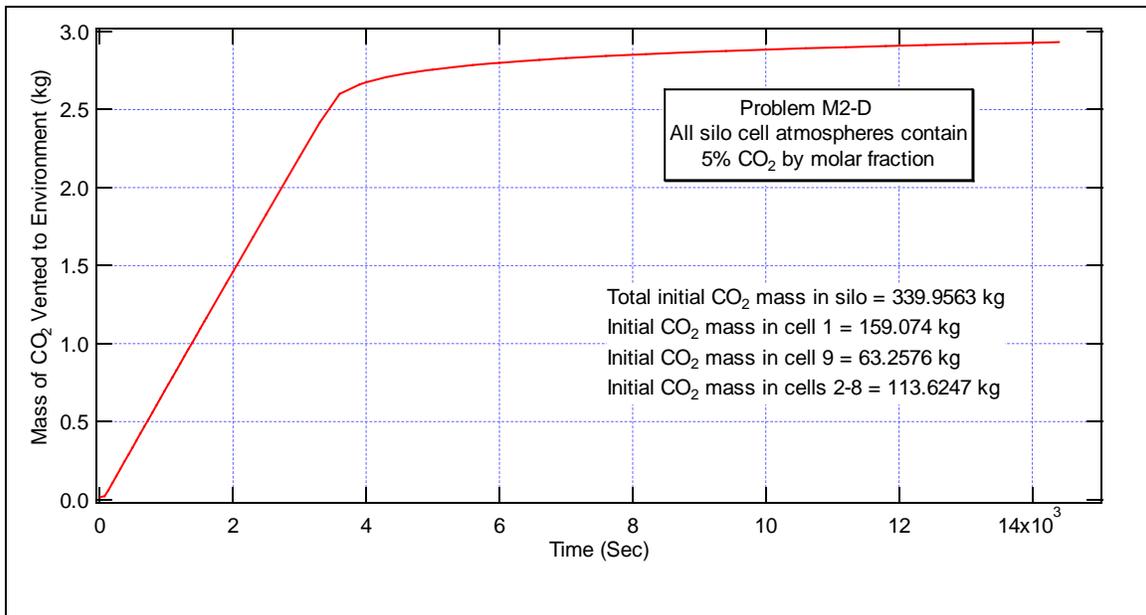


Figure A-3. Mass of CO<sub>2</sub> vented to the environment versus time.

Table A-3. Mass of CO<sub>2</sub> vented to the environment versus time for Problem M2-D.

Time (s)	CO <sub>2</sub> in Cell 10 (kg)	Time (s)	CO <sub>2</sub> in Cell 10 (kg)
10	0.016170	3,900	2.66138
70	0.021368	4,000	2.67535
130	0.0606967	4,300	2.70795
190	0.106139	4,600	2.73212
250	0.151547	4,900	2.75134
310	0.196920	5,000	2.75696
370	0.242260	5,300	2.77205
430	0.287565	5,600	2.78505
490	0.332836	5,900	2.79647
550	0.378073	6,000	2.79999
610	0.423276	6,300	2.80982
670	0.468445	6,600	2.81871
730	0.513580	6,900	2.82683
790	0.558682	7,000	2.82939
850	0.603749	7,600	2.84343
910	0.648783	8,200	2.85565
970	0.693783	8,800	2.86645
1,030	0.738749	9,400	2.87614

## ATTACHMENT A CONTAIN CODE CALCULATIONS AND RESULTS

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Time (s)	CO <sub>2</sub> in Cell 10 (kg)	Time (s)	CO <sub>2</sub> in Cell 10 (kg)
1,090	0.783682	10,000	2.88492
1,150	0.828581	10,600	2.89295
1,210	0.873446	11,200	2.90035
1,270	0.918278	11,800	2.90721
1,330	0.963076	12,400	2.91360
1,390	1.00784	13,000	2.91958
1,450	1.05257	13,600	2.92520
1,510	1.09727	14,200	2.93050
1,570	1.14193	14,400	2.93221
1,600	1.16425		
1,900	1.38699		
2,000	1.46105		
2,300	1.68269		
2,500	1.82998		
2,800	2.05024		
3,000	2.19663		
3,300	2.41553		
3,600	2.59989		

The results were divided by the total initial CO<sub>2</sub> mass in the problem to give the fraction that was vented to the outside. Figure A-4 shows the percent of the CO<sub>2</sub> vented to the environment. The resulting fraction that was vented at 14,400 seconds (4 hours) was 0.8728%.

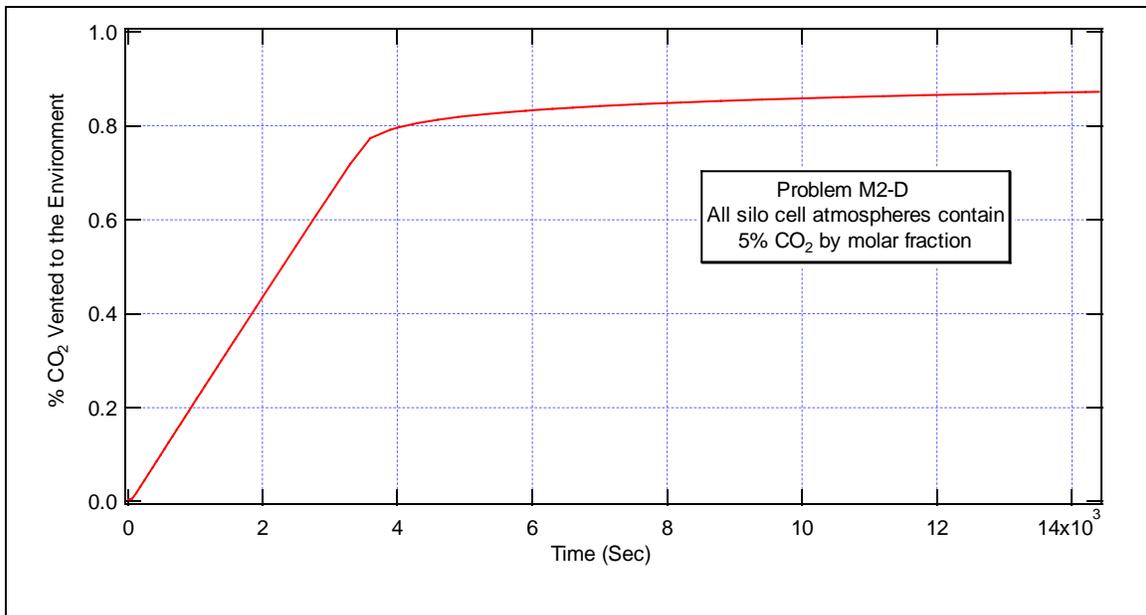


Figure A-4. Percent of the CO<sub>2</sub> vented to the environment as a function of time.

The calculation was repeated with 10%, 15%, and 20% CO<sub>2</sub> by molar fraction. Table A-4 shows the atmospheric molar fractions for each case. The fraction of the CO<sub>2</sub> vented changed only slightly, decreasing as the molar fraction increased. Table A-5 shows the final results for each case. The fraction vented is not sensitive to the molar fraction of CO<sub>2</sub> used; it decreases slightly as the molar fraction is increased.

**ATTACHMENT A**  
**CONTAIN CODE CALCULATIONS AND RESULTS**

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Table A-4. Atmospheric molar fractions.

Gas	O <sub>2</sub>	N <sub>2</sub>	Ar	CO <sub>2</sub>
5% CO <sub>2</sub>	0.199025	0.741	0.009975	0.05
10% CO <sub>2</sub>	0.18855	0.702	0.00945	0.1
15% CO <sub>2</sub>	0.178075	0.663	0.008925	0.15
20% CO <sub>2</sub>	0.1676	0.624	0.0084	0.2

Table A-5. CO<sub>2</sub> vented versus molar fraction CO<sub>2</sub>.

<b>Problem M2-D</b>		<b>5% CO<sub>2</sub></b>
<b>Time (s)</b>	<b>0.000</b>	<b>14,400</b>
<b>Cell #</b>	<b>CO<sub>2</sub> Mass (kg)</b>	<b>CO<sub>2</sub> Mass (kg)</b>
1	159.074	159.072
2	16.2321	16.2319
3	16.2321	16.2319
4	16.2321	16.2319
5	16.2321	16.2319
6	16.2321	16.2319
7	16.2321	16.2319
8	16.2321	16.2318
9	63.2576	60.3289
10	0	2.93221
Total	335.9563	335.95631
<b>% CO<sub>2</sub> vented =</b>	<b>0.8728</b>	

<b>Problem M2-E</b>		<b>10% CO<sub>2</sub></b>
<b>Time (s)</b>	<b>0</b>	<b>14,400</b>
<b>Cell #</b>	<b>CO<sub>2</sub> Mass (kg)</b>	<b>CO<sub>2</sub> Mass (kg)</b>
1	318.149	318.145
2	32.4642	32.4638
3	32.4642	32.4638
4	32.4642	32.4638
5	32.4642	32.4638
6	32.4642	32.4638
7	32.4642	32.4638
8	32.4642	32.4636
9	126.515	120.718
10	0	5.80438
Total	671.9134	671.91378
<b>% CO<sub>2</sub> vented =</b>	<b>0.8639</b>	

**ATTACHMENT A  
CONTAIN CODE CALCULATIONS AND RESULTS**

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<b>Problem M2-F</b>		<b>15% CO<sub>2</sub></b>
<b>Time (s)</b>	<b>0</b>	<b>14,400</b>
<b>Cell #</b>	<b>CO2 Mass (kg)</b>	<b>CO2 Mass (kg)</b>
1	477.223	477.217
2	48.6963	48.6957
3	48.6963	48.6957
4	48.6963	48.6957
5	48.6963	48.6957
6	48.6963	48.6957
7	48.6963	48.6957
8	48.6963	48.6953
9	189.773	181.165
10	0	8.61876
Total	1007.8701	1007.87026
<b>% CO<sub>2</sub> vented =</b>	<b>0.8551</b>	

<b>Problem M2-G</b>		<b>20% CO<sub>2</sub></b>
<b>Time (s)</b>	<b>0</b>	<b>14,400</b>
<b>Cell #</b>	<b>CO2 Mass (kg)</b>	<b>CO2 Mass (kg)</b>
1	636.298	636.29
2	64.9284	64.9276
3	64.9284	64.9276
4	64.9284	64.9276
5	64.9284	64.9276
6	64.9284	64.9276
7	64.9284	64.9276
8	64.9284	64.9266
9	253.03	241.668
10	0	11.3774
Total	1343.8268	1343.8276
<b>% CO<sub>2</sub> vented =</b>	<b>0.8466</b>	

**ATTACHMENT B**  
**GAS DATA APPLIED IN CONTAIN MODEL**

**Gray (1963), pp. 4-103, -108, -114,**  
**CO<sub>2</sub>**

**Density at 300 K, 1 atmosphere**

$$\rho_0 = 1.977 \times 10^{-3} \text{ g / cm}^3$$

$$\frac{\rho}{\rho_0} = 0.9088 \text{ @ 300 K, 1 atm}$$

$$\rho = 1.796697 \times 10^{-3} \text{ g / cm}^3$$

**Specific Heat  $C_p$**

$$C_p/R = 4.513 \text{ @ 300K, 1 atm Reference: 0.188921 J/gK}$$

$$C_p = 0.852600 \text{ J/gK}$$

**Air**

$$\rho_0 = 1.29304 \times 10^{-3} \text{ g / cm}^3$$

$$\frac{\rho}{\rho_0} = 0.9102 \text{ @ 300 K, 1 atm}$$

$$\rho = 1.1769 \times 10^{-3} \text{ g / cm}^3$$

**Specific Heat  $C_p$**

$$C_p/R = 3.5059 \text{ @ 300K, 1 atm Reference: 0.287041 J/gK}$$

$$C_p = 1.006337 \text{ J/gK}$$

### ATTACHMENT C WIND MODELING IN THE CONTAIN MODEL

The CONTAIN model includes wind pressure as another driving force for transporting the radon (CO<sub>2</sub>) to the environment.

Wind pressure effects are evaluated using the American Society of Heating, Refrigeration, and Air-Conditioning Engineers methodology and are applied to the model environment cells. The building wind pressure is calculated from the dynamic head equation written in the form:

$$\Delta P_w = 0.0129 c_w \rho \frac{v_w^2}{2}$$

where:

$\Delta P_w$  = Localized air pressure change due to wind (inches water)

$c_w$  = Wind pressure coefficient (dimensionless)

$v_w$  = Wind speed (mph)

$\rho$  = Air density (lb/ft<sup>3</sup>)

The wind coefficient  $c_w$  varies with wind direction in relation to building surfaces. With wind impinging normally on a wall,  $c_w = +0.7$ . Wind parallel to a wall produces a wind coefficient of  $-0.35$  and on the downwind side of the building a coefficient of  $-0.4$  is used. With these wind coefficients, the pressure difference across the facility was calculated and analysis was performed for each wind direction.

## ATTACHMENT D VENT GEOMETRY CALCULATIONS

### Unloading Manhole

Path length = 4" = 0.1016 m

$$A_2 = \pi \left[ \frac{12.25 \text{ in}}{12 \text{ in / ft}} (0.3048 \text{ m / ft}) \right]^2 = 0.3041512 \text{ m}^2$$

$$A_1 = \pi \left[ \frac{12.00 \text{ in}}{12 \text{ in / ft}} (0.3048 \text{ m / ft}) \right]^2 = 0.2918635 \text{ m}^2$$

$$\text{Gap Area} = A_2 - A_1 = 1.22877 \times 10^{-2} \text{ m}^2 \quad A / L = 0.120942 \text{ m}$$

### Gooseneck

Path length = 5 ft = 1.524 m

Diameter = 6 in., Radius = 3 in.

$$A = (0.25 \text{ ft} \times 0.3048 \text{ m / ft})^2 = 1.8241 \times 10^{-2} \text{ m}^2 \quad A / L = 0.011969 \text{ m}$$

### Instrument manhole

Path length = 4 in. = 0.1016 m

$$A_2 = \pi \left[ \frac{3.125 \text{ in}}{12 \text{ in / ft}} (0.3048 \text{ m / ft}) \right]^2 = 1.979326 \times 10^{-2} \text{ m}^2$$

$$A_1 = \pi \left[ \frac{3.00 \text{ in}}{12 \text{ in / ft}} (0.3048 \text{ m / ft}) \right]^2 = 1.8241468 \times 10^{-2} \text{ m}^2$$

$$\text{Gap Area} = A_2 - A_1 = 1.551792 \times 10^{-3} \text{ m}^2 \quad A / L = 0.0152735 \text{ m}$$

**ATTACHMENT E  
PROBLEM INPUT DECK**

Page 1 of 5

&& Model #M2-D for K-65 Silo Simulation

&&

&&

control

ncells = 10

ntitl = 11

ntzone = 10

numtbg = 2

maxtbg = 50

nengv = 3

nmtrac = 1

eoi

&&

material

compound n2 o2 ar conc h2ov ss co2

trackmat co2

eoi

&& -----Title-----

title

K-65 Silo Airflow Simulation.

The domed headspace is simulated by a cylinder with equal volume.

Cell 1 contains the debris - assumed to have 0.7porosity. The volume of the cell counts only the portion available to gas.

The connections of cell 9 (headspace) to the outside environment are by three engineered vents: one for the Unloading Manhole, one for the Gooseneck, and one for the Instrument Manhole. Cells 2 through 8 are for observing gradients in the gas. All internal cells contain air with 0.05 molar fraction of CO<sub>2</sub>. CO<sub>2</sub> is tracked in the problem. The environmental cell contains only air.

&&

&&

&& -----Edit Times-----

times 30000.0 0.0

0.1 5. 10.0

1.0 20. 1600.0

1.0 100. 2000.0

1.0 100. 2500.0

1.0 100. 3000.0

1.0 100. 4000.0

1.0 100. 5000.0

1.0 100. 6000.0

2.0 100. 7000.0

4.0 200. 14400.0

&& -----Specify Thermal Reactor---

Thermal

&& -----Output frequency-----

**ATTACHMENT E  
PROBLEM INPUT DECK**

Page 2 of 5

shortedt = 3

&amp;&amp; ---Long edit output variables desired ----

longedt = 3

&amp;&amp; -----Flow Data-----

flows

implicit

perror = 0.1

&amp;&amp; -----Flow paths -----

&amp;&amp; Turbulent flow coefficients (CFC) for closed hatches

&amp;&amp; were calculated using the methodology found in

&amp;&amp; CALC-906-NA-000013 by Robert Colwell. For open paths

&amp;&amp; a value of CFC = 0.01 was assigned since turbulence for

&amp;&amp; flows through large openings should be minor.

&amp;&amp; -----Internal Layers of the Silo-----

area(1,2) = 466.98 &amp;&amp; (silo cross section) bottom layer to second layer

avl(1,2) = 1556.6 &amp;&amp; Assumes 0.3m path length

cfc(1,2) = 0.01 &amp;&amp; Arbitrary choice for an open path

topen(1,2) = 0.0

area(2,3) = 466.98 &amp;&amp; (silo cross section) second layer to third layer

avl(2,3) = 1556.6 &amp;&amp; Assumes 0.3m path length

cfc(2,3) = 0.01 &amp;&amp; Arbitrary choice for an open path

topen(2,3) = 0.0

area(3,4) = 466.98 &amp;&amp; (silo cross section) third layer to fourth layer

avl(3,4) = 1556.6 &amp;&amp; Assumes 0.3m path length

cfc(3,4) = 0.01 &amp;&amp; Arbitrary choice for an open path

topen(3,4) = 0.0

area(4,5) = 466.98 &amp;&amp; (silo cross section) fourth layer to fifth layer

avl(4,5) = 1556.6 &amp;&amp; Assumes 0.3m path length

cfc(4,5) = 0.01 &amp;&amp; Arbitrary choice for an open path

topen(4,5) = 0.0

area(5,6) = 466.98 &amp;&amp; (silo cross section) fifth layer to sixth layer

avl(5,6) = 1556.6 &amp;&amp; Assumes 0.3m path length

cfc(5,6) = 0.01 &amp;&amp; Arbitrary choice for an open path

topen(5,6) = 0.0

area(6,7) = 466.98 &amp;&amp; (silo cross section) sixth layer to seventh layer

avl(6,7) = 1556.6 &amp;&amp; Assumes 0.3m path length

cfc(6,7) = 0.01 &amp;&amp; Arbitrary choice for an open path

topen(6,7) = 0.0

area(7,8) = 466.98 &amp;&amp; (silo cross section) seventh layer to eighth layer

**ATTACHMENT E  
PROBLEM INPUT DECK**

Page 3 of 5

avl(7,8) = 1556.6 && Assumes 0.3m path length  
cfc(7,8) = 0.01 && Arbitrary choice for an open path  
topen(7,8) = 0.0

area(8,9) = 466.98 && (silo cross section) eighth layer to ninth layer  
avl(8,9) = 1556.6 && Assumes 0.3m path length  
cfc(8,9) = 0.01 && Arbitrary choice for an open path  
topen(8,9) = 0.0

engvent

from = 9 to = 10  
varea = 1.2288e-02 && (Unloading Manhole) ninth layer to exterior  
vavl = 0.12094 && Assumes 0.1016m path length  
vcfc = 0.01 && Arbitrary choice for an open path  
vtopen = 0.0  
eoi

from = 9 to = 10  
varea = 1.8241e-02 && (Gooseneck) ninth layer to exterior  
vavl = 0.011969 && Assumes 1.524m path length  
vcfc = 0.01 && Arbitrary choice for an open path  
vtopen = 0.0  
eoi

from = 9 to = 10  
varea = 1.5518e-03 && (Instrument Manhole) ninth layer to exterior  
vavl = 0.0152735 && Assumes 0.1016m path length  
vcfc = 0.01 && Arbitrary choice for an open path  
vtopen = 0.0  
eoi

&& -----Print optional outputs-----

prflow

&& -----Begin cell inputs-----

cell = 1 && Bottom layer containing radioactive material  
control  
eoi

geometry 1771.29 3.7931 && (gas volume only - ignores solids)  
&& Assumes cylinder portion of silo is 2/3 full of material  
atmos = 4 99422.05 293.0  
o2 = 0.199025 n2 = 0.741 ar = 0.009975 co2 = 0.05  
eoi

&& -----End of cell 1 inputs

cell = 2 && Layer 2 (first layer above radioactive material)  
control  
eoi

**ATTACHMENT E  
PROBLEM INPUT DECK**

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geometry 180.744 0.38705  
atmos = 4 99422.05 293.0  
o2 = 0.199025 n2 = 0.741 ar = 0.009975 co2 = 0.05  
eoi  
&& -----End of cell 2 inputs-----

cell = 3 && layer 3 (second layer above radioactive material - all gas)  
control  
eoi  
geometry 180.744 0.38705  
atmos = 4 99422.05 293.0  
o2 = 0.199025 n2 = 0.741 ar = 0.009975 co2 = 0.05  
eoi  
&& -----End of cell 3 inputs-----

cell = 4 && layer 4 (third layer above radioactive material - all gas)  
control  
eoi  
geometry 180.744 0.38705  
atmos = 4 99422.05 293.0  
o2 = 0.199025 n2 = 0.741 ar = 0.009975 co2 = 0.05  
eoi  
&& -----End of cell 4 inputs-----

cell = 5 && layer 5 (fourth layer above radioactive material - all gas)  
control  
eoi  
geometry 180.744 0.38705  
atmos = 4 99422.05 293.0  
o2 = 0.199025 n2 = 0.741 ar = 0.009975 co2 = 0.05  
eoi  
&& -----End of cell 5 inputs-----

cell = 6 && layer 6 (fifth layer above radioactive material - all gas)  
control  
eoi  
geometry 180.744 0.38705  
atmos = 4 99422.05 293.0  
o2 = 0.199025 n2 = 0.741 ar = 0.009975 co2 = 0.05  
eoi  
&& -----End of cell 6 inputs-----

cell = 7 && layer 7 (sixth layer above radioactive material - all gas)  
control  
eoi  
geometry 180.744 0.38705  
atmos = 4 99422.05 293.0  
o2 = 0.199025 n2 = 0.741 ar = 0.009975 co2 = 0.05  
eoi  
&& -----End of cell 7 inputs-----

**ATTACHMENT E  
PROBLEM INPUT DECK**

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cell = 8 &amp;&amp; layer 8 (seventh layer above radioactive material - all gas)

control

eoi

geometry 180.744 0.38705

atmos = 4 99422.05 293.0

o2 = 0.199025 n2 = 0.741 ar = 0.009975 co2 = 0.05

eoi

&amp;&amp; -----End of cell 8 inputs-----

cell = 9 &amp;&amp; layer 9 (Head Space - eighth layer above radioactive material)

control

nsoatm = 1

nspatm = 50

numtbc = 1

maxtbc = 50

eoi

geometry 704.372 1.5084 &amp;&amp; (equivalent cylinder)

atmos = 4 99422.05 293.0

o2 = 0.199025 n2 = 0.741 ar = 0.009975 co2 = 0.05

eoi

&amp;&amp; -----2.98077E-03 Megawatt Fire Simulation-----

source = 1

h2ov = 5

iflag = 2

t = 0.0 100. 3500. 3600. 4000.

mass = 0.0 1.0e-10 1.0e-10 0.0 0.0

enth = 0.0 2.98077e+13 2.98077e+13 0.0 0.0

eoi

&amp;&amp; -----End of cell 9 inputs-----

cell = 10 &amp;&amp; Outside Atmosphere

control

eoi

geometry 1.0e10 20.

atmos = 3 99420.30 293.0 &amp;&amp; 1.751 Pa below ambient (lateral wind)

o2 = 0.2095 n2 = 0.78 ar = 0.0105

eoi

&amp;&amp; -----End of cell 10 inputs-----

eof