

# ORAU TEAM Dose Reconstruction Project for NIOSH

Oak Ridge Associated Universities I Dade Moeller & Associates I MJW Corporation

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## **PUBLICATION RECORD**

EFFECTIVE DATE	REVISION NUMBER	DESCRIPTION
02/01/2006	00	New document to establish the technical basis for the development of a radiation exposure matrix for Allied Chemical Corporation Plant. First approved issue. Training required: As determined by the Task Manager. Initiated by Cindy W. Bloom.
10/01/2007	01	Approved Revision 01 initiated to add text in Section 2.0 and remove information regarding nonuranium radionuclides in Sections 3.0 and 5.0 to incorporate SEC evaluation report recommendation to not reconstruct nonuranium internal dose. Moved Figure 2-1. Incorporates formal internal and NIOSH review comments. Added reference for employee statement regarding X-rays. Constitutes a total rewrite of the document. The recently designated residual period is identified. Section 5.0, Estimating Exposures From Residual Radioactivity, expanded to address dose reconstruction methodologies for exposures received during the residual period. Added Attributions and Annotations section and additional references. Training required: As determined by the Task Manager. Initiated by Bernard M. Olsen.

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## ACRONYMS AND ABBREVIATIONS

ACCP AEC AP AWE	Allied Chemical Corporation Plant U.S. Atomic Energy Commission anterior-posterior atomic weapons employer
Ci cm	curie centimeter
DOE dpm	U.S. Department of Energy disintegrations per minute
EEOICPA	Energy Employees Occupational Illness Compensation Program Act of 2000
ft	feet
hr	hour
IMBA IREP	Integrated Modules for Bioassay Analysis Interactive RadioEpidemiological Program
keV kg KOH	kiloelectron-volt, 1,000 electron-volts kilogram potassium hydroxide
L	liter
m MeV mL mrem	meter megaelectron-volt, 1 million electron-volts milliliter millirem
nCi NIOSH NRC	nanocurie National Institute for Occupational Safety and Health U.S. Nuclear Regulatory Commission
ORAU	Oak Ridge Associated Universities
pCi	picocurie
RMC	Radiation Management Corporation
SEC	Special Exposure Cohort
U.S.C.	United States Code
yr	year
μCi μg	microcurie microgram

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§ section or sections

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### 1.0 PURPOSE AND SCOPE

Technical basis documents and site profile documents are not official determinations made by the National Institute for Occupational Safety and Health (NIOSH) but are rather general working documents that provide historic background information and guidance to assist in the preparation of dose reconstructions at particular sites or categories of sites. They will be revised in the event additional relevant information is obtained about the affected site(s). These documents may be used to assist NIOSH staff in the completion of the individual work required for each dose reconstruction.

In this document the word "facility" is used as a general term for an area, building, or group of buildings that served a specific purpose at a site. It does not necessarily connote an "atomic weapons employer [AWE] facility" or a "Department of Energy [DOE] facility" as defined in the Energy Employees Occupational Illness Compensation Program Act of 2000 [EEOICPA; 42 U.S.C. § 7384I(5) and (12)]. EEOICPA, as amended, provides for employees who worked at an AWE facility during the contract period and/or during the residual contamination period.

Employment at an AWE facility is categorized as either (1) during the contract period (i.e., when the AWE was processing or producing material that emitted radiation and was used in the production of an atomic weapon), or (2) during the residual contamination period (i.e., periods that NIOSH has determined there is the potential for significant residual contamination outside of the period in which weapons-related production occurred). For contract period employment, all occupationally derived radiation exposures at covered facilities must be included in dose reconstructions. NIOSH does not consider the following exposures to be occupationally derived:

- Radiation from naturally occurring radon present in conventional structures
- Radiation from diagnostic X-rays received in the treatment of work-related injuries

For residual contamination period employment, only the radiation exposures defined in 42 U.S.C. § 7384n(c)(4) [i.e., radiation doses received from DOE/U.S. Atomic Energy Commission (AEC)-related work] must be included in dose reconstructions. Radiation dose received from DOE/AEC-related work includes: (1) radiation from radon consistent with NIOSH policies for including such radiation in the contract period; and (2) medical screening X-rays, but not diagnostic X-rays for the treatment of work-related injuries. It should be noted that: (1) under subparagraph A of 42 U.S.C. § 7384n(c)(4), radiation associated with the Naval Propulsion Program is specifically excluded from the employee's radiation dose; and, (2) under subparagraph B of this section, radiation from a source not covered by subparagraph A that cannot be reliably distinguished from radiation that is covered by subparagraph A is considered part of the employee's radiation dose. This site profile covers only exposures resulting from nuclear weapons-related work. Exposures resulting from non-weapons-related work, if applicable, will be covered elsewhere.

A decision has been made that internal dose from nonuranium radionuclides cannot be reconstructed with sufficient accuracy for employees of the Allied Chemical Corporation Plant (ACCP) during the period 1959 through 1976, so only internal dose from uranium is considered in Section 3 of this site profile. The Secretary of Health and Human Services (HHS) has designated a single class of employees from ACCP for inclusion in the Special Exposure Cohort (SEC) (Leavitt 2007). The class of employees for which NIOSH cannot estimate radiation doses with sufficient accuracy includes all AWE employees who were monitored or should have been monitored for exposure to ionizing radiation while working at the ACCP in Metropolis, Illinois, for a number of workdays aggregating at least 250 workdays from January 1, 1959, through December 31, 1976, or in combination with workdays within the parameters established for one or more other classes of employees in the SEC.

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Section 2.0 of this document describes the Allied site and its history including information about the radiological processes and source terms as well as the radiological controls and monitoring practices. Sections 3.0 and 4.0 discuss internal and external dose assessment, respectively. Section 5.0 provides information for assigning dose during the residual radiation period beginning in 1977, after the cessation of AEC related operations.

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 SITE DESCRIPTION AND OPERATIONAL HISTORY

The information that follows applies to the period of AEC operations at ACCP in Metropolis, Illinois, from January 1, 1959, to December 31, 1976, which involved AEC-contracted conversion of uranium ore concentrates to uranium hexafluoride. Opening ceremonies for the Allied Chemical's fluorine plant occurred in October 1958 (Sloop 1978). The original license issue date was December 17, 1958 (NRC 2003, p. 8-9). Four claims (Claim Numbers redacted) include bioassay results in December 1958, and one of these from December 15, 1958, has a positive result that indicates that uranium exposures might have occurred in late 1958, before the DOE Office of Worker Advocacy-established covered period. The plant was closed temporarily on June 30, 1964, and AEC (1966) reported that it might reopen in 1966. A review of available worker dosimetry records indicated that some workers might have been laid off or transferred around this period, but that other workers were still on the site. Documentation reviewed indicates that weapons related residual contamination exists outside the listed operational period (NIOSH 2006b). Residual contamination from prior weapons-related activities is indistinguishable from contamination period has been established as being from January 1, 1977 through the present (NIOSH 2006b).

Detailed information about buildings and processes is provided in Sections 2.1, 2.2, and 2.3 and is based on descriptions written after the period of operations covered by this document. Individuals who worked at ACCP indicated that there was little variation in the processes over time. Because individual dosimetry results are available for the vast majority of workers at ACCP, the reconstructed doses will be based on the specific individual dosimetry.

The ACCP radiological source term started with the receipt of uranium ore concentrates. Some longlived uranium progeny were included in the concentrates (e.g., <sup>230</sup>Th and <sup>226</sup>Ra). Uranium chemical forms included oxides, fluorides, and hexafluorides, which meant that exposures could have been to uranium solubility types F, M, and/or S.

The ACCP was also known as General Chemical Division, Allied Signal Metropolis Plant, and was later purchased by Honeywell.

### 2.1 SITE DESCRIPTION

"On February 4, 1957, the Allied Chemical and Dye Corp. announced selection of Metropolis III., as the site of its plant to process 5,000 tons of  $U_3O_8$  a year under contract with the [Atomic Energy] Commission" (AEC 1957). The official plant startup date appears to be January 1, 1959, but there are indications that uranium and uranium workers might have been on the site in late 1958. One individual reported participating in opening ceremonies at Allied Chemical's fluorine plant at Metropolis, Illinois in October 1958 (Sloop 1978), which indicates that uranium exposure could have occurred as early as October 1958.

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Allied Chemical operated its  $UF_6$  production facility using a dry conversion process (versus a wet solvent extraction process) to supply  $UF_6$  feed for the Paducah Gaseous Diffusion Plant through June 30, 1964, under an AEC contract (Perkins 1982). In addition, Allied Chemical was sampling uranium concentrates for other entities as of 1982.

On June 30, 1964, the plant was temporarily closed. AEC (1966) reported that the plant "may be reopened in 1966 for uranium hexafluoride production." Perkins (1982) noted that ACCP resumed operations in February 1968, but the available dosimetry records indicate that radiological exposures were still occurring at the plant between June 30, 1964, and February 1, 1968. Some of the claimant interviews and external dosimetry records show that a number of workers were on the site during the shutdown, and hiring was occurring in 1967, although these records also indicate that a number of workers were laid off in July 1964. The available records do not indicate what activities might have been ongoing in the plant during the shutdown period. However, based on U.S. Department of Labor job descriptions associated with one claim (Claim Number redacted), some processing operations might have continued at least on a sporadic basis.

The ACCP U.S. Nuclear Regulatory Commission (NRC) license (NRC 2003, p. 8-2) states:

The Allied Signal Metropolis Plant is located on approximately 1,000 acres of land in Massac County at the southern tip of Illinois, along the north bank of the Ohio River. The site perimeter is formed by US Highway 45 to the north, the Ohio River to the south, an industrial coal blending plant to the west and privately owned, developed land to the east. Plant operations are conducted in a single fenced-in, restricted area covering 59 acres in the north-central portion of the site.

The plant is now owned by Honeywell and is still providing UF<sub>6</sub> to gaseous diffusion plants for use in the commercial sector. The 1982 building locations are shown in Figure 2-1.

The following description of the buildings is excerpted from ACCP licensing documentation (NRC 2003, pp. 8-8 to 8-9). The list was originally numbered 8.5.1 through 8.5.8, but has been simplified to 1 through 8 below.

Most of the uranium processing equipment is housed in a six-story structure termed the Feed Materials Building where essentially all of the steps in the  $UF_6$  manufacturing process are conducted. Other areas and buildings in which operations are conducted involving the handling or processing of significant quantities of source material include the following:

- 1. A Sampling Plant which receives samples or concentrates for uranium assay and moisture content.
- 2. The Sodium Removal (pretreatment) and Uranium Recovery Facilities which are housed in buildings where high sodium content ore concentrates are treated to remove sodium impurity, and where materials are reprocessed to recover additional uranium.
- 3. The KOH [potassium hydroxide] muds washing facility which removes fluorides and KOH from the potassium diuranate muds generated in the fluorination scrubber system. The washed potassium diuranate is then processed through Sodium Removal. The wash liquors are neutralized at EPF [environmental protection facility, this facility probably did not exist during the early years].

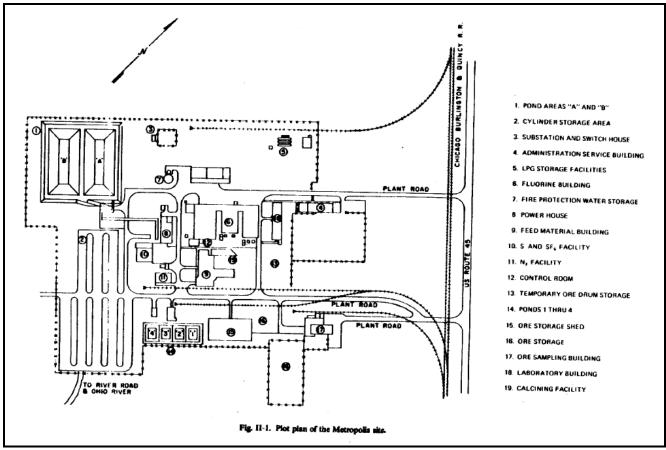


Figure 2-1. Plot plan of ACCP Metropolis, Illinois (Perkins 1982, p. 3).

- 4. The Calcining Facility which dries the incoming feed material and recovered uranium as the first step in ore preparation.
- 5. The Pond Mud Calciner Drier where hard/wet ore concentrates and KOH Muds are processed prior to packaging for blending with additional ore concentrates at the Feed Materials Building for conversion to UF<sub>6</sub>.
- 6. The Laboratory Building which houses facilities for conducting process control, product, and radiological control analyses.
- 7. The Cylinder Wash Building where  $UF_6$  product cylinders are periodically washed and hydrostatically tested prior to reuse.
- 8. Outdoor pads for storage of drums of ore concentrates and other uranium bearing materials, as well as UF<sub>6</sub> product cylinders.

Additional plant facilities which are involved directly in the UF<sub>6</sub> manufacturing process but do not involve the handling of any significant quantities of source material include a fluorine manufacturing building, a fluoride waste treatment facility with four large CaF<sub>2</sub> settling tanks, a powerhouse, a reductor off-gas incinerator, and two small uranium settling ponds to collect any uranium spills.

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Allied Chemical was also involved in the manufacture of 30,000 pounds per week of liquid fluorine, 1,200 tons per year of sulfur hexafluoride, 2,500 pounds per week of antimony pentafluoride, and 10,000 pounds per week of iodine pentafluoride (Perkins 1982). These materials were not radioactive.

### 2.2 PROCESS DESCRIPTION

The conversion process from ore concentrates (yellowcake) to  $UF_6$  is described below, and a process flow diagram is presented in Figure 2-2. As of 1982, the processing capacity was 14,000 tons of uranium per year and the plant had processed more than 100,000 tons of uranium from domestic and foreign uranium concentrates (Perkins 1982).

As part of an evaluation of environmental control technologies for nuclear fuel [uranium hexafluoride  $(UF_6)$ ] conversion facilities, Perkins (1982) described the conversion of yellowcake to uranium hexafluoride at ACCP. Although some changes in the process might have occurred since the first days of operation, it is likely that these changes would have been related to ore sampling techniques, ventilation and filtration controls, and waste disposal practices, and that the general process was similar throughout the plant's history. Process descriptions from Perkins (1982) follow.

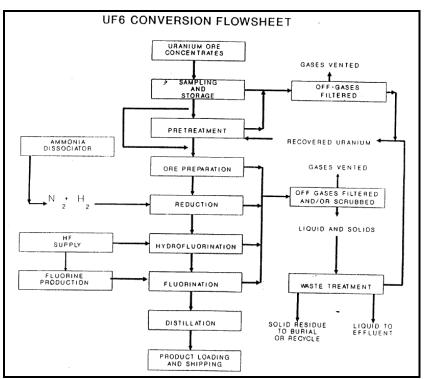


Figure 2-2. Process steps for conversion from ore concentrates (yellowcake) to  $UF_6$  (NRC 2003, p. 9-18).

#### Introduction

The Allied facility uses the fluoride volatility process to produce  $UF_6$  from uranium concentrates. The facility does not accept uranium slurries.

When the 55-gallon-drummed concentrate is received it is weighed, sampled, and analyzed for moisture content. After feed preparation, mechanically sized particles

 $(U_3O_8)$  are reacted with hot cracked ammonia to form uranium tetrafluoride  $(UF_4)$  or green salt. Next, the green salt is combined at high temperature in a fluidized bed with gaseous fluorine to form the gas  $UF_6$ . Finally, the  $UF_6$  off-gas is further purified by condensation and fractional distillation.

Each of these steps, with the resulting waste and effluent streams, is discussed in detail in the following sections.

#### Receiving and Sampling

Uranium concentrates are received at Allied Chemical Corp. Plant in 55-gallon drums. The drums are stored outside on specially constructed pads designed to allow rainwater to drain to centralized collection sumps, which is then pumped to uranium spill control ponds.

Drums are weighed, and a falling stream method is used to obtain a representative sample. The airborne dusts generated in the sampling process are removed through the use of two baghouses in series, which discharge through a stack. Empty drums are air cleaned and the discharge runs through two baghouses before being discharged to a stack.

Concentrates that are found to contain a high percentage of sodium are pretreated in the sodium-removal facility.

#### Sodium Removal (Pretreatment)

Because sodium forms a compound that causes caking and sintering in the fluorination fluid beds, any incoming uranium concentrates containing sodium and all the uranium concentrates produced by the uranium recovery sections of the UF<sub>6</sub> facility must first have the sodium or potassium removed before they can be processed. These concentrates are treated by reaction with a solution of ammonium sulfate in four counter current vessels to make  $(NH_4)_2U_2O_7$  for further processing. The chemical reaction is

$$Na_2U_2O_7 + (NH_4)_2SO_4 \rightarrow (NH_4)_2U_2O_7 + Na_2SO_4$$

The liquid effluent contains the excess  $(NH_4)_2SO_4$ ,  $Na_2SO_4$ ,  $K_2SO_4$ , and uranium and small quantities of <sup>226</sup>Ra and <sup>230</sup>Th, selenium, molybdenum, and other trace impurities. The effluent is sent to the two uranium spill control ponds. Uranium contaminated storm water is also discharged to these ponds. The sludges are allowed to settle, and the decantate is discharged to the main discharge outfall for the plant.... When the minimum freeboard is reached (approximately 2 ft) on a pond, the pond is dredged and the sludge is sent to the pond sludge calciner.... Pond liners are inspected and repaired at this time. In addition, all pond liners are underlain by a gravel layer that allows any seepage to drain to a leak detection sump....

The  $(NH_4)_2U_2O7$  (ADU) [ammonium diuranate] is removed from solution in the last reaction vessel and is sent to the main feed preparation section of the plant.

#### Feed Preparation

In the feed preparation circuit, sodium-free uranium concentrate, either from the pretreatment section or from the sampling section, is first fed into a calciner where water and ammonia are removed. The off-gases containing ammonium, sulfur dioxide, uranium, and contaminant particles pass through two baghouses in series. Particulates recovered by blowback of the baghouses are collected and sent back into the blending feed stream...

The calcined material is blended and then agglomerated, dried, crushed, and sized before being sent to the reduction circuit. Wet off-gas streams join the off-gases from the calciner, whereas dried off-gases pass through two baghouses in series before discharge.... Material collected by the dry stream baghouse is fed into the blending feed stream.

#### Reduction

In the reduction circuit, Allied operates two trains in parallel, both having the same basic design. The  $U_3O_8$  from the feed preparation circuit is fed into the reductor vessel where hot cracked ammonia ( $N_2$  and  $H_2$ ) and additional nitrogen mix with the feed to form a fluidized bed. The  $U_3O_8$  is reduced to  $UO_2$ , which is withdrawn from the bottom of the bed. Off-gases from the reactor include  $H_2$ ,  $N_2$ ,  $H_2S$ ,  $AsH_3$ , vaporized S,  $SeH_2$ , and particulates composed of  $UO_2$ , unreacted  $U_3O_8$  and reduced compounds of impurities originally in the concentrate....

The off-gases pass through two porous metal filters in series, a sulfur condenser, and incinerator....

Particulates collected by cleaning the filters are sent to the uranium recovery section.

Off-gases from the oxide vacuum cleaner are sent through cyclone and two baghouses in series before discharge.... Residues from these collection devices are sent to the uranium recovery section.

#### Hydrofluorination

Again in the hydrofluorination circuit, two trains in parallel are in use. The UO<sub>2</sub> from reduction is fed into two reactor vessels in series, which use vaporized hydrofluoric acid and N<sub>2</sub> to fluidize the UO<sub>2</sub> and permit HF to react with the material to form UF<sub>4</sub>. The off-gases contain the excess HF needed to give good conversion to UF<sub>4</sub> (10% excess or more), volatile SiF<sub>4</sub>, BF<sub>3</sub>, and some of the molybdenum and vanadium (which were present as impurities in the concentrate) as volatile fluorides and oxyfluorides, and any remaining sulfur as H<sub>2</sub>S<sub>4</sub>. These gases first pass through two sets of porous carbon filters per set, where the unreacted UO<sub>2</sub> particles, entrained UF<sub>4</sub> particles, and any other particulate matter are removed. The gases then pass through two venturi water scrubbers where the H<sub>2</sub>O in the off-gases condenses, and finally through a venturi KOH scrubber and a packed tower using KOH scrubbing liquid.

The residue from the clean carbon filters is sent to uranium recovery. Liquid coming from the venturi water scrubbers contains hydrofluoric acid and is sent to the acid neutralization treatment plant, where lime is added to precipitate the fluorine as  $CaF_2$ .

The uranium content of this stream is reported to average less than 5 ppm [parts per million]. Silicon, boron, and some molybdenum, and vanadium, which were originally in the concentrate, are expected to be in the sludge. The liquid from the KOH venturi and packed tower is sent to the liquid treatment systems.

The vacuum cleaning off-gases from the cleaning activities necessary in the hydrofluorination section are routed through a cyclone and two baghouses before discharge.... Residues from the dust collectors are sent to the uranium recovery section.

#### Fluorination

The green salt,  $UF_4$ , produced in the hydrofluorination process is fed into one of two fluid bed reactors used in parallel. Again, Allied uses two trains at the works. The bed material is CaF<sub>2</sub>, and unreacted UF<sub>4</sub>. The vessels are operated at temperatures of approximately 1000° F, and good temperature control is required. Fluorine, F<sub>2</sub>, from an on-site fluorine production facility is introduced into the reactor vessels to convert UF<sub>4</sub> into gaseous UF<sub>6</sub>.

The off-gasses, including uranium  $UF_6$ ,  $F_2$ , HF, unreacted  $UF_4$ , and other particulates, and volatilized impurities ... are first cooled before passing through two sets of sintered nickel filters, each containing two filters. The material recovered as the filters are cleaned is stored, and then sent to the uranium recovery section.

The stream exiting from the filters has the UF<sub>6</sub> removed by condensation in a set of three cold traps used in series. Any uncondensed UF<sub>6</sub>,  $F_2$ , HF and other volatiles passing from the cold wraps are removed from the gas stream by a KOH spray tower, a KOH pack tower, and finally a KOH coke box.

Carbon dioxide in the spent scrubber liquor reacts with the uranium and potassium as follows.

 $2(UO_3)^{4-} + 6CO_2 + 6OH + 2K^+ \rightarrow K_2U_2O_7 + 6CO_3^{-2-} + 3O_2 + 3H_2O$ 

The  $K_2U_2O_7$  is settled from the KOH solution and sent to the pretreatment facility.

Decantate from the uranium settling section is sent to the KOH regeneration system as indicated previously.... The KOH is then sent back to the scrubbing system. In addition to  $CaF_2$  compounds of uranium, vanadium, and molybdenum, trace quantities of compound of silica, carbon sulfur, and other trace contaminants are expected to be in the sludge....

The condensed material in the three cold traps in series is melted and drained to the still feed tanks. Entrained hydrogen fluoride is vaporized during melting and passes to the off-gas system.

From the still feed tanks, the liquid is fed to a low-temperature boiler bubble cap column in which the impurities are volatilized and exit from the top of the column....

The liquid uranium hexafluoride passes from the low boiler column into the bubble cap, high boiler column. In this column, the  $UF_6$  is volatilized and exits from the top of the

column. Non-volatilized impurities are removed from the bottom of the column and stored.

The gaseous  $UF_6$  from the column is condensed into two cold traps operated in series. The  $UF_6$  is transferred to the shipping cylinders by melting the  $UF_6$  and allowing the material to drain into the cylinders.

Because impurities build up on the recycled  $CaF_2$  bed material used in the fluorination reactors, part of the bed material must be withdrawn periodically. This "ash" [also referred to as spar or bed material] along with the fluorination-cleaned filter residue, is drummed and stored for a minimum of 6 months to permit the uranium daughters <sup>234</sup>Th and <sup>234</sup>Pa to decay. The ash is then sent to the uranium recovery section. A total quantity of about 0.1 ton of ash per ton of uranium processed is produced, and it contains approximately 1.8% of the total uranium processed and most of the uranium daughters originally present in the concentrate.

The dusts produced by withdrawing bed material from the reactors are passed through a cyclone and two baghouses in series before discharge.... The ash vacuum cleaner systems also discharge from this stack. Residue from cleaning these particulate removal devices is treated similarly to the spent bed material....

The feed preparation, reduction, hydrofluorination, and fluorination circuits are all located in the feed material building. This building has a complete air changeout approximately once every 5 minutes....

Washdown of the feed materials building is pumped from collection sumps into the uranium recovery ponds.

The transfer lock to fluorination discharges through two baghouses.... Again, dust collection residue is sent to uranium recovery.

#### Uranium Recovery

There are several sources of uranium-containing wastes produced by the facility, and it is desirable to recover the uranium from these wastes. Thus, uranium-containing dried sludges, spent bed material, filter residue, and various scraps and dust are sent to the uranium recovery circuit.

Off-gases from storage before processing vent through the 3-1 stack. The main contaminants in the stack off-gases should be radon and radon daughters, if the radium originally in the concentrate feed goes into the ash (as was assumed in the previous section).

The material from storage is first dumped and ground. Dusts from this operation are recovered in a baghouse....

The finely ground material is leached using a sodium carbonate solution to solubilize the uranium as the tricarbonate complex....

After leaching, the pregnant solution is filtered. Lime is used to aid in precipitation of any fluoride, and the solution is again filtered. Sludge from the filters is dried. Off-gases from the dryer pass through the dumping and grinding off-gas baghouse.

The dried sludge probably contains all nondecayed uranium daughters, some residual uranium, and some fluoride compounds. This sludge is drummed and sent to a licensed radioactive waste disposal facility. Approximately 1500 tons of this waste, containing a maximum of 46.9 Ci is produced yearly.

The uranium is precipitated from the solution using NaOH. The uranium is filtered and washed and then sent to the sodium removal section.

#### Cylinder Wash

Cylinders are returned to Allied from the enrichment facilities. These cylinders contain residual  $UF_6$  and the daughters of uranium that have "grown in" while the cylinders containing  $UF_6$  were in storage. The daughters in general are found plated out on the cylinder walls.

After any residual  $UF_6$  is removed from the cylinder, the cylinder is washed, using a solution of  $Na_2CO_3$  to remove any impurities from the walls and to solubilize the uranium. The wash solution is filtered to remove the unleached solids, and the pregnant solution is pumped to join the pregnant solution in the uranium treatment section.

The solid residue from the filters contains daughter products of uranium, principally <sup>234</sup>Th and <sup>234</sup>Pa, and is stored on-site in drums until disposal in a licensed waste disposal facility.

#### Sludge Dryer

As previously described, the process generates several sludges that contain uranium. Before being sent for uranium recovery, these sludges are dried in a calciner. Offgases containing  $SO_2$ , HF and particulates from the calcining operation pass through a baghouse and a water spray tower before discharge....

The spent scrubber water, which contains uranium, fluoride compounds, and some sulfur compounds, is sent to the uranium recovery ponds.

The calcined sludges are drummed and stored until they can be processed in the uranium recovery section.

### 2.3 SOURCE TERM

The plant produced  $UF_6$  from uranium ore concentrates initially for gaseous diffusion facilities and later also for commercial fuel fabrication facilities. The uranium conversion process at the plant has remained essentially unchanged since its inception. In 1982, Perkins (1982) reported that the concentrates initially contained 70% to 75% uranium by mass, and these concentrates included impurities that were dependent on the mines and mills from which the yellowcake came.

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Uranium chemical forms at ACCP included all solubility types (i.e., F, M and S). The initial design processing capacity was 5,000 tons of uranium in a year, and by 1982 the throughput capacity had been increased to 14,000 tons of uranium in a year in what was essentially a continuous chemical process. As of 1982, more than 100,000 tons of uranium had been processed (Perkins 1982).

The initial milling of the uranium before arrival at ACCP resulted in disequilibrium of uranium progeny in the concentrate. Exposure to the nonuranium radionuclides occurred at the front end of the processing, beginning with receipt. Increased concentrations of other radionuclides in relation to uranium were found in the fluorination bed ash and other solid waste streams, such as the sludge. Environmental air measurements in the late 1970s showed ratios of <sup>226</sup>Ra and <sup>230</sup>Th in relation to uranium that were larger than the concentrate ratios but lower than complete equilibrium ratios.

Radon concentrations would probably have been elevated during the initial opening of the drums of concentrate, in the feed area, and in work areas where radium would have concentrated. No measurements of radon concentrations have been found at this time.

Perkins (1982) summarized uranium, thorium, and <sup>226</sup>Ra activities in ACCP effluents and wastes from the 1970s. These values, listed in Table 2-1, were used to estimate ratios of the activity of other radionuclides to uranium in the effluent. The reported concentrate ratio was based on an average of yellowcake feed ratios from 33 mills (Perkins 1982, Table II-8). The equilibrium activity ratio of <sup>230</sup>Th to uranium in uranium ore would be about 0.4886; the milling of the ore usually reduces this ratio by almost a factor of 100 by removing the <sup>230</sup>Th from the concentrate. It should be noted that <sup>232</sup>Th comprises a portion of the radioactivity in the uranium concentrates, and although <sup>232</sup>Th was not specifically reported for most effluents at ACCP, the water effluents for 1975 and 1976 showed annual average <sup>232</sup>Th:U-nat and <sup>230</sup>Th:U-nat activity ratios of 0.076 and 0.017, respectively.

	Average			Largest ratio		
Source	Th-nat:U-nat	Th-230:U-nat	Ra-226:U-nat	Th-nat:U-nat	Th-230:U-nat	Ra-226:U-nat
Concentrate (yellowcake feed averaged over 33 mills)	ND	5.20E-03	ND	ND	ND	ND
Uranium recovery solid waste (dried sludge)	3.58E+00	1.90E+01	1.11E+00	ND	ND	ND
Solid nonsludge waste	ND	5.27E-03	2.75E-04	ND	ND	ND
Outdoor air	ND	6.70E-02	1.97E-03	ND	1.67E-01	8.46E-03
Air effluent	ND	1.70E-02	2.70E-04	ND	4.75E-02	3.38E-04
Water effluent	ND	3.99E-03	2.04E-02	ND	1.58E-02	5.79E-02

Table 2-1. Radionuclide activity ratios in relation to uranium based on reported activities in ACCP concentrates, wastes, air, and effluents.

ND: No data provided in Perkins (1982)

Processing at ACCP results in further separation of uranium and its progeny, and this can result in increased activity ratios of nonuranium radionuclides in some areas in the plant. The largest disequilibrium ratios during the operational period are associated with the solid waste (dried sludge) from the uranium recovery facility. As stated above, these materials were barreled and shipped offsite as radioactive waste. AEC and DOE inventory reports use a designation of JSA to identify ACCP. DOE reports that the Rocky Flats Plant shipped 71 kg (160 pounds) of depleted recycled uranium to ACCP in 1975, which is a very small fraction of the annual inventory (which might have been as high as 10 to 28 million pounds) processed at ACCP (RFETS 2000, Appendix A, p. 27). A second mention of the same shipment indicates that the 71 kg were shipped to Allied Chemical Corporation in Idaho Falls, Idaho (RFETS 2000, Appendix B, p. 2). Another reference (Author unknown ca. 2000) referred to a shipment of 22,060 kg from Rocky Flats Plant to ACCP between 1955 and 1964, but this appears

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to be an error due to a mix-up between the next designation in the report, LAB, with JSA. It is assumed that contaminants in recycled uranium were not significant contributors to ACCP workers' radiation dose.

ACCP is licensed by the NRC to have sealed Cs-137 sources up to 100 millicuries each. These are used as level control devices and are unlikely to contribute to internal dose.

## 2.4 SAFETY

Specific radiological safety measures implemented in the beginning of ACCP operations are not available but, as of 1958, radiological control programs in general had become more sophisticated. Dosimetry is available from the earliest days of covered operations for all employees working in the controlled area. The following information is based primarily on reference material dated after 1972, and it should be noted that the criteria to prompt radiological control activities could have differed in the early years.

Engineered confinement was and is the primary source of contamination control used at ACCP. Drums, process vessels, lines, etc., are used to confine the materials. When unconfined uranium is handled, procedures call for ventilation controls to reduce exposures (Author unknown 1985, p. 1).

An exception was requested from the NRC requirement to post and label individual radioactive material areas and containers, which was replaced by a requirement to train workers (NRC 2003, p. 11-1). Signs state that any area or container in the plant may contain radioactive materials.

Protective clothing and shoes or shoe covers were provided to employees and visitors to ACCP to ensure that personnel were not inadvertently contaminated with uranium compounds as of 1982 (Perkins 1982), and probably from the earliest days of operations. This did not apply to administrative areas where contamination would not be expected.

Current ACCP procedures, discussed below, include four contamination measurement and control parameters:

- Visual observation of contamination
- Air concentration measurements
- Surface contamination measurements
- Decontamination procedures

## 2.4.1 <u>Visual Observation of Contamination</u>

Allied Chemical noted that natural uranium compounds have a low specific activity, characterized by a high ratio of mass to activity. The ACCP uranium compounds are highly colored, which provides an immediate method for detecting gross surface and air contamination. For instance, yellowcake was yellow and UF<sub>6</sub> releases appeared as white smoke due to the decomposition products,  $UO_2F_2$  and HF. Procedures from 1985 stated that if employees detected small spills, they were responsible for cleanup (Author unknown 1985). Larger leaks or spills that resulted in visible accumulation of uranium on equipment or on the floor required immediate decontamination, activation of the respirator lights, and possibly shutdown of the unit. The foremen were to prepare a health physics incident report and notify the health physics shift technician to initiate special air sampling and investigation as appropriate. Health physics personnel made a daily inspection of uranium operating areas to ensure proper decontamination and contamination control. These procedures were consistent with contemporary industrial health practices for uranium facilities (DOE 1988).

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### 2.4.2 <u>Air Activity Measurement of Contamination</u>

As of 1985, stationary breathing zone samples were collected daily from 57 stations in the uranium hexafluoride building, 10 stations in the sampling plant, 2 stations in the drum dumper, 3 stations in sodium removal, and 2 stations in the uranium recovery unit. Summaries of some of these measurements can be found in the available references. As of 1985, respirators were required when the average gross alpha air concentration on a floor exceeded 4E-11  $\mu$ Ci/cm<sup>3</sup> or when any four air samples on the floor exceeded that concentration. In addition, as of 1985, any single air sample result that exceeded 1E-10  $\mu$ Ci/cm<sup>3</sup> required investigation and preparation of a health physics incident report (Author unknown 1985, p. 3).

Although some air concentration data are available for the 1970s and later, these data are not used to estimate worker intakes because of the abundance of bioassay data.

## 2.4.3 Surface Contamination Measurements

Surface contamination measurements in uranium and nonuranium areas are made routinely. Removable contamination is determined by taking a wipe or smear of 100 cm<sup>2</sup> on a surface. Each smear is counted for alpha radioactivity and reported in dpm/100 cm<sup>2</sup> removable alpha contamination. The plant lunchroom and offices or control rooms are surveyed weekly, as are the locker rooms and personnel exit areas. Uranium processing areas such as the Feed Materials Building, Sodium Removal, Uranium-Recovery Building, and Sampling Plant are surveyed monthly. Other nonuranium processing areas are surveyed quarterly (Author unknown 1985; Author Unknown 2002a).

As of 1985, decontamination is to be initiated when removable alpha contamination exceeds 5,000 dpm/100 cm<sup>2</sup> in the uranium processing areas. All other areas are to be decontaminated when removable alpha contamination exceeds 200 dpm/100 cm<sup>2</sup> (Author unknown 1985).

One summary of Health Physics data for the last half of 1991 (Wilkins 1992) noted that 1.7% of the 2,002 weekly smears exceeded the weekly limit of 200 dpm/100 cm<sup>2</sup> and that the highest result was 923 dpm/100 cm<sup>2</sup> in the lunchroom on a table. One of the 570 monthly smears and none of the 163 quarterly smears exceeded the limit.

### 2.4.4 Decontamination

Allied Chemical has detailed procedures for decontaminating spills or leaks found in the plant. These procedures generally include roping off the area, sweeping or vacuuming the contaminated material, and then washing the contaminated surface. During these procedures protective clothing is to be used and monitoring is to be performed (Author unknown 1985; Author unknown 2002b).

### 2.5 INCIDENTS

The claims include some information about incidents, but details are few and there are no dates or references to particular incident reports. Occasional leaking valves or inadequate packing resulted in material releases.

### 2.6 PHYSICAL EXAMINATIONS – X-RAYS

No documented ACCP requirement for occupationally required medical X-ray examinations has been found. More than half of the claimant interviews indicate that chest X-ray examinations were routine and a number of the interviews indicate the frequency was annual. The examinations were offered

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annually, but were optional by 1980, and would have been done at Massac Memorial Hospital or perhaps a local doctor's office according to one employee, who began work at ACCP in the 1970s (Bloom 2005).

# 2.7 SUMMARY OPERATIONAL PERIOD ASSUMPTIONS, WORKDAYS, WORK HOURS, WORK CATEGORIES

Because bioassay and film badge data were used to estimate internal and external exposures during the AEC uranium operations, an estimate of the number of workdays or work hours per year is not necessary.

No attempt has been made to sort workers into exposure categories. Individual dosimetry appears to be available for most workers.

### 2.8 RESIDUAL CONTAMINATION

Uranium concentrate processing continued at ACCP after 1976, and differentiation between residual contamination from the AWE period and contamination due to later and/or ongoing operations is not feasible.

## 3.0 ESTIMATING INTERNAL EXPOSURE

As stated in Section 1.0 above, a decision has been made that internal dose from nonuranium radionuclides cannot be reconstructed with sufficient accuracy for employees of ACCP during the AEC operations period 1959 through 1976. The primary source of internal radiation exposure at ACCP was uranium dust produced from the processing of uranium concentrates to produce uranium hexafluoride. It is assumed that the uranium was of natural enrichment before 1977, although there is some indication that the ACCP source term included a small amount of depleted uranium. Although the uranium ore concentrate contained uranium progeny of dosimetric interest as well as thorium (assumed to be <sup>232</sup>Th and <sup>228</sup>Th), only uranium dose is estimated for ACCP claims for the period January 1, 1959 through December 31, 1976.

### 3.1 URANIUM

During the processing of uranium concentrates at ACCP, workers might have been exposed to a variety of uranium chemical forms that encompass all lung absorption types: F, M, and S. Although some process steps might have had more or less exposure to a given lung absorption type, it is not clear how well separated these areas were. In addition, workers might have worked in multiple uranium process areas. The selection of absorption type should depend on the organ of interest. The specific activity of the natural uranium and the isotopic mass and activity fractions are listed in Table 3-1 [1]. For organ dose estimates, the uranium activity can be assumed to be from  $^{234}$ U [2]. A conversion factor is provided to adjust results reported as  $\mu g$  of  $^{235}$ U to total uranium activity in picocuries.

Isotope	Activity	Activity Mass Activity ratio Mass rat fraction fraction to U-235 to U-235					
1301000	Haction	пасноп	10 0-200	10 0-235			
U-234	0.4886	5.37E-05	21.4	0.00745			
U-235	0.0228	7.20E-03	1	1			
U-238	0.4886 9.93E-01 21.4 138						
Useful factors							
U-natural pCi/µg 0.683							

U (pCi): U-235 (µg)	94.8

## 3.1.1 Uranium Urinalysis Data

Individual uranium urinalysis began as early as December 1958 and was done in house. It appears that there are bioassay data for all workers. The worker records show that bioassays were typically collected at hire and periodically thereafter. Allied Chemical notes that fluorimetry was used for analyses until 2002, when kinetic phosphorescence analysis (KPA) began (Rogers 2006). Urinalysis data for 1989 to 1992 are missing, and ACCP reported that it was assumed that the missing records might have been water-damaged beyond recovery.

In the early years, uranium urinalysis results reported in microcuries per milliliter ( $\mu$ Ci/mL) are misstated; the units are actually micrograms per liter ( $\mu$ g/L). The recording limit of 10  $\mu$ g/L is found in the early records and is consistent with the default detection threshold for uranium urinalysis assumed to be 10  $\mu$ g/L based on a reported sensitivity of 5 to 10  $\mu$ g/L for uranium fluorimetry urinalysis in the early years (Wilson 1958). Beginning in late 1974, a recording limit of 5  $\mu$ g/L appears to have been in use. Allied Chemical reported that its minimum detection level (MDL) was 2  $\mu$ g/L in the early 1980s, but no records of MDL are available before that. In 1985, ACCP noted that routine urinalysis samples were to be left after 24 to 96 hours of absence from the workplace, but it is unknown when this procedure was implemented. In addition, as of 1985, ACCP employees were encouraged to leave special urine samples before their next work shift in response to known or suspected airborne exposures (NRC 1993, p. C-14). In later years, it was noted that a result equal to 30  $\mu$ g/L was cause for investigation.

On the computer printouts, results are sometimes noted in the comments section rather than the bioassay measurement section.

### 3.1.2 <u>Uranium In Vivo Data</u>

Lung counts are available for many workers beginning in late 1978, which is after the covered operational period. Lung counts appear to have been performed on an annual or biannual frequency until the mid-1980s. They appear to be less frequent in later years, but this could indicate that only some workers were required to have annual lung counts in later years.

ACCP claims include a letter that reports use of a uranium lung-counting system from Radiation Management Corporation (RMC) and analysis of the spectra using a software package called RMCWBC, which was based on the Oak Ridge National Laboratory program ALPHA-M (Rogers 2006). The counting system includes an RMC-designed shadow shield and four detectors, which appear to consist of thin Nal(TI), and thicker CsI(Na) crystals. The diameter is reported as 5 feet, but is probably 5 inches. Before analysis of the spectra, the background is automatically subtracted. A REMCAL standard man equivalent phantom was used to calibrate the system. Radionuclides include <sup>235</sup>U, <sup>234</sup>Th, <sup>137</sup>Cs, and <sup>40</sup>K.

Specific ACCP criteria are given for judging the data and can be used for considering the relative validity of a result (Rogers 2006):

- Fit should be less than 3.0 and usually ranges between 0.5 and 1.5. An ideal fit would be 0.
- The shift value should range between +2.0 and -2.0; values outside the range of ±3.0 are carefully reviewed before they are accepted. MCA zero level and/or gain adjustments are warranted when the shift value exceeds ±2.0.

• The gain should ideally be 1.0 and a gain of 0.90 to 1.10 is considered acceptable. Adjustment to the gain and/or zero might be needed if the value is outside this range.

The analysis program reportedly prints a table showing all radionuclides found and their estimated activities and relative 2-standard-deviation counting error in terms of a percent error. Allied Chemical reports that results of <sup>235</sup>U are given in µg and results for other radionuclides are given in nanocuries; however, some records show <sup>234</sup>Th results in µg. Results with errors greater than 100% (results less than 2 times their counting standard deviation) are considered questionable. The reported <sup>235</sup>U minimum detection level was noted as 63 µg, which was equated to 32% of the maximum permissible lung burden. The maximum permissible lung burden was noted as 194.3 µg of <sup>235</sup>U or summed as 26,974 µg of total uranium (Author unknown 1986, p. 11). In later years, it was noted that a result equal to 50% of the maximum permissible lung burden was cause for investigation.

Unless there is a prominent low-energy peak or the detector ratios vary by more than 0.025, the detected uranium is assumed to be in the lung.

In general, the urinalysis results should be used to estimate intakes and the lung counts can be used as supporting data. The lung counts do not directly measure total uranium and, although height and weight are recorded and could have been used to estimate chest wall thickness, there is no indication that chest wall thicknesses were measured directly.

To convert <sup>235</sup>U mass to total uranium activity, it is assumed that the uranium is of natural enrichment, and so the <sup>235</sup>U mass in  $\mu$ g is multiplied by 2.16 pCi/ $\mu$ g (the specific activity of <sup>235</sup>U) and divided by 0.0228 (the activity fraction of <sup>235</sup>U), a total factor of 94.8, to obtain the total uranium activity in picocuries [1].

To convert <sup>234</sup>Th to total uranium activity, it is assumed that the <sup>234</sup>Th is in equilibrium with both <sup>238</sup>U and <sup>234</sup>U, and so the mass of <sup>234</sup>Th in micrograms is multiplied by 2.32E+10 pCi/µg and divided by 0.4886 to obtain the total uranium activity in picocuries [3]. If the <sup>234</sup>Th is given in nCi, it can be converted to picocuries of uranium by dividing by 4.886E-4. Note that with a 24.1-day half-life, <sup>234</sup>Th might not be in equilibrium with its uranium parent depending on when and where an exposure occurred.

The <sup>40</sup>K and <sup>137</sup>Cs reported in lung counts are not likely to be associated with workplace exposures; <sup>40</sup>K is ignored and any positive <sup>137</sup>Cs results are evaluated against the background levels due to fallout. Cesium-137 values associated with fallout are shown in Table 3-2.

A handwritten note (Author unknown 1996) indicates that other whole-body counting systems might have been in use as of 1996, but there were no differences in reported sensitivity.

Year	Body burden (nCi)	Year	Body burden (nCi)
1953	0.27	1966	9.7
1954	1.1	1967	5.6
1955	2.2	1968	3.5
1956	4.3	1969	2.7
1957	5.1	1970	2.7
1958	6.5	1971	2.7
1959	8.1	1972	2.7
1960	6.8	1973	2.7
1961	4.6	1974	1.6
1962	6.0	1975	1.1

Table 3-2. Mean whole body <sup>137</sup>Cs from fallout in the United States.<sup>a</sup>

1963	11	1976	1.6
1964	19	1977	1.1
1965	16		

 a. Mean whole body Cs-137 activity in the United States due to fallout. Adjusted from becquerels to picocuries by multiplying NCRP Report 94(NCRP 1987, Table B.5) results by 27 picocuries per becquerel.

#### 3.2 NOTATIONS ON BIOASSAY RECORDS

Some of the bioassay records include notations and abbreviations. These include:

- Laid-off
- Rehired
- Trnfd [transferred]
- MTO [Morristown Office, corporate headquarters in New Jersey]
- MPLB [maximum permissible lung burden]
- Sample type R [assumed to mean routine]
- Sample type S [assumed to mean special]

# 3.3 OCCUPATIONAL INTERNAL DOSE RECONSTRUCTION ASSUMPTIONS AND SUMMARY

The AEC operational exposure period was from January 1, 1959, to December 31, 1976. As stated in Section 1.0 above, a decision has been made that internal dose from nonuranium radionuclides cannot be reconstructed with sufficient accuracy for employees of ACCP during this period, so only internal dose from uranium is considered in this section.

In general, the urinalysis results should be used to estimate uranium intakes and the lung counts can be used as supporting data. The uranium photofluorimetry urinalysis reporting limit in the early years at ACCP was 10  $\mu$ g/L [4]. The less-than results, when available in the individual dosimetry records, can be assumed to be uranium urinalysis detection thresholds. Uranium can be absorption type F, M, or S. For dose calculations, uranium intakes can be assumed to be <sup>234</sup>U [2].

To estimate uranium intakes from the lung counts, the intake of  $^{235}$ U is determined and then adjusted to total uranium intake activity using the conversion in Table 3-1. The lung detection threshold is 63 µg for  $^{235}$ U and 5,970 pCi for total uranium.

Based on the current information, all workers were monitored for uranium intakes, so at this time no assumptions have been provided for unmonitored workers or unmonitored periods. This section will be developed further, if necessary.

#### 4.0 ESTIMATING EXTERNAL EXPOSURE

External dosimetry results are available for ACCP workers beginning on January 1, 1959, and include results for gamma, beta, and neutron exposures. Allied Chemical reports that the data before 1970 are handwritten and the records do not include the company or method of measurement. Beginning in 1969, R.S. Landauer Jr. & Company provided film badge dosimetry services and measured gamma and beta radiation. In about April 1976, Eberline Instrument Company provided thermoluminescent dosimeter services. Neither Landauer nor Eberline reported doses below 10 mrem.

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Film badge detection thresholds in the early days were assumed to be as high as 50 mrem for gamma and 60 to 80 mrem for beta. A detection threshold of 40 mrem has been recommended for this Project (ORAUT 2006).

The majority of photons from natural uranium have energies in the range of 30 to 250 keV, so this analysis assumed workers were exposed to photon energies in the 30- to 250-keV range, which is consistent with NIOSH (2006c). Nonpenetrating dose from natural uranium consists primarily of electrons with energies above 15 keV. Because the uranium is mixed with material that can produce neutrons from an alpha particle collision, the neutron dose is also considered. For consistent presentation, exposure or dose is reported as:

- Penetrating, assumed to be associated with photons of energies 30 keV or greater
- Nonpenetrating, assumed to be associated with photons of energies less than 30 keV or with electrons
- Neutron, assumed to be associated with neutrons of energies of 0.1 to 2 MeV

The earliest available ACCP external dosimetry results are dated January 1, 1959. Badge exchange frequencies at ACCP were typically monthly in the available records. In the 1980s, hourly employees exchanged badges monthly and salaried employees changed badges quarterly.

Although some records have results recorded as neutron doses, in later years no records of neutron doses are available. Most of the results appear to be zeros, and it is not clear if these represent actual dosimeter results or were entered because there was no result. The Bridgeport Brass site profile (ORAUT 2005a) notes that the minimum detectable neutron dose reported in the early 1960s was 60 mrem.

A reasonable neutron dose estimation which is favorable to the claimant is based on a 500-pound drum that contains 75% by mass of natural uranium. It is assumed that the alpha particles collide with a fluorine target. Using the method described in (ORAUT 2005b), the neutron dose is 3.28E-04 rem/hr, calculated at 1 foot from a drum. A worker who is 1 foot from such a drum for 2,000 hr/yr would be exposed to a dose rate of 6.56E-01 rem/yr. This document considers this estimated neutron dose rate to be an upper bound (maximum estimate) of the ACCP neutron dose rate. The mode neutron dose rate is estimated as 5.47E-03 rem/yr, which is the result of applying an occupancy of 3 hours/week over 50 weeks to the estimated dose rate at 3 feet from a drum [5]. The minimum neutron dose rate is estimated as 0 rem/yr. These estimated minimum, mode and maximum neutron dose rates are to be used only for reconstructing doses during the listed operational period, and applied in IREP as a triangular distribution. During the residual period, the weapons-related source materials (uranium, fluorine targets) needed to produce neutrons are not presumed to be present in amounts significant enough to warrant the estimation of neutrons.

## 4.1 OCCUPATIONALLY REQUIRED MEDICAL X-RAY

No definitive information was found that indicated the type or frequency of occupationally required medical X-ray examinations, although the claimant interviews support an assumption of an annual chest X-ray examination. The type and projection of X-ray examination should be based on current Oak Ridge Associated Universities (ORAU) Team guidance, which at this time is a PA radiographic chest examination. Organ doses can be obtained from the current revision of ORAUT-OTIB-0006, *Dose Reconstruction from Occupationally Related Diagnostic X-Ray Procedures* (ORAUT 2005c).

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### 4.2 MISCELLANEOUS INFORMATION ABOUT EXTERNAL DOSE

This section includes external dose information that could be of value for specific dose reconstructions. This analysis did not consider such information generically because of its limited applicability or because of limited details.

The fluorination bed ash contains short-lived progeny, which results in elevated beta and gamma radiation fields in the vicinity of the ash. Workers who handled this material or washed drums or cylinders could receive extremity doses that are significantly higher than those measured by dosimeters worn on the trunk of the body.

# 4.3 OCCUPATIONAL EXTERNAL DOSE RECONSTRUCTION ASSUMPTIONS AND SUMMARY

Table 4-1 summarizes assumptions about occupational external doses during the covered period January 1, 1959, to December 31, 1976, at ACCP.

Exposure		
mode	Exposure type	Annual exposure
	Penetrating (photons, 30– 250 keV, AP, acute)	Based on individual dosimetry
Uranium processing	Non-penetrating (electrons >15 keV, acute)	Based on individual dosimetry
	Neutron 0.1 to 2 MeV, triangular	6.56E-01 rem max; 5.47E-03 rem mode; 0.00 rem min.
PA chest radiographic examination	Initial plus one exam per year	See ORAUT-OTIB- 0006 (ORAUT 2005b)

Table 4-1.	External exp	osure summary.
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## 5.0 ESTIMATION OF EXPOSURE FROM RESIDUAL RADIOACTIVITY

ACCP continued to operate after the covered AWE period ended on December 31, 1976. NIOSH has determined the residual radiation period to be from January 1, 1977 to present (NIOSH 2006b). As stated in Section 2.8, differentiation between residual contamination from the AWE period and contamination due to later and/or ongoing ACCP operations is not feasible. The following information provides a method for estimating exposures during the residual radiation period due to uranium and nonuranium residual contamination.

Internal exposures due to intakes of uranium after 1976 can be estimated using personnel monitoring data which was collected during this period (in vivo and in vitro data, as applicable). Table 5-1, below can be used to account for intakes of nonuranium radionuclides during the residual radiation period. The values in this table are based on the highest reported ratio in the outdoor air and air effluent pathways reported in Table 2-1. The use of these pathways during the residual period is appropriate since they are representative of the isotopic distribution of the resuspended materials to which workers present during the residual period would be exposed [6]. Since no values are reported for Th-232, the water effluent value (reported in section 2.3 were used).

largest reported ratio in relation to uranium in Table 2-1).		
U-natural	1	
Th-232	0.076	
Th-230	0.167	
Th-228	0.076	
Ra-226 0.00846		
Pb-210	0.00846	
Po-210	0.00846	

Table 5-1. Source term activity ratios relative to uranium (based on the

In order to account for the fact that only a portion of the internal exposure during the residual contamination period is from resuspended residual contamination versus exposure due to continued site operations, calculated intakes are adjusted by a weighting factor to account for the continued depletion of the pre-1977 source term during the residual period.

Decrease in particulate resuspension with time has been well documented in experimental studies in outdoor environments (Sehmel 1980, Till 1983). Measured resuspension factor "half-lives" in the range of 35 days to years have been reported (Sehmel 1980). Models for this effect have been proposed in the form of a constant (steady state component) and a second component with an exponential term. For example, Linsley reported an expression (Linsley 1978):

> K(t in days) =  $[10^{-6} \exp^{(-0.01t)} + 10^{-9}]m^{-1}$  with the  $10^{-6}$  factor being replaced by  $10^{-5}$ for periods of "regular disturbance by vehicular or pedestrian traffic."

Fewer data are available on the variation of resuspension factors with time in indoor environments. However, Healv recommends a decay constant value of 0.1 d<sup>-1</sup> which represents the effects of source depletion with time (Healy 1971). While no experimental studies were identified for indoor facilities, an exponential decrease in resuspension is expected to occur due to conservation of mass and the depletion of easily suspended contaminants.

Current project guidance (ORAUT-OTIB-0004) establishes a source term depletion factor of 1% of the surface activity per day. Use of this 1% depletion factor is favorable to claimants, based on the depletion behavior reported above, however, to account for the observed steady-state resuspension condition, this factor is held constant after 1979. The factors to account for depletion of the pre-1977 source term during the residual period are presented in Table 5-2.

> Table 5-2, Adjustment factors to account for depletion of pre 1977 source term during the residual contamination period.

Year	Factor
1977	1
1978	0.03
1979 - present	0.0007

Examples of the application of the Table 5-1 activity ratios and Table 5-2 depletion adjustment factors to estimate intake rates are presented in Attachment A.

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To estimate external exposure to residual activity, external dosimetry results that are available after 1976 are used to provide an upper bound of the dose. Since external dose is from both resuspended and fixed contamination, no adjustment to external dose due to depletion of resuspended material is necessary.

To summarize, the information in Table 5-3 (supplemented with the detailed guidance in Sections 3, 4, and 5) is used to account for dose during the residual exposure period.

	<u>г</u>	Period of	
Source	Exposure information	applicability	Annual exposure
Uranium	Internal (alpha,	Beginning	Inhalation intake based on
U-234	chronic)	January 1, 1977	individually calculated results adjusted by factors in Table 5-2.
Nonuranium	Internal (alpha,	Beginning	Inhalation intake for listed
Th-232	chronic)	January 1, 1977	nonuranium radionuclides based
Th-230	-	-	on the assigned uranium intake
Th-228			rate and the nonuranium ratio
Ra-226			values of Table 5-1.
Pb-210			
Po-210			
Uranium	External	Beginning	Use reported dosimetry results
contamination	Penetrating (photons,	January 1, 1977	
	30–250 keV, AP, acute)		
Uranium	External	Beginning	Use reported dosimetry results
contamination	Nonpenetrating	January 1, 1977	
	(electrons >15 keV,		
	acute)		

Table 5-3. Residual radiation exposure summary.

## 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.

- [1] Olsen, Bernard M. MJW Corporation. Senior Health Physicist. July 2007. The data for natural uranium are as presented in the bioassay application, Integrated Modules for Bioassay Analysis (IMBA).
- [2] Olsen, Bernard M. MJW Corporation. Senior Health Physicist. July 2007. This assumption results in estimated organ doses which are favorable to the claimant. The assumption that all uranium available for intake is <sup>234</sup>U will result in slightly higher estimates of organ doses than if natural uranium is assumed. For a dose reconstruction employing best estimate methodology, natural uranium can be assumed.
- [3] Olsen, Bernard M. MJW Corporation. Senior Health Physicist. July 2007. The activity fraction of <sup>234</sup>U and of <sup>238</sup>U (0.4882) in natural uranium are as presented in the bioassay application, Integrated Modules for Bioassay Analysis (IMBA). The specific activity

of <sup>234</sup> Th (2.32 E+10 pCi/µg) and half lives of radionuclides are listed in any standard handbook of radiation protection quantities, e.g. Shleien, B., 1992, *The Health Physics and Radiological Health Handbook*, revised edition, Scinta, Inc., Silver Spring, MD.

- [4] Olsen, Bernard M. MJW Corporation. Senior Health Physicist. July 2007. The increased sensitivity of the urinalysis methods used at ACCP over time is evident from the individual bioassay monitoring records of ACCP claimants. Results less than the level of detection for urinalysis results, in units of micrograms per liter (µg/L), are reported as < 10 µg/L through approximately mid 1974; < 5 µg/L from then through approximately late 1978; and < 2 µg/L after late1978.
- [5] Olsen, Bernard M. MJW Corporation. Senior Health Physicist. August 2007. Surveys of UF6 storage cylinders more enriched than those at Allied Chemical, and studies of work practices in moving those cylinders, have been made at the Portsmouth Gaseous Diffusion Plant, as described in ORAUT-2006d. It was estimated in ORAUT-2006d that a worker would take on average 3 hours/week, for 50 weeks in a year, to transport these cylinders, and that the average neutron dose rate to the worker would be that at approximately 1 meter from the cylinder surface. The upper bound neutron dose rate developed in Section 4.0 is 3.28E-04 rem/hr at 1 foot from the cylinder. At 3 feet from the cylinder, the neutron dose rate would be estimated at 3.28E-04 rem/hr [(1 ft)<sup>2</sup> / (3 ft)<sup>2</sup>], or 3.64E-05 rem/hr. The average neutron dose rate, applying the 150 hr/year occupancy methodology of ORAUT-2006d (3 hr/wk for 50 wk/yr), is 5.47E-03 rem/yr.
- [6] Guido, Joseph S. MJW Corporation. Senior Health Physicist. July 2007. The airborne effluent and ambient air data from Perkins 1982 was collected between 1976 and 1979 and as such are representative of the isotopic distribution at the start of the residual period.

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# ATTACHMENT A APPLICATION OF RESIDUAL DOSE FACTORS

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For the purpose of demonstration of the application of residual dose factors two example cases are provided. In all cases, the internal dose is first assessed using the worker's urine and lung count data (if available) during the entire employment period, or up to date of diagnosis. Once an appropriate set of intake regimes are determined, adjustments are made as follows:

- Break the intake regimes into the following time periods:
  - o pre 1977
  - o **1977**
  - o **1978**
  - o 1979 onward.
- Adjust post 1976 uranium intake rates using appropriate factor in Table 5-2 (based on intake year)
- Add intakes for uranium progeny based on ratios in Table 5-1 for post 1976 periods

#### Example 1.

A chronic uranium intake of 100 pCi/d is to be assigned from June 1, 1972 through March 25, 1983. This example is typical for assignment of missed dose only.

- Break this single chronic intake into appropriate time periods:

0	June 1, 1972 – December 31, 1976	100 pCi/d (Uranium)
0	January 1, 1977 – December 31, 1977	100 pCi/d (Uranium)
0	January 1, 1978 – December 31, 1978	100 pCi/d (Uranium)
0	January 1, 1979 – March 25, 1983	100 pCi/d (Uranium)

- Adjust the post 1976 uranium intake rates based on factors in Table 5-2

0	June 1, 1972 – December 31, 1976	100 pCi/d (Uranium)
0	January 1, 1977 – December 31, 1977	100 pCi/d (Uranium)
0	January 1, 1978 – December 31, 1978	3 pCi/d (Uranium)
0	January 1, 1979 – March 25, 1983	0.07 pCi/d (Uranium)

- Add intakes for uranium progeny for post 1976 intake regimes using appropriate ratios in Table 5-1.

#### ATTACHMENT A APPLICATION OF RESIDUAL DOSE FACTORS Page 2 of 3

Intake period	Radionuclide	Intake rate, pCi/day
June 1, 1972 – December 31, 1976	U-234	100
	U-234	100
	Th-232	7.6
	Th-230	16.7
January 1, 1977 – December 31, 1977	Th-228	7.6
	Ra-226	0.846
	Pb-210	0.846
	Po-210	0.846
	U-234	3
	Th-232	0.23
	Th-230	0.5
January 1, 1978 – December 31, 1978	Th-228	0.23
	Ra-226	0.025
	Pb-210	0.025
	Po-210	0.025
	U-234	0.07
	Th-232	0.0053
	Th-230	0.0117
January 1, 1979 – March 25, 1983	Th-228	0.0053
	Ra-226	0.0006
	Pb-210	0.0006
	Po-210	0.0006

### Example 2.

Acute uranium intakes are to be assigned as follows: 10,000 pCi on March 5, 1973; 100,000 pCi on January 15, 1977; and 100,000 pCi on February 15, 1978. Uranium intakes are assigned:

- March 15, 1973 10,000 pCi (Uranium)
- o January 15, 1977 100,000 pCi (Uranium)
- o February 15, 1978 100,000 pCi (Uranium)
- Adjust the post 1976 uranium intake rates based on factors in Table 5-2

0	March 15, 1973	10,000 pCi (Uranium)
0	January 15, 1977	100,000 pCi (Uranium)

- February 15, 1978 3,000 pCi (Uranium)
- Add intakes for uranium progeny for post 1976 intake regimes using appropriate ratios in Table 5-1.

# **ATTACHMENT A APPLICATION OF RESIDUAL DOSE FACTORS**

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Intake date	Radionuclide	Intake, pCi
March 15, 1973	U-234	10,000
	U-234	100,000
	Th-232	7,600
	Th-230	16,700
January 15, 1977	Th-228	7,600
	Ra-226	846
	Pb-210	846
	Po-210	846
	U-234	3,000
	Th-232	228
	Th-230	501
February 15, 1978	Th-228	228
	Ra-226	25
	Pb-210	25
	Po-210	25

Note that for this example, it is also necessary to calculate missed dose (as in Example 1) and perform a missed versus fitted comparison, in accordance with section 5.5.3.1 of ORAUT-OTIB-0060. This comparison should be performed based on the adjusted intakes (considering Table 5-1 and 5-2 adjustments, as appropriate).