



ORAU TEAM Dose Reconstruction Project for NIOSH

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ACRONYMS AND ABBREVIATIONS

ACCP	Allied Chemical Corporation Plant
ADU	ammonium diuranate
AEC	U.S. Atomic Energy Commission
cc	cubic centimeter
Ci	curie
DOE	U.S. Department of Energy
dpm	disintegrations per minute
ft	foot
hr	hour
keV	kilovolt-electron, 1,000 electron volts
kg	kilogram
L	liter
m	meter
MeV	mega-electron volts, 1,000,000 electron volts
ml	milliliter
mrem	millirem
nCi	nanocurie
NIOSH	National Institute for Occupational Safety and Health
NRC	Nuclear Regulatory Commission
pCi	picocurie
ppm	parts per million
U.S.C.	United States Code
WLM	working level month
y	year
α	alpha particle
γ	gamma
μ Ci	microcurie
μ g	microgram

1.0 INTRODUCTION

Technical Basis Documents and Site Profile Documents are general working documents that provide guidance concerning 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 the National Institute for Occupational Safety and Health (NIOSH) 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 facility" (AWE facility) or a "Department of Energy facility" as defined in the Energy Employees Occupational Illness Compensation Program Act of 2000 [EEOICPA; 42 U.S.C. Sections 7384l(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 radiation exposures must be included in dose reconstructions. 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/AEC-related work) must be included in dose reconstructions. 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.

2.0 SITE DESCRIPTION AND OPERATIONAL HISTORY

The information that follows applies to a period of AEC operations at Allied Chemical Corporation Plant (ACCP) located in Metropolis, Illinois from January 1, 1959, to December 31, 1976, involving AEC-contracted conversion of uranium ore concentrates to uranium hexafluoride. Opening ceremonies for Allied Chemical's fluorine plant at Metropolis, Illinois occurred in October 1958 (Sloop 1978). The original license issue date was December 17, 1958 (NRC 2003, p. 8-9). Four claims (numbers redacted) include bioassay results in December 1958, and one of these from December 15, 1958, has a positive result indicating that uranium exposures might have occurred in late 1958 prior to 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 indicates that some workers might have been laid off or transferred around this period, but that other workers were still onsite. This analysis assumed that the residual contamination period was from January 1, 1977, through the present.

Detailed information regarding 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 Allied Chemical Corp. Plant, indicate there was little variation in the processes over time. Because individual dosimetry results are available for the vast majority of workers at Allied Chemical, the reconstructed doses will be based on the specific individual dosimetry. To account for intakes of unmonitored radionuclides, bounding radionuclide fractions based on measurements in later years were used to estimate intake ratios of these radionuclides relative to uranium. Overtime, it is believed that the separations of uranium and associated radionuclides would have become more

efficient and that the radionuclide fractions derived from measurements in later years would overestimate exposures to non-uranium radionuclides in the earlier years.

The ACCP radiological source term started with the receipt of uranium ore concentrates. Some long-lived uranium progeny were included in the concentrates, e.g., thorium-230 and radium-226. Uranium chemical forms include oxides, fluorides and hexafluorides, which mean that exposures could have been to uranium solubility types F, M and/or S.

Allied Chemical Corporation Plant 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 Ill., 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 onsite in late 1958. One individual reported participating in opening ceremonies at Allied Chemical's fluorine plant at Metropolis, Illinois in October 1958 (Sloop 1978), indicating perhaps that uranium exposure could have occurred as early as October 1958.

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 Atomic Energy Commission contract (Perkins 1982). It was also noted that 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 Allied 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 dosimetry records show that a number of workers were onsite 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 still have been ongoing in the plant during the shutdown period, although based on DOL job descriptions associated with one claim (number redacted), some operations associated with processing might have continued at least on a sporadic basis.

Allied Chemical Corp.'s NRC license (NRC 2003) 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 still providing UF_6 to gaseous diffusion plants for use in the commercial sector. The 1982 building locations are shown in Figure 2-1. A description of the buildings is excerpted from Allied Chemical Corp.'s licensing documentation (NRC 2003, pp. 8-8 to 8-9). The list was originally numbered from 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 muds washing facility which removes fluorides and KOH from the potassium diuranate muds generated in the fluorination scrubber system. The

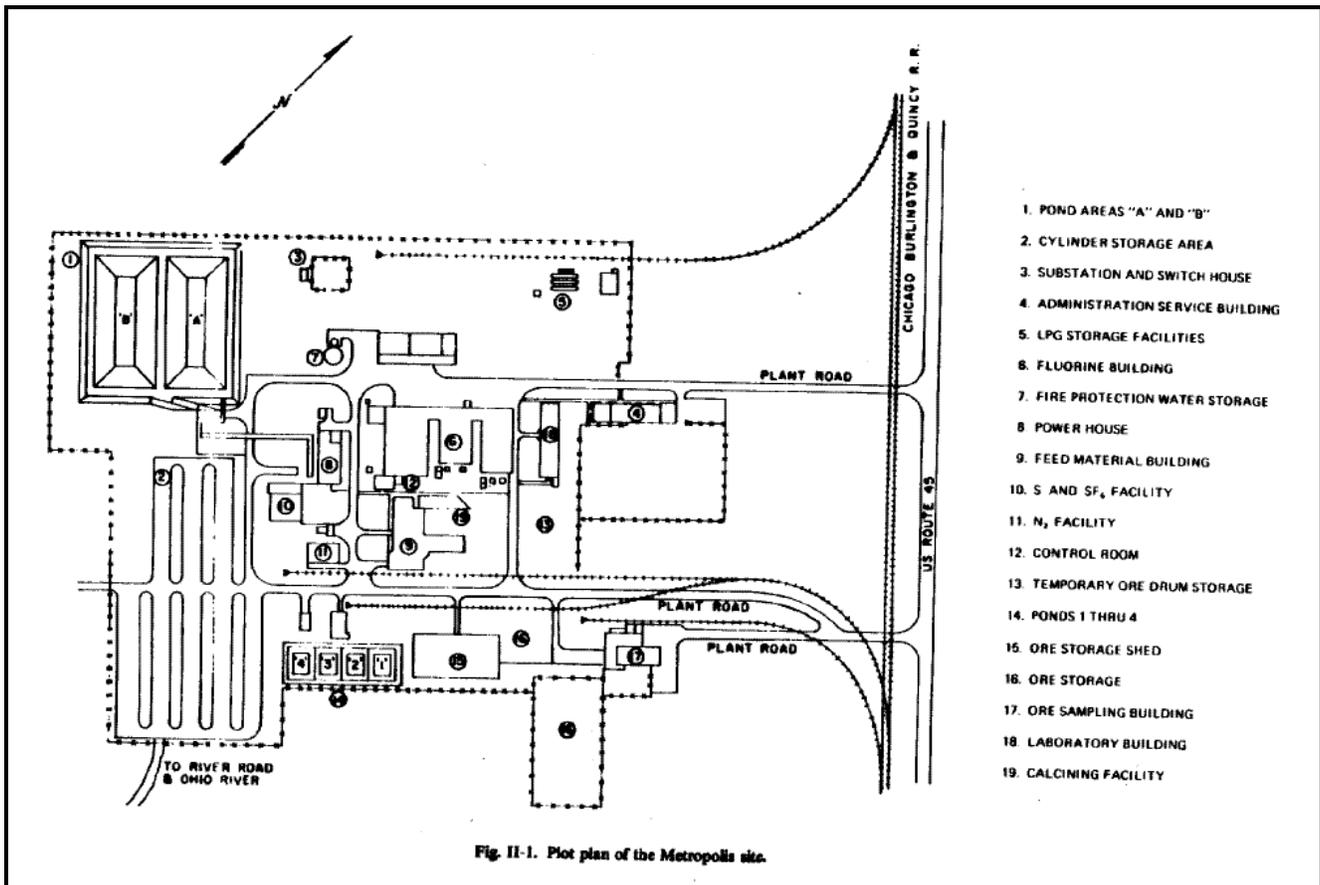


Figure 2-1. Plot plan of Allied Chemical Corp. Plant in Metropolis, IL (Perkins 1982, p. 3)

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].

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 UF₆.*
6. *The Laboratory Building which houses facilities for conducting process control, product, and radiological control analyses.*
7. *The Cylinder Wash Building where UF₆ 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 UF₆ product cylinders.*

Additional plant facilities which are involved directly in the UF₆ 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 CaF₂ settling tanks, a powerhouse, a reductor off-gas incinerator, and two small uranium settling ponds to collect any uranium spills.

Allied was also involved in the manufacture of 30,000 pounds per week of liquid fluorine, 1200 tons per year of sulfur hexafluoride, 2500 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 (yellow cake) to UF₆ 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 annually and the plant had processed over 100,000 tons of uranium from both domestic and foreign uranium concentrates (Perkins 1982).

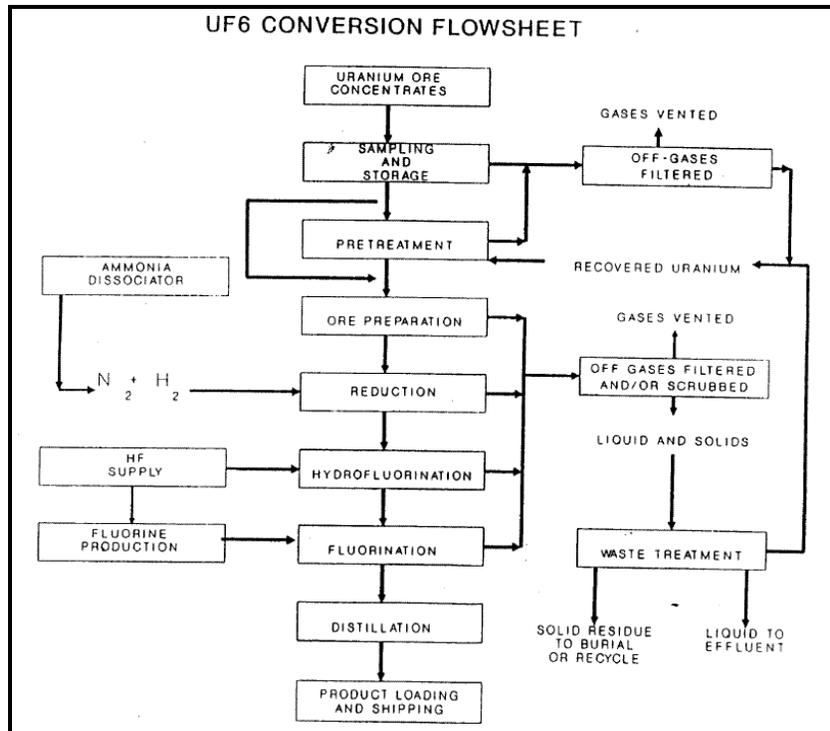


Figure 2-2. Process steps for conversion from ore concentrates (yellow cake) to UF₆ (NRC 2003)

As part of an evaluation of environmental control technologies for nuclear fuel conversion (UF₆) facilities, Perkins (1982) described the conversion of yellow cake to uranium hexafluoride at the Allied Chemical Corp. Plant. 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/filtration controls and waste disposal practices, and that the general process was similar throughout the plant's history. Process descriptions from Perkins (1982) follow.

Introduction

The Allied facility uses the fluoride volatility process to produce UF₆ 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 (U₃O₈) are reacted with hot cracked ammonia to form uranium tetrafluoride (UF₄) or green salt. Next, the green salt is combined at high temperature in a fluidized bed with gaseous fluorine to form the gas UF₆. Finally, the UF₆ 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

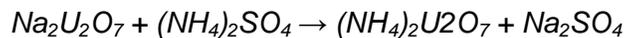
rainwater to drain to centralized collection sumps, which is then pumped to uranium spill control ponds.

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 UF₆ 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 (NH₄)₂U₂O₇ for further processing. The chemical reaction is



The liquid effluent contains the excess (NH₄)₂SO₄, Na₂SO₄, K₂SO₄, and uranium and small quantities of ²²⁶Ra and ²³⁰Th, 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 (NH₄)₂U₂O₇ (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_2 from reduction is fed into two reactor vessels in series, which use vaporized hydrofluoric acid and N_2 to fluidize the UO_2 and permit HF to react with the material to form UF_4 . The off-gases contain the excess HF needed to give good conversion to UF_4 (10% excess or more), volatile SiF_4 , BF_3 , 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_2S_4 . These gases first pass through two sets of porous carbon filters per set, where the unreacted UO_2 particles, entrained UF_4 particles, and any other particulate matter are removed. The gases then pass through two venturi water scrubbers where the H_2O 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. 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

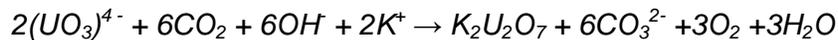
The green salt, UF_4 , produced in the hydrofluorination process is fed into one of two fluid bed reactors used in parallel. Again, Allied uses two trains at the works. The bed material is CaF_2 , and unreacted UF_4 . The vessels are operated at temperatures of

approximately 1000° F, and good temperature control is required. Fluorine, F₂, from an on-site fluorine production facility is introduced into the reactor vessels to convert UF₄ into gaseous UF₆.

The off-gasses, including uranium UF₆, F₂, HF, unreacted UF₄, 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 UF₆ removed by condensation in a set of three cold traps used in series. Any uncondensed UF₆, F₂, 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.



The K₂U₂O₇ 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 CaF₂ 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 UF₆ 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 UF₆ from the column is condensed into two cold traps operated in series. The UF₆ is transferred to the shipping cylinders by melting the UF₆ and allowing the material to drain into the cylinders.

Because impurities build up on the recycled CaF₂ 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 ²³⁴Th and ²³⁴Pa 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 UF₆ and the daughters of uranium that have "grown in" while the cylinders containing UF₆ 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 ^{234}Th and ^{234}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. Off-gases 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 compound, 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 where the yellow cake was derived.

Uranium chemical forms at Allied Chemical Corp. Plant 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, over 100,000 tons of uranium had been processed (Perkins 1982).

The initial milling of the uranium prior to arrival at Allied Chemical Corp. Plant resulted in disequilibrium of uranium progeny in the concentrate. Exposure to the non-uranium radionuclides occurs at the front end of the processing, beginning with receipt. Increased concentrations of other radionuclides relative to uranium are 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 Ra-226 and Th-230 relative 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) summarizes uranium, thorium and radium-226 activities in Allied Chemical effluents and wastes from the 1970s. These values, shown in Table 2-1, are 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 Th-230 to uranium in uranium ore would be about 0.4886; it can be seen that the milling of the ore usually reduces this ratio by almost a factor of 100, by removing the Th-230 from the concentrate. It should also be noted that Th-232 comprises a portion of the radioactivity in the uranium concentrates, and although Th-232 wasn't specifically reported for most effluents at Allied, the water effluents for 1975 and 1976 show annual average Th-232:Th-230 activity ratios of 0.076 and 0.017, respectively.

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

Source	Average			Largest ratio		
	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 (yellow cake feed averaged over 33 mills)		5.20E-03				
Uranium recovery solid waste (dried sludge)	3.58E+00	1.90E+01	1.11E+00			
Solid non-sludge waste		5.27E-03	2.75E-04			
Outdoor air		6.70E-02	1.97E-03		1.67E-01	8.46E-03
Air effluent		1.70E-02	2.70E-04		4.75E-02	3.38E-04
Water effluent		3.99E-03	2.04E-02		1.58E-02	5.79E-02

Processing at Allied Chemical Corp. Plant results in further separation of uranium and progeny, and this can result in increased activity ratios of non-uranium radionuclides in some areas in the plant. The largest disequilibrium ratios are associated with the solid waste (dried sludge) from the uranium recovery facility and these are used to derive a source term that would result in the largest internal dose. In reality, it is unlikely that any one worker would be consistently exposed to these disequilibrium ratios for an entire career. It is assumed that the activity of Th-228 is in equilibrium with Th-232. To derive the source term in Table 2-2, it is assumed that the short-lived progeny of internal dosimetric interest are in equilibrium with their closest long-lived parent.

Table 2-2. Source term activity ratios relative to uranium (based on the largest reported ratio relative to uranium in Table 2-1).

U-natural	1
Th-232	3.58
Th-230	19.0
Th-228	3.58
Ra-226	1.11
Pb-210	1.11
Po-210	1.11
Pa-231	0.887
Ac-227	0.887

AEC and U.S. Department of Energy (DOE) inventory reports use a designation of JSA to identify Allied Chemical Corp. Plant. DOE (Dalton 2000) reports that the Rocky Flats Plant shipped 71 kg (160 pounds) of depleted recycled uranium to Allied 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 Allied. A second mention of the same shipment indicates that the 71 kg were shipped to Allied Chemical Corp. in Idaho Falls, Idaho. Another reference (Author unknown c. 2000) referred to a shipment of 22,060 kg from Rocky Flats to Allied Chemical Corp. between 1955 and 1964, but this appears 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 Allied workers' radiation dose.

Allied 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 Allied's 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. 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 Allied Chemical. Drums, process vessels, lines, etc. are used to confine the materials. When unconfined uranium is handled, procedures call for ventilation controls to reduce exposures. (Allied c. 1985, p. 27).

An exception was requested from the NRC's requirement to post and label individual radioactive material areas and containers, and is replaced by worker training (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 Allied Chemical to assure 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 Allied 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 Visual Observation of Contamination

Allied noted that natural uranium compounds have a low specific activity, characterized by a high ratio of mass to activity. The Allied Chemical uranium compounds are highly colored, which provides an immediate method for detecting gross surface and air contamination. For instance yellow cake was yellow and UF₆ releases appear as white smoke due to the decomposition products, UO₂F₂ and HF. Procedures from 1985 stated that when employees detect small spills, they are responsible for cleanup. 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 even shutdown of the unit. The foremen are 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 make a daily inspection of uranium operating areas to ensure proper decontamination and contamination control.

2.4.2 Air Activity Measurement of Contamination

As of 1985, stationary breathing zone samples were collected daily from 57 stations in the uranium hexafluoride building, 10 in the sampling plant, 2 in the drum dumper, 3 in sodium removal and 2 in

the uranium recovery unit. Summaries of some of these measurements can be found in the available references. As of 1985, wearing of respirators was required when the average gross alpha air concentration on a floor exceeded $4 \text{ E-11 } \mu\text{Ci/cc}$ gross or when any 4 air samples on the floor exceeded that concentration. In addition, as of 1985, any single air sample result which exceeded $1 \text{ E-10 } \mu\text{Ci/cc}$ required investigation and preparation of a health physics incident report (Allied c. 1985).

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

2.4.3 Surface Contamination Measurements

Surface contamination measurements in uranium and non-uranium areas are made routinely. Removable contamination is determined by taking a wipe or smear of 100 cm^2 on a surface. Each smear is counted for alpha radioactivity and reported in $\text{dpm}/100 \text{ cm}^2$ removable alpha contamination. The plant lunch room 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 non-uranium processing areas are surveyed quarterly.

As of 1985, decontamination is to be initiated when removable alpha contamination exceeds $5000 \text{ dpm}/100 \text{ cm}^2$ in the uranium processing areas. All other areas are to be decontaminated when removable alpha contamination exceeds $200 \text{ dpm}/100 \text{ cm}^2$.

One summary of Health Physics data (Wilkins 1992) for the last half of 1991, noted that 1.7% of the 2,002 weekly smears exceeded the weekly limit of $200 \text{ dpm}/100 \text{ cm}^2$, and the highest result was $923 \text{ dpm}/100 \text{ cm}^2$ in the lunch room 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. Generally these procedures include roping off the area, sweeping up or vacuuming up the contaminated material and then washing off the contaminated surface. During these procedures protective clothing are to be used and monitoring is to be performed.

2.5 INCIDENTS

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

2.6 PHYSICAL EXAMINATIONS – X-RAYS

No documented Allied Chemical 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 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.

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

Because bioassay data 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 important.

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 Allied Chemical Corp. Plant 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

The primary source of internal radiation exposure at Allied Chemical Corp. Plant was uranium dust produced from the processing of uranium concentrates to produce uranium hexafluoride. It is assumed that the uranium was of natural enrichment prior to 1977, although there is some indication that Allied's source term included a small amount of depleted uranium. The uranium ore concentrate also contained uranium progeny of dosimetric interest as well as thorium (assumed to be Th-232 and Th-228).

3.1 URANIUM

During the processing of uranium concentrates at Allied, 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 may 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 shown in Table 3-1. For organ dose estimates, the uranium activity can be assumed to be from U-234. A conversion factor is also provided to adjust results reported as μg of U-235 to total uranium activity in pCi.

Table 3-1. Natural uranium's specific activity and isotopic fractions.

	Activity Fraction	Mass Fraction	Activity ratio to U-235	Mass ratio to U-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 data began as early as December 1958 and was done in house. It appears that most workers will have bioassay data. The claimant file records show that bioassays

were typically collected at hire and periodically thereafter. Allied notes that fluorimetry was used for analyses until 2002, when kinetic phosphorescence analysis (KPA) began. Urinalysis data for the period 1989 to 1992 are missing, and Allied 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 units of microcuries per milliliter ($\mu\text{Ci/ml}$) are misstated, and the units are actually in units of micrograms per liter ($\mu\text{g/L}$). The recording limit of 10 $\mu\text{g/L}$ is found in the early records and is consistent with the default detection threshold for uranium urinalysis assumed to be 10 $\mu\text{g/L}$ based on a reported sensitivity of 5 to 10 $\mu\text{g/L}$ for uranium fluorimetry urinalysis in the early years (Wilson 1958). Beginning in late 1974, a recording limit of 5 $\mu\text{g/L}$ appears to have been in use. Allied reported that their MDL was 2 $\mu\text{g/L}$ in the early 1980s, but no records of MDL are available prior to that. In 1985, Allied Chemical Corp. Plant noted that routine urinalysis samples were to be left following 24 to 96 hours of absence from the workplace, but it is unknown when this procedure was first implemented. Also, as of 1985, Allied employees were being encouraged to leave special urine samples prior to their next work shift in response to known or suspected airborne exposures (NRC c. 1993, pdf p. 50). In later years, it was noted that a result equal to 30 $\mu\text{g/L}$ was cause for investigation.

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

3.1.2 Uranium In Vivo Data

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 may be that only some workers were required to have annual lung counts in later years.

Allied 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, based on the Oak Ridge National Laboratory program, ALPHA-M. The counting system includes an RMC-designed shadow shield and four detectors, which appear to consist of thin NaI(Tl), and thicker CsI(Na) crystals. The dimensions are reported as 5 feet in diameter, but are probably 5 inches in diameter. Prior to analysis of the spectra, the background is automatically subtracted. A REMCAL standard man equivalent phantom was used to calibrate the system. Radionuclides include U-235, Th-234, Cs-137 and K-40.

Specific Allied criteria are given for judging the data and can be used for considering the relative validity of a result:

- 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 may be need if the value is outside this range.

The analysis program reportedly prints a table showing all radionuclides found and their estimated activities and relative two standard deviation counting error in terms of a percent error. Allied reports

that results of U-235 are given in μg and results for other radionuclides are given in nanocuries; however, some records show Th-234 results in μg . Results with errors greater than 100% (results less than two times their counting standard deviation) are considered questionable. The reported U-235 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 U-235 or summed as 26,974 μg of total uranium (Allied 1985). 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 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 U-235 mass to total uranium activity, it is assumed that the uranium is of natural enrichment, and so the U-235 mass in μg is multiplied by 2.16 pCi/ μg (the specific activity of U-235) and divided by 0.0228 (the activity fraction of U-235), a total factor of 94.8, to obtain the total uranium activity in pCi.

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

The K-40 and Cs-137 reported in lung counts are not likely to be associated with workplace exposures, and can usually be ignored for the purpose of dose reconstruction.

A handwritten note (Honeywell c. 1998, pdf page 2) indicates that other whole body counting systems might have been in use as of 1996, but there were no differences in reported sensitivity.

3.2 OTHER RADIONUCLIDES

Bioassays for non-uranium radionuclides do not appear to be in the claim files. To account for intakes of other radionuclides (except radon) handled incidentally to the uranium processing, the ratios relative to uranium activity in Table 3-2 can be used to estimate upper bound intakes relative to uranium. These ratios are reported for process streams where non-uranium radionuclides are concentrated. It is unlikely that any worker was exposed to this source term for their entire Allied Chemical Corp. Plant career, but these numbers represent a bounding estimate of intakes relative to uranium.

Table 3-2. Assumed types and ratios for intakes of other radionuclides relative to uranium intake activity.

Radionuclide	Type	Activity ratio relative to uranium
Th-232	M, S	3.58
Th-230	M, S	19.0
Th-228	M, S	3.58
Ra-226	M	1.11
Pb-210	F	1.11
Po-210	M	1.11
Pa-231	M	0.887
Ac-227	F, M	0.887

3.3 RADON

No radon air concentration data have been found to date. Radon exposures were probably most significant in the receiving, sampling and feed areas and in the waste storage area, where radium source terms would have been more significant. It can be reasonably inferred that the radon concentrations would not have been higher than the high, but not atypical, concentrations during processing at Linde and Mallinckrodt, where African ores were being wet-processed in the 1940s and 1950s. A concentration of 100 pCi/L, with a fraction of equilibrium of 0.4 would equate to 4.8 working level months per year (WLM/y).

3.4 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 N.J.]
- MPLB [maximum permissible lung burden]
- Sample type R [assumed to mean routine]
- Sample type S [assumed to mean special]

3.5 OCCUPATIONAL INTERNAL DOSE RECONSTRUCTION ASSUMPTIONS AND SUMMARY

The assumed operational exposure period ran from January 1, 1959, to December 31, 1976.

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 Allied Chemical Corp. Plant was 10 µg/L. Uranium may be either absorption type F, M or S. For dose calculations, uranium intakes can be assumed to be U-234.

To estimate uranium intakes from the lung counts, the intake of U-235 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 U-235 and 5970 µg for total uranium.

Intakes of other radionuclides are accounted for by calculating the uranium intake(s) from the uranium urinalysis, and then applying the fractions in Table 3-2 to estimate intakes of other radionuclides. The dose distributions are assumed to be constant for the other radionuclides.

A high estimate (constant distribution) of radon exposure is 4.8 WLM/y.

Based on the current information, all workers were monitored, 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 Allied Chemical Workers beginning January 1, 1959, and include results for gamma, beta and neutron exposures. Allied Chemical reports that the data prior to 1970 is 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 TLD dosimetry services. Neither Landauer nor Eberline reported doses below 10 mrem.

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 2004).

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 claimant favorable. 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, and
- 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 Allied Chemical Corp. Plant external dosimetry results are dated January 1, 1959. Badge exchange frequencies at Allied 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 neutron doses are available. Most of the results appear to be zeroes and it is not clear if these represent an actual dosimeter result or are just written because there was no result. The Bridgeport Brass site profile (ORAUT 2005) notes that the minimum detectable neutron dose reported in the early 1960s was 60 mrem.

A quick and probably high estimate of neutron dose is estimated based on a 500-pound drum containing 75% by mass of natural uranium. It is assumed that the alpha particles collide with a fluorine target. Using the method described in ORAUT 2005, the neutron dose calculated at one foot

from a drum is 3.28E-04 rem/hr. A worker, who is one foot from such a drum for 2000 hours per year, would be exposed to a dose rate of 6.56E-01 rem/y. This document considers this estimated neutron dose rate to be an upper bound of the Allied neutron exposure rate.

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 of X-ray should be based on current ORAU Team guidance. Organ doses can be obtained from the current revision of ORAUT-OTIB-0006, *Technical Information Bulletin: Dose Reconstruction from Occupationally Related Diagnostic X-Ray Procedures* (ORAUT 2003).

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 result in elevated beta and gamma radiation fields in the vicinity of the ash. Workers handling this material or washing 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 assumption regarding occupational external doses during the covered period January 1, 1959 to December 31, 1976 at Allied Chemical Corp. Plant.

Table 4-1. External exposure summary.

Exposure mode	Exposure type	Exposure or dose rate	Basis	Exposure time assumption	Annual exposure	IREP distribution
Uranium processing	Penetrating	-	-	-	Based on individual dosimetry	-
	Non-penetrating	-	-	-	Based on individual dosimetry	-
	Neutron	0.328 mrem/hr	Calculation	2000 work- hr/y	6.56E-01 rem	Constant
Medical X-ray		Initial plus one exam per year	-	-	See ORAUT-OTIB-0006, (ORAUT 2003)	

- Use generic external dose reconstruction guidance, when summary information is unavailable.

5.0 ESTIMATION OF EXPOSURE FROM RESIDUAL RADIOACTIVITY

Allied Chemical Corp. Plant continued to operate after the AWE covered period ended on December 31, 1976. NIOSH (2004) noted that a significant potential for residual contamination remained after 1976. It is not feasible to estimate exposures to residual activity from workplace measurements during continued operations; however, it is reasonable to estimate internal intakes due to residual contamination after 1976 by assuming that up to one additional year of intake occurred during the residual period as shown below.

During operations with uranium, airborne contamination can be caused directly from operations with uranium bearing materials as well as by the resuspension of surface contamination. This airborne

contamination can be removed by settling on floors or other surfaces, or by ventilation (both engineered and natural building ventilation). The surface contamination levels can be increased directly by radioactive material processing/handling or by the settling of airborne contamination. This level is decreased by various incidental means such as general housekeeping or tracking of contamination out of the area on equipment or personnel.

To estimate the airborne contamination from residual contamination, it was assumed that during uranium operations, these factors reached an equilibrium that caused the airborne concentration and surface contamination to remain at a constant level. Once the contamination-generating source, the uranium recovery operation, was removed, the other factors then caused the mass of uranium to decrease at some rate due to the remaining removal factors.

The analysis of residual intakes assumes that 1) uranium operations directly accounted for half the airborne concentration in the facility during operations, (i.e., half of the previously calculated airborne exposure is assumed to come from resuspended materials) and 2) all the remaining removal mechanisms eliminated only 1% of the uranium mass per day. With these assumptions in place, an equation can be written for the airborne concentration as a function of time.

$$C(t) = 0.5C(0) \times e^{-0.01t}$$

where:

- $C(t)$ = Airborne activity at time t (pCi/m³)
- t = time since uranium operations ended (days)
- $C(0)$ = Concentration during operations (dpm/m³)
- 0.01 = fraction of material removed per workday

When this is multiplied by the breathing rate of 9.6 m³ per 8-hour workday, a daily intake is calculated. This equation can then be integrated from the end of uranium operations to infinity to determine the total amount of uranium that could be inhaled from residual contamination. This integration results in:

$$I = 9.6 \times 0.5 \times C(0) / 0.01$$

where:

- I = total intake from inhalation of residual activity (dpm)
- 9.6 = breathing rate (m³/8-h workday)

A total uranium inhalation intake of 480 times the air concentration was assumed to occur during the residual contamination period. This compares to an estimate of annual intake during the operational period of 2,400 m³/y of air breathed times the initial concentration, which would result in an intake 5 times larger. This indicates that an estimate of inhalation intake during the residual period will be accounted for by assuming one additional year of intake at the calculated intake rate prior to 1977 for the individual. This assumed inhalation intake from residual contamination is sufficiently large to account for dose from ingestion intake.

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, and this is assumed to be a constant distribution.

Neutron doses from residual contamination are assumed to be insignificant.

To summarize, the following information in Table 5-1 is used to account for dose during the residual exposure period.

Table 5-1. Residual radiation exposure summary.

Source	Exposure information	Period of applicability	Annual exposure	IREP distribution
Uranium and progeny contamination	-	Beginning January 1, 1977	Add up to one additional year of inhalation intake based on individually calculated results	Constant
Uranium contamination	Penetrating (Photons, 30-250 keV, AP, acute)	Beginning January 1, 1977	Use reported dosimetry results	Constant
Uranium contamination	Nonpenetrating (Electrons >15 keV, acute)	Beginning January 1, 1977	Use reported dosimetry results	Constant

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