ORAU Team Dose Reconstruction Project for NIOSH Basis for Development of an Exposure Matrix for the Mallinckrodt Chemical Company St. Louis Downtown Site and the St Louis Airport Site, St. Louis, Missouri, Period of Operation: 1942–1958	Document Number: ORAUT-TKBS-0005 Effective Date: 03/10/2005 Revision No.: 02-A Controlled Copy No.: ________ Page 1 of 240
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RECORD OF ISSUE/REVISIONS

ISSUE AUTHORIZATION DATE	EFFECTIVE DATE	REV. NO.	DESCRIPTION
Draft	08/18/2003	00-A	New document to establish the technical basis for the development of a radiation exposure matrix for the Mallinckrodt Chemical Company St. Louis Downtown Site, St. Louis, Missouri. Initiated by Janet L. Westbrook.
Draft	10/17/2003	00-B	Draft revision to incorporate NIOSH review comments. Initiated by Janet L. Westbrook.
10/24/2003	10/24/2003	00	First approved issue. Initiated by Janet L. Westbrook.
Draft	03/04/2004	01-A	Revision to incorporate comments on Section 8.0 and to incorporate additional lately captured information. Initiated by Janet L. Westbrook.
Draft	02/16/2005	01-B	Revision to incorporate comments on Sections 7.0 and 8.0; to incorporate additional lately captured information regarding site work and external and radon exposure rates; to incorporate neutron exposure information; to incorporate information about the St. Louis Airport Site (SLAPS); and to provide additional guidance to dose reconstructors. Section 7.0, regarding external dose, remains on hold. Initiated by Janet L. Westbrook.
Draft	03/08/2005	01-C	Incorporates NIOSH comments. Initiated by Janet L. Westbrook.
03/10/2005	03/10/2005	01	Approved Issue of Revision 01. Initiated by Janet L. Westbrook.
Draft	8/16/2005	02-A	Initiated revision to reflect comments of the ABRWH

ACRONYMS AND ABBREVIATIONS

AEC	Atomic Energy Commission
AP	anterior-to-posterior
Ci/L	curies per liter
D&D	decontamination and decommissioning
DCF	dose conversion factor
DWE levels	time-weighted daily average exposure levels
EEOICPA	Energy Employees Occupational Illness Compensation Program Act
FMFL	fluorinated MFL
HASL	Health and Safety Laboratory (of the Atomic Energy Commission)
HVL	Half-value layer
LOD	limit of detection
MAC	Maximum Allowable Concentration
ME or MEP	Minor Elements Production
MED	Manhattan Engineer District
MFL	MgF_2 liner
MgX	magnesia concentrate feed
MPC	maximum permissible concentration
mR	milliroentgen
mrad	millirad
mrep	millirep
NBS	National Bureau of Standards
NCRP	National Council on Radiation Protection and Measurements
NRC	Nuclear Regulatory Commission
NYOO	New York Operations Office (of the Atomic Energy Commission)
ORGDP	Oak Ridge Gaseous Diffusion Plant
RBE	relative biological effectiveness
RMF	reject MgF_2 material
ROT	rotational
SLAPS or SLAPSS	St. Louis Airport (Storage) Site
SLDS	St. Louis Downtown Site
UNH	uranyl nitrate hexahydrate
VLE	very low enrichment
WL	working level
WLM	working level month

Code terms for the various uranium forms (e.g., TA-7) are given in Table 5 in Attachment A.

1.0 PURPOSE AND SCOPE

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 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” or a “ Department of Energy facility” as defined in the Energy Employee Occupational Illness Compensation Program Act of 2000 (42 U.S.C. § 7384l (5) and (12)).

This technical basis is established for the reconstruction of radiation doses to workers at the Mallinckrodt Chemical Company St. Louis Downtown Site, which refined uranium under contract to the United States government from 1942 to 1958, and for the St. Louis Airport Storage Site, which was received residues from Mallinckrodt operations from 1946 until 1958, after which it was used until about 1967 for residue storage and burial of some material. The two principal purposes of this technical basis document are (1) to provide information sufficient to enable dose reconstructors to estimate claimant-favorable doses for these workers on an individual basis under the provisions of the Energy Employees Occupational Illness Compensation Program Act (EEOICPA) and (2) to allow claimants, federal assessors, and others to understand the information sources and assumptions on which the dose estimations are based.

This document describes exposure conditions in Mallinckrodt Plants 1, 2, 4, 6, 6E, and 7 (including 7E) over the time period of the start of contract operations for the Atomic Energy Commission (and its predecessor agency) through the cessation of these operations, which is different for each plant. The whole period of such operations covers April 1942 through July 1958. Additionally, exposure due to residual contamination left in these plants during and after decontamination and decommissioning is considered for the period 1959-1995 for workers who began work at Mallinckrodt during the period of operations.

This document also describes exosure conditions for Mallinckrodt workers who performed operations at the St. Louis Airport (Storage) Site (SLAPS or SLAPSS), the waste repository site north of the former St. Louis Municipal Airport in Robertson, Missouri, from 1946 to 1958. Additionally, exposure due to residual contamination left at the site and during further work at the site is considered for the period 1959-1962.

In light of the addition of those workers employed at the Mallinckrodt Destrehen Street Facility, from 1942-1948, to the Speical Exposure Cohort, this document only applies to the reconstruction of doses for workers employed at this facility after 1948.

2.0 INTRODUCTION

The Manhattan Engineer District (MED), the predecessor agency of the Atomic Energy Commission (AEC), asked the Mallinckrodt Chemical Works in April 1942 to begin research on uranium refining and processing operations to lead to large-scale uranium production operations (Fleishman-Hilliard 1967; FUSRAP undated b). The work began immediately and by July 1942 Mallinckrodt was producing almost a ton of UO_2 per day (Fleishman-Hillliard 1967; Mallinckrodt 1994; FUSRAP undated b). Later in 1942, Mallinckrodt started production of UF_4 and in July 1943, it started the first uranium metal plant (Fleishman-Hillliard 1967; Mallinckrodt 1994). It is estimated that between 1942 and 1957, Mallinckrodt processed more than 50,000 tons of natural uranium products at the St. Louis facilities (FUSRAP 2002).

The first three Mallinckrodt plants (1, 2, and 4) were not built for the purpose of uranium refining but were converted on an urgent basis from other uses and were intended to operate for only six to eight months. Like most of the early MED plants, the early Mallinckrodt facilities were large-scale expansions of laboratory processes and techniques and were designed without pilot plant studies or any special provision for health and safety (AEC 1951b). Also, temporary types of construction methods were used; even Plant 6, which was completed in mid-1946 and did have the benefit of an engineering pilot plant study, was of temporary-type construction because it was to process only high-grade pitchblende and the available supply was thought to be limited (AEC 1951b). The permanent status of the Mallinckrodt site was not established until 1946 (AEC 1951b; Mason 1958a). Engineered and administrative provisions for health and safety were instituted as high exposures were found in the course of monitoring (Hickey and Dupree 1984). A full-scale health program was not authorized until 1947 and did not get under way until 1948, as a joint AEC-Mallinckrodt effort (Mason 1958a). Film badging started only in late 1945 (except possibly for a pilot project starting in mid-1945) and urinalysis in the summer of 1948 (MCW 1950c) (some urinalysis was done earlier, but there was some question about its validity – see Section 5.3.6). Thus there is little data on radioactivity concentrations in air or on internal and external doses prior to the late 1940's.

Although Mallinckrodt had its own industrial hygiene group and did most of its own safety work (Eisenbud 1975; MCW 1955d; AEC 1950a), AEC's Health and Safety Laboratory (HASL) also did periodic air sampling and other surveys for the operating plants. While some records of the Mallinckrodt surveys survive, they essentially cover the same work and time periods as AEC's surveys, except for some special cases. In this document preferential use is made of the AEC air concentration data because AEC developed and used the standard survey methods. During the early years, film badge servicing, urinalysis, and breath radon analyses were done under the aegis of HASL (AEC-NYOO) (Eisenbud 1975); later, Mallinckrodt took over this work for its own plants and created the corresponding records. Eventually, however, HASL resumed doing the urinalysis and breath radon analysis for a time (MCW 1955d), but eventually Mallinckrodt did its own urinalyses again (ORAU 1983b). It appears that most of the records of this monitoring are available. The external dose records needed for dose reconstruction are mostly available from 1946 on, but are missing for the period 1942-1944 and for most of 1945, and are also missing for the period of some time in 1954 to the end of operations; the internal dose records (as bioassays) are mostly available from 1948 to the end of operations, but are missing for the period 1942-1947. Records for the postoperations decontamination and decommissioning should be found with the Weldon Spring records since film badges were issued from Weldon Spring (MCW 1961b), but to date the Weldon Spring records are not available except by request for individual claim subjects.

The St. Louis Airport Storage Site (SLAPS or SLAPSS) was acquired by AEC in 1946 for use in storing residues arising from pitchblende ore processing (AEC 1959; ORNL 1979). Later, it was also used to store other types of residues and some ore (AEC 1956d; Mallinckrodt 1994). Although

MED/AEC operated the site itself from 1946 until July 1953, it appears that Mallinckrodt and perhaps some Ledoux Company workers actually did the work (MCW 1949d; MCW 1949p; ORAU 1989); Mallinckrodt ran the site under contract to AEC from July 1953 to 1958 and possibly until 1962, when control of the site passed to a private concern that was given a license to take over the stored residues (AEC 1959; ORAU 1989; AEC 1972). Thus the film badge records for airport workers from 1946 until at least 1958 should be found among the Mallinckrodt film badge records; film badge records after that may possibly be among the Weldon Spring records.

In this document, the context for interpretation of the existing records is established, along with the basis on which to determine missing doses for periods in which records do not exist.

3.0 HISTORY OF SITE USE

At their height, uranium-refining operations took place in about 60 buildings on the so-called St. Louis Downtown Site (SLDS), of which about 20 were left after decontamination (Applied Nuclear Safety 1986; FUSRAP 2003a). All of these 20 buildings and most of the other 40 buildings are listed in Table 1 along with their principal uses. (See Attachment A for all tables). A summary chronology of the use of the site is shown in Table 2.

MED was required to store some of the residues produced from the pitchblende ore (AEC 1959) because the ore supplier, the African Metals Company, retained ownership of the radium and valuable metals remaining in the residues. Because of the volume of the residues and the high gamma and radon exposures associated with storage at the Mallinckrodt site, MED acquired a tract of land located in Robertson, Missouri near the St. Louis Municipal Airport (Labert Field) (AEC 1959) to use to store the residues and some of the pitchblende ore (AEC 1959). This storage area was formally called the St. Louis Airport Storage Site (SLAPS or SLAPSS), but in Mallinckrodt and MED/AEC documents it was often referred to simply as “the airport”.

3.1 HISTORY OF USE OF THE ST. LOUIS DOWNTOWN SITE (SLDS)

Plants 1 and 2 were already in existence and used for other operations by Mallinckrodt at the time uranium-refining operations started in 1942 (Mason 1977). MED (1944p) states that as of November 1944, MED operations occupied a small part of Plant 1, part of Plant 2, and all of Plant 4.

Laboratory development work started in April 1942 in Plant 1’s Building 25 (second floor) and in the alley on the southwest side of the building (Mason 1977). (Mallinckrodt 1994 states that work began in Plant 2 in uranium-refining operations began at Plant 2 in April 1942, but the Mason 1977 chronology appears to be more plausible.) Additional laboratory and developmental work at the laboratory level to support Plant 2 and Plant 4 took place in Plant 1, specifically the Building 25 laboratory and the alley between Buildings K1E and 25 (Mallinckrodt 1994).

In Plant 1, Building 25 contained the project offices, while Building P contained the engineering office. MED (1944p) states that Building Z contained other offices; Mason (1977) specified that the early uranium project was a semi-works operation for which administrative services were furnished from Mallinckrodt headquarters in Building Z . Building A in Plant 1 was the base for the plant mechanical department, which provided general maintenance services (Mason 1977; MED 1944p). Mason (1977) states that the laboratory in Building 25-2W provided quality control services.

Facilities for batch production were installed in Plant 2’s Buildings 50, 51, 51A, and 52 to produce uranium trioxide (UO_3) from ore concentrates (Mason 1977; Hickey and Dupree 1984; Mallinckrodt 1994). The concentrates were “digested” (dissolved) in nitric acid in Building 51 to produce uranyl nitrate (Mason 1977; ORNL 1981; Mallinckrodt 1994; Fleishman-Hilliard 1967). The digested liquid was then transferred to Building 52 to be purified by ether extraction to uranyl nitrate hexahydrate (UNH) (MED 1944p; Mason 1977; ORNL 1981; Mallinckrodt 1994); however, sources such as ORNL (1981) suggest that an area outside of Buildings 51 and 52, referred to as 51X and 52X, was covered with a canopy and used temporarily for extraction. The UNH was converted in Building 51A first to UO_3 and then to UO_2 (Mason 1977; ORNL 1981; Mallinckrodt 1994). The UO_2 was apparently packaged in Building 51A (Mason 1977). By July 1942 Mallinckrodt was producing approximately a ton of uranium oxide (UO_2) per day (Mason 1977; Mallinckrodt 1994).

Part of Building 38 was used as the change house for the uranium project workers (Mason 1977). Buildings 40, 45, 45A, and 47 were used as warehouse buildings for raw, in-process, and finished materials (MED 1944p). Building 50 was also used as a utility area, as a tank storage area for liquids to be used in Building 51, as a storage area for incoming feed materials and packaged products, and as a mechanical repair area (Mason 1977; Mallinckrodt 1994).

Building 55 contained the “shotgun” laboratory that tested UO_3 samples (MED 1944l; Mason 1977; ORNL 1981) and also operated as a pilot plant from 1943 until some time in 1947. The shotgun laboratory was designated as a special restricted area because of the RaBe source used for the sample testing (Mason 1977).

Work began in 1942 on production of UF_4 by the high-temperature gas-solid reaction of UO_2 with HF; for a short time, one corner of Building 50 (the “sulfur burner room”) was used for development work on this process (Mason 1977; ORNL 1981).

Experimental processing of high-grade pitchblende ores began in Plant 1 in about July 1944 (Mason 1977; Mallinckrodt 1994). Because of the high radium content of these ores and the residues resulting from the processing, Mallinckrodt wanted to confine the materials to a limited area and to prevent their spread into other plant areas (Mason 1977). Mallinckrodt therefore instituted strict controls of the developmental work and also decided to do pilot-scale work to obtain design data for eventual construction of a production-level refinery (Mason 1977). The laboratory for the testing of radium extraction methods was set up in Building 25-2 and the pilot plant in Building K-1E (Mallinckrodt 1994; ORNL 1981; Hickey and Dupree 1984 (which states that the pilot plant was in Building 25-2)) and in the alley (Fleishman-Hilliard 1967). The radium-free feed liquor that resulted from the Building K-1E digestion and precipitation processes was conveyed in containers by hand cart to Building 52, where it was first treated in vessels outside the building (presumably in the alley) (Mason 1977). The liquid was then taken into Building 52 for conversion as usual to UO_3 and then to UO_2 (Mason 1977). By the end of 1944, Mallinckrodt had obtained sufficient data to begin the engineering design of a pitchblende ore refinery (Mason 1977). Mason (1977) states that “radium-bearing materials were not processed or stored at any time in the Plant 2 buildings”, but consistent with the usage of the term “radium-bearing materials” in various documents from the 1940’s and 1950’s, it may be the high-radium-level residues that are being referred to. Mason (1977) also states that solid residues from the extraction were drummed, stored temporarily, and transferred from Plant 2 to other government sites for scrap recovery.

In the spring of 1945, an annex to Building 52 (presumably 52A) was added to serve as a pilot plant for a continuous countercurrent ether extraction process to replace the existing batch process (Fleishman-Hilliard 1967). It is not clear where it is this plant or the Building K-1E plant that is the one referred to as the “Pilot Plant” in early film badge records; perhaps the term included both. Work at Plant 2 continued until 1946, when the plant was closed in preparation for the startup of the newly built Plant 6 (Mason 1958a; Mason 1977; Eisenbud 1975; ORNL 1981; Mallinckrodt 1994). Mason (1977) also states that when Plant 6 started up, all uranium work at Plants 1 and 2 stopped and the buildings were sealed to await further cleanup (but see below).

Plant 1 was not used after about 1945 (ORNL 1981) or early 1946 (Mason 1977); the offices and laboratories apparently moved to Plant 6. In particular, administrative and related services moved from Building Z to Plant 6 at this time; however, the quality control services moved to a laboratory in Building 400 of Plant 4 (Mason 1977). Although as noted above the Plant 2 operating area was said to have been closed in 1946 when production moved to Plant 6, Mason (1958a) states that milling of UO_3 and unloading of (some) pots was not discontinued at Building 51 (Plant 2) until about 1947 and ORAU (1977) states that this building was closed about January 1947. Thus it can conservatively be assumed that Plant 2 was in limited operation through 1946.

From late November 1942 through April 1943, Plant 4, a lumber sash and door works, was hastily converted for uranium processing (AEC 1951b; Mason 1958a) and dubbed “the metal plant” (AEC 1949b; AEC 1951b). It replaced the uranium metal production work done in Building 25-2 (Hickey and Dupree 1984). Production of uranium tetrafluoride (UF_4) began in Building 400 in April 1943 after the process had been scaled up from the early work in Building 50 of Plant 2 (AEC 1951b; Mason 1977; Fleishman-Hilliard 1967). Production of uranium metal began in Buildings 400 and 401B in July 1943 (AEC 1951b; Mason 1977; Mallinckrodt 1994). Magnesium metal for the process was stored in Building 406, while the residues from the process (e.g., slag) were stored in Building 408 (Mason 1977). The principal production of metal moved from Plant 4 to Plant 6E in 1950 (Mallinckrodt 1994) although some metal was still produced in Plant 4 (AEC 1951b; Hickey and Dupree 1984) and some UF4 was still produced there until perhaps early 1953 (Hickey and Dupree 1984). After the development of a continuous furnace process, UO_2-to-UF_4 production moved to Building 705 of Plant 7 (7N) in late 1951 (overlapping Plant 4’s production) (AEC 1951b; Fleishman-Hilliard 1967; Mason 1977 (which says that Plant 4 UF_4 production was completely shut down in 1951); Hickey and Dupree 1984; Mallinckrodt 1994). In about 1950 or 1951 (ORNL 1981 says 1956, but this is evidently incorrect), Plant 4 was refitted as an experimental development and a metallurgical pilot plant processing uranium metal and was then referred to as the “Pilot Plant” (Mason 1977; ORNL 1981). The “dingot” metal production process was developed and conducted at Plant 4 in the mid-1950’s (AEC 1956a; Fleishman-Hilliard 1967), along with sporadic ordinary metal “derby” production on a developmental basis. Plant 4 was used until 1956 (Mason 1977; ORNL 1981; Mallinckrodt 1994; FUSRAP 2003c).

Due to the need to increase production and also due to the recognition by MED and Mallinckrodt of significant safety problems with dust and external doses (partly arising from the prospective extensive use of radium-containing pitchblende ore (ORNL 1981; ORAU 1983a; Mallinckrodt 1994)), Plant 6 was built in 1945-46 on a large site fronting on Destrehan Street (Mason 1958a; Fleishman-Hilliard 1967; Mallinckrodt 1994); it began operation in July 1946 and was then referred to as “the refinery” (AEC 1949b; AEC 1951b). Mason (1977) states that the choice of this site was based on proximity, security considerations, and space availability for constructing administrative and other support buildings that would enable the uranium operations to function as an independent unit. Most of the administrative offices, laboratories, and support facilities for the uranium refining operations were in fact located there. In early 1946, the ore-to-UO_2 part of the refining process was moved to Building 104 of Plant 6 from Plant 2 and apparently the laboratory work moved to Building 102 from Plant 1, while the UO_2-to-metal production remained at Plant 4 (FUSRAP 2003c; Hickey and Dupree 1984; AEC 1949b; Mason 1958a). However, some reduction of UO_3 to UO_2 appears to have been done at Plant 4 also, perhaps as part of pilot plant operations, as indicated by Mason (1958) and AEC (1950c). Mason (1977) states that expansion of Plant 6 (new construction and production increases) continued through 1954.

In November 1947, Mallinckrodt cast 100 special billets for use by Hanford in an alpha extrusion experiment (AEC 1949d). These special billets contained higher nickel levels than usual and were prepared as part of the research into green salt production, in the belief that the new continuous green salt production reactor might give a product with higher levels of nickel than usual (AEC 1949f). From late 1947 to early 1948, Mallinckrodt was engaged in producing natural uranium castings for the Schenectady zero power pile (AEC 1949d). Also from late 1947 to early 1948, several tanks and centrifugal filters were added to the digest process line to allow the reprocessing of the radium (K-65) and barium sulfate residues and recovery of 95% of the 2% of the original uranium from the ore that was in these two residues (AEC 1949d; AEC 1948g). The K-65 and barium sulfate residues already stored in the St. Louis area were then returned from storage and processed. Subsequently this so-called C-3 process was used on all the K-65 residues produced and the companion process on all the barium sulfate residues produced (AEC 1949d).

The incoming ore arrived by rail and was stored in Building 110; however, in late 1950 or early 1951 an outdoor onsite ore storage area was added for pitchblende ore (Q-11) (AEC 1950i). Building 104 housed the continuous process equipment, which replaced the batch process equipment that had been used in Plant 2, and processed mostly pitchblende ore (Mallinckrodt 1994). The new refinery process was designed to include the new operations developed in the laboratory and pilot plant, i.e., to recover radium and raffinates as separate residues and to remove sulfate as a barium sulfate cake (Mason 1977). In 1949, following laboratory and pilot-scale studies, a second digest line was added to Building 104 to process uranium ore (mill) concentrates (Mason 1977; Mallinckrodt 1994). This appears to have included a magnesia concentrate designated MgX or MGX and a calcium uranate concentrate designated CX, both of which were prepared in the Congo from low-grade tailings (AEC 1951b; AEC 1949f). Building 114 was used for temporary storage of residues until they could be transferred elsewhere (Mallinckrodt 1994). Most of the UO_2 produced at Plant 6 was trucked over to Plant 4, with the rest going by rail to Harshaw and Linde for some years (AEC 1949b). Pitchblende ore use was phased out over the period 1952-1954, after which the pitchblende digest line was placed on standby (Mason 1977). Feed concentrates were used exclusively from 1955 through the end of production in 1958 (Hickey and Dupree 1984).

When equipment was added to Plant 7 to allow continuous UO_3-to-UF_4 conversion, Plant 6 continued to produce UO_2 until early 1953 and then began to produce only UO_3 (Hickey and Dupree 1984; Mallinckrodt 1994). Milling of UO_3 at Plant 6, milling of ore at Plant 6, and pre-digestion ore grinding, conducted initially at Plant 4 and later at Plant 6, were discontinued in 1950, mid-1949, and 1955 respectively (Mason 1958a). Pitchblende ore continued to be used as feed until early 1955 (AEC 1959). In 1957, a pilot-scale fluid-bed denitration reactor from Argonne was installed for testing and developmental improvements at the Destrehan site, presumably at Plant 6 (Fleishman-Hilliard 1967).

In 1949 it had become clear to AEC that process improvements to Plants 4 and 6 were not enough to bring about satisfactory control of dust and other hazards (AEC 1949b; AEC 1951b). As Mallinckrodt reminded AEC, Plant 6 had been built as an ordinary processing plant with no special provisions for health hazards because it was not expected to be used more than eight months (AEC 1951b; MCW 1950t). AEC authorized funding for dust control and mechanization improvements, which were installed in 1949-1950 (see Table 3). Plant 6 was shut down for a time in late 1949-1950 for this purpose. It is not clear what the workers did during the time the plant was shut down, although it seems likely that those who had the necessary skills worked on the installation project.

These improvements were regarded as an interim measure, however (MCW 1950e; AEC 1951b). Mallinckrodt and AEC agreed to build a new pair of plants that were located at the Destrehan Street site (AEC 1949b; Mason 1958a). The first was Plant 6E, the new metal plant, which was constructed from early 1948 to June 1950, began startup operations in June 1950, and went into operation in July 1950 (AEC 1949e; AEC 1951b; Mason 1977). Note that there was a laborers' and hod carriers' strike from 28 March into early April 1949 that slowed construction of Plant 6E (AEC 1949e; AEC 1949d). Metal production (UF_4-to-U metal) operations at Plant 4 moved to Plant 6E, which was from then on referred to in records as "the metal plant". Metal production took place in Building 116 (Mason 1977; Mallinckrodt 1994). An expansion project was started in March 1951 and finished in about September 1951 (AEC 1951b). Building 116C was built in 1954 to recycle magnesium fluoride slag (Mallinckrodt 1994).

The second new plant was Plant 7, the green salt plant, which was constructed from November 1950 to March 1951 and was turned over to Mallinckrodt for operation in March 1951 (AEC 1951b). Note that AEC (1949b) appears to indicate that Plant 7 operations began in late 1952, while Mason (1958a) indicates a 1948 start date and Mason (1977) indicates a 1951 start date. The confusion may be due to the nature of the new direct or continuous UO_3-to-UF_4 process used at Plant 7 and the apparent use of Plant 4 facilities to test out the process, as a pilot plant, and to work out difficulties even after Plant 7 went into production. The buildings involved at first were Buildings 703 through 708 (Mason 1977). At Plant 7, a continuous process replaced the batch-type process used at Plant 4 and the UO_3-to-UO_2 production process at Plant 6, although UO_2 could still be produced as needed (AEC 1951b; Mason 1977); however, Mallinckrodt (1994) states that it was only later that equipment was added to allow the continuous production of UF_4 from UO_3 directly.

Uranium metal recovery and some storage operations were moved to Plant 7 in late 1951 (AEC 1951a; AEC 1951b; Mallinckrodt 1994) or early 1952 (Mason 1958a). Some reversion of UF_4 to UO_2 or UO_3 was done in 1953 and perhaps into 1954 (MCW 1953c). Building 700 was placed in service in 1954 to serve as a warehouse (Mason 1977). A new wet slag (interim residue) recovery operation was added in late 1955 in Building 701 (Mason 1977; Mallinckrodt 1994); this was apparently what was referred to as Plant 7W and may have been a pilot plant (Hickey and Dupree 1984). Some time in 1954 (Mason 1977) or in the late 1950's (Mallinckrodt 1994) machining of (nuclear) reactor core (elements) was done on a temporary basis in a fabrication facility in Building 700. In 1955, very low enrichment uranium (probably only a small amount) as UF_4 was processed at Plant 7 (AEC 1955d) and in August 1956, about 5.5 kg of 20%-enriched uranium was processed, presumably in Plant 7 (MCW 1956e).

Plant 7E, whose sole building (712) was constructed as a temporary facility in 1954-1955 and which was regarded administratively as part of Plant 7, was used from 1955-1957 to process pitchblende raffinate (solids removed during uranium refining by wet filtration) to produce a concentrated thorium solution (FUSRAP undated b, FUSRAP 2003a; Mason 1977 says from 1955-1956) by an acid digestion process similar to the uranium ore digestion (AEC 1955e). The concentrate was sent to the Mound site for further processing.

A new facility to replace the so-called "Ledoux Laboratory" was completed in November 1952 (MCW 1953a). The purpose of this was to provide improved dust control for the sampling and processing of K-65, the radium-containing solid waste. Further improvements were made in 1953 (MCW 1953a). Other sampling and assay work was done, such as the evaluation of slugs produced by National Lead Company of Ohio from extruded rods that had been manufactured by the Bridgeport Brass Company from dingot billets apparently produced by Mallinckrodt (MCW 1956d).

In 1957 or 1958, all uranium refining operations ceased at all the plants (FUSRAP 2003c; DOE 1981) and the work moved to Fernald and Weldon Spring. Eisenbud (1975) stated that all Plant 6 work was transferred to Weldon Spring in March 1957 (steps through UO_3 production, at that time). However, Mason (1958a) stated that some Plant 7 production operations continued up to July 1958, when they were transferred to Weldon Spring; this was probably to use up the store of orange oxide that had already been produced. This is supported by the statement of Mallinckrodt (1994) that the last of the site was put on standby in 1958.

Decontamination and surveys were performed at Plants 1 and 2 in 1948-1950 by Mallinckrodt personnel, applying AEC criteria (ORNL 1981; Mason 1977, which says the work began in 1949); contaminated materials and some contaminated equipment were collected and delivered to AEC for disposal, while the rest of the contaminated equipment was transferred to Plant 6 (Mason 1977). After AEC was satisfied as to the completion status of the decontamination, AEC performed a final survey (Mason 1977), with presumably some touchups by Mallinckrodt and resurveys by AEC. In 1951, AEC released these plants to Mallinckrodt for unrestricted use and no further AEC work was performed there (Mason 1977; DOE 1981). Mallinckrodt (1994) states that further decontamination took place in 1954 and 1970. Some of these buildings were still extant as late as 1990 (Applied Nuclear Safety 1991). Plants 4, 6, 6E, and 7 were surveyed by AEC and Mallinckrodt in 1958-1959 and were decontaminated or taken down in 1959-1961 by Arch Wrecking Company (under contract to AEC), with safety and health coverage by Mallinckrodt (Mason 1977). Mallinckrodt performed a final decontamination survey for AEC during 1960-1961 (Mason 1977). The buildings removed during the takedown included all of the Plant 4 buildings and all wet-process buildings in Plant 6 (ORNL 1981; Mallinckrodt 1994) (see Table 1 for details). The site was released to Mallinckrodt without restriction in 1962 (ORNL 1981). These decontaminations were done to the AEC standards then in force, i.e., not to background levels (ORNL 1981; DOE undated). The remaining Plants 6, 6E, and 7 buildings were decontaminated to modern standards in the 1990's and demolished in 1997 (FUSRAP 2002).

Note that the term "Destrehan Street site" seems to have been used in documents sometimes in reference to all the Mallinckrodt St. Louis facilities and sometimes just to those on the Destrehan Street site proper. In this technical basis, the former interpretation will be used, i.e., Plants 1, 2, 4, 6, 6E, and 7 will be covered. For clarity in using the references, note that Plant 4 fronted on Broadway Street and although Plants 1 and 2 did not, either Plant 4 or all three plants could be referred to as "the Broadway site". Thus the "Broadway" and "Destrehan" terms appear to be a loose reference to geographical location and not necessarily to a division of function or operations. Also, as will be explained later, due to a rotation practice, Mallinckrodt workers were apt to have worked in multiple plants over the course of their employment. In distinguishing between these plants and the SLAPS airport site (see Section 3.2), these plants as a group will be referred to in this technical basis document as the Mallinckrodt St. Louis main site.

To summarize, work performed at the Mallinckrodt St. Louis main site on a continuing basis included the following.

1. Production of UO_2 and UO_3 from ore, with some being shipped to Harshaw and other sites and with some natural and low-enrichment UO_3 coming from Hanford and other sites in the 1950's (DOE 1997; Mallinckrodt 1994; MED 1946a)

2. Production of UF_4, with some being shipped to Harshaw and K-25 (DOE 1997; Mallinckrodt 1994; MED 1946a; FUSRAP undated b)

3. Production of uranium derby metal and vacuum recasting of ingot metal, with the ingots being the final product shipped to other sites (DOE 1997; Mallinckrodt 1994; MED 1946a; FUSRAP undated b)

4. Recovery of scrap uranium metal, some from other sites, such as Hanford (FUSRAP Undated b; DOE 2000; Mallinckrodt 1994)

Other work performed for a limited period or on an occasional or small-batch basis included the following.

1. Production of dingots using a thermite reduction process (Mallinckrodt 1994; Fleishman-Hilliard 1967; AEC 1956a)

2. Machining of metal rods for reactor fuel slugs (Mallinckrodt 1994; FUSRAP undated b)

3. Casting of special billets (ingots) and other special uranium castings (AEC 1949d)

4. Reversion of UF_4 to UO_2 or UO_3 (FUSRAP Undated b, which says UO_2 or U_3O_8; Mallinckrodt 1994; AEC 1954g)

5. Production of UO_2F_2 (FUSRAP undated b)

6. Reprocessing of pitchblende residues to recover uranium (AEC 1949d; AEC 1951b)

7. Recycling of slag for use as liner material in the processing of uranium metal

8. Extraction and concentration of Th-230 from pitchblende raffinate (FUSRAP undated a; FUSRAP undated b; Mallinckrodt 1994; AEC 1955e)

9. Experimental processing of very low enrichment UF_4 (FUSRAP undated b; Mallinckrodt 1994; ORNL 1981) and some 20% uranium metal (MCW 1956e)

10. Production of small research quantities of uranyl nitrate hexahydrate (UNH)(AEC 1951b); conversion of small research quantities of aqueous uranyl nitrate hexahydrate to UO_3 (DOE 2000)

11. Production of U_3O_8 and UO_2 using an experimental continuous denitration furnace (Mallinckrodt 1994)

12. Experimental extraction of uranium using tributyl phosphate (TBP) to replace ether (Mallinckrodt 1994), apparently in a pilot plant circa 1950-1951

13. Miscellaneous analysis and assay work (e.g., MCW 1956d)

DOE (2001b) states that from 1946-1955, personnel provided by Ledoux and Co. of New York performed uranium ore assays at the Mallinckrodt St. Louis main site and FUSRAP (2003b) states that this work was done in the “Ledoux Laboratory” located in Buildings 110A and 111 of Plant 6. However, from MCW (1954a) and other contemporary references, it appears that only the K-65 residue assays were done in the Ledoux Lab, which work had been contracted to Ledoux by African Metals (ORAU 1989a), which retained the rights to the radium in the residue. The Ledoux Laboratory is also mentioned in Mallinckrodt film badge records, so presumably the Ledoux and Co. personnel were badged under Mallinckrodt’s aegis.

MED (1944p) stated that non-MED Mallinckrodt employees and employees of the St. Louis Sash & Door Works (the former operator of Plant 4’s Building 400 in its pre-MED days) had occasion to enter respectively Buildings 45, 45A, and 47 in Plant 2 and “Building 1” at Plant 4 (presumably Building 400) respectively. These employees were presumably not monitored in any way other than having their access controlled for security reasons. They are not considered to be atomic workers for the purposes of this dose reconstruction.

Mallinckrodt processing of black oxide (pre-milled ore), soda salt (sodium diuranate, or $Na_2U_2O_7$), UO_3 (orange oxide), and UO_2 (brown oxide) was done under AEC/MED Contract W7401-Eng 1; UO_2 and UF_4 (green salt) under Contract W7405-Eng 29, 1st Phase; and UF_4, biscuit metal (uranium derbies), slag processing, ingots (billets), croppings, sawdust processing, and other metal production under Contract W7405-Eng 29, 2nd Phase (MED 1945a; MED 1944p).

Starting in 1952, some recycling of uranium was done by the AEC and its contractors nationwide. Thus the question arises whether this was done at the Mallinckrodt facilities in St. Louis. However, Mallinckrodt (as a company) did not begin to receive recycled uranium until 1962, which was after the St. Louis facilities had been shut down and their work had shifted to Fernald and Weldon Spring (the latter run by Mallinckrodt) (FUSRAP 2003b). Also, ORNL (1981) stated that in its pre-survey review of the site, including interviews with Mallinckrodt old-timers, no indications were found that there had ever been any process conducted under AEC contracts involving the purification or working of Th-232, highly enriched uranium, fission products, or byproduct material. Thus it is assumed that no recycled uranium or any of the materials listed by ORNL (1981) was handled at Mallinckrodt's St. Louis facilities as part of AEC work.

In 1949, AEC requested that Mallinckrodt produce in their AEC-owned facilities 200 pounds of uranium metal for "nonproject and educational uses" (MCW 1949h); however, there is no indication that Mallinckrodt ever did so. Mallinckrodt apparently also carried on some commercial processing of euxenite (an ore) at Plant 5 at some point in the 1950's. Because this work was done commercially and not for the AEC (FUSRAP 2003a), it is not considered to fall in the category covered by EEOICPA.

3.2 HISTORY OF USE OF THE ST. LOUIS AIRPORT STORAGE SITE (SLAPS OR SLAPSS)

The main supplier of the pitchblende ore to MED, the African Metals Company, retained ownership of the radium and valuable metals remaining in the residues after the ore was processed (AEC 1959). Thus by contract, MED was required to store the radium-bearing residue (K-65 or gangue lead cake) and the metal-rich barium sulfate cake residue until they could be transferred to African Metals (AEC 1959). From 1942 to 1945, the residues and other solid wastes were stored temporarily at the Mallinckrodt site and then shipped to the Lake Ontario Ordnance Works. Because of the volume of the residues and the high gamma dose rate and radon emanation rate of the K-65 residue, MED obtained consent in March 1946 to use for residue storage purposes a 21.7-acre tract of land located in Robertson, Missouri north of the former St. Louis Municipal Airport (also known as Lambert Field) (AEC 1959). In January 1947, MED obtained title to the property by condemnation (AEC 1959). This storage area was formally called the St. Louis Airport Storage Site (SLAPS or SLAPSS), but in Mallinckrodt and MED/AEC documents it was often referred to simply as "the airport". The SLAPS site was operated by MED/AEC from 1946 until July 1953 (AEC 1959; ORNL 1979), when the operation was turned over to Mallinckrodt (AEC 1959); Mallinckrodt then ran the site under contract to AEC from July 1953 to 1958 and possibly until it was turned over to a private concern that assumed control of it under an AEC license (AEC 1959; ORAU 1989; AEC 1972). Although AEC owned SLAPS, Mallinckrodt workers appeared to have been doing the residue delivery and placement work even in 1946-1953 (MCW 1949d; MCW 1949p).

Initially, it was only the radium-bearing waste residues (K-65), the barium sulfate residues (AJ-4), and the pitchblende raffinates (AM-7 or "airport cake") produced at Mallinckrodt that were stored at SLAPS (Mallinckrodt 1994). Later, SLAPS was also used to store other types of residues and wastes and some ore and to serve as a storage and burial site for contaminated objects and debris (AEC 1959; AEC 1956d; Mallinckrodt 1994; AEC 1948e; AEC 1949m). The ore was stored there as part of AEC's strategic stockpiling program (AEC 1949d). The other types of residues included the residue left after reworking of the AM-7, called AM-9; Colorado (domestic non-pitchblende) ore raffinate; and a residue formed from the precipitate of the first-phase ether extraction columns ("Sperry cake", apparently a synonym for or subset of the AM-7) (AEC 1956e; AEC 1964; Mallinckrodt 1994). Metal scrap, dolomite slag ("C-liner" or "C-oxides"), and tailings of uranium scalping operations from magnesium fluoride slag (the latter two from the derby metal production) were also sent to SLAPS (Mallinckrodt 1994). Some of the debris buried at the site appears to have been from the decontamination and decommissioning of the Plants 1 and 2 area in 1948-1950. From perhaps mid-1948 on, pitchblende ore (Q-11) was stored at SLAPS (MCW 1949p).

In 1948 and 1949, the K-65 residue was brought back from SLAPS in lots to be "reworked" (reprocessed to recover more uranium) (AEC 1959), after which it was sent to the Lake Ontario Ordnance Works (AEC 1959). The drums in which it had been stored were returned to SLAPS for storage. From 1955-1957, most of the AM-7 was brought back from SLAPS to be processed to recover Th-230 as a solution that was sent to Mound; the resulting residue, AM-9, was sent back to SLAPS (AEC 1959). Also in 1955, the tailings (D-701) from the processing of the magnesium fluoride slag produced in the metal plant were stored at the site (AEC 1959) or at SLAPS (AEC 1956d; Mallinckrodt). The barium sulfate cake remained at SLAPS because eventually African Metals relinquished ownership of it (AEC 1959).

In 1952, some of the western part of the SLAPS plot, where contaminated metal and debris was buried (presumably from the decontamination of Plants 1 and 2), was filled in deeply with earth (AEC 1964). In April 1959, a railroad siding and loading facilities were installed (AEC 1959), apparently in preparation for moving out the residues and other material (AEC 1959); the tailings from the magnesium fluoride slag processing were then sent to the Fernald site (AEC 1959). In about 1959-1960, some of the scrap and rubble from the demolition and decontamination of the Mallinckrodt St. Louis site facilities was buried at the west end of the site (AEC 1972; ORNL 1979). At some point, a building consisting of a changeroom, a shower, and office space was moved to the site (AEC 1959; AEC 1964); it is not clear when this occurred, but since earlier references mention only a truck wash pad as a cleanup area, the building was likely not installed until the burial activities of 1959-1960 took place.

In March 1962, AEC issued an invitation to bid on the residues and on September 1962, AEC issued a license to the successful bidder, a commercial entity, to possess and remove 125,000 tons of uranium and thorium residues stored at SLAPS and at the licensee's processing facility. Although this is taken to be the end point for dose reconstruction purposes, some further history of the site will be given below in case it is later necessary to choose a different end point.

The 1962 license was terminated in February 1966. This was possibly due to the licensee's going out of business (AEC 1967). A new license was issued to a new purchaser in 1966. In 1966-1967, this purchaser moved the residues from SLAPS (AEC 1967, FUSRAP 2004a) to a site at Latty Avenue in Hazelwood, Missouri (AEC 1967); this was said to have required ten dump trucks operating for five months to move a total of 100,000 tons of residue (AEC 1967).

AEC apparently proposed to the City of St. Louis that the city take over the site, specifically the city-owned airport management organization. Thus in 1969-1970, under a November 1969 acquisition permit from AEC and with some radiological support from the City of St. Louis Health Department, the St. Louis (Lambert) Airport Authority demolished the structures on the site and excavated some of the contaminated earth (AEC 1969; AEC 1972; ORNL 1979; FUSRAP 2004a). Excavated material, including some residual barium sulfate residues, was trucked to the AEC quarry at Weldon Spring (AEC 1969; AEC 1972), while the demolished structures, including the concrete floors and pads, were buried at the site and clean fill was placed over the entire site (AEC 1972). More fill was placed over several areas following a radiation survey performed in December 1969 to meet the requirements of the permit (ORNL 1979). In November 1971, AEC representatives performed a satisfactory final radiological survey of the site (AEC 1972; ORNL 1979).

In 1973, AEC relinquished ownership of the site to the St. Louis Airport Authority by quit claim deed (FUSRAP 2004a). After that, access was controlled by the St. Louis Airport Authority and casual entry was precluded (ORNL 1979). Over the period from about 1972 through 1978, the site served as an isolation or buffer space on the side of a runway approach at the adjacent airport and it was also used as a dump site for nonradioactive rubble from the airport; the fence remained in place, but the area was not patrolled (AEC 1979). Without maintenance, the site deteriorated, in that erosion occurred and minor contamination was transported to the drainage ditch that surrounded the site outside the fence and to the creek that traversed the lower end of the site (AEC 1979). This was shown in the radiological characterization surveys performed by Oak Ridge National Laboratory (ORNL) for AEC in November 1976 and August 1978 (ORNL 1979; AEC 1979). On the basis of these surveys, it was decided that remedial action was required (AEC 1979; FUSRAP 2004a) and in 1981 the drainage ditches along the north side of the site were designated for remedial action under FUSRAP (2004a).

The residues that had been sold and taken out of the site went to the Latty Avenue (St. Louis) site to await processing by the purchaser; however, the purchaser went bankrupt before processing actually began. Under a Nuclear Regulatory Commission (NRC) license, the Cotter Corporation then dried the residues in a kiln at the Latty Avenue site and sent them off to their Colorado facility. However, the Latty Avenue site was not properly decontaminated before the NRC license was terminated and the site was sold to a private purchaser. After an ORNL survey at the Latty Avenue site, the owner was advised by NRC to stop property development and the NRC asked the Department of Energy (DOE) to allow materials from the future decontamination of the site to be taken to the Weldon Spring Plant (AEC 1979). However, DOE considered the Weldon Spring raffinate pit to be unsuitable for permanent disposal of these materials. NRC then suggested that the material be returned to SLAPS and DOE agreed, providing that this could be done in an environmentally acceptable manner (AEC 1979).

Eventually, the US Congress, via the Energy and Water Development Appropriate Act of 1984 (Public Law 98-360), directed DOE to reacquire and stabilize the SLAPS site and to use it as a storage and disposal site for the Latty Avenue materials (Bechtel 1987; DOE 1986). Although it postponed acquisition of the site even after being directed to do so by Congress, DOE had begun to have routine monitoring of SLAPS done by Bechtel National Incorporated in 1983 and as of 1987 was planning to dispose of the waste materials from the Hazelwood Interim Storage Site (including the Latty Avenue materials) at SLAPS eventually (Bechtel 1987). In October 1989, the Environmental Protection Agency (EPA) placed SLAPS on the National Priorities List, which meant that the cleanup would proceed according to the guidelines of the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) (FUSRAP 2004a). DOE then began to work with EPA to determine how to clean up the site and where to put the contaminated soil (FUSRAP 2004a). The two agencies signed an agreement establishing an environmental review process and remediation schedule, with one of the options to be considered being storage of the SLAPS and HISS materials at SLAPS; DOE declined to accept SLAPS back from the City of St. Louis until the CERCLA review process was completed (FUSRAP 2004a).

In 1990, the St. Louis Board of Aldermen approved a plan to transfer SLAPS to DOE on condition that a permanent disposal cell for radioactive wastes would not be constructed on the site (FUSRAP 2004a). In 1994, DOE formulated a remediation plan and presented it to the St. Louis community. The community rejected it because it included a disposal cell at SLAPS, contrary to the terms of transfer enacted by the board of aldermen; the site was therefore never transferred to DOE and it appears unlikely that it will ever be (FUSRAP 2004b). However, as of July 2004, FUSRAP was working to develop a Record of Decision to document the final cleanup remediation plan for the North County sites, including SLAPS – a plan that did not include a disposal cell at SLAPS (FUSRAP 2004b).

4.0 DESCRIPTION OF THE MALLINCKRODT URANIUM REFINING PROCESS, OTHER PROCESSES, AND SLAPS STORAGE AND DISPOSAL ACTIVITIES

It is important to understand the Mallinckrodt uranium refining process in order to understand the radiological hazards, to follow the changes in source terms and exposure potential, and finally to estimate the doses to individual workers. The basic process will be described here. Then particulars will be discussed for three cases: the early wartime period, the late wartime and early postwar period, and the later postwar period. Details regarding the types and quantities of ore, uranium products, and residues are shown in Table 4; Table 5 is provided as a reference for technical terms and keywords. An additional table is given within the text of Section 4.4 to illustrate the variety of feed forms that was used in later years.

It is also important to understand the nature of the work performed at SLAPS in order, again, to understand the radiological hazards, to follow the changes in source terms and exposure potential, and finally to estimate the doses to individual workers. The discussion of SLAPS work is included in the discussion of Mallinckrodt-produced residues (Section 4.7 below).

4.1 THE BASIC PROCESS

First the ore was prepared for uranium extraction, as follows. Milled ore, as ore concentrate (“black oxide”), was taken from its storage location(s), thawed if necessary in an enclosed “thaw oven” in the thaw house, and dried. It was then digested in nitric acid in a digestion vessel or tank (MED 1946a; AEC 1949b; AEC 1967; Fleishman-Hilliard 1967). Sulfuric acid was added to the solution in the vessel to precipitate the radium and lead as sulfates (MED 1946a; AEC 1949b; AEC 1951b; AEC 1967; Fleishman-Hilliard 1967). The uranium was left in solution as uranyl nitrate and the precipitate was filtered out (MED 1946a; AEC 1949b; Fleishman-Hilliard 1967) using a string-discharge rotary vacuum filter (AEC 1967) referred to as a Feinc because of the name of its manufacturer(Federal Engineers Incorporated or FE Inc). The filtered solids formed a radium-bearing residue referred to as the K-65 residue (AEC 1949b; AEC 1967) or GLC (“gangue lead cake”) (MED 1946a). Next, a slurry of barium carbonate was added to the uranyl nitrate solution to remove the sulfates, when present (MED 1946a; AEC 1949b; AEC 1951b; AEC 1967; Fleishman-Hilliard 1967). To remove the solids, the mixture was run in a continuous solid-bowl centrifuge (also referred to by the name of its manufacturer, Bird) (MED 1946a; AEC 1949b; AEC 1967). The uranium remained in solution as uranyl nitrate and the precipitate formed a barium sulfate cake. The uranyl nitrate solution -- the “liquor” -- was then boiled to concentrate it (MED 1946a; AEC 1951b). Calcium nitrate was added to the solution to assure nitrate saturation (MED 1946a), then the solution was filtered to remove any solids formed. The acidity of the solution was adjusted as needed by adding acid (AEC 1967; Fleishman-Hilliard 1967).

Second, uranium oxide as UO_3 was produced from the solution, as follows. In the two-step ether extraction process, diethyl ether was added to the solution in extraction columns; the double extraction was to remove the acid-insoluble molybdenum (AEC 1951b). The first step was the acid ether extraction, including ether addition, nitric acid addition, and re-extraction into water (also called a “water wash”), while the second step was a neutral ether extraction followed by re-extraction into water (MED 1946a; AEC 1949b; AEC 1951b; AEC 1967; Fleishman-Hilliard 1967). Eisenbud (1975) stated that in the ether extraction process, the isotopes of thorium and protactinium were left in the “aqueous phase” while the uranium was stripped off by the solvent (ether), so presumably the thorium and protactinium were separated from the uranium in the first step of the ether extraction process, before or as part of the water wash.

After the ether extraction was complete, the uranyl nitrate extract solution was boiled to the molten salt to form uranyl nitrate hexahydrate (“hex liquor” or “OK hex liquor”) (MED 1946a; AEC 1949b; Fleishman-Hilliard 1967). The molten salt was then directed in batch form to gas-fired denitration “pots” or boildown vessels, which were continuously heated and agitated so as to produce the dissociation of the hexahydrate and the formation of UO_3 (also called orange oxide or QM-2) (MED 1946a; AEC 1949b; Fleishman-Hilliard 1967). The UO_3 lumps were often broken up by crude mashing, but there was also a formal double grinding process requiring the material to be moved to the grinding area (MED 1944o).

Third, the UO_3 was placed on thin trays, placed in airtight boxes, and loaded into a batch electric (muffle) furnace to react with dissociated (cracked) ammonia to form UO_2 (also called brown oxide or LF-9), a step that took about 7 hours (MED 1949b; MED 1946a; AEC 1949b; Fleishman-Hilliard 1967). Usually the UO_2 was further processed at Mallinckrodt, but some was shipped to non-Mallinckrodt sites such as Harshaw (DOE 1997).

Fourth, the UO_2 was converted to UF_4, as follows. The UO_2 was removed from the fiber containers and placed into large stainless steel drums for temporary storage (AEC 1949b). As needed, it was removed onto steel (later monel) trays and weighed (MED 1946a; AEC 1949b; Fleishman-Hilliard 1967). The trays were then set into airtight graphite or nickel boxes and placed in a hydrofluorination reactor (MED 1946a; AEC 1949b; Fleishman-Hilliard 1967). (Note that this was not a nuclear/atomic type of reactor but rather a chemical reaction vessel.) In the reactor, hydrogen fluoride gas (HF) was passed over the UO_2, forming UF_4 (also called green salt or TA-7) and water (MED 1946a; Fleishman-Hilliard 1967). The UF_4 was unloaded and put through a pulverizer or mill and then a blender, followed by packing into metal containers (AEC 1949b). Most of the UF_4 was further processed at Mallinckrodt, but some was regularly shipped to non-Mallinckrodt sites (DOE 1997; AEC 1949b); MED (1946), however, implies that all of the UF_4 produced at Mallinckrodt was also further processed there. Excess HF was neutralized with lime or potassium hydroxide solution to recover uranium (Mason 1977).

Fourth, the UF_4 was converted to uranium metal, as follows. The UF_4 was blended with magnesium powder and mixed in a tumbler (AEC 1949b; Fleishman-Hilliard 1967; Mason 1977). A special firing container (the “bomb”) was lined with a refractory material, variously reported to be lime (1945, per MED 1946a); dolomite (from 1946 on, per FUSRAP undated a, or 1942 on, per Fleishman-Hilliard 1967); or recyclable magnesium fluoride from about 1954 on (FUSRAP undated a; Fleishman-Hilliard 1967). The bomb was “jolted” (shaken) on a mechanical jolter until the liner was packed sufficiently hard. The mixture (the “charge”) was then placed in the bomb. After the bomb was sealed, it was placed in a gas-fired furnace and heated until it “fired”; the magnesium reduced the UF_4 to uranium metal in this process (MED 1946a; AEC 1949b; Fleishman-Hilliard 1967). The molten mass formed a “derby” or “biscuit” of solidified metal as it cooled (MED 1946a; AEC 1949b; Fleishman-Hilliard 1967). (The derby form was also called KB-2 and there were subtypes of derbies called Number 1 Number , etc.) After the bomb cooled, the derby was taken out by “breaking out” the bomb shell (Mason 1977) and the slag on the derby was chipped off pneumatically, crushed, and sent to uranium salvage (recovery), first to Vitro and eventually to onsite recovery (MED 1946a; AEC 1949b; Fleishman-Hilliard 1967; Mason 1977).

Finally, the metal derbies were remelted and cast in an induction-heated, high-vacuum furnace (a process called vacuum recasting). In this process, the derbies were placed in a crucible, the crucible was placed over a mold, and the crucible-mold assembly was placed in a quartz shield, sealed, evacuated, and placed into the furnace (MED 1946a; Fleishman-Hilliard 1967). There was a stopper rod at the bottom of the crucible that was removed when the critical temperature was reached, allowing the molten metal to flow into the mold and form a cylindrical ingot or billet (MED 1946a; AEC

1949b; Fleishman-Hilliard 1967). (The billet form was also called YM-5.) The billet was then removed from the mold by separating its parts. The porous impurity-heavy top section was cropped off and sent for recovery; the other surfaces were cleaned of liner ("C-liner") slag and other impurities (MED 1946a; AEC 1949b; Fleishman-Hilliard 1967). A sample was taken from the billet by power hacksaw (AEC 1949b) and the billets were sawed to suitable lengths (presumably only when necessary) (Mason 1977). The billets were then packed, stored temporarily in Building 400 (Mason 1977), and shipped to the appropriate non-Mallinckrodt site for further processing (MED 1946a; AEC 1949b; Mason 1977).

The nitric oxides produced as offgasses in the pots were sent to a recovery system (MED 1946a; AEC 1949b; FUSRAP 2003b) where the nitric oxides were converted back to nitric acid. The GLC residue and the barium sulfate cake were usually leached with sodium carbonate to remove the residual uranium (AEC 1967).

The precipitate from the first extraction column was on occasion dewatered using a Sperry filter press, producing a supernate and a batch of cake called Sperry cake (AEC 1967). The supernate from the press and the aqueous uranium tails from the wash were de-etherized and treated with a hydrated lime slurry (AEC 1967). This was passed through a continuous rotary vacuum leaf filter (referred to by the name of its maker, Niagara); the supernate was discharged to the local river and the limed fraction became AM-7, called "airport cake" (AEC 1967), from its being stored for several years at the AEC's SLAPS site that had been an airfield. Note that the Sperry cake appears to be a subset of the AM-7 or perhaps synonymous with it. Also, several other types of cake (e.g., the barium sulfate cake) were also referred to loosely as "airport cake" due to their being sent there for storage.

4.2 THE WARTIME PERIOD (APRIL 1942– APRIL 1945)

In the first months of refining by Mallinckrodt, a different extraction process from the ether extraction was used since the latter had not yet been developed. No information could be found as to the details of the first process; however, the ether extraction process seems to have been in the process of development at Mallinckrodt from April 1942 on and in use from July 1942 on (DOE 1997; AEC 1967; Fleishman-Hilliard 1967), so the first process could have been used for about three months at most (mid-April to mid-July). The magnesium reduction process for metal production was developed by Iowa State by about mid-July 1943 (DOE 1997) and Mallinckrodt established the first metal plant , Plant 4, during the same month (Fleishman-Hilliard 1967). Mason (1977) stated that there were no wet processes in Plant 4 (ever).

Initially, the work consisted primarily of the production of UO_2 and UO_3 from mined ore marginally milled (FUSRAP undated b); some preliminary milling to "black oxide" (a form of U_3O_8 concentrate) thus had to be done, some of it at Mallinckrodt (DOE 1997). The ore had to be ground to "four mesh" size for optimum digestion as well, if the ore particle size was larger (MCW 1947). Little information is available as to how or where the early milling and grinding operations were performed, but it was apparently in Plant 2 (see Table 1), later in Plant 4 (AEC 1945f; AEC 1945h; AEC 1945i), and still later in Plant 6 (e.g., MCW 1950e). Some soda salt was also used as feed material (DOE 1997). Note that the radium-lead precipitation step was necessary for the high-radium pitchblende ores, but not necessarily for other types of ore.

Plant 4 began production-level UO_2-to-UF_4 conversion in April 1943 and production-level UF_4-to-metal production in July 1943. The ore$\rightarrow UO_3 \rightarrow UO_2$ conversion continued to be done in Plant 2. MED (1942) stated that the ether extraction was done by a completely enclosed process of "recirculating" and that a solution of purified uranyl nitrate was boiled in large open receptacles to decompose the nitrate and produce the UO_3. The UO_3 was then ground and sifted to yield the appropriate size for "roasting" in a furnace to produce the UO_2.

AEC (AEC 1951b) stated the following. The process of refining pitchblende ore by using nitric acid, including the radium removal technique and acid extraction were developed by Yale University but expanded to production scale by Mallinckrodt during this period; also, double extraction to remove molybdenum when pitchblende ores were processed and the balance of the processing to UO_3 were developed by Mallinckrodt. The UO_2 hydrofluorination process was developed by DuPont and Harshaw, while the process for reduction of UF_4 to uranium metal was developed by Iowa State College at Ames. The research for the continuous UO_3-to-UF_4 process was done by Mallinckrodt.

4.3 THE EARLY POSTWAR PERIOD (MAY 1945–DECEMBER 1949)

As noted above, processing of significant quantities of pitchblende ores began at Mallinckrodt in about May 1945 and accelerated with the start of Plant 6 in 1946.

During and after the war, African and Canadian ores were milled to black oxides elsewhere and at Mallinckrodt (DOE 1997, Mason 1958a); at Mallinckrodt, this moved from Plant 2 (apparently) to Plant 4 (AEC 1945f; AEC 1945h; AEC 1945i) and then to Plant 6 after the latter began operation in 1946. Because of the dust hazards of the pitchblende ore, AEC had Mallinckrodt develop a method to permit automatic unloading and loading of the pitchblende ore in the grinding operation (AEC 1948f). Mason (1958a) gives data showing very high dust levels for ore milling, so the process was apparently not an entirely enclosed one. Ore milling stopped in mid-1949 (Mason 1958a) and after that all ore arrived mill-processed. The ore delivered to Mallinckrodt after that apparently still had to be ground to some extent (e.g., AEC 1947a).

The continuous countercurrent ether extraction process was developed in 1945-1946 and put into production with the start of Plant 6 in 1946 (Fleishman-Hilliard 1967). This eliminated the necessity of moving the intermediate materials batchwise from one process location to another as had been necessary at Plant 6; rather, the materials passed from vessel to vessel or filter via piping. There was apparently still some possibility of "splatter or slopping over" at some point if the chemical reaction was too violent, but this would not be true of the column extraction process because it was completely enclosed (AEC 1949e). The sulfur removal step was added when ores containing high levels of sulfur were used. However, the removal of products such as UO_3 and UO_2 was usually done by manual scooping and the removal of waste products, such as the filtered-out solid wastes and recovery products, was usually done by manual methods such as scraping.

As noted earlier, Plant 6 was shut down for a time in 1949-1950 for modifications; it seems likely that the end of ore milling coincided with this shutdown. Mechanization improvements during this period decreased the amount of manual and close-in handling of ore and the other uranium forms, but there was still a significant amount, especially with regard to the intermediate solid forms (UO_2, UO_3, and UF_4) produced as particles and chunks of material. Also, during the first year of operation (1946-1947), it had become apparent that not all of the uranium in the pitchblende ore was being dissolved in the nitric acid step, but was remaining in the "K-65" or gangue sludge residue that was to be stored and returned to African Metals Company; a process was developed and installed in 1948-1949 to allow greater capture of the uranium from the ore (see Section 4.7).

It was realized that the UO_2-to-UF_4 production in Plant 4 was particularly problematic from a dust point of view and AEC therefore had Mallinckrodt embark on an experimental program to produce a continuous-process reactor for the production of UF_4 (AEC 1948f).

4.4 THE LATER POSTWAR PERIOD (1950–1958)

Milled ore was sent from Middlesex to Mallinckrodt up to 1955, when the Middlesex work was transferred to the Fernald plant , which thereafter was the sender of milled ore to Mallinckrodt ((DOE 1997). An Ore Room and addition were added by 1950 to Plant 6. Early on, there was grinding of ore in a rod mill and opening, dumping, cleaning, and closing of drums (MCW 1950u); later, the duties of the operator included opening (deheading) and cleaning ore drums (AEC 1953). Hence this was not simply a storage area. During 1950, the average concentration of raw materials arriving at Mallinckrodt dropped to less than 50% (as U_3O_8), compared to ~65% during 1947-1949 (AEC 1951b). This was due to a gradual decrease in the content of the pitchblende and to the introduction of the magnesia concentrate MgX, which was produced from low-grade tailings. The new feeds had higher impurity content and the MgX contained 25-30% moisture compared to the normal pitchblende content of less than 3%. In order to maintain the production level, a new extraction column was designed and installed by Mallinckrodt in December 1950 to permit direct solvent extraction of the MgX slurry without prior filtration.

During the period 1950-1951, Plant 6E and Plant 7 went into operation as previously described and a number of further process and equipment changes were made to reduce exposures, principally in the area of dust control and mechanization (see Table 3). The main reason for the provision of new plant space was to reduce exposures, but it is notable that in each case, production increased markedly after a new plant was added, sometimes to several times the original anticipated capacity (Mason 1958a). Thus the exposure-reduction changes were sometimes successful and sometimes not. In late 1950 or 1951, in order to increase production, Plant 6E was placed in six-day, three-shift operation as quickly as new crews could be trained (AEC 1951b), while Plant 7 was designed to operate on a six-day, three-shift schedule.

AEC (AEC 1951b) stated the following. In 1951, the Plant 4 UO_2-to-UF_4 conversion equipment consisted of six large gas-fired furnaces containing five reactor banks each. The individual reactors were composed of a graphite tray assembly inside of a steel shell. The Plant 4 UF_4-to-biscuit equipment included six individual gas-fired furnaces, and the biscuit-to-ingot conversion equipment included four high-vacuum induction furnaces. The Plant 6E UF_4-to-biscuit equipment included 16 electrically heated induction furnaces and the biscuit-to-ingot equipment included six ultrahigh-vacuum furnaces. In Plant 6E, operations were mostly remotely controlled and/or totally enclosed, for health and safety reasons. In Plant 7, there were three parallel reactor banks, each made up of one UO_3-UO_2 reactor tube and three UO_2-UF_4 reactor tubes, with the four reactors making up the bank operated in series. Each reactor tube was equipped with a ribbon screw that moved the material continuously through the reactor into the feed area for the next tube. A single UO_3-UO_2 tube was installed to provide for the production of UO_2 alone. The later expansion called for additional UO_2-UF_4 tubes to be added.

Some time prior to August 1954, the Ore Room and K-65 sampling operations in Plant 6 appear to have ended, probably due to the end of high-grade ore processing. The Plant 6 pilot plant was constructed in 1949-1951 and began operation at some point during that period (Fleishman-Hilliard 1967); references to it started to appear in AEC air dust study reports by 1953 (AEC 1954c). The purpose of this pilot plant was said to be process and product improvement (Fleishman-Hilliard 1967). An AEC dust exposure report giving time-and-motion information states that a Pilot Plant 6 technician was cleaning old MgF_2 out of a kiln, putting in new MgF_2, and scooping Anaconda feed (sodium diuranate) into a digestion tank (AEC 1956b). Thus it is likely that the pilot plant was working on the development of improved methods for various parts of the refining processes. The research laboratory was constructed and put into operation during the same period as the pilot plant (Fleishman-Hilliard 1967).

The Plant 6E operations became more enclosed and more mechanized over the years. In 1955, the billet (ingot) postprocessing grinding and inspection method in Plant 6E was described as follows (MCW 1955q); it is likely that this was the method used from the start but with the addition, over time, of the grinding enclosure for operation. The operator moved the billet from the water cooling tank into grinding position by means of a remote-controlled hoist. Through canvas gloves installed in the enclosure wall, he ground off the fin (raised line) from the mold joint. Using a remote-controlled piston, he then ejected the billet from the grinding rollers on the scale through a small opening in the enclosure. Using a rag, he wiped off the excess black oxide as he rotated the billet slowly, checking it for defects. He then moved the billet by hoist onto the billet cart to move it to the saw for a sample to be taken.

An alternative method of producing the metal form was developed at Plant 4 in the mid-1950's. This process (AEC 1956a; Fleishman-Hilliard 1967) took the UF_4 to the final cast (ingot/billet) form in one step instead of two, eliminating the need for recasting and the associated impurity contamination. The result was a massive single ingot, called a dingot, weighing about 3300 pounds. In this process, a bomb was lined at the bottom with MgF_2, a mandrel (mold liner support) was inserted into the bomb, more MgF_2 was poured into the space between the bomb shell (wall) and the mandrel, and the bomb was jolted to pack the liner hard between the mandrel and shell. The UF_4 and MgF_2 were blended and put into a drum, which was then capped and removed to a charging station. The drum was inverted over the station insert collar, the drum cap valve was opened, and the charge was allowed to flow into the bomb. During this step, the operator used a long stainless steel rod to "pole" the charge down into the bomb; the operator also used a mechanical rammer to tamp it down for maximum density. Recycled slag containing MgF_2 and U was added to the top of the charge and tamped. Finally, the exterior of the bomb was vacuumed and a steel lid applied. The bomb was removed by hoist to a "Hevi-duty" furnace, where the thermite (metal-metal reduction reaction) took place.

Because of the size and purity of the dingot, the postcasting processing differed from that of the derby and billet. The bomb was cooled in air, then transferred to a breakout enclosure where it sat over a downdraft-ventilated floor grill. The bomb was inverted over the grill and the contents jolted out onto the grill. The slag liner was broken with a mechanical sledge hammer and swept onto the grill and down into a hopper. A conveyor took it to a grinder, from which it was discharged into drums and taken for reprocessing (presumably in Plant 6E -- see below). The dingot was cleaned with a pneumatic chipping hammer over the grill and then removed to a machining area, where it was "scalped" (had the outer surface trimmed off) on the top and sides with a vertical turret lathe. If samples were needed, it was then taken to a saw area and sections were cut off with mechanical hacksaws. Finally, the dingot was put into a salt bath and heated for several hours, then put on the bed of a 100-ton forging press that had a mechanical manipulator for positioning. It was pressed into a slab for several minutes, then returned to the salt bath for about an hour for reheating. This process was repeated four more times, with the piece being rotated on the press between passes. The forged slab was then quenched and taken to storage.

Fleishman-Hilliard (1967) implies that Mallinckrodt also produced extruded billets from these dingots, at least on an experimental basis. Once extruded into a much longer and thinner cylinder, the billet was cut into shorter lengths.

Plant 7 was built not only to increase production of UF_4, but also to take advantage of the new continuous process using a "stirred bed" reactor for producing it (Mallinckrodt 1994, Fleishman-Hilliard 1967). At some point not long after this process went into operation, the process was modified to allow production of UF_4 continuously from a UO_3 feed instead of a UO_2 feed, thus eliminating a transfer step (Mallinckrodt 1994, Fleishman-Hilliard 1967). At this point, UO_3-to-UO_2 production at Plant 6 seems to have ceased or at least to have decreased significantly.

By 1955, Mallinckrodt was using a variety of feed forms from many different sources to produce the various uranium product forms. The table below (AEC 1955a) illustrates this for a typical period in 1955; figures are in tons.

Feed forms and quantities used for a period in 1955.

To be produced:	**January**	**February**	**March**	**April**	
QM-2	500	465	535	510	
TA-7	280	355	320	320	
TM-5	205	240	270	260	
Shipments to NLO:					
QM-2	235	155	170	170	
YM-5	All finished metal of the metal plant plus all shippable metal from the pilot plant				
TA-7	0	100	100	45	
Feed type	**January consumption**	**January 31 inventory**	**February receipts**	**March receipts**	**April receipts**
Q-11	76	8	86	---	---
MgX	26	10	40	64	30
South African	---	---	129	80	90
Portuguese	21	---	40	---	20
Colorado soda salt	46	12	50	50	50
Beaver Lodge	23	31	28	28	28
Vitro	9	---	---	---	---
Colorado black	92	12	50	50	50
Canadian black	43	17	25	25	25
NLO Recycle/Scrap Plant	27	15	20	20	20
MCW	18	---	15	15	15
Miscellaneous	---	---	---	---	---
Dissolver	24	77	20	20	20
Sawdust	5	6	5	5	5

A list of process cells, dust collectors, and tanks appears in MCW (MCW 1955g).

4.5 OTHER PROCESSES

Some uranium recovery operations consisted of processing solid scrap wastes, such as the portions of billets removed as assay samples, to recover the valuable uranium and thus to maximize the uranium obtained per unit quantity of ore. This was probably done in the early years by digestion of the scrap in nitric acid (FUSRAP 2003a). Sawdust (from the sawing of uranium metal billets) was stored under oil until it could be processed in this way and converted to ingots (Fleishman-Hilliard 1967; MED 1944o; MED 1944p); the sawdust was also recovered by oxidizing it at high temperature to U_3O_8, and was then sending it back to Plant 6 as a feed material (AEC 1949g). As mentioned earlier, some uranium was recovered from neutralizing excess HF in the UF_4 production process (Mason 1977).

In early 1954, a slag separation plant was built at Plant 6E, apparently as a six-level wing (116C) on Building 116, to recover most of the uranium content from the MgF_2 slag produced there in the UF_4-to-derby operation (AEC 1959, AEC 1954f). The slag was fed into a jaw crusher on the first level, then the crushed slag was conveyed on a “vibro conveyor” to a bucket elevator and on to a roll mill hopper on the 6th level. From there the slag was gravity-fed via a roll mill feeder into a series of roll mills and screens on the middle levels. Reject material from the last mill screen flowed by gravity feed into a reject hopper and then to a reject drum on the 1st level. Discharge streams from the roll mill and the roll mill screens were fed into a ball mill. The discharge from the ball mill was conveyed by a bucket elevator to a series of air classifiers; after passing through these, the separate product and reject streams flowed by gravity feed to the respective drums on the first level.

In late 1955 or early 1956, operations to process slag began in Building 701 in Plant 7. This building was then called the Slag Separation Plant or the Slag Processing Plant. According to AEC (1956d), the feed material was uranium concentrate from reject MgF_2 material (RMF). The RMF was charged from drums into a skip hoist that discharged into a hopper serving a rod mill. In the mill, the RMF was crushed and water was added to make a slurry. This was passed through a mechanical screen shaker; the part that did not pass through was recycled through the rod mill and the part that did pass went onto three successively smaller-sized Wolfey gravity feed tables. The tables separated the uranium-bearing slurry from the rest of the slurry, with the former being allowed to run off the last table into a screw conveyor. The slurry was then fed into a drum. This drummed material was referred to as “U-Con” or “U-CON”. The non-uranium-bearing slurry was pumped to settling tanks and then to a rotary filter; the resulting cake (D-701) was discharged into dumpsters (buggies) and taken to the SLAPS waste storage site.

Little information could be found about shipping, receiving, and storage operations and about the milling of the UO_3, except that it was apparently done in Plant 2 until the work moved to Plant 6 (Mason 1958a). See Sections 5.2.1-5.2.3 for more details. The ore areas and residue storage areas appeared to be separate from the product warehouse areas, however.

Similarly, regarding the reversion of UF_4 to UO_2 or UO_3 (or U_3O_8) there was little information except for a short reference in an AEC Plant 7 air dust study report (AEC 1954g) to the work of a panel board operator’s work: the operator was said to vacuum “C-31 material” from a drum into a “reverter”. What the C-31 material consisted of was unclear, but often the “C-” designated scrap material or waste material captured in a dust collector. Later such AEC reports also mention that the reverter was being used by this operator (AEC 1955d, AEC 1956b) but not an earlier report (AEC 1952a), so presumably this work continued from at least 1953 (the date of the AEC 1954g survey) to the end of Plant 7 operation.

There was no information about processing very low enrichment (VLE) uranium beyond what was given in Section 3.0 above, except for a reference in an AEC air dust study report for Plant 7 in which it was stated that the subject AEC survey covered “health and safety problems existing during production and processing green salt, with the added operation of processing enriched uranium”; the panel board operator was said to include among his observed duties “charging enriched UF_4 into hopper” and “replacing enriched material drum and sample bottle” (AEC 1955e). Since there was no further mention of any of these activities in later such reports, it appears that this operation may have been performed for only a short period of time, e.g., to use up excess UF_4 from another site. There is no indication that Mallinckrodt itself produced the VLE UF_4. In any case, the operations described appear to have been of a routine process nature.

There was no information at all about the production of UO_2F_2 or the processing of (supplied) uranyl nitrate hexahydrate, both of which functions were mentioned by only a single reference source (see the summary list of work in Section 3), with no details given. There was information (AEC 1955d) regarding an experimental process begun in Plant 7 in 1956 in which MgF_2 liner (MFL) was treated to reduce the hydrogen content for use in dingot bombs. This created fluorinated MFL (FMFL, perhaps also called D-30). However, this process appeared not to involve any radioactivity except what would likely have been present in the ambient air due to other processes. Also, Fleishman-Hilliard (1967) commented that the reason for adding a little UO_2F_2 to a bomb was to slow down the process thermally, resulting in a better separation of slag and metal.

An experimental permit was issued in July 1956 that allowed the pickling of 14 tons of high-carbon scrap metal in Plant 6 , which was done on 29-30 July 1956 without the health and safety department's knowledge (MCW 1956f). It is not known if any more of this work was done.

Mallinckrodt (1994) estimated that small-volume batch and experimental uranium-related processes constituted less than .3% of the total uranium produced. Some of the processes considered in this estimate were the conversion of slightly (very low) enrichment feed materials to metal or uranium nitrate liquor, extraction of Th-230 from pitchblende raffinate, production of U_3O_8 and UO_2 using an experimental continuous denitration furnace, experimental extraction of uranium using tributyl phosphate to replace ether, production of uranium metal dingots, and recycling of slag for use as liner material. This supports an assumption that only a negligible part of total processing activities involved such short-term and usually low-volume processes.

Regarding specific laboratory sampling and assay processes, there is limited information available. This is given in Section 5.2.4.

4.6 ORES AND OTHER FEED FORMS

In the early years, the ore arrived as a milled concentrate from Canada (Mallinckrodt 1994), consisting mostly of black oxide (U_3O_8) (Mallinckrodt 1994; Fleishman-Hilliard 1967); MED (1945) stated that the Eldorado mine site in Port Huron, Canada, was a source of black oxide to Mallinckrodt. These concentrates were produced at offsite uranium mills and were free of radium and its decay products (Mallinckrodt 1994). That is, in the early years, Mallinckrodt did not process the high-radium-containing ore of later years. Although the uranium was mostly supplied to Mallinckrodt in already milled form (as black oxide), some milling and pre-processing of high-grade uranium ore was performed at Mallinckrodt (FUSRAP 2003c).

The Vitro plant in Canonsburg, Pennsylvania produced sodium diuranate ($Na_2U_2O_7$), also called soda salt, some of which was sent to Mallinckrodt for refining to UO_2 and UO_3 (DOE 1997, MED 1945a; MED 1943e); other soda salt may have come from Belgium and Port Hope, Ontario (MED 1943e). This appears to have been digested in the same fashion as the black oxide. Soda salt and other non-ore feeds were referred to as "soluble feed".

Other wartime ore sources were pitchblende ores from Canada (Radium City in the Great Bear Lake Area and Port Hope in Ontario) (DOE 1997; Eisenbud 1975). Later in the war, domestic ores were used also; the principal Colorado source during the war was not the carnotite ore itself, but tailings from vanadium mining and milling, shipped as a concentrated sludge (Eisenbud 1975).

In about 1944, AEC wanted to increase production of uranium, not only by increasing capacity but by using high-grade pitchblende ores. However, such ores had the disadvantage that they contained a high level of radium as a decay product of the uranium. This entailed significant gamma and airborne exposure hazards. It was because of the limitations of Plant 2 with respect to capacity and hazard that Plant 6 was designed and built (Mallinckrodt 1994). Another consideration was that crude concentrates and pitchblende ores tended to have undesirably high levels of sulfur, which necessitated the extra precipitation step for removal (Fleishman-Hilliard 1967). The high-level ore was used as a principal feed material from May 1945 until early in 1955 (AEC 1956e; AEC 1959), when the plant switched to processing various types of concentrates. However, MED (1943e) remarked in 1943 that there appeared to be more "radiation" in the U_3O_8 being received by Mallinckrodt from Port Hope, suggesting that pitchblende ore concentrates may have been used as feed prior to May 1945. It is known that experimental processing with pitchblende ores was done in 1944 at Plant 1 (Mallinckrodt 1994), so this may explain the 1943 reference.

Most of the high-grade pitchblende ore processed by Mallinckrodt was obtained by AEC as a concentrate from the Belgian Congo in 1944 (AEC 1967), the so-called African ore. There is some disagreement as to how it came to St. Louis. DOE (1997) states that from 1943 on, the receiving and storage facility operated at Middlesex, New Jersey (DOE 1997) assayed, crushed, riffled, and redrummed the ore as it came into the United States, then sent it to the various refineries, including Mallinckrodt (AEC 1949b, Eisenbud 1975). MED (1945n) states that the African ore came through the Eldorado (Port Huron, Canada) area for processing before being sent on to the US refineries. AEC (1967) states that the concentrate was shipped from the Belgian Congo to St. Louis in 55-gallon drums (but this could mean that it came by way of Middlesex). Whatever the case, it was not likely that significant milling of this ore (as opposed to simply grinding to somewhat smaller size) was done at Mallinckrodt. In any case, it arrived by rail in carload lots and was stored in Building 110, the warehouse (Mason 1977).

After World War II, foreign uranium ore was supplied from the Belgian Congo, Canada, Australia, South Africa, Portugal, and other nations. Besides the foreign ore, domestic carnotite ores began to be mined directly, milled elsewhere, and then sent as ore concentrate (mainly U_3O_8) to Mallinckrodt. Early postwar domestic ore supply areas included Uravan, Durango, Grand Junction, and Naturita, Colorado, and Monticello, Utah (DOE 1997). Once the US began to stimulate domestic mining and milling in 1948, the proportion of domestic ores used appears to have increased; thus the overall concentration of uranium in the ore may have decreased over time. The ore was usually stored in the warehouse or in another dedicated area on site, but presumably because of the volume of the pitchblende ore (Q-11) and its hazards, from about mid-1948 on at least some of it was stored at SLAPS (AEC 1949d) in a dedicated shed or shack (MCW 1949g; AEC 1949d, which states that ten 25-ton lots were shipped to Mallinckrodt in May 1949 and an additional twelve lots were sent to SLAPS).

From 1955 on, ore concentrates were mostly used. This included some Belgian Congo ore tailings concentrates (resulting from the leaching of raw ore during pre-processing in the Belgian Congo in previous years before the ore was shipped as a concentrate) (AEC 1951b). Soda salt appears to have been sent from several sites; a description of some work indicated that Fernald, Durango, and Anaconda diuranate were being handled by a soluble feed operator (AEC 1956b) and as noted Vitro also supplied soda salt until it was shut down.

4.7 RESIDUES AND EFFLUENTS

The radium-bearing pitchblende residues (wastes) were always stored separately from the non-radium-bearing residues and the barium-bearing and other types of cake (AEC 1967); thus the external exposure implications of handling residue waste depended on what kind of waste it was.

From 1942-1945, MED solid wastes were stored temporarily at the Mallinckrodt site and then shipped to the Lake Ontario Ordnance Works in Lewiston, New York (Mallinckrodt 1994). The radium-bearing pitchblende residues (K-65 or gangue lead cake) were shipped by rail to the Middlesex, New Jersey repository for storage for a time, up to 1946 (AEC 1949b). But from the time AEC acquired the SLAPS site in 1946 and up through 1948, all residues produced at Mallinckrodt were stored temporarily in Building 114 of Plant 6 until they were shipped by truck to SLAPS (MCW 1949p; AEC 1967; Mason 1977; Mallinckrodt 1994). From 1948 to at least 1949, the K-65 residues were shipped to Middlesex (AEC 1949b) or to the Lake Ontario Ordnance Works (AEC 1959)after being drawn back from SLAPS and reprocessed. After about 1949, K-65 was shipped to the Lake Ontario Ordnance Works (FUSRAP undated a; AEC 1959; AEC 1949b); although Mallinckrodt (1994) says that these were still sent to SLAPS up to 1956, it was clearly only the other types of residues and wastes that were sent after 1949, not the K-65. Some uranium scrap, including the mainly magnesium fluoride slag immediately above the derby (in the metal reduction step) was sent to the Vitro Manufacturing Company for uranium recovery (AEC 1949b).

Initially, it was only the K-65, the barium sulfate residues (AJ-4 or “airport cake”), and the pitchblende raffinates (AM-7) produced at Mallinckrodt that were stored at SLAPS (Mallinckrodt 1994). The K-65 was trucked in drums (AEC 1947a), while the barium sulfate (AJ-4) and pitchblende raffinate (AM-7) cakes were collected in dumpster-type containers, loaded into AEC dump trucks, and transported on a daily basis to SLAPS (Mallinckrodt 1994). The K-65 drums were stored in a dedicated shed structure (AEC 1947a), but the other residues were simply dumped on the ground in piles (AEC 1947f; AEC 1947a; AEC 1947e; AEC 1949b) because they were insoluble in water and were thus regarded as fairly immobile (AEC 1947a).

Later, SLAPS was also used to store other types of residues and wastes and some ore and to serve as a storage and burial site for contaminated objects and debris, such as empty drums, Rauschig rings from the ether extraction columns, bituminous floor materials, and some materials from the decontamination and decommissioning of Plants 1 and 2 (AEC 1959; AEC 1956d; Mallinckrodt 1994; AEC 1949m). As noted earlier, the other types of residues included the AM-9 residue left after reworking of the AM-7 to recover thorium; Colorado (domestic carnotite and other low-grade non-pitchblende) ore raffinate (AM-10); and the precipitate residue of the ether extraction columns (“Sperry cake”, apparently a subset of the AM-7) (AEC 1956e; AEC 1967; AEC 1964; Mallinckrodt 1994). There were also small amounts of other raffinates present at the SLAPS waste storage site from the processing of lower-grade uranium ores (AEC 1967). The material obtained from processing carnotite and other low-grade domestic ores was kept separate from the K-65 (AEC 1967).

Metal plant waste, such as metal scrap, dolomite slag (“C-liner” or “C-oxides”), and tailings of uranium scalping operations from magnesium fluoride slag were also sent to SLAPS (Mallinckrodt 1994). The C-liner consisted was mainly used and nonrecyclable dolomite liner with less than 2% uranium content (FUSRAP undated b; AEC 1949b) and resulted from the separation of slag in the reduction (bomb) step in the metal plant (first in Plant 4, later Plant 6E). This waste was created until early 1953 when the dolomite liner was replaced by a recycled magnesium fluoride liner (FUSRAP undated b). There was another bomb waste called “C-special” (AEC 1949b), which may be the same as the C-liner slag since it also originated in the bomb furnace. Yet another bomb waste was the recyclable magnesium fluoride slag that remained after “scalping” the uranium content from the magnesium fluoride slag, beginning in 1955; this was called C-701 (FUSRAP undated a).

Within the Mallinckrodt plants, most “product” (UO_3, UO_2, and UF_4) was packaged into 75-lb paper sacks and then into a barrel. There was usually a fine film of uranium material clinging to the sacks, so they were burned in an incinerator and the ashes sent to recovery (MED 1944o). Also sent to a recovery system were floor sweepings and spillages, swept into sumps; scrapings off the walls and equipment; and dust collected in collectors, usually by vacuum systems (MED 1944o).

Some reprocessing of residues was also done to recover uranium. AEC (1951b) states that following the development and installation of a sodium carbonate/sodium bicarbonate leach process in 1948-1949 to recover more of the uranium , the K-65 produced up to that point was brought out of storage at SLAPS in lots and reprocessed in about 1949 (MCW 1949o and AEC (1959) imply 1948-1949), resulting in a final uranium content in the residue of 0.05%. This was apparently done in the same vessels and in the same general manner as ore would be processed, including heating the K-65 drums in the thawing oven used to thaw ore drums (MCW 1949o). The new residue was sent to the Lake Ontario Ordnance Works (AEC 1959) to await return to African Metals and the corroded drums in which the K-65 had been stored were returned to SLAPS for storage (AEC 1949m). At least some of the barium sulfate cake was reprocessed similarly (AEC 1951b; AEC 1948g); however, the resulting barium sulfate residue was taken back to SLAPS because (eventually) African Metals relinquished ownership of it (AEC 1959). Some of the Sperry Cake, a good source of protactinium-231, was brought back from SLAPS and shipped to Mound, which processed approximately 20 tons (about eighty 55-gallon drums) of it and obtained approximately two grams of protactinium-231 (AEC 1967).

In late 1954 or early 1955, a new subplant, called Plant 7E, was established. This was part of what was referred to as the Minor Elements Production (ME or MEP) facility that did smaller-scale processing and development in Plant 7. The purpose of 7E was to process some of the AM-7 (pitchblende) residues that had been in storage at the SLAPS site. (FUSRAP undated a; AEC 1967; FUSRAP undated b). Starting in 1955, the thorium-bearing AM-7 raffinate residue was brought back from storage at SLAPS, stored at Plant 6, and conveyed by dumpster from Plant 6 to Plant 7E as needed (AEC 1955b). The processing was first done on a crash basis in early 1955, apparently on a laboratory level (AEC 1955c; ORAU 1991), to produce a solution containing several hundred grams of thorium. Mallinckrodt later undertook the process on a pilot scale, but insisted on only a limited number of workers and strict safety precautions (ORAU 1991). The resulting residue, AM-9, was sent back to SLAPS (AEC 1959).

AEC (1955b) described the thorium solution extraction process as follows. The AM-7 was first digested in nitric acid in a tank and filtered. The resulting “liquor” was then processed in a TBP (solvent) contactor, forming an aqueous phase and a thorium-bearing TBP phase. The TBP phase was treated with hydrofluoric acid, leaving another aqueous phase, a stripped TBP phase, and thorium fluoride (apparently in solution). The thorium fluoride was sent to the Hot Lab in Plant 6, where it was treated with aluminum nitrate ($Al(NO_3)_3$) and a pentaether-ether mixture, forming thorium nitrate ($Th(NO_3)_4$) and impurities. The thorium nitrate was stripped from the impurities, yielding the solution that was sent to Mound. The various waste streams were treated in several ways (e.g., the aqueous fractions were treated with lime). This left various cake, slurry, and water filtrate forms; the first two types were sent to storage and the latter to the sewer. The main residual cake, called AM-9, was sent back to storage at SLAPS. From the beginning of 1955 into 1957, a total of 350 tons of the AM-7 was processed (AEC 1959; AEC 1967). (ORAU 1991 says the processing was performed in 1958, but this seems unlikely). Note that the Th-230 was also called “ionium” and was referred to that way in Mallinckrodt records.

As noted earlier, significant earthmoving operations to cover buried contaminated items were done in 1952 (AEC 1964); the tailings from the magnesium fluoride slag processing were sent to the Fernald site in 1959 (AEC 1959); and in 1959-1960, some of the scrap and rubble from the demolition and decontamination of the Mallinckrodt St. Louis site facilities was buried at the site (AEC 1972; ORNL 1979). There were separate sets of sewers running from Plants 1 and 2, Plant 4, and Plants 6, 6E, and 7 out to the streets, under the streets, and out to the Mississippi River (Mallinckrodt 1994). In 1949, about 3 million gallons a day of liquid effluent classed as alkaline filtrate was being sent to the Mississippi River out a drain pipe, presumably a sewer; this was mostly cooling water, but some was process waste containing up to 12 pounds per day of uranium (AEC 1949b). A small additional amount of acid waste flowed to the Mississippi via an acid sewer (AEC 1949b); Mason (1977) states that excess HF (from UF_4 production) was neutralized with lime or potassium hydroxide and sent to process sewers, so it is unclear what the acid referred to by AEC (1949b) might have been. Mallinckrodt (1994) stated that a 1956 description of Plant 6/6E/7 operations showed that 12,000 gallons per day of raffinate filtrate from the Plant 6 pitchblende extraction was being discharged to the sewer. These lines were apparently underground for the most part, but it is possible that some of them were aboveground where they ran inside the site (see the Table 1 Notes column).

5.0 RADIOLOGICAL CHARACTERISTICS, CONDITIONS, CONSIDERATIONS, AND AVAILABLE DATA

As AEC (1950a) observed, radiation measurements or evaluations of dust exposure for the purpose of dose determination were not made in the Mallinckrodt plants in the first few years of operations because it was expected that the processing of uranium ores and compounds would involve little risk of radiation injury. This was because of the low specific activity of uranium and because of what was thought at the time to be the temporary nature of the work. Thus only a few instantaneous measurements were made and time-weighting was generally not considered. However, when AEC's New York Operations Office (NYOO), which oversaw the Mallinckrodt work, evaluated the potential hazards (which at this time included those of radium-bearing pitchblende ores), they determined them to be "considerable" with respect to the long term (AEC 1950a). NYOO and Mallinckrodt began a program of workplace and personnel monitoring.

AEC and Mallinckrodt had already begun to issue film badges in 1946 (AEC 1950a), with apparently a small-scale effort begun in late 1945. To this was added breath radon determinations in 1945 and a formal dust measurement program in 1948 (AEC 1950a). Although urinalysis for uranium was done as a "screening experiment" in acute and chronic exposures as early as 1944 (MED 1944j), a routine urinalysis program appears to have begun only in 1947 also or possibly 1948 (MCW 1950c) (see Section 5.3.6).

Some extrapolation of existing data to cover unmonitored periods is necessary, as AEC itself tried to do (AEC 1950a). Also, data must be analyzed to allow missing dose to be calculated for individual workers where there are gaps in the monitored period. The sections below provide information as to the available data and other information that will allow this to be done.

AEC thought that as a result of improvements planned for 1949 and early 1950, there would be no whole-body radiation exposures greater than 300 mrep/week in Plant 6 and the dust concentrations would be reduced to the AEC's "preferred level" of 50 $\mu g/m^3$, or 70 dpm/m^3 (AEC 1949b). AEC was also expected that construction of a new metal plant (Plant 6E), in which UF_4 would be reduced to metal as was currently done at Plant 4, would produce satisfactory (occupational) environmental conditions (AEC 1949b). It was also expected that in 1951, the new UO_2-to-UF_4 plant (Plant 7) would further reduce exposures (AEC 1949b). However, with the increase in production, these goals were not met in all cases, although there were some successes and although doses and air concentrations did decrease overall. The effects of the various plant changes and improvements were reflected in the airborne and external exposure levels, as shown in the text and tables below.

An AEC effort in evaluating dose to workers is documented in AEC (1950a), which is a report of an attempt by AEC in late 1949 to estimate the cumulative exposures of Mallinckrodt workers at Plants 4 and 6 who were working at any time between July 1942 and 1 October 1949 and who had more than six months of exposure to radioactive materials. The base data came mostly from Mallinckrodt through its health physics staff, who did some calculations and sent them to AEC (e.g., MCW 1950c). This study is important because it was apparently AEC's first large-scale study of chronic exposure to all types of workers; ORAU (1977) commented that this was the best information available about individual employment dates, jobs, and dust concentration during the period since more than 90% of all full-time uranium workers were included; also, the exposures were chronic and potentially sizeable -- production was in thousands of tons per year and the Mallinckrodt main facilities were "factories, not laboratories" (ORAU 1977). Of the 650 workers included in the study, 99 were working in Plant 4, 294 were working in Plant 6, and about 250 had previously terminated (ORAU 1977).

However, the potential usefulness of this data is lessened by the fact that the Plant 4 work records were somewhat deficient and Mallinckrodt was thus not able to evaluate past dust exposures for workers who had terminated employment or who had transferred out of the AEC project work prior to 1 October 1949 (MCW 1950c). Also, some of the estimates were difficult for Mallinckrodt to do because the particular type of work was variable (e.g., maintenance work) (MCW 1950c). Mallinckrodt used the data of 1948 to calculate exposures prior to 1948 although they thought that many of the exposures were likely somewhat higher (MCW 1950c). Finally, since reported dust concentrations were based on a gross alpha measurement rather than on a chemical (element-specific) basis, the reported concentrations and the doses calculated from them might not always indicate a strictly uranium exposure; it was known that some workers, such as those working with K-65 residue, were exposed to radium dust (beyond that normally present in uranium dust as per uranium-radium parent-daughter ratio) (MCW 1950c). Still, correlation of the urinary uranium concentrations with calculated dust exposures was considered to be good for Plant 6, although poor for Plant 4 (MCW 1950c).

AEC's estimates for the dose to the lung were based on air samples of alpha-emitting dusts (translated to a daily weighted average exposure level); to the bone, on breath radon analysis (to determine the fixed radium burden); and to various organs, on film badge data (AEC 1950a). ORAU (1977) stated that there was a Mallinckrodt lifetime tolerance dose exposure level, but did not state what the level was; however, it did report that by October 1949, 52 employees had accumulated 100% of this level, with 45 exceeding 100% of it and 10 exceeding 200% of it.

Plant 6 was the most hazardous area in terms of dust levels, radon levels, and external gamma dose. Both Mallinckrodt and AEC made periodic studies of the various areas and continuous incremental improvements were made over the years of operation. An AEC consultant noted in 1949 that film badge records were well kept and the external exposures could be seen to correlate with the amount of radium in the plant (in ores and residues); despite problems with airbornes, progress had been made in reducing external dose and airborne dust levels (AEC 1949k). However, in October 1955, a Mallinckrodt health and safety official was asking an operations manager what had happened to Plant 6, listing in detail the many areas that were dirty and had leaking or nonfunctional equipment and the many questionable practices and violations of requirements (e.g., doors left open and respirators not being worn) that were resulting in unnecessary exposures to workers, particularly dust exposures (MCW 1955d). This official stated that many of these items had been pointed out repeatedly, to no avail. It appears that by this time it was clear that Plant 6 production would be moved elsewhere within a few years and either Mallinckrodt or AEC felt that it was not worthwhile to make improvements in the plant or even to maintain it adequately. Thus although the various hazards were progressively identified and addressed over the course of the years of operation and exposure levels did go down significantly, in the last few years some hazards were allowed to continue for extended periods before being addressed and some exposures were seen to go up. Even so, it appears that some studies and improvements were made very late in the life of the plant (e.g., the cleanup following the January 1957 study of radon and gamma levels in the Cloth Storage Room (MCW 1957)). Because of this somewhat up and down safety history, it is important to consider what was going on at the plants in any given year in order to assess the dose to an individual.

5.1 UNITS, LIMITS, AND RECOMMENDATIONS

The external exposure (dose) units used by MED/AEC during most of the relevant period were milliroentgen (mR) for gamma doses and millirep for beta doses, with the rep being equal to 0.93 rad; the abbreviations in the film badge and other records were mr and mrep respectively. Air exposures were expressed as disintegrations per minute per cubic meter sampled or inhaled (dpm/m^3), while radon exposures were usually given in units of 10 x 10^{-10} pCi/L. AEC also expressed exposures in terms of "tolerance" levels set on the basis of assumed safe levels; a tolerance level was equal to a given external dose level in mR per week, mrep per week, dpm/m^3, etc., but the given value changed with time as AEC set the safe levels lower and lower. The tolerance was regarded as a recommendation rather than a rigid limit, but AEC pressed the contractors to get and keep exposures to below the tolerance level. The units and the tolerances are further explained below.

In the early days of the war, MED decided that its tolerance levels of external exposure for uranium processing would be 0.1 R per eight-hour day or 0.6 R per week for gammas and 0.5 R per eight-hour day or 3.5 R per week for betas (MED 1943c, MED 1945b). This was based on the findings of the University of Chicago that the gamma dose rate from an infinite plane source (a 2-π geometry) of uranium metal was 0.04 R per eight-hour day and for a room of uranium metal (a 4- π geometry) it was 0.09 R per eight-hour day (MED 1943c). Later in the war, the limits were set at 700 mR per week to the whole body and 3500 mrep per week to the hands (MED 1945b, AEC 1949b; AEC 1950b). MED (1945b) stated that the entire dose for one week of either type of radiation could be received in one day or less; AEC (1950b) stated that the limit was 700 mrep per week "each of beta and gamma", as accepted by the University of Rochester in processing film badges (i.e., Rochester did not flag reported doses as above tolerance if the weekly beta and gamma doses were each below 700 mrep). MCW (MCW 1950f) states that the tolerance level as 12.5 mR/hr, i.e., the hourly average for the 0.1R per eight-hour day; presumably this was according to AEC usage as well.

At some point, when NYOO had assumed the job of reading the film badges, the tolerance level appeared to have been lowered again to 500 mrep per week, which Mallinckrodt continued to interpret as applying to either beta or gamma but not to the total (AEC 1950b). However, Mallinckrodt used a control level of 150 mrep per week, called the "preferred level" in its 1946-1952 film badge records. The number that was compared to this level was the sum of the gamma dose in mR (i.e., mr in the records) and the beta dose in mrep, as registered by the film badge, apparently with no adjustment of the mrep by the factor of 0.93. In 1947, the basic dose limit was said to be 0.1 rep/day and the relative biological effectiveness (RBE) for alphas to be 10 (Hursh 1975); probably this was true earlier as well.

In anticipation of the lowering of the radiological dose standards (recommended limits) by the national expert committees such as the National Council on Radiation Protection and Measurements (NCRP), AEC began making changes in the plants in about 1948 to meet new "maximum permissible levels" of 300 mR/week for whole body irradiation and 1500 mrep/week for beta radiation to the hands (AEC 1949b). An AEC internal memo dated January 1949 (AEC 1949c) states that the Plant 6E design by Singmaster & Breyer was intended to meet the following AEC-specified criteria: whole-body beta dose less than 250 mrep per week (based on an maximum permissible (tolerance) level of 500 mrep per week) and beta dose to the hand less than 500 mrep/week (based on a maximum permissible level of 1500 mrep per week). It was noted that AEC considered these criteria to be the minimum requirements and did not include any factors of safety beyond the "acceptable" (i.e., tolerance levels). New shielding added in 1948-1949 was designed on the basis of 50 mR per day of gamma radiation (MCW 1950e; AEC 1947b).

In August 1949, AEC established and circulated to its contractor personnel a tolerance level of 300 mrep per week, which was to be taken as the total gamma plus beta dose to the whole body. However, Mallinckrodt misunderstood that the 300 mrep limit was to be applied to the total of beta and gamma and interpreted it as the limit for either beta or gamma (AEC 1950b). Finally, in January 1950 AEC made it clear to Mallinckrodt that the limit applied to the total beta plus gamma (AEC 1950b)

In a 1950 letter to AEC, Mallinckrodt summarized AEC's tolerance levels in the postwar era, as given in the table below (MCW 1950t). In MCW (MCW 1950l), Mallinckrodt suggested that AEC use absolute units instead of tolerance units to report exposures because the tolerance values were apt to change with time and this lead to confusion in interpreting older records.

Summary of AEC tolerance levels in the postwar years.

Date	Beta tolerance level, mrep/week	Gamma tolerance level, mR/week	Total beta + gamma tolerance level, mrep/week
Before June 1948	700	700	1,400
June 1948 - August 1949	500	500	1,000
August 1949 - January 1950	300	300	600
January 1950 on	---	---	300

In mid-1950, AEC agreed to allow Mallinckrodt to interpret the 300 mrep total gamma plus beta whole-body limit as being taken as the average weekly dose over a three-month period, thus allowing the 300 mrep to be exceeded in some weeks (AEC 1950e); this was apparently based on the fact that Mallinckrodt was already using a system of personnel rotation to reduce doses (MCW 1949p; AEC 1950b). However, AEC at the same time suggested that 150 mrep per week be taken as a recommended limit for most purposes and that a weekly dose of 600 mrep be exceeded only in exceptional cases. It should be noted that in 1953, a design contractor was stating that the design criteria for ventilation and dust control equipment his company had put in at Mallinckrodt and Harshaw included a maximum weekly exposure of 300 mR of gamma radiation, with actual design predicated on half that to allow for a safety factor in unusual circumstances (Miller 1953). Subsequently, Mallinckrodt gave as the “tolerance cumulative dose” limits in use in 1955 as beta, 500 mrep per week, whole or part body; gamma, 300 mR per week, whole or part body; sum of beta and gamma, 500 mrep per week, whole or part body; and 1500 mrep per week, extremity (MCW 1955).

During the early days of wartime uranium processing, AEC/MED’s acceptable levels of exposure for the uranium processing plants for dust in air were 500 $\mu g/m^3$ for insoluble uranium salts and 150 $\mu g/m^3$ for soluble salts (AEC 1949b). In 1944 MED determined that a standard was needed for uranium dusts and adopted the air maximum permissible concentration (MPC) level for lead, 150 $\mu g/m^3$, as the interim standard (Hursh 1975).

In 1949, a University of Rochester scientific group suggested an air MPC of 50 $\mu g/m^3$ for soluble uranium forms based on (chemical) injury to the kidney and an air MPC for insoluble forms based on radiation injury to the lung (Hursh 1975). In 1953 the NCRP recommended in National Bureau of Standards Handbook 52 (quoted in Hursh 1975) a limit of 73 $\mu g/m^3$ for both soluble and insoluble forms; it was adopted. These were occupational standards that correspond to a 40-hour week (the number of hours that Hursh (1975) uses in conversions in his discussion of the history of standards).

By 1949, AEC had set a “preferred level” (also referred to as a tolerance level) of 50 $\mu g/m^3$ for uranium dust, assuming a routine exposure of 8 hours a day, 6 days a week (AEC 1949b). This was taken to be equivalent to 70 dpm/m^3 for alpha and is based on animal studies (Hursh 1975). For dosimetry reference, Appendix A gives the basis for this figure. This was later referred to as the Maximum Allowable Concentration (MAC) and was still in use as of 1953 (Miller 1953). In early 1955, AEC appears to have adopted a MAC of 100 dpm/m^3 for alpha, as AEC (1955e) stated in an air dust study report. In a 1958 report, AEC gave the limit for natural uranium, either soluble or insoluble, in air as 5×10^{-11} µCi/ml for 40 hours/wk (i.e., occupational) and 1.7×10^{-12} µCi/ml for continuous occupancy (AEC 1958, Table I). In 1959 the AEC also recommended against respirator use except in emergency situations (AEC 1949b), suggesting that before the relevant period of Mallinckrodt work, extensive use of respirators was still tolerated as a means of minimizing exposure.

However, note that in a 1958 paper by an AEC-NYOO safety official, while the wartime MAC was given as 500 µg/m^3, agreeing with the information above, the “present MAC” (i.e., in 1958) was said to be 110 dpm/m^3 instead of the 70 dpm/m^3 of other references (Breslin 1958). This may be because of the so-called “double curie” or “special curie” uranium radioactive content unit that the NCRP and others used prior to about 1 January 1974. The special curie was defined for the natural uranium mix and was taken to be 2.05 times the nominal number of disintegrations of U-238 because it included allowances for the U-234 and U-235 content (NCRP 1974). If the 110 dpm/m^3 was based on a 48-hour week, this would be equivalent to 65 dpm/m^3, or about 70 dpm/m^3, based on a 40-hour week and multiplying by 2.05 to convert from the special curie to the regular curie. The NCRP MPC value that corresponded to the special curie was given as 6×10^{-11} “special” µCi/cm^3, which agrees with the value of 5×10^{-11} given in the paragraph above if the former was based on a 48-hour week. (The NCRP abandoned use of this unit in 1973 because it caused confusion.) It does not appear that the former use of this unit has caused any disparity or confusion in the interpretation of data used in this document, but this potential problem should be noted.

The “generally accepted level” for radium fixed in the body (as a body burden) was taken to be 0.1 µCi (MED 1946c). In the highest concentrations handled by Mallinckrodt (i.e., of the pitchblende ores), 0.1 µCi corresponded to about 0.5 g of ore (MED 1946c). Since the radium was present in insoluble or only slightly soluble form, the original MED value for allowable dust concentrations of ore was 150 µg/m^3 (MED 1946c) (as mentioned above). When the tolerance for other kinds of dust was raised to 500 µg/m^3, MED apparently did not mean for the ore and radium-bearing waste dust tolerance level to rise to 500 µg/m^3 as well, but Mallinckrodt apparently assumed so, based on the statement in MED (1946b) that the maximum allowable concentration for chronic exposure for U_3O_8, UO_2, and UF_4 had been raised from 150 µg/m^3 to 500 µg/m^3, effective June 1945. Thus in late 1946, a Mallinckrodt manager told MED that they were using 150 µg/m^3 as the tolerance level for (apparently unground) ore dust, but 500 µg/m^3 for U_3O_8, UO_3, UO_2, and UF_4; MED (1946c) soon told them to return to using 150 µg/m^3 for uranium compounds and “5 x 10^{-11} grams radium per cubic meter [sic]” for ore residues with the uranium removed. As noted, above the tolerance level was raised to 50 µg/m^3, equivalent to 70 dpm/m^3, by 1949; this is also the figure given as the tolerance level for the radium-bearing residue in 1953 (MCW 1953b).

There was a separate tolerance level of 40 µg/m^3 set for the “shotgun” residue dust (i.e., for sampling and work done in the shotgun laboratory), according to MED (1945d); this applied only if the concentration of the Th-234 and Pa-234 decay products (called TI1 and TI2 in the reference) had not been increased by concentration to more than 400 times the equilibrium value (presumably secular equilibrium).

Prior to 1941, there was no standard for occupational radon exposure. In 1941, the United States Advisory Committee on X-Ray and Radium Protection recommended 10^{-11} Ci/L (curies per liter) as the acceptable occupational radon level, based on a 40-hour work week (Akerblom 1999). (This would be 10 picocuries/liter in the units most used today.) This standard was published as a handbook by the National Bureau of Standards (NBS 1941). According to Raabe (2002), the standard was for ventilated rooms in which work was done with radium (e.g., luminous dial painting) and did not include consideration of radon daughter products. An AEC report (AEC 1949b) stated explicitly that the acceptable radon level for “environmental” air (room or outside air breathed by occupational workers) in AEC-sponsored facilities was taken to be 10^{-10} Ci/L, i.e., ten times higher than the NBS standard; the report refers to this level as the “ “maximum permissible concentration” “ (page 14) and also as the “preferred level” (page 20). However, a value of 1 x 10^{-12} Ci/L was also mentioned as the tolerance level in 1950 (AEC 1950k). A ventilation design contractor stated in 1953 that the radon design criteria limit for work for Mallinckrodt and Harshaw was 10^{-8} Ci/m^3 of air, or 10^{-11} Ci/L of air (Miller 1953). Thus it is claimant-favorable to assume that the standard that was applied during most of the period in which Mallinckrodt uranium refining took place was 10^{-10} Ci/L (100 pCi/L) and that this applied to radon only and not to the daughters.

Regarding effluents, AEC proceeded on the basis that it was acceptable for liquid and solid effluents to have concentrations up to one order of magnitude greater than natural background (AEC 1949b). AEC "recommend[ed] that neighborhood air levels for these radioactive materials [containing uranium and radium] should not exceed 1% of the levels used within the plants" (AEC 1949b). No information is available as to Mallinckrodt's approach to effluent control, e.g., whether it followed the AEC recommendation.

For workers employed in years during which no (radiological) urinalysis was done, reference may be made to AEC (1950a), which was a cumulative exposure estimate study done by AEC of Mallinckrodt workers who had been employed between July 1942 and October 1949. AEC's estimates for the dose to the lung were based on air samples of alpha-emitting dusts (translated to a daily weighted average exposure level); to the bone, on breath radon analysis (to determine the fixed radium burden); and to various organs, on film badge data. Because in dose reconstruction different assumptions are made than AEC made and thus these calculations will have to be redone, their results are not repeated here. However, dose reconstructors should be aware of this report in the case of workers who began MED/AEC work before urinalyses were routinely done and for whom AEC's "back-calculation" estimate may be found listed in dose records as simply an accumulated dose for the pre-monitoring period of operation. It should be noted that workers were not identified by name or work category in this report but the tabulations that produced the bottom-line figures for the report may be available by worker name in dose reconstruction project files.

Also with regard to this report, it should be noted that the calculations of lung dose did not include a gamma contribution because AEC deemed it negligible compared to the dose from airborne particle inhalation; that AEC did not include radon dose because they could not estimate an average concentration and they assumed that airborne particle inhalation would dominate; that AEC assumed all the uranium to behave like UO_2 in the lung; and that AEC assumed that biological equilibrium existed since the start of employment. AEC did include the external dose in the bone dose calculation. It should also be noted that AEC thought that the exposures in the unmonitored years were "at least as severe as they were found to be at the time of our initial studies" (in early 1947). They stated that conditions probably were not more favorable and may have been "moderately" more severe. Thus they thought that their extrapolations could possibly be somewhat nonconservative.

5.2 RADIOACTIVITY CONTENT AND HANDLING OF THE ORE, URANIUM PRODUCTS, AND RESIDUES

5.2.1 Ores and Other Feeds and the Initial Process Steps

The origin of the ores is important in considering source terms at Mallinckrodt facilities because the content of uranium in the ores was greatly variable. Pitchblende ores contained high levels of radium-226 and other radiologically significant daughter products, while other ores and feed materials typically did not. Ra-226 (in equilibrium with its daughter products) constitutes a significant gamma source and thus produced most of the external whole-body dose received by the Mallinckrodt workers, while Th-234 and Pa-234, both beta emitters, produced most of the extremity dose. In addition, radon and radioactive dusts were released in storage and processing, resulting in internal dose due to inhalation, with the radon releases again being highly correlated to the radium content. Since the concentration of radium and other daughters present in the ore, processed uranium, and processing residue at any given time depended most strongly on the concentration of uranium in the ore, on a per-ton-of-ore-processed basis (ignoring process differences), the various doses received depended on where the ore originated.

Pitchblende ores from the Belgian Congo (the so-called "Congo ore" or African ore), supplied by the Belgium-based African Metals Company, had average concentrations of 25% (Eisenbud 1975) or 30% (Mason 1977) uranium by weight, up to a maximum of 65-70% (DOE 1997; Dupree-Ellis et al. 2000). Other early ore sources were pitchblende ores from Canada (Radium City in the Great Bear Lake Area and Port Hope in Ontario) (DOE 1997), containing uranium concentrations of about 10% (Eisenbud 1975). Later in the war, domestic ores were used also. The wartime domestic supplies were actually tailings from vanadium mining and milling (Eisenbud 1975); although the original ore had uranium concentrations of less than 1%, the tailings were shipped as a 20% sludge concentrate (Eisenbud 1975). (Note that there is some confusion among the various references with regard to ore specifications, in that some give the percentage as applying to uranium alone and some as applying to U_3O_8. Since the percentage is by weight and the uranium is by far the larger weight constituent of U_3O_8, the differences are not significant.)

Ores from the Belgian Congo had average concentrations of up to 100 milligrams of Ra-226 per ton of ore (Dupree-Ellis et al. 2000; Eisenbud 1975), possibly up to 135 milligrams per ton (AEC 1949b). (Mason (1977) gives 300 milligrams per ton, but this appears to be in error.) Thus there could be a significant dose rate from the ore when it was in drums or when it was being loaded into other containers and hoppers. As previously noted, the ore was dried before use; this was a necessary condition for optimal processing, but the dust levels created during drying and later handling were high. A 1947 study done for AEC by the University of Rochester evaluated the relative amounts of uranium and radium in ore (see Section 5.3.1). The radon in the Belgian Congo ore was also significant because it built up over time in containers and enclosed spaces. When drums, enclosed storage areas, the thaw house, etc., were opened, a worker could be enveloped in the escaping radon. Table 4 gives more information about the quantities and radiological characteristics of the ore.

As stated earlier, most of the pitchblende processed by Mallinckrodt was obtained as a concentrate from the Belgian Congo by AEC in 1944 and was shipped to St. Louis in 55-gallon drums from whatever North American receiving point was distributing it at a given time. After the war, feed materials were usually packed into 30- or 55- gallon steel drums at the mills and shipped by rail in full carload lots (Mason 1958a). It can be inferred from Mason (1958a) that a rough conversion is about 15,000 tons of uranium ore per 100,000 drums, so that each ton represented about 7 drums to be dumped, sampled, and processed.

In 1942, there was some contact (i.e., it was open to the air and potentially could be touched) with non-pitchblende uranium in the form of U_3O_8 when it was mixed with nitric acid for digestion (MED 1942). As noted earlier in Section 3.1, the pilot plant operations in Building K-1E yielded a liquor that was conveyed by hand cart to vessels outside Building 52 and as suggested by Mason (1977), the further transfers also appear to have involved some closeup, manual handling. But when the pitchblende ore began to be used, contact was reduced as much as possible. To minimize personnel exposure, the ore (storage) room (addition) was divided by brick piers into corridors, with each corridor being wide enough to hold four drums of ore side by side, with a layer of four more drums stacked on top of them (MED 1946c). It was thought that this design would eliminate the need for workers to pass between or close to stacks of ore during the ore storage and transfer operations. However, due to the blocking of some corridors and the filling of others because of the volume of ore drums, it was after all necessary for workers to pass (close) by the ore drums stored in the partly filled corridors (MED 1946c). As of at least 1947, barrel handling of ore barrels was done with a forktruck, thus eliminating nearly all of the necessity of manual handling of the barrels (AEC 1947a).

In 1948-1949, ventilation and remote control systems were installed in the Ore Room and its addition. This helped to reduce the radon concentrations in the operating areas, except for a point in the Ore Room at the junction of the two conveyor systems: radon levels were found to be high and to remain high for nearly half an hour after pitchblende drums were opened (MCW 1949m). Operators were not supposed to be near the drums after they were opened, but operators sometimes still had to enter the Ore Room or the conveyor alleys while drums were standing open on the conveyor because of operating difficulties. These difficulties included the need to make frequent visual inspections of the flow through the Syntron feeder mechanism into the rod mill, incorrect placement of a drum on the conveyor, failure of conveyor stops, and the jamming of some fiber (cardboard) drums on the conveyor. In the case of the second and third items, the operator had to crawl into the conveyor alley to remedy the problem and in the case of the fourth item, had to go into the Ore Room to remove the open drum manually.

In February 1950, a study was done of gamma and radon exposure to three workers conducting a special pitchblende ore inventory (MCW 1950g). This included moving 1.5 lots (probably a total of 9 drums) by forktruck, weighing the ore drums, checking the numbers, and counting all the ore drums present (not just the checked drums). They received gamma doses of 45-75 mR, compared to the then-tolerance dose of 60 mR per day. Radon exposures averaged 1.02 x 10^{-10} Ci/L, which was higher than normal because the ventilation system was operated in a special mode to facilitate radon sampling for the study. It was concluded that these exposures were acceptable if the special inventory was done monthly, but if more extensive inventories were done, rotation of personnel and extra planning would be necessary.

In 1950, it was observed during a study that it was very dusty in the Ore Room proper and that while the rod mill (grinder) was running it was necessary for the operator to enter once for 1.5 minutes to go to the top of the skip hoist and dislodge the ore stuck in the supply drum; this was done using a hammer (AEC 1949e). Other than that, the operator’s time in the Ore Room was spent opening, dumping, cleaning, and closing the drums, with the average number handled being 22 per shift over about four days (MCW 1950u). In 1954, it was noted that operators were still being forced to enter the Ore Room frequently, due to the ineffective drum-handling system: drums would hang up on the conveyor and have to be pushed free manually (MCW 1954c). Also, large drums had to be removed from the skip hoist by hand because the system could not handle large drums (MCW 1954c).

Contamination control instructions were not always enforced in these known dusty operations. For example, in 1954 operators in the Ore Room were observed to be loosening lids on the ore drums on the conveyor before moving the drums to the skip hoist, which allowed dust to come out under the lids as the drums were moved; similarly, these operators took drums from the skip hoist and placed them on pallets with the lids positioned only loosely and without the clamps being fastened on (MCW 1954c). Drums of ore were also observed to be sitting in the Scale House without lids on, with the lids and clamps being put on in the open area of the Scale House or even outside and not on the conveyor as procedure required (MCW 1954c). The reason for the irregularities with the drum lids and clamps was that by this time many of the drums, having been used and re-used, were old and slightly out of true, so that the lids had to be beaten on and the clamps were fastened on only with great difficulty (MCW 1954c). When the feeder (conveying) screws that removed the ore from the hopper became blocked, ore had to be scooped from the hopper by hand; it was noted at least once in 1954 that this was done without respirators (MCW 1954c). The dust released was not confined to the ore area, since the doors to the Ore Room and Ore Room addition were often left open through negligence or for convenience (MCW 1954c).

Similar issues arose in the MgX area, used from 1949 on. Various memos and an August 1953 report (MCW 1953d) expressed concern about the MgX area, in particular the inadequate design and maintenance of the conveying facilities that created dust levels in the breathing zone and general area up to 300-350 dpm per m^3. This report noted that the drum washer was not working properly, so that there were high levels of dust when the workers hammered the ring clamps onto the drum; the airflow into the exhaust on the operator's platform was so inadequate that dust often came out of the opening when the operator released the mechanical "grab"; the operator had to go to the top frequently to push the feed material down the hopper into the screw feeder with a paddle, with dust coming out of the hopper opening as he did so. Also, the housing was warped, the gasketing material was loose, and there were broken windows in the drum washer housing (MCW 1953d). The problems had still not be solved in 1954, when leakage was observed coming from the feed hopper to the tanks at the feeder screw housing because of flanges that were not securely fastened and from loose packing at the screw shaft; the tops of the digestion tanks were thus covered with this dust (MCW 1954c).

In 1947, apparently after some months of experience in working with the high-level pitchblende ores, Mallinckrodt wanted to install steel shielding around several of the digestion tanks along the catwalks (platforms) and around the Feinc press filters that produced the K-65 waste (AEC 1949f). However, AEC rejected shielding around Tank M-14 because sludge was shoveled into the tank from the platform and the shielding would impede the shoveling (AEC 1949f). AEC apparently rejected all of the extra shielding eventually (MCW 1947). Mallinckrodt protested, stating that they had agreed to fewer provisions for health protection than they thought advisable in the design of Plant 6, on condition that the provisions would be added "if trouble later developed"; the difficulty was that after the plant was built and high-level pitchblende ores were being handled, AEC and Mallinckrodt disagreed as to what constituted "trouble developing" (MCW 1947). Thus although some shielding appears to have been added in 1948-1949 (AEC 1949b, MCW 1950e), notably for the ore grinding and C-3 filtration operations, the relatively high dose rates from the digestion tanks and the Feinc "bowls" were a continuing personnel protection problem at Mallinckrodt. This was noted on an inspection of the Mallinckrodt facilities by an AEC consultant, who stated that shielding was reaching a practical limit because with the tanks and vessels being shielded, the existing piping had become an important source of dose and the piping was not easily shielded (AEC 1949k). Further, rearranging the piping was not an option because the pitch and length of the sections was important to the process (AEC 1949k).

Dust and radon could be a continuing problem even in some of the "wet" areas. It is known that one reason for the spread of dust and radon was that doors to the cells containing the process equipment were often left open through negligence or for convenience; as late as 1954, a Mallinckrodt internal safety report gave as examples the leach and wash (Oliver) cells, the Feinc cells, and the M-14 cells (MCW 1954c). Tank lids were also left open unnecessarily, most notably the centrifuges, which were sometimes opened on one shift for cloth changing and not closed until the next shift (MCW 1954c); this allowed K-65 residue to dry out and drift out of the centrifuge and onto its outer surfaces and the floor. Also, as equipment became worn, there could be substantial leakage out various openings. In the case of the centrifuges, leakage was observed around the shaft, plow, and scoop openings at the top; this leakage was often allowed to collect and dry out (MCW 1954c).

From about mid-1948 on, at least some pitchblende ore (Q-11) was stored at SLAPS (AEC 1948e, MCW 1949p). Apparently at least one forktruck driver (to move the drums) and one truck driver were required; possibly a sampling worker was needed at times. Guards were said to have been "maintained" at the site from 1946 to 1951 (AEC 1959), apparently constantly; after that, it appears that guards were present only for routine patrols and actual worker entry into the site for deliveries or removals of ore. It is not clear whether the storage workers and guards were always Mallinckrodt employees or not; e.g., Mason (1977) states the raffinate cake and barium sulfate cake collected in dumpsters in Plant 6 were taken away in dump trucks "which AEC operated". However, Mallinckrodt warehouse workers and guards appeared to have been doing the residue delivery and placement work even in 1946-1953 (MCW 1949d; MCW 1949p), when AEC was running the site. There are notations of “airport” in urinalysis and film badge records, so it may be possible to determine from such records whether and when a particular individual did work at the site.

Material was taken between the main Mallinckrodt site and SLAPS by truck (Mallinckrodt 1994; AEC 1947a; MCW 1949p), so it can be inferred that ore drums were received by rail at Mallinckrodt and then trucked to SLAPS. The ore drums were stored on a concrete pad next to the K-65 shed (the pad also formed the floor of the shed) (AEC 1948j). Mallinckrodt (MCW 1949p) restricted the time that warehouse workers spent at SLAPS because of the hazards from the K-65 drums and because of other ore work at the main Mallinckrodt site, such as unloading ore drums from railcars. The task of obtaining ore drums from the airport pad involved three trips per week (MCW 1949p) from Plant 6 (where the warehouse and yard workers were based) to SLAPS; apparently at least one forktruck driver (to move the drums) and one truck driver were required. Because of the proximity to the K-65 storage in the enclosed shed, it appears that the K-65, with its much higher content of Ra-226 than the ore, may have contributed non-negligibly to the exposure of the ore workers.

It should be noted that in the later years, many booths or walk-in areas were called “enclosures”. These were generally not entered bodily by the operator, unless there was a malfunction of the equipment; rather, he inserted his arms into such enclosures through openings or used remotely operated devices to perform work within the enclosure. Some “stations” were actually enclosures such as this, some were hoods, and some were semi-enclosed booths.

5.2.2 Uranium Products

Once the Ra-226 was removed following the digestion step and the vessel(s) had been vented, the gamma dose rates were much lower and the radon (which arose from the radium) was no longer an issue in processing (except for the radium-bearing residue). Radium and radon would again build up to significant levels from the uranium parent, but this took more time than the apparent typical digestion-to-shipout time at Mallinckrodt. The principal hazard was thus the uranium-bearing dust.

The main hazard after the radium-bearing residue was removed was dust, since a fraction of the uranium salts and oxides tended to aerosolize when dry and when handled. Initially, somewhat crude precautions were taken to control dust during handling and there was extensive manual handling of uranium salts and oxides in the dry form (Eisenbud 1975); in 1942, it was noted that contact could be made with UO_3 when removing it from the open receptacles (boilers) to transfer it to the furnaces, and with UO_2 when packing it. For example, in Plant 6 (and presumably in its predecessor plants 1 and 2), the UO_3 dry powder was unloaded from the reaction pots by hand-scooping (Mason 1958a), i.e., manually using handheld scoops. The UO_3 was scooped into a drum through a grated funnel that had a vacuum connection on it to draw the UO_3 dust out of the drum (MCW 1956c) (the vacuum connection was probably in use only from the late 1940’s on). The large pieces caught in the grated funnel were broken up by hand, which was always recognized as being a particularly dusty step operation and a principal reason why respirators were supposed to be worn during the operation. In 1954, there was a temporary top that was supposed to be used during the filling of the UO_3 drums to keep the dust done, but since it did not allow the operator to see when the drum was full, it was usually left off (MCW 1954c). The dust on the outside of the drums was supposed to be vacuumed off, but the operators often used a rag to do it (MCW 1954c), which meant that much of the dust went into the air and onto the floor or the conveyor. When the dust collection bag for the UO_3 area was changed, a spool piece was supposed to be installed in the ductwork, presumably to allow for an alternate suction source to keep the dust from coming out, but this was not done even through many bag changes (MCW 1954c).

The drums of UO_3 were taken to the loading area and trays were filled with the material. After being weighed, the trays of UO_3 were placed into the furnaces to be reduced to UO_2 (Mason 1958a). The trays of UO_2 were then unloaded by hand into drums for transport to other areas or sites. This was described in Rochester (Rochester 1948b) as follows. In the tumbling area, the newly formed and cooled UO_2 was placed in tumbling drums and rotated on a tumbler to break up clumps; the drums were taken to the skip hoist, where the tumbling drums were opened and transferred to the hopper of the Fiberpack loader; at the Fiberpack loader, a vibrating chute fed the material into 75-lb containers and the containers were sealed on a roller conveyor; and from the UO_2 production area, the UO_2 containers were trucked to the warehouse for storage.

The major handling improvement of 1949, the installation of pneumatic unloading and conveying systems, was supposed to have eliminated all hand-scooping of UO_2 and UO_3. .However, AEC inspectors repeatedly noted hand-scooping going on until the end of operations at the plant, often due to the failure of equipment such as the vacuum-type UO_3 “gulpers” (AEC 1954d; AEC 1956b). A 1956 Mallinckrodt report (MCW 1956c) explained why: when the pneumatic system was not operational when the UO_3 was ready to be unloaded from the pots, the unloading could not be postponed because acid would condense in the ducts and run into the pot, ruining the batch of UO_3. So it was recommended that while the pneumatic system was out of order, the exposure be reduced somewhat by collecting the large pieces caught in the funnel grate and waiting until the pneumatic system was back in order before breaking them up (MCW 1956c)

UO_2 produced at Plant 6 was trucked over to Plant 4 in small fiber containers (AEC 1949b); no information is given as to how this affected containment of the dust. In Plant 4, there was again extensive hand-scooping and other manual handling of the uranium materials (UO_2, UF_4, and uranium metal) (Mason 1958a). AEC (1947a) states that the UO_2 was hand-scooped into trays and leveled off with a stick; the gloved hand thus came into direct contact with the material. Manual handling was reduced by mechanization in 1948 and 1949, but even so dust levels were considered too high (Mason 1958a). AEC agreed to have Mallinckrodt construct Plants 6E and 7 to replace Plant 4 and part of Plant 6. These plants were even more mechanized and were said to require little (if any) manual handling (Mason 1958a); however, as various AEC air dust study reports indicate (e.g., AEC 1954g; AEC 1955d), this was not always so. AEC (1955e) even reported in 1955 that a Plant 7 operator used a piece of cardboard in lieu of a conventional metal scoop to make up UF_4 weight (in loading a container), with the operator’s (presumably gloved) fingers dipping into the material frequently.

In 1955, a new derby “pickling” facility went into operation in Plant 6. The derbies made in Plant 6E were transported to this facility to be pickled – i.e., cleaned in a dilute acid bath – and then returned to Plant 6E in baskets of eight or metal boxes of twelve (AEC 1955e, MCW 1955p). Prior to being processed in the recast step, the derbies were dried with a flame in a hood, if necessary (AEC 1955e). Some chipping in the chipping enclosure was still necessary (AEC 1955e) because during the drying (whether by air or flame), the surface of the derby would oxidize to black oxide (U_3O_8). Thus the dust in the chipping operation was at least partly U_3O_8. Although the use of the flame for drying was supposed to be discontinued (MCW 1955o), it appears that it did continue (MCW 1955p).

The billet (ingot) was initially low in beta activity because during the melting in the vacuum furnace (i.e., in vacuo), the dross and slag floated to the top of the crucible and the beta-emitting UX1 and UX2 (Th-234 and Pa-234) sublimed and cooled on the underside of the furnace lid (Mason 1977). The dross from the crucibles and the other furnace residues was packaged separately from the slag and the billet (Mason 1977). However, as the billet aged, the UX1 and UX2 started to build up again.

In November 1953, a study was done of Plant 4 recast operation to determine the cause of high film badge readings (MCW 1953e); although Plant 6E made most of the metal, some metal was still produced on a pilot or experimental basis in Plant 4. The following dose-producing practices were found. The furnace jack was warped, so the operator had to enter the furnace enclosure to guide the jack with his foot each time the furnace was loaded. After unloading the furnace, the operator had to weigh the crucible and put it into a drum by hand. The crucible dose rate varied with the number of times used and the type of metal; the crucible in the study read almost 400 mrep/hr (beta plus gamma) before it was loaded because it had been used before (and would read more after the recast was done). Aged metal billets were stored on the floor beside the operator’s desk and were left there until the lead operator or the engineer examined and approved them, a period of one to three days. The operator spent four to six hours sawing “betatron slices” (samples to be sent to Granite City Steel (aka General Steel Castings) in Granite City, Illinois for testing) because the shape of the slices required the operator to spend several minutes setting the metal on the saw. The slices were stored on a table in a storeroom until enough had accumulated for shipment, a period of three to four weeks. When enough slices had accumulated, the operator would examine each closely to determine the weight and lot number stamped on it, which took a total of 30 to 45 minutes.

Besides these practices, MCW (MCW 1953e) noted that in their one run per shift, the same two Plant 4 recast operators had to load the crucibles, clean the furnace, saw the metal, degrease the metal (by hand until some time in late 1953), and load the betatron slices. There was no mechanical method to move the crucible from the furnace to the scale, so one of the operators had to climb physically into the furnace bottom and attach a chain to the billet, remove the fire brick from the billet , remove the empty crucible, and weigh the crucible. Meanwhile, at Plant 6E there were separate groups for each step, i.e., saw operators, furnace operators, crucible loaders, and cage men (who ran the degreasing machine). At Plant 6E, after the metal was sawed, it was kept covered and was stored away from contact with workers. Also, the metal at Plant 6E read fairly low compared to the aged Plant 4 metal. Thus as MCW (MCW 1953e) pointed out, the hazards faced by the Plant 4 recast workers were not being considered to the same degree as the Plant 6E recast workers. It is not clear whether the subsequent film badge readings went down because changes were made or because the type of operation was reduced or discontinued, but it appears that the handling of the crucible was minimized thereafter.

5.2.3 Residues and Other Wastes and Reprocessing

Because of the splitting of chemical forms in the course of processing, the various wastes had content that was concentrated or depleted in particular isotopes, depending on where in the process the wastes were formed. Details are given in Table 4 and in the text as specific isotopes are discussed. But in general, it can be stated that the waste was disposed of promptly because otherwise storage space would have been exhausted. Thus once the waste was taken out of the process equipment or area, exposure to the waste was limited to the radioactivity existing in it at the time it was created or collected, with the exceptions of the K-65 drums that might be stored until they were transported to SLAPS, the slag that was recycled in the later years, the low-level solid wastes (such as rags) that might be accumulated prior to incineration; and the residues brought back from SLAPS for reprocessing. In the case of the K-65 drums, the exposure would be to radon emanating from the drums and to the gamma radiation from the radium; in the case of the slag, the exposure would be to dust and mostly beta radiation; in the case of the low-level solid wastes, which were probably kept in dumpsters or similar large bin-type containers, the exposure would be dust and external radiation; and in the case of the reprocessable residues (e.g, the AM-7), the exposure would be to dust and external radiation. It does not appear that any of the wastes had before disposal sufficient time to build up daughters much beyond what was already in the waste, except to some extent the residues brought back from SLAPS.

The residues dumped on the ground or stored in drums at SLAPS are a special case, since they sat there for a number of years, with only a few workers entering the storage area intermittently. In the case of the residues dumped on the ground, the exposure would be to dust and to some degree from external radiation and in the case of the drummed K-65 residue, it would be to radon and gamma radiation.

A condition placed on the ores from the Belgian Congo by African Metals was that the Ra-226, the Ra-226 daughters, and the lead and precious metals be extracted, stored, and returned to African Metals (AEC 1967; AEC 1949b). Thus the Mallinckrodt process included steps to extract these materials as a separate residue from the bulk of the ore residue; this was the radium- and lead-bearing K-65 residue, also called gangue lead cake or GLC. K-65 and other wastes were trucked to SLAPS but sent by train to sites outside Mallinckrodt. Radiation levels from the K-65-containing railcars and trucks exceeded those permitted under the regulations of the day, so shipments had to be made under special permits granted by the Interstate Commerce Commission (forerunner of the Department of Transportation) (AEC 1949b).

A 1947 dust study done for AEC by the University of Rochester evaluated the relative amounts of uranium and radium in the K-65 residue (see Section 5.3.1). It was reported in February 1948 that the K-65 contained about 600 mg of radium per ton and about 0.2% uranium; at that time it was being sent to the Lake Ontario Ordnance Works at an estimated rate of 6000-12000 pounds per day (AEC 1949m). AEC (1949b) stated that as much as 100 grams of Ra-226 contained in the K-65 residues was produced per month at the Mallinckrodt facilities. This was in fact the quantity produced in December 1948 (AEC 1949b). It was reported that 200 grams had been transported at one time to Middlesex (AEC 1949b), meaning that up to this quantity had been in storage at the Mallinckrodt site or at SLAPS and had had to be loaded for transport at one time.

In addition to the K-65 residues, there were other solid and liquid residues and wastes, as reported in February 1949 (AEC 1949m). The other two major solid refinery residues were the barium sulfate or AJ-4 residues and the pitchblende raffinate or AM-7 residues. The barium sulfate contained about 4 x 10^{-9} grams of radium per gram of residue and about 0.1% uranium; because of the low radioactivity content, it was being dumped on the ground at SLAPS at the rate of about 6800 pounds per day. The AM-7 consisted of various metal hydroxides with about 0.1% uranium; it too was dumped on the ground at SLAPS at an estimated rate of 23,000 pounds per day. The liquid wastes (from the refinery processes) contained about 0.002% uranium; they were discharged through a sewer to the Mississippi River at an estimated rate of 3000 gallons per day, with about 12 pounds per day of it being uranium. Miscellaneous waste byproducts of the UF_4 production process were sent to Vitro for recovery of uranium, but in relatively insignificant quantities, while the slag from the uranium metal production process contained about 0.3% uranium and was stored at the airport. All of the various processes generated contaminated scrap metal, with the most problematic, in terms of quantity and degree of contamination, being Raschig rings from extraction columns, corroded drums in which K-65 had been stored before reprocessing, and floor materials (such as bituminous floor coverings being replaced).

The K-65 residues were stored in drums. Some of the waste sent to SLAPS was said to have been hand-packed by Mallinckrodt workers into barrels (drums) (Eisenbud 1975). A concrete-lined pit (“the swimming pool”) was constructed to store the drums in, but was never used “due to health reasons” (i.e., the dose rates and perhaps the radon concentrating in the pit would be at excessively high levels) (AEC 1959) and because the corrosion of the K-65 drums would have made retrieval from a pit difficult (MCW 1949g). However, (AEC 1959) states that the pit was used to store tailings from the slag reprocessing operation from 1955 on. The same storage design and precautions as for the ore were followed for the radium-containing waste (MED 1946c), except that the drums may have been kept in the open until the storage shed was built in about August 1947 (AEC 1947a). (See also Section 5.2.1. above.) Shield walls were included in this to divide it into smaller areas (AEC 1947a). Workers who performed operations where the K-65 and ore were stored at the Mallinckrodt main St. Louis site and at SLAPS (i.e., the warehouse/yard workers) were placed on a rigid time schedule to keep them from exceeding the tolerance dose (MCW 1949g; MCW 1949s; MCW 1949p; MCW 1950v). For example, MCW (1949s) states that a warehouse worker (including forktruck drivers) could, in one week, unload one ore car and make one trip to SLAPS involving K-65 handling and other duties within 50 feet of the K-65/ore pad or he could work loading one railcar with K-65 drums and make one trip to SLAPS ditto; an additional restriction was that no more than 2 hours per week could be spent at SLAPS, with only one hour per day actually working at the pad. As Mallinckrodt and AEC recognized (MCW 1949g), prior to the reprocessing of the K-65, corrosion of the drums was a severe problem: although the K-65 was insoluble, stream pollution at SLAPS was a possibility, but repackaging the K-65 would involve significant worker dose. MCW (MCW 1949g) suggests that some of the “sand” (inferentially, the AM-7 residue, as furnace sand with the radioactivity deposited on it) was being put into the K-65 drums, presumably to lower the dose rate from individual drums.

Concern was also expressed in 1949 about the bulldozer and crane operators working at the fresh and aged raffinate (AM-7) heaps at SLAPS (MCW 1949g). This was studied and it was found that the beta dose rate at the cab of a bulldozer pushed all the way into a heap of raffinate was considerably below tolerance. Since the bulldozer operator averaged less than 8 hours per week on the raffinate heap work, this operator’s exposure was a small fraction of tolerance. These workers were thus not issued film badges.

As noted in Section 4.7, most or all of the K-65 was brought back in drums from SLAPS and reprocessed starting in early 1948. In January 1949, the gamma dose to the hands of operators opening drums of return K-65 was studied by placing film badges on their wrists (MCW 1949e). See Section 5.4.1 for results. No correction of the high hand dose situation was made, however, because the K-65 drum opening was done over a limited period of time (i.e., once all the K-65 stored at SLAPS had been brought back and reprocessed, the operation was over) and it was not realized how high the doses were until the study was done late in the reprocessing campaign (MCW 1949e).

The K-65 waste residues included not only the residues themselves, but also the used cloths from the Feinc press filters; drum disposal was the only solution for disposal of the cloths, since (1) they were impregnated with the radium-lead salt sludge and thus would not burn and (2) they were made of plastic fibers and thus would not dissolve in acid (AEC 1949f). It was reported that three used filter cloths, stored in two 55-gallon drums, produced 30 times the tolerance level dose rate at contact with the drums (AEC 1949f). Thus the cloths and the operations associated with them were of major concern with respect to external exposures in Plant 6.

In 1949, a study was done of the beta and gamma exposures associated with preparing, repairing, cleaning, and changing Feinc filter cloths by the cloth operators (MCW 1951g). Due to the recent reduction in the external dose tolerance levels by AEC, it had been recommended that an additional (third) worker be added to the rotation schedule for this job. Major revisions as a result of the 1949 study were that a basket was provided for carrying the used cloths in, that the filters were partly cleaned by the area operator before the cloth operator began work, and that a fourth worker was to be added to the rotation schedule in May 1950. However, the cloth operators did not like to use the basket because it was hard to haul it up the stairways of the filter platforms and they still had to spend some time cleaning the filter. Also, during the summer of 1950 several dozen workers were transferred from Plant 6 to Plant 1 (see Section 5.0) and this disruption of operations and the need to retrain new workers delayed the assignment of new people to the rotation, which was not done until

November 1950. This also increased the two principal cloth operators' doses significantly over the other cloth operators' doses.

As a result, it was agreed in 1950 that the area operators would better clean the filters and blankets in preparation for the cloth operators' work, the cloth change interval would be increased from two days to three, and a fifth worker would be added to the rotation (MCW 1951g; MCW 1951a). In January 1951, a new study was done of beta and gamma exposures associated with preparing, repairing, cleaning, and changing filter cloths (MCW 1951a); this included cloths for the Feinc, C-3, and Recovery filters. Dose rates measured with instruments were combined with time measurements to estimate doses; the gamma doses were said to agree well with film badge readings but not the beta doses, due to the technical difficulties of field measurements of beta radiation. These results were said to be comparable to those found in the 1949 study, although some of the tasks the cloth operators were doing in 1949 had been given to the area operators to do (MCW 1951a), but clearly the weekly doses had increased at the time of the 1951 study. Also, the operators were still not using the basket provided because of its bulkiness. But waiting in the cell for work to start and other work practices that increased the "close to Feinc" (distance of less than three feet) time had been changed and the cloth change interval was increased from two days to three (MCW 1951a), so the doses were expected to decrease.

There were varying quantities of miscellaneous waste arising from various sources. For example, rags and paper with small amounts of uranium forms and residues were created from cleaning and wiping procedures, such as the black oxide from wiping derbies, billets, and mold strips and the residue from wiping the outsides of drums before transport. Other types of contaminated materials included packaging, sweeping compound, old uniforms and gloves, and like items that were used in the process areas or were in contact with radioactive matter. All of these types of materials could be burned in an incinerator outside Building 115 at Plant 6 or outside Building 403 in Plant 4 (Mallinckrodt 1994) and the ash could be processed or sent elsewhere for recovery. The filter cloths were also incinerated (MCW 1951a), at least at some periods of operation.

Dust from the various processes was collected mostly in bag-type filters, although some was collected with a rotoclone (a water spray type of collector) and run into a recovery tank (MED 1944o). Dumping these filters, i.e., dumping the bags of dust into drums, was recognized as a high-dust operation (e.g., AEC 1954a , AEC 1954d, AEC 1954c). Mallinckrodt estimated that bags were changed about every six weeks for the Plant 6 Pot Room dust collector and that it took 1.5-2 hours per bag change, for an downtime percentage of 0.4% (MCW 1955r). Occasionally a bag would break, obviously resulting in even higher dust levels. For these reasons, respirators were supposed to be worn but were not always (e.g., AEC 1954e). There was also a possibility of increased external dose from dealing with the collectors. For example, in 1956 a maintenance worker received an external dose higher than usual, as shown by his film badge reading; this was found to be mostly due to the eight hours he spent repairing one of the Hoffman collectors that served the recast (YM-5 or billet) area (MCW 1956j). This collector handled the beta-active black oxide that resulted from the recast activities (MCW 1956j).

Dust was a problem even with empty drums. Mallinckrodt reported to AEC in 1950 that used K-65 drums being returned to Mallinckrodt in ATMX railcars did not always have lids and were not always cleaned (MCW 1950n). This resulting in dispersal of the residual dust throughout the car during shipment, to such an extent that Mallinckrodt collected 100 pounds of it in one railcar after it was unloaded; there was contamination of the receiving dock as well. Dust concentrations in the railcar were in excess of 250 times tolerance (i.e., more than 17,500 dpm/m^3) were found in the general area of the railcar and about 27 times tolerance in the worker's breathing zone, but the radon concentrations did not exceed 20% of tolerance and the gamma radiation did not exceed 15% of tolerance. AEC appears to have resolved this by directing the drum sender (presumably Lake Ontario Ordnance Works) to correct the situation.

It was noted in January 1955 that for a considerable period, radon concentrations of 1-8 x 10^{-10} Ci/L had been detected in the Plant 6 Cloth Storage Room (MCW 1955j). After much detective work, it was found that the wall near the M-50 (tank) sump was saturated with “material” that had come from the sump and from the Wash Oliver cell (presumably by liquid leakage and/or by deposition via air currents). Holes in the wall and ceiling of the Cloth Storage Room created a draft from the sump area across the accumulated material and up into the upper area of the room. This situation appears to have been remedied once discovered. However, in December 1956, the Cloth Storage Room was again studied because of the wide variation in the radon readings taken during weekly checks; Over 1 August to 31 December 1956, the 18 weekly measurements of radon level varied from less than 1.0 x 10^{-12} Ci/L (i.e., nondetectable by Mallinckrodt methods) to 1.11 x 10^{-10} Ci/L, with the average being 0.5 x 10^{-10} Ci/L and the median being 0.265 Ci/L (MCW 1957). It was found that there were two major radium-containing deposits in the floor and a wall of the room. While the highest gamma reading was only 2 mR/hour, in accessible areas, it was thought that the reading would be higher in the nonaccessible areas behind equipment. Thus the migration of material from adjacent areas appears to have been a continuing phenomenon.

It was recognized that radon levels at the K-65 and ore storage area at SLAPS were high also; along with the gamma problem, the radon problem necessitated restrictions on the time a worker could spend there (MCW 1949g). Mallinckrodt was supposed to be taking radon samples at SLAPS, but after one set had been taken someone mistakenly stopped taking them (AEC 1949j). An AEC safety representative then averaged the sixty-plus samples taken up to then to produce results that could be graphed so as to see the rough falloff with distance (AEC 1949j).

The ore drum thawing and drying oven – apparently a large walk-in room with steam coils for heating – was used to thaw the K-65 drums before processing and even for temporary storage of dozens of these drums (MCW 1949o). Not only did radon emanate from the drums during heating, but because the drums were apparently opened for the heating, K-65 had been spilled onto the coils and was contributing radon on a continuing basis. This could not all be cleaned off by a vacuum cleaner. High radon levels were seen outside the oven during the heating, so a fan and stack were installed in the back of the thawing oven to draw away the radon from the drums during thawing (MCW 1949o). However, this produced no improvement. It was discovered when the heating coils were turned off, even when the fan was on, air was drawn in under the oven door and forced out over the top of the door, where it eddied in front of the oven before being drawn in again under the door. This phenomenon kept radon concentrations up inside and outside the oven. Although the time spent by workers in the oven was short and their daily weighted exposure was below the radon tolerance level, it was believed that it was not safe to operate at tolerance levels for extended periods (MCW 1949o). Thus the thawing oven ventilation had to be revised and the coils taken apart for cleaning.

As noted earlier, in 1955-1957 Mallinckrodt processed AM-7 residues (see Table 4) and produced 3600 gallons of a concentrated thorium nitrate solution that was sent to Mound (AEC 1967; DOE 2002). AEC (1967) states that Mound purified and concentrated approximately a kilogram of thorium-230 from this material, but DOE (2002) states that although 500 grams was produced and an additional 500 grams ordered, the latter was apparently never produced. The claimant-favorable assumption will be made that the entire 1 kg was produced. Thus assuming a high separation percentage, in the original 350 tons of AM-7 during processing there were approximately the one kilogram of Th-230, about 9 kilograms of total thorium, and 0.015 kilogram of uranium. This is consistent with the report by Figgins et al. (1962) that in the solution there were 29 ppm of Th-232 and 3.8 ppm of Th-230, i.e., 11.6% (weight) Th-230 by isotope.

When AEC asked Mallinckrodt to establish a pilot plant to process more of the thorium-bearing residue, Mallinckrodt refused, on the basis that the health hazards of thorium were not well understood (AEC 1955c). One concern was that because the process involved only solutions (i.e., the liquid form), exposures would mainly arise from accidental releases or leaks, which would not be easily detectable by ordinary industrial hygiene measures; hence Mallinckrodt wanted some way to detect exposure via urinary or other biological measurements (AEC 1955c). AEC agreed to study the biological half-life of thorium in the lung and bone following inhalation of thorium fluoride and nitrate salts; they were also to study the urinary clearance rate (AEC 1955c). Mallinckrodt approached the Los Alamos site because it would be receiving the thorium, but could elicit only the information that thorium could be considered equivalent to plutonium on a curie basis (ORAU 1991). AEC agreed that the pilot plant would be built and operated as if it were to process plutonium (AEC 1955c) and the pilot plant began operation at MEP on 25 February 1956 (ORAU 1991). Because the health hazards were on a "speculative level", Mallinckrodt restricted the number of workers on the project and took strict precautions as to handling, contamination control, and access (ORAU 1991). However, there does not appear to be any information available as to how the urinalyses were analyzed.

The "breathing zone" operations in the thorium processing were the removal of the raffinate cake (formed in the processing) in dumpsters and the opening of the contactor tank to adjust the concentration of the process material and to add laboratory waste materials (AEC 1955d). While AEC (1955d) found that exposures of the Plant 7E workers were always below the MAC (note that since gross alpha was measured, the thorium and its alpha-emitting daughters were in fact measured in air sampling), it was also pointed out that the pilot plant was then in the startup phase of operations and that thus the 1955 air survey was possibly not representative of mature operating conditions (AEC 1955d).

It is not clear how long it had been since the first residue sent in 1946 to the SLAPS waste storage site had been produced in the refining process. That is, this type of residue had been produced since ether extraction started in July 1942, but it is not clear whether the residues produced between 1942 and 1946 were all stored at Mallinckrodt and then shipped to SLAPS, or whether some of the older residues had been shipped elsewhere. The claimant-favorable assumption will be made that the waste was all produced in 1942 and sat in containers for 15 years, until 1957; this will maximize the radium and radon content. As Table 6 shows, although the Th-230 and Th-232 daughters would have had some time to build up, only the Th-232 daughters would be nearing equilibrium with the parent. The maximum Ra-226 content over the 2.25 possible years of processing (i.e., the 15-year maximum) was 0.158 mCi (ignoring decay of Ra-226) and the maximum Rn-222 content was 0.158 mCi (ignoring decay of Rn-222).

Regarding the protactinium processing of residues, there is no suggestion in any reference that the Sperry cake used had been processed in any way at the St. Louis downtown site; the waste appears to have been shipped out to Mound directly from SLAPS. Thus, it will be considered that there are no radiological implications of protactinium processing associated with the Mallinckrodt downtown site.

AEC (1960) states that the SLAPS waste, also called the airport residue, was evaluated for sale or disposal in about 1960 and were sampled even before that. The pitchblende (K-65) residues were auger-sampled in June 1953; barium sulfate cake, Colorado raffinates (from domestic ores), and miscellaneous ores were also considered. See Table 4 for quantitative information.

It is not clear whether the storage workers and guards were always Mallinckrodt employees or not, but Mallinckrodt workers appeared to have been doing the residue delivery and placement work even in 1946-1953 (MCW 1949d; MCW 1949p) and there are mentions of "airport" in urinalysis records. ORAU (1989) suggests that Ledoux Company laboratory workers also worked there, probably doing sampling. Material was taken to and from the Mallinckrodt St. Louis site by truck (Mallinckrodt 1994; AEC 1947a; MCW 1949p). Guards were maintained at the site from 1946 to 1951 (AEC 1959); after that, it appears that guards were present only for routine patrols and actual worker entry into the site.

At the time that planning for SLAPS decontamination was being planned in about 1964, the residues still stored on the site were the pitchblende raffinate (AM-7), Colorado ore residues, leached and unleached barium sulfate cakes (AJ-4), miscellaneous residues (but not K-65), and the C-oxides (AEC 1964). The C-oxides were then begin shipped to Fernald for reprocessing and the rest were to be offered for sale. In addition, there were several hundred tons of contaminated metal and debris at the site (AEC 1964).

5.2.4 Sampling and Laboratory Activities for All Forms

As noted earlier, other feeds besides ore were used from the mid-1950's on. The drums of these alternative feeds were usually sampled (e.g., for oil analysis) in the Plant 6 warehouse by either Analytical Laboratory workers (MCW 1956h) or warehouse workers. The process as performed by the laboratory workers was described in December 1956 as follows (MCW 1956h). Three drums were opened and a small amount was scooped out of each and put into a bottle, which was labeled and taken to the laboratory. Air samples of this activity were taken with the warehouse exhaust fan running and the large doors (panels) to the outside open. Air movement was found to be almost nil and the dust created by the sampling lingered in the air for an extended period. The average airborne level during sampling was 18,000 alpha dpm per cubic meter. A recheck of sampling was done under the same conditions but from the residue drum where the samples taken at the origination point were put after analysis there; this drum held the samples from the entire lot shipped and so could be assumed to be representative of the lot. Readings during the residue drum sampling showed an average level of 8530 dpm per cubic meter. Using the all-purpose hood in the Old Boiler House showed levels of 3460 and 584 dpm per cubic meter respectively for the two types of drums. Since the residue drum was at most half full and was a single dust source, it was clearly preferable to sample that way. It is not clear whether the change to sampling the residue drum and using the Old Boiler House hood was made, but it is likely that it was.

In 1955 feed material had come from or was expected to come from Beaver Lodge, South Africa, and Portugal (the latter as a concentrate) (MCW 1955m). A problem identified with this material was that it contained scrap iron, cake materials, and large pieces of uranium. Obtaining a representative sample of the feed would thus require preliminary separation of the extraneous material. It was necessary to process this feed and more like it at the rate of 200 drums per day, hence construction of a suitably ventilated sampling station was not an option in the short term. It appears that at least in the short term Mallinckrodt sampled these drums with an auger in the ventilated sampling bays of the Plant 6 warehouse.

In the early days of the project, the shotgun laboratory in Building 55 of Plant 2 used a 100-mg RaBe source to test UO_3 samples using neutron absorption techniques; hence it had to be designated as a special restricted area for access control purposes (Mason 1977). It appears that the RaBe source was not used after some early point, since (MED 1944m) and other documents state that such samples were prepared at Mallinckrodt and then sent elsewhere for analysis. It may therefore be assumed that use of the RaBe source for this purpose had ended by September 1944 (the date of MED 1944m).

MED (1944m) stated that UO_2 shotgun samples (possibly the reference meant UO_3) were prepared by dissolving 25 lbs of UO_2 in acid and extracting uranium nitrate hexahydrate in a four-step ether process; the remaining water layer, which also contained nearly all of the Th-234 and Pa-234 (UX1 and UX2), was run through a separating funnel, then evaporated to dryness in a dish, first by the chemist using a handheld blast burner, then in a muffle furnace. The sample remaining, which averaged 3-5 grams, was ground by hand in a mortar and poured into a bottle. The accumulation of sample bottles, usually three to twelve per day, was sent away daily for analysis. In 1954, the shotgun samples were made up in the Dry Material Sample Room (or Powder Sample Room), apparently because of equipment availability, but the work was performed by the Shotgun Sample Lab personnel and not the Dry Material Sample Room personnel (MCW 1954b). There was also a Metal Sample Room (MCW 1954b), presumably for assaying the finished metal forms.

Prior to August 1955, an average of two Mallinckrodt UO_3 samples were run daily in the Shotgun Laboratory and film badge readings were under the applicable limits (MCW 1955e); usually only one worker was needed for this. However, from August 1955 into October 1955, a campaign was performed to run UO_3 samples from other sites as well; these other-site samples constituted more than half of the total and were mostly from Fernald but also from Beaver Lodge, Port Hope, “Canada”, and “Pilot Plant” (apparently not a Mallinckrodt pilot plant). The workload increased by a factor of three and temporary workers were hired for a few months to do the work. With one Mallinckrodt worker and one temporary per shift to do two Mallinckrodt samples and four other-site samples per day, film badge readings went up significantly – one badge read 5810 mrep in one week in August and another read 2040 mrep in September, mostly beta. The hot plate hood, the muffle furnace, and the evaporating dish in the cooling and scrapedown hood were found to be contaminated with material producing mostly beta radiation, while a can of pellets under the sample press was giving off a significant gamma dose rate.

The situation was studied (MCW 1955e). It was found that there was no significant radiation reading in the laboratory assay of UO_3 until the laboratory crucible containing the sample was placed on the hot plate; as the sample was evaporated, the reading rose rapidly. The source of the radiation was the dried residues on the crucible. For the Mallinckrodt-produced samples, the average reading of 10 samples was 1500 mrep/hour at 1” above the top of the crucible and 1000 mrep/hour at 2”; the respective averages for the four “Pilot Plant” samples checked were 1800 and 1200 and for the ten Fernald samples they were >12,500 and 12,000 mrep/hour respectively. (Apparently the maximum that the Zeus instrument could read was 12, 500 mrep/hour, so the Fernald 1” readings were undoubtedly much higher.) A film badge set at 2 feet over the scale on the main work bench and about 5 feet from the boildown and crucible cooling hoods for a week read 1000 mrep beta and 50 mR gamma. A time-and-motion study showed that a laboratory worker handled the sample in some way for a total of 11 minutes after boildown, i.e., after significant readings could be expected to appear.

The Shotgun Laboratory was decontaminated by the Mallinckrodt decontamination group, with the contaminated items either cleaned or disposed of; a Lucite shield was provided to put in front of the crucibles during cooling and a steel box was provided to put the crucibles in during cooling and storage (MCW 1955e). Respirators were required to be worn during the scraping, grinding, and palletizing of the samples. A film badge placed in the same area as the first one had been read 1500 mrep/hour beta and 95 mR/hour gamma after two weeks, indicating that the cleanup and other corrective measures had been only partly successful.

K-65 sludge was sampled to determine its radium content 24 hours after it was produced, put into metal drums and stored in an adjacent warehouse (AEC 1949f). The sampling appears to have been done in the warehouse by the warehouse workers in the early years, but in a sampling room in the Scalehouse in later years; the assay was done in the Ledoux Lab by Ledoux company workers (MCW 1954a, ORAU 1989a).

In March 1955, a survey of the Scalehouse Sampling Room was made to evaluate dust, radon, and external exposure hazards from sampling K-65 drums (MCW 1955n). At this time, drums of K-65 were sampled once a day. Approximately 24 drums were moved, four at a time on a pallet, by forktruck from the storage bay to one of the K-65 sampling bays. Two samples were taken from each drum using a thief sampler inserted by means of the sampling holes (as described above); after the sample was deposited in a pan, the thief was cleaned by scraping it and knocking it against the sample rack, putting dust into the air. The pans of material were accumulated inside the sampling area until all the drums had been sampled, after which the pans were taken into the Scalehouse area and put on a rack to await transport to the Ledoux Laboratory. The operator then cleaned the floor of the sample room. It was found that during the 80 minutes of the sampling work period, the total external exposure to an operator from sampling a lot of 24 drums was 3.4 mR gamma and 3.8 mrep beta; the average total beta-gamma dose rate was 5.5 mrep/hour, the peak radon level was less than 0.5×10^{-10} Ci/L and the average was less than 0.25×10^{-10} Ci/L; and the average dust level was 1870 dpm per cubic meter, with both the general area and the operator breathing zone concentration during the actual sampling being about 2300 dpm per cubic meter. It was also found that air was frequently forced into the Sampling Room through the sampling holes in the floor, especially with a north or west wind. This air carried with it dust from its passage across the stored drums of K-65 and thus contributed strongly to the dust concentration in the room despite the nominal adequate exhaust flow, which was exhausted at floor level to draw down the dust from the sampling pans. It was recommended that respirators be worn, which apparently was not being done previously.

In 1953, the Ledoux Lab technician spent three whole days and two half days in the Ledoux Lab and two half days in the shotgun (assay) lab (MCW 1953b). At this time, the K-65 (radium-bearing) residue testing work in the Ledoux Lab consisted of compositing residue, drying it in an oven, blending it in a blender, redrying it in a dry box, mixing it in a tumbler, and then presumably testing it. The sources of dust in this operation were identified as using the grinding hood, when dumping pans of K-65 for grinding; weighing out (since there was some spillage and some need to scrape off the bottoms of the pans); cutting the sample (ground K-65 was cut through a riffle); screening (on the “Ro-tap”); and using the jaw crusher (for AM-7 residue only), for which there was no hood (MCW 1954a). The sink used for washing pans had no sump, so the wash water went to the sewage; splashing from the sink was viewed as a source of the contamination on the floor (MCW 1954a). At one point, the exhaust fan door was locked closed until the chain assembly could be repaired, hence ventilation was impaired (MCW 1954a). The cabinets interiors were found to be very dusty and the hood bases and walls, which were made of transite, had visible signs of absorbing K-65 (MCW 1954a). There was an associated Furnace Room, presumably for drying, in which respirator wearing was recommended (MCW 1953b) and the K-65 was said to be weighed on a pan on a scale “through the hole in the Furnace Room wall” (MCW 1953b). However, the heat from the ovens (furnaces) created enough draft to draw the radon up out of the lab and out over the roof (MCW 1954a). All of this information suggests a significant amount of manual handling of the residue and uncertain control of contamination, even after a new laboratory was built in 1952 and improvements were made in 1953 (MCW 1953a).

When the “U-Con” process was set up to recover uranium as a slurry from reject MgF_2 material, the U-Con began to be sampled in the Slag Separation area of Plant 7 and the samples processed in a minimal preliminary fashion in the Analytical and LeDoux Laboratories (MCW 1955L). Four samples per day were taken and transported together in open stainless steel pans in a coaster wagon to the Shotgun Laboratory. They were processed in a hood with a left-right sliding panel, the purpose of which was to keep dust out of the laboratory technician’s breathing area. The technician reached around the panel and used a small hand scoop to remove some of the U-Con and place it in a grinder in the hood. After the sample was ground the grinder pan was emptied back into the sample pan. Then the sliding panel was moved to allow the dust in the grinder to be brushed out into the grinder pan. Finally the panel was slid back and the grinder pan was re-emptied back into the sample pan and the sample pan was removed from the hood. The grinding and grinder brushing took a total of 8.7 minutes and was repeated for each of the four samples per day. With preparation work, the technician thus spent about an hour per day in the Shotgun Laboratory. A dust study showed the average breathing zone concentration to be 20 dpm per cubic meter and the general area concentration to be 23 dpm per cubic meter over the entire four-sample operation, indicating that the open pans were more of a problem than the grinding itself, because of the hood.

A check of the Shotgun grinding hood was also performed. This showed that the flow volume was 234 cubic feet per minute and the air velocity averaged 75 linear feet per minute over the face of the hood, only somewhat under the 100 linear feet per minute that might be typical of such a hood today.

5.2.5 Safety, Health, Decontamination, and Laundry Facilities

A respirator decontamination facility was added, presumably in Plant 6, in April 1950 (MCW 1950e). It is unclear where this was done previously.

Although most contaminated equipment could be repaired in situ or in the Mallinckrodt repair shop -- which was not dedicated to AEC work but served the entire Mallinckrodt general site – some equipment had to be sent out to other companies' facilities for repair (AEC 1948h). AEC approved this practice based on its requirement that all items should be decontaminated if possible to "zero wipe" (i.e., no detectable removable contamination) and on the assumption that any resulting contamination on floors and machines would not be significant. AEC also stated that air samples should be taken of representative repair operations in the Mallinckrodt shop and that areas immediately adjacent to the Mallinckrodt shop should be monitored for contamination in order to verify that surface contamination was not being tracked out (AEC 1948h); it is not clear if this was done or not.

MED (1942) stated that both contaminated and clean clothing were laundered on the premises, in separate laundries; however, AEC (1947a) stated that contaminated protective clothing from all areas was sent to public laundries, contrary to other AEC sites' practice. Use of public laundries appears to have ceased at AEC direction (as suggested by AEC 1947b). An onsite laundry with "clean side, dirty side" flow was installed some time prior to April 1950 (MCW 1950e).

5.3 INTERNAL DOSE CONSIDERATIONS

The primary route of internal exposure was via inhalation of airborne particulates, although radon was a considerable problem in some areas. It was noted in December 1942 (MED 1942) that the primary precautions taken against radiological hazards were against inhalation of the dust. The wet operations did not generate as significant a level of airborne particulates as the dry operations (Mason 1958a). Thus the operations that were likely to produce airborne particulates were those in which the uranium-containing material was dry or was heated, although airborne contamination could occur even when the material was not open to the room air because some of the equipment leaked (Caplan and Mason 1952). Material that had settled on the floor and other surfaces could also be resuspended in the course of operations.

Very little internal exposure data (in dose units) is available. Measurements of airborne concentrations were used per se as indicators of potential exposures and of the need for changes in processes and equipment (as suggested by AEC 1950a). Urinalysis appears to have been performed as a means of retroactively checking for acute exposures or for the onset of damage due to chronic exposure, rather than a dose measurement per se, although some dose calculations were apparently done by AEC for study or verification purposes. In addition, as discussed below, the particle size and solubility of the various uranium forms were still being investigated experimentally by researchers in the field, so that the assumptions that were used at that time in calculating internal doses might not be acceptable based on current knowledge. Therefore in this technical basis document, potential internal exposure is presented in terms of airborne concentration data (and the derived inhaled amounts) and surrogate urinalysis results, rather than internal doses, so that dose reconstructors may estimate individual internal doses using modern methods.

5.3.1 Particle Size, Solubility, and Composition Considerations; Air Sampling Methods

A discussion of the thinking of the time regarding particle size and solubility is included in the discussion below in case questions arise regarding typical operations or regarding notes in the urinalysis and other records.

The uranium refining operations at Mallinckrodt produced nominally insoluble uranium compounds, e.g., UO_2, UF_4, and uranium metal (Lippmann 1958, regarding solubility); no patently soluble forms, such as UF_6, appear to have been produced. However, as some of the literature in the years between 1958 and about 1975 showed, some supposedly insoluble particles produced in these operations seemed to behave like soluble particles (e.g., Lippmann 1958, Heatherton 1975, Archer et al. 1975). More recently, ICRP 71 (ICRP 1995a) states that studies of UF_4 show behavior consistent with Type F in some cases and Type M in other cases, while ICRP 68 (ICRP 1995b) recommends Type M for UF_4. Yet autopsy data from deceased workers showed a far lower concentration of uranium and thorium in lung and other tissues than would have been expected based on the average airborne concentrations inhaled. This led Eisenbud (1975) and others to postulate that the particle behavior formulation at the time was incorrect: they conjectured that due to the high density of the uranium and daughters, particles of respirable diameter behaved like lighter particles of much larger diameters.

A study of air dust particle sizes in the UO_3-to-UO_2 area was done in 1948 for AEC by the University of Rochester (1948b). Both filter paper dust samples and cascade impactor samples were taken at the tumbling area (where the newly formed and cooled UO_2 was place in tumbling drums and rotated on a tumbler to break up clumps); at the skip hoist (where the tumbling drums were opened and transferred to the hopper of the Fiberpack loader); at the Fiberpack loader (where a vibrating chute fed the material into 75-lb containers and the containers sealed on a roller conveyor); and at the warehouse (where the UO_2 containers were stored). The sampling rate of the cascade impactor was 14 liters per minute; it is unclear what the filter paper sampling rate was. All samples were analyzed for uranium by the fluorometric method and the results were found to be well within the accuracy range of the method. Particle size distributions showed normal curves and to be in excellent agreement with the theoretical curves. The results are shown in Table 7.

Also shown in Table 7 are some measurements reported by Sanders (1975). Sanders (1975) also provides a useful general observation that foundry operations (those in which uranium oxides are produced) have the highest percentage of airborne particles in the lower lung-inhalation range, while reduction operations (UF_4 to metal) have the smallest percentage.

Eisenbud (1958) reported that "It has been shown that in these [uranium processing] plants [the uranium processing plants] the mass median diameter was about 2 μ [microns]". But he also noted that while the peak for alveolar retention is 1-2 μ for dust of unit density (i.e., 1 g/cm^3), uranium oxide dust has a density of 9 or 10 g/cc, so that a 1-2 μ particle would behave as though it were a 3-6 μ particle of unit density. An AEC report on uranium mills stated that ore concentrates (the form that arrived at uranium refineries such as Mallinckrodt) had a "mass medium [sic] size" of 2.5 μ (AEC 1958). Some uranium refinery workers were said to have been exposed to UO_3 dust with a mass median diameter of 2 µ; however, with a density for U compounds in the range of 9-10 g/cm^3, the effective aerodynamic diameter was much larger than 2 µ, possibly in the range of 5-6 µ (Eisenbud 1975).

Schwendiman et al. (1975) cited the results of a study by Lippmann and Harris regarding the application of size-selective samplers in the uranium industry. Lippmann and Harris performed a sampling survey of six different uranium processing plants. They used a two-stage setup: following a cyclone separator, the first stage collected 100% of particles ≥ 10 µm AED, 75% of those = 5 µm, 50% = 3.5 µm, 25% = 2.5 µm, and 0% of those ≤ 2 µm, while the second stage, a filter, collected the rest of the particles. They found that less than 15% of the airborne material collected on the second (the respirable-particle) filter. Schwendiman et al. (1975) noted that this agreed fairly well with the ICRP assumption of a size distribution such that 25% deposits in the lower respiratory passages and, for insoluble compounds, only half of the 25% (or 12.5%) would be retained with biological half-life of 120 days: only 10% of the samples in the study had more than 25% collected on the second filter. Categories studied included U_3O_8, UO_3, UO_2, ore concentrate, and mixed scrap.

Because of the lack of specific information regarding all types of particle sizes at the Mallinckrodt plants, the ICRP 66 default deposition parameters (ICRP 1994) should be used to estimate internal doses for Mallinckrodt workers. However, where there are particle size issues in specific cases, the information above and in Table 7 should be of general application even when the listing is for depleted or enriched rather than for the natural uranium used at Mallinckrodt.

Regarding the isotopic and chemical composition of the dusts, there is only a little Mallinckrodt-specific information. In 1947 AEC had the ore and residue dust studied by the University of Rochester, which investigated the uranium and radium content of filter paper after use in air sampling (Rochester 1948a); it was stressed in the University of Rochester's report on the results that the samples were taken not to determine health hazards directly (i.e., by inferring the air concentration from worker positions) but rather to obtain a heavy concentration of each material so as to compare the relative concentrations in the dust. Thus each concentration was taken at the maximum emission point (in fact the centrifuge was partially opened for the sample-taking), without reference to actual worker position or breathing zone. The precision of the measurements was said to be better than +/-10% and each figure in the given represented at least three analyses. See the table below for the results and the inferred curie ratios. As can be seen, the U/Ra ratio was approximately 1 in the ore, indicating equilibrium of the uranium and at least its first few daughters; 0.01 in the gangue (GLC or K-65) residue, indicating removal of most of the uranium; and intermediate during the centrifuging of the residue to separate the liquid and solid (Ra-bearing) fractions.

Comparison of the uranium and radium content of ore and gangue (K-65) residue samples.

			Uranium		**Radium**		**U-238 Ci/ Ra-226 Ci**
Compound	**Area**	**Operation**	**ug of sample**	**u/m^3**	**ug of sample**	**u/m^3**	**Ratio**
Ore (U_3O_8)	Ore Room	Crushing and milling ore	540	2,540	182	850	1.0
			520	2,440	184	860	0.95
			260	1,220	102	480	0.86
			1,010	4,750	307	1,440	1.1
			760	3,560	220	1,030	1.2
Gangue residue	Scalehouse Sampliing Room	Sampling residue	21	148	665	4,700	0.011
			17	120	555	3,920	0.010
	Centrifuge	Centrifuging residue	10	71	25.5	180	0.13
			11	78	18	130	0.21

The precision of the measurements was better than +/-10%. Each figure above represented at least three analyses.

Observations at a large uranium processing mill (probably Anaconda) showed that although the potential for dust creation in the dry processing steps was clear, the wet processing steps, such as grinding, leaching, separation, and precipitation could create aerosols by agitation or by transfer of solutions and slurries. Even so, in the wet processing areas airborne uranium was never a major problem, i.e., local hooding and exhaust ventilation were not required in order to keep the dust concentration below allowable levels. The physical characteristics of the airborne particulates in the dry and the wet areas were thought to be similar; thus it was assumed that nearly all of the mass of the particle was siliceous material, clay, or a mixture of the two. The mass of the particles was found to be less than one percent of the total mass. From specific nuclide analyses done over several years, they concluded that secular equilibrium existed at least through Ra-226 in the ore at the time of mining. Specific nuclide analyses of particles taken in various areas of the mill showed that the U-238:Th-230:Ra-226 ratio was 1:1:0.3 and was fairly constant regardless of the source of the aerosol or dust. But in the yellowcake section of the mill, where the ore had been processed to become mostly U_3O_8, the ratio was 1:0.01:0.001, indicating that these products were removed in the processing (Wilde 1975). However, as discussed in the paragraph above, at least the one study of what was probably Belgian Congo ore at Mallinckrodt showed that secular equilibrium down to just above radon had been re-established by the time of processing at Mallinckrodt. Thus it seems best to assume that equilibrium did exist in the ore.

AEC began taking air dust samples for particulate alpha emitters in 1943 and Mallinckrodt appears to have been taking dust samples and sending them to NYOO for evaluation from about 1943 on (MED 1944j). The various AEC air dust study reports state that an established AEC protocol was followed, but the documentation for what the protocol was is lacking. However, some information is known about Mallinckrodt's and AEC's methods. MED (1943d) stated that the sample rate for measurements in the second quarter of 1943 was 3 ft^3/min, with either 30 or 120 ft^3 sampled, depending on the area. Mallinckrodt (MCW 1946g) stated that dust was measured with a "precipitron and ion-meter". For the data reported by MCW (1949d), dust samples were collected on 1-1/8" Whatman #41 filter paper disks, using a modified Fischer pump, at 0.5 ft^3/min, over a period of 45 seconds to 30 minutes depending on conditions and job time. The disks were counted on a parallel plate alpha counter such that statistical variations would be no more than +/-10% at the 0.9 confidence level. It was stated that a few of the low-level samples might have errors of +/-20%. Data from 1946 lectures by A. A. Jarrett of Clinton Laboratories were used in the error calculations. AEC (1949g) also states that Mallinckrodt was using a flow rate of 0.5 ft^3/min, with a Fisher and Porter rotometer. AEC (1950a) stated that it was using 1-1/8" Whatman filter paper disks and a handheld air sampler with a collection rate of 15-20 lfm to take its samples at Mallinckrodt.

From reports of sampling at other sites and from unspecific mentions in papers and reports about Mallinckrodt, it appears that typical practice was for the air to be drawn through filter paper (usually Whatman #41, widely used in the time period) and counted on an alpha scintillation counter (e.g., the laundry samples of Utnage 1958b). AEC in-plant air sampling was done by collection on Whatman #41 filter paper and counting of total alphas; a correction for self-absorption in the filter paper was applied (Eisenbud 1975). The results were reported as alpha dpm/m^3 (Eisenbud 1975). The AEC, in sampling stack and environs air at various sites including Mallinckrodt, used Whatman #41 filter discs with "standard sampling equipment and techniques normally employed by HASL" and counted them on scintillation counters (Weinstein 1958). In 1958 at the Oak Ridge Gaseous Diffusion Plant, the Whatman #41 paper was used with fixed counting equipment, for a counting and collection efficiency of 30% (Becher 1958). Since the methods of the time seem to have been fairly uniform (with HASL setting the standard), it is assumed that the AEC and Mallinckrodt measurements were taken consistent with these references.

In 1948, some filter paper samples were taken at Mallinckrodt, conveyed by an AEC inspector to Clinton Laboratories in Oak Ridge to be counted for alpha and beta-gamma activity, and then returned by the inspector to Mallinckrodt to be counted for alpha activity there, apparently as a comparison test (AEC 1948a). The samples were taken in the ore milling area, where ore drums were opened and the ore was ground to the desired consistency for later digestion; thus it can be inferred that the samples showed uranium in equilibrium with its daughters down to radon (as suggested by the results of Rochester 1948a – see table above). The counting equipment was said to be a parallel plate counter at Mallinckrodt, but was unspecified for Oak Ridge. For both the Oak Ridge and Mallinckrodt alpha counting conditions (with identical geometries), the absorption loss was assumed to be 30% and the geometry factor to be 52% (with the extra 2% above the 50% being ascribed to backscatter of the alpha particles). For the Oak Ridge beta-gamma counting conditions, the absorption loss was assumed to be 30% and the geometry factor to be 10%; the beta-gamma counting apparatus at Mallinckrodt was being repaired, so no beta-gamma measurements were made there. The (alpha?) background at Oak Ridge was 4 cpm, while the background at Mallinckrodt was 20 dpm. The conversion factor for alpha was given as 2.75 dpm per cpm and for beta-gamma as 14.3 dpm per cpm. (Although these figures were backward in the text of the reference, the data sheets show the correct correspondence.) In 1949, Mallinckrodt (MCW 1950g) stated that it was assuming the counter geometry to be 52% and the filter paper absorption to be 30%.

AEC (AEC 1948a) noted that the samples taken could not be read at Mallinckrodt for several days after they were brought back from Oak Ridge because the parallel plate apparatus and the counting room had to be decontaminated and the background reduced from 110 cpm to 20 cpm. The agreement for the alpha samples was fairly good, in general mostly within 25% and in many cases much better. For a subsequent set of samples counted at Mallinckrodt (with the background now down to 6 cpm), the same AEC inspector reported (AEC 1948b) that the formula used to convert from alpha cpm to microcuries per cubic meter was as follows, with the 0.3 and the 0.52 being the absorption and geometry factors respectively and the 0.5 being the intake rate of the sampling pump:

$$\#\ \mu\text{Ci/m}^3 = \frac{\#\ \text{cpm} \times 35.3\ (\text{ft}^3/\text{m}^3)}{(1 - 0.3) \times .52 \times 3.7 \times 10^{10}\ (\text{dis/sec-Ci}) \times 60\ (\text{sec/min}) \times .5\ (\text{ft}^3/\text{min}) \times 1 \times 10^{-6}\ (\text{Ci}/\mu\text{Ci})}.$$

A report issued about the same time by AEC to Mallinckrodt gives the results of dust samples taken in an unspecified Mallinckrodt location and counted apparently at NYOO (AEC 1949i). Counts are given in dpm per cubic meter, with alpha counts given for all samples and both alpha and beta-gamma counts given for about half the samples. These latter show alpha-to-beta-gamma ratios ranging from 0.98 to 6.44, with most values in the range 2.5-3.4.

John Harley, former director of HASL, told an epidemiological studies group (ORAU 1983b) that the equipment used to do the air dust surveys was not very good. He said that HASL had confidence in readings greater than or equal to 25 ug/m^3, but in the range of 10 ug/m^3, the results were thought to be “shaky”. He added that although most of the production sites did not do breathing zone sampling (only general air sampling) and some even resisted it, Mallinckrodt did do breathing zone sampling (e.g., MCW 1949d).

The AEC’s HASL staff was committed to the time-weighted average as being most representative of total exposure. As Glauberman and Harris (1958) put it, “HASL has found from experience that the multiple-sample time-weighted average exposure procedure is the most accurate....The GA [general area] sample normally will tend to underestimate an operator’s exposure and the BZ [breathing zone] sample to overestimate it, but by time-weighting the average concentrations for both types of samples an operator's exposure may be closely evaluated...[this method] yields reasonably reproducible results". In this technical basis document, time-weighted averages will be preferred to static measurements or short-term maxima as being representative of worker exposures. Besides that, AEC was interested in using the feasibility of the levels of urinary excretion of uranium as a control metric in production and processing facilities (AEC 1949d); to do this, it was necessary to correlate the urinary measurements with the dust exposures, which could not be done if only general, instantaneous dust levels were used. The time-weighted averages for Mallinckrodt are documented in a series of AEC air dust study reports (e.g., AEC 1954b) and in some Mallinckrodt reports (e.g., MCW 1949d).

5.3.2 Airborne Dust Levels

Eisenbud (1975) stated that "above all other types of exposure, it was the airborne alpha-emitting dust that was the cause of greatest concern." This was particularly true in the sampling and crushing of ore and in the mechanical and manual handling of dry uranium salts and oxides (Eisenbud 1975). Dupree-Ellis et al. (2000) stated that daily average uranium dust concentrations of up to 100-200 times the maximum allowable concentration of 50 $\mu g/m^3$ were measured in poorly ventilated processing areas. An industry-wide survey showed the average concentration to be 5000 dpm/m^3, which an AEC-HASL official stated was "a conservative estimate of the levels that existed from 1942 to 1948" (Harris 1958).

Mason (1958a) stated that while no regular dust sampling program was in effect at Mallinckrodt during 1943-1947, enough samples were collected to show that concentrations were high by 1958 standards, that concentrations of 50 to 100 times the MAC level of 70 dpm/m^3 were not uncommon, and that some operations produced concentrations up to 1000 MAC for a few minutes at a time. AEC also stated many employees were exposed to elevated dust levels for years (AEC 1949b); AEC estimated that the inhalation of many Plant 4 operators involved in UF_4 production was 27 grams or more of uranium, as compared to what it termed a "life tolerance" of 6 grams, and that an additional two years' of exposure would add about 2 more grams (MCW 1949b).

The high levels were of concern to both MED and Mallinckrodt. MED (1944h), in transmitting to Mallinckrodt the results of dust samples taken in June 1944 in the bomb and furnace areas, noted that the results were high and that either ventilation in those areas should be improved or respirator use should be required. Mallinckrodt too was concerned by the high levels, for example the fact that dust concentrations around the various crushing, grinding, and packaging operations consistently exceeded the tolerance levels by a factor of 1-300 (MED 1944k). As a result, Mallinckrodt hired a consulting engineering firm to put together a re-design proposal for dust control (MED 1944k).

MED (1944n) stated the following. In September 1944, MED and Mallinckrodt representatives met to discuss proposals for ventilation improvements in the green salt and metal production areas. It was decided that the ventilation in the green salt furnace room did not need improvement because there were 30 air changes per hour and because the "smell of HF [hydrogen fluoride] was faint". But on the other hand, the green salt unloading, grinding, milling, and blending operations were so dusty that it was decided to do them henceforth under hoods in a continuous sequence, instead of as separate operations, and the blending would be done mechanically instead of manually. Also, the bomb loading, jolting, unloading, and charge blending and the biscuit chipping, all of which were carried out in close proximity, would be provided with separate, localized ventilation. Finally, the green salt blending operation would be redesigned to use a tumbler-type mixer instead of manual mixing, after which time the blender table would no longer be used. Thus in about 1944-1946, significant incremental improvements in dust control were made that reduced initial production-level dust concentrations in the green salt area.

Respirators were said to be required “for practically all plant operations” in 1946-1948 (Mason 1958a) but respirators were not used consistently (see Section 5.3.3 below).

Major improvements in dust control and ventilation were made at Mallinckrodt in 1949 under the new AEC health program, such as the installation of pneumatic unloading and conveying equipment in Plant 6 process areas that eliminated most hand-scooping and thus that mode of exposure to dust (Mason 1958a). However, while there was a marked reduction in dust levels, the improvement was not what had been hoped for in some areas, especially with respect to the handling of the UO_3 (Mason 1958a). In 1950, for example, it was reported that the air supply system in the Ore Room was such that there was a significant outward flow through the drum opening/closing area shield (MCW 1950u); in 1953, an AEC inspector reported that there were many small openings between the operating area and the drum storage “alleys” in the Ore Room addition; as a result, winds blew into the area and upset the ventilation air balance, causing dust to be blown into the operators’ area (AEC 1953). Even the most modern plant, Plant 7, had dust problems: in 1954 an AEC inspector noted that there was a fine film of UO_3 on supposedly clean drums and that as cans of UF_4 were transferred from a hooded enclosure to a conveyor, a green dust cloud could be seen to escape from under the lid of nearly every can (AEC 1954f).

Similarly, in 1948-1950, tighter adminstrative controls were instituted and much of the manual handling of UO_2, UF_4, and uranium metal in Plant 4 was eliminated, but dust concentrations did not get down to satisfactory levels (Mason 1958a). Even the building of the new Plant 6E and Plant 7 did not completely eliminate the problem: the uranium was never contained well enough that it ceased to create airborne levels of concern in the plant air and in the (multi-building) plants in general (Mason 1958a). A Mallinckrodt official remarked that it had been a constant battle to keep airborne levels at 1 MAC or less (Mason 1958a). An AEC safety official speaking of workers at all the AEC uranium-refining plants (Breslin 1958) reported that even in 1951 approximately half of the workers were exposed to average concentrations above the MAC then in force (which he said was 110 dpm/m^3, although AEC (1954f) indicates that it was 70 dpm/m^3 until at least the end of 1954). He also stated that in 1956, 6% still were above the MAC; the percentages for those exposed to average concentrations greater than 1800 dpm/m^3 were 4% and 1% respectively. Finally, he noted that while airborne control in these plants was largely achieved by 1955, there had been a retrogression resulting from a large increase in production volume.

Tables 8 and 9 give a trend overview of airborne uranium concentrations measured over the years at Plants 4, 6, 6E, and 7 (Mason 1958a). The concentrations are given as multiples of the “preferred level” at the time of measurement (1948), i.e., multiples of 70 dpm alpha per m^3. It appears from the reference that they represent typical or representative concentrations rather than maxima. In AEC’s measurements, they did not separate the uranium and radium components of the alpha activity, i.e., they counted gross alpha (AEC 1950a). This was Mallinckrodt’s practice as well (MCW 1950c; MCW 1955d); also, beta-gamma activity analysis was not routinely done (MCW 1955d). Thus Tables 8-16 must be assumed to be gross alpha measurements, whoever took the measurements. There were no continuous air monitors at the Mallinckrodt site, at first because there was no commercial monitor available (MCW 1950e) and later because the results were thought to be of doubtful value (MCW 1955d).

Table 10 presents some early air sampling data taken by MED. The samples represent single point measurements and no correction was applied for exposure time. Thus unless some assumptions are made as to time spent in the given area performing the given operation, only a bounding or conservative dose calculation can be performed with this data. However, there are some after-operation or "not in operation" measurements among the data that could help to characterize an average level, if needed.

In this technical basis document, AEC's data are used preferentially because AEC set the standard of measurement for the uranium processing sites and because AEC's figures for the most exposed workers are typically higher than Mallinckrodt's. Thus using AEC's numbers is claimant-favorable, in general. The AEC data are mostly from a series of dust studies that AEC did on a nearly annual basis from about 1948 on. This is presented, with a few additions from Mallinckrodt reports to fill gaps, in comparative form in Table 11 and in comprehensive form in Tables 12 through 16.

Table 11 presents the results of airborne dust surveys made in Plant 4 by, respectively, AEC-NYOO's Medical Division in May 1948 and Mallinckrodt safety officials in September-October 1948. These results are given as time-weighted daily average levels (called DWE levels by AEC) of dust exposure by plant. Table 11 shows that AEC and Mallinckrodt's data were in general agreement (AEC (1949b) termed it "excellent agreement"), although there were some differences. Mallinckrodt (MCW 1949d) stated that while the Mallinckrodt and AEC results did differ significantly in some instances, it was thought that none of the differences was highly significant except in the case of the office workers; Mallinckrodt (MCW 1949d) also noted that the (overall) lower results of the Mallinckrodt studies were due to Mallinckrodt's taking more control samples and counting low-activity samples for longer times to reduce statistical error.

One notable difference is for "Cage handling", where the AEC-measured level is given as 2.7 times the tolerance level and the Mallinckrodt-measured level is given as 52 times tolerance, or 189 and 3640 dpm/m3 respectively. The ratio of these two values is about 20. In the original report in which AEC reported its values (AEC 1848l), the job description and summary listing of job categories and DWEs does not include a mention of the cage handling function; there is only a mention of the "Saw man" as taking billets to the cage area. However, in the original report in which Mallinckrodt reported its values and where the comparision table (assembled by Mallinckrodt) first appears (MCW 1949c), there is a category called "Cage man". Thus it appears that the job category of cage man did not exist until mid-1948 or after, i.e., after the AEC study was made. Mallinckrodt seems to have put together an estimate for the "Cage handling" category from the AEC data as some sort of composite of the values for two or more of the other categories, probably including the slag handler (1.6 times the tolerance level). However, given the general functions of the cage man as later described in other dust studies, the cage man's exposure was more likely to be similar to that of the saw man (15.8 times tolerance according to AEC) and the chipper (26.8 times tolerance according to AEC).

Further, an inspection of the detailed job sheet for "Cage man" in the Mallinckrodt report shows that about two-thrids of his dust exposure was due to a single activity, "Dumping D-7". What D-7 was is not defined in any available Mallinckrodt document but it appears to be a form of slag dust.

Dumping D-7 is again mentioned in the Mallinckrodt dust study done in December 1949, but the average levels reported here were reduced by a factor of about 1000 from those of the earlier Mallinckrodt study. This reduction is most likely attributable to admininistrative measures taken in 1949 to reduce dust levels in the highest-level activities, but it could also have been due to malfunction of the sampling apparatus in the earlier study, as explained below.

Also, the high value listed in the 1948 Mallinckrodt study (MCW 1949c) for "Dumping D-7" appears to have been thrown out in the calculation of the DWE by Mallinckrodt. This may have been due to a belief that the sampler malfunctioned in taking that sample. It should be noted that the average level in the next highest area of Plant 4 as measured in this study was less than half this value and that the known highly dusty operations reported on by Mallinckrodt (MCW 1949d) for Plant 6 for October-November 1948, such as ore room, pot room work, and UO_2 handling activities, typically had highest-sample levels that were lower by 50% or more than the MCW (1949c) “Dumping D-7” value. (The pot room and UO_2 handling had only single samples.) Because Mallinckrodt did not include this value in the calculation of the average, probably because they thought it suspect, it is possible that the other D-7 measurements apparently taken at the same time may also have been higher than the true values. Thus the value for “Cage handling” attributed to AEC by Mallinckrodt may in fact be more correct.

Thus the AEC-Mallinckrodt discrepancy in the “Cage handling” category may be due to the job’s being new with the Mallinckrodt study, to a mis-estimation of the cage man’s exposure from the AEC data, or to a malfunction of the sampler. AEC and Mallinckrodt obviously did not think this discrepancy was significant at the time. However, because it is not possible to determine this from the information available (including missing pages in the project copy of the AEC report (AEC 1948l)), the claimant-favorable assumption is made that the Mallinckrodt value was the more correct for 1948 and the cage man’s exposure value was used in Tables 29B and 31B to calculated inferred intakes.

An additional note about the dust levels measured in 1948-1949 and their applicability to previous years is in order. It is clear from the inclusion in Table 1 of Mason (1958a) of 1946 through 1950 figures (repeated in this technical basis document as Table 8) that 1946 and 1947 data was available that Mason thought appropriate for comparison to 1948, 1949, and 1950 data, or at least that Mason believed (by his repetition or plateauing of the same values throughout 1946-1948) that the 1948 data was representative of the 1946 and 1947 operations as well. This can be inferred for Plant 4 as well from Table 2 of Mason (1958a) (repeated in this technical basis document as Table 8). Thus although the report argues from the paper’s Table 1 that significant improvements were made by late 1948 through 1950 that reduced dust levels considerably, this is irrelevant to a significant conclusion that can be drawn from Mason’s tables: that the 1948 data was representative of the 1946-1947 data.

This is also borne out by other information from Mason (1958a). This paper states that the dust concentrations “in some operations” in 1942-1943 “were considerably higher than present standards”. However, the paper was likely written in 1958 (the conference at which the paper was given took place in October 1958), so the “present standards” were those of the late 1950’s. Mason might arguably have made the same comment about 1946-1948 since he also stated of the 1946-1948 operations in Plant 6 that “during initial operations dust control was minimal, and it can be seen that air concentrations were high during the period 1946 through 1948, when respirators were required for practically all plant operations”. While Mason stated that the new health program was authorized in 1947 and got under way in 1948, he also said that one of the first projects of the new Mallinckrodt Health Department was “a thorough analysis of the dust data already accumulated and the immediate collection of additional data to enable an estimation of dust exposure already received by operation and maintenance personnel”. Thus even in 1948, apparently no immediate engineered improvements were undertaken. Mason went on to say that only in 1949, under the new health program, “immediate steps were taken to install good ventilation and dust control and to initiate process improvements”. This implies that the point at which dust levels began to go down significantly in Plant 6 was in 1949, as is also shown from Mason’s (1958a) Table 1. Regarding Plant 4, Mason stated that mechanization was installed in 1948 and 1949, but since he gave the same figures for 1943-1947 as for 1948, it must be concluded that either he considered the 1948 values to bound the 1943-1947 ones or the values he used for 1943-1948 (from whichever year) corresponded to those that were representative of the entire period.

Mason (1958a) referred to his Tables 3 and 4, showing Plants 7 and 6E values respectively (repeated in this technical basis document as Table 9), as showing an improvement of “a factor of 60 compared to 1943 figures and by a factor of 8 compared to 1950 figures" for Plant 7 versus Plant 6 and a reduction factor "of about 20 compared to 1943 figures and 8 compared to 1950 figures" for Plant 6E versus Plant 4. These explicit comparisons too suggest that Mason had information as to the early (1943) dust levels and that his tables reflect that. Mason also plotted his data from 1943 on, again typically showing a plateau for the years 1943-1947.

Mason (1958a) noted that "no regular dust sampling program was in effect during 1943 through 1947, but sufficient samples were collected to show that airborne uranium concentrations were high by present standards; concentration of 50 to 100 times the present MAC [70 dpm/m3] were not uncommon, and some operations produced concentrations up to 1000 MAC for a few minutes". However, such levels were also measured in the later dust studies, e.g., near the end of the paper, Mason commented that it was "evident that personnel did work in fairly high concentrations in the early days of the operation, and that the exposures received depended partly on the effectiveness of the respirator program". However, he appears to consider the early days to be 1942-1947 or (by his tables) 1942-1948.

This does not mean that the dust exposures in about 1949-1951 were not also high (above the preferred level), although not usually as high as in 1948, as was shown by the following table of data. Some dust sample measurement data from 1948 and 1950, in multiples of the PL*.

Survey date	**May 1948**	**Nov 1948**	**Jul-Aug1950**
Survey performed by	AEC	Mall	Both
General information about the studies			
Number of workers in areas studied	170	279	406
Percent at less than the PL*	31	43	61
Percent at 1-3 PL	20	32	31
Percent at 3-5 PL	23	3	5
Percent at 5-10 PL	13	13	0.7
Percent at 10-15 PL	2	0	0
Percent over 15 PL	12	9	2.5
General Categories (in multiples of the PL)			
Average exposure	53	12	2.1
High exposure	660	195	20**
Ore Room operations	71	195	4.9
Digest/Feinc/feed makeup/C-3, centrifuge operations	3.4 - 12	9.8	1.1 - 2.2
Pot Room operations	460	111	4.8
Raffinate and recovery operations	5.2	3.9	1.8 - 2.3
Rockwell (brown) furnace operator	350	76/45	20/3.1
Brown oxide packer	560	161	5.2
Warehouse worker	3		1.4
General maintenance and mechanics	1.8		1.3
Boiler House		0.63	0.62
Digest pilot plant	3.5	1.3	1.5
Experimental continuous furnace (pilot plant)	Not in operation yet	Not in operation yet	122
Ledoux Lab K-65 sampler		30	20
Shotgun Lab		0.34	3.4
Production offices		1.4	0.2
MCW offices	0.7	0.08	0.013
AEC offices	0.5	0.11	0.013
Guards	1.1		0.5
Health office	0.2	0.1	0.16
Other Information			
Ore Room: cleaning ore drums, dpm/m^3	Range 1710-127000, avg 64400, for 30 min	---	Range 253-496, avg 374, for 64 min
Ore Room: general air, operating, dpm/m^3	Range 275-8920, avg 2590, for 125 min	---	Range 700-5750, avg 1954, for 20 min

Figures are from AEC (1948l), MCW (1949d), and AEC (1950o). Dashes in the table above indicate the range given in AEC (1950o), while slashes indicate two types of workers of the same title, e.g., day shift versus night shift workers.

* The PL is the preferred level, equal to 70 dpm/m^3. All dust sample measurements are in multiples of the preferred level (PL, or 70 dpm/m3), except as indicated.

** This figure does not include the exposure of the three Experimental Continuous Furnace workers' exposure at 122 PL, although their exposures were included in the calculation of the percent over 15 PL.

Here we see that in 1948 there were areas where the exposures were indeed "50 to 100 times the present MAC [i.e., 50-100 times the preferred level of 70 dpm/m^3, or 3500-7000 dpm/m^3]" and that some operations, e.g., the ore room drum cleaning, clearly did produce concentrations up to 1000 MAC (70,000 dpm/m^3) for periods of minutes at a time. Thus it appears that the 1948 figures do fit Mason's description of what "high" was.

Some comparisons were made between spot dust levels measured in 1943-1946 (Table 10) and the corresponding ones measured from 1947 on, with the point being to verify that for the presumably peak or near-peak points of individual operations, the 1943-1946 dust levels were generally no greater and were usually less than they were later, after Plant 6 was in full operation. This too supports the conclusion that the 1948 figures, prior to installation of significant upgrades in Plant 6, would bound the experience in the earlier years.

Data similar to that in the table above can be shown to exist for Plant 4 (i.e., for 1948 and before), so that the "50-100 times" quotation appears to apply for Plant 4 as well. Also, in AEC (1949c), a memorandum from early 1949, an AEC official summarized the hazards in Plant 4, which he said were to be solved by the completed designs for Plant 6E, including dust levels of up to 186 times the preferred level. This suggests that the 1948 figures for Plant 4 are also indicative of the exposure levels of earlier years. The spot dust levels for the early years, when compared to those of about 1947-1948, also appear to show that the 1947-1948 levels bound the early years.

It can be concluded that Mason (1958a) and the references supporting the table above show that the 1948 figures did bound or were representative of the figures of the earlier years. Comparisons with the data from earlier years are consistent with this conclusion.

Some detailed information is available about the particular case of the laundry workers circa 1958, when the laundry had operated for at least ten years (Utnage 1958b), as shown in Table 17; some information about the laundry was also found in the various AEC dust study reports.

Information from other sites is helpful in deducing what would be typical at Mallinckrodt. In an AEC report in 1958, the breathing zone concentrations in the final ore concentrates packaging areas of over a dozen mills were evaluated; the concentrations ranged from 0.01 to 5.5 pCi/L, with a median of about 0.15 and a mean of 0.91 (AEC 1958, Table V). These figures suggest levels that might be encountered by Mallinckrodt workers unloading the packages (drums) at the beginning of the refining process. See also Section 5.3.5 below regarding resuspension of surface contamination. Resuspension contributions are assumed to be included in all data cited in this technical basis document since dust levels were typically measured while work was taking place.

No information is available regarding dust exposures at SLAPS. This was likely due to low radioactive dust levels in this open area (i.e., most of the dust collected would have been nonradioactive even in the dump pile areas, except possibly when the dumping was actually taking place) and to the contained nature of the K-65 residue. Because the SLAPS workers likely were subject to higher dust levels in their regular work at the main St. Louis site (e.g., in the warehouse or loading the dumpsters with barium sulfate residues or doing guard rounds in the plant), their dust exposures are based on their main St. Louis site plant exposure and thus are expected to be conservative.

The reported air concentrations generally pertain to those workers directly and continuously involved in uranium refining work. However, Breslin (1958) defined "auxiliary workers" as workers "not directly connected with production but located in or near production buildings,....[including] chemists, engineers, office workers, garage mechanics, outside maintenance personnel, and the like", noting that some of these had occasion to visit production areas in the course of their work while others did not. Even with the limited access, in 1948 about 13% of the auxiliary workers studied were exposed to average concentrations above the MAC (110 dpm/m^3 at that time) and more than 1% to concentrations greater than 440 dpm/m^3 (Breslin 1958). By 1954, none of these auxiliary workers were exposed to average concentrations above the MAC (Breslin 1958), although some of the process workers still were. These statements are illustrated by the data given in the various AEC dust study reports.

The case of the maintenance and craft worker requires special consideration. Area mechanics were assumed to be dedicated to the particular process area or plant, with their work time being mostly spent in and around process areas, and thus their daily weighted average inhalation exposures should be considered to be representative of their residence time in the process areas. Similarly, their radon exposures can be taken to be those representative of their process areas. However, it is more difficult to determine inhalation and radon exposures for maintenance and craft workers who spent part of their time in their shop areas, where contaminated equipment might be brought to them to work on at irregular intervals, and part of their time working on equipment in process areas. As Hickey and Dupree (1984) pointed out, Mallinckrodt Uranium Division maintenance and craft workers were not usually assigned to a particular process (with the exception of area mechanics, which Hickey and Dupree do not note) but served the entire division. Not only Breslin (1958, quoted above) but also MCW (1950c) pointed out the variability of exposure of these workers as compared to the process area workers.

AEC and Mallinckrodt dust studies from 1948 on list at least some maintenance workers besides area mechanics as a category and tabulate the time spent in various activities and areas in some detail (e.g., MCW 1949c; MCW 1949d). However, AEC (AEC 1954b) and MCW (MCW 1949d) stated that the studies they described used only general area samples to estimate the exposures of maintenance workers and not breathing zone samples while they were performing specific operations (unlike the case with the process workers). MCW (1950c) made a similar point with regard to the AEC-directed back-calculation of dust exposures to Plant 4 employees in 1949. Although it is not possible to determine what the temporarily elevated levels in the shops and the resulting somewhat higher exposures were, it can be concluded from the data in the various dust studies that the shop area dust levels were lower than the process area levels, usually decidedly so, and that thus the most significant contribution to the exposures of these workers most likely came from their time spent in process areas. It is assumed that from 1948 on, this time is factored into the exposures of these workers (as part of the daily weighted averaging calculation). However, for the years from 1942-1947, it is appropriate for the purposes of dose reconstruction to determine from the dust study tabulations reasonable and conservative percentages of time spent by the maintenance and craft workers in process areas and to use the percentages to determine the potential exposures during the time spent in the process areas. The table of these percentages and an explanation of their use are given in Section 6.1, Item 11 rather than here (for the convenience of internal dose reconstructors).

Relatively high potential for dust exposure applied not only to those actually present in the dusty buildings and to some extent to those working elsewhere in the plants, but even to those outside the plants. For example, a Mallinckrodt safety official remarked that one reason to revise the ventilation in about 1952 had been that a study of plant effluents showed that “large bursts of dust found their way outside of the plant immediately after filter cleaning” (Harris and Mason 1953). Mason (1958a) also suggests that co-located (nearby but uninvolved) workers were exposed to elevated airborne levels.

In later years, Mallinckrodt was supposed to sample stacks at least once a year, but it was not being done (MCW 1955d). Weinstein (1958) reported on an air sampling study that AEC-HASL did of stack and environs (outside) air at various sites, including Mallinckrodt, in November 1949. They did not take any stack samples at Mallinckrodt, but Weinstein indicated that previous data implied a probable average rate of emission of uranium from the Mallinckrodt stack(s) of about 0.011 g/sec, with a flow rate of about 20,000 cfm. About 52 tons of uranium as metal was estimated to have been discharged in the stack effluents since the beginning of operation. While nearly every reported (outside) concentration at Mallinckrodt was below maximum permissible levels, it was observed that “1000 feet would circumscribe the MAC” level (out from the plants), i.e., within a few hundred feet of the plant(s) the MAC might be exceeded. The MAC given in the version of 10 CFR 20 in force at the time was 1.7 × 10^{-12} µCi/ml air for continuous exposure, or 2.5 µg/m^3, which presumably was the “public” MAC and not the occupational MAC. This would help explain the somewhat elevated weighted average concentrations even for workers who did not enter production areas.

Based on the information in the references cited in this and previous sections, Table 18 was set up to help dose reconstructors interpret claimant submissions and the Mallinckrodt records. Table 18 lists job titles obtained from AEC and Mallinckrodt reports and from film badge and urinalysis records; in addition, it gives a geometry factor set appropriate for each job title (for later use as described in Section 7).

Table 18 is to be used with Tables 19 through 22 and later tables to help determine the exposure to an individual worker when bioassay data for the worker is missing or is conflicting and when comparable worker bioassay data (see Section 6) is insufficient. Tables 19 through 22 were derived from condensing Tables 12 through 16 and from other sources; the data they contain thus represents average daily weighted air concentrations in nearly all cases. Additional data for individual sampling and laboratory operations is given in Section 5.2.4. All of the data in the tables mentioned are based on a natural uranium mixture, with the exception of thorium processing, as discussed below.

The process in which AM-7 residue was converted into a concentrated thorium nitrate solution, as explained in Section 4.7, is a special case. No information was found regarding the particle size of this residue, its tendency to be aerosolized, etc. However, the digestion and extraction process appears to be similar to the basic uranium ore digestion and extraction process and so it presents no novel operational or processing features. The processing appears to have been entirely of liquids (AEC 1955c). Besides the AEC-measured data given in Table 16, there is some data from the Mound end of the processing (Mound 1956): the maximum and average air concentrations in the ionium (Th-230) “high-risk” part of the Mound processing area were 48.1 x10^{-10} and 16.3 × 10^{-10} µCi/cm^3 respectively. In the exhaust line of the hood in which the work was done the maximum and average concentrations were 384.9 × 10^{-10} and 38.8 × 10^{-10} µCi/cm^3 respectively. It is not known whether the work at Mallinckrodt was done in a hood and since the nature of the Mound work was further acid digestion and extraction (DOE 2002), the Mallinckrodt exposures were potentially higher.

Also, it must be noted that the concentrations reported by Mound (1956) were of ionium, i.e., Th-230, not of total thorium. Thus the associated source term must include the Th-232 known to have been in the solution as well. From Section 5.2.3, there was 11.6% Th-230 weight in the original residue, hence the Th-230/Th-232 ratio will be assumed to be 0.116/0.884, or 0.131. This gives a Th-232 source to be added to the Th-230 source term (see Table 6). Although the internal dose per curie intake is generally somewhat higher for Th-232 than that for Th-230, the specific activity (Ci per g) of Th-230 is over 10^5 times that of Th-232. Thus the Th-232 contribution to the dose is likely to be negligible.

5.3.3 Respirator Use

An undated MED reference that is assumed to be of 1942-1944 vintage (MED undated a) stated that respirator use was mandatory when the process required; MED (1942) and MED (undated a) stated that this was especially necessary for the UO_3 and ore grinding and sifting operations respectively.

An MED reference from May 1945 shows layouts indicating the areas of required respirator wear (MED 1945n). A Mallinckrodt manager stated in 1946 (MCW 1946g) that although dust levels had been high, exposures were being reduced not only by improving the dust systems but also by having workers keep their respirators on all the time that they were working in high-dust areas (thus implying that this may not always have been the practice). An AEC inspector noted in 1947 that although respirators were supposed to be worn in high-dust areas such as the ore grinding and orange production area, they were seen to be worn around the neck or even not at all in areas immediately adjacent to the cited areas; these adjacent areas were visibly covered with finely divided particles that could clearly be made airborne when swept off the floors and walls by drafts (AEC 1947a). Mason (1958a) stated that MED and Mallinckrodt agreed in the early years of the work that production would proceed on a priority basis, with the understanding that in high dust areas extensive use of respirators would need to be made. Thus during 1946-1948, respirators were used for “practically all” plant operations. Mason (1958a) commented that the exposures received depended partly on the effectiveness of the respirator program (at that time).

Mallinckrodt’s policy in the later years was that routine respirator use was not acceptable practice and that they were a temporary expedient for unusual conditions only (MCW 1955d). The requirements for them were spelled out in standard operating procedures (MCW 1955d). Even so, AEC inspectors noted instances where visible dust clouds were present or they measured clearly significant dust levels, yet the operators were not wearing respirators ((AEC 1954b; AEC 1954c; AEC 1954e; AEC 1954f; AEC 1954g; AEC 1955e).

A Mallinckrodt official observed in 1958 that in the plants they tried to keep below the MAC, but that if the concentration were greater than 1 MAC for a specific operation, they would not necessarily require the operator to wear a respirator (Utnage 1958c). He explained that this would depend on the worker’s integrated exposure, taking all operations into consideration, and that if some short-term high alpha concentrations in the air were found, they made it a practice to have personnel wear respirators temporarily until the situation was corrected. He concluded that Mallinckrodt did not subscribe to the use of respirators as standard control equipment. Breslin (1958) stated that the time-weighted average exposures measured by AEC-NYOO did not include corrections for respirator use and so should be viewed as potential exposure; however, he also asserted that in very few cases would these be substantial overestimates “as the use of respirators was inadequate and spotty”. This is borne out by the various AEC dust studies that, as noted above, pointed out cases of significantly elevated dust levels where respirators were not being worn.

MED (1946b) gave acceptable types of respirators for use in uranium processing work, as follows. For use in U_3O_8 dust, the MSA “Comfo” 2101 with the All Dust 2133 filter was acceptable; for use in UO_2 dust, the MSA “Comfo” 2101 with the Dust & Mist 2130 filter, the MSA Dustfoe 2147 with the Dustfoe 2138 filter, or the Willson 770 2119 with the Fume 2123 filter; and for use in UF_4 dust, the Duper 46 2111 with the 2124 filter or the MSA “Comfo” metal frame 2101 with the Dust & Mist 2139 filter. A 1955 list of Plant 6 process cells, dust collectors, and tanks, together with the then-current uses of the tanks, appears in MCW (MCW 1955g); maximum emergency nonsurvey stay times (i.e., for urgent access and not inspections) and the general respiratory protection (mask) types are also given in this reference. It appears that the stay times are based on external dose rate, not on inhalation considerations.

In the absence of any firm figures on respirator use and efficacy at Mallinckrodt, it will be assumed that respirators were not reliably used.

5.3.4 Radon

As noted earlier, radon levels could be significantly elevated in enclosed areas where material containing uranium daughter products was stored. ORAU (1989a) stated that a short burst exposure was more typical than a sustained exposure and this appears to be borne out by contemporary descriptions of the various operations in which radon might be released.

Radon measurements by Mallinckrodt and AEC were reported in units of 10^{-10} Ci/L. It is clear from MCW (MCW 1955k) (which, despite its lack of company heading and date, is assumed to be a Mallinckrodt procedure circa 1955) that for both the "environmental" radon measurements and the breath radon measurements, it was just the radon and not the radon daughters in the air that was counted. That is, the procedure specifies that the air sample be alpha-counted for 14 hours, i.e., allowing the radon to come to equilibrium with its daughters Po-218 and Po-214, and that the first two hours of counting are to be discarded, i.e., discarding the contribution of the radon daughters already in the air at the time of sampling. The data from 2 to 14 hours is then extrapolated back to zero. As also stated by MCW (MCW 1955k), the chamber standarization value is given as a theoretical 266 counts per hour per pCi of "radon in equilibrium with its daughters", with half the counts due to the radon and the other half due to the two daughters. The radon decays are assigned a counting efficiency of 100% in the ionization chamber, but the daughters are assigned an efficiency of only 50% because they deposit on the walls of the chamber. Thus in that laboratory and with that type of chamber, the count rate was actually 220 counts per hour per pCi of radon in equilibrium with its daughters, for a effective total efficiency of 83%. However, because the net counts per hour is divided by the chamber standardization value, the result is in pCi of radon alone, not radon and its daughters.

Operations involving radon potential were examined by the Mallinckrodt health physicist and supervision to determine if there was any step in which transient exposure to flood (sudden high) radon concentrations were a possibility (MCW 1950e). From perhaps the mid-1940's on, radon samples were taken once a week in high-radon areas (MCW 1950e). This included the outdoor yard areas, from which measurements Mallinckrodt concluded that some outdoor area was always downwind and that the Plant 6 outdoor background (based on "ordinary" emissions to the outdoor plant atmosphere) was on the order of 1 x 10^{-11} Ci/l. (MCW 1949n). AEC (1949g) stated that Mallinckrodt had constructed several radon measuring devices for its own use and were in fact in the process of constructing one for NYOO. The standard used by Mallinckrodt was made up by the National Bureau of Standards (AEC 1949l). AEC (1949g) also reported that the author of the reference, an AEC inspector, had visited the airport (SLAPS) with Mallinckrodt health personnel and had assisted them in taking radon samples by using an extension pole device designed by Mallinckrodt; samples could thus be taken at levels as high as 20 feet above the ground. It seems clear that based on (1) the long history of radon sample taking at Mallinckrodt, with the start preceding by some years the processing of high-radium and thus high-radon pitchblende ores, and (2) the apparent competence of the Mallinckrodt health and safety and instrument people at developing and using radon-measuring instruments, the radon measurements taken can be regarded as reasonably representative of the conditions.

Some early radon measurements from 1945 are shown in Table 23. The principal locations where difficulty was found in 1946 in keeping radon concentrations below tolerance levels were the railcars during unloading of ore drums and in the ore and residue storage area (MED 1946c).

MED (1946c) noted that a railcar could be unloaded fast enough that the then-current ventilation methods and restrictions spent on time in the railcars were usually sufficient to keep below tolerance levels. Still, the radon problem became more acute with the heavy use of pitchblende ore and in 1948 AEC provided or arranged for new railcars to be used that could accommodate installation and deinstallation of fans at the ends of the cars at the delivery points (MCW 1948h). Presumably the openings were covered with panels while the railcar was in transit. In October 1948, Mallinckrodt tested the efficiency of the ventilation in one of these new railcars (MCW 1948h). The door was left open two feet overnight, then closed for three hours until the start of the test. The fans were started and at subsequent intervals the radon level was measured at two points at the center of the car halfway between the door and either end. The result was that the radon level fell from an average of 7.03 x 10^{-10} Ci/L at time zero to about 0.5 x 10^{-10} Ci/L after about 3.5 hours, reaching the 1.0 x 10^{-10} Ci/L point some time around 20 minutes, although the levels fluctuated at and between points. While the ventilation was seen to be effective, two drawbacks were noted. First, the gamma dose rate at the top and side of the car was 20-32 mR/ hr and it took two or three workers about 1.5 hours to install the fans, so that the workers who installed the fans received 30-50 mR, according to pocket chamber readings. Second, because the total time to unload a car increased to more than two hours with the addition of the fan installation step, the car could be on the siding for an extra day before being unloaded and thus people passing by and working near the car received extra exposure. Hence Mallinckrodt recommended that the fans be left permanently installed in the railcars (MCW 1948h). It is not known if AEC allowed this.

The problem was more complicated in the storage area because worker occupancy times were of necessity longer. There was no forced-draft ventilation in the ore storage area at that time, only a natural exchange of air provided mostly by a four-foot opening on either end of the building. The result was that radon samples likely averaged on the order of the maximum permissible concentration (1 x 10^{-10} Ci/L) or higher. These openings appear to have been permanent, since AEC (1947e) states that the (each) whole end of the building could be removed, but if not, a large opening still remained.

In the Scalehouse Sampling Room, samples of K-65 residue were taken from open K-65 drums sitting in the sampling bays on the level below the Sampling Room (MCW 1948j). This was done with a thief sampler through holes in the Sampling Room floor. Although there were several small exhaust fans that were supposed to draw the radon away from the sampling workers, the turbulence caused by a unit heater resulted in the radon being distributed throughout the room. Also, although most of the time the exhaust fans maintained adequate control over the open pans into which the drawn samples were deposited, at times the turbulence also drew radon from the thief and the pans as the sample was being dumped out. The concentrations found in the Scalehouse itself were very erratic, showing that here too the radon was being carried in momentary eddy currents about the room. High concentrations usually coincided with the pulling out of the drums on pallets from the storage bins and the removal of the drum lids. It was noted that a worker could be present in the Scalehouse and the Sampling Room for up to 4 hours on some days.

Besides the ore and residue handling and storage areas, some processing equipment was also associated with elevated radon levels. In August 1948, a ventilating fan was installed in the wall of the K-65 centrifuge room to exhaust thermally hot air (MCW 1948l). However, this created a suction from the neighboring Wash Oliver (filter press) room into the centrifuge room, drawing appreciable quantities of radon off the Wash Oliver filters and into the centrifuge area. To solve this problem, a door was installed between the two rooms. Prior to the installation of the fan, the overall average radon concentration is the centrifuge room was 0.83 x 10^{-10} Ci/L; after the installation of the fan, it was 1.25 x 10^{-10} Ci/L; and after the installation of the door it was 0.62 x 10^{-10} Ci/L. Further ventilation improvements were apparently made in the centrifuge area.

A significant short-term exposure to radon would occur when the Feinc Niagara filter press, which was used to produce the high-radium K-65 residue, was opened for the washing of the plates (MCW 1950b). This occurred about 12 times a day when the processing was "operating on digest" because the plates could not effectively be washed down by remote control in this regime. It was necessary for a worker to open the press and hose down the cake. Approximately once a week, the plates became "blinded" (clogged) and had to be manually removed for cleaning with caustic (MCW 1950b). When the Niagara was opened, the radon concentration in the near area would suddenly go up to as high as 34 times tolerance, then went back to tolerance level over about 5 minutes. As the last of the cake was washed into Tank M-289, the radon concentration would double again because of the venting of M-289 as the pipe cleared and air from the tank was displaced back to the open Niagara and thus to the room. The concentration fell back quickly to nondetectable levels when the line to M-289 was closed. Radon levels at the small tank where the plates were cleaned, however, showed low or nondetectable levels.

Radon concentration in enclosed spaces in MED/AEC refineries produced levels of up to 10^{-8} to 10^{-7} Ci/L, which were eventually significantly lowered by ventilation improvements (Eisenbud 1975). After these improvements were made at Mallinckrodt in about early 1949, all Mallinckrodt operating areas in Plant 6 were found by AEC to be below the preferred level of 10^{-10} Ci/L, with the exception of the K-65 return (drying) oven, the ore thaw house, and the Wash Oliver cell, as shown by some of the results in Tables 24 and 25 which cover the years 1947-1949 and 1950-1957 respectively. However, Mallinckrodt (MCW 1949m) reported the results of measurements taken in the Ore Room and its addition soon after ventilation and remote handling equipment was installed; some of these results are shown in Table 24. The improvements significantly reduced radon levels in most areas, but there were still some operational problems (see Section 5.2.1). Because as MCW (MCW 1949m) states the radon hazard was greatest near newly opened drums and because the ventilation system apparently was not very effective at taking care of this hazard for close approaches to the drums (i.e., the radon die-off to less than 1×10^{-10} Ci/L after a drum was opened took 25 minutes), radon was a continuing concern in the ore drum storage and handling areas. The operational problems that forced operators to approach open drums appear to have been corrected thereafter.

Most of the information regarding radon levels from 1947 on comes from the weekly "MZ" radon reports (Mz was a code for radon) (MCW 1955d; MCW 1947-1957). Additional data are shown in the table below (from AEC 1949b, Table 1), which gives typical radon exposures for different areas at the Middlesex facility over the years 1944-1949 (covering the early years of pitchblende use). These would be comparable to the railcar unloading and Ore Room operations at Mallinckrodt.

Middlesex ore storage worker radon exposures, January 1944–January 1949.

Area	Level		
	Min	Avg	Max
Railcar unloading	200		250
Drying room	1	6.7	21.7
Storage area	0.1	0.8	2.9
Sampler rooms	0.1	0.2	0.5
Crushing area	0.1	0.2	0.9
Crusher pit	0.3	0.8	1.3
Sampling lab	<0.1	<0.1	<0.1
Weigher's booth	<0.1	<0.1	<0.1

Radon levels are given in multiples of the radon MPC or preferred level of 10^{-10} Ci/L. Use of blower-equipped railcars starting in about early 1949 reduced levels to below the MPC.

Tables 24 and 25 also give information that has been found about the radon levels in the plants other than Plant 6. However, because in the plant with the greatest exposure potential (i.e., Plant 6) and in SLAPS the levels were lower than tolerance (1×10^{-10} Ci/L) in all but very enclosed areas and because the thorium and radium were removed in the production of UO_3, it can be assumed that the radon levels were well below the tolerance level in areas where ore and pre-UO_3 residues were not present.

A further item regarding radon content of the residues was noted by AEC (1947e). A certain percentage of the radon produced during decay would be adsorbed on the surface of the ore particles (the percentage depending on the particle size of the ore, the temperature, the moisture level in the ore, etc.). When the ore was digested in acid, the adsorbed radon was released and the gamma activity of the ore would drop. But when the radium was precipitated as a sludge, as was done at Mallinckrodt, the precipitate would adsorb some of the radon and there would thus be an increase in gamma activity as the precipitate "aged". This meant that the radon content of the digest vent exhaust, the residue, etc., could be variable depending on process conditions.

Regarding the processing of residues to concentrate thorium, it is clear from the data in Table 26 that the average concentration of radon (Rn-220 and Rn-222), under process-applicable and claimant-favorable assumptions, was about eight times the AEC "preferred level" of 10^{-10} Ci/L. It was also well above the 3.0 $\times10^{-12}$ Ci/L that represented an "undetectable" amount for the AEC during the relevant period of time (as per Eisenbud 1975).

ORAU (1989a) estimated the radon exposures of approximately 184 Mallinckrodt Plant 6 workers as part of a larger study. They calculated the working level (WL) for each job title using the mean radon concentration value from all available surveys in 1946-1957 for each job title's assigned work area; exposure estimates were then calculated in units of working level months (WLMs) for each job title. It was assumed that an equilibrium factor of 0.25 applied for radium and its daughters, based on plant and memoranda, other documents, and findings from uranium mine and residential radon studies. Their approach of assigning a WLM value for each job title for a single grouped period of years (rather than breaking it down by years) was due to the lack of survey data for many of the years. The "roving operator" and the production/processing/manufacturing operators were assigned the average area radon levels for the entire Plant 6 area and the highest daily work time breakdowns of all the production processes. Weekly rotation as practiced at Mallinckrodt was handled by assigning an average radon level of all available survey data from those work areas in the rotation plan for that job title. Pertinent results are shown below for information. These results are for the years of pitchblende processing and so would presumably be conservative for the pre-pitchblende years.

Exposure rates of 184 Mallinckrodt workers, based on surveys done in 1946-1957 (ORAU 1989a).

Job title	No. of workers	WLM per month worked
Manufacturing operator	3	.138
Operator, process develop	1	.138
Production operator	2	.138
Roving operator	3	.138
Cleanup operator	8	.127
Maintenance (general)	1	.111
K-65/GLC sampler	1	.102
Pot Room operator	14	.097
K-65/AJ-4 sampler	4	.096
Raffinate operator	9	.092
Barium operator	6	.091
C-3 (Centrifuge) operator	18	.084
Feinc operator	12	.084
Director, technical	1	.077
Mail boy (office)	2	.057
Porter, Production	6	.056
Ore Room operator	16	.055

Job title	No. of workers	WLM per month worked
Recovery operator	13	.052
Guard	3	.047
Porter, Lab	1	.044
Digest operator	2	.042
Pilot Plant operator	4	.041
Clerk, Manufacturing Office	1	.040
Clerk, Production Office	8	.040
Feed operator	2	.038
Filter press operator	3	.038
Plant monitor	1	.030
Health office	2	.020
Engineer, senior mechanical	1	.018
Furnace operator	12	.010
Industrial relations (office)	1	.010
Ether House operator	17	.008
Machinist (instrument)	2	.008
Chemist	4	.003

Note that for use in dose reconstruction, radon concentrations reported in units of Ci/L must be converted to units of working level months (WLM) before they can be used in calculations. One working level (WL) is the total amount of energy given off over a long period of time by the short-lived radon-222 daughters in equilibrium with 100 pCi (10^{-10} Ci) of radon, taken to be in one liter of air. Since the daughters will typically not be in equilibrium with the radon if the ventilation is good, this conversion is not simple, but depends on the ventilation conditions. The reported results are given in Tables 23-25 of this technical basis document in the original units so that the dose reconstructor can readily use whatever equilibrium factor appears to be most appropriate if the factors used in the conversions to annual intakes in this technical basis document (see Section 5.3.8) need to be re-examined for whatever reason.

5.3.5 Surface Contamination

Although surface contamination levels per se are not indicative of airborne contamination levels or external dose rate, they can suggest whether or not a potential for exposure exists. Little surface contamination measurement data appears to have survived, presumably because it could vary so much and because excessive levels, when found, were cleaned up but not necessarily documented.

Mallinckrodt (MCW 1958; MCW 1959) gave the results of a plant-wide surface contamination and external dose rate study that Mallinckrodt did after some preliminary post-operation decontamination of the site. These reports indicate that ground areas adjacent to the production plants were heavily contaminated, with average surface alpha activity of 2500 dpm/100 cm^2 and average beta activity of 2 mrep/hr; high spots of 35,000 dpm/ 100 cm^2 alpha and 15 mrep/hr beta were not unusual. High spots of up to 20 mR/hr gamma were found west of the UF_4 loading dock where ore cars where cleaned prior to 1950. In Plants 6 and 7, the administration building (Building 112) (except for the storeroom and maintenance shop), the Boiler House (115), the Service Building (117) (except for the laundry and the regulated locker room), and the Magnesium Building (708) were not significantly contaminated. The average alpha activity on floors in the excepted areas was 3000 dpm/100 cm^2 and the beta-gamma activity was generally at background levels. In Plant 4, beta activity measured at contact with surfaces in production areas averaged between 10 and 50 mrep/hr with the overall average activity measured at contact with surfaces estimated at a level of 25 mrep/hr; occasional high spots were found up to 80 mrep/hr; activity measured at the three-foot level in the center of production areas ranged from 1 to 5 mrep/hr. Gamma activity measured at contact with surfaces was 0.02-0.9 mrep/hr, with the highest readings being in some yard areas and the slag processing area. The overall plant average measured at the three-foot level in the center of production areas was 0.07-0.1 mR/hr.

Data from this Mallinckrodt study (i.e., the study partly documented in MCW 1958 and MCW 1959, quoted by Utnage (1958a)), also showed fixed floor surface contamination levels of greater than 10,000 alpha dpm/100 cm^2 at various floor locations in the ore sampling area after vacuuming; it was highest at the hopper loading and weighing stations. In the metal reduction area (Plant 6E), the floors of the crucible disassembly areas and the saw areas measured 2200-3300 dpm/100 cm^2 and the center of the maintenance cage (where there was no uranium processing) measured 1200 dpm/100 cm^2. Other stations in the “pitchblende (ore) area” (Plant 6) showed levels of 1000-60,000 dpm/100 cm^2; in the uranium products warehouse, 4500-21,500 dpm/100 cm^2; and in the metal plant (6E), 1000-9000 dpm/100 cm^2. The UF_4 production plant (Plant 7) had no reading higher than 1200 dpm/100 cm^2. Since these figures represent fixed contamination, loose contamination removed by the vacuuming could have been present at far higher levels (Brobst 1958). Thus these figures, while suggestive of where the worst areas were, cannot be considered to be representative of operational total surface contamination levels.

Although as noted above resuspension is assumed to be included in the measured airborne concentrations, some available information about the relationship between loose surface uranium-bearing contamination and airborne concentrations will be summarized here in case it becomes relevant in individual cases. A study was done at the Oak Ridge Gaseous Diffusion Plant to ascertain the relationship between alpha airborne activity and alpha surface contamination (Becher 1958). The air samples were measured with Whatman #41 filter discs and fixed counting equipment, with an overall counting and collection efficiency of 30%. The surface transferable activity was measured with a Samson alpha survey meter, with an overall counting and efficiency factor of 20%. The data, shown in the table below, indicate that the airborne concentrations ranged from 0.36 to 5.05 dpm/m^3 for every dpm/cm^2 of surface contamination, or an air concentration of 3.6 to 50.5 dpm/m^3 for every 1,000 dpm/100 cm^2 of surface contamination.

Ratio of alpha airborne activity to surface contamination (Becher 1958, Table 2).

Source of Data	Ratio*
Two operating areas having the highest contamination levels (1953)	
Shift-length air samples	0.64
Spot air samples	1.9
Plant-wide operations (415 surveys over 9 months in 1958)	
Shift-length air samples	0.36
Spot air samples	5.05
Special test conditions (1953)	
Simulated conditions	13
Worst possible conditions (short periods)	20

* Units of the ratio are dpm/m^3 of air per dpm/cm^2 of surface

Other fixed surface contamination data are given below for various surfaces and pieces of equipment in the laundry (from Table 1 of Utnage 1958b).

Contamination levels on various surfaces and pieces of equipment in the laundry.

Unit	Beta + Gamma, cpm
Presser pad surface	2,000
Cloth hamper surface	5,000
Wooden hamper surface	1,000
Work tables and surfaces	200-1000
Lint ball/dryer lint trap/roof lint trap	4,000/2,000/3,000
Inside washer	300
Inside dryer	1,000-4,000
Under washer	20,000-60,000
Under dryer	100-40,000
Floors, average	300-500
Walls	100-300
Overhead pipes, etc.	300-1000

With surface contamination there is the potential for two modes of exposure other than inhalation. These are ingestion and skin doses from contamination on skin and clothing. Ingestion would most likely take place during eating or smoking breaks. No information is available as to the likelihood of ingestion during eating or the quantities that might have been taken in. However, some information is available on recommended AEC limits on hand contamination and on ingestion as a result of smoking.

A 1947 AEC letter to Mallinckrodt (AEC 1947b) stated that surfaces in the lunchroom should show less than 100 dpm alpha on a smear test and less than 500 dpm alpha per 100 sq cm (fixed plus removable); the maximum hand count should be equivalent to no more than 0.35 mg of uranium on either side of the hand. In 1949, dust samples taken in the lunchroom at various times over one lunch period, showed that the level ranged from 6 dpm per cubic meter with only 7 workers present to 57 dpm per cubic meter when the room was full; the level was 12 dpm per cubic meter at the start of the post-lunch cleaning and then went up to as high as 65 dpm per cubic meter following the cleaning due to dust suspended in the air (MCW 1949d). The same sampling study showed that levels in a conference room in association with a service group (operators') meeting were 4, 22, and 30 dpm per cubic meter respectively before, during, and just after the meeting. In both the lunchroom and conference room, the workers wore cover clothing over their work clothing. In 1951 it was reported that the average dust level in the Plant 6 lunchroom (MCW 1951b) was 1.3 dpm/m^3. Thus although it was stated in MCW (1949d) that the sub-tolerance levels reported were acceptable, clearly the lunchroom levels were reduced over time by better clothing change and contamination control practices.

Smoking areas were typically adjacent to the working areas. In 1951, in response to AEC's pointing out that Plant 6 workers entering the smoking room habitually slapped their dusty gloves and brushing material from their clothing after they entered the smoking room, Mallinckrodt agreed to provide improved ventilation in the smoking room and to put a small vacuum cleaner inside the smoking room for workers to use to vacuum their gloves and clothing after they entered but before they smoked (MCW 1951d). Mallinckrodt also agreed to have greater enforcement by supervisors of the requirement that gloves be removed before smoking (MCW 1951d).

These findings may explain the observation by AEC (1951a) that in a majority of the jobs evaluated at Mallinckrodt, the contribution to dust levels from time spent in the smoking (cigarette break) areas was greater than the contribution of dust from the time spent in the operations area. In 1951 it was reported by Mallinckrodt that air samples taken in the smoking room in Plant 6 were always above tolerance, even after UO_2 production had moved to Plant 7 (MCW 1951b); the average dust level was 116 dpm/m^3, versus 1.3, 4.2, and 66 for the lunchroom, locker room, and change room respectively, and the typical daily individual occupancy of the smoking room was 60 minutes. This all suggests that the contribution to the ingestion of radioactivity by a smoker might be significant, although AEC apparently did not do any studies of this at the time.

Tests done in about 1958 at the Oak Ridge Gaseous Diffusion Plant with UNO_4, UF_2, and UF_4 (Bailey 1958) indicated that when loose uranium-bearing material was placed on the palmar surfaces of the hands, the palmar transfer of uranium from the hands to the cigarette amounted to about 1% of the material on the hands and that inhalation of the material during smoking amounted to less than 1%. They also tested absorption by placing material on the backs of the hands. They concluded that a maximum of 2.5×10^5 dpm on the palm of each hand, for a 20-cigarette-a-day smoker, would be allowable for him not to exceed the maximum permissible inhalation of uranium by cigarettes alone; that a maximum of 2.5×10^5 dpm on each hand would correspond to the maximum permissible ingestion; and that a maximum of 2.5×10^5 dpm on the back of each hand would correspond to the maximum permissible absorption dose. Taking the reciprocal of the sum of the reciprocals, they obtained a maximum of 8.6×10^4 dpm per hand for the total considering all three routes of exposure. They assumed a 15% geometry factor for the hands (based on their instruments) and concluded that the total limiting level was 13,000 cpm per hand.

The allowable amounts given in Bailey (1958) corresponded presumably to National Bureau of Standards Handbook 52 allowable intakes, since they cited Handbook 52 although they did not quote the figures. The allowable amount was assumed to be on the hands continuously for 5 days per week, 8 hours per day. This level or greater is likely to have been on workers' hands at times but not to have been on the hands constantly. Thus while a potentially significant contribution to the dose from hand contamination cannot be ruled out, it seems unlikely that most smokers would have had a sustained level of contamination of this magnitude on the hand during breaks, especially since they were likely to have worn gloves during most of the processes (due to heat, acidic content, etc.). This suggests that ingestion is a negligible source of internal dose during operations, compared to inhalation.

Regarding clothing, Table 27 shows contamination levels and some associated dose rates from clothing (Utnage 1958b). The contamination measurements in cpm were taken with a Thyac beta-gamma meter with thin-wall tube, while the measurements in mrep/hr were taken with an unspecified air ionization chamber. At the time the measurements were taken in 1957, the laundry had been in operation for almost ten years and had never been decontaminated; there were nine laundry workers; and the laundry processed 25,000 coveralls and 25,000 "soft" items (handkerchiefs, socks, and underwear) each month. Clothing used in contaminated or potentially contaminated areas ("regulated areas") was kept separate from clothing used in non-regulated areas. However, regulated-area clothing was worn interchangeably by anybody, so that "a uniform contamination level [was] eventually obtained" (Utnage 1958b). Gloves were discarded when soiled and were treated as contaminated clothing (MCW 1950e).

At the Oak Ridge Gaseous Diffusion Plant (ORGDP), test measurements in 1957 on clothing showed that the highest spot reading was typically about 3.5 times the average reading (Becher 1958); this is probably roughly applicable to Mallinckrodt as well, although the uranium compounds at ORGDP were mostly soluble whereas the Mallinckrodt compounds were mostly insoluble. The ORGDP tests also showed 1620 alpha dpm/cm^2 to be equivalent to 9700 cpm/100 cm^2 as measured on a Samson alpha meter, giving an "efficiency-geometry" factor of 6%. (Note that for the "surface transferable activity" on filter paper used for air sampling also reported in Becher 1958, an efficiency-geometry factor of 20% was assumed for the Samson alpha meter.) Finally, the ORGDP measurements showed that about half the uranium applied to the clothing at the beginning of the test had dropped off within the first two hours of wearing. This suggests that uranium that gets on clothing can come back off it readily and that surface contamination on clothing can contribute to airborne levels via resuspension.

Railcar interiors were invariably found to be contaminated above 2500 dpm/100 cm^2 after unloading uranium oxide, UF_4, or uranium metal at uranium processing plants, even though the sites made an effort to decontaminate them (AEC 1949b). AEC advised that strippable coatings would eventually need to be used (AEC 1949b), but there was no evidence that this was ever done. This suggests that even where closed containers of uranium-bearing materials were being unloaded, it must be assumed that surface contamination was typically present.

Monitoring for surface contamination was done using portable alpha and gamma-beta survey meters and smear tests (MCW 1950e). Tools to be moved from contaminated areas were to be cleaned to a stated level (MCW 1950e), given in 1947 as 100 cpm, presumably beta-gamma (AEC 1947b). If the article could not be decontaminated, it was to be securely wrapped in paper or placed in a clean container and marked with handling instructions. However, these checking and control requirements did not apply to tools belonging to outside contractors, who presumably could take away tools contaminated to any level (MCW 1950e). Alpha hand counting devices were not used after some time prior to 1950, supposedly because AEC-NYOO requested that their used be discontinued (MCW 1950e). As of at least 1950, hands and feet were not checked when workers exited a process area to go to the lunchroom to eat or smoke (MCW 1950e; AEC 1947a), but they had to go through the personnel decontamination cleanup in the change room area, with a strict "clean side, dirty side" regime maintained (MCW 1950e) (see Section 5.5.3). Visitors were not checked for contamination before leaving the plant, at least through about 1950 (MCW 1950e).

5.3.6 Information and Available Data Regarding Urinalyses

Mallinckrodt uranium processing workers were given a pre-employment physical that included an initial urinalysis and a blood count (MED 1942) and they were given an annual physical that included a routine urinalysis and a blood count (MED 1942 (which says it was a "followup" examination); MCW 1955d; Mason 1958a). From about the summer of 1948 on (MCW 1950c), this included a measurement of uranium in the urine. In addition, up to March 1954 some worker classifications had more frequent urinalyses, either every 4 months or every 6 months depending on the worker classification (MCW 1955d; Mason 1958a); after this, time the frequency was no more than semiannual (MCW 1955d). As urinalysis records indicate, some office workers appear to have been given urinalyses, but it is not clear whether this was done on a regular basis.

The Mallinckrodt urinalyses were first performed by the University of Rochester under contract to AEC-NYOO up to about 1948. At one point, according to ORAU (1983b), there were some problems with the running of standards because there had been uranium plateout on a table. This was corrected. In 1948, Barnes Hospital began to do the urinalyses for Mallinckrodt. However, an AEC health official stated (AEC 1948) that while it had been his understanding that the analyses were being done at Barnes Hospital (at Washington University), it turned out that they were being done in laboratories at the Mallinckrodt St. Louis site. This came to light when it was discovered, apparently in late 1947, that some urinalysis samples were contaminated due to contamination in the laboratory. An undated, untitled urinalysis listing found in dose reconstruction project files indicates that closed, blank samples were found to have significant levels of uranium in them, indicating contamination in the laboratory; it was suggested that this might explain the high levels of some of the non-blank (worker) samples. Thus at least the early urinalysis samples must be considered to have been potentially contaminated (i.e., some of the uranium content may have come from the laboratory analyzing them).

Apparently Barnes Hospital resumed doing the urinalyses (AEC 1949g, MCW 1950e). However, in 1949, AEC compared the Mallinckrodt analyses against those for other sites handling similar material and concluded that the results were consistently inexplicably high (MCW 1950e), although Mallinckrodt thought the agreement was acceptable (MCW 1949j). They then sent Mallinckrodt some spiked samples and also had an independent Mallinckrodt chemist prepare a stock solution of known concentration. The spiked samples and samples of the stock solution were sent to Barnes as regular samples, while Barnes standards and samples of the stock solution were sent to NYOO for analysis. AEC also compared Barnes' methods and equipment to standard ones. The conclusion was that the samples were indeed reading too high at Barnes. (It is not clear if the spiked sample comparison referred to in MCW (1949j) is this same set of samples, but if so, the data can be found in MCW (1949j), along with a statement that the reliability of the Mallinckrodt sample testing is +/- 15%). Subsequent data analysis showed a gradual precipitation of uranium in the Barnes standards, which meant that the daily standard curves showed a gradual loss of slope over time, up to 30%. Also, Mallinckrodt had been called three times over the previous year to service the Barnes instrument because of sensitivity loss, when the problem was actually the standard. The maximum error in the urinalyses over the preceding 14 months was estimated to be +89%.

AEC technical personnel thought that the affected data were of doubtful value (AEC 1950l). Still, AEC (1950i) asked Mallinckrodt if the urinalysis data could be salvaged, i.e., if there was a consistent factor that could be applied to all of the subject urinalyses; MCW (1950q) thought that there was not. AEC also recommended that a note regarding the problem should be inserted into the medical files of the affected individuals (AEC 1950b), presumably to aid in the interpretation of the results. It is not clear whether this was done. AEC-NYOO stated that it was not possible for them to take over the urinalyses again since the number of samples to be analyzed was too high for their capacity (AEC 1950l).

It is not clear who did the urinalyses from 1950 to 1954, although MCW (1950m) suggests that this was no longer being done at Barnes Hospital but at AEC-NYOO, presumably at HASL. Also, in September 1951 AEC-NYOO was clearly performing them, but directed AEC St. Louis area representative not to have any more routine samples sent during the last quarter of 1951 because of an “unusual work load” that its chemical laboratory had (AEC 1951c); emergency samples could still be sent, however. In 1954 AEC gave Mallinckrodt permission to perform their own urinalyses (MCW 1954e, ORAU 1983b), presumably in the laboratories at the Mallinckrodt St. Louis site. From the Mallinckrodt Health Office monthly reports, they were analyzing for “X in urine” (i.e., uranium) and it appears that at times there was a significant backlog of overdue analyses, at least in the early 1950’s. Eventually AEC-NYOO (HASL) must have resumed performing the analyses, because a 1955 Mallinckrodt report describing its health program stated that NYOO was doing so (MCW 1955d). This report stated that NYOO was analyzing about 2500 Mallinckrodt urine samples a year and that urine samples that were taken were split, with half going to AEC-NYOO for the radiological analysis and half to Barnes Hospital for the medical analysis.

Referring to urine samples sent to the University of Rochester from any AEC site, ORAU (1983b) stated that samples were collected from workers in four-ounce glass bottles with bakelite caps and shipped to HASL. After 1949, only samples taken within one hour of the start of the weekly shift were accepted (ORAU 1983b).

The radiological analysis was apparently only for uranium content (referred to as “X in urine” or “uranium-in-urine”). However, one set of urinalysis records shows for a single individual and for periods from March to May 1956 the notation “Shoot for radium”. Thus it appears that radium in urine could be analyzed as a special case. It is not clear whether the thorium (ionium) processing workers in Plant 7E had special urinalyses for thorium or not. However, this is a possibility, as suggested by Mallinckrodt’s health and safety director’s obtaining a copy of a Los Alamos Scientific Laboratory procedure on urinalysis for plutonium (LASL 1955) and the director’s noting on his copy that he was told that urinalysis for thorium could be done by the same method as in the procedure.

ORAU (1983b) stated that all urinalyses done at the University of Rochester used the fluorometric method; the urine was dried in a platinum dish, then fluxed with either sodium fluoride or a lithium-calcium fluoride mix and counted. Ross et al. (1975) states that for all AEC contractors before 1961, estimates of lung dose were made on the basis of urinalysis and that this was usually done on the basis of electrodeposition and subsequent counting. AEC (1949l) states that Barnes Hospital (at Washington University in St. Louis) used “fluorometric (photoelectric ultraviolet)” methods in performing urinalyses for Mallinckrodt.

ORAU (1983b) states the following about samples analyzed at the University of Rochester. These samples were run in triplicate, with the results usually being with ± 2 µg/L of one another; if this turned out not to be the case, it indicated that there had been poor fusion of the flux and the samples were re-fused and re-run, which usually corrected the discrepancy. The value recorded was the median value of the three. For insoluble uranium, it was considered that 30 µg/L in the urine corresponded to an air concentration of 50 ug/m^3. The analysts felt confident of readings greater than or equal to 5 µg/L. However, if more confidence was desired for lower-level samples (e.g., for special projects), the urine was concentrated either by ion exchange or by extraction, or more aliquots were run.

The uranium fusion photofluorimetry urinalyses performed by the University of Rochester and the AEC NYOO are known similar to those performed at other AEC facilities. Based on the lack of clear information regarding a detection threshold for Mallinckrodt urinalyses, the default detection threshold for uranium urinalysis may be assumed to be 10 µg/L based on a reported sensitivity of 5 to 10 µg/L for uranium fluorimetry urinalysis in the early years, as was reported for other AEC facilities (Wilson 1958).

Because of the questions regarding the validity of the samples, the apparent variations in sample analysis methods, and even who was doing the analyses, the Mallinckrodt urinalysis data should be used with caution, at least when the data were taken by Barnes prior to about 1951. However, it appears that the errors, if any, are in the conservative (high) direction and thus are claimant-favorable.

Urinalysis records appear to be available, but many appear to be handwritten notes on cards. These are found scattered in various dose reconstruction project files. Fortunately, in about the 1970's, the records were entered into a computer data base for research purposes and have been used in that form since then by Oak Ridge Associated Universities and other research groups. The resulting file has more than 40,000 records (i.e., lines, with each line representing one urinalysis). A "stripped" version (ORAU 2003) is also available, with the names and Social Security numbers removed for privacy reasons.

The large stripped urinalysis file (ORAU 2003) was reviewed for comparable or surrogate worker cases that could be used to produce a table of intakes applicable when bioassay data for an individual is missing or spotty. Cases were selected on the basis of their containing a reasonably uninterrupted series of urinalyses and having reasonably clear notations of job title and/or area worked in. The selected cases were then further evaluated and a subset was extracted for each of various categories of identified locations and operations or position titles. These categories are given in Table 28. Where there were more than two applicable cases for a category, the IMBA program was then used with the assumptions of chronic intake and Type M form and with the data from these cases to produce a category-specific distribution and standard deviation for the typical daily intake, as shown in Table 28; otherwise, the actual data (i.e., for one or two cases) was given. Since there were changes in exposure potential at various times due to process improvements, engineering modifications, or the building of new plants, three periods were established for the determination from the cases of the typical daily intake, as given in Table 28.

Table 28 does not include intakes for workers who processed wastes containing thorium (ionium) because there was not enough urinalysis information for such workers and what there was did not always have a clear indication of work with thorium. It appears that there were few such workers: (1) AEC (1955e) reports studying only six workers in their Plant 7E dust exposure study whereas, e.g.,119 were studied in Plant 6E, (2) few worker cases were found in the large stripped urinalysis file where it was clear that the worker did this type of work, and (3) the thorium worker in every case that was found had worked in other areas where an intake of uranium material was possible. It appears that thorium itself in urine was measured at all, rather than uranium. Thus there likely was not any differentiation in the urine analyses between uranium (and its daughters) and thorium. Hence in dose reconstruction, the urinalysis data from early 1955 on for workers who processed thorium wastes should be examined on a case-by-case basis to see if any special evaluation for thorium was done; if not, then dose estimates from air sampling data may have to be made.

5.3.7 Information and Available Data Regarding Other Types of Bioassay

Breath radon measurements began to be made in June 1945 (as shown by, e.g., MED 1945e and MCW 1946g), but only for workers who worked in areas where there was a potential for radium intake.

MCW (MCW 1950l) gives a list of worker types or occupational areas, along with a rating of the need for workers of each type or in each area to have their breath sampled; most of the Manufacturing (process), Stores (warehouse), and Maintenance workers were in the “Definite” category, while most of the Power Plant, Occupancy, Plant Protection (guard), Research, Laboratory, and Laundry workers were in the “Possible” category and most of the Health & Safety, Office & Administration, and Plant 4 workers had most of their people in the “Improbable” category.

The Massachusetts Institute of Technology (MIT) or the National Bureau of Standards (NBS) did the breath radon analyses for MED/AEC until 1951, when HASL obtained an ion chamber of the type needed to perform the analyses (ORAU (1983b). Initially, samples were taken in the infirmary of the MED/AEC site because it was thought that this area was removed from the production areas; later a respirator with bottled air was used to collect samples in order to control for radon in the ambient air (ORAU 1983b). However, the first breath radon measurements were thought to be worthless by MED because the MED expert concluded that the samples were contaminated at the time they were taken (MED 1945e). As a result, MED directed Mallinckrodt to have breath radon samples taken only in a room away from the plant and only when the employee had not worked for at least 48 hours and while he was still wearing his street clothes (MED 1945e).

Breath radon samples were collected by obtaining one-liter samples of exhaled breath after two days of non-exposure, usually on a Monday morning (AEC 1949g; AEC 1950a; MCW 1950g). The samples were measured at the NBS (AEC 1949g) or at HASL (AEC 1950a, MCW 1955d) by an “automatically recording pulse-counting device” (AEC 1950a). If a sample was over the “tolerance level of 1 x 10^{-12} Ci/L, then a recheck (repeat sample) was made immediately (AEC 1949g). The lower limit (of detection) at HASL was 0.1 pCi/L and there was confidence in readings of greater than or equal to 0.5 pCi/L (ORAU 1983b). In 1946, it was reported that of 40 workers tested, the maximum level was right at the tolerance level of 1 x 10^{-11} Ci/L (MCW 1946g).

In early 1950 AEC apparently became concerned about the high background that seemed to be present where the samples were being taken (MCW 1950g). Mallinckrodt agreed to take test samples elsewhere than in their usual testing area and also to take a room air sample in their normal testing room; these samples were then sent to AEC-NYOO. The normal testing room sample showed a radon content of 0.8 x 10^{-12} Ci/L, which AEC judged was a high background for a breath radon sampling area (AEC 1950k). It is known that in 1950 the Mallinckrodt medical department was located adjacent to the change rooms, which enabled workers to take their physicals after a shower without getting dressed (AEC 1950m); if the breath radon samples were also taken there, that could explain the relatively high background radon. In early 1951, Mallinckrodt (MCW 1951g) reported the results of a further test done at the behest of AEC in which they took breath radon samples from the same four workers in the usual location (background of 0.8 pCi/L) and in a lower-background area (background of 0.2 pCi/L). Mallinckrodt admitted that although they had been told earlier by the (National) Bureau of Standards that the test area background did not matter as long as it was on the same order as the sample, AEC had turned out to be correct in its concerns about the background: the differences between samples taken in the lower-background area and the samples taken in the higher-background area differed by almost exactly the difference between the two backgrounds (MCW 1951g). Mallinckrodt apparently corrected this problem by then using the special respirator-testers provided by AEC ((MCW 1951e); they may also have moved the breath radon sampling location to a lower-background area. Note that Srivastava et al., in their study of uranium miners (Srivastava 1986), found that post-shift measurements (i.e., after breathing ambient radon in the workplace) were 2 to 4 times higher than pre-shift measurements, which confirms the confounding influence of background radon.

A baseline (start of employment) breath radon test was not administered; the routine test during operations was thought to be sufficient (MCW 1950w). In 1949, the nominal breath radon sampling frequency was quarterly (AEC 1949g), which seems to have been the typical frequency for eligible workers (ORAU 1983b). In 1955, AEC-NYOO was analyzing about 500 Mallinckrodt samples a year, taken semiannually at a minimum but about quarterly when permitted by AEC-NYOO sample capacity (MCW 1955d). It was not always possible to obtain a termination breath radon sample because some workers left without notice or because the sample had to be taken after 48 hours of nonexposure, i.e., usually on a Monday (MCW 1950w). Mallinckrodt thought that about 80% of terminating employees had termination medical examinations (MCW 1950w), but it is not clear if this was also true of the breath radon analyses for the relevant workers. Breath radon records are available in scattered form in reconstruction project files; possibly all of it is found in the large urinalysis research file mentioned in Section 5.3.6 above.

AEC considered that many of the early breath radon samples likely represented transient as well as fixed burden and that the background level at the point of sampling (which was generally ignored) was likely to have been relatively high; thus the resulting estimates they made of alpha radiation to the bone based on breath radon measurements would typically be higher than was actually the case (AEC 1950a). ORAU (1989b) quoted a report by the MIT Radioactivity Center that stated that their lower limit of detection for breath radon was 0.006 pCi/L and that this corresponded to a minimum detectable body burden of radium-226 of 0.00059 µCi. ORAU (1989b) also quotes the MIT report as stating that assuming that 65% of the radon generated by the body burden of radium was exhaled, 1 pCi/L of radon in the breath corresponded to approximately 0.1 uCi of Ra-226 in the skeleton, which ORAU (1989b) verified in its own calculation. This can be compared to the assumptions made by Srivastava et al. (Srivastava 1986), who took breath samples of uranium miners using a controlled radon intake respirator apparatus: the equilibrium percentages of the radium-226 body burden were taken to be 33% in the lungs, 39% in cortical bone, 14% in trabecular bone, and 14% in other soft tissues and the corresponding radon escape fractions were taken to be 1.0 for lungs and other soft tissues and 0.7 for bone. Srivastava et al. thus concluded that 84% of the radon produced by decay of the Ra-226 was exhaled, a figure applicable to those working in the mines for several years (i.e., to workers chronically exposed for an extended period).

ORAU (!989a) reported the results of a study of ambient radon exposures to some 184 Mallinckrodt workers. In addition to the lung doses from the ambient radon exposures, they attempted to estimate the contribution from breathing radium dust and used breath radon data and the associated skeletal radium burden as an indicator (as per ORAU (1989b). These estimates are given in ORAU (19889a), but as with the ambient radon exposures, the subjects are listed only by their ORAU study identification numbers.

Whole body and lung counts appear to have been performed rarely if at all, since workers had to be sent to sites outside Missouri for this to be done or a mobile counter would have had to be brought to St. Louis. Hence there were evidently so few (if any) such counts done as to be of little use in reconstructing individual doses, except possibly for those individuals actually counted. However, even individual whole body and lung count data (if any) appear to be unavailable.

5.3.8 Summary Information Regarding No-Bioassay-Based Inhalation, Radon, and Ingestion Annual Intakes (Tables 29A-29F, 30, and 31A-31F)

In order to put the measured nonbioassay-based data presented in the preceding subsections of Section 5.3 into the form most useful for dose reconstruction, these data have been converted into annual intakes.