# Dosimetry Is Key To Good Epidemiology: Workers At Mallinckrodt Chemical Works Had Seven Different Source Exposures

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## ABSTRACT

Mallinckrodt Chemical Works was the earliest uranium processing facility in the Manhattan Project, beginning in 1942. Even then, concern existed about possible health effects resulting from exposure to radiation and pitchblende dust. This concern was well founded as the facility processed Belgian Congo pitchblende ore that was up to 60% pure uranium with high $^{235}U$ content and up to 100 mg of radium per ton. Workers were exposed to external gamma radiation plus internal radiation from inhalation and ingestion of pitchblende dust (uranium, radium, and silica). Multiple sources of exposure were available for organ dose reconstruction to a degree unique for an epidemiologic study. Personal film badge measures available from 1945 captured external exposures. Additional external exposure included 15,518 occupational medical x-rays and 210 radiation exposure records from other facilities outside of Mallinckrodt employment. Organ dose calculations considered organ-specific coefficients that account for photon energy and job-specific orientation of workers to the radiation source during processing. Intakes of uranium and radium were based on 39,451 uranium urine bioassays and 2,341 breath radon measurements, and International Commission on Radiological Protection (ICRP) Publication 68 biokinetic models were used to estimate organ-specific radiation absorbed dose. Estimates of exposure to airborne radon and its short-lived progeny were based on radon measurements in work areas where radium-containing materials were handled or stored, together with estimated exposure times in these areas based on job titles. Dose estimates for radon and its short-lived progeny were based on models and methods recently recommended in ICRP Publication 137. This comprehensive dosimetric approach follows methods outlined by the National Council

on Radiation Protection Scientific Committee 6-9 for the Million Worker Study. Annual doses were calculated for six organs: lung, brain, heart, kidney, colon and red bone marrow. Evaluation and adjustment for individual cumulative measures of pitchblende dust inhalation were made for lung and kidney diseases.

Key words: uranium, radium, dosimetry, epidemiology, alpha particles

## INTRODUCTION

The Mallinckrodt Chemical Works (MCW), located in St. Louis and Weldon Springs, MO, began processing uranium in 1942. The facility produced the uranium oxide needed for the Manhattan Project through processing pitchblende from the Belgian Congo, currently known as Zaire. In 1940, Belgium sent 1,250 tons of pitchblende to Staten Island, New York, to prevent it from falling into the hands of Germany. This ore was extremely rich in uranium, upwards of 70 percent, in contrast to the North American carnotite ore which was less than 1 percent uranium. Hundreds of tons of pitchblende were processed to produce 40 tons of uranium oxide used in the first sustained and controlled nuclear reaction in Chicago in December 1942. It would have taken thousands of tons of carnotite and a much longer time to produce the required uranium oxide for the first atomic pile. Pitchblende ore also was the source of Madame Curie's discoveries of radium and polonium.

The Belgian Congo ore contained up to 100 mg of radium per ton of ore, which increased to 300 mg per ton during processing. Both soluble and insoluble uranium compounds were produced during the predominantly dry chemical process. Throughout processing, potential exposures were to internal and external radiation. Exposure to alpha particle emitting dusts was a concern, but no regular dust sampling program was in effect prior to 1948 and no maximum allowable concentration (MAC) had yet been set. A level no greater than 500 μg $m^{-3}$ was thought to provide adequate protection (Harris 1958). Sufficient samples were collected, however, to show that air-borne uranium dust during this period before 1948 was 100–200 times the MAC of 50 μg $m^{-3}$ established in 1948, and up to 1000 times the MAC in poorly ventilated processing

areas (Mason 1975). Industrial hygiene concerns included silica, sulfuric acid, and the chemical toxicity of uranium (Eisenbud 1975).

A previous epidemiologic study of 2,514 white male workers employed between 1942 and 1966, the entire period of uranium processing operations at MCW, considered only external radiation exposure as measured by film badge monitoring at MCW (Dupree-Ellis et al. 2000). In updating the cohort study, the desire was to include radiation doses to target organs of interest resulting from exposure to uranium as well as radium present in the pitchblende ore.

The MCW cohort is part of the Million Worker Study (MWS) (Bouville et al. 2015). The aim of this epidemiologic study of one million U.S. radiation workers and veterans is to provide scientifically valid information concerning radiation risk when exposure is received gradually over time regarding the health effects attributable to radiation dose. The appropriate dosimetric quantity is annual absorbed dose to the target organ or tissue of the radiation exposure received by the study population. Achieving this aim hinges on reconstruction of organ-specific doses that are both accurate and precise. Scientific Committee 6-9 of the National Council on Radiation Protection and Measurement (NCRP SC 6-9) has produced a report on the comprehensive organ dose assessment for the MWS which covers the specifics of practical dose reconstruction and guidelines to be used for the various epidemiologic studies that are or will be included in the MWS (Bouville et al. 2015). This approach has been used in previous epidemiologic studies of MWS cohorts, specifically Rocketdyne and Mound (Boice et al. 2006; Boice et al. 2011; Boice et al. 2014).

The wealth of contemporary radiation monitoring data available for the MCW workers provided the opportunity to construct annual radiation doses for target organs of interest following the guidelines of the NCRP SC 6-9. These data sources were individual film badge readings recorded during MCW employment; individual film badge readings acquired before or after employment at MCW; annual medical chest x-rays required for continued employment in the uranium processing operations; urinary uranium bioassay data; radon breath samples; contemporary time-weighted job-specific dust measurements calculated for each worker; and air monitoring data for radon and radon progeny.

## Radiation Exposure from Uranium Processing

The uranium process at MCW began with receipt of the ore through to the production of uranium metal (Fig. 1). The uranium ore was sent to MCW in sacks or 55 gallon drums. The level of radon associated with pitchblende ore was so high that boxcar doors had to be left open for 12 to 24 hours before workers were allowed to carry the material into the processing facilities. The yellowcake ore was transported and distributed by various means, including shovels. After it was unloaded, dried, ground, and classified, the ore was dissolved in nitric acid to separate uranium decay chain members. The subsequent combination of the solution with sulfuric acid resulted in precipitation of much of the silica and radium as sulfate. For pitchblende ore the precipitate required subsequent extraction of radium because the company that provided the ore retained ownership of its radium content. The aqueous portion containing the uranyl nitrate was extracted with diethyl ether leaving precipitate of

isotopes of the actinides $^{231}$Pa and $^{227}$Ac (members of the $^{235}$U chain) and $^{230}$Th (in the $^{238}$U chain). Ether was replaced with a mixture of tributyl phosphate and hexane in later years. The wet chemistry portion of the processing ended with uranyl nitrate in the solvent phase being stripped into a water solution which was thermally denitrated to produce uranium trioxide, a dry powder. The dry chemistry phase continued with oxidative reduction to produce uranium dioxide, which reacted with anhydrous hydrogen fluoride to produce uranium tetrafluoride. This product was mixed with magnesium and heated to produce pure uranium metal. The metal was recast before being machined, rolled and extruded to the final required form. During the recasting process, $^{231}$Pa and $^{230}$Th condensed on the inside of the furnace. These two isotopes came to equilibrium in the U metal as it aged (Eisenbud 1975).

Throughout the processing, the potential for exposure existed from gamma and beta radiation as well as radon and alpha-emitting dusts. $^{226}$Ra presented a hazard for gamma radiation exposure while handling the ore, precipitating the ore, and extracting radium from the ore precipitate. The potentially important internal emitters at MCW were all naturally occurring radionuclides, primarily members of the $^{238}$U and $^{235}$U decay chains. Frequent exposures to elevated concentrations of airborne $^{238}$U, $^{235}$U, $^{234}$U, $^{226}$Ra, $^{222}$Rn and its short-lived progeny, and other members of these two chains would have occurred during production of purified uranium compounds and metals from uranium feed materials and the resulting waste products. Much of the work involved extraction of uranium from pitchblende ore and concentration of the extract. Other processes with high potential for intake of $^{238}$U, $^{235}$U, and some chain members included recovering uranium from scrap uranium metal; machining natural uranium metal rods to

make reactor fuel slugs; reverting $UF_4$ to $UO_2$ or $U_3O_{8;}$ producing $UO_2F_{2;}$ and extracting and concentrating $^{230}Th$ from pitchblende raffinate. The separated $^{230}Th$ was sent to the Mound nuclear facility for further processing (Westbrook and Bloom 2010).

## Available Radiation Monitoring Data

Table 1 describes the sources of radiation monitoring data that were used to estimate annual organ-specific radiation doses for each worker. Three sources of external monitoring data were available for estimating organ doses. Individual film badge records obtained from Manhattan Engineer District (MED) film badge files and MCW occupational records from mid-1945 through 1966 were the basis for the annual external radiation doses used in the current study. An average of 6 records was found for over 80% of the 2,514 workers included in the study. Among the 2,011 workers with film badge readings, the percentage with cumulative gamma ray exposure recorded on the film badges in mSv was the following: <5  28.90%; 5-9.99 12.55%; 10-19.99 37.00%; 50-99.99 11.12%; 10-19.99 5.88%; 20+ 4.55%. Over 14,500 medical chest x-rays, required annually for continued employment in uranium processing operations, were found for 1,995 workers and included in the radiation dose assessment. Also included were individual film badge records for time periods before or after MCW employment MCW identified for 43 workers.

Several sources of individual monitoring were available for estimating internal organ doses. Uranium urine bioassay data were available for workers from 1948 to 1966. There were 39,451 urine samples available for study, which is more than 15 urine samples per worker. The 2,341 radon breath samples available for 500 workers from

1948 to 1959 were used to estimate the intake of radium. Limited urinalyses for thorium, presumably for monitoring $^{230}$Th intake, were available for a brief period in 1955.

Air monitoring was used as the primary indicator of exposure to airborne radionuclides. The air monitoring program focused on measurement of uranium isotopes, total alpha, and radon. It included both general air and breathing zone sampling. These sampling data were applied to calendar- and activity-specific, time-weighted breakdowns of specific jobs to develop the total dust exposure received for each day worked in that job. The job-specific total daily dust exposure values were applied to each worker's job history to calculate that worker's total exposure to dust from 1942 through 1952. Worker-specific dust exposure estimates were available for 1,091 workers.

## METHODS

### Dose Reconstruction for External Emitters

*MCW film badge*

The deep dose equivalent, i.e., HP(10), from the film badges worn by the workers from mid-1945 onward was used to calculate individual annual organ doses. For the 20.8 % of working years at MCW for which film badge data were not available, an algorithm was used to assign the annual dose (Watkins et al. 1997). Many of the assigned annual exposures were for the period 1942-1945, i.e. prior to the earliest available film badge records in mid-1945.

To calculate organ doses resulting from the external radiation exposure, the photon energy of the radiation source, the organ-specific conversion coefficients resulting from the energy (ICRP 2010), and the geometry of the exposure of the worker

to the radiation source as he was doing his job were included. The primary external radiation emanating from the pitchblende was from radium with an effective gamma ray energy of 0.83 MeV. Scaling factors, i.e. dose coefficients, for the organs of interest were based on this energy level (Table 2, top). Since complete documentation of what each job entailed was not available, an estimate of the orientation of the worker to the radiation source was assigned based on three job groups common at uranium facilities defined in National Institute of Occupational Safety and Health Dose Reconstruction Project (NIOSH 2007). These three orientations were based on the percentage of time for anterior-posterior (AP) exposure to the radiation source: 25%, 50% or 75% (Table 2, bottom). Since the film badge measurements were annual, these orientation assignments were made for each year of employment. If a worker had jobs that fell into more than one of these labor categories in a given year, the labor category was assigned in the following order based on AP exposure: 50%, 75% and 25%. This order was chosen over other possibilities since it tended to provide a dose that was not consistently over- or under-estimated for the given year. This prioritization also addressed workers who were put on a "rotation schedule" of jobs where the potential for radiation exposure was high for some jobs on the schedule. The workers on these rotation schedules tended to have all 3 labor categories in a given year.

The annual organ dose was calculated as follows:

$$\text{Annual organ dose (mGy)} = FB \times 0.83 \times \Sigma(DC_o \times OB_j)$$

where FB=annual gamma film badge reading in mGy, $DC_o$=organ dose conversion factor for the appropriate photon energy (0.83 Mev), and $OB_j$=geometric orientation of

the body for the specific job. For example, the dose to the brain for a general laborer with an annual film badge reading of 50 mGy would be 27.49 mGy, as follows:

50 mGy x 0.83 x [(0.658 x 0.25) + (0.676 x 0.75)].

*Occupational x-rays*

Many nuclear weapons complex workers were required to undergo medical x-ray examinations as a condition of their employment (Cardarelli et al. 2002; Daniels et al. 2005; Anderson and Daniels 2006; Shockley et al. 2008). The medical x-ray program recommended by the MED included a routine chest x-ray prior to employment, an annual chest x-ray thereafter, and a pelvic x-ray examination for employees handling fluorides. The pelvic x-ray examinations were used in an attempt to detect potential bone changes due to fluorosis (Westbrook and Bloom 2010).

From the early days of operations, the health of workers employed in the MCW uranium processing operations was monitored through an extensive occupational medicine program which included mandatory pre-employment, annual, and termination physical examinations. These physicals routinely included radiographic chest x-rays taken from the posterior-anterior (PA) position prior to 1949 and from the PA and lateral position (LAT) beginning in 1949 through 1966. The LAT chest x-ray was usually performed with the left side of the body toward the x-ray film, to image the heart and aorta. Pelvic x-ray examinations appear to have been given infrequently to MCW employees working with hydrogen fluoride or involved in the uranium oxide to uranium fluoride conversion process during 1942 through 1944. Chest x-ray procedures were made using 14- × 17-inch x-ray film at a standard source-to-film distance of 72 inches.

While no information has been found concerning the type of equipment used to take the x-rays, there is no evidence that photofluorography chest x-ray examinations were ever performed (Dupree-Ellis et al. 2000; Westbrook and Bloom 2010).

The National Institute for Occupational Safety and Health Radiation Dose Reconstruction Program (NIOSH-DR), which estimates work-related radiation exposure for workers with cancer who file claims under the Energy Employees Occupational Illness Compensation Program Act of 2000, established "typical" values for exposures and organ doses for various time periods when no site- or machine-specific information is available (Shockley et al. 2008). The typical values were based on general medical literature from the time period and were designed to be somewhat conservative, i.e. high-sided. Based on typical x-ray machine parameters in use during the period prior to 1970, the estimated entrance dose at the center of the x-ray beam and at the surface of the body in terms of kerma in air (or so-called air kerma) was approximately 2 mGy for a PA chest exam and 5 mGy for a LAT chest exam (Shockley et al. 2008; Thomas 2011). These values for air kerma were then used to estimate the radiation dose from x-rays in various organs of the body based on data contained in ICRP Publication 34 (ICRP 1982), which are for well collimated beams used after 1970. However, the radiographic examinations taken at MCW were all before 1970 and were poorly collimated compared to those taken after 1970. This necessitated the use of some dose estimates from other references for organs that were outside the well collimated beams for the chest x-ray examinations considered in ICRP Publication 34 (Shockley et al. 2008; Thomas 2011).

An electronic file containing information abstracted from reports of MCW occupational medical x-rays was used to estimate the annual external radiation dose a

worker received from these x-rays. Individual records in the electronic x-ray file had various notations concerning the part of the body and view of the films taken. Because chest x-rays were taken during routine medical examinations and outcomes of interest included lung cancer, non-malignant respiratory disorders, and cardiovascular diseases, the exposure to the thorax was considered to be important. Those films with a notation of “chest” or “spine” were used to estimate dose to specific organs of interest. The radiation dose for each chest/spine x-ray was assigned based on the view (PA or LAT). If the chest x-ray view was not given, it was assumed to be a PA view based on the NIOSH-DR report (Thomas 2011), which stated that the PA view was consistently taken as part of the physical exam.

For the 1,995 workers with at least one record of chest/spinal x-ray, the electronic records appeared to be complete with about one entry for each year of employment. Their records were used as they appeared in the electronic file. For the 519 workers with no x-ray records in the electronic file, it was assumed that they received only a PA chest x-ray before 1949 and a PA and LAT chest x-ray from 1949 onward as required for each year employed.

The annual organ-specific dose was determined using the following formula:

Annual organ dose (mGy) = (#PA x organ dose-PA) + (#LAT x organ dose-LAT)

where #PA = total number of PA chest x-rays taken that year, organ dose-PA = dose to the organ given the air kerma for PA x-ray, #LAT = total number of LAT chest x-rays taken that year, and organ dose-LAT = dose to the organ given the air kerma for LAT x-ray. Dose equivalents specific for the organ of interest given by Thomas (2011) were used.

*Exposures received outside MCW employment*

Annual doses, received before or after employment at MCW, were obtained from five additional sources of dosimetry records: DOE Radiation Exposure Monitoring System (REMS); other historic radiation exposure data not included in DOE Radiation Exposure Monitoring System; the Nuclear Regulatory Commission Radiation Exposure Information and Reporting System; Landauer, Inc.; and the Nuclear Test Personnel Review Program for military veterans who participated in the U.S. atmospheric nuclear tests from 1944 to 1962 or the occupation forces of Hiroshima and Nagasaki, Japan. The recorded values for external ionizing radiation were added to the organ dose for the year when the exposure was recorded. This process located additional occupational/military external radiation records for 43 workers (mean, 13.4 mGy) and added 0.575 Gy to the population dose.

## Dose Reconstruction for Internal Emitters

In view of the nature of the operations at MCW and available air monitoring and bioassay data, a substantial portion of the collective dose from internal emitters resulted from intakes of natural uranium and $^{226}$Ra. It is probable that elevated intakes of $^{227}$Ac, $^{230}$Th, or $^{231}$Pa also occurred for some workers, but monitoring data are absent for $^{227}$Ac and $^{231}$Pa and are too sparse for $^{230}$Th to allow internal dose reconstruction.

*Internally deposited uranium*

Internal monitoring data for uranium, generally consisting of measurements of the mass concentration of uranium in urine, were found for 1,925 workers. Annual doses from internally deposited uranium were estimated in cases where uranium intakes were indicated by positive measurements (~88% of the workers monitored for uranium intake).

The urine sampling frequency varied considerably among workers, presumably depending on the potential for exposure associated with a given job or work location. In most cases the frequency was in the range 1-12 urine samples per year. In the present dose reconstruction, the urinary excretion data were interpreted as reflecting chronic exposure to a gradually changing level of uranium in air. The data for the discrete urine samples for a worker were extrapolated to a continuous urinary excretion pattern over the entire monitoring period by linear interpolation of results with time between neighboring collection times (Fig. 2). An interval >1.1 years between two neighboring sampling times was assumed to indicate that the worker was not involved in uranium work during much of that period. In such cases, the average of the bounding urinary uranium measurements was assumed to have occurred for only a 0.25-year period. The intake rate at a given sampling time was based on the assumption that 24-hour urinary excretion represented 90% of the amount of uranium reaching the blood during that 24-hour period. The value 90% is based on predictions of the biokinetic models used in the dose reconstruction, assuming that the concentration of uranium in air does not fluctuate greatly between neighboring collection times.

The biokinetic and dosimetric models used to estimate the time-dependent intake of uranium and the resulting annual tissue doses are those recommended in ICRP Publication 68 (ICRP 1994). Specific information generally was not available regarding the mode of intake or the form of uranium entering the body. It was assumed that all intakes were by inhalation. The authors' assessments of follow-up data for workers accidentally exposed to unknown forms of uranium at other production sites (e.g., Boice et al., 2006) indicate that, more often than not, Type M parameter values yield a better fit to uranium excretion data than do parameter values for Type F or S.Thus, it was assumed that i uranium was inhaled in moderately soluble form (Type M). The ICRP's default particle size (5 μm AMAD) was applied. The biokinetic model for systemic (absorbed) uranium applied in ICRP Publication 68 has been adopted by the ICRP for use in an upcoming report on occupational intake of radionuclides (ICRP Publication 137, in press) . The respiratory and gastrointestinal models used in ICRP Publication 68 have been updated (ICRP, 2006, 2015), but the changes appear to result only in modest differences with regard to reconstruction of doses from inhaled uranium.

*Internally deposited $^{226}Ra$*

Breath radon ($^{222}Rn$) measurements were started at MCW in 1945 for workers with potential radium intake. Typically, one liter of exhaled breath was collected after two days of non-exposure, usually on a Monday morning (Fig. 3). If a sample was above the "tolerance" level of 1 pCi $L^{-1}$ (0.037 Bq $L^{-1}$), a second sample was collected and analyzed soon after the result was reported. Analyses were performed at the Massachusetts Institute of Technology or the National Bureau of Standards until 1951

and subsequently at the Atomic Energy Commissions's Health and Safety Laboratory (HASL). This lab's lower limit of detection was 0.1 pCi $L^{-1}$.

Breath radon data were found for 500 workers. In most cases, the sampling frequency was in the range 1-4 per year. The method of extrapolating data for the discrete breath radon samples to a continuous breath radon pattern over the entire monitoring period was analogous to that described above for uranium, i.e., using linear interpolation with time between adjacent measurements (Fig. 4) and the assumption that an unusually long period between measurements indicated absence from radium work for that period except for one quarter (0.25 years).

The biokinetic models used to estimate intake of radium for a worker based on breath radon data are the models applied in ICRP Publication 68 (1994). It was assumed that all radium intakes were by inhalation and that material was inhaled in moderately soluble form (Type M). The ICRP's default particle size (5 μm AMAD) was assumed. The respiratory and gastrointestinal models and the systemic model for radium used in ICRP Publication 68 have been modified for use in upcoming ICRP documents on occupational intake of radionuclides, but the changes appear to make only modest differences with regard to reconstruction of doses from inhaled radium.

Ideally, biokinetic models would be used to determine a $^{226}Ra$ intake rate and hence a rate of accumulation of $^{226}Ra$ in the body that would give rise to the extrapolated radon breath curves. This is generally not feasible in practice because of the typically high variability in the radon breath data over relatively short time periods. In the present dose reconstruction, the assumption was made that the integrated $^{222}Rn$ activity in breath over the entire monitoring period for a radium worker (typically only a

few years) was proportional to the integrated $^{226}Ra$ in the body over that period and that the $^{226}Ra$ intake rate was constant over that period. The constant $^{226}Ra$ intake rate for an individual worker could then be derived using the biokinetic models of ICRP Publication 68, the worker's continuous breath $^{222}Rn$ curve, and a conversion factor that relates breath $^{222}Rn$ to a typical body content of $^{226}Ra$ over an extended period.

Conversion from $^{222}Rn$ in breath to $^{226}Ra$ in the body was based on the assumption that 1 Bq $^{222}Rn$ per liter of air exhaled during a breath test corresponds to a body $^{226}Ra$ content of 72,000 Bq. This relation is derived from a formula proposed by Srivastava et al. (1986) and is reasonably consistent with predictions of the biokinetic models used in this study assuming extended exposure to airborne $^{226}Ra$. Srivastava and coworkers concluded that the body content of $^{226}Ra$ could be estimated from the rate of exhalation of $^{222}Rn$ by the relationship

$$^{226}Ra \text{ in the body (Bq)} = (C \cdot I) / (\lambda_{Rn} \cdot f)$$

where I is the breathing rate in liters ($h^{-1}$), $\lambda_{Rn}$ is the decay constant of $^{222}Rn$ ($h^{-1}$), C is the concentration of $^{222}Rn$ in the breath sample (Bq $L^{-1}$), and f is the release fraction of $^{222}Rn$ from the body. They suggested the value 0.84 as a typical release fraction f but noted that f can vary from 0.7-1.0, depending on whether most of the $^{226}Ra$ is in bone (relatively low f) or in lungs and other soft tissues (relatively high f). The relationship used here (1 Bq $^{222}Rn$ / L = 72,000 Bq $^{226}Ra$) yields a lower estimate of body $^{226}Ra$ than has been estimated by other authors because of a higher breathing rate, representing a worker during a work period, and a lower value of f (down to 0.63) generally have been used. The relationship between radon exhalation and radium body burden used here takes into account that the worker is at rest during the breath test and thus has a much

lower breathing rate than the reference value for a worker. The value f=0.84 estimated by Srivastava et al. was applied.

Internally deposited radon and progeny

Dose estimates for inhalation of $^{222}$Rn and its short-lived progeny $^{218}$Po, $^{214}$Pb, and $^{214}$Bi were based on models and methods used in ICRP Publication 137 (in press) to derive dose coefficients for radon isotopes and mixtures of their short-lived progeny. The biokinetic models applied in Publication 137 include an updated version of the ICRP's Human Respiratory Tract Model (HRTM) (ICRP, 2015), the ICRP's Human Alimentary Tract Model (HATM) (ICRP, 2006); and systemic models for radon, polonium, lead, and bismuth described in Publication 137. The derived tissue doses depend on the assumed fractional deposition of the radon progeny in different regions of the HRTM, which varies with the form of the progeny. Typically, a portion of airborne radon progeny is attached to existing aerosol particles and a portion is unattached. As discussed in ICRP Publication 137, the attached portion typically exists in part in a "nucleation mode" associated with relatively small aerosols and in part in an "accumulation" or "coarse" mode associated with larger aerosols. Publication 137 provides reference values for the following quantities, which determine fractional deposition of radon progeny in different regions of the HRTM: (a) sizes of the unattached, nucleated, and accumulation modes; (b) relative quantities of these three forms of radon progeny in different settings (indoor workplace, mine, or tourist cave): and (c) an equilibrium factor F (i.e., a measure of the disequilibrium between the radon gas and its progeny) for each setting. In the present

study the dose estimates for inhaled radon and its short-lived progeny were based on the following reference values applicable to an indoor workplace: an equilibrium factor of 0.4; unattached, nucleated, and accumulation fractions of 0.08, 0.184, and 0.736, respectively; and particle sizes of unattached, nucleated, and accumulation modes of 0.001, 0.06, and 0.5 μm AMAD (Activity Median Aerodynamic Diameter), respectively.

Air monitoring data for radon are available for several work areas at Mallinckrodt that had elevated levels of radon for extended periods due to handling or storage of $^{226}$Ra-rich material. These monitoring data were used, together with estimated exposure times for individual workers in these areas based on job titles, to estimate exposure levels to radon progeny in terms of working level months (WLMs). The relative concentrations of radon and progeny in air were based on an equilibrium factor of 0.4. Doses from radon and its short-lived progeny were estimated for 1392 workers. Dose estimates were derived separately for $^{222}$Rn, $^{218}$Po, $^{214}$Pb, and $^{214}$Bi based on models and methods described in ICRP Publication 137 and then combined to derive total annual doses for each worker corresponding to their estimated annual exposures. Analyses indicate that the combined dose estimates are relatively insensitive to the combination of $^{218}$Po, $^{214}$Pb, and $^{214}$Bi used to represent a working level.

*Brain dose*

Estimates of brain dose from internally deposited uranium or radium were based on standard ICRP methodology for estimating dose to a tissue not explicitly addressed in the biokinetic model for a radionuclide. That is, the activity of uranium or radium and

any of their chain members contained in the brain is estimated as the activity in “Other tissue” (the collection of all tissues not addressed explicitly in the biokinetic model) times the reference mass of brain, divided by the reference mass of “Other tissue” for that model. The estimated dose to brain is the self-dose from the activity contained in brain, plus any cross irradiation from the rest of the body (which is zero for alpha particles). Thus, the estimated dose to brain contains the contribution of high and low LET radiations emitted within the brain plus cross irradiation from penetrating radiations emitted from the rest of the body. In the case of the uranium and radium isotopes addressed in the MCW dose reconstruction, the contribution to brain dose from low-LET radiation is negligible so that the brain dose can be considered as an alpha dose to brain. Levels of radium have been detected in brain tissue among workers with known intakes (Hursh and Lovaas 1963), i.e. radium and other alpha-particle emitting radionuclides can cross the blood-brain barrier.

## Exposure to Uranium Dust

Because of the high levels of airborne alpha-emitting dust in the uranium processing operations at MCW, there was a concern about both the radiation and chemical toxicity of uranium to the kidney. Silica associated with the ore was also of concern with regard to the lung.

From the earliest days of uranium operations, respirators were required for almost all operations, but they were not used consistently (Westbrook and Bloom 2010). Both general air and breathing zone sampling for alpha-emitting dust were done frequently beginning as early as 1943. Prior to 1953, worker-specific estimates of daily exposure to dust were calculated in multiples of the MAC (MMAC), which was

established as 50 μg $m^{-3}$ in 1948. Each job was broken down into its specific components with the number of times per day, number of minutes, and dust concentration determined for each component to derive the daily time-weighted average dust exposure (DTWA) for the job. Based on these job-specific DTWAs, annual cumulative levels of dust exposure in MMAC were assigned to each worker based on his job history. These annual cumulative MMACs were converted to mg $m^{-3}$-$year^{-1}$ which was used to evaluate the effect of dust exposure on risk of malignant and non-malignant kidney and lung disease. The job-specific DTWAs were identified for 1,091 of the 1,457 workers employed any time during the period from 1942 through 1952. A job-specific DTWA was constructed from the work history for the 366 workers employed during that period but lacking cumulative DTWA.

An example of the method that was used to convert the MMAC in μg $m^{-3}$ for specific jobs in the DTWAs to an annual cumulative value in mg/$m^3$-year for a given worker is given in Fig. 5. The time period covered in the example spans across three years. For each job title, the MMAC was multiplied by the weeks worked at that job to produce MMAC-weeks. For example for the first job listed in the upper table of Fig. 5, 13 weeks was multiplied by the MMAC of 36.5 to produce 474.5 MMAC-weeks. This was done for each job listed and then summed over all jobs so that the average MMAC-weeks for that time period could be determined (i.e., 4,292.9/94=45.669). Since the time period in the example crosses three years, the average value for MMAC-weeks was multiplied by the number of weeks worked each year to produce a cumulative MMAC for that calendar year. This latter value was divided by 0.05 mg $m^{-3}$ [i.e., (50 μg $m^{-3}$) x $10^{-3}$]

to produce mg $m^{-3}$-$year^{-1}$. When a worker had more than one time period cluster of job title-weeks-MMACs, this process was repeated for each time cluster.

## RESULTS

Descriptive statistics of the contribution of each type of personal radiation monitoring data to the overall dose to the organs of interest are given in Table 3. The highest dose was to the lung with an average of 59.9 mGy and a median of 29.6 mGy. This average was over 12 mGy higher than the average for the heart, which had the next highest dose (average=47.5 mGy; median 23.3 mGy). The average doses for the remaining organs varied between 35 and 40 mGy. The difference between the average and median indicate that the data are somewhat skewed by high exposure excursions. The cumulative dose was greater than 640 mGy for every organ.

In each case, the dose from the external film badge contributed the greatest portion of the total organ dose. This was followed by the contribution of the occupational medical x-rays for all organs except the lung. For the lung, the internal dose contributed by exposure to uranium and radium was the next highest contribution to the total dose.

Table 4 presents the annual cumulative dust exposure estimates for workers employed between 1942 and 1952. The overall average was 15.08 mg $m^{-3}$-$year^{-1}$. The average gradually increased to a maximum of 30.88 mg $m^{-3}$-year in 1944 and then declined to 16.72 mg $m^{-3}$-$year^{-1}$ in 1949. It dropped almost three-fold in 1950 after which it continued to decline to its lowest level of 1.23 mg $m^{-3}$-$year^{-1}$ in 1952.

## DISCUSSION

There is an extensive body of epidemiologic literature addressing the health effects from occupational exposure to ionizing radiation, including uranium, but the quality of the dosimetry supporting this literature varies extensively and hampers the usefulness of the results from these studies (Canu et al. 2008; Zhivin et al. 2014; Till et al. 2017). If important questions such as the tenability of the linear non-threshold assumption or the likely effects of exposure to radiation encountered by astronauts on a mission to Mars are to be addressed, the quality of the dosimetry used in the epidemiology studies is critical to developing the risk estimates needed to answer these questions. The best available methods given the dosimetry data available should be used to conduct these studies. The NCRP SC 6-9 has provided a standard for developing organ doses useful in epidemiologic studies that incorporates the application of dose calculation methods documented by the NCRP and ICRP (Bouville et al. 2015).

The uranium processing operations at MCW were instrumental in the birth of the nuclear age. Despite the urgency of the times and the lack of knowledge of the health effects of the operations, there was concern within the MED about the effects of the radiation exposure on the workers. This is reflected in the wealth of very early monitoring data available for the MCW workers as well as workers at other MED sites. The employment of all five available sources of radiation monitoring data facilitated the application of the methodology endorsed by NCRP SC 6-9 in reconstructing organ doses, from both internal and external radiation sources, useful for an epidemiologic study. In addition, the availability of contemporary measures of

worker specific exposure to alpha-emitting dust allowed the consideration of this type of occupational exposure to be included in the analysis.

In the present study, dose estimates from internal emitters were derived for uranium (assuming natural isotopic ratios of $^{238}$U, $^{235}$U, and $^{234}$U), $^{226}$Ra, and $^{222}$Rn and its short-lived progeny $^{218}$Po, $^{214}$Pb, and $^{214}$Bi. Even with all the monitoring data available, exposures to a number of radioisotopes were not considered. These includethe actinides $^{230}$Th, $^{231}$Pa, and $^{227}$Ac. Only limited monitoring data were available for $^{230}$Th, and data were lacking for $^{231}$Pa and $^{227}$Ac. Depending on their solubility in the lungs, these three alpha-emitting bone-seekers may transfer to blood and accumulate in bone to a significant extent.

It was assumed that all intakes of uranium or $^{226}$Ra were via inhalation of moderately soluble (Type M) material. This assumption may yield substantial under- or overestimates of the residence time of activity in the lungs and hence the lung dose in some cases. For example, $UO_2$ and $U_3O_8$ were both handled at Mallinckrodt. These forms of uranium have been classified as insoluble material (formerly called Class Y and now called Type S) in some ICRP reports (ICRP, 1979, 1995). In ICRP Publication 137 (in press), $UO_2$ and $U_3O_8$ are assigned to a uranium-specific absorption type intermediate to Type M and Type S. Thus, for Mallinckrodt workers whose intakes of uranium were largely to $UO_2$ and $U_3O_8$, the default assumption of Type M material may underestimate lung dose. For workers who mainly inhaled relatively soluble forms of uranium such as $UF_6$ or whose intakes were mainly via ingestion or wounds, the assumption of inhalation of Type M material may yield a sizable overestimate of lung dose.

The uncertainties in doses associated with lack of knowledge of the mode of intake or form of uranium taken into the body are much smaller for systemic tissues than for the lungs. This is because the dose estimates for uranium were based on cumulative losses of uranium in urine over time, which closely reflect the amount reaching the systemic circulation regardless of the mode of intake or the form of uranium entering the body.

Dose estimates for $^{226}$Ra were based on measurements of $^{222}$Rn in breath, which is a more error-prone method of dose reconstruction for systemic tissues than urinary excretion measurements. This is because in some cases the exhaled $^{222}$Rn may have arisen largely from $^{226}$Ra retained in the lungs rather than $^{226}$Ra retained in systemic repositories. Thus, the dose estimates for $^{226}$Ra involve sizable uncertainties for systemic tissues as well as lung tissue.

The dose estimates for inhaled radon and its short-lived progeny involve larger uncertainties than dose estimates for either uranium or $^{226}$Ra. This is because the estimated doses from radon and progeny are based on hypothetical exposures of workers derived from measured radon concentrations in certain work areas and times spent in those areas by workers based on their job titles, rather than on direct exposure data for individual workers. An aim of an epidemiologic study is to estimate risk, which requires doses to be as precise and accurate as possible. Overestimating the dose underestimates disease risk just as underestimating the dose overestimates disease risk. While the methodology endorsed by NCRP SC 6-9 relies on dosimetric and biokinetic models to produce the organ doses, assumptions have to be made to apply the models. In addition, some workers were missing records or

had incomplete records. When this occurred, the doses were imputed. The external radiation exposure data available for the 43 workers before and after employment at MCW did not contain the level of detail available for the same data collected while employed at MCW. This lack of detail decreased the precision and accuracy of the organ doses for those workers. The personal monitoring records for internal radiation exposure available from other sites were not included. In addition, there may be records that have not been identified for some workers. The assumptions that were made, the methods used to impute missing data, and the decisions that were made to use the personal monitoring data collected outside of MCW employment may have affected the dose that is produced for some workers.

While there are some limitations, the extensive collection of radiation exposure data available for the MCW uranium processing workers provided the broadest application to date of the methodology endorsed by the NCRP SC 6-9 for construction of organ doses for use in epidemiologic studies. As the MWS expands to include additional worker cohorts whose doses were constructed using this or similar methodology (Boice et al. 2006), the data across studies can be more validly combined to produce the statistical power needed to appropriately address radiation issues of importance today and into the future, such as projected risk from computed tomography; environmental contamination from nuclear waste or from terrorist intent; compensation issues related to prior exposures; improved protection standards for workers and the public; providing judgment of the safety and radiation risks for astronauts leaving earth orbit; and whether the linear non-threshold model remains appropriate for the purposes of radiation protection.

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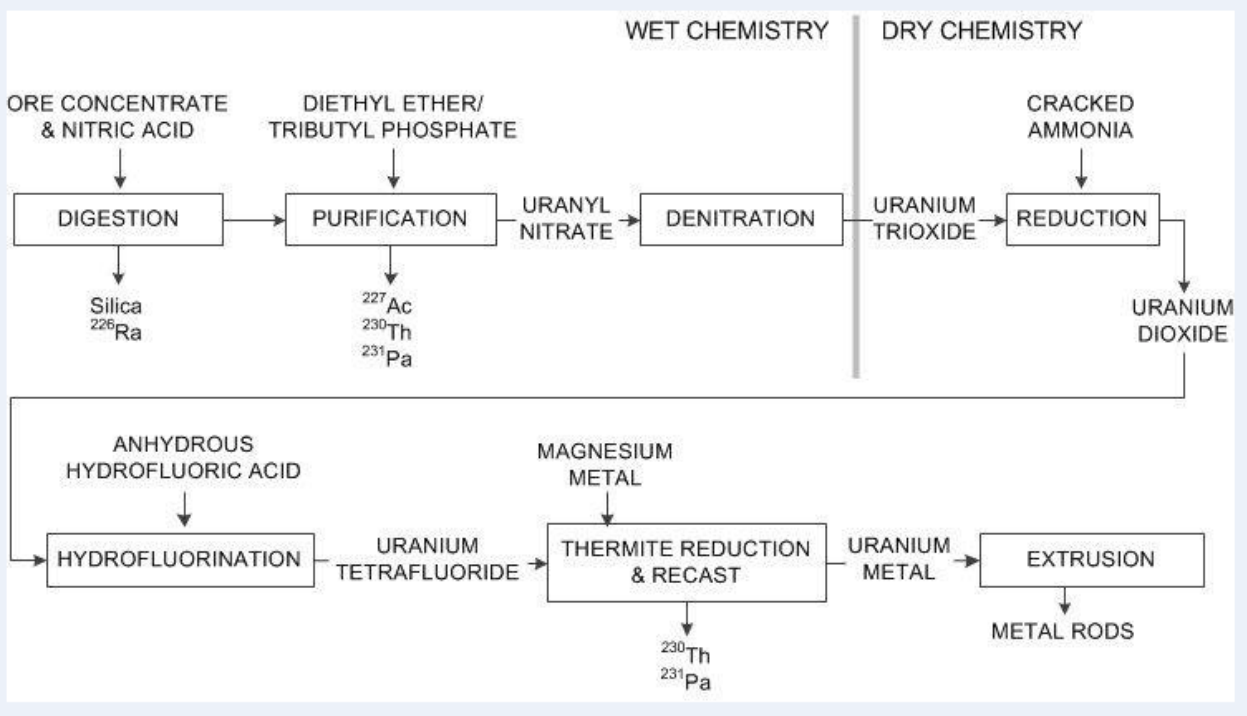
List of Figures

Fig. 1. Uranium processing at Mallinckrodt Chemical Works: 1942-1968*




*Adapted from Mason 1975

Fig 2. Illustration of a dose reconstruction for a worker using his discrete uranium urinalysis samples to construct a continuous urinary excretion pattern over the entire monitoring period by linear interpolation of results between neighboring collection times. The tick mark above 1960, for example, is positioned at 1 January 1960.

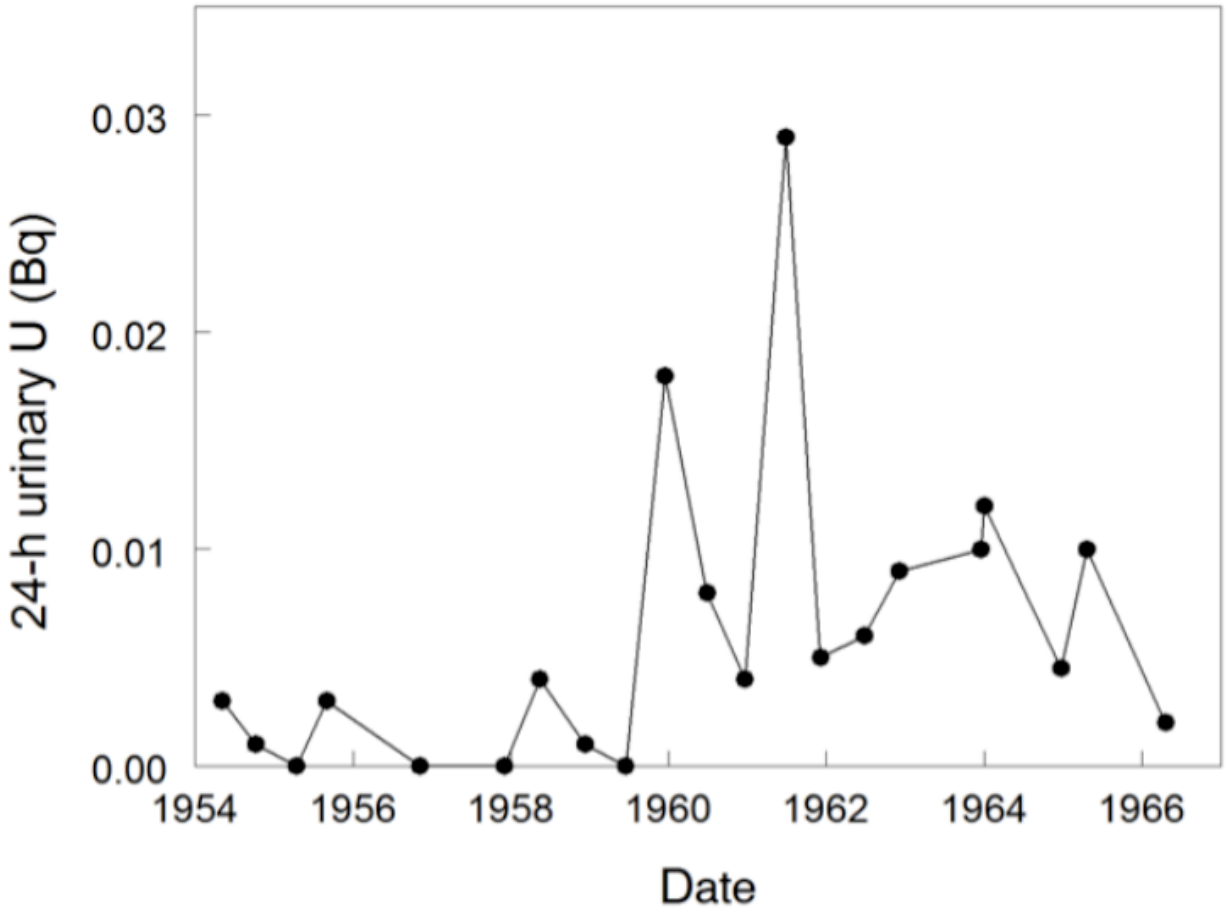


Fig. 3. Radon breath sampling using clean compressed air after 48 hours away from work

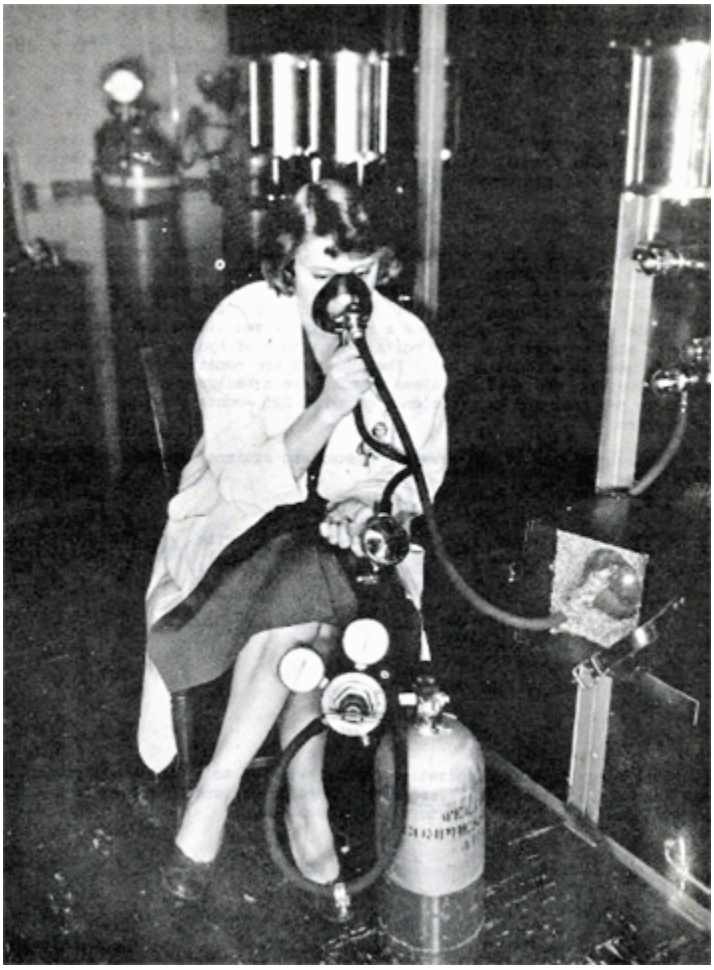


Fig. 4. Illustration of a dose reconstruction for a worker using his discrete breath radon samples to construct a continuous $^{222}Rn$ excretion pattern over the entire monitoring period by linear interpolation of results between neighboring collection times. The tick mark above 1952, for example, is positioned at 1 January 1952.

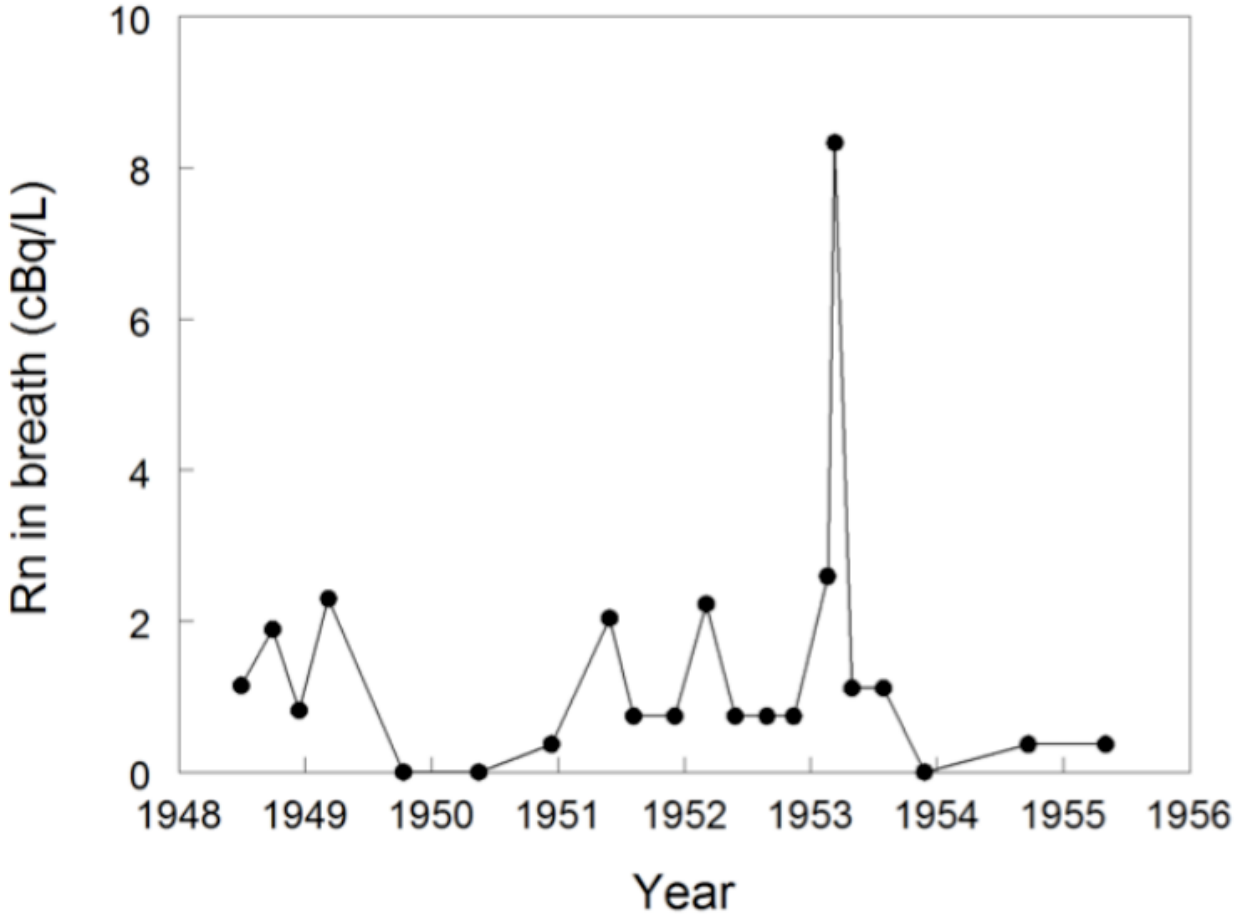


Fig. 5. Example of the conversion of the multiples of maximum allowable concentration in μg $m^{-3}$ on a dust exposure worksheet (top) to annual cumulative dust exposure in mg $m^{-3}$-$year^{-1}$ (bottom) for the period 8 December 1947 through 1 October 1949.*

| Job Title | Total Weeks | MMAC† | MMAC - weeks |
|---|---|---|---|
| CLEANER TOP | 13.00 | 36.5 | 474.5 |
| BOT MAN | 52.00 | 59 | 3068 |
| SAW MAN | 8.00 | 5.1 | 40.8 |
| CAGE MAN | 8.00 | 51 | 408 |
| TOP MAN | 13.00 | 23.2 | 301.6 |
| TOTAL | 94.00 | | 4292.9 |
| Time-weighted average MMAC | 4292.9/94 | | 45.669 |

| Time period split out by year | Weeks worked in year | Cumulative MMAC | mg $m^{-3}$ – $year^{-1}$ |
|---|---|---|---|
| 12/8/1947 to 12/31/1947 | 3 | 137.007 | 6.850 |
| 1/1/1948 to 12/31/1948 | 52 | 2374.788 | 118.739 |
| 1/1/1949 to 10/1/1949 | 39 | 1781.091 | 89.055 |

*Maximum allowable concentration =50 μg/$m^3$
†MMAC=multiples of maximum allowable concentration

Table 1. Radiation monitoring data available for uranium processing workers at Mallinckrodt Chemical Works: 1942-1966.

| Type of data | Number of records | Number of workers | Years covered |
|---|---|---|---|
| MCW film badge[a] | 13,832 | 2,118 | 1945-1966 |
| Occupational x-rays | 14,649 | 1,995 | 1942-1966 |
| Non-MCW film badge/TDL | 210 | 43 | 1943-2003 |
| Uranium urinalysis | 39,451 | 1,925 | 1948-1966 |
| Breath radon | 2,341 | 500 | 1948-1955 |
| Uranium dust exposure worksheets | 6,300 | 1,091 | 1942-1952 |

[a]This includes the film badge records found in Manhattan Engineering District files.

Table 2. Organ dose conversion factors (top) and geometric orientation values for specific job categories (bottom) used in the calculation of specific organ doses resulting from external ionizing radiation exposure to radium (effective gamma energy=0.83MeV).

Organ dose conversion factors

| Organ | AP | ISO |
|---|---|---|
| Brain | 0.658 | 0.676 |
| Lung | 0.831 | 0.601 |
| Red bone marrow | 0.741 | 0.572 |
| Kidney | 0.587 | 0.536 |
| Heart | 0.844 | 0.568 |
| Colon | 0.846 | 0.565 |

Orientation of the body

| Job | Geometry | Percentage |
|---|---|---|
| General laborer | AP | 25 |
| | ISO | 75 |
| Machinist | AP | 75 |
| | ISO | 25 |
| Supervisor | AP | 50 |
| | ISO | 50 |

Table 3. Descriptive statistics for contribution of each type of personal monitoring data to the organ-specific doses in mGy.

| Organ | External-Film Badge/TLD N=2514 | External-Medical xray N=2514 | Internal U and Ra N=1886 | Total External+ Internal N=2514 |
|---|---|---|---|---|
| | Dose (mGy) Average [Std] Median [Max] | | | |
| Lung | 36.0 [68.9]<br>11.2 [724.2] | 10.7 [8.7]<br>7.2 [53.6] | 17.7 [27.1]<br>8.9 [396.0] | 59.9 [90.7]<br>29.6 [829.5] |
| Brain | 33.0 [62.5]<br>10.7 [738.4] | 4.2 [3.6]<br>3.0 [20.0] | 0.067 [0.091]<br>0.037 [1.4] | 37.2 [63.9]<br>15.2 [749.6] |
| Heart | 35.6 [68.4]<br>11.0 [710.8] | 11.8 [9.7]<br>8.0 [59.3] | 0.065 [0.085]<br>0.037 [1.3] | 47.5 [72.9]<br>23.3 [737.6] |
| Kidney | 27.9 [53.1]<br>8.9 [603.5] | 10.7 [8.7]<br>7.2 [53.6] | 0.66 [0.79]<br>0.42 [11.5] | 39.1 [57.7]<br>20.2 [637.4] |
| Colon | 35.6 [68.4]<br>11.0 [711.5] | 2.3 [1.9]<br>1.8 [11.9] | 0.12 [0.19]<br>0.05 [2.6] | 38.0 [69.3]<br>13.4 [713.4] |
| Bone marrow | 32.9 [62.9]<br>10.3 [675.5] | 2.2 [1.9]<br>1.8 [11.1] | 0.48 [0.95]<br>0.15 [9.9] | 35.5 [64.2]<br>12.8 [682.6] |

Table 4. Descriptive statistics for annual cumulative dust exposure in mg $m^{-3} – y^{-1}$: 1942-1952.

| | | Dust Concentration (mg $m^{-3} – y^{-1}$) | | | |
|---|---|---|---|---|---|
| Year | No. of workers | Average | Minimum | Maxium | Standard Deviation |
| 1942 | 82 | 10.77 | 0.01 | 90.47 | 20.62 |
| 1943 | 256 | 25.45 | 0.00 | 399.57 | 49.53 |
| 1944 | 303 | 30.88 | 0.00 | 400.33 | 50.33 |
| 1945 | 293 | 25.27 | 0.00 | 399.57 | 45.55 |
| 1946 | 449 | 21.93 | 0.00 | 283.95 | 42.72 |
| 1947 | 498 | 26.36 | 0.00 | 300.06 | 48.19 |
| 1948 | 533 | 27.90 | 0.00 | 300.63 | 46.93 |
| 1949 | 737 | 16.72 | 0.00 | 246.48 | 34.62 |
| 1950 | 762 | 5.35 | 0.00 | 317.20 | 16.32 |
| 1951 | 738 | 3.18 | 0.00 | 121.87 | 7.08 |
| 1952 | 765 | 1.23 | 0.00 | 36.41 | 2.13 |
| Total | 5416 | 15.08 | 0.00 | 400.33 | 35.78 |