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AIR FORCE SAFETY CENTER**

**Topical Issues for Assessment
of Dose to Palomares Accident
Recovery Workers (1966)**

Steven E. Rademacher



Final

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14. ABSTRACT On 17 January 1966, a B-52 and KC-135 collided during a mid-air refueling operation over southern Spain. Two of the four nuclear weapons detonated upon impact, causing distribution of weapons grade plutonium (WGP), and highly enriched and depleted uranium (HEU and DU, respectively) to the environment. The plutonium is the overwhelming concern for internal radiological exposures. The Department of Defense had about 1,600 personnel supporting the restoration of plutonium-contaminated soils. This report documents provides a topical review of issues pertinent to assessment of doses recommended to response personnel to support Veteran Administration health-based claims. The primary health conditions of concern for inhalation of insoluble plutonium is increased risk of cancer to the lung, liver, and bone. This is based on extensive animal exposure studies and epidemiological studies of former Soviet Union plutonium workers.						
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Executive Summary

On 17 January 1966, a B-52 and KC-135 collided during a mid-air refueling operation over southern Spain. In addition to the mid-air explosion of the KC-135 aircraft, the B-52 suffered damage that caused the plane to break-up mid-air, with the jettison of four nuclear weapons. Two of the four weapons detonated upon impact, which with the influence of winds at the time caused distribution of weapons grade plutonium (WGP), highly enriched and depleted uranium (HEU and DU, respectively), and tritium to the environment. It is important to note that the detonation only involved conventional high explosives; there was no nuclear contribution. The other two weapons were recovered intact – one from land and the other from the Mediterranean Sea. The majority of the residual radiological contaminants were retained in surface soils in the small rural village of Palomares. The primary radiological concern for workers was the inhalation of plutonium.

All four of the KC-135 crew died in the accident, while four of the seven crew of the B-52 survived. A small Air Force (AF) team departed from Torrejon Air Base (AB), Spain, within about two hours of notification of the accident. By the evening, 49 US personnel had arrived at the accident site, with over 650 personnel within a few days. The large majority of military personnel supporting the land-based recovery operation were AF, primarily from two Spanish bases: Torrejon AB and Moron AB. Over the course of the recovery operation, 17 January to 7 April 1966, about 1,600 personnel supported the land portion of work.

In 2000, based upon veteran inquiries of potential health effects from their on-site support to the recovery operations to the Veterans Administration (VA), the AF Surgeon General's Office (AF/SG) contracted with Labat-Anderson, Inc. to assess radiation exposure potential for workers. The primary focus of their work was evaluation of urine sample results from recovery workers. The report also discussed some environmental sampling performed by the Spanish, shortly after completion of the recovery action. The latter information was based on an AF/SG request to assess supporting environmental data. Labat-Anderson provided estimated doses to twenty-six individuals among the 400+ recovery workers that submitted urine samples, as part of an Air Force-directed re-sampling program in the summer of 1966 for Palomares recovery workers. These 400+ individuals had the highest predicted inhalation intakes of plutonium among about 1,400 samples collected by workers on-site. Due to the on-site collection and the gross α -particle analysis laboratory method, a sizeable fraction of these results had questionable utility for dose assessment. The primary concern was cross-contamination of urine samples with plutonium that was not metabolized by the body. Unless sampling methods limit this potential contamination, sample results will be high-biased and misleading. Individuals with the highest predicted intakes based on initial urine sampling results were deemed implausible when balanced against the results of air sampling conducted during the recovery. Other important factors considered were airborne concentrations predicted from historical resuspension studies models, and a large number of initial urine samples from other recovery workers that appeared more reasonably consistent with expected intakes based on airborne sampling results.

High levels of inhaled plutonium have been linked to increased risk of lung, liver, and bone cancer based on animal research studies and epidemiological studies of former Soviet Union plutonium workers (Labutina *et al.* 2013). The majority of claims received by Palomares recovery

workers have been based on malignancies not linked to inhalation of plutonium. Still, other claims are based on non-radiogenic conditions. This is common and experienced by the Safety Center's history of evaluating claims for other occupational exposure radiation cases. Regardless of a recovery workers medical condition, all claims are reviewed with the same level of care, with an appropriate dose recommendation being made to the VA. To date, the AF/SG has received about 30 claims from AF Palomares recovery workers. Additionally, the Safety Center has been requested to offer guidance to the US Army and US Navy for a few claims submitted by their personnel.

Over the years, the Air Force has been questioned for the approach used in estimates and/or the outcome of a VA decision, in particular to Palomares claims. This document provides a discussion of issues pertinent to assessment of doses for the Palomares recovery workers. Due to the issues that have been raised over the years, this report is written in a topical manner. The primary issues relate to estimates of plutonium inhalation intakes (subsequently directly impacting dose estimates) and probability of causation assessments (PoC). Both of these factors are key in VA decisions. The Figure below provides an illustration of the key factors that contribute to the review and adjudication of radiation exposure claims for Palomares recovery workers. The last of the key factors is radiogenic disease assessment. For Palomares recovery workers, assessment of probable dose is impacted by a number of factors shown in the Figure. They fall within three major categories: urine sample results of workers and Palomares residents, air sampling results, and predicted airborne resuspension based on documented ground contamination. All three are deemed mutually supportive to the estimates of probable dose. Due to the extensive reliance on International Commission on Radiological Protection (ICRP) models for the modeling plutonium internal exposures and dose assessments, the report contains detailed information on the evolution of ICRP modeling of plutonium. Other important factors considered by the VA are results from animal and human studies of plutonium exposure, which in part fulfills the VA's requirement to weight sound scientific and medical evidence.

The primary conclusions from the report are summarized.

a) The radiation exposure standard used at the time by the Air Force were based on ICRP Report 2, issued in 1959. Over the intervening 60 years, ICRP has made four changes to their recommendations for internal exposures to workers from plutonium, with the latest published in 2019. In the early years, lack of information on radiation effects and plutonium metabolism in humans was augmented by data from animal exposure studies and prudent conservatism. Since then, knowledge on humans from epidemiological and autopsy studies of exposed workers has allowed refinement of standards. This expanded knowledge is incorporated into the latest ICRP recommendations. Despite these recent updates, there have been only modest changes in the acceptable intakes of plutonium for workers over the past 60 years. Exposure to workers within the limits of these standards protect against deterministic effects of radiation (e.g., reddening of the skin) and maintain probability of stochastic effects (e.g., cancer and genetic) at very low levels. The Air Force has adopted changes in the recommendations made by ICRP.

b) Analysis of initial urine samples submitted by recovery workers had potential for "high-bias" in predicted inhalation intakes of plutonium. This was related to two factors: the existence of α -particle emitters in routine urine excretions from background radiation sources and the potential

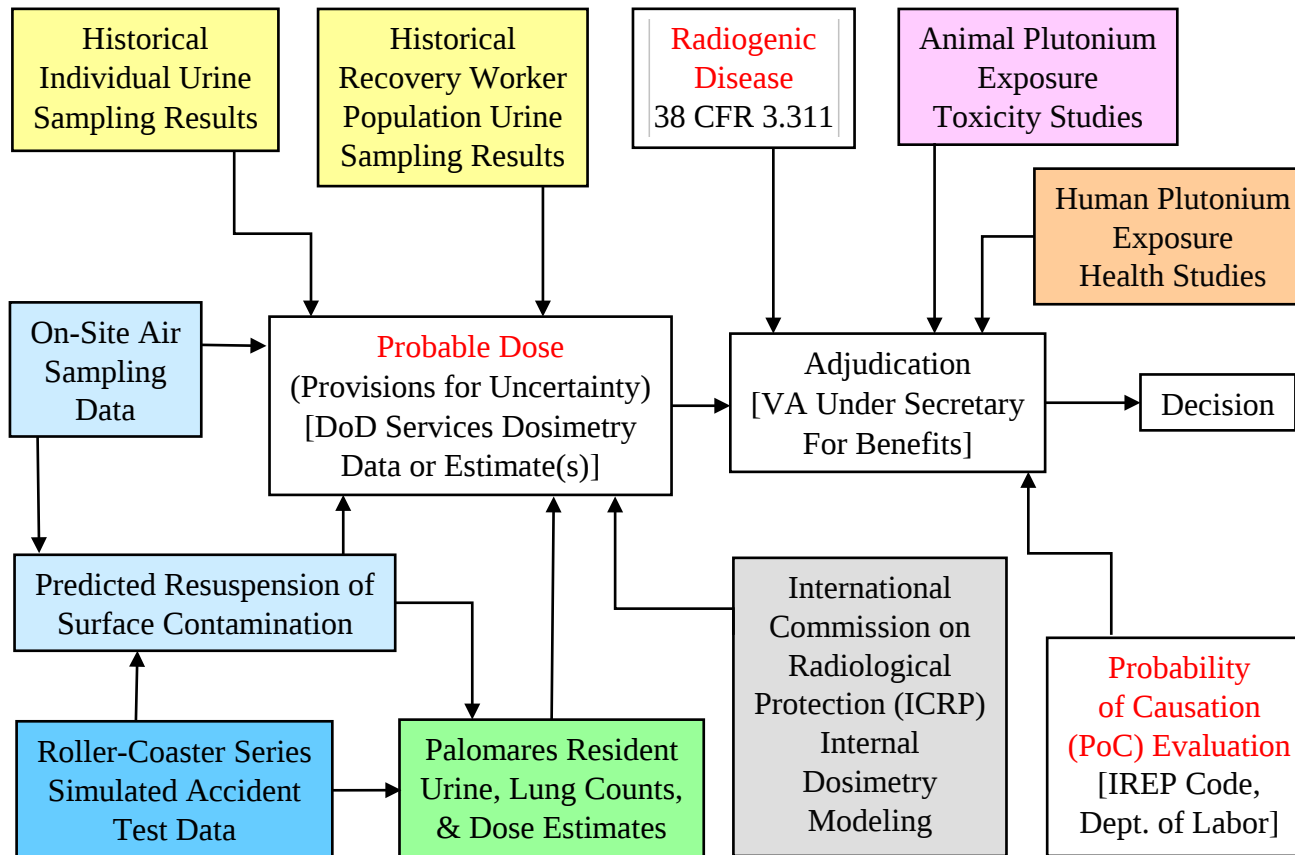


Figure ES. Key Factors in Review and Adjudication of Palomares Recovery Worker Radiation Exposures.

for contamination of urine samples from plutonium that was not internally metabolized by the worker. Despite these shortcomings, the results from these samples provided a sound technical basis for selection of 400+ individuals for participation in a urine re-sampling program. The resampling program used a plutonium-specific analysis method that removed the bias from background α -particle emitters in urine, removed the cross-contamination problem as these samples were not collected on-site, and were collected at a time when urinary excretion values have lower variability in predicted intake, as introduced by the uncertainty in the time of intake. The initial results of the resampling confirmed that high-bias was introduced by a combination of these factors, as the number of individuals within the highest predicted intake categories dropped significantly between the two sampling efforts. The Air Force recommended additional urine sampling on 26 individuals among the group of 400+ that participated in the urine resampling program.

c) In 2000, the Air Force contracted with Labat-Anderson, Inc. to evaluate exposure potential for Palomares recovery workers. Dose assessments were prepared for the high-26 individuals based on two computer-based programs. Doses were recommended based on modeling with the Code for Internal Dosimetry (CINDY), as it implemented the ICRP methodology in current use by the Air Force for its occupationally-exposed workers. Labat-Anderson did not recommend assessment of dose for other individuals due to the greater levels of uncertainty that existed for small intakes predicted by the urine results, and also in part because the other individuals did not submit the number of samples common to the high-26. Among the high-26 individuals, the degree of plutonium inhalation intake by the two individuals with highest intakes is questionable. For one of these individuals, only a single additional sample was provided by the veteran. For the other veteran, highly varied results existed among the samples, the latter two of which were below the detection level of the analytical method. Among the top-third highest predicted plutonium intakes within this high-26 group, all but one were present within one day of the accident. Among the high-26, all but five were present within the first 12 days. Early presence was an important factor in exposure potential. Over time, airborne resuspension of ground-deposited plutonium decreases rapidly. During remedial actions that enhanced airborne resuspension, e.g., plowing and scraping, mitigation by application of water suppression and access restrictions were effective, as observed in air sampling results. As well, air-purifying respirators were worn by some recovery workers during specific activities. This would have made inhalation intakes negligible.

d) The Spanish conducted important medical monitoring of Palomares residents. Projected inhalation intakes were verified by urine analysis in a small fraction of the population. The Spanish, similar to the Air Force, identified problems with plutonium contamination high-biasing results in samples collected from residents while they were in the Village of Palomares. This necessitated adopting a practice of urine sample submission while residents were present in Madrid, where plutonium levels in the environment were substantially lower. An assessment of air sampling conducted during the joint US-United Kingdom plutonium scatter safety studies in Nevada in 1963 was considered for assessment of exposure to Palomares residents from the initial plume transport created by the detonations. It was concluded that it could have been responsible for all or the vast majority of intakes observed in a small fraction of the Palomares residents with verified plutonium intakes.

e) The Air Force, consistent with 38 CFR 3.311, *Claims Based on Exposure to Ionizing Radiation*, provides estimates of radiation dose upon request by the Veterans Administration for personnel with occupational exposure potential. For Palomares recovery workers, the AF/SG in conjunction with our office, developed an approach to estimate doses. For individuals in the high 26 group, it was agreed that doses reported in Labat-Anderson should be provided to the VA, based on ICRP 26/30/48 methodology. For application to organs not covered by this set of ICRP recommendations, it was agreed to use the updated ICRP model in Reports 60/68. AF/SG and our office thought the estimated doses to the lung, liver, and bone surfaces, as deemed appropriate for intakes to the vast majority of recovery workers, likely to receive favorable compensation decisions by the VA for primary cancers originating in these organs. Notably, the Air Force does not tailor dose assessments based upon this consideration, and recognizes that the VA has sole authority for adjudication of claims for radiogenic diseases.

f) For the recovery workers that were not in the high 26, it was recommended by the AF/SG to assign an inhalation intake consistent with the lowest estimate intake among the high 26, which is 34 nCi. This intake level was believed applicable to the vast majority of recovery workers. There was provision to recommend lower intakes for recovery workers that did not have on-site presence, e.g., Navy personnel that provided logistical transports to local ports or were assigned duties on Naval vessels searching for the lost weapon. The vast majority of recovery workers not within the high 26 would be assigned an intake of 34 nCi. This intake level represents a high-sided estimate of intakes among recovery workers that did not have dose estimates prepared within the 2001 Labat-Anderson report. As detailed in this report, this intake level represent an estimated 95th percentile or greater plutonium intake level, and meets the 38 CFR 3.111 requirement for assessment of probable dose with provision for uncertainties. The high-sided dose estimate approach used here is similar to other radiation exposure dose assessment practices used within the DoD.

g) The VA is charged under 3.311(c)(1)(i) to assess whether it is:

*“at least likely as not the veteran’s disease resulted
from exposure to radiation in service,”*

using sound scientific and medical evidence. To this end, there are three key factors of consideration: does the veteran have a radiogenic disease, the probable dose received by the veteran, commonly supported by information provided by the Services, and the probability of causation (PoC) typically assessed by the RadioEpidemiological Program (IREP). To account for uncertainties, the calculated PoC is commonly assessed at the 99% credibility level (CL). This approach lends to factors of 10-fold or greater margins of benefit in assessment of PoC for veterans over 50% PoC and at a 50% CL¹. The primary uncertainty calculated by IREP are those related to the dose-response models. IREP has the capability to jointly incorporate uncertainty in dose estimates and dose-response models. At the 99% CL, uncertainties in dose provided a small impact on PoC, because of the dominance of uncertainty afforded to the dose-response models. Additionally, uncertainties in dose are already applied in a separate phase of the claim adjudication

¹ The 50% PC at the 50% CL meets the 3.311(c)(1)(i), *“at least likely as not the veteran’s disease resulted from exposure to radiation in service,”* definition.

process. The AF recommends high-sided dose estimates for Palomares veteran claims not within the high-26 cohort. IREP uses linear dose-response factors for solid tumors, as applicable to primary lung, liver, and bone cancers. The study of former Soviet Union plutonium workers, which received some of the highest internal exposures of internally-deposited plutonium, found a threshold-like dose-response characteristic for risk of lung cancers when corrected for smoking, a liver cancer dose-response curve with better fits to quadratic and linear-quadratic models, and for bone cancer a threshold-like dose-response curve. The results from these studies provide further conservative influence on compensation decisions.

h) The value of conducting additional medical surveillance for Palomares responders has been an issue of interest since the completion of the Labat-Anderson report. AF/SG has discussed this issue a number of times with our office since, and evaluated appropriate analytical methods. A number of current methodologies are significantly more sensitive for detection of plutonium in urine than the isotopic plutonium method using α -particle spectrometry, as used for the urine resampling program. Also, due to the pernicious retention in some tissues, individuals with plutonium intakes well in excess of typical background intakes would continue to have detectable excretions many decades later. The Air Force declined to recommend an extensive urine resampling program for three primary reasons. First, as noted above, the Air Force believed that dose estimates would likely be favorable to compensation decisions for Palomares responders with primary cancers of the lung, liver, and bone, if they had estimated intake commensurate with the lowest estimated intake of the high 26, i.e., 34 nCi. Second, due to the conservative methods used to estimate plutonium intakes, it was believed that current urine bioassay would not provide any additional benefit to a favorable compensation decision for these three cancers. To the contrary, the results from a current bioassay for individuals with these cancers would more than likely debase the high-sided dose estimates. Lastly, for individuals with cancers not related to deposition and retention in the lung, liver, and bone, there was not any reasonably-expected benefit from a current bioassay. Induction of other cancers are highly unlikely among any of the cohort due to the relatively poor deposition and retention in other tissues, and subsequently small cumulative dose in the organ(s) of interest. Hypothetical intakes would have to be at least a couple of orders of magnitude higher than deemed reasonable for the exposure conditions during the recovery actions to acquire a favorable PoC for many of these cancer types. Intakes of this magnitude, would with little doubt, be detectable by the urinalysis methods available today. Nevertheless, intakes of these magnitudes are deemed implausible and contrary to 38 CFR 3.311 requirement for estimates of probable dose. 38 CFR 3.311 does allow for an independent assessment of dose. Within this provision, it may be appropriate for a veteran to provide an independent assessment of their present day plutonium content in urine excretions. This would allow a veteran to demonstrate an intake of plutonium in excess of that provided by the Air Force.

i) While myeloid forms of leukemia are related to exposures to the red bone marrow, increased risk of these leukemias have not been demonstrated in animal studies with internal plutonium exposures, nor related to internal plutonium exposures in former Soviet Union workers. Many of these workers also had high external radiation exposures in addition to internal plutonium exposures. Lymphomas are also unique. Hodgkin's and chronic sub-types are deemed under 38 CFR 3.311 to be non-radiogenic, though the VA evaluates these on an individual case basis.

Information on unique aspects of assessing dose for lymphoma cases are provided in this report. The only deterministic health conditions recognized in 38 CFR 3.311 are non-malignant thyroid nodular disease and posterior subcapsular cataracts. Neither of these are related to internal plutonium exposures. ICRP Report 103 found little evidence of any excess risk of non-cancer disease below 1 Gy (2,000 rem for α -particle emitting radionuclides), a reiteration of its position in ICRP Report 60. Therefore, while some claimants will have non-radiogenic diseases, there is not a sound scientific and medical foundation for a link to intakes of plutonium for Palomares recovery workers.

Dedication

John C. Taschner, LtCol, USAF, BSC (retired)
Certified Health Physicist

This report is dedicated to the memory of John C. Taschner, to many a good friend and valued health physics colleague. John's leadership of the radioanalytical laboratory team at the US Air Force Radiological Health Laboratory during the Palomares accident provided vital health and environment safety analysis support to the Air Force. The tireless and dedicated work of his team during this accident recovery was responsible for the application of a new urine sample analysis method that provided improved accuracy for assessment of dose to workers. During the accident recovery and after, the laboratory processed nearly 2,000 urine samples. These groundbreaking efforts in the DoD were also critical for the same support during the January 1968 nuclear weapon accident recovery at Thule AB, also an accident involving the dispersal of weapons grade plutonium. John retired from the Air Force in 1974 after serving 21 years. John served nine years in the US Public Health Service as a civilian, as well as nine years in the US Navy, becoming their first civilian Deputy Director for Radiation Safety Programs. Later, John served as a staff health physicist at the Los Alamos National Laboratory, where his wealth of experience was valued as a member of the Hazardous Response Group. John was an atomic veteran, having supported Operation Hardtack II. John had an avid personal and professional interest in nuclear weapons accidents. John and two colleagues researched and co-authored a compendium of nuclear weapons accidents, an important historical document used by the Departments of Energy and Defense. John was a frequent guest lecturer at the Defense Nuclear Weapons School because of his accident response experience and a volunteer docent at the National Atomic Museum in Albuquerque. John was a Health Physics Society Founder's Award recipient and has an annual award in his honor, "John C. Taschner Leadership Award," for senior health physicists with notable contributions to military health physics.

Acknowledgement for Technical Review

Gerald A. Faló, PhD, CHP, Staff Health Physicist within the U.S. Army Public Health Center.

Dr. Faló has been in the Health Physics Division and the Public Health Center since August 1997. Dr. Faló's assignments and projects emphasize occupational and environmental health physics as applied to military deployments. He manages all radiation dose inquiries from DoD personnel exposed to radiation during the Fukushima Daiichi Nuclear Power Station (FDNPS) accident that occurred in 2011. Dr. Faló serves as member of the Operation Tomodachi Registry's Dose Assessment and Recording Working Group, a team of technical experts assessing the radiation dose to DoD military, civilians, and their dependents from the FDNPS accident. Dr. Faló performs radiation dose assessments for Army veterans exposed to radiation from a multitude of sources. He also serves on the MARSSIM and the DOD Ionizing Radiation working groups.

Anthony J. Cagle, LtCol, USAF, BSC, Commander, Bioenvironmental Services Flight, Joint Base Elmendorf-Richardson, AK. LtCol Cagle was a previous Chief of Radiation Health, Office of the Air Force Surgeon General. He has extensive experience managing estimated doses for Department of Veterans' Affairs radiation exposure claims for AF veterans, especially for those pertaining to Palomares veterans. LtCol Cagle also performed dose reconstructions as a staff health physicist at the AF Safety Center.

Kenneth L. Groves. Mr. Groves is a retired CDR, MSC, US Navy with extensive experience in emergency response to radiological incidents, especially those involving nuclear weapons. After 26 years of service in the Navy, Mr. Groves served for 10 years in numerous Environmental, Safety, and Health (ES&H) and Emergency Management and Response (EM&R) programs at Los Alamos National Laboratory (LANL). Mr. Groves retired from the University of California, Office of the President, in 2002 where he served as the Deputy Director of ES&H. He served on the Veterans' Advisory Board on Dose Reconstruction for 10 years and chaired the subcommittee on Communication and Outreach. Mr. Groves is a Fellow of the Health Physics Society.

Alan C. Hale, LtCol, USAF, BSC, CHP, Chief, Radiation Health, Office of the Air Force Surgeon General, is responsible to the AF Surgeon General for management of AF radiation safety programs. LtCol Hale is responsible for managing estimated doses for Department of Veterans' Affairs radiation exposure claims for AF veterans.

Edward F. Maher, ScD, CHP. Dr. Maher is a retired Colonel, USAF, BSC with extensive experience in military health physics and leadership responsibilities over radioanalytical and radiation dosimetry programs. Dr. Maher is a Former Manager, Dose Reconstruction and Estimation Objective, Oak Ridge Associated Universities, National Institutes of Safety and Health Dose Reconstruction Project for the Part B, Energy Employees Occupational Illness Compensation Program Act. Dr. Maher is a past President of the Health Physics Society.

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List of Acronyms

ACS	American Cancer Society
AEC	Atomic Energy Commission
AF	Air Force
AFSEC	Air Force Safety Center
Am	americium
ALL	acute lymphocytic leukemia
AMAD	activity median aerodynamic activity
AML	acute myelocytic leukemia
AMTD	activity median thermodynamic diameter
BB	body burden

BNL	Brookhaven National Laboratory
BOMARC	Boeing Michigan Aeronautical Research Center
BS	bone surfaces
CDE	committed dose equivalent
CED	committed equivalent dose
CFR	Code of Federal Regulations
CIEMAT	Centro de Investigaciones Energéticas Medioambientales y Tecnológicas
CINDY	Code for Internal Dosimetry
CLL	chronic lymphocytic leukemia
DAC	derived airborne concentration
DC	dose conversion
DCF	dose conversion factor
DNA	Defense Nuclear Agency
DoD	Department of Defense
DOE	Department of Energy
DOL	Department of Labor
DTRA	Defense Threat Reduction Agency
DU	depleted uranium
EEOICPA	Energy Employees Occupational Illness Compensation Program Act
EOD	explosives ordnance disposal
ERR	excess relative risk
FDNPS	Fukushima Daiichi Nuclear Power Station
FTA	fission track analysis

GI	gastro-intestinal
HEU	highly enriched uranium
HHS	Health and Human Services
ICRP	International Commission on Radiological Protection
IREP	Interactive RadioEpidemiological Program
LANL	Los Alamos National Laboratory
LN(TH)	thoracic lymph nodes
LUDEP	Lung Dose Evaluation Program
MDA	minimum detectable activity
MPA	Mayak Production Authority
MPBB	maximum permissible body burden
MPOB	maximum permissible organ burden
MPC	maximum permissible concentration
NAS	National Academy of Sciences
NBS	National Bureau of Standards
NCRP	National Council on Radiation Protection and Measurements
NIH	National Institutes of Health
NIOSH	National Institute for Occupational Safety and Health
N-P	naso-pharynx
NRC	Nuclear Regulatory Commission
NTPR	Nuclear Test Personnel Review Program
P	pulmonary

PoC	probability of causation
PHS	Public Health Service
Pu	plutonium
QF	quality factor
RBE	relative biological effectiveness
RBM	red bone marrow
RHL	Radiologic Health Laboratory
SRS	Savannah River Site
T-B	trcheo-bronchial
TEDE	total effective dose equivalent
Th	thorium
U	uranium
UK	United Kingdom
VA	Veterans Administration
WGP	weapons grade plutonium

Topical Issues for Assessment of Dose to Palomares Accident Recovery Workers (1966)

1.0 Introduction.

On 17 January 1966, a B-52 and KC-135 collided during a mid-air refueling operation over southern Spain. In addition to the mid-air explosion of the KC-135 aircraft, the B-52 suffered damage that caused the plane to break-up mid-air, with the jettison of four nuclear weapons. Two of the four weapons detonated upon impact, which with the influence of winds at the time caused distribution of weapons grade plutonium (WGP), and highly enriched and depleted uranium (HEU and DU, respectively) to the environment. The other two weapons were recovered intact – one from land and the other from the Mediterranean Sea. The majority of the residual radiological contaminants were retained in surface soils in the small rural Village of Palomares¹. All four of the KC-135 crew died in the accident, while four of the seven crew of the B-52 survived. A small Air Force (AF) team departed from Torrejon AB, Spain, within about two hours of notification of the accident. By the evening, 49 US personnel had arrived at the accident site, with over 650 personnel within a few days (DNA 1975). The large majority of military personnel supporting the land-based recovery operation were AF, primarily from two Spanish bases: Torrejon and Moron. Over the course of the recovery operation, 17 January to 7 April 1966, about 1,600 personnel supported the land portion of work. Its notable that most Navy personnel supporting recovery operations were assigned duties on Naval vessels supporting the sea search for the unrecovered weapon, sea-based logistics support for the operation, and ocean transport of soil and debris back to the US. Figures A-1 and A-2 provide maps of the Kingdom of Spain and the Province of Almeria where the Village of Palomares is shown.

In 2000, based upon veteran inquiries of potential health effects from their support to the on-site recovery operations, the AF Surgeon General's Office (AF/SG) contracted with Labat-Anderson, Inc. to assess radiation exposure potential. Inquiries by veterans are most commonly initially managed by the Veterans Administration. Labat-Anderson's assessment was dominated by the review and analysis of urine sampling results, as this monitoring method was conducted extensively for recovery workers to assess the potential and extent of WGP inhalation. Labat-Anderson also provided some review of "environmental-based" indices of exposure potential. The findings of their work was published the next year (Labat-Anderson 2001).

Since then, the Air Force has used exposure information from this report and other sets of environmental data to assess exposure potential for recovery worker claims managed through the VA. Over the years, the AF has been scrutinized for the approach used in estimates and/or the outcome of a VA decision. To date, the AF/SG has received about 30 claims from AF Palomares recovery workers. This report describes important factors used by the VA in adjudication of these claims. As discussed in detail later in the document, the three key factors are: estimate of probable dose, radiogenic disease assessment, and probability of causation (PoC) assessment. Because urine

¹ The accident is most commonly referred to as the Palomares Nuclear Weapons Accident.

samples analysis was a primary source of exposure assessment, individual and population urine sample results remain a foundation for assessment of exposure potential, though environmental data is an important source of supporting information, as is monitoring accomplished on Palomares residents. Because WGP causes radiation exposures to individuals by internal distribution and metabolism, International Commission on Radiological Protection (ICRP) models are described in detail, as applicable to plutonium exposure assessments. There have been five distinct sets of ICRP recommendations for plutonium since 1959. Figure ES provides a graphical depiction of the inter-related factors that are key to adjudication of health claims for Palomares recovery workers. Due to the large number of factors, this report is written in a topical manner. These factors considered as a whole support the conclusion that estimates of probable dose are high-sided. When merged with the PoC assessment method, and weighted against sound scientific and medical evidence, a high-degree of “claimant-favorability” is afforded to recovery worker veterans.

2.0 Radiological Monitoring.

Radiological monitoring during the accident recovery was conducted for three primary purposes: determine the extent and magnitude of the contamination on land areas, assess environmental impact (vegetation and water), and personnel safety (urine, nasal swabs, external dosimetry, and air samples). Because WGP dispersed to the environment is primarily an internal exposure hazard, inhalation was the key exposure pathway of concern. For recovery workers, this was due to resuspension of ground-deposited contamination.

Air sampling is an important method to assess exposure potential through the inhalation pathway. Other methods include the assessment of the amounts of plutonium in urine and fecal excretions from individuals with internal burdens of plutonium. For these individuals, small amounts of plutonium would be excreted over time in both urine and feces. Predictive excretion models were not well developed for inhalation exposures at the time of the accident. In the decades since, refinements in excretion models have been promulgated by the ICRP. These predictive models have been developed from plutonium exposure studies in animal models, plutonium worker studies, and importantly with augmentation of data from autopsy studies of plutonium workers. Thirteen-hundred seventy members of the recovery operation provided initial urine samples while on-site (DNA 1975), though a total of 1,571 urine samples were analyzed (Oldand 1966), which would provide for duplicates for some personnel. As will be discussed later, a large number of these personnel provided additional samples after they returned to their base of assignment, some through 1967 (Odland 1968). Four-hundred thirty-nine air samples were collected during recovery operations, as reported by DNA (1975). The majority of the air sampling was conducted during those operations generating the greatest exposure potential, e.g., soil scraping, plowing. Air samples were analyzed for gross α -radiation, with average sample results below 2 pCi m^{-3} ($\mu\text{Ci cm}^{-3}$), the maximum permissible concentration (MPC) established for the plutonium contaminant by the Air Force. ICRP Report 2 (ICRP 1959) provided MPC values for two classes of compounds: soluble and insoluble. Though WGP involved in detonation events is expected to be dominated by dioxide forms (insoluble), the more conservative MPC, applicable to soluble forms was used as a reference value. Figure 1 shows the changes in ICRP MPC and derived airborne concentration (DAC) values for ^{239}Pu over a series of ICRP publications. Since recommendations from ICRP Report 141 (ICRP

2019) have yet been promulgated in a regulatory form, DAC values are not available. Inhalation Class Y under ICRP Report 30 and Type S under ICRP 60/68 was similar to the insoluble form under ICRP Report 2. Inhalation Class W (ICRP Report 30) and Type M (ICRP Report 60/68) refer to moderately soluble forms. ICRP 2 did not have an equivalent solubility class.

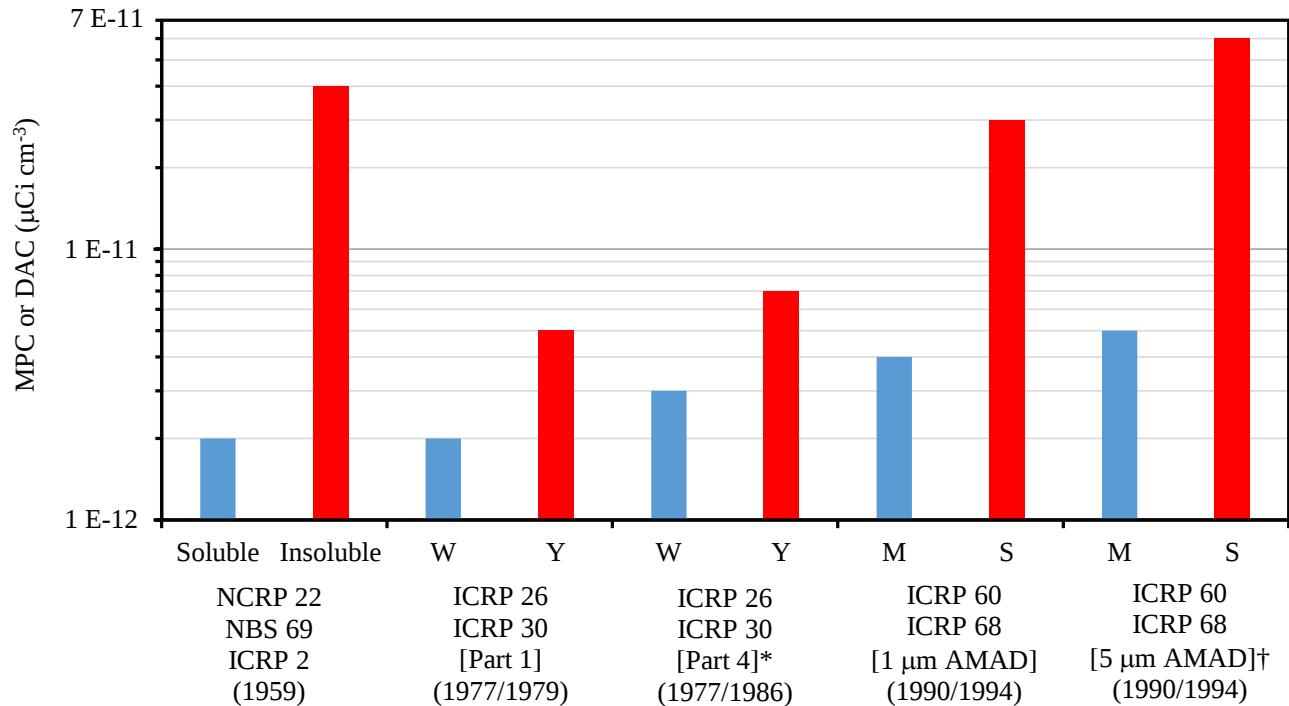


Figure 1. ICRP MPC or DAC Values for ²³⁹Pu for Occupational Exposures.

[* Updated from ICRP 48, †5 μm activity median aerodynamic diameter (AMAD) deemed more appropriate for occupational exposures]. Figure 3 from Rademacher (2019).

The radiological impact to the environment from the Palomares accident involved HEU, DU, and WGP. Due to the relative differences in specific activity of the contaminants, WGP provides the greatest activity. Figure 2 shows the relative specific activity among fissionable materials used in nuclear weapons. For each of the contaminants, the primary internal radiological hazard is due to the α -particle emission. The relative α -particle activity of the combined contaminants is dependent on the relative mass of each material in the weapons that detonated. This information remains classified. The Boeing Michigan Aeronautical Research Center (BOMARC) accident occurred in 1960 and also dispersed WGP, HEU, and DU to the environment. Environmental samples used in support of restoration at the BOMARC accident site provided a ²³⁹⁺²⁴⁰Pu to ²³⁴⁺²³⁵⁺²³⁸U activity concentration ratio estimate of 469 (Rademacher *et al.* 2009). The ratio, as applied to the contaminants at Palomares are expected to be of similar magnitude, though a precise value is classified. Odland *et al.* (1968) reported the presence of ²³⁵U in soil samples analyzed by RHL during the recovery. Sancho *et al.* (2019) also reported the mixed presence of Pu and U in discrete particles from the site. Due to the relatively small uranium activity in the contaminant, these are

deemed negligible compared to the $^{239+240}\text{Pu}$. Hence, radiological health hazards have been assessed for $^{239+240}\text{Pu}$. Due to the similar α -particle energy of these two isotopes of plutonium, they are inseparable by α -spectrometry analyses and carry the same internal radiological health hazard.

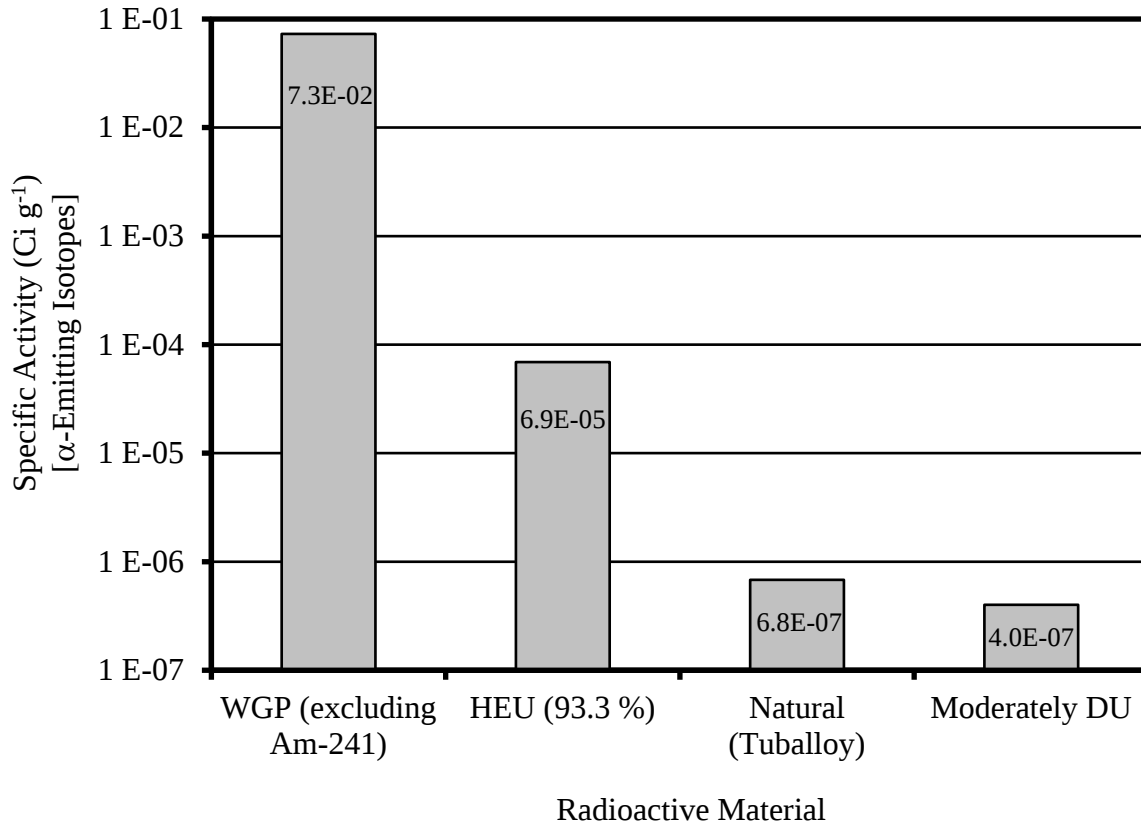


Figure 2. Comparison of Specific Activity of α -Emitting Radionuclides in Radioactive Material Potentially Used in Nuclear Weapons. [Figure 3-1 from Rademacher (2016)].

In their study of Palomares resident exposures to plutonium, the Spanish collected urine samples in a manner similar to that conducted for US military personnel that supported the recovery. The Spanish also conducted direct external measurements over the chest of individuals that were part of their medical follow-up. The detectors used for these measurements cover the lung tissue and other associated lung tissues, e.g., lymph nodes.

3.0 Urine Bioassay Methods.

Assessment of radioactive material content in urine samples is varied dependent on the radiation emissions of the target radionuclides, radioactive emissions of radionuclides in the environment that are expected to be excreted in urine due to routine intakes by individuals, and sensitivity requirements. Some radionuclides can be assessed alternatively by non-radiological methods, e.g., mass spectrometry. WGP contains predominantly by mass ^{239}Pu , with the majority

remaining balance to ^{240}Pu , trace amounts of ^{241}Pu , ^{242}Pu , ^{238}Pu , ^{241}Am , and possibly inert metals for alloy stabilization. Example amounts for the BOMARC accident site are shown in Table A-1. The WGP involved in the Palomares accident are expected to be somewhat similar to that from the BOMARC accident.² With the exception of ^{241}Am : ^{239}Pu , ^{240}Pu , and ^{238}Pu have negligible photon emissions (see Table A-2). For this reason, urine bioassay has commonly been accomplished by assessment of α -particle emissions. For some assessment of ^{239}Pu , ^{240}Pu , and ^{238}Pu in WGP, a relationship between specific isotopes of plutonium and ^{241}Am can be used.³ This is common for assessment of soil and is a reasonable assumption for relatively immobile forms because they remain comingled. The relationships among these radionuclides change over time, as illustrated in Figure A-3.

Within the last few decades, more sensitive methods have been developed. Mass spectrometry has been used extensively by Los Alamos National Laboratory (LANL) and by the Savannah River Site (SRS) for assessment of urine bioassay samples. A fission track analysis (FTA) is another sensitive method that was developed by Brookhaven National Laboratory (BNL). More discussion of urine bioassay methods is based on the plot contained in Figure F-1.

4.0 Urine Bioassay Methods used by the Air Force in 1966 – 1967.

4.1 General.

The AF's Radiological Health Laboratory (RHL), Wright-Patterson AFB, OH, analyzed initial urine samples by a gross α -particle emission screening method (Odland 1966). At the time of the accident, the RHL did not have a plutonium-specific method. As part of the response to the accident, RHL staff developed a method based on consultation with staff from Los Alamos Scientific Laboratory, NM (Taschner 1999). The new method included a plutonium-specific chemical separation and isotopic-specific analysis by α -spectrometry (Odland 1966). The gross α -particle screening method used a gas-flow proportional counter for sample assessments, while the isotopic-specific plutonium analysis used solid-state surface barrier detectors (Odland 1966). The initial urine sample results are summarized in Table 1 by service affiliation and an estimate of the body burden using the Langham system excretion model of that time (Langham 1959)⁴:

$$D = 435Ut^{0.76},$$

where D is the initial systemic body burden (e.g., excludes lung and its associated tissues), U is the amount in a single day urine excretion, and t is the number of days following an acute intake.

² Actual WGP characteristics for individual stockpiled US weapons are classified. The AF Safety Center has not reviewed US estimates for the Palomares site. However, the Safety Center has reviewed estimates of the $^{239+240}\text{Pu}$ to ^{241}Am ratio for soils at the site, as analyzed by Spanish scientists (Sancho et al. 2019; Sancho-Llerandi 2011). The ratios are similar. Chamizo *et al.* (2006) assessed the ^{240}Pu to ^{239}Pu ratio at $6.57 \pm 0.06\%$, which is only 9% higher than the estimated ratio of BOMARC WGP contaminants.

³ This is common for assessment of WGP in soil and is a reasonable assumption for relatively immobile forms.

⁴ The expression was modified over time – some power function exponents have also been listed as 0.77 & 0.78. A 1980 version is described in Langham *et al.* (1980).

Figures A-4 and A-5 illustrate this relationship. The plot in the figure is based on an initial systemic body burden of 0.04 μCi ^{239}Pu . As noted in footnote 5, RHL used a value of 0.044 μCi . The source of the small 10% discrepancy with the ICRP 2 value is not known.

4.2 Initial Screening Urine Samples.

The sampling data in Table 1, is based on a summary by Odland (1966), with updates provided by Odland (1968) in a later published report. The latter data set contained 15 more samples for AF personnel than the early dataset. All were for samples that provided a predicted initial systemic body burden (BB) between 0.09 and 0.99 of the maximum permissible under ICRP 2. Overall, AF personnel submitted the vast majority of samples. Only one of the 20 samples that was predictive of an initial body burden greater than one BB (see Table 1) was for a non-AF individual. The 20 samples, however, represented only 1.3% of the total.

With respect to the initial urine samples, it is important to understand that the majority of these samples were submitted by individuals within two to three weeks after initiation of their on-site work. With the exception of a small number of urine samples collected from individuals that were on-site in the early days of the accident recovery response, samples were collected at the end of an individual's deployment. In an effort to reduce the burden of the deployment, 16th AF planned on individuals rotating after a 21-day period (Air Force 1968). Many personnel volunteered for longer deployments during the 11-week recovery action. Estimates of BB were based on an acute intake on the middle date of the exposure period. Due to the highly varied daily urinary excretion rates within short periods after an acute intake, there is an expectation for much greater uncertainty in the estimate of BB than for urine samples collected at longer periods after the exposure period, according to the Langham model. Table 2 illustrates the predicted daily excretion based on the Langham model for a uniform daily intake over exposure durations of 7, 14, 21, and 28 days, and for the cases of acute intakes on the first day of exposure, the last day of exposure, or the middle of the exposure period. There are number of factors impacting which approach is most appropriate for

TABLE 1. Initial Urine Samples, Alpha Activity. Values from Odland (1966) [Values in Parenthesis from Odland et al. (1968), Only Listed if Different from 1966 Data].

Affiliation	Air Force	Army	Navy	Other	Total
Number Analyzed	1389 (1404)	107	37	38	1571 (1586)
BB* > 1 (100%)	19	1	0	0	20
BB: 0.09 – 0.99	361 (375)	33	5	8	407 (422**)
BB: 0.009 – 0.09	487	23	20	7	537
BB: < 0.009	522	50	12	23	607

* Systemic body burden (e.g., excludes lung and its associated tissues), value of 0.044 μCi ^{239}Pu for D represents one body burden or 100%⁵

** Should be 421, or one sub-category increased by one.

⁵ ICRP Report 2 lists the Maximum Permissible Body Burden (MPBB) of 0.04 μCi (40 nCi), however, for inhalation of soluble forms of plutonium.

estimation of body burden to be discussed latter. For the exposure examples where urine samples were collected immediately at the end of the exposure period, the range of predicted daily excretion values were the greatest as a group, but greatest for the example exposure duration of 28 days. For urine sample collections at 50 and 100 days post exposure, the differences in predicted urine excretion among the different intake models are progressively much lower. Therefore, sampling conducted shortly after an exposure provides superior sensitivity for detection of an intake, but at a compromise to accuracy in the predicted intake. The latter is due to unknown temporal characteristics of the intake for each worker.

TABLE 2. Urine Excretion from Plutonium Intakes for Varied Exposures and Intake Models, Langham Urine Excretion Model (Langham 1959) for One BB, 0.04 μCi $^{239+240}\text{Pu}$ Systemic Intake.

Exposure Duration (days)	Predicted Daily Excretion (pCi d^{-1})				Post Exposure Sample Collection (days)
	Uniform Daily Intake	Acute Intake (First Day)	Acute Intake (Last Day)	Middle of Exposure Period	
7	41.4	21.0	92.0	32.1	0
14	28.5	12.4	92.0	19.9	0
21	22.5	9.1	92.0	14.9	0
28	18.9	7.3	92.0	12.1	0
7	4.4	4.3	4.7	4.4	50
14	4.2	4.0	4.7	4.2	50
21	4.1	3.6	4.7	4.0	50
28	3.9	3.4	4.7	3.8	50
7	2.7	2.6	2.8	2.7	100
14	2.6	2.5	2.8	2.6	100
21	2.6	2.4	2.8	2.6	100
28	2.5	2.3	2.8	2.5	100

4.3 Factors Affecting Intakes.

There are a number of important factors impacting intakes of plutonium for recovery workers. The primary factors are the amount of time each individual was involved in the recovery and the types of duties each individual performed. Some individuals performed duties within the contamination zone, while others had duties supporting the base camp and logistics. In addition, the degree of contamination at locations within the contamination zone was highly varied, as shown in Figure A-6. This plot was prepared from initial 16th Air Force ground surveys conducted early in the recovery. Work conducted in the contamination zone was primarily comprised of survey work (detection), searches for weapon debris and the one unrecovered weapon⁶, decontamination, crop harvesting, soil scraping, and soil plowing. Table A-3a provides a summary of personnel and

⁶ The fourth weapon was eventually located and recovered off the coast in Mediterranean waters.

functions two weeks into the recovery, with weekly numbers for the course of the recovery in Figure A-3b. Soil scraping occurred only within the most highly contaminated area $> 1,200 \text{ kBq m}^{-2}$ ($32 \text{ } \mu\text{Ci m}^{-2}$), which was about 10- to 100-fold higher in surface concentration levels than the middle two contamination contours of Figure A-6. The most highly contaminated area subject to soil scraping encompassed about 5.5 acres (DNA 1975). The type of work being conducted is also important to the degree of contamination resuspension available for inhalation intakes. For dose assessments to personnel supporting the restoration of radiological contamination on Enewetak Atoll, conservative airborne resuspension (mass loading approach) used values of 100, 300, and $600 \text{ } \mu\text{g m}^{-3}$, for quiescent conditions where soil is not purposely disturbed by mechanical operations, e.g., during vegetation grubbing and soil handling activities, and soil excision, respectively (Rademacher 2019). Additionally, as practiced during soil removal activities at Enewetak Atoll, for soil plowing activities and soil scraping⁷ during the Palomares recovery, the soils were heavily watered down an hour prior to the work. This action was enforced to, “prevent resuspension of active material in dust clouds” (Air Force 1968). For soils transport and loading operations, piles of soil were wetted prior to and during active loading (Air Force 1968). Because of this mitigating action, resuspension of contamination may have been less than resuspension under even quiescent conditions.

Another factor affecting resuspension of airborne contaminants is the time between the initial deposition and period of occupancy. Figures A-7 and A-8 provide plots of modeled temporal variance in ground-deposited radiological material resuspension from a number of authors. The first plot is for periods up to 40 years, while the second is for periods up to 20 weeks (140 days) and is more visually useful for application to the exposures under consideration of this report. A key characteristic among the models is the rapid decrease in the initial periods after deposition. The Langham model (Langham 1966, 1969) was based specifically on airborne sampling shortly after conventional explosives detonation tests on nuclear weapons, which is pertinent to the type of dispersion experienced at Palomares. The Langham and Kathren models only covered short periods of monitoring after initial deposition and inference of long-term applicability is inappropriate. The model by Maxwell and Anspaugh. (2011) is a more accurate version of the 2002 Anspaugh model (Anspaugh *et al.* 2002). This model provides an upper and lower bound to account for variability in environmental conditions and contaminants. Dioxides of plutonium dispersed from accidents of this type are expected to have a considerable degree of heterogeneity, due to a large fraction observed in discrete particle form (Rademacher 2016; Iranzo *et al.* 1994 & 1998). Iranzo *et al.* (1998) noted,

“Resuspension factors were calculated from average annual measurements of Pu in soil and air. The data indicate that the resuspension factor decreased exponentially with time from an initial annual average value of about 10^{-7} m^{-1} to around 10^{-9} m^{-1} some months later and to values on the order of 10^{-11} m^{-1} some years later.”

Iranzo *et al.* (1994) noted that the resuspension factors were lowest in air monitoring locations closest to weapon impact points, while they were higher in areas downwind where surface soil

⁷ The term “scraping” was used for soil excavations of the most highly contaminated soils. Scraping was deemed a more appropriate term because only two inches of surface soils were targeted for removal.

contamination levels were significantly lower. This is a logical finding. Aerodynamically larger particles will have greater deposition from the debris cloud in proximity to the impact point, with the finer, aerodynamically smaller particles having greater transport range from the impact point. Subsequently, the aerosols associated with contamination near the impact points will have a greater non-resuspendable fraction than the aerosol associated with the diffuse contamination a greater distance from impact points.

The 2002 Anspaugh and 2011 Maxwell and Anspaugh models provided a central estimate with respective bounding by factors of $10^{\pm 1}$ and $4.2^{\pm 1}$, respectively. The upper and lower bounds of the 2002 Anspaugh model is displayed, while only the central estimate of the Maxwell model. Based on actual site data collected by the Spanish (Iranzo *et al.* 1988), the plutonium resuspension near impact points follows the lower-bound of the 2002 Anspaugh model with an initial resuspension factor near 10^{-6} m^{-1} . This model also provided for an approximate decrease of ten-fold over the first 32 days.

Respiratory protection use also affects intakes. Although paper dust masks were commonly worn by many personnel during the Palomares recovery, the air-purifying respirators were used for some operations and in the early days of the response by the explosive ordnance disposal (EOD) team that worked within the vicinity of the detonation points of the two weapons. The M-17 full-face mask was used by EOD personnel at that time. For soil loading operations, the front loader operator was required to wear an M-17 respirator, while unnecessary personnel were restricted from access to areas within 500 feet of the operation, support personnel were located upwind of the soil loading location, and the dump truck vehicle was required to have its windows closed during any soil transfer operations. These measures would have varying impacts on limiting respiratory intakes. While full-face respirators are commonly assigned a respiratory protection factor of 50, paper dust masks may only provide a protection factor of perhaps two- to four-fold. The Occupational Safety and Health Administration recognizes dust mask use under 29 CFR 1910.134(b). Among air-purifying respirator types are the least protective. Similarly, maintaining a closed-cab environment for dump truck drivers will not provide the same protection level as a full-face respirator, but may be a reasonable measure for low hazard environments.

4.4 Interpretation of Initial Screening Urine Samples.

Early in the recovery operation, after review of site data, RHL became concerned that urine samples were compromised by contamination from the site (Odland *et al.* 1968; Air Force 1968). This initial concern was based on the screening of exterior surfaces of urine sample containers, where 15% of exterior surfaces of samples were well over background levels (Air Force 1968). The potential for this condition was reasonable considering the fact that site workers were expected to collect a sample over a twenty-four hour period, which would necessitate them carrying the sampling container throughout the day, to include the contaminated zone. In the early period of response, appropriate sampling containers were lacking which was favorable to cross-contamination.

Radioactive materials within a urine sample that are not due to plutonium incorporated into systemic circulation from an intake due to either inhalation or ingestion source will cause sample

results to be high-biased, as the gross α -radiation laboratory method is not capable of discrimination of the source of α -emitting radioactive material. There are two primary sources of high-bias for the initial urine samples: Pu at the site inadvertently contaminating samples, and natural background radioactive materials that are part of normal dietary intakes and urinary excretion.

To appreciate the potential for Pu dispersed at the site to inadvertently contaminate urine samples, it is useful to evaluate contamination screening levels established for the recovery. Personnel working in the contamination zone were screened for surface contamination with a PAC-1S, portable α -particle scintillator (DNA 1975). Some locations within the most heavily contaminated areas had soil surface concentrations exceeding 2,000,000 counts per minute (cpm), the maximum reading for this detector (DNA 1975). Individuals were screened to surface contamination levels of 200 cpm⁸, which assuming a 4π detection efficiency of 0.3 is equivalent to 41 pCi (1.5 Bq). Vegetation was screened to a surface contamination level of 400 cpm (82 pCi). Based on Figures A-9 and A-10, a 41 pCi ²³⁹⁺²⁴⁰PuO₂ particle can have a diameter less than 5 μ m.

In review of Figures A-4 and A-5, it is readily apparent that single particles with activity of these magnitudes can readily affect the interpreted activity in daily urine excretion for individuals. For an individual with a systemic BB one-half the ICRP MPBB for ²³⁹Pu, 0.02 μ Ci, with 14 days of exposure with the sample collected at the end of the exposure period and an assumed exposure in the middle of the exposure period would have a predicted urine excretion of about 10 pCi (see Table 2). The addition of 41 pCi of contamination would high-bias the predicted systemic BB by five-fold. An activity of 82 pCi inadvertently contaminating the same daily urine sample would high-bias the result by nine-fold. Cross-contamination of urine samples collected from workers while on-site could have much higher predicted body burdens than shown by these examples, because of the levels of contamination that existed on the site.

The concern for plutonium cross-contamination of urine samples extends beyond initial screening urine samples collected and analyzed by the Air Force in 1966. The Spanish Government between June and October 1966 analyzed urine samples submitted by 59 Palomares residents with the greatest risk of exposure (Iranzo *et al.* 1998). On the day of the accident, 485 people were present in Palomares (Church *et al.* 2000). While the samples were analyzed at a Spanish Government facility in Madrid, the residents collected and submitted the urine samples from Palomares. The results of the analysis suggested possible sample contamination problems (Iranzo *et al.* 1998). With use of the Langham excretion model, the magnitude of potential bias from cross-contamination of these may be higher than the case of the initial samples collected by the AF for its personnel. This is due to the much lower fraction of initial systemic body burden predicted in daily excretion. Figure A-11 provides these fractions for up to 360 days after the intake. For example, at seven days since an acute intake, the fraction is 0.00052, while at 180 days it is 0.000044, about 12-fold lower.

⁸ The PAC-1S has an entrance window of 60 cm².

Due to the concern for cross-contamination, follow-on urine sampling of the Palomares residents required them to travel to Madrid to conduct urine sampling (Iranzo *et al.* 1998). The resampling confirmed some cross-contamination. It was a standard practice in the Department of Energy (DOE) Laboratories and their predecessor entities under the Atomic Energy Commission (AEC) to isolate plutonium workers when they submitted periodic urine samples. BNL in their assessment of potential radiological exposures to the Enewetak people adopted a similar precaution of having individuals submit urine samples on a ship anchored off-shore (Sun *et al.* 1997). The procedure involved 1) controlled handling of sample collection bottles, 2) showering and donning of clean clothes prior to sample collection, and 3) the collection of urine samples while aboard the ship.

The other potential factor responsible for high-bias in initial screening urine samples is naturally-occurring radioactive materials in diet and subsequently in urine excretions of individuals. During the recovery operation, very little published data existed on these natural background sources. In the 1960's, due to world-wide distribution of fallout from atmospheric tests of nuclear weapons and the expansion of nuclear power, greater research efforts were initiated. ICRP Report 23 (ICRP 1975) provides a summary of dietary (including drinking water) intakes and excretions (primarily in urine and feces). Table 3 provides estimates of intake and excretion of naturally-occurring α -particle emitters from ICRP Report 23. Among naturally-occurring radioactive materials, α -particle emissions from ^{238}U and its decay chain, ^{232}Th and its decay chain are most prominent. The estimates of α -particle activity for the thorium and uranium categories only include isotopes in the respective chains, e.g., uranium ($^{238}\text{U} + ^{234}\text{U}$) and thorium ($^{232}\text{Th} + ^{228}\text{Th}$). Alpha emitters in the decay chains of other elements are also expected to be part of intakes and excretions in both urine and feces. Due to the relative immobility of uranium and thorium in biological systems, the greatest portion of the intake in food and fluids is excreted directly into feces without systemic absorption. Among the three categories, the greatest average activity expected in urinary excretion is $^{238}\text{U} + ^{234}\text{U}$.

Figure A-12 and A-13 contain scatterplots of daily urine excretion for a cohort of AF workers analyzed for isotopic uranium. Among the 131 samples, the vast majority are encompassed by the upper-bound value of 0.34 pCi d^{-1} , listed in Table 3. One individual provided a sample with an estimated daily excretion of $^{238}\text{U} + ^{234}\text{U}$ about 2.1 pCi , which demonstrates the potential for some individuals to have unusually high excretion rates compared to a population average. Figure A-13 shows the high degree of variability among those with excretions more representative of ICRP 23 values for uranium. The effect of natural α -particle emitters in urine on the estimates of plutonium intake from initial screening samples is expected to be less than that from plutonium cross-contamination accidentally introduced into samples. Similar to the case for plutonium cross-contamination, the net result will be biased-high estimates of intakes. The effect for any individual sample is not known, however, for the gross α -particle analysis method used for initial screening samples, there will be some inherent high bias. At a minimum, it is a source of positive α -particle detection in urine sample devoid of plutonium from a systemic body burden or cross-contamination.

TABLE 3. Estimates of Intake and Excretion of Naturally-Occurring α -Particle Emitters for Reference Man, ICRP Report 23 (ICRP 23).

Parameter	Units	Radioactive Material		
		Uranium*	Thorium**	Ra-226***
Intakes (food and fluids)	$\mu\text{g d}^{-1}$	1.9	3	2.3×10^{-6}
	pCi d^{-1}	1.3	$0.66^{\S}$	2.3
Intake (Airborne)	$\mu\text{g d}^{-1}$	0.007		
	pCi d^{-1}	0.005		
Urinary Excretion	$\mu\text{g d}^{-1}$	0.05 – 0.5	0.1	8.0×10^{-8}
	pCi d^{-1}	0.034 – 0.34	$0.022^{\S}$	0.08
Fecal Excretion	$\mu\text{g d}^{-1}$	1.4 – 1.8	2.9	2.2×10^{-6}
	pCi d^{-1}	0.95 – 1.2	0.63	2.2

* Naturally-occurring uranium in the environment is comprised of 99.28% ^{238}U , by mass; in soils ^{238}U and ^{234}U are in equilibrium on an activity basis. ** Naturally-occurring thorium in the environment is comprised of 99.98% ^{232}Th , by mass; in soils ^{232}Th and ^{228}Th are in equilibrium on an activity basis. ^{230}Th is in the ^{238}U decay chain with a natural mass abundance of about 0.02%. $\S$ Estimate excludes ^{230}Th . *** ^{226}Ra is in the ^{238}U decay chain, but due to its relatively long radiological half-life of 1,600 y, it and its decay progeny are often treated separately.

TABLE 4. Sources of α -Particle Emitters in Initial Urine Samples, as Analyzed by Gross α -Particle Analysis.

Source	Effect on Predicted Systemic BB of Plutonium for Different Pu Exposure Potential	Discussion Points
Systemic plutonium excreted in urine	Neutral	Sample collected early have best detection efficiency, while samples collected many months after an exposure provide a more precise estimate of systemic body burden
External plutonium cross-contamination	High-Bias	Individuals with highest systemic exposure expected to have greater potential for urine sample cross-contamination
Naturally-occurring radioactive materials in urine	High-Bias	Activity level independent (neutral) to Pu exposure from Palomares work

4.5 Follow-on (Resampling) Urine Samples from Recovery Workers

A follow-on sampling program using an isotopic plutonium analysis method was instituted to better assess potential intakes among individuals with a predicted BB 10% or greater based on initial screening samples that were collected on-site. New urine samples were collected from individuals during a period of 90 to 150 days after collection of the initial sample (Odland *et al.* 1968). RHL

implemented this approach believing these individuals represented those workers with the highest plutonium inhalation potential among recovery workers. As noted above, these sample were suspected to have some degree of plutonium cross-contamination because they were collected while workers were on-site and high-bias from naturally-occurring radiative materials from routine dietary intakes. Table 4 lists the effects and discussion on the effects of these sources of bias to initial urine samples. Plutonium cross-contamination and naturally-occurring radioactive materials in urine are expected to high-bias predicted system BB of plutonium. Due to this potential, it is important to understand how this high-bias is apportioned among the sample from individuals based on their potential for inhalation exposure from plutonium. This is an important factor impacting the RHL's decision to resample those with the highest predicted BB from initial urine samples. It was deemed an appropriate sampling approach to omit workers with less than 10% of the BB, as the resampling effort was designed to add a greater degree of validity and fidelity to the initial screening results. As noted in Table 4, an individual's naturally-occurring radioactive materials in urine are expected to be independent (neutral) to plutonium exposures. In the case of the external cross-contamination of samples by plutonium, those individuals with greater inhalation exposure potential were deemed more aligned with greater potential for cross-contamination of their urine due to their work activities. In some cases, the opposite is possible, but unlikely. Overall, for the cohort of workers, this factor is expected to be neutral as a minimum to affecting samples to a greater degree from those workers with greater inhalation exposure potential.

Table 5 contains a summary of results from the urine resampling effort. The cohort contained samples from 422 individuals, though RHL requested follow-on samples from 409 individuals that submitted initial samples. A small number of resamples arrived at RHL in a leaking condition, which required a request for a replacement sample (Odland *et al.* 1968). Additionally, some of the samples encompassed within the resampling program included individuals that did not submit an initial sample (Odland *et al.* 1968). The resampling program included a little over 25% of the total recovery operation workforce. As discussed above, with the exception of samples included in this group as submitted by individuals that failed to submit an initial sample, this subgroup of workers is expected to be biased toward those that had the greatest exposure potential. In review of the data in Table 5, the resampling program yielded only six individuals (1.4%) with a predicted

TABLE 5. Resampling Program Urine Samples, Isotopic Plutonium. Values from Odland (1966) [Values in Parenthesis from Odland et al. (1968), Only Listed if Different from 1966 Data].

Affiliation	Air Force	Army	Navy	Other	Total
Number Analyzed	328 (375)	30 (33)	8 (7)	7	373 (422)
BB* > 10%	6	0	0	0	6
BB: 1 – 10%	162 (195)	10 (13)	5	0	177 (213)
BB: < 1 %	36 (26)	11	1	1	49 (39)
BB: zero (non-detects)	124 (148)	9	1	6	141 (164)
No. requested analysis	363	33	5	8	409

* Systemic body burden (e.g., excludes lung and its associated tissues), value of 0.044 μCi ^{239}Pu for D represents one body burden or 100%

initial systemic body burden greater than 10% of the MPBB, with 39% of the samples being non-detects (listed as “zero”) and 50% with body burdens between 1 and 10%. The resampling program contained similarly-proportioned distribution among the DoD services and other personnel, as compared to initial samples. Air Force members were about 89% for each dataset. Nevertheless, among individuals with predicted initial systemic body burden greater than 10% of the MPBB, all six were Air Force members, with the highest at 67% of the MPBB.

Figure 3 contains a histogram of predicted initial systemic body burden for the cohort of individuals that provided initial screening urine samples while on-site and for the subset of workers that participated in the resampling program. The bar graph is not completely accurate, as the data in Tables 1 and 5 have slight differences in MPBB bins. For example, Table 1 bins those with 9 to 99% of the MPBB, while Table 5 bins those greater than 10%. Therefore, the bin, 9 to 99%, for resampling should be a little higher, while 0.9 to 9% a little lower.

Table 6 provides the statistical review of resampling for those individuals with predicted initial systemic body burden equal to or greater than 1% of the MPBB. This table contains values from Odland (1966) and 1968 updates (Odland *et al.* 1968). While the resampling program was conducted by RHL between 90 and 150 days after collection of initial samples, some individuals did not submit samples until much later – the greatest lapse between the end of exposure and sample submission was 396 days, though the median and mode were both 140 days. The highest activity in a daily excretion was 1.03 pCi $^{239+240}\text{Pu}$, though the most probable daily excretion was 0.029 pCi and the median was 0.066 pCi.

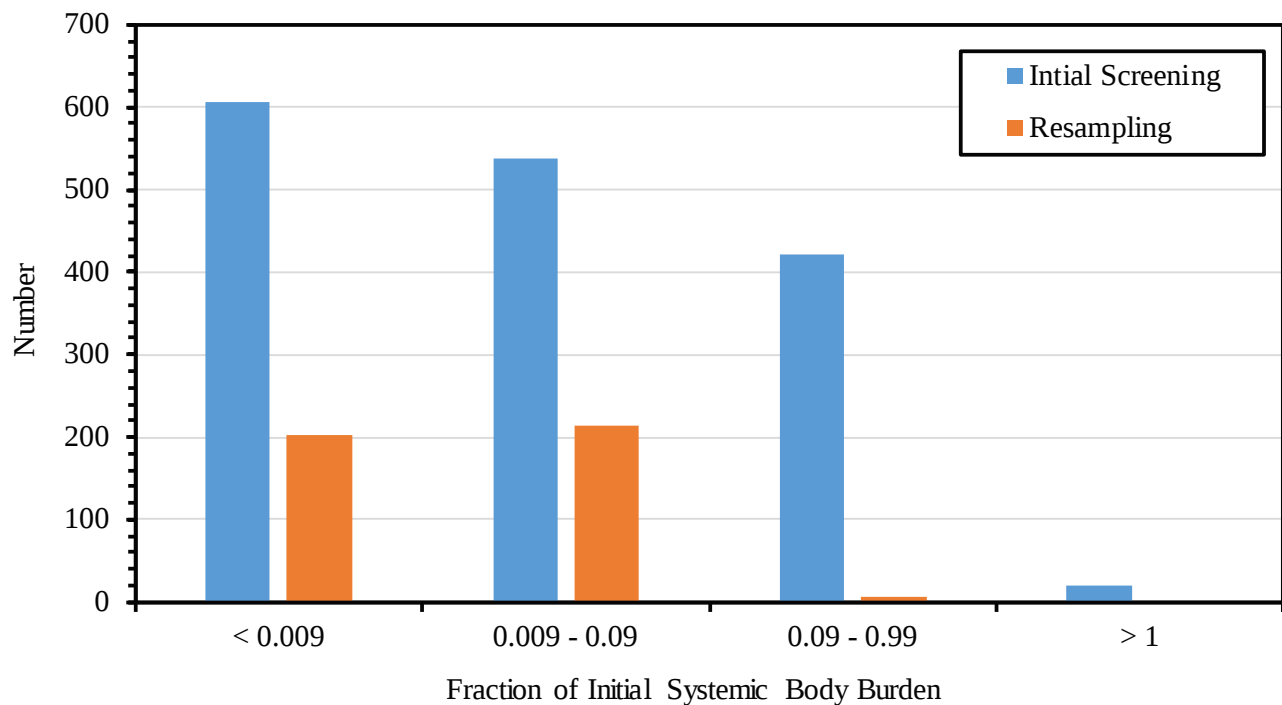


Figure 3. Distribution of Fraction Initial Systemic Body Burden based on Initial Screening and Resamples of Urine [Data from Tables 1 and 5].

RHL recommended additional follow-up on 25 individuals, which had between 7 and 67% of the MPBB. Another service member was added to this group to make the total 26. These individuals were initially requested to submit a total of six additional samples, provided at 2-month intervals over one year (Wallace 1968). However, after a set of three samples were analyzed among most of the 26 participants, it was determined that additional information could not be gained by continuing the program. Among the 26 participants:

- 25 were AF members and one was Army
- 67 samples were processed in total for this sub-cohort
- 3 individuals provided only one sample, two individuals provided only two samples
- one individual declined to provide any additional samples; this individual had a predicted initial systemic body burden of 10% of the MPBB from a screening sample and 16% from a single resampling sample
- two of the 26 individuals were among the 20 with predicted systemic body burdens greater than the MPBB based on initial urine sample results

TABLE 6. Analysis of Body Burden (BB) Greater 1% Group. Values from Odland (1966) [Values in Parenthesis from Odland *et al.* (1968), Only Listed if Different from 1966 Data, 183 Samples in 1966 Dataset, 219 in 1968 Dataset].

Parameter	Mean	St Dev	Mode	Median	Range
$^{239+240}\text{Pu}$ (pCi)	0.093	0.063 (0.114)	(0.029)	0.077 (0.066)	0.026 – 0.390 (0.011 – 1.030)
^{236}Pu spike (% recovery)	76	13	(61)	75 (76)	43 – 109 (113)
Sample volume (liters)	1.3	0.5	(1.1)	1.2	0.29 – 3.1 (3.6)
Elapsed time (days)	147 (178)	25 (77)	(140)	140	110 – 237 (65 – 396)
BB* (%)	4	3 (4)	(3)	3	1 – 16 (67) %

* Systemic body burden (e.g., excludes lung and its associated tissues), value of 0.044 μCi $^{239}\text{-Pu}$ for D represents one body burden or 100%

Table A-4 contains a tabular summary of the results for the 26 individuals from Wallace (1968).

4.6 Labat-Anderson Dose Evaluation Report

Labat-Anderson, Inc, (2001) completed a dose evaluation report with purposes:

- to identify, locate and review the records of the incident, radiation exposure assessments, and other information pertinent to the study.
- to evaluate current methods and models for estimating radiation doses and risks from the intake of radioactive materials contained in nuclear weapons.

- to recommend a methodology for conducting re-evaluation of the available radiation exposure information.
- to evaluate any and all radiation exposure information, such as urine bioassays, nasal swabs, air sampling information, etc. for scientific soundness and possible use in updating the radiation records of the response personnel.
- to perform the update and prepared records for input to the Air Force Master Radiation Exposure Repository.

The one key task completed in the report was a comprehensive dose assessment for the individuals that were part of the high 26 group, based on urinalysis. As noted above, this was the group of individuals with the highest predicted initial systemic intakes among the group of 400+ individuals that participated in the urine resampling program. Labatt-Anderson considered the use of three software codes to estimate ^{239}Pu intakes: a code developed specifically for the AF as modified by an existing US Army code, Code for Internal Dosimetry (CINDY) that met the AF's regulatory requirement to adopt the use of the ICRP 26/30 internal dosimetry recommendations, and Lung Dose Evaluation Program (LUDEP) version 2.06. The latter program used the ICRP's updated lung model detailed in Report 66 (ICRP 1994b) and Report 60 tissue weighting factors (ICRP 1990). Labatt-Anderson noted that using the CINDY code to estimate doses was deemed the primary method due to its adherence to the Air Force's current regulatory requirement for use of ICRP 26/30 and CINDY offered more flexibility to the user. LUDEP, therefore, fulfilled a complementary assessment. In addition to intake estimates for the high 26, the Labatt-Anderson report contained a listing of urine sampling data for all individuals that submitted samples to RHL. A complete summary of estimated ^{239}Pu intakes by both modeling methods are provided in Table A-5 for the cases, by patient⁹ number according to Wallace (1968). In addition, for modeling with CINDY, the committed dose equivalent (CDE) values are provided in accordance with ICRP 26, while for LUDEP, the committed equivalent dose (CED) in accordance with ICRP 60.

Labatt-Anderson (2001) concluded that the estimated intakes were within a factor of two for the majority of the high 26 individuals. These are highlighted in green in Table A-5. Estimated intakes for four individuals were within a factor of three. A summary of the data from Table 7 is contained in Table 7. The range between estimated minimum and maximum intakes among the 26 using CINDY was 35, while 137 for the LUDEP estimates. A key characteristic of the data shown in Table A-4 is the fact that a large majority of the group were on-site early in the response. Beyea and von Hippel (2019) reviewed and questioned how the Air Force used this data set for assessment of doses for veterans that responded to the accident. This issue, as well of consideration of comments made by Beyea and von Hippel will be discussed in greater detail in other parts of this document. Evaluation of the process used by the Air Force, however, is better understood upon review of the evolution of ICRP internal dose assessment methodologies.

⁹ The term "patient" was used by RHL. No inference should be made that the individuals were ill. It is assumed that this term was used because the Commander of the organization at the time was a physician.

TABLE 7. Summary of Labat-Anderson Dose Intake and Dose Assessment for High 26 Individuals.

Parameter	CINDY [ICRP 26/30/48]		LUDEP [ICRP 60/66]	
	Intake (nCi)	CEDE (rem)	Intake (nCi)	CED (rem)
Min	34	10	19	1.3
Median	68	21	86	6.1
Maximum	1,200	370	2,600	180

5.0 International Commission on Radiological Protection (ICRP) for Internal Dosimetry.

5.1 General

The ICRP methodology for application of radiation safety for inhalation exposures to plutonium have evolved over the last 60 years. The first standards for plutonium in humans were based on extrapolation of radium dial painter exposure data supplemented by animal studies with plutonium. Since, these models have been refined numerous times based on both animal and human health studies, and epidemiologic studies. The primary impact of the evolution is improved metabolic data and individual tissue dose estimates. Due to these refinements, the distribution of CED among organs has varied.

Recommendations of the ICRP have been incorporated into US and international standards. The Nuclear Regulatory Commission (NRC) used the recommendations from ICRP Report 2 prior to the Commission being established in 1974 (in the reorganization of the Atomic Energy Commission) up to the early 1990s when it adopted provisions of ICRP Reports 26 and 30. The Air Force followed suit to remain consistent with its radioactive materials licensed by the NRC. The Department of Energy (DOE) adopted the methodology of ICRP Reports 60 and 68 in 2007. ICRP published in Report 103 (ICRP 2007) updates to its recommendations in Report 60. Updates for internal dosimetry modeling were provided in Report 130 (ICRP 2015), with specific information on plutonium in Report 141 (ICRP 2019). The combination of ICRP 103/130/141 has yet to be adopted for use by a US regulatory body.

Radiation protection guidance issued in the United States has evolved over nearly 90 years since the first recommendations were released in National Bureau of Standards (NBS) Handbook No. 15, *X-Ray Protection*, in 1931. Early exposure guidance was designed to protect against observable radiation effects, i.e., reddening and desquamation of the skin, abnormal changes in blood counts, and others. Early guidance focused on protection from external radiation exposures from machine-produced x-rays and radium sources. The first US radiation protection guidance for internally-deposited radionuclides was issued in NBS Handbook No. 27, *Safe Handling of Luminous Compounds*, in 1941. The primary focus of this guidance was protection from ingestion and/or inhalation of dangerous amounts of radium and secondarily protection from inhalation of hazardous levels of radon gas and its daughters. As noted above, the standard for ingestion of radium was modified for applications to internal exposures to plutonium in the 1950's. This guidance continued to provide recommendations for protection from the readily observable short-term effects of

radiation exposure, but added new guidance to protect from the delayed effects of radiation due to the accumulation of radium in the skeleton and exposure to the lung from radon daughters. At that time, important recognized delayed effects were bone necrosis, leukopenia, anemia, and increased risk of osteosarcomas.

Radiation exposure guidance published in the US by the National Committee on Radiation Protection and Measurements (NCRP) in Report No. 22 in 1959, *Maximum Permissible Body Burdens (MPBB) and Maximum Permissible Concentrations (MPC) of Radionuclides in Air and in Water for Occupational Exposure* had a long-lasting influence on radiation protection standards in the US. This report was published as NBS Handbook No. 69, and similar to many previously NBS-published radiation protection handbooks was developed by the NCRP with input from numerous US radiation protection experts and in parallel with international experts, most notably those associated with the ICRP. The MPBB and MPC values contained in NCRP Report No. 22 were the same as those published in ICRP Report 2 (ICRP 1959), with only minor exceptions. These reports met the underlying exposure guidance and objectives of ICRP Report 1 (ICRP 1958). The stated objective of the guidance was “to prevent or minimize somatic injuries and to minimize the deterioration of the genetic constitution of the population.” The common exposure guidance from these reports formed the basis for AEC exposure standards promulgated in 10 Code of Federal Regulation (CFR) Part 20, 1960, which remained largely unchanged until the Nuclear Regulatory Commission (NRC) adopted an updated set of ICRP recommendations in the early 1990s. The ICRP published recommendations for internal exposures to plutonium from inhalation in 1950 (ICRP 1950) and 1955 (ICRP 1955), however, these recommendations were supplanted by ICRP Report 2.

5.2 ICRP Reports 1 and 2

The methodology in ICRP Report 2 was in use during the Palomares accident recovery. A summary of the exposure limits specified in ICRP Reports 1 and 2 are contained in Table B-1. The primary criteria applied to most occupational exposures of this period was the dose equivalent limit to the whole-body, head and trunk, blood-forming organs, and lens of eye, which was 1.25 rem per calendar quarter and 5 rem in a year, but with a provision for higher exposures for adults older than 19 when lifetime occupational exposure history was considered. External radiation dosimetry monitoring results are traditionally used to estimate the deep tissue dose to a monitored individual. For this period, film badge dosimetry was commonly conducted to assess external exposures. Due to the very low external radiation emissions from ground-deposited WGP, only limited external dosimetry monitoring was issued to personnel, and adherence to a whole-body dose limit was not a key radiological health concern.

Internal dose limits were the key radiological health concern, which invoked the methodology in ICRP Report 2. Effective control of radiation exposure to individuals from internally-deposited radionuclides is more complicated than external exposures due varied exposure routes, deposition, and retention in the organs and tissues of the body. Typically, the most important exposure routes are inhalation and ingestion, though in some cases skin absorption and exposure through wounds can be important. For the conditions at the Palomares accident site, inhalation of

WGP was the key exposure pathway. To address these varied conditions, ICRP limited radiation exposures by the critical organ concept,

“A critical organ is defined as that organ of the body whose damage by a given radiation source results in the greatest impairment to the body. Criteria appropriate to the determination of critical organs for external or internal exposure are: (1) the radiosensitivity of the organ, i.e., the organ damaged by the lowest dose; (2) the essentialness or indispensability of the organ to the well-being of the entire body; (3) the organ that accumulates the greatest concentration of the radioactive material; and (4) the organ damaged by the radionuclide enroute into, through, or out of the body.”

This terminology was developed in radiation exposure guidance issued by the NCRP and ICRP in the early 1950's, and was effectively the basis for the internal radiation exposure standard for radium issued in 1941. Because the guidance is developed for individuals with potential for radiation work over an entire lifetime, these limits were developed to ensure that the annual dose to the critical organ did not exceed the acceptable annual limit, cumulated over a 50-y working lifetime. For cases of exposure to bone seeking radionuclides, MPCs could be based on the MPBB due to accumulations in the bone. In these cases, the MPBB was based on equivalency to ^{226}Ra . For ^{239}Pu , inhalation exposures to soluble materials were limited by exposure to the bone, while for insoluble materials exposures were limited by dose to the lung, which had an annual RBE dose limit of 15 rem. RBE, *relative biological effectiveness*, was the modifying factor used in ICRP Report 2 to account for the varied effect on biological endpoints of radiation types. Table B-2 contains a listing for RBE values in ICRP Report 2 and later ICRP reports. In Report 26, the quality factor (QF) term was used, while in Report 60 and 103 the term, *radiation-weighting factor*, w_R , was used. In Report 2, the RBE for α -particles was 10, while for ICRP updates, the value is 20.

As noted earlier in this document, RHL established exposure limits based on ^{226}Ra -equivalent to bone from ^{239}Pu , in effect assuming a soluble chemical form of plutonium. In contrast, accidents of this nature are known to produce an insoluble, $^{239}\text{PuO}_2$ contaminant. The reason this decision was made for Palomares responders is not known. For other exposures to DoD from WGP in the 1960's and 1970's, use of the more limiting maximum permissible concentration for air (MPC_a), as applicable to soluble compounds was also used¹⁰, e.g., at Johnston Island (Rademacher 2016) and Enewetak Atoll (Rademacher 2019). It is possible that it was done to be conservative, as the MPC_a for soluble compounds of WGP is lower than for insoluble (see Figure 1).

Figures B-1 and B-2 provide an illustration of the accumulation of lung and bone burdens of ^{239}Pu from inhalation of insoluble and soluble forms, respectively. The solid black line in the plots represents the accumulation in the respective organ over a 50-y occupational exposure period at an inhalation level equal to the MPC in air. The MPOB for the lung is $0.016\ \mu\text{Ci}$ (16 nCi), while it is about $0.04\ \mu\text{Ci}$ (40 nCi) for the bone. For inhalation exposures of insoluble ^{239}Pu , a near steady-state equilibrium lung burden is achieved in about nine years, while for the bone, a steady-state equilibrium is not achieved within the 50-y occupational exposure timeframe. The ramifications of

¹⁰ Figure 1 notes the MPC_a of 40 and $2\ \mu\text{Ci cm}^{-3}$, for insoluble and soluble forms of plutonium, respectively.

these metabolism characteristics are interesting from a practical exposure standpoint for Palomares response personnel, where support of this project may have been the only occupational exposure potential for the majority of personnel. Each plot also contains the accumulation of lung and bone burdens of ^{239}Pu for a two-month and 12-month exposure period, but after the respective periods, no additional exposure is received. With the exceptions of some Palomares personnel that only visited the site for a few days of the 11-week duration of the response, most were assigned for a few weeks. The illustrated two-month exposure periods is thus high-sided for most personnel. For inhalation exposures of insoluble ^{239}Pu for two- and 12-months, the lung only accumulates about 11 and 50%, respectively of the ICRP Report 2 MPOB. For inhalation exposures of soluble ^{239}Pu for two- and 12-months, the bone only accumulates about 0.4 and 2.2%, respectively of the MPOB. The bar graph in Figure B-3 illustrates how the dose from the lung is distributed from a two-month inhalation exposure at the MPC to insoluble ^{239}Pu , but with no additional exposure. In the year of exposure, the total lung dose is about 1.1 rem, 7% of the annual limit to the lung. The total dose summed over eleven years is nearly 2.5 rem. The green lines in each Figure illustrate the magnitude of intake required to produce a MPOB for the lung and bone from a two-month exposure. For insoluble ^{239}Pu , inhalation exposures would have to be nine-fold higher than the MPC_a , while for a soluble ^{239}Pu , it would have to be 275-times the MPC_a .

The metabolic models used for transport and retention of radionuclides in ICRP Report 2 were relatively simple, compared to the updates made over the last 60 years. The improvements were based on human physiology research, animal and human studies with exposures to radioactive materials, and epidemiology studies. A key factor in the progression of the standards development was the incorporation of conservative margins of safety as a compensatory measure for knowledge gaps that may have existed. Tables B-3 and B-4 contain parameters for the ICRP Report 2 respiratory tract and systemic metabolism, respectively. The respiratory model separated compounds between “readily soluble” and “other” deemed insoluble. The model did not have any provision for aerosol distributions: 25% of the aerosol intake was assumed to be exhaled, with 50% deposition in the upper respiratory tract that is assumed to be cleared in mucous to the GI tract, while 25% is deposited in the lower respiratory tract. Readily soluble materials deposited in the lower respiratory tract is assumed to be cleared rapidly to bodily fluids, e.g. primarily blood. One-half of insoluble aerosol deposited in the lower respiratory tract is assumed to have a delayed clearance. This material is responsible for the calculated dose to the lung. The default retention half-life for deposited material is 120 days, one year for plutonium. Soluble materials were assumed to receive no lung dose. For these materials, dose was limited by tissue retaining materials distributed through systemic circulation or dose to the GI tract. ICRP Report 2 assumed 80% of ^{239}Pu reaching the blood was retained in the bone with a biological half-life of 200 y, 15% retention in the liver with a half-life of 82 y, and 2% to the kidneys. The preferable retention in the bone and liver was initially based on animal studies.

5.3 ICRP Reports 26 and 30

In 1977, the ICRP updated their guidance for occupational exposures to ionizing radiation in Report 26 (ICRP 1977). The objectives remained largely unchanged from those made in 1959:

- the prevention of detrimental non-stochastic (dose threshold for occurrence) effect on the exposed individual,
- limitation of stochastic (probability of effect related to dose) effects on the exposed individual to acceptable levels, and
- limitation of stochastic effects on descendants of the exposed individual (hereditary).

The 1977 ICRP recommendations limited exposure to the whole-body, by applying a tissue-specific weighting factors, w_T , for the dose equivalent received by the various tissues and organs of the body and limiting the summation of the product. Table B-5 contains a listing of the dose equivalent limits, while Table B-6 weighting factors, as well as those from updates to ICRP Report 26 in Report 60 (ICRP 1990) and Report 103 (ICRP 2007). Although the critical organ approach for dose limitation was no longer used by ICRP, the factors used in this approach were incorporated into the organ weighting factors. The higher annual dose equivalent limits of 15 and 50 rem, as applied to individual organs, extremities, and the lens of eye are specified to prevent detrimental non-stochastic (i.e., deterministic) effects. It is notable that the annual whole-body dose limit of ICRP Report 2 is similar to that for the total effective dose equivalent (TEDE) limit of ICRP 26. In addition, each set of standards has higher acceptable limits for extremities, skin, and lens of eye than for the whole-body. In relation to key organs and tissues related to ^{239}Pu , the lung and red bone marrow have the same tissue-weighting factor in Reports 26, 60, and 103. In the latter recommendations (Reports 60 and 103), the w_T for the bone surfaces dropped from 0.01 from 0.03. The weighting factor for the liver is similar for the three sets of recommendations: 0.06 (Report 30), 0.05 (Report 60) and 0.04 (Report 103). Although not of particular concern for internal exposures from ^{239}Pu , the weighting factor for the gonads has progressively declined due to evidenced-based, reduced concern for induction of heritable effect from ionizing radiation.

One of the most important aspects of ICRP Report 30 was introduction of more detailed transport and retention models. Figure 4 provides a generic representation of key components. For inhalation intakes of insoluble forms of ^{239}Pu , key components are the respiratory tract, the liver, and specific tissues of the bone: the endosteal surfaces¹¹ and the bone marrow. The transport compartment encompasses fluid flow through the body, but for practical purposes is dominated by the circulatory system for blood in regard to ^{239}Pu . A diagram of the ICRP Report 30 respiratory tract is shown in Figure B-4. The respiratory tract is a key organ for inhalation of insoluble forms of ^{239}Pu , as it retains materials for prolonged periods that affords accumulation of dose to lung tissue, it serves as a conduit for uptakes into the circulatory system, and clearance to the GI tract by mucociliary action. Although radioactive material transiting through the GI tract have some modeled uptake into the circulatory system, for inhalation exposures to insoluble ^{239}Pu uptakes to the circulatory system are dominated by that absorbed through the lung.

ICRP Report 30 Lung Model

The ICRP Report 30 lung model provides deposition of inhaled aerosols within three general regions of the respiratory tract: naso-pharynx (N-P), trachea-bronchial (T-B), and pulmonary (P).

¹¹ Generally termed simply bone surfaces.

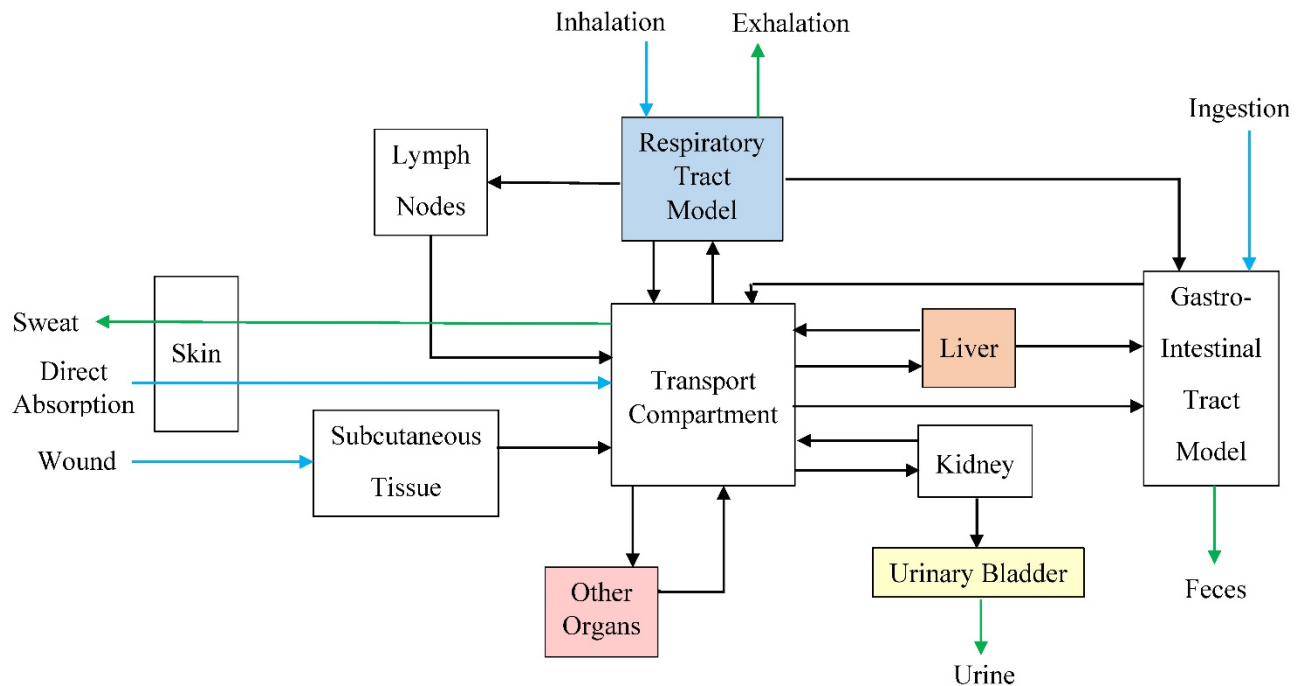


Figure 4. Generic Internal Metabolism Model.

Each of these regions are also annotated on a detailed anatomical model of the lung in Figure B-6. Within each region of the respiratory tract, aerosol deposition is partitioned between material destined for long-term retention and removal to the lymph nodes and body fluids (i.e., circulatory tract for blood) or that destined for clearance to the GI tract. Within each region, total aerosol deposition is dependent on the activity median aerodynamic diameter (AMAD) distribution, as shown in Figure B-5. The diagram shows the range of assumed deposition for aerosol distributions between 0.1 and 20 μm AMAD. For the default aerosol distribution of ICRP Report 30, 1 μm AMAD, the deposition fraction in the N-P region is 0.3, 0.08 in the T-B region, and 0.25 in the P region. Hence, exhalation accounts for 37% of a 1 μm AMAD aerosol intakes. For 5 and 10 μm AMAD aerosol distributions, the exhaled fractions are 11 and 1%, respectively. Partitioning of the aerosol deposition within each region is based on the bio-mobility (transportability) of the compound, binned in one of three inhalation classes: D, W, and Y. Since ICRP did not place any compound of plutonium in Class D, no values are shown for partitioning fractions in Table B-7. Plutonium dioxide was assigned to Class Y, with other common compounds to Class W, e.g., nitrates. Figure 5 contains the retention and clearance of the ICRP Report 30 lung model to an intake of 1 μm AMAD Class Y ^{239}Pu . Due to the exhalation of 37% of the intake, the total for all components shown in the Figure for any time is 63% in the intake. While there is some small fractional initial clearance to the blood, most initial clearance is to the GI tract. The clearance from the lung for later periods is mostly to the blood and to a lesser degree the lymph nodes. The cumulative clearance to the blood from the lung is 5%. Only an insignificant additional amount is introduced in the blood from uptakes of material cleared to the GI tract. Over 99.99% of ^{239}Pu cleared to the GI tract from lung clearance is expected in feces. For this model, the only appreciable

fraction of the initial intake decays after the intake is attributed to retention in the lymph nodes. The modeled retention and clearance for Class W ^{239}Pu is in Figure B-7. Key differences with Class Y material is a much more rapid clearance to the blood, with a cumulative clearance to blood at 12% of the intake, over two-fold higher, and no long-term retention in the lymph nodes.

Figure B-8 contains a plot of retention and clearance for Class Y ^{239}Pu , but for a $5\text{ }\mu\text{m}$ AMAD aerosol. A key difference with the modeled response for a $1\text{ }\mu\text{m}$ AMAD intake is the cumulative clearance to the blood is only 2.5%, one-half the cumulative value for a $1\text{ }\mu\text{m}$ AMAD intake. In comparison to the ICRP Report 2 and ICRP Report 30 lung models, for inhalation intakes of ^{239}Pu , the mean retention time of inhaled aerosols (weighted by exhalation, deposition, and clearance) is 230 days (Class Y, $1\text{ }\mu\text{m}$ AMAD, ICRP 30) versus 66 days for insoluble intakes under ICRP Report 2.

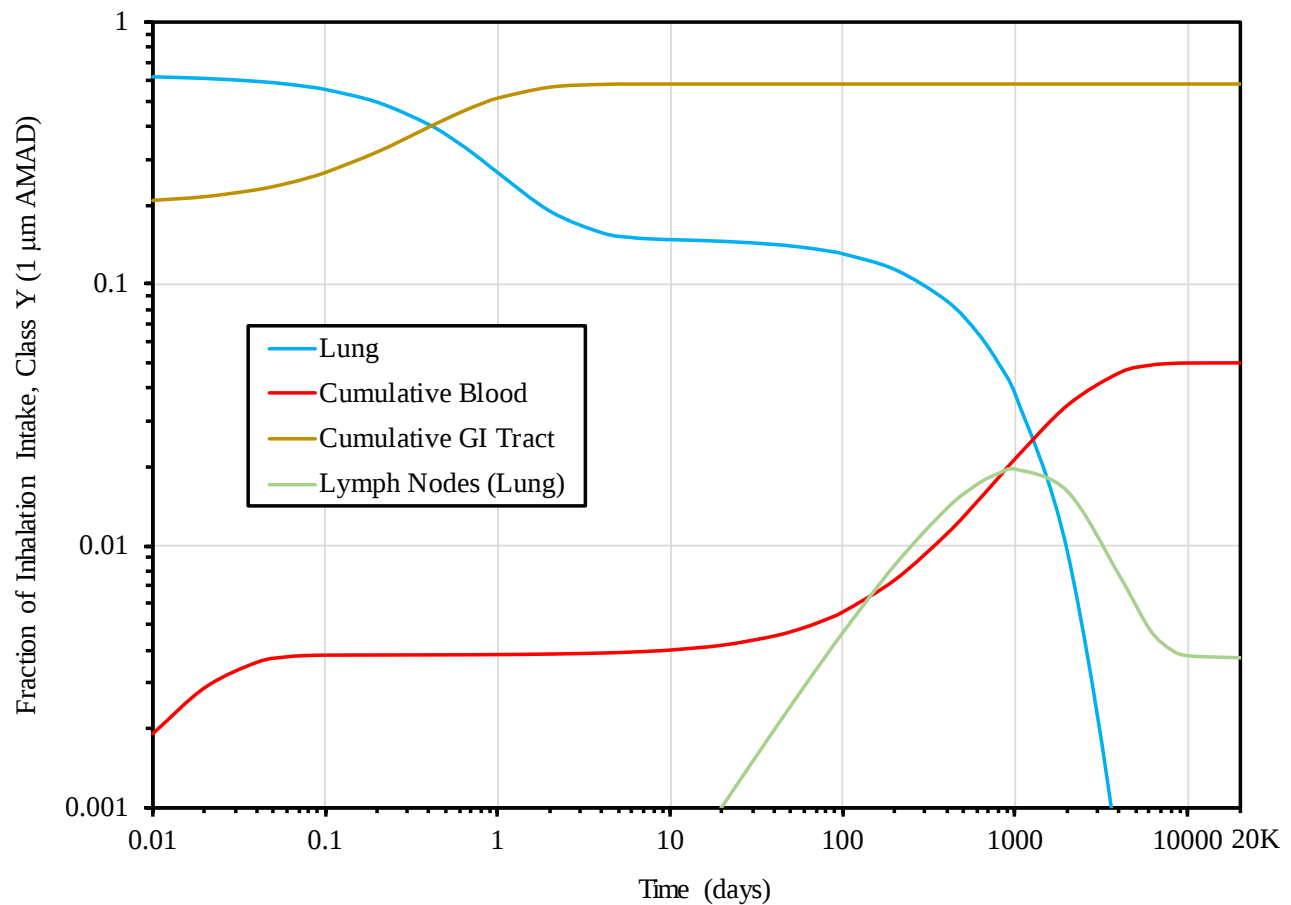


Figure 5. Retention and Clearance for Inhalation Intakes of Class Y ^{239}Pu , Based on $1\text{ }\mu\text{m}$ AMAD Aerosol with ICRP Report 30 Respiratory Model [Radioactive Decay Ignored].

ICRP Report 30 Systemic Deposition

Among the organs of the body subject to deposition and retention of ^{239}Pu from the blood, 45% is deposited in the bone and liver, with 10% uniformly in other soft tissues and early excreta, 0.035% in the testes, and 0.011% in the ovaries. Table B-8 shows these values along with biological retention half-lives in these organs among the various applicable ICRP reports.

For the mineral bone, bone surfaces (e.g., endosteal) and active red bone marrow (RBM), ICRP Report 30 separated bone dosimetry by the predominant deposition pattern of the element. Alkaline earths, e.g., calcium and its analogues: strontium, barium, and radium, are assumed to be uniformly distributed in mineral bone, with an equal partition between cortical and trabecular. For surface seeking elements like plutonium and thorium, the model assumes a uniform deposition over the bones surfaces, mass 120 g, without any burial in mineral bone.¹² Twenty-five percent of the α -particle energy emitted by material on the BS is assumed to be absorbed, regardless of the type: trabecular or cortical BS. For the RBM, the model assumed 50% α -particle energy absorption, but only in the trabecular marrow, which has an assumed mass of 1,500 g. Similarly, the assumption of no burial in the mineral bone matrix, RBM doses were known to be conservative. Since the primary radiological emissions from ^{239}Pu are from α -particles, energy absorption fractions will not be listed for the emissions of other radiations. Biological retention on bone surfaces was assumed to be 100 years, while 40 years for the liver, respectively, about half the values used in ICRP 2. The material transferred from blood to the ovaries or testes is assumed to be retained permanently.

5.4 ICRP Report 48.

ICRP Report 48 (ICRP 1986) provided updated metabolism information on plutonium and related elements. A summary of the changes are contained in Table B-8 along with parameters from other ICRP reports. The key change for plutonium was a reduction in the biological half-life in the bone and liver: dropped by a factor of two from ICRP Report 30, 100 $\rightarrow$ 50 y and 40 $\rightarrow$ 20 y, respectively.

5.5 ICRP Reports 26, 30, and 48 Doses from Inhalation Intake.

Table 8 contains a listing of dose conversion factors (DCFs), tissue weighting factors, weighted dose equivalent values, and dose equivalent values for individual tissues. The values are from FGR 11 (EPA 1988), as scaled to a committed effective dose equivalent (CEDE) equal to the annual limit of 5 rem. The DCF for the liver was calculated from the method in ICRP Report 30. As noted in the Table, the liver is one of the five organs that contributes to the remainder. The weighted contribution from the liver dominates the weighted remainder contribution to CEDE. Other organs contributing to the weighted remainder dose equivalent were part of the GI tract. The largest contribution to CEDE was the lung, with an equivalent dose of 19.1 rem. The highest estimated equivalent dose to a tissue was 48.5 rem to the BS.

¹² ICRP recognized that this assumption was conservative in nature (ICRP 1977).

The estimated dose equivalent values to the breast and thyroid were extremely low because these organs are not among tissues that are assumed to metabolize plutonium within the ICRP Report 30 model. The source of dose to these tissues is from the low-frequency emission penetrating radiations from plutonium in organs that have metabolism of the element. This condition illustrates one of the practical approaches of ICRP Reports 26/30 in radiation safety and shortcomings if it is used for dose reconstruction purposes. For inhalation exposures to ^{239}Pu , the key organs of concern are those that have primary deposition and long-term retention: the lung, liver and bone. Importantly, animal exposure studies and more recent human epidemiological studies have demonstrated a link between ^{239}Pu exposures and increased risk of primary lung, liver, and bone cancers. Despite projected doses to the lung-associated lymph nodes and the RBM, increased risks of leukemia and lymphoma induction have not been linked to inhalation of ^{239}Pu among epidemiological studies of exposed workers. For assessment of dose to organs and tissues with very low deposition and/or poor retention, the models may not provide reasonably accurate estimates for some dose reconstruction purposes. Nevertheless, the simplified assumptions for deposition and retention in these organs or tissues have negligible impacts to their use for applications to radiation safety in workplaces.

TABLE 8. Dose Conversion Factors (DCF), Tissue Weighting Factors, Tissue Dose Equivalent, and Committed Effective Dose Equivalent for Inhalation of Class Y ^{239}Pu , 1 μm AMAD, at the Annual Limit, 16 nCi, FGR 11.

Organ or Tissue	DCF		W_T	Weighted Dose Equivalent (rem)	Fraction of CEDE	Dose Equivalent (rem)
	Sv Bq^{-1}	$\text{rem } \mu\text{Ci}^{-1}$				
Gonads	1.20 E-5	44.4	0.25	0.177	0.035	0.708
Breast	3.99 E-10	0.0015	0.15	3.52 E-06	7.1 E-7	2.35 E-5
RBM	6.57 E-5	243	0.12	0.464	0.093	3.88
Lung	3.23 E-4	1,200	0.12	2.28	0.457	19.1
Thyroid	3.75 E-10	0.0014	0.03	6.62 E-07	1.3 E-7	2.21 E-5
BS	8.21 E-4	3,040	0.03	1.45	0.291	48.5
Liver*	1.49 E-4	550	0.06	0.526	0.105	8.77
Remainder	3.02 E-5	112	0.30	0.535	0.107	1.78
Effective	8.33 E-5	308	1.0	5.00	SUM	-

* Liver is also part of the remainder

5.6 ICRP Report 66 Lung Model and ICRP Report 67 Systemic Model.

After the publication of Report 60 (ICRP 1990), ICRP made updates their lung model in Report 66 (ICRP 1994b) and systemic metabolism in ICRP Report 67. In addition to refinements in lung function modeling for adult workers based on “increased knowledge of the anatomy of physiology of the respiratory tract,” the updated lung model provided provisions for different age groups (ICRP 1994b). While the updated model shown in compartmentalized form in Figure B-9,

with corresponding anatomy in Figure B-6 was more complex in structure to the ICRP Report 30 lung model shown in Figure B-5, ICRP recognized that the “wide availability of personal computers” allowed “the easy use of the model” (ICRP 1994b). The ICRP Report 30 lung model included dose to the lung-associated lymph nodes within total lung tissue dose, while the updated model separated dose to the lymph nodes as a separate tissue¹³. This is an important update with respect to inhalation of ²³⁹PuO₂ due to its limited mobility in the lung and recognized persistence in the lung-associated lymph nodes. Key updates in the model were also made to better characterize dose from radon progeny, though this is not important to this report.

Similar to the ICRP Report 30 lung model, deposition of aerosols are cleared to the blood stream, GI tract, and lymph nodes as shown in Figure B-9. ICRP Report 30 distributed aerosol deposition among eight lung compartments, while the newer model has twelve compartments. Estimated fractional deposition among the compartment are dependent on the activity median thermodynamic diameter (AMTD) according to Figure B-10. Note on the Figure that some compartments have the same modeled deposition and that three compartments in the alveolar region are combined into one for total deposition. To allow comparison to the deposition pattern for the ICRP Report 30 lung model, as shown in Figure B-5, Figure B-11 provides a plot where deposition is consolidated within the three distinct regions. ET (total) is equivalent to N-P as is alveolar to P. This plot also contains an exhaled fraction. The peak deposition fraction is for aerosols with an AMTD about 5 µm. Table B-9 contains the distribution of deposition fractions among each compartment, and transfer rates to other compartments. One key difference with the ICRP Report 30 lung model is longer retention for some fraction of the aerosol deposited in the alveolar region. Transfer of deposited material to the blood stream from each compartment of the lung competes with the particle transport, but is dependent on the specific biochemical characteristics of the aerosol. Details are not summarized in this report, yet a more practical illustration is provided by the cumulative percent of inhaled activity transferred to the blood from the lung (Figure B-12). Curves are provided for 1 and 5 µm AMADs and for Type M and S chemical forms. Table 9 provides a 50-y cumulative percent transferred to blood for the ICRP Reports 30 and 66 lung models. The Table provides values for 1 and 5 µm AMAD, with the time to reach 50% of the 50-y cumulative. Notably, the ICRP Report 30 lung model estimates greater fractions of ²³⁹Pu in the lung clear to the

TABLE 9. Cumulative Percent of Inhaled Activity Transferred to Blood for the ICRP Report 30 and 66 Lung Models at 50-y Post, Acute Exposure.

Aerosol AMAD (µm)	ICRP Report 30			ICRP Report 66		
	Class	Cumulative %	Time (50%)	Type	Cumulative %	Time (50%)
1	W	12	45 min	M	10	80 d
	Y	5	1300 d	S	1.4	1200 d
5	W	13	18 min	M	6.2	9 min
	Y	2.5	660 d	S	0.65	1300 d

¹³ Class Y materials under the ICRP 30 lung model, about one-half of the 50-y cumulative dose to the lung is due to lymph node retention.

blood than the ICRP Report 66 lung model for the respective Class W/Type M and Class Y/Type S combinations. For the comparison between Class Y and Type S, the differences in cumulative fractions cleared to blood are between a factor of three to four. For 1 μm aerosols in the same chemical Class Y/Type S, the time to accumulate 50% is about the same – 3.5 years. For 5 μm aerosols, the Type S compounds under the ICRP Report 60 lung model have estimated 50% accumulated clearance to the blood about twice as long as the Class Y under ICRP Report 30 lung model. ICRP Report 66 recommended 5 μm AMAD aerosols for common workplaces, vice 1 μm , the default recommendation in ICRP Report 30.

ICRP Report 67 provided updates to the ICRP Reports 30/48 systemic metabolism. The model for plutonium is shown in Figure B-13. Three key changes to systemic metabolism are:

- the addition of a second liver compartment to account for tenacious retention of plutonium in the liver,
- a more flexible bone model which allows for transport among the bone surfaces, the trabecular and cortical volume (i.e., mineral bone), and bone marrow, and
- a multi-compartment soft tissue model to account for deposition and retention in organs/tissues with very low deposition and/or poor retention.

While not important to this work, the updated model also provided varied parameters for groups of individuals under 18 years. Table B-8 contains fractional deposition and clearance rates for key tissues involved with systemic metabolism along with values from other ICRP reports. In contrast to ICRP Reports 30/48, the fraction of transfer from the blood to liver is lowered from 45 to 30%. ICRP Report 67 slightly increases the fraction transferred from the blood to bone, but doubled the fraction from the blood to soft tissues and early excreta, 20%. The gonads have the same fractional transfer from the blood as ICRP Reports 30/48, but provides a 10-year retention half-life vice infinite. One objective of the updated model was to increase long-term retention in the liver. Findings from autopsy data on workers exposed to plutonium was an impetus for this change (ICRP 1992). The primary update to metabolism within the skeleton provided for transport of plutonium from the site of initial deposition: the bone surfaces to the marrow and mineral volume, and that within the mineral volume to the marrow. The updates were a major change from the simple assumptions made in the ICRP Report 30 bone model for ^{239}Pu .

ICRP Report 68 (ICRP 1994a) and ICRP Report 71 (ICRP 1995) incorporated recommendations in ICRP Reports 60, 66 and 67. Table B-10 contains a listing of dose coefficient (DC) values for inhalation intakes of Types M and S compounds from an aerosol with a 1 μm AMAD. A histogram of the DC values for Type S are shown in Figure 6. The diagram is quite clear on the prominent organs for dose from inhalation intakes: the respiratory tract, bone surfaces, and liver. The relationship between the committed equivalent doses (CDE) to the BS and RBM is a factor of 20, while for these same organs under ICRP Report 26/30/48, the ratio is a factor of 12.5. The large drop in this ratio is a reflection of the updates in the bone model. With the exception of the kidneys and gonads, the other organs and tissues have identical DC values and arise from the modeled deposition and retention in soft tissues. This was a key addition to the ICRP Report 67 systemic

metabolism model. In review of Table B-12, the weighted contributions from the lungs, 0.64, and liver, 0.12, to effective dose are the largest. From Table 8, the lung contributed 0.46, while the BS 0.29 for the ICRP Reports 26/30/48 from inhalation of Class Y ^{239}Pu , 1 μm AMAD. One source of the difference between these sets of recommendations is the drop in the tissue-weighting factor for the BS from 0.03 to 0.01.

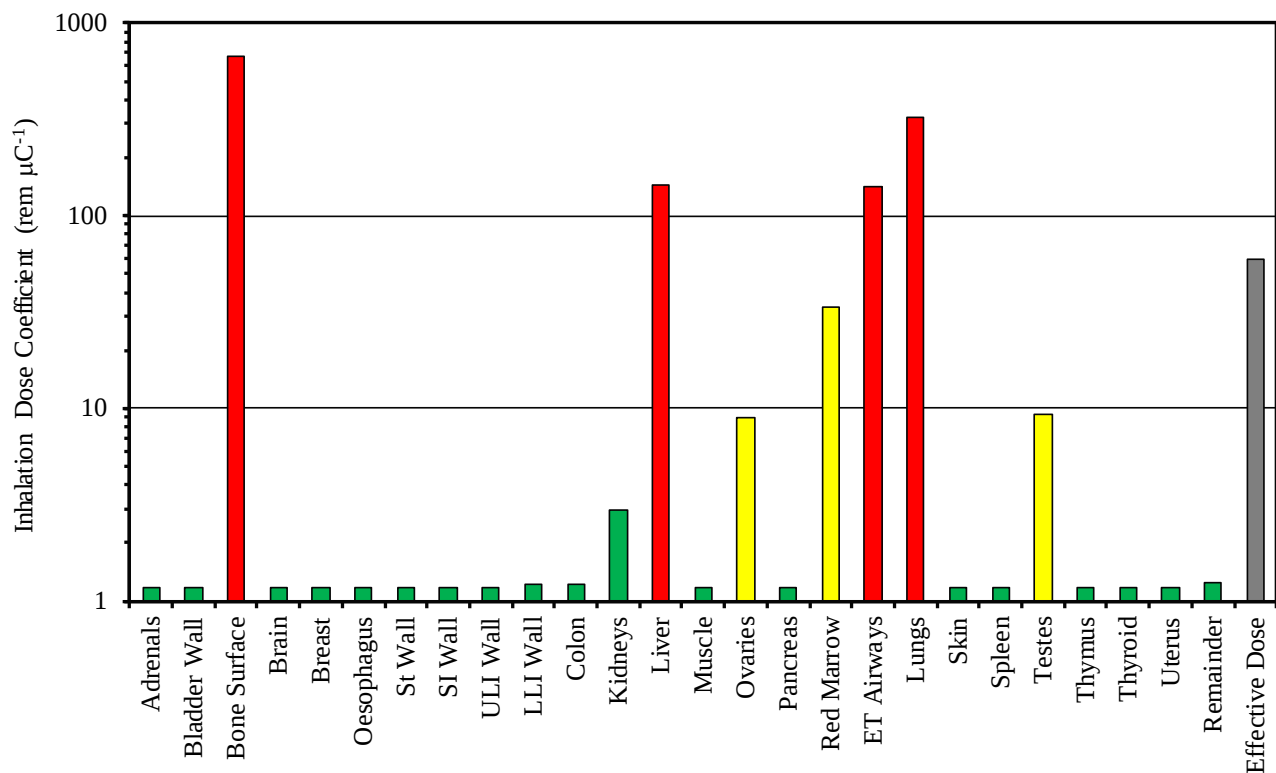


Figure 6. Inhalation Dose Coefficient Values, Inhalation Type S, 1 μm AMAD, Adults, ICRP 71.

5.7 ICRP Reports 103, 130, and 141.

The most recent ICRP updates in plutonium internal dosimetry modeling are based on the tissue weighting factors in ICRP Report 103 (ICRP 2007), updates to the ICRP Report 66 lung model contained in ICRP Report 130 (ICRP 2015), and Pu-specific systemic parameters contained in ICRP Report 141 (ICRP 2019). A key improvement to the methodology introduced in ICRP Report 103 was varied DC values for males and females, with an average DC for effective dose based on the arithmetic mean of the value for each gender. Unless specifically noted in this report, values for males are listed. Values for males will be applicable to the vast majority of Palomares recovery workers. There was minor changes to the respiratory tract of ICRP Report 66. A diagram of the model is shown in Figure B-14. The update model was simplified with a fewer number of compartments than ICRP Report 66 model (Figure B-9). Deposition fractions of aerosols among the

three primary components of the respiratory tract remained the same. Partitioning of deposition among sub-regions remained unchanged, except for the ET. The ICRP Report 66 model partitioned 65% of aerosol deposition in ET₁ and 35% to ET₂, while in the update the partition was evenly split among the two sub-regions. Table B-11 contains the transfer rates for individual compartments. Similar to ICRP Report 66, specific parameters for dissolution of materials are accommodated. ICRP Report 141 provides metabolism for the same three inhalation types: F, M, and S, as used in ICRP Report 66, but provides specific lung metabolism for a number of common compounds of plutonium likely to be encountered in workplaces.

The systemic metabolism model under ICRP Report 130 has similarity to that in ICRP Report 67. One key difference with respect to plutonium metabolism is the addition of a third sub-compartment to the liver, as shown in Figure B-15. In contrast to the ICRP Report 67 metabolism model, the ICRP Report 141 model provides for transfer of 60% of plutonium from the blood to liver and 30% to the bone. This increased partitioning of plutonium to the liver was implemented based on the autopsy studies of former Soviet Union Mayak Production Authority (MPA) workers (ICRP 2019).¹⁴ The ICRP 130 systemic metabolism model provides for transfers from the blood to both bone surface and mineral volume. This is in contrast to ICRP Report 67, which used assumed transfers from the blood to bone surfaces. Details of key transfer fractions and rates are listed in Table B-10. Although the transfer of plutonium from the blood to bone decreased from 50% of systemic circulation to 30%, the transfer rates of material from initial deposition on bone surfaces to mineral volume were decreased (i.e., longer retention). These changes altered the ratios of committed equivalent doses among the key systemic tissues: bone surfaces, red bone marrow, and liver from the previous set of ICRP recommendations for plutonium.

Other changes to metabolism were made in ICRP Report 141 over ICRP Report 67. Though the fraction of plutonium transferred from the blood to gonads remained the same, the retention half-time was reduced from 10 to five years. Other soft tissues retention rates decreased as well, though these tissues, as a whole, have insignificant deposition and retention in the organs affected by systemic distribution. Table B-12 lists DC values for ²³⁹Pu for inhalation Type S and the specific recommendation for PuO₂. In comparison to the values from ICRP Reports 68/71 in Table B-10, noticeable are differences in the tissues covered. The GI tract was limited to four portions, as compared to six under the previous set of ICRP recommendations. Oral mucosa, the heart, the salivary glands, gall bladder, lymph nodes, and prostate were soft tissue additions. Among these, inclusion of the prostate gland is important due to the prominence of prostate cancer incidence in males. Table B-13 contains a summary of American Cancer Society (ACS) cancer statistics for 2010 – 2012, whereby males had a 14% lifetime probability of diagnosis. Addition of the lymph nodes is important because nodes associated with the respiratory tract receive clearance from the tract and long-term retention of insoluble forms of plutonium. Nevertheless, lymphatic precursor cells are distributed among many tissues, and lymphomas have a weak link of induction from ionizing radiation exposures. Previous ICRP models included deposition and retention of radionuclides in the lymph nodes, but did not incorporate the CDE into calculation of effective dose. Table B-12 also contains tissue-weighted DC values, which reflect the contribution to effective dose.

¹⁴ The Mayak Production Authority workers represent a cohort of plutonium workers with the greatest levels of intakes.

Among the tissues, the lungs have the greatest contribution, with the lymph nodes second. Other tissues have varied contributions dependent on the aerosol distribution and lung class of the compound – Type S or PuO₂. Among these tissues, the liver, RBM, and extra-thoracic airways are important. A display of the DC values for individual tissues and effective dose from Table 12 are displayed in Figure 7. The display is for inhalation Type S compounds, 1 μm AMAD aerosols. The same key tissues exist, as shown in Figure 6 from ICRP 71, except for the addition of the lymph nodes. Figure B-16 shows a distribution of DC values for all four cases (e.g., inhalation compound type and aerosol combinations) listed in Table B-12 for key tissues and a few additional tissues that have more prominent frequency of occurrence – bladder (urinary), colon, kidneys, skin, and prostate.

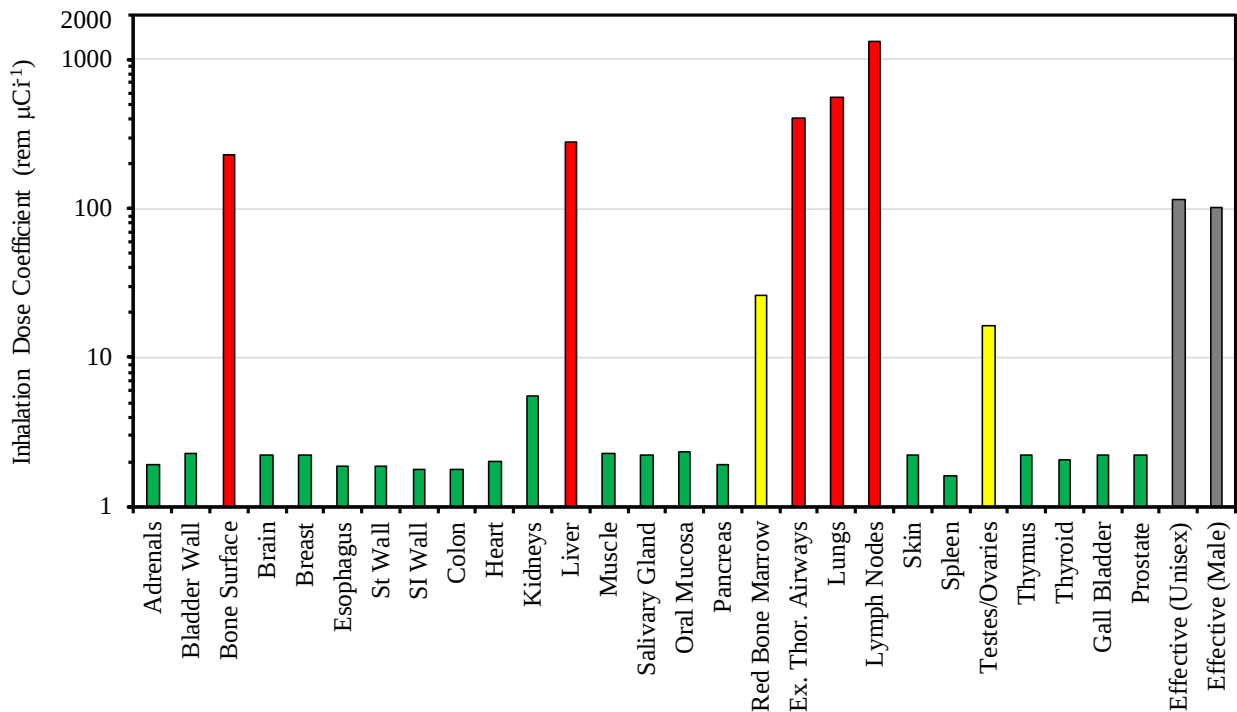


Figure 7. Inhalation Dose Coefficient Values, Inhalation Type S, 1 μm AMAD, Adults, ICRP 141.

One key feature of this histogram is the difference in transfer of plutonium from the lung to systemic circulation based on the aerosol characteristic. Aerosols of 1 μm AMAD afford about twice the transfers to the blood than 5 μm aerosols. The dioxide forms have about five-fold lower transfers than predicted by the Type S model. To the contrary, dioxide forms have longer modeled retention in the lungs, extra-thoracic airways, and lymph nodes than predicted for the Type S model. This leads to higher DC values for the extra-thoracic airways, lungs, and lymph nodes.

For the key organs affected by systemic distributions, Figure 8 provides a histogram of committed effective dose (CED) for the BS, RBM, and liver for three sets of ICRP recommendations for ²³⁹Pu. The ICRP Report 26/30/48 and 60/66/68 combinations provided the BS with the highest

CED among the three, while the ICRP Report 103/130/141 combination provided the liver with the highest CED. This is due to a key change to accommodate greater fractional liver retention observed among autopsies of MPA workers (Birchall *et al.* 2017). For this study, the urine excretion rates and body burdens at the time of autopsy were evaluated for about 500 workers.

5.8 Summary.

The ICRP methodology for application of radiation safety for inhalation exposures to plutonium have evolved over the last 60 years. The first standards for plutonium in humans were based on extrapolation of radium dial painter exposure data supplemented by animal studies with plutonium. Since, these models have been refined numerous times based on both animal and human health studies, and epidemiologic studies. The primary impact of the evolution is improved metabolic data and individual tissue dose estimates. Due to these refinements, the distribution of CED among organs has varied by generation of ICRP recommendation. This provides a conundrum for dose assessments, based on projected doses from urine bioassay data and/or air sampling data, and a discussion point. Should the latest ICRP metabolism models be used for estimates of dose to organs or tissue? Or the set of ICRP recommendation that were initially used for dose assessments? For internal exposures, the refinements recognize improved knowledge and better estimates of dose. However, dose estimates to some organs may be lower than those based on previous ICRP recommendations.

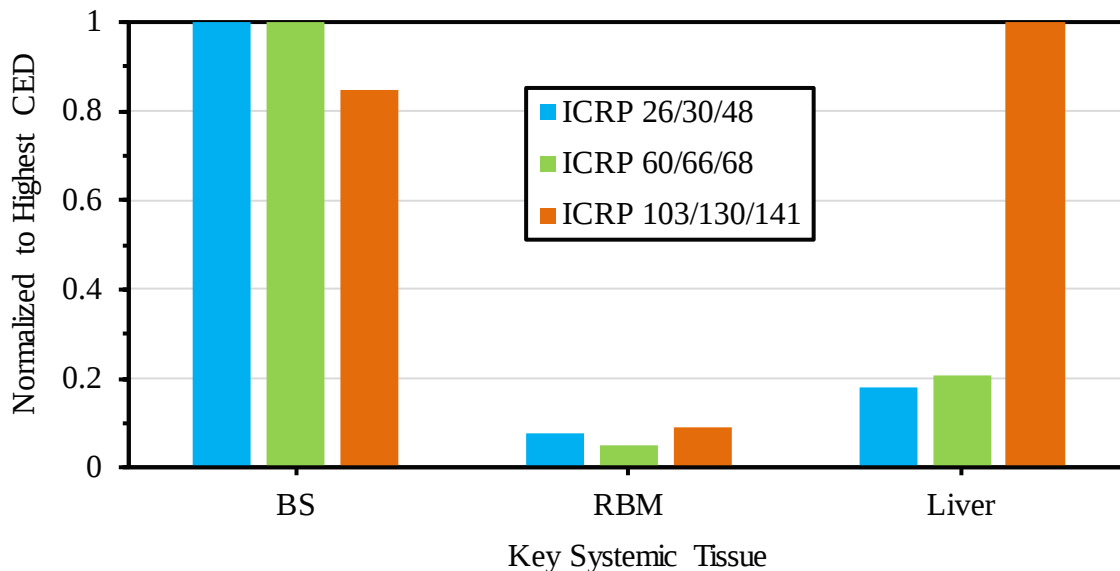


Figure 8. Histogram of Committed Effective Dose¹⁵ for the BS, RBM, and Liver for ²³⁹Pu by Various ICRP Systemic Metabolism Models.

¹⁵ ICRP Report 26 used the term committed dose equivalent (CDE), but is similar to CED in application to the comparison here.

6.0 Environmental Exposure Assessment.

6.1 General.

One goal of Labat-Anderson in their 2001 effort was to evaluate other sources of data to augment that from urinalysis-based exposure assessments. Air sampling data was noted as one source of environmental data. Labat-Anderson reviewed and reported on air sampling conducted by the Spanish in the vicinity of the detonations/plutonium dispersal points. The air sampling program established by the Spanish was accomplished in cooperation and support from the US AEC, then later the Energy, Research and Development Administration and the DOE. Beyea and von Hippel (2019) offered critique of the approach used by Labat-Anderson. Points of their critique were:

a) Dust generated by shoveling contaminated soil and vegetation into barrels, by deep-plowing fields, and by movement of trucks and machinery across fields during the cleanup could have caused much more resuspension of particles than wind.

b) It is well known that resuspension factors decline rapidly with time (Maxwell and Anspaugh 2011). The maximum resuspension coefficient of 10^{-7} quoted by Labat-Anderson for Palomares was measured 6 mo after the accident (Iranzo et al. 1994). This is consistent with measurements made 6 mo after the Chernobyl release, but measurements of resuspension coefficients immediately after the Chernobyl accident were two orders of magnitude higher (Garger et al. 1997).

c) The cleanup effort deliberately attempted to reduce the wind resuspension factor by deep-plowing fields that have been contaminated. The purpose was to redistribute surface contamination through the top 30 cm of soil and thereby make most of inaccessible to the wind.

d) The land contamination level of 1.19 MBq m^{-2} assumed by Labat-Anderson was the level below which cleanup was deemed unnecessary – much less than the contamination levels in the areas where the cleanup took place (Iranzo et al. 1987).

Some points of critique are well taken, though some points of the critique are based on incomplete information and do not take into account some conservative aspects of the example environmental dose estimate provided by Labat-Anderson. First, while surface soil concentrations of the contaminant were greater before remedial action was completed, and though remedial actions can increase resuspension rates over quiescent condition, other factors reduce these concerns. As noted above, two specific groups of individuals did wear air-purifying respirators for their work: EOD personnel and individuals in the vicinity of the operations where soil was being transferred to drums. This is in contrast to the statement by Beyea and von Hippel (2019) quoting Wright Langham, “the manual says you will dress up in coveralls, booties, cover your hair, wear a respirator, wear gloves, yet none of these were done” which premises that no mitigation to airborne resuspension were incorporated into work practices. It was noted above that during soil scraping, plowing, transport and loading operations, soils were wetted, which would greatly reduce resuspension over that expected for unmitigated conditions. Second, deployment periods for most individuals were for

three-week periods, while the Labat-Anderson report high-sided occupancy for 11-weeks, six days per week, and 12 hours per day. In reality, only a small fraction of personnel were present for the entire duration of the recovery. As well, occupancy in the most highly contaminated areas was limited to a small fraction of personnel and very unlikely for prolonged periods during a week. Due to the limited area subjected to soil scraping, these operations were accomplished over a few days per site.

The Labat-Anderson report had limited discussion of environmental data. Over the past fifteen years, the AF Surgeon General's Office in coordination with the Air Force Safety Center has incorporated additional applicable data sources to augment urine bioassay data in their assessment of exposure potential for the Palomares accident recovery. Three important sources of data were the:

- e) air samples collected during the recovery action period,
- f) the medical and dosimetry study on Palomares, Spain residents, and
- g) the Joint US-United Kingdom (UK) Roller Coaster Safety Shots conducted in Nevada in 1963.

Each one of these sources of data provided support to the conclusions drawn from the urinalysis program on exposure potential. Among these, the air sampling conducted during the recovery actions is most important, as it was accomplished during actual work.

6.2 Air Sampling Conducted by 16th Air Force during Recovery.

Earlier in this report, it was noted that four-hundred thirty-nine air samples were collected during the recovery (DNA 1975). The Air Force for disaster responses collected air samples with Staplex air samplers, the designated air sampler used during that period. Samples were field monitored with the Eberline PAC-1S α -particle scintillator. Sampling results were summarized in Appendix VI to Annex C to the *Disaster Control Report – Air Sampling Results*, an Appendix to the 16th Air Force Operations Recovery report (Air Force 1968). Annex C contains a listing of 439 sample results, for samples collected between 19 January to 17 March 1966. Upon review of the results, it was determined that 27 sample results were reported twice, providing only 412 samples vice the reported 439.

Air sampling was conducted during recovery operations that had the greatest potential for airborne resuspension of contaminants, with the exception of a couple of samples collected very early in the recovery action at the base camp. For operations involving mechanical disturbances of soil, e.g., soil scraping, plowing, transfer to drums, air sampling was conducted downwind of the operation to target areas of greater potential for contamination suspension in air. Based on review of the air sampling summary in Annex C, samples collection time appears to have been standardized to 30 minutes. With a typical sampling rate of $55 \text{ ft}^3 \text{ min}^{-1}$ (cfm), total sample volumes would have been about $1,650 \text{ ft}^3$ (47 m^3). The Bioenvironmental Engineering team instituted a procedure to read air filters immediately after collected and again many hours later (commonly the delayed reading

was the next morning). The delayed reading was implemented to allow for decay of radon daughters. Only about one-in-six samples had detectable α -particle emissions, based on the histogram shown in Figure 9. A detailed summary is in Table C-1. Samples with reported airborne concentrations had PAC-1S readings ranging from 50 to 1,700 cpm. The four samples with the highest concentrations were collected during EOD operations early in the response, 20 and 22 January, and soil to barrel filling operations on 16 March. It is important to note that the workers conducting the operations when these samples collected were wearing air-purifying respirators. Among the 11 samples with concentrations greater than 22.5, but less than equal to 54 pCi m^{-3} , eight were also collected during EOD operations, 20 and 22 January, and during soil to barrel filling operations between 14 and 17 March. Among the 12 samples with activity concentrations greater than 9, but less than equal to 22.5 pCi m^{-3} , seven were collected during soil to barrel filling operations between 13 and 17 March; the other five samples were not tied to any specific activity or period. Overall, only ten samples were in excess of the ICRP Report 2, MPC_a value of 40 pCi m^{-3} . MPC_a values, like DAC values, are based on a 2,000 h work year, with an expected dose equal to the annual limit. As such, some air samples with concentrations above the MPC_a is of no consequence.

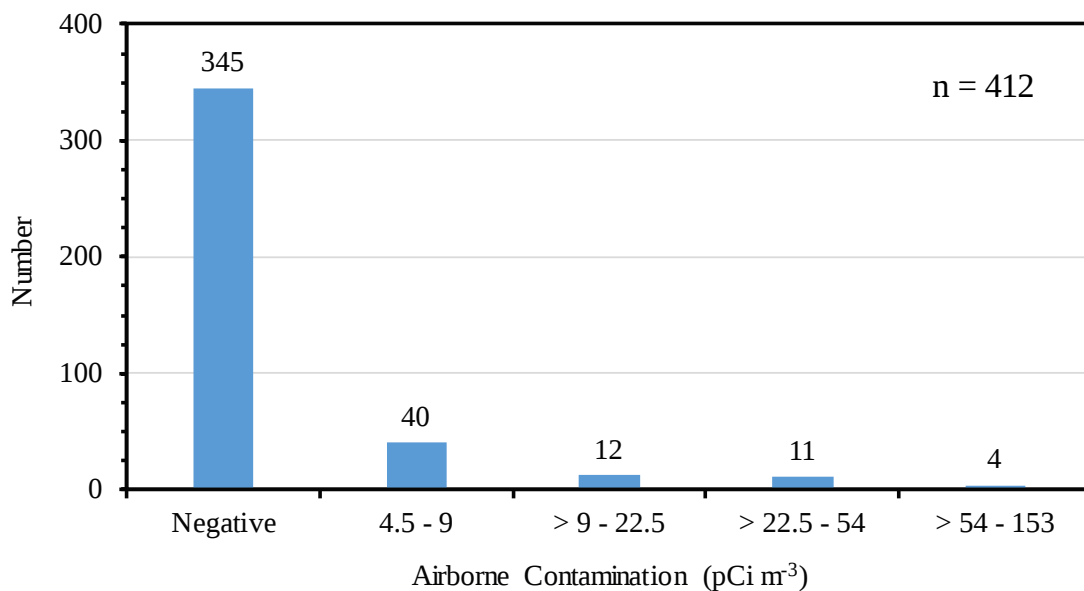


Figure 9. Histogram of Airborne α -Particle Concentrations in Air Samples Collected during the Palomares Recovery Action, 17 January – 17 March 1966.

It is important to note that the gross α -radiation analysis method used to assess air samples would include background source contributions from the naturally-occurring ^{238}U , ^{232}Th , ^{235}U , and their radioactive progeny in each series. This naturally-existing contributions to airborne radioactivity is more important for those samples with lower concentrations of plutonium from resuspension. With the exception of delay-counting, no other deliberate steps were taken to account (e.g., subtract) for α -radiation emitting sources in background. Because of the relatively-low

airborne concentrations of α -radiation detected in the air samples, RHL questioned some of the abnormally-high gross α -radiation levels observed in some initial urine samples submitted by personnel. Nevertheless, the data confirms the reasonably low airborne contamination levels, and the reason the vast majority of individuals that were targeted for the urine resampling effort had negative or very low results confirmed by the resampling.

6.3 Medical and Dosimetry Study on Palomares Residents.

The Spanish Center for Investigation of Energy, Environment and Technology (CIEMAT) in 1966 established a program to evaluate the health and potential exposures to residents of Palomares (Iranzo *et al.* 1998). Over 20 years after the initiation of the program, 896 individuals participated in the program. Initially, 59 residents of Palomares that were believed to have the greatest exposure potential submitted urine samples for analysis in 1966. Due to a similar cross-contamination problem the US observed in the collections of urine samples from veterans in proximity to the source of contamination, the Spanish resampled the same individuals in 1967, but while the residents were in Madrid. At the time of the accident, there were 485 people present in Palomares (Church *et al.* 2000). Among the 59 residents, detectable levels of plutonium were only found in the urine of 23 individuals.¹⁶ After the initial screen of these individuals, 150 residents from the village per year have participated in a medical follow-up, which included medical exams, submission and analysis of a 24-h void urine samples, and an in-vivo chest count. Seven-hundred fourteen individuals submitted at least one urine sample, with 19 submitting 10 or more samples (Iranzo *et al.* 1988). The in-vivo chest count best quantifies the plutonium inhaled and retained in the lung, and material that was subsequently cleared and retained in lung-associated lymph nodes. The urine samples best quantify plutonium released from systemic circulation and retention.

None of the individuals that received in-situ lung counts had a detectable level of ²⁴¹Am or ²³⁹Pu. The minimum detectable activity (MDA) for the system in initial use had a reported sensitivity of about 22 nCi, but higher in obese individuals. The long-term follow-up of Palomares residents by urinalysis is in Table C-2. Fifty-five individuals had detectable levels of plutonium in urine. Forty-five of these individuals were present during the recovery, while 10 resided after the recovery operation. It was noted by Church *et al.* (2000) that the burdens received by residents that were present at the time of the accident and recovery, were likely due to acute intakes, as air sampling conducted by the CIEMAT could not justify the degree of intakes on a long-term exposure basis. Overall, the highest estimated intake among the Palomares residents is less than 64 nCi, assuming inhalation intakes and the use ICRP Report 26/30/48 methodology.

Comparison of the exposure potential to Palomares residents and military members involved in the accident and recovery provide some insight into key exposure factors. Table 10 provides a summary of exposure considerations for each group, based on three phases: the initial plume produced by the two weapons that detonated, exposures during the recovery phase, and post-recovery. Two exposure pathways are listed. In general, due to the relatively poor uptake of plutonium into systemic circulation for material transiting the gastrointestinal tract, the ingestion

¹⁶ CIEMAT established a minimal detectable concentration for these at 0.74 mBq d⁻¹ (0.02 pCi d⁻¹) [Iranzo *et al.* 1998]

TABLE 10. Sources of Plutonium Intake for Palomares Recovery Responders and Residents.

Source	Exposure Pathway	Palomares Residents	Palomares Recovery Responders
Initial Cloud Plume (17 January, AM)	Inhalation	Key intake for residents within downwind plumes; exposure varied dependent on location and distance.	Only applicable to aircrew. One came down on land, while 3 landed in the Mediterranean. All transported from local area to hospitals, then Torrejon AB.
		<u>Mitigating factor:</u> individuals indoors would have much lower intakes than those outdoors.	Insignificant pathway compared to inhalation.
	Ingestion	Insignificant pathway compared to inhalation for this phase.	Insignificant pathway compared to inhalation for this phase.
Recovery Operation (17 January, PM – 11 April)	Inhalation	Exposure potential highly dependent on location during recovery.	Exposure potential highly dependent on type of work conducted during recovery.
		<u>Mitigating factors:</u> - residents minimum separation distance of 500 ft during soil disturbing remedial activities - water suppression actions reduced resuspension potential - access controlled in proximity to craters formed by detonations	<u>Mitigating factors:</u> - water suppression actions reduced resuspension potential for soil disturbing activities - air-purifying respirator use during some activities - practical restrictions of personnel from downwind locations during soil disturbing activities
	Ingestion	Exposure potential highly dependent on location during recovery. Magnitudes of intake generally much smaller than inhalation pathway for ground-deposited plutonium.	Exposure potential highly dependent on location during recovery.
			<u>Mitigating factors:</u> - water suppression actions reduce cross-contamination of clothing during for soil disturbing activities - protective clothing, gloves, booties, and contamination control monitoring
Post-Recovery 11 April and Beyond	Inhalation	Exposure potential highly dependent on locations occupied. Direct support of agriculture in unmitigated contaminated areas, specifically soil disturbing actions, increase potential.	None
	Ingestion	Much smaller than inhalation.	None

pathway is deemed a small contributor to bodily intakes. Members of the Palomares community had the potential for exposure during all three periods considered, however, due to the mitigation measures instituted during the recovery operation, contribution to the total intakes of residents is small. As will be shown in the next section of this report, for those members of the Palomares community present during the transit of the initial cloud plume, exposures from this phase could have been sufficient to explain the entire estimated intake. For the small number of members of the community that were not present the day of the accident, their intakes are most likely attributable to presence and direct participation in agricultural activities, e.g., close proximity during activities that caused soil disturbance. Less potential for exposure is believed to be attributed to similar activities conducted during the recovery phase, as members of the public were restricted from access during these operations and the application of mitigation actions.

In contrast, the only US military members present during the initial plume transport were the four surviving aircrew from the B-52 (DNA 1975). The three that landed in the Mediterranean were brought to a port to the northeast of Palomares. The other surviving aircrew was transported to a local hospital west of Palomares. All four were transported to Torrejon AB, Spain. Major General Wilson, the eventual on-scene commander, flew with two other members from Torrejon AB to the observe the site from the air, about two hours after the B-52/KC-135 collision. Another group of 33 from Torrejon AB arrived at a San Javier, a Spanish Air Base about two hours north-east of Palomares. These two groups along with other personnel travelled by bus from San Javier and arrived at Palomares in the early PM (DNA 1975). Forty-nine US personnel were on-site the day of the accident. Within 7 days, 583 Americans were at the Palomares site.

Limited soil disturbing activities were performed early in the recovery response. As such, resuspension would have limited to foot traffic related to search activities. In review of the information on the high 26 individuals, Table A-4, the majority of personnel were present early in the response; one was present on 17 January. The two individuals with the highest estimated intakes by the CINDY dose modeling code, patients 6 and 21, were present on 18 and 17 January, respectively. Patient 6 had one urine initial sample that was analyzed by the gross α -particle method, with another collected as part of the resampling program - analyzed by α -spectrometry. This patient declined to provide any additional samples. Because of the variability common among samples collected from the same individual, yet separated by only brief periods, the estimated dose for this individual has much higher uncertainty than the other individuals that provided multiple samples. In the case of patient 21, three samples were analyzed by gross α -radiation, while four were analyzed by isotopic plutonium with α -spectrometry. It is clear for this case, why there was a high degree of variability in the estimated intake. The first sample analyzed by α -spectrometry, has an estimated BB of 67%, while the second provided an estimated BB of 3%, and the last two samples being non-detects.

Among early responders, EOD personnel worked in close proximity to (and within) the impact/detonation craters. These areas had the greatest degree of ground contamination and subsequently airborne resuspension. In spite of this, only one of the initial EOD team members was among the high 26. This is likely because air-purifying respirators were used by this group of

personnel. Because of rapidly decreasing resuspension factors for ground-deposited contaminants, the greatest inhalation intakes have an expected attribution to inhalation intakes in the early days of the recovery. While many in the US high 26 group, based on urinalysis results, are above the committed doses observed in Palomares residents, some of the difference may be due to the urine excretion models used. In review of the Iranzo work (Iranzo 1988), it appears their analysis is based on ICRP Reports 26/30/48, similar to estimates made by CINDY (Labat-Anderson 2001).

6.4 Review of Roller Coaster Safety Shots Data in Relation to Palomares Accident.

Operation Roller Coaster was a joint US-United Kingdom non-nuclear research program devoted to studying and better defining the environmental hazards associated with the scattering of plutonium (USA 1965). The tests were conducted on the Tonopah Test Range and the Nellis AFB Bombing Range, NV, in 1963. Four tests were conducted. The first, Double Tracks involved a single nuclear weapon test device, the second Clean Slate I involved nine devices, while Clean Slates 2 and 3 involved devices in an igloo. While each test contained only one device with plutonium, the other devices contained high explosives. The Double Tracks test is the most pertinent as a comparison to the weapons accidents at Palomares. Though the specific details of the test devices used for Operation Roller Coaster and the Palomares accident remain classified, the Double Tracks test remains sufficiently similar to illustrate key issues for this accident. While not critical to the initial contaminants dispersed to the environment, the environmental conditions at the Tonopah Test Range and Palomares are both arid. The wind conditions during the Double Tracks event were lower than those that existed during the Palomares initial plume transport, about 11 knots and 30 knots, respectively. This difference would provide for a more diffuse ground deposition in Palomares compared to Double Tracks. The Roller Coaster studies incorporated extensive air sampling arrays, the majority of which were cascade impactors that allow quantification of the total and respirable fractions of the dispersed aerosols.

The plots in Figures C-1 through C-4 show the reconstructed “concentration time integral” contours from Church *et al.* (1970), based on cascade impactor air samplers. The plots use the term “respirable” which includes only the fraction of the aerosol that encompasses particles with aerodynamic equivalent diameters less than 10 μm , while the other in each set of plots “total air exposure.” The samplers were operated for approximately 45 minutes after the test, with the sampling rate of the cascade impactors, 18 L min^{-1} , being very similar to the inhalation rate of an adult male performing light work. The distances down range from the detonation point were labelled alphabetically from A to R, corresponding to distances from 1,250 to 48,000 ft for the plots. The most extensive sampled range was at ring B, 2,500 feet downrange. From Figure C-1, our interest is related to the concentration time integrals for 10 to 100 $\mu\text{g sec m}^{-3}$. Application of the air sampling rate of 18 L min^{-1} and a specific activity of 0.067 Ci g^{-1} for the plutonium, the projected inhalation intakes for these contours correspond to 12 to 120 nCi. Based on these plots, an individual within these contours would have a predicted respiratory intake within this range, if present for the duration of the air sampling period. This range of intake also corresponds well with estimated intakes for the Palomares residents with positive urine samples (Table C-2). Notably, the projected intake contours would be a little less for Palomares due to difference in wind conditions

between Palomares and Double Tracks. Figure C-5 and C-6 provide distances for the peak respirable and total air exposure for each of the four tests. Figures C-7 and C-8 provide estimated areas encompassed by the respirable and total air exposure. From Figure C-5, the area encompassing 10 to 100 $\mu\text{g sec m}^{-3}$ for the Double Tracks (DT) test is about 8.7 km². This area is about three-fold higher than the extent of the contaminated areas delineated in Figure A-6. These areas encompassed primarily farmland, with some residential homes. The majority of the residences were outside the contaminated areas delineated in Figure A-6. Another important point regarding exposure potential is the fact that it was a holiday the day of the accident; work in the fields was unlikely (Church *et al.* 2000). Hence, under these conditions, it is reasonable to understand why only about 10% of the Palomares population at the time of the accident would have received detectable intakes due to their presence during the initial plume transit. Once the plume had deposited material on ground surfaces, resuspension dynamics is then the factor responsible for airborne radioactive material for later periods.

Figure C-9 contains a plot of aerodynamic equivalent particle diameter vs. cumulative percent plutonium in sampled air for Roller Coaster tests from Friend and Thomas (1965). Particles with aerodynamic equivalent diameters less than 10 μm encompass only about 20% of the total plutonium in the aerosol. These measurements were based on data from ring B, 2,500 ft (0.76 km) from GZ. Figure C-10 provides a plot of the variation of median particle diameter size vs. distance for aerosols generated in the Roller Coaster tests. The plot was reported in Dewart *et al.* (1982). The plot is based on the portion of measured aerosols normalized to the fraction under 10 μm aerodynamic equivalent diameter (AED). While the plot has a primary purpose of estimating deposition characteristics in the lung, which impacts internal dosimetry applications, the plot also provides qualitative information about aerosols characteristics vs. distance from GZ. It is well understood that with increasing distance from the GZ, particles of greater aerodynamic equivalent diameter are more readily deposited closer to the GZ with the aerosol progressively having smaller aerodynamic equivalent diameters with greater distance. Subsequently, the estimate of 20% respirable fraction for a distance of 2,500 ft from GZ, would be lower for distances closer to the GZ, but higher at greater distances.

6.5 Estimated Airborne Concentrations and Inhalation Intakes based on Surface Soil Contamination Levels at Palomares.

Appendix D contains plots that are useful for estimation of inhalation intakes based on surface soil concentrations. The appendix does not provide new information, but rather a synthesis of information already provided in this report. For brevity, no discussion is provided here. Yet some is provided in the Appendix. The synthesized data supports conclusions already drawn on exposure potential for Palomares Recovery workers.

7.0 Assessment of Department of Veterans Administration Claims for Radiation Exposure.

7.1 General.

The seminal event in radiogenic disease compensation occurred in 1977 when the Veterans Administration (VA) Office in Boise, ID, now known as the Department of Veterans Affairs, denied a claim by retired Army Sergeant Paul R. Cooper for service connection of his condition of acute myelocytic leukemia (AML) to radiation exposures he may have received in 1957. Sergeant Cooper participated in Shot Smoky of Operation Plumbbob, an atmospheric nuclear weapon test conducted at the Nevada Test Site (DTRA 2014). The VA decision led to a series of events that ultimately involved the DoD, Department of Energy (DOE), the National Academy of Sciences (NAS), the Department of Health and Human Services, and the White House (DTRA 2014). DoD established the Nuclear Test Personnel Review (NTPR) program in 1978 with an initial task of evaluating exposures from veterans participating in atmospheric testing of nuclear weapons, the majority of which was conducted at the Nevada Proving Grounds (NPG), and the Pacific Proving Grounds (PPG) in the vicinity of Bikini and Eniwetok Atolls.

In 1981, the VA began offering medical care to atmospheric nuclear test participants and veterans that were part of the occupation forces in Hiroshima and Nagasaki (Public Law 1981). In 1982, the Public Health Service (PHS) was tasked with developing radioepidemiological tables for PoC for cancer from radiation exposure (Public Law 1983). In 1984, Congress enacted the Veterans' Dioxin and Radiation Exposure Compensation Act that established standards for compensation of veterans exposed to ionizing radiation from atmospheric nuclear testing and the occupation of Hiroshima and Nagasaki (Public Law 1984). The law also established an advisory committee on environmental matters, and for the NTPR program, it established guidelines for dose reporting. In 1988, Congress provided veteran's with presumptive service connection for a number of cancers to on-site participation in atmospheric nuclear weapon testing and the occupation of Hiroshima and Nagasaki (Public Law 1988). This list includes a number of cancers, but notably did not list chronic lymphocytic leukemia (CLL) and prostate cancer, as these cancers had not been demonstrated to have a causative link to ionizing radiation exposures from studies on radiation-exposed populations. The most important studies considered have been the life-span studies of the atomic bomb survivors of Hiroshima and Nagasaki.

The compensation program for veterans under the NTPR has often been used as a basis for comparison for other radiation-exposed groups. One reason NTPR encompassed a large number of radiogenic cancers was due to the fact that individuals whom supported atmospheric tests of nuclear weapon received the vast majority of their dose from external radiation. The external source of radiation provided reasonably uniform exposures to the internal organs of the body. One exception was the skin, where much higher doses were possible, and primarily due to energetic β -particles emitted by fission product releases from atmospheric tests in air and also deposited on ground surfaces. In contrast to these exposure conditions, internal exposures to plutonium from inhalation, which is key to the exposure potential that existed for the Palomares veterans, are largely limited to the skeleton (e.g., bone surfaces and bone marrow being key), the liver, lung (and other parts of the respiratory tract), and lung-associated lymph nodes. Other tissues of the body receive negligible

doses. These facts have been supported by human and animal studies on exposures to plutonium that date back to the Manhattan Project in the early 1940's.

7.2 Veteran Administration Regulations Applicable to Veteran Exposures to Ionizing Radiation

Title 38, US Code of Federal Regulation (CFR) has two Sections with applicability to ionizing radiation exposure to veterans, §3.309, *Diseases Subject to Presumptive Compensation*, and §3.311, *Claims Based on Exposure to Ionizing Radiation*. As discussed above, veterans meeting exposure conditions and a listed disease are eligible for presumptive compensation. Other veterans with ionizing radiation exposure potential, yet not meeting eligibility requirements of §3.309 can be assessed under the provisions of §3.311. While exposure scenarios under § 3.309 are narrowly-defined, § 3.311 covers any radiation exposure a veteran may receive during the course of his duties.

Assessment of claims under the provisions of §3.311 are reliant upon three key factors: the veteran has a radiogenic disease, the amount of ionizing radiation exposure received by the veteran from his service, and a determination by the Under Secretary of Benefits for the VA that it “is as least as likely as not the veteran’s disease resulted from exposure to radiation in service.” This latter factor is the PoC. Within each of these categories are additional details, as discussed below.

Radiogenic Diseases.

Table 11 contains a listing of radiogenic diseases recognized by the VA under 38 CFR 3.311(b)(2)(i). The majority of conditions recognized are malignancies (e.g., cancer), where occurrence is probabilistic in nature, related to the dose and other biological factors. Induction of malignancies are deemed a stochastic process, as are the induction of genetic effects. Malignancies related to key tissues for deposition and retention of plutonium are highlighted in Table 11. Leukemias are listed, despite the fact that these malignancies have not been linked to animal or human studies of plutonium exposures. As well, lymphomas: acute and chronic lymphocytic leukemia, e.g., ALL and CLL, have special considerations. Hodgkin’s lymphomas are excluded, as were CLL. Recently, there was concurrent action within the Energy Employees Occupational Illness Compensation Program Act (EEOICPA) [ORAU 2012] and the National Institute for Occupational Safety and Health (NIOSH) to add CLL as a radiogenic disease. DTRA modified their standard operating procedures to manage dose assessments for CLL claims (Mannis *et al.* 2013). Lymphomas will be discussed in detail later.

The time of disease on-set is an important consideration for most of the radiogenic diseases, as detailed in Table 11. For most cancers, there is a minimum latency period of five years between the exposure and manifest evidence of disease. Exceptions are for induction of leukemias, which can be manifest at any time after exposure. Primary bone cancers, e.g., most commonly osteosarcomas, must become manifest within 30 years after exposure. The 30-year condition is confounding for assessment of internal exposures to the bone for plutonium, due to its relatively long retention time in bone tissues. The rules listed in 38 CFR 3.311(b)(5)(i)-(iv) were developed in response primarily to ionizing radiation exposure conditions listed 38 CFR 3.309, where doses were

TABLE 11. Radiogenic Diseases, 38 CFR 3.311(b)(2)(i) and (5)(i-iv).

Disease	Conditions	Disease	Conditions
leukemias, except CLL	may become manifest any time after exposure	bone cancer	must become manifest within 30 years after exposure
thyroid cancer	must become manifest 5 years of more after exposure	pancreatic cancer	must become manifest 5 years of more after exposure
breast cancer		stomach cancer	
lung cancer		colon cancer	
liver cancer		kidney cancer	
skin cancer		urinary bladder cancer	
multiple myeloma		prostate cancer	
salivary gland cancers		non-Hodgkin's lymphomas*	
esophageal cancer		ovarian cancer	
non-malignant thyroid nodular disease		parathyroid adenoma	
tumors of the brain and central nervous system		rectal cancer	
any other cancer		posterior subcapsular cataracts	must become manifest 6 months or more after exposure

* Hodgkin's lymphomas excluded. Due to exclusion of CLL, for practical purposes refers to ALL (non-Hodgkin's types). VA currently considers CLL on a case-by-case basis.

incurred over brief periods, and primarily from external radiation sources. Due to the long-term accumulation period of dose to the bone from internally-deposited plutonium, latency period is an ambiguous term. This is an issue for consideration by the VA. Nevertheless, primary bone cancers are very uncommon¹⁷ and have only been associated with high doses, congruent with those associated with medical therapy (Boice 2005).

Another important consideration for the disease on-set period in relation to the accumulation of dose is the case of leukemias. In general, most cases submitted by veterans for compensation related to ionizing radiation exposures are many decades after the exposure period. This exists because most cancer induction is correlated with aging. As such, in assessment of claims for internal emitters, the Air Force commonly reports the 50-y committed doses to organs of interest. As will be shown later, leukemia has a stronger causative link to ionizing radiation exposure for latent periods within 5 years after exposure, but greatly diminishes as latent periods increase. This condition runs in contrast to assumptions made by Beyea and von Hippel (2019), where early on-set leukemias and liver cancers are rated against 50-y committed dose to the RBM and liver, respectively. Within a few years after an acute intake of plutonium, only a small fraction of the 50-y committed dose to the RBM and liver will be realized. Clearly, for early on-set malignancies, 50-y committed dose calculations would be high-sided. Primary exceptions are tissues of the respiratory and GI tracts.

¹⁷ Primary bone cancers account for much less than 1% of all cancers. Osteosarcomas are most common for individuals between ages 10 and 19, while chondrosarcomas are more prominent in adults over the age of 40. (NCI 2008)

38 CFR 3.311(b)(2)(i) also provides for, “any other cancer.” The primary reason for this addition is based on the history of radiogenic disease compensation for atomic veterans in the early 1980’s and National Institutes for Health Radio-Epidemiological Tables developed in 1985. These tables, based upon the Japanese A-bombs survivor cohorts, included prominent cancers, but expanded over the years with additional cancer types. From a practical standpoint, due to the highly-specific tissues for deposition and retention of plutonium, other cancers are not likely to provide sufficient dose for favorable compensation decisions. For exposures from external radiation sources where reasonably uniform exposures to the internal organs of the body occur, the provision for “any other cancer” may be important. Significant external radiation exposures were not possible for Palomares recovery workers; in fact, they were negligible.

A number of conditions recognized by the VA as radiogenic, excluding malignancies or genetic effects, are termed “deterministic effects.” These conditions have historically been attributed to a threshold below which the effect is not observed. The threshold for these effects, though is varied by the individual and condition. The two deterministic effects listed in 38 CFR 3.311(b)(2)(i) are thyroid nodular disease and posterior subcapsular cataracts. Due to limited deposition and retention of plutonium in the thyroid, this condition is not likely related to the Palomares cohort. Cataract induction is related to very-high, shallow-tissue external doses - most commonly from β -particles emitted in fission products decay. This condition is also unlikely related to Palomares radiation exposures. Claims for these conditions are evaluated by the VA.

In addition to the 21 enumerated 'radiogenic diseases' in 38 C.F.R. 3.311(b)(2), VA will apply the procedures in that regulation in claims for "polycythemia vera" and any other disease if the claimant presents competent scientific or medical evidence that the claimed condition (including polycythemia vera) is a "radiogenic disease." 38 CFR 3.311(b)(3)-(4). Deterministic effects, in contrast to stochastic ones, are based on loss of function in a tissue based on a large loss of cells (death) from radiation exposures. For stochastic effects, cells must be modified, yet not killed. Because a large loss of cells is necessary for these effects, these are deemed threshold effects, below which the effect is not observed. Above the threshold, the effect in tissues will have a steep rise in the observation rate, until the dose level reaches the point where the observation rate is 100%. The variation in the dose required to observe the effect is largely based on biological variations. Deterministic effects in the bone were summarized by Rowland (1994) for radium dial painters that received very-high intakes of radium, which concentrates in the bone. Bone necrosis was found in a small group of the studied population of individuals, where the condition was primarily attributed to degradation of blood circulation, yet limited to subjects with ^{226}Ra ingestion intakes greater than 85 μCi (85,000 nCi), when modified with updated methodology for estimating intakes (Rowland *et al.* 1994). Other cases of bone-related deterministic effects from individuals with high intakes of radium were anemia and leukopenia. These cases, nevertheless were related to very-high intakes, where dose to the RBM was substantially higher than expected for any Palomares recovery worker.

Lung fibrosis, as related to high acute intakes of plutonium by the inhalation pathway, was observed in some plutonium workers, yet only for those workers with high-accumulated doses to the lung. In a study of US plutonium workers, Newman *et al.* (2005) reported that for accumulated

doses to the lung in excess of 1,000 rem lung fibrosis could be observed¹⁸. Similar findings were made in the medical follow-up study on MPA plutonium workers (Azizova *et al.* 2020). The MPA worker study involved the review of 188 workers with lung fibrosis, with the median cumulative equivalent dose to the lung from plutonium about 9,000 rem (Azizova *et al.* 2020). The threshold lung CDE for causing this effect is well above that for Palomares recovery workers. For example, a 34 nCi inhalation intake provides a 50-y CDE to the lung of 40.8 rem using ICRP 30/48.

Maher (2020) noted that deterministic effects are uncommonly compensable for EEOICP cases; virtually all of the claims are based on malignancies. A notable exception are non-malignant respiratory conditions, i.e., pneumoconiosis and fibrosis of the lung, but only for individuals that had jobs in uranium mining, milling, and ore transport, where dust loading in the lung could be significant, yet not related to ionizing radiation. In general, the same scientific bases are considered for these and VA radiation exposure cases. ICRP Report 103 (ICRP 2007) noted that despite updates in scientific data on radiation effects, the overall estimate of deterministic effects remained the same as published in ICRP Report 60. ICRP found little evidence of any excess risk of non-cancer disease below 1 Gy (ICRP 2007). For alpha-particle dose to tissue, this would be equivalent to 2,000 rem, though ICRP prefers use of dose values for deterministic effects.

Radiation Dose

The **probable dose**, in terms of the dose type, rate and duration as a factor in inducing the disease taking into account any known limitation in its measurement [38 CFR 3.311(e)(1)] is key for assessment of claims based on exposure to ionizing radiation. Commonly for occupational exposures to radiation, 38 CFR 3.311(a)(2)(iii), *Other Exposure Claims*, a review of available records is made, where historically dose information was documented on a DD Form 1141, *Record of Occupational Exposure to Ionizing Radiation*. Over the last few decades, DoD services have transitioned to other forms. Ionizing dosimetry information documented on the DD Form 1141 was generally limited to external dosimetry monitoring data. Because the DoD possesses dosimetry, detailed information on exposures circumstances, and other records vital to assessment of exposure potential, it has been a practice for DoD services to assist the VA in meeting its responsibility under 38 CFR 3.311 in “preparation of a dose estimate, to the extent feasible, based on available methodologies.” Some examples of DoD support to the VA:

a) Defense Threat Reduction Agency (DTRA)¹⁹ extensive publications on doses to personnel supporting US atmospheric tests of nuclear weapons.

b) DTRA and Joint DoD services evaluation of doses to military members and civilians, and dependents in Japan and on the US Ronald Reagan Carrier Group during the Fukushima Daiichi Nuclear Power Station (FDNPS) accident. Primary dose assessments are contained in Cassata *et al.*

¹⁸ The values were listed in units of dose in the article. The application of a radiation-weighting factor, w_R of 20 was applied to allow ready comparison to dose equivalent (ICRP 26) and equivalent dose (ICRP 60 and 103) values used in this report. w_R values are designed for use in stochastic effect endpoints.

¹⁹ Formerly Defense Special Weapons Agency (DSWA) and Defense Nuclear Agency (DNA).

2012 and Marro *et al.* 2014, with numerous additional supporting documents available to the public on the Department of Defense Environmental Health Surveillance Registry. The US Army Public Health Center address specific veteran queries for the DoD.

c) Personnel assigned to Johnston Atoll because of exposures to low-level residual plutonium from aborted missile launches during Operation Dominic I. Report completed by Rademacher (2016) provides methodology for estimation of dose to personnel assigned to Johnston Atoll from 1963, shortly after missile mishaps and outside the scope of the Nuclear Test Personnel Review (NTPR) coverage, until 2003, when use of the Atoll by the DoD was completed. Though the report was written to support AF estimates of doses to its personnel, the methodology was deemed useful for assessment of dose to other personnel assigned to JA.

d) Clean-up of Enewetak Atoll conducted by multi-service team, Department of Energy personnel and contractors. Two dose assessment reports were recently completed. Rademacher (2019) provides a summary and methodology for assessment of doses to AF personnel, as did DTRA (McKenzie-Carter *et al.* 2020) for 6,000 military personnel.

While many VA claims are readily assessed based on external dosimetry records, the examples listed above are more complicated for a number of reasons. A key factor is the necessity to assess internal dose, which is also an important issue for Palomares recovery workers and Johnston Island workers. For atmospheric testing participants, complicating issues also involve some internal dose aspects, but more importantly external dose issues. Some external dose issues are related to biases in early film badge dosimetry, while others are related to the application of team dosimetry; some personnel were monitored as part of a team, where a single dosimeter was assigned to one team member. For the same participants, greater uncertainties were also related to varied external dose rates produced shortly after detonations in environments accessed by personnel, as compared to some sites, where exposure conditions were constant, e.g., Enewetak Atoll during the 1977 to 1980 cleanup. To address this issue for cases where specific dosimetry information may not exist for a veteran, it has been a common practice for the DoD in its dose assessments to perform “upper-bound dose estimates.” This practice is designed to meet the intent of 38 CFR 3.311(a)(1), “When dose estimates provided pursuant to paragraph (a)(2) of this section are reported as a range of doses to which a veteran may have been exposed, exposure at the highest level of the dose range reported will be presumed.” In common terms, these estimates are often referred to as “high-sided” or “conservative.”

Many methods are employed in dose assessments to ensure dose estimates are “high-sided.” When well known distribution data is involved, it is common to use the 95th percentile of these distributions. This was employed in the analysis of some data in the dose estimates for the Fukushima cohorts and Enewetak. Other methods for exposure conditions where work was conducted over long periods, it is common to assume full work days within an exposure zone, though in reality for some fraction of the day, workers are travelling between staging points and work areas, involved in contamination control monitoring and decontamination, and taking work breaks. For Fukushima dose estimates, exposure rates observed in outdoor locations were used for estimates of dose for entire days, while in reality indoor environments would have had much lower

dose rates due to shielding afforded by building structures. Because outdoor temperatures commonly peaked only to the high 40°F in Tokyo during this period, this assumption is very high-sided. As discussed above, Labat-Anderson (2001) in their estimate of exposure potential made an assumption of an 11-week exposure period, with full exposure in every day for Palomares recovery workers. This unrealistic assumption is clearly high-sided. Another common assumption is application of dose to internal organs, based on external dosimetry results, whereas in reality self-attenuation of the body will afford lower doses to internal organs.

There are two key points to be made regarding “high-sided” dose estimates:

a) In the course of the dose assessment process, numerous high-sided features can be introduced, when combined, they can provide an estimate of dose that is well beyond reason, being in reality “improbable.” This is in direct conflict with the charge made in 38 CFR 3.311(e)(1) to evaluate PoC where “probable dose” is a key factor.

b) The PoC methodology commonly incorporated by the VA also incorporates, “high-sided” uncertainty factors that dwarf the uncertainty factors introduced in the assessment of probable dose. This issue is discussed in the next section.

38 CFR 3.311(a)(3), *Referral to independent expert*, provides a mechanism to “reconcile a material difference between an estimate of dose, from a credible source, submitted by or on behalf of a claimant, and dose data derived from official military records.” The Director of the National Institutes of Health (NIH) selects independent experts to prepare separate radiation dose estimates.

Probability of Causation

Probability of causation considers probable dose and the following additional factors:

- a) the relative sensitivity of the involved tissue,
- b) the veteran’s gender and pertinent family history,
- c) the veteran’s age at the time of exposure,
- d) time-lapse between exposure and onset of the disease,
- e) background cancer rates at age of onset, and gender-specific,
- f) ethnicity for skin cancer evaluation,
- g) smoking history, and
- h) the extent to which exposures to radiation, or other carcinogens, outside of service may have contributed to development of the disease.

The VA Undersecretary of Benefits considers the factors noted above, with sound scientific and medical evidence to determine if:

“at least likely as not the veteran’s disease resulted from exposure to radiation in service.” 38 CFR 3.311(c)(1)(i)

Some radiogenic disease compensation claims handled by the VA are evaluated by the Interactive RadioEpidemiological Program (IREP) computer software (Kocher and Apostoaei 2007). The IREP code used by the VA was developed by the NIH, an agency currently in the Department of Health and Human Services (HHS). A similar IREP code was later developed by NIOSH, also part of HHS. The NIOSH code was similar to the existing NIH code, though it was developed for use by the Department of Labor (DOL) in assessment of claims under the EEOICPA. In the mid- to the later 2000’s, the VA began using the NIOSH version of IREP for most medical opinions (Otchin 2007). IREP calculates a PoC distribution for specific cancer induction sites, based on radiation exposure estimates. PoC values vary by age at exposure and the time between the age at exposure and malignancy diagnosis, e.g., latency or onset period. Land *et al.* (2003) define the “probability that a cancer in an individual was caused by prior exposure to ionizing radiation” - PoC as:

$$PoC = R / (R + B),$$

where R is the probability of incurring a specific cancer due to the radiation exposure and B is the baseline (background) probability of incurring the cancer, absent the radiation exposure. Table B-13 lists recent lifetime cancer rates for the US population, as an illustration. The values used by NIOSH are likely to have some differences. One important difference for any background cancer rate estimate is correction of population statistics for smoking habits, an important factor in lung cancer induction, and to a lesser degree other cancers. As well, the baseline risk is age-dependent; Table B-13 lists estimated lifetime cumulative rate.

IREP calculates PoC on an “estimate of the excess relative risk (ERR) associated with a given radiation dose to an organ or tissue in which a cancer occurred” (Kocher and Apostoaei 2007). ERR values are based on estimates obtained from epidemiological studies of populations exposed to radiation. The Japanese atomic bomb survivor follow-up studies have been a primary source of risk data. IREP applies a radiation effectiveness factor to the type of radiation absorbed in the organ of interest. It is important to note CED values use radiation weighting factors from ICRP Report 26²⁰, 60, and 103, which uses a factor of 20 for the absorption of α -particle energy in tissue. IREP uses a probability distribution of REF for α -particles, as compared to high-energy photons. For individuals using CED values from this report, it is reasonable to assume that the vast majority of CED is attributed to the absorption of α -particle energy. Over 99.8% of the energy emitted in the decay of ²³⁹Pu and ²⁴⁰Pu is due to α -particles. Hence, dose values are simply 1/20th of the CED.

²⁰ CDE for ICRP 26.

Table D-1 contains example PoC calculations for key cancer induction sites from inhalation of insoluble forms of plutonium: lung, liver, and bone. Because of the variability among the approximate 1,600 Palomares recovery workers, the table is specific only to individuals born in 1946, being about 20 years old during the recovery. For brevity, the table only contains a few latency times: 12, 22, 32, 42, 50, and 52 years. As noted previously primary cancers of the lung, liver, and bone in adults are have expected onset later in life. IREP includes the ability to introduce uncertainty in dose in addition to that associated with the individual tissue and organ risk coefficients. Table D-1 only includes the assumption of 10% coefficient of variation (CV) in dose, using a normal distribution. Cases 1 to 9 are applicable to lung cancer.

Case 1 provides an example PoC for a case meeting the 38 CFR 3.311(c)(1)(i) “*at least likely as not*” criterion for radiation exposure to a veteran with lung cancer that never smoked. This is equivalent to the PoC value of 50% at the 50% credibility level (CL). The effective dose for this case is 285 rem, based on a 42-y latency (onset delay). For the Table, four other CL are observed: 1%, 5%, 95% and 99%. The higher the CL, the greater degree of favorability is granted to the veteran. This provides “benefit of doubt” in the presence of uncertainty. Although not dictated by law, the VA commonly grants compensation when the probable dose exceeds the PoC of 50% at the 99% CL. The DOL provides a similar approach for EEOICPA cases. Case 2 provides an example of calculations for this case, but with the same age of exposure, latency period, and cancer condition as Case 1. The difference in effective dose between these two cases is 14-fold. Importantly, the use of “high-sided” estimates of veteran’s doses, this well meets the “benefit of doubt” intent of the VA.

The other seven lung cancer cases illustrate variations in PoC calculation in IREP for latency period and smoking status. Cases 3 through 7 use all of the same conditions as Case 2, except latency period. Cases 3 and 4, latency of 52 and 32 years, respectively, have the same CED as the 42-year of Case 2. Cases 5 and 6, latency of 22 and 12 years, respectively, have lower dose levels, 18 and 10.8 rem. For the short latency periods, cumulated effective dose will be somewhat lower than the 50-y CED for the lung, while this is less important for the longer latency periods. Cases 7 through 9 are the same as Case 2, except the smoking status: former smoker, 10-19 cigarettes/day, and 20-39 cigarettes/day, respectively. For a former smoker, the 50% PoC at the 99% CL is 30.8 rem, about 50% higher than the case for a never smoker, while the current smokers are 33.5 and 34 rem, for Cases 8 and 9, respectively.

Cases 10 through 14 provide example PoC values for primary cancer of the liver. Case 10 is for a PoC value of 50% at the 50% CL, where the effective dose is 92.4 rem. Case 11 has all conditions the same as Case 10, except for a 99% CL. For this case, the effective dose is 4.9 rem, nearly 19-fold lower. As shown by cases 12 through 14, no difference is observed with Case 11 for latencies from 22 to 52 years. An important observation for the effective dose in these cases is the fact that 4.9 rem is below the annual dose limit for whole-body occupational exposures, 5 rem, observed by the NRC, DoD, and DOE. For myeloid leukemias, e.g. AML and CML, for short latency periods, dose levels can be even lower (Kocher and Apostoaei 2007).

Cases 15 and 16 shown in Table D-1 are for primary cancers of the bone. Case 15 is for a 42-y latency, with a PoC at 50% and a 50% CL, where the effective dose is 277 rem. As compared to Case 16, with similar provisions, except a 99% CL, the effective dose is 24.9, 11-fold lower.

Some other examples are provided for the urinary bladder, kidney, nervous system tissue, and CLL. While not tissues of concern for internal plutonium exposures, they provide additional information for the reader. The urinary bladder and kidney have similar dose values as the bone, while nervous system tissues and CLL have extremely-high dose levels at the 50% CL, and 1,030 and 2,050, respectively. These are reflective of the relatively weak link of ionizing radiation exposure to induction of these cancers. A striking feature is the ratio of the dose levels for CLL: 38:1 for 50 to 99% CL, whereas, the ratios are substantially lower for other cancer sites. The ratio is a reflection of a high degree of uncertainty in epidemiological data.

In summary, primary cancers in key induction sites are afforded significant “benefit of doubt” through use of a 50% PoC at the 99% CL in IREP.

IREP and Linear Risk Coefficients for Solid Cancers – the MPA Cohort Experience

Calculations of excess relative risk (ERR), as used in IREP for solid cancers, follow a linear function of dose:

$$ERR = \alpha D,$$

where α is the risk coefficient in units of inverse dose, and D is dose (Kocher 2007). For leukemias, IREP uses a linear-quadratic function of dose (Kocher 2007):

$$ERR = \alpha(D + D^2).$$

Since the primary cancers in key induction sites are solid tumors, the linear model is of interest here.

Animal studies confirmed induction of primary lung, liver, and bone cancers from inhalation exposures of insoluble forms of plutonium (IARC 2012), with an extensive history detailed in Stannard (1988). Studies on US and British plutonium workers did not find statistically significant differences in cancer induction risks (ATSDR 2010). Frankly, the US and British plutonium worker health studies were limited in statistical power because the workers did have significant body burdens and the total number of workers were limited (Sokolnikov *et al.* 2008). The study of workers at the MPA demonstrated statistically significant increases of primary cancers of the lung, liver, and bone based on plutonium intakes, after adjustments for confounding risk of external radiation exposure. No increased risk of leukemia was noted among the cohort of workers exposed to plutonium (IARC 2010). In comparison of US and British plutonium workers to their Soviet counterparts, the difference in epidemiological finding was of no surprise when one compares the residual $^{239+240}\text{Pu}$ activity burden in the liver from autopsy study of workers. Figure 10 shows a comparison of US and MPA plutonium worker liver burdens from autopsy. The median concentration of $^{239+240}\text{Pu}$ among livers studied in MPA workers was about 400-fold higher than among the livers studied in USTUR cases (USTUR 2012), though there is some overlap in the data.

The MPA worker studies are the most valuable for occupational exposures to plutonium, with applicability to other plutonium workers and those involved with the Palomares recovery. Though these studies confirmed long-held scientific evidence of risk for induction of lung, liver, and bone cancers from animal studies (Wilson *et al.* 2010), the risk coefficients are of particular interest to this work. These studies are particularly important because they involve identical exposure conditions, not being reliant upon comparison to acute external radiation exposures that formed the basis for Hiroshima and Nagasaki atomic bomb survivor-based risk coefficients. The MPA worker cohort has two important confounding factors: a high degree of smoking among MPA male workers and high intakes of alcohol, of which are important to cancer risk estimates from plutonium to the lungs and liver, respectively. Most germane to this work, the assumption of linear risk coefficients may not be appropriate at low cumulative effective doses to the lung, liver, and bone.

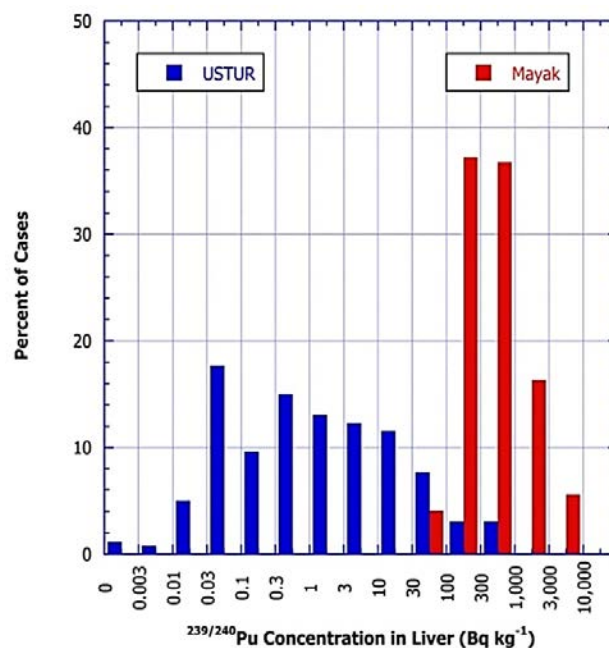


Figure 10. $^{239+240}\text{Pu}$ Concentration in Liver Compared for USTUR and MPA Workers [Figure 14, USTUR (2012)].

a) Lung cancer. Lung cancer was found to have a statistically significant increase in the relative risk of lung cancer with increasing cumulative dose to the lung from α -radiation for doses in the range of 0.1 – 0.2 Gy for MPA workers when corrected for smoking (Labutina *et al.* 2013). At this range and above, the relationship was linear. In terms of effective dose, as used by the ICRP for α -particle emitters, this range would be 200 – 400 rem. Similar findings were previously documented by Tokarskaya *et al.* (1997), where the authors noted, “the dose-response relations for plutonium incorporation is more intricate: it has a non-linear threshold character.” Additional work was conducted by Tokarskaya *et al.* (2002) on the MPA cohort, where the authors concluded that “low-dose risk estimates for radiation-induced lung cancer derived without adjusting for the influence of

cigarette smoking could be greatly overestimated.” A more recent pooled analysis of MPA and British plutonium workers found a linear correlation between lung cancer and plutonium burdens (Gillies *et al.* 2017). The study, however, did not correct for smoking habit.

b) Liver cancer. Labutina *et al.* (2013) found a statistically significant correlation between plutonium body burdens and liver cancer, but no correlation to smoking habit. Both a linear-quadratic and quadratic risk model fit the data better than a linear risk model.

c) Bone cancer. Labutina *et al.* (2013) noted that there were only two cases of malignant neoplasms of bone and associated connective tissue with respect to internal dose to bone surfaces with doses above 1 Gy, 200 rem using an ICRP radiation weighting factor of 20 for α -particles. The analysis by the authors noted increasing risk with increasing dose, yet the trend was not statistically significant. The limited observation of bone cancers among the MPA cohort is related to the relative insensitivity of the bone from induction of sarcomas from internally-deposited radionuclides. Raabe (2012) notes a practical threshold in studies of ^{226}Ra and ^{239}Pu injected into animals, and follow-up studies on US and British radium dial painters. IARC (2000) notes, “Tissues that are apparently less susceptible or in which cancers are induced only at relatively high-doses include the brain, **bone**, uterus, skin, and rectum. Some cancers have not been linked convincingly to exposure to radiation; these include CLL, Hodgkin disease, multiple myeloma, non-Hodgkin lymphoma, and cancers of the cervix, testes, prostate, pancreas, and male breast.”

In summary, it is clear that the assumption of a linear risk coefficient for induction of primary cancers of lung, liver, and bone, is conservative in its use in IREP for PoC values of 50% at the 99% CL. This assumption also provides additional “benefit of doubt” favor to veterans, though it is embedded within the IREP methodology.

8.0 Air Force Evaluations of Palomares Recovery Worker Doses

8.1 2001.

The first significant effort conducted by the Air Force in assessment of Palomares recovery workers since the 1968 assessment of urine results from recovery workers was contracting Labat-Anderson, Inc. to re-evaluate the urine results among other tasks, discussed above. In addition to this work, the AF/SG and the Air Force Institute for Environment, Safety and Occupational Health Risk Analysis evaluated modern methods of urine bioassay. These initial efforts were due to veteran claims for compensation due to occupational exposures to radiation. Prior to this time, the AF/SG infrequently received inquiries on radiation exposures received by veterans. The vast majority of DoD veteran claims based on radiation exposure were managed by DTRA for veterans eligible under the NTPR program.

8.2 2005.

The Air Force Safety Center (AFSEC) began assisting the AF/SG in assessment of veteran radiation exposure claims from materials related to nuclear weapons and testing. Prior to this time,

AFSEC primarily assisted AF/SG in claims related to veterans performing nuclear weapon maintenance work. In assessment of radiation exposure claims for Palomares recovery workers, the AFSEC evaluated:

- a) a veteran's presence and type of support to the recovery,
- b) urine sample results, as documented in Labat-Anderson (2001),
- c) air samples collected by 16th Air Force during the recovery (summarized earlier in this report),
- d) the type of medical condition reported by the veteran to the VA²¹, and
- e) exposure potential based on the extensive monitoring of Palomares residents that were present during and after the aircraft mishap, and the levels of airborne contamination based on activity concentrations in the soil surfaces (see Figure A-6). These details have been noted previously in this report.

In 2005, AFSEC also began evaluation of other potential occupational exposures to AF veterans from plutonium:

- f) personnel assigned to Enewetak Atoll between 1959 and 1973, and during the 1977 to 1980 cleanup (Rademacher 2019),
- g) personnel assigned duties on Johnston Atoll after 1962 (Rademacher 2016),
- h) personnel participating in the recovery actions from the 1968 Thule AB nuclear weapons accident,
- i) personnel participating in the recovery actions from the 1960 McGuire AFB nuclear weapons accident involving a nuclear-tipped, BOMARC missile (Rademacher 2009), and
- j) a few additional suspected exposure categories.

In assessment of Palomares recovery worker VA claims, AFSEC would adopt the estimated dose provided by Labat-Anderson (2001), if a veteran was within the high 26 group of veterans. Assessments of dose would necessitate additional analysis, if one of these individuals had induction of a malignancy within a tissue not covered by ICRP 26/30. It is notable, that AFSEC has only evaluated two cases of veterans in the high 26 group: one had a malignancy in an organ covered by ICRP 26/30, while the other did not have a one of the 21 enumerated radiogenic disease in 38 CFR 3.311(b)(2)(i). For the former, the dose values for tissues covered by ICRP 26/30 were recommended to the VA, as listed by Labat-Anderson (2001).

²¹ AFSEC always pays keen attention to those conditions deemed related to internal plutonium exposures.

AFSEC has assessed less than two dozen total Palomares responder veteran cases. Three of the cases have been members of other DoD services. For the cases where veterans had on-site presence, yet they were not within the high 26 group of veterans, AFSEC adopted the method of assigning an intake based on the lowest estimated intake among the high 26, based on the Labat-Anderson (2001) report. As noted previously, and shown in Table A-4, 34 and 19 nCi intakes were applicable to this approach, dependent on the modeling (Labat-Anderson 2001). AFSEC followed this approach based on numerous factors documented in Table 12.

As detailed in the Table, AFSEC implemented measures to maintain the integrity of the dose estimates provided to the high 26 individuals, as these values were already published in the Labat-Anderson report for 26 veterans. As primary cancers of the lung, liver, and bone originated in organs covered by the ICRP 26/30 methodology, AFSEC recognized that if a veteran had one of these conditions, there would have been a consistent assessment of dose. AFSEC recognized that the dose estimates for these organs may be sufficient for favorable VA compensation decisions, though it did not conduct an analysis. Nevertheless, AFSEC does not estimate doses based on this factor, nor does it make compensation recommendations to the VA; the VA has sole authority for these decisions.

For all veterans with on-site participation, regardless of whether they were part of the high 26 or not, AFSEC was limited in their ability to estimate doses for some organs or tissues that were not detailed by the ICRP 26/30 methodology. In these cases, the ICRP 60/68 methodology was used, as this set of ICRP recommendation had provision for dose estimates for a larger group of organs. It is important to note that the DOE for EEOICPA cases uses the ICRP 60/68 methodology and that this methodology is currently being used in the DoD dose estimates for other groups, e.g., cohorts of US personnel from the FDNPS accident, veterans with duties on Enewetak Atoll prior to and during the 1977 – 1980 cleanup, and others. It is important to recognize that due to changes in the ICRP metabolic models, differences will exist in the calculated doses. This fact was recently pointed out by Beyea and von Hippel (2019). Nevertheless, as noted in the Table, the dose estimates for organs that are not covered by ICRP 26/30 are too low based on expected intakes and not likely to provide usable data to the VA for claim adjudication. Most of these organs have DC values over two orders of magnitude (100-fold) lower than the key organs at risk from inhalation exposure to plutonium, and are also commonly related to cancer types which have not been convincingly linked to exposure to radiation (IARC 2000).

8.3 2013.

In the FY 2014 National Defense Authorization Act §1059, the Secretary of the Air Force was directed to submit to the Committees on Armed Services of the Senate and the House of Representatives a report on “the implementation of the recommendations of the Palomares Nuclear Weapons Accident Revised Dose Report released by the Air Force in April 2001.”²² A reasonable

²² A reasonable number of claims by AF veterans to VA are accompanied by Congressional interest on an annual basis. This Act of Congress was initiated by one Palomares recovery veteran, yet was seeking information applicable to all Palomares recovery veterans.

TABLE 12. AFSEC Approach for Use of Lowest Estimated Intake Among Palomares Recovery Worker, High-26 Documented in Labat-Anderson (2001).

Provision	Rationale(s)
Assignment of Labat-Anderson dose estimate for high 26 Palomares recovery veterans.	<u>Cancer site at tissue covered within ICRP 26/30.</u> Assignment of tissue/organ dose, as documented in Labat-Anderson. Although ICRP published updates in internal dose methodology, AF continued to use ICRP 26/30, based on desired consistency with 10 CFR Part 20 for AF operations under NRC rules.
	<u>Cancer site at tissue not-covered within ICRP 26/30.</u> Assignment of tissue/organ dose based on updated ICRP, e.g. ICRP 60/68. 1) Use of alternate ICRP methodology deemed appropriate due to lack of ability to address all organs and tissues within ICRP 26/30. 2) Used estimated inhalation intake equivalent for use in ICRP 60/68 method, based on the estimated intake from ICRP 26/30 in Labat-Anderson (2001). Some differences existed between the estimated intakes for high 26 individuals between the two methods illustrated in Labat-Anderson, and summarized in Table A-4. 3) AFSEC recognized the differences in the estimated intakes between the two methods. The differences were deemed to be relatively small and have no impact on final adjudication of claims, as ICRP 26/30 organs contained the lung, liver, and bone. Among the four sets of ICRP recommendations, as applied to plutonium, there are variabilities in estimated doses to the metabolic organs. Cancers originating within other organs, as illustrated within the discussion of all sets of ICRP recommendations, are unlikely to provide favorable compensation decisions because intakes were too low to produce sufficient dose.
	<u>Other cases.</u> Other methods of dose assessment may be used to provide a dose recommendation to the VA for unique cases. Because of experience addressing 1,000+ VA claims, most of these cases are expected for non-radiogenic medical conditions.
Assignment of Labat-Anderson dose estimate for other than high 26 Palomares recovery veterans.	<u>Intake estimates.</u> 1) Evaluate if a veteran had on-site presence, based on submission of a urine sample, documentation in veteran records, or other source of information. Based on review of some cases, veterans provided material support to the recovery, but did not have on-site presence. 2) Recommend a plutonium inhalation intake of 34 nCi. This level of intake was based on the lowest intake among the high 26 defined in Labat-Anderson (2001) using the ICRP 26/30 method. a) Estimated intake was deemed high-sided, based on levels of plutonium contamination in soils, accepted resuspension rates, mitigation methods, e.g., targeted air-purifying respirators and water suppression, air sampling conducted during the recovery, and estimated body burdens of Palomares residents. (Environmental Evidence)

TABLE 12. AFSEC Approach for Use of Lowest Estimated Intake Among Palomares Recovery Worker, High-26 Documented in Labat-Anderson (2001), continued.

Provision	Rationale(s)
Assignment of Labat-Anderson dose estimate for other than high 26 Palomares recovery veterans.	<u>Intake estimates, continued.</u> b) The high 26 individuals had urine excretions that represented an estimate of systemic body burdens in excess of the 95 th percentile of for recovery all workers. The high 26 was based on a urine resampling program that included in excess of four-hundred workers. The twenty-six individuals is approximately 5% of the 400+ individuals, in essence the 95 th percentile. The 400+ plus individuals selected for resampling were among those with the greatest potential systemic body burdens among the nearly 1,600 initial urine samples collected. AFSEC recognized that use of the Langham urine excretion model was not as accurate for inhalation exposures ²³ compared to urine excretion models developed later. Nevertheless, these initial screening samples were effective at choosing those individuals with the greatest potential for intakes. Effectively, the combination of choice selection of the highest 400+ from initial screening and the high 26 from the resampling effort provided an estimated intake greater than the 95 th percentile. As discussed above, the 95 th percentile is a common bench-mark adopted for “high-sided” dose estimates. c) It was recognized that for the individual, there could be some variability in samples day-to-day. However, the process of selecting 26 individuals among over 400+ individual participating in the urine resampling program was robust to this source of uncertainty. d) Urine sample collected a number of months after an acute exposure are expected to provide less error in estimate systemic body burden that those collected shortly after a suspected acute intake. e) Due to the high-degree of cross-contamination identified in the initial screening urine samples, they were deemed of lesser credibility in estimates of intake than the samples collected in the resampling program. This was noted in Labat-Anderson (2001)
	<u>Cancer site at tissue covered within ICRP 26/30.</u> Use an estimated intake of 34 nCi with use of ICRP 26/30 dose conversion coefficients, based on the ICRP 48 updates.
	<u>Cancer site at tissue not-covered within ICRP 26/30.</u> Use an estimated intake of 34 nCi with use of ICRP 60/68 dose coefficients. Similar to discussion above.
	<u>Other cases.</u> Other methods of dose assessment may be used to provide a dose recommendation to the VA for unique cases. Because of experience addressing 1,000+ VA claims, most of these cases are expected for non-radiogenic medical conditions.

²³ Odland (1966) documented this same scrutiny, yet it was noted that these initial screening sample would have effectively screened those with exposures and those without.

fraction of claims by veterans to the VA are accompanied by Congressional interest on behalf of a constituent. In response to Congressional requests, staff from AFSEC and the AF/SG discussed key issues. The key issues discussed was the methodology used by AFSEC to estimate plutonium intakes, the possibility that current day urine bioassay could be used provide greater confidence in previous estimates, and the history of dose estimates for Palomares recovery veterans.

A summary of conclusions and path forward was provided by the AF/SG to the VA (Ashworth 2013) and to Congress (Travis 2014). Key items noted in these documents are summarized:

a) The AF would report doses using methodologies from ICRP 26/30 and 60/68. This was deemed of necessity due to precedent established in Labat-Anderson (2001), but the necessity of the newer ICRP recommendations when ICRP 26/30 did not meet the AF's obligation to the VA in providing dose estimates.

b) Use an intake range of 1,100 to 34,000 pCi (1.1 to 34 nCi) for remaining responders that were not within the high 26. AFSEC recommended a range of intakes to accommodate assignment of intakes to responders where no evidence existed for on-site exposure potential. These estimates would be appropriate for some small groups of individuals, e.g., Navy personnel that transported supplies to local ports, yet did not have on-site exposure potential and Navy personnel that were in vessels supporting the search for weapon #4, which was eventually discovered in early April. For individuals that submitted urine samples, yet were not within the high 26, it has been the practice to assign 34 nCi intakes to these individuals.

c) The AF rectified some dose estimates among the 22 Palomares recovery veteran cases received by the AF/SG to date. Some of the early Palomares recovery veteran cases evaluated by the AF used ambient air monitoring results, yet did not provide organ-specific doses. The 22 veterans whose dose estimate was affected by the updated were notified. Though these exposures may have been inferred from the environmental data referenced in Labat-Anderson (2001), the two organizations had extensive discussion of the additional environmental data that was pertinent and summarized in this report, in addition to the results of the monitoring program on Palomares residents.

d) The AF declined to recommend an extensive urine resampling program, though:

- i) the current methods are significantly more sensitive than the methods used in the mid- to latter-1960s and
- ii) individuals with plutonium intakes well in excess of typical background intakes would continue to have detectable excretions many decades later.

This recommendation was based on a number of factors:

- iii) dose estimates for the high 26 and for individuals with "high-sided" estimated

intakes of 34 nCi would likely receive a favorable compensation decision for primary cancers of the lung, liver, and bone,

iv) due to the conservative methods used to estimate plutonium intakes, it was believed that current urine bioassay would not provide any additional benefit for a favorable compensation decision for lung, liver, and bone cancer; to the contrary, the results from current bioassay for individuals with these cancers, would more likely debase the “high-sided” estimated intake, and

v) individuals with cancers that were not related to deposition and retention in the lung, liver, and bone would not have any reasonably-expected benefit from current bioassay, as these cancers are highly unlikely among any of the cohort due to the relatively poor deposition and retention in other tissues, and subsequently low cumulative dose in the organ(s) of interest. Hypothetical intakes would have to be at least a couple of orders of magnitude higher than deemed reasonable for the exposure conditions during the recovery actions to acquire a favorable PoC for many of these cancer types. Further, some cancer types have not been convincingly linked to radiation exposure. Despite the unlikelihood of a sufficient plutonium intake to produce a favorable compensation decision, under 38 CFR 3.311(a)(3), the VA could assess a present day urine sample analysis for a veteran, as recommended through “referral to an independent expert.”

AFSEC developed a plot to illustrate the potential sensitivity of α -spectrometry (used in 1966 and 1967) versus a more sensitive method currently being used at LANL, as applicable to Palomares recovery veterans. The plot is in Figure F-1, with regression of estimated daily excretion levels against predicted inhalation intakes. Two sets of intake are provided: various indexed values of the high 26, based on the Labat-Anderson treatment with the CINDY code and annual acceptable inhalation intakes from ICRP Reports 2, 26/30/48, and 60/68. The predicted daily excretion rates from ICRP 30/48 (listed as NUREG-4884) and ICRP 68 are provided for PuO₂ inhalation intakes, Class Y and Type S, respectively. Clear from the plot, the LANL method of urine bioassay analysis has sufficient sensitivity for quantification of a 10 nCi intake, assuming a ICRP Report 68 excretion and a 1 μ m AMAD. This would encompass individuals with intakes at the levels assumed for the high 26, but would not be sensitive for the majority of Palomares recovery workers that were expected to have intakes much lower.

9.0 Additional Topics of Interest.

9.1 The Lymphatic System and Lymphoma [Summary Excerpted from Rademacher (2016)²⁴]

The human lymphatic system functions as an accessory route for transport of fluids in the body back to the blood stream and an important part of the immune system. The lymphatic system is comprised of the lymphatic organs, vessels, and the circulating lymph tissue. Under ICRP 23, the

²⁴ Minor editing.

lymphatic system has a mass of 2,200 g, which excluding the spleen, thymus, and tonsils has a mass of 1,996 g. The thymus and bone marrow are responsible for the production of lymphocyte tissue. In contrast to most solid tumors, for lymphomas, the location of irradiation is not always linked to the site of metastasis. This is particularly the case for circulating cells in the lymphatic system. With respect to inhalation exposures to insoluble chemical forms of plutonium, dose to thoracic lymph nodes [LN(TH)] is high compared to even the lung, liver, and bone surface, though these lymph nodes comprise only about 1.5% of the mass of entire lymphatic system. This is due to the long-term retention of plutonium deposited in the LN(TH). Among other organs in the lymphatic system, dose to the RBM is relatively high from internal plutonium exposures, while the spleen, thymus, and lymph nodes (outside the respiratory tract) do not have a proclivity for retention of plutonium or americium, and subsequently substantially lower dose than that received by the RBM.

For dose assessments under ICRP Report 68, only a 0.001 tissue-weighting factor for the dose contribution to the lymph nodes is used in calculation of dose to the ET and thoracic airways, because they are assumed to have a very low radiation detriment (Eckerman 2012). Mettler and Upton (1995) categorize malignancies to the lymphatic system in the group of tissues with very low or absent susceptibility to radiation induction. This was noted previously in this report from IARC (2000) and in Stannard (1988).

In ICRP Report 103 (ICRP 2007), the lymphatic nodes are handled as a separate remainder tissue that is one of 13 for each gender. Nevertheless, with averaging of the 13 remainder tissues, each remainder tissue receives an effective weighing factor of ~ 0.01 . Further, with the LN(TH) comprising only about 1.5% of the total lymph tissue mass outside of the spleen, thymus, and tonsils, dose to the LN(TH) is deemed relatively insignificant compared to other detriment from internal plutonium dose. ICRP recognized that for many circumstances dose was distributed in a heterogeneous manner, but deems detriment from stochastic effects (i.e., cancer, genetic effects) in all parts of that organ or tissue can be correlated with dose averaged over the entire tissue with sufficient accuracy (ICRP 2007). For unique circumstances like radon daughter product deposition on the bronchial mucosa or plutonium deposited on bone surfaces, the ICRP addressed these issues in specific organ (i.e., skeleton) or system (i.e., respiratory tract) models (ICRP 2007).

Historically, under the EEOICPA, cases where a worker has a lymphoma, medical reviews were performed, without specification of a specific organ dose (ORAU 2003). More recent guidance for EEOICPA cases (ORAU 2012) contains internal dose target organs for consideration, based on the site of metastasis in the lymphatic system and the metastasis type. The VA approach in evaluating cases of lymphoma may have some similarities and differences in the manner used under EEOICPA cases.

The recent addition of CLL cases under the EEOICPA involved a detailed review of the lymphatic system to radiation dosimetry for CLL by Specialists in Energy, Nuclear and Environmental Sciences (SENES), Oak Ridge, TN (Apostoaie and Trabalka 2012). The Apostoaie and Trabalka (2012) work concentrated on potential dose to precursor B-cells, as this group of cells were deemed to represent those cells within the lymphatic system that were potential precursors to CLL. Because these cells are located throughout the lymphatic system, assessment of dose and ultimately PoC is complicated by the varied distribution among individuals, affected by age, gender status, and other factors (Apostoaie and Trabalka 2012).

A brief review of some information from this work is useful in illustrating the problem. Dose coefficient (DC) values for organs key to internal plutonium exposure and the lymphatic system from inhalation of Type S $^{239+240}\text{Pu}$ are listed in Figure 11. Obvious is the ten-fold higher DC for the LN(TH) as compared to the lungs, yet the very low DC's for other organs/tissues in the lymphatic system – thymus, spleen, remainder lymph nodes, other. Figure 12 contains a point estimate of percentage of precursor B-CLL cells in various organs/tissues, though the Apostoaei and Trabalka (2012) work treated the distribution in a probabilistic manner. Among the organs/tissues with the largest fraction of precursor B-CLL cells, only the RBM was among those organs with a reasonably high DC for inhalation of Type S $^{239+240}\text{Pu}$. The results of the Apostoaei and Trabalka (2012) probabilistic evaluation of inventory-weighted 50-y CED DC's for precursor B-CLL cells and B-cells are shown in Figure 13 against DC's for key organs. The plot demonstrates the high variability in potential DC factors for calculation of PoC's for CLL. Use of the DC for LN(TH) for calculation of PoC in CLL cases is highly conservative, but unrealistic and lacking scientific defense (Apostoaei and Trabalka

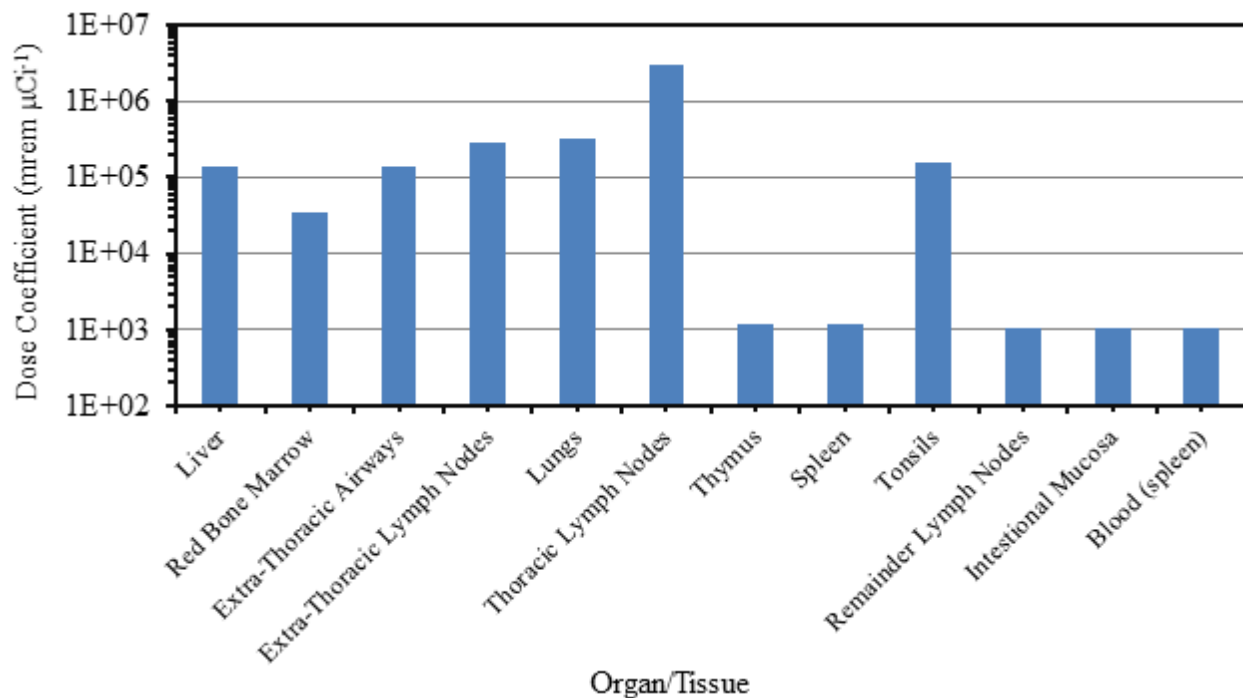


Figure 11. Dose Coefficients for Organs Key to $^{239+240}\text{Pu}$ Internal Exposure and the Lymphatic System, data from Apostoaei and Trabalka (2012) for Inhalation Intakes, ICRP Report 66, Type S.

2012). Application of the LN(TH) DC in PoC calculation for some lymphomas may have a similar shortcoming. The discussion here helps illustrate the complications in assessment of PoC for lymphatic metastases.

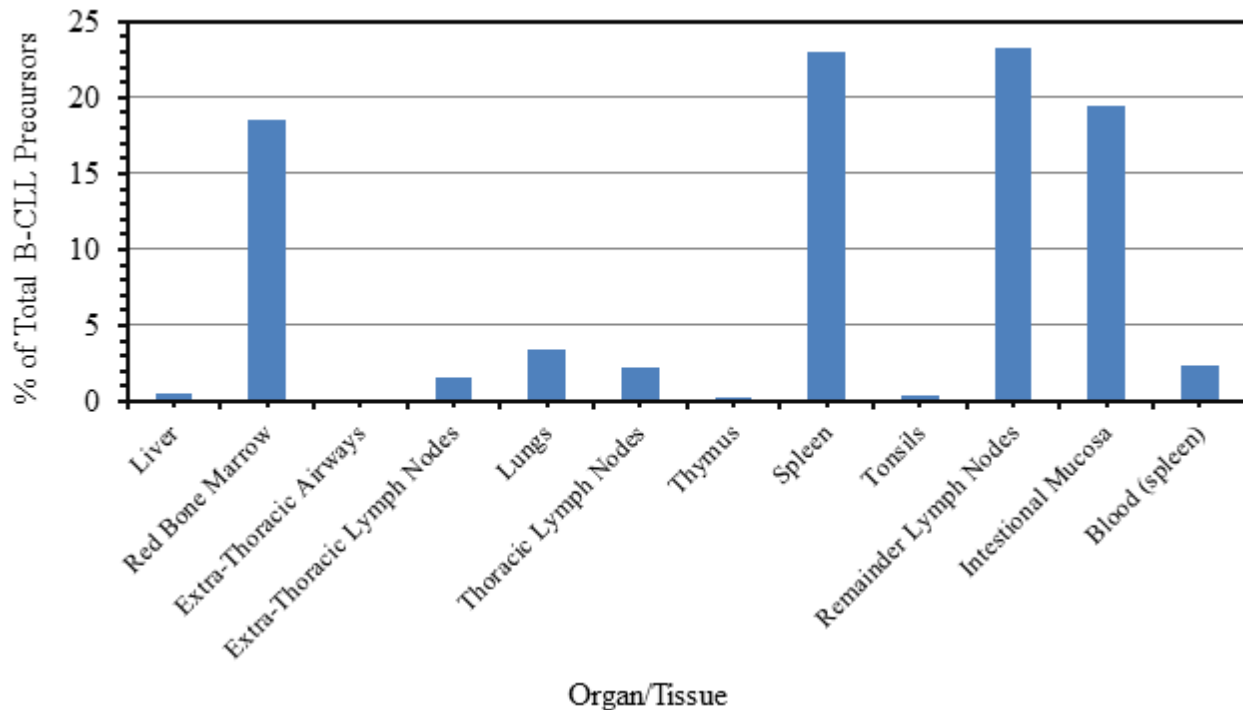


Figure 12. Point Estimate, Percent of Total B-CLL Precursors to Organs Key to Internal Plutonium Exposure and the Lymphatic System, data from Apostoaei and Trabalka (2012).

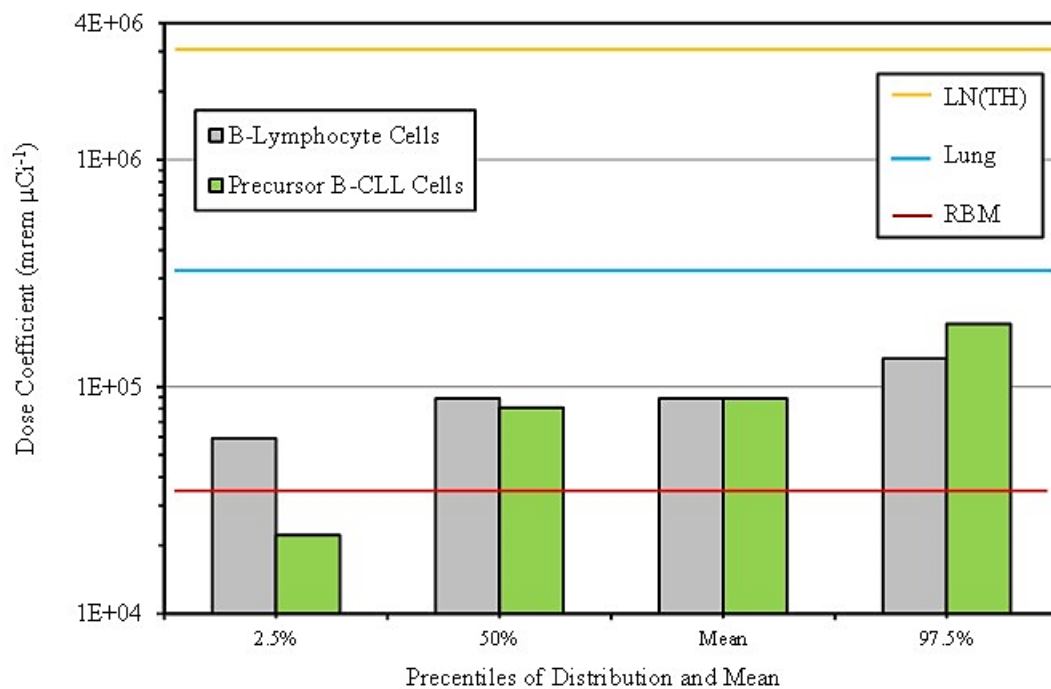


Figure 13. Percentiles of Probabilistic Inventory-Weighted Average 50-y CDE Dose Coefficient Distributions for B-Cells and Precursor B-CLL Cells, and Key Organ Dose Coefficients for Inhaled $^{239+240}\text{Pu}$, Type S, data from Apostoaei and Trabalka (2012).

9.2 Prostate Cancer.

Prostate cancer is the most common cancer among men after skin cancer, with over 50% of new cases diagnosed after 65-y of age. While prostate cancer is listed by the VA as radiogenic in 38 CFR 3.311, the prostate is relatively insensitive to ionizing radiation, as compared to other tissues (Mettler and Upton 1995). Because the ICRP Report 60/68 set of recommendations did not list the prostate gland, under EEOICPA, it is recommended to apply dose received by the organ with highest dose among those that do not have specific-metabolic models for the radionuclide of interest. For internal dose from plutonium, the appropriate “surrogate” organ was the testes. ICRP Report 103 provides DC values for the prostate, with them being the same as the thymus, skin, brain, and others. Therefore, in lieu of the previous practice of using the testes, it is recommended to use the DC for thymus (or others), as listed in ICRP Report 68, as a surrogate for AF dose estimates.

9.3 Urine Excretion and Other Internal Dosimetry Plots.

Appendix F contains a number of internal dosimetry distribution plots, with key plots for urine excretion. For brevity, no discussion is provided here. Appropriate discussion is contained in the Appendix.

10.0 Summary.

This report provided a discussion of pertinent issues related to the Air Force’s methodology developed to estimate doses for Palomares recovery veterans. The approach maintains limited use of the extensive urine bioassay data collected from recovery workers during on-site work, but a greater degree of confidence in a urine resampling program conducted on a large fraction of workers after completion of the recovery. The Air Force recognized that many initial samples had cross-contamination issues and high bias from α -particle emitter found naturally in urine. Follow-on resampling supported this conclusion. A subset of 26 workers with the highest predicted inhalation intakes were asked to participate in additional sampling, beyond the off-site sampling of 400+ workers. Estimated doses from this group of 26 workers provided a high-sided basis for estimation of intakes for other workers that did not participate in an additional phase of urine resampling.

The estimated inhalation intakes of plutonium, derived from analysis of urine analysis date, are supported by air sampling conducted during recovery, the concentrations of plutonium in surface soils produced by the detonation/dispersal events (paired with resuspension factors), and inhalation intakes by Palomares residents receiving medical follow-on assessments. The estimated intakes are directly correlated to estimated doses.

The methodology for assessments of metabolism and doses have been based upon ICRP recommendations. These recommendations have evolved four times since publication of Report 2 in 1959. Methodologies in Report 2 were used for initial radiation safety evaluations for recovery workers. The recommendation in Report 2 for plutonium were based largely on animal research studies and prudent conservatism. Since, ICRP recommendations for plutonium metabolism and dose estimates have been refined by substantial data from study of workers exposed to plutonium,

with the most substantial source being from MPA workers. Differences among the ICRP recommendations, as applied to plutonium are modest. This report contained a detailed summary of changes over the past 60 years.

High levels of inhaled plutonium have been linked to increased risk of lung, liver, and bone cancer based on animal research studies and epidemiological studies of former Soviet Union plutonium workers. The study of US and British plutonium workers did not find statistically significant differences in cancer induction risks (ATSDR 2010). These plutonium worker health studies were limited in statistical power because the workers did have significant body burdens and the total number of exposed workers were limited.

The Air Force adopted reporting doses to the high 26 individuals based on the Labat-Anderson report dose summaries if a veteran had a medical condition in an organ covered by the ICRP 26/30 methodology. The lung, liver, and bone are covered within this methodology. For other organs, use of the ICRP 60/68 was recommended, because it contained a more extensive coverage of organs. For individuals not in the high 26, the AF applied the lowest intake among the estimated intakes for the high 26, with the exception for those workers without on-site presence. The same practice of applying ICRP 26/30 estimates of dose for covered organs, and ICRP 60/68 for cases with a disease applicable to an organ not covered by ICRP 26/30. The assignment of dose to individuals not in the high 26 is consistent with requirements stated in 38 CFR 3.311 for a determination of “probable dose” with accounting for uncertainties, and represents an estimated 95th percentile or greater plutonium intake level.

AF/SG and our office thought the estimated doses to the lung, liver, and bone surfaces, as deemed appropriate for intakes to the vast majority of recovery workers, are likely to receive favorable compensation decisions by the VA for primary cancers of the lung, liver, and bone. Nevertheless, the Air Force does not tailor dose assessments based upon this consideration. The VA has sole authority for these decisions.

Overall, application of conservative, high-sided dose estimates paired with IREP calculated dose values at the 50% PoC, 95% CL provides significant benefit of doubt in favor of veterans. Furthermore, the evaluation of MPA workers demonstrated that the assumption used in IREP of a linear dose-response relationship for solid tumors is overly conservative at the lower levels of dose to the lung, liver, and bone. The uncertainties in dose estimates are dwarfed by the uncertainty provisions applied by IREP for uncertainties in the dose-response relationship.

The Air Force has not recommended an extensive current day urine resampling program. The three primary reasons are:

- conservative dose assumptions would likely be favorable for compensation decisions for Palomares responders with primary cancers of the lung, liver, and bone if they had estimated intake commensurate with the lowest estimated intake of the high 26 or higher; additional bioassay would likely debate high-sided dose estimates,

- current urine bioassay is unlikely to provide any additional benefit to a favorable compensation decision for these three cancers, and
- for individuals with cancer not related to deposition and retention in lung, liver, and bone, there was not any reasonably-expected benefit from a current bioassay.

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Appendix A

General Site Information, Source Term Information, and Urine Analysis Data



Figure A-1. Map of Spain with US Military Bases in 1966 and Village of Palomares.



Figure A-2. Almería Province Map.

TABLE A-1. Isotopic Composition of WGP in BOMARC Weapon Based on Los Alamos National Laboratory Estimates and Soil Analyses for 1958.
[Table A-1 from Rademacher *et al.* 2009]

Isotope	Mass Percent	α -Activity Percent	Radiological Half-life (y)
Pu-238	0.0099	2.3	87.74
Pu-239	93.7	80.1	24,110
Pu-240	5.6	17.6	6,560
Pu-241	0.47	Not Applicable	14.35
Pu-242	Negligible	Negligible	376,000

TABLE A-2. Major Radiation Emissions of WGP Constituents.
[Table A-2 from Rademacher *et al.* 2009]

Radionuclide	α -Particle Energies (MeV) & Frequency	β -Particle Energies (MeV) & Frequency	Photon Energies (MeV) & Frequency
Pu-239	5.155 (0.733) 5.143 (0.151) 5.105 (0.115)	None	0.113 (0.0005) 0.014 (0.044)
Pu-240	5.168 (0.735) 5.123 (0.264)	None	0.054 (0.0005) 0.014 (0.11)
Pu-241	None	0.021 (1.00)	None
Am-241	5.486 (0.852) 5.443 (0.128) 5.388 (0.014)	None	0.014 (0.427) 0.0595 (0.359) 0.026 (0.024)

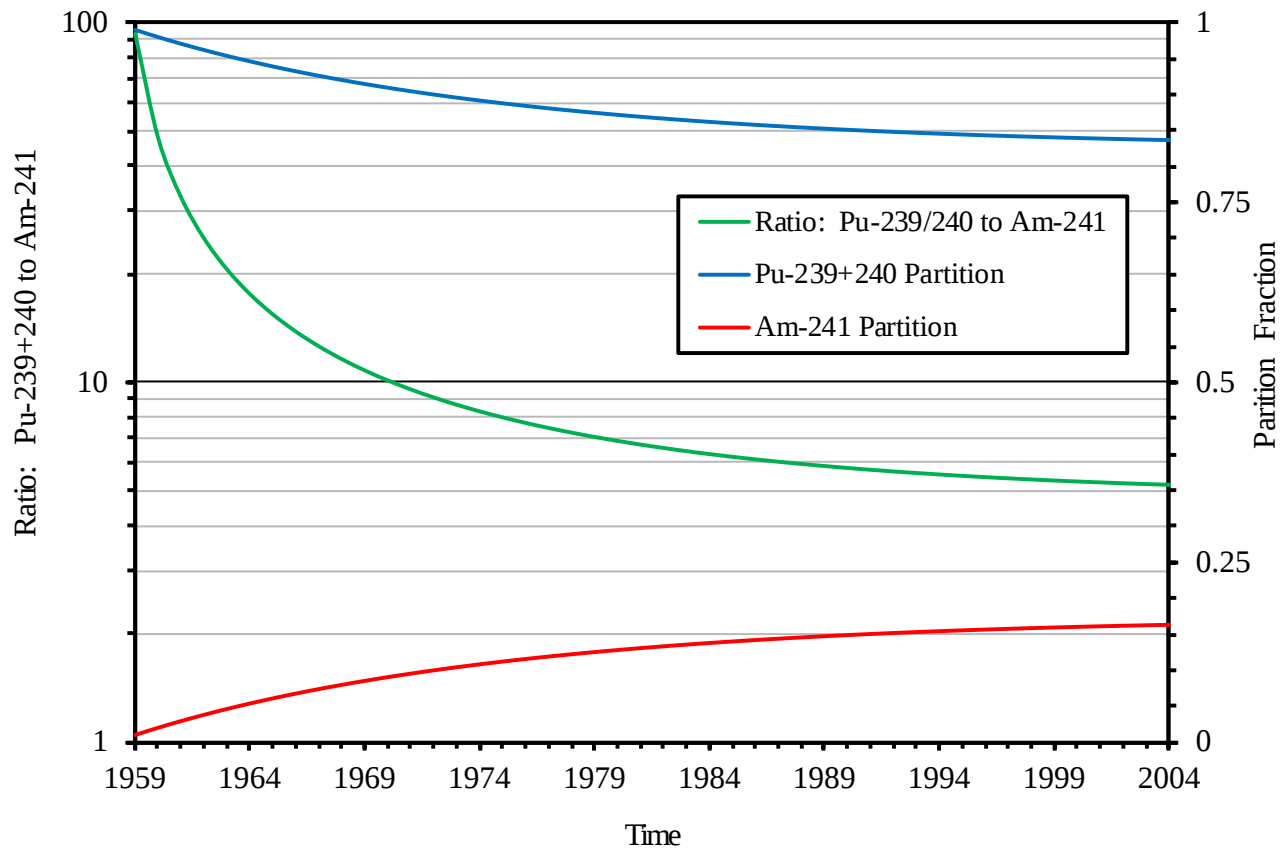


Figure A-3. $^{239+240}\text{Pu}$ and ^{241}Am Partition Fractions and $^{239+240}\text{Pu}$ and ^{241}Am Ratio Estimates for WGP in Soil at BOMARC Site. [Data from Table A-1 (1958)]

TABLE A-3a. Personnel and Functions, 31 January 1966 (DNA 1975).

Number	Function	Number	Function
200	Ground search	2	Helicopter operations
200	Detection, decontam., harvesting	41	Air police
23	Accident investigation board	7	Information and public relations
23	Civil eengineering	19	Navy ordnance disposal
30	Campl support	4	Technical representatives
6	Legal claims	7	Army engineers
5	Medical	36	Transportation
58	communications	20	Command and staff

TABLE A-3b. Personnel at Camp Wilson and San Javier (Air Force 1968).

Week	Camp Wilson Americans	Spanish (less Guardia Civil)	San Javier Americans	Total
17 - 23 Jan	49	0	1	50
24 - 30 Jan	583	0	50	633
31 Jan – 6 Feb	665	37	73	775
7 – 13 Feb	666	25	53	744
14 – 20 Feb	632	36	51	719
21 – 27 Feb	661	36	47	744
28 Feb – 6 Mar	618	33	50	701
7 – 13 Mar	522	33	42	597
14 – 20 Mar	471	32	31	535
21 – 27 Mar	330	0	31	361
28 Mar – 3 Apr	144	0	28	172
4 – 7 Apr	34	0	12	46

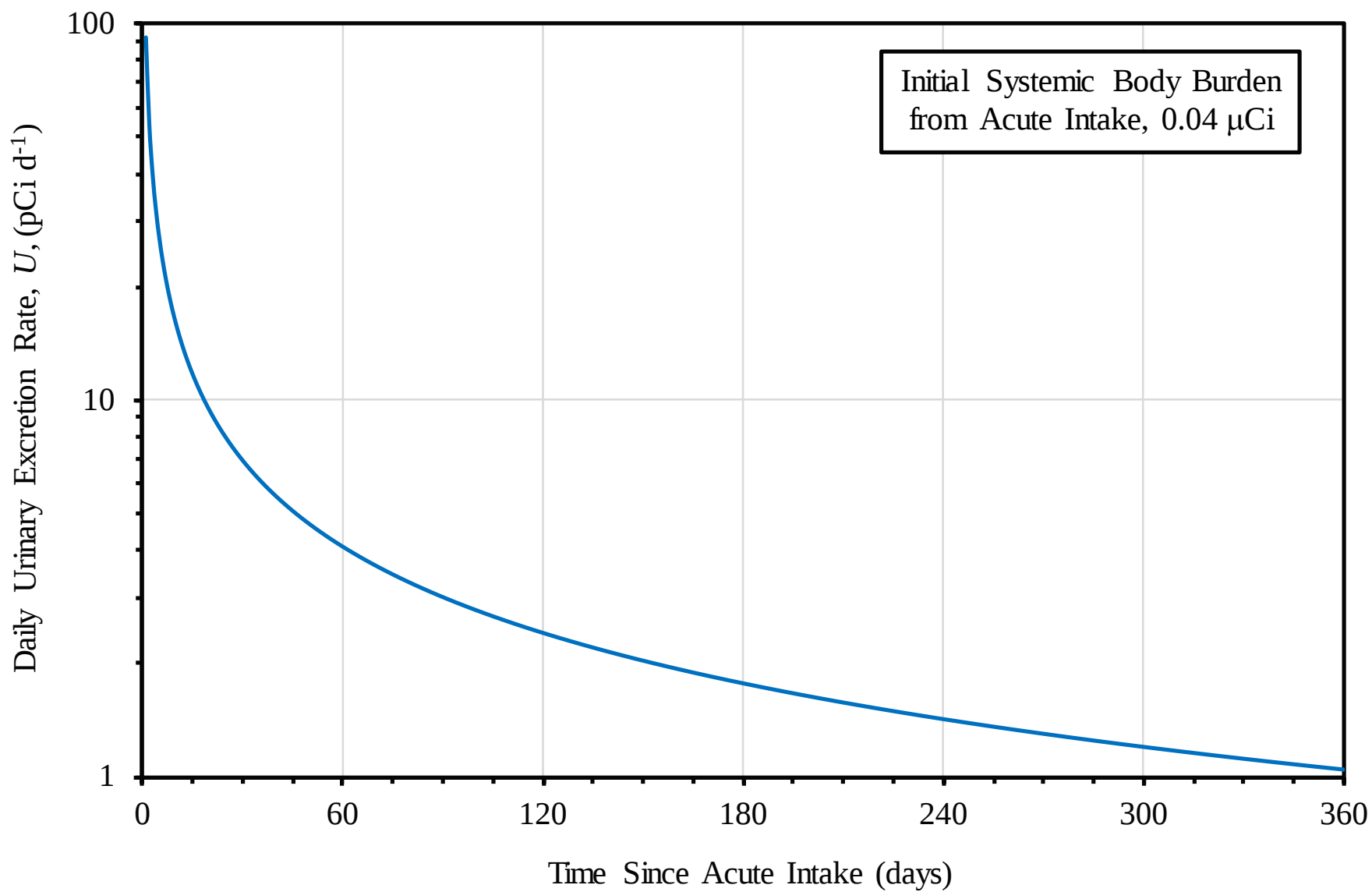


Figure A-4. Daily Urinary Excretion Rate, U , After Acute Intake, for Initial Systemic Body Burden of 0.04 μCi ^{239}Pu .

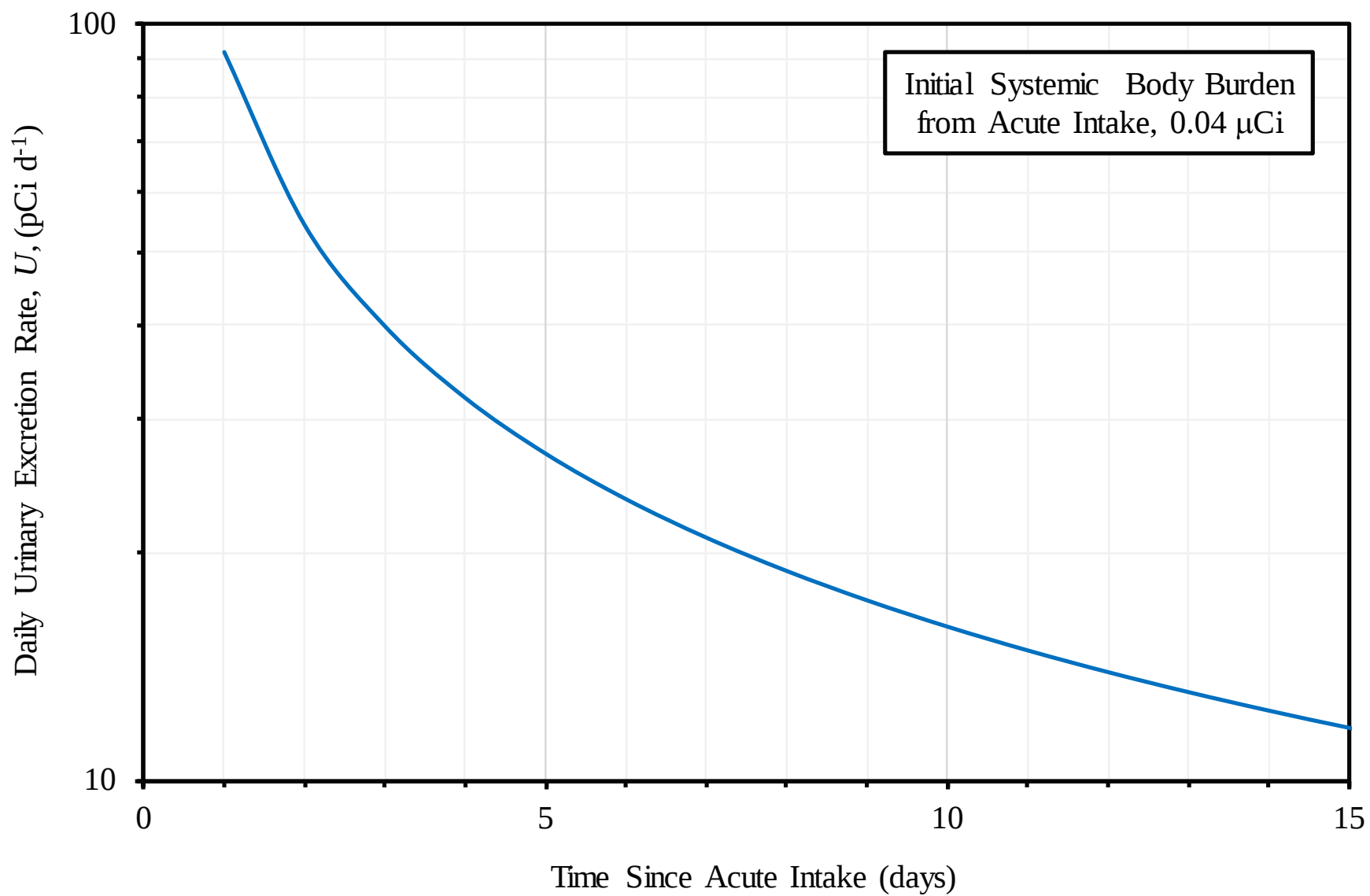


Figure A-5. Daily Urinary Excretion Rate, U , After Acute Intake, for Initial Systemic Body Burden of 0.04 μCi ^{239}Pu , Identical to A-2, for 15-day Period (Langham Model).

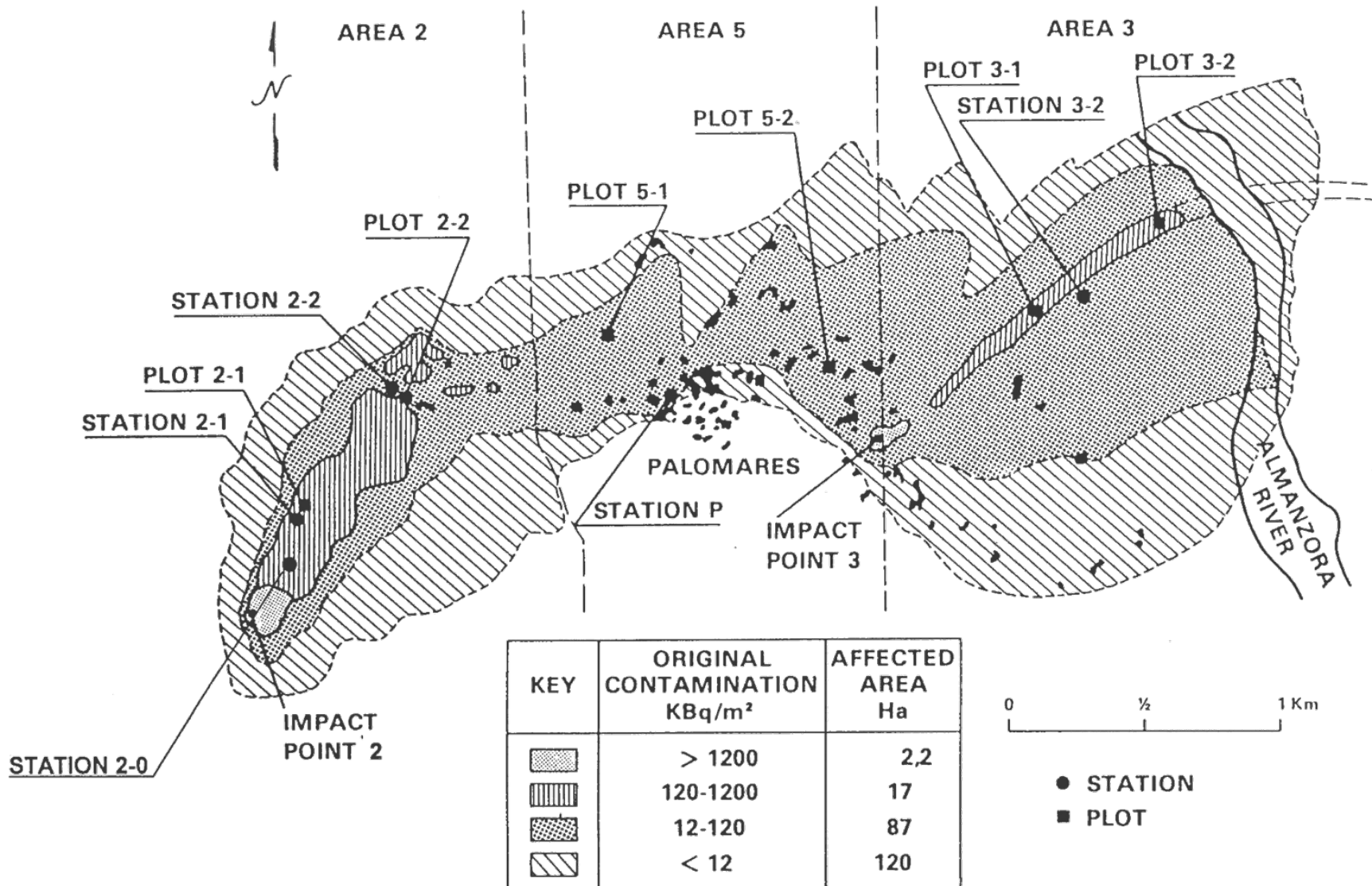


Figure A-6. Environmental Sampling Stations Established by the Spanish to Monitor Airborne Contamination Overlaying Initial $^{239+240}\text{Pu}$ Soil Surface Contamination Contours (27 nCi KBq^{-1} ; $1 \text{ Ha} = 2.47 \text{ acre}$). [Figure 8 from Iranzo *et al.* 1998]

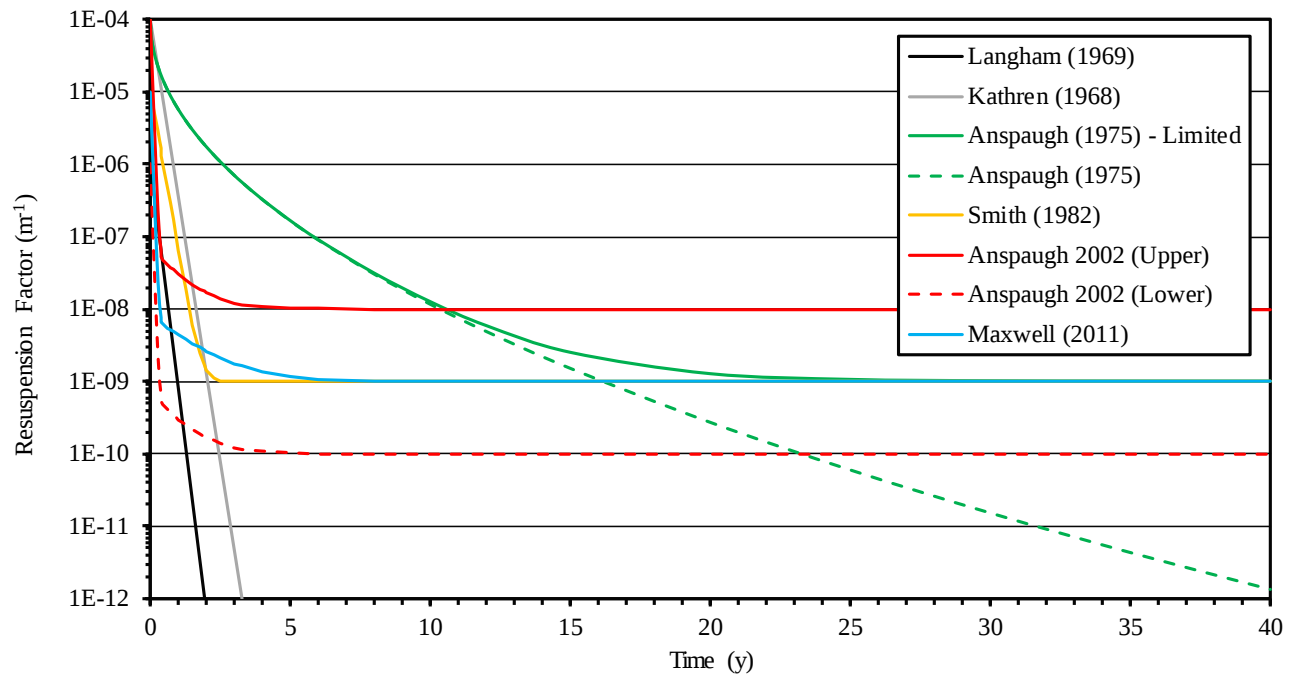


Figure A-7. Resuspension Factors of Multiple Authors, up to 40 Years Post Initial Deposition.

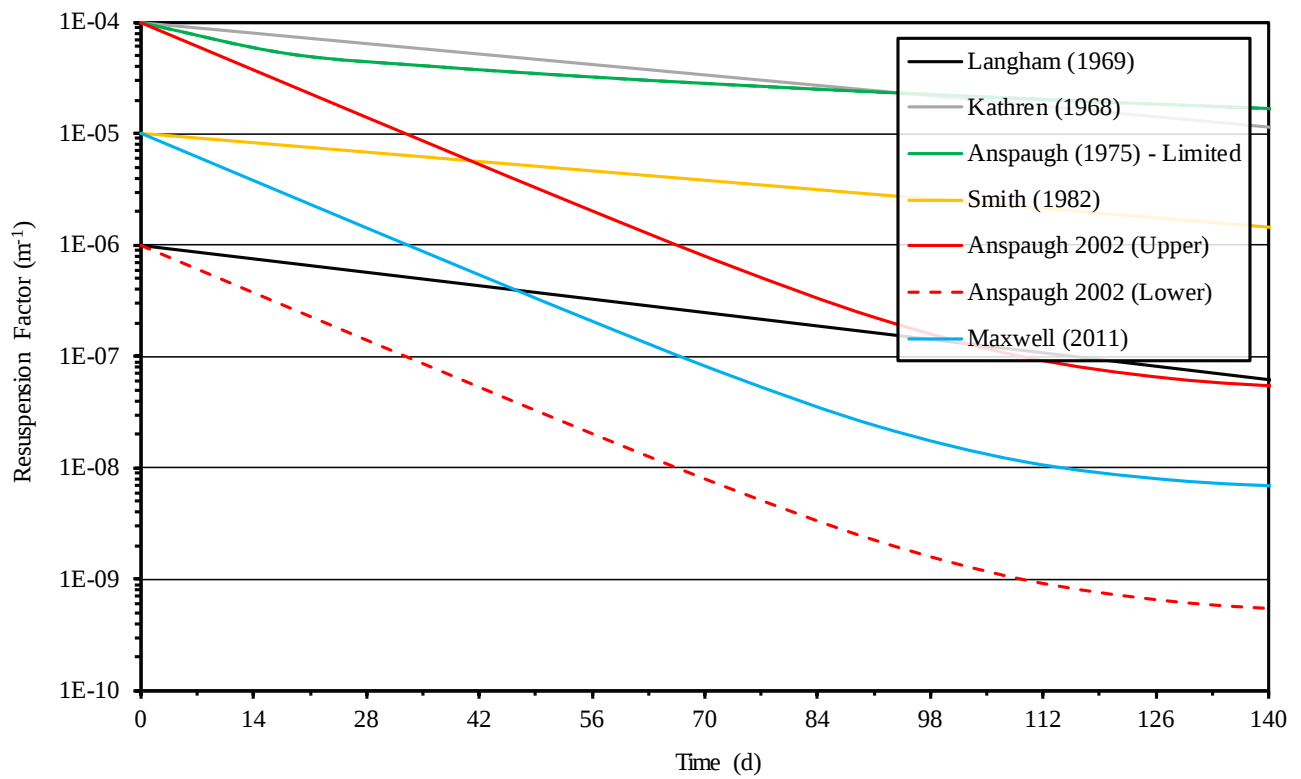


Figure A-8. Resuspension Factors of Multiple Authors, up to 140 Days Post Initial Deposition.

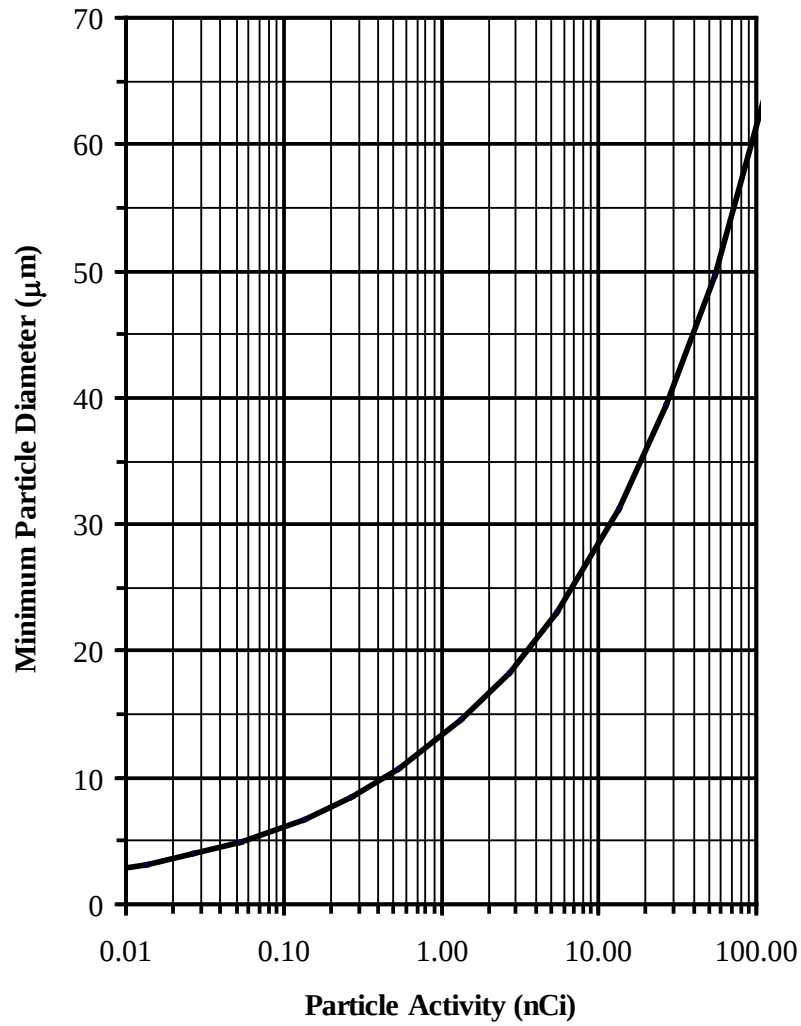


Figure A-9. Minimum $^{239+240}\text{PuO}_2$ Particle Diameter, Assuming Spherical, and $\rho = 11.5 \text{ g/cm}^3$ (Low Range). [Figure A-4 from Rademacher *et al.* 2009]

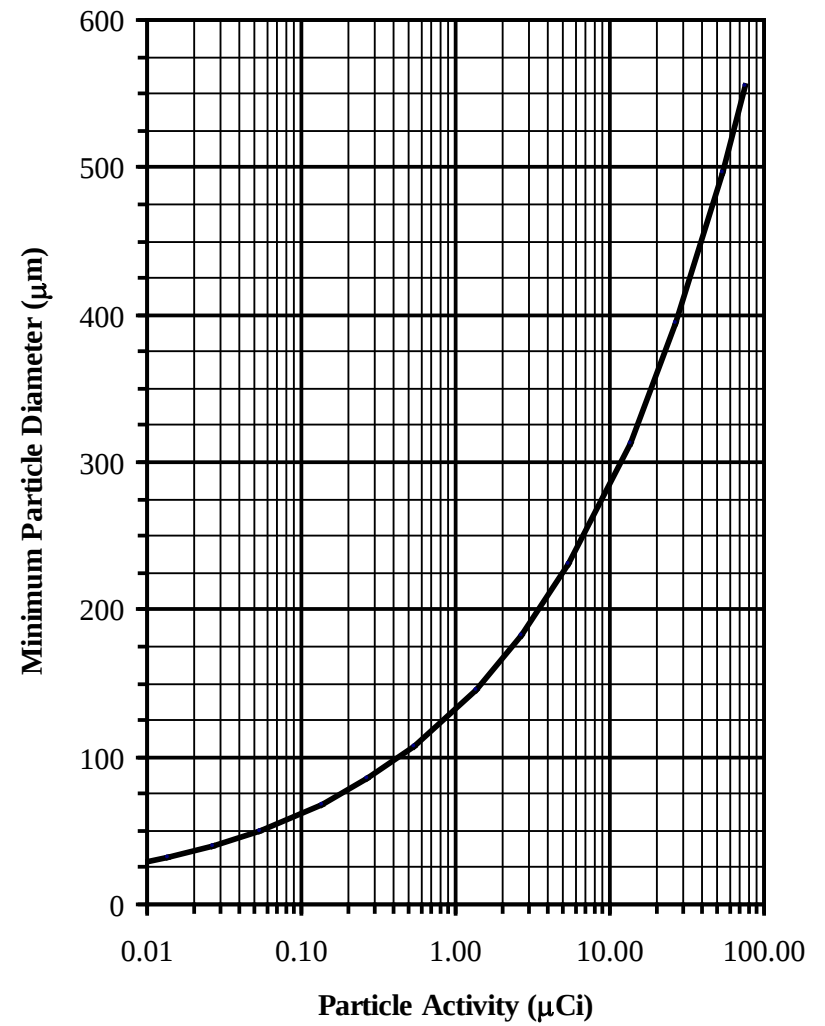


Figure A-10. Minimum $^{239+240}\text{PuO}_2$ Particle Diameter, Assuming Spherical, and $\rho = 11.5 \text{ g/cm}^3$ (High Range). [Figure A-5 from Rademacher *et al.* 2009]

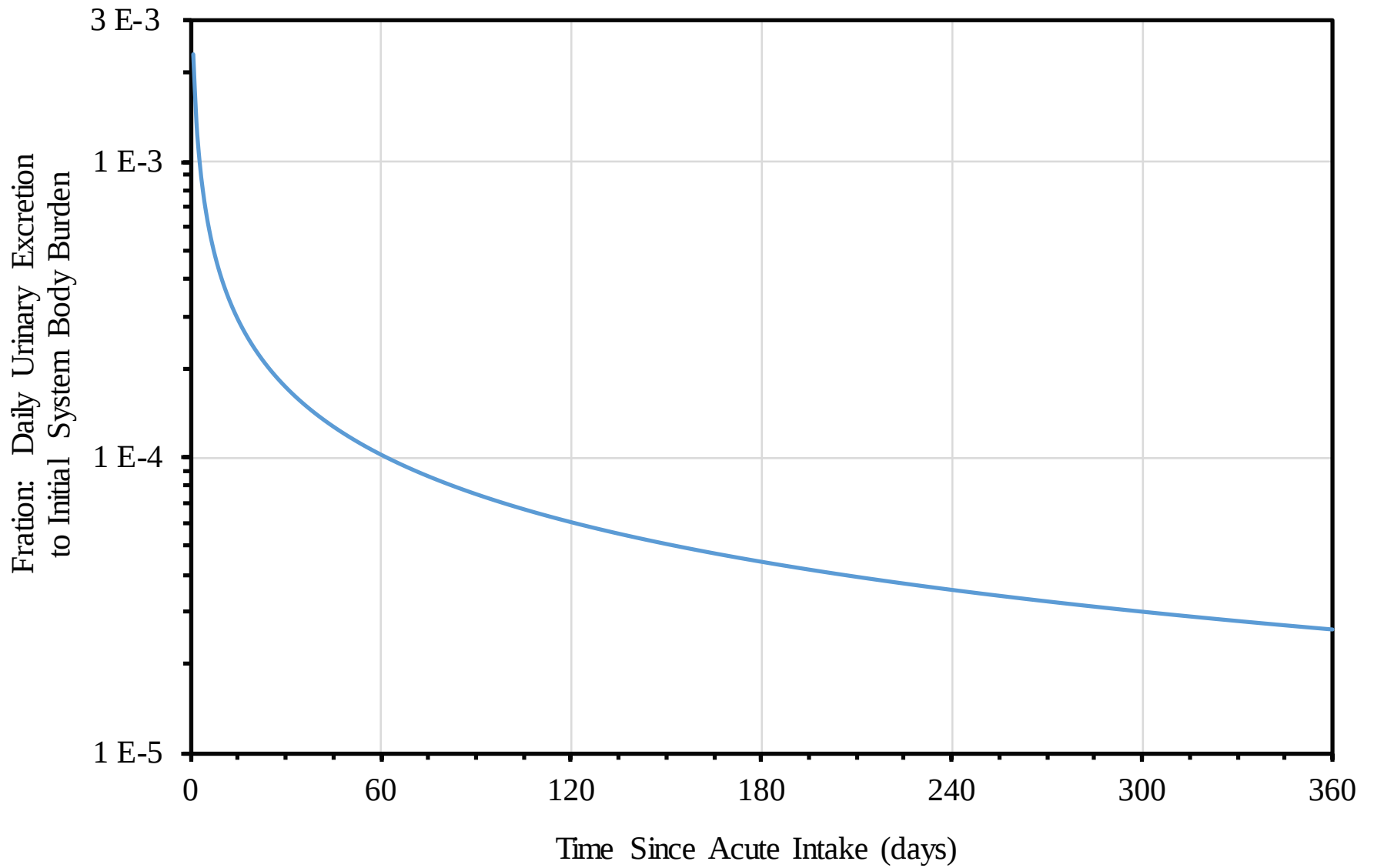


Figure A-11. Fraction of Daily Urinary to Initial Systemic Body Burden (Langham Model).

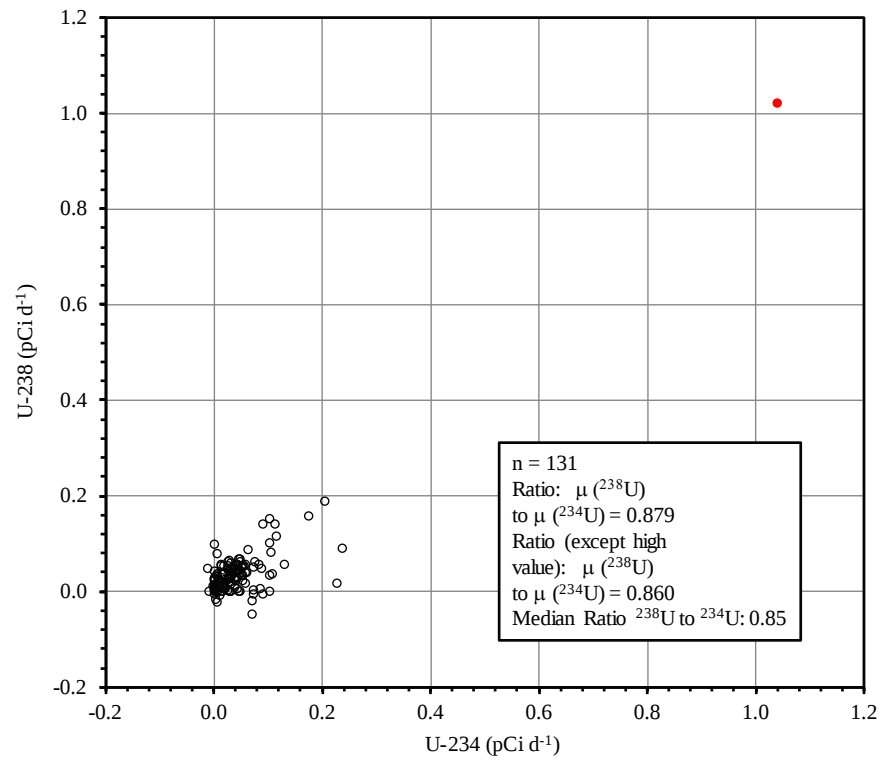


Figure A-12. Regression of ^{238}U to ^{234}U in Urine Samples from Workers Handling Depleted Uranium at Eglin AFB [Generated in Support of Work Documented in Rademacher *et al.* (2017)]

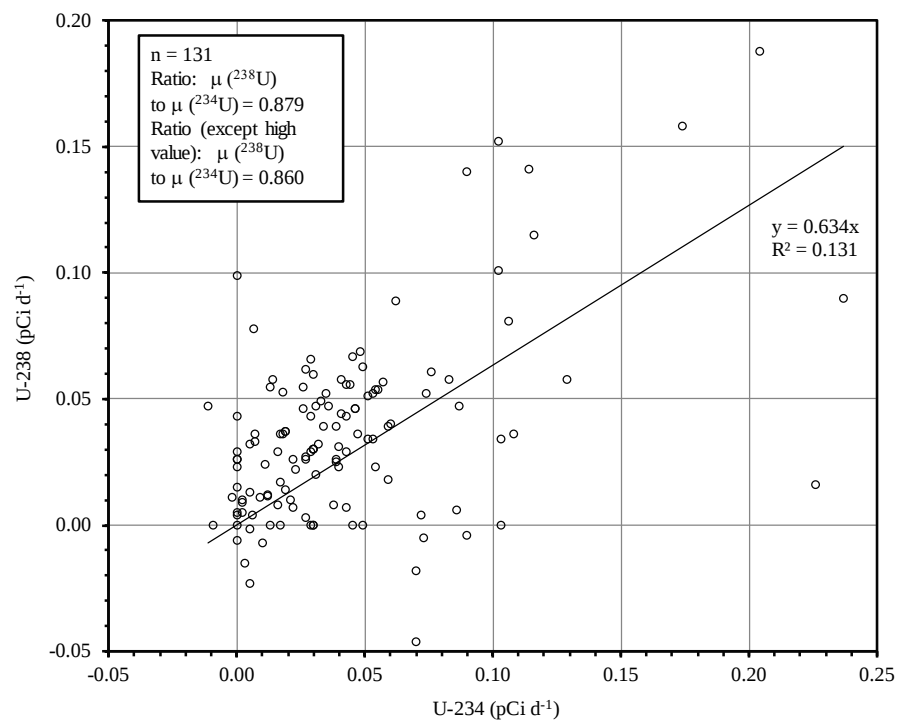


Figure A-13. Data from Figure A-3, Omitting Sample with Highest Total Uranium.

TABLE A-4. High 26 Bioassay Results for Palomares Responders Reanalysis Follow-up, Summarized from Wallace (1968).

Patient	Sample 1		Sample 2		Sample 3		Sample 4		Sample 5		Sample 6		Sample 7		
	Date	% BB	Date	% BB	Date	% BB	Date	% BB	Date	% BB	Date	% BB	Date	% BB	
1	30Mar66	LE	7Oct66	16	20Jan67	0	17Apr67	0	14Aug67	0					
2	13Feb66	48	13Jun66	9	19Jan67	3	24Apr67	0	20Aug67	0					
3	10Mar66	LE	22Apr66	41	28Jun66	0	20Jul66	8	28Jan67	0	21Apr67	0	10Oct67	0	
4	14Apr66	9	8Jun66	7	20Sep66	7	21Feb67	3	30Apr67	0					
5	3Mar66	LE	20May66	18	8Sep66	8	15Jan67	0	10Apr67	0	9Aug67	0			
6	4Feb66	10	22Jun66	16											
7	17Feb66	34	15Aug66	9	28Feb67	2									
8	13May66	14	26Sep66	7	19Jan67	3	17Apr67	0	14Sep67	0					
9	7May66	18	28Jul66	7	22Feb67	2	2May67	0	7Sep67	0					
10	25Feb66	29	8Jun66	7	21Jan67	2	8Apr67	0	4Aug67	0					
11	6Apr66	10	4Sep66	7	13Jan67	2	8Apr67	2	4Aug67	0					
12	28Feb66	25	28Jul66	11	1Sep66	8	20Jan67	0	17Apr66	0	16Aug67	0			
13	7Mar66	25	26May66	7	20Jan67	3	17Apr67	0	10Aug67	0					
14	30Aug66	7	13Jan67	4	14May67	0	23Aug67	0							
15	10May66	16	1Jun66	9	7Feb67	2									
16	13Feb66	400	8Apr66	20	20Oct66	10	13Jul67	0							
17	17Feb66	17	6Jun66	7	23Jan67	2	9May67	0	17Aug67	0					
18	7Mar66	15	6Oct66	11	9Mar67	0	13Apr67	5	9Aug67	0					
19	27Jan66	12	18Jun66	8	12Feb67	2	9Apr67	0	13Aug67	0					
20	13Jul66	9	24Jan67	6	26Apr67	1	10Aug67	0							
21	10Feb66	100	3Mar66	10	26Apr66	17	10Oct66	67	26Jan67	3	9May67	0	20Sep67	0	
22	17Feb66	23	8Jun66	7	19Jan67	3	17Apr67	0	10Sep67	0					
23	3Mar66	15	1Sep66	7	2Feb67	0	5Jun67	0	15Sep67	0					
24	25Jan66	21	6Apr66	15	26Oct66	11	19Jan67	1	5May67	0	15Aug67	0			
25	11Feb66	25	24Jul66	7	17Jan67	3	25May67	0	4Sep67	0					
26	20Jul66	8	20Jan67	3	19Apr67	0									
LE	Lab Error				Gross α -Particle Analysis										

TABLE A-5. High 26 Urine Bioassay Intake Estimates from Labat-Anderson (2001).

Patient	Estimated On-site Dates		Surname*	Modeling Code			
	Start	Stop		CINDY [ICRP 26/30/48]		LUDEP [ICRP 60/66]	
				Inhalation Intake (nCi)	CEDE (rem)	Inhalation Intake (nCi)	CED (rem)
1	29 Jan	29 Feb		68	21	22	1.6
2	18 Jan	4 Feb		86	26	210	15
3	24 Jan	14 Feb		63	19	19	1.3
4	18 Jan	20 Mar		62	19	77	5.4
5	29 Jan	19 Feb		65	20	790	55
6	18 Jan	6 Feb		560 – 1,200	170 - 370	2,600	180
7	18 Jan	6 Feb		160	49	130	8.8
8	18 Jan	1 Apr		110	34	92	6.5
9	18 Jan	5 Feb		42	13	140	9.5
10	18 Jan	19 Feb		64	20	37	2.6
11	6 Feb	28 Feb		55	17	91	6.4
12	18 Jan	11 Feb		44	14	29	2.0
13	18 Jan	13 Feb		76	23	160	11
14	1 Feb	2 Apr		72	22	62	4.4
15	31 Jan	8 Apr		180	55	79	5.6
16	18 Jan	4 Mar		210	65	700	49
17	18 Jan	3 Feb		66	20	130	9.3
18	18 Jan	15 Feb		68	21	20	1.4
19	18 Jan	21 Jan		69	21	82	5.7
20	20 Jan	11 Feb		34	10	65	4.5
21	17 Jan	30 Jan		100 - 350	31 – 110	1,100	79
22	22 Jan	9 Feb		71	22	57	4.0
23	18 Jan	19 Feb		44	14	33	2.3
24	18 Jan	18 Mar		58	18	75	5.2
25	18 Jan	6 Feb		64	20	160	11
26	18 Jan	6 Feb		99	30	160	12
	Intake within 2x		* Excluded from this version of report due to Privacy Act restrictions, 5 U.S.C. 552(1)				

Appendix B

Radiation Exposure Standards and ICRP Internal Dosimetry Information

TABLE B-1. Occupational Exposure Standards in ICRP Reports 1 and 2.

Applicable Organ/Tissue	Dose Limit (rem)	
	Calendar Quarter	Calendar Year
Whole-body, head and trunk, blood-forming organs, gonads, lens of the eye*	1.25 [3]	5 [5 (N-18), N is age]
Skin of whole-body, cornea of the eye, bone [†]	8	30
Hands and wrists, or feet and ankles	20	75
Forearms	10	30
Thyroid	8	30
Other organs, tissues, and organ systems	4	15

* Acceptable dose for all adults listed first, higher limits listed in parentheses acceptable for adults 21 and older, but must required consideration of previous lifetime exposure history

[†] Radium-equivalent provisions for internal emitters applicable to bone

TABLE B-2. Radiation Weighting Factors, w_R , or Relative Biological Effectiveness (RBE) for ICRP 2 and Quality Factor for ICRP 26.

Radiation Type	ICRP 2	ICRP 26	ICRP 60	ICRP 103
Photons	1	1	1	1
Electrons and muons	1 [‡]	1	1*	1*
Alpha particles, fission fragments, heavy ions	-	20	20	20
Alpha particles	10	-	-	-
Recoil atoms	20	-	-	-
Neutrons	-	10	-	2.5 - 20 [§]
Neutrons < 10 keV	-	-	5	-
Neutrons: 10 to 100 keV	-	-	10	-
Neutrons: > 100 keV to 2 MeV	-	-	20	-
Neutrons: > 2 to 20 MeV	-	-	10	-
Neutrons > 20 MeV	-	-	5	-
Protons, other than recoils, energy > 2 MeV	-	-	5	-
Protons and charged pions	-	10 [★]	-	2

[‡] 1.7 for electrons with energy < 30 keV * Special considerations for auger electrons

[§] Continuous function, peak of 20 at 1 MeV ★ Other singly-charged particles of rest mass greater than one amu

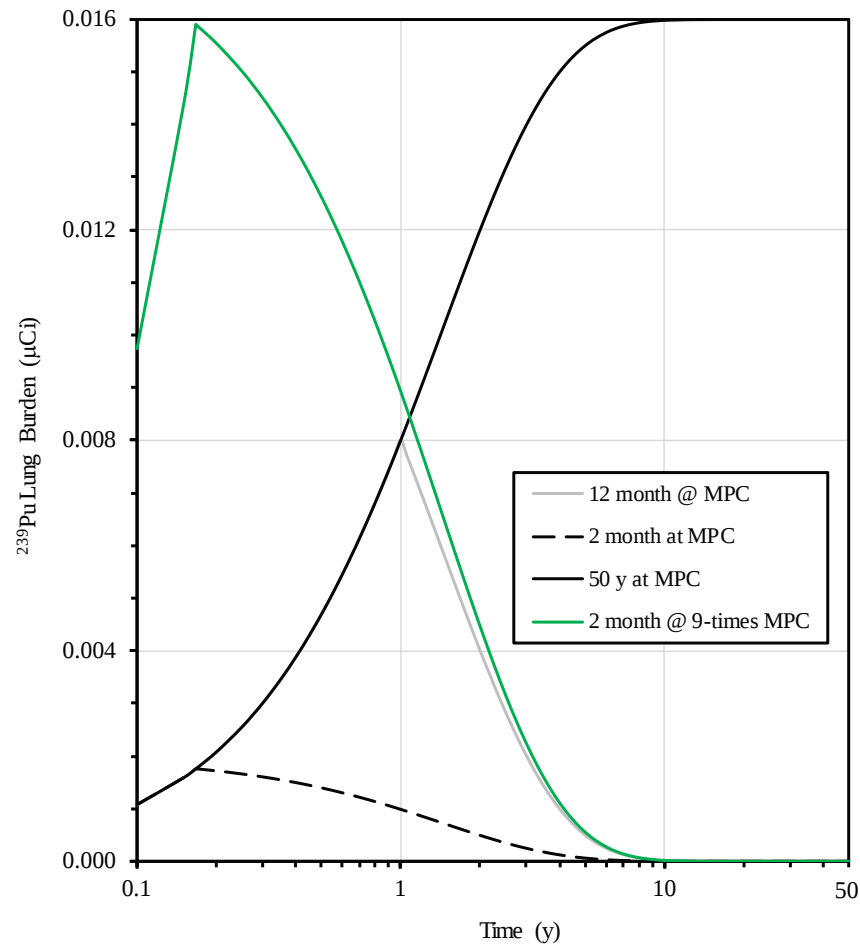


Figure B-1. Lung Burdens for Various Inhalation Exposures to Insoluble ^{239}Pu using ICRP 2 Metabolism.

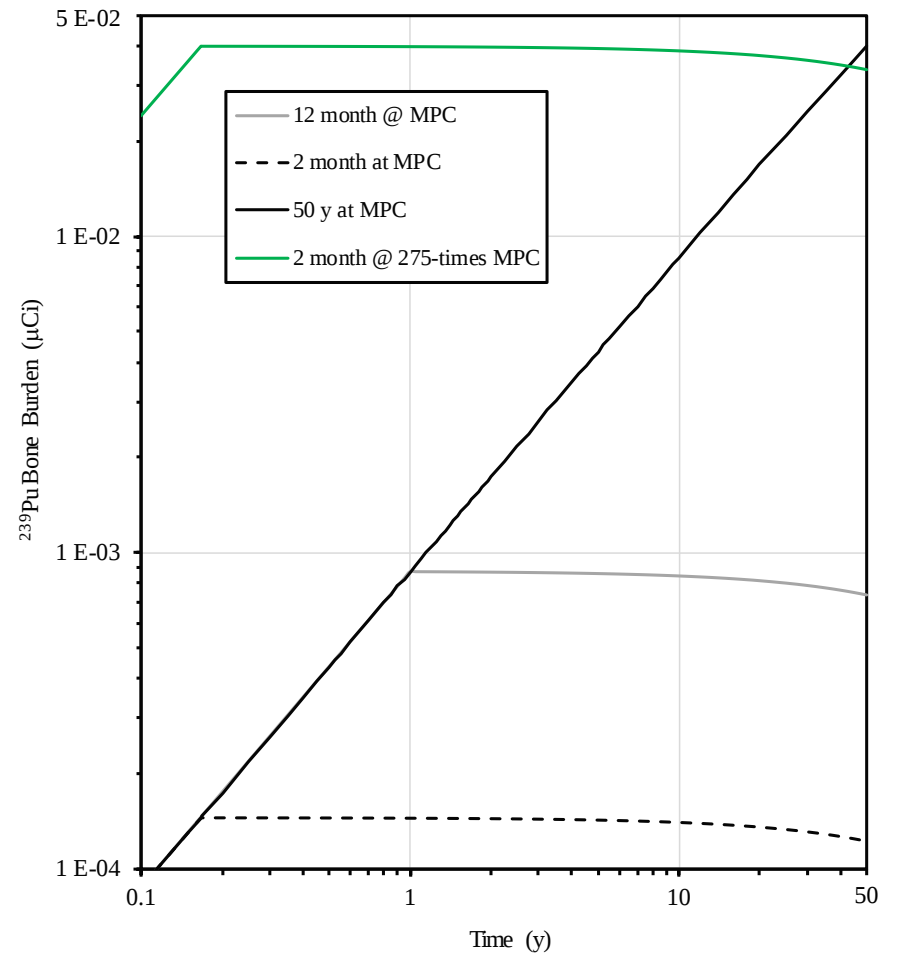


Figure B-2. Bone Burdens for Various Inhalation Exposures to Soluble ^{239}Pu using ICRP 2 Metabolism.

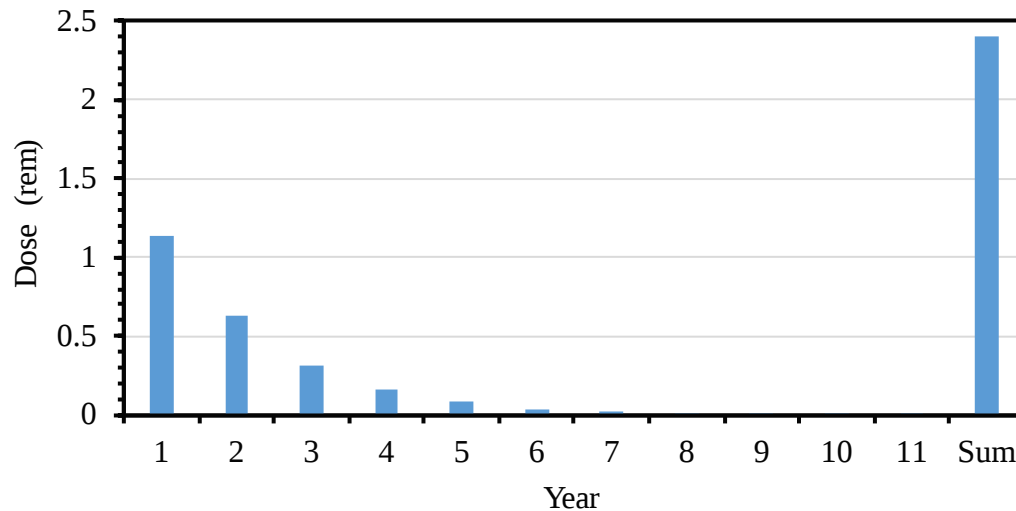


Figure B-3. Annual Lung Doses from 2-month Inhalation Exposure in Year 1 at MPC for Air to Insoluble ^{239}Pu Using ICRP 2 Metabolism.

TABLE B-3. ICRP 2* Respiratory Tract, Adapted from Table 10 (ICRP 1959).

Distribution	Readily Soluble Compounds (%)	Other Compounds (%)
Exhaled	25	25
Deposited in upper respiratory tract passages and subsequently cleared to the gastrointestinal tract by mucous [§]	50	50
Deposited in the lower respiratory tract passages and subsequently cleared to bodily fluids through the respiratory tract	25	0
Deposited in the lower respiratory tract passages and subsequently cleared to the gastrointestinal tract by mucous (e.g., prompt clearance)	0	12.5
Deposited in the lower respiratory tract passages and subsequently cleared to bodily fluids through the respiratory tract (e.g., delayed clearance)	0	12.5
Retention half-time for aerosol deposited in lower respiratory tract passages and subsequently cleared (delayed clearance)	Other Compounds	Half-life (d)
	Default	120
	Plutonium	365
	Thorium	1,461

* Unchanged from ICRP 1955 recommendation (ICRP 1955)

§ Clearance to gastrointestinal tract is assumed to occur within 24 hours

TABLE B-4. ICRP 2 Biological System Parameters, General and for Pu (ICRP 1959).

Parameter	Value	Parameter	Value
Fraction of Pu uptake to blood from GI tract	3×10^{-5}	Fraction of inhalation intake reaching bone (readily soluble)	0.2
Fraction of Pu in blood to bone	0.80	Half-life of Pu in bone	200 y
Fraction of Pu in blood to liver	0.15	Half- life of Pu in liver	82 y
Fraction of Pu in blood to kidneys	0.02	Half- life of Pu in kidneys	88 y

TABLE 5. ICRP Report 26 Dose Equivalent Limits (ICRP 1977).

Application	Annual Limit
Total Effective Dose Equivalent (TEDE)	5 rem (50 mSv)
Deep Dose Equivalent & Committed Effective Dose Equivalent (CEDE)	50 rem to an individual organ or tissue, except lens
Lens of Eye	15 (150 mSv)
Skin	50 (500 mSv)
Extremities	50 (500 mSv)

TABLE 6. Tissue Weighting Factors, w_T .

Tissue	ICRP 26	ICRP 60	ICRP 103
Gonads	0.25	0.2	0.08
Breast	0.15	0.05	0.12
Red bone marrow (RBM)	0.12	0.12	0.12
Lung	0.12	0.12	0.12
Thyroid	0.03	0.05	0.04
Bone surfaces (BS)	0.03	0.01	0.01
Colon	-	0.12	0.12
Stomach	-	0.12	0.12
Bladder	-	0.05	0.04
Esophagus	-	0.05	0.04
Liver	-	0.05	0.04
Brain	-	-	0.01
Kidney	-	-	-
Salivary glands	-	-	0.01
Skin	-	0.01	0.01
Remainder	0.30*	0.05 [§]	0.12 [‡]

* Five remaining tissues with the highest dose equivalent values, $w_T = 0.06$ for each tissue

§ Remaining tissues: adrenals, brain, upper large intestine, small intestine, kidney, muscle, pancreas spleen, thymus, and uterus. If a single tissue among remainder tissues has an equivalent dose greater than one with a specified w_T , then a w_T of 0.025 shall be applied to that tissue with 0.025 applied to the average of the other remaining tissues.

‡ Remaining tissues: adrenals, extrathoracic (ET) region, gall bladder, heart, kidneys, lymphatic nodes, muscle, oral mucosa, pancreas, prostate (♂), small intestine, spleen, thymus, and uterus/cervix (♀). Arithmetic mean of 13 remaining tissues.

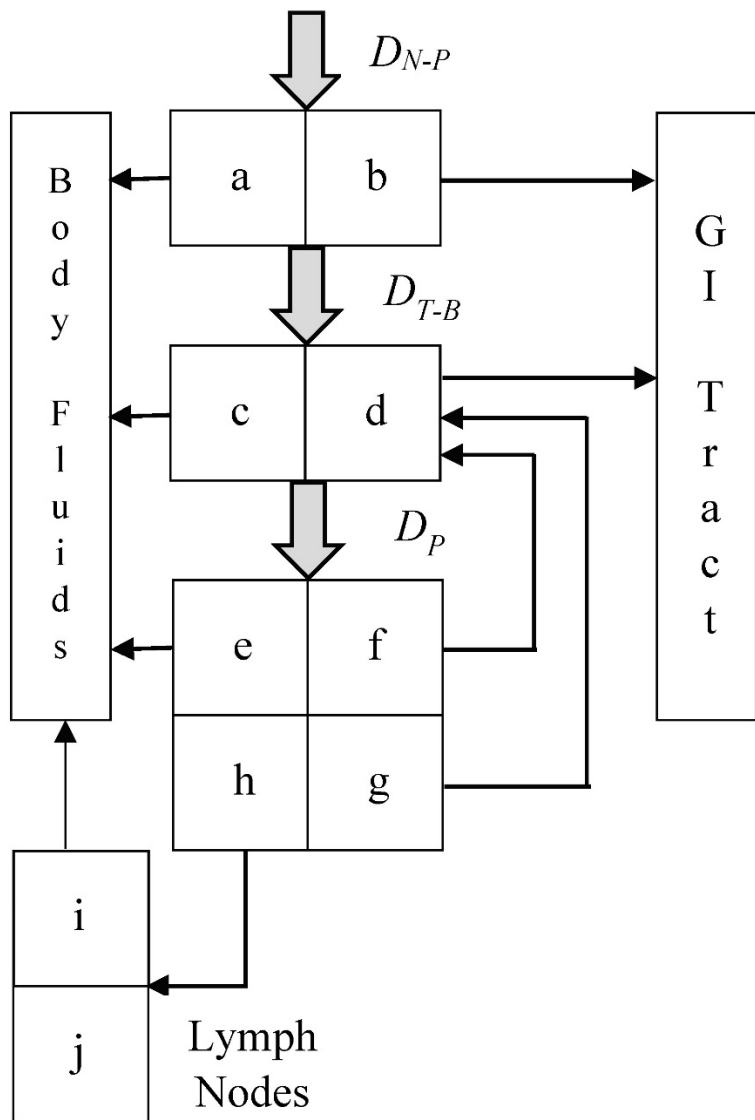


Figure B-4. ICRP Report 30 Respiratory Tract Model Diagram.

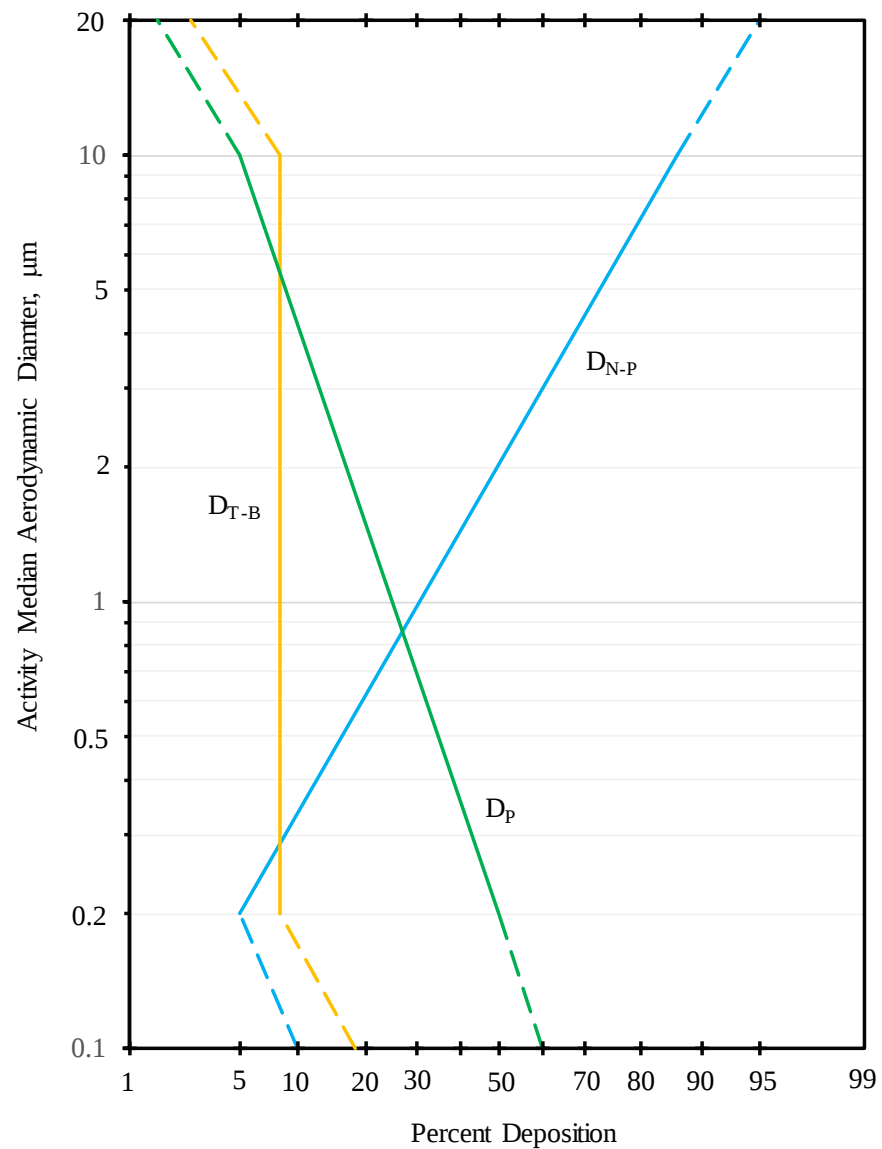


Figure B-5. ICRP Report 30, Aerosol Deposition Model for Regions of Respiratory Tract.

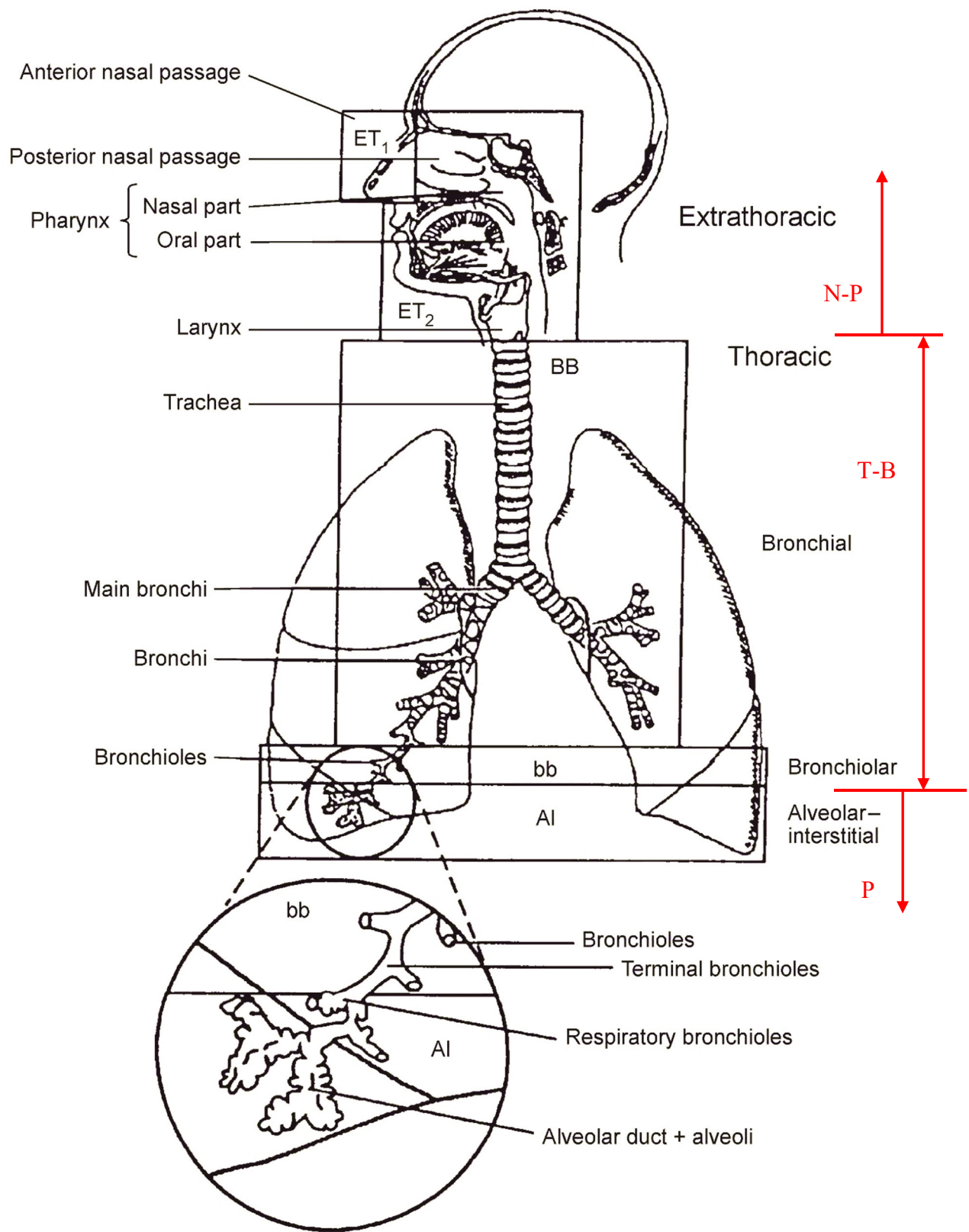


Figure B-6. Detailed Diagram of Respiratory Tract
Following Detail of ICRP Report 66 (ICRP 1994).

TABLE B-7. Deposition Fractions and Retention Half-Life for Inhalation Class W and Y under ICRP 30 Respiratory Tract Model for Aerosols with 1 μm AMAD.

Respiratory Tract Region [Deposition for 1 μm AMAD]	Compartment	Inhalation Class			
		W		Y	
		$\tau_{1/2}$ (d)	Fraction	$\tau_{1/2}$ (d)	Fraction
Naso-Pharynx [$D_{N-P} = 0.3$]	a	0.01	0.1	0.01	0.01
	b	0.4	0.9	0.4	0.99
Tracheo-Bronchial [$D_{T-B} = 0.08$]	c	0.01	0.5	0.01	0.01
	d	0.2	0.5	0.2	0.99
Pulmonary [$D_P = 0.25$]	e	50	0.15	500	0.05
	f	1.0	0.4	1.0	0.4
	g	50	0.4	500	0.4
	h	50	0.05	500	0.15
Lymphatic	i	50	1.0	1000	0.9
	j	NA	NA	∞	0.1

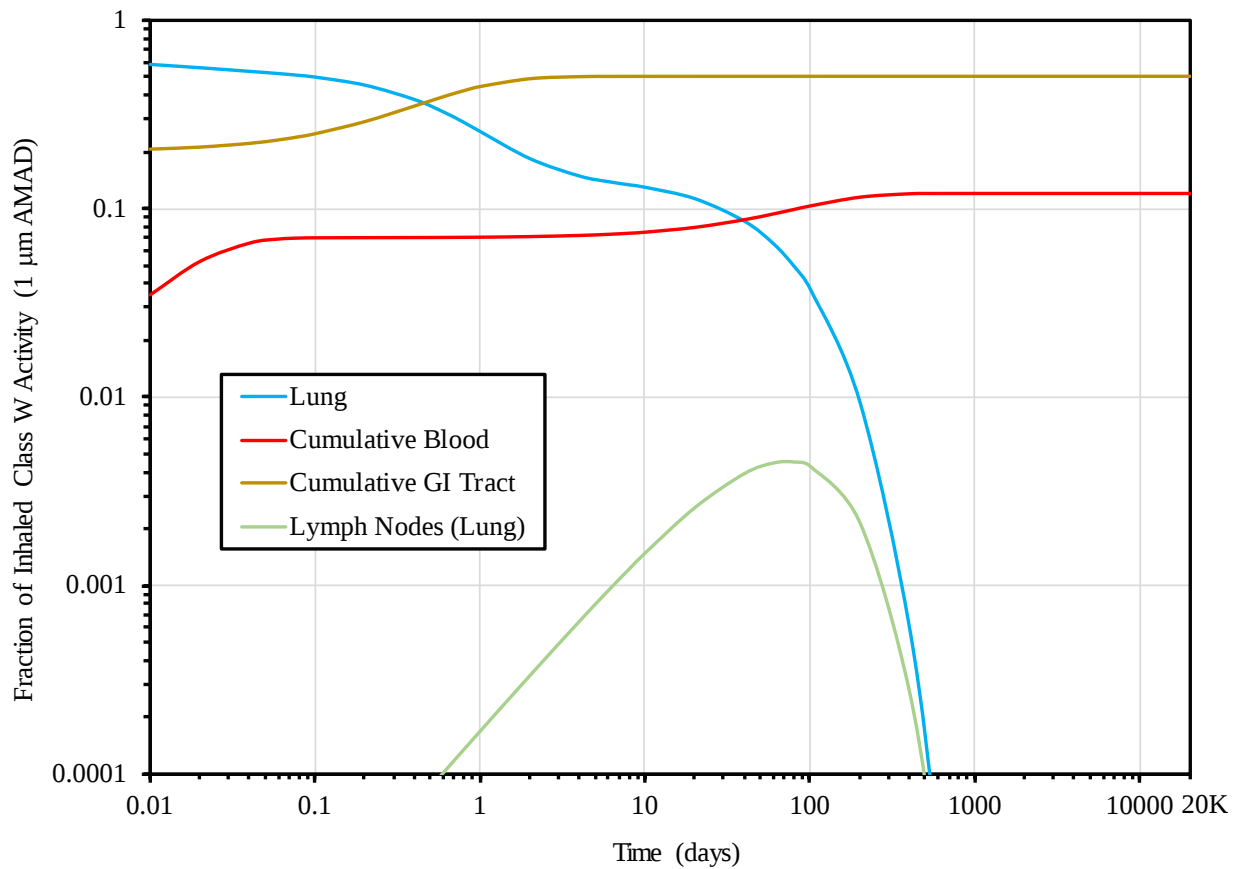


Figure B-7. Retention and Excretion for Inhalation Intakes of Class W ^{239}Pu , Based on 1 μm AMAD Aerosol with ICRP Report 30 Respiratory Tract Model [Radioactive Decay Ignored].

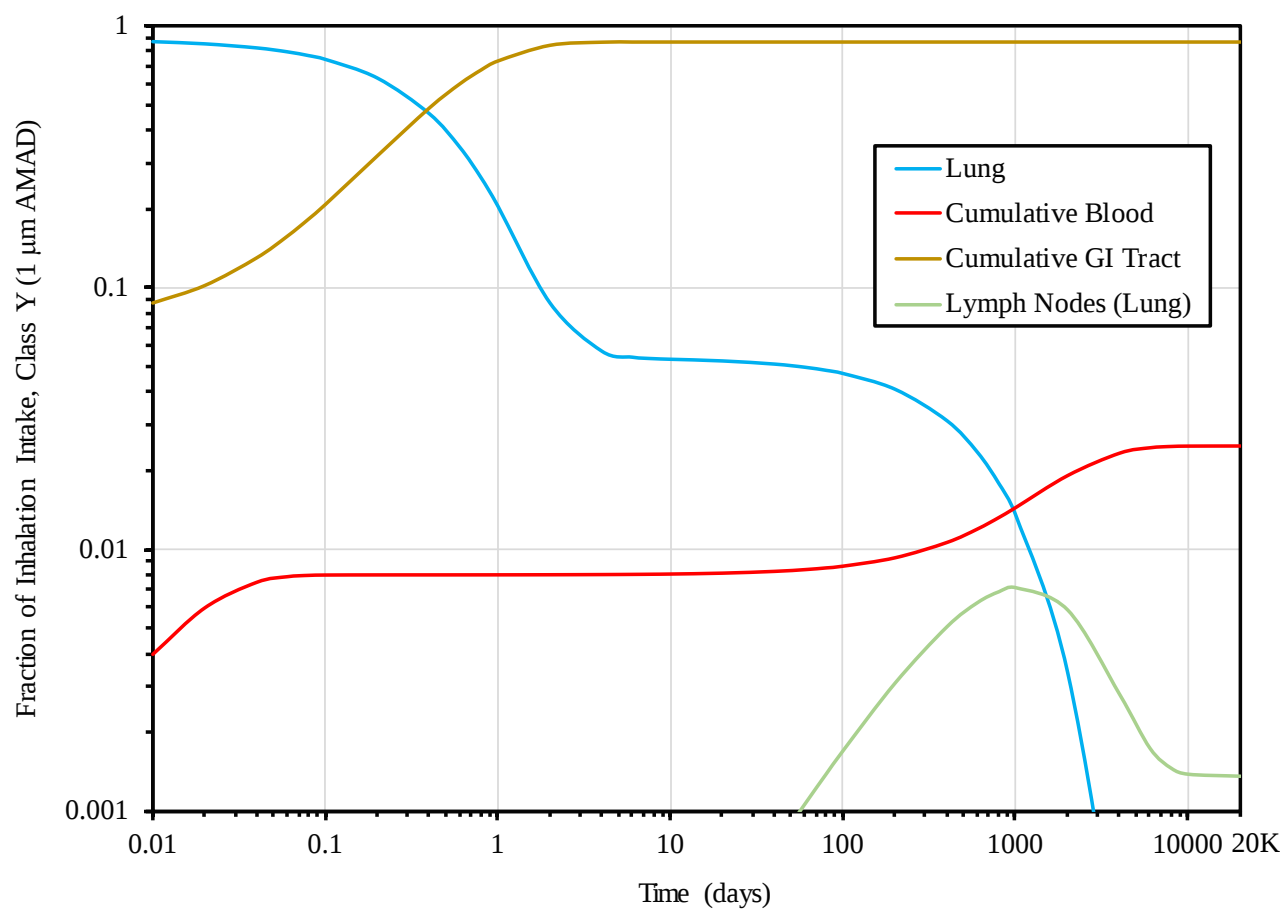


Figure B-8. Retention and Excretion for Inhalation Intakes of Class Y ^{239}Pu , Based on 5 μm AMAD Aerosol with ICRP Report 30 Respiratory Tract Model [Radioactive Decay Ignored].

TABLE B-8. General Systemic Distribution of Plutonium from Blood for Various ICRP Models.

Parameter	ICRP Report							
	No. 2	No. 30	No. 48	No. 56	No. 67		No. 141	
Fraction to bone (or surfaces) from blood	0.80	0.45	0.45	0.45	0.50		0.30	
Fraction to liver from blood	0.15	0.45	0.45	0.45	0.30		0.60	
Liver 1 st compartment from blood	NA	NA	NA	NA	0.30		0.60	
Liver 2 nd compartment from blood	NA	NA	NA	NA	0		0	
Liver 3 rd compartment from blood	NA	NA	NA	NA	NA		0	
Fraction to kidneys from blood	0.02	-	-	-	0.005		0.0005	
Fraction to soft tissues from blood*	-	0.10	0.10	0.10	0.20		0.10	
Fraction to testes from blood	-	0.00035	0.00035	0.00035	0.00035		0.00035	
Fraction to ovaries from blood	-	0.00011	0.00011	0.00011	0.00011		0.00011	
Half-life in bone (or surfaces)	200 y	100 y	50 y	50 y	-		-	
Trabecular surface to volume	-	-	-	-	7.7 y		15.4 y	
Trabecular surface to marrow	-	-	-	-	3.8 y		3.8 y	
Cortical surface to volume	-	-	-	-	4.6 y		93 y	
Cortical surface to marrow	-	-	-	-	23 y		23 y	
Trabecular volume to marrow	-	-	-	-	3.8 y		3.8 y	
Cortical volume to marrow	-	-	-	-	23 y		23 y	
Cortical/trabecular marrow to blood	-	-	-	-	0.25 y		0.25 y	
Half-life in liver	82 y	40 y	20 y	20 y	-		-	
Liver 1 st compartment	NA	NA	NA	NA	14.3 y	to GI	2.1 y	to GI
					1.1 y	to liver 2	0.42 y	to liver 1
Liver 2 nd compartment	NA	NA	NA	NA	9 y	to blood	1.25 y	to blood
							5 y	to liver 2
Liver 3 rd compartment	NA	NA	NA	NA	NA	NA	15 y	to blood
Half-life in kidneys	88 y	-	-	-	1.4 y		15 y	
Half-life in soft tissues*	-	-	-	-	-		-	
Slow turnover	-	-	-	-	100 y		15 y	
Intermediate turnover	-	-	-	-	2 y		1.4 y	
Half-life in testes	-	∞	∞	∞	10 y		5 y	
Half-life in ovaries	-	∞	∞	∞	10 y		5 y	

* Includes early excreta

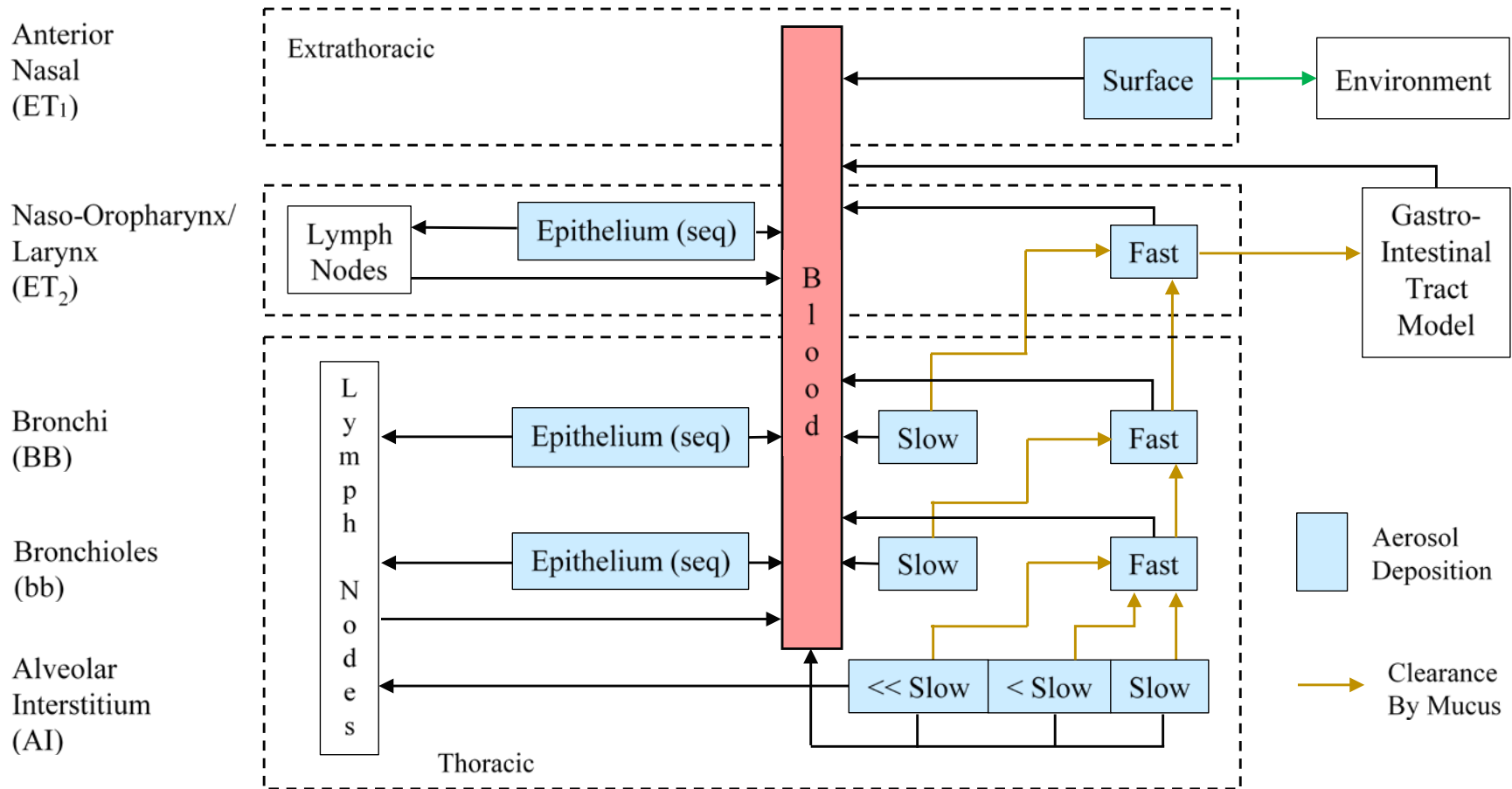


Figure B-9. ICRP Report 66 Respiratory Tract Model Diagram.

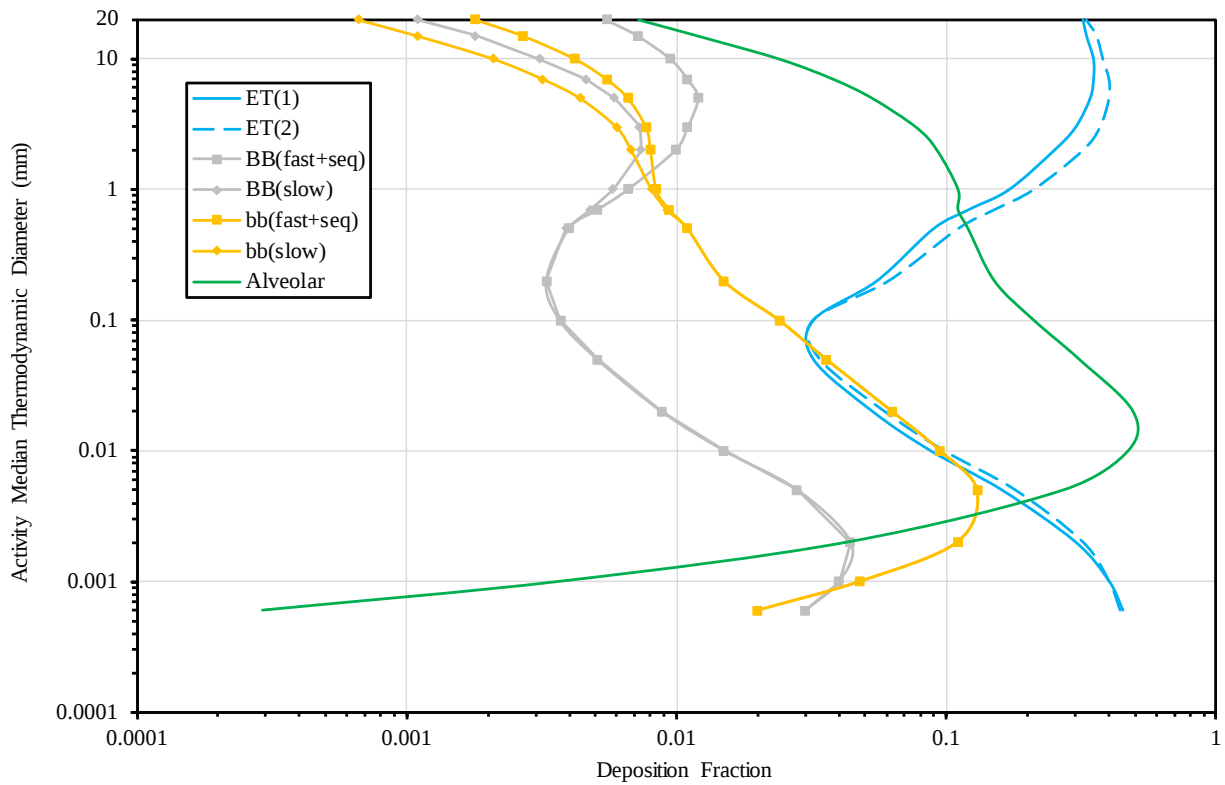


Figure B-10. ICRP Report 66, Aerosol Deposition for Regions of Respiratory Tract.

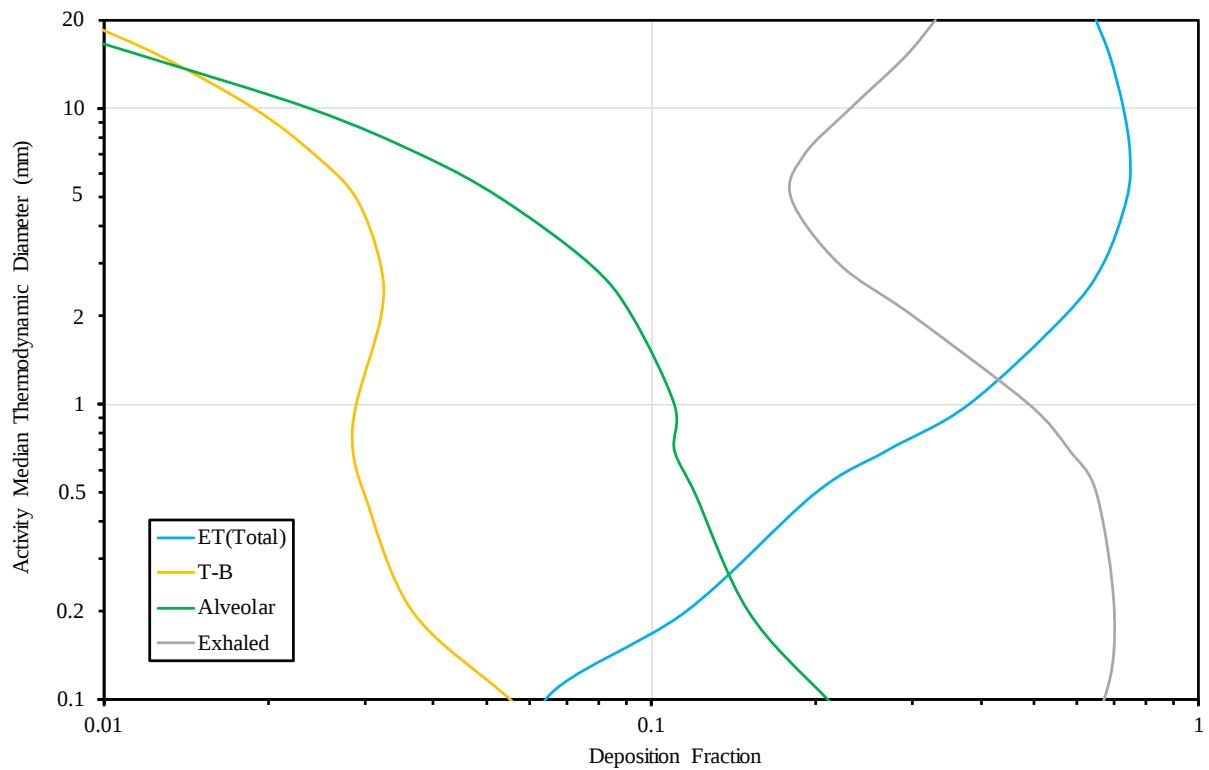


Figure B-11. ICRP Report 66, Aerosol Deposition for Consolidated Regions of Respiratory Tract.

TABLE B-9. Deposition Fractions and Retention Half-Life for Compartments of ICRP 66 Respiratory Tract Model for Aerosols.

Compartment	Compartment Deposition Fraction	Figure B-9 Notation	Clearance Disposition Compartment	Transfer Rate (d ⁻¹)	Retention Half-Life (d)
ET ₁	1	Surface	Environment	1	0.69
ET ₂	0.9995	Fast	GI Tract	100	0.0069
ET _{seq}	0.0005	Epithelium (seq)	LN _{ET}	0.001	693
LN _{ET}	NA	Lymph Nodes	-	-	-
BB ₁	$0.993 - f_s$	Fast	ET ₂	10	0.069
BB ₂	f_s	Slow	ET ₂	0.03	23.1
BB _{seq}	0.007	Epithelium (seq)	LN _{TH}	0.01	69
bb ₁	$0.993 - f_s$	Fast	BB ₁	2	0.35
bb ₂	f_s	Slow	BB ₁	0.03	23
bb _{seq}	0.007	Epithelium (seq)	LN _{TH}	0.01	69
AI ₁	0.3	Slow	bb ₁	0.02	35
AI ₂	0.6	< Slow	bb ₁	0.001	693
AI ₃	0.1	<< Slow	bb ₁	0.0001	6930
LN _{TH}	NA	Lymph Nodes	-	-	-

* $f_s = 0.5$ for $d_{ae} \leq 2.5 \sqrt{\rho/\chi} \mu\text{m}$, where ρ is density (g cm^{-3}), χ is particle shape factor, and d_{ae} is aerodynamic equivalent diameter; $f_s = e^{-0.63(d_{ae}\sqrt{\rho\chi}-2.5)}$ for $d_{ae} > 2.5 \sqrt{\rho/\chi} \mu\text{m}$; for PuO_2 density $\sim 9.5 \text{ g cm}^{-3}$; $\chi = 1.5$ recommended

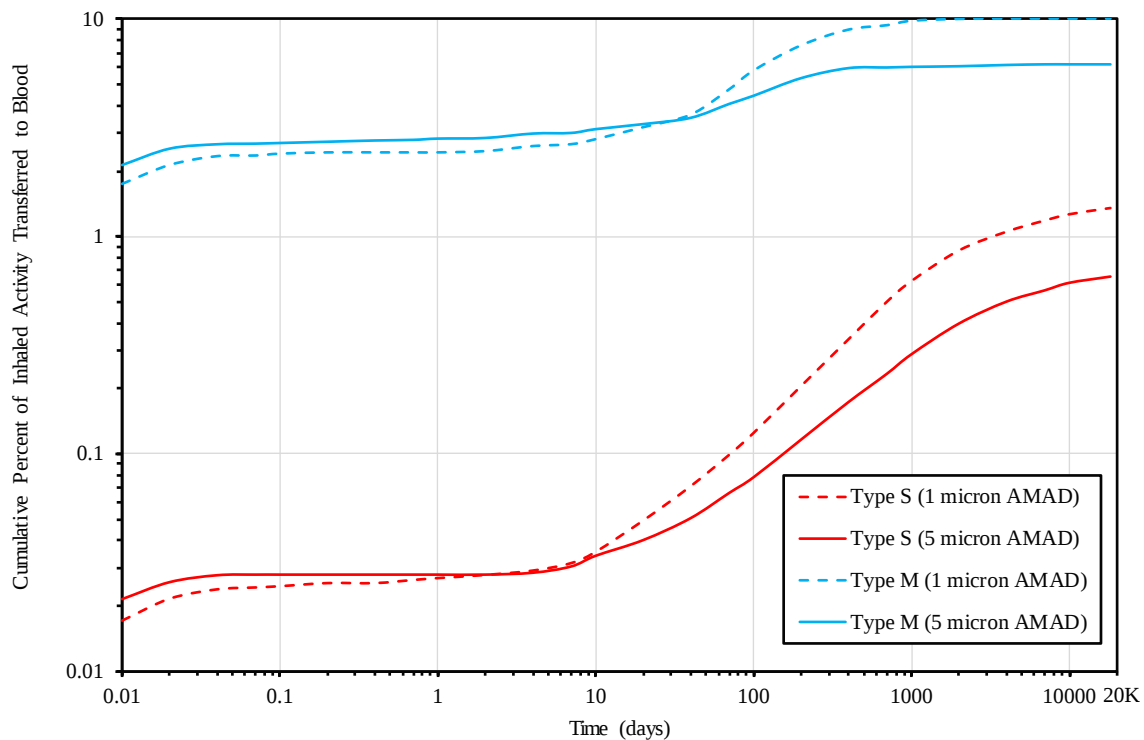


Figure B-12. ICRP Report 66, Cumulative Percent of Inhaled Activity Transferred to Blood, Adapted from Figure 25 of ICRP Report 66 (ICRP 1994).

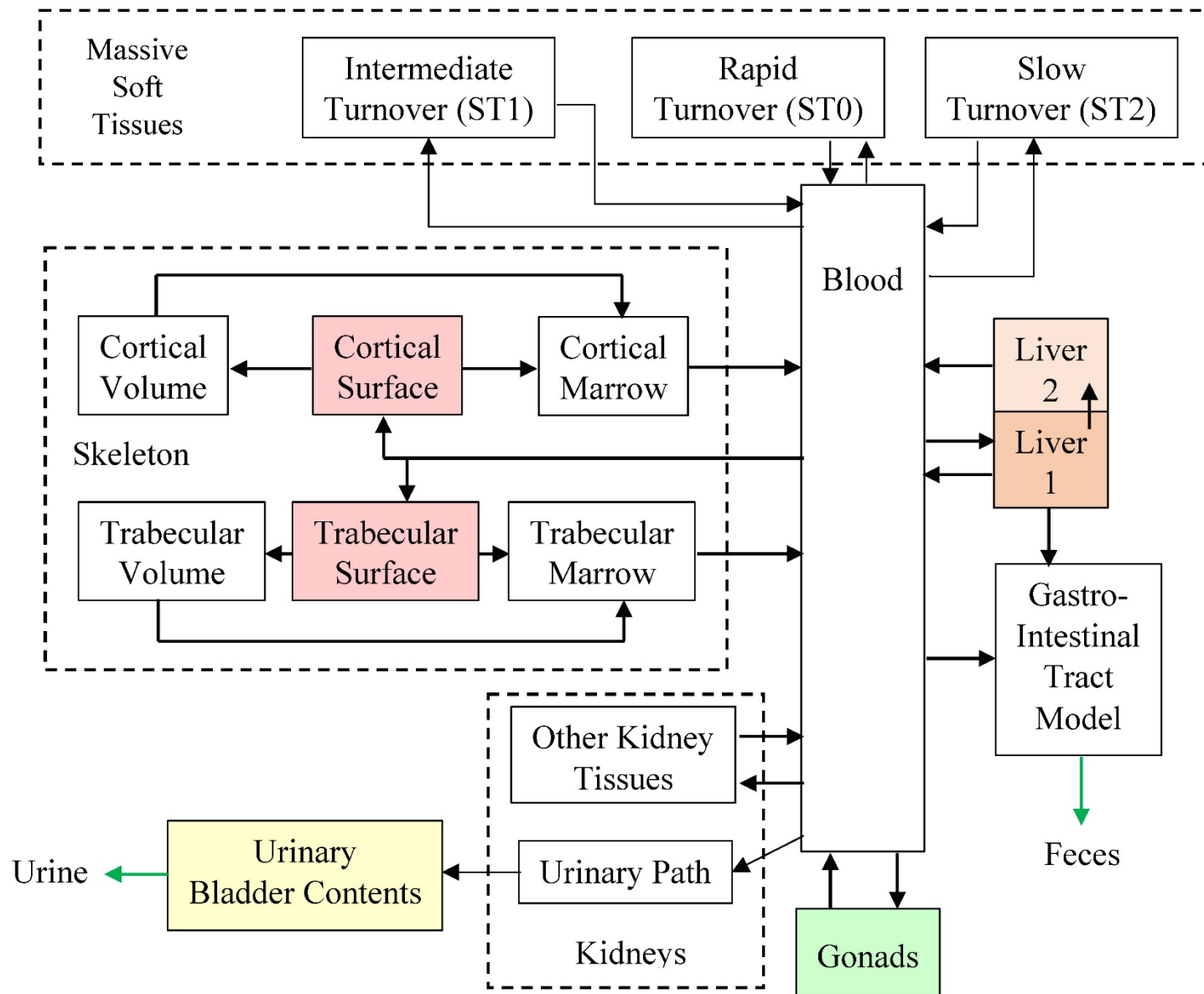


Figure B-13. ICRP Report 67 Systemic Metabolism Model for Plutonium.

TABLE B-10. Inhalation Dose Coefficients, ^{239}Pu , 1 μm AMAD (ICRP Report 71), 5 μm AMAD (ICRP Report 68, Effective Only) and Committed Equivalent Organ/Tissue Doses for 75 nCi Acute Inhalation Intake.

Organ/Tissue		Committed Equivalent and Effective Doses (rem μCi^{-1})		W_T	Type S		
					Committed Equivalent Dose [CED] (rem) for 75 nCi	Weighted CED (rem)	Fraction of CED
		Type M	Type S				
Adrenals		1.0 E+1	1.2 E+0	-	0.09	-	-
Bladder Wall		1.0 E+1	1.2 E+0	0.05	0.09	4.44 E-03	0.00098
Bone Surface		5.6 E+3	6.7 E+2	0.01	50.0	0.500	0.11
Brain		1.0 E+1	1.2 E+0	-	0.09	-	-
Breast		1.0 E+1	1.2 E+0	0.05	0.09	4.44 E-03	0.00098
GI Tract	Esophagus	1.0 E+1	1.2 E+0	0.05	0.09	4.44 E-03	0.00098
	St Wall	1.0 E+1	1.2 E+0	0.12	0.09	1.07 E-02	0.0024
	SI Wall	1.0 E+1	1.2 E+0	-	0.09	-	-
	ULI Wall	1.0 E+1	1.2 E+0	0.01	0.09	8.89 E-04	0.00020
	LLI Wall	1.0 E+1	1.2 E+0	0.01	0.09	9.17 E-04	0.00020
	Colon	1.0 E+1	1.2 E+0	0.12	0.09	1.10 E-02	0.0024
Kidneys		2.4 E+1	3.0 E+0	0.01	0.22	2.22 E-03	0.00049
Liver		1.2 E+3	1.4 E+2	0.05	10.8	0.542	0.12
Muscle		1.0 E+1	1.2 E+0	0.01	0.09	8.89 E-04	0.00020
Ovaries		7.4 E+1	8.9 E+0	0.2	0.67	0.133	0.029
Pancreas		1.0 E+1	1.2 E+0	-	0.09	-	-
Red Bone Marrow		2.7 E+2	3.4 E+1	0.12	2.5	0.303	0.067
Respirat. Tract	Ex. Thor. Air	3.3 E+1	1.4 E+2	0.01	10.6	0.106	0.023
	Lungs	1.2 E+2	3.2 E+2	0.12	24.2	2.90	0.64
Skin		1.0 E+1	1.2 E+0	0.01	0.09	8.89 E-04	0.00020
Spleen		1.0 E+1	1.2 E+0	-	0.09	-	-
Testes		7.8 E+1	9.3 E+0	0.2	0.69	0.139	0.031
Thymus		1.0 E+1	1.2 E+0	-	0.09	-	-
Thyroid		1.0 E+1	1.2 E+0	0.05	0.09	4.44 E-03	0.00098
Uterus		1.0 E+1	1.2 E+0	-	0.09	-	-
Remainder		1.0 E+1	1.3 E+0	-	0.09	-	-
Effective Dose		1.9 E+2	6.0 E+1	-	-	4.54	-
Effective (5 μm AMAD)		1.3 E+2	3.1 E+1	-	-	-	-

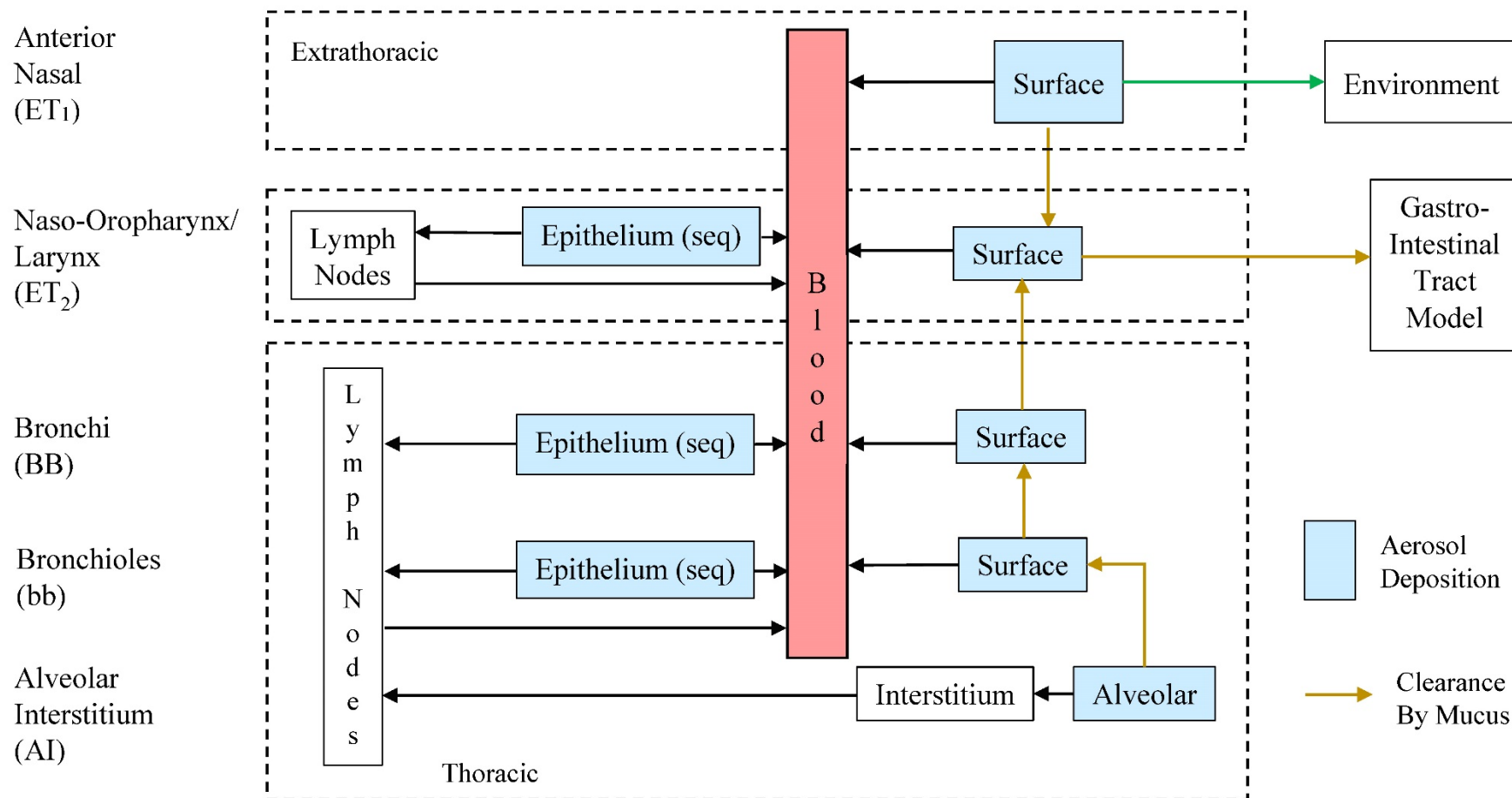


Figure B-14. ICRP Report 130 Respiratory Tract Model Diagram.

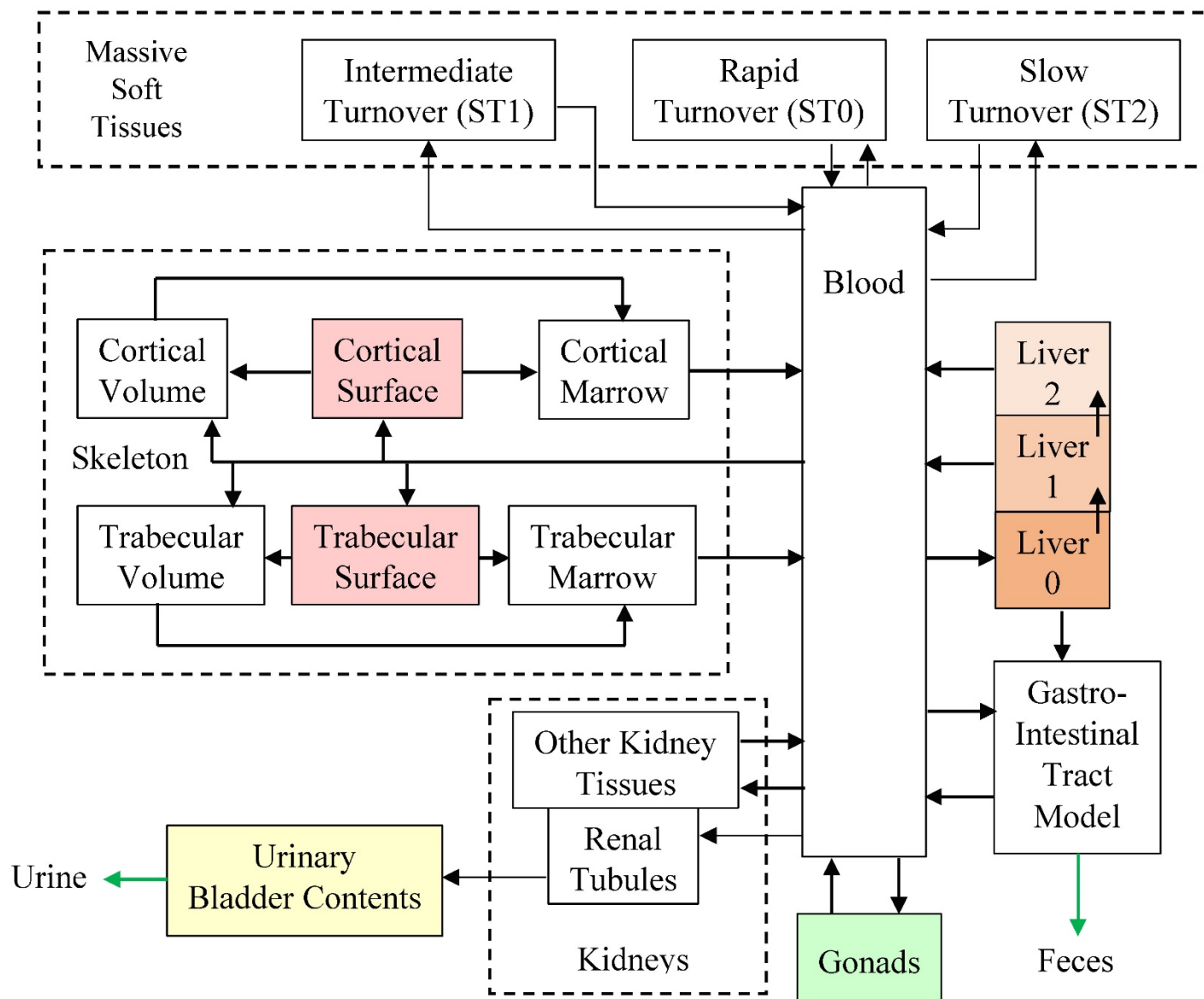


Figure B-15. ICRP Report 141 Systemic Metabolism Model for Plutonium, excluding Detail of Two-Component Blood.

TABLE B-11. Deposition Fractions and Retention Half-Life for Compartments of ICRP 130 Respiratory Tract Model for Aerosols.

Compartment	Compartment Deposition Fraction	Figure B-14 Notation	Clearance Disposition Compartment	Transfer Rate (d^{-1})	Retention Half-Life (d)
ET ₁	1	Surface	Environment	0.6	1.16
			ET' ₂	1.5	0.46
ET' ₂	0.998	Surface	GI Tract	100	0.0069
ET _{seq}	0.002	Epithelium (seq)	LN _{ET}	0.001	693
LN _{ET}	NA	Lymph Nodes	-	-	-
BB'	0.998	Surface	ET' ₂	10	0.069
BB _{seq}	0.002	Epithelium (seq)	LN _{TH}	0.001	693
bb'	0.998	Surface	BB ₁	0.2	3.47
bb _{seq}	0.002	Epithelium (seq)	LN _{TH}	0.001	693
ALV	1	Alveolar	bb'	0.002	347
			INT	0.001	693
INT	NA	Interstitium	LN _{TH}	0.00003	23100
LN _{TH}	NA	Lymph Nodes	-	-	-

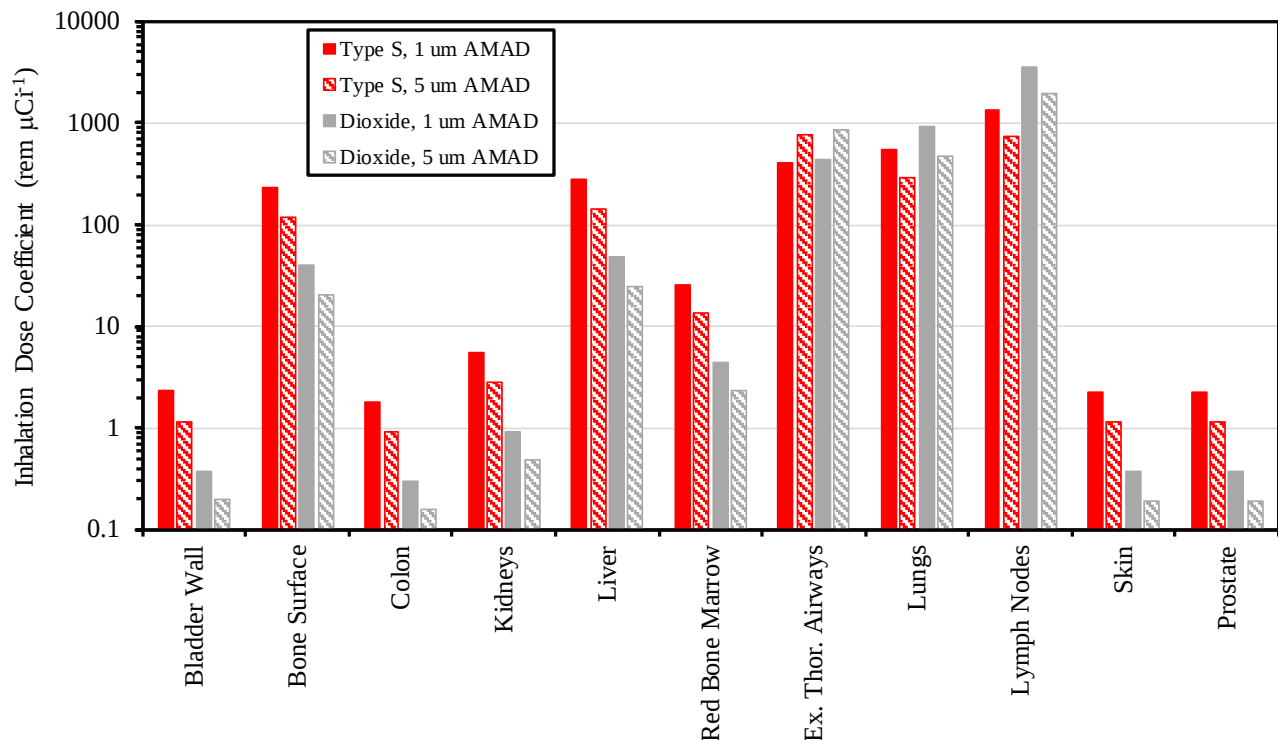


Figure B-16. Inhalation Dose Coefficient Values of Key Tissues for Inhalation Type S and PuO₂, 1 and 5 μm AMAD Aerosols, Adults, ICRP 141. Values Courtesy of Keith Eckerman (2020).

TABLE B-12. Inhalation Dose Coefficients and Weighted Fractions of Effective Dose for ^{239}Pu , ICRP Reports 103, 130, and 141.

Organ/Tissue		Committed Equivalent Dose (rem μCi^{-1})				W_T	Weighted Fraction of Effective Dose			
		Type S ($f_A = 5 \times 10^{-6}$)		PuO ₂ of Pu/U Oxides ($f_A = 2 \times 10^{-6}$)			Type S ($f_A = 5 \times 10^{-6}$)		PuO ₂ of Pu/U Oxides ($f_A = 2 \times 10^{-6}$)	
		1 μm	5 μm	1 μm	5 μm		1 μm	5 μm	1 μm	5 μm
Adrenals		1.92	0.96	0.322	0.167	0.0092	0.0002	0.0002	0.00002	0.00002
Bladder Wall		2.29	1.15	0.370	0.196	0.04	0.001	0.001	0.0001	0.0001
Bone Surface		229	118	40.7	20.4	0.01	0.023	0.020	0.003	0.002
Brain		2.22	1.15	0.370	0.192	0.01	0.0002	0.0002	0.00002	0.00002
Breast		2.22	1.15	0.370	0.192	0.12	0.003	0.002	0.0003	0.0003
GI Tract	Esophagus	1.85	0.93	0.315	0.163	0.04	0.001	0.001	0.0001	0.0001
	St Wall	1.85	0.93	0.311	0.159	0.12	0.002	0.002	0.0002	0.0002
	SI Wall	1.78	0.93	0.303	0.155	0.0092	0.0002	0.0001	0.00002	0.00002
	Colon	1.78	0.93	0.303	0.155	0.12	0.002	0.002	0.0002	0.0002
Heart		2.00	1.04	0.340	0.174	0.0092	0.0002	0.0002	0.0000	0.0000
Kidneys		5.55	2.81	0.925	0.481	0.0092	0.001	0.0004	0.0001	0.0001
Liver		281	144	48.1	24.4	0.04	0.11	0.098	0.013	0.011
Muscle		2.26	1.15	0.370	0.196	0.0092	0.0002	0.0002	0.00002	0.00002
Salivary Glands		2.22	1.15	0.370	0.192	0.01	0.0002	0.0002	0.00002	0.00002
Oral Mucosa		2.33	1.18	0.407	0.204	0.0092	0.0002	0.0002	0.00002	0.00002
Pancreas		1.92	0.96	0.322	0.167	0.0092	0.0002	0.0002	0.00002	0.00002
Red Bone Marrow		25.9	13.3	4.44	2.33	0.12	0.031	0.027	0.004	0.003
Respirat. Tract	Ex. Thor. Air	407	777	444	851	0.0092	0.037	0.12	0.027	0.092
	Lungs	555	292	925	481	0.12	0.65	0.60	0.73	0.68
Lymph Nodes		1330	740	3550	1960	0.0092	0.12	0.12	0.22	0.21
Skin		2.22	1.15	0.370	0.192	0.01	0.0002	0.0002	0.00002	0.00002
Spleen		1.59	0.81	0.270	0.137	0.0092	0.0001	0.0001	0.00002	0.00001
Testes		16.3	8.14	2.81	1.44	0.08	0.013	0.011	0.0015	0.0014
Thymus		2.22	1.15	0.370	0.192	0.0092	0.0002	0.0002	0.00002	0.00002
Thyroid		2.04	1.04	0.348	0.178	0.04	0.0008	0.0007	0.0001	0.0001
Gall Bladder		2.22	1.15	0.370	0.192	0.0092	0.0002	0.0002	0.00002	0.00002
Prostate		2.22	1.15	0.370	0.192	0.0092	0.0002	0.0002	0.00002	0.00002
Effective Dose (males)		102	58.9	151	85.4	-	-	-	-	-

TABLE B-13. Lifetime Probability (Percents) of Developing* and Dying from Cancer for 23 Sites, 2010 – 2012, American Cancer Society, Surveillance Research (ACS 2016).

Site	Males		Females	
	Developing	Dying	Developing	Dying
All Sites †	42.1	22.6	37.6	19.1
Brain & ONS	0.7	0.5	0.5	0.4
Breast	0.1	< 0.1	12.3	2.7
Colorectal	4.7	2.0	4.4	1.8
Esophagus	0.8	0.8	0.2	0.2
Hodgkin lymphoma	0.2	< 0.1	0.2	< 0.1
Kidney & renal pelvis	2.0	0.6	1.2	0.3
Larynx	0.6	0.2	0.1	< 0.1
Leukemia	1.8	1.0	1.2	0.7
Liver & intrahepatic bile duct	1.3	0.9	0.5	0.5
Lung & bronchus	7.2	6.3	6.0	4.9
Melanoma of skin‡	3.0	0.5	1.9	0.2
Myeloma	0.9	0.5	0.6	0.4
Non-Hodgkin lymphoma	2.4	0.9	1.9	0.7
Oral cavity & pharynx	1.6	0.4	0.7	0.2
Ovary	--	--	1.3	1.0
Pancreas	1.5	1.4	1.5	1.3
Prostate	14.0	2.6	--	--
Stomach	1.1	0.5	0.7	0.3
Testis	0.4	< 0.1	--	--
Thyroid	0.6	0.1	1.7	0.1
Urinary bladder§	3.8	0.9	1.1	0.3
Uterine cervix	--	--	0.6	0.2
Uterine corpus	--	--	2.8	0.6

* For those who are cancer free.

† All sites excludes basal cell and squamous cell skin cancers and in-situ cancers except urinary bladder.

‡ Statistics are for whites.

§ Includes invasive and in-situ cancer cases.

Appendix C

16th Air Force, Air Sampling Data during Palomares Recovery and Other Environmental Data

TABLE C-1. Air Sampling Data during Palomares Recovery.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)*	(pCi m ⁻³)	
19-Jan	1st Camp				1	1030					Negative
19-Jan	1st Camp				2	1100					Negative
20-Jan	#3				1-3	1322			20	9.0	100
20-Jan	#3				2-3	1515			340	153	1700
20-Jan	#3				3-3	1550			80	36.0	400
21-Jan	#3				4-3	1035				0.0	Negative
21-Jan	#3				5-3	1030				0.0	Negative
21-Jan	#3				6-3	1155			20	9.0	100
21-Jan	#3				7-3	1155			20	9.0	100
21-Jan	#3				8-3	1345					Negative
21-Jan	#3				9-3	1345					Negative
22-Jan	#3				10-3	1150					Negative
22-Jan	#3				11-3	1150					Negative
22-Jan	#3				12-3	1155					Negative
22-Jan	#3				13-3	1155					Negative
22-Jan	#3				14-3	1550			100	45.0	500
22-Jan	#3				15-3	1550			340	153	1700
23-Jan	#3				16-3	1140					Negative
23-Jan	#3				17-3	1140					Negative
23-Jan	#3				18-3	1227					Negative
23-Jan	#3				19-3	1301					Negative
23-Jan	#3				20-3	1428					Negative
24-Jan	#3				21-3	1045					Negative
24-Jan	#3				22-3	1226					Negative
24-Jan	#3				23-3	1324					Negative
25-Jan	#3				25-3	958			10	4.5	50
25-Jan	#3				26-3	1057			10	4.5	50
25-Jan	#3				27-3	1057					Negative
27-Jan	#3				1-3	1015					Negative

TABLE C-1. Air Sampling Data during Palomares Recovery, continued.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)	(pCi m ⁻³)	
27-Jan	#3				2-3	1015					Negative
27-Jan	#3				3-3	1130					Negative
27-Jan	#3				4-3	1130					Negative
27-Jan	#3				5-3	1400					Negative
27-Jan	#3				6-3	1400					Negative
27-Jan	#3				7-3	1440					Negative
28-Jan	#3			5171	8-3	945	1015	30			Negative
28-Jan	#3			5642	9-3	1000	1030	30			Negative
28-Jan	#3			5171	10-3	1030	1100	30			Negative
28-Jan	#3			5171	11-3	1330	1400	30			Negative
28-Jan	#3			5642	12-3	1330	1400	30			Negative
28-Jan	#3			5171	13-3	1415	1445	30			Negative
28-Jan	#3			5642	14-3	1415	1445	30			Negative
28-Jan	#3			5642	15-3	1500	1530	30			Negative
29-Jan	#3			5642	16-3	1030	1100	30			Negative
29-Jan	#3			5171	17-3	1030	1100	30			Negative
29-Jan	#3			5642	18-3	1105	1135	30			Negative
29-Jan	#3			5642	19-3	1310	1340	30			Negative
29-Jan	#3			5171	20-3	1310	1340	30			Negative
29-Jan	#3			5642	21-3	1355	1425	30			Negative
29-Jan	#3			5171	22-3	1355	1425	30			Negative
31-Jan	#3			5642	23-3	930	1000	30			Negative
31-Jan	#3			5171	24-3	930	1000	30	40	18.0	200
31-Jan	#3			5642	25-3	1030	1100	30			Negative
31-Jan	#3			5171	26-3	1030	1100	30			Negative
31-Jan	#3			5642	27-3	1400	1430	30			Negative
31-Jan	#3			5171	28-3	1430	1430	30			Negative
31-Jan	#3			5642	29-3	1445	1515	30			Negative
1-Feb	#3			5642	30-3	1100	1130	30	20	9.0	100

TABLE C-1. Air Sampling Data during Palomares Recovery, continued.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)	(pCi m ⁻³)	
1-Feb	#3			5171	31-3	1100	1130	30			Negative
1-Feb	#3			5642	32-3	1315	1345	30	10	4.5	50
1-Feb	#3			5171	33-3	1315	1345	30			Negative
1-Feb	#3			5642	34-3	1400	1430	30			Negative
1-Feb	#3			5171	35-3	1400	1430	30			Negative
2-Feb	#2			5171	1-2	1000	1030	30			Negative
2-Feb	#2			5642	2-2	1000	1030	30			Negative
2-Feb	#2			5171	3-2	1045	1115	30			Negative
2-Feb	#2			5442	3-2	1045	1115	30			Negative
2-Feb	#2			5171	5-2	1120	1150	30			Negative
2-Feb	#2			5642	6-2	1120	1150	30			Negative
2-Feb	#3			5171	7-2	1430	1500	30			Negative
2-Feb	#3			5642	8-2	1430	1500	30			Negative
2-Feb	#3			5642	9-2	1500	1530	30			Negative
3-Feb	#3			5171	1-3	1425	1505	30			Negative
3-Feb	#3			5642	2-3	1425	1505	30			Negative
3-Feb	#3			5642	3-3	1510	1540	30			Negative
3-Feb	#3			5171	4-3	1510	1540	30			Negative
3-Feb	#3			5171	10-2	910	940	30			Negative
3-Feb	#3			5642	11-2	910	940	30			Negative
3-Feb	#3			5171	12-2	1000	1030	30			Negative
3-Feb	#3			5642	13-2	1000	1030	30			Negative
3-Feb	#3			5642	14-2	1100	1130	30			Negative
3-Feb	#3			5171	15-2	1100	1130	30			Negative
4-Feb	#3			5642	1-3	1015	1045	30			Negative
4-Feb	#3			5171	2-3	1015	1045	30	10	4.5	50
4-Feb	#3			5171	3-3	1045	1115	30	20	9.0	100
4-Feb	#3			5642	4-3	1045	1115	30	20	9.0	100
4-Feb	#3			5171	5-3	1130	1200	30	10	4.5	50

TABLE C-1. Air Sampling Data during Palomares Recovery, continued.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)	(pCi m ⁻³)	
4-Feb	#3			5642	6-3	1130	1200	30	20	9.0	100
4-Feb	#3			5171	7-3	1345	1415	30			Negative
4-Feb	#3			5642	8-3	1345	1415	30	10	4.5	50
4-Feb	#3			5171	9-3	1420	1450	30	10	4.5	50
4-Feb	#3			5642	10-3	1420	1450	30	15	6.8	75
4-Feb	#3			5171	11-3	1500	1530	30			Negative
4-Feb	#3			5642	12-3	1500	1530	30	10	4.5	50
5-Feb	#3			5642	13-3	940	1010	30			Negative
5-Feb	#3			5171	14-3	940	1010	30	10	4.5	50
5-Feb	#3			5642	15-3	1020	1050	30	50	22.5	250
5-Feb	#3			5642	16-3	1020	1050	30	90	40.5	450
5-Feb	#3			5642	17-3	1220	1250	30			Negative
5-Feb	#3			5171	18-3	1220	1250	30			Negative
5-Feb	#3			5171	19-3	1315	1345	30			Negative
5-Feb	#3			5642	20-3	1315	1345	30	20	9.0	100
5-Feb	#3			5642	21-3	1415	1445	30	10	4.5	50
5-Feb	#3			5171	22-3	1415	1445	30	10	4.5	50
5-Feb	#3			5642	23-3	1530	1600	30			Negative
5-Feb	#3			5171	24-3	1530	1600	30			Negative
7-Feb	#2			3516	1-2	1400	1430	30			Negative
7-Feb	#2			3516	2-2	1530	1600	30			Negative
7-Feb	#2			3516	3-2	1605	1635	30			Negative
7-Feb	#2			6972	1-2	1330	1400	30			Negative
7-Feb	#2			6572	2-2	1400	1430	30			Negative
7-Feb	#2			6572	3-2	1500	1530	30			Negative
7-Feb	#2			6972	4-2	1500	1530	30			Negative
7-Feb	#2			5642	25-2	920	950	30			Negative
7-Feb	#2			5171	26-2	920	950	30			Negative
7-Feb	#2			5642	27-2	1010	1040	30			Negative

TABLE C-1. Air Sampling Data during Palomares Recovery, continued.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)	(pCi m ⁻³)	
7-Feb	#2			5171	28-2	1010	1040	30			Negative
7-Feb	#2			5171	29-2	1200	1300	30			Negative
7-Feb	#2			5642	30-2	1200	1230	30			Negative
7-Feb	#2			5642	31-2	1355	1425	30			Negative
7-Feb	#2			5171	32-2	1355	1425	30	10	4.5	50
7-Feb	#2			5171	33-2	1440	1510	30			Negative
7-Feb	#2			5642	34-2	1440	1510	30			Negative
8-Feb	#3				1	1430	1500	30	10	4.5	50
8-Feb	#3			7098	2	1430	1500	30	100	45.0	500
8-Feb	#3			7090	3	1500	1530	30	60	27.0	300
8-Feb	#3			3449	4	1500	1530	30	10	4.5	50
8-Feb	#3			7089	5	1530	1600	30	10	4.5	50
8-Feb	#3			7090	6	1530	1600	30			Negative
8-Feb	#3			5642	1	945	1015	30			Negative
8-Feb	#3			5171	2	945	1015	30			Negative
8-Feb	#3			5171	3	1030	1100	30			Negative
8-Feb	#3				4	1400	1400	30			Negative
8-Feb	#3				3	900	930	30			Negative
8-Feb	#3				4	1000	1030	30	20	9.0	100
8-Feb	#3				5	1530	1600	30			Negative
9-Feb	#3			7089	7-3	830	900	30	20	9.0	100
9-Feb	#3			7090	8-3	830	900	30	20	9.0	100
9-Feb	#3			7089	9-3	915	945	30	30	13.5	150
9-Feb	#3			7090	10-3	915	945	30	10	4.5	50
9-Feb	#3			7090	11-3	1000	1030	30	10	4.5	50
9-Feb	#3			7089	12-3	1000	1030	30	10	4.5	50
9-Feb	#3			7089	33-3	1045	1115	30	20	9.0	100
9-Feb	#3			7090	14-3	1045	1115	30	10	4.5	50
9-Feb	#3			7089	15-3	1410	1440	30	10	4.5	50

TABLE C-1. Air Sampling Data during Palomares Recovery, continued.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)	(pCi m ⁻³)	
9-Feb	#3			7070	16-3	1410	1440	30	10	4.5	50
10-Feb	#3			7089	17-3	850	920	30			Negative
10-Feb	#3			7090	18-3	850	920	30			Negative
10-Feb	#3			7090	19-3	1130	1200	30			Negative
10-Feb	#3			7089	20-3	1130	1200	30			Negative
10-Feb	#3			7089	21-3	1315	1415	30			Negative
10-Feb	#3			7090	22-3	1345	1415	30			Negative
10-Feb	#3			7089	23-3	1430	1500	30	10	4.5	50
10-Feb	#3			7090	24-3	1430	1500	30			Negative
10-Feb	#3			7089	25-3	1515	1545	30			Negative
10-Feb	#3			7090	26-3	1515	1545	30			Negative
9-Feb	#2				7-2	1530	1600	30			Negative
11-Feb	#2			7089	1-2	1515					Negative
11-Feb	#2			7090	2-2	1515					Negative
11-Feb	#2			7089	3-2	1520	1550	30	10	4.5	50
11-Feb	#2			7090	4-2	1520	1550	30			Negative
11-Feb	#2			7089	5-2	1600	1630	30	10	4.5	50
11-Feb	#2			7090	6-2	1600	1630	30			Negative
12-Feb	#3			7080	1-3	830	900	30			Negative
12-Feb	#3			7090	2-3	830	900	30			Negative
12-Feb	#3			7089	3-3	915	945	30			Negative
12-Feb	#3			7089	4-3	915	945	30			Negative
12-Feb	#3			7089	5-3	1030	1100	30			Negative
12-Feb	#3			7090	6-3	1110	1140	30			Negative
12-Feb	#3			7089	7-3	1315	1345	30			Negative
12-Feb	#3			7089	8-3	1350	1420	30			Negative
12-Feb	#3			7089	9-3	1430	1500	30			Negative
14-Feb	#3			5171	1-3	940	1010	30			Negative
14-Feb	#3			5171	2-3	1015	1045	30			Negative

TABLE C-1. Air Sampling Data during Palomares Recovery, continued.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)	(pCi m ⁻³)	
14-Feb	#3			5171	3-3	1050	1120	30			Negative
14-Feb	#3			5171	4-3	1400	1430	30			Negative
14-Feb	#3			5171	5-3	1435	1505	30			Negative
14-Feb	#3			5171	6-3	1510	1540	30			Negative
15-Feb	#3			5171		900	930	30			Negative
15-Feb	#3			7089		900	930	30			Negative
15-Feb	#3			5171		935	1005	30			Negative
15-Feb	#3			7089		935	1005	30			Negative
15-Feb	#3			5171		1010	1040	30			Negative
15-Feb	#3			7089		1010	1010	30			Negative
15-Feb	#3			7089		1050	1120	30			Negative
15-Feb	#3			5171		1050	1120	30			Negative
15-Feb	#3			5171		1125	1155	30			Negative
15-Feb	#3			7089		1125	1155	30			Negative
15-Feb	#3			7089		1315	1345	30			Negative
15-Feb	#3			5171		1315	1345	30			Negative
15-Feb	#3			5171		1350	1420	30			Negative
15-Feb	#3			7089		1350	1420	30			Negative
15-Feb	#3			7089		1445	1515	30			Negative
15-Feb	#3			5171		1445	1515	30			Negative
16-Feb	#3			5171		900	930	30			Negative
16-Feb	#3			7089		900	930	30			Negative
16-Feb	#3			5171		945	1015	30			Negative
16-Feb	#3			7089		945	1015	30			Negative
17-Feb	#3			5642	1	845	915	30			Negative
17-Feb	#3			5171	1a	835	905	30			Negative
17-Feb	#3			7089	1b	830	900	30			Negative
17-Feb	#3			5642	2	930	1000	30			Negative
17-Feb	#3			5171	2a	955	1025	30			Negative

TABLE C-1. Air Sampling Data during Palomares Recovery, continued.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)	(pCi m ⁻³)	
17-Feb	#3			7089	2b	1030	1100	30			Negative
17-Feb	#3			5642	3	1350	1420	30	10	4.5	50
17-Feb	#3			5171	3a	1030	1130	30			Negative
17-Feb	#3			5642	4	1425	1455	30			Negative
17-Feb	#3			5171	4a	1335	1405	30			Negative
17-Feb	#3			5079	4b	1405	1435	30			Negative
17-Feb	#3			5642	5	1500	1530	30			Negative
17-Feb	#3			5171	5a	1410	1440	30	40	18.0	200
17-Feb	#3			7089	5b	1440	1510	30			Negative
17-Feb	#3			5642	6	1535	1605	30			Negative
17-Feb	#3			5171	6a	1445	1515	30			Negative
17-Feb	#3			7089	6b	1515	1545	30			Negative
17-Feb	#3			5972	17	1905	1935	30			Negative
19-Feb	#3			6972	1	840	910	30			Negative
19-Feb	#3			7089	1a	830	900	30			Negative
19-Feb	#3			6972	2	912	943	30			Negative
19-Feb	#3			7089	2a	920	950	30			Negative
19-Feb	#3			6972	3	1320	1350	30			Negative
19-Feb	#3			7089	3a	955	1020	30			Negative
19-Feb	#3			6972	4	1355	1425	30			Negative
19-Feb	#3			7089	4a	1030	1100	30			Negative
19-Feb	#3			6972	5	1425	1455	30			Negative
19-Feb	#3			7089	5a	1310	1340	30			Negative
19-Feb	#3			6972	6a	1350	1420	30			Negative
21-Feb	#3			6972	1	940	1010	30			Negative
21-Feb	#3			5171	1a	900	930	30			Negative
21-Feb	#3			7089	1b	900	930	30			Negative
21-Feb	#3			5972	2	1015	1045	30			Negative
21-Feb	#3			5171	2a	940	1010	30			Negative

TABLE C-1. Air Sampling Data during Palomares Recovery, continued.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)	(pCi m ⁻³)	
17-Feb	#3			7089	2b	1030	1100	30			Negative
17-Feb	#3			5642	3	1350	1420	30	10	4.5	50
17-Feb	#3			5171	3a	1030	1130	30			Negative
17-Feb	#3			5642	4	1425	1455	30			Negative
17-Feb	#3			5171	4a	1335	1405	30			Negative
17-Feb	#3			5079	4b	1405	1435	30			Negative
17-Feb	#3			5642	5	1500	1530	30			Negative
17-Feb	#3			5171	5a	1410	1440	30	40	18.0	200
17-Feb	#3			7089	5b	1440	1510	30			Negative
17-Feb	#3			5642	6	1535	1605	30			Negative
17-Feb	#3			5171	6a	1445	1515	30			Negative
17-Feb	#3			7089	6b	1515	1545	30			Negative
17-Feb	#3			5972	17	1905	1935	30			Negative
19-Feb	#3			6972	1	840	910	30			Negative
19-Feb	#3			7089	1a	830	900	30			Negative
19-Feb	#3			6972	2	912	943	30			Negative
19-Feb	#3			7089	2a	920	950	30			Negative
19-Feb	#3			6972	3	1320	1350	30			Negative
19-Feb	#3			7089	3a	955	1020	30			Negative
19-Feb	#3			6972	4	1355	1425	30			Negative
19-Feb	#3			7089	4a	1030	1100	30			Negative
19-Feb	#3			6972	5	1425	1455	30			Negative
19-Feb	#3			7089	5a	1310	1340	30			Negative
19-Feb	#3			6972	6a	1350	1420	30			Negative
21-Feb	#3			6972	1	940	1010	30			Negative
21-Feb	#3			5171	1a	900	930	30			Negative
21-Feb	#3			7089	1b	900	930	30			Negative
21-Feb	#3			5972	2	1015	1045	30			Negative
21-Feb	#3			5171	2a	940	1010	30			Negative

TABLE C-1. Air Sampling Data during Palomares Recovery, continued.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)	(pCi m ⁻³)	
21-Feb	#3			7089	2b	940	1010	30			Negative
21-Feb	#3			6972	3	1310	1340	30			Negative
21-Feb	#3			5171	3a	1025	1055	30			Negative
21-Feb	#3			7089	3b	1310	1340	30			Negative
21-Feb	#3			6972	4	1352	1423	30			Negative
21-Feb	#3			5171	4a	1400	1430	30			Negative
21-Feb	#3			7089	4b	1352	1423	30			Negative
21-Feb	#3			6972	5	1440	1510	30			Negative
21-Feb	#3			5171	5a	1435	1505	30			Negative
21-Feb	#3			7089	5b	1435	1505	30			Negative
21-Feb	#3			6972	6	1520	1550	30			Negative
21-Feb	#3			5171	6a	1510	1540	30			Negative
21-Feb	#3			7098	6b	1520	1550	30			Negative
22-Feb	#3			6972	1	843	912	29			Negative
22-Feb	#3			5171	1a	835	905	30			Negative
22-Feb	#3			7089	1b	845	915	30			Negative
22-Feb	#3			6972	2	915	945	30			Negative
22-Feb	#3			5171	2a	910	940	30			Negative
22-Feb	#3			7089	2b	920	950	30			Negative
22-Feb	#3			6972	3	950	1020	30			Negative
22-Feb	#3			5171	3a	945	1015	30			Negative
22-Feb	#3			7089	3b	955	1025	30			Negative
22-Feb	#3			6972	4	1040	1110	30			Negative
22-Feb	#3			5171	4a	1020	1050	30			Negative
22-Feb	#3			7089	4b	1030	1100	30			Negative
22-Feb	#3			5171	5	1320	1350	30			Negative
22-Feb	#3			6972	5a	1315	1345	30			Negative
22-Feb	#3			7089	5b	1320	1350	30			Negative
22-Feb	#3			5171	6	1400	1430	30			Negative

TABLE C-1. Air Sampling Data during Palomares Recovery, continued.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)	(pCi m ⁻³)	
22-Feb	#3			6972	6a	1350	1420	30			Negative
22-Feb	#3			7089	6b	1400	1430	30			Negative
22-Feb	#3			5171	7	1445	1515	30			Negative
22-Feb	#3			6972	7a	1430	1500	30			Negative
22-Feb	#3			7089	7b	1440	1510	30			Negative
22-Feb	#3			7089	8	1525	1555	30			Negative
23-Feb	#3			7089	8b	1525	1555	30			Negative
23-Feb	#3			5642	1	1330	1400	30			Negative
23-Feb	#3			5642	2	1405	1535	30			Negative
23-Feb	#3			5642	3	1445	1515	30			Negative
23-Feb	#3			5642	4	1525	1555	30			Negative
23-Feb	#2			5642	1	900	930	30			Negative
23-Feb	#2			6972	1a	930	1000	30			Negative
23-Feb	#2			5171	1b	930	1000	30			Negative
23-Feb	#2			5972	2a	1010	1040	30			Negative
23-Feb	#2			5171	2b	1005	1035	30			Negative
23-Feb	#2			5972	3a	1045	1115	30			Negative
23-Feb	#2			5171	3b	1040	1110	30	40	18.0	200
23-Feb	#2			5972	4a	1120	1150	30			Negative
23-Feb	#2			5171	4b	1115	1145	30	20	9.0	100
24-Feb	#3			7089	1	830	900	30			Negative
24-Feb	#3			6972	1a	855	925	30			Negative
24-Feb	#3			5171	1b	845	915	30			Negative
24-Feb	#3			7089	2	905	955	30			Negative
24-Feb	#3			6972	2a	930	1000	30			Negative
24-Feb	#3			5171	2b	920	950	30			Negative
24-Feb	#3			7089	3	940	1010	30			Negative
24-Feb	#3			6972	3a	1015	1045	30			Negative
24-Feb	#3			5171	3b	1350	1420	30			Negative

TABLE C-1. Air Sampling Data during Palomares Recovery, continued.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)	(pCi m ⁻³)	
24-Feb	#3			7089	4	1020	1050	30			Negative
24-Feb	#3			6972	4a	1100	1130	30			Negative
24-Feb	#3			7089	4b	1351	1421	30			Negative
24-Feb	#3			7089	5	1430	1500	30			Negative
24-Feb	#3			5171	5a	1440	1510	30			Negative
24-Feb	#3			6972	5b	1355	1425	30			Negative
24-Feb	#3			7089	6	1505	1535	30			Negative
24-Feb	#3			5171	6a	1515	1545	30			Negative
24-Feb	#3			6972	6b	1430	1500	30			Negative
24-Feb	#3			7089	7	1545	1515	30			Negative
24-Feb	#3			5171	7a	1555	1625	30			Negative
24-Feb	#3			6972	7b	1500	1530	30			Negative
24-Feb	#3			7089	8	1620	1650	30			Negative
24-Feb	#3			5171	8a	1630	1700	30			Negative
24-Feb	#3			6972	8b	1540	1610	30			Negative
25-Feb	#3			7089	1	830	920	30			Negative
25-Feb	#3			5642	1a	855	925	30			Negative
25-Feb	#3			6972	1b	845	915	30			Negative
25-Feb	#3			7089	2	930	1000	30			Negative
25-Feb	#3			5642	2a	930	1000	30			Negative
25-Feb	#3			6972	2b	920	950	30			Negative
25-Feb	#3			7089	3	1020	1050	30			Negative
25-Feb	#3			5642	3a	1025	1055	30			Negative
25-Feb	#3			6972	3b	1015	1045	30			Negative
25-Feb	#3			7089	4	1055	1125	30			Negative
25-Feb	#3			5642	4a	1125	1155	30			Negative
25-Feb	#3			5642	5	1330	1400	30			Negative
25-Feb	#3			7089	6	1400	1430	30			Negative
25-Feb	#3			7089	7	1435	1505	30			Negative

TABLE C-1. Air Sampling Data during Palomares Recovery, continued.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)	(pCi m ⁻³)	
25-Feb	#3			6972	7a	1410	1440	30			Negative
25-Feb	#3			7089	8	1515	1545	30			Negative
25-Feb	#3			6972	8a	1445	1515	30			Negative
25-Feb	#3			6972	9	1520	1550	30			Negative
26-Feb	#3			7089	1a	1320	1350	30			Negative
26-Feb	#3			7089	2a	1400	1430	30			Negative
26-Feb	#3			5642	3a	1400	1430	30			Negative
26-Feb	#3			6972	4a	1410	1440	30			Negative
26-Feb	#3			7089	5a	1435	1505	30			Negative
26-Feb	#3			5642	6a	1435	1505	30			Negative
26-Feb	#3			6972	7a	1445	1515	30			Negative
26-Feb	#3			7089	1	840	910	30			Negative
26-Feb	#3			6972	2	840	910	30			Negative
26-Feb	#3			5642	3	845	915	30			Negative
26-Feb	#3			7089	4	915	945	30			Negative
26-Feb	#3			6972	5	920	950	30			Negative
26-Feb	#3			5642	6	925	955	30			Negative
26-Feb	#3			7089	7	950	1020	30			Negative
26-Feb	#3			6972	8	955	1025	30			Negative
26-Feb	#3			5642	9	1005	1035	30			Negative
27-Feb	#3			5642	1	1330	1400	30			Negative
27-Feb	#3			7089	2	1330	1400	30			Negative
27-Feb	#3			5642	3	1440	1440	30			Negative
27-Feb	#3			1087	4	1410	1440	30			Negative
27-Feb	#3			5642	5	1440	1510	30			Negative
27-Feb	#3			7089	6	1440	1510	30			Negative
27-Feb	#3			5642	7	1525	1555	30			Negative
27-Feb	#3			7089	8	1525	1555	30			Negative
1-Mar	#3			5642	1	840	910	30			Negative

TABLE C-1. Air Sampling Data during Palomares Recovery, continued.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)	(pCi m ⁻³)	
1-Mar	#3			7089	2	845	915	30			Negative
1-Mar	#3			6972	3	845	915	30			Negative
1-Mar	#3			5642	4	920	950	30			Negative
1-Mar	#3			7089	5	920	950	30			Negative
1-Mar	#3			6972	6	920	950	30			Negative
1-Mar	#3			6972	7	955	1025	30			Negative
1-Mar	#3			7089	8	955	1025	30			Negative
1-Mar	#3			5642	9	950	1020	30			Negative
1-Mar	#3			5642	10	1025	1055	30			Negative
1-Mar	#3			7089	11	1030	1100	30			Negative
1-Mar	#3			6972	12	1030	1100	30			Negative
1-Mar	#3			6972	13	1315	1345	30			Negative
1-Mar	#3			5642	14	1315	1345	30			Negative
1-Mar	#3			7089	15	1310	1340	30			Negative
1-Mar	#3			6972	16	1355	1425	30			Negative
1-Mar	#3			5642	17	1355	1425	30			Negative
1-Mar	#3			6972	18	1440	1510	30			Negative
1-Mar	#3			6972	19	1440	1510	30			Negative
1-Mar	#3			5642	20	1540	1610	30			Negative
1-Mar	#3			6972	21	1540	1610	30			Negative
2-Mar	#3			5642	1	950	1020	30			Negative
2-Mar	#3			6972	2	950	1020	30			Negative
2-Mar	#3			7089	3	945	1015	30			Negative
2-Mar	#3			6972	4	1115	1145	30			Negative
2-Mar	#3			5642	5	1115	1145	30			Negative
2-Mar	#3			7089	6	1120	1150	30			Negative
2-Mar	#3			6972	7	1320	1350	30			Negative
2-Mar	#3			7089	8	1320	1350	30			Negative
2-Mar	#3			7089	9	1410	1440	30			Negative

TABLE C-1. Air Sampling Data during Palomares Recovery, continued.

Sampling Date	Site	Location Descriptor†		Staplex Serial #	Sample Number	Sampling Time		Sampling Time (min)	Airborne Concentration		PAC-1S (cpm)
		Primary	Secondary			Start	End		(dpm m ⁻³)	(pCi m ⁻³)	
2-Mar	#3			6972	10	1405	1435	30			Negative
2-Mar	#3			6972	11	1440	1510	30			Negative
2-Mar	#3			7089	12	1445	1515	30			Negative
2-Mar	#2			6972	13	1545	1615	30			Negative
2-Mar	#2			7089	14	1545	1615	30			Negative
7-Mar	#2			7089	1	1440	1510	30			Negative
7-Mar	#2			7089	2	1442	1512	30			Negative
7-Mar	#2			7089	3	1515	1545	30			Negative
12-Mar	#2			7089	12-1	1530	1600	30			Negative
12-Mar	#2			7089	12-2	1600	2630	30			Negative
12-Mar	#2			7089	12-3	1730	1800	30			Negative
12-Mar	#2			7089	12-4	1817	1847	30			Negative
12-Mar	#2			7089	12-5	2030	2100	30			Negative
13-Mar	#2			7089	13-1	1620	1650	30	20	9.0	100
13-Mar	#2			7089	13-2	1800	1830	30	30	13.5	150
14-Mar	#2				14-1	640	740	30	40	18.0	200
14-Mar	#2				14-2	830	900	30	100	45.0	500
14-Mar	#2				14-3	1527	1557	30			Negative
14-Mar	#2				14-4	1645	1715	30			Negative
14-Mar	#2				14-5	1830	1900	30			Negative
14-Mar	#2				14-6	2000	2030	30			Negative
14-Mar	#2				14-7	2030	2100	30			Negative
15-Mar	#2				15-1	1130	1200	30	35	15.8	175
15-Mar	#2				15-2	1900	1930	30	50	22.5	250
15-Mar	#2				15-3	2130	2200	30	70	31.5	350
15-Mar	#2				15-4	1430	1500	30	30	13.5	150
16-Mar	#2				16-1	1015	1045	30	90	40.5	450
16-Mar	#2				16-2	1235	1305	30	50	22.5	250
16-Mar	#2				16-3	1400	1430	30	160	72.1	800

TABLE C-2. Estimated Committed Effective Doses based on Urinary Excretion Data Assuming an Acute Inhalation Intake using ICRP Reports 26/30 Methodology [Iranzo 1988].

Estimated Committed Effective Dose Equivalent		Number of Individuals	Inferred Inhalation Intake, ICRP 26/30, Lung Class Y*
mSv	rem		
< 20	< 2	659	< 6.4 nCi
20 – 50	2 – 5	22	6.4 – 16 nCi
50 – 100	5 – 10	22	16 – 32 nCi
100 – 150	10 – 15	6	32 – 48 nCi
150 - 200	15 - 20	5	48 – 64 nCi

* Some Palomares members were children at the time of the accident.

TABLE C-3. Distance from Ground Zero for Sampling Arcs Established for Operation Roller Coaster Tests in 1963 (USA 1963).

Arc	Distance (ft)	Arc	Distance (ft)	Arc	Distance (ft)
A	1,250	F	7,500	K	15,000
B	2,500	G	8,750	L	17,500
C	3,750	H	10,000	N	23,500
D	5,000	I	11,500	P	35,000
E	6,250	J	13,000	R	48,000

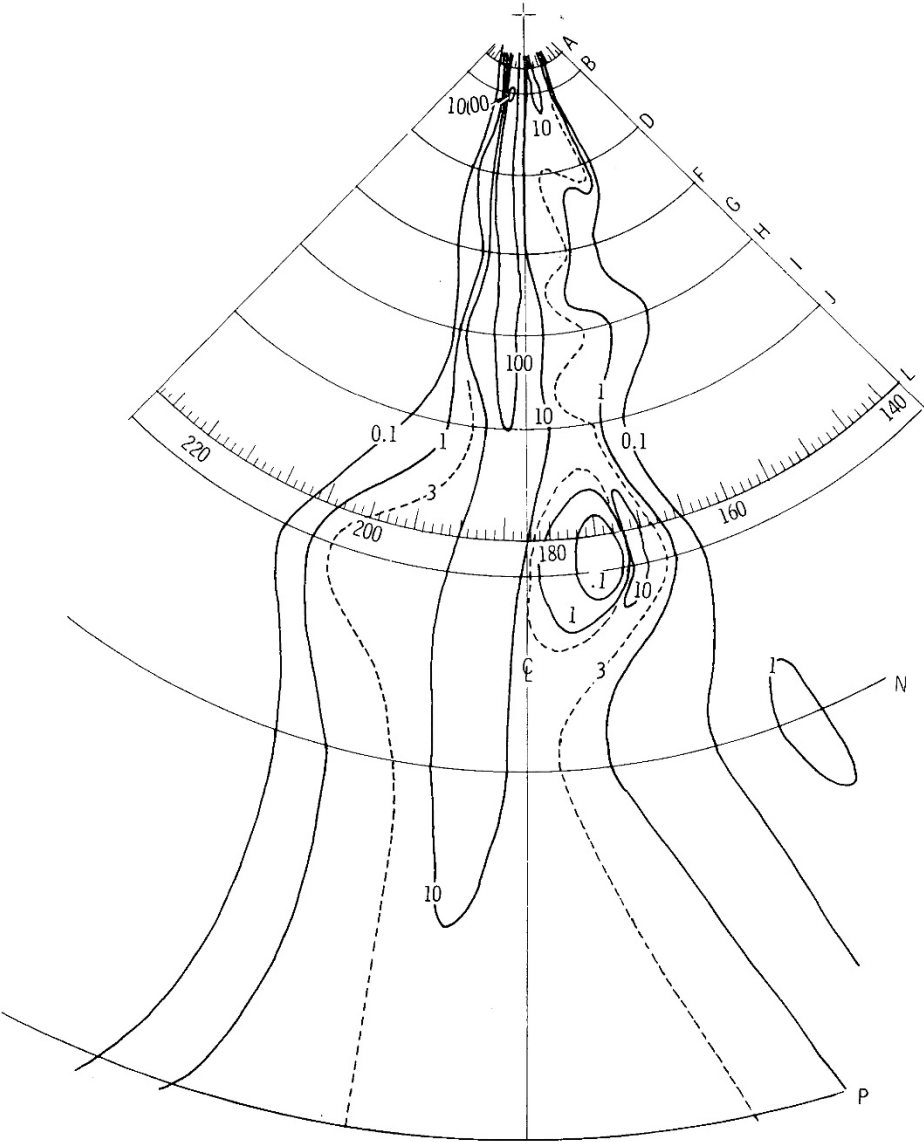


Figure C-1. Double Tracks Respirable Contours, $\mu\text{g sec m}^{-3}$
[Figure 5 from Church *et al.* 1970].

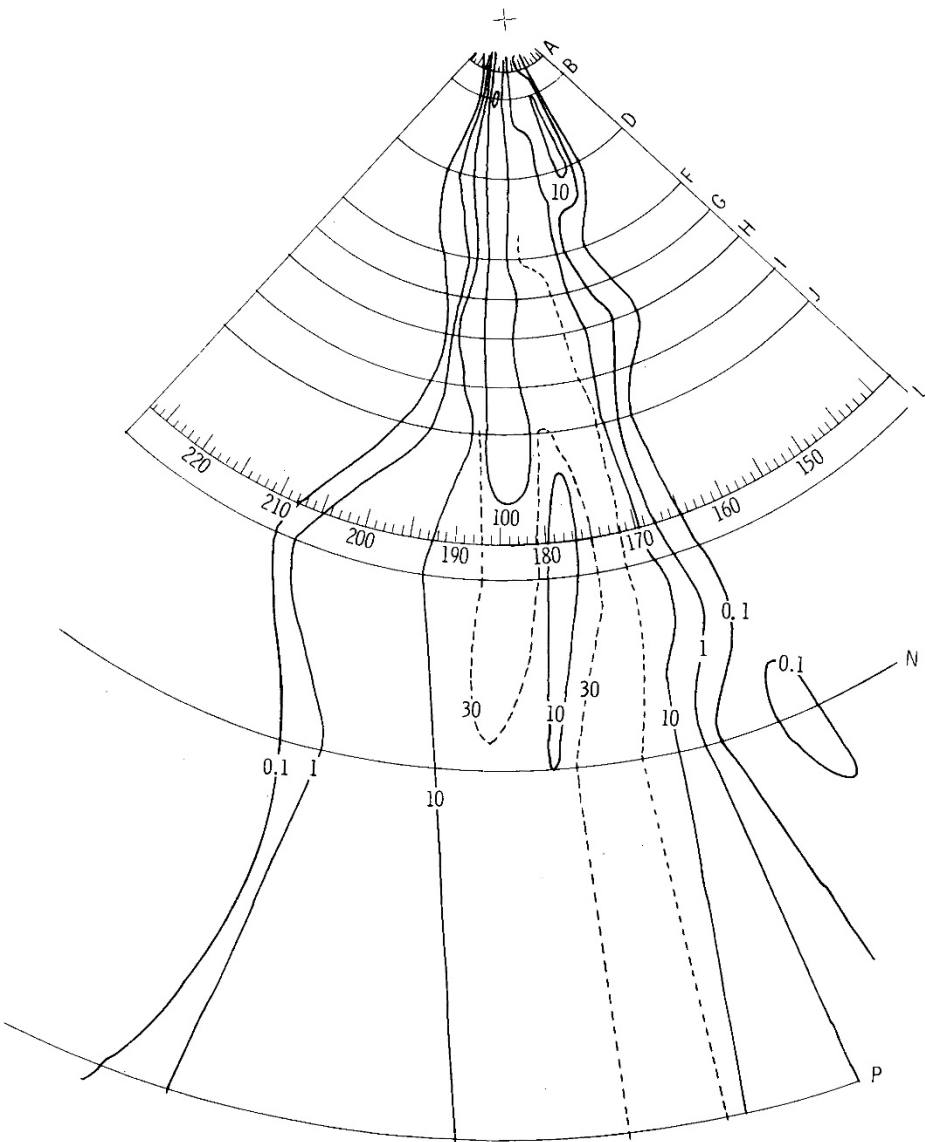


Figure C-2. Double Tracks Total Air Exposure Contours, $\mu\text{g sec m}^{-3}$
[Figure 7 from Church *et al.* 1970].

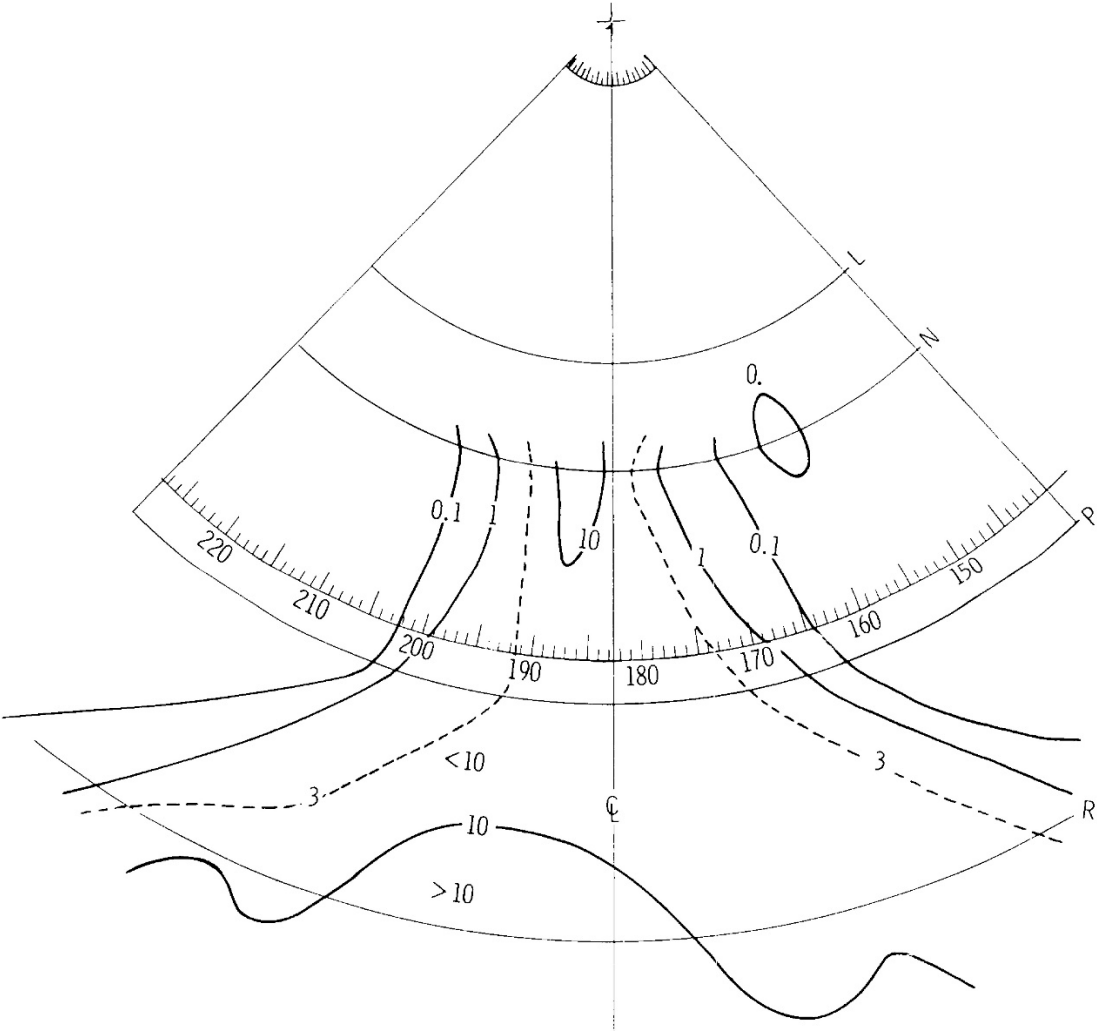


Figure C-3. Double Tracks Respirable Contours, $\mu\text{g sec m}^{-3}$
[Figure 6 from Church *et al.* 1970].

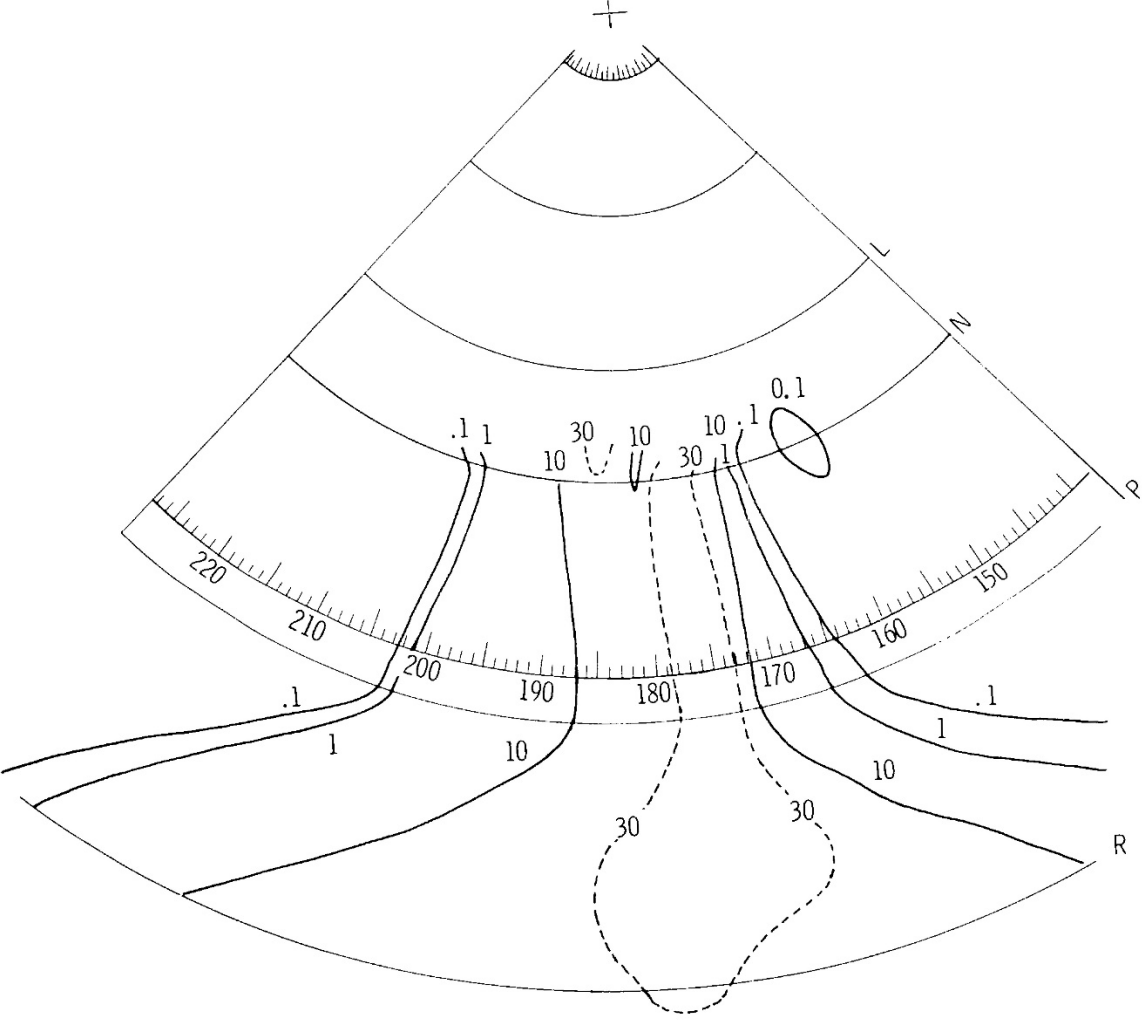


Figure C-4. Double Tracks Total Air Exposure Contours, $\mu\text{g sec m}^{-3}$
[Figure 8 from Church *et al.* 1970].

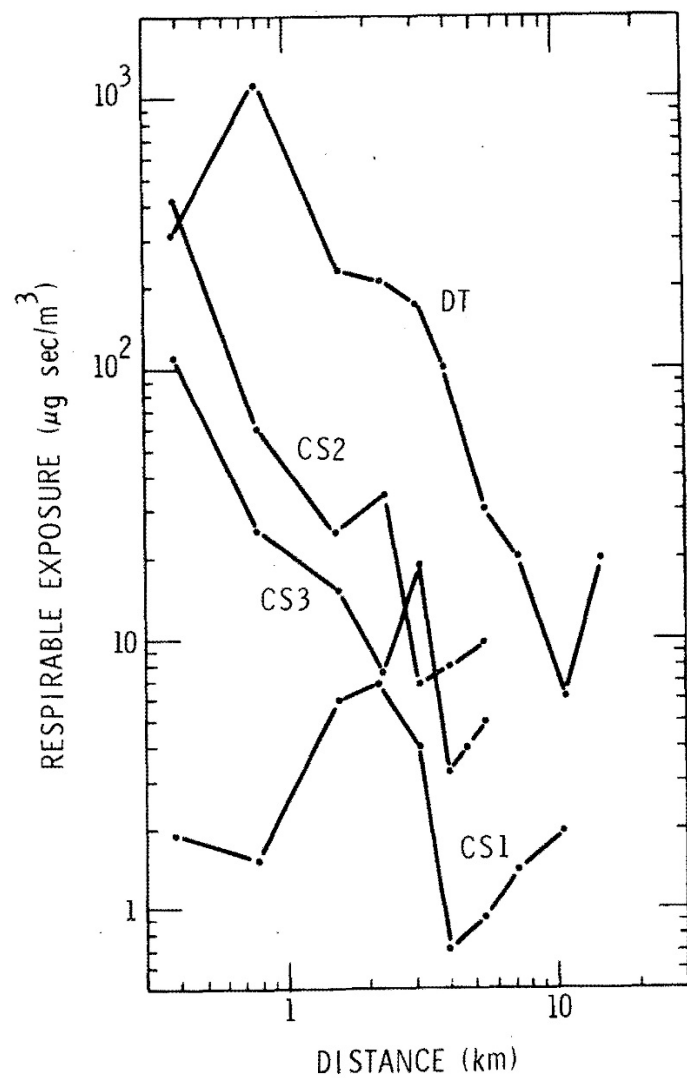


Figure C-5. Peak Respirable Exposure Contours, $\mu\text{g sec m}^{-3}$, vs. Distance from Ground Zero for Roller Coaster Tests [Figure 15 from Church *et al.* 1970].

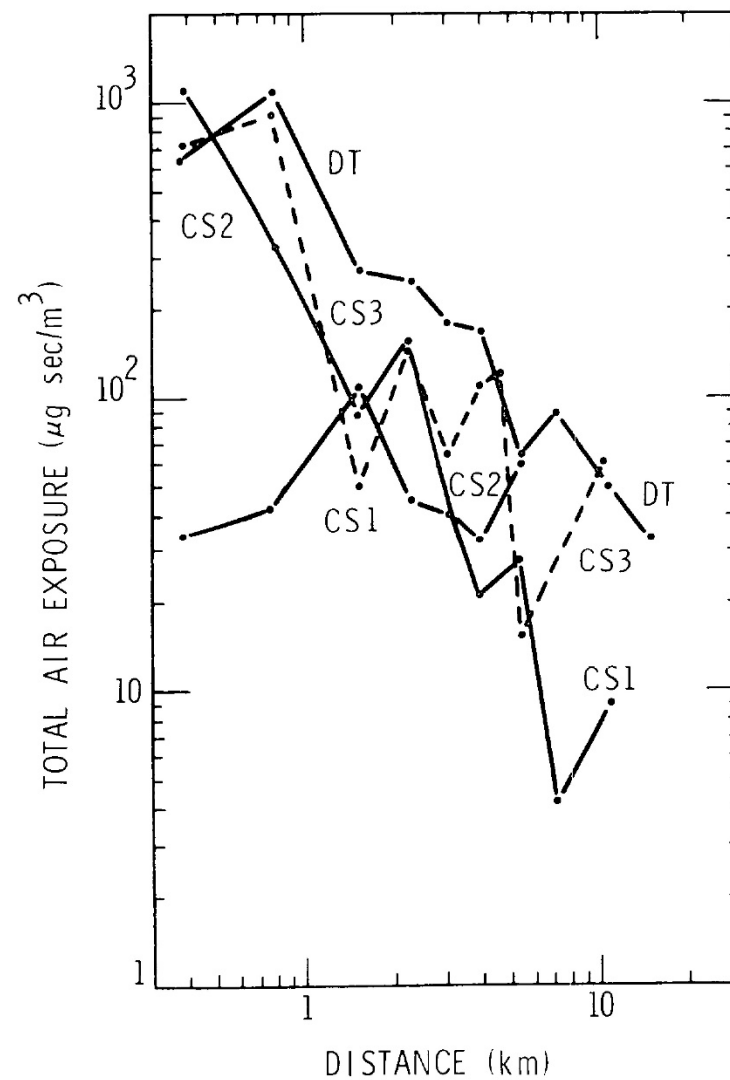


Figure C-6. Peak Total Air Exposure Contours, $\mu\text{g sec m}^{-3}$, vs. Distance from Ground Zero for Roller Coaster Tests [Figure 16 from Church *et al.* 1970].

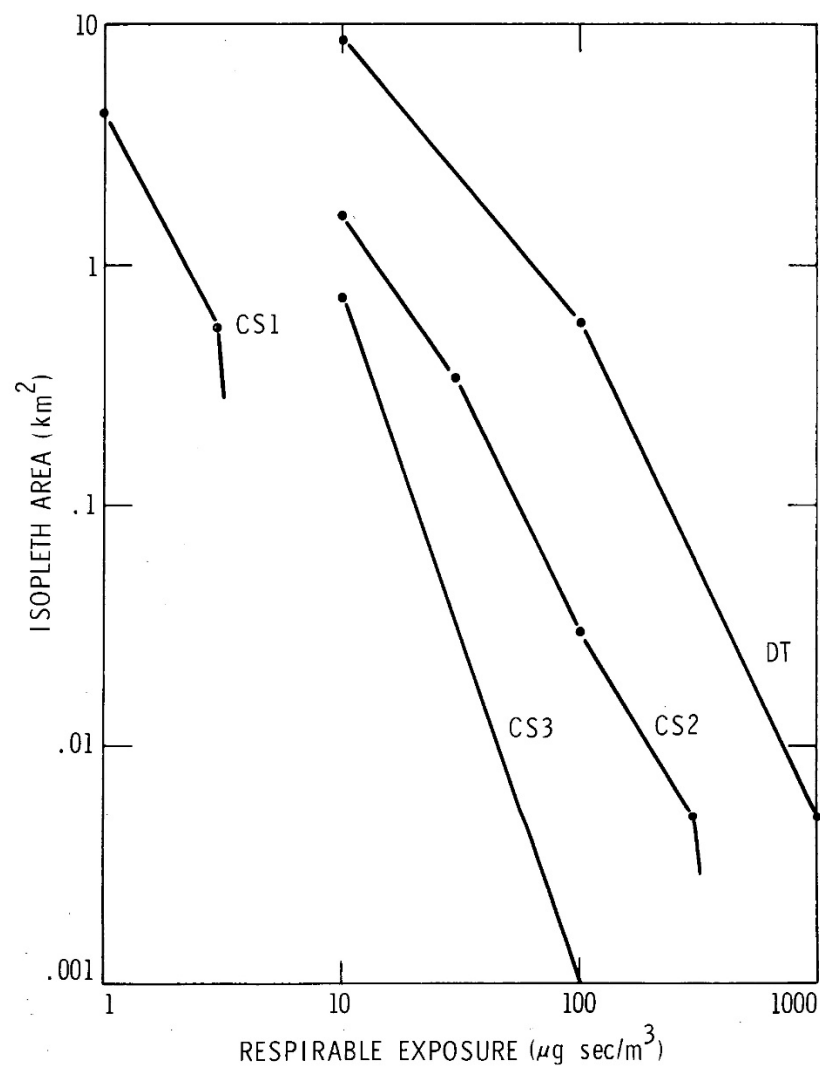


Figure C-7. Isoleth Areas, km², vs. Respirable Exposure, μg sec m⁻³, for Roller Coaster Tests [Figure 17 from Church *et al.* 1970].

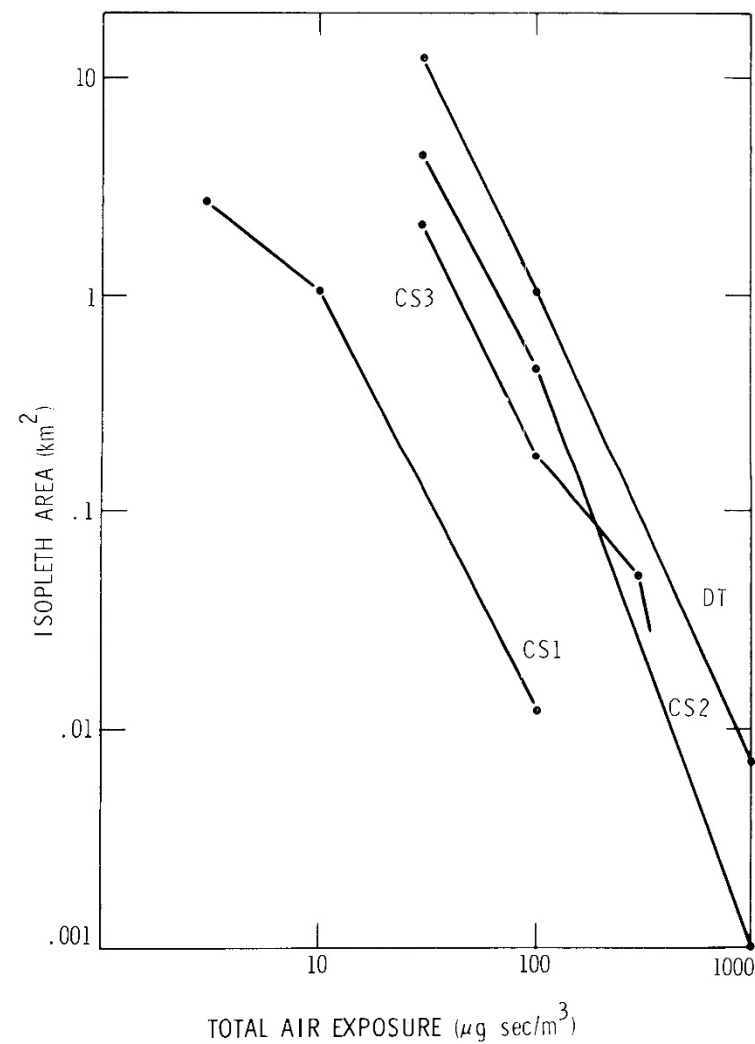


Figure C-8. Isoleth Areas, km², vs. Total Air Exposure, μg sec m⁻³, for Roller Coaster Tests [Figure 18 from Church *et al.* 1970].

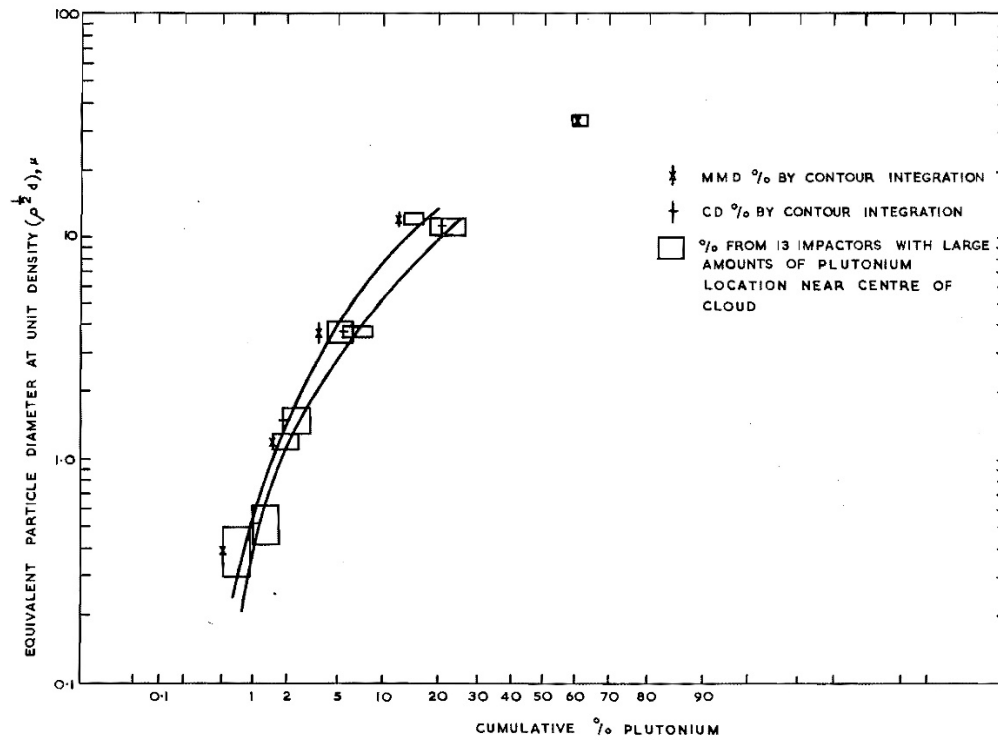


Figure C-9. Double Tracks Particle Size Distribution from Casella Impactors [from Friend and Thomas (1965)].

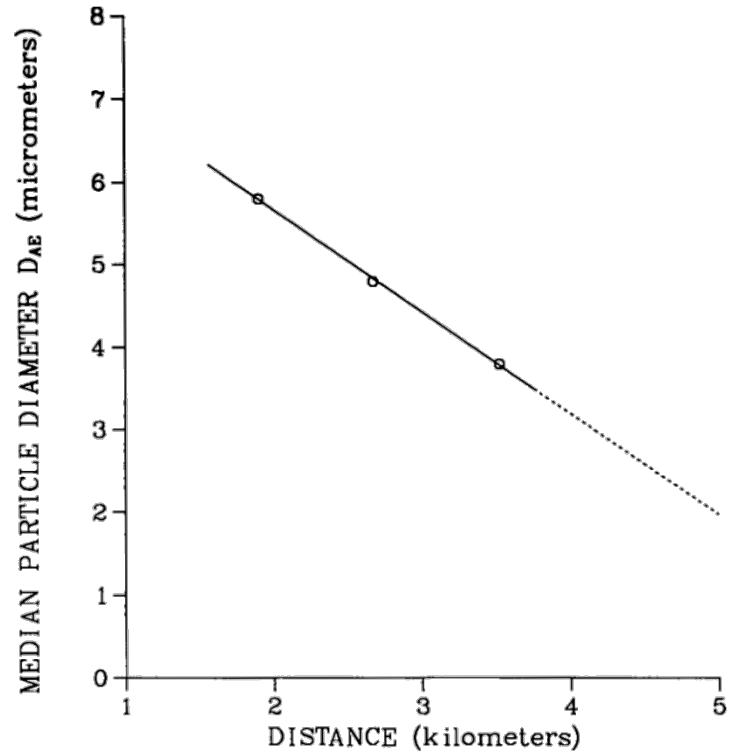


Figure C-10. Variation of Median Particle Diameter with Distance for Operation Roller Coaster Test Detonations (Dewart *et al.* 1982).

Appendix D

Estimated Airborne Concentrations and Inhalation Intakes based on Surface Soil Contamination Levels at Palomares

The resuspension models displayed in Figures A-7 and A-8 are useful in predicting airborne concentrations of surface-deposited contaminants over time. As discussed earlier, the Langham model was developed based on air sampling conducted shortly after plutonium safety detonation tests at the Nevada Test Site prior to the Operation Roller Coaster tests. For this model, resuspension is initially lower than the central value of the Maxwell and Anspaugh (2011) model. This is logical since the Maxwell and Anspaugh model was developed for applicability to a wide variety of circumstances, e.g., plutonium scatter from conventional explosives detonation mechanisms, the scattering of material from nuclear detonations, and others. It was noted earlier, that based on Operation Roller Coaster tests, locations closer to the point of detonation are expected to have a distribution of particles aerodynamically larger than for the contaminants deposited progressively at greater distances. For the plots provided in this Appendix, the Maxwell and Anspaugh (2011) will be used. A key point to its use is the understanding that resuspension rates are likely to be lower than predicted by the model for distances close to the detonation point. Other important points, discussed earlier are also important factors:

- mechanical disturbances will enhance resuspension rates and
- the mitigation methods of water spray and saturation of soils prior to mechanical disturbance will reduce resuspension rates.

Figure D-1 contains a plot of predicted airborne concentration for the three highest ground contamination concentrations contour boundaries from Figure A-6. Some areas had ground contamination levels greater than $32 \mu\text{Ci m}^{-2}$, though this represented less than 1% of the contaminated area. The area with contamination levels between 3.2 and $32 \mu\text{Ci m}^{-2}$ was 7.5% of the total. The areas encompassed within each zone is an important factor for airborne concentration of contaminants. Work within the zone with the greatest contamination levels was limited during the initial response to primarily survey personnel and EOD. EOD undoubtedly spent the greatest amount of time within this zone in the early stages of the recovery. Soil scraping operations were initiated later. The air sampling results in Table C-1 had only a couple of samples with concentrations slightly above 150 pCi m^{-3} . Since air sampling was directed at activities with the greatest expected potential for airborne concentrations, this finding is likely due to a few factors:

- very little work was conducted within the most highly contaminated zones in the initial stages of the recovery (except EOD, whom used air-purifying respirators),
- when air sampling was conducted during operations in more highly contaminated zones that afforded mechanical disturbances, mitigation was effective, and
- ground contamination may be less than predicted by the model.

The search for the lost weapon was a high priority for a large number of personnel. In addition to searches outside the contamination zones, these were also conducted within the contamination zone delineated in Figure A-6. Nevertheless, over 90% of the contaminated areas had relatively lower surface soil contamination levels, with subsequently low predicted resuspension.

Figure D-2 contains a plot of predicted daily inhalation intakes for the three ground contamination contours used in Figure D-1. For the plot, an inhalation rate of $1.5 \text{ m}^3 \text{ h}^{-1}$ is assumed, with an exposure duration of four hours. The exposure duration of four hours was used to infer that individuals only worked four hours a day, rather it was used because time spent in the more highly contaminated zones was limited during periods when certain tasks were accomplished. For example,

during soil scraping and loading, teams were rotated during the course of a work day. A key aspect of the plot is the daily inhalation intake on day 1. A four hour work duration in an area with contamination equal to $32 \mu\text{Ci m}^{-2}$ provides a predicted intake of 1.8 nCi, though 10 days later, the predicted intake is one-half as high. This plot clearly demonstrates the supposition provided earlier in the report that the greatest potential intakes were likely for work conducted very early in the recovery. At about 5 weeks into the recovery, the predicted inhalation intakes were about 10-fold lower than that predicted for day 1. Overall, the plot of projected intakes support the urine sampling analysis that only a small fraction (5% or less) of the recovery workers had inhalation intakes above 34 nCi $^{239+240}\text{Pu}$.

The plots within Figures D-3 through D-6 provide the same predicted daily inhalation intakes of Figure D-2, but with the integrated intake for individual work weeks. During the recovery, on-site work was accomplished six days a week. These plots are useful for reviewing estimated intakes for recovery workers, based on their period of presence at Palomares. As noted earlier, with exceptions, most workers were present for three week periods. In the case of Figure D-3, applicable to the $32 \mu\text{Ci m}^{-2}$ surface contamination, the sum of predicted inhalation intakes for the first three weeks is 18 nCi, while for the fourth thru six being 4 nCi, about 4.5-fold lower.

Figure D-6 provides estimates for the lowest contamination contour $0.32 \mu\text{Ci m}^{-2}$ and is useful for estimation of inhalation intakes for an individual that was present, yet did not have duties in the more highly contaminated zones. For a case of this type, it is reasonable to multiply the daily intake by a factor of six to cover a 24-hour period. The inhalation intake for the first three weeks, using this approach is 1.1 nCi. This value is equivalent to the lower intake value recommended for recovery workers that did not perform on-site work (see § 8.3).

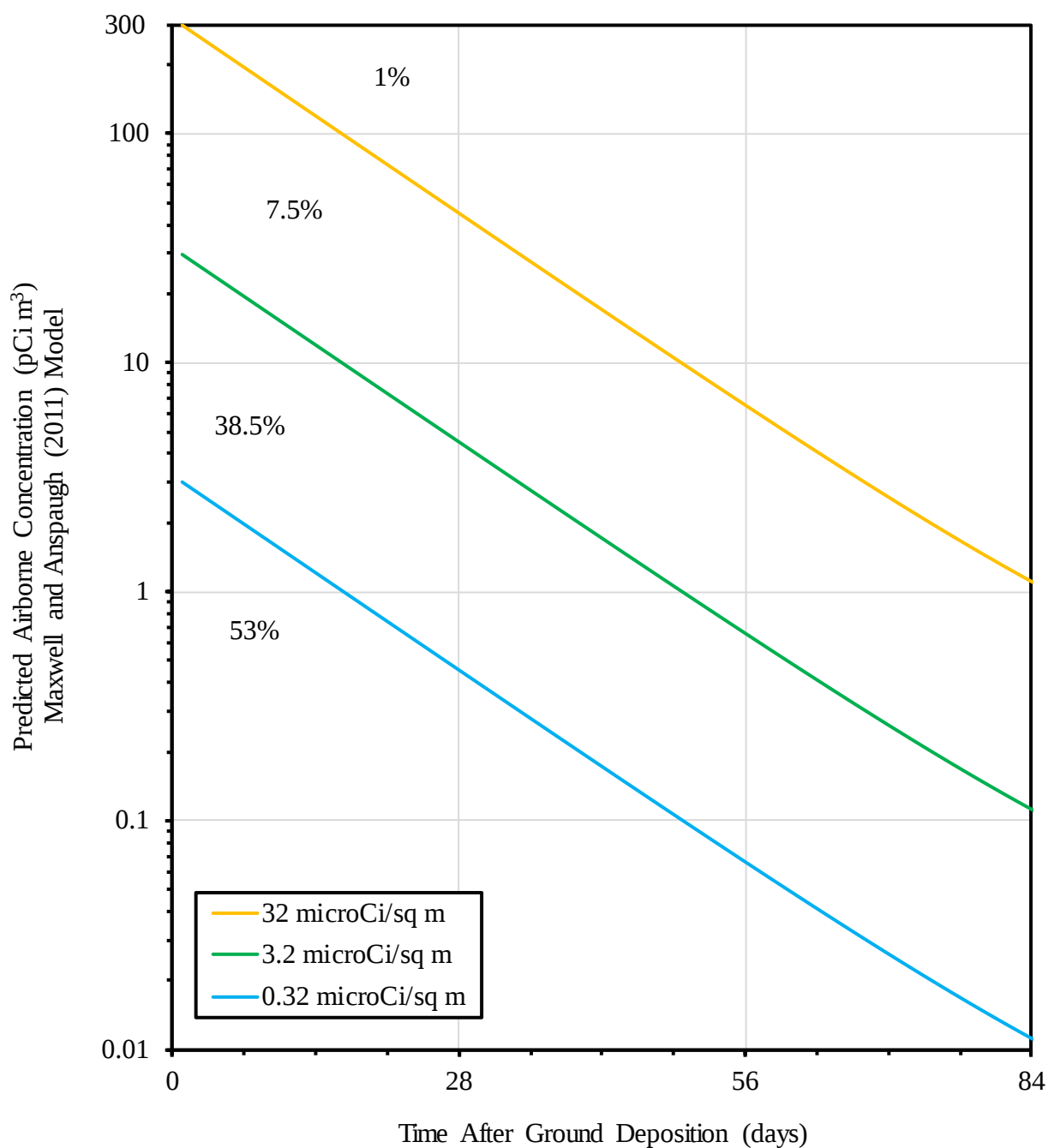


Figure D-1. Predicted Airborne Concentration for Various Times After Ground Deposition of Plutonium Contamination, Based on the Three Highest Ground Contamination Concentration Contour Boundaries of Figure A-6 and the Maxwell and Anspaugh (2011) Resuspension Model [Percent Values Annotated Represent Contribution to Total Contaminated Area from Figure A-6].

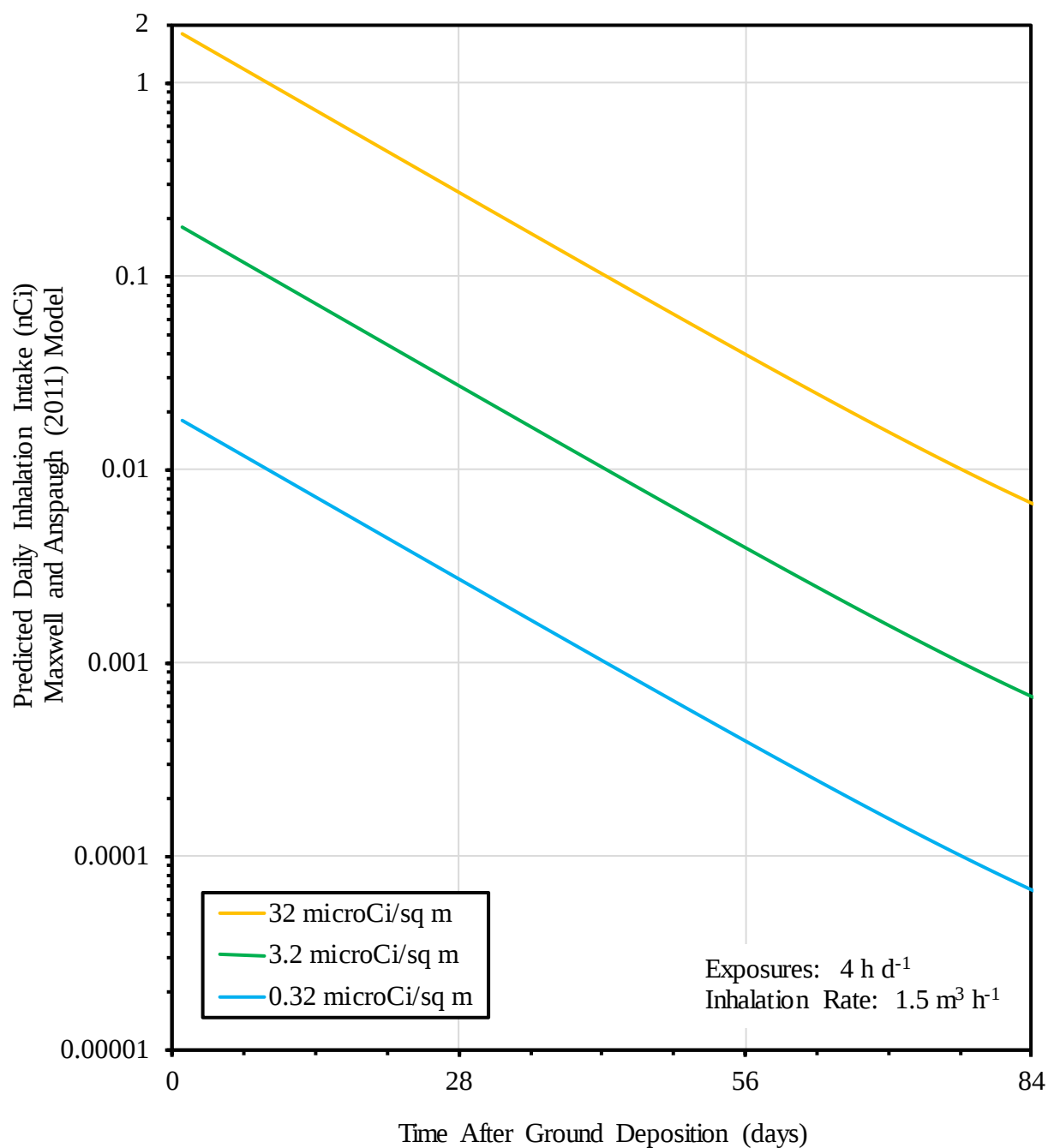


Figure D-2. Predicted Daily Inhalation Intakes for Various Ground Surface Contamination Levels with Resuspension Predicted by Maxwell and Anspaugh (2011) Model, Inhalation Rate of 1.5 m³ h⁻¹ and an Exposure Duration of 4 h d⁻¹.

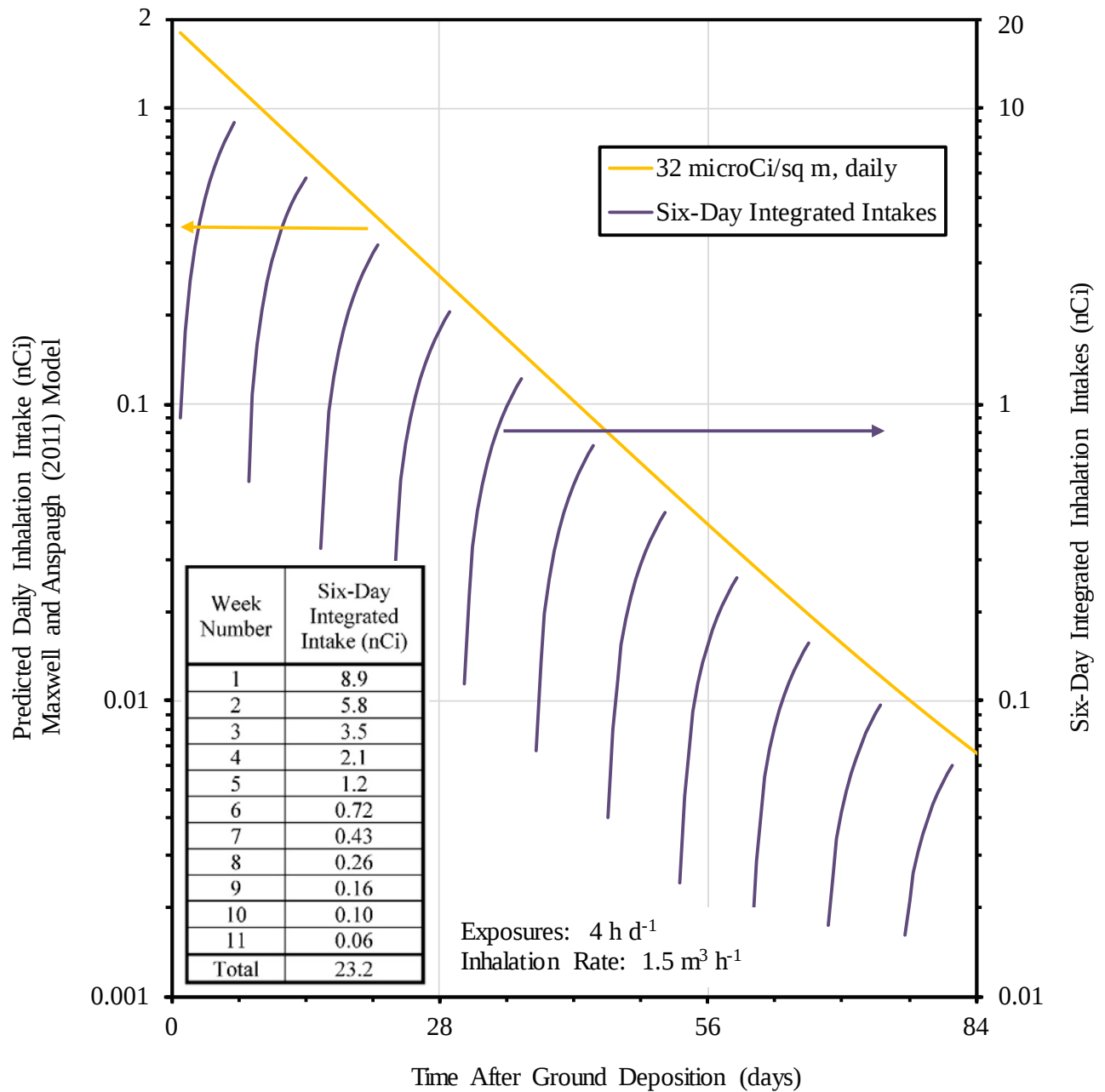


Figure D-3. Predicted Daily Inhalation Intakes for 32 $\mu\text{Ci m}^{-2}$ Surface Contamination with Resuspension Predicted by Maxwell and Anspaugh (2011) Model, Inhalation Rate of 1.5 m³ h⁻¹ and an Exposure Duration of 4 h d⁻¹. Integrated Intakes for Individual Six-Day Work Periods.

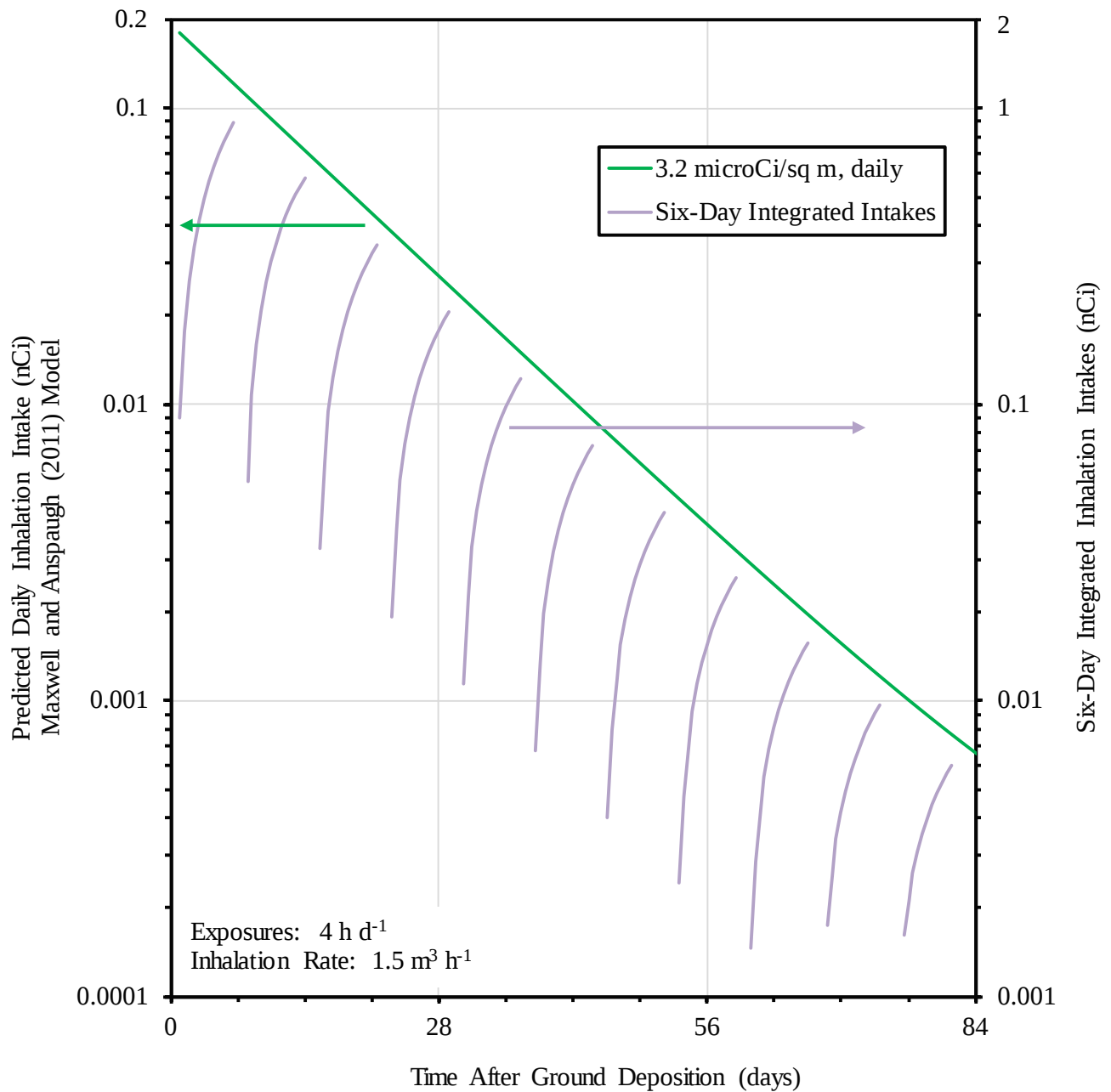


Figure D-4. Predicted Daily Inhalation Intakes for 3.2 $\mu\text{Ci m}^{-2}$ Surface Contamination with Resuspension Predicted by Maxwell and Anspaugh (2011) Model, Inhalation Rate of 1.5 $\text{m}^3 \text{h}^{-1}$ and an Exposure Duration of 4 h d^{-1} . Integrated Intakes for Individual Six-Day Work Periods.

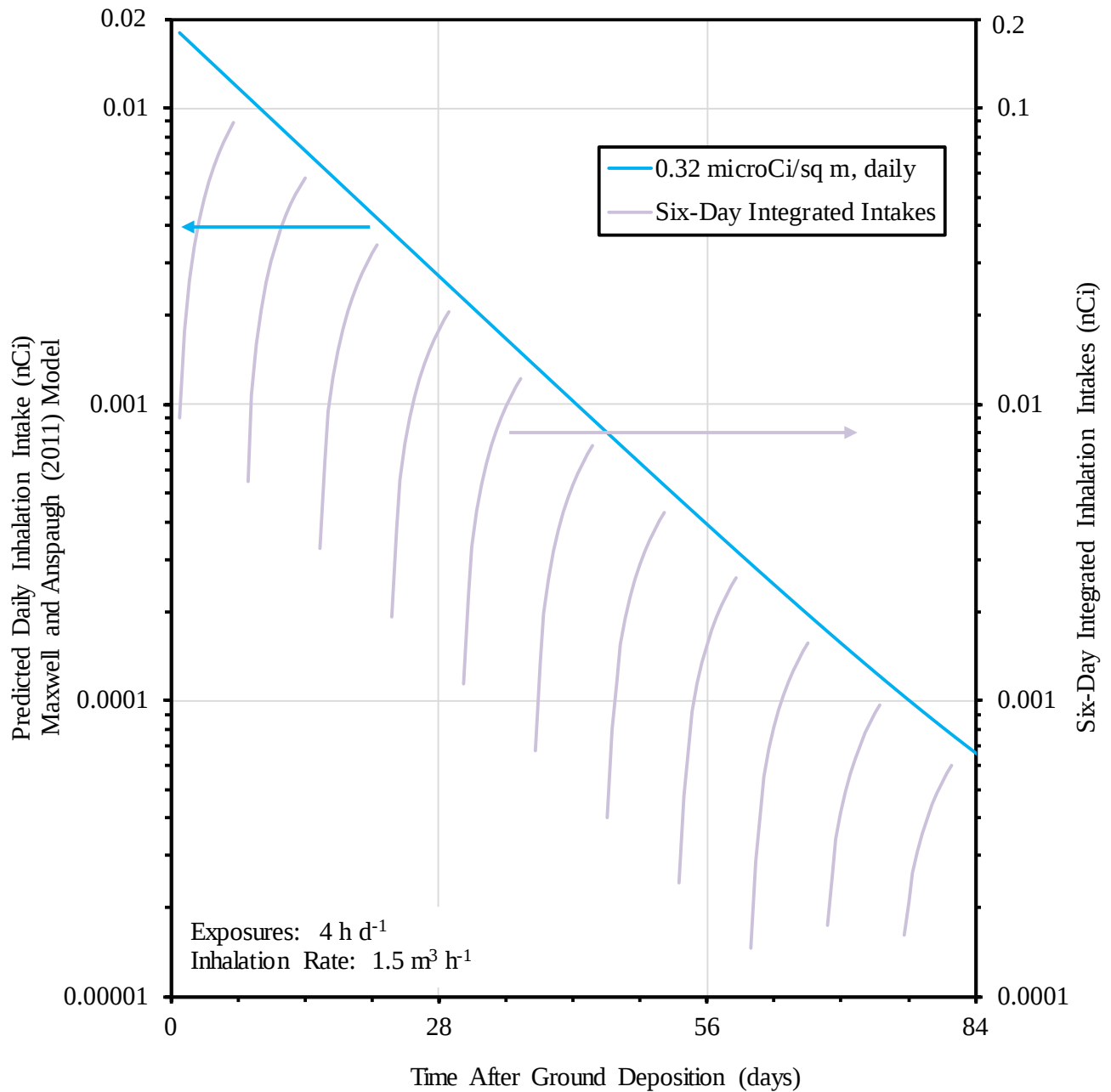


Figure D-5. Predicted Daily Inhalation Intakes for $0.32 \mu\text{Ci m}^{-2}$ Surface Contamination with Resuspension Predicted by Maxwell and Anspaugh (2011) Model, Inhalation Rate of $1.5 \text{ m}^3 \text{ h}^{-1}$ and an Exposure Duration of 4 h d^{-1} . Integrated Intakes for Individual Six-Day Work Periods.

Appendix E

IREP Version 5.8.2 Example Calculations

TABLE E-1. Example IREP Version 5.8.2 PC Calculations for α -Particle Radiation, Year of Exposure – 1966, Year of Birth – 1946, 10% Standard Deviation in Dose (Normal Distribution).

Factor	Example Cases								
	1	2	3	4	5	6	7	8	9
Diagnosis Year	2008	2008	2018	1998	1988	1978	2008	2008	2008
Latency (years)	42	42	52	32	22	12	42	42	42
Dose (rem)	285	20.8	20.8	20.8	18	10.8	30.8	33.5	34
Cancer Site	lung	lung	lung	lung	lung	lung	lung	lung	lung
Smoking	never	never	never	never	never	never	former	10-19 cig/d	20-39 cig/d
CL (1 %)	4.3%	0.32%	0.32%	0.32%	0.65%	0.57%	0.55%	0.48%	0.44%
CL (5 %)	11.4%	0.93%	0.93%	0.93%	1.4%	1.2%	1.24%	1.19%	1.11%
CL (50 %)	50%	6.8%	6.8%	6.8%	7.4%	7.0%	7.1%	7.1%	7.2%
CL (95 %)	86.6%	32.0%	32.0%	32.0%	31.2%	30.3%	31.4%	32.5%	32.6%
CL (99 %)	93.2%	50 %	50 %	50 %	50 %	50 %	50 %	50 %	50 %

Factor	Example Cases								
	10	11	12	13	14	15	16	17	18
Diagnosis Year	2008	2008	2018	1998	1988	2008	2008	2016	2016
Latency (years)	42	42	52	32	22	42	42	50	50
Dose (rem)	92	4.9	4.9	4.9	4.9	277	24.9	1030	59.3
Cancer Site	liver	liver	liver	liver	liver	bone	bone	nervous	nervous
Smoking	NA	NA	NA	NA	NA	NA	NA	NA	NA
CL (1 %)	4.40%	0.24%	0.24%	0.24%	0.24%	6.12%	0.58%	0.00%	0%
CL (5 %)	9.9%	0.6%	0.6%	0.6%	0.6%	13.8%	1.4%	5.1%	0%
CL (50 %)	50%	5.1%	5.1%	5.1%	5.1%	50%	8.3%	50.0%	5.5%
CL (95 %)	87.9%	27.9%	27.9%	27.9%	27.9%	85.6%	34.9%	88.8%	31.3%
CL (99 %)	94.9%	50%	50%	50%	50%	91.8%	50%	94.5%	50.0%

CL = credibility level

TABLE E-1. Example IREP Version 5.8.2 PC Calculations for α -Particle Radiation, Year of Exposure – 1966, Year of Birth – 1946, 10% Standard Deviation in Dose (Normal Distribution), continued.

Factor	Example Cases									
	19	20	21	22	23	24	25	26	27	28
Diagnosis Year	2016	2016	2016	2016	2016	2016	2016	2006	1996	1986
Latency (years)	50	50	40	30	20	50	50	40	30	20
Dose (rem)	320	27.4	27.4	27.4	18.8	236	22.2	22.2	22.2	15.5
Cancer Site	urine bladder	urine bladder	urine bladder	urine bladder	urine bladder	kidney	kidney	kidney	kidney	kidney
Smoking	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
CL (1 %)	4.89%	0.44%	0.44%	0.44%	0.44	7.0%	0.70%	0.70%	0.70%	0.70%
CL (5 %)	12.0%	1.16%	1.16%	1.16%	1.2%	14.2%	1.5%	1.5%	1.5%	1.6%
CL (50 %)	50.0%	7.9%	7.9%	7.9%	7.8%	50.0%	8.6%	8.6%	8.6%	8.6%
CL (95 %)	86.3%	35.0%	35.0%	35.0%	34.7%	85.0%	34.7%	34.7%	34.7%	34.7%
CL (99 %)	92.1%	50.0%	50.0%	50.0%	50.0%	91.4%	50.0%	50.0%	50.0%	50.0%

Factor	Example Cases	
	29	30
Diagnosis Year	2016	2016
Latency (years)	50	50
Dose (rem)	2050	53.5
Cancer Site	CLL	CLL
Smoking	NA	NA
CL (1 %)	0.0%	0.0%
CL (5 %)	0.0%	0.0%
CL (50 %)	50.0%	2.5%
CL (95 %)	93.9%	28.5%
CL (99 %)	97.5%	50.0%

CL = credibility level

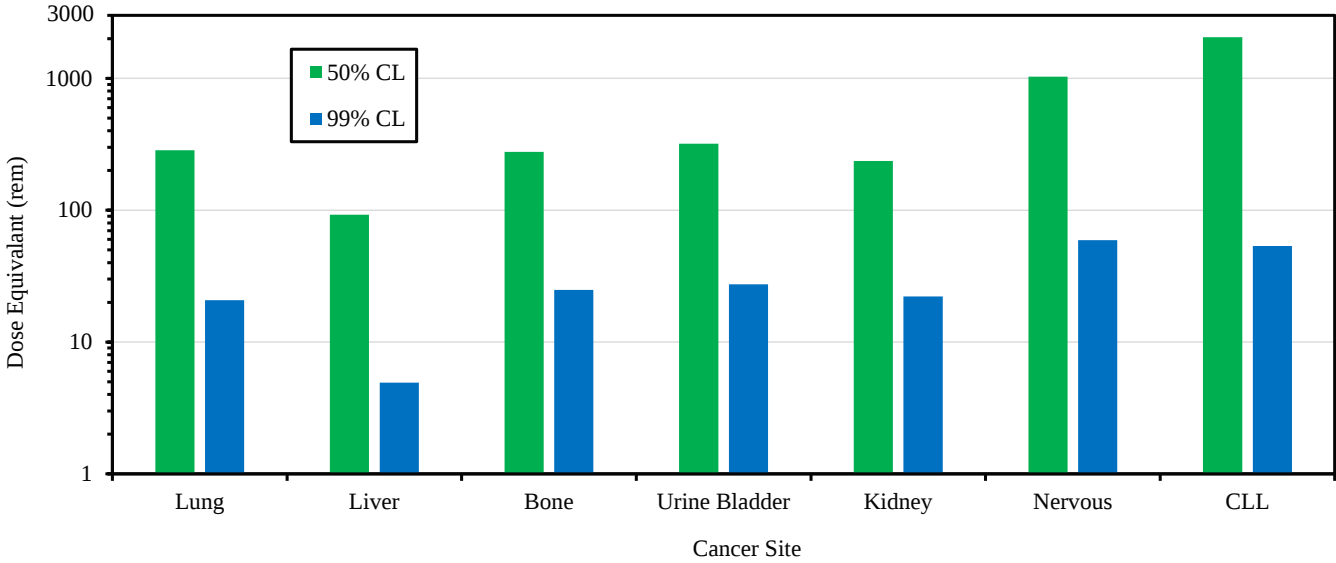


Figure E-1. Histogram of Dose Equivalent Values for Various Cancer Sites at the 50 and 99% Credibility Levels, 50-y Latency.

Appendix F

Urine Bioassay Information

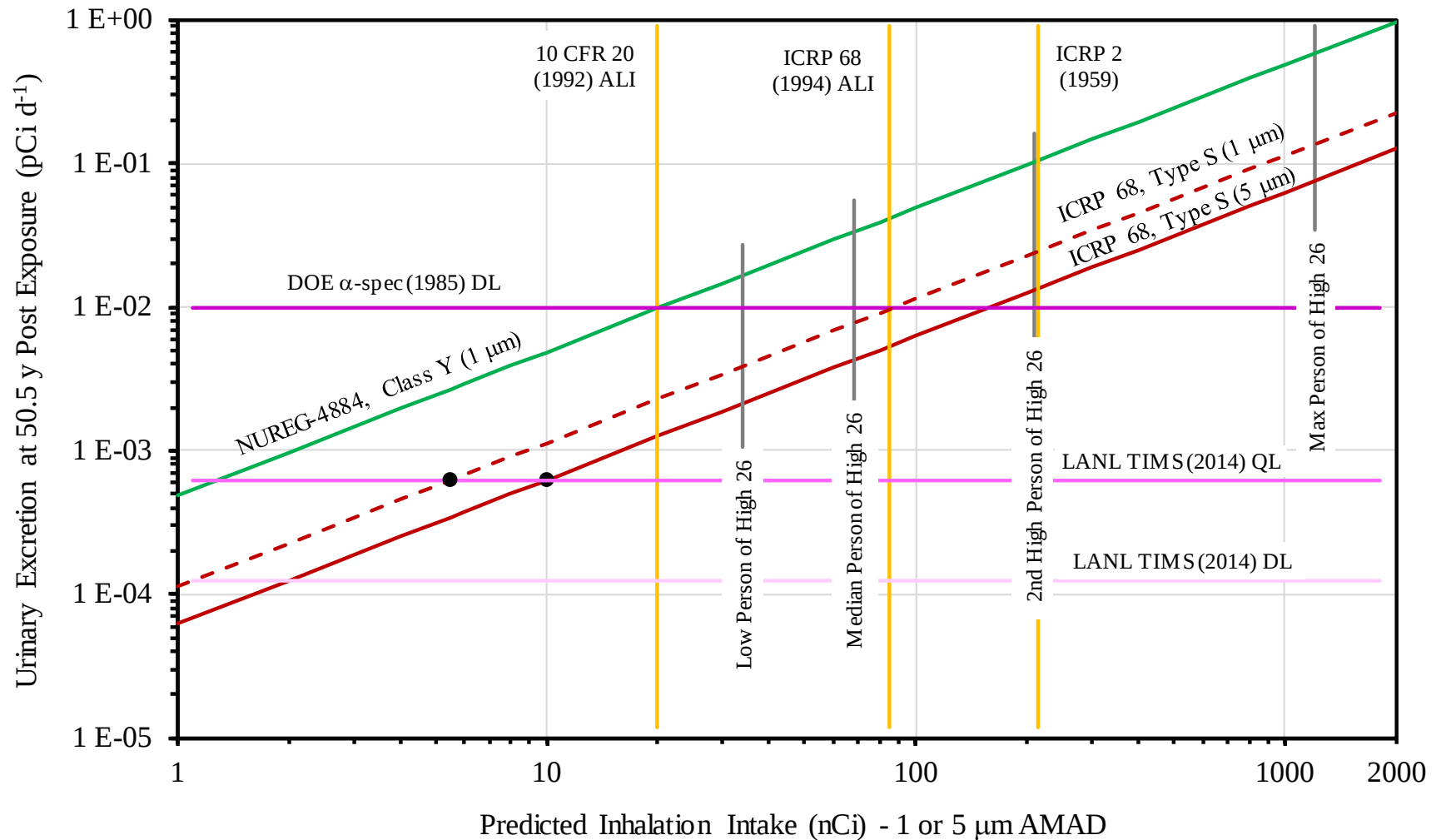


Figure F-1. Plot Illustrating the Predicted Daily Urine Excretions for an Acute Intake of PuO_2 at 50.5 y Post Intake, with Annotation of Sensitivity of LANL Thermal Ionization Mass Spectrometry (2014) and α -Particle Spectrometry (1985), for Both ICRP Report 30, Inhalation Class Y (NUREG-4884), $1 \mu\text{m}$ AMAD, and ICRP Report 68, Inhalation Class S, 1 and $5 \mu\text{m}$ AMAD. Annual Inhalation Exposure Intakes Overlaid for ICRP Report 2, Insoluble (1959), ICRP Report 26/30/48 [listed as 10 CFR 20 (1992)], and ICRP Report 60/68 (1994).

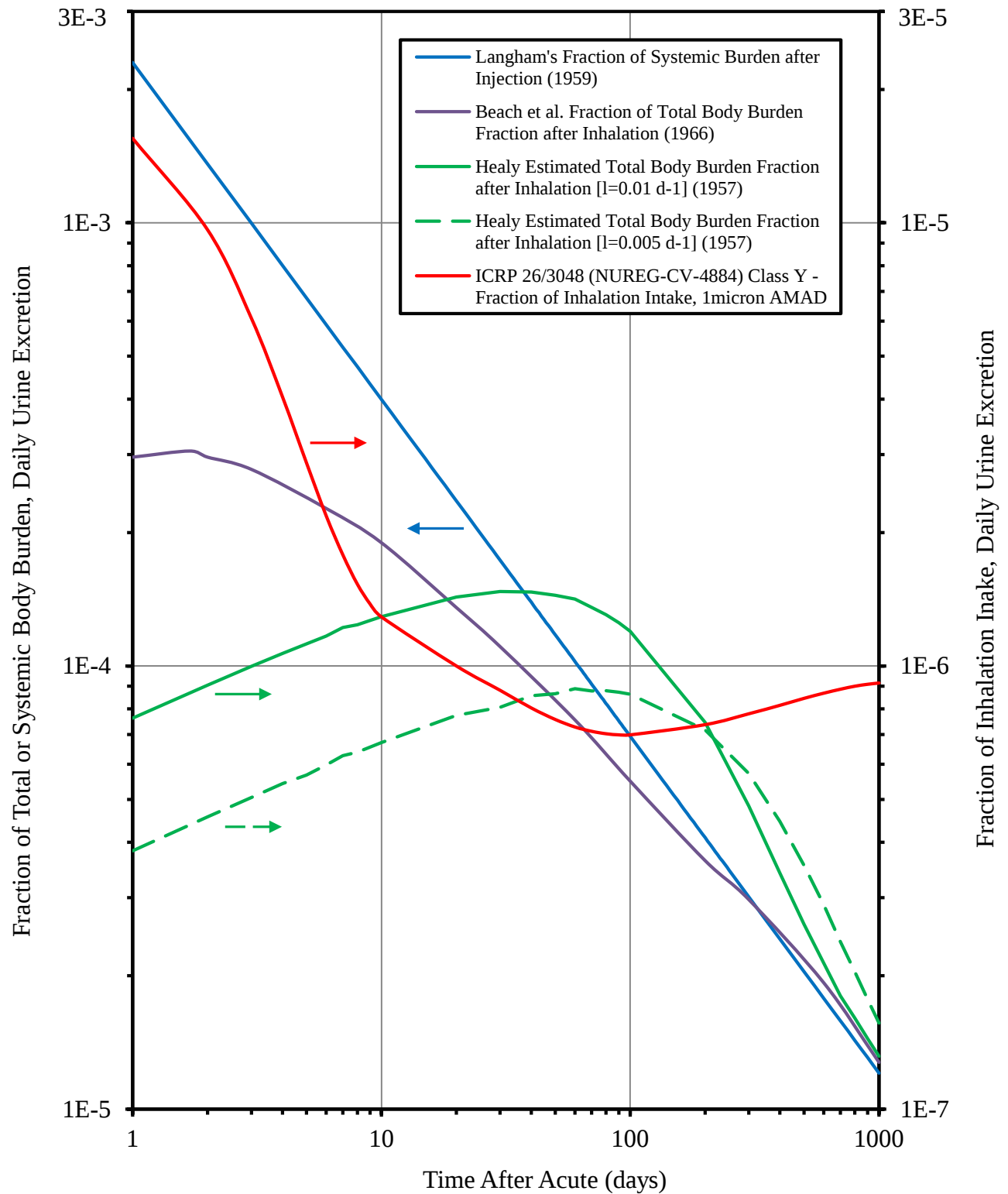


Figure F-2. Daily Urine Excretion Models after Acute Intakes, Langham (1959) from Human Injection Studies, Healy (1957) for Inhalation, Beach *et al.* (1966) for Inhalation.

Discussion Notes for Figure F-2. The plot contains estimated daily urine excretions from four early models. Important factors in comparison of the models are the denominators that fractional daily urine excretion estimates are made. The Langham model was discussed extensively earlier in the report, due to its application to the Palomares responders. This model related urine excretion to systemic body burdens. In contrast the Beach *et al.* and Healy models relate urine excretion to total body burden, while the ICRP 30/48 model is related to the inhaled activity. The Beach *et al.* model was based on the Langham model, modified by clearance of plutonium from the lung to the blood stream. Clear from the modification is provision for early (fast) and slower clearance. The NUREG/CR-4884 Model (NRC 1987) was produced for ICRP Report 26/30/48. The complete set of metabolism plots from NUREG/CR-4884 are contained in Figures F-3 and F-4. In comparison of the Langham Model and ICRP 30/48 excretion models, both have a similar trend of decreasing excretion levels up to about 100 days.

Discussion Notes for Figures F-5. The complete set of metabolism plots from Potter (2002) are contained in Figure F-5 for an acute inhalation intake. They summary points: about 48% that is inhaled is eventually cleared to the GI tract (about 90% of the clearance occurs within about 30 d), about 18% of the inhaled activity is exhaled, retention in the thoracic lung decreases slowly over the first few years, and the daily urinary excretion rates are somewhat uniform after a month to a few years.

Discussion Notes for Figures F-6 through F-9. The plots contains 50-y CED (ICRP 60/68 and 103/130/141) or CDE (ICRP 30/48) for the RBM, BS, liver, and lung for daily urinary excretions, normalized to a pCi. Fractional urinary excretion values are from NUREG/CR-4884 [ICRP 30/48], Potter (2002) [ICRP 68, 5 μm AMAD], Thomas (2020) [ICRP 68, 1 μm AMAD], and Leggett (2020) [ICRP 141]. There are many key points to be made regarding the comparisons. First, the primary assessment of urine results in the resampling program were for samples collected about four months after the recovery up to a year. Over this period, the ICRP 30/48, 68, and 141 models are relatively flat, though there are subtle differences. For each organ, there are differences in estimated CED or CDE for the key organs. These values for RBE and BS are progressively lower for the use of ICRP 30/48 to 68, and 141. Therefore, use of the ICRP 30/48 DCF are more conservative for these organs, though the progressive updates made by ICRP were deemed more representative of dose to these tissues. For the liver, ICRP 68 estimates of CED are lower than the similar estimates from ICRP 30/48 and ICRP 141. The latter were similar for periods from 100 days to one year. Estimated 50-y committed doses were similar for ICRP 30/48 and ICRP 141, Type S, within the same period. For the ICRP Report 141 $^{239}\text{PuO}_2$ and mixed oxide lung Type , estimated 50-y committed dose to the lung would be about 10-fold higher than Type S. This category is considered a super Type S, having more pernicious retention than Type S. Upon consideration of measured and predicted airborne concentrations, Type S is deemed more representative.

Discussion Notes for Figure F-10. The plot contains 50-y CED (ICRP 60/68 and 103/130/141) or CDE (ICRP 30/48) for the RBM, BS, liver, and lung based on an inhalation of 1 nCi ^{239}Pu . The plot offers the perspective of doses from an inhalation standpoint, rather than from the urine bioassay perspective, as illustrated in Figures F-6 through F-9.

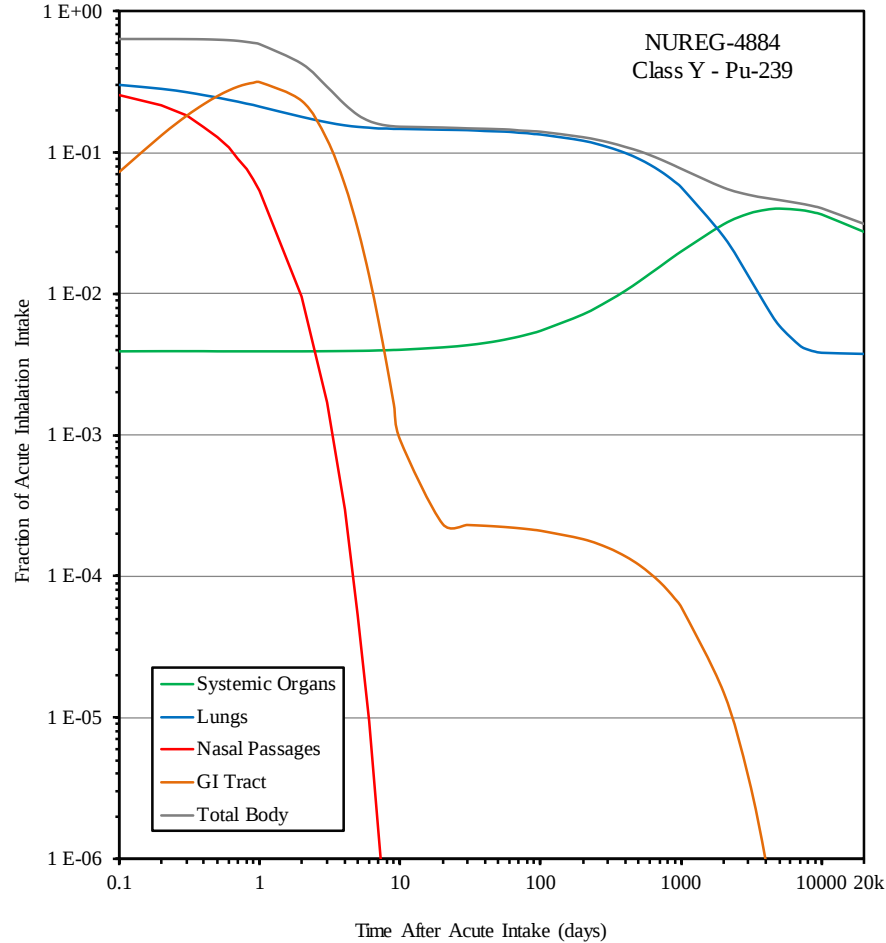


Figure F-3. ICRP 30/48 Metabolism for Class Y Inhalation, $1 \mu\text{m}$ AMAD (NUREG/CR-4884).

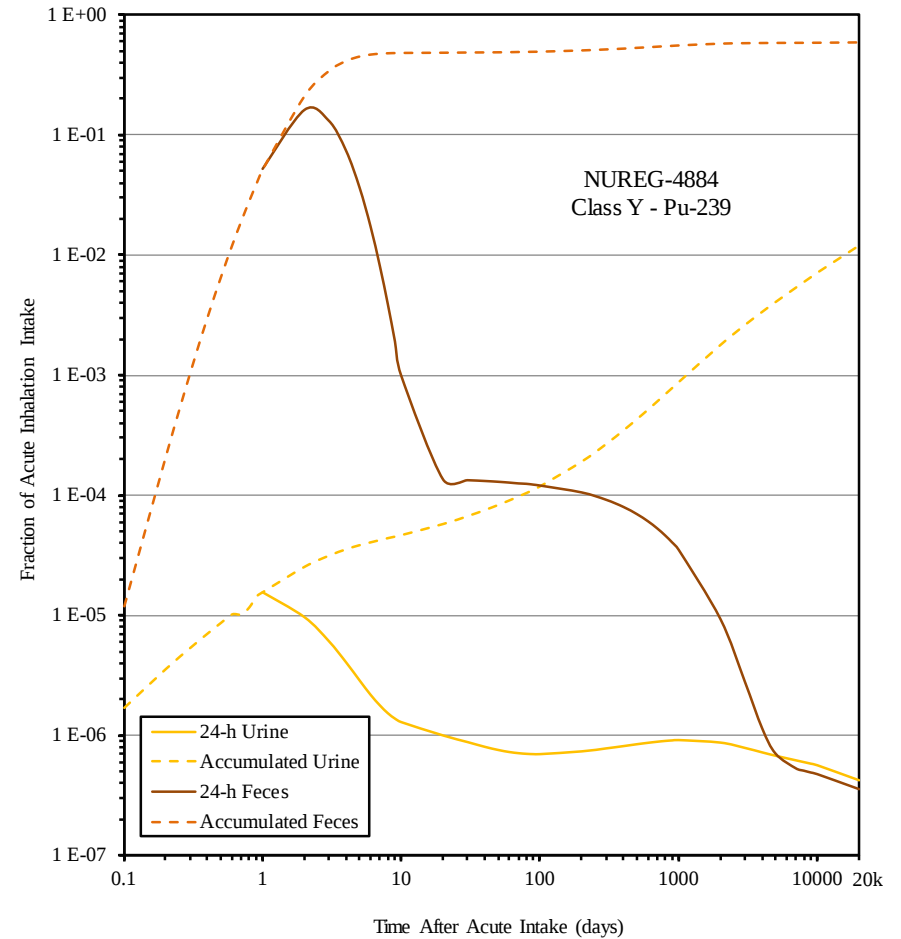


Figure F-4. ICRP 26/30/48 Excretion for Class Y Inhalation, $1 \mu\text{m}$ AMAD (NUREG/CR-4884).

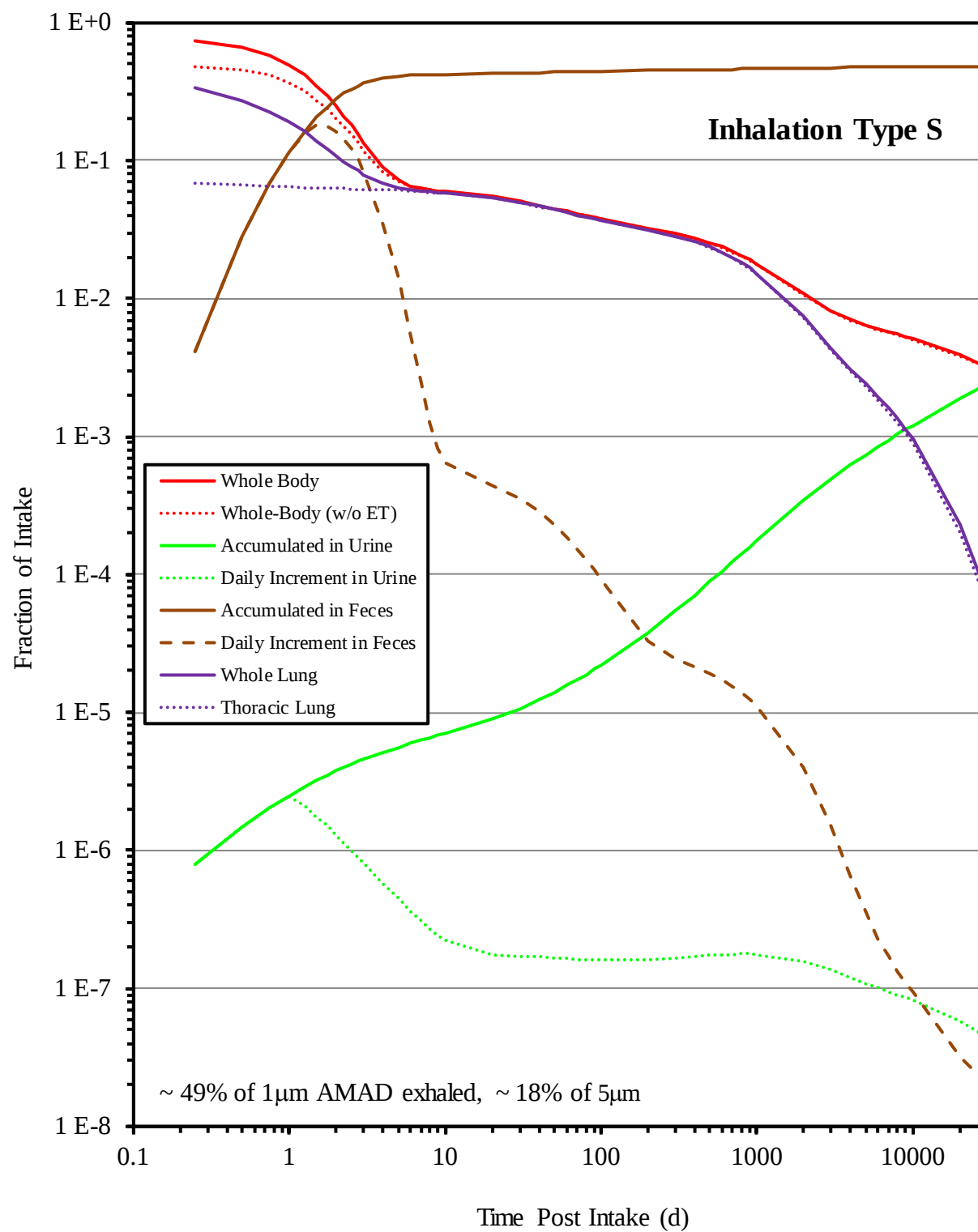


Figure F-5. ICRP 68 Metabolism for ICRP 66 Type S Inhalation, 5 μ m. [from Potter (2002)].

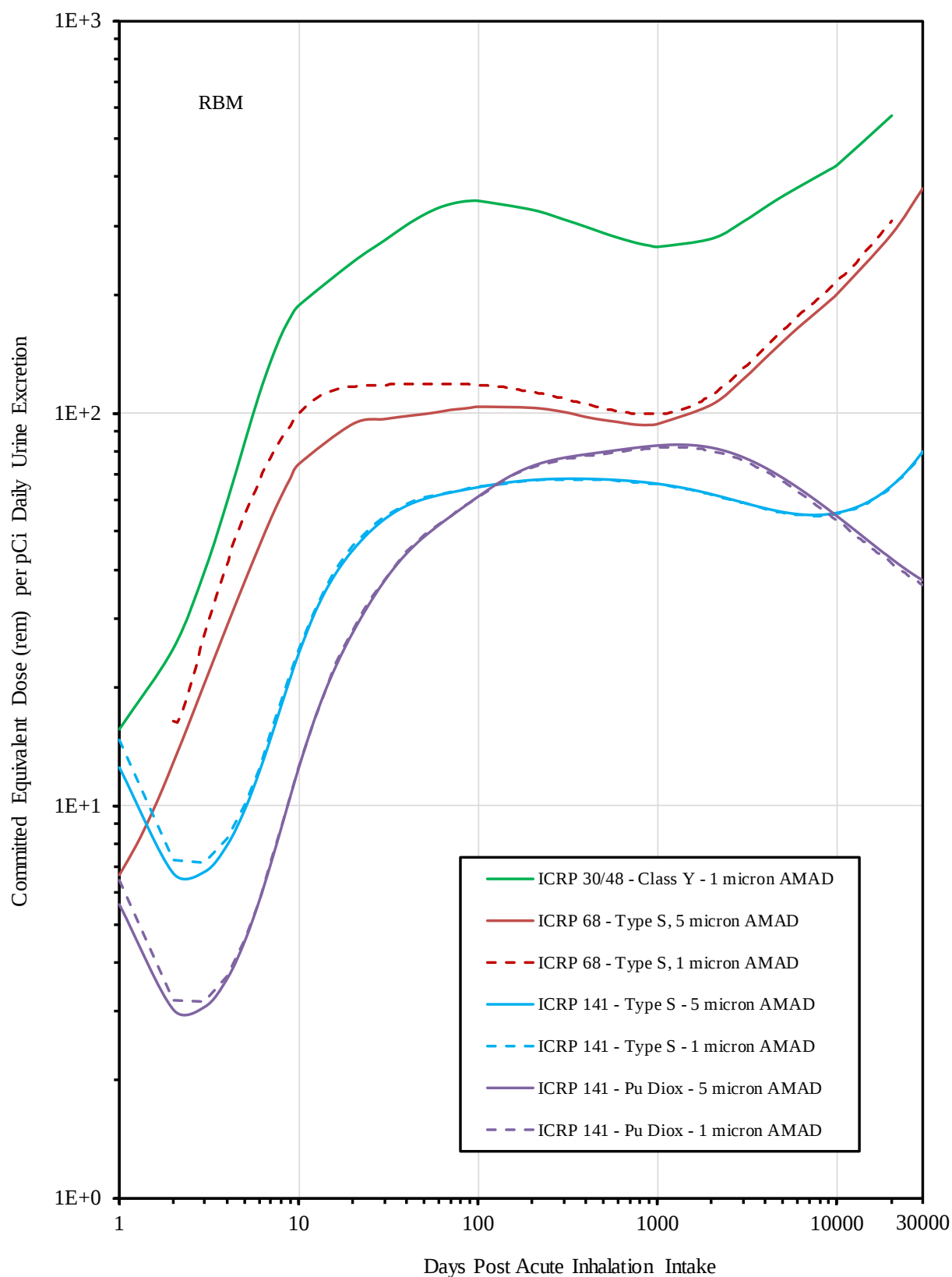


Figure F-6. Committed Equivalent Dose (or Dose Equivalent for ICRP 26) to RBM, Normalized to 1 pCi ^{239}Pu in Daily Urine Excretion for ICRP Reports 30/48, 68, and 141.

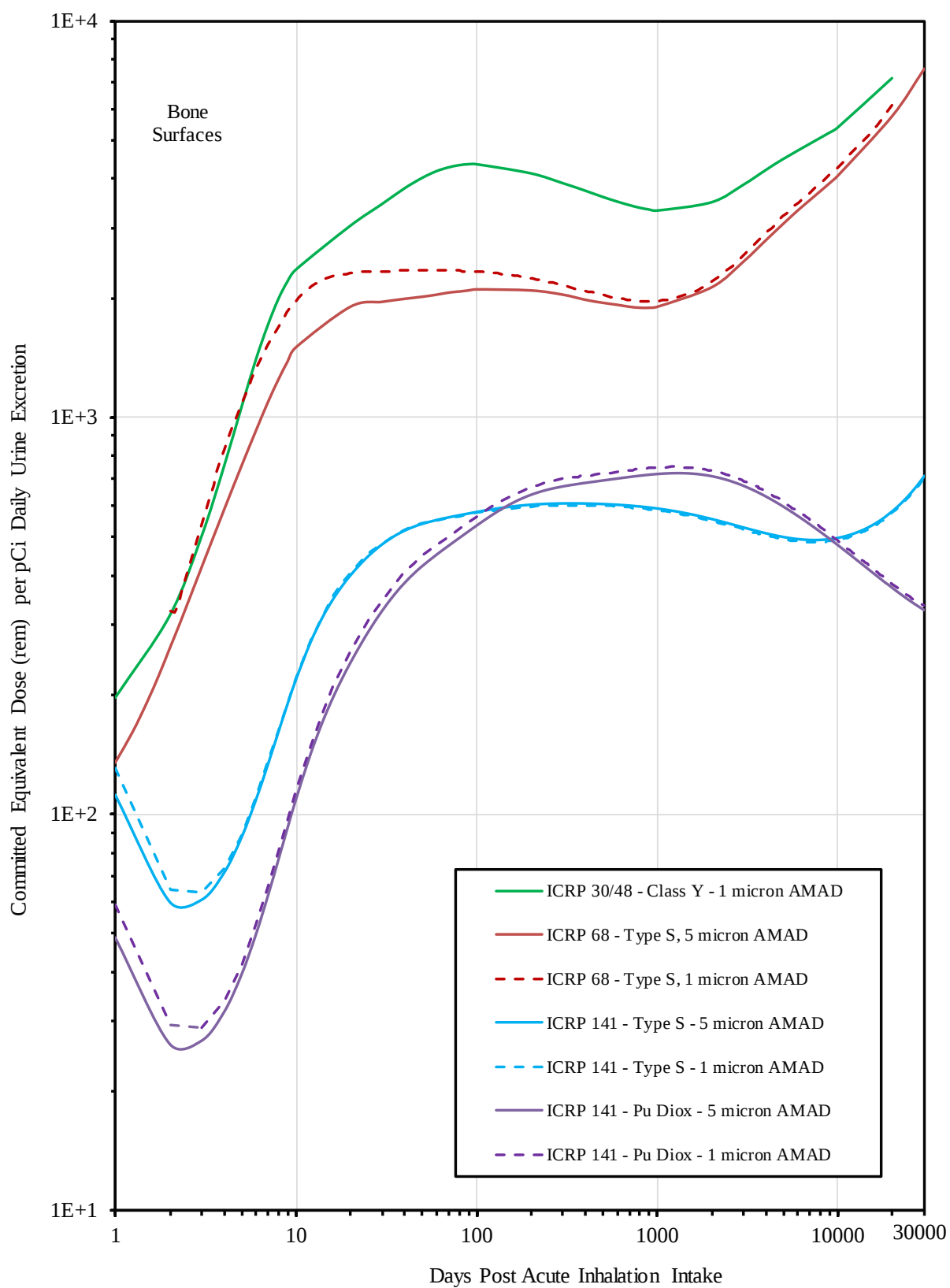


Figure F-7. Committed Equivalent Dose (or Dose Equivalent for ICRP 26) to BS, Normalized to 1 pCi ^{239}Pu in Daily Urine Excretion for ICRP Reports 30/48, 68, and 141.

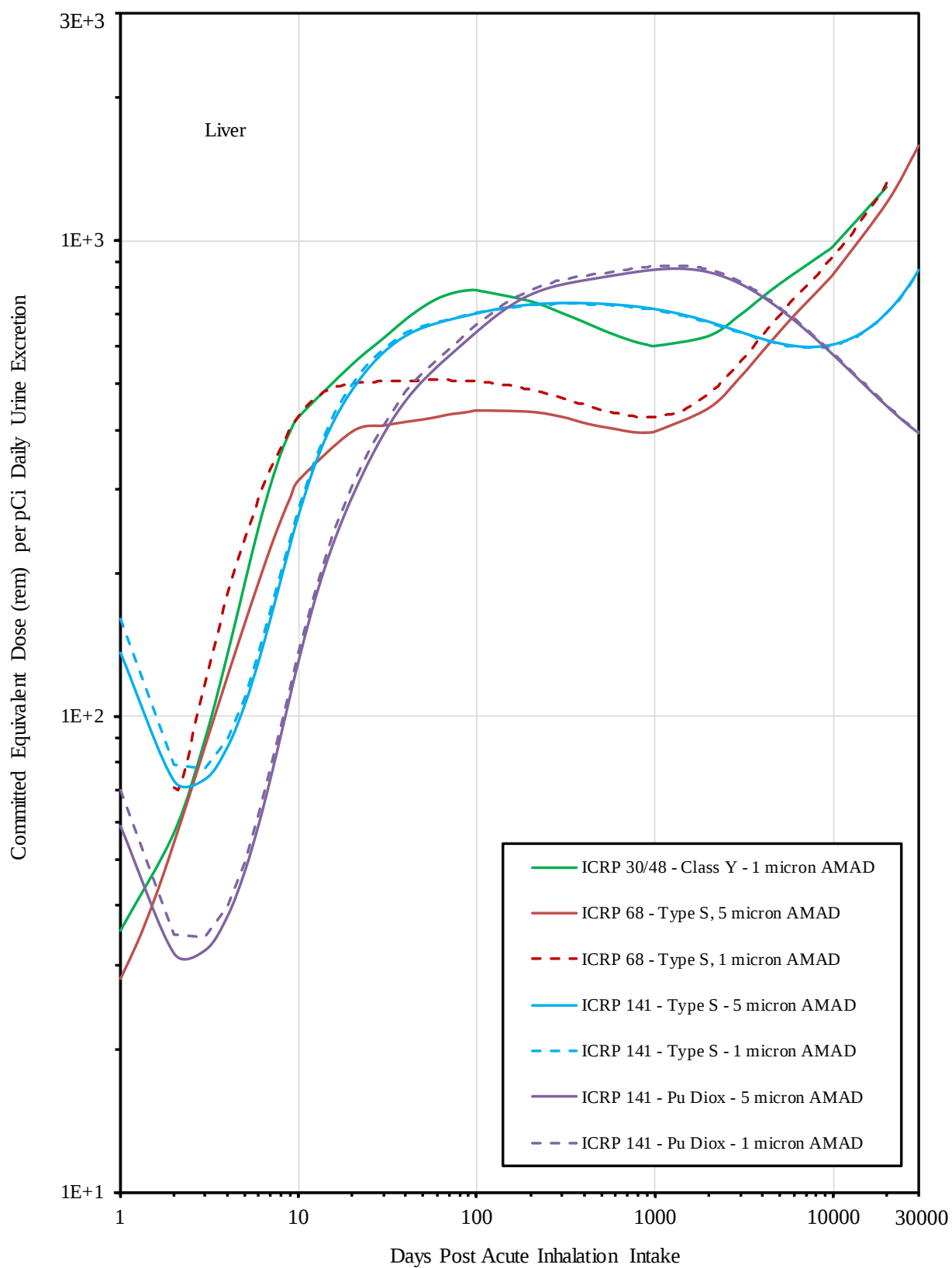


Figure F-8. Committed Equivalent Dose (or Dose Equivalent for ICRP 26) to Liver, Normalized to 1 pCi ^{239}Pu in Daily Urine Excretion for ICRP Reports 30/48, 68, and 141.

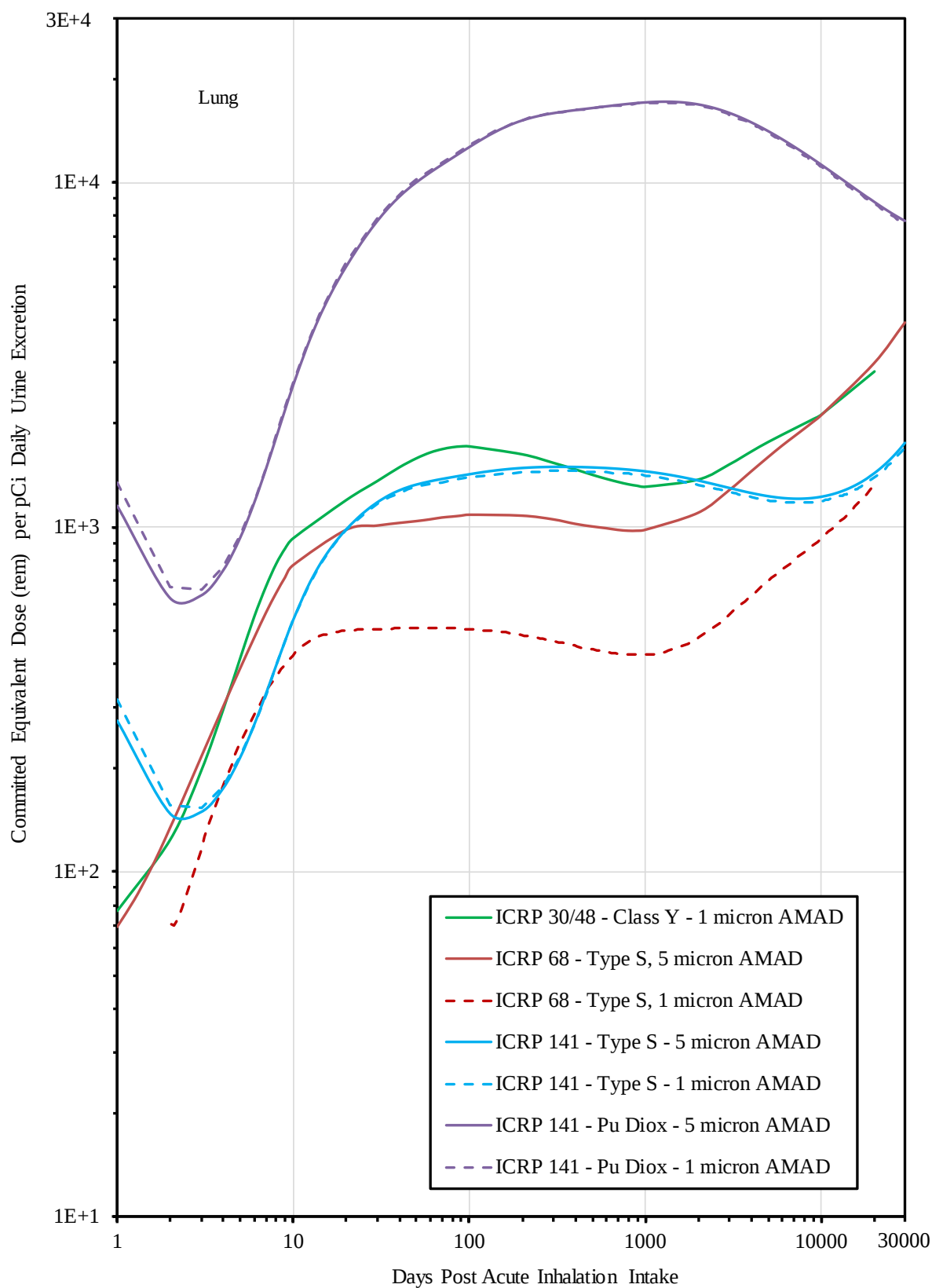


Figure F-9. Committed Equivalent Dose (or Dose Equivalent for ICRP 26) to Lung, Normalized to 1 pCi ^{239}Pu in Daily Urine Excretion for ICRP Reports 30/48, 68, and 141.

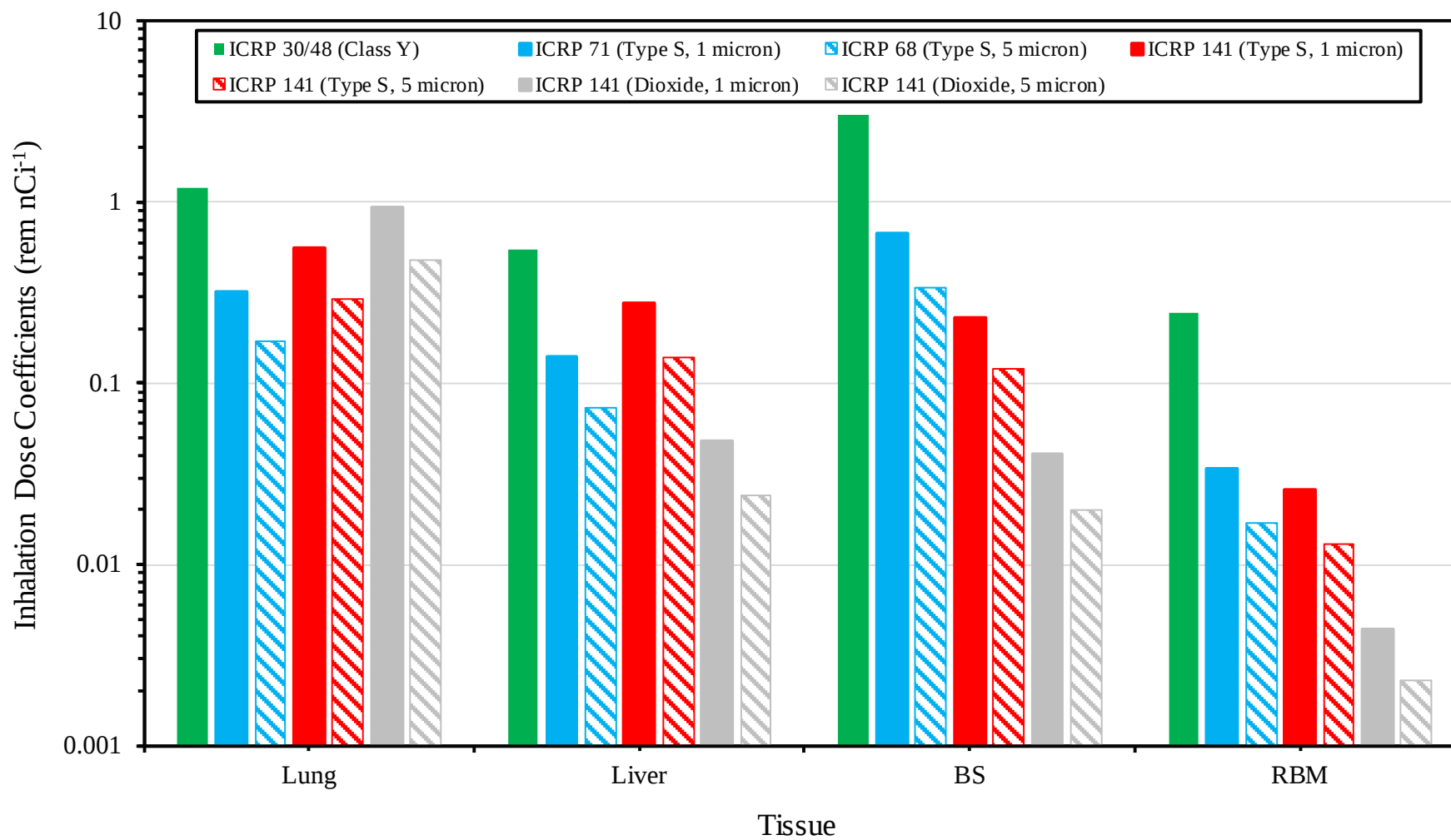


Figure F-10. Inhalation Dose Coefficients for ICRP Reports 30/48, ICRP 68/71, and ICRP 141 for Lung, Liver, Bone Surfaces (BS), and Red Bone Marrow (RBM) for ^{239}Pu .