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ANALYSIS OF WORKER CONSEQUENCES FROM A POTENTIAL ACCIDENTAL RELEASE OF SPECIAL NUCLEAR MATERIAL FROM A RECENTLY COMMISSIONED GLOVEBOX AT IDAHO STATE UNIVERSITY

By

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To the Graduate Faculty:

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LIST OF ACRYONIMS

AED	Aerodynamic Equivalent Diameter
AGS	American Glovebox Society
ALARA	As Low as Reasonably Achievable
ALI	Annual Limit on Intake
AMAD	Activity Mean Aerodynamic Diameter
ASTM	American Society for Testing and Materials
CEDE	Committed Effective Dose Equivalent
CFR	Code of Federal Regulations
DAC	Derived Air Concentration
DDE	Deep Dose Equivalent
dpm	decay per minute
FFTF	Fast Flux Test Facility at PNNL
HDBK	Handbook
HEPA filter	High Efficiency Particulate Air filter
IAEA	International Atomic Energy Agency
ICRP	International Commission on Radiological Protection
INL	Idaho National Laboratory
LLD	Lower Limit on Detection
MFC	Materials and Fuels Facility
NALI	Non-stochastic Annual Limit on Intake
NESHAPS	National Emission Standard for Hazardous Air Pollution
NFF	Nuclear Fuel Fabrication

NUREG	Nuclear Regulatory Commission Regulation
NUREG-CR	NUREG-Contractor
PNNL	Pacific Northwest National Laboratory
RISE Complex	Research and Innovation in Science and Engineering Complex
RSO	Radiation Safety Officer
SALI	Stochastic Annual Limit on Intake
SNM	Special Nuclear Material
TECDOC	Technical Document
TEDE	Total Effective Dose Equivalents
UR	Uranium Recovery
US DOE	United States Department of Energy
US EPA	United States Environmental Protection Agency
USNRC	United States Nuclear Regulatory Commission
WG	Water Gauge
WL	Working Level
WLM	Working Level Month

GI Tract	Gastrointestinal Tract
P	Pulmonary region
NP	Naspoharyngeal region
TB	Tracheobronchial region

ABSTRACT

An integrated safety analysis is carried out on a glovebox that was installed in the Research and Innovation in Science and Engineering (RISE) Complex to develop an assessment of possible dose equivalents to workers in the RISE Glovebox Laboratory and workers in the adjacent Crystal Growth Laboratory. Pertinent United States Nuclear Regulatory Commission (US NRC), Environmental Protection Agency (EPA) and Department of Energy (DOE) regulations and guidance documents are reviewed in the context of the RISE Glovebox. The NRC specified performance requirements for the glovebox that workers will not be exposed to more than 25 rem from the RISE Glovebox in the case of accidental release of uranium and/or plutonium, and persons outside the controlled radiation are consisting of the Glovebox Laboratory and the Crystal Growth Laboratory shall not be exposed to more than 5 rem. The dose equivalents were calculated using calculation methods recommended by the NRC and the DOE and by using the RESRAD-BUILD code. It was determined that the RISE Glovebox satisfies the performance requirements of the NRC.

CHAPTER 1 INTRODUCTION

1.1 Statement of the Problem

Single crystals of uranium oxide and plutonium oxide have been the subject of research interest for a variety of nuclear and non-nuclear reasons- mostly for its semiconducting properties and potential application for photovoltaic devices. Cutting-edge research being conducted at Idaho State University is developing single bulk crystals of uranium dioxide. The objective of this project is to better understand nuclear fuels, and to develop quantitative models of how nuclear fuels behave during the fission process, heat transport in nuclear fuel and eventually neutronics studies. The reason that this nuclear fuel research must be performed using single crystals is to eliminate the randomness that is inherently present in the nuclear fuel pellets that are commonly used in power and research reactors.

The fabrication of these uranium oxide single crystals involves crushing pellets of depleted uranium oxide originally from the Fast Flux Test Facility (FFTF) of the Pacific Northwest National Laboratory (PNNL). A new glovebox was installed at the Research and Innovation in Science and Engineering (RISE) Complex in order to allow for the grinding and polishing of the single crystals. The glovebox was designed to minimize the prospect of accidental releases of uranium oxide powder. Accidents, though must be considered and contingencies must be made in case an accident does happen. An important part of contingencies is an analysis that spells out the hazards that are present

in the RISE Complex, the accidents that can possibly occur, accidents that require further study and the results of said studies.

This thesis seeks to address possible accidental releases that can occur with the RISE glovebox at present. Inhaled and external doses were calculated using the RESRAD-BUILD code. The RESRAD-BUILD calculations were compared to "hand calculations" using the methods detailed in the US NRC NUREG-CR 6410, Nuclear Fuel Cycle Facility Accident Analysis Handbook and the UE DOE HDBK 3010-94, Airborne Release Factors/Rates and Respirable Fractions for Non-reactor Nuclear Facilities. Assumptions made in the course of this thesis were of a conservative nature; hence, the doses should be consider a worst-case scenario in the event of an accidental release of radioactive materials from the RISE Glovebox.

1.2 Benefits of this Thesis

10 CFR 70, "Domestic Licensing of Special Nuclear Material[1]" Subpart 61, "Performance Requirements" and Subpart 62, "Safety Program and Integrated Safety Analysis" require an integrated safety analysis in relation to critical amounts of special nuclear material, and specifies performance requirements for systems involving special nuclear materials that high-consequence and intermediate consequence accidents are minimized. This thesis undertakes the integrated safety analysis that is mandated in 10 CFR 70.

The most obvious benefit of this thesis is that an estimate of the external and inhaled dose equivalents in the event of an accidental release of radioactive materials from the RISE glovebox. The dose information is necessary to demonstrate that the glovebox is compliant with the requirements of 10 CFR 70.61 and 70.62. Furthermore, the dose information would be necessary to determine necessary medical treatments if it ever was the case that an accidental release of radioactive materials from the glovebox did occur.

1. Special nuclear material (SNM) is defined as plutonium and uranium enriched in the 233 and 235 isotopes. Critical masses of special nuclear materials are deemed to be 420 g of plutonium, 520 g of uranium-233, 700 g of uranium-235 or 1500 g of uranium-235 if it is enriched below 4%, or 420 g of any combination of fissile nuclides (10 CFR 70).

CHAPTER 2 BACKGROUND

2.1 Glovebox Preliminaries, Regulations and Standards

One of the more significant radiation safety issues in any facility that handles special nuclear materials, such as uranium enriched in the 233 and 235 isotopes and plutonium is the prevention of the spread of airborne radioactive materials- be it a uranium mill, uranium conversion facility, fuel cycle facility, fuel fabrication facility, reprocessing facility, or a radionuclide production facility. Once airborne radioactive materials are inhaled, a variety of health consequences that depend on the type and quantity of radionuclide inhaled, the chemical form of the radionuclide and particulate size are to be expected. The simplest solution to prevent the spread of airborne radioactive materials is to isolate them from the occupational and general environment. Isolation of radioactive materials has the additional benefit of reducing the dose equivalents of workers.

There are four methods that are commonly employed to isolate radioactive materials from the environment: fume-hoods, gloveboxes, tong-boxes and hot-cells. Fume-hoods are most appropriate for small amounts of radioactive materials of low to moderate specific activity; IAEA recommends that the face velocity comparable to or greater than the capture velocity, which is a function of the size of the particle. When this condition for the fume-hood cannot be satisfied or when one has a need to completely isolate the material being used from the worker breathing zone, then it is necessary to use a glovebox. A tong-box is used when working with several curies of radioactive material

and the extra distance is needed for shielding from the strong gamma-ray fields. It is often made from aluminum or steel, fitted with leaded glass for viewing operations. Hotcells are used for kilocurie amounts of radioactive materials, are constructed from concrete and equipped with master-slave manipulators (IAEA- TECDOC-1340, 2003).

Two standards pertaining to gloveboxes dedicated to plutonium work that are of significance to the nuclear industry in the United States are the American Glovebox Society AGS-G006-2005, "Standard of Practice for the Design and Fabrication of Nuclear-Application Gloveboxes," and the American Society for Testing and Materials (ASTM) C852-09, "Standard Guide for Design Criteria for Plutonium, Gloveboxes." Both Standards cover materials used in glovebox construction, criticality prevention, radiation protection, ventilation and gas service systems, gloveports and gloves (AGS, 2005; ASTM, 2009). Both standards specify that the glovebox shall be capable of preventing accidental release of radioactive materials during both normal and abnormal operation and mitigate negative effects of abnormal situations as much as maximally possible. Safety considerations that are common to both standards include the following:

- that sharp edges and points and crevices be absent from the interior of the glovebox
- that the glovebox operate at a negative pressure, or the pressure inside the glovebox is less than the pressure in the room
- the glovebox is electrically grounded
- the atmosphere inside the glovebox is filtered before entering into glovebox and as atmosphere exits the glovebox

- the gloveports should facilitate easy replacement of gloves, which should be located in such a way as to facilitate work and maintenance activities
- gloves should be selected to minimize the potential corrosion, eliminate to the most feasible extent the permeability of the chemical form of radioactive material to the hands of the workers, and by maximizing the manual dexterity of glovebox workers
- the glovebox should be well-lit, and that the illumination system should minimize glare for the glovebox workers
- the ventilation system shall have sufficient capacity so as to ensure that the glovebox shall remain at negative pressure relative to local atmospheric pressure
- HEPA filters shall be installed at every atmosphere exit and inlet of the glovebox to filter out any radioactive particles that possibly could be present
- Fire detection devices and fire suppression systems shall be installed
- Radiation shielding shall be provided as needed

The AGS standard is a considerably more detailed standard than the ASTM standard. The AGS standard specifies that the glovebox and gloveports shall be constructed of stainless steel types 304, 304L, 316, 316L, aluminum of the 6000 series alloys or a fire-retardant and high-strength fiberglass. It specifies that gloveports shall be attached by direct welding to the glovebox or by gaskets and bolts. Inert atmospheres are recommended when work is to be done involving uranium and/or plutonium, in part to prevent risk of fires occurring inside the glovebox (AGS, 2005; ASTM, 2009).

Regulatory Need and Requirements for Glovebox Safety Analysis

The United States Nuclear Regulatory Commission (US NRC) regulates uranium mills, conversion facilities, enrichment facilities, fuel fabrication facilities and reprocessing facilities in part through 10 CFR 70, "Domestic Licensing of Special Nuclear Materials" 10 CFR 70 and regulates the ownership, transport and use of special nuclear materials in the United States. Special nuclear material (SNM) is defined as plutonium and uranium enriched in the 233 and 235 isotopes. Critical masses of special nuclear materials are deemed to be 420 g of plutonium, 520 g of uranium-233, 700 g of uranium-235 or, 1500 g of uranium-235 if it is enriched below 4%, or 420 g of any combination of fissile nuclides (10 CFR 70).

Subpart H, Additional Requirements for Certain Licensees Required to Possess a Critical Mass of Special Nuclear Materials, applies to facilities that process enriched uranium, fabricate uranium fuel or fuel pellets, process enriched uranium hexafluoride, enrich uranium, process plutonium, or any other activity that the Commission deems could significantly affect public health and safety. Facilities that process special nuclear materials are mandated to have an integrated safety analysis. The safety analysis must address the following items (10 CFR 70.62):

- Radiological hazards pertaining to the processing of licensed material at the facility
- Chemical hazards of the licensed material
- Facility hazards that can impact the safety of licensed material and increase

radiological risk

- Potential accident scenarios that can result in the release of licensed material
- Consequence and likelihood of accident scenario
- Mitigation methods.

The NRC stipulates performance requirements that facilities processing critical amounts of special nuclear materials must meet (10 CFR 70.61):

- The risk of each credible high-consequence event must be limited, using engineering and/or administrative controls to the extent that the probability of occurrence is highly unlikely; high consequence events are defined as resulting in

(1) an acute total effective dose equivalent (TEDE) in excess of 100 rem (1 Sv) to workers

(2) an acute TEDE in excess of 25 rem (0.25 Sv) to anybody outside the controlled area

(3) an intake in excess of 30 mg in soluble form to anybody outside the controlled area

(4) acute chemical exposure resulting from the licensed material or hazardous chemical resulting from processes involving licensed materials that could

(a) threaten the life of workers

(b) lead to irreversible and other serious long-term health effects in anybody outside the controlled area

- The risk of each credible intermediate-consequence event must be limited, using engineering and/or administrative controls to the extent that the probability of occurrence is unlikely; intermediate-consequence events are defined as resulting in:

(1) an acute total effective dose equivalent (TEDE) in excess of 25 rem (0.25 Sv) to workers

(2) an acute TEDE in excess of 5 rem (0.05 Sv) to anybody outside the controlled area

(3) 24-hour averaged exposure in areas outside the controlled area should not exceed 5,000 times the DAC value listed in Appendix B of 10 CFR 20

(4) acute chemical exposure resulting from the licensed material or hazardous chemical resulting from processes involving licensed materials that could

(a) lead to irreversible and other serious long-term health effects in the workers

(b) lead to transient health effects in anybody outside the controlled area.

Risks of criticality must also be minimized, under normal and abnormal operational conditions, subcritical amounts of licensed material must be used in any processes using approved margins of subcriticality for safety (US NRC 10 CFR 70).

2.2 Regulatory Aspects of Uranium, Transuranic and Irradiated Material Processing

Exposure Limits

10 CFR 20 (Title 10, Chapter 1 of the Code of Federal Regulations, Part 20) prescribes exposure limits for uranium and plutonium based on both isotopic form and chemical form. Exposure limits based on chemical form are informed by the solubility class of the uranium and plutonium compounds. 10 CFR 20 makes reference to ICRP 30, whose solubility classes are D (days), W (weeks) and Y (years). The solubility classes depend on the retention time in the lungs; compounds that have a half-life of 10 days or less are Class-D; compounds that have a half-life between 10 and 100 days are Class-W, while compounds that have longer half-lives are Class-Y. 10 CFR 20 classifies UF_6, UO_2F_2, $UO_2(NO_3)_2$ as Class-D compounds, UO_3, UF_4 and UCl_4 as Class-W compounds, and both UO_2 and U_3O_8 as Class-Y compounds; all plutonium compounds are Class-D except for PuO2, which is Class-Y. (Turner, 2007; 10 CFR 20). The NRC limitations for uranium, plutonium and related compounds are listed in Table 2.1.

	Solubility Class	**Related Compounds**	**ALI (µCi)**	**DAC (µCi/m^3)**	**Air Effluent Limits (µCi/m^3)**	**Water Effluent Limits (µCi/m^3)**
U-235, U-238	D	$UO_2(NO_3)_2$ UO_2F_2 UF_6	1.0 (bone surface) 2.0 (else)	$6.0x10^{-10}$	$3.0x10^{-12}$	$3.0x10^{-7}$
	W	UF_4 UCi_4 UO_3	0.4	$3.0x10^{-10}$	$1.0x10^{-12}$	
	Y	UO_2 U_3O_8	0.3	$2.0x10^{-11}$	$6.0x10^{-14}$	
Natural U	**Solubility Class**	**Related Compounds**	**ALI (µCi)**	**DAC (µCi/m^3)**	**Air Effluent Limits (µCi/m^3)**	**Water Effluent Limits (µCi/m^3)**
	D	$UO_2(NO_3)_2$ UO_2F_2 UF_6	10.0 (bone surface) 20.0 (else)	$5.0x10^{-10}$	$3.0x10^{-12}$	$3.0x10^{-7}$
	W	UF_4 UCi_4 UO_3	0.8	$3.0x10^{-10}$	$1.0x10^{-12}$	
	Y	UO_2 U_3O_8	0.05	$2.0x10^{-11}$	$6.0x10^{-14}$	
	Solubility Class	**Related Compounds**	**ALI (µCi)**	**DAC (µCi/m^3)**	**Air Effluent Limits (µCi/m^3)**	**Water Effluent Limits (µCi/m^3)**

	D	Everything else	0.006 (bone surface) 0.001 (else)	$3.0x10^{-12}$	$2.0x10^{-14}$	$3.0x10^{-7}$
	Y	PuO_2	0.02	$7.0x10^{-12}$	$2.0x10^{-14}$	

Table 2.1. List of Annual Limits on Intake (ALIs), Derived Air Concentrations (DACs) and release limits for uranium and plutonium

US Environmental Protection Agency Regulations

The US Environmental Protection (EPA) regulations for drinking water have the following limits on radioactive materials: 15 pCi/L for gross alpha-emitting radionuclides, with the notable exceptions of 30μg/L for uranium, 15 pCi/L for Rn-226 and Rn-228. Gross β- and γ-emitting radionuclides are limited to an annual dose equivalent of 4 mrem to the total human body or any human organ; two notable exceptions are tritium (H-3) and strontium-90, which are 20,000 pCi/L and 8 pCi/L respectively (US EPA, 40 CFR 141).

40 CRF 61, "National Emission Standards for Hazardous Air Pollutants," commonly referred to as NESHAPS delineate limits on industrial air pollution for all industries. The subparts of most interest to the nuclear industry are Subparts H, I, Q and T. Subparts Q and T are the National Emission Standards for radon emissions from Department of Energy (DOE) facilities and for disposal of uranium mill tailings

respectively; emissions are limited to 20 pCi/L in both Subparts. . The limits on radionuclide effluent releases to the air for DOE facilities appears in Subparts H and Q; Subpart Q regulates radon emissions to the air, while Subpart H regulates all other radionuclide emissions to the air. Generally, all effluent releases to the air are to be limited such that exposure to members of the general public of radiation shall be limited to 10 mrem in any given year (US EPA, 40 CFR 61).

Air samples should be taken from all existing point sources that have the potential of causing doses to the general public in excess of 1 mrem in any given year, which is in excess of 10% of the National Emission Standard of 10 mrem annually. Sampling locations are to be selected at stacks and vents where the emission flow is moving in a known direction, and at locations where radionuclides are well mixed. The frequency of sampling will depend on the variability of the effluent flow rate. Acceptable sampling techniques include direct measurements of emissions flowing through stacks using inline radiation detectors and grab samples serving as a quality assurance check. Air samples taken at critical receptor sites that may be used in lieu of air-dispersion calculations (US EPA,40 CFR 61; Glissmeyer and Davis, 2000).

The US EPA mandates an annual NESHAPS report from DOE facilities, which is due to the EPA regional office and headquarters on 30 June. Items in the annual NESHAPS report shall include the name of the facility, list of radioactive material used, description of radioactive materials handling and processing, list of stacks and vents from which radioactive materials can escape into the environment, effluent controls, nearby

residences, schools, farms, and businesses, values used in all user-supplied input parameters for computer models, brief description of all construction and modifications in the previous year, and signatures of all facility directors, corporate officers and public officials. Any non-compliance facilities must report to the EPA Administrator on a monthly basis until the EPA Administrator determines that the facility is back in compliance. (US EPA, 40 CFR 61).

Health Physics Surveys

The Research and Innovation in Science and Engineering (RISE) Complex has research that involves the fabrication of single crystals of uranium dioxide (UO_2), and this research does involve UO_2 dusts. This following discussion concerns health physics surveys, radiological monitoring and surveillance, air monitoring, and dosimetry that are required to be carried out in uranium mines and recovery facilities, and uranium enrichment facilities and nuclear fuel fabrication facilities. This discussion is relevant to this thesis because the RISE Complex has similar radiological safety concerns.

The United States Nuclear Regulatory Commission (US NRC) and the Department of Energy (DOE) have occupational safety and environmental regulations on the processing of uranium, transuranics and irradiated materials. The US NRC has issued Regulatory Guide 8.24, "Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication," and Regulatory Guide 8.30, "Health Physics Surveys in Uranium Recovery Facilities." The uranium recovery (UR) facilities covered by

Regulatory Guide 8.30 include uranium mills, heap leach and in-site leaching facilities, ion-exchange recovery facilities, and can be extended to other related UR and uranium conversion facilities. Regulatory Guide 8.24 covers uranium enrichment and nuclear fuel fabrication (NFF) facilities. Regulatory Guide 8.30 does not cover surveys to prevent radioactive releases to unrestricted areas, or effluent releases (except for surveys of skin, clothing, shoes, equipment and packages), and Regulatory Guide 8.24 does not consider the processing of uranium-233, the attributes of the health physics staff or how to handle instrumentation, personnel dosimetry or bioassay programs. The regime of health physics surveys described in both NRC Regulatory Guides 8.24 and 8.30 overlap, though slight differences of details are present. Given that the radiological hazards for the RISE Glovebox have attributes both of a UR and a fuel fabrication facility, it is most appropriate to treat these Regulatory Guides together.

Air surveys are part of the regime of health physics surveys that are to be done in uranium processing facilities. Air sampling is to be done to demonstrate compliance with the requirements of 10 CFR 20.1201, determine airborne radioactivity zones, verify engineering and process controls and the need for increased radiological surveillance and possibly respiratory protection, and if releases are as low as reasonably achievable (ALARA). General principles are that air sampling should be representative of the air in which radiation workers will function, a mix of fixed location and breathing zone lapel air samplers should be used. Additionally, appropriate corrections for overestimates of the airflow rate onto the filter should be made to account for the buildup of uranium and other radioactive materials. The frequency of air surveys is to be commensurate with the

work being done, the amount of material being processed and the scope of engineering and process controls and the procedures used to protect workers from uranium uptakes and external exposures. The operational state of major equipment shall be recorded. Airborne radioactivity areas are typically those areas where the airborne uranium concentration exceeds a quarter of the ALI averaged over the number of hours in any one week in which radiation workers will be functioning (40 hours in a workweek is assumed), and this commonly comes out to be $5x10^{-11}$ μCi/mL for uranium ore dust and $1x10^{-10}$ μCi/mL for yellowcake. Generally, any yellowcake drying and packaging area will be considered to be airborne radioactivity areas. Radon and its progeny should be considered in these determinations. (Regulatory Guide 8.24, "Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication"; Regulatory Guide 8.30, "Health Physics Surveys in Uranium Recovery Facilities")

Regulatory Guide 8.30 stipulates that fixed location air samples should be collected at a height of three to six feet and positioned between the source and radiation workers, and shall be taken while work functions are occurring. The derived air concentration (DAC) for gross alpha contamination is $6x10^{-11}$ μCi/mL, applying to uranium-234m uranium-235, uranium-238, thorium-230 and radium-226; however, if alpha spectrometry or fluoroscopic methods are used to analyze air samples, then the DAC for natural uranium of $3x10^{-11}$ μCi/mL applies. Surveys in airborne radioactivity areas are to be done as 30 minute grab samples on a weekly basis. Surveys in uncontrolled areas (free of airborne radioactivity) as 30 minute grab samples on a monthly basis. A lower limit of detection (LLD) of $3x10^{-12}$ μCi/mL (or 4.5 mg/m^3) is

required for instrumentation. Yellowcake surveys do require lapel air samplers, especially for Class-D (soluble) uranium baked below 400 °C, weekly 30 minute grab samples. Limits on the air concentration of Class-D uranium compounds are limited by uranium's chemical toxicity to the kidneys, while concentration limits for Class-W and Class-Y uranium compounds are limited by radiation dose to the lung. When handling yellowcake, protective clothing such as coveralls, shoe covers (or special shoes) and thick rubber gloves are strongly encouraged to prevent yellowcake spread to uncontrolled areas; if protective clothing is not discarded, then it should be laundered in an NRC or agreement state approved laundry facility. Both uranium recovery (UR) and nuclear fuel fabrication (NFF) facilities must contend with radon progeny. Regulatory Guide 8.30 allows for either direct measurements of radon or measurements of radon progeny, but recommends measurements of radon progeny as the dose from radon progeny is much greater than the dose from radon itself. Radon levels in UR facilities are not to exceed 0.3 working levels[2]. Radon surveys are to be conducted monthly when radon levels are 0.03 working levels or less and performed weekly if radon levels exceed 0.08 working levels. Analytical methods for analyzing radon samples include alpha spectrometry analysis codes, or gross-alpha counting immediately after the sample was collected and 24 hours after the initial measurement, giving radon progeny sufficient time to decay away (Regulatory Guide 8.24, "Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication"; Regulatory Guide 8.30, "Health Physics Surveys in Uranium Recovery Facilities").

2 A working level (WL) is the equilibrium concentration of radon and radon progeny in 1 liter of air that will result in the emission of 1.3×10^5 MeV of alpha energy, and a working level month (WLM) is exposure to 1 WL for 170 working hours in a month (10 CFR 20).

Air monitors have special mention in Regulatory Guide 8.24. Air monitors are to be used to alert radiation workers when uranium concentrations become unexpectedly high, as their principal function is to alert workers to take immediate action to protect themselves from airborne uranium. The uranium intakes are most likely to occur between the onset of the unexpected uranium release and the initial annunciation of air monitor. Hence, it is necessary to shorten this response time as much as possible. Several approaches are to use as little tube plumbing as possible, use tubing materials that will not absorb uranium, placement of the air monitor as close to the uranium as possible. It is necessary to make the alarm set point to minimize false alarms as much as possible, lest workers lose confidence in the air monitoring system (Regulatory Guide 8.24, "Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication"; Regulatory Guide 8.30, "Health Physics Surveys in Uranium Recovery Facilities").

Surveys for both external beta and gamma radiation are to be performed in uranium recovery, enrichment and fuel fabrication facilities. External beta and gamma radiation surveys are useful to determine which areas are radiation areas, and to document significant changes over time of radiation fields in the facility, and to detect undesirable buildup of radioactive materials in equipment; preoperational benchmark surveys should be performed to determine a baseline survey. Regulatory Guide 8.30 stipulates that quarterly surveys for external beta and gamma radiation are to be performed in all radiation areas on a quarterly basis and semiannually in all areas of the UR facility. Radon can build up in various circuits of a UR facility and both uranium

enrichment and fuel fabrication facilities have special radiological hazards associated with pellet trays, fuel rods, fuel rod bundles and storage areas where attention should be given to enrichment, daughter products contaminants and cladding. Uranium enrichment and fuel fabrication facilities have an additional concern of neutron radiation and neutron reaction with fluorine-19. Beta radiation surveys are particularly useful in uranium recovery facilities to determine the extent of protective clothing and other needed personal protective equipment (i.e., rubber gloves) and in refining operational procedures to reduce exposure to beta radiation. Beta surveys in UR facilities should be performed on the surface or at short distances, while gamma surveys are to be performed at 30 cm (12 in) (Regulatory Guide 8.24, "Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication"; Regulatory Guide 8.30, "Health Physics Surveys in Uranium Recovery Facilities").

Surveys for removable radioactive contamination are necessary to ensure that equipment, processes and procedures are sufficient in containing uranium in its controlled areas and to prevent unnecessary external exposure or internal uptake of uranium. Regulatory Guide 8.30 states the most important source of removable contamination in UR facilities is yellowcake and uranium ore dust. Sweeping of such uranium contamination is not suggested, as sweeping can suspend said dust into the air and create an even worse situation; it is preferable instead to hose down the yellowcake or ore dust on a contaminated surface or object into a floor sump or to use a vacuum cleaner with a filtered exhaust. The International Atomic Energy Agency (IAEA) suggests an alpha-contamination limit of 10^{-3} $\mu Ci/cm^3$ (or 2 mg/ cm^3 of natural uranium or 220,000

dpm/100 cm^3) on floors, walls, benches, clothing and so forth; this amount of yellowcake is visible to the naked eye (provided that yellowcake rooms are painted with contrasting colors), thus allowing for a visual survey. The NRC recommends a swipe survey for which contamination can easily be transferred to a dry smear paper and counting said paper with a gas-flow alpha-beta proportional counter, alpha scintillation counters or thin-window

Geiger-Mueller counters. Uncontrolled areas in UR facilities should be surveyed weekly to verify that uranium is confined to controlled areas (Regulatory Guide 8.24, "Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication"; Regulatory Guide 8.30, "Health Physics Surveys in Uranium Recovery Facilities").

Both Regulatory Guides 8.24 and 8.30 give extensive mention of removable surface contamination on workers, protective clothing and equipment. When radiation workers are leaving a controlled area, it is necessary that they survey their hands, clothing and shoes before leaving the controlled area with an alpha-survey instrument. Contamination limits are 1000 dpm/100 cm^2 on skin and clothing, and 5000 dpm/100 cm2 for soles of shoes. Shoes are to be scrubbed if above the contamination limit; workers must shower if they are contaminated, and their clothing must be laundered in an NRC or agreement state licensed laundry. It is suggested that special protective clothing and shoe covers or special shoes should be issued to radiation workers when they are about to enter a controlled area, especially for areas that have potential for extremely high radiation levels. Equipment that is to be removed from a controlled area must be surveyed and has a contamination limit less than 1000 dpm/100 cm^2 (Regulatory Guide

8.24, “Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication”; Regulatory Guide 8.30, “Health Physics Surveys in Uranium Recovery Facilities”).

Dosimetry is an important issue in any radiological workplace. Personal dosimetry is required for all radiation workers who are likely to receive an annual dose of 10 percent of any NRC dose limit, as stated in 10 CFR 20.1502. The total effective dose equivalent (TEDE) is the sum of the deep dose equivalent (external dose) and the committed effective dose equivalent (CEDE), which measures the internal uptake; in the event of an internal uptake, there are two methods that should be used to assess CEDE: one using the annual limit on intake (ALI) and the other using the derived air concentration (DAC). The CEDE from the ALI is calculated as:

$$CEDE = 5*I_i/ALI_{i,E} \tag{2.1}$$

where i is a specific radionuclide, I_i is the measured quantity of radionuclide i over one calendar year, and $ALI_{i,E}$ is either ALI for the radionuclide; whichever ALI is more restrictive, be it the stochastic ALI (SALI) or the non-stochastic ALI (NALI). The DAC is calculated from the ALI as

$$DAC = ALI / 2.4x10^9, \tag{2.2}$$

where $2.4x10^9$ m^3 is the volume of air inhaled by reference person in one calendar year

(10 CFR 20); the CEDE then becomes

$$CEDE = \frac{(5\,rem)C_i * t}{2000\,DAC - hours} \tag{2.3}$$

where C_i is the concentration of radionuclide i that the worker was exposed to and 2000 is the number of working hours in one calendar year (2,000 DAC-hours incurs a dose equivalent of 5 rem), and t is the time in units of hours of the duration of exposure. The NRC normally does not accept results of air, beta/gamma or other surveys in lieu of personnel dosimetry to verify compliance with dose limits. An exception can be made in the event of a dosimeter being lost or damaged; estimates of the dose using results of survey data combined with appropriate occupancy factors; these records must be maintained for as long as the NRC license is in effect. The health physics staff should promptly review all dosimetry data, in part to identify negative trends and potentially hazardous situations and to ensure that ALARA is maintained (Regulatory Guide 8.24, "Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication"; Regulatory Guide 8.30, "Health Physics Surveys in Uranium Recovery Facilities").

Action levels should be set slightly above the normal background fluctuations that occur during normal operation for uranium ore dust, yellowcake and beta/gamma external radiation. Yellowcake should have an additional set of action levels when lapel air monitors are used. The Radiation Safety Officer[3] (RSO) should investigate whenever any

3. The Radiation Safety Officer, or an equivalent position oversees the radiation protection program.

action level is exceeded and take appropriate corrective action to prevent such future incidents. The RSO should also investigate any worker's time-averaged exposure, calculated either by ALI or DAC that are in excess of a quarter of any annual dose limit, or if any worker receives an external radiation contamination in excess of 1000 dpm/100 cm^2 and take the appropriate corrective actions to such prevent future incidents. Additionally, the RSO should have action levels for abnormally low concentrations of airborne radioactivity (uranium ore dust, yellowcake, radon and its progeny, or other source of radioactivity), and this can indicate equipment malfunction or procedure error and create a worse radiological problem elsewhere in the facility; the RSO should investigate whenever the low concentration action level is breached and take appropriate corrective action to prevent such future incidents. The licensee shall report all action level breaches to the NRC as soon as possible, in accordance with 10 CFR 20, Subpart M (Regulatory Guide 8.24, "Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication"; Regulatory Guide 8.30, "Health Physics Surveys in Uranium Recovery Facilities").

When transporting tools, equipment, scrap from controlled areas to uncontrolled areas, removable contamination surveys should be assumed to be contaminated until shown otherwise; it is suggested that items are contaminated if they have 1000 dpm/100 cm^2 or more. It is good practice to avoid as much hand manipulation of items being moved from controlled areas as possible; it is suggested that items and material be transported via carts, conveyor belts or other mechanical equipment. Packages containing radioactive material should only be opened in fume hoods, glovebox or some other

properly ventilated facility. When licensees are intend to release package for shipment, the package's external surfaces must be surveyed for removable contamination and total radioactivity and have contamination levels in accordance with Department of Transportation's limits, which are 2,200 dpm/100 cm^2 beta-gamma and 20 dpm/100 cm^2 alpha for shipment in non-exclusive use vehicles and 22,000 dpm/100 cm^2 beta-gamma and 2,200 dpm/100 cm^2 alpha in exclusive-use vehicles; additionally, surveys and labeling for packages containing radioactive materials must comply with 10 CFR 71, "Packaging and Transportation of Radioactive Material." Yellowcake packages should be washed down after being filled before survey and shipment (Regulatory Guide 8.24, "Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication"; Regulatory Guide 8.30, "Health Physics Surveys in Uranium Recovery Facilities").

Survey instruments should be on a routine maintenance and calibration cycle to ensure that they properly functioning. It is not adequate to calibrate survey instruments with built-in check sources, nor is it adequate to perform an electronic calibration not involving radiation check sources. Each survey instrument should be calibrated at two points on every frequently used linear scale; Regulatory Guide 8.24 specifies 20% and 80% of each linear scale, while Regulatory Guide specifies 33.3% and 66.7% of the linear scale; logarithmic scales only need one calibration point near the midpoint. Survey instrument that have digital readout scales that have electronic or manual scale switching should be treated as a linear scale meter, while survey meters that do not have scale switching should be treated like a meter with an instrument with a logarithmic scale.

Survey instruments are to be calibrated on an annual basis, and they are to have ±10 accuracy according to Regulatory Guide 8.24, but ±20% accuracy according to Regulatory Guide 8.30 (Regulatory Guide 8.24, "Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication"; Regulatory Guide 8.30, "Health Physics Surveys in Uranium Recovery Facilities").

Surveys of the ventilation system are to be performed monthly by the health physics staff, on fume hoods and gloveboxes. Checks that should be done on fume hoods include checks of the airflow velocity at hood entrances and other exhausted enclosures and close-capture points are sufficient to preclude escape of airborne uranium- taking into consideration the density and material particle distribution, and it minimize potential for uranium intake. A properly calibrated thermoanemometer or velometer should be used. The average airflow velocity is 45 m/min (150 ft/min) and should be measured at five different locations in the fume hood. Manometers or other pressure check devices should be present in the fume hood to measure pressure drops across filters to give early warning of airflow reduction; the pressure should be monitored throughout work in the fume hood. Work should be immediately terminated if the airflow velocity falls below 30 m/min (100 ft/min), as airflow disturbances may indicate unacceptable filter loading exhaust fan malfunctions or other issues with the fume hood. Gloveboxes should always be kept under negative pressure. Additional surveys, beyond routine glovebox surveys should be performed whenever any work that can compromise the glovebox integrity (such as changing a glove) is performed (Regulatory Guide 8.24, "Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication"; Regulatory

Guide 8.30, “Health Physics Surveys in Uranium Recovery Facilities”).

Another important part of any health physics program is surveillance of the radiological workplace in which the health physics staff functions, and surveillance of the radiological workplace is one of the very most important components of any radiation protection program. The health physics staff should have as detailed a knowledge of each operation in the facility, the processes, equipment and procedures as is practical. This will allow the health physics staff to identify ways to prevent or minimize occupational exposures to external radiation and uranium intakes, select the most appropriate survey times and methods, and to allow for adequate preparation for action to be taken in cases of equipment breakdown or emergency (Regulatory Guide 8.24, “Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication”).

Periodic surveys of uncontrolled areas should be performed to verify and document that airborne uranium is being confined to controlled areas. The frequency of surveys of uncontrolled areas is to be consummate with the type of work in the facility, the quantity of radioactive materials being processed and the scope of engineering and process controls, specific protective facilities and procedures that are in place at the facility. It is suggested that break rooms, lunchrooms, snack bars, cafeterias, vending machines, offices, furniture and select random floor locations in uncontrolled areas be subject to survey for removable radioactive contamination on a periodic basis. If escape of uranium is determined to have occurred from analysis of swipe data, then corrective action must be taken as soon as is possible; contaminated areas must be decontaminated and surveillance of uncontrolled areas must become more frequent until a negative trend

is reestablished. It is also suggested that the transport of uranium not occur in uncontrolled areas, or that procedures be established that eliminate or minimize uranium contamination when it is transported through corridors (Regulatory Guide 8.24, "Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication").

Records must be kept of radiation surveys and dosimetry reports of radiation workers. Dosimetry reports on external dose equivalents in units of rem or mrem to relevant organs, air concentrations in dpm/100 cm^2 or μCi/mL, surface contamination in units of..., uptakes of radioactive materials in terms of μCi (mg acceptable for uranium-234, uranium-235 or uranium-238) and the percentage of ALI which should reference bioassay data. Additionally, records showing results of air sampling sufficient to identify potential hazards used to justify the selection of a particular respiratory protection system, records showing the results of bioassays to evaluate intakes, records of whole body count bioassays, external dose rate equivalents, records of results of measurements and calculations used to determine individual intakes and resulting internal doses, and records of release of radioactive effluents into the environment are to be kept in the permanent file until the NRC terminates the operating license of the facility (Regulatory Guide 8.24, "Health Physics Surveys during Enriched Uranium-235 Processing and Fuel Fabrication"; Regulatory Guide 8.30, "Health Physics Surveys in Uranium Recovery Facilities").

2.3 Safety Analyses of other Nuclear Material Gloveboxes

Gloveboxes are used in in the nuclear industry for a variety of radiochemical separations and other chemical reactions when a large amount of radioactive material being processed or if the specific radionuclide has a high specific activity, or if the chemical properties of a radionuclide-containing compound requires an inert atmosphere or vacuum conditions. The US NRC requires that a safety analysis be performed on all infrastructure designed to handle and/or process special nuclear material. Many facilities that handle, process and treat special nuclear materials have affiliations with the US DOE, and DOE L and/or Q clearances often are required for admission to such facilities, access to information about processes inside such facilities. Hence, it is difficult to easily find comprehensive information about special nuclear material gloveboxes; yet, general information about special nuclear material gloveboxes is still available (NUREG 1821).

The details and the means of a safety analysis in included in NUREG-1821, Final Safety Evaluation Report on the Construction Authorization Request for the Mixed Oxide Fuel Fabrication Facility (MOX FFF) at the Savannah River Site, South Carolina, hazard evaluations for specific components of the MOX FFF, such as gloveboxes are not included, and the results of accidental releases of radioactive materials are not included in NUREG 1821. Four accidents involving gloveboxes at the MOX FFF are over-temperature accidents, over- and under-pressurization accidents, breach of containment accidents, and dynamic exhaust failure accidents; all four types of accidents involve loss

of confinement and possible dispersal of radioactive materials. Over-temperature accidents, for the purposes of the MOX FFF complex involve excessive heat generated by the AP electrolyzer, control system failure, electrical isolation failure, or loss of cooling to process equipment, etc. sufficient to cause high-temperature damage to glovebox panels and disperse radioactive material. Breach of containment accidents could result from small breaches in the structure of the glovebox (for reasons other than an over-temperature accident), or a backflow through the nitrogen or helium lines; the material at risk is the entire radioactive material inventory inside the glovebox at that moment; this accident can be prevented by maintaining negative pressure inside the glovebox. The dynamic exhaust failure accident can result when negative pressure inside the glovebox is lost or when a severe perturbation in the gas flow occurs. The material at risk in all accident scenarios is the entire radioactive inventory present inside the glovebox; accident analysis deemed all four accident scenarios to be above the criteria of 10 CFR 70.61(c) (NUREG 1821).

Dosimetry calculations for accident scenarios were carried out using the 5-factor formula, which is discussed in Section 4.2 of this thesis. The dosimetry results of the accident scenarios that would result in radiation exposures were not included in NUREG 1821 (largely for reasons of national security), but the NRC staff agreed with the conclusions in the construction authorization request (CAR) for the MOX FFF that all components therein satisfy the requirement of 10 CFR 70.61, that “the plant site description relating to safety assessment was found to be adequate” and “the safety assessment team description was found to be adequate” (NUREG 1821).

Review of the analysis of the MOX FFF glovebox safety analysis gives insights to carrying out a safety analysis of the RISE glovebox, and comparison of work activities in both gloveboxes is insightful for the undertaking of the safety analysis of the RISE glovebox. There are no processes carried out in the RISE glovebox at this moment that would generate heat sufficient to cause a high-temperature breach of the glovebox structure. Over- and under-pressurization accidents are of concern to the RISE glovebox. A computer-controlled mass-flow-controller, at the time of the defense of this thesis is being designed to control the pressure inside the glovebox. The use of negative pressure for gloveboxes is standard practice for radioactive material gloveboxes. The accident scenario that was deemed to be of most concern with the RISE glovebox was a dynamic exhaust failure at the same time that the RISE glovebox had a breach in the gloveport or in a glove.

2.4 Internal Dosimetry

Particle Kinetics and Internal Uptake of Uranium Dioxide Dust

The internal dose of a UO_2 dust uptake strongly depends on the size of the particles constituting the UO_2 dust. Particles in air are influenced primarily by gravitational settling and diffusion, which are aerodynamic and thermodynamic properties respectively. Diffusion predominates for particles smaller than 0.1 μm while gravitational

settling predominates for particles greater than 1 μm. Other means of particle deposition include impaction and interception. When considering larger particle for which aerodynamic effects predominate, the particle is being pulled down by the gravitational force, but there is also the frictional force that resists the gravitational force, and a terminal velocity is ultimate observed which is used as a measure of settling of the particle onto the ground and is sometimes called the settling velocity. Almost always, it is the case that dust particles are highly irregular in their structure, and it is impossible for most practical purposes to handle this situation theoretically. It is therefore more convenient to develop a type of effective aerodynamic diameter, which is more convenient even from an experimental perspective. The aerodynamic diameter, which is normalized to a sphere of unit density, is convenient because knowledge of particle density is not necessary. One way to normalize various sizes and densities of particles is called the activity mean aerodynamic diameter (AMAD), and it is the effective diameter used to in the ICRP 30 lung model and successive ICRP lung models. If the particle's chemical composition and actual diameter are known, then the AMAD can be calculated as:

$$\frac{d_{AMAD}}{d_{actual}} = \sqrt{\frac{\rho_{actual}}{\rho_{unit}}} \tag{2.4}$$

where d_{AMAD} and d_{actual} are the AMAD diameter and actual diameter of the particle, ρ_{actual} and ρ_{unit} are the particle density and 1 g/cm^3 respectively. UO_2, which has a density of 10.97 g/cm3, and an actual diameter of 10 μm, will have an AMAD of 33.12 μm. UO_2 particles, if they escape from the glovebox have an AMAD of about three times the

"physical radius." This will be an important mitigating factor in the safety analysis (Cember and Johnson, 2008).

The depth of penetration of any dust or aerosol into the respiratory tract is a function of the AMAD of the particle; absorption of the dust is dependent on the chemical composition of the dust. Particles with an AMAD > 5 μm will typically be caught by nasal hair on impact and be filtered out; particles that are not filtered out by the nasal hairs are not likely to be able to follow the sharp bends in the respiratory tract and deposit in the nasopharyngeal surface, largely due to the size of the inhaled particle (Cember and Johnson, 2008).

Particles with an AMAD < 10 μm will have less inertial impact and are more likely to be carried deep into the lung. The air in the alveoli is relatively stationary, given that only a small fraction of the alveoli air is exchanged with incoming air during the breathing process. Therefore, particles that are carried deep into the respiratory tract still have the opportunity to settle out via gravitational settling, though the probability of this occurring reaches a minimum at 0.5 μm; Brownian motion and other thermodynamic effects predominate for particles smaller than 0.1 , and as a result they are more likely to collide with alveolar walls. The likelihood of deposition of particles deep into the respiratory tract is at a maximum for particles of 1 to 2 μm and is at a minimum for particles of 0.1-0.5 μm. A graph showing the percent deposition of particles as a function of aerodynamic diameter in various parts of the respiratory tract is shown in Figure 2.1 (Cember and Johnson, 2008).

The retention of particles in the lung is a function of the physical and chemical properties of the particles and the physiological properties of the lungs. The retention of particles in the lungs is of great importance in determining the radiological (or chemical) hazards associated with the dust material because the time of residence in the lungs, the rate of dissolution and the total dose to the lungs. There are three mechanisms by which dusts and other foreign materials are cleared from the respiratory tract: ciliary clearance, solubility and absorption of particles in the lungs, and phagocytosis. Ciliary clearance is the only clearance mechanism associated with the upper respiratory tract; it works through the rhythmic beating of the ciliated epithelial cells (also known as the respiratory epithelium or cilia) that moves mucous toward the throat, and is also known as the ciliary escalator. Particles that are purged from the respiratory tract via the ciliary escalator will either be coughed out or enter into the gastrointestinal (GI) tract. The ciliary escalator is able to move mucous at a speed of 2 mm/min in the bronchi and 3 cm/min in the trachea (Cember and Johnson, 2008).

Some dusts are soluble in the lungs, and these will be absorbed into the capillary bed across the alveolar membrane. Most inhaled particles which are soluble in the lungs that are carried into the lungs will be absorbed into the blood and transported to other organs wherein the can be absorbed. It should be noted that solubility in lungs may not always correspond to solubility in water. Cember notes two examples that illustrate the differences; HgS is one of the most insoluble (in water) molecules known to science; yet 1 μm ^{209}HgS particles are found in significant quantities in urine samples in rats. $CeCl_3$

intratracheally inject into rats was found to be a very long retention time, because of Ce bonding to tissue protein in the lung (Cember and Johnson, 2008).

Phagocytosis is the process in which macrophages engulf foreign objects and remove them either through the ciliary escalator and ultimately passing into the GI system, or through entrance into the lymphatic system. The rate of phagocytosis of dusts is strongly dependent on the nature of the dust particle. Phagocytes loaded with radioactive dusts can be trapped in the sinuses of the cheobronchial lymph nodes and remain therein for extended periods of time, giving a higher dose to the lymph node than to the lungs. Interestingly enough, it has been shown that the rate of phagocytosis is slower for radioactive dusts than for non-radioactive dusts of the same chemical composition, physical form and size distributions (Cember and Johnson, 2008).

The retention of inhaled particles in the lungs is commonly fit by a series expansion of exponential curves that includes at the least two components: one for inhaled particles of a short residence time (several hours) and the other on the order of days, which itself may be an expansion of exponential functions. The retention function has the form

$$Q(t)=Q_1 e^{-k_1 t}+Q_2 e^{-k_2 t}+.... \tag{2.5}$$

where Q_1 and Q_2 are the dust concentrations deposited in the upper and deep respiratory tracts respectively, and k_1 and k_2 are the clearance rates for upper and deep respectively (Cember and Johnson, 2008).

International Commission for Radiation Protection (ICRP) 30 Lung Model

The International Commission for Radiation Protection (ICRP) releases updated internal dosimetry models on a semi-regular basis, with the most recent model ICRP 103 having been released in only the last few years. The US NRC however, still uses the ICRP 30 internal dosimetry model, published in 1979. The IRCP 30 lung model takes into account the observation that particle deposition in the respiratory tract is a function of airflow dynamics in the respiratory tract, the size distribution of particles being inhaled, the location within the respiratory tract that on which particles deposit, and both chemical and physical properties of the inhaled particles. The ICRP 30 lung model is designed to take into account particle size distributions between 0.2 and 10 μm. The organ weighting factors used in 10 CFR 20 are taken from ICRP 30; hence any dosimetry calculations must refer to ICRP 30 (Cember and Johnson, 2008).

The dose equivalent is calculated as the energy absorbed by the organ or tissue per radioactive transformation multiplied by the number of radioactive transformations divided by the mass of the organ or tissue in which the radioactive material is located, or mathematically:

$$H=1.602\times10^{-13}\ \Sigma U_S\times SEE(T\leftarrow S)\ , \tag{2.6}$$

where H is the dose equivalent, U_S is the number of radioactive transformations occurring

inside the organ or tissue, and SEE($T \leftarrow S$) is the specific equivalent energy per transformation absorbed by the the organ or tissue; S denotes the source organ, or the organ that took the internal dose, and T denotes target organs. U_S is defined as

$$U_s = \frac{A_S(0)}{\lambda_E}\left(1 - e^{-\lambda_E t}\right) \tag{2.7}$$

where $A_S(0)$ is the initially deposited activity in the compartment and λ_E is the effective decay constant, which itself is the sum of the radioactive decay constant and the biological clearance decay constant. The dose is summed over every radiation i. The number of transformation itself is defined as

$$SEE(T \leftarrow S) = \frac{\sum_i n_i E_i \varphi_i(T \leftarrow S)}{m_T} \tag{2.8}$$

where n_i is the mean number per transformation , E_i is the energy of the radiation in the organ or tissue, $\varphi_i(T \leftarrow S)$ is the fraction of the radiation that is absorbed in the target organ or tissue from the source tissue or organ, and m_T is the mass of the organ or tissue. Alternatively, ICRP 30 provides several tables of for each organ and fractions of radiation that will go to other target organs, as a function of the species of radionuclide and the energy of the radiation (Cember and Johnson, 2008).

The solubility of inhaled radioactive particles is accounted for in the ICRP 30 internal dosimetry model by creating three particle solubility classes: D, W and Y. Class

D particles have a dissolution rate (clearance half-life) on the order of days or fraction of a day; Class W particles have clearance half-lives on the order of weeks (or months); Class Y particles have retention rates in the lungs on the order of years. It is interesting to note that only Class Y particles are known to be retained permanently in the lymphatic system. The ICRP 30 model treats the lung and the pulmonary lymph node as a single organ with the activity in the lung and the pulmonary lymph node being added together and the sum mass of the lungs and the pulmonary lymph node used to calculate the doses from inhaled radioactive particles (Cember and Johnson, 2008).

The three generic classes of particle solubility were developed by the ICRP as an alternative approach to overcome the inherent impracticality of having to calculate clearance half-lives for every particle of every chemical composition and of every AMAD. The ICRP 30 lung model divides the respiratory tract into three regions: the nasopharyngeal region (NP), the tracheobronchial region (TB) and the pulmonary region (P); it is in the pulmonary region that is treated as the deep respiratory region where gas exchange with blood takes place. A diagram showing the physiological lung model is shown in Figures 2.2 and 2.3 (Cember and Johnson, 2008).

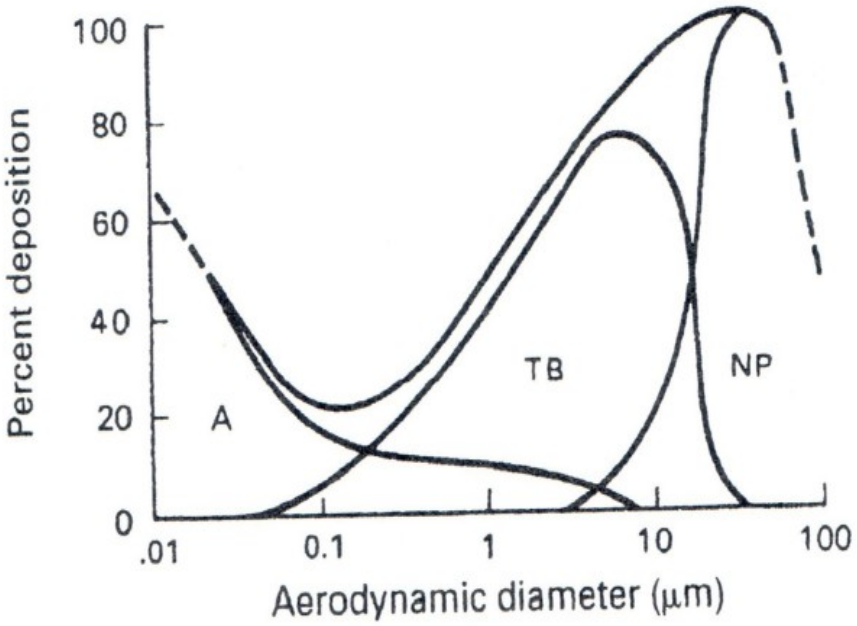

Figure 2.1. Particle deposition in the respiratory tract as a function of the particle size. This graph is from P.E. Marrow, Evaluation of inhalation hazards based upon the respirable dust concept and the philosophy and application of selective sampling. Association of Industrial Hygienists of America Journal. 1964: 25(3): 213-236. The region denoted as "A" is the alveolar region, which is referred to as the pulmonary region (P) in ICRP 30. Notice that deposition in the P region is virtually nonexistent for particle sizes beyond 10 μm AMAD. Particle deposition prevails in the tracheobronchial region (TB) for particles of 0.2 to 12 nasopharyngeal AMAD. Deposition of particles larger than 20 μm AMAD is virtually restricted to the nasopharyngeal region (NP). Taken from Cember and Johnson, 2008.

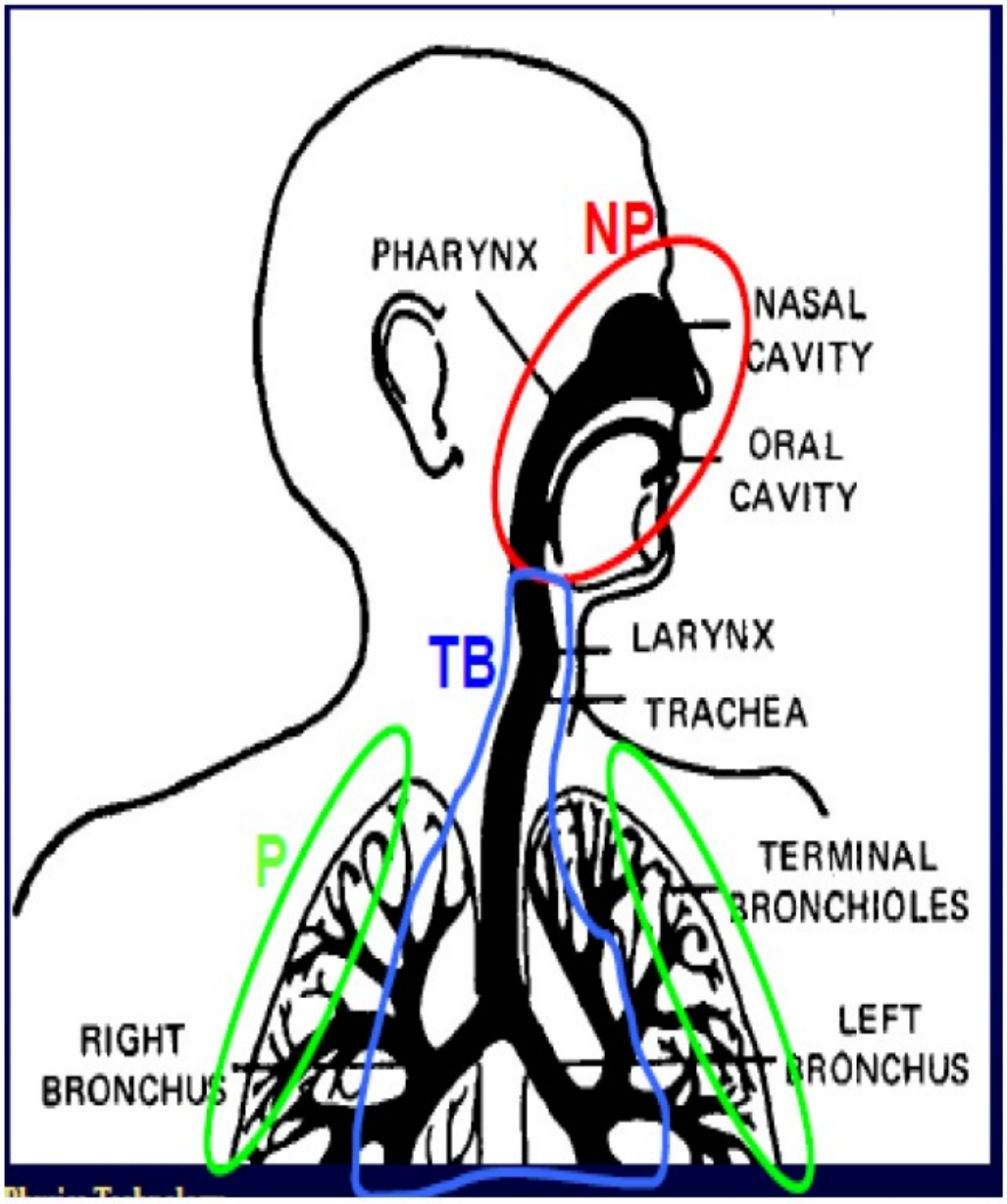

Figure 2.2 ICRP 30 Lung Model. The Nasopharyngeal region (NP) takes in the nose, nasal cavity and the oral cavity. The Tracheobronchial region (TB) takes in the larynx, trachea, and the larger branches inside the lung. The Pulmonary region (P) takes in the right and left bronchi and the alveolar areas where the gas exchange with blood takes place. Taken from the US Nuclear Regulatory Commission, Health Physics Technology training document.

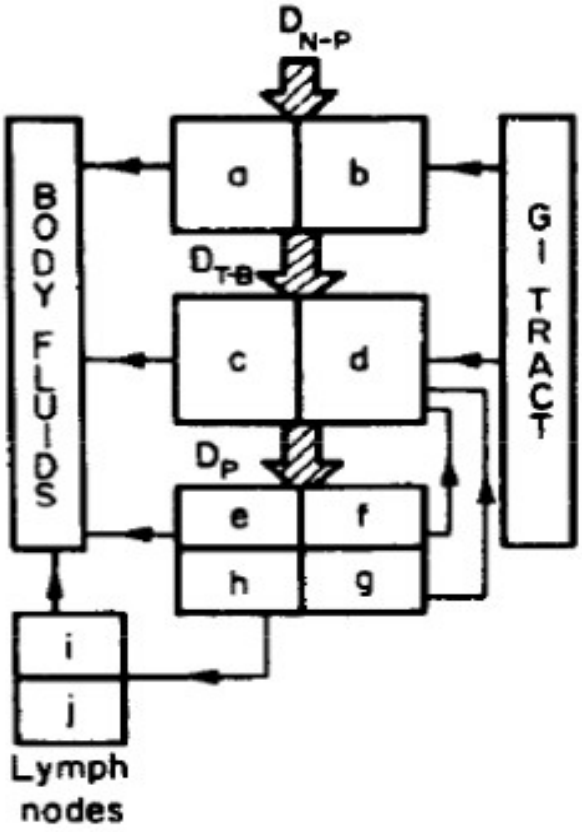

Figure 2.3. ICRP 30 Lung Model Compartments. Compartments *a, c, e, h* and *i* represent particles that are dissolved and absorbed into the bloodstream. Compartments *b, d, f* and *g* represent particles that are mechanically transferred out of the respiratory tract by means of the ciliary escalator and introduced into the gastrointestinal tract via swallowing. Taken from Cember and Johnson, 2008.

Particles deposition in the respiratory tract is primarily a function of the particle size, expressed in terms of the AMAD, and the region of the respiratory tract in which the particle deposits. It is noteworthy that particle deposition in the TB region is mostly constant for a particle size range from 0.2 to 10 μm (AMAD); particle deposition in the pulmonary region decreases linearly with increasing particle size, which particle deposition in the NP region increases linearly with increasing particle size. If particles of AMAD of 20 μm or greater are inhaled, it is assumed that all the deposition will occur in the NP region. The particle deposition model in ICRP 30 lung model is independent of the chemical composition of the particles. Using particles of 1 μm AMAD as an example, 30% of particles will be deposited in the NP region, 8% of particles will be deposited in the TB region; 25% in the P region, with the remainder exhaled (Cember and Johnson, 2008).

The ICRP 30 lung model defines different components that represent a distinct clearance mechanism that occurs in the respiratory tract. The NP region is represented by two compartments *a* and *b*; Compartment *a* represents the fraction of particles that are dissolved and directly absorbed into the bloodstream, while Compartment *b* represents the fraction of particles that are cleared into the GI tract via swallowing. The TB region is likewise represented by two compartments *c* and *d*; Compartments *c* and d represent similar mechanisms for the NP region respectively, though Compartment *d* specifically represents the ciliary escalator and swallowed into the GI tract. The pulmonary region is more interesting, in that it is represented by four different compartments. Compartment *e* represents dissolution of particles and introduction into the bloodstream, like Compartments *a* and *c*. Compartments *f* and *g* represent two different mechanisms that

reintroduce particles into the TB region and ultimately swallowed into the GI tract; Compartment *f* represents a mechanical transport, which is presumed to be unbalanced forces during respiratory excursions (act of breathing), and Compartment *g* represents the fraction of macrophages that will be introduced to the GI tract via the ciliary escalator. Compartment *h* represents the fraction of the inhaled particles that are transferred into the lymphatic system. The lymphatic system itself has two compartments *i* and *j*, which represent the fraction dissolved and absorbed into the bloodstream and the fraction that remain in the lymphatic system permanently. Table 2.5 shows physiological data for these compartments (Cember and Johnson, 2008).

Particles that are caught in the ciliary escalator and swallowed will ultimately pass through the GI tract and out of the body. Particles that are dissolved and absorbed into the bloodstream will be transported throughout the body. The ICRP 30 model has developed specific absorbed fractions from one organ to every other organ and tissue in the entire body, which is a function of the energy of the radiation. It is not necessary to calculate transfer compartment coefficients for each and every particle size; the ICRP 30 lung model does allow one to calculate a dose equivalent for a 1 μm AMAD particle, and then to rescale the dose for different size particles:

$$\frac{H_{50}(r)}{H_{50}(1\,\mu m)} = f_{NP}\frac{D_{NP}(r)}{D_{NP}(1\,\mu m)} + f_{TB}\frac{D_{TB}(r)}{D_{TB}(1\,\mu m)} + f_{P}\frac{D_{P}(r)}{D_{P}(1\,\mu m)} \quad , \tag{2.9}$$

where H_{50} are the committed dose equivalents to the 1 μm AMAD and the r μm AMAD particles, f_{NP}, f_{TB} and f_P are the fractions of radionuclide that are retained in the NP, TB or

P regions and can be found in Figure 2.5, and D_{NP}, D_{TB} and D_P are the deposition fractions of inhaled particles in the NP, TB and P regions respectively and listed in the graph in Figure 2.4 (Cember and Johnson, 2008).

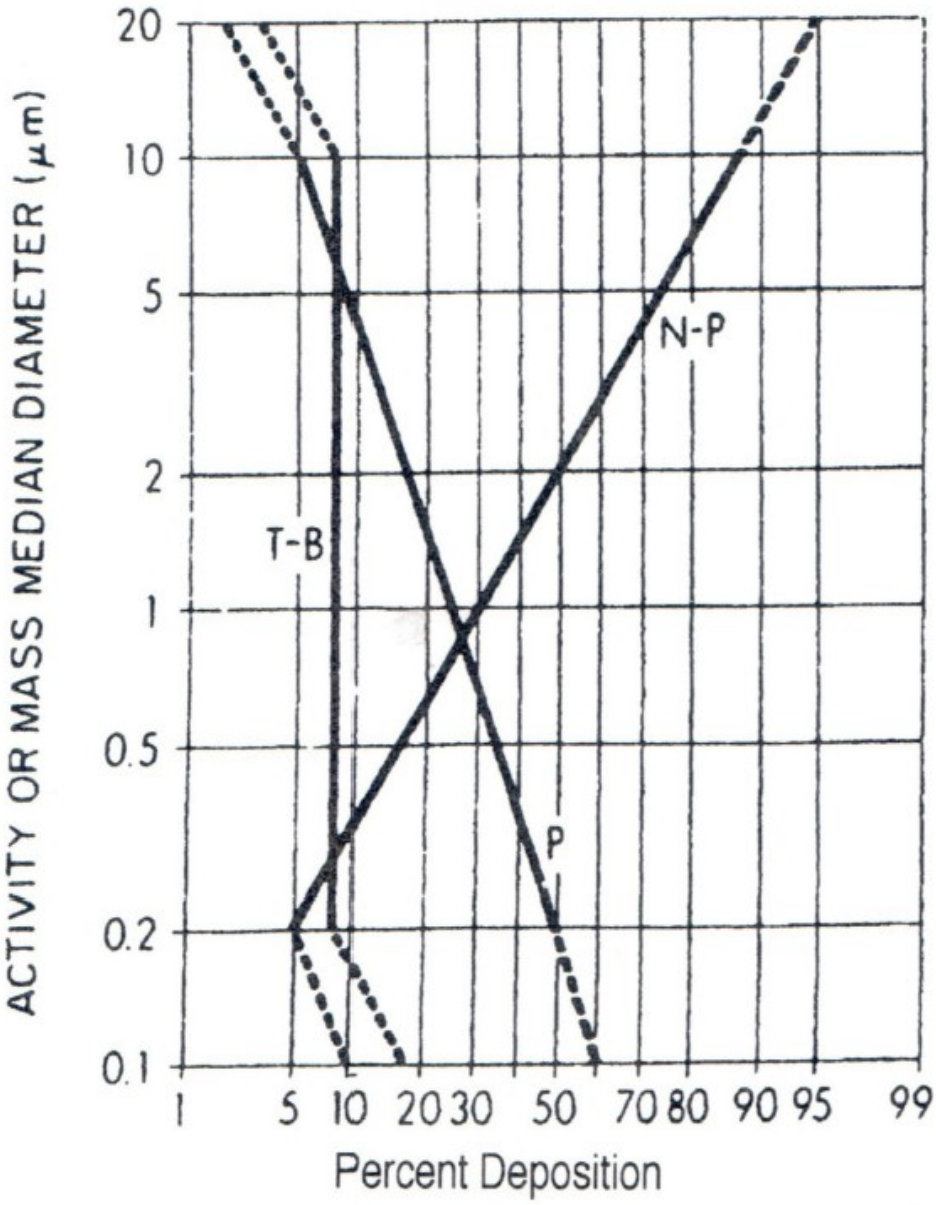

Figure 2.4. The ICRP 30 particle deposition model. Deposition in the TB region is relatively constant for the particle range of 0.2 to 10 μm. Particle deposition in the P region decreases as particle size increases, while particle deposition in the NP region increases with particle size, and it can be assumed that 100% of particles greater than 20 μm AMAD will deposit in the NP region. Taken from Cember and Johnson, 2008.

REGION	COMPARTMENT	CLASS					
		D		W		Y	
		T	F	T	F	T	F
NP (D_{NP} = 0.30)	a	0.01	0.5	0.01	0.1	0.01	0.01
	b	0.01	0.5	0.40	0.9	0.40	0.99
TB (D_{TB} = 0.08)	c	0.01	0.95	0.01	0.5	0.01	0.01
	d	0.2	0.05	0.2	0.5	0.2	0.99
P (D_P = 0.25)	e	0.5	0.8	50	0.15	500	0.05
	f	n.a.	n.a.	1.0	0.4	1.0	0.4
	g	n.a.	n.a.	50	0.4	500	0.4
	h	0.5	0.2	50	0.05	500	0.15
L	i	0.5	1.0	50	1.0	1000	0.9
	j	n.a.	n.a.	n.a.	n.a.	∞	0.1

Figure 2.5. This table lists the clearance half-lives (T) and the fraction of inhaled particles that will be transported to either the GI tract or the bloodstream (F) for each compartment of the ICRP 30 lung model. This table was developed for 1 μm AMAD particles. One can rescale internal doses for a different particle size. Taken from Cember and Johnson, 2008. There is a difference of notation between this table and Equation 2.9; the symbols f_{NP}, f_{TB} and f_P are used in Equation 2.9, which is denoted by F_{tissue} in this table.

CHAPTER 3 HAZARD EVALUATION

Uranium-Plutonium Glovebox Hazard Analysis

The US NRC NUREG/CR-6410, Nuclear Fuel Cycle Facility Accident Analysis Handbook and US DOE HDBK 3010-94, "Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Accidents" describe a safety analysis for a variety of nuclear fuel cycle facilities, giving several examples of how to perform a safety evaluation for eight sample accidents. Questions to be asked in a hazard evaluation include:

1. What hazardous materials are present and under what conditions? What hazards are present? What process hazards are present at the facility? What chemical hazards are present at the facility?
2. What accidents can happen that result in a release of hazardous materials? What negative impact could there be on radiation workers, non-radiation workers, the general public, the environment and other nearby facilities?
3. What engineering and process controls are present to prevent or mitigate such accidental releases?
4. What modifications could be made to the engineering and process controls to either prevent or further mitigate accidental releases of hazardous materials?
5. Is further analysis required for any such accidental releases? If so, what analysis is needed?

This safety analysis is being conducted for the Research and Innovation in Science and Engineering (RISE) Complex, located at 1999 Alvin Ricken Drive in Pocatello, ID. The RISE Complex and its location are shown in Figures 3.1a and 3.1b. The RISE Complex is a nuclear research facility primarily for the Idaho State University Department of Nuclear Engineering and Health Physics, though it is open for other academic departments. The primary research focuses at this moment in time at the RISE Complex are development of novel radiation detection technology and nuclear fuel research. The nuclear fuel research requires the use of a glovebox to isolate uranium from the occupational environment and the external environment. The nuclear fuel research poses potential radiological hazards that would be realized in the event of an accidental release of uranium.

Currently, the UO_2 Crystal Project is interested in the structural and materials properties of UO_2 solids, including but not limited to heat transport, mechanical properties, and phonon structure. Future studies of UO_2 single crystals will include neutronics studies, such as the effects of fission products on the structural and materials properties of crystalline nuclear fuel; hence, a criticality safety analysis is not being pursued in this safety analysis.

Figure 3.1a. The RISE Complex. Taken from the Idaho State University website.

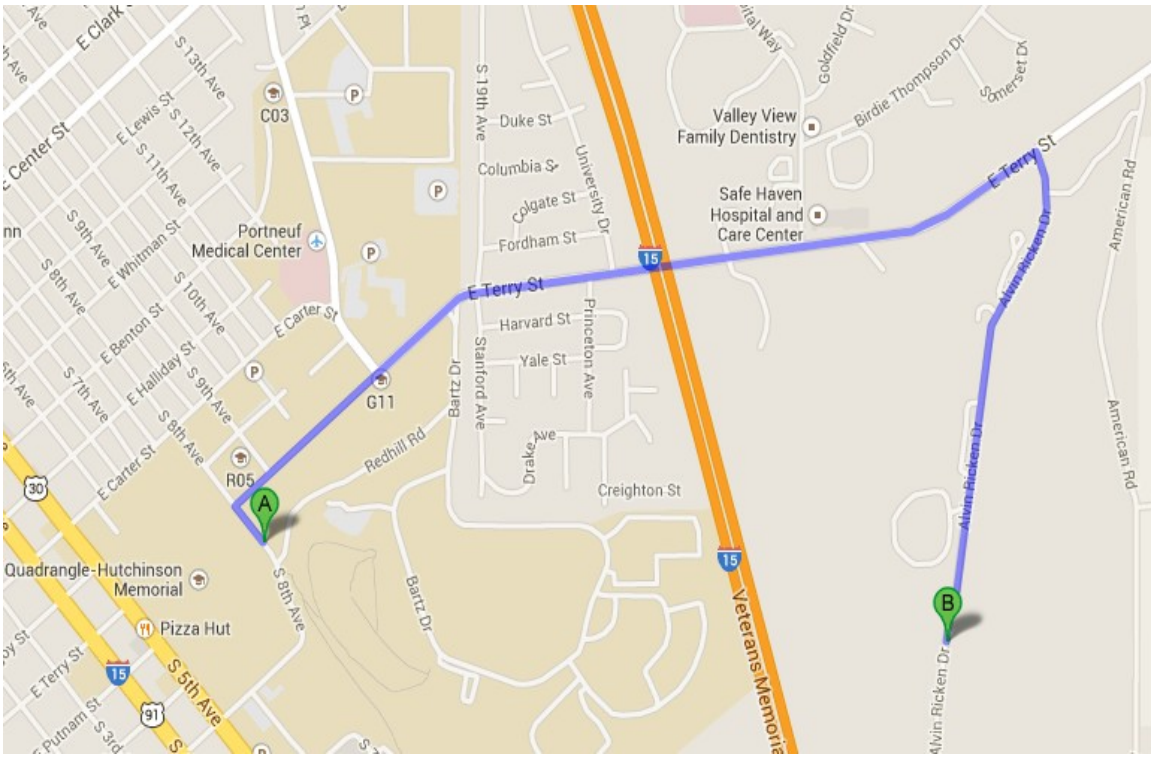

Figure 3.1b. The location of the RISE Complex (marker B) in relation to the Idaho State University campus (marker A). Taken from Google Maps.

Hazardous Materials Present in the RISE Complex

The principle hazardous material present is UO_2. The uranium is depleted; hence, there is no criticality concern arising from the UO_2 itself. The primary hazards associated with uranium generally are nephrotoxicity, external radiation from x-ray and gamma radiation, and alpha radiation in the case of internal exposures. When nuclear fuels are enriched in uranium-233, uranium-235 or plutonium-239, there is then also the possibility of an accidental criticality. However, fissile nuclides currently are not being used at this time.

Though the best-known hazard of depleted UO_2 is that it is radioactive, the UO_2 hazard of most concern its nephrotoxicity. If UO_2 is sufficiently free with substantial fractions less than 5 μm AMAD and sufficient energy is added to that powder to cause it to become dispersable in the local atmosphere, it is quite possible that a significant internal dose could result if one inhales air that is contaminated with UO_2 powder. Depending on the size of the UO_2 particle, some UO_2 can deposit into the lungs while the remainder will pass through the digestive and excretory systems. What UO_2 is not inhaled or ingested will possibly cause an external dose equivalent to the worker. The radioactive decay chain form uranium-238 is shown in Table 3.1.

Processes Involving Uranium

The process of forming the UO_2 crystals, presents a set of physical hazards. The UO_2 material that is being used for this project are sintered pellets of depleted UO_2 pellets that were originally made for the Fast Flux Test Facility (FFTF) at Pacific Northwest National Laboratory (PNNL) and was shipped to the Materials and Fuels Complex (MFC) at the Idaho National Laboratory (INL) in the 1970s. The sintered UO_2 pellets are first loaded into the Glovebox (see Figures 3.2 through 3.5). These pellets are reduced into a fine power using a custom-built hammer-mill (seen in Figure 3.4), generating a fine powder of UO_2 (diameter on the order of hundreds of microns). This powder is sifted in a sieve (seen in Figure 3.4) to remove larger grained particles (<150 microns), and the remaining smaller particles are pressed in a set of dyes to form a new pellet. Figure 3.2 shows the Glovebox; Figure 3.3 shows the Glovebox antechamber; Figure 3.4 shows the hammer-mill and the sifter, and Figure 3.5 shows the pellet-presser.

The pellet, which typically has a diameter of 1.5 inches is moved to an adjacent laboratory, the Crystal Growth Laboratory, and is initially sintered in a horizontal furnace. The purpose of sintering is to provide a skin over the UO_2 pellet; the horizontal furnaces are shown in Figure 3.6. The sintered pellet is then placed in a crystal furnace (which is of a Bridgman process), and the powder is RF-heated to form a single crystal of UO_2. The crystal furnace, seen in Figure 3.7 is a modified Bridgman-type crystal-growth chamber. There are two sleeves that comprise the Crystal Furnace: the outer sleeve that is visible, and an inner sleeve. There is a quartz tube contained inside the inner sleeve which

is encircled by RF coils; it is in this quartz tube where the pelletized UO_2 powder is RF-heated to form the single crystal. The Bridgman crystal growth technique requires different pressures for different materials; oxygen is used as the forming gas in this process. The part of the inner sleeve that is outside the quartz tube is cooled with nitrogen gas. The outer sleeve is water-cooled. The Bridgman crystal growth process is discussed in Appendix A. The crystal furnace is shown in Figure 3.7.

Symbol	Radiation	Half-life	Decay Product
U-238	alpha	4.46×10^9 years	Th-234
Th-234	beta	24.1 days	Pa-234
Pa-234	beta	1.17 min	U-234
U-234	alpha	2.47×10^5 years	Th-230
Th-230	alpha	8×10^4 years	Ra-226
Ra-226	alpha	1,602 years	Rn-222
Rn-222	alpha	3.82 days	Po-218
Po-218	alpha	3.05 min	Pb-214
Pb-214	alpha	27 min	Bi-214
Bi-214	beta	19.7 sec	Po-214
Po-214	beta	1 msec	Pb-210
Pb-210	alpha	22.3 years	Bi-210
Bi-210	beta	5.01 days	Po-210

Table 3.1. The Uranium-238 decay chain. Taken from the New York State Department of Health, Bureau of Environmental Radiation Protection

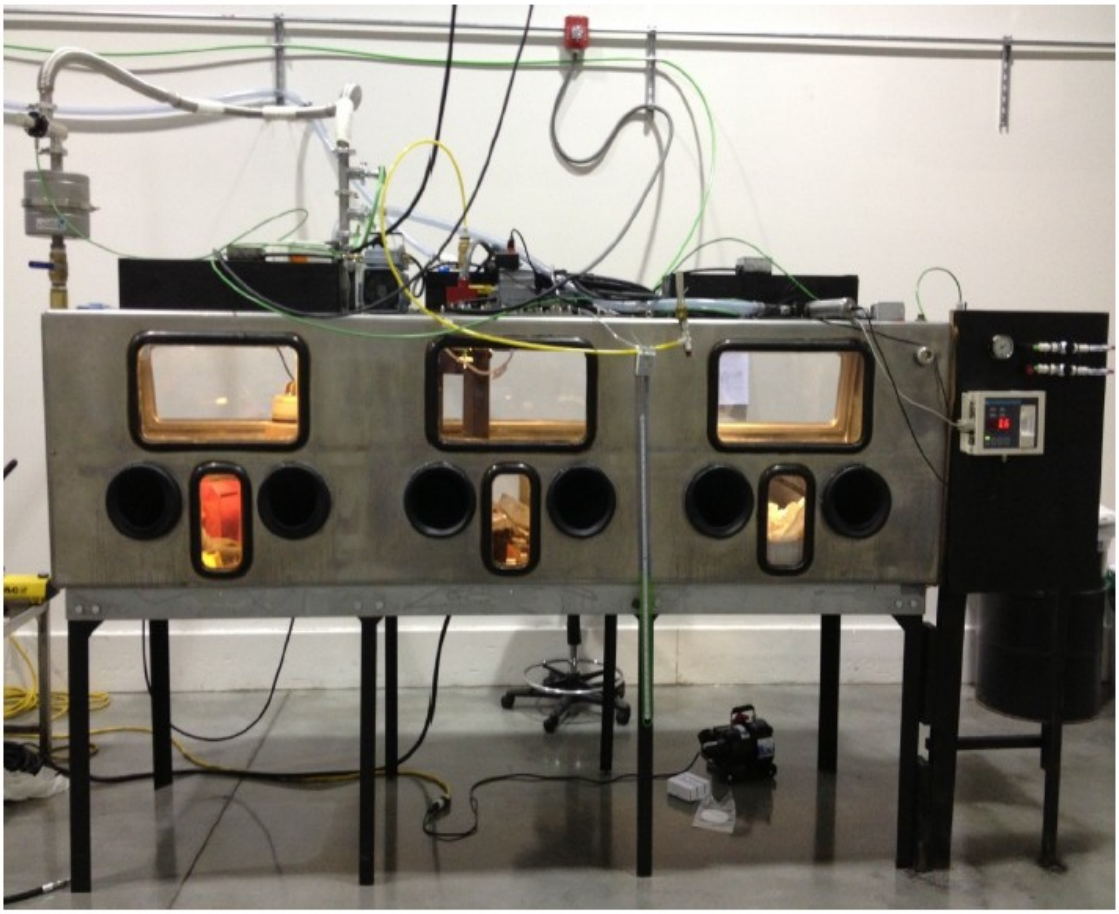

Figure 3.2. The large-volume Uranium glovebox in which most work with depleted uranium dioxide is performed. One can see the nitrogen plumbing and the vacuum system, and the HEPA filter that was installed to catch uranium dioxide particles.

Figure 3.3. The antechamber in which materials and/or tools are transferred into and out of the glovebox. The inside of the glovebox is always to be kept under negative pressure during work involving the glovebox. The antechamber is always pumped to a vacuum when loading materials and/or tools into the glovebox, and as a means to secure the

glovebox. If an object or material is being moved into the glovebox, the item is loaded into the antechamber and sealed shut, then the antechamber is taken to a vacuum state, where it can be loaded into the glovebox itself. Conversely, if an object is being moved out of the glovebox, then, the antechamber is taken to room pressure, so that the item can be removed. Items leaving the glovebox are decontaminated or are taken to the radiological waste receptacle.

Figure 3.4. The pellet crusher. Sintered pellets of the dimensions that would be found in nuclear fuel rods provided by the Materials and Fuels Complex (MFC) of the Idaho National Laboratory (INL) are introduced into the pellet crusher and reduced to a fine

powder. Also seen in this picture is a mechanical sieve that will filter out any uranium dioxide particles greater than 100 μm.

Figure 3.5. The pellet presser. The crushed up uranium oxide powder is pressed into a pellet using this press. There is a hydraulic pump located on the outside of the glovebox that is used to apply the pressure to form the pellet.

(a) (b)

Figure 3.6. Horizontal Furnaces used to scinter UO_2.

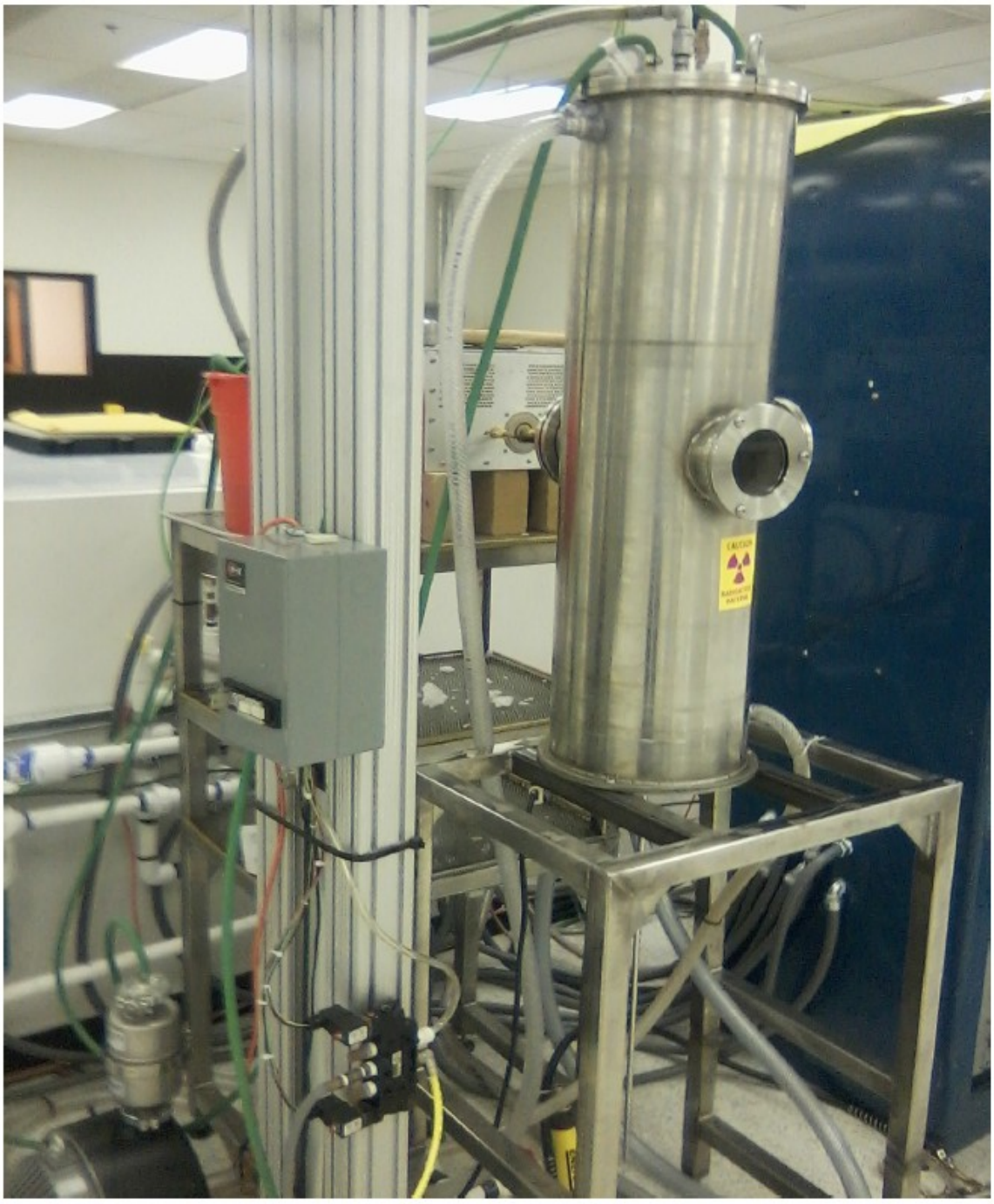

Figure 3.7. The Crystal Furnace. This is the Bridgman-type RF-heated induction chamber where the pellet is crystallized.

Potential UO_2 Release Accidents

It should be noted that every accident scenario presented in this section will require further analysis, and the steps in the analysis will be described- the goal to which this thesis is dedicated. It is also worthwhile to consider a few accident scenarios that currently are not of concern, at this moment. Some accident scenarios that have been ruled out of consideration, at least for the time being include:

A: Explosions inside the glovebox: The only organic materials present in the glovebox are steric acid, used to line the dyes that form UO_2 pellets, and acetone used as a solvent. Furthermore, the atmosphere inside the glovebox is a nitrogen atmosphere; hence, explosions are not of concern. Should there ever be an occasion where it was proposed that oxygen gas (or even air) be introduced into the glovebox, then an explosive situation would be present and another safety analysis would need to be performed before this could be allowed to conceive. It is conceivable that metallic uranium and plutonium that can react in the presence of air (and especially oxygen) may be processed in the glovebox in the future; hence, this is the reason why nitrogen is used in the glovebox.

B. Criticality: Since it is the case that at present in time, only depleted UO_2 is used inside the glovebox; hence, there is no current need to evaluate criticality accidents. The studies that UO_2 crystals are currently being undertaken are independent of the isotopic concentrations of uranium. Future plans for the uranium oxide project call for growing uranium single-crystals containing uranium-235, uranium-233 and/or plutonium-239 in

concentrations sufficient for criticality would be fabricated; hence, in the future a criticality analysis will be needed.

C. Internal exposures from wounds: There are no sharp edges present inside the glovebox, in accordance with the recommendations of the plutonium glovebox standards. Sharp objects are used to access the sintered UO_2 pellets. The presence of sharp objects and sharp edges allows for the possibility that a glove could be torn, and give opportunity for UO_2 dust to escape into the glovebox laboratory. It would be prudent to simply not have any sharp objects and sharp edges inside the glovebox; this however, is unavoidable.

D. Tears to the gloves: Even in the absence of objects objects, gloves can tear apart if part of a glove becomes entangled in the pellet-presser or another piece of equipment and the glovebox worker pulls the glove away with a force sufficient to cause a glove to fail. This scenario is most probable of all accident scenarios.

The four scenarios that can result in accidental releases of radioactive material from the glovebox and under consideration in this thesis are breaches to the glove, breaches to the forearm cover and a breach of confinement involving the antechamber. The accident analysis will calculate inhaled and external dose equivalents for several concentrations of UO_2, PuO_2, U-metal and Pu-metal in air: 1 pCi/g, 33 pCi/g, 67 pCi/g, 100 pCi/g, 10^4 pCi/g and 10^6 pCi/g; the rationale for selecting these air concentrations will be explained in Section 4.3 of this thesis. Dose equivalents were calculated for a worker at the glovebox and another worker at some distance away from the glovebox,

either at the far end of the Glovebox Laboratory or in the Crystal Growth Laboratory. The dose equivalent will depend on the amount of UO_2 released into the glovebox laboratory, the particle size distribution, and the amount of UO_2 dust that is inhaled, with the remaining UO_2 dust contributing to the external dose equivalent.

Because of the (shielded) sharp objects used to open the rods containing the sintered depleted UO2 pellets, it is possible that a glove could tear, and a puncture could occur. If a glove failed the same moment the glovebox went into positive pressure, UO_2 dust could escape through the glove into the glovebox laboratory.

Engineering and Process Controls Available

HEPA (High Efficiency Particulate Air) filters are frequently the final line of defense to prevent the release of radioactive particulates and aerosols into the air. HEPA filters have four different mechanical mechanisms by which they capture particulates; inertial impaction, interception, gravitational settling, and diffusion and Brownian motion. Inertial impaction occurs when the presence of a filter fiber forces the gas flow to curve around the filter. The ability of a particle (or aerosol) to successfully navigate around the fiber depends on the size and the velocity of the particle. Larger particles, greater than 1 μm AED (Aerodynamic Equivalent Diameter) directly impact the surface of the HEPA filter. Interception is the process in which a particle comes within one particle radius of a fiber; the particle comes into contact with the fiber and is captured due to its finite size.

Interception is the only HEPA filter collection process that is independent of the gas flow through the filter. Gravitational settling is the process of the drag force working against the gravitational force, thus causing the particle to reach a terminal settling velocity. Terminal settling velocity increases rapidly with particle size. Brownian motion of small particles is sufficient to enhance the probability of their collision with a filter fiber; this is a special case of diffusion to the surface of the filter fiber. Brownian motion and diffusion are connected; at any point particles are undergoing Brownian motion, but due to the particle concentration gradient, there are always particles diffusion from regions of higher concentration to regions of lower concentration of said particle. There is however, a diameter of particle that has the highest probability of penetrating through the HEPA filter, which typically is from 0.1-0.2 μm AED for a standard 2-foot by 2-foot HEPA filter with a flow rate of 1000 cfm (cubic feet per minute) and pressure of 1-inch WG (water gauge) - which is standard in the United States. NUREG-CR 6410 states that most good modern HEPA filters made from glass fiber filters can withstand pressures of up to 80-inches WG, but a loss of binder failure can occur at pressures of 10-inches WG (NUREG-CR 6410; Hinds, 1999).

An adaptive-algorithm mass-flow controller is currently being programmed and constructed to regulate the pressure inside the glovebox. The current procedure for regulating the pressure inside the glovebox is a manual knob attached to a pressure gauge. The mass-flow controller should be capable of regulating the glovebox pressure such that the pressure inside never goes positive and should not exceed 8-inches water gauge (WG); since binder failure can occur at 10 inches WG, a limit of 8 inches WG

provides a margin of error to prevent binder failure (NUREG-CR 6410).

Visual inspection of the HEPA filters would partly enable a determination of the binder quality. However, this would not yield any information about the quality of the filter matrix, nor would this give any information about UO_2 loading in the HEPA filter. One improvement that could be made to the inspection regime of the HEPA filters would to survey the HEPA filters with an appropriate beta and/or gamma sensitive survey instrument. Moreover, any breakthrough of the HEPA filters would result in triggering the alpha-detector which is in the vacuum line after the HEPA filter, which would immediately shut off the entire system.

CHAPTER 4 Computational Methods

The execution of the safety analysis that is the subject of this thesis involved two different methods to calculate external and inhaled doses from potential accidental releases of radioactive materials from the glovebox. The first method was to use the freely distributed radiological assessment code RESRAD-BUILD, that is maintained by the Radiation and Chemical Risk Management program area of the Environmental Science Division of Argonne National Laboratory. RESRAD-BUILD is one of several codes that constitutes the family of RESRAD codes. The second method was a set of "hand calculations" described in the DOE HDBK-3010-94, "Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities" and the NRC NUREG-CR 6410, "Nuclear Fuel Cycle Facility Accident Analysis Handbook."

4.1 Description of Physics of the RESRAD-BUILD Code

RESRAD-BUILD is capable of calculating transport of radionuclides in up to three rooms, which is shown in Figure 4.1. Rooms 1 and 3 are assumed to not be connected; therefore, any air movements between rooms 1 and 3 cannot be programmed into any RESRAD simulations. The air flow rates- in units of m^3/hr between rooms Q_{ji}, obeys conservation of mass, and steady-state conditions are assumed; hence,

$$\sum_{\substack{j=1 \\ j\neq i}}^{3} Q_{ji} = \sum_{\substack{j=1 \\ j\neq i}}^{3} Q_{ij} \tag{4.1a}$$

and it is seen that

$$Q_{10}+Q_{12}=Q_{01}+Q_{21}$$

$$Q_{20}+Q_{21}+Q_{23}=Q_{02}+Q_{12}+Q_{23}; \tag{4.1}$$

$$Q_{30}+Q_{23}=Q_{03}+Q_{23}$$

summing equations 4.1b, one obtains

$$Q_{10}+Q_{20}+Q_{30}=Q_{01}+Q_{02}+Q_{03}. \tag{4.1c}$$

It is concluded that the total airflow into a particular room is equal to the total airflow leaving said room. Air-exchange rates λ^a_i and λ^b_i can be defined for a particular room and the building respectively from the airflow rates (RESRAD_BUILD Manual):

$$Q_{10}+Q_{20}+Q_{30}=Q_{01}+Q_{02}+Q_{03} \quad . \tag{4.2a}$$

$$\begin{aligned} \lambda_i^a &= \frac{\sum_{\substack{j=1 \\ j\neq i}}^{3} Q_{ji}}{V_i} = \frac{\sum_{\substack{j=1 \\ j\neq i}}^{3} Q_{ij}}{V_i} \\ \lambda_0^a &= \frac{\sum_{j=1}^{3} Q_{i0}}{\sum_{j=1}^{3} V_i} = \frac{\sum_{j=1}^{3} Q_{0i}}{\sum_{j=1}^{3} V_i} \end{aligned} \tag{4.2b}$$

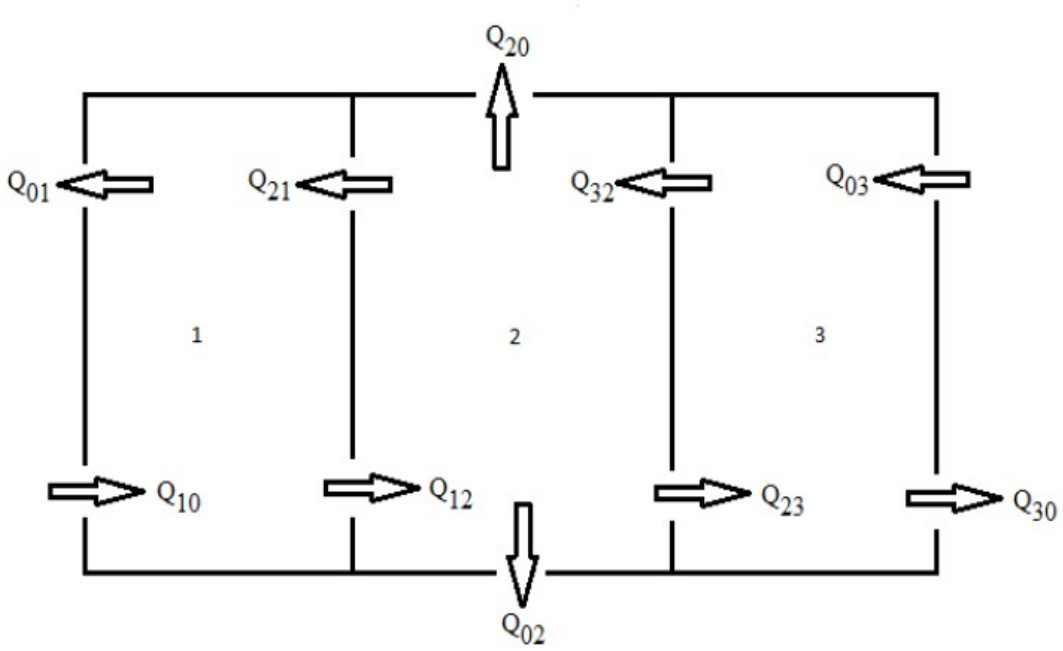

Figure 4.1. Schematic describing airflow rates in RESRAD-BUILD. Rooms 1 and 3 do not connect under any circumstance in RESRAD-BUILD; therefore Q13 = Q31 = 0.

Indoor Air Concentration Model

The concentration of radionuclide *n* in room *ι* at any moment in time *t* is given by

$$V_i \frac{d\,C_i^n(t)}{dt} = \lambda_{rn} V_i C_i^{n-1}(t) + \sum_{\substack{j=0 \\ j \neq i}}^{3} [Q_{ji} C_j^n(t)] - C_i^n(t) \sum_{\substack{j=0 \\ j \neq i}}^{3} Q_{ij} + I_i^n(t) - S_i^n(t) \quad , \qquad (4.3)$$

where,

$C_i^n(t)$ is the concentration of radionuclide *n* (daughter *n* in a radioactive decay series) in room *ι* at time *t* in units of pCi/m^3;

λ_{rn} is the radioactive decay constant for radionuclide *n* (with *n-1* representing the radioactive parent of radionuclide *n*)

$I_i^n(t)$ is the injection of radionuclide *n* into the air, and

$S_i^n(t)$ is the sink term, that represents loss of radionuclide *n* (RESRAD-BUILD Manual).

The term on the left side, $V_i \frac{d\,C_i^n(t)}{dt}$, represents the net instantaneous accumulation/depletion of radionuclide *n* within a room of volume V_i. The first term on the right side, $\lambda_{rn} V_i C_i^{n-1}(t)$, represents the generation of radionuclide *n* via

radioactive decay of parent *n-1*. The second term, $\sum_{\substack{j=0 \\ j\neq i}}^{3} [Q_{ji} C_j^n(t)]$, represents the inflow of radionuclide *n* from other rooms containing said radionuclide. The third term, $C_i^n(t) \sum_{\substack{j=0 \\ j\neq i}}^{3} Q_{ij}$, represents the loss of radionuclide *n* to other rooms (RESRAD-BUILD Manual).

The injection term $I_i^n(t)$ itself is the sum of two terms

$$I_i^n(t) = I_{Si}^n(t) + I_{Ri}^n(t) \quad , \tag{4.4a}$$

where $I_{Ri}^n(t)$ denotes the resuspension term which depends on the resuspension rate λ_R and the deposition velocity, and $I_{Si}^n(t)$ is a source-dependent terms that represents direct injection of radionuclide *n*, mechanical removal (erosion rate) and must be specifically evaluated for each source and operational scenario. A more detailed consideration of the resuspension term $I_{Ri}^n(t)$ requires an explanation of the particle deposition model that is used in the RESRAD-BUILD code. The differential equation used to calculate particle deposition is

$$A_i \frac{d\, C_{di}^n(t)}{dt} = v_d\, A_i C_i^n(t) - \lambda_{rn} A_i C_{di}^n(t) - \lambda_R A_i C_{di}^n(t) \quad , \tag{4.4b}$$

where $C_{di}^{n}(t)$ denotes the surface concentration of radionuclide *n* at time *t* on surface *ı* in units of pCi/m^2

$C_{i}^{n}(t)$ is the concentration of radionuclide *n* in air in at time *t* on surface *ı* in units of pCi/m^3

v_d is the deposition velocity, in units of m/hr

λ_R is the resuspension factor

A_i is the surface area in units of m^2

The left-hand term denotes the total instantaneous accumulation/depletion of radionuclide *n* on given surface *ı*. The first term on the right-hand side denotes the addition of mass due to dust deposition, while the middle and last terms denote depletion of radionuclide *n* via radioactive decay and resuspension respectively. Using the steady-state assumption, one obtains

$$v_d C_{di}^{n}(t) - \lambda_{rn} C_{di}^{n}(t) - \lambda_R C_{di}^{n}(t) = 0 \quad ,$$

or

$$C_{di}^{n}(t) = \left(\frac{v_d}{\lambda_{rn} + \lambda_R}\right) C_{i}^{n}(t) \quad . \tag{4.4c}$$

The surface concentration is directly related to the resuspension term $I_{Ri}^{n}(t)$:

$$I_{Ri}^{n}(t)=\lambda_R A_i C_{di}^{u}(t) \quad ,$$

or

$$I_{Ri}^{n}(t)=\left(\frac{\lambda_r}{\lambda_{rn}+\lambda_R}\right)\lambda_{di} V_i C_i^{n}(t) \quad , \tag{4.4d}$$

with the deposition rate $\lambda_{di}=v_d A_i / V_i$. The source-dependent term $I_{Si}^{n}(t)$ represents direct injection of radionuclide *n* into room *ι*, and is given as

$$I_{Si}^{n}(t)=\frac{10{,}000\, ARF\, \epsilon\, \rho_{bs} C_s^{n}(t)}{24} \quad , \tag{4.4e}$$

where: *ARF* is the airborne release fraction, and is explained in the discussion of the hand-calculations

ε is the erosion rate, and is in units of cm/day

A_s is the effective surface area, in units of m^2

ρ_{bs} is the bulk density of the source material, in units of g/cm^2

$C_s^{n}(t)$ is the air concentration of the source material radionuclide *n* at time *t*, in units of pCi/g

10,000 is a conversion factor between cm^2 and m^2

24 is a conversion factor between hours and days.

The sink term $S_i^n(t)$ represents the loss of airborne radionuclide *n* due to radioactive decay and surface deposition, and is represented mathematically as (RESRAD-BUILD Manual):

$$S_i^n(t)=\lambda_{rn}V_iC_i^n(t)+\lambda_{di}V_iC_i^n(t) \quad \text{, or}$$

$$S_i^n(t)=(\lambda_{rn}+\lambda_{di})V_iC_i^n(t) \quad . \tag{4.4f}$$

Substituting Equations 4.4 into Equation 4.3, one obtains

$$V_i\frac{d\,C_i^n(t)}{dt}=\lambda_{rn}V_iC_i^{n-1}(t)+\sum_{\substack{j=3\\ j\neq i}}^{3}[Q_{ji}C_j^n(t)]$$
$$-C_i^n(t)\sum_{\substack{j=3\\ j\neq i}}^{3}Q_{ij}+[\frac{\lambda_R\lambda_{di}}{\lambda_{rn}+\lambda_r}-(\lambda_{rn}+\lambda_{di})]V_iC_i^n(t)+I_i^n(t) \tag{4.5}$$

Using the steady-state approximation, conservation of mass and taking into consideration all three rooms, a matrix formulation of the indoor air quality model can be obtained (RESRAD-BUILD Manual):

$$A \times x = b \quad , \tag{4.6a}$$

where

$$x = \begin{bmatrix} C_1^n(t) \\ C_2^n(t) \\ C_3^n(t) \end{bmatrix} \quad , \tag{4.6b}$$

$$b = \begin{bmatrix} I_{S1}^n(t) + Q_{01} C_0^n(t) + \lambda_{rn} V_2 C_1^{n-1}(t) \\ I_{S2}^n(t) + Q_{02} C_0^n(t) + \lambda_{rn} V_2 C_2^{n-1}(t) \\ I_{S3}^n(t) + Q_{03} C_0^n(t) + \lambda_{rn} V_3 C_3^{n-1}(t) \end{bmatrix} \quad , \tag{4.6c}$$

$$A = \begin{bmatrix} A_1 & -Q_{21} & 0 \\ -Q_{12} & A_2 & -A_{32} \\ 0 & -Q_{23} & A3 \end{bmatrix} \quad , \tag{4.6d}$$

and

$$\begin{aligned} A_1 &= \left(\lambda_{rn} + \lambda_{d1} - \frac{\lambda_r \lambda_{d1}}{\lambda_{rn} + \lambda_R}\right) V_1 + \sum_{\substack{j=0 \\ j \neq 1}}^{3} Q_{j1} \\ A_2 &= \left(\lambda_{rn} + \lambda_{d2} - \frac{\lambda_r \lambda_{d2}}{\lambda_{rn} + \lambda_R}\right) V_2 + \sum_{\substack{j=0 \\ j \neq 2}}^{3} Q_{j2} \\ A_3 &= \left(\lambda_{rn} + \lambda_{d3} - \frac{\lambda_r \lambda_{d3}}{\lambda_{rn} + \lambda_R}\right) V_3 + \sum_{\substack{j=0 \\ j \neq 1}}^{3} Q_{j3} \end{aligned} \quad . \tag{4.6e}$$

One can consult Appendix a: Indoor Air Quality Model for further details of this physics.

The total committed effective dose equivalent (CEDE) is calculated as

$$H_i^n(t) = f_{inside} f_i t_E < C_i^n(t) > IR\, DCF^n \quad , \tag{4.7}$$

where $H_i^n(t)$ is the CEDE

f_{inside} is the fraction of the time spent inside for the exposure

f_i is the fraction of the time spent in room $\imath$

t_E is the duration of exposure, in days

IR is the inhalation rate, in units of m^3/s

$< C_i^n >$ is the average concentration of radionuclide n over the exposure duration t_E in units of pCi/m3

DCF^n is the dose conversion factor, in units of mrem/pCi .

Calculation of the External Dose

RESRAD-BUILD has several methods to calculate external doses, including external doses from contaminated volume sources, line sources, point sources, and from contaminated dusts in air. The nature of potential accidental radioactive exposures in the

RISE Complex will most probably arise from escaped UO_2 powders, the contaminated dust scenario is the one that is most pertinent in our case. The external dose that arises from radioactive dusts in the air is calculated by

$$D^n_{i,subm}(t) = (ED/365) F_{inside} F_i < C^n_i(t) > DCF^n_i \quad , \tag{4.8}$$

where $D^n_{i,subm}(t)$ is the external dose arising from submersion in a radioactive dust contaminated cloud from radionuclide *n* in compartment *i* at time *t* in units of rem

ED is the exposure duration in days and 365 are the days of the year

F_{inside} is the fraction of the time spent inside

F_i is the fraction of the time spent in compartment *i*

$< C^n_i(t) >$ is the average concentration of radionuclide *n* over the exposure duration *ED* starting at time *t* in compartment *i* in units of mrem/year per pCi/m^3. This is calculated as the integrated concentration divided by the integrated time (RESRAD-BUILD Manual).

The committed effective dose equivalent (CEDE) for all rooms is the sum of each $H^n_i(t)$. The deep dose equivalent is the sum of of each $D^n_{i,subm}(t)$. The total effective dose equivalent (TEDE) is the sum of the CEDE and the DDE (RESRAD-BUILD Manual).

Particle Size Distributions in RESRAD-BUILD

As mentioned previously, RESRAD-BUILD does not directly accept inputs for particle sizes. If one is interested in computing doses as a function of particle sizes, then one must use a relation between the particle size and deposition velocity. Deposition velocity v_d itself is formally defined as

$$v_d = \frac{amount\ of\ radionuclide\ deposited\ \ per\ square\ centimenter\ \ per\ second}{concentration\ of\ \ radionuclide\ per\ cubic\ centimeter\ above\ surface} \quad . \tag{4.9}$$

The terms in definition of deposition velocity are Common units for deposition velocity are cm/s and m/s, with the latter being the RESRAD-BUILD input, and the deposition velocity incorporates all relevant physical and chemical properties of a radionuclide into a single observable parameter. Deposition velocity is most practical for particle 10 μm and smaller, as it is in this region that the settling velocity is negligible (Eisenbud and Gesell, 1997; Zannetti, 1990).

4.2 RESRAD-BUILD User Interface

Dose equivalents resulting from an accident resulting in the release of radioactive materials were directly calculated, and simulations were run using RESRAD-BUILD. RESRAD-BUILD is frequently used in decommissioning work, but RESRAD-BUILD is well-suited for assessing doses from releases of radioactive materials from glove boxes

and fume hoods. The RESRAD-BUILD input interface has the appearance shown in Figure 4.2.

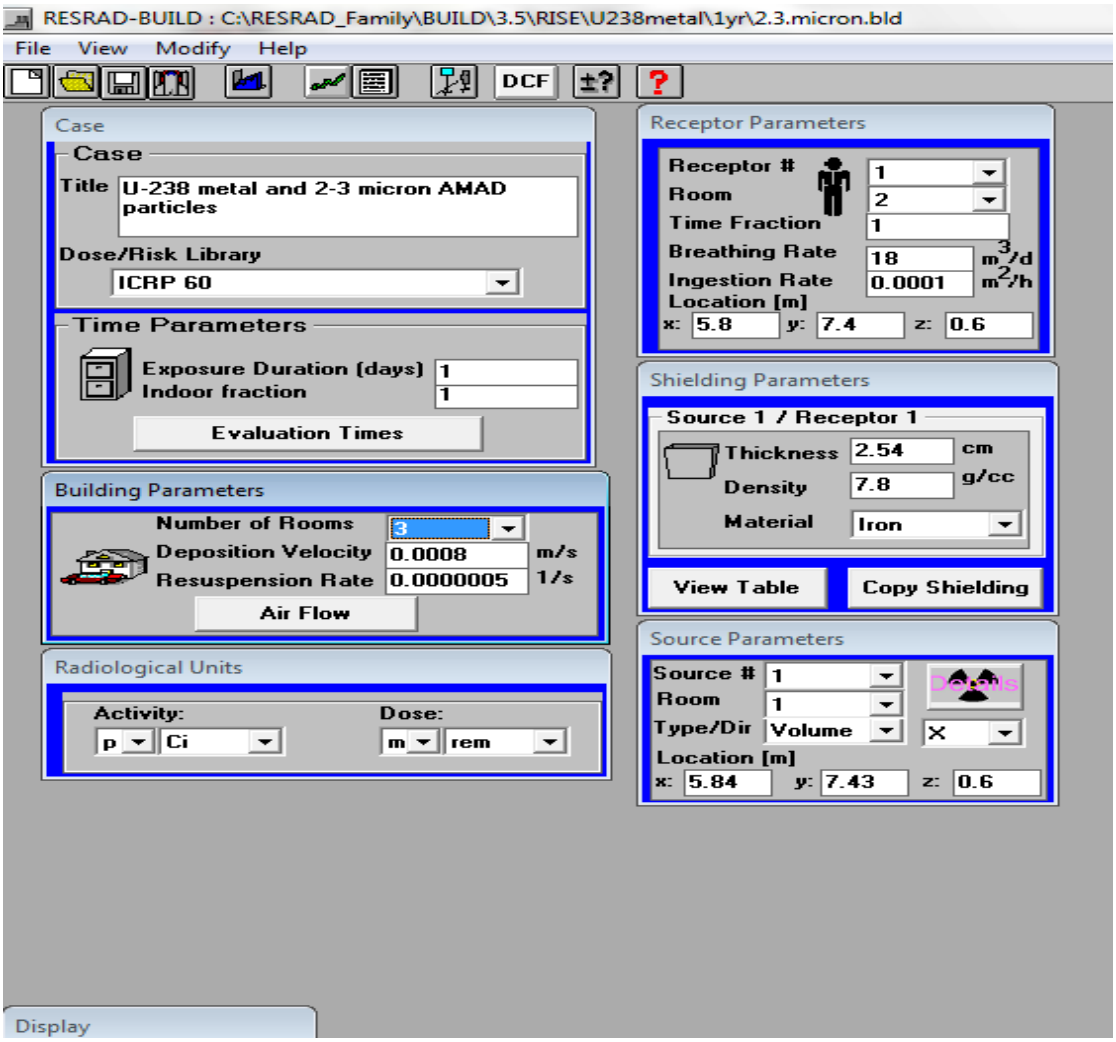

Figure 4.2. RESRAD-BUILD input interface.

Several windows in RESRAD-BUILD can be expanded: the Source Parameters window wherein the species of radionuclide(s), radionuclide concentration, and material density can be defined in Figure 4.3.

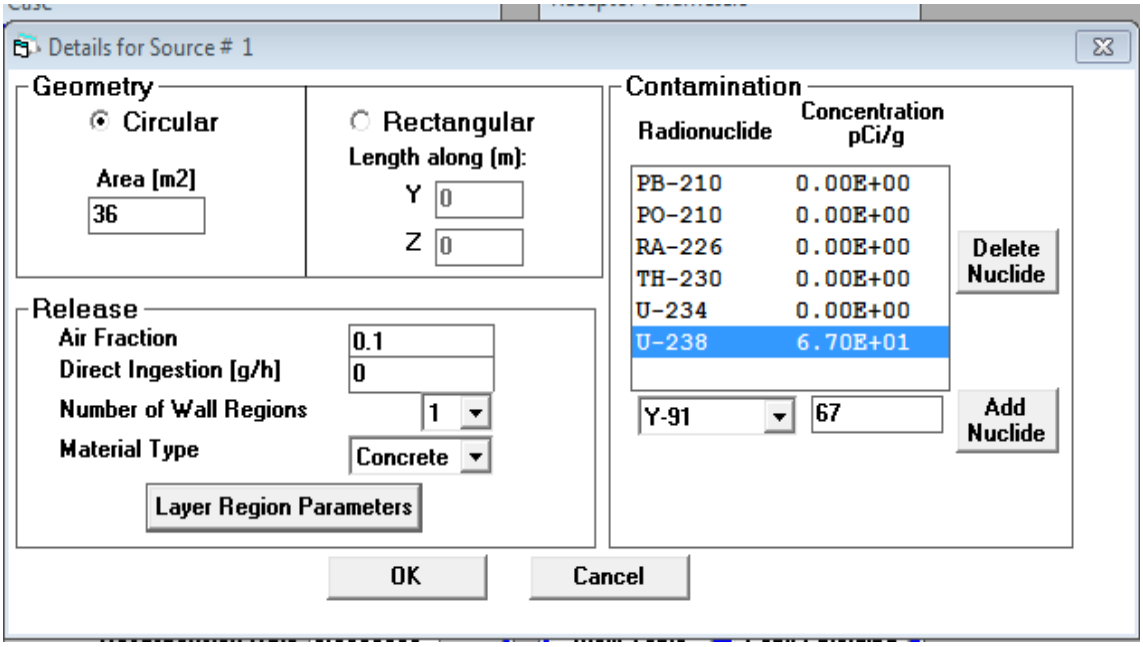

Figure 4.3a. Radionuclide Definition Window.

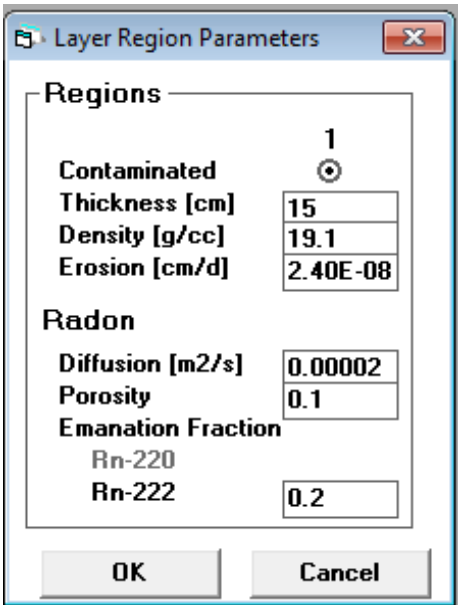

Figure 4.3b. The material properties of the radionuclide under study can be entered into the "Layer Region Parameters" window, which allows for the case of concrete the escape of radon.

Another window that can be opened up in RESRAD-BUILD is the “Evaluation Times” window, in which one can calculated total doses of a desired number of years in Figure 4.4.

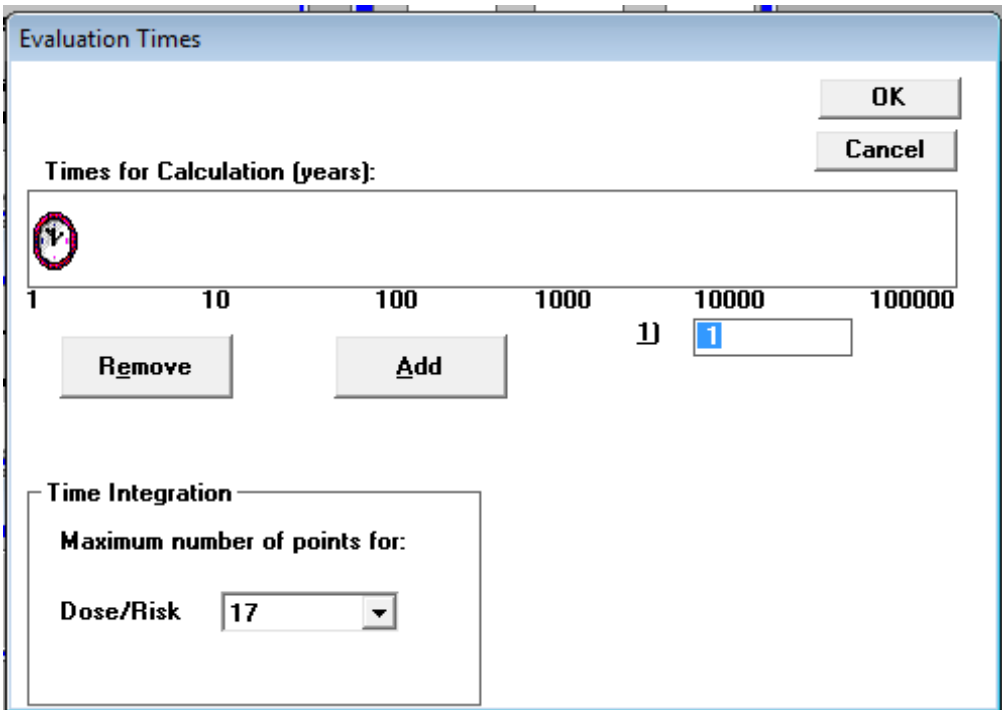

Figure 4.4. “Evaluation Times” Window that allows for the desired number of years to be entered, and also allows for needed input parameters for risk evaluations to be entered into RESRAD-BUILD calculations.

RESRAD-BUILD allows uses to model up to three rooms for a simulation. Rooms can be arranged as three parallel boxes vertically or horizontally, or they can be all adjacent (in an “L-configuration”) with each other. One potential downside with the RESRAD-BUILD code is that one is limited to three different rooms and is limited to specific

geometries. Figure 4.5 shows the user-input windows for entering the room parameters for RESRAD-BUILD.

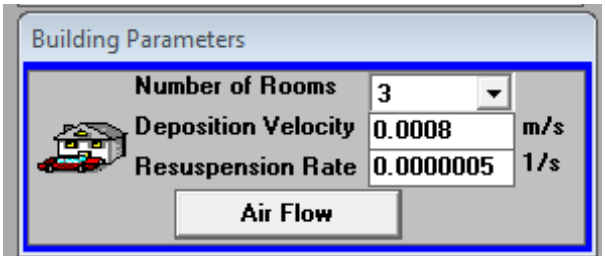

Figure 4.5a. The "Building Parameters" window. Notice that particle size is not an explicit input parameter either in the Building Parameters or "Air Flow" pop-up window. One must relate particle size to a desired deposition velocity in order to ascertain information about dose as a function of particle size.

Along with imputing the room geometry, one can also input the air-flow parameters in this section of RESRAD-BUILD. Air-flow parameters come equipped with another window that allows for more detailed air-flow kinetics to be incorporated into simulations. RESRAD-BUILD can model up to three rooms. Appendix A of the RESRAD-BUILD manual, and discussed in Section 4.1 of this thesis describes the mathematical model for the air-flow kinetics in great detail.

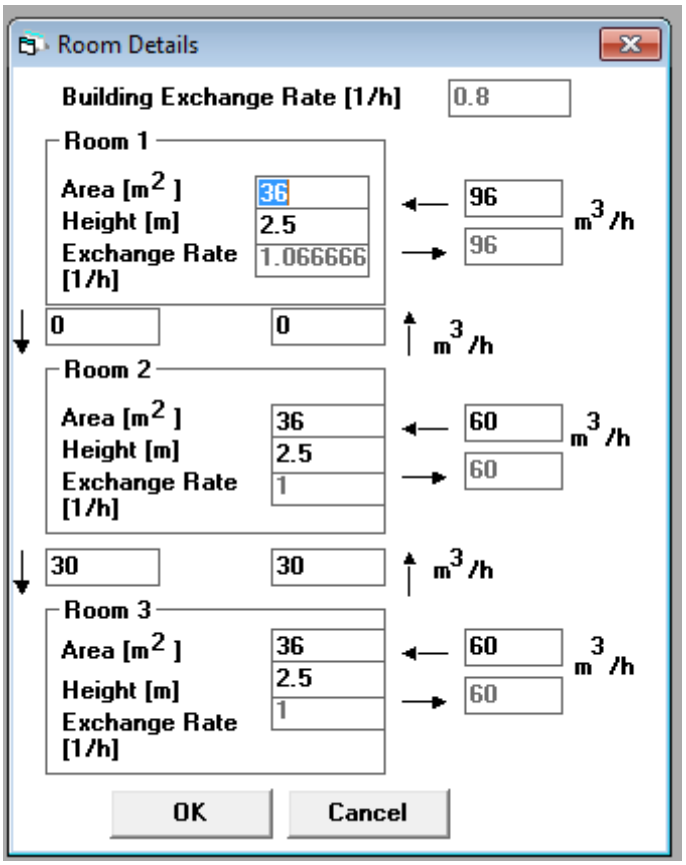

Figure 4.5b. Room Details.

The basic input for radioactivity units in RESRAD-BUILD is air-concentration of a given radionuclide, either in Ci/mL air or in Bq/mL air. One disadvantage of RESRAD-BUILD is that one does not simply enter the amount of a radionuclide directly in units of mass or in particle-size; rather, one has to calculate equivalent air concentration for either mass or for particle size. However, the RESRAD-BUILD manual does provide a number of data sets relating particle size to deposition velocity, which is one of the airflow parameters that is an input into RESRAD-BUILD. Viewed in another way, an advantage of using air concentration of radionuclide the input parameter in RESRAD-BUILD is that one can

directly relate dosimetry calculations to results from air sampling analysis. Table 4.1 shows a data set taken from Abt et. al. (2000).

Particle Size (μm)	Deposition Velocity (m/s)
0.2-0.3	5.40E-04
0.3-0.4	5.10E-04
0.4-0.5	4.74E-04
0.7-1.0	7.30E-04
1 – 2	6.60E-04
2 – 3	8.00E-04
3 – 4	1.00E-03
4 – 5	1.20E-03
5 – 6	1.30E-03
6 – 10	2.10E-03

Table 4.1. Deposition Velocity as a Function of Particle Size.

4.3 Five-Factor Formula Calculations

Both documents, NRC NUREG-CR 6410 and DOE HDBK 3010-94 use the same approach for calculating doses from accidental releases of radioactive materials; NUREG-CR 6410 refers to this as the "Five-Factor Formula." The first step in this approach is to calculate a "source term:"

$$ST = MAR * DR * ARF * RF * LPF \tag{4.10}$$

where

ST: The "Source Term" is the amount of material (expressed in terms of either mass or activity) that can be expected to be released in the case of an

accidental release of radioactive materials. The "Source Term" is the quantity that is used to calculate inhaled doses.

MAR: The "Material at Risk" represents what for the purposes of this thesis is the amount of radioactive material that is present inside the glovebox, and like the source Term can be represented in terms of either mass or activity- also known as the radioactive inventory.

DR: The "Damage Ratio" denotes the amount of MAR that is affected by the accident resulting in the release of radioactive material. The DR is assumed for the purposes of conservatism to be unity

ARF: The "Airborne Release Fraction," which denotes the fraction of the radioactive inventory that actually is released into the air in the event of an accidental release of radioactive material into the atmosphere.

Since the process of forming uranium oxide crystals involves RF-heating of a fine powder to form a crystal, the ARF for this situation was calculated for that of a free-fall spill of a powder. The ARF in this case is given as:

$$ARF = 0.1604 \frac{m^{0.125} h^{2.37}}{\rho^{1.02}} \qquad (4.11)$$

where m is the mass of the material involved in the accident, kg

h for the purposes of the thesis is the length of the glovebox,

ρ is the density of the material, kg/m^3.

RF: The "Respirable Fraction" denotes the fraction of radioactive material in the air that once inhaled is able to pass into the respiratory system. RF is significant only for particles of 10 micron AMAD and less, and this assumption is made for the purposes of conservatism in this thesis, RF is assumed to be unity (NUREG-CR 6410).

LPF: The "Leak Path Factor," and denotes the fraction of airborne radioactive material that is able to escape through various confinement systems. It can represent radioactive material that is capable of escaping through a HEPA filter, or escape through a barrel in the event that a barrel containing radioactive materials is punctured. A conservative assumption made in this calculation is that the entire airborne release fraction (ARF) will escape out of the glovebox; therefore, the LPF in this thesis is assumed to be unity.

It is necessary that all input parameters for the Five-Factor Formula have the same units as the input parameters present in the RESRAD-BUILD simulation, in order to facilitate direct comparison of the two calculations. The input concentrations of 1 pCi/g, 33 pCi/g, 67 pCi/g, 100 pCi/g, 10^4 pCi/g and 10^6 pCi/g allow for U-238 metal, UO_2, Pu-239 metal and PuO_2 to be treated in a consistent manner for the calculations, and have the

advantage of being compared directly to air sample data. The disadvantage is that air concentration is not useful for making safety or operational decisions. Since air concentrations correspond to material that would be present in the laboratories in the event of an accidental release, the air concentrations correspond to the Source Term, ST. Using the room parameters for the RESRAD-BUILD simulations (such as the air-exchange rate and the volumetric flow rates), the ST was calculated as:

$$ST = \left(\frac{pCi}{g}\right) * \left(\frac{g}{mL}\right) * \left(10^{-6}\frac{Ci}{m^3}\right) * \left(volumetric\ flow\ rate\ \frac{m^3}{hr}\right) * \left(air\ exchange\ rate\ hr^{-1}\right) \quad (4.12)$$

A lower estimate of the MAR can be back-calculated by

$$MAR = ST/(DR*ARF*RF*LPF), \quad (4.13)$$

and this information is presented in Table 4.2. The reason that only a lower estimate of the MAR can be given is that the particle size distribution for the material inside the glovebox is ultimately not knowable. It is hence, necessary to assume a worst-case scenario that all the material could be dispersable in the air, and it is necessary to make this worst-case assumption for both the LFP and the RF.

Concentration (pCi/g)	U-238 metal		UO_2	
	MAR (g)	**ST (g)**	**MAR (g)**	**ST (g)**
1	4.00E-01	1.77E-03	2.27E-01	1.77E-03
33	8.53E+00	5.85E-02	4.85E+00	5.85E-02
67	1.59E+01	1.19E-01	9.01E+00	1.19E-01
100	2.25E+01	1.77E-01	1.28E+01	1.77E-01
1.00E+04	1.27E+03	1.77E+01	7.19E+02	1.77E+01
1.00E+06	7.12E+04	1.77E+03	4.04E+04	1.77E+03
Concentration (pCi/g)	**Pu-239 metal**		**PuO_2**	
	MAR (g)	**ST (g)**	**MAR (g)**	**ST (g)**
1	5.21E-04	9.47E-09	3.21E-04	9.47E-09
33	1.11E-02	3.13E-07	6.85E-03	3.13E-07
67	2.06E-02	6.35E-07	1.27E-02	6.35E-07
100	2.93E-02	9.47E-07	1.81E-02	9.47E-07
1.00E+04	1.65E+00	9.47E-05	1.02E+00	9.47E-05
1.00E+06	9.26E+01	9.47E-03	5.71E+01	9.47E-03

Table 4.2. Original inventory and source term of U-238 metal, UO_2, Pu-239 metal, and PuO_2 in units of grams, when converted from RESRAD-BUILD air concentrations. MAR denotes the material at risk and ST denotes the source term that actually is released in an accidental release of radioactive material.

Once the source term has been determined, in terms of both mass and activity, it is then possible to calculate the inhaled dose that can be expected from the calculated source terms. The radioactive material escaped in the glovebox laboratory is assumed to immediately mix into the air; hence, the concentration is simply the source term per volume of the glovebox laboratory (taken to be 145 m^2 and 13 m high). This concentration is multiplied by the breathing rate and the time of exposure to calculate the amount of radioactive material inhaled, in terms of activity. The dose was calculated by using dose conversion factors published in EPA Report 11.

Mathematically, this is

$$H = \frac{ST}{V_{Glovebox\ Lab}} * BR * t_{exposure} * DCF \tag{4.14}$$

where

H is the dose, in units of mrem

ST is the source term, in units of μCi

$V_{glovebox\ Lab}$ = volume of the Glovebox Laboratory

BR is the breathing rate, assumed to be 20 L/min

$t_{exposure}$ is the exposure time, assumed to be 1 hour

DCF is the dose conversion factor, in units of mrem/μCi (taken from EPA Report 11).

CHAPTER 5 RESULTS AND DISCUSSION

The doses resulting from an accidental release of radioactive materials from the glovebox was calculated using two different methods. The first method was to calculate doses using the RESRADBUILD code produced by Argonne National Laboratory. The second method was to use the calculation specified in both NUREG/CR-6410 and the DOE HDBK-3010-94, and explained in Section 4.3 of this thesis.

5.1 Results of RESRAD-BUILD Simulations

Parameters Used in RESRAD-BUILD Calculations

The input parameters used in these simulations were provided by Dr. David LePoire of Argonne National Laboratory, and represents a general model for a glovebox and fume hood. It cannot be guaranteed that the air flow parameters in these input files correspond well to the air flow dynamics in the Glovebox laboratory, with the exception of the deposition velocity values that correspond to the particle sizes. Measurements of the air-flow dynamics would involve a complete analysis of the ventilation system throughout the entire RISE Complex, and this was simply beyond the scope of this thesis, due to time constraints. The airflow and room parameters were adjusted to fit the characteristics of the Glovebox Laboratory, the floor area in the Glovebox laboratory is 145 m^2 and assumed to be 13 m high; the volumetric air flows were assumed to be 2 m^3/hr between

the room and between the glovebox and the Glovebox Laboratory, and between the Glovebox Laboratory and the Crystal Growth Laboratory. The erosion used in the 2.4×10^{-5} cm/d (LePoire, 2013).

Only the particle sizes between 0.2 to 0.3 μm, 0.7 to 1.0 μm, 2 to 3 μm and 6 to 10 μm were examined during the course of the RESRAD-BUILD simulations; Appendix B shows that there was little difference between dose equivalents as a function of particle size generally. Doses were calculated for one year. A robust 50-year deep-dose equivalent (DDE), committed effective dose equivalent (CEDE) and total effective dose equivalent (TEDE) would require an accurate measurement of the air-flow dynamics that was well beyond scope of this thesis and not possible with the time allowed for this thesis. A one year DDE, CEDE and TEDE were calculated, as to have an idea of what doses would be expected in the case of an accidental release of radioactive material from the RISE Glovebox.

Doses were calculated for one radiation worker working at the glovebox, and another worker at a distance of 7.42 m (157 ft) away from the glovebox. The default distance represents a distance consistent with the distance between the glovebox and another worker on the other end of the Glovebox Laboratory or the Crystal Growth Laboratory that it was acceptable to use for the calculations. The calculated doses did however, show a strong influence on the concentration of radioactive material in the air.

Uranium-238 Metal Simulations

All dose calculations were plotted on a log-log scale, with the air concentration being the x-axis (abscissa) and the appropriate dose being the y-axis (ordinate). A general trend that was noticed with the RESRAD-BUILD simulations was a lack of dependence of the inhaled doses as a function of the particle size. Granted, particles larger than 10 μm AMAD were not considered, nor were particle smaller than 0.2 μm AMAD. The largest TEDE, naturally was observed for a source term (ST) of 1773 g, which was 151 mrem for the glovebox worker. The worker away from the glovebox had a higher total dose than the glovebox worker for all concentrations, and for U-238 metal the dose at was 3,320 mrem (3.32 rem). The TEDE is shown in Figure 5.1.

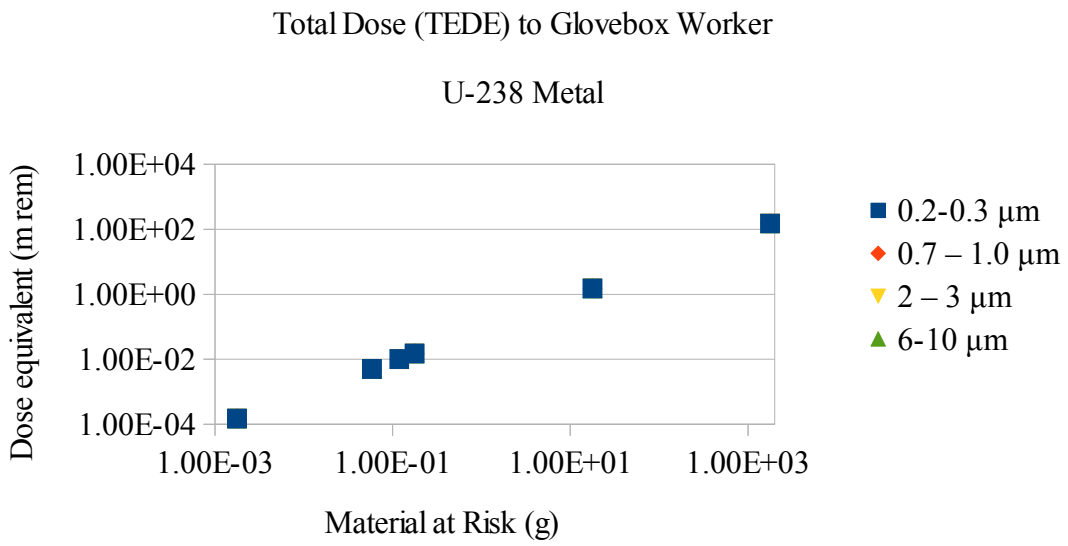

Figure 5.1a. TEDE to Glovebox Worker. The dose for a source term of 1773 g was 53.2 mrem.

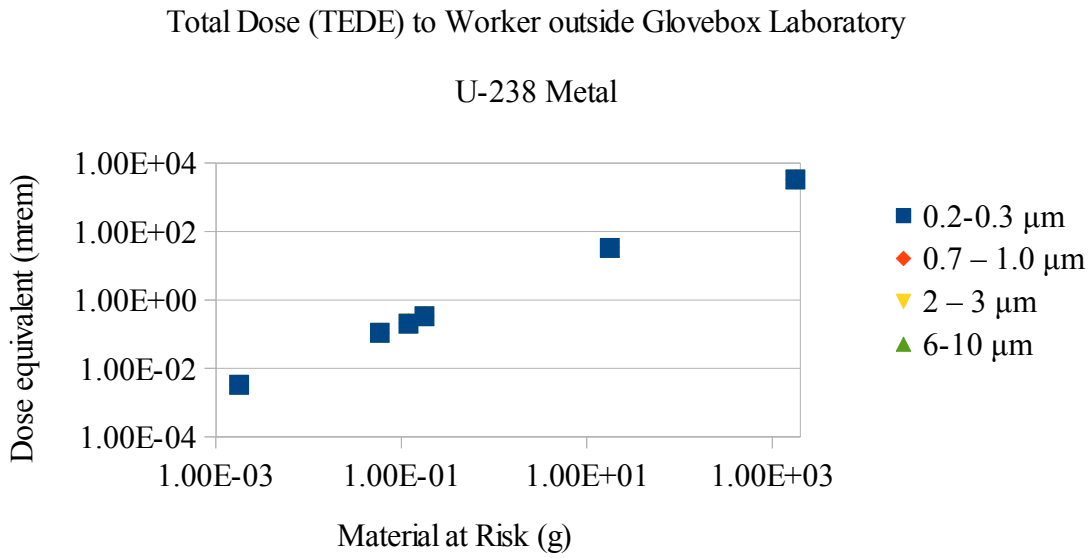

Figure 5.1b. TEDE to worker away from Glovebox. The maximum dose for a source term of 1773 g was 3320 mrem (3.32 rem).

The general trend continued, that the RESRAD-BUILD simulations was a lack of dependence of the inhaled doses as a function of the particle size. Granted, particles larger than 10 μm AMAD were not considered, nor were particle smaller than 0.2 μm AMAD. The doses increased with the concentration of the radioactive material, as is to be anticipated. The DDE for the glovebox worker at a ST of 1773 g, which was 47.3 mrem, while the DDE for the worker away from the glovebox for the same concentration was 1.45 mrem, and is shown in Figure 5.2.

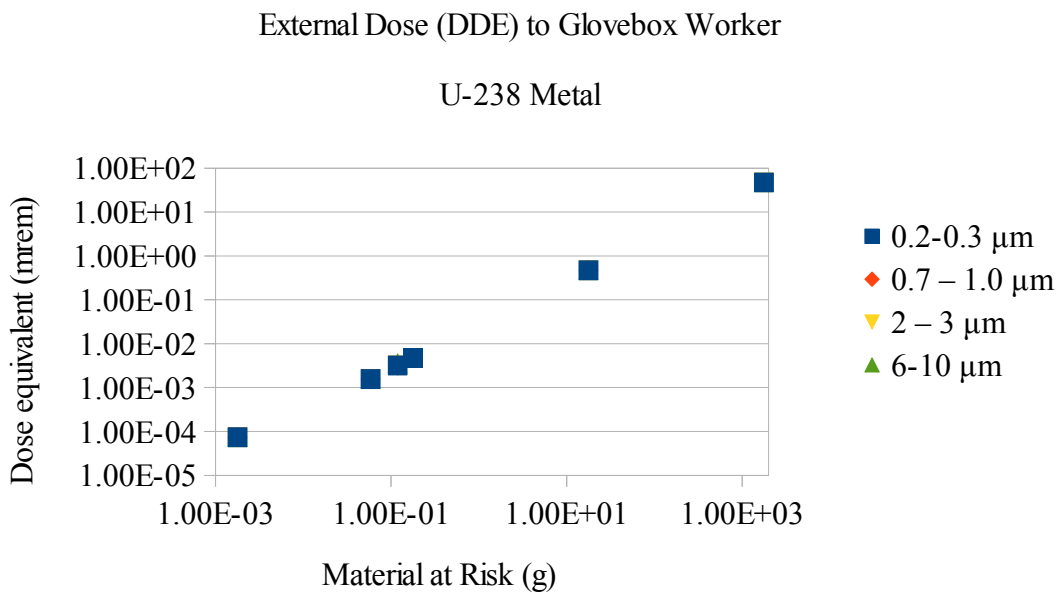

Figure 5.2a. DDE to Glovebox worker

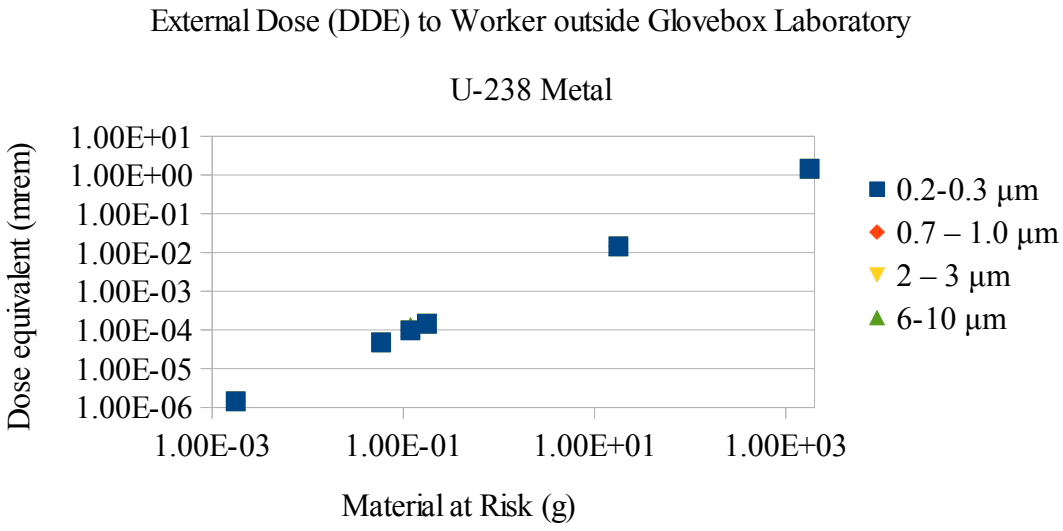

Figure 5.2b. DDE to worker away from Glovebox

The general trend continued, that the RESRAD-BUILD simulations was a lack of dependence of the inhaled doses as a function of the particle size. The doses increased with the concentration of the radioactive material, as is to be anticipated. The glovebox worker had an CEDE of 104 mrem for a ST of 1773 g, while the worker away from the glovebox had an CEDE equal to the total dose, and are shown in Figure 5.3.

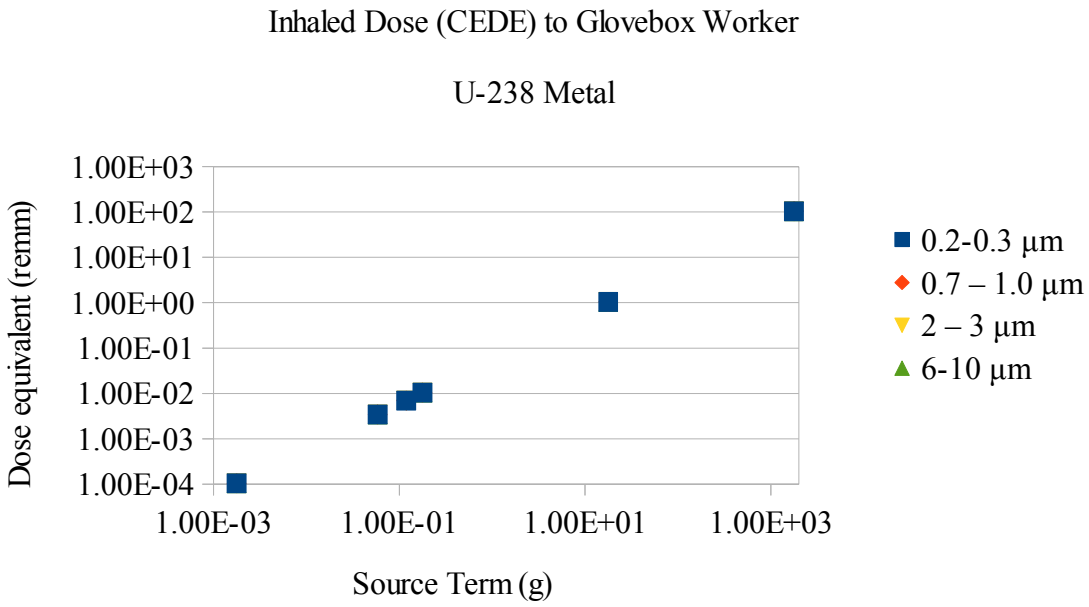

Figure 5.3a. CEDE to Glovebox Worker

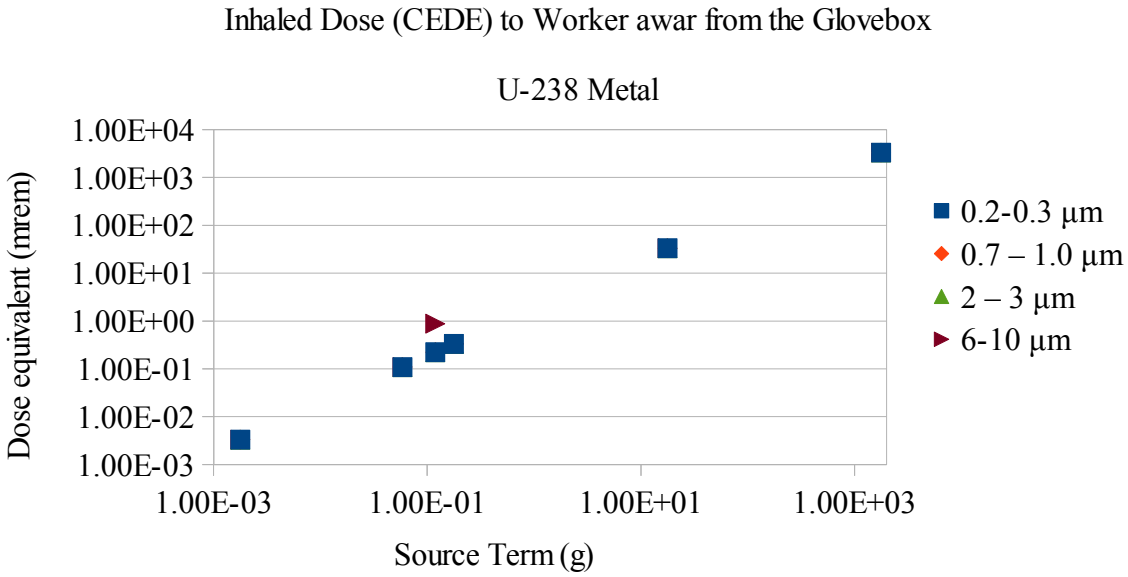

Figure 5.3b. CEDE to worker away from Glovebox

Uranium Oxide Simulations

The systematics, such as a lack of dependence on particle size that were observed for uranium-238 metal were also observed for UO_2. The doses from UO_2 were slightly smaller for than for U-238 metal; this is probably because of the smaller density of UO_2 compared to uranium-238 metal, and that not every atom in is a uranium atom. The largest TEDE from a UO_2 release was 107 mrem for a ST of 1773 g for the glovebox worker, compared to a 1,910 mrem (1.91 rem) for a worker away from the glovebox, both of which were largely inhaled doses, and is shown in Figure 5.3.

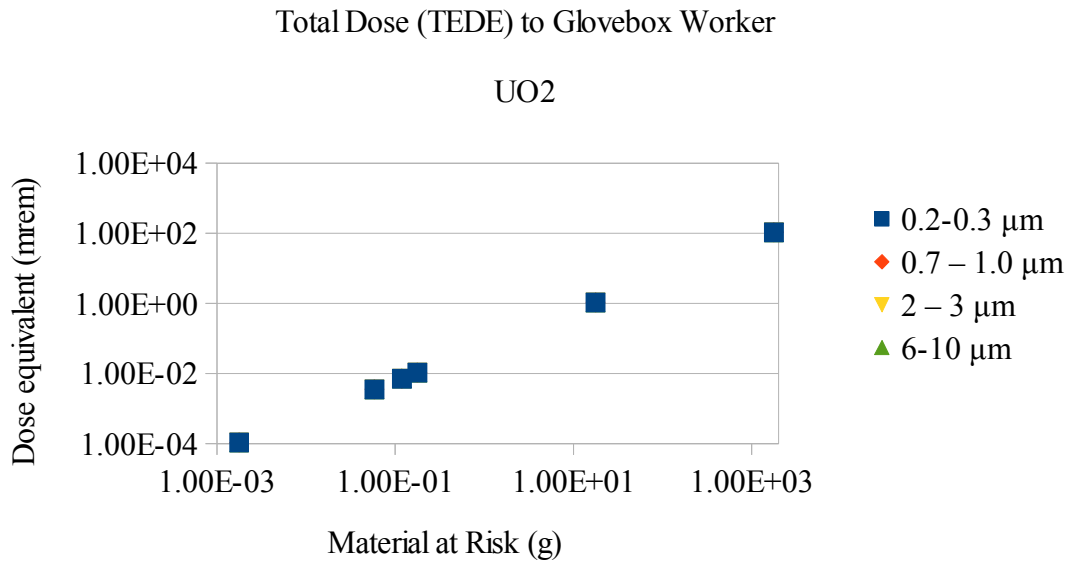

Figure 5.4a. TEDE to Glovebox Worker.

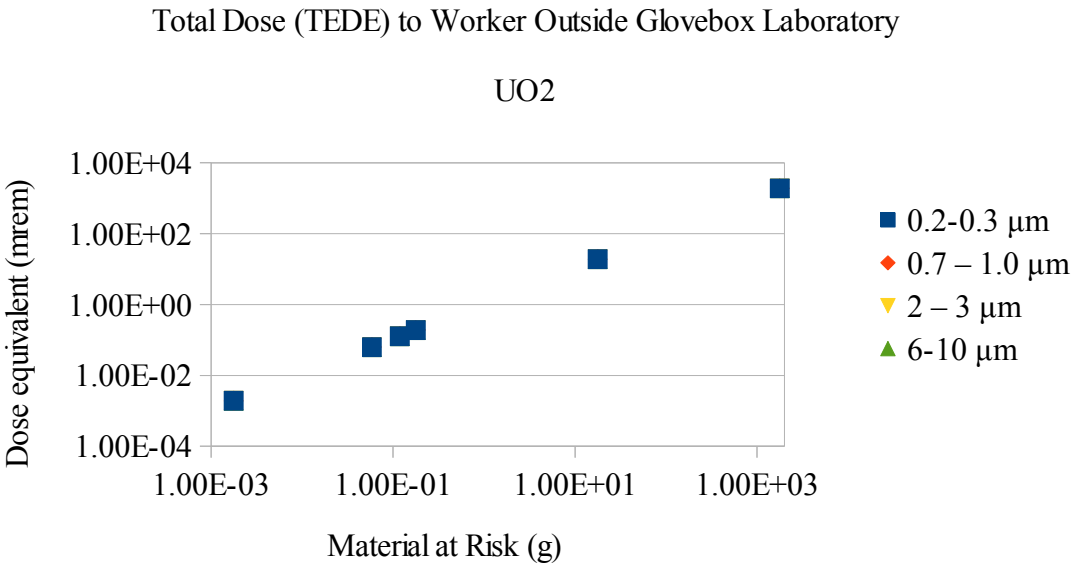

Figure 5.4b. TEDE to Worker away from Glovebox

The general trend continued for the external dose, that the RESRAD-BUILD simulations was a lack of dependence of the inhaled doses as a function of the particle size. The doses increased with the concentration of the radioactive material, as is to be anticipated. The DDE were 47.3 mrem for the glovebox worker and 1.45 mrem for a worker away from the glovebox, and is shown in Figure 5.5.

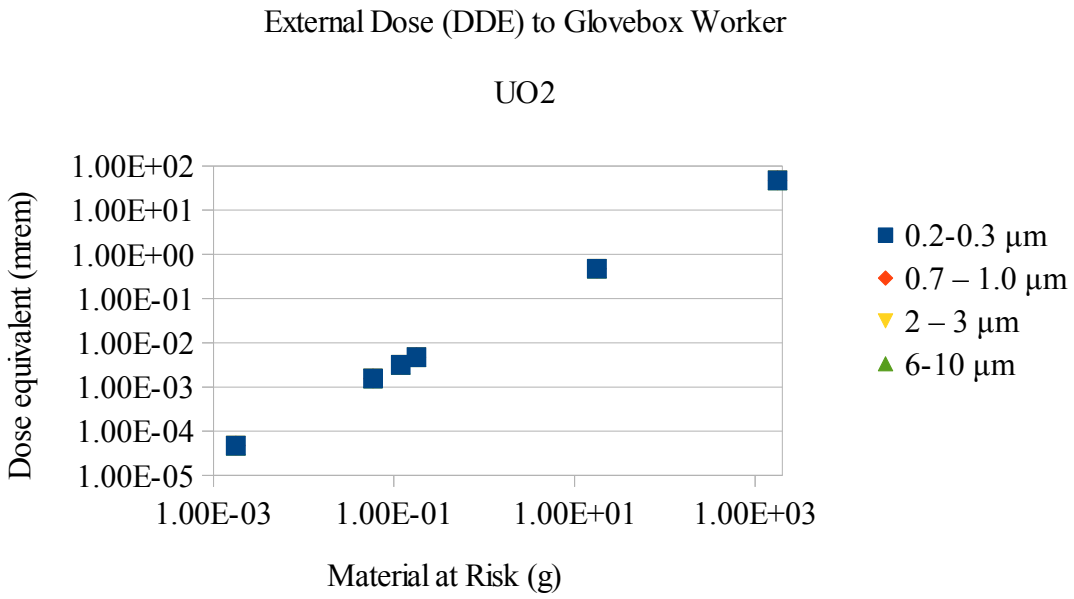

Figure 5.5a. DDE to Glovebox Worker

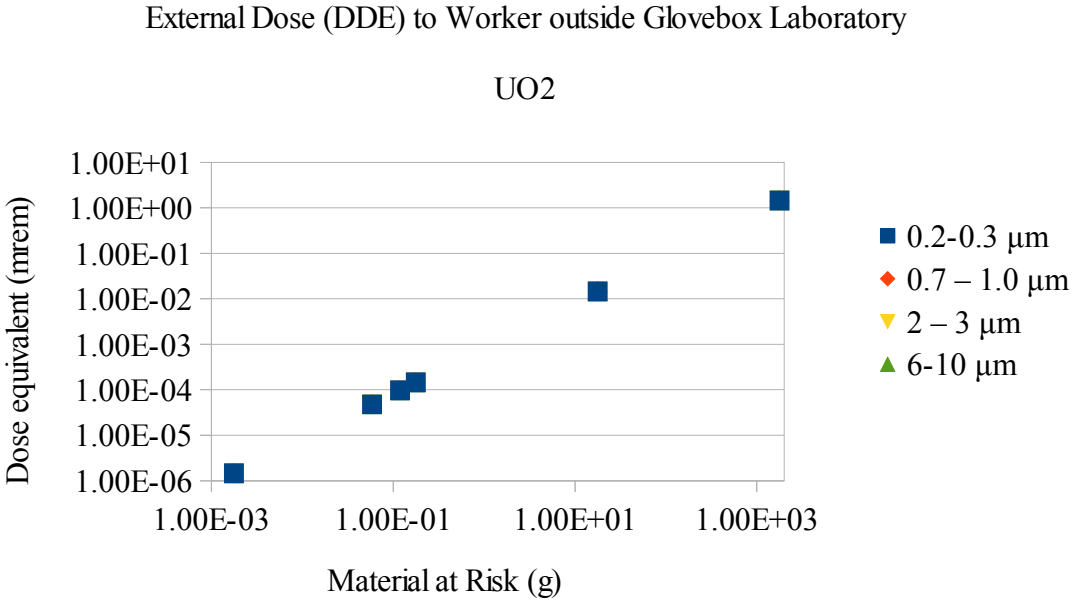

Figure 5.5b. DDE to Worker away from Glovebox

The general trend continued for the inhaled dose, that the RESRAD-BUILD simulations was a lack of dependence of the inhaled doses as a function of the particle size. The doses increased with the concentration of the radioactive material, as is to be anticipated. The glovebox worker had an CEDE of 59.3 mrem, while the worker away from the glovebox had an CEDE almost equal to the total dose, and is shown in Figure 5.6.

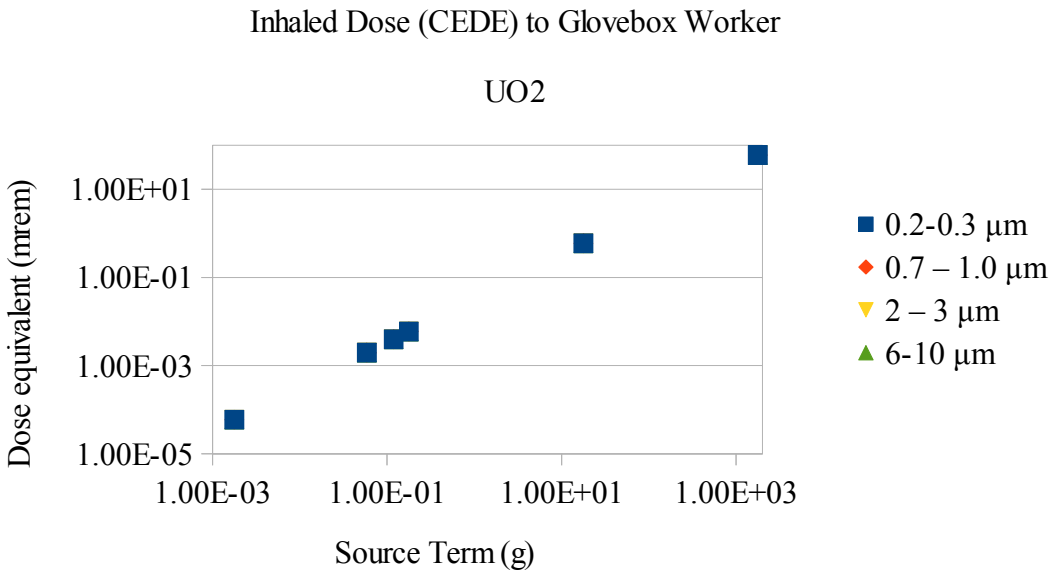

Figure 5.6a. CEDE to Glovebox Worker

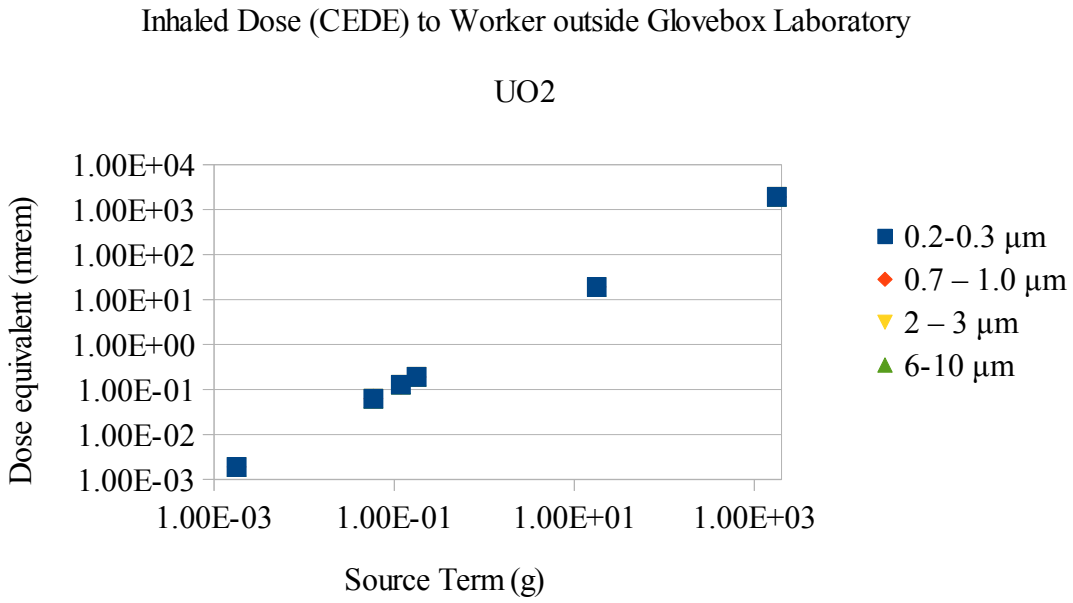

Figure 5.6b. CEDE to Worker away from Glovebox

Plutonium-239 Metal

Owing partly to the significantly higher specific activity of plutonium-239 than uranium-238, it is logical to expect that doses resulting from accidental plutonium releases would be higher than for accidental uranium releases, and this was observed as plutonium doses were 3.5 to 4 times greater than the doses resulting from uranium exposures. Generally, the DDE resulting from accidental plutonium releases were significantly smaller than the CEDE; indeed, the DDE are negligible compared to the CEDE for both the glovebox worker and the worker away from the glovebox; for a ST of 9.47 mg, the glovebox worker had a dose of 392 mrem while the worker away from the glovebox had a dose of 12,500 mrem (12.5 rem), and is shown in Figure 5.7.

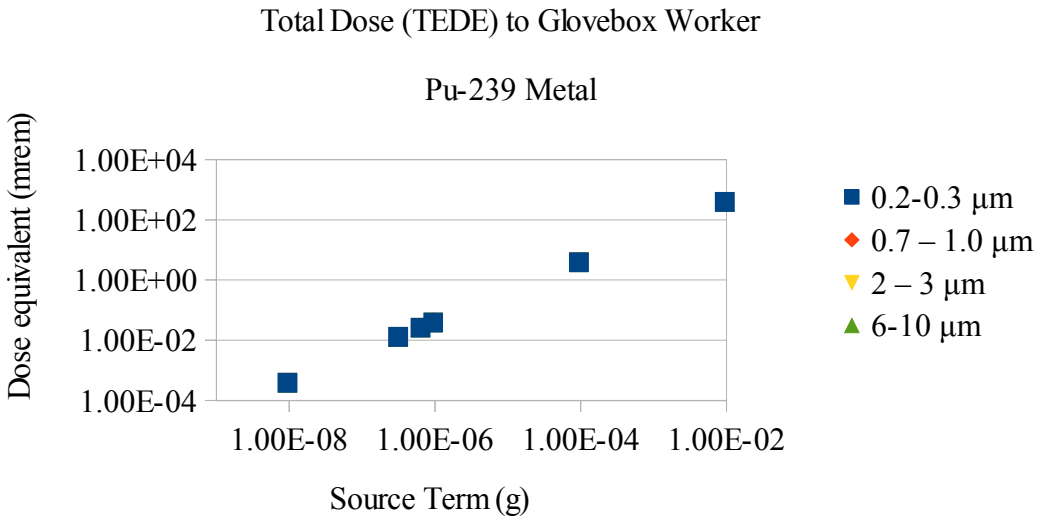

Figure 5.7a. TEDE to Glovebox Worker

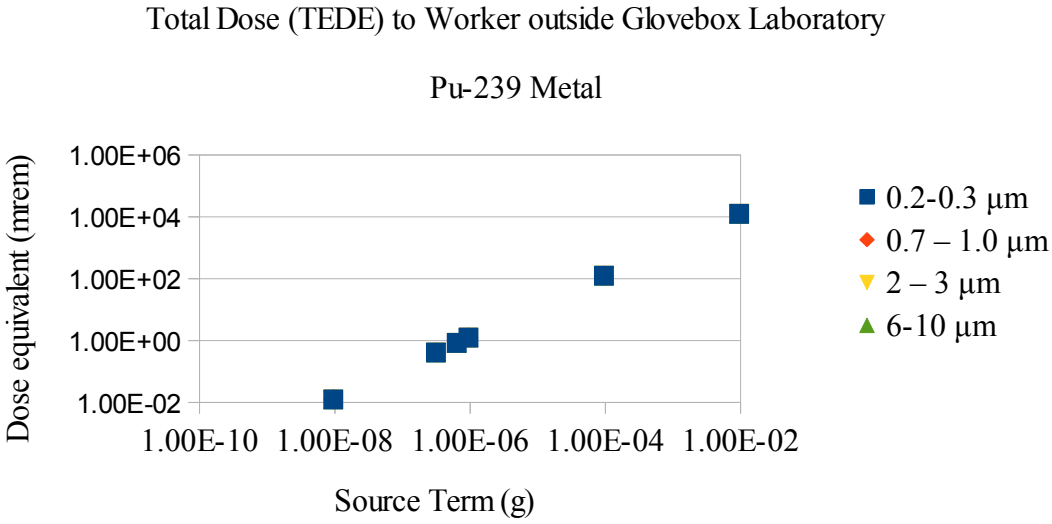

Figure 5.7b. TEDE to Worker away from Glovebox

The general trend continued for the external dose, that the RESRAD-BUILD simulations was a lack of dependence of the inhaled doses as a function of the particle size. DDE for Pu-239 exposures were negligible, with a glovebox worker receiving an external dose of 0.00516 mrem (5.15 μrem) and a worker away from the glovebox receiving a dose of 0.00111 mrem (1.11 μrem) for a ST of 9.47 mg, and is shown in Figure 5.8.

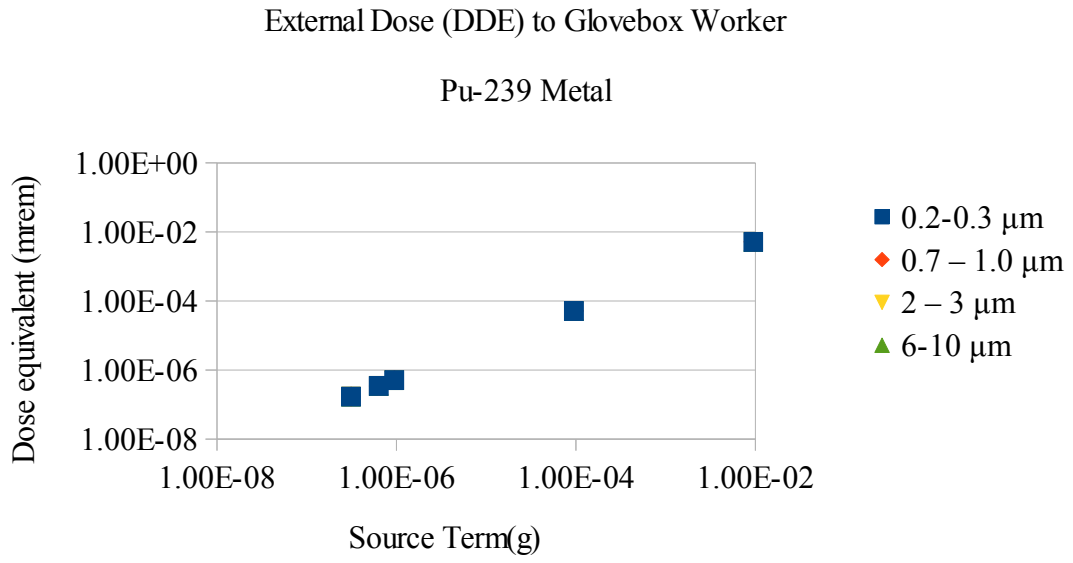

Figure 5.8a. DDE to Glovebox Worker.

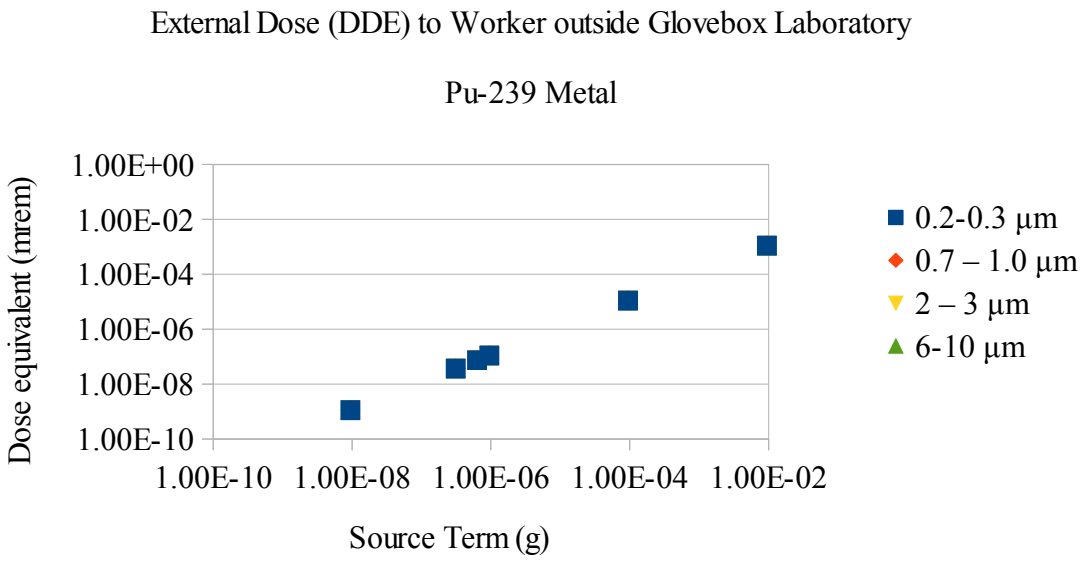

Figure 5.8b. DDE to Worker away from Glovebox.

The general trend continued for the inhaled dose, that the RESRAD-BUILD simulations was a lack of dependence of the inhaled doses as a function of the particle size. The inhaled doses were the majority constituent of the the total dose (TEDE); for a Pu-239 ST of 9.47 mg, the glovebox worker received an inhaled dose of 391 mrem while a worker away from the glovebox received an inhaled dose of 12,5000 mrem (12.5 rem), and is shown in Figure 5.9.

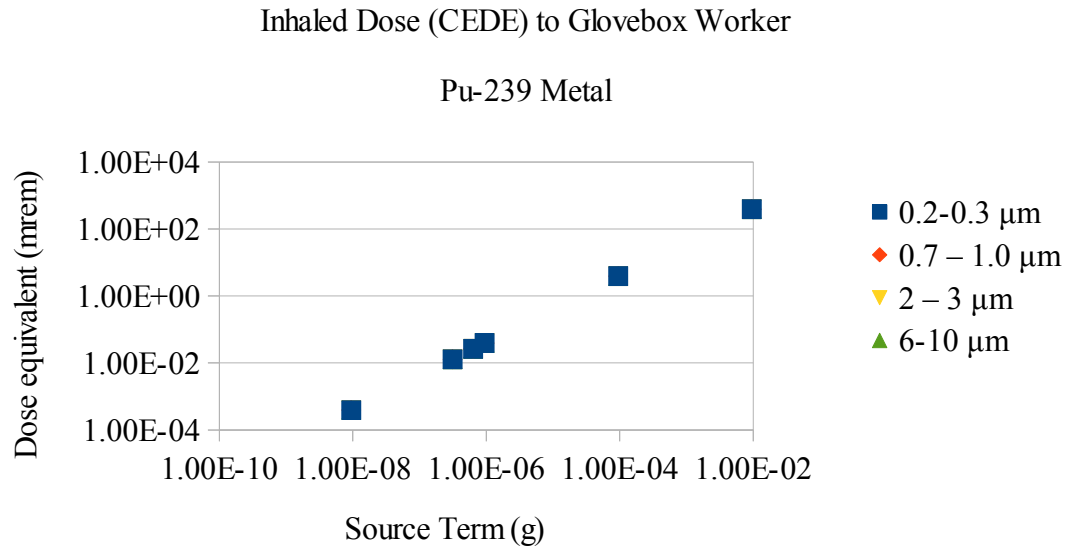

Figure 5.9a. CEDE to Glovebox Worker

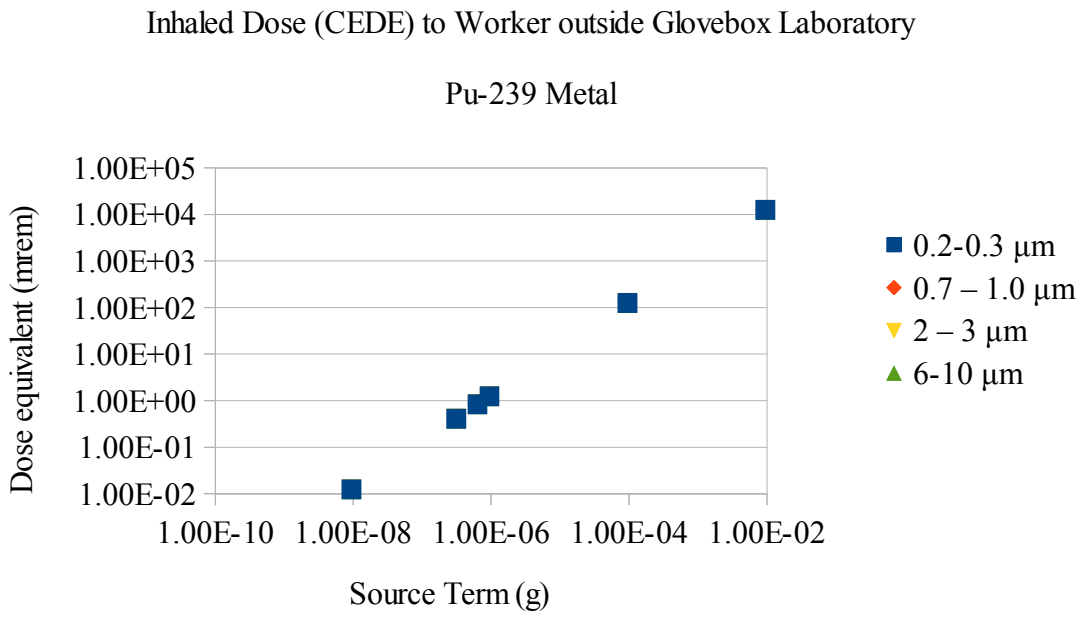

Figure 5.9b. CEDE to Worker away from Glovebox.

Plutonium Oxide Simulations

The results of the RESRAD-BUILD simulations for plutonium oxide were similar to those of plutonium metal. The systematics of PuO2 releases were similar to those of Pu-239 metal in that the DDE was negligible with respect to the CEDE, and the CEDE can be treated as the TEDE. The systematics of accidental PuO_2 also shared characteristics of UO_2 releases, in that the doses are slightly smaller than for Pu-239 metal. The glovebox worker received a dose of 227 mrem at a ST of 9.47 mg was 10.6 μrem, while the worker away from the glovebox received a dose of 7270 mrem (7.27 rem), and is shown in Figure 5.10.

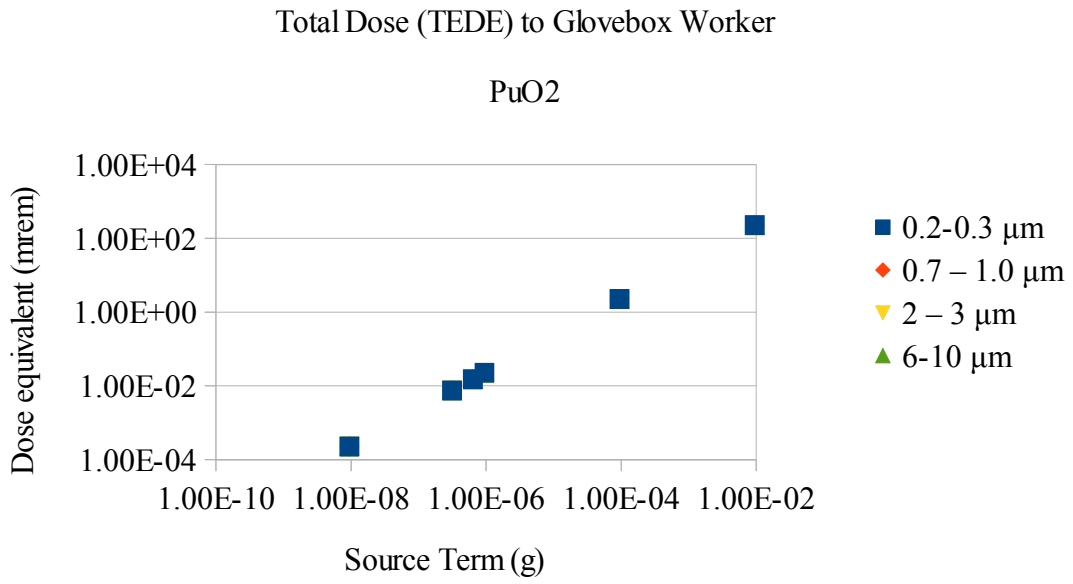

Figure 5.10a. TEDE to Glovebox Worker.

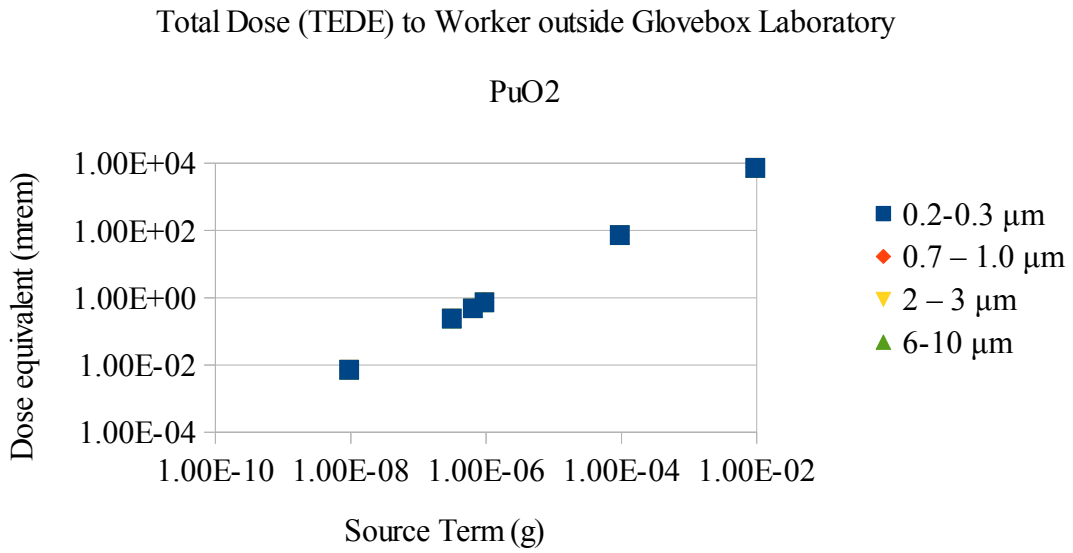

Figure 5.10b. TEDE to Worker away from Glovebox.

The general trend continued for the external dose, that the RESRAD-BUILD simulations was a lack of dependence of the inhaled doses as a function of the particle size. The DDE for Pu-239 exposures were negligible, with a glovebox worker receiving an DDE of 0.00516 mrem (5.16 μrem) and a worker away from the glovebox receiving a dose of 0.00133 mrem (1.33 μrem) for a ST of 9.47 mg of Pu-239, and is shown in Figure 5.11.

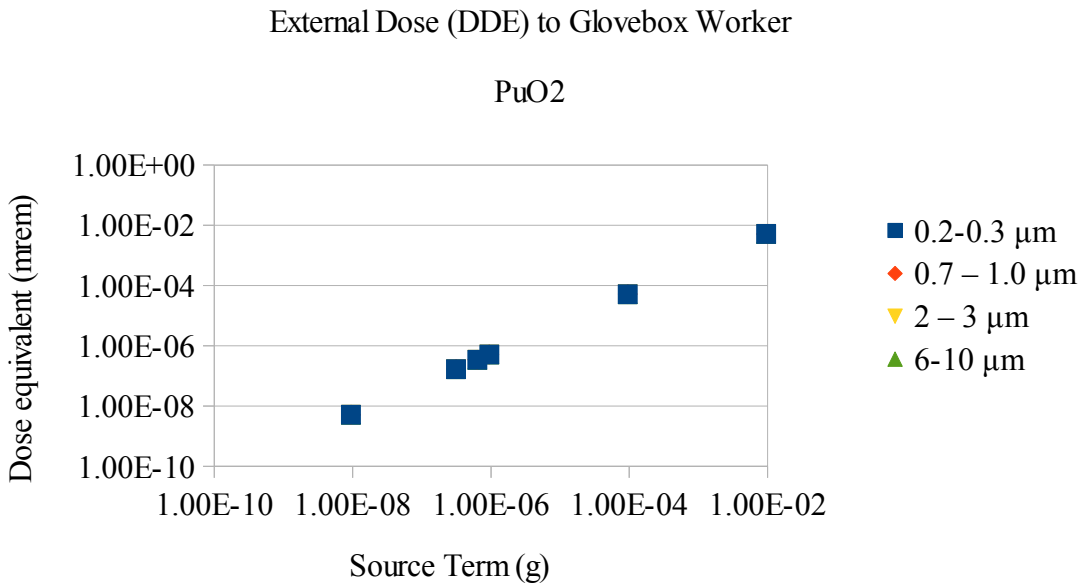

Figure 5.11a. DDE to Glovebox Worker.

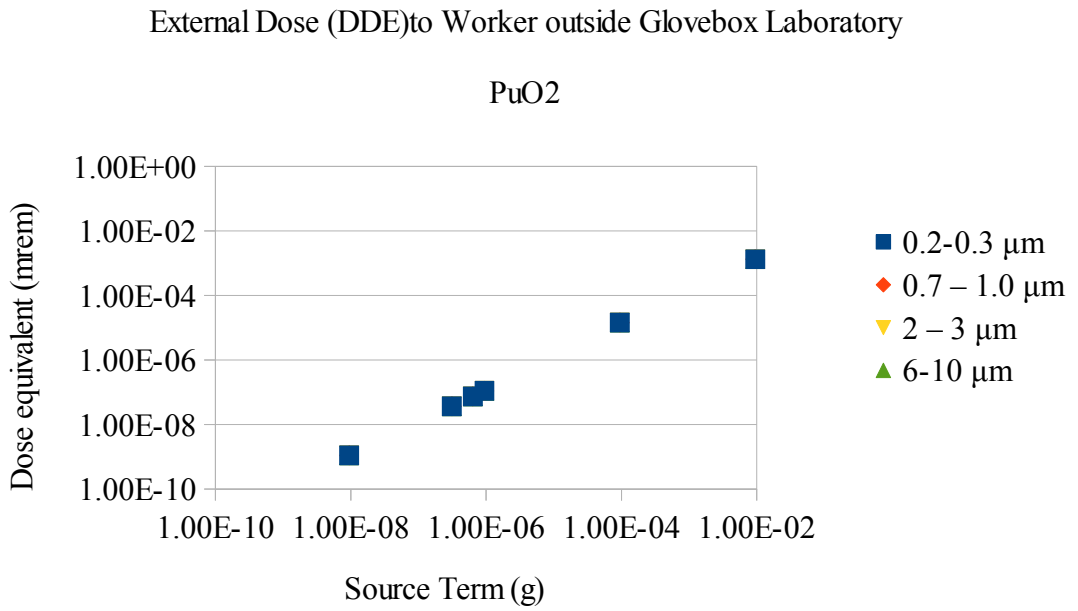

Figure 5.11b. DDE to Worker away from Glovebox.

The general trend continued for the inhaled dose, that the RESRAD-BUILD simulations was a lack of dependence of the inhaled doses as a function of the particle size. The CEDE were the majority constituent of the the total dose (TEDE); for a Pu-239 ST of 9S.47 mg, the glovebox worker received an inhaled dose of 227 mrem while a worker away from the glovebox received an inhaled dose of 7,26000 mrem (7.26 rem), and is shown in Figure 5.12.

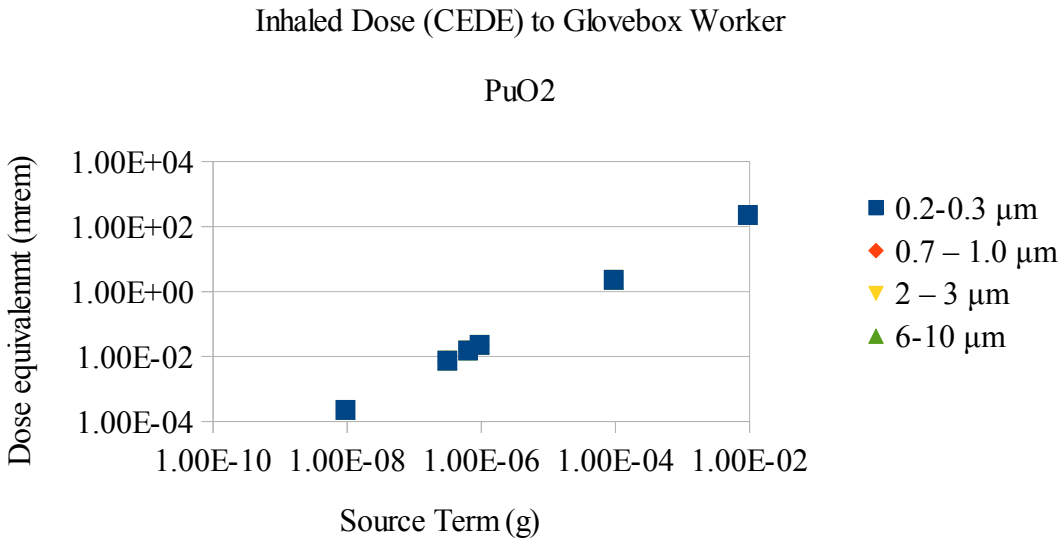

Figure 5.12a. Inhaled Dose to Glovebox Worker.

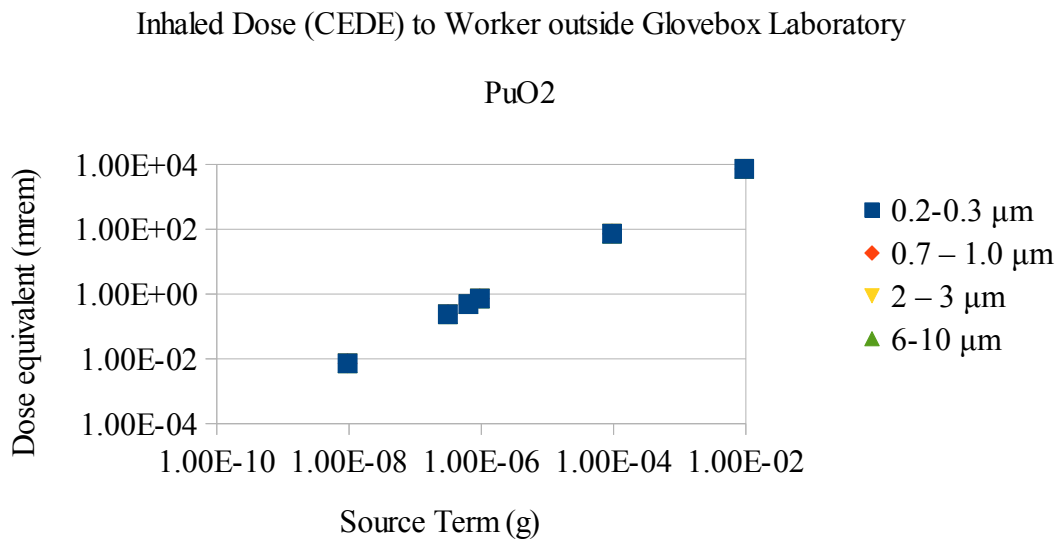

Figure 5.12b. Inhaled Dose to Worker away from Glovebox.

5.2 Results of "Hand Calculations"

The results from the "hand calculations" were within an order of magnitude of the RESRAD-BUILD simulations. The "hand-calculations" considered several of the same air-flow parameters as the RESRAD-BUILD simulations, but assume a linear dependance on all parameters. The "hand calculations" still were within an order of magnitude of the RESRAD-BUILD simulations. The doses as a function of source term (escaped radioactive inventory) is shown in Figure 5.

It should be noted that airflow parameters that were used in both the Five-Factor Formula calculations and the RESRAD-BUILD simulations are not well understood, because there was no opportunity to measure these parameters. The numerical results for all concentrations are shown in Table 5.1. The results of the Five-Factor Formula calculations that assumed an immediate and homogeneous mixing of released radioactive inventory into the room, were in between the RESRAD-BUILD results for the glovebox worker and the worker away from the glovebox. The two calculation approaches were in best agreement for the glovebox worker away from the glovebox for PuO_2 and UO_2, while in the case of the metals the Five-Factor Formula results were in between the results of the glovebox worker dose and the dose for the worker away from the glovebox for lower concentrations but came into agreement for the results of the worker away from the glovebox as the concentration increased. However, in all cases the doses at low concentration were in the μrem range.

The particle size range utilized in this hazard analysis, that the particle size distribution did not have a significant effect on inhaled doses. The deposition velocity is a difficult parameter to quantify; hence, it is difficult to correlate particle size to deposition velocity. Therefore, any significant deviation in doses as a function of deposition velocity would be inherently suspect. The lack of a strong dependence of doses as a function of particle size is consistent with the ICRP 30 inhalation model, which has a constant fraction of deposition in the tracheobronchial region of the respiratory system from 0.2 to 10 μm AMAD.

It makes more sense that doses would depend more strongly on air-flow parameters present in the Glovebox Laboratory and the Crystal-growth Laboratory, such as room air-exchange rates and the building air-exchange rate. It was not possible to take measurements of either parameter in the time frame of this thesis. The air-flow dynamics in the Glovebox Room and the Crystal-Growth Laboratory along with the air-exchanges between the two rooms were not known when carrying out this safety analysis, and values were assumed for both the "hand calculations" and the RESRADBUILD simulations. The errors in the air-flow dynamics will lead to errors in the inhaled doses that cannot reasonably be dismissed.

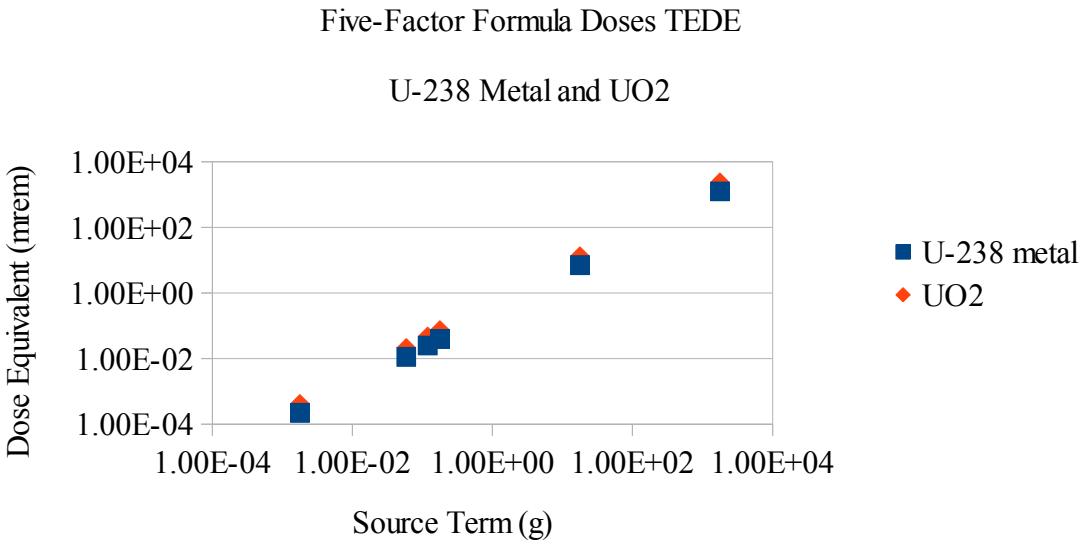

Figure 5.13a. Five-Factor Formula Calculation of Doses as a Function of Escaped Uranium Material Inventory

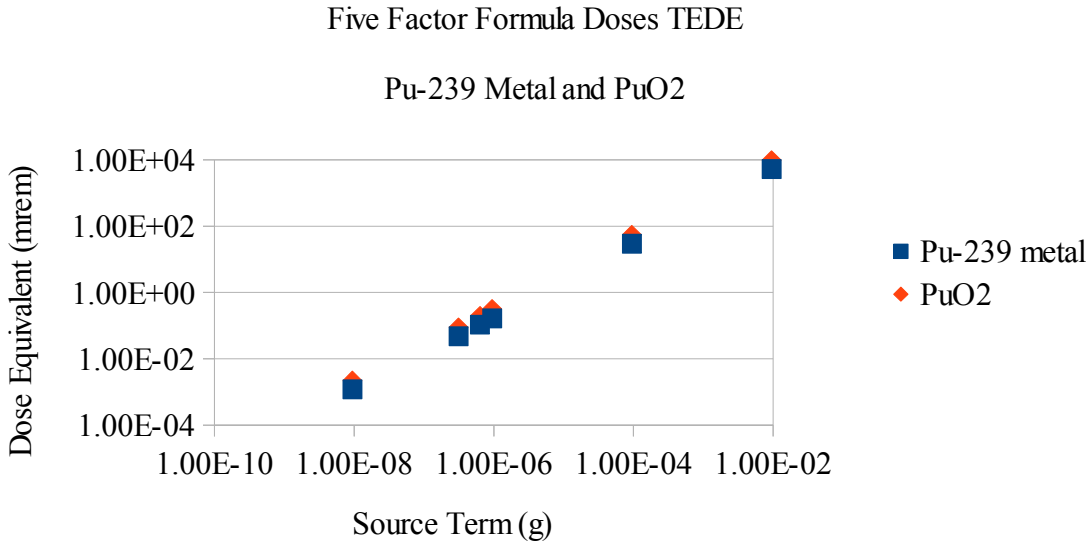

Figure 5.13b. Five-Factor Formula Calculation of Doses as a Function of Escaped Plutonium Material Inventory

U-238 metal				
	ST (g)	**"Five-Factor" TEDE (mrem)**	**RESRAD Glovebox Worker TEDE (mrem)**	**RESRAD Worker away from Glovebox TEDE (mrem)**
	1.77E-03	2.22E-04	1.51E-04	3.32E-03
	5.85E-02	1.13E-02	4.99E-03	1.10E-01
	1.19E-01	2.51E-02	1.01E-02	2.14E-01
	1.77E-01	3.95E-02	1.51E-02	3.32E-01
	1.77E+01	7.02E+00	1.51E+00	3.32E+01
	1.77E+03	1.25E+03	1.51E+02	3.32E+03
UO_2				
	ST (g)	**"Five-Factor" TEDE (mrem)**	**RESRAD Glovebox Worker TEDE (mrem)**	**RESRAD Worker away from Glovebox TEDE (mrem)**
	1.77E-03	4.14E-04	1.07E-04	1.91E-03
	5.85E-02	2.12E-02	3.53E-03	6.29E-02
	1.19E-01	4.69E-02	7.16E-03	1.28E-01
	1.77E-01	7.36E-02	1.07E-02	1.91E-01
	1.77E+01	1.31E+01	1.07E+00	1.91E+01
	1.77E+03	2.33E+03	1.07E+02	1.91E+03
Pu-239 metal				
	ST (g)	**"Five-Factor" TEDE (mrem)**	**RESRAD Glovebox Worker TEDE (mrem)**	**RESRAD Worker away from Glovebox TEDE (mrem)**
	9.47E-09	1.20E-03	3.91E-04	1.25E-02
	3.13E-07	4.81E-02	1.29E-02	4.13E-01
	6.35E-07	1.07E-01	2.62E-02	8.39E-01
	9.47E-07	1.67E-01	3.91E-02	1.25E+00
	9.47E-05	2.98E+01	3.91E+00	1.25E+02
	9.47E-03	5.29E+03	3.91E+02	1.25E+04
PuO_2				
	ST (g)	**"Five-Factor" TEDE (mrem)**	**RESRAD Glovebox Worker TEDE (mrem)**	**RESRAD Worker away from Glovebox TEDE (mrem)**
	9.47E-09	2.21E-03	2.27E-04	7.27E-03
	3.13E-07	8.86E-02	7.50E-03	2.40E-01
	6.35E-07	1.97E-01	1.52E-02	4.87E-01
	9.47E-07	3.18E-01	2.27E-02	7.27E-01
	9.47E-05	5.48E+01	2.27E+00	7.27E+01
	9.47E-03	9.75E+03	2.27E+02	7.27E+03

Table 5.1. Comparison of RESRAD-BUILD and Five-Factor Formula Results

CHAPTER 6 CONCLUSIONS AND RECOMMENDATIONS FOR FUTURE WORK

6.1 Conclusions

RESRAD-BUILD offers several advantages over the hand-calculations in performing safety analyses over hand-calculations. One of the more important advantages is that one can also consider air-flow dynamics in different rooms at the same time when performing a safety analysis in RESRADBUILD, whereas, one generally must consider air-flow dynamics to be identical for all rooms in hand calculations. Conversely, if air-flow dynamics parameters are not known, then one can have an inaccurate result. Alternatively, one could use RESRAD-BUILD to design a ventilation system that would minimize the spread of radioactive materials in the event of an accidental release from the glovebox.

10 CFR 70.61 mandates that credible high-consequence accidents be very unlikely, which is defined as doses to glovebox workers in excess of 100 rem, and 25 rem to anybody outside the controlled area. Even for the worst-case scenario that radioactive material releases of 1 μCi/g, there was no dose to a glovebox worker close to 100 rem, as the highest observed dose to a glovebox worker was 9.75 rem in the case of an accidental PuO2 release, and the worker away from the Glovebox received a dose of 7.3 rem. This dose is not likely to threaten the life of exposed glovebox, or cause irreversible health effects to either worker. 10 CFR 70.61 also stipulates that credible intermediate-

consequence accidents shall be unlikely, which is defined as a dose of 25 rem to glovebox workers and 5 rem to anybody outside the controlled area. The doses for the worst-case PuO2 satisfies the NRC requirements for a credible intermediate-consequence accident.

6.2 Recommendations for Future Work

The hazard analysis that was carried out in this thesis was limited to U-238 in metal and oxide chemical forms, Pu-239 in metal and oxide chemical forms, and to fission products. Accidental criticality incidents were not considered in this thesis. Since current uranium oxide crystal research at the RISE Complex is focused on understanding the mechanical properties, structural properties, thermodynamic properties and other materials science parameters of crystal nuclear fuel, it is not necessary at this juncture to consider criticality safety. However, when the neutronic properties of crystal nuclear fuel are considered, it will be necessary to perform a criticality evaluation of processes in the glovebox. It is entirely conceivable that PuO_2 will be mixed with UO_2 in this glovebox once the Uranium Crystal Project reaches this level.

The current research process involves grinding sintered depleted uranium oxide pellets in a custom built grinder. Resulting uranium oxide powder is selected using a sieve that filters out powder of a desired diameter or smaller. The distribution of particle sizes resulting from the production of the uranium oxide powder is not known. A

determination of the particle size distribution from this grinding process would allow for a more accurate calculation of doses resulting from an accidental release of radioactive material from the glovebox. The particle size distribution could be determined by x-ray diffraction or electron microscopic analysis.

Work that will be essential is a comprehensive analysis of the ventilation system in the entire RISE Complex. If radioactive material was ever to escape out of the RISE Glovebox, it is unknown how radioactive material would travel throughout the ventilation system throughout the RISE Complex at the time this thesis was completed. Simple diffusion of radioactive material out of the Glovebox Laboratory and into the Crystal Growth Laboratory was assumed to be the mode by which radioactive material would be dispersed; this assumption was made because the characteristics of the ventilation system is not known, yet at the same time, a complete analysis was not achievable in the time allotted for this thesis.

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APPENDIX A PROCESSES OF CRYSTAL GROWTH AT THE RISE COMPLEX

A.1 Bridgman Single Crystal Growth Technique

All single crystal growth techniques involve the melting of powdered material, in one manner or another. When a multi-component powder is melted, the impurities will have a different migration in the melt matrix and the different components will solidify at different rates (segregation coefficient). For example, when fabricating carbon doped aluminum oxide crystals (for optically stimulated luminescence [OSL] dosimeters), the carbon atoms will have a different segregation coefficient from the aluminum and oxygen. Also, the "solute" atoms will not fit as easily in the matrix of the "solvent" material, and solute atoms will preferentially segregated out of the bulk mix. Sometimes, "solute" atoms are undesired contaminants (such as anything other than germanium in a high purity germanium [HPGe] gamma spectrometer), and this preferential segregation of "solute" atoms is desirable. Other instances, such as carbon in OSL dosimeters or thallium in sodium iodide scintillators, the "solute" atoms are highly desirable dopants necessary to affect certain physical properties in the crystal, and preferential segregation in these cases is highly undesirable. Care must be taken in all cases to stabilize the liquid-solid interface in either case because dendrites resembling fir trees can form at the interface, affecting the microstructure and the performance and physical properties of the final crystal (Naumann, 2009).

The solidification process does have directionality. In an unidirectional solidification process, the melt is pushed through a cooling system, or a powdered material is heated. A diagram is unidirectional solidification is shown in Figure A.1, which depicts a metal alloy.

The Bridgman crystal growth technique is the most commonly used crystal growth technique involving unidirectional solidification. This crystal growth technique was developed by Dr. Percy Bridgman of Harvard University early in the twentieth century. The original approach was simple and merely involved moving a quarts ampoule (tube containing the powdered material to be melted and crystallized) though a cylindrical furnace. A later refinement was made the the Bridgman crystal growth technique that includes a cold zone and an adiabatic zone between the cylindrical furnace and the cold zone, which has the effect of providing straightened out heat flow and better control of the solidification interface; this is referred to appropriately as the Bridgman-Stockbarger technique. The primary advantages of the Bridgman technique is that it allows for control of the solidification rate, position of the solidification front and the thermal gradients used in solidification (Naumann, 2009).

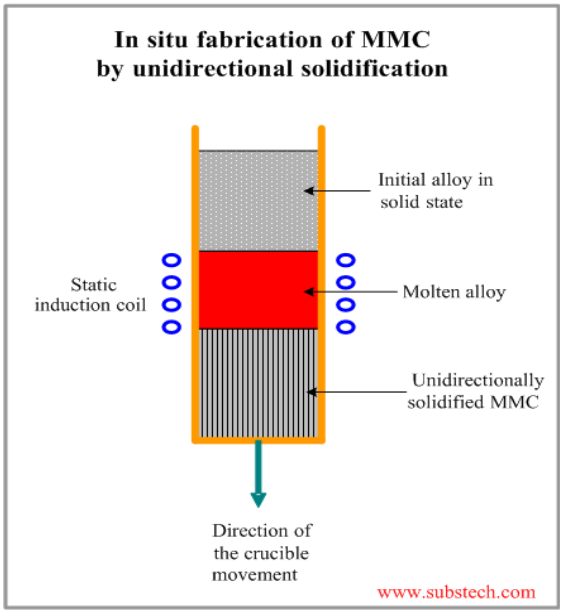

Figure A.1. Unidirectional Solidification of a metal alloy. Taken from

http://www.substech.com/dokuwiki/doku.php?id=in-

situ_fabrication_of_metal_matrix_composites

written by Dr. Dimitri Kopelivich.

A.2 UO_2 Crystal Growth Process

A variation of the Bridgman crystal growth, called the pressure growth process is used at the RISE Complex to grow and fabricate the single UO_2 crystals. This process was originally developed by Jeff Nause of Cermet Industries to grow high-quality single crystals of zinc oxide (ZnO), wafers from which are used to fabricate gallium nitride (GaN) blue lasers (for DVD players), GaN blue diodes, among other devices. The advantages of this pressure growth technique is that it provides large, high-quality ZnO wafers at low cost for the mentioned devices.

The pressure growth technique that is used at the RISE Complex to grow UO_2 single crystals, was developed by Jeff Nause to grow single crystals of zinc oxide (ZnO). The pressure growth technique is a Bridgman-type of crystal growth process that involves RF-heating of UO_2 powder to melting. The RF-heating generates induced fields in the UO_2 powder, which produces eddy currents which in turn produce Joule heating in the UO_2 powder until a molten phase is achieved. The crucible holding the melting UO_2 powder is cold-walled, such that part of the melt material forms a solid thermal barrier with the molten material. The cooled material prevents the melt from coming into contact with the cooling surface and hence, prevents contamination problems and crucible reactivity, irrespective of the temperature of the UO_2 melt. The melting process takes place in high pressure environments ranging from 1 to 100 atmospheres, preventing the evolution of volatile components and decomposition of UO_2 molecules into atomic species that would otherwise occur in normal pressures (D. C. Reynolds, L. C. Litton, D.

C. Look, J. E. Hoelscher, and B. Claflin, T. C. Collins, J. Nause and B. Nemeth, 2004).

A summary of the advantages and disadvantages of the pressure growth technique for ZnO will reveal information that would be useful for understanding potential advantages and disadvantages for using this technique for the fabrication of UO_2 single crystals. Advantages of the pressure growth technique are that it produces a high-purity material at fast growth rates (1-5 mm/hr) and is capable of growing crystals of a 7.5 cm diameter or larger, capability of growing single crystals of different crystal orientations on account of the three-dimensional nature of growth, and in situ control of doping. Disadvantages of the pressure growth technique are that it produces crystals of low-angle grain boundaries[4] and does have impurities on the order of parts per million (Morkoç and Özgür, 2009).

4 A grain boundary is an interruption in the regular periodicity of crystals. Grain boundaries can scatter light photons as well as electrons and holes traversing the grain boundary. Grain boundaries constitute problems for designers of semiconductor or optoelectronic devices who want for semiconductor devices to have uniform electronic and optical properties (Naumann, 2009).