# Draft

#### **ADVISORY BOARD ON**

### **RADIATION AND WORKER HEALTH**

## National Institute for Occupational Safety and Health

### **REVIEW OF THE NIOSH SITE PROFILE FOR THE**

### AMES LABORATORY

Contract No. 200-2009-28555 SCA-TR-SP2013-0044, Revision 0

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## **Acronyms and Abbreviations**

Advisory Board or Board	Advisory Board on Radiation and Worker Health
AEC	U.S. Atomic Energy Commission
ALRR	Ames Laboratory Research Reactor
AWE	Atomic Weapons Employer
сс	cubic centimeter
$\mathrm{cm}^2$	square centimeter
Ci	curie
CFR	Code of Federal Regulations
D	day
D&D	decontamination and decommissioning
DOE	U.S. Department of Energy
dpm/m <sup>3</sup>	disintegrations per minute/per cubic meter
EEOICPA	Energy Employees Occupational Illness Compensation Program Act of 2000
ERDA	Energy Research and Development Administration
FMPC	Feed Materials Production Center
GM	geometric mean
GSD	geometric standard deviation
HEPA	high-efficiency particulate air
hr	hour
ICRP	International Commission on Radiological Protection
IMBA	Integrated Modules for Bioassay Analysis
ISU	Iowa State University
kg	kilogram
kV	kilovolt
L	liter
LAT	lateral
lbs	pounds
$m^3$	cubic meter
MDA	minimum detectable activity
MDL	minimum detection level

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MED	Manhattan Engineer District			
MeV	megaelectron-volt, 1 million electron-volts			
Mg	magnesium			
mg	milligram			
mL	milliliter	milliliter		
mR	milliroentgen	milliroentgen		
MRD	minimum recordable do	se		
mrem	millirem			
MW	megawatt			
N/A	Not applicable			
NCRP	National Council on Rad	diation Protection and Measu	irements	
NIOSH	National Institute for Occupational Safety and Health			
ORAUT	Oak Ridge Associated Universities Team			
ORNL	Oak Ridge National Laboratory			
pCi/d	picocurie per day			
POC	probability of causation	probability of causation		
R&D	research and developme	research and development		
S	second			
SC&A	S. Cohen and Associates	s (SC&A, Inc.)		
SEC	Special Exposure Cohort			
SRDB	Site Research Database	Reference		
TBD	technical basis documen	ıt		
Th	thorium			
ThF <sub>4</sub>	thorium-fluoride			
ThO <sub>2</sub>	thorium dioxide			
TLD	thermoluminescent dosi	meter		
T.N.T.	trinitrotoluene			
U	uranium	uranium		
$U_3O_8$	uranium oxide			
$UF_4$	uranium tetrafluoride			
U.S.C.	United States Code			
yr	year			
ZnCl	zinc chloride			

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β	beta		
γ	gamma		
μCi	microcurie		
μg	microgram		
§	section or sections		

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# **1.0 EXECUTIVE SUMMARY**

This draft report presents S. Cohen and Associates' (SC&A, Inc.) evaluation of the *Site Profile for the Ames Laboratory*, ORAUT-TKBS-0055, Rev. 3 (ORAUT 2012a). This draft report was prepared at the request of the Advisory Board on Radiation and Worker Health (Advisory Board). Authorization for the preparation of this report is described in Section 2 of the report.

As part of our evaluation, SC&A also reviewed numerous other documents that were considered relevant including the following:

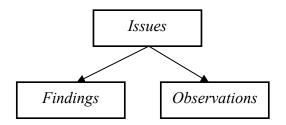
- Select documents that were referenced in the Ames Laboratory Site Profile
- Documents contained in the National Institute for Occupational Safety and Health (NIOSH) Site Research Database (SRDB)
- Relevant reference texts and scientific studies cited in the open literature
- Personal memoirs/manuscripts; worker interviews; and memoranda/letters that provide critical insight on attitudes, work conditions/practices and events that surround the Ames Project

### 1.1 TECHNICAL APPROACH AND REVIEW CRITERIA

The approach used by SC&A to perform this review includes, but is not limited to, the procedural protocols described in *Standard Operating Procedure for Performing Site Profile Reviews* (SC&A 2004). Approved by the Advisory Board on March 18, 2004, SC&A's protocol reflects the following review criteria:

- (1) Completeness of data sources
- (2) Technical accuracy
- (3) Adequacy of data
- (4) Consistency with other site profiles
- (5) Regulatory compliance

Deficiencies pertaining to these review criteria are noted as "issues." Our review of the Ames Laboratory Site Profile identified a total of 23 issues. Twenty-two (22) of the issues are regarded as "findings" and represent deficiencies that may require correction due to their potential adverse impact(s) on dose reconstruction. A single issue designated as an "observation" corresponds to an issue with limited significance for affecting dose reconstruction.



The purpose of this review is to provide the Advisory Board with an independent assessment of issues that surround the Ames Site Profile. Findings identified in our review are expected to

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provide the Advisory Board with a **preliminary** overview of potential issues that may impact the feasibility of dose assessment.

SC&A's draft report with its preliminary findings will subsequently undergo a multi-step resolution process. Resolution includes a transparent review and discussion of draft findings with members of the Advisory Board's Work Group and select personnel representing NIOSH/Oak Ridge Associated Universities Team (ORAUT). This resolution process is intended to ensure that each finding is evaluated on its technical merit in a fair and impartial manner.

### 1.2 SUMMARY OF ISSUES

SC&A's review of ORAUT-TKBS-0055, Rev. 03 (ORAUT 2012a), principally focused on the following three sections of the Ames Laboratory Site Profile:

- Section 4.0 Occupational Environmental Dose
- Section 5.0 Occupational Internal Dose
- Section 6.0 Occupational External Dose

Of the 22 preliminary findings identified in this review, 9 findings pertained to Occupational Environmental Dose, 11 to Occupational Internal Dose, and 2 to Occupational External Dose.

A brief statement for each of the 22 findings is presented below. However, the reader is cautioned that nearly all findings are based on a substantial body of data in the text that is further supported by the attached appendices. A more complete understanding and judgment of merit in behalf of these findings may, therefore, require a full review of this draft report.

## 1.2.1 Occupational Environmental Dose

**Finding #1:** Derived environmental intakes of U and Th, as given in Table 4-7 of the TBD, are improperly referenced and appear without technical basis.

**Finding #2:** NIOSH provides no basis for the **"assumed" losses** of 0.1% of U and Th to the environment and fails to identify a value for resuspension.

**Finding #3:** NIOSH's selection of personnel at the Ames Laboratory Research Reactor (ALRR) Facility as the target population is inappropriate and results in dose estimates that are not claimant favorable.

**Finding #4:** NIOSH's selection of data from the 1961 survey conducted at the Synchrotron Facility defines fenceline dose rates that are 1 to 2 orders of magnitude lower than other measurements reported in the survey when beam direction was shifted from a **westerly** to **easterly** direction.

**Finding #5:** SC&A concludes that the 1961 survey measurements, which were limited to gamma dose rates, were incomplete and may have substantially underestimated total exposure by excluding the contribution of particulate radiation.

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**Finding #6:** Statements in Section 4.3 of the Ames Site Profile are factually incorrect. For all years for which **section** reported environmental sampling data (1974, 1975, 1976), there were two sampling sites (not one, as stated by NIOSH) that provided data in behalf of air concentrations and corresponding dose estimates.

**Finding #7:** The nearly "instantaneous" 100-fold reduction of U and Th environmental intakes that represent the transition of the uranium and thorium metal production facilities at the end of 1953 to research and development (R&D) facilities in 1954 are improperly modeled. Also not included in the model are the contribution of blowouts to environmental contamination and the persistence of these radionuclides in the environment post-1953.

**Finding #8:** Environmental intakes cited in Table 4-7 of the Ames technical basis document (TBD) are based on unsupported assumptions and model parameters, which moreover are inconsistent with parameter values used to model worker intakes inside the hot laboratory described in Section 5.0 of the TBD.

**Finding #9:** Uranium and Th blowouts represent significant environmental events that should be included in Section 4.5 of the Ames TBD for the assessment of environmental exposures.

## **1.2.2 Occupational Internal Dose**

**Finding #10:** Available empirical bioassay and air-sampling data for Annex 1 workers are substantially higher than modeled/surrogate data assigned by NIOSH.

**Finding #11:** NIOSH further minimized the intake value of 853 pCi/d for Annex 1 production workers by assigning the "distribution" as a constant.

**Finding #12:** Default intake rates defined in Table 5-8 of the Ames TBD are improper for absorption Types F or S.

**Finding #13:** The scaling of uranium intake values based on (1) facility and (2) job function is without technical support and conflicts with statements given in the Ames Site Profile.

**Finding #14:** Although NIOSH briefly acknowledged the occurrence of "frequent fires and explosions" associated with the production of uranium metal, no attempt was made to assess potential intakes of these episodic events.

**Finding #15:** Technical Basis for Estimating the Maximum Plausible Dose to Workers at Atomic Weapons Employer Facilities, ORAUT-OTIB-0004, Rev. 03 (ORAUT 2006), is referenced for estimating non-operational intakes. OTIB-0004 was canceled before Rev. 03 of the Ames Site Profile (ORAUT 2012a) was issued. Moreover, the much higher intake values for inhalation and ingestion during non-operating years (i.e., 1954–1976) are inconsistent with intake values for operating years (1942–1953) as given in Table 5-8 of the Ames TBD.

**Finding #16:** NIOSH's approach for deriving estimates of thorium intakes from residual contamination post-1954 does not make the best use of survey data reported by Klevin (1952) and use of ORAUT-OTIB-0070, Rev. 01 (ORAUT 2012b).

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**Finding #17:** Given the availability of credible data, NIOSH's exclusion of thorium exposure in dose reconstruction is not justified.

**Finding #18:** Due to the fact that very little is known about the design features and technical specifications of the Hot Lab and the absence of worker monitoring/facility survey data, the applicability of NUREG-1400 (Hickey et al. 1993), Section 1.2, for use in dose reconstruction lacks technical merit and credibility for a facility that operated between 1943 and 1951.

**Finding #19:** Although Section 5.4.2 of the Ames TBD is titled, "Fission Product Intakes from Research Reactor Operations and Decontamination and Decommissioning," NIOSH restricted intakes to two activation products that are arbitrarily based on a 1977 Energy Research and Development Administration (ERDA) gross beta air concentration limit.

## **1.2.3 Occupational External Dose**

**Finding #20:** By means of documented anecdotes/testimonials regarding potential frequencies of blowouts, technical data for a specific blowout documented at Feed Materials Production Center (FMPC), and reasonable assumptions, SC&A derived significant U and Th intakes and associated organ doses that are applicable to workers at the Ames Laboratory, but were not considered/included in ORAUT-TKBS-0055 (ORAUT 2012a).

**Finding #21:** SC&A's concerns about the use of the same surrogate data sources and questionable assumptions for deriving external dose for Ames' workers exposed to uranium closely parallel those related to uranium intakes as cited in Findings #10 and #13.

**Findings #22:** Given the availability of highly credible and site-specific data for deriving external doses from thorium exposure, NIOSH's decision to exclude said exposures/doses is not justified.

### 1.3 ORGANIZATION OF THE REPORT

This draft report was written and organized as a "stand-alone" document by including summary information/data from various sources used to support our findings in the text of this report. For most of the 22 findings, additional information is provided by the attached appendices that either represent select portions or the full text of documents considered relevant to our findings.

Findings identified by SC&A are confined to Sections 4.0, 5.0, and 6.0 of this report and closely track the topical sequence of Sections 4.0, 5.0, and 6.0 of the Ames Laboratory Site Profile.

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# 2.0 INTRODUCTION AND SCOPE OF AUDIT

Under the Energy Employees Occupational Illness Compensation Program Act of 2000 (EEOICPA) and federal regulations defined in Title 42, Part 82, *Methods for Radiation Dose Reconstruction Under the Energy Employees Occupational Illness Compensation Program*, of the *Code of Federal Regulations* (42 CFR Part 82), the Advisory Board on Radiation and Worker Health (Board) is mandated to conduct an independent review of the methods and procedures used by NIOSH and its contractors for dose reconstruction. As contractor to the Board, S. Cohen and Associates (SC&A, Inc.) has been charged to support this effort by independently evaluating site profiles as requested by the Board that correspond to specific facilities at which energy employees worked and were exposed to ionizing radiation.

In a teleconference held by the Advisory Board on February 7, 2013, SC&A was tasked to review/evaluate the Site Profile for Ames Laboratory (ORAUT-TKBS-0055, Rev. 03), which was issued on January 3, 2012. The Board's request for the review of ORAUT-TKBS-0055, however, was made under circumstances that differed from previous reviews performed by SC&A, as briefly explained below.

# 2.1 A SUMMARY OF EVENTS THAT PROMPTED REVIEW OF ORAUT-TKBS-0055, REV. 03

<u>Revisions to the Ames Site Profile</u>. The first Site Profile for Ames Laboratory, ORAUT-TKBS-0055, Rev 00 (ORAUT 2007a) was issued on June 22, 2007. Since then, the Ames Site Profile was revised four times: Rev. 00 PC-1 on August 20, 2008 (ORAUT 2008); Rev. 01 on December 18, 2009 (ORAUT 2009); Rev. 02 on January 14, 2011 (ORAUT 2011); and Rev. 03 on January 3, 2012 (ORAUT 2012a).

While some of the aforementioned revisions resulted in an increase in assigned dose, others decreased the dose, and still others decreased in an earlier revision and increased in a subsequent revision, as summarized below:

- Revision 01 increased uranium intakes for researchers in the Ames Chemistry Building from August 1942 through December 1953. This change remained in Revisions 02 and 03. Revision 01 also added lateral (LAT) exposure dose estimates to occupational medical exposure.
- External dose for unmonitored workers for some job categories before 1946 increased in Revision 01, remained the same in Revision 02, but increased again in Revision 03.
- External dose for unmonitored workers between 1946 and 1953 decreased for **all** job categories and locations in Revision 01, remained the same in Revision 02, but increased in Revision 03.
- Revision 02 increased uranium intakes for **all** employees in the Chemistry Building for the period January 1954 through May 1976. These higher intakes remained unchanged in Revision 03. **All occupational medical exposures were eliminated in Revision 02** and Revision 03.

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<u>Special Exposure Cohort (SEC) Classes Added</u>. During this time period, NIOSH added classes to SEC-00038 in 2006 (NIOSH 2006), SEC-00075 in 2007 (NIOSH 2007), and SEC-00166 in 2010 (NIOSH 2010) to cover three separate groups of employees based on work location and job description. While the classes added in 2006 and 2007 included specific workers performing specific tasks in designated buildings, the 2010 class determined that the information available about worker job description, work location, or movement about the site was insufficient to determine if an employee worked in the affected area(s).

In 2011, NIOSH designated a fourth class [SEC-00185 (NIOSH 2011)] that encompasses all previous Ames SEC periods from August 13, 1942, to December 31, 1970, and designates all Ames employees (including predecessor agencies, contractors, and subcontractors) who were employed for a number of workdays aggregating at least 250 workdays. In Section 1.3.4 of ORAUT-TKBS-0055, Rev. 03 (ORAUT 2012a), NIOSH states the following:

... This SEC, SEC-00185 does not make any new information available for the feasibility of performing dose reconstructions between the dates of August 13, 1942, and December 31, 1970. ...

... NIOSH has determined that site-specific and claimant-specific data available for Ames Laboratory for this **entire** period are insufficient to enable it to determine that a **specific work group was not** potentially exposed to radioactive material releases or possible subsequent contamination. Based on this information, NIOSH has determined that the previously proposed SEC class definitions cannot be based on or limited to job titles or duties. [Emphasis added.]

It must also be noted that SEC-00185 was revised to change the covered period's start date from January 1, 1942, to August 13, 1942, the start of the Manhattan Engineer District (MED), known later as the Manhattan Project.

In behalf of these four SEC classes, NIOSH in Revision 03 of ORAUT-TKBS-0055 (ORAUT 2012a) provides the following statements and guidance for the dose reconstruction of Ames workers who were employed after December 31, 1970, as well as for workers who were employed prior to 1970 but who are not eligible for inclusion in SEC-00185:

Although NIOSH cannot bound doses for certain areas and periods as described in the SECs, internal and external data that become available for an individual claim (and that can be interpreted using existing NIOSH dose reconstruction processes or procedures) and applicable dose reconstruction methods that are defined in Sections 4, 5, and 6 of this site profile, will be used to complete partial dose reconstructions for workers who worked during the SEC periods but are not eligible for the SEC.

This site profile provides internal and external exposures that might coincide with work periods that fall within the SEC periods. There are varying types of exposures that can be applicable during the SEC periods to dose reconstructions for employees who do not qualify for the SEC(s). The periods in which doses can

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and cannot be reconstructed for a particular claim should be identified when writing the Dose Reconstruction Report. For dose reconstruction of claims with employment during the SEC period (see Section 1.3.4), **all Ames employees are included** and the dose reconstruction is a "partial reconstruction" by default. This designation should be included in the Dose Reconstruction Report. [Emphasis added.]

Tables 2-1, 2-2, and 2-3 summarize NIOSH's assessment regarding the feasibility of performing partial dose reconstructions from internal and external exposures for workers **ineligible** for SEC status. Workers **ineligibility** for SEC status may be due to (1) cancer type, and/or (2) employment prior to 1970 of fewer than 250 workdays.

For ineligible workers, a partial dose reconstruction may, therefore, be based on internal and external sources defined in the second column of Tables 2-1, 2-2, and 2-3 and described in Sections 4, 5, and 6 of Revision 03 of the Ames Site Profile (ORAUT 2012a).

Table 2-1. Feasibility Findings for SEC-00038, August 13, 1942 throughDecember 31, 1954

Source of Exposure	Dose Reconstruction is Feasible	Dose Reconstruction is NOT Feasible
Internal dose:		
Uranium	X	
Thorium/plutonium		Х
Thoron		Х
External dose:		
Uranium beta/gamma	X	
Thorium/plutonium beta/gamma		X (except 1953 and 1954)
Neutron		Х
Occupational medical x-ray	X	

# Table 2-2. Feasibility Findings for SEC-00075, January 1, 1955 throughDecember 31, 1970

Source of Exposure	Dose Reconstruction is Feasible	Dose Reconstruction is NOT Feasible
Internal		
Th-232 and progeny		Х
Ambient environmental	Х	
External		
Gamma	Х	
Beta	Х	
Neutron	N/A	
Ambient environmental	Х	
Occupational medical x-ray	Х	

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# Table 2-3. Feasibility Findings for SEC-00166, January 1, 1955 throughDecember 31, 1960

Source of Exposure	Dose Reconstruction is Feasible	Dose Reconstruction is NOT Feasible
Internal dose:		
Uranium and progeny	Х	
Thorium and progeny	Х	
Other radionuclides (Research		Х
Building)		Λ
External dose:		
Gamma	Х	
Beta	Х	
Neutron	Х	
Occupational medical x-ray	Х	

### 2.2 SCOPE AND OBJECTIVES OF AUDIT

SC&A's past approach to the review of other site profiles closely followed the protocol described in *Standard Operating Procedure for Performing Site Profile Reviews* (SC&A 2004). For the Ames Laboratory Site Profile, this standard review process is considered inappropriate due to the fact that, for earlier time periods of facility operations, Ames workers were either not monitored for exposure or available records pertaining to worker monitoring; claimant-specific job duties and work locations; process descriptions and source-term data were insufficient to perform **complete** dose reconstructions. Hence, for the period of August 13, 1942, to December 31, 1970, NIOSH recommended the addition of the SEC-00185 class that includes all workers for all areas of the Ames Laboratory.

SC&A's review of the Ames Laboratory Site Profile for the period of August 13, 1942, through December 31, 1970, will, therefore, assess and evaluate Sections 4, 5, and 6 of the Ames Site Profile, as well as all claimant-specific and facility-specific data that are available and considered appropriate for a partial dose reconstruction in behalf of individuals who do not qualify for inclusion in the SEC. This may include the review of documents that under more normal circumstances would have a low priority among other available sources and would, therefore, not be given consideration for use in dose reconstruction. Among these are a 1952 Atomic Weapons Employer (AEC) survey (Klevin 1952), personal memoirs/manuscripts, worker interviews, and memoranda that provide a critical insight on attitudes, work conditions/practices, and events that surround the Ames Project.

For facility operations after 1970, SC&A's review of Sections 4, 5, and 6 of ORAUT-TKBS-0055, Revision 03, will follow the traditional protocols (SC&A 2004) with the following review objectives:

- (1) Completeness of information and data sources for all workers and time periods of facility operation
- (2) Technical accuracy and reliability of the data
- (3) Claimant favorability in instances when assumptions, surrogate data, and/or coworker models are used

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### (4) Consistency with other site profiles

(5) Compliance with Federal Regulations pertaining to EEOICPA and with guidance documents established by NIOSH and its contractors

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# 3.0 RELEVANT BACKGROUND INFORMATION

### 3.1 SITE/FACILITY DESCRIPTIONS

The Ames Laboratory is located on the Iowa State University (ISU) campus in Ames, Iowa. Although the Ames Laboratory was not formally established until 1947 by the AEC, work at Iowa State College began in 1942 when Dr. Frank Spedding, a professor of physical chemistry at the college, agreed to establish and direct a chemical and metallurgical research program that complemented the MED's Metallurgical Laboratory in Chicago.

In the beginning, the Ames Project was first and foremost an academic laboratory that was concerned with metallurgical research, studies on physical/chemical properties that investigated such parameters as melting points/viscosities of metals, chemicals reactivity, and separation of select fissile metal compounds and their reduction to pure elemental metals. The vast majority of personnel involved in research activities were academics that included professors, graduate students, and technical support personnel with varying knowledge in chemistry and other sciences.

Within months, Ames researchers were successful in developing a chemical reduction process for converting uranium tetrafluoride (UF<sub>4</sub>) to a highly purified uranium metal. Because this process was efficient and cost effective, the Ames Project was expanded to include the largescale production of purified uranium and later thorium metal. Thus, research and production became an integrated operation in which the production processes were an extension of research efforts, and continuing research reflected failures and problems encountered at the production level. To some extent, problems encountered in the production phase were likely affected by the need to hire workers from the local community who were neither familiar with the hazardous nature of materials employed nor the basic protocols of industrial safety.

## 3.1.1 Uranium and Thorium Facilities

For research and development and for the production of uranium and thorium metal, a total of four campus buildings were utilized by the Ames Project I. These included the following:

- <u>Chemistry Building</u> (Gilman Hall) was utilized for analytical research and process development
- <u>Physical Chemistry Annex I</u> was used for the production of 1,000 tons of uranium ingots
- <u>Physical Chemistry Annex II</u> was used for the recovery of uranium turnings and production of 300 tons of uranium ingots
- <u>Metallurgical Building</u> (Wilhelm Hall) was used for various research and the production of thorium metal

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## 3.1.2 Other Facilities

Other facilities with significant potential for radiation exposure included the following:

- <u>Physics Building</u> was used for R&D that supported other work at Ames. Work included the development of analytical equipment, the operation of an 80-MeV synchrotron, and studies of nuclear fission and the shielding properties of various materials.
- <u>Research Building</u> (Spedding Hall) started operation in 1951 and was used to investigate the properties of various metals. This facility included a 150-kV accelerator that produced 14 MeV neutrons; a hot canyon and hot cell; and a glovebox.
- <u>Synchrotron Building</u> housed two electron accelerators (up to 80 MeV) that upon interaction with a target produced high-energy gammas that in turn interacted with nuclei to release neutrons, protons, and alphas. This facility operated between 1949 and 1971 and was decommissioned in the early 1990s.
- <u>Ames Laboratory Research Reactor</u> (ALRR) was a 5-MW, heavy-water-moderated reactor that employed 93% enriched U-235 and started operation in 1965. Research included various material studies and analysis of decay products of nuclear fission. The reactor operated until 1977 and was decommissioned in 1981. The primary radiological concern at this facility was airborne tritium.

### 3.2 MAJOR OPERATIONS AND PROCESSES

The two major operations that contributed to radiation exposures among personnel at the Ames Laboratory involved the production of purified uranium (1942–1945) and thorium metal (1945–1953).

## 3.2.1 Uranium Metal Production

<u>Chemistry Building</u>. Initial protocols for the purification of uranium metal were tested in the **Chemistry Building**. The Chemistry Building consisted of a basement and first floor. Besides basic chemical and metallurgical research on uranium and its compounds, a key research directive was to develop a chemical reduction process that would lend itself to the large-scale production of highly purified uranium metal.

Up until 1942, virtually no uranium metal in its pure form had been produced in significant quantities. At the time, uranium was generally available in the form of an impure uranium oxide that required initial purification by ether extraction. At the beginning of the Ames Project, it was thought that the oxides of uranium could be reduced to form a salt slag and a purified uranium metal. Early attempts to reduce uranium oxide by hydrogen, carbon, aluminum, magnesium, and calcium met with limited success and low yields, due to temperature melting problems of uranium oxide and interactions with crucibles used to contain the chemical mixture.

In these reduction experiments, oxygen presented the principal obstacle in reducing the uranium to pure metal. This obstacle was overcome in August 1942, when uranium oxide  $(U_3O_8)$  was

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replaced with  $UF_4$  (i.e., green salt) and successfully reduced with a calcium liner. Within weeks, research produced pure uranium ingots weighing several pounds. In total, about 4,000 pounds of purified uranium metal had been produced by the research staff in the Chemistry Building late in 1942.

The successful reduction of UF<sub>4</sub> to pure uranium metal by Ames researchers led to a contract with Iowa State to produce 100 pounds of uranium per day in a pilot plant until such time that companies like Mallinckrodt and DuPont could integrate the Ames process into their own plant.

Because research and process development of the Ames Project were performed/supervised by trained chemists, who at a minimum understood the chemical toxicity, volatility, and potentially explosive reactivity of materials under investigation, standard laboratory precautions were taken. Moreover, the Chemistry Building with its ventilation hoods was designed for work with hazardous materials. Thus, on a relative scale, radiological exposures (in particular internal exposures associated with inhalation and ingestion) were likely lower among researchers that included faculty members, graduate students, and laboratory support personnel.

This conclusion is based on personal interviews with Dr. F.H. Spedding, Director of the Ames Project, and others by Carolyn S. Payne and reported in her 1992 PhD thesis, "The Ames Project: Administrating Classified Research as a Part of the Manhattan Project" (Payne 1992). In one of the many interviews with Dr. Spedding, C.S. Payne (1992) reported the following comments:

As long as the chemists were involved in research with the various elements, typical laboratory precautions were taken . . . ventilation hoods were being used . . . [and] there were few examples of safety breaches or carelessness by the scientists at the Ames Project. The **production area** though presented quite a different problem. Scientists generally had security clearance, so they knew with what they were working . . . [but] workers often from the community had to be hired who were often unfamiliar with even routine practices. [Emphasis added.]

Dr. Spedding's comments were further supported by comments made by Dr. during during an interview with SC&A (Dr. A. Makhijani) on March 12, 2006 (see Appendix A for full transcript of interview).

*Dr. Makhijani: Did you go to Little Ankeny [where uranium metal was manufactured in large amounts]?* 

Dr. First I went to predecessor of little Ankeny. At this place they had a way of preparing a steel cylinder and putting a lime liner in it. Then they put in  $UF_4$  mixed with magnesium and put some lime freshly baked so as to decompose the hydroxide. That was the way the metal was prepared. Then they put in  $UF_4$ . After the heating, the reduction occurred and uranium metal separated out. Some of the guys who worked there refused to wear masks. They would wear them when the director was there and then take them off when he left. They were farm boys and did not worry about the green powder. There was this one fellow;

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<u>Annex I ("Little Ankeny"</u>). For the uranium production pilot plant, a small World War I wooden structure was selected. This facility that resembled a house was located on the southeastern edge of the campus, and at one time served as a women's gymnasium, a popcorn laboratory used by the college's agricultural department, and, in 1942, was used for storage.

Important to an understanding of radiological environments that were encountered by uranium (and subsequently thorium) production workers between the years 1943 and 1954 is a brief description of this building that would officially be called the **Physical Chemistry Annex I** and nicknamed by workers as "Little Ankeny" after a war munitions plant in Ankeny, Iowa.

Figures 3-1 and 3-2 show the simple wooden structure that, after a series of additions, became known as Annex I or Little Ankeny.



Figure 3-1. Physical Chemistry Annex (Little Ankeny) North View

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Figure 3-2. South View of Little Ankeny

In her PhD thesis, titled *The Ames Project: Administrating Classified Research as a Part of the Manhattan Project*, C.S. Payne (1992) provided the following description:

... The College gave the building to the project and immediately the chemists had the dirt floor in the garage area replaced with concrete so that casting could take place in this area. The chemists set up the reduction laboratory in the original part of the building where the popcorn lab had been located. The building shortly began to expand in a most curious pattern. The **porch was used for the especially dirty work**, the least secret of the process. However, when it became too cold to work on the porch, a canvas would be added followed by a crude set of walls and finally a new roof. A new porch appeared and the process repeated itself...

After the building became available, a machine shop at the production site became the second necessity. Wilhelm\* heard of a small machine shop owned and managed by Bill Maitland for sale in Ames west of Grand Avenue near the railroad. Maitland made garden tools normally, but he could no longer obtain the metal he needed because of war-time restrictions on material priorities. Wilhelm examined the shop contents and discovered that Maitland would sell all his tools and equipment for \$8,000. After consulting with Spedding, both men contacted Maitland and bought the entire shop, moving the equipment along with Bill Maitland to the campus production building. . . . [Emphasis added.]

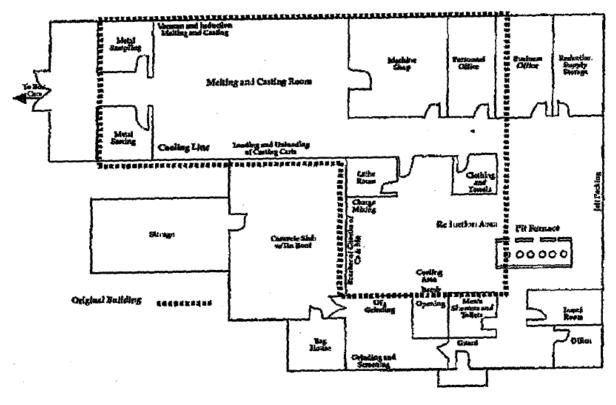
Production equipment, unlike lathes, motors, and small tools from Maitland, was much harder to procure. For example, reduction furnaces were especially hard

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to obtain. The small reduction furnace in the Chemistry Department used to produce most of the metal earlier was not big enough for a large-scale production plant. Luckily for the Ames operation, the Metallurgical Laboratory had ordered two 40,000 watt reduction furnaces for what they called "Site B," but when the Ames pilot plant needed to be established as a production facility, those furnaces were diverted to Ames. Mixers and grinders for processing metals like calcium and later magnesium and vacuum casting apparatus were also purchased from various producer...

\* Harley A. Wilhelm was a metallurgist and professor of chemistry at Iowa State and was Associated Director.

A floor plan of the original structure that, along with subsequent additions became known as Annex I or "Little Ankeny," is provided in Figure 3-3.





(Note: Heavy dotted line identified original buildings.)

Uranium metal production at Annex I began in January of 1943. Ames received the UF<sub>4</sub> from three sources—Mallinckrodt, DuPont, and Harshaw. Production rose from **3,600 pounds per week early in January 1943 to about 5,600 pounds in the last week of that month**. During this time, reduction of UF<sub>4</sub> to uranium metal was performed by means of either calcium or magnesium. However, by March, magnesium became the reductant of choice.

Peak production of uranium metal was reached in July 1943, with **130,000 pounds per month**. During peak production periods, **Ames operated 24 hours per day and 7 days per week**. By

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December 1945, a total of 2,000,000 pounds (or 1,000 tons) of uranium ingots had been produced at Ames, and UF<sub>4</sub> reduction and uranium metal casting shifted from Ames Laboratory to three companies that included Electromet, Mallinckrodt, and DuPont.

In addition to the reduction of  $UF_4$  to uranium metal, Figure 3-3 also identifies numerous other critical processes that were performed at the Annex I facility. These included the following:

- In preparation of the reduction process, UF<sub>4</sub> and Mg had to be "ground and chopped" into smaller pieces.
- UF<sub>4</sub> and Mg were subsequently screened, and ferrous contaminants were removed by magnets.
- Following a successful reduction and the formation of a uranium "biscuit," the biscuit was stamped and sent to the melting and casting room.
- In the casting process, a vacuum induction furnace heated/melted the biscuit in a crucible, which discharged the molten uranium into a graphite mold in the shape of rods that varied between 1.5 to 5 inches in diameter and 20–30 inches in length.
- A cropping was cut from one end of the rod for physical and chemical evaluation, and each rod was stamped with an ID number and crated for shipment.

Based on the above-cited information, it is reasonable to conclude that Iowa State's uranium production facility was neither designed for nor equipped with the necessary safety systems needed to process large quantities of uranium and minimize worker exposure, as acknowledged by C.S. Payne (1992) in the following statements:

There was a basic conflict trying to balance safety with accomplishing the work in time to win the war . . . [and] much of the early work was **not** done under the best of conditions . . . For example, most of the tools that had been obtained from Bill Maitland's garden shop in down-town Ames were hand-driven so power apparatus had to be adapted and added to them. Also many of the grinders, cutting mills, and machining tools were originally manufactured for other industrial purposes and naturally did **not** have all the necessary safety features for working with uranium. It took months to obtain fans that were needed for proper ventilation in the building and since much of the work took place in **hot months** without the luxury of air conditioning, **respirators** and **masks**, though required for particularly dusty work, were sometimes **discarded** for worker comfort. [Emphasis added.]

Failure to enforce basic safety precautions among process workers at Annex I was also acknowledged by a second process, who recalled the following:

I was acting as either **actions** or **actions** for a crew of from six to sixteen people. . . In a situation of that type it often falls on the immediate supervisor to make some decisions with his own judgment. I would say that we were perhaps guilty on erring on the side of, "well, lets get the job done and not worry too much about this or that safety rule". . . (C.S. Payne 1992)

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According to statements provided by Dr. F.H. Spedding in 1967 and acknowledged by C.S. Payne (1992), there were some people on the Ames Project who ignored all safety rules, as given in the following:



Beginning in 1943, Annex I was used for process development, as well as early production of thorium metal.

<u>Chemistry Annex II</u>. In September of 1943, concurrent with the production of uranium metal from virgin material, Ames developed a uranium recovery program from scrap metal turnings produced at Ames and at all other MED sites. Uranium recovery was done in the Chemistry Annex II Building, a one-story brick building, also known as the former Credit Union Building. In April 1944, this development work became a production operation.

In the recovery process, metal turnings were first collected in barrels and examined by hand to identify uranium turnings. Segregated uranium turnings were then subjected to a magnetic separator that removed smaller iron and other ferrous impurities before being sent to a cutting machine. After being washed, rinsed, dried, and again passed over a magnetic separator, the cleaned uranium turnings were pressed into briquettes (about 1-inch thick and 4.25 inches in diameter). Lastly, briquettes were taken to the casting room to be melted and cast into ingots.

Over a 2-year period, Ames recovered in excess of 600,000 pounds of scrap uranium using this process. In December of 1945, this recovery process was taken over by Metal Hydrides in Massachusetts and by a recovery facility at the Hanford Site in Washington.

## 3.3 THORIUM AND METAL PRODUCTION

<u>Metallurgy Building</u>. Interest in thorium (Th-232) as a source for producing the fissile U-233 by the Th-232 ( $\eta$ , $\gamma$ ) U-233 reaction began at Iowa State as early as 1943. Here too, the Ames Project served the dual role of analytical R&D and implementation of a pilot production program. Key research studies involved the chemical separation of thorium from uranium contained in monazite sand and the reduction of thorium oxide (ThO<sub>2</sub>) or ThF<sub>4</sub> to pure metal.

In 1944, preliminary tests successfully demonstrated that the addition of zinc chloride (ZnCl) as a booster resulted in the formation of a zinc-thorium alloy, which when heated to high temperatures under vacuum produced purified thorium metal.

Thorium biscuits were subsequently melted and recast either into ingots of up to 150 pounds or machined into suitable targets for use in atomic piles. By December 1946, Ames had produced 4,500 pounds of thorium metal. Research and development on thorium were conducted in both

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the Chemistry Building and Annex I. In other words, during 1944 and 1945, thorium production took place in the same building as large-scale uranium production; thereafter, production shifted to the Metallurgy Building until 1954, when production was halted. In total, 65 tons of purified and recast thorium metal were produced under the Ames Project.

Table 3-1 summarizes major activities and locations for the Ames Project between 1942 and 1954, when metal production/casting ceased.

Building	Dates	Major Activities
Chemistry Building	1942–1954 ?	• Basic chemical/metallurgical research pertaining to uranium, thorium, plutonium, beryllium, cerium
	1942–1945	<ul><li>Chemical/metallurgical research on uranium</li><li>Process development for uranium metal production</li></ul>
	1943–1944	<ul><li>Process development for uranium recovery from turnings</li><li>Thorium reduction experiments and process development</li></ul>
Annex I	1943–1945 1943–1949	<ul> <li>Uranium metal reduction/production and casting of uranium</li> <li>Process development and early production of thorium metal</li> </ul>
Annex II	1944–1947	• Recovery and casting of uranium metal from turnings
Metallurgy Building	1947–1954	Production and casting of thorium metal

 Table 3-1. Major Activities Associated with the Ames Project

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## 4.0 REVIEW OF SECTION 4.0 OF THE AMES SITE PROFILE: OCCUPATIONAL ENVIRONMENTAL DOSE

In the <u>Introduction</u> of Section 4.0, NIOSH provides the following comments:

Occupational environmental dose refers to the dose received by workers **on the** site but outside facilities (e.g., buildings). . . .

Occupational environmental dose was not measured (direct radiation dosimeters) until 1953, when workers were badged (Martin 2006a,b), and it was **not** calculated from **environmental media** concentrations until 1962. **Sources** of potential environmental exposures (releases to the environment) were not measured until 1962 (Voss 1963). [Emphasis added.]

Sections 4.1 through 4.5 of the Ames TBD provide estimates of environmental external and/or internal exposures from the following source terms during their times of operation:

- Uranium/Thorium Production Period (1942–1952)
- Synchrotron Operations (1949–1971)
- Ames Laboratory Research Reactor (ALRR) Site (1965–1977)
- Ames Laboratory R&D Activities (1942–Present)
- Significant Environmental Event

Based on various assumptions and models, NIOSH estimated/derived annual external doses (mrem/yr) and daily intakes (pCi/d) that are presented in Tables 4-6 and 4-7 of ORAUT-TKBS-0055 (ORAUT 2012a) and reproduced below as Tables 4-1 and 4-2.

#### **Table 4-1. Environmental External Doses**

Location	Dates	Dose (mrem/yr) <sup>a</sup>	Distribution	
Skyshine from Synchrotron Building	1949–June1971	25	Constant	
Ar-41 from ALRR	1965–1977	1	Constant	
All other R&D Buildings (see list in Section 4.4)	All	Negligible	N/A	

(To be applied only to unmonitored workers)

<sup>a</sup> The energy range for all environmental external dose is assumed to be 100% 30–250 keV.

Dates Radionuclide/adsorption		Intake (pCi/d)	Distribution
August 1942–1953	U (assume U-234); Type M or S	5	Constant
1954-present	U (assume U-234); Type F,M, or S	0.05	Constant
June 1954–April 1953	Th-232, Ra-228, Th-228; all Type M	0.07 each	Constant
1954-present	Th-232, Ra-228, Th-228; Type M or S	0.0007 each	Constant
1965–1977	Tritium	2,700	Constant
1943–1981	Fission products per Table 4-2	Per Table 4-2	Constant

<sup>a</sup> Apply the environmental intakes in this table if no occupational intakes are applied for the same radionuclide and the same period in accordance with the instructions in Table 5-8.

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Presented below is SC&A's assessment of these environmental exposure estimates for credibility, claimant favorability, and completeness.

### 4.1 URANIUM AND THORIUM INTAKES

Section 4.1 of the Ames TBD offers the following explanation for the derived upper-bound environmental intakes of (1) 5 pCi/day for U (modeled as U-234, Type M or S) and (2) 0.07 pCi/day each for Th-232, Th-228, and Ra-228 for years 1942–1954:

There were no documents found that stated the room and hood ventilation stacks on the facilities had filters. **Concentrations of uranium dust were measured in the operation rooms (Voss 1978)**. To estimate a bounding dose outside the facilities, it was **assumed** that losses of 0.1% of the uranium or thorium as dust in a facility were emitted continuously and dispersed from ground level in accordance with local and regional meteorological conditions (see Figure 4-1) (Voss 1981) and a standard Gaussian atmospheric dispersion computer model (Napier et al. 2004). When **resuspension** is included, the daily intake rate of uranium (modeled as <sup>234</sup>U, type M or S) was 5 pCi. This intake applies outside Physical Chemistry Annex 1 for August 1942 through August 1945 and outside Physical Chemistry Annex 2 for 1944 through 1954. In addition, daily intakes of <sup>232</sup>Th, <sup>228</sup>Th, and <sup>228</sup>Ra at 0.07 pCi each apply to Annex 1 for June 1946 through 1949 and to the Metallurgy Building for 1950 through April 1953. [Emphasis added.]

### SC&A's Comments and Findings

From the statement cited above, SC&A interprets the derivation of 5 pCi/d for U-234 intake and the 0.07 pCi/d intake for Th-234, Th-228, and Ra-228 to be principally based on the following:

- (1) <u>Measured</u> uranium dust in operations rooms (associated with Annex 1, Annex 2, and Metallurgy Building), as reported by Voss (1978);
- (2) An assumed continuous loss of 0.1% to the outside; and
- (3) The "inclusion of resuspension."

A review of Voss (1978), *Environmental Monitoring at Ames Laboratory Calendar Year 1978*, reveals a 58-page report that as its title suggests is limited to environmental sampling results for the year 1978. The focus of the 1978 sampling effort includes liquid and air releases of fission/activation products from the ALRR. The report neither makes reference to uranium and thorium metal production nor cites data for air concentrations for indoor air, as implied by NIOSH.

**Finding #1:** Derived environmental intakes of U and Th, as given in Table 4-2 above are improperly referenced and appear without technical basis.

For a more credible derivation of thorium values, SC&A recommends the use of data cited in a 1952 AEC survey (Klevin 1952). Klevin (1952) reported (1) a daily weighted thorium air

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concentration of 530 dpm/m<sup>3</sup>, and (2) a daily weighted maximum thorium air concentration of  $3,100 \text{ dpm/m}^3$  for the Metallurgical Building (see page 3 of Appendix B).

**Finding #2:** NIOSH provides no basis for the **"assumed" losses** of 0.1% of U and Th to the outside and fails to identify a value for resuspension.

### 4.2 SYNCHROTRON OPERATIONS

Although the synchrotron operated from 1949 to 1971, no dosimeters were provided to staff at the facility until after 1952, and no routine  $\gamma/\eta$  dose-rate measurements were made outside of this facility. In 1961, a facility survey was conducted that included a gamma dose-rate survey at the fenceline that surrounded the Synchrotron facility (Ames 1961). To estimate external exposure from the operation of the synchrotron, NIOSH stated the following:

The ALRR (now the Applied Science Complex) is about 750 ft from the nearest part of the Synchrotron Building, and construction or operations at the two facilities overlapped, at a maximum, from 1962 to 1971. Skyshine from both **heavy particles** and **photons** decreases at rates equal to or greater than the reciprocal of the square of the distance (1/r2) from accelerator facilities (NCRP 2003). If it is conservatively assumed that all of the radiation field measured in the 1961 survey was from skyshine, the dose rate at the ALRR would have been less than 0.13 mrem/hr during the worst-case operation and less than 0.013 mrem/hr during routine operations. On the main campus of the University, the dose rate would have decreased to less than about 0.00025 mrem/hr during routine operations.

It is favorable to claimants to use the environmental external dose from synchrotron operations at the ALRR for full-time exposure (2,000 hr/yr) for all locations at 25 mrem/yr for the period from 1949 to 1971. [Emphasis added.]

Fenceline readings used by NIOSH are defined in Table 4-1 of the Ames Site Profile and as a convenience to the reader are reproduced herein as Table 4-3. As noted in Table 4-3, fenceline doses corresponded to the beam directed **west**.

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# Table 4-3. Survey of Fenceline Gamma Dose Rates around the Synchrotron Facility with<br/>the Beam Directed West

Location	Dose rate (mrem/hr) <sup>a</sup>	Location	Dose rate (mrem/hr) <sup>a</sup>		
1	0.75-1	14	1.8		
2	3.5	15	1.8		
3	7	16	1.8		
4	6	17	1.6		
5	5.5	18	1.5		
3	7	19	1.5		
7	4.75	20	1.75		
8	3.5	21	1.6		
9	2.6	22	1.6		
10	1.5	23	1.5		
11	2	24	0.75-1		
12	2	25	4.5		
13	1.8				

<sup>a</sup> Background reading in the beam direction (see location 3-6) before turning on the beam was 0.5 to 1.0 mrem/hr.
Source: Ames 1961, p. 7

#### SC&A's Comments and Findings

SC&A questions NIOSH's selection of model parameters, as well as the claim that, "It is favorable to claimants to use the environmental external dose from synchrotron operations at the **ALRR for full-time exposure (2,000 hr/yr) for all locations** at 25 mrem/yr for the period" [Emphasis added.] for the following reasons:

 <u>Selection of the target population</u>. In Section 4.0, NIOSH defined Occupational environmental dose as ". . . the dose received by workers on the site but **outside facilities** (e.g., buildings)." [Emphasis added.]

Based on this definition, the target population would be limited to maintenance workers, groundskeepers, security personnel, etc., who were assigned fulltime to **outdoor** duties at the ALRR at a distance of 750 feet from the source.

If such persons existed at the ALRR, they would also have existed at much closer distances that include areas at or within the fenceline that **surrounds the Synchrotron Facility** (see Exhibit #4-1).

Distances from the Synchrotron to the fenceline that surround this facility are included in the 1961 survey data and are provided here as Exhibit 4-2. For the 25 survey points, fenceline distances range from **55 to 325 feet** and with dose rates ranging from <1 mR/hr to 7 mR/hr. For a full-time exposure (2,000 hr/yr), these dose rates would convert to annual doses of 2,000 mR to 14,000 mR. Dose rates within the fenceline must, therefore, be assumed even higher.

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Exhibit 4-1. 1961 Survey Synchrotron

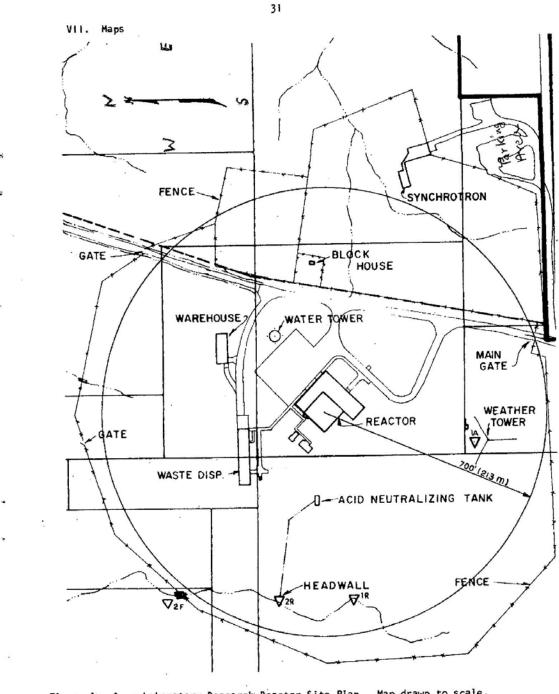


Figure 1: Ames Laboratory Research Reactor Site Plan. Map drawn to scale.

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Exhibit 4-2.	Fenceline Do	se Rates for	r Synchrotron	Beam Directed	West

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(2) <u>Choice of Survey Data</u>. Section 4.2 of the Ames Site Profile states:

The fenceline gamma dose rates measured in the radiation survey ([Ames 1961]) were **worst case** and were produced with the maximum beam current on a target and **direction** that would produce maximum dose rates at the fenceline [Attribution#2]. ... [Emphasis added.]

SC&A's review of the Ames (1961) survey data, however, identified other dose-rate measurements that are substantially higher than those selected by NIOSH. Exhibit 4-3 identifies dose rate measurements:

... recorded with the main beam projected northeast-ward direction. The beam was detected along the cyclone fence 55' west of the northeast corner of the fence that surrounds the synchrotron site. The power level during the run of this survey was 60 to 65 MeV. [Emphasis added.]

Exhibit 4-3 identifies multiple **fenceline** dose rates greater than 100 mrem/hr, which implies that NIOSH's "claimant-favorable" dose of 25 mrem per **year** would have been received in less than **15 minutes** spent at these locations.

(3) <u>Accuracy and Completeness of Survey Data</u>. Footnote "a" in Table 4-1 in ORAUT-TKBS-0055 (ORAUT 2012a) (and reproduced above as Table 4-3) states that "Background reading in the beam direction (see locations 3-6) **before** turning on the beam was 0.5 to 1.0 mrem/hr" [Emphasis added].

<u>Accuracy</u>. It is uncertain what this "background reading" represents, since this would translate to an **annual** dose between 4,400 and 8,900 mrem, which is more than 100-fold higher than ambient dose rates in the U.S. On the assumption that the observed background levels of 0.5 to 1 mrem/hr were correct, one explanation may be the contribution of stored radioisotopes produced at the synchrotron.

<u>Completeness</u>. Survey dose-rate measurements cited in the 1961 survey (Ames 1961) were limited to gamma radiation. As stated in Section 2.1.8 of the Ames Site Profile:

... The synchrotron room housed two electron accelerators that could project electrons up to **80 MeV** onto a target, which produced **highenergy gamma rays** that interacted with nuclei to release **neutrons**, **protons**, and **alpha** particles (Ames 1967). The accelerators were operated from a Control Room where there was a safety gate that prohibited access to the synchrotron room when the beam was on (Ames 1967). In many cases, the products of these reactions were **radioactive**, ... In addition, the synchrotron was used to probe nuclear structures and to provide **radioisotopes** for nuclear spectroscopy. .. [Emphasis added.]

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Findings Associated with Environmental Exposures Associated with the Operation of the Synchrotron

**Finding #3:** NIOSH's selection of personnel at the ALRR Facility as the target population is inappropriate and results in dose estimates that are not claimant favorable. At a minimum, survey data/dose rates cited in Exhibit 4-2, which reflect Synchrotron fenceline measurements, should be considered.

**Finding #4:** NIOSH's selection of data from the 1961 survey conducted at the Synchrotron Facility defines fenceline dose rates that are 1 to 2 orders of magnitude lower than other measurements reported in the survey when beam direction was shifted from **westerly** to **easterly** direction. For bounding estimates of environmental doses, fenceline data cited in Exhibit 4-3 should be considered.

**Finding #5:** SC&A concludes that the 1961 survey measurements, which were limited to gamma dose rates, were therefore incomplete and may have substantially underestimated total exposure by excluding the contribution of particulate radiation.

### 4.3 AMES LABORATORY RESEARCH REACTOR OPERATIONS (1965–1977)

The 5-MW, heavy-water-moderated research reactor is about 1.5 miles northwest of the ISU campus. Operation of the reactor resulted in airborne emissions of H-3, Ar-41, and small quantities of fission and activation products.

In behalf of environmental exposures associated with ALRR operations, Section 4.3 of the Ames Site Profile states the following:

... not all workers at the ALRR have recorded doses. In addition, environmental doses from gaseous effluents released from the operating reactor were not monitored. However, environmental doses to the public from airborne releases were calculated and reported (Voss 1975, 1976, 1977). The only air monitoring station in the vicinity of the reactor was on the roof of the reactor building, as shown in Figure 4-3.

Environmental Monitoring at Ames Laboratory: Calendar Year **1974** was the first annual report to provide gamma spectroscopy of environmental media samples (Voss 1975); subsequent annual reports (Voss 1976, 1977) provided similar results. The average release estimates from these reports were used to determine the estimated environmental dose to offsite workers from gaseous releases from reactor operations. From the effluent data, it was shown that the contribution to radioactivity in air from ALRR operations consisted principally of <sup>41</sup>Ar and tritium (Voss 1975, 1976, 1977). An atmospheric dispersion model, which used annual average meteorological data for Ames and an exposure model (Napier et al. 2004; [Napier 2006]), was used to determine external dose rates from the <sup>41</sup>Ar and inhalation intake estimates for the tritium.

*At the fenceline location with the highest dose from gaseous effluents*, the average annual dose to a person for the entire year (8,760 hours) was estimated

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to be 4.2 mrem from  ${}^{41}$ Ar during the years of reactor operation. An offsite worker who worked full time at this location would not have been exposed for more than 2,080 hr/yr, which would result in a submersion dose of about **1 mrem/yr**.

At the fenceline location with the highest concentration of tritium effluents, the average annual intake of tritium to a person for the entire year (8,760 hours) was estimated to be about 2.9  $\mu$ Ci/yr during the years of reactor operation. An offsite worker who worked full time at this location would not have been exposed for more than 2,080 hr/yr, which would result in an intake of tritium of about 0.7  $\mu$ Ci/yr or 2,700 pCi/d. [Emphasis added.]

## SC&A's Comments and Findings

SC&A reviewed the referenced annual environmental reports, including the first, *Environmental Monitoring at Ames Laboratory: Calendar Year 1974* (Voss 1975). Contrary to statements contained in Section 4.3 of the Ames Site Profile (and cited above), Voss (1975) provided the following statements and data:

## From Section B. Environmental (pp. 17 and 18 of Voss 1975):

Air samples are collected from **two different locations** at the Ames Laboratory on a daily (except weekends) basis for the purpose of monitoring the particulate radioactivity content of the atmosphere. **The sites are located at the ALRR weather tower site and the roof of Spedding Hall**. . . . The specific isotopes for which the filter papers are examined are: Be-7, Ce-144, Cs-137, Nb-95, Ru-106, plus any other unknown which might be observed. [Emphasis added.]

Exhibits 4-4 and 4-5 identify the two air sampling locations as: **1A** for the **Weather Tower** location, which is about 500 feet from the ALRR, and **2A** for Spedding Hall, which is just beyond the 1-mile radius of the ALRR.

Summary data for airborne activity levels for these two sampling locations are cited in Tables 7 and 8 of Voss (1975) and reproduced herein as Exhibit 4-6. The data imply that at Site 2a (Spedding Hall), which is more than 1 mile from the ALRR, air concentrations were substantially higher than at the 1A location within the fenceline perimeter of the ALRR.

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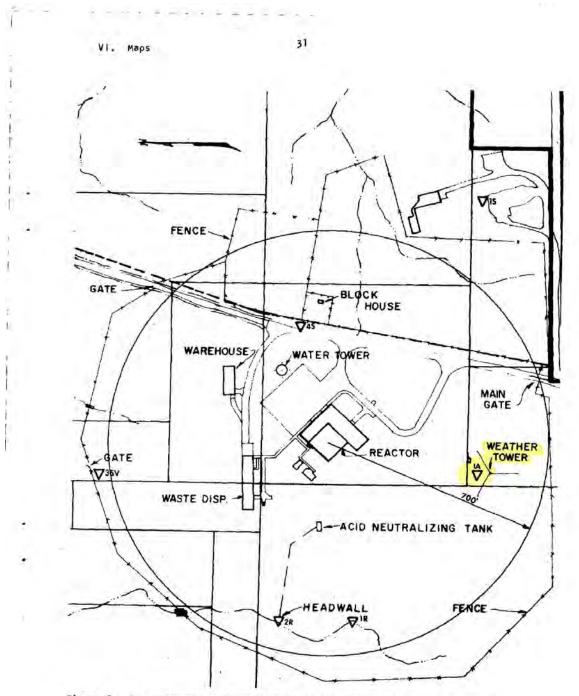


Exhibit 4-4. Air Sampling Locations for 1A for the Weather Tower

Figure 1: Ames Laboratory Research Reactor Site Plan. Map drawn to scale.

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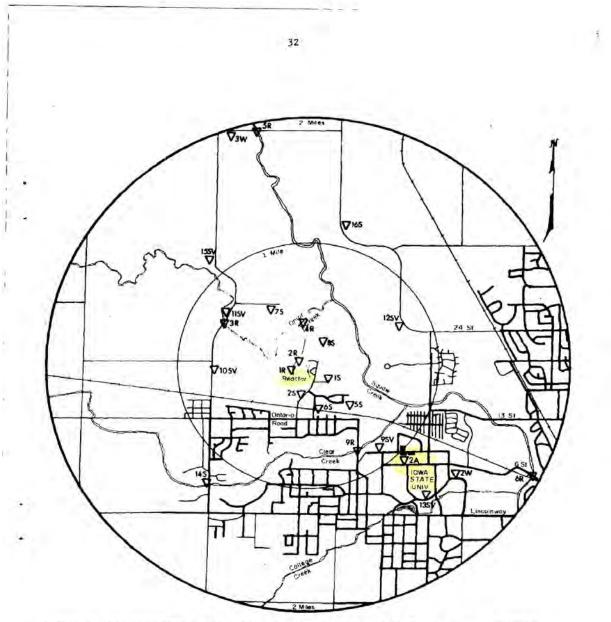


Exhibit 4-5. Air Sampling Locations for 2A for Spedding Hall

Figure 2: Environmental Sampling Sites Within Two Miles of ALRR. Map drawn to scale.

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### Exhibit 4-6. Summary Data for Airborne Activity Levels for These Two Sampling Locations are Cited in Tables 7 and 8 of Voss (1975)

Table 7 Gamma Analysis of Air Samples (10 <sup>-15</sup> µci/ml)					
Isotope	ALRRSi Ave.	te IA % CG	Spedding' Ave,	HallSite 2A % CG	
Be-7	90.70	$2.27 \times 10^{-4}$	125,20	3.13 × 10 <sup>-4</sup>	
Ce-144	18,80	$9.40 \times 10^{-3}$	19.90	9.95 x 10 <sup>-3</sup>	
Cs-137	5.36	1.07 × 10 <sup>-3</sup>	2.70	5.40 × 10-4	
Nb-95	234.20	7.81 × 10-3	220.50	$7.35 \times 10^{-3}$	
Ru-106	2.65	$1.32 \times 10^{-3}$	8.20	4.10 × 10-3	

Table 8

Radioactivity in Air (10<sup>-12</sup> µci/ml)

		Number of Samples	Beta	% CG	Alpha	<u>% CG</u>
ALRR-	-Site IA Ave. High Low	237	0.11 0.40 0.01	0.11	0.0018 0.0181 0.0	9.00
	ing Hall e 2A Ave High Low	171	0.15 0.63 0.02	0.15	0.0038 0.0846 0.0	19.00

#### From Section C. Impact (p. 26 of Voss 1975):

From the effluent data it has been shown that the contribution to the environmental radioactivity in air from Ames Laboratory operations consisted principally of Ar-41 and tritium.

Applying principles of meteorological diffusion to the stack effluent . . . the [following] estimates have been made at the exclusion fence:

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Individual dose at Fence Line for 1974

	<u>Millirem (max)</u>	<u>Millirem (Avg)</u>
Ar-41	2.86	2.15
H-3 (tritium)	0.05	0.04
TOTAL	2.91	2.19

**Finding #6:** Statements in Section 4.3 of the Ames Site Profile are **factually incorrect**. For all years for which Voss reported environmental sampling data (1974, 1975, 1976), there were **two** sampling sites that provided data in behalf of air concentrations and corresponding dose estimates.

**Observation #1:** Doses derived by NIOSH are lower than those reported by Voss. Admittedly, both sets of doses are small, which makes these differences of limited significance.

### 4.1 ENVIRONMENTAL DOSES FROM RESEARCH AND DEVELOPMENT FACILITIES (1942–PRESENT)

NIOSH identified a total of seven facilities as R&D facilities that are located on the main campus of ISU. In behalf of these facilities, NIOSH stated the following:

#### U and Th Exposures

... there was no monitoring of radiation exposure or contamination of the personnel or environmental areas around and among these facilities. As research facilities, the quantities of radioactive materials involved in the R&D work were small compared to the production facilities.

... Principal sources would have been **uranium**, **thorium**, and **fission products** from the hot canyon/hot cell in the Research Building. Because of the smaller amounts of radioactive materials in the R&D facilities in comparison with those in the production facilities, the releases from the R&D facilities were assumed to be **one one-hundredth of the releases from the production facilities**. That assumption resulted in daily environmental intakes of  $5 \times 10^{-2}$  and  $7 \times 10^{-4}$  pCi/d for uranium and thorium, respectively. ... for workers exposed around the R&D facilities, a daily intake of  $5 \times 10^{-2}$  pCi of  $^{234}$ U and  $7 \times 10^{-4}$  pCi each of  $^{232}$ Th,  $^{228}$ Ra, and  $^{228}$ Th ... [Emphasis added.]

#### **Exposures to Fission Products**

A hot laboratory was operated in the Chemistry Building, but was replaced in 1951 by a "hot canyon/hot cell" in the Research Building. In the 1940s, the hot laboratory was used to study extraction of plutonium from irradiated uranium by means of ion exchange columns.

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... The uranium and plutonium were separated from one another and from the fission products using 1 kg samples of uranium, from the pile, which had an activity of 5 curies.

*Releases were estimated from this operation by making the following assumptions (see Bihl 2006 for details of the calculation):* 

- A total annual throughput of the laboratory of 50 Ci/yr
- An airborne fraction of 0.002 for boiling liquids
- Filtration efficiency for average particle sizes of **99.5%** [high-efficiency particulate air (HEPA) filters were just being developed during this period]

These assumptions produce a total release of fission products of  $5 \times 10^8$  pCi/yr.

Using the least dispersive approach recommended by the National Council on Radiation Protection and Measurements (NCRP) in Publication 123 (NCRP 1996), the ground-level annual average air concentration would have been 13.2 pCi/m<sup>3</sup>. Using the inhalation rate of 2,400 m<sup>3</sup>/yr and converting to a daily intake results in 87 pCi/d.

Using the recommended fission product ratios for 180-day cooled fuel from Fission and Activation Product Assignment for Internal Dose-Related Gross Beta and Gross Gamma Analyses ([ORAUT 2007b]), an 87-pCi/d intake of fission products is assigned to specific radionuclides as listed in Table 4-2.

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Radionuclide	<b>Relative fraction</b>	Intake (pCi/d)
Ce-141	0.0221	1.92
Ce-144	0.2191	19.1
Cs-134	0.0054	0.470
Cs-137	0.0208	1.81
Eu-155	0.0014	0.122
Fe-55	0.0172	1.50
Nb-95	0.2492	21.7
Pm-147	0.0546	4.75
Ru-103	0.0321	2.79
Ru-106	0.0844	7.34
Sr-89	0.0558	4.85
Sr-90	0.0157	1.37
Y-91	0.0911	7.93
Zr-95	0.1311	11.4

Table 4-2. Annual environmental fission product intakes from the hot laboratoryin the Chemistry Building (1943–1981)

Because maximizing assumptions were used for most of the parameters in this analysis, the distribution is an upper bound (constant). [Emphasis added.]

SC&A's Comments and Findings Pertaining to Environmental Exposures from R&D Activities

Given the paucity of empirical environmental monitoring data, NIOSH's decision to employ surrogate data and models that are based on assumptions/extrapolation is commendable, as long as the resultant estimates of external doses and intakes are fundamentally sound and favor the claimants.

SC&A's review of Section 4.4 of the Ames Site Profile identified findings pertaining to (1) intakes of U and Th, and (2) intakes of fission products, as explained below.

<u>Environmental Intakes of U and Th</u>. As quoted above, NIOSH twice states that "... as research facilities, the quantities of radioactive materials involved in the R&D work were small compared to the production facilities."

Among the seven facilities identified as R&D facilities is **Wilhelm Hall** (formerly the Metallurgy Building). As described in Section 2.2.2 of the Site Profile (and briefly discussed in Section 3.2 of this report), Wilhelm Hall was the principal facility for the **production of thorium** that also experienced numerous blowouts. Environmental releases during the production periods and **blowouts** undoubtedly resulted in a steady **buildup** of Th-232/-228 and Ra-228 until the end of 1953. Table 4-7 of the Site Profile identifies environmental intakes during this time (i.e., 1943–1953) of **0.07 pCi/d** for each of the three radionuclides.

The inhalation intake of 0.07 pCi/d would correspond to the combined air concentration representing the (1) continuous airborne releases from Wilhelm Hall as the operating research facility and (2) the resuspension of past ground contamination representing years of production. Thus, the transition from operational to R&D status in 1953 to 1954 and the corresponding 100-fold reduction of intake from 0.07 pCi/d to 0.0007 pCi/d for the three nuclides does **not** account for the long-term persistence of these nuclides as ground contaminants and their

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resuspension. [Note, for **indoor environments**, NIOSH defines the transition from facility operation to post-operational time periods by means of a depletion factor as given in ORAUT-OTIB-0070 (ORAUT 2012b).]

Inspection of Table 4-7 of the Ames Site Profile shows the identical "instantaneous" 100-fold reduction of U intakes from 5 pCi/d to 0.05 pCi/d during the transition from 1953 and 1954, respectively.

**Finding #7:** The nearly "instantaneous" 100-fold reduction of U and Th environmental intakes that represent the transition of the uranium and thorium metal production facilities at the end of 1953 to R&D facilities in 1954, as defined in Table 4.7, are improperly modeled. Also not included in the model are the contribution of blowouts to environmental contamination and the persistence of these radionuclides in the environment post-1953.

<u>Environmental Intakes of Fission Products</u>. Estimates of environmental intakes from the operation of the **Hot Laboratory** as defined in Table 4-2 of the Site Profile were principally based on five modeled parameters. The only parameter that was based on documented information involved the quoted statement taken from page 40 of Fulmer (1947):

A hot laboratory, capable of handling 5 curies through the **adsorption process**, was designed and built. The method proved successful in a number of runs. The uranium and plutonium were separated from one another and from the fission products using 1 kg samples of uranium from the pile, which had an activity of 5 curies. (Fulmer 1947) [Emphasis added.]

The remaining parameters used in the derivation of environmental intakes of fission products were based on assumptions stated in a memorandum authored by Bihl 2006 (Subject: "Estimated Releases from the Hot Laboratory in Ames Chemistry Building").

Enclosed herein as Appendix C is the Bihl 2006 memo, which provides the following **assumed** parameters:

- (1) An annual throughput of 50 pCi/y (or about 10 kg of uranium from the pile)
- (2) An airborne release fraction of 0.002
- (3) The hot lab air-exhaust was filtered with an efficiency of 99.5% for all particle sizes
- (4) An air concentration that is based on the NCRP Report 123 (NCRP 1996) model for the Atmospheric Screening Level I: Vent Air (which includes the default value of 0.3 m<sup>3</sup>/s exhaust ventilation rate)

Justification for the **assumed** annual throughput of 50 Ci fission products and fission-product release fraction of 0.002 was based on the following statements:

Since this was an **experimental lab**, **not** a production facility, it was **assumed** that the throughput was 10 times the maximum inventory, or 50 Ci/yr. [Emphasis added.]

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And:

It was assumed due to activities in the hot lab described as experiments on techniques for separating the plutonium out of the irradiated fuel, that .002 of the inventory went airborne (from DOE Handbook 3010 worst case for boiling liquid, page 3-1 [DOE 1994].

## Questions Pertaining to Annual Throughput, Release Fraction, and Filter Efficiency

SC&A's review of the 1947 report (Fulmer 1947) titled, *History of the Ames Project Under the Manhattan District to December 31, 1946*, suggests that (1) the throughput of 50 Ci/yr of fission products assumed by Bihl (2006) may have been considerably higher, and (2) the assumed airborne fraction of 0.002 for "worst case/boiling liquids" does not apply to the extraction method(s) used at Ames and may not represent the "worse case."

Enclosed herein as Appendix D are pages 36 through 42 of the Fulmer (1947) report, which provide a description of the **dry fluoride** process used at Ames for the separation of plutonium from highly **volatile** fission products and uranium (see pages 38–40).

Regarding annual throughput quantities, Fulmer (1947) states the following (Appendix D, bottom of page 40):

A hot laboratory, capable of handling 5 curies through the **adsorption** process, was designed and built . . .

After appreciable amounts of plutonium became available from the piles it appeared that it would have to be obtained in the metallic state and extremely free from light element impurities, if it was to be used for weapons. An extensive "purity" program was set up in cooperation with the various sites and the Ames **Project took part in this program**. [Emphasis added.]

Don Bihl's unsupported assumption that the airborne release fraction of fission products was subject to a filter with an efficiency of "**99.5% for all particle sizes**" (before release to the environment) must be questioned in context with statements made by Dr. During an interview with Dr. Makhijani (SC&A), which included questions of his involvement in the plutonium separation at the Ames Laboratory, Dr. During shared the following comments:

Arjun: Could you tell me about the plutonium separation research?

*Dr. We received uranium metal we made in our laboratory. We made tons of it.* 

Arjun: That was non-irradiated material.

Dr. Yes. It was irradiated in Chicago. We would take some of those irradiated metal slugs and dissolve them and extract out the fission products and separate them to prepare the technique for isolating the plutonium from it. There were others in the country doing separation research also: Berkeley, Oak Ridge

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and maybe some at Los Alamos. I went to the Bureau of Standards in DC and they had a small plutonium research program going. Then I went to Columbia where they had leftovers from the time Fermi was there. There was a small program at MIT. ...

Arjun: You did not have glove boxes when you worked with irradiated material?

*Dr. We had crude devices – a hood with a stream of air. We knew better than to breathe that stuff.* [Emphasis added.]

Lastly, model parameters assumed by Bihl (2006) for deriving **environmental** intakes of fission products (described in Section 4.4) should also be compared to the model used by NIOSH in Section 5.4.1 of the Ames Site Profile. Section 5.4.1, "Fission Product Intakes from Early Fuel Research," derives **worker intakes within the hot laboratory** by means of a model described in NUREG-1400, *Air Sampling in the Workplace* (Hickey et al. 1993). The equation from NUREG-1400 is based on the following:

$$I = Q \times 10^{-6} \times R \times C \times D$$

where

- I = intake
- Q =source term release for 1 year = assumed to be 50 Ci
- R = release fraction = 0.01
- C = confinement factor = 0.1 assuming material was handled in some containment
- D = dispersibility factor = 10 for heating or chemical reactions

A comparison of parameters used by Bihl (2006) for modeling **environmental intakes** versus those defined in NUREG-1400 (Hickey et al. 1993) and selected for **worker intakes** within the hot laboratory identify (1) a 5-fold difference for the release fraction (i.e., 0.01 versus 0.002) and (2) a 20-fold difference in the "confinement" factor (i.e., 0.1 versus the 0.005% filter failure for particulate removal).

**Finding #8:** Environmental intakes cited in Table 4-2 are based on unsupported assumptions and model parameters, which moreover are inconsistent with parameter values used to model **worker intakes inside the hot laboratory** described in Section 5.0 of the TBD.

# 4.2 SIGNIFICANT ENVIRONMENTAL EVENT

Regarding significant environmental releases/exposures, NIOSH limited its discussion to the following event:

The only significant environmental event in the history of the Ames Laboratory was the release to the environment from operations that occurred from July 1951 through August 1952. ...

... Most Ames Laboratory workers were not exposed to the radioactive materials released during this event and were not involved in responding to the event. ...

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*Thus, no additional dose is recommended for occupational environmental dose for any workers.* [Emphasis added.]

These statements **conflict** with earlier statements contained in Section 2.3 of the Ames TBD that include the following:

There were **frequent small explosions and fires** associated with the uranium and thorium production operations (Payne 1992) cited as many as six small fires in a single day; these fires contributed to work-area contamination and potential airborne radioactive material exposures. No records were found to indicate that air sampling or contamination control was associated with these fires. [Emphasis added.]

Additionally, NIOSH's limited reference to Payne (1992) understates the magnitude of these events and their contribution to environmental releases and worker exposures that involved fires and/or explosions during the reduction phase of UF<sub>4</sub> and ThF<sub>4</sub> to metal and subsequent grinding/machining of U and Th ingots.

SC&A's awareness of these discrete radiological events with a high potential for significant radiation exposures at the Ames Laboratory were prompted by a two-volume document that represents a doctoral dissertation authored by Carolyn Stilts Payne and submitted to the Graduate faculty of ISU in 1992. Part 1 of the dissertation is titled, *The Ames Project: Creation, Organization, and Purposes of the Ames Project;* and Part 2: *Issues of Administration*. It should be noted that Dr. Payne's thesis was accepted by the Graduate Committee and is archived in the U.S. Library of Congress.

At the time of the Ames Project, little was known about the behavior and properties of materials and the chemical processes employed in the reduction of  $UF_4$  and  $ThF_4$  to pure metal biscuits. The reduction reaction was highly exothermic, in which internal temperatures reached levels well above the 1,500°C melting point of the steel container. Thus, it was critical that the refractor liner, which separated the reactants from the steel container, was not breached. As noted below, the use of wet material and/or improper lining of the bomb retorts resulted in numerous **fires** and **explosions**.

In her dissertation, Dr. Payne provides several accounts of discrete radiological events, as summarized below:

Mr. Premo Chiotti was working with Dr. Wilhelm and me [i.e., Dr. F. Spedding] on the reduction of thorium fluoride to thorium metal. Mr. Chiotti was adding a booster to the reaction in a room a few doors down the hall from my office. Suddenly there was a terrific explosion which blew out several of the windows in the front of the chemistry building. When I came out of my office to see what had happened, the corridor was filled with dust about six feet above the floor to the ceiling. I was relieved to see that Mr. Chiotti had not been injured, but he looked very dazed and was pacing up and down the corridor. As I passed him, I heard him muttering, "I must have misplaced that decimal point, I must have misplaced that decimal point." (Ref. 324)

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... Improper handling or lack of attention to properly lining the bomb retorts could cause blowout problems when the uranium reaction came into contact with the steel or iron in the bomb containers. ... Some days that was quite a chore; there were at least six explosions in one day because some wet raw lime being mixed in the bomb retort containers adversely affected the reduction experiment. (Ref. 267)...

Once an explosion blew out the south wall of Little Ankeny in the early hours of the morning; by then explosions were so **commonplace** that the **workers** went outside and pushed the wall back in as far as they could . . .

Incidentally, that was the day that several secretaries threatened to resign and one Army officer received a rather suspicious wound. Secretaries, who were at an office attached to the production plant, had to pass through the firing pit area in order to get outside the building. After that series of explosions, they were wary of staying any longer in a potentially dangerous work environment. Spedding, however, convinced all but two of them to stay after he promised to strengthen the wall between the office and the operations area and to cut a door to the outside directly from their office. That same day Major H.A. Savigny, an Army officer who also happened to be the Area Engineer came to investigate the problem after the third explosion. While he was there, another explosion occurred, and, of course, he immediately ran for the door. As he was talking to someone a few moments later, he suddenly grabbed his leg, and a small piece of metal fell from a burned hole in the seat of his pants. Others, however, thought it might be somewhat hard to justify his "bravery" since it was apparent what he was doing when he was injured. (Ref. 268) [Emphasis added.]

... Fires were also a danger at several steps in the process. Magnesium could shoot a flame several feet in length sometimes setting anything in its path on fire. Until the proper insulation techniques were learned, **uranium cutting or machining caused fires** when the cutting blade struck such a hard metal. Controlling these special chemical fires with lime or graphite became **a common practice that every worker** had to learn. (Ref. 365). [Emphasis added.]

Occasionally, more than plant security was threatened by secrecy. Because the chemicals were volatile, **frequent fires erupted**. Since the Ames fire department could not come into the buildings that housed the production plant or the research activities because of secrecy requirements, the College allowed the firemen and equipment to come, but remain outside in the event a fire went out of control. Luckily, the workman [sic] were always able to use the lime and powdered graphite around the production building to squelch any flames (Ref. 267). [Emphasis added.]

The above-cited references in Dr. Payne's thesis correspond to the following sources:

Ref. 267 – Kooser, 8; Frank H. Spedding. "Explosions," Spedding Manuscript, 4-5; Daane, Spedding. Wilhelm Interview, 1967, 25.

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Ref. 324 – Frank Spedding, "Humorous Story Concerning Explosions and Education," Spedding Manuscript, 2.

Ref. 365 – Frank Spedding, interview with Hacker 1980, 18-19; Frank Spedding, "The Day the Wall Blew Out of Little Ankeny," Spedding Manuscript; Frank Spedding, "Explosions," Spedding Manuscript.

Ref. 268 – Frank Spedding. "The Green Hornet," Spedding Manuscript, 3-4. The story was also repeated in varying detail in the following sources: Adolf Voigt interview with the author 1990, 6; Spedding, Wilhelm, Daane interview 1967, 15–16.

The limited role of the Ames fire department and the response of secretaries were acknowledged and referenced to in cartoons that were illustrated in the local newspaper, as shown in Figures 4-1 and 4-2.



Figure 4-1. Cartoon about the Fires in the Reduction and Casting Processes and the Role of the Local Firemen

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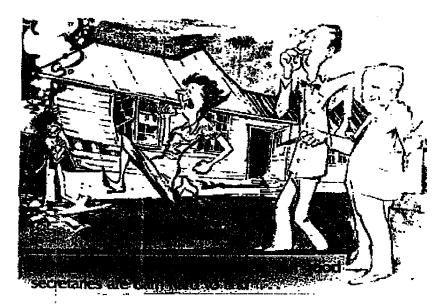


Figure 4-2. Cartoon about Frequent Explosions and Keeping Secretarial Staff on the Ames Project

**Finding #9:** U and Th blowouts represent significant environmental events that should be included in Section 4.5 of the Ames TBD for the assessment of environmental exposures.

Numerous interviews with and anecdotal accounts by Dr. Frank Spedding, Director of the Ames Project, as well as other coworkers, consistently describe bomb explosions and fires as commonplace, routine events during the years of the Ames Project (see Appendix E, "Telephone Interview with Mr.

Explosions and fires involved many kilograms of either uranium or thorium metal that released substantial quantities of radioactivity in the form of metal vapor and/or micro-particulates within the work area and environment.

In summary, the absence of documentation/records pertaining to blowouts should not be used to dismiss these events as insignificant; on the contrary, the absence of formal documentation of these events and assessment of their radiological impacts on workers and the environment should serve as evidence of radiological standards and practices that were lax and poorly enforced, especially during the earlier years of the Ames Project.

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# 5.0 ESTIMATES OF OCCUPATIONAL INTERNAL DOSE

Section 5.0 of the Ames Laboratory Site Profile states that radionuclides of interest for internal dose include **uranium**, **thorium**, **tritium**, and **fission products**. However, based on available information, NIOSH determined the following:

- (1) For the period August 13, 1942, through December 31, 1954, internal doses from the **production** and **casting** of **thorium metal** cannot be reconstructed
- (2) For the period January 1, 1955, through December 31, 1970, internal doses from maintenance and renovation activities of the thorium production areas of Wilhelm Hall (aka the Metallurgy Building) cannot be determined for all workers at Ames Laboratory
- (3) For the period January 1, 1955, through December 31, 1960, internal doses from radionuclides other than uranium (i.e., fission products) cannot be determined for any worker

# 5.1 ESTIMATES OF URANIUM EXPOSURE FROM INHALATION AND INGESTION

Estimates of uranium intakes derived by NIOSH are given in Tables 5-1, 5-2, and 5-3 of the Ames Laboratory Site Profile. Tables 5-1 and 5-2 identify intakes for personnel assigned to the **Chemistry Building** and **Annex 1**, respectively, and were based on data cited in **Christofano and Harris (1960)**. Table 5-3 provides data for uranium scrap recovery at the Annex 2, which is based on data from **Battelle (2011)**. For convenience to the reader, these tables have been reproduced below:

Table 5-1.	<b>Chemistry</b>	Building	Uranium	Intakes (pCi/d)
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Period	Inhalation	Ingestion	
Aug 1942–December 1953	8.5 <sup>a,b</sup>	0.09 <sup>b</sup>	

a. No data were available for determination of intakes in the Chemistry Building; therefore, it was assumed that research activities would have one-hundredth the intake of production activities, since uranium metal production was moved to the Physical Chemistry Annex 1.

b. Values are for workers **assumed** to work in research or production full time. For supervisors, **assume** one-quarter of the intake; for all other employees (clerical, janitorial, security, etc.), **assume** one-tenth of the supervisor's intake.

#### Table 5-2. Physical Chemistry Annex 1 Uranium Intakes (pCi/d)

Period	Inhalation	Ingestion
Aug 1942–December 1945	853 <sup>a</sup>	8.7ª

a. Values are for workers **assumed** to work in research or production full time. For supervisors, **assume** one-quarter of the intake; for all other employees (clerical, janitorial, security, etc.), **assume** one-tenth of the supervisor's intake.

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Period	Inhalation	Ingestion
January 1942–December 1950	6,061ª	124 ª
January 1951–December 1953	5,556ª	114ª

Table 5-3. Physical Chemistry Annex 2 Uranium Intakes (pCi/d)	Table 5-3.	<b>Physical Chemistr</b>	y Annex 2 Uranium	Intakes (pCi/d)
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a. Values are for workers **assumed** to work in research or production full time. For supervisors, **assume** one-quarter of the intake; for all other employees (clerical, janitorial, security, etc.), **assume** one-tenth of the supervisor's intake.

In support of these estimated uranium intakes, NIOSH stated the following:

**Because it is not clear** if there were clerical, janitorial, or nontechnical personnel and other types of researchers working in these buildings, **and it is not known** what precautions might have been taken for contamination control, it can be **assumed** that all individuals who worked in the buildings had some potential for exposure to uranium.

The data in Tables 5-1 and 5-2 were derived from data in Christofano and Harris (1960). In Christofano and Harris (1960), there is a description of the process for metal reduction that is similar to the process used at Ames Laboratory for production of uranium metal (Fulmer 1947). The primary difference appears to be that at Ames Laboratory the process used granulated calcium metal and at AWE sites the process used magnesium. Fulmer (1947) describes the process using magnesium, and it appears to be similar enough to be representative of the intakes at Ames Laboratory. [Emphasis added.]

... The intakes in Tables 5-1, 5-2, and 5-3 were compared to the **few actual bioassay** results found for workers in approximately the same period. Chapter 7 in Stone (1951), "Uranium Excretion Studies," provided data from a series of uranium bioassays obtained from Ames Laboratory workers in 1944 and 1945. Of special interest was a series of samples from the supposedly highest exposed worker at Ames Laboratory and samples from the most highly exposed group of workers at the Laboratory (21 samples from 11 workers)... The **average bioassay** result for the group of highest exposed workers was 75 µg/L. Assuming chronic intake for 1 year before the bioassay, the estimated intakes were:

- Absorption type F: 390 µg/d, 260 pCi/d
- *Absorption type M: 1,670 µg/d, 1,100 pCi/d*
- Absorption type S: 45,400 µg/d, 31,000 pCi/d. [Emphasis added.]

#### SC&A Comments

SC&A reviewed the modeled data representing Tables 5-1, 5-2, and 5-3 in context with the limited **bioassay** data referenced by NIOSH [i.e., Chapter 7 in Stone 1951; also referenced was Ferretti et al. 1951] as well as other data that include **toxicological data** and **spot air sampling data**.

<u>Assessment of Tables 5-1, 5-2, and 5-3 from the Ames TBD</u>. Estimates of uranium intakes at each of the three facilities are not only based on surrogate data, but their uncertainty is further

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enhanced by multiple layers of **assumptions** that are purely speculative, as illustrated in the following example:

- (1) Data in Table 5-2 are based on surrogate data defined in Christofano and Harris (1960) for **production** workers at Annex 1.
- (2) As defined in **footnote a** of Table 5-1, intakes at the **Chemistry Building** are defined for full-time researchers and are "assumed" at 1/100<sup>th</sup> the intake of full-time production workers at Annex 1 defined in Table 5-2 (or 853 pCi/d/100 = 8.5 pCi/d).
- (3) For **supervisors** at the **Chemistry Building**, daily intakes are **assumed** at 25% of the fulltime research or 8.5 pCi/d/4 = 2 pCi/d (see footnote "b" of Table 5-1); and for **all other employees**, daily intakes are **assumed** at one-tenth ( $1/10^{\text{th}}$ ) of the supervisor's intake (or 2 pCi/d/10 = 0.2 pCi/d.

Similar reduced, fractional intakes for **supervisors** and **all other employees** are defined for Annex 1 and Annex 2.

Assessment of Bioassay Data Reported by Ferretti et al. (1951). Between September 1943 and March 1945, a total of 48 workers at the Ames Laboratory were evaluated for urinary excretion of uranium that principally resulted from exposure to  $UF_4$  salt at the Annex 1 facility. To avoid contamination, urine sample collection involved the following protocol:

Upon leaving work **Saturday noon**, the **men** were given a clean bottle enclosed within two large envelopes. They were instructed to remove the outer envelope at home after carefully washing their hands. The second envelope was to be opened after bathing **Sunday evening** or **Monday morning**. The urine sample was to be passed into the bottle after again washing the hands ... [Emphasis added.]

The 48 subjects were grouped by their supervisors into four groups that reflected the potential for uranium exposure. Group 1 was expected to be highest and Group 4 the lowest. Grouping was based on the following criteria:

Those who were probably exposed to the greatest amount were classed as group 1; those exposed to the next highest amount made up group 2; those with very little but continuous exposure constituted group 3; and those with only occasional incidental exposure formed group 4. [Emphasis added.]

Data for the maximally exposed Group 1 are shown in Table 5-4 below. It should be noted that for the 11 individuals, there were 21 data points, which reflect some individuals who were assessed more than once at intervals of a few weeks or months. For example, case (worker) # was assessed for times and case # was assessed for times, with each having a maximum excretion value of  $\mu g/L$ .

Case (worker) # was subjected to additional evaluation, since this individual was believed to have had more exposure than any other individual on the Ames Project. His exposure began in and ended from an ended from  $\mu$  g to  $\mu$  g of uranium per liter. (Note: Case # of Group 1 most likely

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represents the individual as referenced by						
, as well as by						

A summary of bioassay results for all 48 Ames workers are shown in Table 5-5.

## Table 5-4. Uranium Excretion in Personnel, Group 1

Sample	Case	Amount, µg/liter
1	1	40
2	1	96
3	2	52
4	3	86
5	3	50
6	4	100
7	4	44
8	4	70
9	4	200
10	5	123
11	5	96
12	5	74
13	6	84
14	6	200
15	6	73
16	7	48
17	7	40
18	8	29
19	9	25
20	10	12
21	11	31
		75 (avg)

(Source: Ferretti et al. 1951)

 Table 5-5.
 Uranium Concentrations among 48 Ames Workers

(Source: Ferretti et al. 1951)

Group No.	No. of Subjects	No. of Samples	Avg. Urine Conc. (µg/L)	Max. Urine Conc. (µg/L)
1	11	21	75	200
2	20	26	46	130
3	11	14	16	33
4	6	6	<5	7

<u>Assessment of Toxicological Data</u>. During this period (1943–1944), select workers were also evaluated by medical personnel who were concerned about the toxicological effects of uranium, fluorine, and magnesium on function. Analyses focused on sulfur, sugar, albumin, F+, and other ionic species in blood/urine. In a report dated June 1, 1944, by Samuel Schwartz, MD, analyses in behalf of function individuals classified as being in the "heavy exposure"

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group suggested values indicative of abnormal functions (Schwartz 1944). In his **conclusions**, Dr. Schwartz stated that:

In general, fortunately, the tests indicate less abnormality than I would have expected from the amount of exposure these men are getting. The one exception to this statement is the almost consistent elevation of serum sulfur which is indicative of probably slight disfunction. If function tests almost uniformally normal. In only the heaviest exposure group is there significant change in the metabolism.

While these observations are highly suggestive of **radiological** impacts associated with uranium exposure, the **toxicological** effects of uranium, as well as several other agents (including F+), cannot be ruled out. For this reason, non-radiometric analyses are of limited value to dose reconstruction.

<u>Air Sampling during Uranium Metal Production</u>. Routine air sampling in the workplace can provide credible estimates of internal doses from the inhalation of airborne contaminants. This is especially true for **breathing-zone** air samples. SC&A's review of available documents showed data for only 12 air samples taken in May, June, and July of 1943. However, among the 12 area spot-samples taken, only 6 specifically identified uranium as the analyte. Table 5-6 below identifies the results of the 6 uranium area air samples by dates, location, and work activity at the Annex 1 facility.

Table 5-6. Uranium Air Concentrations associated with the Production of<br/>Uranium Metal at Annex 1

Dete	Work Location	Operation	Air Concentration		
Date	WORK Location	Operation	$(\mu g/m^3)$	(pCi/m <sup>3</sup> )	
6/16/1943	Near Jolter	Bomb loading	53.3	36	
6/16/1943	Near Micropulverizer	Grinding (lime, slo-set)	1,080	738	
6/16/1943	Near Riffle	Mg processing	85	58	
7/10/1943	Sample Room	Metal grinding	63.5	43	
7/12/1943	Cut-off Room	Metal cutting	420	328	
7/12/1943	Slag Room	Opening bomb	153	105	

The range for area air concentrations associated with the **routine** production of uranium metal at Annex 1 varied from 36.4 pCi/m<sup>3</sup> to 738 pCi/m<sup>3</sup> with an average of 218 pCi/m<sup>3</sup>. On the **unconservative** assumption that area air-sampling values approximate breathing zone air concentrations, the **average** daily intake is 2,100 pCi, with a **maximum** value of about 7,100 pCi for grinding operation. These two values should be compared to Table 5-2 of the Ames TBD, which shows a **maximum** value of 853 pCi/d.

## SC&A Findings

Uranium intakes were defined in Tables 5-1, 5-2, and 5-3 for three facilities (i.e., Chemistry, Building, Annex 1, and Annex 2, respectively) for workers who were assumed to be "maximally" exposed. Based on **job descriptions** (e.g., supervisors, clerical, janitorial, security, etc.), intakes are assumed to be 0.25 and 0.025, respectively of intake values cited in each of the

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three tables. NIOSH based these values on **surrogate** data derived from data in Christofano and Harris (1960), Battelle (2011), and multiple levels of unsupported assumptions.

NIOSH sought validation for these data by comparing intakes in Tables 5-1, 5-2, and 5-3 of the Ames TBD to the few actual bioassay results reported by Ferretti et al. in Chapter 7 in Stone (1951). For comparison, **bioassay** data selected by NIOSH involved the **average** urine excretion value of **75 \mug/L** (or **51.225 pCi/L**) for the maximally exposed workers designated as Group 1. IMBA runs yielded the following intakes:

- Absorption Type F: 260 pCi/d
- Absorption Type M: 1,100 pCi/d
- Absorption Type S: 31,000 pCi/d

Based on these results, NIOSH stated:

*If the geometric mean of the data is used, the estimated intakes are smaller* . . . *For type M, this range is still consistent with the intakes in Tables 5-1, 5-2, and 5-3.* [Emphasis added.]

Uranium intake values defined in Tables 5-1, 5-2, and 5-3 of the Ames TBD are re-introduced with other data in Table 5-8, "Default intakes for Ames Laboratory (if no bioassay results are available)," in the Ames Site Profile. Findings cited below reference data provided in Table 5-8 of the Ames Site Profile, which is reproduced herein as Table 5-7.

**Finding #10**. <u>Available empirical bioassay and air-sampling data</u> for Annex 1 workers are substantially higher than modeled/surrogate data assigned by NIOSH.

For absorption **Type M**, the **average** excretion concentration value of 75  $\mu$ g/L (or 51.225 pCi/L) yields an intake of 1,100 pCi/d. This value is 29% higher than NIOSH's assumed value of 853 pCi/d for Annex 1 production workers, as given in Table 5-2 of the Ames TBD.

Equally, empirical air sampling data cited in Table 5-6 above would yield an **average** intake of 2,100 pCi/d and a maximum intake of over 7,000 pCi/d for Annex 1 workers exposed to various chemical forms of uranium.

**Finding #11:** NIOSH further minimized the intake value of 853 pCi/d for Annex 1 production workers by assigning the "distribution" as a constant (see Table 5-7 below).

By assigning a "constant" distribution, NIOSH implied that the 75  $\mu$ g/L (or 51.225 pCi/L) value was a **maximal** excretion value, when in fact it was the **average** excretion value for Group 1 (see Tables 5-4 and 5-5 above).

**Finding #12:** Default intake rates defined in Column 6 of Tale 5-7 below are improper for absorption Types F or S.

It is technically incorrect for an intake value that represents (or is assumed to represent) absorption Type M to be used for absorption Types F or S. For example, NIOSH calculated an intake of **31,000 pCi/d** in behalf of the **average excretion** value of 75  $\mu$ g/L (or 51.225 pCi/L)

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excretion value reported by Ferretti et al. (1951). Based on guidance provided in Table 5-8 (Exhibit 5-1), the dose reconstructor would, nevertheless, be expected to select the 853 pCi/d intake value for absorption Type S in behalf of an Annex 1 production worker.

## Table 5-7. Default intakes for Ames Laboratory

(if no bioassay results are available)

Job Category		Dose cal	culation para	ameters		IREP input parameters	
or task/Building	Period	Material	Mode	Absorption Type of f1	pCi/d	Distribution Type	Parameter 1
Researcher/Chemistry Building <sup>a</sup>	Aug 1942– Dec 1953	Natural uranium	Chronic inhalation	F, M, or S	8.5	Constant	Dose
Researcher/Chemistry Building <sup>a</sup>	Aug 1942– Dec 1953	Natural uranium	Chronic ingestion	0.02 with F, M; 0.002 with S	0.09	Constant	Dose
All employees in Chemistry Building	Jan 1954– May 1976	Natural uranium	Chronic inhalation	F, M, or S	4.1	Constant	Dose
All employees in Chemistry Building	Jan 1954– May 1976	Natural uranium	Chronic ingestion	0.02 with F, M; 0.002 with S	0.68	Constant	Dose
Researcher in hot lab/Chemistry Building	1943–1951	Fission products	Chronic inhalation	Use most favorable to the claimant, for Sr-90 use F	See Table 5-7	Constant	Dose
Researcher, production technician, anyone involved daily with uranium in Annex 1 <sup>a</sup>	Aug 1942– Aug 1945	Natural uranium	Chronic inhalation	F, M, or S	853	Constant	Dose
Researcher, production technician, anyone involved daily with uranium in Annex 1 <sup>a</sup>	Aug 1942– Aug 1945	Natural uranium	Chronic ingestion	0.02 with F, M; 0.002 with S	8.7	Constant	Dose
All employees in Annex 1	Sep 1945– Dec 1953	Natural uranium	Chronic inhalation	F, M, or S	17.5	Constant	Dose
All employees in Annex 1	Sept 1945– Dec 1953	Natural uranium	Chronic ingestion	0.02 with F, M; 0.002 with S	1.6	Constant	Dose
Researcher, production technician, anyone involved daily with uranium in Annex 2 <sup>a</sup>	Jan1944– Dec 1953	Natural uranium	Chronic inhalation	F, M, or S	6,061 through 1950, 5,556 from 1951 to 1953	Constant	Dose
Researcher, production technician, anyone involved daily with uranium in Annex 2 <sup>a</sup>	Jan1944– Dec 1953	Natural uranium	Chronic ingestion	0.02 with F, M; 0.002 with S	124 through 1950; 114 from 1951 to 1953	Constant	Dose
All employees in Annex 2	Jan 1954– 1972	Natural uranium	Chronic inhalation	F, M, or S	124.7	Constant	Dose
All employees in Annex 2	Jan 1954– 1972	Natural uranium	Chronic ingestion	0.02 with F, M; 0.002 with S	11.2	Constant	Dose
Anyone routinely in Wilhelm Hall (Metallurgy Building)	1955–present	Th-232	Chronic inhalation	M or S	See Table 5-5	Constant	Dose
Anyone routinely in Wilhelm Hall	1955–present	Th-232	Chronic ingestion	5E-4 with M; 2E-4 with S	See Table 5-5	Constant	Dose
Anyone routinely in Wilhelm Hall	1955–present	Ra-228	Chronic inhalation	М	See Table 5-5	Constant	Dose
Anyone routinely in Wilhelm Hall	1955–present	Ra-228	Chronic ingestion	0.2	See Table 5-5	Constant	Dose

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Job Category	Dose calculation parameters					IREP input parameters	
or task/Building	Period	Material	Mode	Absorption Type of f1	pCi/d	Distribution Type	Parameter 1
Anyone routinely in Wilhelm Hall	1955-present	Th-228	Chronic inhalation	M or S	See Table 5-5	Constant	Dose
Anyone routinely in Wilhelm Hall	1955-present	Th-228	Chronic ingestion	5E-4 with M; 2E-4 with S	See Table 5-5	Constant	Dose
Reactor workers and D&D workers	1965–1981	Tritium	Total of all modes	N/A	N/A	Lognormal, see Table 5-6 for GSD	Dose; see Table 5-6
Workers involved with D&D of reactor, including former reactor workers	1978–1981	Co-60 or Zn-65	Chronic inhalation	Type S	3,300	Constant	Dose
Workers involved with D&D of reactor, including former reactor workers	1978–1981	Co-60 or Zn-65	Chronic ingestion	Type S	660	Constant	Dose
Anyone routinely in Applied Science Center	1982-present	Tritium	Total of all modes	N/A	N/A	Constant	8.6 mrem/yr

<sup>a</sup> Values are for workers assumed to work in research or production full time. For supervisors, assume 0.25 of the intake; for all other employees (clerical, janitorial, security, etc) assume one-tenth of the supervisor's intake (0.025 of the intake in the table).

**Finding #13:** The scaling of uranium intake values based on (1) facility and (2) job function is without technical support and conflicts with statements given in the Ames Site Profile.

For example, NIOSH states that "... for workers involved in research ... in the **Chemistry Building**, an exposure of **one-tenth** of the workers involved in the **production** operations is assumed ..." Values for supervisors were assumed at 0.25 (or <sup>1</sup>/<sub>4</sub>) of the intake of researchers/ production workers, and for all other employees (clerical, janitorial, security, etc.) assume 0.025 (or  $1/40^{\text{th}}$ ).

This rigid scaling of intakes based on location and job description imposes unreasonable burden on the dose reconstructors and conflicts with the following statements contained in Section 1.3 of the Site Profile:

NIOSH added classes to the SEC in 2006, 2007, and 2010 to cover three separate groups of employees based on work location and job description. While the classes added in 2006 and 2007 included specific workers performing specific tasks in designated buildings, the 2010 class determined that the information available about worker job description, work location, or movement about the site was insufficient to determine if an employee worked in the affected area(s). [Emphasis added.]

Should data pertaining to job function become available, scaling factors that more closely resemble the ratios of excretion values reported by Ferretti et al. (1951) for Groups 1, 2, and 3 should be considered (see Table 5-5 above). Based on group averages, intake ratios of 1.0, 0.6, and 0.2 are more likely to represent production workers, supervisors, and all others.

For ingestion intakes of uranium, NIOSH stated that ". . . The rationale used in **inhalation intakes** for **reduced fractions** for other workers is applied **to ingestion intakes** as well." Thus,

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SC&A findings associated with inhaled intakes of uranium apply equally to ingestion intakes as well.

**Finding #14:** Although NIOSH briefly acknowledged the occurrence of "frequent fires and explosions" associated with the production of **uranium** metal, no attempt was made to assess potential intakes of these episodic events. Potential doses from **uranium** blowouts are discussed in combination with **thorium** blowouts in Section 5.7 below.

# 5.2 RESUSPENSION DURING PERIOD WITH NO URANIUM OPERATIONS

To estimate exposure during **non-operations** for the Chemistry Building, NIOSH stated the following:

... The daily intake rates for the Chemistry Building can be estimated by **reducing** the generic exposure estimate from ORAUT ([2006]) by a factor of 10 to **account for** the **conclusion** of the work, the standard laboratory precautions that were in place, and the production time at about 50% of full time in the Chemistry Building. [Emphasis added.]

**Finding #15:** ORAUT (2006) referenced above for estimating non-operational intakes corresponds to ORAUT-OTIB-0004, Rev. 03, *Technical Basis for Estimating the Maximum Plausible Dose to Workers at Atomic Weapons Employer Facilities*. It should be noted that OTIB-0004 was canceled before Rev. 03 of the Ames Site Profile [ORAUT-TKBS-0055 (ORAUT 2012a)] was issued. Moreover, the much higher intake values for inhalation and ingestion during **non-operating years (i.e., 1954–1976)** make no sense when compared to intake values for **operating years (1942–1953)**, as given in Table 5-8 of the Ames TBD (see Table 5-7 above) and briefly summarized below:

For non-operating years (1954–1976):

All Workers in Chemistry Building

- Inhalation of 4.1 pCi/d
- Ingestion of 0.68 pCi/d

# For the **operating** years (1942–1953), intake for the **maximally** exposed **Researcher** identifies:

- Inhalation of 8.5 pCi/d
- Ingestion of 0.09 pCi/d

Note: During the same **operating years**, **Supervisors**, and **all others** would be assigned the following intakes:

Supervisor:

- Inhalation: 8.5/4 = 2.1 pCi/d
- Ingestion: 0.09/4 = 0.0225 pCi/d

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All Others:

- Inhalation: 8.5/40 = 0.2125 pCi/d
- Ingestion: 0.09/4 = 0.00225 pCi/d

# 5.3 THORIUM EXPOSURE FROM THORIUM CONTAMINATION AFTER 1954

The Ames Laboratory Site Profile restricted its assessment of thorium exposures to **residual contamination** following the production of 65 tons of thorium metal in 1954. This decision was based on the conclusion ". . . that it was not feasible to complete dose reconstruction for internal dose for thorium, plutonium, and thoron for 1942 through 1954 (NIOSH 2006) and, therefore, internal dose is **not** addressed here for those radionuclides during those years."

Inhalation intakes from residual contamination for years 1955 through 2010 are principally based on past survey data reported in a 1998 report (Hokel 1998 et al.) titled, *An Assessment of the Causes, Mitigation Efforts, and Current Status of Th-232, U-238, and Beryllium Contamination in Wilhelm Hall at Ames Laboratory.* 

NIOSH summarized survey data contained in Hokel et al. (1998) as follows:

# From Section 5.2 (pp. 39-41)

... Starting in 1984 and continuing through the early 1990s, surveys were conducted in Wilhelm Hall to determine locations of contamination left from the early production years. This information is in Hokel et al. (1998). The following discussion using data from Hokel et al. and Klevin provides an estimate for intakes by workers in Wilhelm Hall from 1955 to the present. A summary of the air concentrations and daily inhalation and ingestion intakes is provided in Table 5-5.

All the data in Hokel et al. (1998) were reviewed and considered. Much of the data was related to locations that were hard to access and considered not to be an inhalation issue. There was one set of data that had floor surveys made in 1988, but these locations all had fixed activity, indicating that this was from beta radiation (see Appendix 6 of Hokel et al.). For this reason, the recommendation for assessing dose from thorium from 1954 through the present was provided. The accessible areas of the building, including rooms, air ducts, hallways, stairwells, transformer rooms, etc., were surveyed starting in 1984. The data in Hokel et al. present an overview of the survey results. In 1996, some measurements were made using an Alpha Continuous Air Monitor in the subbasement pipe tunnels and in large vertical void spaces in the stairwells. All of these results were less than background for thorium, thoron, or radon (Hokel et al. 1998). Therefore, the results used for this estimation are from a **pipe tunnel** survey that showed removable contamination on smears ranging from background (3 dpm) to 1,224 dpm. These numbers are high for generally accessible areas and with the possibility of the contamination becoming resuspended. Although other survey numbers in the report with removable

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contamination are higher, they are in locations that are inaccessible (in a drain line), hard to reach (inside a drawer), or small (edge of a sink), or the contamination was fixed. [Emphasis added.]

SC&A reviewed the Hokel et al. (1998) survey report for its credibility and use in assessing inhalation intakes for the years 1955 through 2010. Exhibit 5-1 corresponds to page 13 of Hokel et al. (1998) and identifies the following salient information:

- The "Pipe Tunnel Special Surveys of East Tunnel" was conducted on November 15, 1995, or more than 40 years after Wilhelm Hall was used for the production of thorium metal.
- The purpose of the "Special surveys" of the **Pipe Tunnel** was to test the effectiveness of the "water wash down method."
- The location of this survey (i.e., the pipe tunnel) has little to no relevance to areas of Wilhelm Hall where workers were engaged in the **production** of thorium metal (see photos #10 and #11 of Exhibit 5-1).
- The statement that "initial analyses showed removable contamination levels on the smears ranging from background levels (approximately 3 disintegrations per minute (dpm)) to 1,224 dpm." (The obvious deficiency in these two contamination levels is the unit area that was smeared was not identified. It can only be assumed to be 100 cm<sup>2</sup>.)

For deriving a "removable contamination" value, NIOSH adopted the "1,224 dpm" value cited in Exhibit 5-1; and due to the uncertainty regarding the surface area sampled, increased the value to 2,000 dpm for an **assumed** sampling area of 100 cm<sup>2</sup>, as described in the <u>Attribution and</u> <u>Annotation</u> statement #19 of the Ames TBD.

For estimating thorium inhalation quantities at Wilhelm Hall on November 15, 1995, NIOSH employed the 2,000 dpm/100 cm<sup>2</sup> as follows (p. 41 of the Ames Site Profile):

A removable surface concentration of 2,000 dpm/100 cm<sup>2</sup> of thorium in equilibrium with its progeny was assumed. This means the  $^{232}$ Th activity was approximately 200 dpm/100 cm<sup>2</sup>. Applying a resuspension factor of  $10^{-4}$ .

Air concentration<sub>Th-232</sub> =  $(200 \text{ dpm}/100 \text{ cm}^2)(10^{-4}/\text{m})(100 \text{ x } 100^2) = 2 \text{ dpm}/\text{m}^3$ 

The value of **2** dpm/m<sup>3</sup> on November 15, 1995, when the survey of the pipe tunnel occurred, represents an upper bound. [Emphasis added.]

For estimating the starting air concentration at the end of the production period (i.e., 1955), NIOSH stated the following (p. 41 of TBD):

Because survey information in the building just after cessation of the thorium operations was **not** available, the **Klevin data** were used to represent air concentrations in **1955** with the **caveats** provided below: ...

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Reference to the "Klevin data" corresponds to a survey conducted by the AEC in 1952 and authored by Paul B. Klevin. This survey report titled, *Ames Research Laboratory Occupational Exposure to Thorium and Beryllium*, is enclosed herein as Appendix B. Data selected by NIOSH from Klevin (1952) involved **average general air samples** as given in Table III (page 11) with exclusion of Room 303 (Drying and Calcining Area) and the Seminar Room/(Lunch Room). Using the 95<sup>th</sup> percentile of the geometric distribution of remaining air concentration values cited in Table III of Klevin (1952), NIOSH calculated an air concentration of 479 dpm/m<sup>3</sup> for airborne concentration at Wilhelm Hall in 1955.

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#### Exhibit 5-1. Locations Selected by NIOSH for Determining Removable Contamination Levels

(Source: Hokel et al. 1998, page 13)

Pipe Tunnel Special Surveys of East Tunnel (Test Areas)

To test the effectiveness of the water wash down method we tested a small area of the East tunnel prior to washing down the entire tunnel. On November 15, 1995, (between the initial September survey and before washing the entire East tunnel) special smear surveys were taken in a section of the north side of the east pipe tunnel about 40 feet in total length. This section of the tunnel was divided into four locations (A-D), each approximately 10 feet in length (See Figure 1 and photo #10). Smear locations were numbered with grease pencil in each of the four 10-foot test areas, so that the same areas could be identified for subsequent smears. Subsequent smears were taken after each wash down in the immediate vicinity of the original smears in order to get a close approximation of the effectiveness of the wash downs. [NOTE: The subsequent 100 cm<sup>2</sup> smear samples were taken just to the right, left, above or

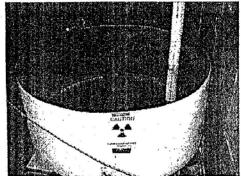


Photo #9 Holding tank for wash down residue and water.

below of the previous samples, since any removable contamination would have theoretically been removed by the previous smear in that location. So, the smears were not *exactly* in the same location, but very close].

The first wash down of these test areas was performed on November 27, 1995. Low-pressure water spray was used to wash down all pipes, walls and the' floors. The residue was washed into the large holding tank in the lower equipment room. Smear surveys of the washed areas were taken again on November 28 in approximately the same locations as the original smear surveys.

The second wash down was performed on December 4, 1995. Smear surveys were taken a third time on December 5, again near the same locations as the original smear surveys. After the second wash down, the asbestos covering on many of the large overhead pipes in the tunnels was starting to show signs of degradation from the water spray.

Analyses of the smears taken *before* the water wash downs (initial smears) in these areas indicated levels of contamination that were significantly above acceptable limits for removable radiation as defined in Appendix D. The initial analyses showed removable contamination levels on the smears ranging from background levels (approx. 3 disintegrations per minute (dpm)) to 1224 dpm. Analyses of the smears taken after the first wash down indicated a substantial reduction in the contamination levels, but still in excess of Appendix D limits. Analyses of the smears taken after the second

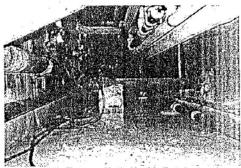


Photo # 10 - Test areas, North side of the East pipe tunnel.

wash down indicated some additional reduction in the contamination, but not as dramatic as the after the first wash down, and the levels of contamination still exceeded Appendix D limits. Overall, the data indicate

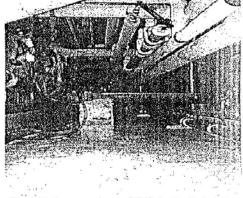


Photo # 11 -- Same general area as # 10, North side of the East pipe tunnel. Shows more detail of the pipes near ceiling.

that the second wash down did not add significantly to the effectiveness of the decontamination effort. Although the wash downs performed in the test areas appear to have reduced the overall amount of contamination, most of the values from smear locations

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From these two data points (representing air concentrations on January 1, 1955, and November 15, 1995), NIOSH derived a removal rate (i.e.,  $\lambda$ ) value of  $3.7 \times 10^{-4}$ /day or 0.134/yr that was used for estimating daily intakes (inhalation and ingestion) for years 1955 through 2010 as given in Table 5-5 of the TBD. For 1955, Table 5-5 recommends an air concentration of 448 dpm/m<sup>3</sup> for Th-232.

### SC&A Comments and Findings

NIOSH's approach for modeling intakes from residual contamination during the non-operational period of 1955 through 2010 was based on two data points that were separated in time by 14,929 days.

For November 15, 1995, the data point of **2 dpm/m<sup>3</sup>** of air represents a single adjusted smear sample taken in the **East Pipe Tunnel**, which is located in the **sub-basement** of Wilhelm Hall. Based on location, this smear sample has a **limited** relevance to those areas of Wilhelm Hall that were utilized for the production of thorium and resultant residual contamination levels during the non-operational period of 1955 through 2010.

The second data point assigned to January 1, 1955, was derived from general air samples taken in March of 1952 (see Table III of Appendix B). These data define thorium air concentrations based on alpha activity for **thorium** that assumes equilibrium of Th-232 with Th-228. Thus, the 95<sup>th</sup> percentile air concentration of 479 dpm/m<sup>3</sup> assigned to Th-232 at t=0 by NIOSH is a factor of 2 too high. This error applies to all values defined in Table 5-5.

Although the Klevin (1952) time-weighted air concentrations for workers were reviewed by NIOSH, these data were dismissed as **not** relevant (p. 41 of TBD):

Klevin (1952) did a **thorough** survey during operations, including breathing-zone task-specific air concentrations, time-weighted air concentrations for various workers, and general room air concentrations. ...

The Klevin air concentrations for specific tasks during production and the timeweighted averages are **not** relevant for Wilhelm Hall after 1954. [Emphasis added.]

Surprisingly, for the 22 workers assessed by Klevin (1952), their average time-weighted air concentration to thorium was 530 dpm/m<sup>3</sup>. For Th-232, this would correspond to only 275 dpm/m<sup>3</sup>, which is slightly more than one-half the value of 479 dpm/m<sup>3</sup> assumed by NIOSH. Additionally, a review of Appendix B identifies several other credible options by which the Klevin (1952) **operational survey data** in combination with ORAUT-OTIB-0070, Rev. 01, *Dose Reconstruction during Residual Radioactive Periods at Atomic Weapons Employer Facilities* (ORAUT 2012b), can be used to derive exposure estimates from residual contamination.

**Finding #16:** NIOSH's approach for deriving estimates of thorium intakes from residual contamination post-1954 does not make the best use of survey data reported by Klevin (1952) and use of ORAUT-OTIB-0070, Rev. 01 (ORAUT 2012b).

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## 5.4 THORIUM EXPOSURES DURING OPERATIONAL PERIODS THAT WERE <u>NOT</u> CONSIDERED BY NIOSH

Justification for the decision **not** to consider/assess thorium exposure during the operational period of 1942 through 1954, which produced 65 tons of thorium, was explained in Section 1.3.1 of the TBD, which states:

*NIOSH*... found that the monitoring records, process descriptions, and source term data available [for thorium/plutonium and thoron (see Table 1-1)] are not sufficient to perform complete dose reconstructions... [Emphasis added.]

The inverse of these statements and Table 1-1 of the TBD suggest that for **uranium**, a complete dose reconstruction can/will be performed using available **monitoring records, process descriptions,** and **source term data**.

However, NIOSH's method for deriving **uranium**, as evaluated by SC&A in Section 5.1 above, employed a combination of surrogate data [i.e., data derived from Christofano and Harris (1960); and Battelle (2007)] and layers of unsupported assumptions defined in footnotes assigned to Tables 5-1, 5-2, and 5-3. The relative merit by which NIOSH derived uranium exposures should, therefore, be compared to thorium data considered suitable for dose reconstruction, as discussed below.

# 5.4.1 Thorium Bioassay Data

Four datasets were found to contain bioassay data for thorium representing urine samples collected on the following dates: April 13, 1952; May 12, 1952; April 11, 1953; and May 11, 1953. As discussed below, the significance of dates for these four datasets is their **temporal** relationship to the AEC's radiation survey conducted on March 18–21, 1952 (Klevin 1952).

Enclosed herein as Appendix F are redacted copies of the four datasets, which identify sample dates, task(s) performed by each individual, and observed concentrations of thorium in urine samples. (Note: Datasets #1 and #2 report urine concentrations in the units of  $\gamma$ /Liter of urine, and datasets #3 and #4 in the units of  $\gamma$ /200 cc of urine). Table 5-8 provides summary data in behalf of the four bioassay datasets. Tasks which corresponded to the highest urine excretion values included calcining and hydrofluorination, metal reduction, final casting, and solution of Th nitrate (see Appendix F). At this time, the quantitative interpretation of thorium urine concentrations must await an understanding of the unit value of " $\gamma$ ," based on the fact that the colorimetric analysis assumes that " $\gamma$ " equates to microgram (µg).

(see Appendix F)					
Set #	Sampling Date	No. of Samples	Thorium Range (γ/L)	Urine Conc. (γ/L) Average	
1	4/13/1952	5	15–35	21	
2	5/12/1952	15	10-80	23.7	
3	4/13/1953	7	0–15	5.7	
4	5/11/1953	15	0–90	13.7	

# Table 5-8. Summary of Urine Bioassay Data for Thorium

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A relative comparison of thorium concentration values among the four sets identifies a marked reduction in exposure between sets #1 and #2 versus sets #3 and #4. Undoubtedly, this difference can be explained by the impacts of the radiological survey and resultant recommendations made by the AEC on March 18–21, 1952, but not issued as a report until July 14, 1952 (Klevin 1952).

As stated on page 3 of the 1952 Survey Report:

This survey was made with the following objectives in mind:

- 1. To gather data from which estimation of the **daily weighted average exposure** can be determined for the personnel working on the AEC project;
- 2. To suggest the **physical** and **procedural** changes which **should** be made in order to **correct excessive** exposure. [Emphasis added.]

<u>AEC Recommendations</u>. Pertaining to the **second objective**, the AEC audit team cited a total of 36 recommendations (see pp. 10 through 13 of Appendix B), which were facility engineering controls for specific processes involving (1) thorium production, (2) calcining and hydrofluorination, (3) handling of thorium crude (ThF<sub>4</sub>), and (4) metal casting. AEC recommendations pertaining to procedural changes included the **introduction of the most basic health physics practices and protocols for worker safety** (see recommendations #29–#36).

# 5.4.2 AEC Survey Data

Independently or in combination with bioassay data summarized above, quantitative survey data reported by the AEC in 1952 for the Ames Laboratory provide key information that satisfies regulatory requirements for dose reconstruction defined in §82.14 of 42 CFR 82.

Among applicable data provided in the AEC Survey Report are the following:

- <u>Breathing Zone Air Samples</u> that are facility-specific and **task-specific** for a total of **54 operations** that involved the production of thorium metal (see Table II and Figures #2 through #8 in Appendix B).
- <u>An Assessment of Daily Weighted Average Inhalation Exposures to Thorium/Thoron</u> among 22 Ames workers representing a total of 15 different assigned tasks in the production of thorium metal (see Table I in Appendix B).
- <u>Average Air Concentrations for Thorium and Thoron Based on General Air Sampling</u>. General air sampling was performed at discrete locations of Wilhelm Hall. Locations critical in the production of thorium included Rooms 203, 303, 307, and 33 (see Table III and Figures #2 through #8 in Appendix B).
- <u>Location- and Task-Specific Smear Contamination and Dose Rate Data</u> for Rooms 303, 204, and 33 (see Table IV in Appendix B).

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# 5.4.3 Thorium Processes and Source Term Data

In a 1947 report titled, *History of the Ames Project under the Manhattan District to December* 31, 1946, E.I. Fulmer, Assistant Director of the Ames Project, provided a comprehensive summary of processes and source terms for thorium (Fulmer 1947). Relevant sections of the report include II, Section 1.4 – Production and Casting of Thorium Metal; II, Section 2 – Fabrication of Uranium and Thorium; II, Section 4.2 – Thorium Alloy Studies; and III, Section 4 – Thorium Chemistry including purification of thorium compounds by liquid-liquid extraction.

In Section 1.4 of the report, Fulmer provided the following information regarding the production of thorium metal.

Soon after the large scale bomb reduction of uranium was in successful operation, similar approaches were made on the reduction of thorium. In August, 1943, attempts were made to reduce a number of thorium compounds in a bomb. The first attempts were unsuccessful due to the high melting point of thorium and the great stability of its compounds. Later, small amounts of thorium were produced by reducing  $ThF_4$  with metallic calcium using iodine as a "booster". The yields were low and the metal was obtained in small pellets which were very difficult to recast into solid metal. In August, 1944,  $ZnCl_2$  was tried as a "booster" and solid biscuits of thorium-zinc alloy were obtained in good yields. Within three months the conditions necessary for good yield had been well enough established to allow expansion of the process to the use of a reduction bomb 6 inches in diameter. ... By June 1946, most of the details had been worked out successfully and the bomb reduction of thorium fluoride was ready for expansion to large scale production.

The process, as [sic] use in December, 1946, was a metallothermic reduction of thorium fluoride by metallic calcium. Zinc chloride was used to provide additional heat, to give a more fusible slag, and to form a low-melting allow of thorium which would collect in the form of a solid biscuit. The reduction was carried out in an iron bomb 7 inches in diameter and 45 inches long. This bomb was lined with a layer of dolomitic oxide compacted into place around a steel mandrel with a pneumatic jolter. The charge was placed in the bomb which was then closed and the reduction started by preheating in a gas-fired furnace. The bomb was allowed to cool after the reaction and the biscuit of thorium-zinc alloy removed and cleaned. This method produced a biscuit of about 39 pounds of thorium-zinc alloy with better than 96 per cent [sic] yield of thorium. ... By December 31, 1946 over **4500 pounds of thorium** had been cast for shipment to other sites. [Emphasis added.]

Production of thorium metal continued at Annex I (i.e., Little Ankeny) until 1949. Starting in 1947 and continuing through the end of 1953, production and casting of thorium metal was also conducted in the Metallurgy Building. Total production of thorium at both facilities amounted to 65 tons (or 130,000 pounds).

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In summary, based on the availability of urine bioassay data, the comprehensive AEC survey of 1952, and detailed process and source-term data, SC&A concludes the following:

In the regulatory hierarchy of data considered suitable for dose reconstruction, available data for the reconstruction of internal doses associated with the production of **thorium** metal are considerably higher than data used by NIOSH in the dose reconstruction for uranium cited in Tables 5-1, 5-2, and 5-3 of the Ames TBD.

**Finding #17:** Given the availability of credible data, NIOSH's exclusion of thorium exposure in dose reconstruction is not justified.

# 5.5 TRITIUM EXPOSURES

Section 5.3 of the Ames TBD evaluated tritium exposures associated with the ALRR, which was a 5 MW heavy-water-moderated reactor that operated from February 1965 through December 1977. Decontamination and decommissioning of the ALRR was completed in 1981.

Personnel at the ALRR were monitored for tritium and should have bioassay and dose records. In the event that records are either incomplete or unavailable, NIOSH constructed a coworker tritium dose model for years 1965–1981, as given in Table 5-6 of the Ames TBD.

## SC&A Comments

SC&A has not been given access to the raw data that defines estimates of tritium exposures for unmonitored coworkers. On the assumption that available tritium monitoring records were objectively evaluated, SC&A has no comments/findings.

# 5.6 FISSION PRODUCT INTAKES

Worker exposure to fission products at the Ames Laboratory may have come from two sources: (1) a Hot Lab that was operated in the Chemistry Building and (2) the decontamination and decommissioning of the ALRR.

# 5.6.1 Use of NUREG-1400 for Modeling Intakes at the Hot Lab

Between 1943 and 1951, workers at the **Ames hot laboratory** separated plutonium from uranium and their associated fission products. In the absence of empirical survey/bioassay data, Table 5-7 of the Ames TBD provides modeled estimates based on a generic formula defined in NUREG-1400: *Air Sampling in the Workplace* (Hickey et al. 1993):

$$I_p = Q \times 10^{-6} \times R \times C \times D \qquad \qquad Eq. \ 5.6.1$$

where,

Q = total quantity of unencapsulated material R = release fraction C = confinement factor D = dispersibility

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### SC&A Comments and Findings

The intent of NUREG-1400 is to assist licensees in establishing **air sampling programs** that conform with recommendations in the 1992 Regulatory Guide 8.25, Revision 1, *Air Sampling in the Workplace* (NRC 1992), and the regulatory requirements stated in 10 CFR Part 20. Thus, modifying factors for the intake  $I_p$  are based on **licensed facilities** that reflect current-day timeframes of operation. For example, NUREG-1400 states that the **10**<sup>-6</sup> factor cited in Equation 5.6.1 represents a rule of thumb that applies ". . . when **normal** precautions are taken [and] a worker is not likely to have an intake  $I_p$  exceeding 10<sup>-6</sup> of the material being handled . . ." [Emphasis added].

As previously referenced in Section 4.4 of this report, a mere glimpse of the design and operational conditions that describe the Hot Lab in the 1940s was offered by Dr. **Section 2010** in response to questions during an interview with SC&A (see Appendix A):

Arjun: You did not have glove boxes when you worked with irradiated material?

*Dr. We had crude devices* – *a hood with a stream of air. We knew better than to breathe that stuff.* [Emphasis added.]

**Finding #18:** Due to the fact that very little is known about the design features and technical specifications of the Hot Lab and the absence of worker monitoring/facility survey data, the applicability of NUREG-1400, Section 1.2, for use in dose reconstruction lacks technical merit and credibility for a facility that operated between 1943 and 1951.

# 5.6.2 Intakes of Fission Products from Decontamination and Decommissioning of the Ames Laboratory Research Reactor

Section 5.4.2 of the Ames TBD states:

During D&D operations, radiation protection **appeared to be acceptable for the time**... There is no evidence that bioassay for other radionuclides [i.e., fission projects] was performed. However, during D&D of the reactor, which included dismantlement, cutting, grinding, etc., it can be expected that some intakes from **activation** products occurred. [Emphasis added.]

This research reactor operated with 93% enriched U-238 fuel for a period of 12 years (1965–1977). Thus, in addition to activation products, one must also expect **substantial** contamination from fission products. For estimates of intakes, NIOSH stated:

A reasonable estimate of intakes from D&D of the reactor can be made by using the gross beta air concentration limit from 1977 ( $1 \times 10^{-9} \,\mu Ci/mL$ ) (ERDA 1977) based on the most conservative beta emitter ( $^{90}Sr$ ). [Emphasis added.]

Intakes for D&D workers were further adjusted by means of an assumed occupancy factor of 0.5 (or 1,000 hours/year) with supervisory and other personnel receiving **one-fourth** of the intake of D&D workers.

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Inspection of the Ames TBD Table 5-8 ("Default intakes for Ames Laboratory (if no bioassay results are available") (see Exhibit #5-1 above) identifies that intakes from D&D activities of the ALRR are restricted to **two activation products**: **Co-60 or Zn-65 as Types S**.

**Finding #19:** Although Section 5.4.2 of the Ames TBD is titled, "**Fission Product** Intakes from Research Reactor Operations and Decontamination and Decommissioning," NIOSH restricted intakes to two **activation products** that are arbitrarily based on a 1977 ERDA gross beta air concentration limit. SC&A regards the default intake (that is limited to a choice of one of two **activation products** as surrogate for fission products) as qualitatively incomplete and quantitatively without technical basis/support.

# 5.7 FAILURE TO ADDRESS URANIUM AND THORIUM BLOWOUTS FOR THE ASSESSMENT OF EPISODIC INTAKES

In two previous draft reports, *Review of the Ames Laboratory Special Exposure Cohort (SEC) Petition SEC-0038* (SC&A 2006) and *An Assessment of Worker Eligibility Criteria For Less Than 250-Days Employment: Ames Case Study* (SC&A 2007a), SC&A identified to the Advisory Board the relatively common radiological incidents of chemical explosions or "blowouts" at the Ames facility in context with the 250-workday requirement. In response to SC&A's concern, the Board appointed an ad hoc work group chaired by Dr. James Melius to further evaluate this issue. The work group requested SC&A to (1) review all available records/sources that would establish the frequency of such events, and (2) provide scoping calculations that would assess reasonable estimates of potential internal exposures associated with a single event. A third SC&A draft report was issued in June 2007 in order to satisfy this request. A full text of the third SC&A draft report titled, *The Relevance of the 250-Workday Requirement to Potential Exposures Associated with a Single Blowout* (SC&A 2007b), is enclosed herein as Appendix G. As a convenience to the reader, salient elements of this report (Appendix G) are summarized below.

# 5.7.1 Quantities of Uranium and Thorium Metal Produced

Data regarding the potential quantities of uranium and thorium that might have become volatilized and airborne as a result of blowouts during the reduction of UF<sub>4</sub>/ThF<sub>4</sub> to metal at Ames Laboratory can be found in a 1947 document titled, *History of the Ames Project Under the Manhattan District to December 31, 1946* (Fulmer 1947). This document was compiled by E.I. Fulmer, who served as Assistant Director to Dr. F.H. Spedding, Director of the Ames Project. Sections 1.1 and 1.4 of the report provide the following information pertaining to the reduction of UF<sub>4</sub> and ThF<sub>4</sub> to pure metal.

<u>Uranium</u>. Section 1.1 of the report emphasizes that the principal objective of the Ames Project was the production of uranium metal, which originally was based on the chemical reduction of  $UF_4$  by calcium metal in a refractory-lined steel bomb. In the first quarter of 1943, however, calcium was replaced by magnesium for the reduction, and by July 1943, uranium metal production reached **130,000 pounds per month**. Production of uranium metal was performed exclusively in the remodeled one-story wooden building identified as the Physical Chemistry **Annex I**.

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Uranium production ceased by January 1, 1945, after producing a total of about **2,000,000 pounds**. Over the 2-year production period, reduction of UF<sub>4</sub> commonly involved steel pipes measuring 6 inches in diameter by 36 inches in length (however, larger pipes of up to 10 inches in diameter by 36 inches in length were also used). A successful reduction yielded a biscuit that was typically **42 pounds of uranium metal**.

In Section 1.4 of the report, Fulmer (1947) provided the following information regarding the production of thorium metal.

Soon after the large scale bomb reduction of uranium was in successful operation, similar approaches were made on the reduction of thorium. ... By June 1946, most of the details had been worked out successfully and the bomb reduction of thorium fluoride was ready for expansion to large scale production.

The process, as [sic] use in December, 1946, was a metallothermic reduction of thorium fluoride by metallic calcium. ... This method produced a biscuit of about 39 pounds of thorium-zinc alloy with better than 96 per cent [sic] yield of thorium. ... By December 31, 1946 over 4500 pounds of thorium had been cast for shipment to other sites. [Emphasis added.]

On the **un**conservative assumption that the total of 4,500 pounds of pure thorium had been produced between June 1946 and December 1946 by means of 39-pound thorium-zinc biscuits with ~96% yield of thorium, a **minimum** of **120 reduct**ions may be assumed for the 7-month period (or about 4 reductions per week).

Production of thorium metal continued at Annex I (i.e., Little Ankeny) until 1949. Starting in 1947 and continuing through the end of 1953, production and casting of thorium metal was also conducted in the Metallurgy Building. Total production of thorium at both facilities amounted to 65 tons (or 130,000 pounds).

Furthermore, assuming that the size of the bomb retorts remained constant after December 1946, a total of around 3,500 ThF<sub>4</sub> bombs were reduced between June 1944 and December 1953, yielding an average of seven reductions per week.

# 5.7.2 Estimated Frequency of Blowouts

As noted in Appendix G and Section 5.7 above, official statements made by Dr. Frank Spedding attest to the fact that uranium and thorium blowouts were episodic events that were not uncommon during the entire production period.

From Spedding's statement that "...I remember one night we had an explosion that blew the whole south end of the building out and being an old wooden building, when things quieted down we all went outside and shoved the wall back in again and went to work" [Emphasis added], it is reasonable to conclude that this and other blowouts were not perceived as potential radiological threats, since no attempt was made to limit the time of exposure or to mitigate subsequent exposure by decontamination efforts, engineering controls/building modification, or the use of respiratory protection.

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Dr. Spedding's assessment of these episodic events and their casual acceptance was confirmed in a separate interview conducted by SC&A with a former Ames Laboratory employee,

, on January 3, 2007. Transcript of the interview is enclosed herein as Appendix E. In response to the question of the frequency of blowouts, stated "... Oh, I would say maybe once a week" (see pp. 2–3 of Appendix E).

## 5.7.3 Assessment of Airborne Contamination Levels Associated with Blowouts

Although no radiological incident/investigative reports were found in behalf of the **Ames Laboratory**, there was a blowout incident at **Fernald on April 5, 1954**, which provides data that are applicable to Ames.

Summarized below are descriptions and data contained in a report issued by J.H. Noyes et al. (1954) on April 5, 1954, titled *Committee Investigative Report of Thorium Blender Incident* – *March 15, 1954*.

## From Section 2.0:

**On March 15, 1954**, personnel were attempting to blend a batch of thorium fluoride, calcium metal and zinc chloride preparatory to the reduction of the charge. After some delay in starting the blending operation and after unsuccessful attempts to properly seat a Gemco valve on the blender, a puff of dust appeared at the mouth of the blender; then a short flame; followed at 3:13 p.m. by a sheet of flame that extended horizontally from the blender a distance of about 45 feet over an arc segment of 38 feet. The duration of the flame is estimated at less than 10 seconds during which time two persons... received serious burns which subsequently proved fatal, and two others received minor burns. Three additional persons received minor hand burns while assisting the injured. Physical damage to the equipment is estimated at about \$700. Approximately **50 lb. of thorium were unaccounted for following the incident.** The building was vacated and all activities stopped until an adequate investigation established such factors contributing to the accident as an examination of the building and equipment could offer. [Emphasis added.]

## From Section 7.1:

The blending of a charge of thorium fluoride using calcium metal and zinc chloride is **similar to the process utilized by Iowa State College, at Ames, Iowa**... [Emphasis added.]

*The charge that was being blended when the incident occurred consisted of the following:* 

100.0	lb. thorium fluoride
10.0	lb. zinc chloride
35.9	lb. calcium metal

... Examination of the blender after the accident showed it to be nearly empty...

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These data suggest that 50 lbs (or about 50%) of the 100 lbs thorium charge were likely to have been volatilized and dispersed by the high temperature that characterizes this exothermic chemical reduction of ThF<sub>4</sub>. Thus, data involving a thorium blowout at FMPC in 1954 were used to model internal exposures from inhaling airborne contaminants in the first few minutes following a blowout, and from resuspension of residual surface contamination for a period of 30 days (or 193 workhours). This time interval was selected on the **un**conservative assumption that the frequency of blowouts may have occurred on a monthly bases.

Applying FMPC data to Ames, a 50% volatilization of a typical UF<sub>4</sub> or ThF<sub>4</sub> charge would have released about 21 lbs of U and about 19.5 lbs of Th into the air/environs at the Ames Annex I/ Little Ankeny facility.

By means of these quantities, building dimensions for Annex 1, and assumptions stated in Section 6.0 of Appendix G, SC&A derived the internal dose estimates for a single thorium or uranium blowout as given in Tables 5-9 and 5-10, respectively.

Radior	Radionuclide/		unt Bone Surface				Lu	ng		
	bility	Inhaled (µCi)	1 yr	5 yr	10 yr	30 yr	1 yr	5 yr	10 yr	30 yr
Th-232	Type S	4.4E-02	0.1	1.3	3.6	13.7	2.6	5.1	7.3	11.4
Th-228	Type S	4.4E-02	0.3	1.4	2.0	2.0	29.3	34.2	34.2	34.2
Th-232	Type M	4.4E-02	4.6	26.0	53.7	153	1.8	2.0	2.0	2.3
Th-228	Type M	4.4E-02	12.2	39.0	43.9	45.8	19.5	21.2	21.2	21.2
Total			12.7	67.7	103	214.5	53.2	62.5	64.7	69.1

 Table 5-9. Internal Dose Estimates for a Thorium Blowout\* (rem)

\* Dose estimates include inhalation exposures resulting from the first 5 minutes of a blowout and from resuspension of contaminants for a 30-day work-period. Not included are exposures to kidneys, liver, and other organs.

**Bone Surface** Amount Lung Radionuclide/ Inhaled **Solubility** 10 yr 30 yr 1 yr 5 yr 1 yr 5 yr 10 yr 30 yr (µCi) U-238 Type S 0.186 0.012 0.043 0.076 0.14 12.4 17.9 19.9 22.7 U-235 0.009 0.002 0.94 Type S 0.001 0.004 0.01 0.67 1.06 1.17 U-234 Type S 0.186 0.014 0.050 0.080 0.15 15.8 21.9 24.7 27.5 U-238 0.760 1.44 8.94 8.94 8.94 Type M 0.186 0.360 1.030 8.26 U-235 Type M 0.009 0.020 0.040 0.07 0.050 0.47 0.47 0.47 0.47 U-234 Type M 0.186 0.400 0.820 1.100 1.65 11.0 11.0 11.0 11.0 1.71 3.43 50.94 Total 0.81 2.34 61.15 66.07 71.78

 Table 5-10. Internal Dose Estimates for a Uranium Blowout\* (rem)

\* Dose estimates include inhalation exposures resulting from the first five minutes of a blowout and from resuspension of contaminants for a 30-day work-period. Not included are exposures to kidneys, liver, and other organs.

Exposure to the lungs and bone surface from a single incident suggests substantial doses that increase with time, as shown in Table 5-3 and Table 5-4 above. The selection of 5-, 10-, and 30-year periods corresponds to critical time intervals between exposure and the induction period for cancer development.

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**Finding #20:** By means of documented anecdotes/testimonials regarding potential frequencies of blowouts, technical data for a specific blowout documented at FMPC, and reasonable assumptions, SC&A derived significant U and Th intakes and associated organ doses that are applicable to workers at the Ames Laboratory, but were not considered/included in ORAUT-TKBS-0055.

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# 6.0 OCCUPATIONAL EXTERNAL DOSE

In Section 6.0 of the Ames TBD, NIOSH stated the following:

Workers at Ames Laboratory received external radiation doses between **1942 and 1952** that were largely unmonitored. Pocket chambers were [sic] available that might have been used to monitor external doses, but very few records could be found . . .

An upper bound of the total external exposures cannot be made for SEC-00038 workers because external doses from beta and gamma radiation resulting from exposure to thorium and its daughters or plutonium cannot be reconstructed due to a lack of information on the percentage of thorium daughter in-growth (up to 1954, when thorium operations ended) (NIOSH 2006). However, external dose from potential exposure to uranium from 1942 to 1953 can be estimated. [Emphasis added.]

## 6.1 UNMONITORED EXTERNAL DOSE

In Section 6.3.1 of the Ames TBD, NIOSH derived external doses for **uranium** exposure using the same **surrogate data** and **assumptions** that had been used for deriving estimates of uranium intakes, as given by the following:

To estimate reasonable external doses from the uranium processes before 1953, the methods described in three documents (Battelle 2011, Christofano and Harris 1960, and [ORAUT 2010]) were used. These documents are representative of the potential external exposures encountered at Ames Laboratory because the processes developed there were similar to processes at AWE sites...

*Exposure levels in these documents were converted to annual doses. These doses are provided for the Chemistry Building, Physical Chemistry Annex 1, and Physical Chemistry Annex 2 in Tables 6-4, 6-5, and 6-6 below.* 

## SC&A's Comments and Findings

**Finding #21:** SC&A's concerns about the use of the same surrogate data sources and questionable assumptions for deriving **external dose** for Ames' workers exposed to uranium closely parallel those related to **uranium intakes**. These concerns must be addressed, but for the purpose of expediency will not be repeated here.

However, a concern that needs further discussion is NIOSH's failure to include external doses from exposure to **thorium** (1) for years 1943–1949 during early production of thorium metal in Annex 1 and (2) for years 1947–1953 that correspond to large-scale production and casting of thorium metal at the Metallurgy Building (aka Wilhelm Hall).

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## Justification for the Inclusion of Thorium External Exposure

SC&A questions both the need for and purpose of NIOSH's statement that "... An **upper bound** of the total external exposure **cannot be made**... due to the lack of information on the percentage of thorium daughter in-growth...." [Emphasis added.]

It is likely that the "age" of separated thorium that was processed at the Ames Laboratory was highly variable, as suggested by statements contained in a letter authored by Allen P. Skoog M.D., Head of the Ames Laboratory Safety Group, to Dr. Spedding dated September 12, 1952 [Dr. Skoog's letter (Skoog 1952) is enclosed herein as Appendix H].

In his five-page letter, Dr. Skoog responded to numerous issues and suggestions raised by the AEC in their survey of the Ames Laboratory in March of 1952, including problems associated with "aged" thorium:

## From pp. 3-4

In connection with thorium and uranium processing the health physicists work in close conjunction with the production groups in all health matters. It should be called to the attention of all concerned that the operations in the production of thorium metal at the Ames Laboratory are not stable industrial processes. The area actually is a research pilot plant in which new developments are constantly being made. At present this pilot plant, when in operation, is called upon to do the work of several times the originally-designed capacity. One of the major problems in the mesothorium disposal aspect of our general problem lies in the raw material T.N.T. Originally the T.N.T. was shipped directly from the Lindsay Light and Chemical Corporation and was newly manufactured. Since the first scale-up of thorium production, this basic material has been shipped by the New York Office from its Middlesex, N.J., storage area. Recently, after we noticed increased activity, we were informed that they were shipping material which might have been in storage up to 6 yrs. The radioactive elements, especially mesothorium, have built up by a factor of 5 to 10 in this older T.N.T. It is these problems that cause most of our headaches. [Emphasis added.]

In the absence of more definitive information regarding the "age" of the separated thorium and the degree of Ra-228 ingrowth, an **upper bound** estimate of external exposures would conservatively **assume full equilibrium**. However, the need for the use of an upper bound/full equilibrium assumption becomes irrelevant when empirical dose rate measurements are **available**.

Included in the March 18–21 radiological survey of the Metallurgical Building at the Ames Laboratory, the AEC assessed both beta and gamma dose rates at key locations of the thorium production/operational areas, as given in Table IV of Klevin (1952) (see Appendix B). For example, shown as Exhibit 6-1 are beta and gamma dose rates associated with thorium reduction, casting, machining, and storage. These and other dose rate measurements cited in Table IV represent empirical **site-specific**, **area-specific**, and **task-specific thorium dose rate** 

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**measurements** that are available for the reconstruction of external dose for years 1943 through 1952.

In the hierarchy of data that may be used in the reconstruction of external dose, SC&A concludes that the credibility of available data for deriving external thorium doses exceeds those used by NIOSH for deriving external doses from uranium exposures.

**Findings #22:** Given the availability of highly credible and site-specific data for deriving external dose from thorium exposure, NIOSH's decision to exclude said exposures/doses is not justified.

## 6.2 COWORKER DOSE MODEL – 1952 TO THE PRESENT

Table 6-7 of the Ames TBD identifies coworker doses for the years 1952 to 2005 at the 50<sup>th</sup> and 95<sup>th</sup> percentile for unmonitored workers with potential exposure to beta, gamma, and/or neutron sources. The derivation of annual coworker doses was based on the following statements contained in Section 6.3.1.2 of the Ames TBD:

Extensive dosimetry records have been found for Ames Laboratory workers; however, many of the records for 1965 to 1981 do not identify the person receiving the radiation dose. If a worker was monitored but cannot be identified in the dosimetry records, that individual must be considered unmonitored and assigned a dose in each year for which no clearly identified records exist.

The coworker data study for Ames Laboratory included all available dosimetry records from 1952 through 1981 (Martin 2006c). All dose results were analyzed, including zeros and blank values, to determine the 50<sup>th</sup>- and 95<sup>th</sup>-percentile doses for each year for beta, gamma, and neutron exposures (McCartney 2006). The results of the analysis are summarized in Table 6-7. The missed dose recommended for monitored workers in Table 6-8 was added to the 50<sup>th</sup>- and 95<sup>th</sup>-percentile values in Table 6-7. Specifically, half of the maximum annual missed doses were added to the reported annual doses, except the reported positive doses, in which case the maximum missed dose was reduced by the dose corresponding to one badge exchange (because it is not possible that all individual badge results were zero if a positive annual dose was reported). [Emphasis added.]

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## Exhibit 6-1. Table IV of Klevin 1952

## TABLE IV (Cont'd)

Ares	Todation 1	Radiation Alpha d/m/100cm <sup>2</sup>	Heamiremen Beta mreps/hr	Gamma
Thorium Ex- traction (Rm 203)	Corridor Outside Th. Extraction Room	5,000	0.02	0.11
Calcining & Hydrofluoring- tion	General Ekgd. Radiation Room	0	0.3	0.5
eron	Floor	30,000	0.05	1.8
	ThO <sub>2</sub> Tray (6 <sup>1</sup> / <sub>2</sub> x 11 x 23) Inside Outside	<del></del>	3.0 0	8.5 5.0
	ThFL Tray (Fresh from Furnace) (11 x 11 x 30) Top Bottom		10 0.3	4.0 4.0
	fhF), fray (cooled for 17 hrs) Top Bottom		1 0.75	3.0 2.0
	THOI Tray (62 x 11 x 23) Top Bottom		2.0 0.1	6.5 3.0
	General Room Bkgd.		0	0.2
(Room 33)	Floor around Mixer	<del></del>	0	1.0
	5-gal. can ThFL Cover off-Top Material Cover on-Top material	100,000 2,000	9.5 0	9.8 11.5
	ThFh Can Storage Area Side - 7 cans	2,000	0	22
	Crude Billet	100,000	4.5	5.5
	Crude Billet Storage Area (Flogr)	20,000	2.0	1.0
	for a second			
***	Juno Contaminated.			
		SECP		
				USA 012

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## SC&A Comments and Findings

Our review of raw dosimetry records and NIOSH's assembly of these records in a spreadsheet format (McCartney 2006) confirms NIOSH's claim of "extensive dosimetry records" and the fact that "... many of the records for 1965 to 1981 do **not** identify the person receiving the radiation dose" [Emphasis added]. For example, a review of dosimetry records for 1966 show that of the 241 badged personnel, only 42 (or 17.4%) individuals could be identified by name. Nevertheless, annual doses were derived in behalf of all badged personnel, which proved useful in the construction of the coworker model summarized in Table 6-7 of the Ames TBD.

In addition, SC&A selected a small number of individuals in order to verify the assignment of a total annual dose against raw recorded data and NIOSH's assigned dose from potential missed dose defined either by minimum recordable dose (MRD/2) or by minimum detection limit (MDL/2).

Our limited review verifies that potential missed doses assigned to monitored persons who represent the coworker dose model correctly used one-half of the "**minimum reportable dose**" (MRD/2) as well as the recommended **minimum detection level** (or MDL/2) cited in Table 6-1 for the various time periods.

Regarding Table 6-7 for assigning dose to unmonitored workers or unmonitored periods for monitored workers, SC&A has no findings.

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# APPENDIX A: INTERVIEW WITH DR. MARCH 12, 2006, BY DR. ARJUN MAKHIJANI, SC&A

Draft notes sent to Dr. for approval on March 13, 2006. Written corrections received on March 16, 2006, and incorporated.

This account of the interview is **not** verbatim. The conversational style has been retained to give a flavor for how the interview was conducted and of the nature of the conversation.

Dr. Arjun Makhijani explained that he was a part of a team that was providing the Advisory Board with technical support on the DOE worker compensation law, and that he was at the beginning stages of looking at the petition for a Special Exposure Cohort filed by

and others for the Ames Laboratory. They discussed a good time for the interview and Dr. decided to give an interview then and there.

Dr. Makhijani: I understand that you worked on plutonium separation research at Ames Laboratory. I have found information about the uranium and thorium processing, but not much about the plutonium separation work. Could you tell me about it?

Dr. **Constitution**: All our work was written up in a series of reports with numbers that had CC in them. Our studies on the fission products were described there.

Dr. Makhijani: Do you have these CC reports?

Dr. **1** No. I was not allowed to take them. Secrecy at the time. You should try to find them. They may be at the Iowa State University archives.

Dr. Makhijani: Could you tell me about the plutonium separation research?

Dr. : We received uranium metal we made in our laboratory. We made tons of it.

Dr. Makhijani: That was non-irradiated material.

Dr. Yes. It was irradiated in Chicago. We would take some of those irradiated metal slugs and dissolve them and extract out the fission products and separate them to prepare the technique for isolating the plutonium from it. There were others in the country doing separation research also; Berkeley, Oak Ridge and maybe some at Los Alamos. I went to the Bureau of Standards in DC and they had a small plutonium research program going. Then I went to Columbia, where they had leftovers from the time Fermi was there. There was a small program at MIT.

Dr. Makhijani: Did you have any air-monitoring data or urinalysis or other such data for the people in the lab doing plutonium research?

Dr. : We did have one		who took
care of testing all of us in the analytical c	chemistry laboratory.	tested for uranium and also
thorium, and would take blood samp	les.	. After the
project,	. You might try to loca	te about the

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monitoring. [In his review of the interview, Dr. informed Dr. Makhijani that had passed away.]

Dr. Makhijani: Was that monitoring for uranium and thorium in the production and testing areas for metal production?

Dr. : Yes.

Dr. Makhijani: Do you remember any sampling for bioassay or air monitoring in the chemistry labs where the plutonium separation research was done?

Dr. : Actually we did not pay too much attention to the toxicology of it [in the separations research areas]. We knew we should not be breathing it. We did not wear masks all the time but we wore them at the times when we were doing something like burning scrap uranium and converting it to black oxide. There was particulate matter that went up the hood. I am sure that carried fission products.

Dr. Makhijani: You did this burning with irradiated uranium?

Dr. : Yes, we practiced first on non-irradiated uranium. I was the who prepared and practiced uranium hydride. I had the idea that if I can find a way to make uranium hydride, then we could separate the uranium from the fission products. But that turned out to be wrong. The hydride particle size is so big that the amount of FP you could separate was too tiny. You had to actually dissolve the uranium chemicals to get a good separation. That was the reason for the loss of interest on uranium hydride.

Dr. Makhijani: How much irradiated material did you get per batch and how hot was it?

Dr. : Can't give quantitative figures, but we handled roughly 100 grams at a time. The shipments were half a kilogram or so.

Dr. Makhijani: What about the level of irradiation? Were the slugs in lead containers?

Dr. : Yes they were. We often drove to Chicago and brought them back in the trunk of our car. There were three of usbook, and myself. [See Dr. All Things Nuclear, p. 206.] I'll tell you a story that's been written. The three of us were driving back from Chicago one day in February 1943 or 1944. Around 3 a.m., we had a flat tire out there on the Iowa prairie. We got out in the snow – there was snow coming down – but the jack was out of order. It would only lift the car up three or four inches. Then the three of us had the same idea at the same time. We jacked up the car three or four inches; then we put the uranium cylinders under the axle and lowered the car down on them. Then we raised the car some more, put another stack of uranium rods, and then jacked up the car again. Fortunately the length of the cylinder was more than the jack would raise the car so we were able to do the job. Otherwise I might still be out there on the Iowa prairie. And I learned something very important about uranium. It is very good to hold up cars.

Dr. Makhijani: How long did the plutonium separation research work go on at Ames?

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Dr. **We**: Well, it started in about 1943 and continued as long as I was there, which was until 1946 when I had to decide; what am I going to do? Am I going to work the rest of my life with plutonium? I knew **Mathematical Start S** 

Dr. Makhijani: Do you still teach there occasionally?

Dr. : I did , but after they don't call on me.

Dr. Makhijani: Did you ever measure the level of radiation of the rods you were working on in your research? Was there ever a GM tube that was put on them to get an idea of the level?

Dr.  $\Box$  : Once I used a sample of uranium that was not clad in aluminum. It was bare uranium metal that had been exposed in a reactor. Under these conditions, you start out with bright shiny uranium that turns brown and gray and black as U<sub>3</sub>O<sub>8</sub> builds up. I took some of that [black oxide] and scraped it off and then I measured the total radioactivity per milligram and I compared that with the radioactivity of unoxidized metal. The oxide had a lower level. I made the correction for the oxide part. You know the eight atoms of oxygen and three of uranium in U<sub>3</sub>O<sub>8</sub> – and then they came out to be the same. That told me that no segregation of fission products took place during oxidation.

Dr. Makhijani: Do you remember the number?

Dr. : No.

Dr. Makhijani: You did not have gloveboxes when you worked with irradiated material?

Dr. **We** had crude devices – a hood with a stream of air. We knew better than to breathe that stuff.

Dr. Makhijani: Did you go to Little Ankeny [where uranium metal was manufactured in large amounts]?

Dr.  $\mathbf{T}$ : First I went to predecessor of little Ankeny. At this place they had a way of preparing a steel cylinder and putting a lime liner in it. Then they put in UF<sub>4</sub> mixed with magnesium and put some lime freshly baked so as to decompose the hydroxide. That was the way the metal was prepared. Then they put in UF<sub>4</sub>. After the heating, the reduction occurred and uranium metal separated out. Some of the guys who worked there refused to wear masks. They would wear them when the director was there and then take them off when he left. They were farm boys and did not worry about the green powder. There was this one fellow;

Dr. Makhijani: Does that say that Iowa farm boys are healthy and strong?

Dr. : Yes. Too bad is not available to interview. He died recently.

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Dr. Makhijani: So it seems like the conditions in the uranium metal production areas must have been very dusty?

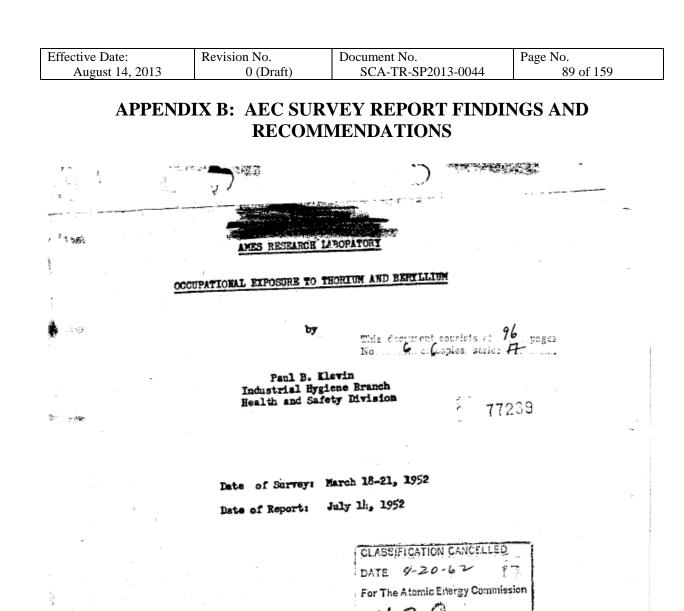
Dr. Yes. Yes,  $UF_4$  was of most concern. The air was also dusty from lime. That's good, because that gave a matrix to sweep up the smaller amounts of radioactive materials. There were hand brushes to clean the area. I don't remember vacuum cleaners being used though.

Dr. Makhijani: I think I have the preliminary information I need at this stage. I will send you the interview for checking to correct any errors I may have made.

Dr. : I hope you don't mind my stories and jokes.

Dr. Makhijani: Not at all. On the contrary, I will leave them in. They make the interview more interesting. Thanks so much for your time and for giving me an interview on the spur of the moment?

End of interview.



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TABLE I - Daily Weighted Average Exposure TABLE II - Tabulation of Avg. Breathing Zone Samples TABLE III- Tabulation of Avg. General Air Samples TABLE IV - Radiation Measurements

Fig. 1 - Chart of Avg. Daily Weighted Exposures Figs. 2 to 8 - Operating Area Layouts Showing Breathing Zone and General Air - Thorium & Thoron - Values

### APPENDIX A

### Job Analysis Sheets

#### APPENDIX B

Sample Record Sheets (copies #1 & #6 only)

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#### Scope

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This is a report of a preliminary survey performed during the period March 18-21, 1952 at the Ames Metallurgical Laboratories, Iowa State College. This survey, made in response to a request from the Chicago Operations Office, covered the health and safety problems existing during refining and thorium metal production. In addition, a brief study was made of several beryllium operations.

#### Purpose 1.1

- exposure can be determined for the personnel working on the AEC projects
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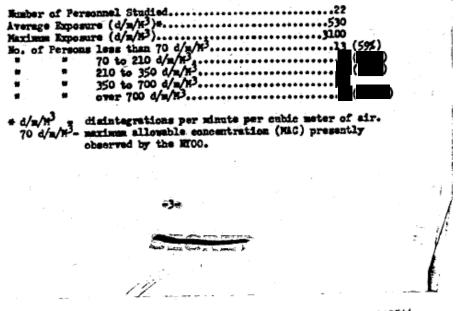
To suggest the physical and procedural changes which should be made in order to correct excessive exposures.

#### Regults of Study

#### 1. Thorium

Of the twenty-two employees studied in the thorium production areas, ( ) were exposed to thorium concentrations exceeding the maximum permissible level; employees were exposed to concentrations of 791 to 3100 alpha disintegrations per minute per cubic meter of air  $(d/m/M^3)$ . A complete breakdown of the daily weighted thorium exposure of the production personnel is as follows:

## Susmary of Daily Weighted Thorium Exposure



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This survey was made with the following objectives in minds 1. To gather data from which an estimation of the daily weighted average

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The following tables summarize the thoron exposure of the Ames production personnels

### b. Summary of Daily Weighted Thoron Exposure

Mamber of Personnel Studied	
Average Exposure (d/m/H3)	10 <sup>4</sup>
Maximum Exposure (d/m/M2) 3 x 10	4
No. of Persons less than 2.2 x 104 d/m/N3#	
Humber of Personnel Studied	

# 2.2 x 10<sup>h</sup> d/m/M<sup>3</sup> MAC for thoron.

### 3. Beryllium

of the personnel manufacturing crucibles were studied and were found to be exposed to acceptable daily weighted beryllium concentration of 0.95 V/M<sup>3</sup>. However, several of the operations studied exposed these technicians to concentrations exceeding the AEC maximum concentration for a single exposure by 6 to 8 times. additional beryllium sample was found to be 16 times the allowed single sample concentration.

No outdoor neighborhood survey was made since the nature of the beryllium operations and the small amounts of material handled did not constitute a problem.

#### L. Rediction Measurements

External radiation measurements made in each of the thorium operating areas showed excessive amounts of radiation present in both the storage and loading areas. The highest radioactivity found was 22 mr/hr gamma, greater than 20 mreps/hr bets (2610% B- 3 Survey Neter) and 100,000 d/m/100cm<sup>2</sup> alphs.

Included in the tabulation of the radioactivity measurements are values showing both growth and decay of thorium daughters.

Recommendations are included in this report which should reduce existing airborne contamination and external radiation at the various operations and in the general laboratory area,

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### Method of Study

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1. Sampling Procedures

a. Thorium

Dust samples were collected on 1-1/8" Whatman ful filter paper, using a 0.5 c.f.m. Universal air sampler. The sample collection period varied from thirty seconds to two hours, depending on conditions of operation and dust loading.

The dust samples collected divided into the following categories:

- (1) General air samples a sample obtained of a general area or room atmosphere,
- (2) Breathing some samples a sample obtained in the actual breathing zone of an operator during the performance of a particular task.
- b. Thoron

It is not possible with portable equipment to measure thoron concentrations directly; therefore, the concentration of the thoron daughter products, as collected with airborne dust, is used as an indication of thoron activity. The dust collection media and apparetus is the same as described above.

c. Beryllium

Beryllium dust samples were collected on 10 cm. Bureau of Mines All Dast Filter, BM #2133, using a 35-45 c.f.m. high volume air sampler. Periods of sampling varied from one minute to 30 minutes, depending on same conditions as mentioned above.

## 2. Job Analysis Sheets

The job analysis sheets give a detailed analysis of the operational time relationship of each employee on the project. This consisted of a statement of the total time spent on a particular job with an additional breakdown as to the number of minutes each task is performed per day. In addition, the average thorium alpha concentration and the thoron daughters' alphas, as obtained from the sample record sheets, are recorded. The average alpha concentration per 9 hour day is determined by dividing the alpha concentration times total time by the total number of minutes per day.

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### 3. Analytical Procedure

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Because of the unusual decay characteristics of the thorium sories, it would be well at this point to indicate briefly the analytical methods which we used to derive the results expressed in the report:

 Samples were counted on an alpha scintillation counter assuming a 30% self-absorption.

- b. Samples were counted as soon as received in the laboratory. The time of counting of samples varied from 1 to 3 days after sampling. The first count was then extrapolated back to time of sampling.
- c. All samples were re-counted after one week to determine the presence of long-lived material. Counting procedure was identical to the first count.
- d. The second count was subtracted from the extrapolated first count to yield the thoran daughter concentration; the second count being the thorium concentration. No evaluation of airborne bata or gamma emitters was made.
- e. Beryllium samples, which were taken at a rate of about 0.5 cubic meters per minute, were analyzed on a solution photo-fluorimeter measuring the fluorescence of the beryllium morin complex.

It is not possible to measure the activity of therea gas directly with field equipment. Therefore, the measurement of therea concentration must be carried out indirectly by determining the alpha activity of the therea daughter products collected with airborne dust. Since the half-life of  $Po^{210}$  (Th A) is only 0.16 sec., the alpha activity that is used is that of the daughter products following Po<sup>212</sup> (Th B) which has a half-life of 10.6 hours.

If the system is in radioactive equilibrium, the equivalence abown will be one daughter alpha disintegration per thoron disintegration. This allows counting at any reasonable time after sampling and calculation of the activity at the time of sampling by simple extrapolation. The long-lived alpha activity due to thorium may be obtained by counting after decay of the daughter products, and this activity may be subtracted from the total to obtain the daughter product activity.

The extrapolation of the daughter product activity at the time of counting to the activity at the time of sampling is based on the decay of Th B, which is the slowest rate in the series following theren. The activity due to the long-lived thorium is determined by recounting the sample after allowing the daughter products to decay.

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		Ap a Daughter product activity at sampling time
		A1 = Total activity at first count
		A2 z Thorium activity at second count
		A = A1 - A2 = Daughter product activity at first count
		λ = Decay constant for ThB
<b>2</b> 90		t z Time after sampling
	Then,	$A = A_{o}e^{-\lambda t}$
	-	And A, may be calculated.
	The rat:	s of A to A for several times after sampling are given below:

2	days	•	•	•	••	•	•	•	•	•	•	•	•	•	٠	.0450
3					••											.0051
Ŀ		•	•	•	••	•	•	•	•	•	٠	•	•	٠	•	.0018

The shortcomings of the method are the assumption that radioactive equilibrium exists and the assumption of complete collection of the daughter products on airborne dust. However, it is the best method available for field use at the present time.

### Process Description

- 1. Thorium
  - (a) Thorium Refining & Metal Production
    - (1) Solution and Precipitation Stage

Thorium received as a mitrate in drums is weighed out and dissolved in dilute mitric acid and omalic acid. Thorium oxalate, the precipitate, is filtered in an Einco press. The thorium oxalate is dumped into 30 gal. drums in a damp state and scaled.

(2) Calcination and Hydrofluorination Stage

The damp thorium explate is transferred to trays and dried. The dried thorium explate is weighed out in trays and calcined in an electric muffle furnace at 1100° F. The trays containing thorium oxide are cooled and dumped into a loading bood. The thorium oxide is transferred to flat type trays, weighed and then placed into reactors for hydrofluorination to thorium tetrafluoride. The thorium tetrafluoride is cooled and packaged in 5 gal. containers.

(3) Metal Reduction

Calcium reduction to the thorium biscuit is accomplished by adding a mixture of sinc chloride, calcium fluoride and thorium

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tetrafluoride to a dolemite-lined bomb and gas firing. The biscuits are unloaded from the bomb and conveyed manually to the thorium casting area.

(h) Thorium Metal Casting

By two successive vacuum furnace castings, the impure thorium biscuit is desinced and terminally purified to the thorium billet.

(5) Machining operations are then performed on each of the billets. These operations include saving, turning and milling and cropping.

#### 2. Beryllium

Beryllium operations consist of the manufacturing of beryllium shapes and crucibles from raw beryllium oxide.

#### Discussion

In order to interpret the data of this report, it is necessary to have criteria for judging whether or not an exposure is potentially hasardous. For this purpose, the following maximum allowable concentrations are suggested:

1. Thorium:

Although there is no generally accepted MAC for thorium, we are tentatively using 70  $d/m/M^3$ , which is the level which has been in use for some time for insoluble uranium compounds.

2. Thorons

As in the case of thorium, there is as yet no generally accepted MiC but the Conference of Governmental Industrial Hygienists has tentatively proposed  $10^{-11}$  curies per liter, the value used in this report. This is equivalent to 2.2 x  $10^4$  d/m/M<sup>3</sup>,

3. Beryllium:

The Commission recommendations for control of beryllium hasards are:

a. The in-plant stmospheric concentration of beryllium at beryllium operations should not exceed 2 micrograms per cubic meter as an average concentration throughout an 8 hour day.

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Even though the daily average might be within the limits of recommendation a, no personnel should be exposed to a concenъ. tration greater than 25 micrograms per cubic meter for any period of time, however short. c. In the neighborhood of an AEC plant handling beryllium compounds, the average monthly concentration at the breathing sone level should not exceed 0.01 microgram per cubic meter. In the course of the study, it was found that all thoron daughters exposures and, therefore, the probable thoron exposure, except that of the HF operators, were below the permissible limit and that one only slightly above. The thorius exposures, on the other hand, were found to be high in several cases. The operators working with thorium mitrate tetrahydrate, thorium oxalate and the calcined oxalate, in general, had exposures ranging to as high as h0 times this level, the maximum expos-ure occurring to the HF operators of  $3100 \text{ d/m/H}^3$ . These individual values are given in Table I. Table II, which shows the breakdown of operations giving the individual exposures for each, indicates that the marinum exposures occurred on the tray handling operations where values of 12,000 and 17,000 d/m/M3 were found. These are single sample walkes while those above are daily weighted average exposures, The following points were made in the meeting between Kr. Harris of this office and various of the laboratory personnel: 1. In general, ventilation which is supplied to the areas appears to be adequate. Most of the operations could be modified or controls supplied without major expense. 2. Housekeeping in the industrial areas is poor and no adequate means for housekeeping is provided. 3. Tracking through the laboratory is quite general and off-plant tracking appears to be likely. Compressed air hose cleaning and man-cooling fans probably add sig-Ŀ. nificantly to the dispersion of airborne dust-Table III, which gives the average general air concentrations, indicates very high concentrations in Room 303 (drying and calcining area) with moderately high concentrations in various areas of Room 33. is far as it was possible to investigate it during the period of study, the weighted averages of exposures to beryllium are satisfactory. Individual high exposures were found, however, which should be corrected. USA 012518

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The radiation measurements which were taken indicate values of gamma radiation up to 22 mr/hr. It can be seen from Table IV that older thorium billets show radiation buildup. For example, a reading taken 6" from the long side of 12 boxes of billets showed 16 mr/hr when the billets were 3 weeks old, 9 mr/hr near billets which were one week old.

Several recommendations are included in the report. If these recommendations are followed, it should be possible to reduce all exposures to within acceptable limits.

## Recommendations

In order to reduce airborne contamination in the thorium and beryllium areas, the following recommendations are presented:

- 1. Thorium Production
  - a. Thorium Extraction Area

Rec. #1 Provide additional ventilation at site of loading thorium nitrate tetrahydrate into hopper. Unloading of thorium nitrate tetrahydrate should be performed inside same hood. This hood should also include all weighing apparatus required for the extraction operation and should be adequately ventilated.

- Rec. #2 Eliminate broom sweeping. Use vacuum cleaner which should be exhausted out of doors.
- Rec. #3 Eliminate use of cooling fans.
- Rec. fb Provide wall exhaust fan behind alurry tanks.
- b. Calcining and Hydrofluorination Area
  - Rec. #5 Local exhaust ventilation should be provided at front of dryers and calcining furnaces.
  - Rec. #6 A buggy should be devised to transport trays from loading hood to dryer and calciner and HF furneces and back.
  - Rec. #7 Tray weigh scale should be incorporated with the loading hood.
  - Rec. #8 Cooling areas and tables should be adequately ventilated or hooded.

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See Los Nor 2 View		confines of a hood			
		Eliminate all broo	eaner should be used in this on and foxtail sweeping.		
	Rec. #1	1 All drums when not	t in use should be kept lidde	ed.	
C.	Thorium				
. *	Rec. #1	- P Whith Into owing	d enclosure at site of drum of der. Enclosure should be pro- door, glove openings and cove		
	Rec. 🎵	3 Ventilation should charging ends of a	d be provided at charging an mixer.	d dis-	~
	Rec. #1	-	stem should be installed for	use in	
	Rec. #1	5 Provide local exh bomb capping area	enst ventilation at bomb loa S.	ding and	
	Rec. A	6 Vacuum clean tops ing same to toppi	and sides of bomb before tr ng and capping areas.	ensport-	
đ.	Metal C		÷	F	
	Rec. #1	7 Provide floor gri ing desinced bill	ll type exhaust hood at site ets from graphite pots.	of dump-	
	Rec. 1	8 Provide local exh desinced billet i	anst ventilation at site of nto beryllium crucible.	loading	
	Rea. F		acuum cleaning system in the ta and machining and berylliv t area (Rooms 29-22 and 15)	rius or 674-	
•	Rec. #	20 Eliminate use of parts.	air hosing when cleaning off	furnace	
•	Rec.	21 Provide exhaust 1 clean out area.	mood for furnace unloading, I	erts and	
	Rec. #	22 Exhaust discharge into ventilation system.	e portable vacuum cleaner non system or change to central	r in use vecuum	
			-11-		ť
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Rec. #23 Discharge the effluent of the Minney vacuum pump into the ventilation system.

. Machining Areas

Rec. #24 Provide local exhaust ventilation at milling machine, Room 22.

Rec. #25 Provide exhaust ventilation at saw and lathe in Room 15.

#### 2. Beryllium Operations

Rec. #26 Confine all loading and weighing operations within a ventilated hood.

Rec. #27 Provide ventilated enclosure at site of dumping BeO and lime charge into large mixer.

Rec. #28 Provide ventilation at discharge of large mixer.

3. Operating Criteria

Rec. #29

- 1. The following operating criteria should be met in the Metallurgical Building:
  - a. The daily average personnel exposure to long-lived thorium alphas should be less than 70 d/m/M3.
  - b. Gamma radiation maximum or whole body or any part (except hands or forearms) should not exceed 300 mr/wk.
  - c. Bats plus gamma, whole body or any part (except hands or forearms) should not exceed 500 wreps/wk, no pore than 300 of which should be gamma.
  - d. The hands and forearms should be less than 1000 mr gamma or 1500 mreps beta plus gamma.
  - e. Weekly average concentration of therium dust should be no greater than 0.7 d/m/H<sup>3</sup> at all places beyond the site perimeter. (Based on 1% of in-plant preferred lovel)

#### Rec. #30

2. A weekly radiation survey of all production, locker and lunch room facilities should be made. In addition, a reachly "apot" survey should be made in the other Metallurgy Building facilities.

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Rec. #31

Suitable radiation warning signs should be provided in all drum, tray and metal storage areas. The practice of roping or sectioning off of areas where known direct radiation hasards are present should be encouraged.

L. General

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Rec. #32 Install a personnel monitoring service which will include film badge service, radiation monitoring, etc.

- Rec. #33 Provide work clothes, cover shoes and shoes and hats for all operating personnel.
- Rec. #34 Provide two room change lockers, one for contaminated clothing and the other for non-contaminated street clothes.

Rec. #35 Provide shower facilities for personnel.

Rec. #36 Provide supervisory control to insure adequate housekeeping throughout the operating areas.

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NOTICE: This document has been reviewed to identify and redact any information that is protected by the Privacy Act 5 USC §552a and has been cleared for distribution.

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DATES WE	TABLE	I EXPOSURE - AMES I	ABORATORY
406	No. of Employees		Concentrations
		Thorium	Thoron
Einco Unloader	1	7.7	22,000
Extraction Loader Operator	1	111	19,000
Drying Operator	1	1500 -	1,400
HF Operators	2	3100	30,000
Foreman, Thorium Crude	1	84	3,800
Bomb Unloaders	2	հր	3,100
Borb Loaders	2	852	14,000
Packing & Jolting	2	69	1,800
Weigh Man	1	791	19,000
Poreman Metal Casting	1	0.89	1,900
Casting Operator	1	0.06	1,000
Half-time Dezincing Operators	3	49	1L,000
Casting & Dezincing Operators	3	66	19,000
Thorium Milling Opera- tor	1	61	6,h00
Thorium Machining Operation	1	276	2,000
TOTAL PERSONNEL	22		
AVERAGE DAILY WEIGHTED	CONCENTRATION	530	13,000

\* Thorium data extrapolated to time of sampling.



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TABLE II

<u> </u>	made	194	Ε	1	-	
Box 194	March 18	Sec.2	k	10	5.000	ŗ.

## TABULATION OF AVERAGE BREATHING ZONE SAMPLES - AMES

Ope	oration	Avg. Concent Thorium	tration (d/m/M <sup>3</sup> ) Theren	No. of Samples
The	rium Extraction			
1.	Unloading 190# Fibre Pak carton of TNT in hopper	1170	h6,000	3
2.	Loads up correct weight THT in 3 small cartons	3000	73,000	3
3.	Dumps Cartons containing TMT into tank	0	15,000	2
<b>L</b> .	Discharging thorium Cx- ylate into 30 gal. drums	7	52,000	3
The	rium Calcination & Hydrofl	uorination		
1.	Losding (1) tray(s) thori orylate (in hood) from 30 gal. drum (outside hood) a conveying to driver	ŀ	0	Ŀ
2.	Unloading 2 trays from drier, conveying to hood & dumping trays	1770	10,000	3
3.	Loading therium explate into 2 trays for cal- ciming	1730	7,000	3
<b>L</b> .	Transporting therium Oxy- late trays from load hood to storage area		25,000	2
5.	Sweeping up thorium oxy- late and thorium oxide in hood using broom and vacuu cleaner		0	1
6.	Unloading calcining fur- nace removing & trays thorium axide	6000	100,000	2

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TABLE II (Cont'd)

		Avg. Concentration (d/m/M <sup>3</sup> )		No. of	
Coe	ration	Thorium Thoron			
	Loading calcining fur- nace with h treys thorium oxylate	11800	30,000	2	
8.	Unloading 2 trays thorium oxide in hood (weighs trays and then damps in hood)	10800	125,000	2	
9.	Unloading one drum thor- ium oxide in large hood	7650	360,000	2	
10.	Leading & treys with thorium oxids in large hood	5800	117,000	3	
11.	Weighing h trays, making up weight and trans- porting to open storage area	2070	27,000	3	
12.	Levelling off therium order in 6 trays	12000	120,000	2	
13.	Sweeps up thorium exide and thorium exylate	1000	0	1	
ц.	Removing 12 thorins oxide trays from cool- ing area (Bench) and inserting trays in fur- nace. Scaling furmace.	132	9 <b>,</b> 600	3	
15.	Unloading h large trays thorium oxide into large hood. Weigh & dump.	16800	0	1	
16.	Unloading therium fluor- ide furnace. Opening 3 furnaces.	270	33,000	1	
17.	Unloading 12 trays thorius fluoride in sets of h onto buggy carts	78	19,000	3	
18.	Transporting 12 trays on 3 buggies from HF furnace room to calcining room for cooling. Unloading buggy	<b>r</b>	19,000	2	
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- 1; -		TABL	Ave. Concer	d) ntration (d/m/M3)	Ho. of	-
Oper	ation		Thorium	Thorop	Samples	
-	fluoride with for		51.00	215,000	2	
20.	fluoride	g 3 trays thorium from cooling tighs & dumps into hood, haumering tray	3720	hh0,000	3	•
21.	into 5 s	thorium fluoride gal. lined cans, k scals can	7300	180,000	3	
The	rius Cro	de .				
1.	Dumping	1 5-gal. drum of fluoride into grad	er hhoo	100,000	3	
2.	Emptyin gal. dr	g hooper into 30	C	0	3	
3.	chlorid	sixer with sinc e, thorius fluor- alcium fluoride	7600	130,000	3	-
4.	tharits	ng mine chloride, fluoride & cal- uoride from mixer	7400	150,000	3	
5.	Loading ThFh, Z	bomb-liner with a Cl and CaFl charg	e 5200	80,000	3	
6.	Ding 41	rting bend to top- es, top, add e plug, limes & comb	2130	10,000	3	T T
7.	Opening loading bomb	1 boab and un- line from top of	٥	٥	3	
	Drillin bomb	ng out crucible from	• •	0	3	
			5	DE CARAL		
				a long R	USA	012526

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	<u></u>	TABLE 1	I (Cont'd)	
<b>0</b> p	eration .	Nyg. Con Thorium	centration (d/m/M <sup>3</sup> ) Thoron	No. of Samples
9,	Tamping & dumping alag from bomb	0	0	3
10.	Chipping slag from billet with hasmor & chisel	0	5,000	3
11.	Removing slag with air bacmer	0	0	. 3
12,	Cleaning work area	0	0	3
Net	al Casting			
1.	Unlosding Desinced Bil	let		
1.	Removing furnaces by cart to boist, removin quarts cover tube & insulation bricks	e Q	13,000	3
2.	Remove 2 graphits pots containing 2 desinced hillets. Unloads by dumping on floor, weig & removes to chipping area		80,000	3
3.	Chips 2 billsts & blow off dust with air hose		0	3
в.	Loading Desinced Bills	La		η.
1.	Inserting 2 demineed billets into Be cruci- ble, adds thorium scru and places top on cru- cible	b 263	10,300	3
2.	Placing quarts tube around Be crucible. Adds graphite between quarts tube & crucible. Places quarts cover tube on furnace. Air			
	hosing unit.	0	17,000	3
	×	i	STORET,	

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TABLE II (Cont'd)

o,	SECRET perstion	Avg. Concent: Thorium	ration (d/m/m <sup>3</sup> ) Thoron	No. of Samples
3	. Removing furnaces to remeit areas. Secures cover tube with scalar	0	o	3
4.	Removes quarts furnace top, graphite top & vacuums furnace	o	3,500	2
5.	Removes quarts inner tube graphite. Blows off fur- nace	0	3,000	2
6.	Unloading furnace insula- tion & thorium metal casting	o	16,000	2
7.	Cleaning off furnace parts brick insulation	250	76,500	2
a.	Cleaning out inside fur- nace cover tube with scraper, brush & air hoss	0	0	ż
9.	Cloaning graphite heater pots that hold crude bis- cuit	0	17,000	2
10.	Loading 2 crude biscuits into pots, place insula- tion brick around pots, place quarts tubes on fur- nace. Air hoses.	29	1,600	2
Kao	hining			
1.	Milling or cropping therius metal pieces	<b>7</b> 9	6,100	3
2.	Sawing therium billst	310	1,700	3
	5 C			

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TABLE III

TABULATION OF AVERAGE GENERAL AIR SAMPLES - AMES

Location	Avg. Come Thorium	entration d/m/M <sup>3</sup> Thoron	ho. of Samples
Thorium			
Extraction Area (Rm. 203)	9	17,500	8
Drying & Calcining Area (Rm 303)	1780	1,700	h
Hydrofinorination Area (Rm 307)	34	0	5
Therium Crude Area (Rm 33)	<b>9</b> 5	3,600	11
Bomb Storage Area (Rm 33)	195	8,500	2
Metal Casting Room (Rm 29)	1	1,600	6
East Control Panel (Bm 29)	o	600	2
West Control Panel (Rm 29)	0	700	2
Furnace Cleanout Area (Ra 22)	0	6,800	3 .
Corridor outside Rm 203	0	1,100	2
Seminar Room (Lunch / rea)	ò	2,800	2
Jolting Area (Rm 33)	38	3,300	2
Lime Packing & Tamping Area (Rm 33)	195	8,500	2

Beryllium

Be Furnace & Preparation Area (Rm 28)

0.4 ug/m3

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### TABLE IV

### SECTOR I RADIATION MEASUREMENTS

r <b>82</b>	Location	Alpha c/a/100 cm <sup>2</sup>	Beta Beta mreps/hr	Games wr/hr
				,
traction	General Ekgd. Radiation (Room)	0	0.1	0.35
(Rm 203)	THT Weigh Scale	34,000	<b>Q.6</b>	1.9
	THT Drum Storage Area			
	Top Drum	2,000	0	15.0
	Side Drum	3,000	1.5	17.0
	Floor	5,000	0	2.0
	THI Load Hopper (on THI)	50,000	6	<b>1</b> h
	Floor	21:,000	0	0.95
	* TNT Tank #1 (Espty)			
	Inside	0	0.3	0.5
	Outside	õ	0	0
	TMT Tank #1 (Full)			
	Inside	0	1.0	6.0
	Outside	õ	0	5.5
	Step in front of 3 Slarry			,
	Tanks	32,000	0.1	0.6
	Einco Filter (ThOx) material	50,000	2.5	1.5
	55 gal. Drum ThOx Half Full	0	0	4.5
	55 gal. Drum ThOx Full	50,000	5.0	7.0
	Filter Press			
	Top	3,000	0	0.1
	Side	3,000	0.8	0.7
	Process Floor			
	Avg. Reading at Filter			·
	Press, Rimco & Slurry	34,000	0.1	0.75
	Tank Areas			
	Work Bench	3,000	0.05	0.1
	Sink	15,000	0	1.5
	104 & Juno used for all measured	monts	-	
B 0 20.	nasurements for Tanks #2 and #2	3		

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TABLE IV (Cont'd)

Lres .	"Location"		Heamiremen	
		d/n/100cm <sup>2</sup>	Beta mreps/hr	Gamma mir/ha
Thorium Ex- traction (Rm 203)	Corridor Outside Th. Extraction Room	5,000	0.02	0.11
Calcining & Hydrofluoring-	General Bkgd. Radiation Room	o	0.3	0,5
c10n	Floor	30,000	0.05	1.8
	Th02 Trey (62 x 11 x 23) Inside Outside		3.0 0	8.5 5.0
	ThFL Tray (Fresh from Furnace) (11 x 11 x 30) Top Bottom		10 0.3	4.0 4.0
	ThFL Tray (cooled for 17 hrs) Top Bottom		1 0.75	3.0 2.0
	THOI Tray (62 x 11 x 23) Top Bottom		2.0 0.1	6.5 3.0
	General Room Bkgd.		0	0.2
Room 33)	Floor around Mixer		0	1.0
	5-gal. can ThFL Gover off-Top Material Cover on-Top material	100,000 2,000	9.5	9.8 11.5
	ThFh Can Storage Area Side - 7 cans	2,000	0	22
	Crude Billet	100,000	4.5	5.5
	Crude Billet Storage Area (Flogr)	20,000	2.0	1.0

\*\*\* Juno Contaminated.



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TABLE IV (Cent'd)

90 S		Baddatt	Rediction Measurement		
Area	Location a	Alpha	Beta	Garran	
		d/m/100cm2	mreps/hr	mr/ar	
	Average Desinced Billet				
Th. Netal	Botton		3.5	9.5	
Reduction &			Off Scale	10.5	
Casting	Top Side		2.5	5.0	
	Average Cast Billet (Fresh)	)	1.0	5.0	
	Bottom		1.0	<b>b.</b> 0	
-	Middle				
	Top		6.0	7.0	
e	Average Cast Billet (20 hrs old)			_	
	Bettom		1.0	1.0	
	Hiddle		3.0	1.0	
	Top		1.0	3.5	
Generator	General Room Bigd.		0	0.3	
Thorium Mach	- Pinished Billet Storage				
ining Shop	Area				
	12 Boxes (billets) 1 wk c	14		•	
	Length (Contact)		2.0	16	
	End (Contact)		0	12	
	Longth (6")		2.0	9.0	
	End (6")		0	7-0	
	Length (1º)		2.0	6.5	
	End (1')		0	F*0	
	12 Bores (billets) 3 wks	old			
	Longth (Contast)		1.0	Off Seals	
	End (Contest)		1.0	1 <u>1</u>	
	Length (6*)		1.0	16	
-	End (6")		1.0	8.0	
	Length (1)		1.0	9.5	
	End (1')		1.0	6.5	
	Floor between Crude & Hetal				
Corridor	Hachining (after washing)	15,000	0.5	0.05	
	Steps to Locker	3,000			
	•				

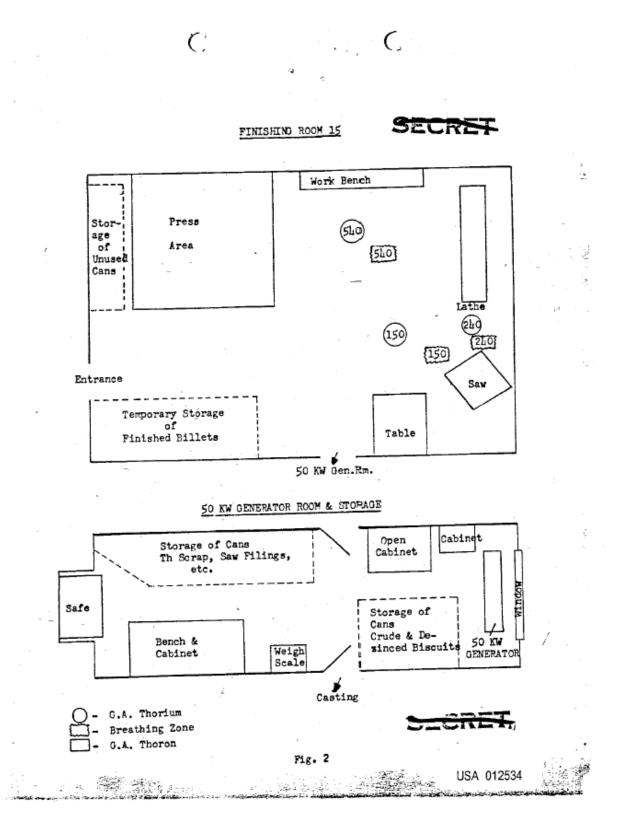
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• •		WEIGHTED AVE	RAGE EXPOSURE	(MAC)	SECR	
	Occupation	0 1.0 2	.0	5.0	<u>ULCR</u>	10,0
		· ·				
	Eimco Unloader					-
	Extraction Loader Operator	1				1
	Drying Operator					44
	HF Operators Foreman, Thorium Crude	1				्र देख इ इ इ
	Bomb Unloaders	2		-	to ]	2.2
	Bomb Loaders					
	Packing and Jolting					10 11 20 1
	Weigh Man					
,	Foreman, Metal Casting	<b>P</b>				
	Casting Operator					DAT
	Half-time De- zincing operato					AM N
	Casting and De- zincing Operato Thorium Milling					EIGH
	Operator		1			ABOR
	Thorium Machin- ing Operation			•		DAILY WEIGHTED AVERAGE EXPOSURE AMES LAEGRATORY
					-	AGE B
	-					XPost
	Total Personnel				22	RE
				LEGEND Thori	um (	
				Thor	on Room	£
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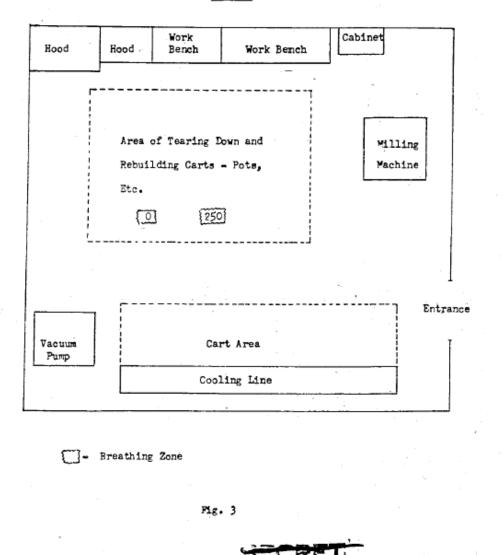
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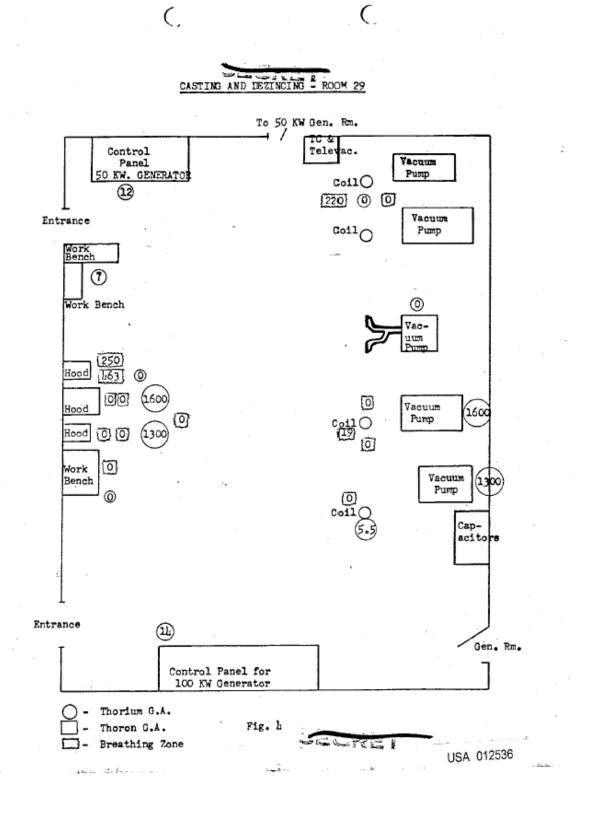
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#### CARTS SET UP & TORN DOWN FOR INGOTS & COOLING LINE

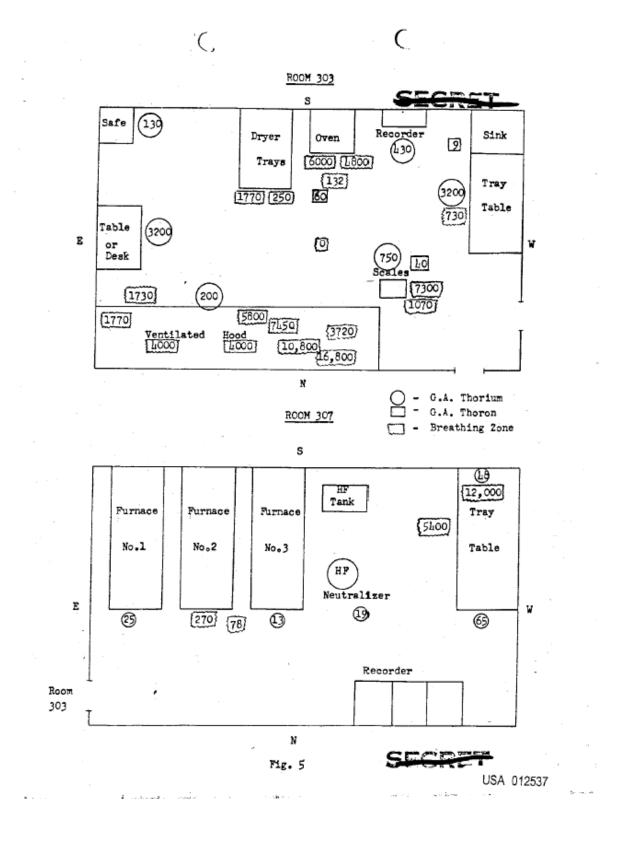
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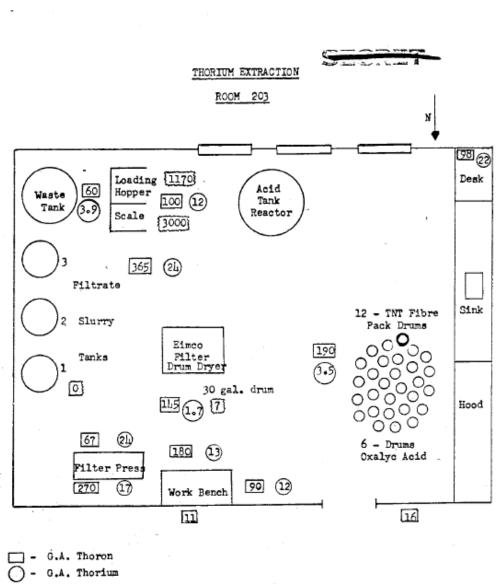


Fig. 6

Breathing Zone

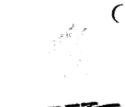


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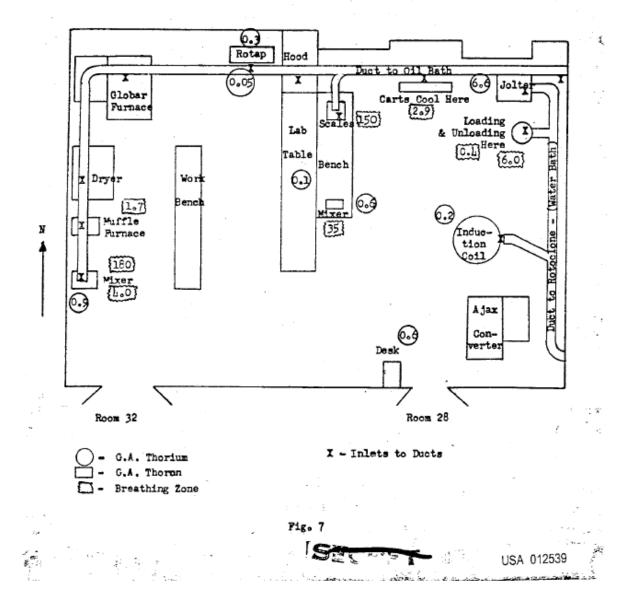
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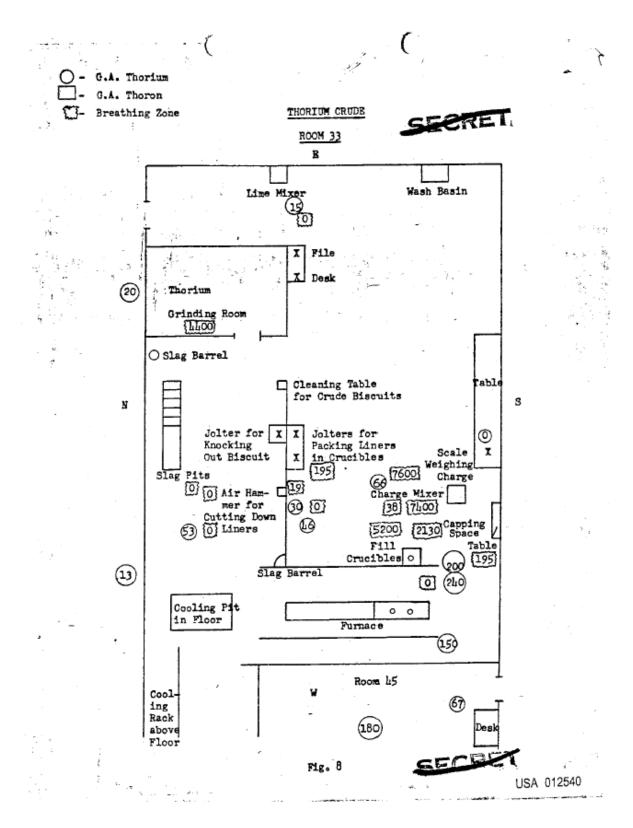
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#### BERYLLIUM OPERATION



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# APPENDIX C: 2006 BIHL MEMO

# Battelle

Project No.

Internal Distribution

The Business of Innovation

Date December 29, 2006

To NIOSH Dose Reconstruction Project Files

From Don Bihl DBihl

Subject Estimated Releases from the Hot Laboratory in Ames Chemistry Building

Maximum inventory allowed in the hot lab was 5 Ci (Spedding 1947).

It was assumed due to activities in the hot lab described as experiments on techniques for separating the plutonium out of the irradiated fuel, that .002 of the inventory went airborne (from DOE Handbook 3010, worst case for boiling liquid, page 3-1 [DOE 1994]), that the hot lab exhaust was filtered. HEPA filtration efficiency for particulates was 99.95% (penetration of 5E-4) (HEPA filtration minimum standard in early HEPAs, Burchsted et al 1976); however, because HEPAs were under development in the late 1940s, it is unlikely the Ames hot lab used a HEPA filter. Other filters, such as woven fiberglass filters, were available and were almost as good except for the smallest particles. (The 99.95% efficiency for HEPAs is for a particle size of 0.3  $\mu$ m; the efficiency for other particle sizes is better.) Considering all particle sizes, a 99.5% efficiency was consider reasonable. Since this was an experimental lab, not a production facility, it was assumed that the throughput was 10 times the maximum inventory, or 50 Ci/yr.

Total release was (5E13 pCi/yr)(.002)(5E-3) = 5E8 pCi/yr.

Using X =  $F^*Q/V$  from NCRP 123, where X is the annual average air concentration at ground level in pCi/m<sup>3</sup>; F is fraction of time plume goes in any one occupied direction = 0.25; Q is release rate per second; V is flow rate of the exhaust system in m<sup>3</sup>/s with a default of 0.3.

Q = 5E8 pCi/yr/3.15E7 s/yr = 15.9 pCi/s

 $X = .25*15.9/.3 = 13.2 \text{ pCi/m}^3$ 

Total activity breathed is X\*2400 m<sup>3</sup>/yr breathed = 3.17 E4 pCi/yr.

IMBA needs the daily intake, so 3.17E4/365 = 87.0 pCi/d.

Another approach is to compare the release fractions from the hot lab to the early, unfiltered releases from dissolution of irradiated fuel at Hanford. For particulates that unfiltered release fraction was estimated to be 2.1E-5 (Till et al 2002). This results in a annual release of (5E13 pCi/yr)(2.1E-5) = 1.05E9 pCi/yr, essentially within a factor of 2 of the prior estimate. Considering that the Ames hot lab was on a college campus, it is unlikely that the releases were totally unfiltered.

OTIB-054 was developed to help interpret intakes of fission products from irradiated fuel when air samples are just listed in terms of gross beta. Table E-2 from OTIB-054 gives the relative ratios of fission products that deliver significant dose at several cooling times (10, 40, 180, 365 d). Of

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these the 180-d cooling time was used under the assumption that short-cooled fuel with lots of iodine still present would not have been shipped to Ames.

	Default			
Radionuclides	10 d	40 d	180 d	1 y
Ba-140	0.0334	0.0299		
Ce-141	0.0388	0.0887	0.0221	0.0818
Cc-144	0.0209	0.0704	0.2191	0.3023
Cs-134	0.0004	0.0014	0.0054	0.0119
Cs-137	0.0016	0.0054	0.0208	0.0507
Eu-155	0.0001	0.0004	0.0014	0.0031
Fe-55	0.0022	0.0061	0.0172	0.0312
I-131	0.6904	0.2366		
La-140	0.0383	0.0297		
Nb-95	0.0368	0.1374	0.2492	0.1170
Pm-14/	0.0044	0.0151	0.0546	0.1214
Ru-103	0.0296	0.0739	0.0321	
Ru-106	0.0071	0.0265	0.0844	0.1461
Sr-89	0.0248	0.0693	0.0558	0.0119
Sr-90	0.0013	0.0042	0.0157	0.0387
Y-91	0.0304	0.0887	0.0911	0.0297
Zr-95	0.0396	0.1162	0.1311	0.0543

Table E-2. Default source term for intake calculations.

So the intake of each radionuclide is just the fraction from the table under the 180 d column times the total release of 87.0 pCi/d.

Radionuclide	Relative	Intake,
	fraction	pCi/d
<u>Ce-141</u>	0.0221	1.92
Ce-144	0.2191	19.1
Cs-134	0.0054	0.470
Cs-137	0.0208	1.81
Eu-155	0.0014	0.122
Fe-55	0.0172	1.50
Nb-95	0.2492	· 21.7
Pm-147	0.0546	4.75
Ru-103	0.0321	2.79
Ru-106	0.0844	7.34
Sr-89	0.0558	4.85
Sr-90	0.0157	1.37
Y-91	0.0911	7.93
Zr-95	0.1311	11.4

Because maximizing assumptions were used for most of the parameters in this analysis, the distribution is an upper bound (constant). The most claimant favorable absorption type for each radionuclide, as listed in ICRP 68, should be used with the exception that only type F should be used for strontium.

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Reviewed by Bruce Napier, 12/29/2006

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by means of HCl. This process was developed on a pilot-plant scale, at Ames, to produce kilogram quantities, and turned over to the Madison Square Area with a veiw to employment for large scale development at Oak Ridge.

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#### 3. Plutonium Chemistry

One of the major problems facing the Plutonium Project in 1942, was to devise a number of processes for separating plutonium which it was contemplated to produce the proposed piles, from the fuel uranium and from the fission products which would be formed during the reaction. These processes had to be simple, capable of being engineered on a large scale and capable of operation under remote control with a minimum of operational maintenance.

The radioactivity of the fission products was expected to be so great that it would be necessary to conduct all operations behind thick concrete shields and the equipment was expected to be so containinated at the start of the operation that any subsequent repairs, replacement or maintenance would also have to be done by remote control. Furthermore, due to the great value of the plutonium its recovery had to be practically a hundred per cent and it was necessary to obtain the material extremely pure and free from residual redioactivity from the fission products. The problems were made more difficult by the fact that the plutonium was expected to be extremely poisonous in the form of the dust and that large amounts of it could not be accumulated in one place without danger of explosion. The problem was still further complicated by the fact that almost no plutonium existed nor could it be obtained in any quantity until the piles were constructed and the required processes performed.

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The only plutonium available at that time consisted of a few micrograms which could be obtained by cyclotron bombardment. Therefore, one of the first problems facing the chemists was to develop chemical and ultra-microchemical techniques which would enable the chemists to determine the physical and chemical properties of the salts, solutions and metal using such minute quantities. The major emphasis on plutonium chemistry was centered at Chicago. However, California and Ames also contributed to this program.

A number of processes for separating decontaminated plutonium were worked out on the project on a laboratory scale. Finally, one of these processes, the bismuth phosphate process primarily developed at Chicago, was selected for development to large scale production at Hanford. The alternative processes, although several of them could have been successfully stepped up with further work, were deemphasized or discontinued at that time in order to advance the selected process. A number of these alternative processes were important as insurance and seemed capable, in the long run, of being cheaper and more efficient than the one adopted. It seems likely in the future that several of these will be put into operation in connection with the development of peacetime atomic energy power piles.

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Conferences were held in Chicago almost weekly, at which the information obtained in the several laboratories was correlated. It is accordingly impossible to say that any process was solely developed by a single laboratory. In one process a given laboratory might play a major role while the reverse might be true for another process. The Ames Project took an active part in this program.

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In the summer of 1943, the Ames laboratory became interested in a dry process for separating plutonium from the fission products and uranium. The process was based on the preparation of uranium hydride from the metal, followed by conversion to the tetrafluoride and then to the hexafluoride gas. Subsequent reduction of the uranium hexafluoride to the tetrafluoride, after separation from plutonium, was to provide a compound which could be readily reduced to metallic uranium. This process was the outgrowth of an earlier one, developed at Chicago, in which the uranium was converted into UF6 by reactions producing first  $U_3O_8$ , then  $UO_2$ ,  $UF_4$  and finally  $UF_6$ . A different temperature (500-750°C) was required for each step and some of the gases were highly corrosive, for example the HF-1:20 mixtures. The discovery of the hydride suggested an alternative procedure with many advantages. The tetrafluoride could be produced in two steps, hydride formation and hydrofluorination, both at 250°C, with dry HF the only corrosive gas. Studies on this procedure showed it to be feasible though conversion of large amounts of U to UF4 was slow. A simultaneous reaction procedure was also tried in which both gases were allowed to attack the uranium at the same time. This was also shown to be feasible though again slow due to caking of the reaction products around the uranium metal. During the process the rare gases of the fission products were liberated and it was demonstrated that they could be recovered from the other gases for experimental use, if desired.

Since the final reaction, conversion of  $UF_4$  to  $UF_6$ , required fluorine gas, methods for producing it were studied. A number of electrolytic cells using XF-2MF as electrolyte and operating at a medium temperature (75-90°C) were designed and built. Principal features of the design were a trichlorethylene bath for temperature regulation, a rectang ular iron -39-

tank as container and cathode, a nickel plate as anode and the highly resistant polytetrafluoroethylene plastic (Teflon) as gasket and insulator material. They were considerably better than anything then in the literature, although extensive work in another part of the Manhattan District, with which we had no contact, had preceded this.

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One of the chief problems in the dry fluoride process was the question of the volatility of the higher fluoride of plutonium. Work was done on this problem by the Ames group which contributed to our knowledge but no clear-cut understanding yet exists as to the volatility, stability and formula of the higher fluorides of plutonium.

In the dry fluoride process the uranium and a few of the fission products were removed from the rest of the fission product impurities and from plutonium by volatilization of their higher fluorides. Thus a large percentage of the radioactivity could be collected with the plutonium in a highly concentrated form. The uranium and the volatile fluorides of the fission products could then be separated by fractional volatilization or other methods; the uranium could be recovered, in a decontaminated state, in a form which could be directly reduced to the metal by the procedure described in the Metallurgical section of this report. The specific separation of radioactive columbium was studied at Ames as representative of those volatile fission products whose fluorides could be removed from the plutonium at the temperatures employed. Only preliminary work was completed on the separation of plutonium from non-volatile fluorides by distillation with subsequent collection of plutonium fluoride. It should be pointed out that this separation could be effected by other means than volatilization. During this period, attention was called to the fact that the dry fluoride

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process might be adapted, with further development, to the recovery of uranium from the uranium wastes of the bismuth-phosphate process.

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Considerable work was done at Ames on a metallurgical process for the separation of plutonium which is described in the Metallurgical section of this report.

An alternative process, worked on at Ames, was the development of a separation method based on the adsorption column procedure developed at another site, but with additional steps using a complexing reagent. The reagent which proved successful, 7-iodo-3-hydroxyquinoline-5-sulfonic acid, known as ferron, forms a stable complex with Pu which, in a certain pH range, is not adsorbed by the column of ion-exchange resin which does adsorb most of the fission products.

Since the plutonium and the bulk of the fission products had already been separated from the uranium by the previous process, these materials could be deadsorbed, the plutonium could then be complexed with the reagent and repassed through the column at the appropriate pH. Under these conditions the fission products would again be adsorbed but the plutonium would pass through without being adsorbed. This process led to a high decontamination of the plutonium. A hot laboratory, capable of handling 5 curies through the adsorption process, was designed and built. The method proved successful in a number of runs. The uranium and plutonium were separated from one another and from the fission products using 1 kg. samples of uranium, from the pile, and which had an activity of 5 curies.

After appreciable amounts of plutonium became available from the piles it appeared that it would have to be obtained in the metallic state and extremely free from light element impurities, if it was to be used for weapons. An extensive "purity" program was set up in cooperation with

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the various sites and the Ames Project took part in this program.

One of the several lines of action, carried out at Ames, was to conduct an exhaustive study into the organic complexes of plutonium. A number of these complexes should be extractable into organic solvents and it was hoped that one of these would be specific with respect to plutonium or at least highly selective. The most successful reagents were derivatives of disalicylalethylemediamine which led to complete extraction of the plutonium. Such a procedure would have been quite effective had not the "purity" program been cancelled six months after its initiation. An attempt was made to use these reagents in a decontamination process, but they did not give sufficient separation from some of the fission products. Some of these complexes offer considerable promise as reagents to be used in the liquid-liquid separation processes for plutonium which are now being developed.

Weighable amounts of neptunium were available much later than in the case of plutonium. Thus, although it was known that plutonium had stable valences of 3, 4 and 6 with a less stable one of 5, similar information was not available for neptunium. Experiments with tracer neptunium were designed to determine whether there was a stable valence lower than 4. Rather surprisingly it appeared from the tracer experiments that a value intermediate between the lower valence (4) and the higher one (6) was stable in aqueous solution. It was suggested that Np<sup>+5</sup> existed, a suggestion which was substantiated elsewhere when micro amounts of neptunium became available.

Since the handling of micro and ultra-micro quantities was a necessity, some thought was given to the design of equipment specifically intended for such use. Chief among the results was a magnetically controlled quartz fibre balance which could be operated externally to the system,  $\chi^1$ 

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weighings being made by means of a potentiometer. Development was also made of a magnetic susceptibility balance using quartz fibre suspension.

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#### 4. Thorium Chemistry

#### 4.1 General Chemistry

As soon as the first piles were in operation, and it was generally realized that thorium could be used to extend the available sources of fissionable material, it became necessary to enlarge the then rather meager basic knowledge concerning the chemical, physical and nuclear properties of this element. Practically all of the research programs which had been set up for uranium studies had to be paralleled for thorium. The Ames Project played a major role in this field, specializing in studies on the properties of inorganic compounds of thorium. Among the results which have been reported in numerous project notes, reports and papers, it might be mentioned that the thorium-hydrogen system was found to consist of two different hydrides.

#### 4.2 Purification of Thorium Compounds by Liquid-Liquid Extraction

Late in the summer of 1944, when the Ames Project started work on the large scale production of thorium metal, research was initiated for developing a method of preparing thorium salts free from elements having high neutron-capture cross-sections. Thorium salts available commercially were sufficiently pure with respect to all of the undesirable elements except the rare earths. Hence, the problem became one of separating the rare earths from thorium.

Two approaches were considered in solving the problem: one to find a suitable complexing agent which would facilitate the separation and

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# APPENDIX E: TELEPHONE INTERVIEW WITH MR. ON JANUARY 3, 2007

Behling: Today is January 3<sup>rd</sup>, 2007. This is Dr. Hans Behling, and I am interviewing Mr. Will you please identify yourself for the record and perhaps give us your mailing address?

: This and my address is

Behling: Mr. **Sector**, let me just ask a few questions that will clarify what your role was at Ames Laboratory? Can you give us the years in which you were employed?

: Well, I am not really sure, but I believe that I started there in either and I was there for years.

Behling: So, you were there from approximately through

: Yes.

Behling: Can you give us a brief description and is it possible that you might have had several types of jobs, but can you give us an overview as to what type of work you were doing at that time?

: I worked at what they called the Building, and we got I believe thorium, but I am not really sure. It was a white substance that was put into trays that were long and narrow and then they were put into a furnace to be heated and dried.

Behling: Were you a member of the team? In other words, were you considered a worker?

: Yes.

Behling: Okay. And you believe that the material in question was thorium.

: Well, I was told that later on, but I really don't know.

Behling: As a matter of record, I do believe that it had to be thorium because the uranium production stopped before your time of employment. So, I agree with your recall that it was likely thorium. Let me ask you, when you were working as a **second of** worker, were you given any special training about handling the material? Were you given respirators to wear during the working hours? Were you given anti-contamination clothing or gloves? Can you describe the working conditions?

: I was given none of the stuff you just mentioned. We did have to take a shower at the end of the day, and we had khakis and a T-shirt or shirt that they laundered and furnished to us and that is about it.

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Behling: So there were no anti-contamination clothing or gloves that you wore other than the khakis and T-shirt that were given to you and washed by the Ames Laboratory facility – that was the only precaution taken?

: Right.

Behling: And the shower at the end of a day.



: That is right.

Behling: Did people routinely take showers or do you know people who decided that they were not interested in taking a shower?

: Well, the people that I worked with – we all had to take a shower, and we just automatically did it.

Behling: Do you remember ever being monitored for internal exposure? In other words, was there any attempt to assess what you may have breathed in in terms of thorium that would be measured by various assays? Today, for instance, we would use urinalysis; we would ask you to give us a urine sample and that would be analyzed for thorium or, in some cases, fecal analysis where a 24-hour stool sample would be analyzed for thorium. Do you remember ever being given any of these tests—urinalysis, fecal analysis to assess what you might have had in your body?



: No, we were never given anything like that.

Behling: Okay. Let me talk about specific events, which, as I had mentioned to you, are part of my interest here in talking to you. And that is, when we talk about discrete events, I am talking about unusual events that are not necessarily part of the normal working environment. Events that I am specifically looking to get some information on involve blowouts or explosions, as well as potential fires that might have resulted. Do you remember any of those particular incidents during the timeframe when you were working – explosions, blowouts, fires?

: Yes, basically there would be an explosion then a blowout where it would knock out an outside wall of a building.

Behling: Let me ask you again, focusing on these discrete events—explosions, blowouts— what happened when there was such an event? Was there an attempt to get people out of the building or did you stay in the building? Were you asked in some instances to assist with the fire? What happened during these explosions? And, I am going to ask you to describe not only what you may have been doing but perhaps other coworkers who were sharing in that same work environment.

: Well, I have to go back quite a ways; no, we stayed right in the building and we helped with the cleanup; and we seemed like we took it for granted that there could be an explosion.

Behling: During these explosions, do you remember, was there visible contamination that you could see in the air, such as a cloud of dust, material that was airborne that you were breathing

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in? Do you remember anything about the explosions that stands out in terms of visual things contamination that involved airborne dust or anything like that?

: I am afraid that I cannot bring myself to remember anything like that.

Behling: Was there any attempt to decontaminate after such an event? In other words, were there vacuum cleaners used to pick up the dust that might have been dispersed into the work environment or any attempt to clean up surface areas of any contamination that might have resulted from the explosion? Do you recall any clean-up activities?



Behling: So, by in large, you would continue working unless you were involved in putting out the fire or restoring a wall that may have been knocked out. But in essence, you did not run out of the building and avoid any potential exposures.

: Correct.

Behling: Do you have any feel—and I am obviously asking a question that might be very difficult for you-but do you have any feel for how often these events occurred?

: Oh, I would say maybe once a week.

Behling: Once a week?

: I would say that – yes.

Behling: Is that correct? Let me ask you something else, during the timeframe when you said , do you remember an inspection that was done by that you were there between and the Atomic Energy Commission in March of 1953? Do you recall that particular event? There was a three-day inspection that was conducted by the Atomic Energy Commission. They assessed the working conditions in various facilities, including the Building, by the Atomic Energy Commission. Do you remember that inspection?

: No, I am afraid I don't.

Behling: Do you keep in touch with any of the coworkers that you worked with during that timeframe assuming that they may still be alive?

: Well, the fellows that I worked with in the room where we , thorium, I guess it was, I'm sure have passed on by now. They were older than I was by quite a bit. But, no not from the end of it, I do not recall socializing with them.

Behling: So, right now, you do not have anyone that you stay in touch with that you worked with during that timeframe.



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Behling: Let me ask you this, and I guess I should have asked earlier, are you a claimant yourself?



: Am I what?

Behling: Are you a claimant? In other words, have you filed a claim for any health effects—cancer—that is associated with your work?

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Behling: I needed to ask that question, and I am **an end of** to hear that your answer is **a**. Well, unless you have something else to add, I thank you for your time; but let me give you my phone number, if you think of something that I should have asked or you failed to mention, let me give you my phone number. It is area code **and the end**.

# :

Behling: That is correct. And, my first name is Hans H-A-N-S and last name Behling, B as in boy E-H-L-I-N-G.

: B-E-H-L-I-N-G.

Behling: Yes. I think that you have given me the information that I am looking for. And, I will forward to you by mail a transcript of this interview so that you have assurance that I did not say anything that you did not state in this interview; and, if you have any additional comments or questions, do call me.



Behling: Well, I appreciate your cooperation, Mr.

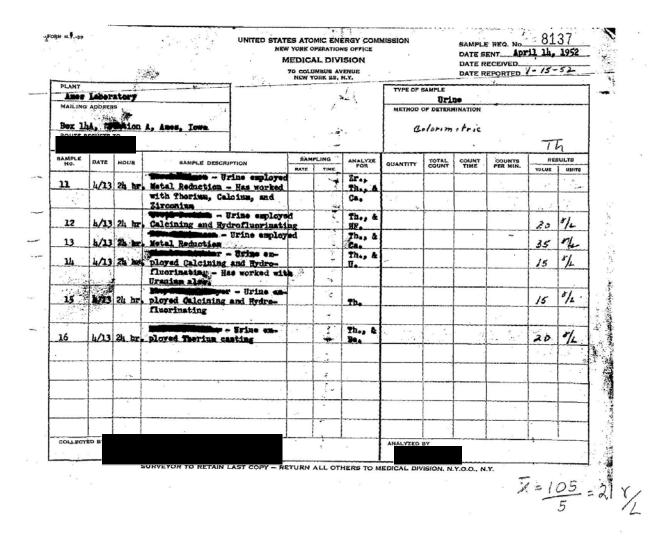
: Well, I am glad that I could help.

Behling: Thank you so much.

: Thank you.

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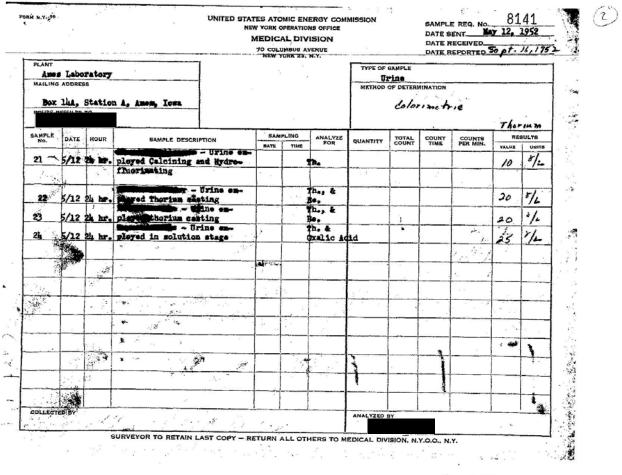
# **APPENDIX F: THORIUM BIOASSAY DATA FOR AMES WORKERS**



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# APPENDIX G: DISCUSSIONS RELATING TO THE 250-WORKDAY REQUIREMENT FOR SEC STATUS

# **WORKING DRAFT**

### THE RELEVANCE OF THE 250-WORKDAY REQUIRMENT TO POTENTIAL EXPOSURES ASSOCIATED WITH A SINGLE BLOWOUT

Contract No. 200-2004-03805 Task Order No. 5

Prepared by

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June 2007

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# **1.0 INTRODUCTION AND STATEMENT OF PURPOSE**

For Special Exposure Cohort (SEC) class eligibility, regulations defined in Title 42, Part 83, Section 83.13 of the *Code of Federal Regulations* (42 CFR Part § 83.13) currently specify a minimum aggregate of 250 workdays. An exception to this requirement is provided in §83.13(c)(3)(i), which involves exposure to "...discrete incidents likely to have involved exceptionally high levels of exposures, such as nuclear criticality incidents or events involving similarly high levels of exposures resulting from the failure of radiation protection controls." [Emphasis added.]

In two previous draft reports (*Review of the Ames Laboratory Special Exposure Cohort (SEC) Petition SEC-0038* issued in June 2006 and *An Assessment of Worker Eligibility Criteria* issued in January 2007), SC&A identified to the Advisory Board the relatively common radiological incidents of chemical explosions or "blowouts" at the Ames facility in context with the 250-workday requirement. In response to SC&A's concern, the Board appointed an ad hoc working group chaired by Dr. James Melius to further evaluate this issue. The working group requested SC&A to (1) review all available records/sources that would establish the frequency of such events, and (2) provide scoping calculations that would assess reasonable estimates of potential internal exposures associated with a single event. The enclosed draft report attempts to satisfy this request.

# 2.0 RELEVANT BACKGROUND INFORMATION

Data regarding the potential quantities of uranium and thorium that might have become volatilized and airborne as a result of blowouts during the reduction of UF<sub>4</sub>/ThF<sub>4</sub> to metal at Ames Laboratory can be found in a 1947 document entitled, *History of the Ames Project Under the Manhattan District to December 31, 1946.* This document was compiled by E.I. Fulmer, who served as Assistant Director to Dr. F.H. Spedding, Director of the Ames Project. Sections 1.1 and 1.4 of the report provide the following information pertaining to the reduction of UF<sub>4</sub> and ThF<sub>4</sub> to pure metal.

Section 1.1 of the report emphasizes that the principal objective of the Ames Project was the production of uranium metal, which originally was based on the chemical reduction of UF<sub>4</sub> by calcium metal in a refractory-lined steel bomb. In the first quarter of 1943, however, calcium was replaced by magnesium for the reduction, and by July 1943, uranium metal production reached 130,000 pounds per month. Production of uranium metal was performed exclusively in the remodeled one-story wooden building identified as the Physical Chemistry Annex I, known locally as Little Ankeny.

Uranium production ceased by January 1, 1945, after producing a total of about 2,000,000 pounds. Over the 2-year production period, reduction of UF<sub>4</sub> commonly involved steel pipes measuring 6 inches in diameter by 36 inches in length (however, larger pipes of up to 10 inches in diameter by 36 inches in length were also used). A successful reduction yielded a biscuit that was typically 42 pounds of uranium metal.

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In Section 1.4 of the report, Fulmer provided the following information regarding the production of thorium metal.

Soon after the large scale bomb reduction of uranium was in successful operation, similar approaches were made on the reduction of thorium. In August, 1943, attempts were made to reduce a number of thorium compounds in a bomb. The first attempts were unsuccessful due to the high melting point of thorium and the great stability of its compounds. Later, small amounts of thorium were produced by reducing ThF<sub>4</sub> with metallic calcium using iodine as a "booster". The yields were low and the metal was obtained in small pellets which were very difficult to recast into solid metal. In August, 1944, ZnCl<sub>2</sub> was tried as a "booster" and solid biscuits of thorium-zinc alloy were obtained in good yields. Within three months the conditions necessary for good yield had been well enough established to allow expansion of the process to the use of a reduction bomb 6 inches in diameter. . . . By June 1946, most of the details had been worked out successfully and the bomb reduction of thorium fluoride was ready for expansion to large scale production.

The process, as use in December, 1946, was a metallothermic reduction of thorium fluoride by metallic calcium. Zinc chloride was used to provide additional heat, to give a more fusible slag, and to form a low-melting allow of thorium which would collect in the form of a solid biscuit. The reduction was carried out in an iron bomb 7 inches in diameter and 45 inches long. This bomb was lined with a layer of dolomitic oxide compacted into place around a steel mandrel with a pneumatic jolter. The charge was placed in the bomb which was then closed and the reduction started by preheating in a gas-fired furnace. The bomb was allowed to cool after the reaction and the biscuit of thorium-zinc alloy removed and cleaned. This method produced a biscuit of about 39 pounds of thorium-zinc alloy with better than 96 per cent yield of thorium. . . . By December 31, 1946 over **4500 pounds of thorium** had been cast for shipment to other sites. [Emphasis added.]

On the **un**conservative assumption that the total of 4,500 pounds of pure thorium had been produced between June 1946 and December 1946 by means of 39 pounds thorium-zinc biscuits with ~96% yield of thorium, a **minimum** of 120 reductions may be assumed for the 7 month period (or about 4 reductions per week).

Production of thorium metal continued at Annex I (i.e., Little Ankeny) until 1949. Starting in 1947 and continuing through the end of 1953, production and casting of thorium metal was also conducted in the Metallurgy Building. Total production of thorium at both facilities amounted to 65 tons (or 130,000 pounds).

Furthermore, assuming that the size of the bomb retorts remained constant after December 1946, a total of around 3,500 ThF<sub>4</sub> bombs were reduced between June 1944 and December 1953, when thorium production ceased.

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# **3.0 FREQUENCY OF BLOWOUTS**

As previously noted in two separate reports issued to the Advisory Board, official monitoring records and radiological incident reports for Ames Laboratory are essentially non-existent. Since the issuance of these reports, further efforts have yielded no additional documentation that would quantify the number or frequency of blowouts, fires, and other radiological incidents.

However, previous SC&A reports included excerpts of official statements made by Dr. Frank Spedding, Director of Ames Laboratory (as well as by others), that uranium and thorium blowouts were commonplace events, as given in the following statements:

## Interview with Dr. Frank H. Spedding by Dorothy Kehlenbeck, July 5, 1961 (page 7):

- Interviewer: *It seems to me I remember there were a few explosions over in Little Ankeny, too, weren't there?* [Emphasis added.]
- Spedding: Yes, these caused quite a bit of concern. We were reducing the **uranium** with magnesium and the magnesium, of course, as you know, when ground into finely divided state is the active ingredient in flash powder. The chopped up magnesium was mixed with uranium salt and put in a steel container and then heated in a furnace. And occasionally the magnesium and the uranium would start alloying with the steel container, punching a hole in it and giving rise to an explosion which was intensely brilliant, just as if they'd shot off a ton of flash powder inside the building. And the building actually appeared to glow and expand, although actually it didn't, it was a brilliant light coming out of it and this caused a good deal of comment. Well, one day the explosions were really set off, we found later, by moisture. We were buying line [sic] from St. Louis, having it hard fired, and then having it set up here. But in the spring things became moist and the shipments got damp coming to Ames so we suddenly had a *flurry of explosions*. Fortunately in chemistry, my experience has been that whenever anyone expects that there might be an explosion, nobody gets hurt. It's only when you don't expect something that you get hurt so our men were usually down behind a wall when the explosion took place and it did no harm. Although I remember one night we had an explosion that blew the whole south end of the building out and being an old wooden building, when things quieted down we all went outside and shoved the wall back in again and went to work. Another difficulty I had with explosions was, during this same period we had employed a number of girls to do the business part of the project and when we had six of these in one day and set the building afire they all resigned in mass, although I did persuade about half of them to stay with us assuring them that the explosions were under control and that they would not damage them. [Emphasis added.]

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# Excerpts from F.H. Spedding Manuscript 2, *Humorous Story Concerning Explosions and Education*

*Mr.* Premo Chiotti was working with Dr. Wilhelm<sup>1</sup> and me [i.e., Dr. F. Spedding] on the reduction of thorium fluoride to thorium metal. Mr. Chiotti was adding a booster to the reaction in a room a few doors down the hall from my office. Suddenly there was a terrific explosion which blew out several of the windows in the front of the chemistry building. When I came out of my office to see what had happened, the corridor was filled with dust about six feet above the floor to the ceiling. I was relieved to see that Mr. Chiotti had not been injured, but he looked very dazed and was pacing up and down the corridor. As I passed him, I heard him muttering, "I must have misplaced that decimal point, I must have misplaced that decimal point."

These testimonials support the following assumptions:

- (1) Blowouts involving the reduction of UF<sub>4</sub> and ThF<sub>4</sub> were episodic events that were not uncommon during the entire period of facility operation.
- (2) From Spedding's statement that "...I remember one night we had an explosion that blew the whole south end of the building out and being an old wooden building, when things quieted down we all went outside and shoved the wall back in again and went to work," it is reasonable to conclude that this and other blowouts were not perceived as potential radiological threats, since no attempt was made to limit the time of exposure or to mitigate subsequent exposure by decontamination efforts, engineering controls/ building modification, or the use of respiratory protection.

# 4.0 ASSESSMENT OF AIRBORNE CONTAMINATION LEVELS ASSOCIATED WITH BLOWOUTS

Although no radiological incident/investigative reports were found in behalf of the Ames Laboratory, there was a blowout incident at Fernald on April 5, 1954, which provides data that are applicable to Ames.

Summarized below are descriptions and data contained in a report issued by J. H. Noyes et al. on April 5, 1954, entitled *Committee Investigative Report of Thorium Blender Incident – March 15, 1954*.

## From Section 2.0:

On March 15, 1954, personnel were attempting to blend a batch of thorium fluoride, calcium metal and zinc chloride preparatory to the reduction of the charge. After some delay in starting the blending operation and after unsuccessful attempts to properly seat a Gemco valve on the blender, a puff of

<sup>&</sup>lt;sup>1</sup> Dr. Harley Wilhelm was the Associate Director of the Ames Laboratory and Mr. Premo Chiotti was a scientist at the Ames Laboratory when it was under contract to the U.S. Atomic Energy Commission.

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dust appeared at the mouth of the blender; then a short flame; followed at 3:13 p.m. by a sheet of flame that extended horizontally from the blender a distance of about 45 feet over an arc segment of 38 feet. The duration of the flame is estimated at less than 10 seconds during which time tow persons... received serious burns which subsequently proved fatal, and two others received minor burns. Three additional persons received minor hand burns while assisting the injured. Physical damage to the equipment is estimated at about \$700. Approximately **50 lb. of thorium were unaccounted for following the incident.** The buildings was vacated and all activities stopped until an adequate investigation established such factors contributing to the accident as an examination of the building and equipment could offer. [Emphasis added.]

#### From Section 7.1:

The blending of a charge of thorium fluoride using calcium metal and zinc chloride is similar to the process utilized by Iowa State College, at Ames, Iowa... [Emphasis added.]

*The charge that was being blended when the incident occurred consisted of the following:* 

100	lb. thorium fluoride
10	lb. zinc chloride
35.9	lb. calcium metal

... Examination of the blender after the accident showed it to be nearly empty...

These data suggest that 50 lbs. (or about 50%) of the 100 lbs. thorium charge were likely to have been volatilized and dispersed by the high temperature that characterizes this exothermic chemical reduction of ThF<sub>4</sub>.

Applying these data to Ames, a 50% volatilization of a typical  $UF_4$  or  $ThF_4$  charge would have released about 21 lbs. of U and about 19.5 lbs. of Th into the air/environs at the Ames' Annex I/ Little Ankeny facility.

### 5.0 ESTIMATES OF AIRBORNE CONTAMINATION LEVELS

Figure 1 provides a floor plan of Little Ankeny. The heavy dotted line identifies the original building. Within the heavy dotted line, the lower right-hand side identifies the "Reduction Area." Unfortunately, the floor-plan does not include a scale that would permit an assignment of physical dimensions of the reduction area and surrounding rooms. However, available photographs, reproduced herein as Figures 2 and 3, depict the north and south view of the building and offer a crude measure of the building's physical dimensions. Based on relational dimensions of windows, doors, and automobiles (parked on the north side), an estimate of about 800 ft<sup>2</sup> is appropriate for the reduction area; for the entire building (inclusive of add-ons), an area

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of about 6,000  $\text{ft}^2$  is estimated. The low profile of the building further suggests a standard ceiling height of 8 to 9 feet.

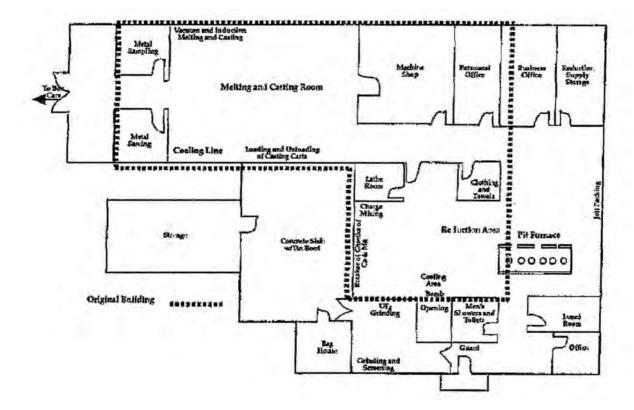


Figure 1. Floor Plan of Little Ankeny Production Facility for Uranium and Thorium



Figure 2. Physical Chemistry Annex (Little Ankeny) North View

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Figure 3. South View of Little Ankeny

### 6.0 ASSUMPTIONS USED FOR MODELING AN ACUTE INTAKE/DOSE FROM A BLOWOUT

Potential inhalation exposures from blowouts associated with the reduction of UF<sub>4</sub> or ThF<sub>4</sub> at Ames Laboratory are affected by a host of parameters. While some parameters can be defined empirically from historical data, others require reasonable but claimant-favorable assumptions. For estimating potential workers' doses, the following model is proposed.

- (1) Assume one blowout per month for the entire period of facility operations.
- (2) For either U<sub>nat</sub> or Thorium, a typical charge of 40 lbs. is assumed. This value is based on historical data for Ames.
- (3) For a blowout, 50% of the charge is assumed to have been reduced to metal and 50% is assumed to remain in the fluoride form.
- (4) Due to the explosive nature of a blowout (i.e., blown-out windows, doors, etc.), it is assumed that as much as 75% of the charge is released to the outside, with only 25% remaining indoors.
- (5) For the metallic form, solubility Type S is assumed; for the fluoride form, solubility Type M is assumed.
- (6) Based on the documented radiological incident data at Fernald, essentially all of the charge is assumed to escape from the bomb retort.

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- (7) For simplicity, about 10 lbs. (or 4,540 g) of natural uranium or Th-232 (in equilibrium with Th-228) are suspended uniformly in air at time zero throughout Annex I building having an interior volume of about 50,000 ft<sup>3</sup> (or ~1,350 m<sup>3</sup>). This would yield an initial air concentration of 3.36 g/m<sup>3</sup> of U<sub>nat</sub> or Th-232/-228.
- (8) Due to the intense heat generated by the blowout, we will assume a 5-minute period for contaminants to condense and settle out. During this 5-minute period, the worker is assumed to have a breathing rate of 2 m<sup>3</sup>/hr with an air concentration of 3.36 g/m<sup>3</sup>, as discussed in bullet #7 above. (In behalf of a radiological incident at Pantex, NIOSH assumed a breathing rate of 3 m<sup>3</sup>/hr for a period of 5 minutes (see ORAUT-TKBS-0013-5).
- (9) In the absence of any cleanup effort, the uniform deposition of 4,540 g of  $U_{nat}$  or Th-232/-228 is assumed to have deposited onto 6,000 ft<sup>2</sup> (or 540 m<sup>2</sup>), yielding a surface concentration of 8.4 g/m<sup>2</sup>.
- (10) Due to operation of machinery, human activity, building ventilation, and continued production of uranium and thorium metal, resuspension of contamination from floors and machinery would continue to create airborne levels for the next 30 days (or the next blowout).

For the intermittent period of 30 days between successive blowouts, the following empirical data may be used to estimate the air concentration and inhalation of thorium that is likely to have reached a **steady-state** contamination level resulting from successive blowouts.

In a 1952 AEC survey/audit conducted at the Ames Laboratory, the working environment of 22 production workers was evaluated. A breakdown of the daily weighted **thorium** exposure of the production personnel were as follows:

Average Weighted Daily Exposure:	$530 \text{ dpm/m}^3$
Max Weighted Daily Exposure:	$3,100 \text{ dpm/m}^3$

For modeling the 30-day post-episodic exposure, SC&A will use the **un**conservative **average** value of 530 dpm alpha per m<sup>3</sup>, and further make the **un**conservative assumption that the thorium alpha activity was contributed equally between Th-232 and Th-228.

For the post-incident period of 30 days, a normal breathing rate of  $1.2 \text{ m}^3/\text{hr}$  is assumed for a 9-hour workday, yielding a total of 193 workhours.

Estimate of Thorium Intakes Per Blowout

• First 5 minutes =  $(3.36 \text{ g/m}^3)(2 \text{ m}^3/60 \text{ min.})(5 \text{ min.})$ = 0.56 g total thorium

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The intake of 0.56 g thorium is equal to the intake of 0.06  $\mu Ci$  of Th-232 and 0.06  $\mu Ci$  of Th-228.

• For balance of 30 days =  $(530 \text{ dpm/m}^3)(1.2 \text{ m}^3/\text{hr})(193 \text{ hr}/30 \text{ days})$ = 122,748 dpm/30 days

Assuming an equilibrium condition, an intake of 0.028  $\mu$ Ci is estimated for Th-232 and for Th-228.

Further, assuming that one-half of the thorium existed as metal (or oxide) with a solubility Type S and one-half existed as ThF<sub>4</sub> with Type M solubility, the following dose estimates are derived.

Table 1. Internal Dose Estimates for a Thorium Blowout<sup>\*</sup> (rem)

Radionuclide/		Amount		Bone S	Surface			Lu	ng	
Solut		Inhaled (µCi)	1 yr	5 yr	10 yr	30 yr	1 yr	5 yr	10 yr	30 yr
Th-232	Type S	4.4E-02	0.1	1.3	3.6	13.7	2.6	5.1	7.3	11.4
Th-228	Type S	4.4E-02	0.3	1.4	2.0	2.0	29.3	34.2	34.2	34.2
Th-232	Type M	4.4E-02	4.6	26.0	53.7	153	1.8	2.0	2.0	2.3
Th-228	Type M	4.4E-02	12.2	39.0	43.9	45.8	19.5	21.2	21.2	21.2
Total			12.7	67.7	103	214.5	53.2	62.5	64.7	69.1

\* Dose estimates include inhalation exposures resulting form the first 5 minutes of a blowout and from resuspension of contaminants for a 30-day work-period. Not included are exposures to kidneys, liver, and other organs.

Estimates of Uranium Intakes Per Blowout

٠	For the first 5 minutes	$= (3.36 \text{ g/m}^3)(2 \text{ m}^3/60 \text{ min.})(5 \text{ min.})$
		= 0.56 g Natural Uranium

The intake of 0.56 g U<sub>nat</sub> is equal to the following activities:

 $U-238 = 0.186 \ \mu Ci$  $U-235 = 0.009 \ \mu Ci$  $U-234 = 0.186 \ \mu Ci$ 

• For the 30-day post-incident period, the uranium air concentration will **un**conservatively be scaled to the thorium data. This assumes a time-weighted average air concentration of 1.2 mg/m<sup>3</sup>. For a breathing rate of 1.2 m<sup>3</sup>/hr and 193 work-hours, an intake of 0.278 g is estimated for the 30-day post-incident period. The intake of 0.278 g of U<sub>nat</sub> equals the following activities:

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 $\begin{array}{l} U\text{-}238 \ = \ 0.093 \ \mu\text{Ci} \\ U\text{-}235 \ = \ 0.0044 \ \mu\text{Ci} \\ U\text{-}234 \ = \ 0.093 \ \mu\text{Ci} \end{array}$ 

Table 2. Internal Dose Estimates for a Uranium Blowout* (rem)	Table 2.	<b>Internal Dose</b>	<b>Estimates</b>	for a	Uranium	<b>Blowout</b> *	(rem
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Radio	Radionuclide/			Bone S	burface			Lu	ing	
Solubility Inhaled (µCi)		Inhaled (µCi)	1 yr	5 yr	10 yr	30 yr	1 yr	5 yr	10 yr	30 yr
U-238	Type S	0.186	0.012	0.043	0.076	0.14	12.4	17.9	19.9	22.7
U-235	Type S	0.009	0.001	0.002	0.004	0.01	0.67	0.94	1.06	1.17
U-234	Type S	0.186	0.014	0.050	0.080	0.15	15.8	21.9	24.7	27.5
U-238	Type M	0.186	0.360	0.760	1.030	1.44	8.26	8.94	8.94	8.94
U-235	Type M	0.009	0.020	0.040	0.050	0.07	0.47	0.47	0.47	0.47
U-234	Type M	0.186	0.400	0.820	1.100	1.65	11.0	11.0	11.0	11.0
Total			0.81	1.71	2.34	3.43	50.94	61.15	66.07	71.78

\* Dose estimates include inhalation exposures resulting form the first five minutes of a blowout and from resuspension of contaminants for a 30-day work-period. Not included are exposures to kidneys, liver, and other organs.

# 7.0 SUMMARY CONCLUSIONS

The Ames Laboratory operated on the Iowa State University campus between 1942 and 1953. In the beginning, the Ames Project was primarily a metallurgical research laboratory that investigated the chemical properties of various heavy metals that were of interest to the Manhattan Project. A major focus was the reduction of uranium and thorium compounds to pure elemental metal.

Within months, Ames researchers pioneered a chemical reduction process for converting  $UF_4$  to a highly purified uranium metal. Because this process was efficient and cost effective, the Ames Project was expanded to include the large-scale production of purified uranium and later thorium metal. Thus, research and production became an integrated operation in which production processes were an extension of research efforts, and ongoing research reflected failures and problems encountered at the production level.

Of the many difficulties affecting worker health and safety was the ability to safely execute the highly exothermic reduction of uranium and thorium in devices called bomb retorts. Chemical impurities and trace amounts of water resulted in blowouts or chemical explosions that frequently also resulted in fires.

Formal documentation or investigation of these incidents do not exist, but are acknowledged in personal interviews and memoirs by key personnel, including the Director of the Ames Project.

After 1953, the production of uranium and thorium metal by means of the Ames protocols was transferred to other facilities, including the Feed Materials Production Center (FMPC) in Fernald, Ohio. Data involving a thorium blowout at FMPC in 1954 was used to model internal exposures from inhaling airborne contaminants in the first few minutes following a blowout, and

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from resuspension of residual surface contamination for a period of 30 days (or 193 workhours). This time interval was selected on the assumption that the frequency of blowouts may have occurred on a monthly bases.

Exposure to the lungs and bone surface from a single incident suggests substantial doses that increase with time, as shown in Table 1 and Table 2 above. The selection of 5-, 10-, and 30-year periods corresponds to critical time intervals between exposure and the induction period for cancer development.

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## APPENDIX H: MEMORANDUM FROM DR. SKOOG TO DR. SPEDDING

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in An	PATE FED 1 1978; For the Atomic Inergy Consistent			
	ROJAL MOREN for the	September	er 12, 1952	
	Dr. F. H. Spedding, Director Ames Laboratory	21 Gai		
	Ames, Iowa			
	Subject: ENVIRONMENTAL HEALTH CON	DITIONS - AMES LABORATOR	Ϋ́Υ.	
	Dear Dr. Spedding:			
	I have before me a copy of a repor Donald M. Gardiner, Staff Biologis to A. Tammaro, Manager. The repor Conditions - Ames Laboratory" was within the domain of the Chicago ( sent to W. W. Lord, Area Manager, Sanitary Engineer, USAEC, Washingt Division of Biology and Medicine, Director, Division of Reactor Deve D. J. Casey, Director, Engineering	et, Technical Services Di et entitled "Environments for distribution essenti Operations Office and cop Ames Laboratory, A. E. G con, Dr. Walter D. Claus, USAEC, Washington, L. R. Copment, USAEC, Washingt	vision al Health ally Dies were Horman, Hafstad,	
	Said report, which was recently br out of line with certain facts tha time now to put the record straigh	t it is necessary that I		
	The lack of proper sequence and cl with misunderstanding on the part the picture and falsely indicate a Laboratory Health Physics Group. to Mr. Gardiner's blunt statement program at Ames is lacking in dire	of Mr. Gardiner, tend to laxity on the part of t Strong objection must be that the Health and Safe	muddle he Ames rade	
	In the Tirst two paragraphs of the to analyze, and to generally evalu thorium handling. This brief trea certain quantitative aspects, is s assumed as a working basis for all thorium handling and no implicatio regard to the Ames Laboratory.	ate, the hazards connect tment, although question ufficiently general to b AEC laboratories concer	ed with able in e ned with	
	A report furnished by Dr. George H Biology and Medicine, USAEC, Washi paragraph under <u>Resume</u> , was receiv	ngton, referred to in th	e first	
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A copy of this report is attached. Please note that the last paragraph states "Other details of the medical program will be covered in my general report of our visit to the Ames Laboratory." These details referred to have not yet been received. Neither has a report from Mr. Eisenbud ever been received here.

All recommendations referred to in the report of Dr. Hardie were put into effect at the Ames Laboratory by January 1, 1952. This report has only one reference (Paragraph 5) to Radio Physics and Industrial Health Programs.

The specific recommendations referred to as made and discussed with Dr. Spedding and me were covered in the Hardie report with the exception of two points. One was a request for an Environmental Health Survey and the other the hiring of a secretary for the Medical Department. The request for the Environmental Health Survey made by W. W. Lord on January 18, 1952, was the result of the request for this survey made by Dr. Spedding on January 14, 1952. The secretary was put to work on February 13, 1952, and this was about as early as interviewing, clearing and hiring machinery would allow.

In the first paragraph on page two of the Gardiner report the statement "that health monitoring activities for radiation had been neglected and that personnel exposure records had neither been made or kept" is grossly in error. Since the Health Physics Group did the monitoring and kept the records, and since no exposure approaching tolerance levels had been observed, the Medical Department did not have the records concerning these activities on file in its office. The Health Physics Group had the records of exposure readings on the production workers dating back to June, 1951, in its files. In light of the misunderstanding occasioned by inspection from outside sources, duplicate records are now on file in the Medical Department office. Mr. Gardiner probably based his opinion on the records in the Medical Department files at the time, although he did visit the Health Physics Group.

During the requested survey of March 19 to 21, a short conference was held by Mr. Harris and members of the Safety Group of the Ames Laboratory. The deficiencies discussed referred to three main items. (1) Housekeeping in general was fair but could be improved. (2) Ventilation in general was more than adequate but could be employed better on certain operations. (3) There was apparently some tracking of radioactive dust between the operating areas. Mr. Harris pointed out at this conference that remedial measures could be taken better upon receipt of the written report which would contain evaluations of the findings of the survey.

On a number of occasions attempts were made to obtain the results of the survey in order that the recommendations could be carried out. Two of these occasions were as follows: (1) At the

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Industrial Health Conference held at Cincinnati on April 24 and 25, 1952, when Mr. Klevan was asked concerning this report. He stated that the report was not yet finished and he offered no recommendations at that time. Mr. Klevan informed me that the reason for delay in completing his report was due to the fact that the members of the New York Operations Office Health and Safety Group were busy gathering and documenting data on the Nevada bomb test which was in progress. Work on the report necessarily had to be sidetracked due to the more pressing business concerned with the Nevada activities. (2) At the Industrial Physicians Conference of the AEC and its contractors held at Rochester, New York, on May 15 and 16, 1952, Dr. George Hardie, of the Division of Biology and Medicine, USAEC, Washington, had no information concerning the status of the Environmental Health Survey report. The reasons for Dr. Hardie having no information on the data of the survey have been explained above.

The survey was made, as stated, on March 19 through 21 but the first report entitled "Amos Research Occupational Exposure to Thorium and Beryllium" was dated July 14, 1952, and came to my attention on July 28, 1952. From the above information it is quite obvious that since the suggested corrective measures were not definitely known until July 20, 1952, some of these stops could not be taken until that date. It should be emphasized, however, that continuous corrective steps were being taken as the process evolved, including some of those recommended later in Mr. Cordiner's report. This accounts for the delay in placing certain corrective measures in effect during the period referred to by Mr. Cardiner as a period when only "lip service" was given to the problem. Also, we are still waiting for a report on the determination of radioactive material in the wrines of the men employed in thorium production. These samples were submitted, by the Ames Laboratory, to the Health and Safety Group of the New York Operations Office in April, 1952. It is readily understood by such a research organization as Ames Laboratory that there are difficulties in establishing methods of determination for detection of the products of the decay chain in urine. I realize that this takes time and I appreciate all the help given to the Ames Laboratory.

While on July 18 the thorium operations were interrupted due to a failure in the HF processes, on the 25th, not the 28th, Dr. Wilhelm closed down the oxalate precipitation step which effectively soon brought the entire thorium operation to a standstill. Let it be understood that this interruption of the oxalate precipitation process was based solely on the findings of our own Health Physics Group concerning mesothorium. This problem had neither been stated nor implied by any visiting Health or Safety survey group. Since we ourcelves desired to accomplish certain corrective neasures concerning surface contamination and dust problems, it was felt that the shutdown provided us with an opportunity to institute these measures. Therefore, the receiving of the report from the New York Operations Office was not the cause of churdown, as it in itself did not suggest these drastic actions in order to institute the suggested corrective measures. We are glad to receive the

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assistance of experts on these problems and to get their constructive recommendations, but regardless of how well we meet such recommendations the resumption of production will not take place until our own Health Physics Group is satisfied that we have a satisfactory solution to our health problems.

One could readily infer from Mr. Gardiner's report that the activity data connected with the sewage problem came from some outside Health Survey report. Let it be emphasized that all data in regard to radioactivity in the sewage included in Mr. Gardiner's report were very tentative and were supplied by the Ames Laboratory and not by visiting Health Survey groups.

In connection with Annex 1, this building has not been dormant. Annex 1 has been in continuous operational use for one process or another in certain areas since late in 1942 when the original uranium production plant was set up. We realize that it is not a desirable operational area and we have for a number of years continued to ask for an adequate replacement building where pilot plant operations could be concentrated and properly controlled. We operate our calcium grinding equipment and some semi-works operations for other processes in this building, but we know of no group of twenty employees that are relining furnaces, moving in new equipment and preparing for production in early September in this building.

The numerous suggestions outlined in the letter of Mr. Cardiner have been studied and are discussed below.

#### HEALTH PHYSICS

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Develop a thorough and rigorous health physics program that will

(a) "Control the flow of radioactive materials in the laboratory."

In regard to this suggestion, we would like it called to the attention of the authorities that all radioactive materials with the exception of thorium and uranium are under the direct control of our Health Physics Group. Artificial radioisotopes are received by this group and issued to proper personnel with instructions as to their use. The Health Physics Group also monitors both personnel and areas involved.

In connection with thorium and uranium processing the health physicists work in close conjunction with the production groups in all health matters. It should be called to the attention of all concerned that the operations in the production of thorium metal at the Ames Laboratory are not stable industrial processes. The area actually is a research pilot plant in which new developments are constantly being made. At present this pilot plant, when in operation, is called upon to DEMIIAL do the work of several times the originally-designed MIY EFORMATION capacity. One of the major problems in the mesothorium disposal aspect of our general problem lies in the ray

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CONFILENTIAT. SECURITY INFORMATION F. H. Spedding 5 material T.M.T. Originally the T.M.T. was shipped directly from the Lindsay Light and Chemical Corporation and was newly manufactured. Since the first scale-up of thorium production, this basic material has been shipped by the New York Office from its Hiddleson, N. J., storage area. Recently, after we noticed increased activity, we were informed that they were shipping material which might have been in storage up to 6 yrs. The radioactive elements, especially mesothorium, have built up by a factor of 5 to 10 in this older T.N.T. It is these problems that cause most of our headaches. All shipments of thorium metal from the Ames Laboratory have been made under the supervision of the Health Physics Group. During the past five to six years occasional surveys for radioactivity have been made throughout the process areas at points where high levels of activity might be suspected. (b) "Establish and enforce standards of lab operations to reduce personnel exposures to sub-tolerance levels." Concerning this subject it should be noted that no operation at the Ames Laboratory has subjected the personnel to above-tolerance levels of external radiation. Determination of exposure to possible internal radiation, that is dust sampling, has been delayed until the arrival of the necessary equipment for sampling and analysis. Dust samples have been taken at breathing-zone levels for operators of all processes in the thorium production during the past week. The data on the tests are not yet available. (c) "Adequately gather and document exposure records on all personnel." Dosimeter readings have been taken and recorded regularly, by the Health Physics Group of the Ames Laboratory, on the production workers since June 1951. Since it was the practice of the Health Physics Group to maintain their own records and notify me only in case any tolerance levels were approached, I was unaware of the data recorded in its files. At the present time all records of personnel exposure from June 1951 to date are being documented in the Medical Department records. (d) "Continuously monitor laboratory sewage system (drains traps - etc.) and all effluents." This suggestion is being studied by the Health Physics Group of the Ames Laboratory as to its feasibility of operation. CONFIDENTIAL

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 $\mu$  (d) SECURITY ELECTRON 6 F. H. Spedding INDUSTRIAL HEALTH "Institute an Industrial Mealth program that will materially reduce the level of airborne toxic contaminants by" (a) "Evaluating hazards in thorium toxicology." The literature has been surveyed on the topic of thorium toxicology and is found to be both conflicting and incom-

plete on the ideas expressed. It is a known fact that the Wolsbach Kantle Company has been working with thorium for over 30 years and no case of internal radiation damage has been uncovered in its employees. It is our impression at the Ames Laboratory that complete toxicological studies on therium have been carried out by the staff at the University of Rochester. Their recent report dated November 13, 1951, on the "Acute Toxicity on Inhaled Thorium Compounds" gave negative findings for mortality, urinary protein, blood N.P.N. and urea, plasma fibrinogen and serum proteins. Histological examinations revealed no pathological changes. I am sure that it is not the intention of this suggestion by Mr. Gardiner that we set up at the Ames Laboratory any toxicological group for the study of thorium to xicity, since this is a difficult and expensive field of research which requires the service of a highly trained specialist not found at Ames. Also to do this requires laboratory space, which we do not have, as well as equipment and supplies.

(b) "Establishing safety standards and practices for production personnel."

These have been established as far as is practical for this type of operation. With regard to one particular aspect, that of surface contamination, the situation seems to be as follows: the information on tolerance levels suggested in the literature concerning surface contamination is scanty. The only information we have is from the Argonne National Laboratory which states that "one to two thousand counts per minute per 100 square contineters is a safe level of operation. At 20,000 counts per minute per 100 square centineters, a general cleanup of the area is in order. At 50,000 counts per minute per 100 square centimeters, the area is to be evacuated." We have attempted to keep below two thousand counts per minute as a tolerance level in the production areas and find that this is practically impossible, especially on some of the processing equipment. A telephone communication between Dr. Skoog and Mr. W. B. Harris of the Health and Safety Group of the New York Operations Office revealed that a number of environmental Health surveys have been made recently in radium-dial painting establishments. These

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data have not yet been published. Mr. Harris feels that there is no correlation between the measured surface contamination and the air samples at breathing-zone level. Spot checks on several industries handling this type of material have indicated that it is not their practice to keep surface contamination on equipment down to these levels.

Thus it can be seen that it is difficult to establish rational standards concerning surface contamination. We do have a figure of 70 disintegrations per minute per cubic meter as a standard level for airborne dust. According to the Environmental Health Survey there were only a few areas in which above-tolerance values were observed. Revisions in operating procedure and ventilation will reduce the airborne dust in these locations to well below these somewhat arbitrary tolerance values.

(c) "Procure and distribute safety equipment such as protective clothing, respirators, etc."

These measures have been in effect in this Laboratory since 1942.

(d) "Establish 'housekeeping' standards,"

The steff of the Safety Group of Ames Laboratory has been unable to interpret exactly what is meant by this statement. It has always been the policy of the Ames Laboratory to maintain and to emphasize good housekeeping. Our Safety Group has been constantly warning the group leaders whenever bad housekeeping was found. With research activities where the conditions are constantly changing, it is inevitable that occasionally, in some areas, bad housekeeping will occur. Whenever such occasions arise they are corrected as soon as feasible.

#### VENTILATION

"Improve the individual room and building ventilation, using the aid of a consultant if necessary, by"

(a) "Reviewing the air supply and exhaust capacities of entire building system."

The general air supply as far as we can determine is better than the usual practices of nost chemical laboratories with which we are familiar. The special air supply and exhaust systems were designed by Leopold, of Philadelphia, and have been reviewed by the Amos Laboratory engineering division and other competent consultants on several occasions and found to be more than adequate, based on limits set by any recognized authority.

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