ENVIRONMENTAL IMPACTS OF THE RELEASE OF A TRANSURANIC ACTINIDE, AMERICIUM-241, FROM A CONTAMINATED FACILITY

J. Wang and P. Merry-Libby.
Environmental Research Division
Argonne National Laboratory
9700 S. Cass Avenue
Argonne, Illinois 60439

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ABSTRACT

Americium-241 is widely used as a radiation source, but it also has some potential risk if taken into the body because of its high dose conversion factor. Although the radiotoxicity of americium-241 is small compared to other transuranic actinides, its effects on the reproductive system and on development of the placenta are more damaging than the effects of plutonium-239. In Ohio, a gemologist's laboratory was contaminated with americium-241. Prior to decontamination of the laboratory, potential radiological impacts to the surrounding environment were assessed. A hypothetical fire accident resulting in a unit release (1 curie) was assumed. Potential radiological impacts were simulated using an atmospheric dispersion and dosimetry model with local meteorological data, population census data, and detailed information regarding the neighborhood. The results indicate that there could have been a significant impact on nearby residents from americium-241 via atmospheric dispersion if a major catastrophic release had occurred prior to decontamination and decommissioning of the laboratory.

INTRODUCTION

During the 1970s, a laboratory facility in a house near Newark, Ohio (Figure 1), was used for irradiation of diamonds and other gemstones to induce color changes for better market value. Americium-241 (powdered oxide form) was the principal source of radiation. Operations authorized under a license of the U.S. Nuclear Regulatory Commission (NRC) were discontinued in the early 1980s. A radiological survey of the facility was conducted in 1983 (1). Major americium contamination (fixed and removable) was located in the hood of the old glove box in the restricted laboratory area. Some minor contamination was found on the floor, on equipment surfaces of the restricted area, in the sink drain system, in the holding tank, and in the ventilation system filter (1). A previous estimate indicated that the amount of americium contamination in the laboratory was approximately 150 mCi (2). Later, after decontamination operations began, 22 to 24 curies of americium-241 were seized from the gemologist's facility, and the confiscated radioactive material was sent to a nearby U.S. Department of Energy (DOE) facility (3).

Prior to decontamination, the gemologist's house was occupied by tenants. The laboratory area where the americium was used was locked and designated as a restricted area. However, a fire or other accident might have caused a release of americium to the environment.

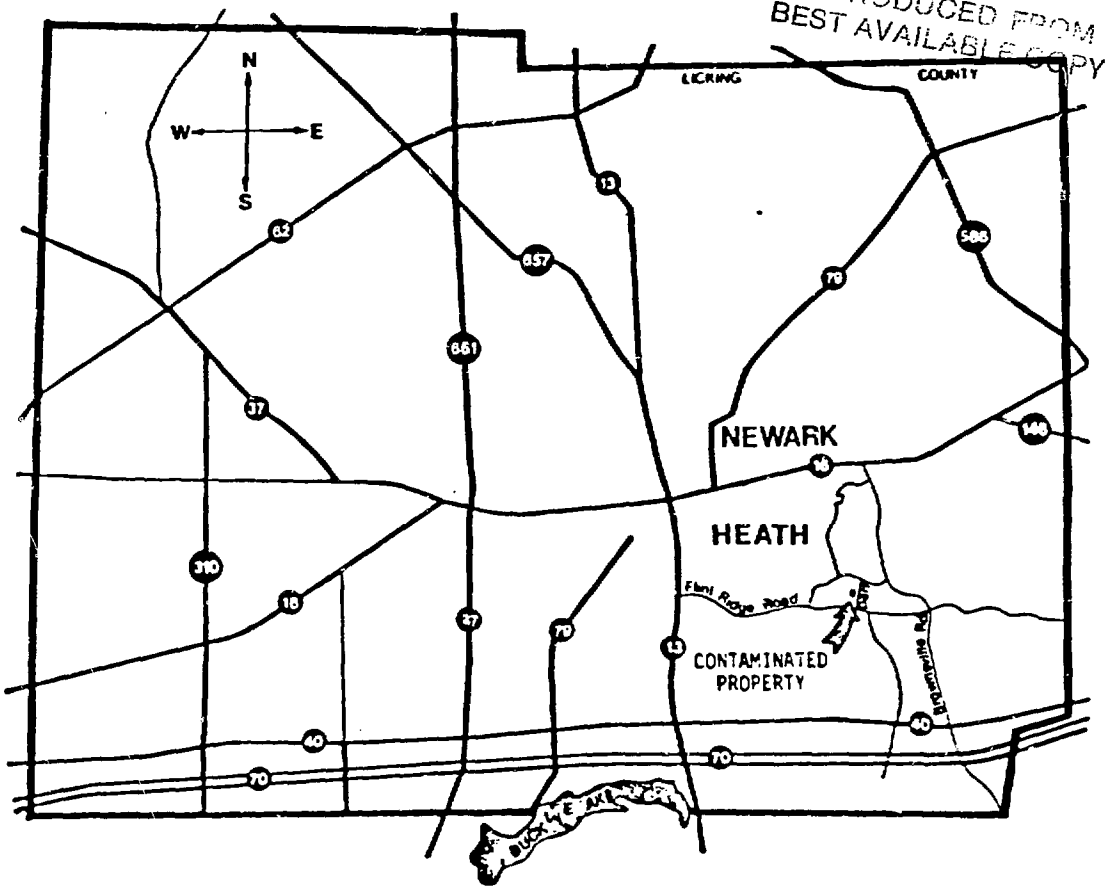

Figure 1. Map of Licking County, Ohio, Indicating the Location of the Americium-Contaminated Property. Source: Modified from Reference 1.

CHARACTERIZATION OF AMERICIUM-241

Americium exists in all oxidation states from II to VII, but the trivalent (III) state is most common. Most trivalent compounds can be converted to highly oxidative tetravalent americium compounds by ignition. The density of americium (III) oxide (hexagonal) is 11.75 g/cm^3 and that of americium (IV) dioxide (cubic) is 11.68 g/cm^3. The value of 11.7 g/cm^3 is used for the dose commitment evaluation in this study.

Americium-241 has a physical half-life of 458 years and an effective half-life in bone of approximately 140 years. The assumed effective half-lives in the whole body and liver are 100 and 40 years, respectively (4). Americium-241 transforms to neptunium-237 by emitting alpha particles of two distinct energies: 5.49 MeV (85%) and 5.44 MeV (13%). The major photons emitted by americium-241 are gamma rays of 60 keV (36%) and 26 keV (100%) and conversion L X-rays of neptunium-237$_m$ with energies centered at approximately 18 keV.

The major pathways of concern for exposure of humans to americium are inhalation, ingestion, and skin wound. The critical organs are liver, gonads,

red bone marrow, and bone surface (5). Americium-241 differs somewhat from other transuranium actinides in its metabolism and radiotoxicity; it has a smaller radiotoxicity and a shorter biological turnover time (faster biological clearance). However, the effects of americium-241 are more damaging than plutonium-239 with regard to the reproductive system and development of the placenta, especially in causing intra-uterine death of embryos and postnatal detrimental effects (6).

Americium oxide powder can be readily dispersed in the atmosphere because of its fine particle size (7-9). If no significant particle agglomeration occurs, the particle size can be approximated by a log-normal distribution with limited variance. In this analysis, a representative activity median aerodynamic diameter (AMAD) of 2.2 microns, with a geometric standard deviation of 2, is assumed (7,8).

POTENTIAL DOSES TO THE GENERAL POPULATION AND INDIVIDUALS AT NEARBY LOCATIONS IN THE EVENT OF A FIRE

Assumptions and Methodology

A severe fire in the laboratory area could cause the release of americium-241 particles to the atmosphere. The dispersion of americium would depend on the severity of the fire, the particle sizes, and the meteorological conditions at the time of the fire. Doses to the general public and to individuals at nearby locations would depend on the amount and pattern of the dispersed americium, the population distribution, and the locations and activities of the nearby individuals. The following assumptions are made to estimate the potential risk to humans in the event of a fire:

1. Release of americium-241 to the atmosphere during the fire is normalized to be 1 curie. The average particle density is assumed to be 11.7 g/cm^3 (see previous discussion).

2. Dispersion in the atmosphere is estimated based on the yearly average meteorological conditions at Columbus, Ohio, which is located about 50 km from the gemologist's laboratory.

3. The population distribution of 1.6 million people living within 80 km of the contaminated property is based on 1980 census data.

4. Some nearby individuals for whom radiological doses are calculated were identified by NRC based on onsite observations and a local tax map (Figure 2).

A computer program developed at Argonne National Laboratory [a modified version of the UDAD computer code (10)] is used to estimate dispersion of the americium and potential radiation doses to the general public and to individuals at nearby locations. Four potential pathways of americium-241 dispersion are evaluated: (1) inhalation of americium particles, (2) direct radiation from immersion in a cloud of americium particles, (3) direct radiation from americium deposited on the ground, and (4) ingestion of americium via the food chain. Doses to the general public are reported as the 100-year dose commitment, and doses to individuals are reported as the 50-year dose commitment. For simplicity, the term "dose" is used as an interchangable term for "dose commitment" in this analysis.

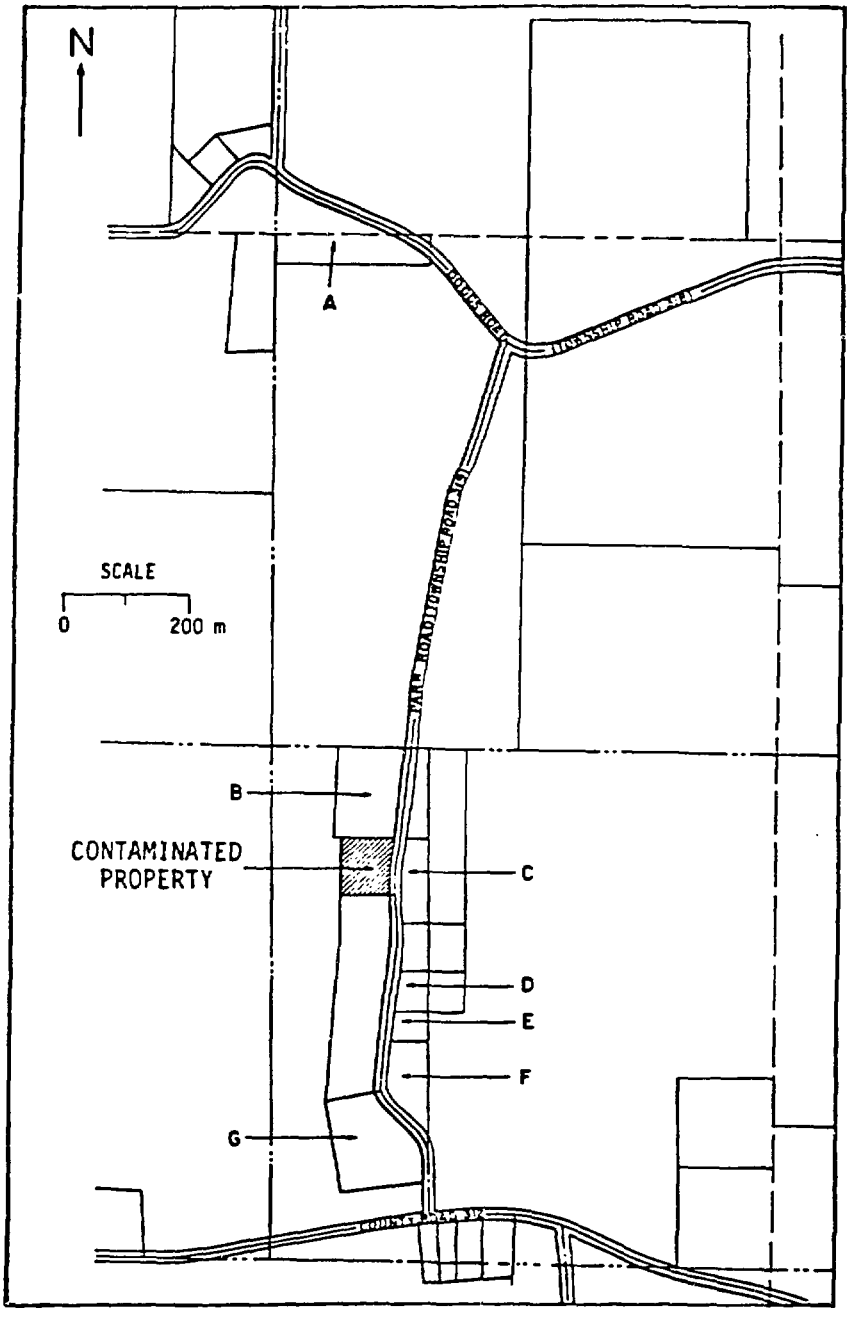

Figure 2. Location of Some Nearby Residents to the Contaminated Property for Whom Radiological Doses Are Calculated. Source: Tax Map, Franklin Township (Twp. 1; Rg. 11), June 1, 1970 (Revised November 4, 1983), and Onsite Observations, December 7, 1984.

Estimated Doses to the General Public

The inhalation and ingestion pathways would account for more than 99% of the potential doses to the general public (population doses) (Table 1). The highest doses would occur in the skeleton (bone surface), red marrow, and liver. The effective population dose would be 1.1×10^4 person-rem (110 person-Sv) per curie release of americium-241, which is approximately equal to 7% of the natural background radiation dose that the same 1.6 million people would receive in one year. However, distribution of the population dose would not be uniform throughout the area analyzed (within 80 km of the source). Most of the population dose would be concentrated near the source. For example, 50% of the whole-body population dose via the inhalation pathway is predicted to occur within 0.5 km of the gemologist's property.

The projected americium-241 concentrations in air (Figure 3) show a pattern of decreasing concentration with distance from the source. Due to the compartmental approach of the model, the spatial distributions of all other pathways are similar to the inhalation pathway (see Figure 3) but differ by a concentration factor. Also, the decreases are fairly uniform in all directions. This can be expected because the annual frequency of wind occurrence in all 16 directions is very uniform, with an average of 6.25% and a standard deviation of 2.43%. For brevity, the air concentrations are presented for only four directions (N, E, S, and W) in Figure 3. On the log-log plotting, all lines have approximately the same slope of -1.9, fitting closely to an inverse square law. This means that doubling the distance would decrease the americum-241 concentration in air by about 73%.

Estimated Doses to Individuals at Nearby Selected Locations

Potential doses per 1 curie release of americium-241 at seven selected nearby locations are given in Table 2; for comparison, the regulatory limits (11) are also given. The doses to the skeleton (bone surface) and red bone marrow are the highest among the selected organs. At all seven locations, individuals would receive a red marrow dose exceeding the NRC regulatory limit of 500 mrem/yr (5 mSv/yr). Except for location A, farthest from the source (about 1 km), all others would have skeletal doses exceeding the regulatory limit. The highest bone doses (34.6 rem [346 mSv] to the red marrow and 33.0 rem [330 mSv] to the skeleton) would be received at location C, just across from the road from the source. At most selected locations, the doses to other organs would also exceed the regulatory limits if there were 1 curie of americium released during the fire.

Discussion

The analysis presented herein is constrained by the assumption that there is 1 curie of americium-241 released from the contaminated laboratory during a fire. In fact, this source term is a product of two independent factors, the amount of americium-241 present at the laboratory and the fraction of that americium-241 released into the air during the fire. As discussed previously, the amount of americium-241 could be more than 20 curies. However, any variation in the two factors--amount of americium-241 present and fraction released during a fire--could directly affect the amount of pollutant available for atmospheric transport and would modify the assessment of radiological impacts.

The estimated doses are actually estimated "risks" of doses based on the release and dispersion of 1 curie of americium-241, assuming the "probability"

Table 1. Estimated 100-Year Dose Commitments to the General Public‡a

	Dose Commitment (organ-rem/curie)				
			External		
Organ	Inhalation	Ingestion	Ground	Cloud	Total
Whole body	1.01×10^{3}	3.63×10^{3}	4.61×10^{0}	9.07×10^{-6}	4.63×10^{3}
Liver	5.14×10^{3}	1.93×10^{4}	2.64×10^{0}	5.21×10^{-6}	2.44×10^{4}
Gonads	2.65×10^{2}	9.60×10^{2}	4.05×10^{0}	7.93×10^{-6}	1.23×10^{3}
Lung	1.90×10^{2}	1.47×10^{3}	3.25×10^{0}	6.35×10^{-6}	1.91×10^{3}
Red marrow	2.37×10^{4}	7.27×10^{3}	8.00×10^{0}	1.56×10^{-5}	3.10×10^{4}
Skeleton	2.49×10^{4}	9.07×10^{4}	8.60×10^{0}	1.71×10^{-5}	1.15×10^{5}

‡a Dose commitments to the population within 80 km of the contaminated property assuming a release of 1 curie (see text).

Table 2. Dose Commitments to Individuals at Nearby Locations‡a

Organ	Regulatory Limit‡b (mrem/yr)	Dose Commitment (mrem/Ci)‡c		
		A	B	C
Whole body	5×10^{2}	3.55×10^{2}	1.09×10^{4}	1.4×10^{4}
Liver	5×10^{2}	1.82×10^{3}	5.59×10^{4}	7.13×10^{4}
Gonads	5×10^{3}	9.33×10^{1}	2.87×10^{3}	3.67×10^{3}
Lung	5×10^{2}	5.31×10^{2}	2.06×10^{4}	2.64×10^{4}
Red marrow	5×10^{2}	6.80×10^{3}	2.58×10^{5}	3.30×10^{5}
Skeleton	1.5×10^{3}	8.80×10^{3}	2.70×10^{5}	3.46×10^{5}

	Dose Commitment (mrem/Ci)			
Organ	D	E	F	G
Whole body	3.71×10^{3}	2.89×10^{3}	1.85×10^{3}	1.27×10^{3}
Liver	1.89×10^{4}	1.47×10^{4}	9.80×10^{3}	6.53×10^{3}
Gonads	9.73×10^{2}	7.60×10^{2}	4.87×10^{2}	3.36×10^{2}
Lung	7.00×10^{3}	5.45×10^{3}	3.49×10^{3}	2.41×10^{3}
Red marrow	8.73×10^{4}	6.80×10^{4}	4.36×10^{4}	3.01×10^{4}
Skeleton	9.20×10^{4}	7.13×10^{4}	4.59×10^{4}	3.17×10^{4}

‡a See Figure 2.

‡b Based on Reference 11 (10 CFR 20) and Reference 12 (Table 4.15).

‡c Reported as 50-year dose commitments per curie release of americium-241.

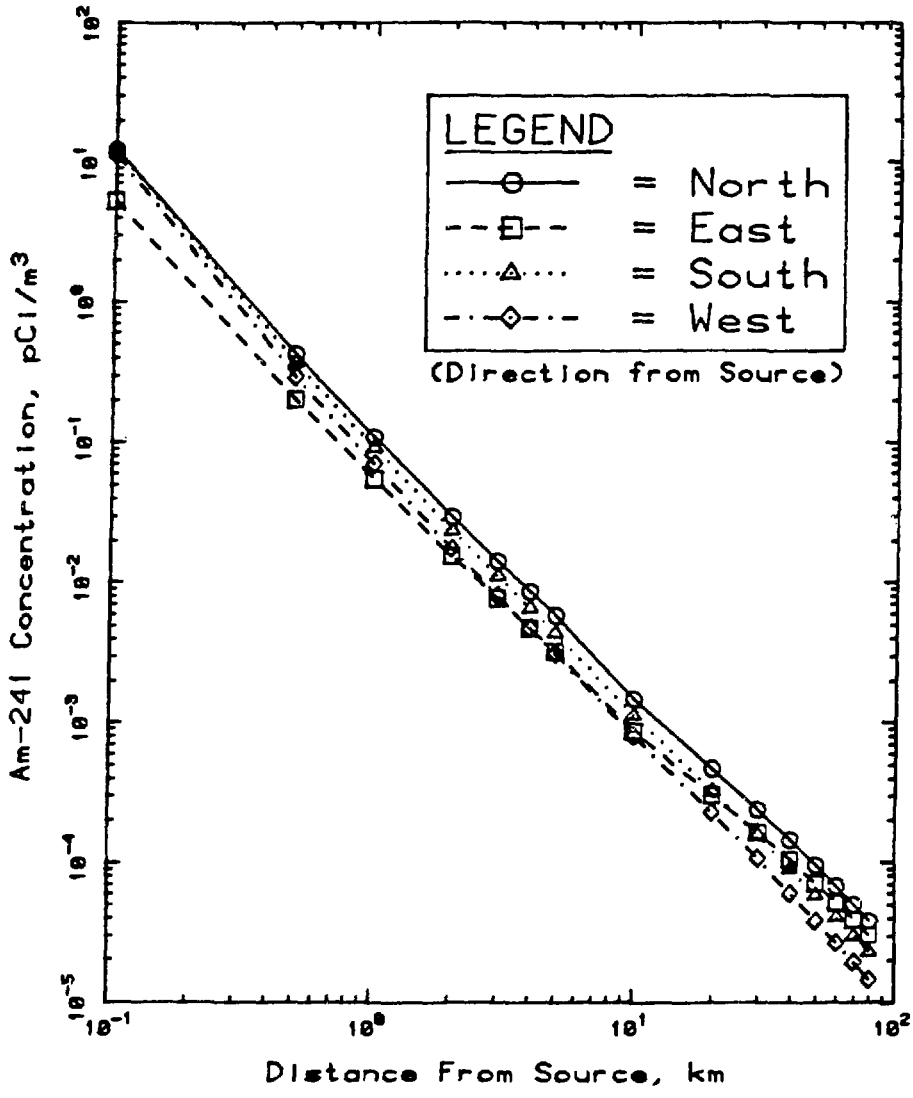

Figure 3. Americium-241 Concentration in Air from Release of One Curie.

of various weather conditions at Columbus, Ohio. Doses--especially individual doses--could be more or less, depending on weather conditions at the time of the fire, and actual weather conditions at the site may differ from those at Columbus. For example, there are a small hill and a small ditch behind the contaminated laboratory (on the west side) that may cause significant micro-meteorological variation. This local effect may alter the potential doses, especially for nearby individuals. However, site-specific meteorological data would be needed to make a more accurate dose assessment.

Firefighters or other persons who might be closer to the fire could possibly receive even higher doses than the nearby individuals. This would depend upon how much time such a person spent near the fire, whether or not the person was downwind of the fire, and whether or not the person used a protective breathing apparatus.

During a fire, americium (III) oxide would be oxidized to americium (IV) dioxide. Tetravalent americium is easily hydrolyzed to form complex ions (13) and experimental work has shown that americium dioxide is somewhat soluble (7). Airborne americium can be washed out substantially by rain. If it is assumed that (a) it rains during or immediately after the hypothetical fire, (b) most of the released americium is confined within 1 km of the contaminated property and is dissolved in rain water, and (c) there is approximately a 1.8-mm (0.07-in.) rainfall, the rainwater would have an average concentration that is at the maximum permissible concentration of 4 pCi/mL. [Note: A 1.8-mm rainfall is approximately 2% of the monthly average rainfall of 78 mm at Columbus, Ohio (14).]

Because the computer model used in this analysis is basically an air pollution model incorporating radiological metabolic models, the water pathways (groundwater and surface water) are not analyzed in this study. If the water pathway were significant, the ingestion dose would be higher. Further study is required to determine the significance of the water pathway.

ACKNOWLEDGMENT

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