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ENVIRONMENTAL IMPACT OF A TERATOGENIC ACTINIDE: A CASE STUDY OF AMERICIUM-241

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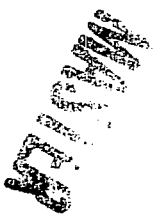

ABSTRACT

Americium-241 is widely used as a radiation source, but it also has some potential risk if taken into the body. 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. A previous report based on a worst-case scenario involving a hypothetical fire accident in a contaminated facility indicated that there could have been a significant impact on nearby residents from a unit release of americium-241 via atmospheric dispersion. However, because the facility is located in a rural region where most drinking water supplies are drawn from private wells, it is believed that deposition of americium-241 from the atmosphere might also have impacts via the groundwater pathway by infiltration of rainwater. In this analysis, a three-dimensional analytical mathematical model is used to assess several aspects of americium-241 contamination of groundwater, including radioactive transformation, advection, dispersion, and soil sorption. Simulation results indicate that no significant

radiological impacts would occur to the nearby residents via the groundwater pathway.

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 U.S. Nuclear Regulatory Commission (NRC) license were discontinued in the early 1980s. A radiological survey of the facility was conducted in 1983.[1] Major americium contamination (fixed and removable) was found 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]

Prior to decontamination of the facility, the gemologist's house was occupied by tenants. The laboratory area where the americium-241 was used

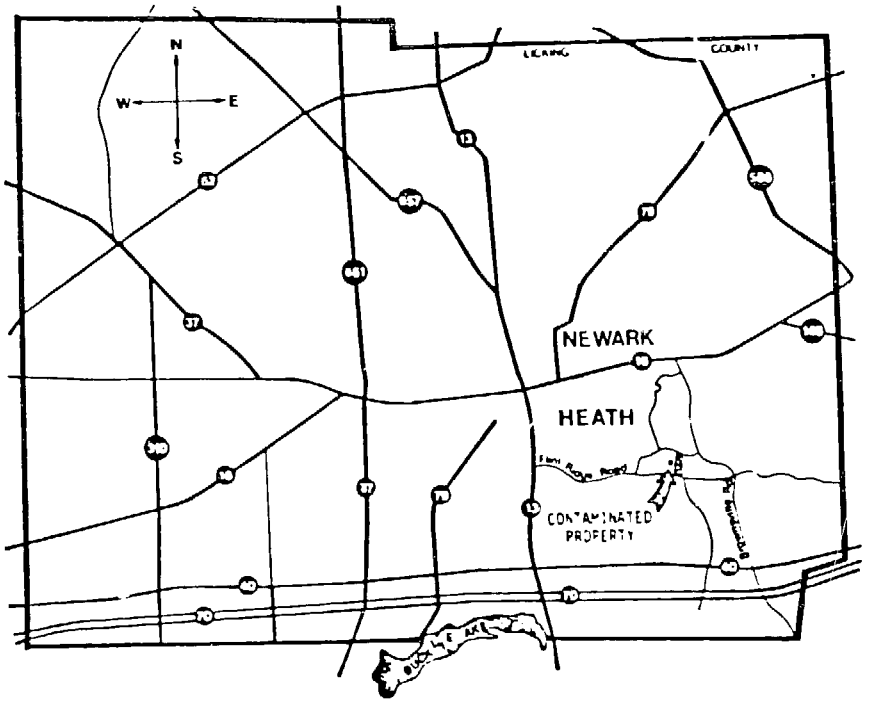

Figure 1. Map of Licking County, Ohio, Indicating the Location of the Americium-Contaminated Property. Source: Modified from Reference 1.

was locked and designated as a restricted area. However, a fire or other accident might have caused the release of americium to the environment. and this scenario is analyzed in this study.

CHARACTERIZATION OF AMERICIUM-241

Americium exists in all oxidation states from II to VII, but the trivalent (III) state is most common. Many trivalent compounds can be converted by ignition to highly oxidative tetravalent americium compounds that are soluble in water.[2]

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.[3] 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 68 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.[4] 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.[5]

Americium oxide powder can be readily dispersed in the atmosphere because of its fine particle size.[6-8] A previous study[9] indicated that there could have been significant radiological impacts to the surrounding environment via the air pathway under a hypothetical severe fire accident with unit (normalized) release of americium-241. However, because of the rural location of the contaminated facility and the reliance on groundwater for drinking water supplies, the deposition of americium-241 from the atmosphere might also have significant radiological impacts via the groundwater pathway by infiltration of rainwater. Therefore, this study focuses on the groundwater transport of americium-241 and its subsequent

radiological impacts to the surrounding general public.

EXISTING GEOHYDROLOGIC CONDITIONS

The ground surface in the vicinity of the site rises from north, east, and west at about 354 m (1161 ft) mean sea level (MSL) to about 323 m (1060 ft) MSL at the site. The surface of the site slopes south and southwest toward Claylick Creek at an elevation of about 290 m (950 ft) MSL. Soils in the site area are classified as Hanover-Londonville Association.[10] The Hanover soils are silty, well drained and formed in loam glacial till. Londonville soils are loamy, well drained, and formed in glacial till overlying sandstone bedrock on uplands. The site is underlain by unconsolidated glacial deposits of clay, silt, sand, and gravel with a water table of about 5 m (16 ft) below the ground surface. The glacial deposits are underlain by the Black Hand sandstone that is a massive coarse-grained quartz sandstone with water tables ranging from 5 to 20 m (16 to 66 ft) below ground. Both the glacial deposits and the Black Hand sandstone are major sources of groundwater in the region. Groundwater yields may range from 0.9 to 38 L/s (15 to 600 gpm).[11] Information regarding

movement of groundwater in the site area is not yet available. Groundwater in the unconsolidated glacial deposits and the Black Hand sandstone probably follows surface contours and hence probably flows from north to south.

MODEL DESCRIPTION AND INPUT DATA

In this study, a three-dimensional solute transport model, AT123D, developed at Oak Ridge National Laboratory[12] is adapted to calculate groundwater concentrations at particular times. Several locations are selected for assessing nearby individuals (see Figure 2). A list of input data for the groundwater migration analysis is given in Table 1. The release rate of americium-241 from the contaminated area (contaminated from airborne americium-241 due to fire, which will be discussed later) is estimated by the ion-exchange model.[13] The release rate of americium-241 is determined by radioactive transformation and leaching of americium-241. Because of the uncertainties relative to hydrogeological conditions at the site, selected values of input parameters in Table 1 tend to be conservative. The groundwater flow is assumed to be under steady-state and isothermal conditions. Groundwater movement is conservatively

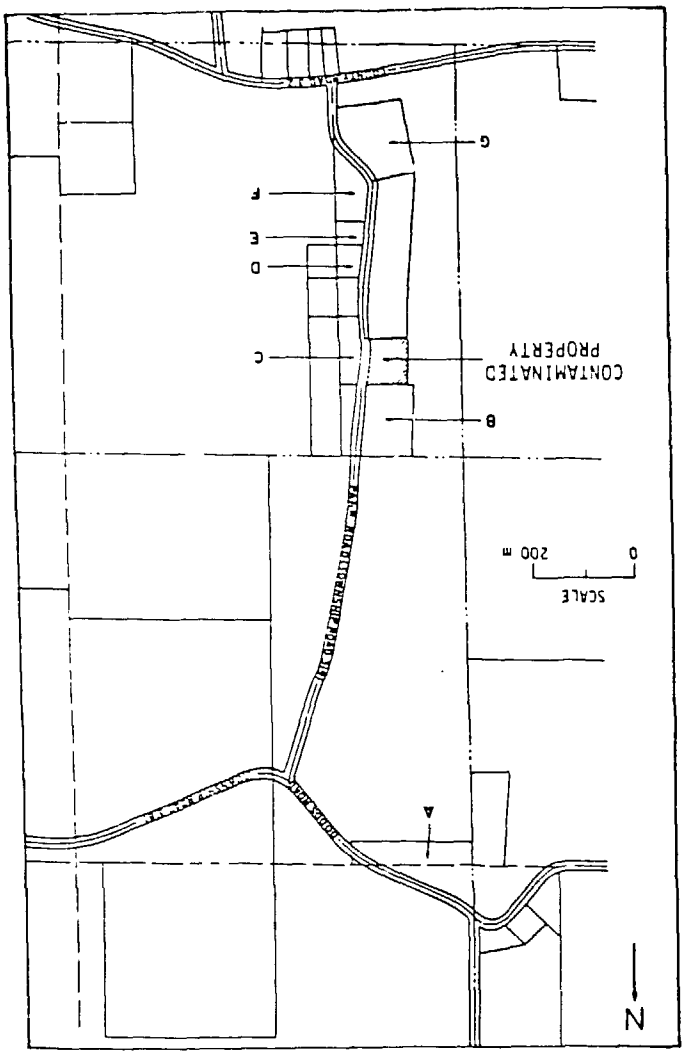

Figure 2. Location of Some Nearby Residents to the Contaminated Property for Whom Radiological Doses are Calculated. Source: Tax Map, Franklin Township (Twp. 1; Rg. 11), June 1, 1970 (Revised November 4, 1983), and Onsite Observations, December 7, 1984

Table 1. Parameters Used for the Calculation of Americium-241 Concentrations in Groundwater

Parameter	Unit	Raffinate Pits
Annual precipitation	m/yr	1.02
Evapotranspiration rate	m/yr	0.81
Infiltration rate	m/yr	0.2
Effective porosity	-	0.1
Bulk density of soil	kg/m^3	1,700
Hydraulic gradient	-	0.04
Hydraulic conductivity	m/s	10^{-4}
Longitudinal dispersivity	m	1.0
Transverse dispersivity	m	0.1
Vertical dispersivity	m	0.1
Distribution coefficient	m^3/kg	0.085
Radioactive transformation constant	1/yr	1.51×10^{-3}
Distance from lower boundary of waste field to water table	m	5
Distance from water table to bottom of resident intruder's well	m	1

assumed to be unidirectional in the north-south direction. The potential contamination of groundwater in the burial site under existing conditions for the accidental fire scenario are analyzed by determining the ratio of the americium-241 concentration in groundwater (C) to the americium-241 concentration in the contaminated soil (S). The concentration of americium-241 in groundwater can then be computed by multiplying this ratio by the radionuclide concentration in the contaminated soil.

A potential pathway for radiation exposure in the vicinity of the contaminated property is direct ingestion of contaminated groundwater. Americium-241 will be leached out of the contaminated soil by infiltration of rainfall and carried into the groundwater system. The source on the ground surface after accidental fire, which is deposited from airborne americium-241, is a major factor in determining the americium-241 concentrations in the groundwater system.

The spatial distribution of deposited americium on the ground from the air can be approximated in a power of -1.9 along the distance from the source.[9] To obtain a conservative analysis, all

the release (1 curie) of americium-241 is assumed to fall within 3 km (10,000 ft). Because the groundwater transport model could handle only simple geometry (e.g., rectangular) and uniform areal source, some further treatment of the source term (deposited americium on the ground surface) is needed. A "birthday-cake" source (Figure 3) is assumed, i.e., three different layers of uniform square source are superimposed (all concentric to the contaminated facility). The bottom layer is a 6 km × 6 km area. Because the farthest distance of the nearby individual selected in the radiological impact analysis is about 1 km, the middle layer is chosen to be a 2 km × 2 km square. Similarly, the top layer is chosen to be 0.2 km × 0.2 km because of the distance of the closest nearby individual. Because of the assumed isotropic areal source, equivalent circles are used to calculate the source concentration for the corresponding square areas which are the input for the groundwater model. Assuming the same areas, the radii of three equivalent circles are calculated to be 3.39, 1.13, and 0.113 km for the bottom, middle, and top areal sources, respectively. If the initial depth of americium in the surface is only 1 cm with a soil density of 1.7 g/cm^3, the corresponding equivalent concentrations in the three layers (assuming

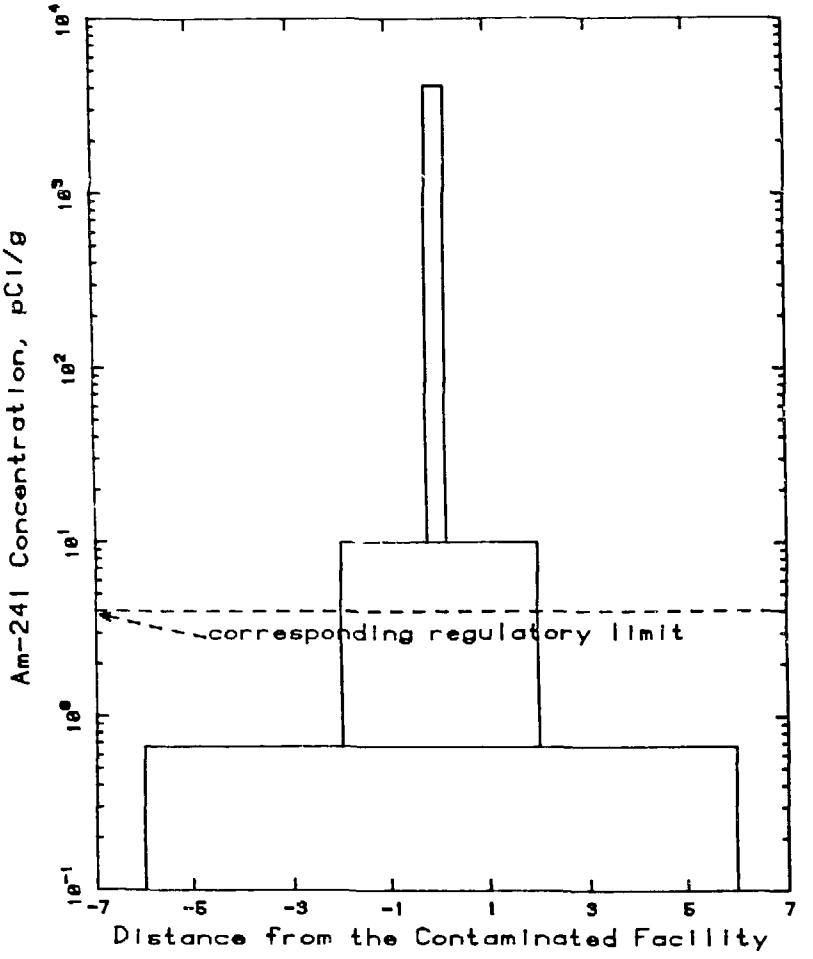

Figure 3. Area Source of Americium-241.

uniform distribution for each area) are 0.676, 10.1, and 4150 pCi/g, respectively. For comparison, the corresponding soil guideline is 4.0 pCi/g for americium-241 (100 dpm/100 cm^2 for the average of the top 15-cm surface).[14]

Another parameter that is important in determining the concentrations of americium-241 in groundwater is the distribution coefficient, Kd. This coefficient represents the ratio of the concentration of a radionuclide adsorbed on soil particles to the radionuclide concentration of the percolating water in the saturated waste material. The values of Kd for specific sites vary depending on soil properties, chemical content of the solution, pH of the water, and nature of the elements. The Kd value for the contaminated property site has not yet been determined through laboratory testing. However, the literature Kd values for americium-241 range from 0.085 to 40 m^3/kg.[15] In this paper, the Kd value of 0.085 m^3/kg is conservatively used for the analysis of americium-241 migration in the groundwater system.

RESULTS AND CONCLUSIONS

The time and spatial variations of americium-241 concentrations in the subsurface aquifer system (Table 2) beneath the study area are calculated using the parameters given in Table 1 under an accidental fire scenario with unit release (1 curie) of americium-241.

The model simulations indicate that the estimated distributions of the americium-241 concentrations are limited only in the areas beneath the bottom of the contaminated property. The maximum concentration locations would occur around the area within a radius of 60 m (197 ft). The maximum depth of americium-241 migration would be less than 6 m (18 ft), less than 1 m (3 ft) below the groundwater table. Results of the analysis of americium-241 migration along the groundwater flow direction (horizontally) also indicate that the americium-241 concentrations would reduce to zero at a distance of about 5 m (16 ft) from the boundary of the contaminated soil. The delay time for radionuclides to migrate from the bottom of the contaminated soil to the groundwater table would be about 3,600 years.

Table 2. Predicted Americium-241 Concentrations Within a Depth of One Meter‡a at Contaminated Areas of Nearby Residents‡b

Nearby Residents‡c	Concentrations at Various Times (pCi/L)						
	0.1 yr	1 yr	10 yr	50 yr	100 yr	150 yr	200 yr
A	0.01	0.01	0.01	0.01	0.009	0.008	0.0076
B	0.01	0.01	0.01	0.01	0.009	0.008	0.0076
C	4.0	4.0	3.94	3.71	3.38	0.54	0.0098
D	0.01	0.01	0.01	0.01	0.009	0.008	0.0076
E	0.01	0.01	0.01	0.01	0.009	0.008	0.0076
F	0.01	0.01	0.01	0.01	0.009	0.008	0.0076
G	0.01	0.01	0.01	0.01	0.009	0.008	0.0076

‡a The contaminant would remain in the infiltrating water in the unsaturated zone because the groundwater table is 5 meters deep.

‡b Values of hydrological parameters used for the calculations are given in Table 1. The maximum concentration locations would occur at the centers of the contaminated properties.

‡c See Figure 2 for locations of the nearby residents.

Because the delay time is relatively long compared to the half-life of americium-241, 458 years, the concentrations of americium-241 would be reduced to 0.04% solely by radioactive transformation effect even without considering other mechanisms, e.g., dispersion and advection. The contaminated region would remain in the unsaturated zone, i.e., the americium-241 would not reach the groundwater table (potential source of drinking water) within 3,600 years. Also, the concentration peak of americium-241 would occur long before reaching the groundwater table; therefore, the appearance of americium-241 in groundwater (after 3,600 years) would be monotonically decreasing with time.

The concentrations of americium-241 in the infiltrating water in the unsaturated zone over a period of 200 years at contaminated areas of nearby residents are presented in Table 2. The concentrations for all nearby residents would fall below 0.01 pCi/L at the end of 200 years. Using the ICRP dose conversion factors[4] and an assumed water intake of 2.5 L/d, it is estimated that 0.01 pCi/L of americium would result in a dose of only 0.02 mrem/yr. The americium-241 would not reach the groundwater for about 3,600 years, and the

concentratioon of americium-241 in ingested groundwater at 3,600 years would be at least 3 orders of magnitude less than that at 200 years because, from 200 to 3,600 years, the americium-241 would decrease by a factor of 1,700 due to radioactive transformation. The dose from ingested groundwater would be at least 8 orders of magnitude less than the dose from the atmospheric pathway,[13] or 7 orders of magnitude less than the current regulatory limit of 500 mrem/yr.

Therefore, based on the mathematical simulation for migration of radionuclides via groundwater transport, the potential health effects to individuals and the surrounding general public from the groundwater pathway due to radioactive release from the contaminated laboratory under such a fire scenario would be insignificant.

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