

*POTENTIAL OGALLALA AQUIFER  
IMPACTS OF A HYPOTHETICAL  
PLUTONIUM DISPERSAL ACCIDENT  
IN ZONE 4 OF THE PANTEX PLANT*

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H. J. TURIN  
I. R. TRIAY  
W. R. HANSEN  
W. J. WENZEL

Los Alamos  
Los Alamos National Laboratory  
Los Alamos, New Mexico 87545

LOS ALAMOS NATIONAL LABORATORY



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## 1 -- EXECUTIVE SUMMARY

Pantex Plant and DOE Albuquerque Field Office staffs are currently preparing a Safety Analysis Report and Environmental Assessment describing current weapon staging and proposed component interim storage operations in Zone 4 of the Pantex Plant, near Amarillo, Texas. The State of Texas has expressed concern over the potential consequences to the Ogallala Aquifer of an accidental plutonium release. Members of the Los Alamos National Laboratory Earth and Environmental Sciences Division have prepared this report describing the potential for aquifer contamination should plutonium be released to the environment within an 80-km radius of the Pantex Plant.

The following assumptions were used in preparing the groundwater impact analysis:

- Surface soils would be decontaminated to a maximum radiation level of  $0.2 \mu\text{Ci}/\text{m}^2$  following the hypothetical accident.
- Surface transport processes may increase soil concentrations ten-fold, to  $2.0 \mu\text{Ci}/\text{m}^2$ , before infiltration takes place.
- Recharge to the Ogallala Aquifer is focused at playa lake beds. Playa lake recharge rates are approximately 3 cm/year, ten times the High Plains average.
- The Ogallala Aquifer water table may be encountered as shallow as 50 feet beneath the land surface within the study area.
- The entire unsaturated zone exhibits a plutonium sorption coefficient of 100 mL/g, approximating the sorption of clean Ogallala sand.

With these conservative assumptions in place, two analyses were performed. A non-dispersive piston-flow model indicated that significant plutonium levels might be encountered in a 50-foot deep aquifer after approximately 76,000 years. A second, more realistic analysis incorporating dispersion showed that even with unrealistically low dispersivity values, peak plutonium concentrations in the 50-foot aquifer would never exceed the most restrictive drinking water dose limits. With more realistic dispersivity values, or deeper water tables more typical of the study area, peak plutonium concentrations in the aquifer would be orders of magnitude below dose limits. Neither analysis showed significant impacts to deeper aquifers.

Additional complicating factors have also been analyzed. These include colloidal plutonium transport, preferential flow, the effects of perched aquifers, opportunities for "short-circuit" flow through abandoned wells or other conduits, and the fate of daughter products. Although it is difficult to quantify these factors accurately, most are expected to have little if any negative impact on the Ogallala Aquifer. Colloidal transport is perhaps the most uncertain process in this category, but a field experiment at a nearby location suggests that colloidal transport will not enhance radionuclide transport enough to significantly affect groundwater quality in the Ogallala Aquifer.

The final conclusion of these analyses is that the hypothetical plutonium dispersal accident does not pose a significant threat to the Ogallala Aquifer.

## 2 -- STATEMENT OF PROBLEM

The hypothetical accident leading to dispersal of plutonium to the environment around Pantex is a high-temperature fire caused by a jet plane impact into a Zone 4 storage igloo containing nuclear weapons components, and subsequent ignition of jet fuel. These conditions may cause surface deposition of air-borne particulate plutonium across a large area downwind of the plant. The exact dimensions and location of this plume will depend on local meteorological conditions at the time of the fire, but much of the plutonium will likely fall on the Southern High Plains and Central High Plains of Texas. These are the recharge areas for the Ogallala (or High Plains) Aquifer, the primary water source for much of the Texas Panhandle. This aquifer is of immense economic significance, and potential threats to the resource should be carefully analyzed and, if necessary, minimized.

This report describes the potential impacts of the hypothetical accident on water quality in the Ogallala Aquifer. The results presented here are based on a thorough review of existing information on local and regional hydrogeology, new laboratory sorption studies on local geologic materials, and transport and dosimetry modeling. Although this is not a formal "worst-case" analysis, we have consistently made conservative assumptions to maximize the probability of identifying any real threats to the Ogallala Aquifer.

### 3 -- PLUTONIUM TRANSPORT TO THE OGALLALA AQUIFER

Any potential impacts to the Ogallala Aquifer would entail transport of contaminants from their deposition site at the ground surface down to the water table. This section of the report briefly outlines the processes that control this transport and describes the conceptual model that underlies the quantitative calculations discussed in the following sections.

#### Regional Setting

In the Panhandle section of Texas, the Ogallala Aquifer underlies the Southern and Central High Plains (Figure 3-1). These provinces are topographically-isolated plateaus bordered by the Pecos and Canadian River valleys to the west and the Caprock Escarpment to the east. The Southern High Plains are separated from the Central High Plains by the Canadian Breaks, an eroded landscape along the Canadian River.

The Ogallala Formation, primary host of the Ogallala Aquifer, is Tertiary in age and consists of eolian and alluvial material derived from the Southern Rocky Mountains to the west. Aquifer materials range from gravel to clay in grain size, and include some post-depositional caliche layers. The Ogallala Formation has been eroded and is no longer present along the Canadian and Pecos Rivers, or on the Rolling Plains, east of the Caprock Escarpment.

The Ogallala Formation is overlain by up to 27 m of Quaternary-age eolian silt and sand that comprise the Blackwater Draw Formation (Holliday, 1990). The major soil on the uplands in the study area is Pullman clay loam, derived from the underlying fine-textured eolian sediments (Unger and Pringle, 1981). The lower horizons of the Pullman soils are characterized by manganese-iron films on the surfaces (Allen et al., 1972). The High Plains surface is characterized by numerous playa lakes, small surface depressions found throughout the study area. The soil underlying the playa lakes is primarily Randall clay, sometimes associated with soils of the Estacado, Mansker, Lofton, and Lipan series (Unger and Pringle, 1981). Based on X-ray diffractometry, Allen et al. (1972) report that the clay mineralogy of the Randall soil consists mainly of poorly organized montmorillonite (associated with fine clay) with considerable quantities of illite, and smaller quantities of kaolinite (associated with coarse clay). Silt mineralogy is dominated by quartz and feldspars.

Groundwater in the Ogallala Aquifer occurs under unconfined or water-table conditions. Regional groundwater flow is from west to east, reflecting the regional topography. Recharge to the aquifer occurs through infiltration of rainfall and surface water through the Blackwater Draw Formation (Nativ and Smith, 1987). Recharge is probably focused at playa lakes, small surface depressions ubiquitous throughout the study area. The question of focused recharge will be discussed further below.

Natural discharge from the Ogallala Aquifer historically occurred through seeps and springs along the boundaries of the High Plains, particularly along the Eastern Caprock Escarpment and

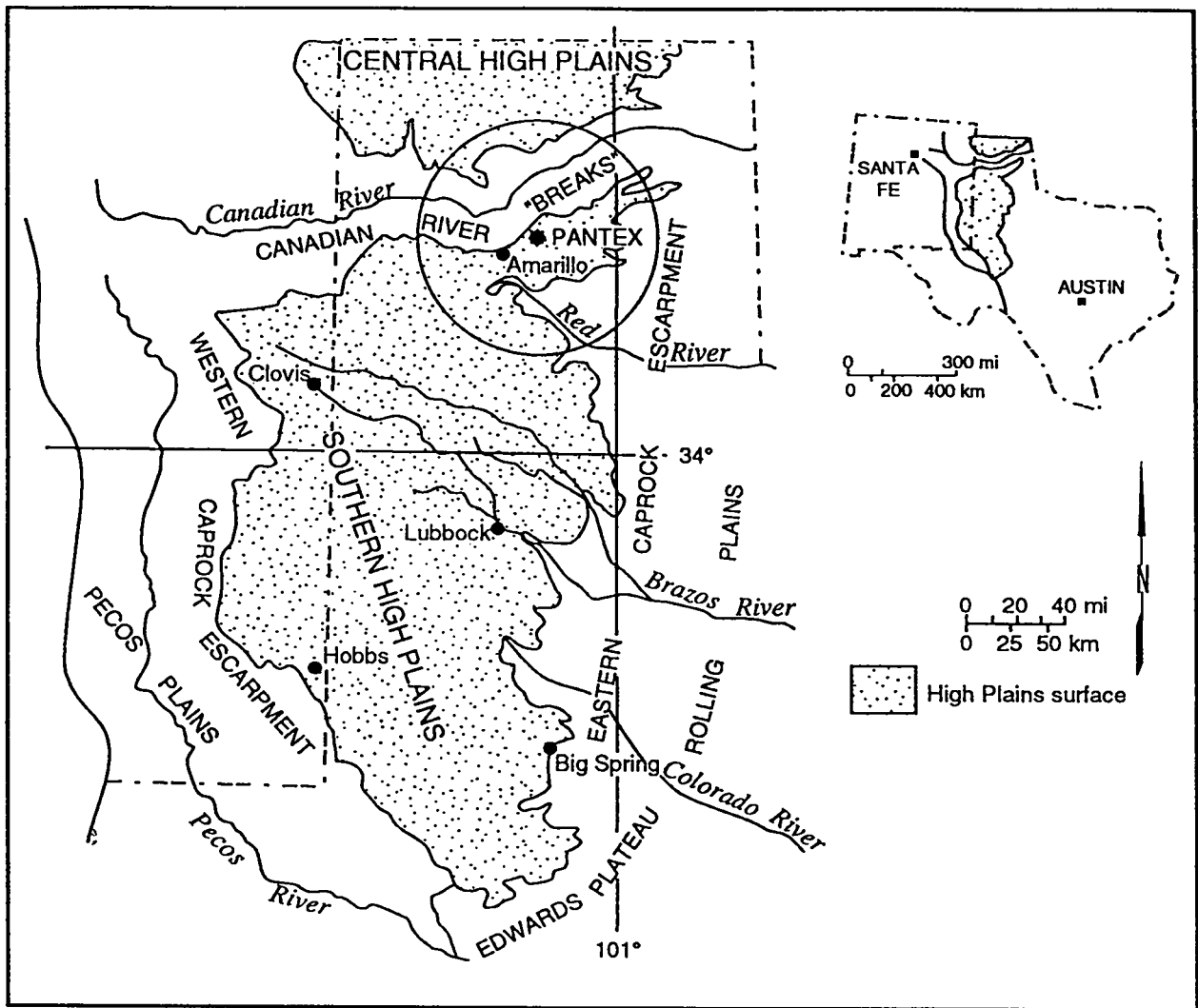


Figure 3-1. Central and Southern High Plains of Texas, showing the 80-km radius study area (adapted from Gustavson et al., 1990).

the Canadian Breaks. Some leakage into underlying formations is also possible. Since massive groundwater pumpage for agriculture began after World War II, many of these natural discharge points have stopped flowing, and today, groundwater pumpage is by far the most significant discharge route from the Ogallala Aquifer (Nativ and Smith, 1987). Current pumpage rates far exceed recharge, and the resulting groundwater mining is leading to declining water tables in most of the study area (Ashworth, 1991).

Water chemistry in the Ogallala aquifer and in the unsaturated zone beneath playas is generally quite good, typically a mixed-cation / bicarbonate water with 200-500 mg/L total dissolved solids and a pH of about 7.5 (Purtymun and Becker, 1982; Wood and Ostercamp, 1984).

## Recharge to the Ogallala Aquifer

Because of the economic importance of the Ogallala Aquifer and concern over water level declines, numerous researchers have studied the aquifer's recharge rate (Table 3-1). As Stone and McGurk (1985) pointed out, recharge can be considered on various spatial scales, ranging from local through areal to regional. At the local scale, recharge is likely to vary widely from spot to spot over a distance of meters, due to variations in local rainfall, surface slope, and soil properties. Over a larger area, these local variations will tend to average out, and a representative recharge rate may be applied over the entire extent of a particular landscape setting, such as playa lakes or uplands. Finally, a weighted average of different areal recharge rates may represent an overall regional recharge rate for a region the size of the Southern High Plains.

Table 3-1. Recharge rate estimates for the Southern High Plains.

Recharge Estimate	(cm/year)	Location	Source
< 0.5 in/yr	< 1.3	Regional, SHP	C.V. Theis, cited by Aronovici and Schneider, 1972
0.25 cm/yr	0.25	Regional, SHP	Wood and Petraitis, 1984
4 - 5 cm/yr	4 - 5	Playa annuli, SHP	Wood and Petraitis, 1984
0.11 in <sup>3</sup> /yr	0.28	Regional, northern SHP	Luckey, 1984
< 3/16 in/yr	< 0.48	Regional, TX High Plains	Knowles, 1984
0.75 mm/yr	0.075	Blackwater Draw Fm., NM	Stone and McGurk, 1985
4.36 mm/yr	0.436	Sand Dunes, NM	Stone and McGurk, 1985
12.22 mm/yr	1.222	Playas, NM	Stone and McGurk, 1985
1.5 mm/yr	0.15	Regional, NM	Stone and McGurk, 1985
0.3 cm/yr	0.3	Regional, SHP	Ostercamp and Wood, 1987
1.3 - 8 cm/yr	1.3 - 8	Maximum, SHP	Nativ and Riggio, 1990

SHP: Southern High Plains



Numerical model calibrations have yielded regional recharge estimates for the High Plains ranging from 0.25 - 0.3 cm/year (Luckey, 1984; Wood and Petraitis, 1984; Ostercamp and Wood, 1987). This relatively narrow range suggests that this number is fairly accurate.

Local recharge is more variable. After years of controversy, there appears to be a consensus building that the playa lakes of the High Plains focus recharge in a relatively small area, and that recharge in the uplands between playas is relatively insignificant or even nonexistent (Wood and Ostercamp, 1984; Wood and Petraitis, 1984; Stone and McGurk, 1985; Nativ and Smith, 1987). Local recharge rates in the playa basins must therefore significantly exceed the regional averages cited above. Local recharge estimates range from 1.2 cm/year for playas in eastern New Mexico (Stone and McGurk, 1985) to 4-5 cm/year for playa annuli (Wood and Petraitis, 1984). By analyzing groundwater tritium concentrations, Nativ and Riggio (1990) calculated a maximum estimated recharge rate at 1.3 - 8 cm/year, but pointed out that the higher rates are found in the southern part of the Southern High Plains, south of the present study area. For the purposes of this project, we shall assume that recharge rates beneath playas in the study area consisting of the northern part of the Southern High Plains and the southernmost part of the Central High Plains is approximately 3 cm/year.

### **Thickness of the Unsaturated Zone**

Water movement through the unsaturated zone between the ground surface and the water table in arid regions is often quite slow. The thickness of the unsaturated zone is therefore an important parameter controlling an aquifer's susceptibility to contamination. Although no recent contour maps showing depth-to-water for the study area were available, a comparison of a recent water-table elevation map (Ashworth, 1991) and USGS topographic maps suggest that typical depths to the water table in the 80-km radius study area are on the order of 200 feet. In the immediate vicinity of the Pantex Plant, the Ogallala water table is much deeper, 400 feet down, primarily due to a zone of depression caused by the City of Amarillo well field. In parts of Randall and Swisher counties, south of Pantex, water tables are shallower, in some cases as shallow as 50 feet.

Hydrogeologic studies in the immediate vicinity of the Pantex Plant have revealed a local perched aquifer above the main Ogallala Aquifer (Purtymun and Becker, 1982; Texas Bureau of Economic Geology, 1992). The lateral extent and continuity of this secondary aquifer is not yet known. Possible effects of the perched aquifer on the potential for contamination of the Ogallala Aquifer will be discussed in Section 5.

### **Definition of the Contaminant Source Term**

An important step in this impact analysis is a determination of the quantity and location of plutonium dispersed by the hypothetical accident that could impact the Ogallala Aquifer. Only materials that fall on the recharge area of the aquifer have that potential. The recharge area for

the Ogallala Aquifer is the relatively flat High Plains surface shown in Figure 3-1. The Ogallala Formation has been completely eroded away along the Canadian River Valley and on the Rolling Plains east of the Caprock Escarpment, and the Ogallala outcrop areas along the margins of the High Plains are groundwater discharge areas.

In the event of the hypothetical accident, the overall size and specific location of the affected area will depend on local meteorological conditions. Predominant wind directions in the Amarillo area are to the north-northeast (Dewart et al., 1982), but for this project, we have conservatively assumed that the contamination plume may extend in all directions for a distance of up to 80 km from the Pantex Plant. Contamination that may pose a risk to the Ogallala Aquifer will therefore be assumed to be limited to material falling onto the High Plains surface within a 80-km radius of Pantex.

If the hypothetical accident occurs, the affected area will be decontaminated to a maximum radioactivity level of  $0.2 \mu\text{Ci}/\text{m}^2$ . As mentioned earlier, recharge to the Ogallala Aquifer is focused at playa lakes scattered across the High Plains Surface. Precipitation runoff collects in these playa lakes, and may be expected to carry particulate and sorbed plutonium as well. Wind transport may also redistribute and locally concentrate plutonium at the playa lakes. To account for these surface processes, we have assumed that a ten-fold concentration of plutonium may occur between decontamination and infiltration, resulting in a soil contamination level of  $2.0 \mu\text{Ci}/\text{m}^2$  at the Ogallala recharge areas in the playa lakes. This is a very conservative assumption -- a study of  $^{137}\text{Cs}$  levels in soil affected by fallout from the Trinity Test showed that after 32 years, surface processes had locally concentrated contaminants by only a factor of 1.5 - 2 (Hansen and Rodgers, 1985).

### Radionuclide Geochemistry and Sorption Properties

The material dispersed to the environment by the hypothetical accident would be weapons-grade plutonium, a mixture of plutonium isotopes with a trace of americium (Table 3-2).  $^{239}\text{Pu}$  dominates the mixture on a mass basis, but because of its greater specific activity,  $^{241}\text{Pu}$  is the primary activity source. The decay chains and daughter product half-lives for the various nuclides are shown in Figure 3-2. The potential elements of concern are plutonium, americium, neptunium, and uranium.

#### Plutonium Solution Chemistry and Sorption

The plutonium solubility reported by Nitsche (1990) in an oxidizing groundwater with a high bicarbonate content at  $25^\circ \text{C}$  and  $\text{pH } 7 - 8.5$  is approximately  $10^{-7} \text{ M}$ ; the plutonium oxidation states are:  $\text{Pu(V)}$  ( $\sim 70\%$ ),  $\text{Pu(VI)}$  ( $\sim 20\%$ ) and small amounts of  $\text{Pu(IV)}$  and  $\text{Pu}$  polymer. The main species at  $\text{pH } 8.5$  are  $\text{Pu(V)}$  and  $\text{Pu(VI)}$  carbonate complexes and  $\text{Pu(IV)}$  hydrolysis products; at  $\text{pH } 7$  the main species are  $\text{PuO}_2\text{CO}_3$  and uncomplexed  $\text{PuO}_2^+$ . A review of the sorption literature presented by Meijer (1992) indicates that plutonium has a high affinity for quartz and feldspar. Triay et al. (1992) reported plutonium distribution sorption coefficients

Table 3-2. Isotopic composition of weapons grade plutonium (Wenzel and Gallegos, 1982).

Nuclide	Weight % in Weapons Grade Plutonium Mixture	Activity in Mixture (Ci/g mixture)
$^{238}\text{Pu}$	0.05	0.0087
$^{239}\text{Pu}$	93.6	0.0575
$^{240}\text{Pu}$	6.0	0.0136
$^{241}\text{Pu}$	0.4	0.448
$^{242}\text{Pu}$	0.5	$1.95 \times 10^{-6}$
$^{241}\text{Am}$	~0.02	$6.48 \times 10^{-4}$

( $K_D$ ) for clays in an oxidizing groundwater at a pH of approximately 8.5: the  $K_D$  values for smectites, illite, and kaolinite are in the range of  $6 \times 10^2 - 1 \times 10^4 \text{ mL/g}$ . High affinities of actinides for oxide minerals are reported by Meijer (1992) and Triay et al. (1991c). The sorption mechanism for actinides onto oxide minerals seems to be correlated with hydrolysis constants for these actinides in solution (Meijer, 1992).

Plutonium transport through the unsaturated zone is the major risk under evaluation in this report, and is primarily controlled by the degree of plutonium sorption onto local soils and aquifer materials. Because of the importance of this sorption process, a series of laboratory

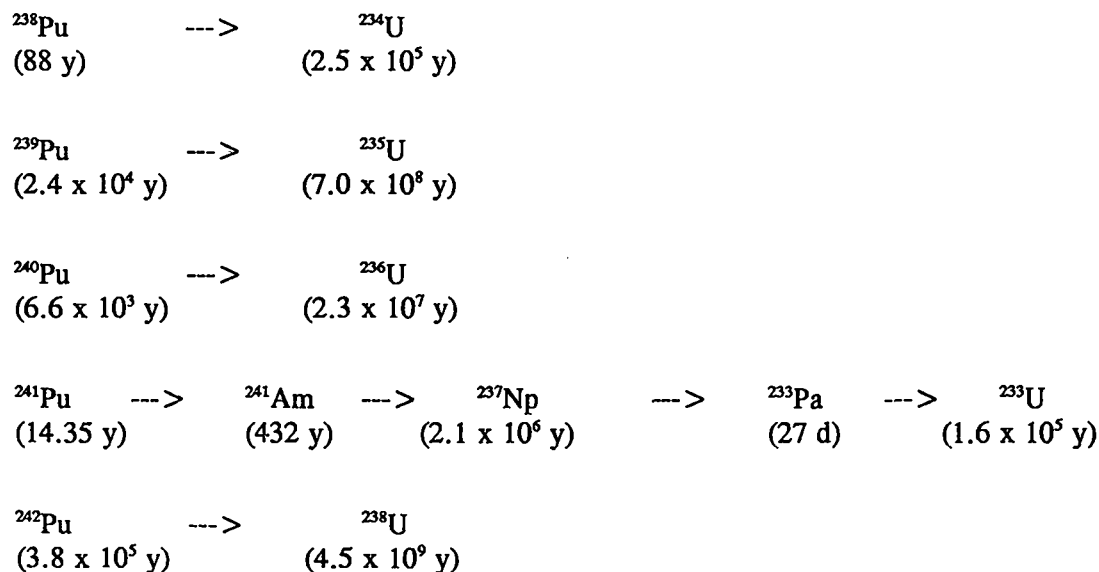


Figure 3-2. Decay chains of weapons grade plutonium components.

batch sorption studies were performed at Los Alamos National Laboratory, using actual plutonium solutions and geologic samples from the vicinity of the Pantex Plant. Studies were conducted using plutonium in various oxidation states, in both deionized and Ogallala Aquifer water, and samples of the Pullman Clay Loam and Ogallala Sand. A full description of the sorption experiments can be found in Appendix A. The plutonium sorption distribution coefficient ( $K_D$ ) measured for the Pullman soil was on the order of  $10^3$  mL/g; the sorption coefficient for the Ogallala sand was on the order of  $10^2$  mL/g. These experimental results are in good agreement with published results which indicate that plutonium sorption distribution coefficients for similar soils and water are on the order of  $10^2$  to  $10^4$  mL/g (Ames and Rai, 1978; Thomas, 1987; Meijer, 1992, Triay et al., 1992). Unfortunately, no samples of the Randall playa soils were available for experimentation, but based on their soil properties (Allen et al., 1972), it is likely that the Randall soils would sorb plutonium more strongly than the Pullman soils tested. Using the Pullman results is therefore a conservative assumption.

### **Americium Solution Chemistry and Sorption**

The americium solubility reported by Nitsche (1990) in an oxidizing groundwater with a high bicarbonate content at  $25^\circ$  C and pH 7 - 8.5 is approximately  $10^{-9}$  M; americium exists in the III oxidation state. The main species at pH 8.5 are Am(III) carbonate complexes; at pH 7 the main species are  $\text{AmCO}_3^+$ ,  $\text{AmOH}^{+2}$ , and  $\text{Am}^{+3}$ . The sorption distribution coefficients for americium are extremely high ( $> 10^3$  mL/g) in groundwaters regardless of the sorbing mineral phase (Thomas, 1987; Triay et al., 1991b; Meijer, 1992). In fact, americium has been shown to sorb even to non-geologic materials, such as the walls of the containers used for sorption experiments (Triay et al., 1991b).

$^{241}\text{Am}$  is a daughter product of  $^{241}\text{Pu}$ , which is a minor constituent of the weapons mix (Table 3-2). Although  $^{241}\text{Am}$  has a relatively high specific activity, its high sorption suggests that it poses only a minimal risk to the aquifer.

### **Uranium Solution Chemistry and Sorption**

Uranium has a high solubility under oxidizing conditions, near neutral pH, in a bicarbonate environment. Uranium exists in the VI oxidation state. The main species in the pH range from 7 - 8 are probably uranyl and U(VI) carbonate complexes. Meijer's review on sorption (1992) indicates that uranium will have a sorption distribution coefficient larger than  $10^2$  mL/g on oxide minerals, clays, and quartz. Uranium seems to sorb to clays by ion exchange. Uranium sorption decreases at higher pH values ( $> 8$ ) due to the formation of U(VI) carbonate complexes (Meijer, 1992).

The long half-lives of most uranium isotopes and uranium's low toxicity relative to plutonium suggest that it is unlikely to pose a significant risk compared to plutonium.

## Neptunium Solution Chemistry and Sorption

The neptunium solubility reported by Nitsche (1990) in an oxidizing groundwater with a high bicarbonate content at 25° C and pH 7 - 8.5 is approximately  $10^{-5}$  M; neptunium exists in the V oxidation state. The main species in the pH range from 7 to 8.5 are  $\text{NpO}_2^+$ ,  $\text{NpO}_2\text{CO}_3^-$ , and  $\text{NpO}_2\text{OH}$ . The sorption distribution coefficients for neptunium are small (on the order of 0 - 50 mL/g) for quartz, feldspar, montmorillonite, and calcite (Thomas, 1987; Triay et al., 1991c; Meijer, 1992). However, neptunium sorbs strongly to metal oxides in the pH range from 7 - 8.5 with a distribution coefficient larger than  $10^3$  mL/g (Triay et al., 1991c). Consequently the oxide mineral coatings in the Pantex soils are expected to retard neptunium transport by sorption.

$^{237}\text{Np}$  is the only neptunium isotope on the decay chains shown in Figure 3-2. It is in the decay chain of  $^{241}\text{Pu}$ , which is a very minor mass constituent of the weapons material (Table 3-2). This low concentration, coupled with its long half-life, limit the risk posed by  $^{237}\text{Np}$ .

It is apparent that of these four elements, plutonium poses by far the greatest risk. The groundwater impact analysis will therefore be confined to plutonium. However, potential effects of the daughter products will be discussed briefly in Section 5, and analyzed in Section 6.

## Conceptual Transport Model

At this point, we have developed a conceptual model that describes our vision of the environmental fate and transport of plutonium dispersed by the hypothetical accident. The jet-fuel fire may disperse fine particulate plutonium downwind of the Pantex Plant. A prompt decontamination effort will reduce radiation levels to  $0.2 \mu\text{Ci}/\text{m}^2$ , but surface runoff and wind transport may concentrate contamination at playa lakes, where surface soil radiation levels may be as high as  $2.0 \mu\text{Ci}/\text{m}^2$ . Surface water infiltrating through this contaminated soil will tend to carry contamination down towards the Ogallala Aquifer, with an average recharge rate of 3 cm/year. Recharge water will reach the Ogallala water table at a depth of 50 - 400 feet, possibly after interacting with one or more perched aquifers on the way down. Sorption interactions with both surficial materials and the unsaturated portion of the Ogallala Formation will retard the movement of plutonium relative to the infiltrating water. During the transport time, radioactive decay will constantly reduce plutonium concentrations. The purpose of the quantitative analyses described in the next section is to estimate if and when plutonium will reach the Ogallala water table, and if so, at what concentration.

## 4 -- ANALYSIS OF GROUNDWATER IMPACTS

Starting with the conceptual transport model just described, we can estimate plutonium arrival times and concentrations at the Ogallala water table. Two alternative calculation methods have been used: a simple non-dispersive piston-flow model, and a more complex model based on the one-dimensional advection-dispersion equation.

### Piston Flow Results

Under the assumptions of piston flow, a "packet" of infiltrating water moves downward towards the water table without interacting with water packets above or below. If we conservatively assume that the entire plutonium surface loading infiltrates with a single year's recharge, we can then envision a contaminated layer of 3 cm of water moving downward through the unsaturated zone completely intact. This contaminated layer will reach the water table as a single unit during a single year at some time in the future, while the preceding and following year's infiltration will be completely uncontaminated. Although this model is clearly unrealistic, it is very simple conceptually and computationally, and provides a useful bounding case of transport behavior.

With the piston-flow model, infiltrating water moves downward with a velocity  $v_w$ :

$$v_w = \frac{I}{\theta}$$

where:

- $v_w$  = Downward water velocity (cm/year)
- $I$  = Infiltration or recharge rate (cm/year)
- $\theta$  = Soil volumetric water content (cm<sup>3</sup> water / cm<sup>3</sup> soil)

Due to sorption effects, plutonium moves slower than water by a factor known as the retardation factor,  $R_{Pu}$ :

$$v_{Pu} = \frac{v_w}{R_{Pu}}$$

where:

- $v_{Pu}$  = Downward plutonium velocity (cm/year)
- $R_{Pu}$  = Plutonium retardation factor

Assuming linear sorption and local equilibrium conditions, the retardation factor can be related to the sorption distribution coefficient for plutonium on the materials in question:

$$R_{Pu} = 1 + \frac{\rho_B K_D}{\theta}$$

where:

$\rho_B$  = Soil bulk density (g/mL)  
 $K_D$  = Sorption distribution coefficient (mL/g)

To calculate travel times and concentrations for plutonium, all of these quantities must be measured or estimated. As described previously,  $K_D$  has been measured directly using laboratory batch studies on Pantex-area geologic materials. Soil bulk density has not been measured directly, but does not vary much under natural conditions, and has been estimated at 1.5 g/mL. Volumetric moisture content, on the other hand, can vary quite widely. Some preliminary geophysical logs near Pantex suggest a mean subsurface moisture content of about 0.15, but also show high variability (Texas Bureau of Economic Geology, 1992).

Fortunately, it can be shown that for highly-sorbed species such as plutonium, the moisture content does not significantly affect solute velocities. For a highly-sorbed species,

$$R_{Pu} \approx \frac{\rho_B K_D}{\theta} \quad , \quad \text{when } K_D \gg 1$$

in which case

$$V_{Pu} = \frac{V_w}{R_{Pu}} = \frac{I/\theta}{\rho_B K_D / \theta} = \frac{I}{\rho_B K_D}$$

Using this last equation, we can calculate plutonium velocities directly, without estimating moisture contents.

Using a recharge rate of 3 cm/year, a soil bulk density of 1.5 g/mL, and a conservative (low-end)  $K_D$  of 100 mL/g, we obtain a downward plutonium velocity of 0.02 cm/year. For water table depths of 50, 200, and 400 feet (15.2, 61.0, 121.9 meters), plutonium travel times are 76,000, 305,000, and 610,000 years, respectively. Based on the 24,400 year half-life of  $^{239}\text{Pu}$ , the longest-lived component of the contaminant mix, during this period concentrations will decrease to 11.5%, 0.02%, and  $3.0 \times 10^{-8}$  of the initial concentration. Again, using our conservative assumption that the entire 2.0  $\mu\text{Ci/L}$  plutonium loading is incorporated into one

year's 3-cm recharge, we can calculate that the initial plutonium concentration will be  $6.7 \times 10^4$  pCi/L. By the time this material reaches the water table at depths of 50, 200, and 400 feet, the concentration will have decreased to 7700 pCi/L, 13 pCi/L, and  $2.0 \times 10^{-3}$  pCi/L, respectively.

Under the piston flow scenario, these concentrations are assumed to occur within a narrow layer of the aquifer, sandwiched between uncontaminated water. The only significant route from the aquifer to a receptor is via a water-supply well, which will likely be screened over a large vertical interval. The contaminated groundwater will therefore be diluted within the well, before it reaches any receptors. The degree of dilution can be estimated by comparing the thickness of the contaminated layer to a typical screen length in a water supply well. Assuming a typical aquifer porosity of 0.3, the 3 cm of contaminated water will form a layer 10 cm thick in the aquifer. A high-capacity water-supply well will typically have at least 50 feet (15 m) of screen. Mixing within the well will therefore dilute the recharge water by a factor of 150, decreasing plutonium concentrations at the well head to 51 pCi/L, 0.09 pCi/L, and  $1.0 \times 10^{-5}$  pCi/L for the three different depth-to-water scenarios. Only the 50-foot scenario result exceeds any recognized drinking water dose limits (Appendix B).

We have shown that even using the physically unrealistic and extremely conservative piston-flow model for plutonium transport, plutonium concentrations in water delivered to potential receptors will be well below dose limits over most of the study area. The only area at risk is shallow groundwater regions south of Pantex, directly opposite predominant wind directions. In order to further determine the extent of risk, we have conducted a more complex analysis, using the advection-dispersion equation.

### Advection-Dispersion Results

In the real world, packets of water do not travel intact through porous media for thousands of years. Rather, mixing processes on various scales tend to mix and homogenize the water as it travels. This effect is known as dispersion, and includes molecular diffusion, mixing within single soil pores (hydrodynamic dispersion), and mixing processes induced by large-scale aquifer property variations (macrodispersion). Transport with a dispersion component can be described using a partial differential equation known as the advection-dispersion equation, and the magnitude of dispersion is measured using an aquifer property known as dispersivity, with units of length.

The advection-dispersion formulation is mathematically more complicated than the piston-flow model, and computer codes are often used for solutions. One such code is CXTFIT (Parker and van Genuchten, 1984). CXTFIT solves the one-dimensional advection-dispersion equation for solutes that display linear sorption and first-order decay. CXTFIT simulations were performed for the same plutonium transport scenarios described for the piston-flow model, using two different dispersivities. A small dispersivity of 1 cm is typical of hydrodynamic dispersion effects, such as might be observed in a column experiment using clean uniform sand. The larger



dispersivity of 1 m might result from macrodispersion effects observable during a field experiment in actual aquifer materials.

The results of these simulations for a 50-foot and a 200-foot deep water table are shown in Figure 4-1. These results show that even with the lower dispersivity and a shallow water table, peak plutonium concentrations after 76,000 years are approximately 1.1 pCi/L, lower than the strictest plutonium dose limits for drinking water. With the higher, more realistic dispersivity value, peak concentrations arrive sooner, but are lower, roughly 0.2 pCi/L. Peak concentrations at the deeper 200-foot water table are about three orders of magnitude lower. 400-foot water table simulations were not conducted, but peak plutonium concentrations would clearly be several orders of magnitude lower still.

The advection-dispersion analysis shows that dispersion effects will reduce peak plutonium concentrations below any drinking water dose limits, even for the low dispersivity / shallow water table scenario. With a more realistic dispersivity value and a more typical deeper water table, peak concentrations are decreased several orders of magnitude further.

3

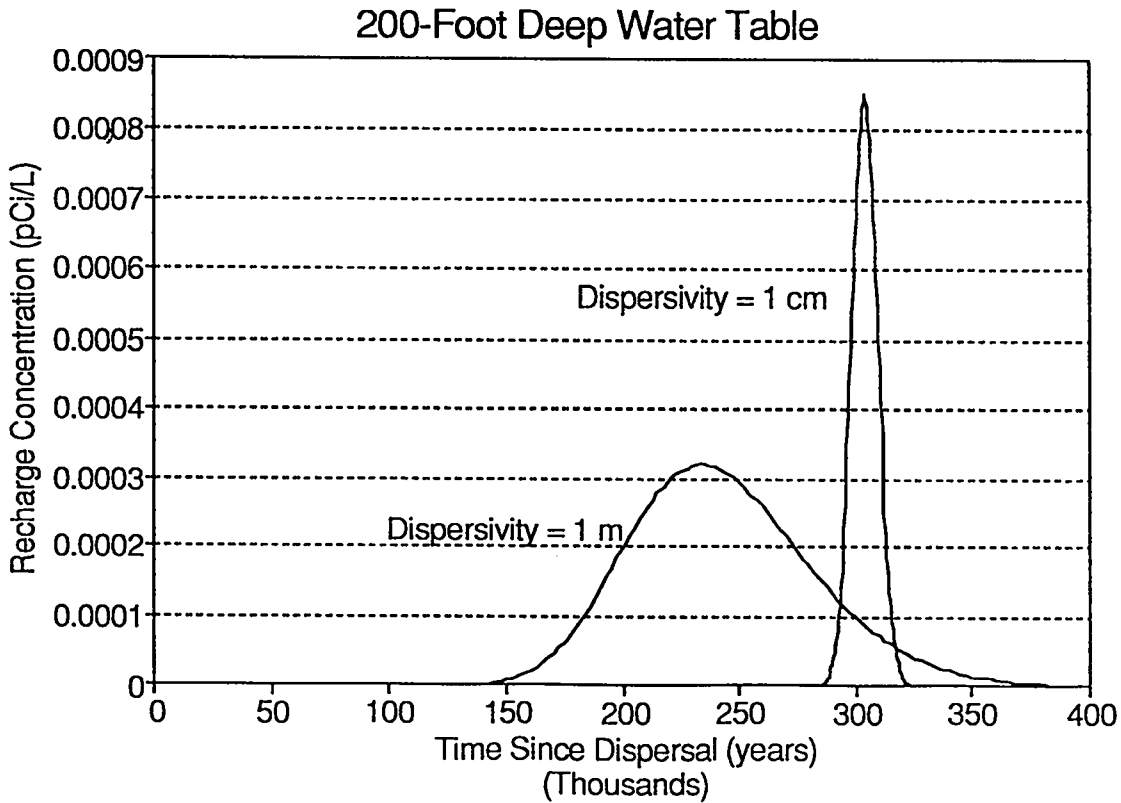
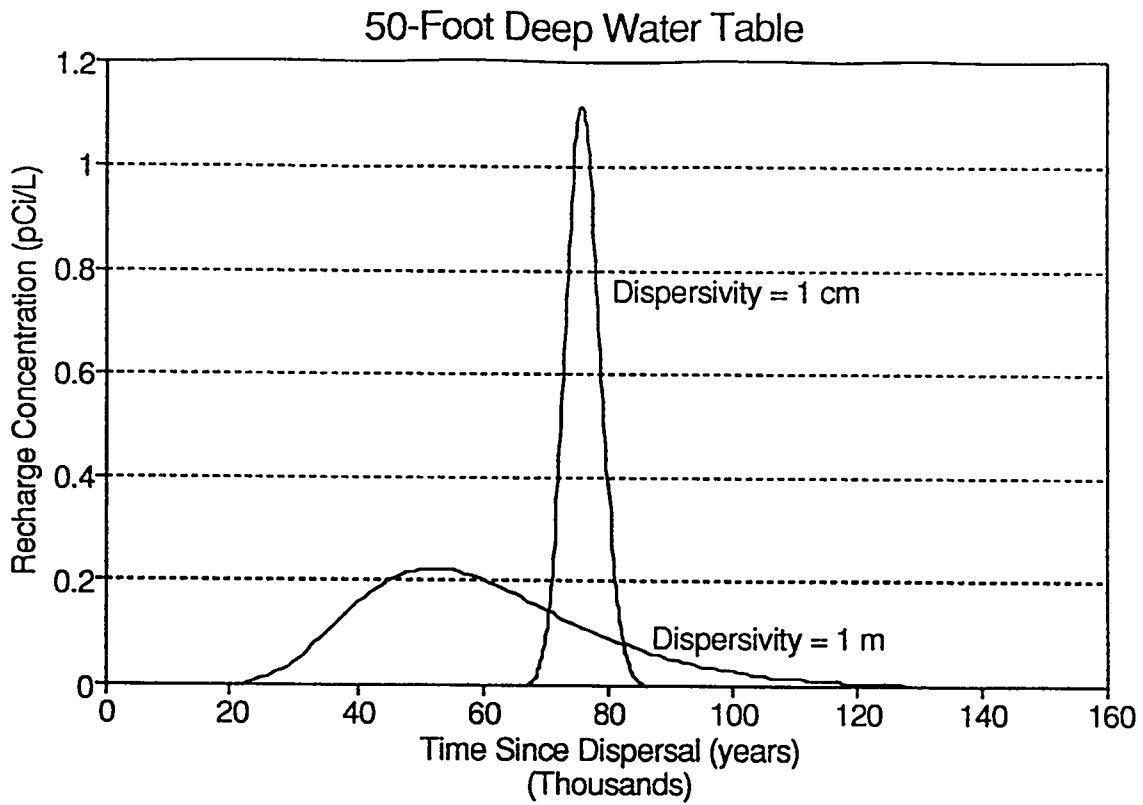


Figure 4-1. CXTFIT simulation results for plutonium transport to a 50-foot and 200-foot deep aquifer.

## 5 -- POTENTIAL COMPLICATIONS

The preceding sections focused on "well-behaved" solute transport processes. There are a number of potential complicating factors that may affect plutonium transport to the Ogallala Aquifer. These factors unfortunately tend to be difficult to quantify accurately, but in the following paragraphs, some of these factors will be described, and their effects approximated.

### Colloidal Transport

Colloids are extremely small ( $< 2\mu\text{m}$ ) solid particles that may be able to sorb contaminants and carry them in moving water. Actinides can form several types of colloids in natural groundwaters (Choppin, 1988; Kim, 1991). "Real colloids" can be formed as a result of the aggregation of hydrolyzed actinide ions. "Pseudocolloids" can be formed by the sorption of actinides onto natural colloids that exist in the groundwater. Triay et al. (1991a) have provided evidence for the formation of real plutonium colloids at pH values of 1 and greater. Pseudocolloids can be formed by the attachment of plutonium or americium onto the colloids in the Ogallala aquifer. Likely natural colloids have been identified by Wood and Petraitis (1984) and Wood and Ostercamp (1987), who invoke clay colloids coated with organic material infiltrating through the unsaturated zone in their hydrologic model of the playa lake basins.

The effects of colloids on the subsurface transport of contaminants has been reviewed by McCarthy and Zachara (1989). Many researchers have invoked colloidal transport to explain the observed movement of contaminants when it exceeds transport predictions. Penrose et al. (1990) observed plutonium and americium mobility through a shallow aquifer in Los Alamos (a semiarid region); they postulate that the observed transport (which exceeds predictions based on laboratory data) is due to the irreversible sorption of plutonium and americium to colloidal material.

The effect of colloidal filtration by a porous medium (McDowell-Boyer et al., 1986) is a likely mechanism for retardation of colloids traveling through the Ogallala formation. However, this filtration effect is difficult to quantify in the absence of a detailed characterization of the colloids in the groundwater and a detailed hydrologic description of the transport pathway.

The most relevant paper relative to likely conditions in the study area after an accidental dispersal of plutonium is the work of Goss et al. (1973). In this study Goss and co-workers performed recharge experiments in Bushland, Texas, on the Southern High Plains just 50 km west of the Pantex Plant. These investigators eluted radioactively-tagged clay colloids through a recharge basin. The basin was constructed to maximize the potential for colloidal transport by removing the slowly permeable surface of the Pullman clay loam and exposing the calcareous underlying sediments, observed to have large pores (ranging from .1 to 1 mm). After infiltration of the radioactively tagged clay colloids in 72.5 ft of water (equivalent to 730 years of natural recharge, assuming 3 cm/year), 35% of the colloids were captured in the top six inches, 50% in the top 18 inches, 75% in the top 4 feet, and there was no evidence of any

colloid transport below the top nine feet. In a second experiment, in which the large pores were destroyed by cultivation, they found that after 52.2 ft of water infiltrated (530 years of natural recharge), over 90% of the radioactively tagged colloids were retained in the upper 1 inch and there was no evidence of colloid transport below the top 30 inches. Based on these results it is not likely that colloid transport is a dominant transport mechanism for the migration of contaminants to the Ogallala Aquifer.

### **Perched Aquifers**

As mentioned previously, there is a known perched aquifer above the Ogallala Aquifer in the immediate vicinity of the Pantex Plant (Purtymun and Becker, 1982; Texas Bureau of Economic Geology, 1992). The lateral extent and continuity of this layer are not known, and it was completely neglected during the travel time and concentration analyses described previously. This was a conservative decision, because it is clear that a perched water zone, if present, would act as an impediment to downward transport for two reasons. First, the existence of perched water indicates that there is a relatively impermeable perching layer present that is impeding downward flow. Second, the perched water constitutes an additional reservoir of water within the unsaturated zone, which implies increased travel times for a given recharge rate. Therefore, a perched aquifer, if present beneath the hypothetical dispersal area, would actually decrease the risk to Ogallala groundwater relative to that calculated here.

The shallow perched aquifer itself would be more vulnerable to contamination than the deeper Ogallala aquifer, and in the event of a plutonium-dispersal accident, monitoring of perched-aquifer water quality would be prudent.

3

### **"Short-Circuit" Recharge Paths**

Under the conceptual model described earlier, primary protection of the aquifer is provided by slow flow and transport through the unsaturated zone. It is possible that artificial recharge conduits could bypass much of the unsaturated zone, thereby "short-circuiting" this protective layer. Two major types of short-circuit paths are unintentional flow down along improperly-constructed or abandoned water wells; and intentional flow to the Ogallala Aquifer as part of an artificial recharge project.

Unintentional downward flow through water wells is unlikely to constitute a major problem, because the high silt content of surface water in the area tends to quickly clog aquifer materials encountered. In fact, this clogging effect stymied almost all early attempts at intentionally recharging the Ogallala Aquifer with playa lake water (Urban et al., 1988). The risk of short-circuiting along improperly-constructed or abandoned wells could be further minimized by identification and grouting of these wells, possibly in conjunction with the post-dispersal decontamination effort.

Recent advances in recharge techniques using geotextile filters have made artificial recharge of the Ogallala Aquifer a real possibility, which will become ever more attractive as the water table continues to fall (Urban et al., 1988). A successful artificial recharge project may conceivably provide a means of moving plutonium down to the Ogallala Aquifer rapidly and in potentially hazardous concentrations, although the same geotextile filter that traps silts may trap most colloidal or polymeric plutonium. It is therefore imperative that in the event of an accident that disperses plutonium into the environment, active groundwater recharge projects be monitored and shut down if necessary.

The potential risks posed by these "short-circuit" recharge paths are purely local in scale, and may be minimized by implementing the described measures in the event of the accident.

### Preferential Flow

The term "preferential flow" encompasses different phenomena that all cause accelerated transport -- solute velocities greater than that predicted using the simple infiltration rate / moisture content relationship described earlier. Macropore flow is one obvious example, in which water rapidly infiltrates via burrows, root casts, mud cracks, or other visible soil features. Immobile water trapped in dead-end pores can also cause accelerated transport, as can microporosity within individual soil grains. Gish and Shirmohammadi (1991) have edited a large collection of papers describing recent research in this field.

The magnitude of accelerated transport observed in actual field studies varies widely, from none (Biggar and Nielsen, 1976; Van De Pol et al., 1977; Jury et al., 1982) to factors on the order of 2 (Bowman and Rice, 1986a; 1986b; Turin, 1992). In one experiment, solutes moved roughly 5 times the predicted rate (Rice et al., 1986). The degree of preferential flow seems to depend primarily on soil type and infiltration patterns.

It is very difficult, if not impossible, to accurately determine the importance of preferential effects on plutonium transport in the study area without conducting detailed field studies. To get some idea of the sensitivity of the results of the advection-dispersion analysis to accelerated transport, two additional CXTFIT simulations were performed for the 50-foot water table case, using twice the calculated water velocity. Results are shown in Figure 5-1. As might be expected, higher transport velocities result in faster plutonium arrival at the water table. Shorter elapsed time before arrival means less radioactive decay, so peak concentrations are higher. Even so, under these strongly conservative assumptions and a dispersivity of 1 cm, predicted peak plutonium concentrations exceed only one of the three applicable drinking water dose limits (Appendix B), and with a dispersivity of 1 m, no dose limits are exceeded. These results indicate that even if preferential flow effects double predicted velocities, adverse impacts on the Ogallala Aquifer are likely to be minimal.

## Preferential Flow Effects 50-Foot Deep Water Table

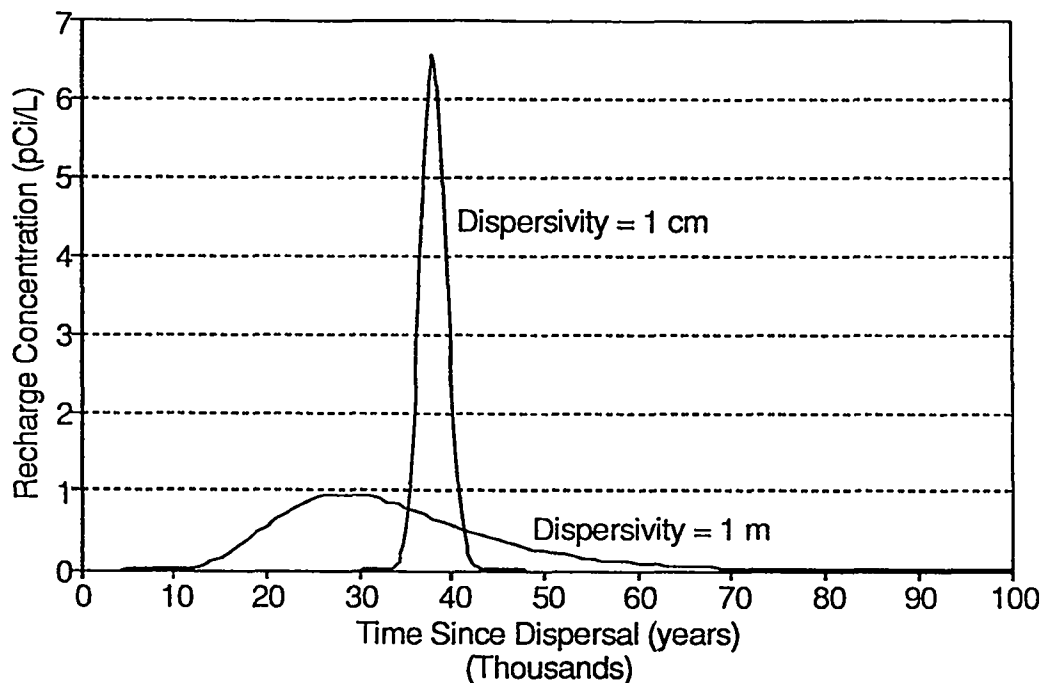


Figure 5-1. CXTFIT simulation results for plutonium transport to a 50-foot deep aquifer, with accelerated transport due to preferential flow.

### Multidimensional Flow

Both the piston-flow and advection-dispersion models assume one-dimensional downward flow through the unsaturated zone. Approximately one-dimensional flow and transport would occur if recharge was uniform across the surface of the High Plains, but as pointed out earlier, recharge is focused at the playa lakes, and relatively unimportant elsewhere. Therefore, the assumption of one-dimensional flow is not strictly valid. In this case, however, one-dimensionality is a conservative assumption, and the true multidimensional nature of the flow system will decrease the impact on the aquifer below that predicted here, for two major reasons. Transverse (horizontal) dispersion in the unsaturated zone will cause increased dilution of the contaminated recharge water, and horizontal spreading of the plume will increase travel time to the water table, in turn allowing more radioactive decay en route.

## **Dropping Groundwater Levels**

The groundwater impact analyses were performed assuming that 1990 water levels would persist into the future. The present rate of water-table decline indicates that this is clearly not likely. In fact, in much of the study area, the water table is dropping faster than the calculated plutonium velocity, implying that the plutonium will never catch up and reach the water table. Obviously, the present rate of water-table decline cannot persist for even a hundred years, and certainly not for the tens of thousands of years discussed in this analysis, because the Ogallala Aquifer would be pumped dry long before. Predicting actual future water levels in the Ogallala is beyond the scope of this report, but it seems likely that as long as the Ogallala Aquifer remains the primary water supply for agriculture, residential, and industrial uses in the Texas Panhandle, water levels are likely to continue to fall. In this case, using present-day water levels is a conservative assumption, because at the hypothetical future date of the plutonium-dispersal accident, and certainly by the time plutonium would have infiltrated down to the water table, depths to water will have increased, affording additional protection to the aquifer.

## **Decay Products**

As mentioned in Section 3, plutonium poses the primary risk in this hypothetical situation, and was therefore the only element considered in the preceding analyses. The conclusion that minimal risk is posed by plutonium contamination suggests that none of the other potential contaminants pose a significant risk. However, as a check of this assumption, the RESRAD model, which incorporates decay-product transport, was used to calculate total doses caused by the hypothetical accident. Results are presented and discussed in the following Section.

3

## 6 -- DOSIMETRY AND RESRAD MODELING OF PLUTONIUM MIGRATION

A check of the results of the previous groundwater impact analyses was carried out using a model developed for estimation of dose from residual radioactivity. The model, RESRAD, was developed for determination of dose to man from all pathways of migration of radionuclides (Gilbert et al., 1989). The model is recommended by DOE Order 5400.5 for determination of doses and cleanup levels for residual radioactivity in the environment. RESRAD is a complex model that allows the user to select site-specific parameters for estimation of dose. The result is a more realistic estimation of the radiation dose, estimated as Committed Effective Dose Equivalents (CEDE) that takes into consideration the specific climate and land use parameters of the contaminated area.

For this project, several scenarios were developed to check on the potential for plutonium migration to the water tables in the area of Pantex. These scenarios included three different aquifer depths: 400 feet, 200 feet, and 50 feet.

The first RESRAD simulation determined the CEDE resulting from a soil contamination level of  $0.2 \mu\text{Ci}/\text{m}^2$ , using the default values of the model and standard conditions described for the EPA's proposed standard for transuranic elements in soil. The output indicated a committed dose equivalent of 10 mrem/year for the heavy dust loadings in air used by EPA, confirming the behavior of the model. The doses in this scenario were dominated by resuspension of plutonium attached to soil particles -- water pathways contributed an insignificant amount of radiation dose from plutonium in the environment. Additional calculations using RESRAD for conditions of a plume of plutonium contamination at the EPA screening limit dispersed over a wide area results in the same conclusion, that the migration of plutonium to an aquifer is not a significant source of radiation risk.

Tables 6-1 and 6-2 list the site-specific distribution coefficients ( $K_d$ ) and initial contaminant isotopic composition used in the calculations. The isotopic mix used was weapons grade plutonium (Table 3-2).  $^{241}\text{Pu}$  is the most abundant isotope, but emits beta particles. The radioactive decay product  $^{241}\text{Am}$  is an alpha-emitter that appears wherever and whenever  $^{241}\text{Pu}$  decays.

Table 6-3 lists the resulting breakthrough times for first arrival of contaminants at different depth aquifers, and Table 6-4 lists estimated maximum dosages for the groundwater pathways. Each scenario simulated has individual doses calculated for inhalation of air and dusts, consumption of foods grown on the contaminated soils, meat and milk produced in the contaminated area, and drinking water from an aquifer below the contaminated zone. Radioactive decay products are included in the calculations. Estimation of the time of maximum dose rate is made by carrying out dose calculations over 10,000 years. In all cases, the dose was dominated by inhalation of resuspended plutonium followed by ingestion of plutonium attached to soil particles on foodstuffs.



Table 6-1. Distribution coefficients used for RESRAD simulations.

<u>Medium</u>	<u>Element</u>	<u>K<sub>D</sub> (mL/g)</u>
Contaminated Soil	Pu	3000
	Am	1000
Top meter of soil	Pu	3000
	Am	1000
Unsaturated zone	Pu	100
	Am	1000
Aquifer sands	Pu	100
	Am	1000

Table 6-2. Initial contaminant mixture used for RESRAD simulations.

<u>Nuclide</u>	<u>Activity (pCi/g)</u>
<sup>238</sup> Pu	1.62
<sup>239</sup> Pu	10.7
<sup>240</sup> Pu	2.53
<sup>241</sup> Pu	83.6
<sup>242</sup> Pu	0.0004
<sup>241</sup> Am	0.121

The water dependent pathways did show one case of 0.001 mrem/year from migration of <sup>237</sup>Np, a decay product of <sup>241</sup>Am, after 30 years for an aquifer 50 feet below the surface. <sup>237</sup>Np is conservatively assigned a K<sub>D</sub> of 0, indicating that the material moves with water with no adsorption on the soils. The calculated dose for all other scenarios and times was zero. The amount of transuranic elements was not sufficient to contaminate the water enough to produce a radiation dose greater than 0.001 mrem/year for the large area of contamination at low levels. RESRAD calculations resulted in zero dose for water pathways for all other depths to the water table over a 10,000-year period.

The final case set up for calculations by RESRAD was that of a playa that received runoff containing suspended sediments over time. In this case, the playa acts as an accumulation point for radioactively contaminated sediments. This type of concentration was demonstrated within

Table 6-3. Contaminant breakthrough times predicted by RESRAD for various aquifer depths.

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<u>Depth to Aquifer</u>	<u>Breakthrough Time</u>
400 feet	190,000 years
200 feet	110,000 years
50 feet	56,000 years

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Table 6-4. Maximum water pathway doses predicted by RESRAD for various aquifer depths.

---

<u>Depth to Aquifer</u>	<u>Maximum Dose (mrem/yr)</u>
400 feet	0.0
200 feet	0.0
50 feet	0.001

---

the Trinity Test fallout zone by Hansen and Rogers (1985), who measured accumulations of radioactive fallout  $^{137}\text{Cs}$  in New Mexico playas that were 1.5 to 2 times the concentrations in surrounding slopes after 32 years. For this calculation, the playa size and watershed area were based on Playas 1 and 2 at the Pantex Plant. The aquifer was conservatively assumed to be located at 50 feet below the surface. The inventory of plutonium and americium isotopes was multiplied by 10 to reflect a conservative concentration of the residual radionuclides in playa sediments. For this scenario, the maximum dose calculated was 0.01 mrem/year after 30 years, from  $^{241}\text{Am}$  formation of  $^{237}\text{Np}$ . After 100 years the dose was zero. This conservative case indicates that an insignificant radiation dose would be produced by the residuals resulting from the hypothetical accident.

To summarize, in every RESRAD simulation, the doses estimated were zero or well below any applicable standard for radioactive contamination of aquifers or drinking water.

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## APPENDIX A

### Laboratory Plutonium Sorption Study

#### *<sup>239</sup>Pu-Solutions Preparation:*

Three <sup>239</sup>Pu oxidation states were studied: IV, V, and VI. Two waters were used for preparing solutions: deionized water and water typical of the Ogallala aquifer (filtered through a 0.05 mm polycarbonate filter). Solutions were prepared by adding an aliquot of a well-characterized Pu(IV), Pu(V) or Pu(VI) acidic stock to deionized or Ogallala water. The concentration of all plutonium solutions was  $10^{-7}$  M with the exception of the Pu(IV) solution in the Ogallala water which was  $10^{-8}$  M. The reason for this difference is that Pu(IV) acidic stock was added to the Ogallala water and most of the plutonium precipitated out (probably due to Pu(IV) polymer formation). The resulting  $10^{-8}$  M plutonium solution was stable with no further loss of plutonium after centrifugation for 4 hours at 12,000 rpm (28,000 g). Aliquots of all the plutonium solutions prepared were centrifuged for 4 hours at 12,000 rpm and the initial plutonium concentrations never differed from the plutonium concentrations after centrifugation by more than 6%.

#### *Soil Samples:*

Soils samples from the Ogallala Formation along Highway 136 (sample numbers 3A and 3B) and the A Horizon of the Pullman soil (sample numbers 1A and 1B) were air-dried and sieved to obtain particles in the size range from 75 - 500  $\mu$ m. For experiments involving the Ogallala water, soil samples were preconditioned with the water by equilibrating the soil with the Ogallala water (in the ratio of 1 g to 20 mL) for 24 hours. The phases were separated by centrifugation for 1 hour at 12,000 rpm (28,000 g). The pre-conditioned soil was used for the sorption experiments. The pH and Eh of the water after equilibration with the soil was measured.

#### *Sorption Procedure:*

The sorption procedure utilized in these studies consists of:

- 1) equilibrating a soil sample with an aliquot of a plutonium solution (in the ratio of 1 g to 20 mL) for 48 hours,
- 2) separating the phases by serial centrifugations totaling 4 hours at 12,000 rpm (28,000 g).
- 3) measuring the pH and Eh of the water after sorption,
- 4) determining the amount of plutonium sorbed to the soil by liquid scintillation counting, and

5) calculating  $K_D$ , defined as:

$$\frac{\text{moles of plutonium sorbed to solid phase per g of soil}}{\text{moles of plutonium in groundwater per mL of solution}}$$

*Analytical Procedure for  $^{239}\text{Pu}$  Determination:*

Liquid scintillation counting consists of counting the  $^{239}\text{Pu}$  solution three times for fifteen minutes in a Packard Liquid Scintillation Counter with a window setting from 25 to 800 KeV. The liquid scintillation samples consist of an aliquot of the plutonium solution and an aliquot of make-up water totaling 6 mL, and 14 mL of Ultima Gold Liquid Scintillation Fluor. The efficiency of the counter was determined by counting an aliquot of the  $^{239}\text{Pu}$  standard SRM-949F. The counter was found to be 100% efficient.

*Results*

The results of the sorption experiments are given in Tables A-1 and A-2. The equilibration among different oxidation states of plutonium in solution is slow; however only one optimal plutonium speciation in each of the waters is expected once the plutonium solution attains equilibrium. The water chemistry in these experiments is expected to be controlled by the type of soil used. Consequently, sorption is fairly independent of initial plutonium oxidation state and water used. The reason for the higher plutonium sorption distribution coefficients for the Pullman soil is its high clay content. The Ogallala sand consist of quartz and feldspars which have a lower affinity for plutonium than clay. The results for Pu(VI) in the Ogallala water are different from all the other results due the change of pH during the experiment (from 8.2 to 7.4). Actinides are well known for having a sorption edge in the pH region from 7-8. The reason for the change in pH in the case of Pu(VI) in the Ogallala water was probably an experimental artifact; the pH of the Pu(VI) solution in the Ogallala water was not adjusted correctly after the aliquot of the Pu(VI) acidic stock was added to the water. The most representative sorption results are those obtained with Pu(V) solutions since Pu(V) is the expected plutonium oxidation state in this type of water. Consequently, even if the plutonium solution did not attain equilibrium prior to use in the plutonium experiments, most of the plutonium in the solutions should have been in the V oxidation state.



Table A-1. Plutonium sorption onto Pullman soil. Initial Ogallala water: pH 8.2, Eh 240 mV. After pre-conditioning: pH 7.7, Eh 220 mV.

Initial Plutonium Oxidation State	$K_D$ (mL/g)		$^{239}\text{Pu}$ Solution after Sorption	
	Sample 1A	Sample 1B	pH	Eh (mV)
<i>Deionized Water</i>				
IV	$4 \times 10^3$	$3 \times 10^3$	8.1	190
V	$3 \times 10^3$	$4 \times 10^3$	8.3	200
VI	$3 \times 10^3$	$5 \times 10^3$	8.1	200
<i>Ogallala Aquifer Water</i>				
IV	$2 \times 10^3$	$2 \times 10^3$	7.9	240
V	$3 \times 10^3$	$4 \times 10^3$	8.0	230
VI	$8 \times 10^2$	$9 \times 10^2$	7.4	230

Table A-2. Plutonium sorption onto Ogallala sand. Initial Ogallala water: pH 8.2, Eh 240 mV. After pre-conditioning: pH 8.1, Eh 220 mV.

Initial Plutonium Oxidation State	$K_D$ (mL/g)		$^{239}\text{Pu}$ Solution after Sorption	
	Sample 3A	Sample 3B	pH	Eh (mV)
<i>Deionized Water</i>				
IV	$3 \times 10^2$	$5 \times 10^2$	8.3	220
V	$6 \times 10^1$	$1 \times 10^2$	8.3	220
VI	$7 \times 10^2$	$8 \times 10^2$	8.3	220
<i>Ogallala Aquifer Water</i>				
IV	$4 \times 10^2$	$5 \times 10^2$	8.2	220
V	$1 \times 10^2$	$2 \times 10^2$	8.3	230
VI	$3 \times 10^2$	$4 \times 10^2$	7.6	240

## APPENDIX B

### Recommended Drinking Water Dose Limits for Plutonium Ingestion

#### A. DOE and EPA Drinking Water Dose Limits

The Committed Effective Dose Equivalent (CEDE) is the sum of the dose equivalents from  $^{239}\text{Pu}$  alpha radiation deposited internally to various tissues in the body, each multiplied by the appropriate ICRP 26 weighting factor. The CEDE is expressed in units of mrem.

In DOE Order 5400.5-II.1a the public dose limit is defined as "the exposure of members of the public to radiation sources as a consequence of all routine DOE activities shall not cause, in a year, an effective dose equivalent greater than 100 mrem." Derived Concentration Guides (DCGs) are given in 5400.5 for ingestion of two liters of water a day based on a CEDE of 100 mrem for plutonium taken into the body for one year. Plutonium is poorly absorbed from the gut and chemical forms influence the absorption. DOE gives an  $f_1$ , the fraction absorbed from the gut into the body, of .00001 for plutonium oxides and hydroxides, .0001 for plutonium nitrates, and .001 for all other chemical forms of plutonium in the environment. The DOE DCGs are 2000 pCi/L for oxides and hydroxides and 30 pCi/L for "other" chemical forms of plutonium. To be conservative we will choose  $f_1$  of .001 since we do not know the long term chemical form of plutonium in this ecosystem even though the initial form is probably an oxide. Once we assume  $f_1$  then the  $^{239}\text{Pu}$  Dose Conversion Factor (DCF), in mrem CEDE/pCi ingested, can be derived based on the ICRP 26 weighting factors, metabolism, and deposition patterns in the body after absorption from the gut. DOE/CH-8901 gives a DCF of .0043 mrem/pCi for an  $f_1$  of .001. This gives the DOE DCG:

$$\begin{aligned}x \text{ pci/L} &= (y/365d)(d/2L)(1/DCF)(100\text{mrem/yr Public Limit}) \\ &= (1/365)(1/2)(1/.0043)(100) \\ &= 30 \text{ pci/L in drinking water}\end{aligned}$$

DOE 5400.5-I3 also states " a higher dose limit, not to exceed the 500 mrem effective dose equivalent recommended by ICRP as an occasional annual limit, may be authorized for a limited period if it is justified by unusual operating conditions." Therefore, under "unusual" conditions a  $^{239}\text{Pu}$  drinking water limit of 150 pCi/L could be acceptable and approved by DOE.

The Safe Drinking Water Act regulated by EPA under 40CFR141, National Interim Primary Drinking Water Regulations, prescribes radionuclide concentration limits for public drinking water systems. The limits in public water systems (PWS) for  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  are explicitly given in 40CFR141.15 as "(a) Combined radium-226 and radium-228: 5pCi/L." Back calculation of this limit to mrem CEDE/y gives:

$$\begin{aligned}\text{mrem CEDE/y} &= (2L/d)(365d/y)[(2.5\text{pCi }^{226}\text{Ra/L})(.0011\text{mrem/pCi}) + \\ &\quad (2.5\text{pCi }^{228}\text{Ra/L})(.0012\text{mrem/pCi})] \\ &= 4.2 \text{ mrem CEDE/y}\end{aligned}$$

The EPA regulations further state in 40CFR141.26-a1i that "a gross alpha particle activity measurement may be substituted for the required radium-226 and radium-228 analysis provided that the measured gross alpha particle activity does not exceed 5 pCi/L at a confidence level of 95 percent." The regulations also develop other limits for beta and gamma emitting nuclides based on the 4mrem CEDE/y.

DOE Order 5400.5-II.1d adopts the SDWA regulations by stating "It is the policy of DOE to provide a level of protection for persons consuming water from a public drinking water supply operated by the DOE, either directly or through a DOE contractor, that is equivalent to that provided to the public by the public community drinking water standards of 40 CFR Part 141. These systems shall not cause persons consuming the water to receive an effective dose equivalent greater than 4 mrem in a year. The dose limit is the effective dose equivalent to individuals whose exclusive source of drinking water contains a radionuclide, or a mixture of radionuclides, at a monthly average level of four percent of the appropriate DCG value." This <sup>239</sup>Pu limit for PWS can be derived as follows:

$$\begin{aligned}x \text{ pCi } ^{239}\text{Pu/L} &= (y/365)(d/2L)(1/.0043)(4\text{mrem/y}) \\ &= 1.3 \text{ pCi } ^{239}\text{Pu/L of drinking water}\end{aligned}$$

#### B. Recommendations for Limiting Doses

Three dose limits are recommended for plutonium ingested exclusively from the drinking water pathway, one at 1.3 pCi/L for PWS using 4 mrem CEDE/yr, the second at 30 pCi/L for the "worst case plausible scenario" for the family farm scenario addressed in DOE/CH-8901 based on 100 mrem CEDE/yr, and a third at 150 pCi/L based on 500 mrem CEDE/yr "unusual circumstances" explained in 5400.5 IV.3b. We recommend usage of all three limits for this exercise because the remediation and subsequent residual contamination of areas in the square kilometer range will have public water systems, private farms with their own wells, and hot spot unusual circumstances requiring application of the three limits based on risk, cost/benefit optimization, and the site specific scenario.

#### C. References

DOE Order 5400.5. "Radiation Protection of the Public and the Environment", Effective May 1990.

EPA Title 40 CFR 141. "National Interim Primary Drinking Water Regulations", (Safe Drinking Water Act, Amended 1988).

Gilbert et al., June 1989. "A Manual for Implementing Residual Radioactive Material Guidelines", DOE/CH-8901, Argonne National Laboratory.

ICRP, 1977. Recommendations of the International Commission on Radiological Protection. ICRP Publication 26. Pergamon Press, Oxford.