



# Oregon

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September 6, 2011

Ms. Paula Call  
U.S. Department of Energy  
PO Box 550, A7-75  
Richland, WA, 99352

Dear Ms. Call:

On July 19, 2011, Oregon submitted written comments on the *Draft Proposed Plan for the Remediation of the 200-CW-5, 200-PW-1, 200-PW-3, and 200-PW-6 Operable Units, DOE/RL-2009-117, Revision 0*. This letter provides additional formal comments on Oregon's behalf.

Our July 19 comments expressed strong concerns with the *draft Proposed Plan* for proposing to leave potentially large amounts of plutonium in the three waste sites that make up the High-Salt Waste Group. The *draft Proposed Plan* contends this would be protective because plutonium will not move in the Hanford subsurface and therefore will pose no threat to people or the environment.

Our July letter referenced Oregon's disagreements with the U.S. Department of Energy's (DOE) position that plutonium does not move in the Hanford subsurface. Those disagreements have been well documented through correspondence and meetings over the past several years. Through this additional comment letter, we wish to further emphasize our concern that plutonium in the subsurface at Hanford is likely mobile. We are attaching a summary of recent literature on this topic, including extensive bibliographic material, to support our argument. We hope that DOE and Hanford regulators will review these materials and will seriously consider a more protective remedy for the High-Salt Group waste sites.

During the more than 20 years that Oregon staff have been involved in the Hanford cleanup, we have observed tremendous growth in the scientific understanding of the environmental chemistry of plutonium. The chemistry of plutonium and other actinides in the soil has been shown to be tremendously complex, and the improvements in understanding this chemistry have been significant. The attached materials provide an overview of recent research on the environmental geochemistry of plutonium. Among key findings noted in the attached discussion, with regard to the fate and transport of plutonium:

- The solution chemistry of plutonium is complex, and is strongly affected by three major factors – redox conditions, pH and carbonate concentration.
- Plutonium commonly exists in equilibrium in more than one valence state. No single valence dominates for long, as valence changes with subtle changes in soil conditions.

- Plutonium readily forms stable complexes with carbonate and hydroxyl ions, especially in high pH, carbonate-rich environments like those that occur in Hanford soils and groundwater. Complexes can be soluble at concentrations well above water quality standards.
- The complexes may be dissolved or colloidal, with colloids ranging from nanometer- to micron-sized particles. Complexes and colloids can be very mobile in water; transport in soils over distances greater than one kilometer has been observed at several locations.

There remains considerable uncertainty regarding the precise mechanisms that control the movement of plutonium in any individual soil environment, and in the resulting rate and amount of movement. There is, however, virtually no debate in the scientific community about the fact that plutonium is mobile in soil and groundwater environments. The issue is not whether plutonium moves in soil and groundwater, but rather how much, and how fast, it is moving.

Given the complexities of plutonium aqueous chemistry, it should be recognized that simplistic tools that have been used in the past to predict plutonium mobility, such as  $K_d$ s, are not adequate for predicting plutonium dynamics in the complex environments of soils and waste sites or for predicting protectiveness. It is therefore essential to develop better quantitative tools to estimate the rate and amount of movement, in order to support a sound assessment of the protectiveness of any proposed remedial action at Hanford for waste sites with a significant inventory of plutonium.

The science and chemistry described in the attached discussion and citations directly challenge the assertions that plutonium is not mobile, and that leaving substantial amounts of plutonium in the Hanford subsurface is protective. We strongly urge DOE as the Site owner, and the U.S. Environmental Protection Agency as lead regulator for the waste sites considered in the *draft Proposed Plan*, to carefully review these materials and to reconsider the proposed remedies for waste sites in the High-Salt Waste group.

Please contact Dale Engstrom of my staff (503-378-5584), with any questions or comments.

Sincerely,



Ken Niles  
Nuclear Safety Division Administrator

Cc Stuart Harris, Confederated Tribes of the Umatilla Indian Reservation  
Russell Jim, Yakama Nation  
Gabriel Bohnee, Nez Perce Tribe  
Max Power, Chair, Oregon Hanford Cleanup Board  
Susan Leckband, Chair, Hanford Advisory Board  
Hanford Natural Resource Trustee Council

## ATTACHMENT

### Discussion of research related to the movement of plutonium in the environment

Research conducted by the U.S. Department of Energy (DOE), its national laboratories, and by others mostly outside of Hanford note there is great uncertainty associated with plutonium mobility. The following documents raise questions about previous assertions that plutonium in the subsurface is not mobile. The range of authors includes many of the principal experts in the United States and in the world in this field of chemistry, variously called “actinide chemistry,” “f-element chemistry,” “transuranic chemistry” and other names.

With regard to *Plutonium Chemistry*:

As noted by Contardi et al.<sup>6</sup> “even in situations where plutonium transport has been documented it is often difficult to determine what mechanism was responsible.” Duff said in her summary in 2001<sup>7</sup> that “sorption, co-precipitation and oxidation state speciation behavior of Pu on geologic materials is poorly understood.” Also noted by Duff<sup>7</sup> was that “a once highly stable form of Pu contamination may eventually become a very active source-term to the surrounding environment.”

Plutonium released to the environment continues to be a major concern at a number of DOE facilities. Los Alamos National Laboratories<sup>2</sup> found that the fate and transport of plutonium in the subsurface is strongly influenced by “coupled physical and biogeochemical processes” present at these contaminated sites. Under groundwater conditions, Pu(IV), Pu(V) and Pu(VI) are the most available and stable oxidation states, with Pu(V) and Pu(VI) fractions generally remaining in solution and Pu(IV) usually present in the solid phase as sparingly-soluble oxyhydroxides and surface sorbed species. However, plutonium nearly always exists in many valence states in equilibrium at the same time. No single valence dominates the valence mix for long except under very controlled conditions that do not exist in the natural environment. Plutonium equilibrates under natural conditions in several valence states, changing from one valence to another in reaction to subtle changes in soil conditions and location. Oxidized plutonium species Pu(V) and Pu(VI) are present in solutions at low concentrations under oxic environmental conditions, but are highly soluble and play an important role in defining overall transport behavior of plutonium.”<sup>1</sup>

Given the importance of plutonium’s oxidation state to its mobility, workers at EPA<sup>27</sup> and others find it necessary for risk assessments to carefully consider all of the factors that may influence oxidation state at a given site. This task is complicated by the fact that the oxidation state of plutonium depends both on how the contaminant was originally formed and released to the environment, as well as on the environmental conditions it is exposed to following placement.<sup>8,9,14,19,20,24,26,27</sup>

Most major plutonium mobility research has found that, in addition to adsorption, a number of other processes are known to be important to determine plutonium mobility. These processes

include such things as changes in the oxidative state of the plutonium through redox transformations<sup>10</sup>, transport of plutonium on or in at least seven different types of colloidal particles<sup>25</sup>, formation of charged and neutral chemical complexes with even greater mobility, and precipitation or co-precipitation of solid contaminant phases.<sup>10,17,25</sup>

According to Clark, et. al.<sup>4,5</sup>, plutonium chemistry is extremely complex. Clark et al<sup>5</sup> found that the dominant aspects of plutonium mobility usually involve carbonate and other complexes that are soluble at levels well above standards, that are mobile and that contribute to movement of plutonium. Plutonium electrochemistry likewise plays an important role resulting in negatively and positively charged complexes<sup>10</sup>.

“For example, as discussed in the article “The Chemical Interactions of Actinides in the Environment” (Runde (2000)<sup>20</sup>, beginning on page 392) if plutonium is accidentally released into the environment, its chemical properties will determine to a large extent whether its transport will be retarded by precipitation from solution or sorption to a mineral surface or whether it will migrate freely as a soluble molecular species.”

#### *More about the importance of the Redox State:*

Choppin and Morgenstern<sup>3</sup> conclude that “in most natural systems plutonium is always found in the +4 and +5 oxidation states.” And that, “while dissolved plutonium can be in either the +4 or +5 redox state, most adsorbed plutonium is found to be in the +4 redox state. The most important property of plutonium with respect to its environmental behavior (mobility) is its oxidation state because solubility, hydrolysis, complexation, sorption and colloid formation reactions differ significantly from one oxidation state to another.”

The +5 and +6 oxidation states have been found to typically be more mobile in groundwater than the +3 or +4 redox states. For example, an 11 year study at the Savannah River Site<sup>10</sup> found that, if the oxidation of plutonium in the environment was not considered, the mobility of the radionuclide would be underestimated by approximately three and a half times. The electrochemistry of plutonium ensures that there will nearly always be several valence states present in equilibrium, resulting in a large fraction of the plutonium being in more mobile valence forms.

#### *Carbonate complexes:*

The EPA<sup>27</sup> notes that, “at pH values above 7, the mobility of plutonium can be influenced by concentrations of dissolved carbonate and hydroxyl ions” and that the “hydroxy-carbonate complexes are among the strongest complexes of plutonium known to exist in the environment. These complexes can inhibit the adsorption of plutonium, and thus increase its mobility in the +4 and +5 oxidation states”. The EPA concludes that “These data suggest that plutonium would be most mobile in high pH carbonate-rich ground waters (like Hanford).”<sup>27</sup>

Carbonate and bicarbonate are common anions present in significant concentrations in many natural water environments (Clark et al. 1995<sup>5</sup>). They are exceptionally strong complexing agents for plutonium and the actinide ions in general. Ions (plutonium) that normally exhibit quite low solubilities in near-neutral solutions can be complexed by carbonate ligands and, through the formation of anionic complexes, become much more soluble. Carbonate complexes have an important role in the migration of plutonium ions from a nuclear waste repository or an unplanned release contamination.

The plutonyl-carbonate system can also be quite complicated in that it consists of several different complex ions in equilibrium with one another and with the aqueous ion or hydrolyzed species, depending on solution conditions. Under dilute solution conditions, compounds of composition  $\text{PuO}_2(\text{CO}_3)_0^0_{(\text{aq})}$ ,  $\text{PuO}_2(\text{CO}_3)_2^{2-}$ , and  $\text{PuO}_2(\text{CO}_3)_3^{4-}$  have all been reported (Clark 2000<sup>4</sup>). These reported compounds all have varying amounts of solubility and mobility in the natural environment.

#### *Organic complexes:*

Dissolved plutonium also forms complexes with many naturally occurring organic ligands such as acetate, citrate, formate, fulvate, humate, lactate, oxalate, and tartrate; as well as with synthetic organic ligands such as EDTA and 8-hydroxyquinoline derivatives.<sup>5</sup> Though the naturally occurring ligands, humate and fulvate are mildly acidic, their principal impact on plutonium mobility is through complexation resulting in neutral or mildly charged dissolved complexes or organic colloids.<sup>4</sup>

Rai, Serne and Moore<sup>18</sup> suggested that the degradation of organic agents originally in the waste will have changed the environmental chemistry of plutonium, probably resulting in inhomogeneous migration of plutonium over time.

#### *About nano-particles and colloidal mobilization:*

Thorsten Schafer<sup>22</sup> reported at the MIGRATION 2009 conference in Kennewick, Washington that it has been demonstrated at several hydrogeological sites in Europe and North America that the mobility of natural or artificially introduced strongly sorbing radionuclides can be enhanced under certain hydrogeochemical conditions by the presence of mobile colloids or the more *en vogue* nanoparticles.

Wilson and Soderholm<sup>29</sup> of the Argonne National Laboratory said “Colloidal metal oxide phases are known to have significant roles in transport and migration of metal contaminants in the environment. Recently published studies have demonstrated the association of plutonium with geochemical colloidal phases is responsible for enhanced transport of plutonium in groundwater systems. Much less understood is the role that eigencolloids (or nanoparticles) of plutonium oxide have in subsurface water transport.”

Soderholm et al<sup>25</sup> also report that plutonium and other light actinides (thorium, uranium and neptunium) naturally and spontaneously form nanometer scale clusters which are negatively charged, stable, fairly soluble, and very mobile. These form the basis of what was formerly called “plutonium polymer” when formed in high concentration from disposed strong acid solutions.

Work at Lawrence Livermore National Laboratory<sup>28</sup> demonstrates that colloid-like nanoparticles in groundwater have been shown to facilitate migration of several radionuclides: plutonium, Cesium, Europium and Cobalt. However, the exact type of nanoparticles and the speciation of the associated radionuclides have remained unknown. These chemical associations with nano-scale particles in the size range <100nm may facilitate transport, and may be responsible for the migration of fissiogenic and actinide elements in groundwater.<sup>18</sup>

Laboratory batch and column experiments at Los Alamos National Laboratory<sup>2</sup> evaluating the generation of calcite colloids and the transport of Pu(VI) by such colloids through saturated alluvium revealed that colloid generation is strongly influenced by flow transects. During the first ~60 days of flushing, as the flow rate increased by a factor of 12, colloid generation increased by a factor of ~6, while increasing the flow rate by a factor of ~3 doubled the colloid generation rate. Results of these experiments indicated that Pu(VI) sorption onto the calcite colloids is strong and almost instantaneous. This in turn suggests a significant potential for colloidal-facilitated transport of Pu(VI) under the hydrogeochemical conditions investigated so far.

Recent field studies by Lawrence Livermore National Laboratories<sup>28</sup> have demonstrated subsurface transport of plutonium over kilometer length scales where the plutonium is associated with colloidal particles and not with the dissolved fraction. Yet, despite these observations and a body of experimental and modeling studies, a comprehensive understanding of the mechanism of colloid-facilitated transport of plutonium remains elusive.

Penrose et al. in a study published in 1990<sup>15</sup> at Los Alamos National Laboratory as part of the DOE’s Subsurface Science Program found that colloidal transport of plutonium was important. Plutonium and americium were found in samples drawn from wells as far as 3.39 kilometers down gradient from the point at which waste was discharged into Mortandad Canyon.

The potential for colloidal transport to affect the mobility of contaminants like plutonium was recognized more than 50 years ago. While the colloid transport pathway has been known for some time, the interest of researchers in the ability for colloids to enhance the mobility of plutonium in the environment was heightened by the discovery that plutonium from at least one nuclear weapons test at the Nevada Test Site had migrated as much as 1.3 kilometers in approximately 30 years<sup>11,12</sup>. Kersting et al., the researchers who reported this discovery, concluded that “models that either predict limited transport or do not allow for colloid-facilitated transport significantly underestimate the extent of radionuclide migration.” Additional experiments have affirmed the conclusion that the mobility of plutonium and other transuranic elements could be much higher than earlier models would have predicted due to adsorption on colloids.<sup>1,6,13,16,21,22,23,25,28,29</sup>

In addition to the findings of Kersting et al. at the Nevada Test Site, Santschi et al.<sup>21</sup> at Rocky Flats found that most of the  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{241}\text{Am}$  transported from contaminated soils to streams occurred in the particulate and colloidal phases. They went on to conclude that colloidal plutonium formation can be one of the most important vectors for enhancing plutonium dispersion at Rocky Flats. Santschi et al.<sup>21</sup> found that at Rocky Flats the mobile plutonium was associated with organic (humic or fulvic) rather than with the more abundant inorganic (iron oxide and clay) colloids and that remobilization of colloid-bound plutonium during soil erosion events was enhanced by the presence of humic and fulvic acids.

The U.S. EPA noted that the oxidation state of dissolved plutonium has itself been found to be dependent on the colloidal organic carbon content in the system.<sup>27</sup>

Choppin and Morgenstern<sup>3</sup> note that the mechanism of the formation of actinide associative colloids has been shown to be closely related to the hydrolysis of the actinide ions” and the strong tendency for plutonium in the +4 oxidation state to undergo hydrolysis thus favors its sorption onto colloidal particles.

Additionally, under the environmental conditions present in Hanford soil waters (circa a pH of 8.5 and redox of 0.5 with substantial carbonate), the expected dominant valence state for plutonium is the more mobile pentavalent V state, rather than the less mobile tetravalent IV state. This increases the proportion of plutonium in the environment that would be expected to be in sparingly soluble anionic carbonate complex forms at solution concentrations well above the appropriate health protection and environmental standards.<sup>10,17,20</sup>

The findings presented above are but a selected few of the many articles and thoughts available from DOE National Laboratories, U.S. University researchers and respected actinide chemists from around the world. The overall opinion that is repeatedly articulated is that plutonium, whether by chemical-compound, valence changes, or attachment to colloids and nanoparticles, is mobile in the natural environment.

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