



**COLORADO**

**Hazardous Materials  
& Waste Management Division**

Department of Public Health & Environment

## Review of potential radiation doses during construction of the Jefferson Parkway

June 2020

## EXECUTIVE SUMMARY

During the Cold War, Colorado's Rocky Flats Plant produced nuclear weapons components for the United States' nuclear arsenal. This included the production of plutonium pits. Unfortunately, environmental releases, fires, and spills occurred throughout its operation. In 1989, the Rocky Flats Plant was added to the U.S. Environmental Protection Agency's National Priorities List of Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA or the "Superfund law") sites needing environmental investigation and remediation.

Today, the Rocky Flats Plant is gone; the CERCLA investigation and cleanup ended in 2006. The Rocky Flats Site entered post-closure and has been divided into three distinct parts:

- (1) the U.S. Department of Energy-retained "Central Operable Unit," the site of the former Rocky Flats Plant, which has been in post-closure since 2006;
- (2) the U.S. Fish and Wildlife Service's Rocky Flats National Wildlife Refuge; and,
- (3) the Jefferson Parkway Authority's narrow transportation corridor, along the west side of Indiana Street.

During the site cleanup, the Refuge and Parkway transportation corridor were part of the "Peripheral Operable Unit," largely the Rocky Flats Plant's former security buffer zone. The cleanup investigation and analysis determined that this area was suitable for unlimited use and unrestricted exposure. That means this land can be used as a wildlife refuge and transportation corridor. While it has long been known small amounts of plutonium from Rocky Flats are present in these areas, and offsite, data and analysis showed these areas met regulatory requirements. The health risk associated with remaining radionuclides is very small. The Colorado Department of Public Health and Environment's Colorado Central Cancer Registry studies have not detected an overall pattern of cancers tied to Rocky Flats. However, interest in Rocky Flats remains strong.

A number of soil sampling events occurred in 2019 in both the U.S. Fish and Wildlife Service's Rocky Flats National Wildlife Refuge and the separate transportation corridor along Indiana Street, which belongs to the Jefferson Public Parkway Highway Authority (JPPHA). Multiple soil sampling events, conducted for different parties, were performed in 2019:

- The U.S. Fish & Wildlife Service tested Rocky Flats National Wildlife Refuge soils along potential trails;
- JPPHA tested soils throughout the Jefferson Parkway transportation corridor;
- Third parties, like Dr. Michael Ketterer and the Rocky Mountain Peace and Justice Center of Boulder, also collected soil for analysis from select areas.

One of the results from one of these sampling events showed a higher than previously recorded level of plutonium in the transportation corridor of 264 pCi/g (picoCuries per gram of soil). All other sampling results were near or below previously detected levels. Nonetheless, the single high result raised questions regarding the potential radiation dose from plutonium that a construction worker and nearby resident could receive during construction of the JPPHA project.

This report details the analysis performed by the Colorado Department of Public Health and Environment (CDPHE) to evaluate the potential radiation dose based on the new soil sampling information from 2019. The following steps were taken during this analysis:

- 1) CDPHE has produced an interactive map of the soil sampling results available on our website at: [Rocky Flats 2019 Soil Sampling Locations](#)
- 2) CDPHE collaborated with Colorado State University to review some of the data and the sampling and analysis plan from the recent sampling events.
- 3) The CDPHE Radiation Program used a regulatory dose assessment tool, called RESRAD (Residual Radioactivity), to perform an assessment of the potential radiation dose from plutonium for a construction worker and nearby resident during the JPPHA project. This tool is specifically designed to help determine the allowable residual radioactivity in site cleanup and has successfully been used in the past to evaluate potential doses from windblown plutonium in the area of Rocky Flats. For more information on RESRAD you can go to: <https://www.evs.anl.gov/research-areas/highlights/resrad.cfm>. Actual data from the recent sampling events, as well as other conservative scenarios, was used in the dose assessment.
- 4) CDPHE prepared a review of select, recent scientific literature on the differences between the risk of exposure to a radioactive particle versus the homogeneous concentration that is used in dose assessments like RESRAD.

#### **Parkway Authority's transportation corridor soil-sampling**

The Parkway Authority's soil samples were collected in numbers and at locations at a 13 to 38 times greater density than the historic soil sampling practices (*Draft Sampling and Analysis Plan, Jefferson Parkway Right-of-Way Rocky Flats National Wildlife Refuge*, Engineering Analytics, Inc., April 2019).

Not every bit of soil must be tested to understand what levels of contamination are or are not present in the environment. Environmental scientists often gather and test soil samples in a grid or spread-out pattern that provides representative data of actual environmental soil conditions. Because plutonium may be unevenly distributed in the environment, it is important to collect a range of samples. Here, that was done. Two different laboratory preparation methods were used to then analyze the soil samples. Data showed a lot of test results within the anticipated range and a single outlier, with a result of 264 pCi/g of plutonium.

It is important to remember since the Plant has been closed and removed, the amount of plutonium in the environment is not expected to have changed since cleanup. However, these soil tests are useful to compare to cleanup test data as a "double check." The data from the many 2019 soil sampling events is consistent with the cleanup investigation and other soil sampling studies.

The statistical methodology and historic soil sampling rationale is presented in the 2002 *Actinide Migration Evaluation Pathway Analysis Summary Report*: "Although an extensive program exists to sample RFETS surface soils for actinides, it is not feasible to collect soil samples from every location at the Site. Therefore, to estimate actinide concentrations in soils at locations that have not been sampled, it is necessary to use data from adjacent locations that have been sampled. Various computerized estimation techniques have been developed for this purpose."

This data was subsequently published in 2007 by the members of the Actinide Migration Panel in the *Journal of Alloys and Compounds* (Clark et al, 2007). This figure below visually illustrates the expected ranges of soil sample results based on geostatistical modelling and the recent Parkway soil samples appear to generally validate the projected modelled results along the eastern side of this figure. Note: this figure is a depiction of pre-remediation conditions; the former 903 Pad and Lip Area are not reflective of current site conditions.

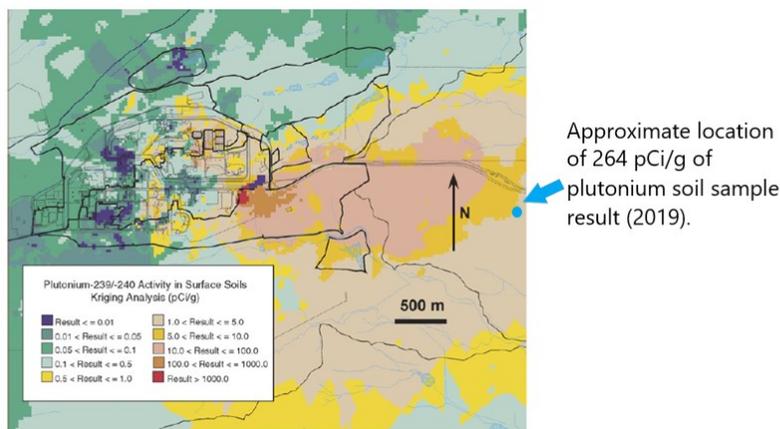


Fig. 3. Because it was not feasible to sample surface soil at every location, and geostatistical modeling technique known as Kriging was applied to the plutonium surface soil data for  $^{239/240}\text{Pu}$  to estimate concentrations in surface soil. The “hot spot” of  $^{239/240}\text{Pu}$  concentrations in excess of 1000 pCi/g at the 903 Pad is shown in red. A clear plume of  $^{239/240}\text{Pu}$  contamination that tracks roughly with the prevailing winds from NW to SE is evident from the data. This figure represents conditions at Rocky Flats prior to soil remediation actions.

Credit: Clark, D.L., Choppin, G.R., Dayton, C.S., Janecky, D.R., Lane, L.J. and Paton, I., 2007. Rocky Flats closure: The role of models in facilitating scientific communication with stakeholder groups. *Journal of alloys and compounds*, 444, pp.11-18.

### Review of data generated from recent soil-sampling

Dr. Ralf Sudowe, Associate Professor, in the Department of Environmental & Radiological Health Sciences at Colorado State University provided an *Evaluation of Sampling, Analytical Procedures and Results* relating to the Jefferson Parkway sample set.

Jefferson Parkway soil sampling was performed along the Jefferson Parkway transportation corridor, an approximate 2.5 miles by 300-foot area along the eastern border of the U.S. Rocky Flats National Wildlife Refuge. Laboratory analysis of soil samples was performed to determine the concentration of radionuclides present in the soil at the transportation corridor, where highway-related construction is expected to occur. Soil sampling was performed in May 2019 (Phase 1), June 2019 (Phase 2), and August 2019 (Step-Out Samples). A total of 467 soil samples from the Parkway footprint were analyzed. This number includes field duplicates, samples taken at two different depth intervals below the surface as well as samples processed using two different digestion methods, acid leaching and carbonate fusion, respectively.

ALS Global Laboratories analyzed the soil samples for americium-241, isotopic plutonium, and isotopic uranium using alpha spectroscopy. Analyses were performed at their facility in Fort Collins, Colorado. The soil samples were prepared, or digested, using two different methods. As

a result of applying two different digestion methods, two results were obtained for each sample.

The two different digestion methods yielded similar results, providing no evidence for the presence of refractory plutonium (which refers to plutonium that was exposed to a high-fired or high-temperature environment). The highest  $^{239/240}\text{Pu}$  activity concentration of 264 pCi/g was found in a soil sample taken at sampling point 765+00, collected on June 14, 2019, at a depth of 0-2 inches below the surface. This data was reported to the CDPHE on August 16, 2019. The corresponding archived sample was analyzed, and a  $^{239/240}\text{Pu}$  activity concentration of 1.52 pCi/g was measured. As a result of this elevated sample result found during Phase 2 of the sampling effort, additional step-out sampling was performed on August 8, 2019 in a 20-foot spaced grid pattern, creating a box of 25 sample points around the 765+50 sample location which had previously shown the highest activity. The  $^{239/240}\text{Pu}$  activity concentration in the samples that were digested using the acid leaching technique ranged from 0.17 - 2.25 pCi/g and the carbonate technique ranged from 0.105 - 2.9 pCi/g.

Other entities also conducted soil sampling in 2019, like the U.S. Fish and Wildlife Service. For brevity, not every report is discussed in detail in this summary. Of note, however, in 2019, Dr. Michael Ketterer, the Rocky Flats Downwinders, and Rocky Mountain Peace and Justice Center collected a select number of soil samples in the Parkway transportation corridor. This information was shared with the public and CDPHE. Dr. Ketterer conducted additional analyses in 2020. In a letter dated April 24, 2020, Dr. Michael Ketterer declined to provide additional information to CDPHE regarding his and the Rocky Mountain Peace and Justice Center's 2020 Parkway and offsite soil sampling project. However, Dr. Ketterer shared some of his conclusions. In particular, Dr. Ketterer reported his soil testing results "are well below the 50 pCi/g  $^{239+240}\text{Pu}$  remedial standard..."

Dr. Ketterer first presented 2019 soil sample results in an *Interim Report* to Dr. LeRoy Moore with Rocky Mountain Peace and Justice, dated September 10, 2019. The September report explained July 2019 sampling and results from six soil collection locations in the Parkway transportation corridor area. Findings of this report are a "consistent baseline  $^{239+240}\text{Pu}$  activity of 4.76 pCi/g." Subsampling "enabled the detection of  $\text{PuO}_2$  particles ranging from 3.2 to 109 pg, having equivalent diameters of 0.82 to 2.62  $\mu\text{m}$ ."

In summary, results of soil sample analyses confirm that the majority of plutonium is associated with the top 2 inches of the surface soil. Apart from the one Parkway transportation corridor result of 264 pCi/g of plutonium in surface soil, no other reported soil samples exceeded the 50 pCi/g of plutonium historical cleanup action level.

### **Radiation dose assessment**

The CDPHE Radiation Program used RESRAD, a regulatory dose assessment tool, to perform an assessment of the potential radiation dose from plutonium for a construction worker and a nearby resident during the JPPHA project. This tool is specifically designed to help determine the unacceptable levels of residual radioactivity in site cleanup and has successfully been used in the past to evaluate potential doses from windblown plutonium in the area of Rocky Flats. The highest recorded sample result from the recent sampling events (264 pCi/g) and other conservative assumptions were used in the dose assessment. The highest calculated dose was to

a construction worker - 11.52 millirem/year. The highest calculated dose to a nearby resident was about 2.6 millirem/year.

Comparing the results of the RESRAD evaluation to regulatory standards, the radiation control program would not consider the potential doses to the road workers or the nearby residents as a result of these activities to be an undue hazard to public health from a radiologic hazard perspective. From a regulatory perspective, two standards regarding public dose were considered. A limit of 25 millirem per year for an individual exposed to a site that has been released for unrestricted use, as well as a 100 millirem per year dose limit for exposures to licensed operations. The calculated potential doses, even in the most extremely conservative worst case scenario, are significantly less than these standards.

Specifically, the Radiation Program used RESRAD to model a “worst-case” scenario where soil was presumed to contain an activity concentration of 264 pCi/g of plutonium down to a depth of 2 meters. Even in this extremely conservative scenario, calculated radiation doses are well below levels that would indicate a need for oversight.

Considering the potential dose to members of the public from this activity in relation to the regulatory thresholds that dictate the lawful requirements applicable to situations that involve radioactive materials, the radiation control program would not require a license for these activities.

### Literature review

The Colorado Department of Public Health and Environment performed a literature review of peer-reviewed research concerning questions of interest about plutonium associated with the Rocky Flats Site) and the proposed Jefferson Parkway Public Highway Authority transportation corridor project. Specifically, we looked at how the size, shape, and distribution of plutonium in the environment may or may not impact human health. We also looked at how plutonium might enter the human body, and how it behaves once inside a person’s body. This was part of a larger effort where CDPHE consulted outside experts and academics about radiation; performed additional radiation dose modeling to check whether construction is safe; and, reviewed recent plutonium soil sampling data from 2019.

The historic analysis published in a *Health Physics* article authored by McDowell and Whicker (1978) provide reporting of PuO<sub>2</sub> characteristics that correlate with Dr. Ketterer’s findings. McDowell and Whicker sized 1,700 particles with mean PuO<sub>2</sub> equivalent diameters ranging from 0.20, 0.25, and 0.2 μm. Follow on work using a different method to scan particles resulted in the largest particle size with a 6.86 μm equivalent diameter.

Dr. David Wood is an interested area resident and physicist. Several articles referenced in Dr. Wood’s white papers and memos are summarized in this literature review (primarily Dr. Emily Caffrey et al. article from 2017, Charles and Harrison, 2007, Burkart and Linder, 1987, and Harrison, 2003). Many of the articles Dr. Wood evaluated were also considered as part of this literature review.

Together, these efforts paint a consistent picture: remaining Rocky Flats plutonium in the Jefferson Parkway transportation corridor and offsite poses a small risk, well within regulatory limits for radiation. This conclusion is consistent with previous findings and the cleanup process.

Attachments:

1. Evaluation of Sampling, Analytical Procedures and Results relating to the Jefferson Parkway Sample Set, Dr. Ralf Sudowe, CSU
2. RESRAD Assessment of Potential Radiological Dose Resulting from Road Work Activities at the Jefferson Parkway, James Grice, CDPHE
3. Rocky Flats Literature Review, CDPHE

# Evaluation of Sampling, Analytical Procedures and Results relating to the Jefferson Parkway Sample Set

**Dr. Ralf Sudowe**

Associate Professor

Department of Environmental & Radiological Health Sciences  
Colorado State University

## 1. Sampling

All sampling was conducted in accordance with the Sampling and Analysis Plan (SAP), Jefferson Parkway Right-of-Way Rocky Flats National Wildlife Refuge, April 2019 Rev 0.0, prepared by Engineering Analytics, Inc. for the Jefferson Parkway Public Highway Authority and Terracon Consultants, Inc.

The goal of the SAP was to generate soil sample results that meet standard quality requirements and that would allow comparison to the risk assessment values used at the site. For this purpose, a method to collect soil samples and a depth(s) at which to collect the soil samples was selected that would result in radionuclide data that could be compared to historic data. Based on previous experience, Engineering Analytics, Inc. used the Visual Sample Plan (VSP) software package, developed by Pacific Northwest National Laboratory (PNNL), to estimate the sample density that was required to fulfill the project's objective. VSP is a software tool that supports the development of a defensible sampling plan based on statistical sampling theory and the statistical analysis of sample results to support confident decision making. VSP incorporates a variety of sampling designs, most importantly those described in the Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM), in its programming. The MARSSIM manual provides detailed guidance on how to demonstrate compliance with dose or risk-based regulations or standards. It is a multi-agency consensus document that was developed collaboratively by four Federal agencies having authority and control over radioactive materials: Department of Defense (DOD), Department of Energy (DOE), Environmental Protection Agency (EPA), and Nuclear Regulatory Commission (NRC). The MARSSIM's objective is to describe a consistent approach for planning, performing, and assessing surface soil final status surveys to meet established dose or risk-based release criteria.

The sampling of soils was performed along the Jefferson Parkway Right-of-Way (ROW), an approximate 2.5 miles by 300-foot area along the eastern border of the Rocky Flats National Wildlife Refuge. Sampling was performed to determine the concentration of radionuclides present in the soil at the ROW, where highway-related construction is expected to occur. Soil sampling was performed in May 2019 (Phase 1), June 2019 (Phase 2), and August 2019 (Step-Out Samples).

As part of Phase 1, soil samples were collected from 20 locations positioned at approximately equal distances along the ROW at depth intervals from 0-2 inches and 6-8-inches below the surface. During Phase 2, soil samples were collected at random locations at a frequency of 1 sample per 0.3 acre. During this phase, samples were taken only at a depth of 0-2 inches below the surface. When the step-out samples were collected in August 2019, only one sample taken at a depth of 6-8 inches was analyzed, all other step out samples were collected from a depth of 0-2 inches below the surface. The decision to only sample the soil to a depth of 2 inches was based on studies in the peer-reviewed literature that show that plutonium in the form of plutonium dioxide is insoluble and typically retained within the top two inches of undisturbed surface soil.

At each sampling location, the personnel involved in the sampling removed vegetation and large materials (i.e. rocks) from an approximate 4x4-inch area. The soil was collected with a clean, stainless steel tool from the surface down to a depth of 2 inches. Care was taken to retain any soil that was attached to plant roots. The soil was sieved in the field using a Number 10 (2 mm) stainless steel sieve to further remove any coarse-grained material. The soil was subsequently mixed thoroughly in a stainless-steel bowl, divided into three approximately equal parts, and a sub-sample of approximately 200 to 300 grams was placed in a new and certified clean container for laboratory analysis. A second portion of approximately the same mass was placed in the same type of container and archived. After the initial sample was collected from a depth of 0-2 inches, the hole was extended to a depth of 6 inches. Any loose soil was removed from the base of the hole, and a second soil sample was collected from an approximate 4x4-inch area started at a depth of 6 inches going down to a depth of 8 inches. The soil samples taken from a greater depth were processed in the same manner as those taken from a depth of 0-2 inches. All samples were then hand delivered to the analysis laboratory under a completed and signed chain-of-custody record.

## **2. Analytical Procedure**

The soil samples were analyzed for americium-241, isotopic plutonium and isotopic uranium using alpha spectroscopy by ALS Global Laboratories (ALS) at their facility in Fort Collins, Colorado.

ALS is a commercial analytical laboratory that provides a wide range of environmental testing capabilities, including radiochemical testing. The laboratory holds a current Radioactive Materials Handling License and is certified under the National Environmental Laboratory Accreditation Program (NELAP) according to their 2009 TNI Standard through the State of Utah Department of Environmental Quality. In addition, it is also accredited with the United States Department of Energy Consolidated Audit Program-Accreditation Program (DOECAP-AP) according to ISO/IEC 17025:2005 "General Requirements for the competence of Testing and Calibration Laboratories" and the DOE Quality Systems Manual for Environmental Laboratories Version 5.1.1, February 2018. ALS participates in performance evaluation tests or proficiency tests for alpha, beta and gamma emitting radionuclides provided by Waters ERA at least twice a year.

The soil samples were analyzed in a destructive manner. Upon receipt and transfer of custody of the project samples from EA to ALS, the samples were logged into a Laboratory Information Management System (LIMS). The field samples were then processed according to ALS SOP 736, Representative Laboratory Subsampling – Radiochemistry. This included drying the samples at  $105\pm 5^{\circ}\text{C}$  for a minimum of 16 hours and then milling the dried soil by machine-shaking two approximately 60-gram aliquots in separate half-pint steel canisters each containing 5 half-inch steel ball bearings for 15 minutes. This milling process produces a 200-mesh powder. The powdered soil from both containers was combined and thoroughly mixed to produce aliquots for subsequent processing and analysis. At this point, splits of each sample were created and packaged for third-party analysis and archival purposes. The samples were subsequently digested using two different methods. The first one consisted of acid dissolution of the sample in a mixture of nitric acid and hydrofluoric acid with an optional second step involving these acids and additional hydrochloric acid (ALS SOP 773 - Total Dissolution of Solids for the Radiochemical Determination of Actinides and other Non-Volatile Radionuclides). The second dissolution method involved fusing the sample in a mixture of equal parts of boric acid, potassium carbonate, and sodium carbonate at high temperatures after an initial treatment with hydrofluoric acid (ALS SOP 768 – Preparation of Soil Samples by Sodium Carbonate Fusion). The second digestion method has the advantage of being able to dissolve high-fired, refractory plutonium. However, it is limited by the amount of material that can be digested (1 gram) compared to the 2-10 grams of sample that can be used for the acid dissolution. This can result in a

higher Minimum Detectable Activity for the samples. As a result of applying two different digestion methods, two results were obtained for each sample. After the dissolution, americium, plutonium, and uranium were initially separated from the digested/fused samples using ion exchange (ALS SOP 778 - Uranium, Plutonium and Americium/Curium (Partial) Sequential Separation by Ion Exchange). Any americium-241 present in the samples was then further purified using extraction chromatography with proprietary resins available from Eichrom Technologies, Inc. (ALS SOP 751 - Actinides – Americium/Curium Separation - Purification by TRU™ and TEVA™ Spec Column). After the separation, the purified radioisotopes were co-precipitated with lanthanum fluoride. The precipitate was filtered and retained on a suitable membrane, which was subsequently mounted on a planchet for quantification by alpha spectroscopy. The mounted samples were then assayed by alpha spectroscopy with solid-state detectors together with method blanks and laboratory control samples according to ALS SOP 714 - Analysis of Alpha Emitting Radionuclides by Alpha Spectroscopy. ALS reported the test results as both an electronic data deliverable (EDD) format (MS Excel spreadsheet) and a printable PDF laboratory report. The laboratory report included a quality control (QC) package consisting of case narrative, receipt documentation and chain-of-custody, individual sample results, and laboratory QC sample results for blanks, duplicates, and laboratory control samples.

All Standard Operation Procedure (SOPs) used by ALS for processing and analyzing the samples are based on established, validated methods and published standards provided by ASTM International, the Environmental Measurements Laboratory and the U.S. Environmental Protection Agency. The quality assurance and quality control methods applied by ALS were consistent with current industry standard.

### 3. Results

A total of 467 soil samples from the Parkway footprint were analyzed. This number includes field duplicates, samples taken at two different depth intervals below the surface as well as samples processed using two different digestion methods, acid leaching and carbonate fusion, respectively. Most of these samples were collected during Phase 2 of the sampling effort.

During Phase 1, a total of 47 soil samples were collected. Twenty-five of these samples were collected at a depth of 0-2 inches below the surface, including five field duplicates. The  $^{239/240}\text{Pu}$  activity concentrations in the samples that were digested using an acid leaching technique ranged from 0.008 – 1.36 picocuries per gram (pCi/g) with an average of 0.469 pCi/g. The  $^{239/240}\text{Pu}$  activity concentrations in the samples that were digested using a carbonate technique ranged from 0.003 – 2.61 pCi/g with an average of 0.536 pCi/g. While some of the sample results obtained for samples with carbonate fusion were greater than the results for the corresponding sample treated with acid leaching, other samples showed a lower activity concentration. There is no clear overall trend evident between the two digestion methods that would indicate the presence of refractory plutonium. Twenty-one samples were collected at a depth of 6-8 inches below the surface, two of which were field duplicates. The  $^{239/240}\text{Pu}$  activity concentrations in the samples that were digested using an acid leaching technique ranged from 0.008 – 0.54 pCi/g with an average of 0.096 pCi/g. The  $^{239/240}\text{Pu}$  activity concentrations in the samples that were digested using a carbonate technique ranged from 0.001 – 0.76 pCi/g with an average of 0.109 pCi/g. These results confirm that the majority of the plutonium is associated with the top 2 inches of the surface soil. Again, the two different digestion methods yielded similar results, providing no evidence for the presence of refractory plutonium.

During Phase 2, a total of 318 soil samples taken at a depth of 0-2 inches below the surface were analyzed. Out of these sample results, 102 were obtained from samples processed with the acid leaching digestion. The highest  $^{239/240}\text{Pu}$  activity concentration of 264 pCi/g was found in a soil sample

taken at sampling point 765+00, collected on June 14, 2019, at a depth of 0-2 inches below the surface. This sample also exhibited the highest  $^{241}\text{Am}$  activity concentration of 32.2 pCi/g, indicating the presence of the beta-emitting radioisotope  $^{241}\text{Pu}$ . The corresponding archived sample was analyzed, and a  $^{239/240}\text{Pu}$  activity concentration of 1.52 pCi/g was measured. The  $^{239/240}\text{Pu}$  activity concentrations in the remaining samples that were digested using the acid leaching technique ranged from 0.011 – 2.06 picocuries per gram (pCi/g) with an average of 0.419 pCi/g. A total of 216 sample results were obtained from samples digested using the carbonate fusion technique. The sample with the highest  $^{239/240}\text{Pu}$  activity concentration in this sample set was collected at sampling location 777+30 on June 17, 2019 at a depth of 0-2 inches below the surface. It had a  $^{239/240}\text{Pu}$  activity concentration of 11.3 pCi/g. The overall  $^{239/240}\text{Pu}$  activity concentrations in the samples that were digested using a carbonate technique ranged from 0.002 – 11.3 pCi/g with an average of 0.757 pCi/g.

As a result of this elevated sample result found during Phase two of the sampling effort, additional step-out sampling was performed on August 8, 2019 in a 20-foot spaced grid pattern, creating a box of 25 sample points around the 765+50 sample location which had previously shown the highest activity. The samples were taken at a depth of 0-2 inches below the surface and three field duplicates were collected as well. Each step-out sample was again processed using both the acid leaching and the carbonate fusion technique, resulting in a total of 56 analytical results. The  $^{239/240}\text{Pu}$  activity concentration in the samples that were digested using the acid leaching technique ranged from 0.17 – 2.25 pCi/g with an average of 0.925 pCi/g. The  $^{239/240}\text{Pu}$  activity concentration in the samples that were digested using a carbonate technique ranged from 0.105 – 2.9 pCi/g with an average of 0.856 pCi/g.

## Memorandum

TO: Jennifer Opila, Director, Hazardous Materials and Waste Management Division, Colorado Department of Public Health and Environment

FROM: James Grice, Radiation Control Program Manager, Hazardous Materials and Waste Management Division, Colorado Department of Public Health and Environment

DATE: 4/23/2020

SUBJECT: RESRAD Assessment of Potential Radiological Dose Resulting from Road Work Activities at the Jefferson Parkway

*Please note that this document is not intended to be a complete risk assessment but rather a radiological dose assessment specific to road workers and offsite residents as a result of anticipated road work activities at the Jefferson Parkway. This assessment is being performed for the purposes of making a regulatory determination regarding program oversight of activities and the potential necessity for radioactive materials licensing.*

## Background

The radiation control program has statutory authority to issue licenses pertaining to radioactive materials. Requirements regarding the licensing of radioactive materials are generally found within the rules pertaining to radioactive materials and based on potential hazard to public health and safety or property. The potential radiological exposure hazard to members of the public is evaluated when considering radioactive materials. The basis for restrictions and requirements within the regulations is a limitation on potential radiological exposure or dose to individual members of the public.

When a situation exists involving radioactive materials and the potential for radiological dose to members of the public, the radiation control program evaluates and assesses that potential and makes decisions on the application of controls or requirements to ensure that no undue hazard shall result.

The radiation control program may use multiple methods to evaluate a situation from a radiation safety or health physics perspective to assess the radiological hazard or potential radiological exposures. These methods may come in the form of regulatory requirement applications, scientific and regulatory guidance and reference document review, computer model use, which use health physics calculations to evaluate potential doses, or other manual health physics calculations performed directly by staff members. When situations call for a site specific type evaluation of a complex system, computer models are typically employed.

The RESRAD computer models, developed at Argonne National Laboratory, were developed to analyze potential human, animal and plant radiation exposures from the environmental contamination of residual radioactive materials. The computer models use pathway analysis to evaluate radiation exposure and associated risks, and to derive cleanup criteria or authorized limits for radionuclide concentrations in the contaminated source medium. The RESRAD computer models are widely used by regulatory agencies, the risk assessment community, and universities in more than 100 countries around the world for these purposes. RESRAD-OFFSITE is a computer model used to assess radiation exposures of a human receptor located on top of or at some distance from soils contaminated with radioactive materials

while RESRAD-ONSITE strictly assesses radiation exposures of a human receptor located on top of soils contaminated with radioactive materials.

## Jefferson Public Parkway Highway Authority (JPPHA)

In 2019, the Jefferson Public Parkway Highway Authority (JPPHA) began soil sampling in advance of a construction project within the transportation corridor along Indiana Street adjacent to the U.S. Fish and Wildlife Service’s Rocky Flats National Wildlife Refuge. One of the results from these sampling events showed a higher than previously recorded level of plutonium for the transportation corridor. This result raised questions regarding the potential radiation dose from plutonium that a construction worker and nearby resident could receive during construction of the JPPHA project.

After this finding, the radiation control program performed an assessment of the potential hazard regarding radiological exposure, or dose, as a result of roadwork activities that are planned for that area.

## RESRAD Assessment Parameters

The radiation control program used both of the RESRAD models mentioned above to assess potential radiological exposure to road workers as well as off-site residents as a result of these activities.

RESRAD models allow for certain site specific parameters to be utilized within the model to provide the best estimate of potential radiological exposure.

The tables below provide a summary of the parameters used within the models during this assessment.

RESRAD-ONSITE was used to evaluate potential dose to road workers during the project.

RESRAD-ONSITE Input Parameter	Value	Unit	Source/Comment
Pathways	External Gamma Inhalation Soil Ingestion	NA	These are the potential exposure routes that would apply to a road worker scenario
Soil Concentrations	264 or 5	picoCuries per gram	Maximum value from Phase 2 sampling or Exposure Point Concentration
Initial principal radionuclide	Pu-239	NA	Plutonium is the radioactive material of concern
Calculation Times	1, 3, 5	years	Project expected to be completed within 5 years
Area of Contaminated Zone	407628	square meters	Approximately 2.77 miles by 300 feet
Wind Speed	4.20218	meters per second	CDPHE Technical Services Program, Air Pollution Control Division - Rocky Flats North sample point 2019 hourly wind speed. Average of 8607 records
Precipitation	0.4	meters per year	Colorado average (Nationalatlas.gov)
Inhalation Rate	9782	cubic meters per year	RESRAD Data Collection Handbook, September 2015 - TABLE 7.2.4 Reference Values for Inhalation Rates of Sedentary and Heavy Workers, ICRP (2002) --- Heavy Worker value used
Mass loading for inhalation	0.00015	grams per cubic meter	40CFR50.6 National primary and secondary ambient air quality standards for PM10.

RESRAD-ONSITE Input Parameter	Value	Unit	Source/Comment
Exposure Duration	5	years	Project expected to be completed within 5 years
Indoor time fraction	0		All work hours outdoors
Outdoor time fraction	0.2283		2000 hours per year
Soil Ingestion	73	grams per year	EPA Exposure Factors Handbook, EPA/600/R-17/384F, September 2017, Table 5-1. Recommended Values for Daily Soil, Dust, and Soil + Dust Ingestion, Soil + Dust General Population Upper Percentile (200 mg/day x 365 days/year x 1/1000 grams/milligram= 73 grams per year)

RESRAD-OFFSITE was used to evaluate potential dose to hypothetical adult and youth (infant and ages 1, 5, 10, and 15) offsite resident farmers residing directly adjacent to the work area during the project.

RESRAD- OFFSITE Input Parameter	Value	Unit	Source/Comment
Pathways	External Gamma Inhalation Soil Ingestion Plant Ingestion Meat Ingestion* Milk Ingestion Aquatic Foods* Drinking Water		All exposure routes are considered for residential scenario. *The Meat and Aquatic Food ingestion pathways were not activated for the infant assessment.
Soil Concentrations	264 or 5	picoCuries per gram	Maximum value from Phase 2 sampling or Exposure Point Concentration
Initial principal radionuclide	Pu-239	na	
Calculation Times	1, 3, 5	years	Project expected to be completed within 5 years
Area of Contaminated Zone	407628	square meters	2.77 miles by 300 feet
Precipitation	0.4	Meters per year	Colorado average (Nationalatlas.gov)
Inhalation Rate	Adult - 8979 Infant - 2591.5 Age 1 - 2920 Age 5 - 5037 Age 10 - 6059 Age 15 - 7993.5	Cubic Meters per year	EPA Exposure Factors Handbook, PA/600/R-09/052F, September 2011, Chapter 6, Table 6-1. Recommended Long-Term Exposure Values for Inhalation (males and females combined) 95th Percentile (Adult 24.6 cubic meters per day x 365 days/year = 8979 cubic meters per year) (Infant--7.1 cubic meters per day x 365 days/year = 2591.5 cubic meters per year // Age 1-- 8 cubic meters per day x 365 days/year = 2920 cubic meters per year // Age 5-- 13.8 cubic meters per day x 365 days/year = 5037 cubic meters per year // Age 10-- 16.6 cubic meters per day x 365 days/year = 6059 cubic meters per year // Age 15-- 21.9 cubic meters per day x 365 days/year = 7993.5 cubic meters per year)
Mass loading for inhalation	0.00015	grams per cubic meter	40CFR50.6 National primary and secondary ambient air quality standards for PM10.
Exposure Duration	5	Years	Project expected to be completed within 5 years
Soil Ingestion	73	grams per year	EPA Exposure Factors Handbook, EPA/600/R-17/384F, September 2017, Table 5-1. Recommended Values for Daily Soil, Dust, and Soil + Dust Ingestion, Soil + Dust General Population Upper Percentile (200 mg/day x 365 days/year x 1/1000 grams/milligram= 73 grams per year)

When parameters are not specifically listed in the above tables it means the default values within the RESRAD model were used.

The values for parameters listed within the tables were based on site specific conditions, established information where applicable, and regulatory requirements. The remaining parameters were established by selecting highly conservative values from a range of recommended values developed and produced by the U.S. Environmental Protection Agency as well as the U.S. Department of Energy Argonne National Laboratory Environmental Science Division. These previously established values were developed to be used in risk assessments and in some cases the RESRAD computer models specifically.

Finally, the parameter that is most specific to this particular situation is the concentration of plutonium within the soils at the site. Two different approaches were used regarding this particular parameter.

In an effort to provide a worst case scenario type assessment, the first approach was to run the model with a soil concentration of 264 picoCuries per gram of plutonium. This provides an extremely conservative estimate of potential dose in the case and assumes that all of the soil at the site was equivalent to the concentration found in the one elevated sample in 2019. Past sampling results, as well as those established subsequent to the elevated finding, would indicate that this is not the case but this was considered in order to provide context for a worst case scenario.

The models were run also with the more commonly used method of establishing an Exposure Point Concentration (EPC) which is the 95% Upper Confidence Limit (UCL) on the mean concentration of plutonium found in the surface soil collected. This means that there is only a 5% chance that the actual mean concentration of plutonium in soil is higher than the EPC value. This is also considered to be a conservative assumption.

## RESRAD Model Results

The results of the models for each plutonium concentration are presented below. These results are the predicted potential radiological dose values for the individuals potentially impacted by the activities.

Plutonium Concentration at 264 pCi/g

Individual	Result (mrem/year)
Road worker	11.52
Offsite resident (Adult)	1.977
Offsite resident (Youth Max)	2.589

Plutonium Concentration at 5 pCi/g

Individual	Result (mrem/year)
Road worker	0.2183
Offsite resident (Adult)	0.03745
Offsite resident (Youth Max)	0.04903

## Conclusion

As a result of the evaluation, the radiation control program would not consider the potential doses to the road workers or the nearby residents as a result of these activities to be an undue hazard to public health from a radiologic hazard perspective.

From a regulatory perspective, two standards regarding public dose were considered. A limit of 25 millirem per year for an individual exposed to a site that has been released for unrestricted use, as well as a 100 millirem per year dose limit for exposures to licensed operations. These potential doses, even in the most extremely conservative worst case scenario, would be significantly less than these standards as demonstrated in the tables below.

Plutonium Concentration at 264 pCi/g

Individual	Result (mrem/year)	% of dose criteria for unrestricted use	% of public dose limit for licensed operations
Road Worker	11.52	46.08%	11.52%
Offsite Resident (Adult)	1.977	7.91%	1.98%
Offsite Resident (Youth Max)	2.589	10.36%	2.59%

Plutonium Concentration at 5 pCi/g

Individual	Result (mrem/year)	% of dose criteria for unrestricted use	% of public dose limit for licensed operations
Road Worker	0.2183	0.87%	0.22%
Offsite Resident (Adult)	0.03745	0.15%	0.04%
Offsite Resident (Youth Max)	0.04903	0.20%	0.05%

Considering the potential dose to members of the public from this activity compared to the regulatory thresholds that would indicate a need for oversight of situations involving radioactive materials, the radiation control program would not require a license for these activities.

## SUMMARY

The Colorado Department of Public Health and Environment (CDPHE) performed a literature review of peer-reviewed research concerning questions of interest about plutonium associated with the Rocky Flats Environmental Technology Site (Rocky Flats) and the proposed Jefferson Parkway Public Highway Authority transportation corridor project. Specifically, we looked at how the size, shape, and distribution of plutonium in the environment may or may not impact human health. We also looked at how plutonium might enter the human body, and how it behaves once inside a person's body. This was part of a larger effort where CDPHE consulted outside experts and academics about radiation; performed additional radiation dose modeling to check whether construction is safe; and, reviewed recent plutonium soil sampling data from 2019. Together, these efforts paint a consistent picture: remaining Rocky Flats plutonium in the Jefferson Parkway transportation corridor and offsite poses a small risk, well within regulatory limits for radiation. This conclusion is consistent with previous findings and the cleanup process.

## PURPOSE

The statement of the purpose of the literature review is to “prepare a review of scientific literature on the differences between the risk of exposure to a radioactive particle versus the homogeneous concentration that is used in dose assessments like RESRAD.” (November 5, 2019 email). RESRAD refers to the RESRAD family of codes as “developed at Argonne National Laboratory to analyze potential human and biota radiation exposures from the environmental contamination of RESidual RADioactive materials”, from the website <https://resrad.evs.anl.gov/>. RESRAD is mentioned here as a component of the wider CDPHE approach, however the literature review focused on empirical data.

## CRITERIA AND PARAMETERS

The development of CDPHE's purpose was informed by certain findings of a 2017 article authored by Emily Caffrey, PhD and others, *Comparison of homogeneous and particulate lung dose rates for small mammals*, which examined the use of averaging radiation dose over an entire organ or tissue volume. This article considered the effects of hot particles above a certain diameter and determined that for large particles a self-shielding effect in the lungs for small mammals can result in a reduced dose rate to surrounding tissue. As a result, the questions of whether or not there were other equivalent peer-reviewed journal articles or bodies of research that considered this concept, arrived at similar or converse conclusions, or considered larger mammalian and/or human receptors became one focus of the literature review.

Based on Caffrey's line of inquiry and CDPHE's stated goal, the literature review was also structured to determine if there was any recent research that could provide perspective on the activities detected from the soil sampling along the proposed Jefferson Parkway transportation corridor.

## DISCUSSION OF SELECT ARTICLES AND PUBLICATIONS

Between December 2019 and March 2020, CDPHE performed a literature review to evaluate current research across the world and published articles concerning radioactive particles, their behavior in the environment and potential human health impacts. CDPHE conducted this literature review to help assess and respond to concerned citizens' questions regarding recent soil sampling from land once a part of the Rocky Flats Environmental Technology Site, U.S. Department of Energy (DOE), which is now the proposed Jefferson Parkway transportation corridor along Indiana Street. This literature review is

one part of a multi-pronged Jefferson Parkway review project by CDPHE. CDPHE has also reviewed Jefferson Parkway and Rocky Flats National Wildlife Refuge radionuclide soil sampling data and produced a map of the area with the soil results posted on this page <https://www.colorado.gov/pacific/cdphe/rocky-flats>, called 'Interactive GIS soil sample locations map, January 14, 2020'; consulted with academic radiation experts from Colorado State University; and, performed a "worst-case" RESRAD dose modeling analysis to determine possible radiation doses to workers and area residents in downwind communities.

This literature review was limited in scope. The literature review examined documents published from between 2010 through 2019, with some additional historical documents ranging from 1978 to 2009 reviewed for context and comparison. The literature review focused on inhalation as the primary exposure pathway for potential human health effects. In addition, CDPHE reviewed literature describing the ingestion exposure pathway. When evaluating inhalation as the primary exposure route, the size of radioactive particles is the foremost consideration. Generally, the larger the particle, the less likely it is to be available for air suspension and inhaled by a human. Smaller particles can be inhaled, but are usually coughed and expelled out of the body. Only very small particles can penetrate deep into the lung and becoming lodged. The effect of plutonium on human health, as affected by size, has been studied from different perspectives and this review provides a summary of parts of health physics and environmental journals, and other periodicals.

This review only considered peer-reviewed journal articles and government technical reports. CDPHE does not have access to academic databases so Google Scholar was used for most research supplemented by suggestions from professors in the Environmental & Radiological Health Sciences Department at Colorado State University. A query for 'Rocky Flats, Colorado plutonium' yielded 4,650 articles that range in dates starting in the 1960s through the present. This literature review did not encompass all published journals, but narrowly focused on research from the last decade (approx. 2010 to 2019). This review sought to capture authors considered as technical experts on the subject and/or with familiarity with Rocky Flats cleanup. Some historic journal publications were reviewed as needed to provide context. While over 100 articles were considered as part of this review, only 19 published articles are summarized below.

Much of the reviewed research studied plutonium exposures via inhalation and the effects of a *pure* plutonium oxide ( $\text{PuO}_2$ ) airborne particle on the lungs. The scenario in the construction of Jefferson Parkway across former land from the Rocky Flats Environmental Technology Site is more likely airborne particle(s) of plutonium oxide sorbed to a piece of soil or 'plutonium laden' aerosol, that would be disturbed and resuspended in the regrading effort for the initial construction of the parkway. The soil particle diameter (measured in microns ( $\mu\text{m}$ )) would need to be respirable (which for the purpose of this review, will be defined as anything less than 10  $\mu\text{m}$ ).

Based on the articles and publications reviewed, this literature review identified three main concepts related to the purpose and question of this review:

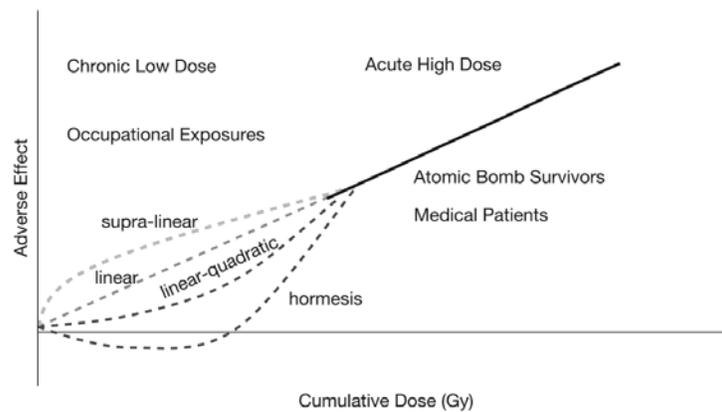
- 1) linear non-threshold (LNT) model approach and dosing;
- 2) generic particle characteristics and biological exposure; and
- 3) historic studies of Rocky Flats

Below are the 20 articles that we reviewed. Following the citation to each article, we briefly summarize the article's conclusions or provide representative quotes from the article.

### Linear Non-Threshold (LNT) Model and the Risk Assessment Approach:

This literature review started by evaluating current opinions of the scientific community as it relates to LNT model approach, because that was the underlying model of the U.S. Environmental Protection Agency (EPA) Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) risk assessment for Rocky Flats. It is important to remember that the LNT model is applied nationally in various environmental contexts. During the Rocky Flats investigation and cleanup process, a CERCLA risk assessment was performed, which considered regulation of radioactively contaminated sites.

The LNT (alternatively, the “nonthreshold”, “no-threshold,” or “no threshold”) model, is a linear regression that projects adverse effect (or carcinogenic risk) to humans for smaller and smaller doses derived by detectable doses (in the figure represented by units in grays (Gy)). The LNT model assumes this relationship is proportional and is the default model used nationally. A supra-linear viewpoint indicates that there may be more adverse effects from smaller doses. The opposite of this viewpoint is hormesis, which suggests there are low doses of ionizing radiation may be beneficial. The LNT model sits between the two viewpoints. The figure below illustrates the LNT viewpoints, adapted from Figure 12D-1, Health Effects associated with dose of ionizing radiation. [Occupational and Environmental Health \(p. 294\), Seventh Edition](#). Edited by B.S. Levy, by Oxford University Press, 2018:



**Figure 12D-1.** Health effects associated with dose of ionizing radiation.

From the EPA’s national *Risk Assessment Guidance, Human Health Evaluation Manual*, “this hypothesized mechanism for carcinogenesis is referred to as ‘nonthreshold’ because there is believed to be essentially no level of exposure to such a chemical that does not pose a finite probability, however small, of generating a carcinogenic response. That is, no dose is thought to be risk-free.” (EPA, 1989)

The *EPA Radiogenic Cancer Risk Models and Projections for the U.S. Population* (EPA, 2011), also known as the “Blue Book,” is the 2011 update of EPA’s methodology for estimating cancer risks from radiation exposure. These updates were based on the National Research Council’s 2006 report, *Biological Effects of Ionizing Radiation (BEIR VII)*, as well as other updated science.

1. Puskin, J.S., 2009, *Perspective on the Use of LNT for Radiation Protection and Risk Assessment by the U.S. Environmental Protection Agency*

This article provides the summary of using the LNT model in risk assessment. The EPA “bases its risk assessments, regulatory limits, and nonregulatory guidelines for population exposures to low level

ionizing radiation on the LNT hypothesis, which assumes that the risk of cancer due to a low dose exposure is proportional to dose, with no threshold. The use of LNT for radiation protection purposes has been repeatedly endorsed by authoritative scientific advisory bodies, including the National Academy of Sciences' BEIR Committees, whose recommendations form a primary basis of EPA's risk assessment methodology.

*2. Shore et al., 2018 Implication of recent epidemiologic studies for the linear non-threshold model and radiation protection, Journal of Radiological Protection*

The *Shore* article reviewed the 2018 National Council on Radiation Protection and Measurements (NCRP) Commentary No. 27 - *Implications of Recent Epidemiologic Studies for the Linear Non-threshold Model and Radiation Protection*. Of the eleven contributing authors providing their expert opinions of the NCRP Commentary No.27, three were actively involved with the closure of Rocky Flats.<sup>1</sup>

This review set to "determine if recent epidemiological studies of low-linear energy transfer (LET) radiation, particularly those at low doses and/or low dose rates (LD/LDR), broadly support the LNT model of carcinogenic risk or, on the contrary, demonstrate sufficient evidence that the LNT model is inappropriate for the purposes of radiation protection." "The Commentary provides a critical review of the LD/LDR studies that are more directly applicable to current occupational, environmental and medical radiation exposure circumstances." "An overview was provided of radiation studies of breast and thyroid cancers, and cancer after childhood exposures. Non-cancers are briefly touched upon in this article, such as ischemic heart disease, cataracts, and heritable genetic effects." For reference, a low absorbed dose is considered to be less than 100 mGy (milligrays), equal to 10,000 millirem (mrem), delivered acutely and a low absorbed dose rate is < 5 mGy/hour (h) (500 mrem/h) for any accumulated dose. For Rocky Flats, the exposure limit for the inhalation of radionuclides is 10 mrem/year, set forth in 40 CFR 61, Subpart H -National Emission Standards for Emissions of Radionuclides Other Than Radon from Department of Energy Facilities.

The Committee judged "that the available epidemiologic data were broadly supportive of the LNT model and that at this time no alternative dose-response relationship appears more pragmatic or prudent for radiation protection purposes." *Shore et al* concluded that the "current data are not precise enough to exclude models that differ from the LNT model, and there is evidence from some datasets that the slope of the dose response at low levels of exposure may be less than that a higher levels."

*3. Cardarelli, J. and Ulsh, B., 2018 - It is time to move beyond the linear no-threshold theory for Low-Dose Radiation Protection, Dose-Response*

This article, published in the July-September 2018 Dose-Response journal, argues against EPA using the LNT model to estimate cancer risk in radiologically contaminated environments. The EPA developed the Radiogenic Cancer Risk Models and Projections for the US Population (2011) document as a tool to help establish cleanup levels based on the LNT model. In Executive Order 13777 (2017), a policy was established to eliminate "unnecessary regulatory burdens" and resulted in a public meeting to solicit proposals. From this meeting, "the Health Physics Society gave verbal comments urging the EPA to reconsider their adherence to LNT." This article contends the EPA policy documents, like the Risk Assessment Guidance for Superfund (RAGS) guidance, were developed to be more consistent with a "chemical risk paradigm." The "risk estimation tools based on the LNT model" would have CERCLA sites "multiply very small doses by large populations to predict excess cancer incidence or mortality."

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<sup>1</sup> John Till, PhD is one of the listed authors of this review. Dr. Till was the primary author of the 2002 'Risks to the public from historical releases of radionuclides and chemicals at the Rocky Flats Environmental Technology Site' and also contributed to the 2002, Rood *et al.* article 'A Model for a comprehensive assessment of the exposure and lifetime cancer incidence risk from plutonium released from the Rocky Flats Plant, 1953-1989'. Helen Grogan, PhD is also a listed author and was also a contributing author to the 2002 Rocky Flats papers. Emily Caffrey, PhD is another one of the listed authors of this article.

Also noted in this article is the Australian Radiation Protection Society's 2008 opinion that "for individual doses less than some tens of millisieverts a year, risk interferences are unreliable and carry a large uncertainty that includes the possibility of zero risk."

In contrast to the Shore *et al.* review of LNT from the 2018 article, Cardarelli, PhD, and Ulsh, PhD, proposed an altogether less conservative approach for the low, low-dose rather (LDDR) exposures. The conclusion of this article presents the findings that scientific literature is showing "that adverse health effects from LDDR radiation exposures are not detectable and there may be a threshold or even a beneficial effect."

The Government Accountability Office (GAO) published a study: 2017 GAO-17-546 on Low-Dose Radiation, Interagency Collaboration on Planning Research Could Improve Information on Health Effects, that summarizes the contrasting opinions of risk, as seen in the two articles above (Shore *et al.* and Cardarelli and Ulsh) from low levels of radiation exposure and request additional collaboration between federal agencies. The study recommended DOE provide leadership and coordination for research in the future. In a 2017 response, DOE disagreed that DOE should take the lead.

One main consideration in the LNT model is the dose. The following articles briefly touch on radionuclide dose.

#### 4. Harrison, J., 2003 - *Carcinogenic risk from hot particle exposures - has ICRP got it right?*

This invited editorial article discusses the "widely used estimates of dose and carcinogenic risk from incorporated radionuclides", provided by the International Commission on Radiological Protection (ICRP), which are based on calculations of average dose. The argument exists that low levels of radiation pose a risk that is not accounted for in averaging, in the case of hot particles. "Doses to the respiratory tract from inhaled radionuclides take account of the position of target cells for the induction of different lung cancer types in the bronchial and bronchiolar airways (ICRP Report 66, 1994)." Thus, target tissue volume can vary from a single layer of cells to a whole organ. But the ICRP concluded that it "considered the available animal and human data on hot particle effects", and the risk "should be estimated on the basis of average dose within the target tissue." "Claims that dose from hot particles can be orders of magnitude more carcinogenic than dose delivered uniformly were judged to be poorly founded."

"Taking account of differences in relative biological effectiveness of alpha particles and low LET radiations, the risk estimates derived were similar despite differences including time-course of dose delivery and the heterogeneity of energy deposition from plutonium-239 oxide particles. While it is important to recognise (sic) the uncertainties associated with these risk estimates, including lifetime risk projection and application to different populations for the Japanese data and estimates for lung target tissues doses from plutonium-239 oxide particles in Mayak workers, the values obtained suggest that hot particle irradiation is not unexpectedly effective."

"It can be concluded that, on current evidence, hot particle effects do not provide a mechanism for dose from environmental levels of artificial radionuclides to be more effective in causing cancer than larger doses from naturally-occurring radionuclides. The ICRP approach of averaging dose to target cells and tissues appears to give reasonably reliable estimates of risk."

#### 5. Grogan, H. et al., 2001 - *Risks of fatal cancer from inhalation of <sup>239</sup>, <sup>240</sup>plutonium by humans: a combined four-method approach with uncertainty analysis*

This study determined a "mortality risk per unit intake of activity via inhalation of a 1- $\mu$ m activity median aerodynamic diameter (AMAD) plutonium aerosol" for different tissues receiving different doses following an inhalation exposure. "The median absorbed dose received per unit activity of inhaled aerosol was found to decline with increasing particle size." Conversely, the absorbed dose per unit activity of inhaled aerosol was therefore increased with decreasing particle size. Plutonium risk

coefficients for lung, liver, bone, and bone marrow were used. "Four approaches based on fundamentally different sources of information" were used: workers exposed to plutonium, persons exposed to low-LET of plutonium, persons exposed to other alpha-emitters, and animals exposed to other alpha-emitters and were "combined to create an overall risk estimate."

This article identifies the primary source of a member of the public 'around Rocky Flats' exposure to plutonium in the atmosphere was "most likely in oxide form." This article concludes "mortality risk per unit intake via inhalation of a plutonium aerosol" for a 1-  $\mu\text{m}$  AMAD. For plutonium aerosol, of the organs studied, the lung represents the "largest risk" with a mortality risk per unit dose median estimate of  $5.3 \times 10^{-7}$  becquerels (Bq)<sup>-1</sup>.

*6. Fritsch, 2004 - Uncertainties in doses due to the number of aerosol particles: Study on <sup>239</sup>PuO<sub>2</sub> using default parameters for workers*

This paper sought to explore pure PuO<sub>2</sub> statistical simulations using the default parameters recommended by ICRP Report 66 (Human respiratory tract model for radiological protection, 1994), but recognized that there are uncertainties in the calculated doses. Uncertainties exist in the "deposition within the respiratory tract regions in terms of activity fraction and number of particles" and also uncertainty in "respiratory tract mechanical clearance." A Monte Carlo exposure simulation was conducted. It proposed the assumption 'that exposure is not aerosol size-dependent (only inhalability is size-dependent" per ICRP Report 66), "a population of different particle sizes was obtained" for a simulated aerosol. The modelling pertained to only PuO<sub>2</sub>, with "a single physical and chemical property of the aerosol." Uncertainties in respiratory tract deposition and doses vary "depending on the specific activity of the compound", number of aerosol particles per unit, as well as aerosol size and density, and therefore could be "significantly different than default values."

**Particle Characteristics and Exposure:**

After considering LNT, this literature review sought to identify peer-reviewed research conclusions regarding that size of particles, "hot particles," and exposure to more heterogeneous scenarios. This section also includes historic peer-reviewed articles for context.

*7. Salbu, B. et al., 2018 - Challenges associated with the behavior of radioactive particles in the environment*

This article contends that due to uneven distribution in the environment and partial dissolution of radionuclides, inventories can be underestimated, and "impact and risk assessment may suffer from unacceptable large uncertainties if radioactive particles are ignored." This paper reevaluates selected sites and samples to review particle characteristics, particle weathering, and "heterogeneities in biological samples to evaluate potential uptake and retention of radioactive particles." The paper characterizes the type of radioactive particle release, including particles released "during low temperature conditions" or corrosion processes and "damages to containment", which is most similar to the conditions from the 903 Pad at Rocky Flats.

This article describes how assessments are traditionally undertaken, where "average bulk mass or surface activity concentrations of radionuclides in environmental compartments." "Localised (sic) heterogeneities such as particles, however, be unevenly distributed, hence representative sampling can be questionable and dissolution of particles prior to measurements may be partial."

Looking at specific particles instead of a bulk sample points to the theory that "radioactive particles are of biological relevance and have generally been ignored when considering risk assessment and impact." Because particles are in the environment and weathering, the process is dynamic and ever-changing and therefore, dose estimates are inadequate for characterizing small high specific activity

particles. Emily Caffrey's work (article summarized below) is referenced in this article with respect to self-shielding, where a homogeneous distribution is assumed for the study.

*8. Caffrey, E. et al. 2017 - Comparison of homogeneous and particulate lung dose rates for small mammals*

This study contends that calculating radiation doses are "unsuitable when the inhaled activity is in the form of hot particles." "As the dose is confined to a small tissue volume, averaging the dose over the entire organ or tissue volume seems erroneous." "plutonium-239+240 was used as the alpha emitting radionuclide" in this study. "It was assumed to be in the form of PuO<sub>2</sub>, consistent with the chemical composition found in the environment at Maralinga, the Nevada Test Site, and Chernobyl."

The study concludes that "analysis methods capable of determining particulate size be employed to allow for more accurate determinations of dose rate." The study also found that the "self-shielding effect is stronger in alpha-emitters for which most of the emissions are shielded in particles above a diameter for approximately 5 µm." "Below this size, on average 85% of the alpha particle energy escapes the particle and deposits in the surrounding tissue."

*9. Burkart, W. and Linder, H., 1987 - Hot particles in the environment - Assessment of dose and health detriment*

This study was a comparison of hot particle exposure versus a homogeneous distribution in the lung. "Only activity deposited in the pulmonary region will lead to a long residence time and hence to considerable doses." "Particles deposited in the tracheobronchial region of the lung will be cleared to the gastrointestinal tract by the mucociliary ladder." "For inhalation, as well as ingestion, theoretical considerations predict a decrease in the effectiveness of the particular radioactivity as compared to monomeric activity." This study concluded, "that radiation from hot particles was never shown to be more radiotoxic than the same activity uniformly distributed in the organ." "Both from theoretical considerations, from animal studies and from human epidemiology, is that wasting of dose on dead cells introduces a safety factor." "Therefore, the general conclusion stressed in review papers, that with increasing inhomogeneity, the detrimental effects of ionizing radiation to the lung tend to decrease." Conversely stated, with homogeneity, effects of ionizing radiation to the lung would then increase.

*NCRP Report 130 - Biological Effects and Exposure Limits for 'Hot Particles', 1999*

"There is currently no dosimetry model designed specifically for evaluating the absorbed-dose distribution to the respiratory tract arising from a single mobile hot particle (Section 8.2 Approaches to Limits in this Report)." "For purposes of this Report, hot particles are considered to be greater than 10 µm but less than 3,000 µm in any dimension (Introduction)."

In Section 6.2.2.2, Respiratory Tract, Hot Particles, this report states that "nonuniform radiation of the lung by low-LET radiation was not demonstrated to be more effective than uniform irradiation." And the "average radiation dose to the lung for evaluating the risk for stochastic effects is supported by animal data for lung tumors." But, "radiosensitivity for cancer induction likely varies over the lung." And "dose should not be averaged over different regions of the respiratory tract because of radiosensitivities" (attributed to Bair, 1991, overview of the ICRP respiratory tract model). "Equivalent dose distribution over the respiratory tract will depend on where the particle deposits in the respiratory tract, its radionuclide content, self-adsorption properties, and its behavior after deposition (e.g. dissolution, particle movement)."

*10. Batuk, et al., 2015 - Multiscale Speciation of U and Pu at Chernobyl, Hanford, Los Alamos, McGuire AFB, Mayak, and Rocky Flats*

The research sought to compare ideal oxidation in a controlled lab setting with actual uranium and plutonium speciation in either bulk soil samples or single particles. Rocky Flats samples were taken from 1) soils from the 903 Pad in 1998 and from 2) the contaminated concrete floor of a building with HNO<sub>3</sub>-based purification lines in 2002. "Bulk measurement with a beam of millimeter (mm) dimension could be placed on different portions" of the Rocky Flats samples. One of the findings was that both uranium and plutonium were "always concentrated in particles without any sign of a more diffuse population." The study observed "equilibrium species as well as finding novel species that would have resulted from unpredicted interactions of plutonium and uranium species with other components of their waste streams and surroundings", as was the case with the Rocky Flats concrete samples. The 903 Pad soil samples principally speciated to PuO<sub>2+x</sub> as presented in Table 1, and are grouped as "relatively well ordered PuO<sub>2+x</sub>" that "equilibrated with O<sub>2</sub> and H<sub>2</sub>O" under ambient conditions. The two Rocky Flats soil samples from the 903 Pad "were known to have been originally in metallic form." In contrast, acidic waste streams promote "unusual speciation and morphology" as is the case with the concrete from a purification process line at Rocky Flats. The authors conclude there can be a "variation in particle composition" due to "chemically diverse soil materials when the putatively acidic solution was neutralized by reacting with the soil components, releasing or activating some that could then possibly combine with the plutonium that was also originally dissolved."

*11. Charles, M. and Harrison J., 2007 - Hot particle dosimetry and radiobiology*

The article discusses hot particles by recognizing variation in particle sizes and then a review of health effects in relation to skin, eye, ear exposures and also ingestion and inhalation exposures. The basis of this paper revolves around fuel fragments (varying "substantially in size but are most typically similar in size to grains of sand") from the Dounreay nuclear site in Scotland, as they are found on a nearby public access beach, Sandside Beach. Because "the spatial dose distribution around 'hot particles' is highly non-uniform", this study concludes that "conventional methods for evaluating dose and for predicting biological effects are inappropriate for 'hot particles'." "'Hot particles' with linear dimensions less than 10 µm are capable of entering the deep lung, and long-term resident of insoluble material may lead to a potential long-term lung cancer risk."

This article presents the following regarding the 'hot particle hypothesis' claim of increased carcinogenicity: "After a decade of intensive experimental and theoretical investigation an ICRP review of the biological effects of inhaled radionuclides in 1980 refuted this claim in the context of inhaled 'hot particles'." "The ICRP has maintained its advice that the use of mean organ or tissue dose is appropriate for the evaluation of carcinogenic risk for radiological protection purposes."

The article concludes that after determining the exposure routes for skin, eye, and lung, "the challenge for regulators remains, as in other situations of environmental contamination, to determine the extent to which remediation is required in the light of potential health effects with a low probability of occurrence."

**Historic Rocky Flats Studies:**

This literature review then considered the historic work published regarding Rocky Flats as it related to soil sampling results and particle size.

*12. Rood, A., Grogan, H., and Till, J., 2002 - A model of comprehensive assessment of exposure and lifetime cancer incidence risk from plutonium released from the Rocky Flats Plant, 1953-1989*

This paper focused on modeling of "ambient air concentrations, surface deposition, and lifetime carcinogenic risk with uncertainty from plutonium released to the air" between 1953 and 1989. "Predicted air concentrations included contributions from site releases and resuspension from contaminated soil." "Release quantities were segregated into four particle-size classes representing <3

$\mu\text{m}$ , 3-10  $\mu\text{m}$ , 10-15  $\mu\text{m}$ , and >15  $\mu\text{m}$ ." Then "fraction of activity values were associated with each size fragment based on measurements" made by other researchers.

Table 7 in the article summarized measured resuspension factors (RFs) that allowed for the calculation of both short-term (newly deposited activity) and long-term (contaminated soil after weathering and redistribution of activity within the soil) values in the U.S. Nuclear Regulatory Commission RF. This data was collected when site remediation activities were occurring at the 903 Pad and represented bare soil surfaces free of vegetation. The mean value reported in Table 8 for short-term RF is  $3.2 \times 10^{-7} \text{ m}^{-1}$  and for long-term RF is  $3.2 \times 10^{-11} \text{ m}^{-1}$ .

This study recognized that when utilizing models for resuspension, there are limitations with a mass loading approach that cannot account for a non-linear "relationship between surface soil plutonium concentrations and concentrations of plutonium on suspended dust." The mass loading model also cannot account for weathering, as it "tends to make activity less susceptible to resuspension."

*13. Till J. et al., 2002 - Risks to the public from historical releases of radionuclides and chemicals at the Rocky Flats Environmental Technology Site*

From the conclusions: "The study analyzed the resuspension of particles during high-wind events to determine if these events could have contributed significantly to the risk to the public from soil contaminated with plutonium on site. This was a key question that had been raised by the public early in the study. The present analysis indicated that risk from the high-wind events was actually lower than previously calculated when the release was evaluated as a steady-state process over a 5-year period. The reason for this finding was that although significant numbers of large particles were resuspended during high-wind events, atmospheric dilution at downwind locations is much greater than during normal environmental conditions."

*14. Little, C. et al., 1980- Plutonium in a Grassland Ecosystem at Rocky Flats*

This study concluded that "wind erosion of contaminated soil was likely the chief mechanism of plutonium transport and that soil contained 99.7% of the plutonium in the system." The study determined that "at all depths, the higher plutonium concentrations were associated with the smaller soil particles or aggregates." The study provides an example of the "high degree of spatial variation" of plutonium distribution from three contiguous soil sample locations, 1 kilometer (km) south of the 903 Pad, with the explanation that "many small plutonium particles agglomerated together with large soil particles."

*15. Little, C. and Whicker, W., 1978 - Plutonium Distribution in Rocky Flats Soils*

This study sought to correlate air data gathered from an "air sampling station S-8 located about 75m southeast" of the 903 Pad with soil samples collected to determine the distribution of plutonium in soils. Soil sampling was initiated along two transects (one 500 meters (m) ESE and the other 250 m SSE) originating close to the 903 Pad following the predominant wind pattern and downslope winds, respectively. The results did "not produce any particular pattern with either depth or particle size range" and the authors concluded there is large variability "in soil plutonium concentrations with regards to depth, particle size distribution, and spatial distribution." The article speculates that  $\text{PuO}_2$  particles "attached to soil particles" and "took the form of easily erodible, agglomerated particles, each containing numerous  $\text{PuO}_2$  and soil particles." The conclusion "that most contaminated particles are very small or single particles" was supported by the autoradiographic studies of the soil.

*16. McDowell and Whicker, W, 1978 - Size Characteristics of Plutonium Particles in Rocky Flats Soil*

This article summarized work that collected soils from approximately 200 m southeast from the 903 Pad. Two autoradiographic techniques were used, 1) to provide information on "occurrence probabilities of relatively large plutonium particles" and 2) "plutonium particle size distributions in soil

samples." Plutonium particles from Rocky Flats are speculated to be "small plutonium particles or plutonium compounds attached to the surface of soil particles." "The largest particle size was 6.86  $\mu\text{m}$   $^{239}\text{PuO}_2$  equivalent diameter using both autoradiographic techniques." "The activity associated with the 6.86  $\mu\text{m}$  is 236 dis/min" or 3.93 Bq (or 106 picocuries (pCi)). The conclusions of this article are that "heterogeneity does exist in plutonium particle concentrations" and "a mechanism may exist that either concentrates plutonium particles in an area, or causes breakdown of plutonium particles into smaller adjacent particles." Therefore "sample aliquots would be expected to exhibit the variability observed in plutonium particle concentrations."

17. Webb *et al.*, 1997 - *A Three-Dimensional Spatial Model of Plutonium in soil near Rocky Flats, Colorado* and 18. Hulse, *et al.*, 1999 - *Comparison of  $^{241}\text{Am}$ ,  $^{239/240}\text{Pu}$  and  $^{137}\text{Cs}$  Concentrations in soil around Rocky Flats*

Webb *et al.* performed 4 transects (60°, 90°, 120°, 150° azimuths from the 903 Pad) up to 19 kms from the 903 Pad and sampled for  $^{239}\text{Pu}$ . Soil types were described as "clay loam, with gravelly sandy loams more frequent near the 903 Pad." The results supported a "relatively uniform concentration of plutonium over the first 2 cm is suggestive of some on-going natural mixing process in that region." The study reported that plutonium concentrations < 5 Bq per kilogram (0.135 pCi/gram) "are much less accurately predicted than higher levels near the contamination source." "Overland or surface flow of precipitation was directly observed on the site in 1995", and "it was hypothesized that erosion may account" for decreases in  $^{239}\text{Pu}$  distribution and apparent loss when compared to studies over a 17-year period. "Evidence of severe erosion was absent from the well-vegetated plots sampled during the current study." Rood *et al.*, 2002 (summarized above) uses the data collected in Webb *et al.* study to model ambient air concentrations, surface deposition, and lifetime carcinogenic risk between the years of 1953 to 1989.

Hulse *et al.* collected soil samples "from 18 on-site and 24 off-site locations along four radial transects, each approximately 20 km long" in the same general transects that Webb *et al.* sampled. "Like  $^{241}\text{Am}$  and  $^{239,240}\text{Pu}$ ,  $^{137}\text{Cs}$  is strongly adsorbed to small soil and clay particles." "Measurements of  $^{137}\text{Cs}$  could reveal much about the expected behavior of  $^{241}\text{Am}$  and  $^{239,240}\text{Pu}$  if it could be demonstrated that these actinides have quantitatively similar behavior to that of  $^{137}\text{Cs}$  in soil around Rocky Flats." "More samples could be analyzed with gamma spectroscopy, which is relatively fast and inexpensive when compared to radiochemical separations and alpha spectroscopy." "Redistribution caused by disturbance and natural erosion following initial dispersion has probably affected current deposition patterns of  $^{241}\text{Am}$  and  $^{239,240}\text{Pu}$  around Rocky Flats." The  $^{241}\text{Am}$  and  $^{239,240}\text{Pu}$  soil concentration ratios "in the top 3 cm of soil appears to increase from 0.18 at on-site locations to 0.36 at off-site locations." The study looked at ratios between  $^{241}\text{Am}$  or  $^{239,240}\text{Pu}$  and  $^{137}\text{Cs}$  and determined that those ratios "proved more useful for delineating the extent and pattern of contamination from Rocky Flats than did activity concentrations in soil." Another estimate from this study was the peak soil concentrations of  $^{241}\text{Am}$  (from decay of  $^{241}\text{Pu}$ ) projected to occur during the year 2032.

19. Paton *et al.*, 2004, *Quantification of Actinide Migration Pathways at Rocky Flats, Colorado*<sup>2</sup>

One of the purposes of this study was to determine actinide (defined as the radioactive metallic elements, and not naturally occurring, which included plutonium-239/240) movement in air, ground water, and surface water at the site. The summary concluded wind and water erosion as the dominant transport pathways for plutonium, but that airborne transport is "more significant than previously suggested in the qualitative conceptual model study." The calculated airborne actinide loads transported off-site were based on air quality and wind data collected from 1997 through 1999, at air monitoring stations at the Rocky Flats perimeter boundaries. The calculated air pathway off-site actinide loads for plutonium-239/240 equals 7E-04 curies per year (Ci/yr) or 7E-16 pCi/yr as presented

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<sup>2</sup> The two primary authors, Ian Paton and Christine Dayton, were members of the Rocky Flats Actinide Migration Panel for approximately 10 years.

in Table 3. The article is careful to note “the air monitoring location with the highest concentration of actinides has levels that are less than 2% of the allowable 10 mrem standard” (40 CFR 61, Subpart H - National Emission Standards for Emissions of Radionuclides Other Than Radon from Department of Energy Facilities).

## CONCLUSIONS

There are three main conclusions from the literature review regarding the behavior of Rocky Flats plutonium in the environment and its potential health impacts:

- (1) remaining Rocky Flats plutonium in the Jefferson Parkway transportation corridor and offsite poses a risk, but it is small and based on CDPHE’s 2020 RESRAD modeling and well within regulatory limits for radiation;
- (2) consistent with findings from the Rocky Flats site investigation and cleanup, Rocky Flats plutonium is heterogeneously distributed in the transportation corridor environment; and
- (3) literature would suggest that while there is no one uniform position on radioactive particle dose assessment, there did appear to be a general tendency to conclude that dose assessments based on homogeneity would be conservative and likely predict a larger dose.

CDPHE recommends the next CERCLA Five-Year Review assess recent scientific literature.

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