UCRL-PROP-225310

Project Pre-Proposal

A Systematic Baseline Study of Internally Deposited Plutonium in Agricultural Workers and Local Residents of Palomares (Almeria District), Spain

Terry Hamilton
Tom Brown

Center for Accelerator Mass Spectrometry
Energy and Environment Directorate
Lawrence Livermore National Laboratory

and

Robin Newmark

Water and Environment Program
Lawrence Livermore National Laboratory

September 2006

This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

A Systematic Baseline Study of Internally Deposited Plutonium in Agricultural Workers and Local Residents of the Almeria District, Spain

Terry Hamilton, Tom Brown and Robin Newmark
Lawrence Livermore Natiional Laboratory
(hamilton18@llnl.gov)

1.0 Background

Residents and agricultural workers living in the vicinity of Palomares, Almeria (Spain) continue to be exposed to residual levels of plutonium contamination in the environment as a consequence of a 1966 mid-air collision between a United States Air Force (USAF) B-52 bomber and a refueling tanker. Of the four nuclear weapons onboard the USAF bomber, two weapons were recovered intact. The chemical high explosive component of the other two weapons detonated on impact and produced plumes of contamination that spread over an area of approximately 226 hectare including the northern edge of the village, farmlands and non-cultivated terrain. Crops and surface soil were removed in the most heavily impacted regions, and the material packaged and shipped to the United States. Crops and vegetation from less contaminated areas were either burned on the beach or buried, and soil plowed to a depth of 30 cm. None of the 1500 residents living in the region at the time of the accident were injured although a small number of villagers suffered from internal contamination due to inhalation exposure (Iranzo *et al.*, 1988). An initial radiological monitoring program involving air, soil and vegetation sampling, urine bioassay and lung counting was immediately established under the authority of the Spanish Junta De Energia Nuclear. The estimated 70-y committed effective dose equivalent (CEDE) to chronically exposed individuals ranged from 20 to 200 mSv (Iranzo *et al.*, 1988). The remaining population received an estimated dose of less than 20 mSv.

2.0 Project Proposal Outline

Today, there is much to be learnt from the situation and recorded experiences at Palomares. The on-going medical and environmental surveillance program conducted by the Spanish Government through the Centro de Investigaciones Energeticas Medioambientales y Technologicas (CIEMAT) has provided invaluable data and information. However, a number of uncertainties still exist in attempting to accurately and reliably characterize the chronic level of plutonium uptake into the local population. Also, evidence linking the need for further remediation of the impact zones with elevated levels of plutonium exposure and uptake in the local population appears to be lacking. At the same time, urine bioassay monitoring of long-term residents-as best that we can tell from available data-continue to show a small number positive detects for plutonium. The minimal detectable amount (MDA) of plutonium based on alpha-spectrometric measurements performed at CIEMAT is reportedly around 700 μBq (DOE 92-234, translation of Spanish document). Average concentrations of plutonium measured in air samples over the past 3 decades do not support findings that people could acquire such high levels of plutonium in their urine through inhalation exposure. Other potential routes of plutonium exposure include ingestion and skin absorption through wounds or open cuts. The presence of hot particles in soils also needs to be considered in predictive dose assessments as do the uncertainties associated with changing land-use patterns. Moreover, it is very clear that this region has undergone tremendous change over recent history. The municipal districts of Cuevas del Almanzora and Vera surrounding the

village of Palomares are attracting more and more tourists with housing and construction activities on the rise. Much of the open terrain is used for intensive agriculture, especially for growing crops such as tomatoes, water melon, grain and alfalfa. The soil-to-crop plutonium concentration ratios for these types of products are of the order of 10^{-3} to 10^{-4} suggesting that the annual CEDE from the ingestion pathway should also be extremely low. However, the historical bioassay program does not provide sufficient sensitivity to substantiate predictive dose assessments based on environmental data. Moreover, the fact that CIEMAT scientists have occasionally detected plutonium in urine bioassay samples at alpha spectrometric detection levels suggests that there are as yet unexplained chronic and/or incremental exposure events that require further investigation. Follow-up action on positive detects in urine appear to be largely limited to the initial 45 people considered to have been chronically exposed at the time of the accident. It should also be noted, classical bioassay monitoring programs based on alpha spectrometry don't have the necessary isotope detection sensitivity to even comply with the latest U.S. Department of Energy implementation of federal regulation 10CFR 835 for *in vitro* bioassay monitoring of plutonium.

The people of Palomares represent a well documented group who have lived and worked in this plutonium-contaminated environment for decades. Consequently, it would be externally valuable to accurately quantify levels of plutonium uptake in the local population using more advanced low-level detection and measurement techniques such as accelerator mass spectrometry (AMS). This information has value in its own right in helping describe the long-term consequences of accidental and/or deliberate dispersals of plutonium within populated areas. In general, the usefulness of urinalysis data for assessing plutonium uptake depends on the mode, time and magnitude of the original intake, the fraction of the internal burden excreted and on the sensitivity of the measurement technique. Urinary excretion of plutonium by the general population will consist of a baseline, long-term excretion rate from any residual systemic plutonium acquired from previous exposures (if any), and a background excretion rate consisting of a prompt and long-term component from exposure to worldwide fallout contamination. A small fraction of the plutonium (about 1%) that enters the blood immediately following an intake is excreted in the first day after absorption. Using modeled long-term urinary excretion rates of about ~10^{-5} per day of the initial uptake and measured whole body burdens of plutonium based on analysis of human tissues collected at autopsy, the background excretion rate of ^{239}Pu from people living in the northern hemisphere is expected to be around 1×10^{-6} Bq (or 1 μBq) per 24 hour void. Any assessment of incremental intakes of plutonium in the general population will therefore require detection sensitivities in the range of μBq of ^{239}Pu or less. Scientists from the Center for Accelerator Mass Spectrometry (CAMS) at Livermore (CA) have a demonstrated capability to perform routine plutonium bioassay measurements using AMS where the typical measurement background ranges from <0.2 to 0.5 μBq of ^{239}Pu per sample.

It is very clear that advanced bioassay monitoring techniques such as those employed at Livermore will enable the U.S. Department of Energy (DOE) to more accurately document and assess the present situation at Palomares rather than relying on predictive dose assessments based on environmental data, and model assumptions of plutonium exposure and uptake. Also, there appears to be a number of fundamental questions that need to be answered before embarking on a major cleanup and

rehabilitation program. For example, as best that we can understand from published reports, the ongoing bioassay program conducted by scientists from CIEMAT does not appear to rule out the possibility of significant incremental and/or chronic public exposures to plutonium within the Palomares region. The level and extent of public exposure to plutonium in the Palomares region needs to be properly defined and documented by comparing quantitative measurements of plutonium urinary excretion from carefully selected subgroups. The objective of this study would be (1) to assess the level of chronic buildup of plutonium in critical population groups such as local residents living near the impact zones and agricultural workers; (2) to develop an accurate and well documented baseline in urinary excretion of plutonium from critical population groups (e.g., those residing close to the impact zones) in order to assess potential future intakes associated with changing conditions and/or remediation activities; and (3) conduct an exposure pathway analysis, and assess the relative routes of plutonium uptake into critical population groups and any associated needs for remediation of sites or introduction of control measures. Under this program, we would like to propose a joint study between CIEMAT and LLNL scientists where bioassay samples are collected and analyzed for plutonium isotopes using CAMS. The selection criteria for various volunteers would be coupled to obtaining accurate and reliable plutonium urinary excretion data on selected cohort populations. These data will provide the basis for a full and defensible assessment of systemic uptake of plutonium associated with the nuclear weapons accident at Palomares–both past and present. The study cohort population groups should include;

Group 1. A Baseline Study Cohort consisting of volunteers living in a region outside of Palomares (e.g., Madrid).

Group 2. A Regional Study Cohort consisting of volunteers (excluding non-agricultural workers and those people with a confirmed intake of plutonium) who reside within the municipal districts of Cuevas del Almanzora and Vera.

Group 3. A Local Study Cohort consisting of volunteers (excluding non-agricultural workers and those people with a confirmed intake of plutonium) living within the village area of Palomares at selected distances from impact zone 3. Group 3 volunteers would also include those people living in the immediate vicinity of impact zone 3 and provide a basis for comparison to future potential exposures resulting from remediation activities.

Group 4. An Agricultural Worker Study Cohort consisting of volunteers agricultural workers (excluding those people with a confirmed intake of plutonium) who preferably live outside of Palomares.

Group 5. A Retrospective Dose Assessment Study Cohort consisting of volunteers with previously confirmed intakes of plutonium.

This project would be initiated through a pilot study involving a subset of Group 3 and 4 volunteers. The pilot study will be used to test sample collection protocols and develop some initial high quality plutonium urinary excretion data from potential maximal exposed population groups, i.e., agricultural workers and those local residents living adjacent to the impact zones. A baseline study on residents living near the impact zones

may also help the Spanish Government respond to litigation action should a decision be made to perform additional cleanup work at these sites. The assumption is that the cleanup program will potentially increase the ambient levels of plutonium in air and also tend to heighten public awareness of the situation. In general, the pilot study will also enable the DOE to make more informed decisions about the potential need for a more comprehensive bioassay monitoring program such as that outlined above and, where applicable, help determine the number of volunteers required to develop a statistically significant dataset to examine specific plutonium exposure/uptake scenarios. Either way, the results of this study will provide more definitive information and answer the underlying question 'is there really a radiological concern at Palomares?' These findings will be critically important in terms of either helping manage public perception about plutonium as the 'most hazardous substance known to man' or in identifying a trend in long-term chronic exposure that cannot be predicted from environmental measurements yet necessitates some type of action related to increasing public awareness and/or performing selective remediation.

Funding requirements (initial Pilot Study only):

AMS measurements on 25 samples + QC	=	$50,000
Technical and Scientific Oversight (including report writing)	=	$25,000
		TOTAL = $75,000

Project Proponents and Contact Information

Dr. Terry Hamilton & Tom Brown
Center for Accelerator Mass Spectrometry
Lawrence Livermore National Laboratory
PO Box 808, L397
Livermore CA 94550-0808
U.S.A.

Telephone	(925) 4226621
Fax	(925) 4236785
Email	hamilton18@llnl.gov

****