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Project Pre-Proposal

Baseline Measurements of Internally Deposited Radionuclides in the U.S. Population

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Statement of Intent (*Why should you care!)*:

Today, nuclear weapons proliferation, illicit trafficking of nuclear materials and the threat of radiological or nuclear attacks on U.S. assets present credible risks to the safety and security of the nation. These risks continue to grow with the end of the Cold War and the increasing need to dispose of legacy waste from nuclear weapons production activities, closure of excess production facilities, environmental cleanup and arms reduction programs.

In the aftermath of a nuclear event, there will be an immediate need to assess, evaluate and classify levels of exposure or exposure potential, and determine the need for medical treatment of the exposed populations. Of particular concern will be the ability to assess exposures to internally deposited radionuclides from inhalation of radioactive dust and debris. What needs to be clearly understood is that there is general lack of comparative baseline radiobioassay data within the U.S. to reliably measure or assess internal exposures to radioisotopes in the event of a radiological attack. Such information will be required to help guide the decision making process to introduce medical countermeasures to reduce potential health risks (short or long term) from any internally deposited material and to manage public hysteria. This includes a lack of comparative baseline data on the present-day whole body burdens of important long-lived, alpha-emitting isotopes of uranium and plutonium. Uranium and plutonium provide the primary fuel for nuclear weapons and will tend to attract the most interest from terrorist groups. New challenges are also arising from the growing accumulation of stored plutonium (and uranium) from both civilian and military operations. By the end of 1997, the global stockpile of separated plutonium included about 170 t of separated plutonium from civilian reprocessing operations and 100 t of excess plutonium from dismantled warheads no longer required for defense purposes. Moreover, the cumulative worldwide amount of plutonium contained in spent fuel from power reactors will exceed 1700 t by 2010.

We propose to fill this information gap and develop high quality *baseline* radiobioassay data on uranium and plutonium isotopes in the U.S. population using urinalysis and blood assay. The database (and the associated standardized collection and analysis protocols stemming from this project) will serve as a benchmark to accurately assess public exposures in the event of a radiological or nuclear attack. An underlying feature of our study design and strategic approach is to enlist emergency responders (firefighters) as volunteers to serve as the baseline study cohort. Consequently, these data will be of value in their own right in helping protect the health of those people who may be charged with serving the nation in a time of need.

Background Information:

The most abundant uranium and plutonium isotopes found in the natural and manmade radiation environment are uranium-234 (^{234}U), uranium-235 (^{235}U), uranium-236 (^{236}U), uranium-238 (^{238}U), plutonium-239 (^{239}Pu) and plutonium-240 (^{240}Pu). All of these long-lived radionuclides emit alpha particles and may be taken into the body by inhalation, ingestion or open-wound absorption. Uranium occurs in nature and, in a purified form, has a number of important industrial and military uses. The principal uranium isotopes of primordial origin are ^{235}U and ^{238}U, with ^{234}U usually present in radioactive equilibrium with ^{238}U. The health effects associated with oral exposure to uranium are usually associated with chemical toxicity to the kidney. Chemical toxicity is independent of isotope and depends only on chemical solubility. Inhalation exposure may lead to radiological toxicity, especially for insoluble uranium compounds. Intakes of uranium across the general population are expected to be highly variable because of variations in the uranium content of food and different sources of drinking water. ^{236}U occurs in nature at ultra-trace concentrations down to $^{236}U/^{238}U$ atom ratios of approximately 10^{-14}. For the purposes of this discussion, the $^{236}U/^{238}U$ content of the natural radiation environment falls below the level of detection by mass spectrometric techniques. However, enhanced levels of ^{236}U are produced in nuclear detonations and in nuclear reactors primarily by neutron irradiation of ^{235}U. Therefore ^{236}U may be useful as an *isotopic fingerprint* to discriminate between natural sources of uranium and intakes associated exposure events following a radiological or nuclear attack. Small amounts of plutonium also exist in nature from spontaneous ^{235}U fission but the worldwide deposit in the near-surface environment is dominated by global fallout deposition from atmospheric nuclear weapons testing. About 11 PBq or 3350 kg of plutonium was globally dispersed by atmospheric nuclear weapons tests conducted during the 1950s. Global fallout deposition has since increased the background whole body burden of plutonium in humans by at least 5 orders of magnitude.

Under this proposal we will develop baseline data on internally deposited plutonium and uranium isotopes in the U.S. population as a basis for assessing potential intakes associated with exposure events following a radiological or nuclear attack. In radiation safety and health, alpha-emitting radionuclides that reach the systemic system of the body are of great concern. Alpha particles have a very short range in tissue and cannot be measured by detectors external to the body, so *in-vitro* bioassay tests have been developed to evaluate intakes based on measured urinary excretion patterns and modeled metabolic behaviors of the absorbed radionuclides. Biokinetic models describing urinary excretion of plutonium have been developed using data from plutonium-exposed workers, plutonium-injected volunteer subjects and experiments with laboratory animals. Systemic plutonium absorbed by the deep-lung or from the gastrointestinal tract after ingestion is either excreted or distributed to other organs, primarily to the liver and skeleton where it is retained over biological half-lives of 20 and 50 years, respectively. Once incorporated into the body, alpha emitters continue to irradiate surrounding tissues until they are totally eliminated by radiological decay or excretion.

Acute uranium or plutonium intakes by the general population following a radiological or nuclear event will most likely come from inhalation exposure or adsorption from open wounds. Intakes of radionuclides via inhalation are studied by means of the respiratory tract model described in International Commission on Radiological Protection (ICRP) Publication 66. The quantity and initial penetration of radionuclides entering into the respiratory tract will vary according to respiratory parameters such as body size, level of activity, respiratory tract diseases and smoking habits. Clearance of inhaled material from the other regions will depend on the competition between particle transport processes to the gastrointestinal tract via the pharynx or to lymph nodes via lymphatic channels, and absorption into blood. The systemic transport of uranium and plutonium in the human body is discussed in ICRP Publication 69.

The usefulness of urinalysis data for assessing uranium and 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. Under normal environmental conditions, urinary excretion of uranium will normally be dominated by intakes associated with ingestion of foods and water. 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 $\sim 10^{-5}$ per day of the initial uptake and measured whole body burdens of plutonium based on analysis of human tissues collected at autopsy, the urinary excretion of ^{239}Pu by the general population is expected to around 1×10^{-6} Bq (or 1 μBq). 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. The amount of plutonium in samples of blood may be even lower but blood assay is potentially beneficial in helping reduce the errors associated with collecting representative '*spot*' urine samples, gives a more direct measure of plutonium entering the systemic system of the body, and helps decrease the risk of sample contamination in an emergency field environment. Comparative measurements between blood and urine could then be used to develop the internal dosimetry for radiological event related intakes.

Implementation Plan:

High quality urinary excretion and blood assay data on plutonium and uranium do not exist for the general U.S. population. Moreover, classical bioassay monitoring programs within the U.S. lack 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. We believe that similar or even improved standards of safety and risk management will be required in assessing the public exposures associated with a radiological or nuclear attack. Routine methods for uranium bioassay provide adequate detection sensitivity based on adopted screening levels but cannot assign a dose to the suspected intake because of the inability to discriminate between anthropogenic and natural intakes of uranium. This project proposal will take advantage of the demonstrated capabilities and existing core expertise within Center for Accelerator Mass Spectrometry at the Lawrence Livermore National Laboratory for ultra low-level actinide detection and measurement, and complimentary expertise in analysis of uranium isotopes using multi-collector inductively coupled mass spectrometry (MCICP-MS).

An inherent feature of our study design is to focus the effort on acquiring baseline urinary excretion and blood assay data on uranium and plutonium from emergency responders (firefighters) and establish fire houses as control radiobioassay collection points around the country. Over the course of this study, blood and urine samples will be obtained from a minimum of 500 firehouses representing not less than 5000 individuals. To minimize travel costs to train fire house personnel in collection protocols, we propose to sample fire houses from selected geographic areas over a 3 to 5 year period starting with California and the Eastern U.S. followed by the Pacific Northwest, Mountain, Hawaii, Alaska, Mid-west and Plains states and the Southern U.S. We also propose to concentrate the sampling program on major urban centers, such as New York, Boston, the Washington Metro-Plex, Atlanta, Chicago, Los Angeles, Denver and the San Francisco Bay Area, since conventional wisdom suggests that a terrorist attack will most likely be centered on a major U.S. city. However, we also anticipate sampling some rural and semi-rural areas with consideration given to the north-easterly tropospheric fallout patterns of atmospheric nuclear tests conducted at the Nevada Test Site (NTS).

A scientific technician will travel to many of the selected major city fire houses to ensure sampling equipment is adequately setup, and provide any necessary background information and training to ensure personnel understand and adhere to sampling protocols. A training video will be produced to send along with collection kits because it will not be feasible or practical to travel to all 500 fire houses. Emergency responders (firefighters) are expected to consist largely of fit young to middle-aged men and women. This is not seen as a limitation but an opportunistic way of collecting information on a fairly well defined subgroup of people across the entire country. Moreover, it is expected that firefighters will have an inherent interest in participating in this project and fire houses provide relatively clean common areas (including bathrooms) for large groups of people who are essentially a captive audience for 2 to 3 day work-shift periods. The sampling strategies and implementation plan will take advantage of the groups experience in bioassay collections in the Marshall Islands. Each fire house has a qualified paramedic on each rotation team who will be responsible for the blood draws and, once trained, for oversight of the urine collection. Blood collections will only be performed at the end of the work shift to ensure the collection does not interfere with the operational status of the firehouse. We also envisage that the training of fire house personnel in bioassay sampling collection and handling will provide invaluable experience in terms of establishing an operational sample collection (screening) program in the event of a radiological or nuclear attack.

Project Deliverables:

(1) The development of a high-quality radiobioassay database (including statistical information and web accessibility) on internally deposited plutonium and uranium isotopes in the U.S. population as basis to accurately and reliably assess potential public exposures (and determine the need for medical countermeasures) in the event of a radiological or nuclear attack on the United States.

(2) To develop, test and provide documented procedures for the collection and handling (and radiobioassay analysis) of composite urine and blood samples, and provide training for fire house personnel located in major urban centers to undertake such work.

(3) Scientific publications on baseline data of internally deposited plutonium and uranium isotopes in the U.S. population based on urinary excretion and blood assay with emphasis on collecting background information on emergency responders.

Budgetary Requirements:

In order to implement this project we will require approximately $6M over a 3 to 5 year period.

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