December 3, 1987

TO: Bart Gledhill

FROM: A. Larry Anderson

SUBJECT: Lung Counter Calibration with Inhaled Nb-92m as a "Mock Plutonium"

Approval is requested from the LLNL Human Subjects Committee for LLNL to participate in radioactivity measurements on approximately eight male subjects involved in a United Kingdom project sponsored and funded by the International Atomic Energy Agency (IAEA). The subjects will inhale particles labeled with Nb-92m in a repeat of an experiment performed in 1980-82 to determine in vivo, the influence of spacial distribution on calibration factors for the internal measurement of inhaled plutonium. In the current experiment, 1 micron particles will be used rather than the 5 micron particles which were used in the earlier study.

BACKGROUND

Many employees at LLNL work directly or indirectly with plutonium and other hazardous radioactive materials. As part of the health and safety program at the Laboratory, the Whole Body Counter provides in vivo measurement services for internally deposited radionuclides in LLNL employees and contractor organizations. These services involve the use of sophisticated counting equipment to measure x-ray emission from the body. Unfortunately, calibration of x-ray detectors for the assessment of plutonium (usually in lungs) is a very difficult process. This has led in the past to widely varying estimates (approaching a factor of three) in counter sensitivity among many laboratories using similar equipment, but with different calibration techniques (Newton 1978).

In an attempt to provide some unity to the problem, a realistic tissue-equivalent human-torso phantom was designed and constructed at LLNL under DOE auspices as a standard for calibration of transuranic radionuclide counting systems at LLNL and other DOE facilities. This phantom contains a human male rib-cage, removeable model organs containing various radionuclides including plutonium, and tissue-equivalent chest plates which can be placed over the torso to simulate humans with a wide range of statures. The phantom was circulated in an intercomparison program to participating laboratories in the United States and Europe. This allowed individual laboratories to prepare similar sets of calibration curves to model the performance of their counting systems. A later version of the phantom contained simulated bone in the rib-cage, and was provided as an option for purchase by any laboratory wishing to have their own individual calibration phantom. Quality control during the production of these phantoms by a commercial vendor was provided by LLNL to insure exact intercomparison between the production units.
One central question relating to the phantoms was how closely they actually simulated internally deposited plutonium in real people. In order to answer this question, an experiment was proposed by the British and later funded by the IAEA to verify the LLNL realistic phantom using volunteers inhaling known amounts of Nb-92m as a practical simulant for plutonium. Nb-92m decays by electron capture, emitting Zr K x-rays at 15.8 keV, similar to those from plutonium at approximately 17 keV. Since Nb-92m has no particulate emission and has a biological half-life of approximately 10 days, the volunteers participating in the program would receive a very low radiation dose.

In the previous work, eight male volunteers of different body shape inhaled an aerosol consisting of 5 micron-diameter polystyrene microspheres labeled with Nb-92m. The volunteers visited two laboratories in the United Kingdom and three laboratories in the United States where their lung contents were assayed by standard lung and whole-body counting techniques. In a later study involving eleven women, additional laboratories in the U.S. were included. The observed counting efficiencies from the volunteers were compared with those predicted by the Livermore phantom when Nb-92m loaded lungs were used for calibration. On the average, the observed efficiencies agreed with those predicted by the phantom; however, a trend in the ratio of observed to predicted efficiencies was noted as a function of body size. This finding may indicate a systematic error in the phantom (due to lung size, distribution, or thorax shape) and needs to be investigated further.

Smaller (i.e. 1 micron) particles will deposit differently within the lung, possibly more toward the periphery. Such a change in distribution could have great effects on lung counting efficiency for external measurements. In such cases, efficiencies predicted by the phantom may be very different from those observed in vivo. Some discrepancies have already been noted involving accidental Pu-238 uptake in men. In addition, studies with Nb-92m in women showed that non-uniform lung depositions compared with the phantom would produce errors in assessment of lung content of up to 300% if Am-241 was involved as the lung contaminant (Gunston 1987). Consequently, further validation of the phantom is required, especially since the LLNL phantom is now being used as an international standard as well as a standard for calibration work at DOE laboratories within the United States.

PROPOSAL

To minimize biological variation, the volunteers who participated in the first male study using 5 micron diameter microspheres will be utilized (as far as possible) in a second experiment using the 1 micron particle size. However, three of the original eight subjects were from the U.S.A. and are not expected to participate in the current experiment. These will be replaced by other volunteers selected in the United Kingdom by Dr. Donald Newton, coordinator of the program. The radiation dose received by the volunteers from alveolar deposition in the lungs is not expected to exceed an estimated weighted committed dose equivalent of 70 µSv (7 mRem). A small additional dose will be incurred from particles deposited elsewhere in the respiratory tract, clearing rapidly via the gut. The experimental protocol has already been approved by the UK equivalent of the LLNL Human Subjects Committee and is attached for your information along with a copy of the current UKAEA Code of Practice for Tracer and Irradiation Studies Involving the Use of Volunteers. Also attached
is the cumulative dose information pertaining to five of the subjects who will be utilized in the current study. This information was supplied by Dr. Donald Newton. We have compared this dose data with standards for a U.S. population, also attached.

A comparison of the results of the two experiments will determine the influence of aerosol deposition pattern in the lungs (as a function of particle size) on external counting rates. As before, the calibration factors obtained in vivo will be compared with those predicted by the LLNL phantom to further validate or establish correction factors for the phantom as a calibration standard.

The proposed research will be performed in collaboration with the Atomic Energy Research Establishment (AERE), Harwell, in the United Kingdom. The Atomic Weapons Research Establishment (AWRE) at Aldermaston will also participate. Participating laboratories in the United States will be restricted to Argonne National Laboratory (ANL), Battelle Pacific Northwest Laboratory (PNL), and the Lawrence Livermore National Laboratory. These U.S. laboratories possess counting systems representative of the general international lung counting community as well as having specific state-of-the-art equipment that other laboratories do not have. For example, the most up-to-date solid state counting system in the world for plutonium lung assay is located at the Battelle Pacific Northwest Laboratory in the U.S.A. At LLNL, a six-inch square phoswich detector array for lung counting is the only one in existence. This system gave the best overall performance in terms average observed/predicted efficiency and standard deviation of any of the systems compared in the first Nb-92m experiment, including the solid state systems (Newton 1984). This was due to the relatively large surface area of the counting array compared with other equipment, making it less dependent on distributional effects within the body. Because of its outstanding performance, this counting system should be included in any future comparative work involving different particle sizes.

Administration of the radioactive particles will be undertaken at AERE in the UK beginning in January 1988. Following rapid clearance of particles not retained in the lungs, the first subject in the study will be available for measurements in the U.S. in February 1988. Additional subjects will then follow throughout the year, extending into 1989. The program is being designed, coordinated, and totally funded by AERE and IAEA. The U.S. laboratories will be involved only in a passive sense, that is in simply counting the subjects after administration of the radioactive particles. No cost to the laboratories will be involved other than personal time required to obtain the counts and to analyze the data.

The information gained from this experiment will be extremely important to the United States. Indeed, some information will not be obtained at all unless the U.S. does participate. For example, performance data on the large surface area array employed at the Lawrence Livermore National Laboratory. PNL also has more sophisticated instrumentation than is available in the UK. LLNL is gearing up with the same type of instrumentation and the experience gained through counting of the Nb-92m subjects will provide LLNL with an improved capability for plutonium lung counting.
A simple interchange of detector calibration data between U.S. and British laboratories, suggested as a possible alternative to U.S. participation in counting the radioactive Nb-92m subjects, would be possible if all detection systems were alike and/or each of the subjects responded in similar fashion to a reference point source which would be used to intercalibrate the detectors. Unfortunately, this is not the case. Each subject is likely to have his own unique distribution of activity in the lungs with calibration data not easily transferable to another system.

By participation in this experiment, we will learn the capabilities and limitations of a phantom that has become an international standard for the calibration of plutonium lung counters. We need to know if the phantom is correct. In many cases, it probably is not. If it is not, we need to know the limitations and how to apply any needed corrections to our data when we do plutonium lung assays for LLNL workers. We also need to inform other DOE facilities which counting systems perform the best for any given situation. There is no better way to do this than to test the phantom against human experience by either counting exposed individuals as a result of an accident (most of these are not useable for primary calibration) or by participating in controlled experiments such as the Nb-92m study, where the materials selected closely simulate plutonium.

Both ANL and PNL have received permission from their management to continue participation in this program. Adding LLNL to the list is strongly recommended, given the degree of input by LLNL as an active participant in the past and the potential benefit to our own whole body counting capability.

REFERENCES

