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SNS SOURCE TERM EVALUATION PROGRAM

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INTRODUCTION

Plutonium-238 is currently used in the form of a pure refractory oxide as a power source on a number of space vehicles that have already been or will be launched during the next few years. Although the sources are designed and built to withstand re-entry into the earth's atmosphere and impact with the earth's surface without releasing any plutonium, the possibility of such an event can never be absolutely excluded. Should the capsule fall into the ocean, the corrosive effect of salt waters must be considered. On land, in addition to the possibility of an immediate release from a fractured capsule, the long-term effects of thermal and humidity cycling must be considered. Should $^{238}\text{PuO}_2$ become mixed in a finely divided form with soil, the resuspension characteristics of the plutonium in conjunction with the soil and the subsequent aerodynamic behavior of the aerosols will be major factors in determination of the fate of the plutonium in the environment.

Thus a range of factors is involved in air analysis of the hazards of plutonium-238 released from SNS power sources. The Biology Department of the Pacific Northwest Laboratories has long and varied experience in the conduct of biological experiments with plutonium compounds, including plutonium aerosol inhalation exposures of animals involving aerosol generation, sampling and characterization, followed by the radiochemical, autoradiographic and other analysis of biological and other samples for plutonium. In addition, a facility is available for the study of interactions between plutonium aerosols and aerosols derived from soil samples. Also available is a large body of biological data from small

and large animals exposed to $^{238}\text{PuO}_2$ and $^{239}\text{PuO}_2$ aerosols over a number of years. These data serve as the base for the computer modeling work conducted for development of predictive dose and hazard estimates of possible future exposure of humans to plutonium aerosols.

The objectives of the present study are confined to four specific tasks:

Task 1 - Aerosol Experimental Studies, which concern interactions between soils and $^{238}\text{PuO}_2$ aerosols which might be created in a space launch abort environment. The specific problem to be solved, analytically and experimentally, is the degree to which the plutonium aerosol source term might be modified in a terrestrial environment and determination of the feasibility of using soil-plutonium aggregates in animal inhalation studies.

Task 2 - Marine Animal Studies, which concern support activities for the SNS underwater experiments at the Navy Undersea Center, including assistance with their technical planning.

Task 3 - Ultrafilterability Studies, concerns in vitro solubility tests of $^{238}\text{PuO}_2$ as a function of time, temperature, suspension concentration and molality of solvent.

Task 4 - Mathematical Modeling, which concerns support of the SNS safety effort to standardize analytical techniques for system risk analysis. The task includes the modeling of biological data obtained from inhalation exposure of rodents and dogs to $^{238}\text{PuO}_2$ aerosols and comparison of the models to that developed for $^{239}\text{PuO}_2$.

PROGRESS DURING FY 1975

Task 1 - Aerosol Experimental Studies

The scope of this task is to study the interactions of $^{238}\text{PuO}_2$ with soils. The work is being carried out in two phases: the first concerns studies of interactions of $^{238}\text{PuO}_2$ aerosols with soil aerosols. The two aerosols are studied separately at first, and are then generated simultaneously and examined for evidence of interactions. The second phase concerns studies of interactions between $^{238}\text{PuO}_2$ and soil when the materials are mixed together, subjected to temperature and humidity cycling, and then aerosolized. Comparisons will be made between the aerosols from each phase of the study.

These studies began in FY 1974 and are being carried out in a low-speed wind tunnel facility which was designed and built for a project concerned with the study of foliar deposition on and uptake of plutonium into plants. The facility has been described in the literature (1,2). Briefly, it is a stainless-steel tunnel having a square cross section varying from 20.3 cm x 20.3 cm at the ends to 30.5 cm x 30.5 cm in the center. The distance from the inlet HEPA filter to the outlet HEPA filter is 178 cm. There is a file-cabinet-type drawer with a 15.2 cm false bottom in the 30.48 cm-square portion of the tunnel for exposing plants to a moving Pu aerosol. The drawer seals to the tunnel wall in the fully open and fully closed positions. (See Figure 1).

The entire wind tunnel is enclosed in an 8 ft x 2 1/2 ft x 2 1/2 ft plexiglass glove box to provide double containment of plutonium aerosols. In operation, a negative pressure differential always exists between the glove box and the room as well as between the tunnel and the glove box.

In these studies soil and $^{238}\text{PuO}_2$ aerosols are generated near the inlet end of the tunnel, isokinetic aerosol samples are taken at several downstream locations, and a number of samples of the settling dust are collected on the tunnel floor.

During FY 1974 13 tests were carried out with variations of four soil types and four wind speeds in the range of 0.4 cm/sec to 22.4 cm/sec. There was no evidence of interaction between the soil and $^{238}\text{PuO}_2$ aerosols. However, variations in the concentration and particle size distribution of the $^{238}\text{PuO}_2$ aerosols resulting from problems in materials handling and generation could have masked the effects of any but strong aerosol interactions.

The work on this task in FY 1975 has been aimed at improvement of techniques of materials handling and aerosol generation and a more careful study of aerosol interactions between two soil types and $^{238}\text{PuO}_2$, at three wind speeds. Experience gained in FY 1974 led us to make certain changes in sampling techniques and equipment and a different scheme was devised to enable us to better examine the effects of interactions of soil upon the $^{238}\text{PuO}_2$ aerosols.

A brief discussion of plans for FY 1976 efforts will be presented later in this report.

Section A - Materials Handling and Aerosol Generation Techniques

The plutonium dioxide used was from the original sample of pure plutonium oxide (PPO) supplied by Los Alamos Scientific Laboratory (LASL). A good part of the effort during the first part of FY 1975 was expended in improvement of procedures for preparation and aerosol generation of

the aqueous suspensions of $^{238}\text{PuO}_2$ particles.

Significant changes in materials handling and $^{238}\text{PuO}_2$ aerosol generation procedures included: (a) removal of much of the large particle and agglomerate fraction of the particles from the suspension by sedimentation, (b) increasing ultrasonic treatment to from 16 to 18 hours before generation and continuing during generation, (c) addition of NH_4OH to the suspension to retard agglomeration, and (d) use of bubbling air to replace the magnetic stirring which could not be used during ultrasonic agitation.

Removal of large particles from suspension was accomplished by settling of the suspension for 1 minute per 3.5 cm of depth. This is approximately the settling rate of a $10\text{ }\mu\text{m}$ sphere having a density of 10.6 g/cm^3 in water. All PuO_2 particles with Stokes diameters of more than $10\text{ }\mu\text{m}$ should be removed from the suspension by this process.

These steps resulted in an increase of ten to twenty times in aerosol concentration as well as in tunnel floor deposition rates with only half the aqueous suspension activity levels used previously. There was a very significant increase in the AMAD of the $^{238}\text{PuO}_2$ aerosols since more of the large particles were aerosolized.

Soil generation and handling techniques were essentially the same as were used in FY 1974 (see Annual Report, 1974).

Section B - Instrumentation and Sample Collection

The same general categories of samples were obtained as in FY 1974; however, the experience gained in FY 1974 led to certain changes in instrumentation and sampling techniques. Filter paper samples and cascade impactor samples were obtained isokinetically, with one

exception which will be explained later.

The filter paper samplers were essentially the same as used in FY 1974, and Figure 2 shows the samplers for the four wind speeds 0.4, 0.9, 5.1 and 22.4 cm/sec studied in FY 1974, although only the 0.9, 5.1 and 22.4 cm/sec speeds were studied in FY 1975.

In FY 1974 the variation in sample rate was achieved by adding dilution air at the impactor inlet so as to maintain a constant total impactor flow rate. This proved to be very difficult to control, especially at the low wind speeds where the sample flow was a small fraction of the total impactor flow. For the Mercer impactor, a set of three sampling nozzles (see Figure 3) were constructed to provide inlet speeds equal to the three wind tunnel speeds (0.9, 5.1 and 22.4 cm/sec) at the normal impactor flow rate. The high flow rate of the Andersen cascade impactor (used for soil aerosol sampling) made it impractical to use nozzles similar to the Mercer's, because the nozzle diameter would be too large even at the 5.1 cm/sec speed. At the 22.4-cm wind speed the full impactor flow entered through the sampling nozzle. At 5.1 cm/sec, 23% of the impactor flow entered the nozzle and 77% was supplied by dilution air. It was not practical to supply 96% dilution air to achieve a 4% sample flow as would be required for isokinetic sampling at 0.9 cm/sec, so the same sampling conditions were used here as for the 5.1-cm/sec wind speed. This was the case only during the low-speed soil-only tests. The Andersen impactor with sampling nozzle is shown in Figure 4.

Collection of samples on the tunnel bottom was expanded by taking additional samples at the sides of the tunnel to determine the degree

of lateral dispersal of the PuO_2 aerosol. Samples were taken upstream of the generator to determine the amount of deposition in that region. Diagrams of sampler locations in the tunnel are shown in Figures 5 through 9.

Isokinetic filter paper samples and sedimentation samples were taken for short periods to obtain amounts of activity low enough for autoradiographic analyses. The filter paper samples were taken in the same manner as the regular filter paper samples except that much shorter collection times were used, ~ 5 seconds compared to 15 minutes. The special sedimentation samples were taken on millipore HAWP filter paper placed on the plunger of a plastic syringe which was pushed through a hole in a rubber stopper inserted in a port in the tunnel wall. A hole was cut in the syringe tube so that when the plunger was depressed the filter paper was located under the hole in the syringe wall, and collected particles settling onto the surface. At the end of the sampling time the syringe plunger was withdrawn, retracting the filter paper from the hole and sealing it. These samples were taken for approximately 1 second. Even with such short sampling times the activity on these samples was too high for good autoradiographic analyses. The upper limit of activity for good analyses is, according to Dr. Soinski of the LFE lab, who examined the samples, about 2 nanocuries per filter paper.

Section C - Sample Analysis

In FY 1974 alpha scintillation was used for $^{238}\text{PuO}_2$ analysis of most samples. This required ashing the samples for removal of organic

material, acid digestion and mixing with a scintillator to prepare for counting. The process is time-consuming, expensive and, due to the difficulty of acid digestion of high-fired PuO_2 , not as reliable as one could hope for. In FY 1974 the Mercer impactor samples were sealed in transparent tape and gamma counted. Since the activity level of our samples is fairly high, this method is reasonably reliable, quick and inexpensive. The reliability was increased by preparation of standard $^{238}\text{PuO}_2$ sources which we counted with each set of samples to ensure that the correct counting efficiency factor was used in computing sample activity. In FY 1975 all $^{238}\text{PuO}_2$ samples were analyzed in this way.

For soil sample analysis in FY 1974 trace metal analysis of the acid-digested samples was used as described above. Considerable difficulty and delay was encountered in the trace metal analysis because of the limited amount of atomic absorption spectrophotometry apparatus available for plutonium contaminated samples. Also, in FY 1974, the analytical results were disappointing due to high background levels of trace metals in the samples. The only available alternative to trace metal analysis was gravimetric analysis. This required sample collection times of up to 7 hours in some cases. For this reason the soil aerosol characterization was obtained from several soil-only aerosol tests, each of several hours duration.

Section D - Description of the Experiment

While considerable improvement was achieved in the efficiency of $^{238}\text{PuO}_2$ aerosol generation, the $^{238}\text{PuO}_2$ suspension concentrations did

not remain constant during generation periods of 20 minutes or longer nor did the concentration and particle size of the generated aerosols. Furthermore, although attempts were made to standardize procedures, considerable variation in $^{238}\text{PuO}_2$ suspension and aerosol characteristics occurred from one day to another.

The day-to-day variations in PuO_2 particle suspensions made it impossible to study PuO_2 aerosols one day and PuO_2 + soil aerosols the next and be able to make reliable comparisons between the two. The only alternative was to make shorter runs with PuO_2 -only, followed as quickly as possible by runs with the PuO_2 + soil aerosol for comparison. Before this, however, we had to establish what changes could be expected from one run to the next with aerosols of PuO_2 only.

An experiment was carried out with each of two soil types at each of three wind speeds. Such an experiment included four tests, each of which consisted of four runs. All the runs of one test were made consecutively, during the same day, using the same aerosol generators, the same soil sample and the same PuO_2 suspension. In two of the tests in an experiment, PuO_2 -only aerosols were generated during all four runs. In the other two tests during runs 1 and 3, only PuO_2 aerosols were generated, while PuO_2 + soil aerosols were generated during runs 2 and 4.

The first two tests of an experiment were intended to establish a pattern of change of PuO_2 aerosol characteristics from run to run in the absence of a soil aerosol. Having established the pattern, the results of runs 1 and 3 were compared with 2 and 4 of the second two tests to determine what modification of the PuO_2 aerosols could be attributed to the presence of a soil aerosol.

As previously stated, the aerosol studies were carried out at three wind speeds (0.9, 5.1 and 22.4 cm/sec) with two soil types. The soils were two of four types supplied by LASL in FY 1974. Soil types studied were identified as "Los Alamos Soil" (Soil 1) and "Finely Divided Clay Soil" (Soil 3).

Characterization of all four types was carried out at the University of Washington Soils Laboratory. The results of this work are presented in Table 4.

The soil aerosols were generated by the Wright Dust Feed Mechanism* operating at a gear ratio of 1 to 1 in each case.

For soil-only aerosol studies, clear water was run through the PuO_2 generator during soil aerosol generation to match the conditions during the PuO_2 + soil aerosol studies.

Aerosol and sedimentation samples were collected in the same locations and in much the same manner as for the PuO_2 aerosols, except that the Andersen impactor was used instead of the Mercer impactor to obtain the particle size distribution. An Ainsworth Model 24N balance** reserved for plutonium-contaminated materials was used to weigh the soil aerosol and sedimentation samples.

Section E - Experimental Results

Three filter paper aerosol samples and the total cascade impactor collection were used to determine aerosol concentrations at four points in the tunnel. From these values a graphical estimate of the aerosol

* Product of L. Adams Ltd., Minerva Road, London, N.W. 10, England.

** Product of Ainsworth, Inc., 5151 South Syracuse St., Englewood, Co 80110

concentration at 125 cm downwind of the generator was made. The sedimentation samples from the tunnel floor were used to construct a plot of the PuO_2 activity deposition rate as a function of tunnel position. From this, a graphical integration yielded a value for the total PuO_2 deposition between the generator and a point 125 cm downwind from it. From these data, the percentage of injected activity still airborne at 125 cm downwind was determined for each run.

Figures 5, 6, and 7 show curves of plutonium activity deposition rate on the tunnel bottom vs. the distance from the generator outlet. The deposition rates decrease noticeably from run 1 to run 4 during a test. Comparison of deposition rates in Figures 4 and 5 (0.9 cm/sec) with those in Figure 6 (22.4 cm/sec) illustrates the much lower deposition rates and greater aerosolization of the plutonium particles at the higher speed.

The activities on the eight stages of the Mercer cascade impactor were measured by gamma counting and these data were computer analyzed by PNL's NEWCAS (3) program for cascade impactor data. This program determines the activity distribution with respect to aerodynamic equivalent diameter (AED), compares the distribution to a log normal distribution and makes the best fit of the data to a log normal distribution for which the AMAD and geometric standard deviation are determined. $\Sigma \chi^2$ values in excess of 9 generally indicate the distribution is not log normal and the GSD is not a valid parameter of the distribution.

Table 1 shows PuO_2 particle size data and percent of injected activity still airborne at 125 cm downwind. Note that the aerosol particle size increases with wind speed. During a test there is

usually a monotonic decrease in AMAD and aerosol concentration with time after the start of generation. Notable exceptions occurred during tests 33 and 37 at 0.9 cm/sec wind speed, where the PuO_2 aerosol AMAD was increased by the presence of the soil 3 aerosol.

The cascade impactor data were examined and the partial aerosol concentrations for particles larger than $2.2 \mu\text{m}$ and smaller than $2.2 \mu\text{m}$ were separately determined. The ratios of these concentrations in runs 2, 3, and 4 to the corresponding values for run 1 of each test were computed. If, for example, the partial aerosol concentration for the smaller particles (Group A) was greater in run 2 than in run 1, the ratio would be greater than 1; if less in run 2 than in run 1, the ratio would be less than 1.

In Table 2 these normalized partial aerosol concentrations are given for PuO_2 aerosols and for PuO_2 + soil aerosols at different wind speeds. The normalized partial concentrations appear to be nearly independent of wind speed for PuO_2 -only aerosols.

Table 3 presents the soil aerosol data for each soil type and wind speed condition. Included are particle size data and aerosol percent at 125 cm downwind of the generator. Figures 8 and 9 are typical soil deposition patterns on the tunnel floor for soils at two different wind speeds.

Section F - Discussion of Results

It is evident from the data in Table 1 that no large amount of $^{238}\text{PuO}_2$ aerosol is scavenged by the soil particles which settle onto the surface a short distance from the generator. When one examines the percentage of injected ^{238}Pu activity in the aerosol at 125 cm downwind

from the generators it is difficult to observe any significant or consistent differences between runs with $^{238}\text{PuO}_2$ -only and $^{238}\text{PuO}_2$ + soil. It is significant that even at a modest wind speed of 22.5 cm/sec, over 90% of the injected Pu activity is still airborne at 125 cm downwind from the source.

Lacking any gross effect on PuO_2 aerosol concentration due to soil interaction, attention was then directed to more subtle effects such as changes in particle size distribution from PuO_2 -only to PuO_2 + soil runs. In the case of tests 33 and 37 (soil #3 + PuO_2 at 0.9 cm/sec), in the PuO_2 -only runs (1 and 3), the $^{238}\text{PuO}_2$ aerosol had smaller AMAD for PuO_2 than it did in runs 2 and 4, where PuO_2 + soil 3 were generated. Such an effect was not evident in the tests with soil 1 at 0.9 cm/sec, nor in any of the tests at higher wind speeds. The higher wind speeds caused greater aerosol dilution and shorter times for interactions to take place.

A scheme was devised to look for still more subtle evidence of aerosol interaction between soil and $^{238}\text{PuO}_2$, as outlined in Section E. Since considerable variability existed in $^{238}\text{PuO}_2$ aerosol concentration and aerosol particle size distribution from one test to another, the aerosol properties observed during runs 2, 3 and 4 were "normalized" to the properties during run 1 of each test. In particular, the aerosol concentrations of particles having an aerodynamic equivalent diameter less than 2.2 μm were determined and normalized as described above. The same was done for the aerosol concentrations of particles having aerodynamic diameters greater than 2.2 μm . All the tests (6 sets of four twenty-minute runs) in which $^{238}\text{PuO}_2$ -only aerosols were generated were examined

and averaged. As can be seen in Table 2 there was a tendency for the large particle concentration to decrease during a test, while there was a small increase in small particle concentration.

It is interesting to note that the data at all wind speeds followed the same general pattern. Runs 2 and 4 with PuO_2 + soil depart significantly from the pattern established for PuO_2 -only runs, and the lower the wind speed the more pronounced the departure. Soil 3 displays a stronger effect of aerosol interaction than soil 1. The mean partial PuO_2 aerosol concentrations relative to the run 1 values are given with standard deviations for the PuO_2 -only runs. Since no more than two tests were conducted at a given wind speed with either soil type, the two values are given (rather than their mean).

The conclusion one might draw from these results is that attachment of some PuO_2 to soil particles results in effectively increasing the AED of the PuO_2 particles. This may have some significance with respect to respirability of the PuO_2 aerosols. To corroborate this finding will require more detailed analysis of the cascade impactor data with weighting of each size fraction according to the ICRP curves for alveolar deposition.

Section G - Future Plans

The second phase of the aerosol studies will begin early in FY 1976. In this phase we will determine the properties of aerosols generated by dry dust techniques from mixtures of $^{238}\text{PuO}_2$ and soils. The aerosol studies will be carried out in the wind tunnel immediately after mixing and again following periods of aging, during which the mixtures will be subjected to a program of temperature and humidity cycling.

Some difficulty has been encountered in satisfying safety requirements in handling $^{238}\text{PuO}_2$ + soil mixtures during the mixing, weighing, temperature and humidity cycling operations. Many of these problems have been ironed out and generation of aerosols of soil + PuO_2 mixtures should begin early in FY 1976.

An aerosol mixer into which both the PuO_2 and soil generators will feed is being designed and built. It is hoped that this will result in improved aerosol mixing and an enhancement of aerosol interactions. This device will also improve the lateral dispersal of the soil aerosols, and will be used for studies with aerosols of PuO_2 + soil mixtures.

The problems of developing a system for microscopic and/or photographic study of aerosol interactions was deemed so complex as to be beyond the reach of the project. Some recent developments in laser microscopy may warrant a reappraisal of the problem.

It appears that soil particles are not efficient scavengers of $^{238}\text{PuO}_2$ particles. The larger PuO_2 particles are probably least affected by the soil particles, therefore it is suggested that further soil- PuO_2 aerosol interactions be limited to $^{238}\text{PuO}_2$ aerosols within the respirable range (i.e. $< 10 \mu\text{m AED}$). Emphasis should be placed on the changes of respirability of the PuO_2 aerosols effected by the presence of a soil aerosol. Inhalation studies could be performed with test animals to study not only the effect of soil aerosol upon the deposition of $^{238}\text{PuO}_2$ but also on the pulmonary clearance rates.

Lower Pu activity concentrations will result in smaller sample activity, which will ease handling problems and improve autoradiographic analysis.

Task 2 - Marine Animal Studies

During the past year, 43 sea cucumbers and one marine snail were received by this laboratory and submitted for radiochemical analysis. The animals were periodically collected from the pure plutonium oxide (PPO) solubility test site off San Clemente Island under the direction of Dr. Frank Kawahara of the Navy Undersea Center, San Diego, California. Numerous sea cucumbers were collected from the ocean floor in that area at varying distance from the vessels in which PPO pellets are contained. Others had been exposed to the PPO in the test vessels for varying lengths of time. In April 1974, two new test vessels with new sand floors and new PPO pellets were put into use. Most of the exposed sea cucumbers analyzed during the past year were from these two vessels. Attempts were made to sample sand and water from those test vessels at periodic intervals. Analysis of sand samples are not yet complete and the data from analyses of water samples presented below are inconclusive (Table 5). Sampling procedures are difficult and are not likely to yield consistent results unless both sand and water could be sampled in a uniform manner prior to each raising of the test vessel.

Alpha activity in sea cucumbers collected outside of and at varying distances from the test vessels are shown in Table 6. Organisms from locations described as "outside cages" were collected in the immediate area of the exposure vessels. They had total body burdens ranging from 573 to 6,359 dpm with the exception of animal 10, whose body burden was

17,738 dpm. Animals collected approximately 100 feet from the cages contained comparable amounts of alpha emitters but two key samples from that group of animals were lost during the ashing process so that total body burdens could be calculated from only three of the five animals. Sea cucumbers listed as having been collected from "Wilson Cove" were obtained approximately 1400 yards from the test area and contained generally lesser amounts of activity with the exception of animal 23 (85,983 dpm). No explanation can be given for the unusually high concentrations of alpha emitters in animals 10 and 23. Precautions against cross contamination are taken when the animals are collected as well as when they are dissected. It is noted that most of the body burdens of those animals was concentrated in the gut and the body walls, as is generally true for all of the sea cucumbers analyzed. Unfortunately, autoradiographs were not made from organs of these two organisms because the high activity levels were not suspected prior to radiochemical analysis. The absence of autoradiographs precludes more specific location of the sites of high activity. Repeat analysis of selected samples is underway as are alpha spectrometric analyses. This combination of analyses should answer two questions: (1) is the high activity a result of the inclusion of a large plutonium particle or aggregate of particles?, and (2) is the activity due to plutonium-238, plutonium-239 or some other alpha emitter of unknown origin?

A 32-gram marine snail was found attached to the exterior of cage #3 when it was raised for the August 1974 inspection. Analysis of the snail revealed a body burden of 192 dpm or 6.0 dpm per gram; however,

there was no way of knowing how long the snail had been in its location.

Alpha activity present in the organs of sea cucumbers which had been confined in test vessels #3 and #4 are presented in Tables 7 and 8, respectively. The numerical designations for the animals are related only to the order in which they were received by this laboratory. The animals were collected between June 1974 and June 1975, and were confined in the test vessels for periods ranging from 49 to 105 days. There is a general trend toward higher body burdens in animals confined in test vessels later in the course of the experiment. Burdens of approximately 0.5 microcuries were present in two animals from each vessel, and one animal from cage 3 (#35) contained more than 1.0 microcuries of alpha activity. No relationship between length of confinement in the vessels and total activity appears to exist, however. Body burdens are concentrated in the stomach, gut, and on the body wall of each animal with some exceptions. Animals from cage 4 had considerably higher activity in the respiratory trees, and two of those animals (#43 and #44), as well as one animal from cage 3 (#42), had very high activity present in the gonads. Selected samples from these two groups of animals are being analyzed for plutonium-239 so that isotopic ratios of plutonium-238 to plutonium-239 can be calculated.

During dissection of the specimens, it was noted that a large number of sea cucumbers had no recognizable gonadal tissue. The gut was absent or considerably atrophied in many of the same animals, and in others. A definite relationship was found between gonadal absence and the time of year in which the animals were collected. Data from Tables 6, 7 and 8 indicated that with few exceptions animals collected between

December and April had no gonads. In some cases, the absence of reproductive capability apparently extends into June, although gonads were present in most animals collected in April and June as well as in the few animals collected in August and September.

Data in Table 6 allows no inference of seasonal variation in the gut size in sea cucumbers. However, it was noted that the guts of several animals which had undergone confinement in test vessels were atrophic. This is illustrated by data in Table 9, which includes mean wet weights and ranges of organs of sea cucumbers from each of the cages. There are at least two possible explanations for the observations: (1) there is a radiation effect on the gut, or (2) confinement in the test vessel somehow limits the grazing ability of the animals. The radiation effect is plausible because of the high alpha activity generally found in the gut if the gut is analyzable. Microscopic examination of gut slides from animals received in the future will be done in an attempt to verify whether a radiation effect exists. Another approach to solution of that problem would be to set up a "control" vessel in which sea cucumbers could be confined without a PPO pellet. If gut atrophy occurred in animals from that cage, a radiation effect in the test vessel animals would seem less likely. There are no apparent differences among mean weights of other organs from the sea cucumbers collected.

Another PPO solubility study was begun in April 1975, with two different test vessels. In vessel #1 a PPO pellet with vented cladding was placed, while vessel #2 contained a PPO pellet with unvented cladding. Two sea cucumbers were placed in each test vessel. Forty-nine days

later, water samples were taken from the vessels and the sea cucumbers were removed. Data from the analysis of organs of the animals are presented in Table 10. The activity present in animals from cage #2 (unvented sample) appears to be confined primarily to their body walls. Animals from cage #1 (vented sample) contained considerably more radioactivity in their respiratory organs and digestive tracts. Water samples from each of the cages contained radioactivity below the background level of the detection instrument.

Task 3 - Ultrafilterability Studies

Little experimental work has been done, but the approach or procedure for testing the ultrafilterability of PuO_2 has been updated.

Several requirements must be explored before the actual long-term experiment can be initiated. Control of pH will be a major problem. It has been found in previous studies that at high activity concentrations, such as 100 $\mu\text{Ci/ml}$, the pH decreases as a function of time. Storage of the suspensions could also be a problem. The ideal would be to have two controlled environments, each having the necessary equipment to provide motion and being large enough to hold all the necessary suspensions. Because of the high level of activity involved, certain radiation safety requirements would have to be satisfied.

Experimental conditions would cover three major variables: concentration, solvent system, and temperature. There will be three concentrations: $C_1 = 100 \mu\text{Ci/ml}$, $C_2 = 100 \text{ nCi/ml}$, and $C_3 = 100 \text{ pCi/ml}$; two solvent systems: $S_1 = \text{seawater}$, $S_2 = \text{distilled water}$; two temperatures, $T_1 = 10^\circ\text{C}$, $T_2 = 37^\circ\text{C}$. These correspond to the temperatures of the ocean bottom and of the human body. It is necessary to have duplicate sets of suspensions plus one backup set, all of which will be treated identically. The backup set will be processed in the same manner as the others, but will not be sampled. Aliquots (2 ml) will be drawn at time intervals of 0, 1, 2, 4, 8, 16 and 32 weeks. On each sampling date three aliquots from each suspension will be placed into specially prepared centrifuge tubes and centrifuged at 2000 rpm for 60 minutes. Concentrations of the sample activity must be determined by counting several suitable aliquots in a liquid scintillation counter. Ideally, the more

aliquots taken from each suspension at each testing date, the better the statistics. The number of aliquots will dictate the amount of original suspension needed. Because of the low level of activity involved, the concentration of the C_3 suspensions will have to be checked to assure reproducibility.

A system has been worked out specifying the numbers of centrifuge tubes, suspension volumes and samples to be run. The smaller items of equipment needed for this task have been ordered.

Task 4 - Mathematical Modeling

The SNS Source Term Evaluation Program required development of a mathematical model to describe the translocation, buildup and excretion of inhaled $^{238}\text{PuO}_2$ in dogs. We started with a model for inhaled calcined $^{239}\text{PuO}_2$ in dogs which has been under development here for several years. This model was operational on hybrid computer and is coded in a digital simulation language called MIMIC, which operates on a CYBER-74 computer.

Since it was suspected early in the project that the model for inhaled $^{239}\text{PuO}_2$ would not match the data for inhaled $^{238}\text{PuO}_2$ and that modifications would be required, the all-digital simulation was eliminated as the development medium in favor of one with an interactive environment providing quick turnaround at reasonable cost. We also substituted a new state-of-the-art digital minicomputer system with a graphic output screen for the analog computer portion of the hybrid computer initially used on the project. (More detailed information is contained in Appendix A of this report).

The early part of the project was devoted to installing the inhaled $^{239}\text{PuO}_2$ model for dogs on the MINERVA (Mobile Interactive Equipment for Research using Visual Assistance) system, creating an integration routine and comparing the results with output from the previous hybrid and digital computer runs. Several graphic displays were created and the model was available for use.

The SNS data was loaded and some initial comparisons with the model were made. The parameters of the model were adjusted to see if an adequate fit of the model to the data could be achieved; it could not. It will be necessary to alter the basic form of the $^{239}\text{PuO}_2$ inhalation

model so that it will match the $^{238}\text{PuO}_2$ data, which will be attempted in the coming year.

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Appendix A

BIOLOGY MODEL DOCUMENTATION

INTRODUCTION

Two biology models have been implemented on the BNW MINERVA System. Both of these models simulate the flow of inhaled plutonium through the organs of the body. The first model was developed by Dionne and Stuart⁽¹⁾ and originally programmed on the BNW hybrid computer. The second was derived from the block diagram of a model developed by the ICRP Task Group⁽²⁾. The simulation programs for both models are quite similar and emphasize user-oriented control and display techniques.

THE MINERVA SYSTEM

The Battelle MINERVA System consists of a PDP-11/35 minicomputer with 24K of core and a Vector General graphics display terminal. Program and data storage is maintained on two removable disk packs which together have a storage capacity of 4.8 million bytes. The system has the capability to produce hard copy output of graphs and data appearing on the Vector General display. The output medium for these displays is either an electrostatic printer/plotter or a CalComp plotter. The major advantage of this system for the two Biology models which are being simulated is fast real-time man-machine interaction.

THE MODEL

Both models were developed using a series of first order linear differential equations. These equations simulate the flow of material

between various organs of the body after initially being inhaled into the lungs. The general form of these equations is

$$(dN/dt)_i = f_k \lambda_j N_j - \lambda_i N_i$$

where

$(dN/dt)_i$ is the time rate of change of burden in the i^{th} organ,

f_k is the fractional uptake parameter. (In the Dionne-Stuart model, $k = i, j$ specifying the fractional uptake by the i^{th} organ from the j^{th} organ. In the ICRP model, k specifies the fractional amount leaving the k subcompartment).

λ_j is the biological elimination rate of the j organ, and

λ_i is the biological elimination rate of the i^{th} organ.

DIONNE - STUART EQUATIONS (5)

Pulmonary

The equation describing the varying biological half-time of the deep lung is

$$\dot{\lambda}^{PV} = -(\lambda)(\lambda^{PV}).$$

λ is the term which affects the variability.

The equation describing the varying portion of the deep lung burden is

$$\dot{N}_{PV} = -\lambda^{PV} N_{PV}.$$

where N_{PV} is a term describing the amount of material. This portion has an initial rapid half-time of 50 days and it continually lengthens, with a half-time of one year (λ). The material leaving the lung through this compartment enters the GI tract. One year after deposition no further material leaves this compartment.

The remaining material in the deep lung is described by the following equation:

$$\dot{N}_{PC} = -\lambda^{PC} N_{PC}.$$

The material leaving the lung by this pathway enters the lymph nodes, blood stream, and GI tract. The biological half-time for this pathway is 4 years. Incorporation of recently acquired data suggests that this may be shortened to 3 years.

$$N_P = N_{PV} + N_{PC}.$$

Lymph Nodes

The uptake of material in the lymph nodes is complicated by the fact that only 30% of the material received by the lymph nodes leaves them for subsequent translocation to systemic organs. Let N_{TB1} represent the amount of material leaving the tracheobronchial lymph nodes and entering the bloodstream, while N_{TB2} represents the amount retained in the tracheobronchial lymph nodes. The rate of change of material in the first compartment is

$$\dot{N}_{TB1} = f_{TB,NPC} \lambda^{PC} N_{PC} - \lambda^{TB1} N_{TB1}.$$

The rate of change of material in the second tracheobronchial lymph node compartment is described by

$$\dot{N}_{TB2} = f_{TB,TB} \lambda^{TB1} N_{TB1}$$

and the total amount of material in the tracheobronchial lymph nodes at any time is

$$N_{TB} = N_{TB1} + N_{TB2}.$$

Similarly, for the abdominal lymph nodes, the rate of change of material in the first compartment is

$$\dot{N}_{A1} = f_{A,NPC} \lambda^{PC} N_{PC} - \lambda^{A1} N_{A1}.$$

The rate of change of material in the second abdominal lymph node compartment is described by

$$\dot{N}_{A2} = f_{A,A} \lambda^{A1} N_{A1}$$

and the total amount of material in the abdominal lymph nodes at any time is

$$N_A = N_{A1} + N_{A2}$$

Blood

The amount of material in blood at any time is the sum of the material going to blood from any organ. Because blood has a fast biological half-time with respect to the rest of the organs in the long-term model it transfers the material immediately, and in the proper proportion, to the organs it feeds.

$$\begin{aligned} \dot{N}_{BL} = & f_{BL,NPC} \lambda^{PC} N_{PC} + f_{BL,TB1} \lambda^{TB1} N_{TB1} + f_{BL,A1} \lambda^{A1} N_{A1} \\ & + f_{BL,BO} \lambda^{BO} N_{BO} + f_{BL,LV} \lambda^{LV} N_{LV} + f_{BL,KI} \lambda^{KI} N_{KI} + f_{BL,OT} \lambda^{OT} N_{OT} \end{aligned}$$

Systemic Organs

The equations describing the buildup and decay of material in the systemic organs are all similar. They are fed by blood and send their material back to blood with long half-times.

Bone

$$\dot{N}_{BO} = f_{BO,BL} \dot{N}_{BL} - \lambda^{BO} N_{BO}$$

Other

$$\dot{N}_{OT} = f_{OT,BL} \dot{N}_{BL} - \lambda^{OT} N_{OT}$$

Liver

$$\dot{N}_{LV} = f_{LV,BL} \dot{N}_{BL} - \lambda^{LV} N_{LV}$$

Kidneys

$$\dot{N}_{KI} = f_{KI,BL} \dot{N}_{BL} - \lambda^{KI} N_{KI}$$

Initial material in the systemic compartments comes from the soluble portion of the material which is deposited in the nasopharynx-tracheo-bronchial compartment of the short-term model.

Excretion

The equations which describe the amount of material excreted are designed to yield the total excreted since the initial uptake.

Urine

$$\Sigma \dot{N}_{UR} = f_{UR,KI} \lambda^{KI} N_{KI} + f_{UR,BL} \dot{N}_{BL}$$

Feces

$$\Sigma \dot{N}_{FE} = f_{FE,LV} \lambda^{LV} N_{LV} + f_{FE,NPV} \lambda^{PV} N_{PV} + f_{FE,NPC} \lambda^{PC} N_{PC}$$

Whole Body

The amount of material in the whole body at any time is simply the sum of the amount of material in all the compartments.

$$N_{WB} = N_P + N_{TB} + N_A + N_{BO} + N_{OT} + N_{LV} + N_{KI}$$

EQUATIONS FOR ICRP MODEL

Nasopharynx

Two differential equations describe the flow of material from this compartment:

$$\dot{N}_{NPA} = -\lambda^{NPA} N_{NPA}$$

$$\dot{N}_{NPB} = -\lambda^{NPB} N_{NPB}$$

where N_{NPA} and N_{NPB} describe the amount of material. N_{NPA} determines the portion passing into the blood while N_{NPB} describes the amount

entering the gastrointestinal tract. The total for the nasopharynx at any time is

$$N_{NP} = N_{NPA} + N_{NPB}$$

Tracheobronchial

There are also two equations describing the flow of material in this compartment. N_{TBC} describes material entering the blood and N_{TBD} describes that for the gastrointestinal tract. These equations are:

$$\dot{N}_{TBC} = -\lambda^{TBC} N_{TBC} \text{ and}$$

$$\dot{N}_{TBD} = -\lambda^{TBD} N_{TBD},$$

and the total for the tracheobronchial compartment is

$$N_{TB} = N_{TBC} + N_{TBD}.$$

Pulmonary

The pulmonary compartment contains four paths for material flow.

Two of these,

$$\dot{N}_{PF} = -\lambda^{PF} N_{PF} \text{ and}$$

$$\dot{N}_{PG} = -\lambda^{PG} N_{PG},$$

describe the flow of material into the gastrointestinal tract, while

$$\dot{N}_{PE} = -\lambda^{PE} N_{PE}$$

describes that entering the blood, and

$$\dot{N}_{PH} = -\lambda^{PH} N_{PH}$$

describes the flow into the lymph nodes. The total amount of material in the pulmonary compartment is given by

$$N_P = N_{PE} + N_{PF} + N_{PG} + N_{PH}.$$

Lung

The material dispersed from the lungs after initial inhalation is the sum of the material in the nasopharynx, tracheobronchial and pulmonary compartments and is given by

$$N_{LU} = N_{NP} + N_{TB} + N_P$$

Gastrointestinal Tract

The GI tract is divided into four compartments: the stomach, the small intestine, the upper large intestine and the lower large intestine. The equations for these compartments describe the buildup and removal of material:

Stomach

$$\dot{N}_{ST} = F_b \lambda^{NPB} N_{NPB} + F_d \lambda^{TBD} N_{TBD} + F_F \lambda^{PF} N_{PF} + F_G \lambda^{PG} N_{PG} - \lambda^{ST} N_{ST}$$

Small Intestine

$$\dot{N}_{SI} = \lambda^{ST} N_{ST} - \lambda^{SI} N_{SI}$$

Upper Large Intestine

$$\dot{N}_{ULI} = \lambda^{SI} N_{SI} - \lambda^{ULI} N_{ULI}$$

Lower Large Intestine

$$\dot{N}_{LLI} = \lambda^{ULI} N_{ULI} - \lambda^{LLI} N_{LLI}$$

Lymph Nodes

The lymph nodes compartment is fed by the pulmonary compartment and drains into the blood. The equation is

$$\dot{N}_{LY} = F_h \lambda^{PH} N_P - \lambda^{LY} N_{LY}$$

Blood

The equation which describes the amount of material in the blood is

$$\begin{aligned}\dot{N}_{BL} = & F_A \lambda^{NPA} N_{NPA} + F_C \lambda^{TBC} N_{TBC} + F_E \lambda^{PE} N_{PE} \\ & + F_i \lambda^{LY} N_{LY} - \lambda^{BL} N_{BL}\end{aligned}$$

The blood compartment feeds the systemic and urine compartments with the following equations:

Systemic

$$\dot{N}_{SY} = F_{SYBL} \lambda^{BL} N_{BL} \quad \text{and}$$

Urine

$$\dot{N}_{UR} = F_{URBL} \dot{N}_{BL}$$

Whole Body

The amount of material in the whole body is given by the sum of all compartments except urine, and is given by

$$N_{WB} = N_{LU} + N_{LY} + N_{ST} + N_{SI} + N_{ULI} + N_{LLI} + N_{BL} + N_{SY}$$

DATA

Experimental data for the Dionne-Stuart model was gathered by D. Craig⁽³⁾. This data is composed of results on 58 dogs, 23 of which were exposed by inhalation to ^{238}Pu crushed microspheres and the remainder exposed by inhalation to $^{238}\text{PuO}_2$ from calcined oxalate. The time span for which this data is available is for the period from 7 to 2133 days with the majority of the data points falling in the lower half of the range. This data consists of burden values for the lungs, tracheobronchial lymph nodes, abdominal lymph nodes, skeleton (bone), liver, kidneys, urine and feces.

The ICRP model is being used to simulate experimental data values collected by J. Ballou⁽⁴⁾ on dogs exposed by inhalation to ^{239}Pu . This data is available for the period between 1 and 100 days. Feces and

urine data were gathered daily for most of the time period while data points for blood, lungs, liver, tracheobronchial lymph nodes, skeleton and residual soft tissue are available only at widely scattered points within the time period.

THE COMPUTER PROGRAMS

The computer programs which simulate both of these models are highly user-oriented. All control of the models is handled using simple commands through a keyboard attached to the Vector General graphics display CRT. These commands allow the user to: set the control mode, modify any of the equation parameters, and choose any two of the equations for display.

In the compute mode the computer solves the differential equations continuously while plotting the two solutions which were selected for display. When the equations have been solved to the end of the simulation, the initial conditions are reset and the solving is restarted. During the compute mode the user is free to choose new equations for display or modify the equation parameters. These changes will be implemented when the current simulation is completed and the initial conditions are reset. This process of continuously monitoring the solutions of the differential equations while modifying equation parameters allows the user to rapidly assess the implications of his modification on the rest of the model.

The design of the computer simulation programs allows a high degree of interaction between the keyboard monitor which accepts and interprets commands and the differential equation solver. This interaction is

obtained by requiring that the differential equation solver interrogate the keyboard monitor after each iteration on the time step. The result of this procedure is that commands inputted from the VG keyboard are responded to very rapidly, thus giving the user the impression that the two modules are operating simultaneously.

SOLUTION METHOD

Both biology models use the modified Euler method. The main consideration for the choice of this method was speed of solution. Also the Euler method is self-starting and, due to the characteristics of the system of equations, is still within the required accuracy constraints.

The modified Euler method is a predictor-corrected method. The predictor used is:

$$Y_{i+1}^{(p)} = Y_i + h Y_i'$$

and the corrector step is

$$Y_{i+1}^{(c)} = Y_i + h(Y_i' + H_{i+1}^{(p)})/2.$$

A variable integration step size was also incorporated into the method. The step size to be used is determined by an estimate of the truncation error as follows:

$$EN+1 = \max_i \frac{Y_{i, n+1}^{(p)} - Y_{i, n+1}^{(c)}}{14 D_i}$$

$$D_i = \max_i (1/Y_{i, n+1}^{(c)}, 1, 1.0)$$

where $Y_{i, n+1}^{(p)}$ is the predicted value for equation i at the $N+1$ step and $Y_{i, n+1}^{(c)}$ is the corrected value for equation i at the same time step.

There are two internal parameters which determine when to change the step size. These are the upper bound $\bar{E}$ and the lower bound $\underline{E}$ on the truncation error. The integration step will be halved, left as it is, or doubled according to the following inequalities:

- (a) If $E_{n+1} \geq \bar{E}$, the interval is reduced to $\frac{1}{2}\Delta t$ or $t_{\min}$, the minimum allowable value of the integration step.
- (b) If $\underline{E} \leq E_{n+1} < \bar{E}$, the integration step remains fixed.
- (c) If $E_{n+1} < \underline{E}$ for four successive steps, the step size is increased to $2\Delta t$ or $\Delta t_{\max}$, the maximum allowable value of the step-size.

METHOD

Three different sets of parameters can be modified while running these simulation models. They are the initial conditions on the differential equations, the fractional uptake parameters, and the biological elimination rates. The procedure followed in attempting to fit the model to the experimental data points involves adjusting the different sets of parameters until the solution curves match the data points. The process is started by first picking some likely candidates for the parameters and then running the model and examining the fit between the actual and predicted results. As the simulation is running, modifications can be made to any of the parameters and the effects can be monitored immediately. Since the effect of changes made to one compartment not only alter the solution curve in that compartment but also in all the compartments chained to that compartment,

the ability to rapidly examine any of the solution curves is essential for the practical use of these simulation models. This ability to rapidly monitor the effects of parameter changes on the simulation was a prime consideration in the design of the programs. Consequently the time required to fit the model to the data is minimized by this (interactive) approach over the other methods of solution.

APPENDIX REFERENCES

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Table 1. ²³⁸PuO₂ Aerosol Data

Test No.	Run No.*	Aerosol Type	Wind Speed cm/sec	Particle Size Data			Aerosol at 125 cm***	
				AMAD μm	GSD	ΣX ² **	Concentration nCi/l	% of Total Activity
19	1	PuO ₂	0.9	5.17	2.59	0.67	4900	62
	2	PuO ₂	0.9	4.41	2.44	1.01	4750	64
	3	PuO ₂	0.9	4.03	2.39	1.50	3650	69
	4	PuO ₂	0.9	3.99	2.52	2.21	3100	76
20	1	PuO ₂	0.9	3.20	2.23	1.59	2700	75
	2	PuO ₂	0.9	2.73	2.10	1.89	2400	82
	3	PuO ₂	0.9	2.45	2.07	2.52	2100	86
	4	PuO ₂	0.9	2.52	2.10	2.95	2000	82
21	1	PuO ₂	0.9	4.87	2.41	1.36	5500	52
	2	PuO ₂ + Soil 1	0.9	5.02	2.33	0.46	4600	49
	3	PuO ₂	0.9	4.33	2.39	1.75	4600	51
	4	PuO ₂ + Soil 1	0.9	4.21	2.28	1.29	5600	59
22	1	PuO ₂	0.9	5.26	2.49	1.47	4800	54
	2	PuO ₂ + Soil 1	0.9	5.14	2.41	1.74	4800	56
	3	PuO ₂	0.9	4.14	2.35	1.40	4300	59
	4	PuO ₂ + Soil 1	0.9	4.16	2.32	2.07	3900	68
23	1	PuO ₂	22.4	7.10	3.51	2.38	270	94
	2	PuO ₂	22.4	6.18	3.60	2.23	220	95
	3	PuO ₂	22.4	5.71	3.45	2.83	270	96
	4	PuO ₂	22.4	5.27	3.38	1.89	265	96
24	1	PuO ₂	22.4	8.34	3.28	1.14	300	93
	2	PuO ₂	22.4	7.49	3.33	0.60	320	95
	3	PuO ₂	22.4	6.01	3.22	0.59	270	96
	4	PuO ₂	22.4	5.50	3.11	1.31	240	96
25	1	PuO ₂	22.4	8.40	4.00	0.89	600	95
	2	PuO ₂ + Soil 1	22.4	7.54	3.73	1.04	750	95
	3	PuO ₂	22.4	6.32	3.71	1.31	750	96
	4	PuO ₂ + Soil 1	22.4	5.26	3.51	1.20	500	94
26	1	PuO ₂	22.4	8.44	3.66	0.53	470	89
	2	PuO ₂ + Soil 1	22.4	5.92	3.26	1.34	420	96
	3	PuO ₂	22.4	4.35	3.16	1.13	280	96
	4	PuO ₂ + Soil 1	22.4	3.15	2.82	4.19	220	97
27	1	PuO ₂	5.02	6.73	3.26	1.23	1580	89
	2	PuO ₂	5.02	6.10	3.19	0.65	1500	89
	3	PuO ₂	5.02	6.03	3.26	0.38	1540	88
	4	PuO ₂	5.02	5.68	3.05	2.40	1500	88

Table 1 (con't). $^{238}\text{PuO}_2$ Aerosol Data

Test No.	Run No.*	Aerosol Type	Wind Speed cm/sec	Particle Size Data			Aerosol at 125 cm***	
				AMAD μm	GSD	ΣX^{2**}	Concentration nCi/l	% of Total Activity
28	1	PuO_2	5.02	6.55	3.26	0.90	930	87
	2	PuO_2	5.02	6.12	3.20	1.19	870	89
	3	PuO_2	5.02	6.33	3.27	1.15	940	86
	4	PuO_2	5.02	6.17	3.31	1.31	1000	87
29	1	PuO_2	5.07	6.67	3.30	1.38	1420	87
	2	PuO_2 + Soil 1	5.07	4.40	2.82	5.63	1350	90
	3	PuO_2	5.07	5.65	3.26	1.98	1190	89
	4	PuO_2 + Soil 1	5.07	4.28	3.39	28.1	1150	90
30	1	PuO_2	5.07	6.67	3.51	1.69	1900	88
	2	PuO_2 + Soil 1	5.07	6.34	3.41	1.54	1500	86
	3	PuO_2	5.07	4.84	3.32	0.50	1550	90
	4	PuO_2 + Soil 1	5.07	4.20	3.18	1.44	1220	89
31	1	PuO_2	5.07	7.51	3.16	1.55	1620	87
	2	PuO_2 + Soil 3	5.07	6.79	2.97	1.27	1700	88
	3	PuO_2	5.07	5.58	3.04	1.02	1310	92
	4	PuO_2 + Soil 3	5.07	5.59	2.97	2.83	1250	90
32	1	PuO_2	5.07	6.88	3.27	0.67	1510	85
	2	PuO_2 + Soil 3	5.07	6.83	3.15	19.49	1300	88
	3	PuO_2	5.07	5.71	3.08	0.80	1420	85
	4	PuO_2 + Soil 3	5.07	5.65	3.00	1.48	1490	87
33	1	PuO_2	0.90	3.67	3.07	4.68	2830	72
	2	PuO_2 + Soil 3	0.90	4.69	3.31	5.51	2960	73
	3	PuO_2	0.90	3.45	2.94	2.12	2950	72
	4	PuO_2 + Soil 3	0.90	4.24	3.28	2.74	3150	74
34	1	PuO_2	0.9	5.13	2.48	23.79	5100	63
	2	PuO_2 + Soil 3	0.9	4.44	2.38	3.15	4760	67
	3	PuO_2	0.9	3.22	2.24	3.98	4360	73
	4	PuO_2 + Soil 3	0.9	3.26	2.22	5.04	3800	78
35	1	PuO_2	0.9	3.95	2.56	2.61	5130	64
	2	PuO_2	0.9	3.18	2.33	3.70	4500	74
	3	PuO_2	0.9	3.41	2.40	3.46	4920	64
	4	PuO_2	0.9	3.05	2.34	4.00	4400	74
36	1	PuO_2	22.6	5.59	2.68	0.91	300	97
	2	PuO_2 + Soil 3	22.6	5.39	2.72	0.64	250	98
	3	PuO_2	22.6	4.91	2.83	1.44	200	97
	4	PuO_2 + Soil 3	22.6	4.29	2.72	1.31	190	97

Table 1 (con't). ²³⁸PuO₂ Aerosol Data

Test No.	Run No.*	Aerosol Type	Wind Speed cm/sec	Particle Size Data			Aerosol at 125 cm***	
				AMAD μm	GSD	ΣX ² **	Concentration nCi/l	% of Total Activity
37	1	PuO ₂	0.9	3.92	2.20	1.37	4900	71
	2	PuO ₂ + Soil 3	0.9	4.17	2.23	1.87	4900	73
	3	PuO ₂	0.9	3.52	2.24	1.60	3780	67
	4	PuO ₂ + Soil 3	0.9	4.29	2.34	1.18	3940	60
38	1	PuO ₂	22.6	7.94	2.92	0.91	581	94
	2	PuO ₂ + Soil 3	22.6	6.88	2.86	1.04	630	95
	3	PuO ₂	22.6	6.36	2.86	1.77	500	94
	4	PuO ₂ + Soil 3	22.6	6.09	2.87	2.51	480	96

* Tests 1 through 13 were carried out in FY 1975. Tests 14 through 18 were mainly for checking out generation technique and are not presented here due to the different generation techniques used.

** A ΣX² test for goodness of fit to a log normal particle size distribution has been made. If ΣX² for a run exceeds 9.23, the distribution is not likely to be log normal.

*** Aerosol properties determined at 125 cm from the generator. % of total activity contained in the aerosol at 125 cm from the generator.

Table 2
Partial Aerosol Concentrations Relative to Run 1 of Each Test

Experimental Conditions		Relative Partial PuO ₂ Aerosol Concentrations									
		Run 2				Run 3				Run 4	
		C _{A2}		C _{B2}		C _{A3}		C _{B3}		C _{A4}	
Aerosol	Wind Speed cm/sec										
PuO ₂ only	All Speeds	1.12 ± 0.10 ⁺		0.88 ± 0.08		1.16 ± 0.08		0.83 ± 0.15		1.10 ± 0.21	
PuO ₂ + Soil 1	0.9	0.98	1.02	1.00	1.02	-	-	0.96	1.23	0.66	0.80
	5.1	0.82	0.84	0.63	0.93	-	-	1.04	-	0.76	0.99
	22.4	1.10	-	0.77	-	-	-	1.16	-	0.34	-
PuO ₂ + Soil 3	0.9	0.68	0.70	0.91	0.93	-	-	0.66	0.90	0.79	0.97
	5.1	0.86	0.88	0.83	1.00	-	-	0.87	1.05	0.76	0.98
	22.4	0.94	1.07	0.89	0.95	-	-	0.94	1.17	0.64	0.81

C_{Ai} = Relative aerosol concentration of Pu activity in PuO₂ particles < 2.2 μm diameter in run i, i = 1,2,3,4.

C_{Bi} = Relative aerosol concentration of Pu activity in PuO₂ particles > 2.2 μm diameter in run i, i = 1,2,3,4.

⁺ = Mean value ± standard deviation. (Applies to PuO₂ runs only. All other entries are the actual values obtained.)

Table 3

Soil Aerosol Characteristics

Test No.	Soil Type	Wind Speed cm/sec	Aerosol Size			Aerosol at 125 cm*		Remarks
			MMAD	GSD	ΣX^2	Concentration mg/l	% of Total	
39	Soil 3	5.07	7.4	2.72	8.84			Pu generator not operated. Humidity and air flow conditions not the same as for PuO ₂ Soil aerosol tests.
43 40	Soil 3	5.07	7.87	3.07	12.39	24	10	Distilled H ₂ O generated in Pu generator to approximate flow and humidity conditions of Soil + PuO ₂ tests.
41	Soil 3	0.9	4.89	2.65	24.8	95	9	"
42	Soil 3	22.4	14.29	3.50	5.92	15	30	"
43	Soil 1	0.9	7.35	2.71	15.3	67	5	"
44	Soil 1	5.07	7.45	2.50	9.5	18	8	"
45	Soil 1	22.4	32.5	4.5	2.1	8	15	"

* Aerosol concentration at 125 cm from the generator and the percent of total soil generated which is still in the aerosol at 125 cm from the generator.

Table 4. Weight Percentages of Particle-Size Fractions of Mineral Matter in SNS Soils.

Fraction	Range of Particle Size, microns	Weight Percent of Mineral Matter in SNS Soils			
		Soil #1 Sandy Loam	Soil #2 Loamy Sand	Soil #3 Clay Soil	Soil #4 Silt Loam
<u>Clay</u>					
Fine Clay	0.08-0.2	0.5	1.2	19.2	1.2
Coarse Clay	0.2-2.0	4.4	2.3	43.8	5.6
<u>Total</u>		4.9	3.5	63.0	6.8
<u>Silt</u>					
Fine Silt	2.0-5.0	3.5	1.0	13.0	1.7
Medium Silt	5.0-20	10.4	1.1	20.4	6.9
Coarse Silt	20-50	27.5	8.8	2.4	50.6
<u>Total</u>		41.4	10.9	35.8	59.2
<u>Sand</u>					
Very Fine Sand	50-100	19.0	7.3	0.8	33.2
Fine Sand	100-250	11.5	51.1	0.3	0.5
Medium Sand	250-500	7.0	16.4	0.1	0.1
Coarse Sand	500-1000	16.2	10.8	0.0	0.2
<u>Total</u>		53.7	85.6	1.2	34.0

Table 5. Alpha Activity in Water Samples Taken
From End of the PPO Solubility Test Vessels

<u>Vessel No.</u>	<u>Date of Sampling</u>	<u>Approximate Time After Initiation of Experiment, months</u>	<u>Activity dpm/sample</u>
3	12-12-74	8.0	< Bkgd*
3	2-27-75	10.5	168
3	4-24-75	12.5	106
3	6-12-75	14.0	< Bkgd
4	12-12-74	8.0	< Bkgd
4	2-27-75	10.5	74
4	4-24-75	12.5	73
4	6-12-75	14.0	< Bkgd

*Background = 4-5 cpm

Table 6. Alpha Activity in Organs of Sea Cucumbers
Collected in the PPO Sea-Water Solubility Test Area

Animal Number	Date Collected	Location	Mouth & Stomach	Gut	Resp. Tree	Gonads	Body Wall	Total dpm/g	Total dpm per animal
10	4-04-75	Outside Cage	< Bkgd	518.5	52.0	< 19.4	.9	56.1	17,738
11	6-06-74	5 - 6 ft Outside Cage	21.7	45.1	34.1	< 13.4	2.9	21.9	6,141
12	6-06-74	5 - 6 ft Outside Cage	17.4	2.8	63.7	< Bkgd	1.6	4.3	573
20	12-12-74	Wilson Cove	46.6	6.8	18.0	-	1.7	5.3	823
21	12-12-74	Wilson Cove	8.0	7.0	135.0	-	1.7	4.0	411
22	12-12-74	Wilson Cove	< Bkgd	16.0	< Bkgd	-	< Bkgd	1.9	337
23	12-12-74	Wilson Cove	8.3	3,900	< Bkgd	-	1.1	581	85,983
24	12-12-74	100 ft from Cages	< Bkgd	114.5	*	-	1.4	11.6	1,080
25	12-12-74	100 ft from Cages	10.4	51.0	117.7	-	0.7	14.2	2,245
26	12-12-74	100 ft from Cages	8.1	41.1	97.3	-	0.7	6.9	1,279
27	12-12-74	100 ft from Cages	< Bkgd	12.6	21.0	-	<u>m</u>	-	-
28	12-12-74	100 ft from Cages	<u>m</u>	68.6	< Bkgd	-	3.0	-	-
33	2-27-75	Outside Cages	62.6	90.5	18.0	-	1.5	29.7	6,359
34	2-27-75	Outside Cages	16.4	21.3	170.0	-	3.6	11.8	1,481
39	4-24-75	Outside Cages	< Bkgd	8.2	< Bkgd	2.5	18.5	14.0	3,543
40	4-24-75	Outside Cages	14.9	12.0	< Bkgd	< Bkgd	2.2	4.5	783

*Included with gut

Table 7. Alpha Activity in Organs of Sea Cucumbers After Exposure to Unclad PPO Pellets in Sea-Water (Exposure Cage 3).

<u>Animal Number</u>	<u>Time Period, days</u>	<u>Mouth & Stomach</u>	<u>Gut</u>	<u>Resp. Tree</u>	<u>Gonads</u>	<u>Body Wall</u>	<u>Total dpm/g</u>	<u>Total dpm per animal</u>
13	62	201.0	-	-	-	367	357	41,039
14	62	371.0	-	-	-	886	850	73,954
15	62	997.0	-	-	61	599	610	63,139
16	55	449.0	-	8.0	< Bkgd	30.5	60	17,094
18	105	11.8	< Bkgd	< Bkgd	< Bkgd	9.8	8.4	2,554
29	85	92.0	2,746	827	-	169	323	38,108
30	85	248.0	7,252	2,590	-	314	835	112,341
35	77	2,038	99,150	1,802	-	761	15,723	2,814,385
36	77	1,731	56,213	7,262	-	2,156	7,768	1,075,864
41	56	922	17,362	202	228	108	1,680	460,350
42	56	3,490	35,932	508	1,533	1.4	3,292	664,963
45	49	24,496	44,533	1,082	-	296	6,830	1,062,080
46	49	427	392	251	56	244	256	31,837

Table 8. Alpha Activity in Organs of Sea Cucumbers After Exposure to Unclad PPO Pellets in Sea-Water (Exposure Cage 4).

<u>Animal Number</u>	<u>Time Period, days</u>	<u>Mouth & Stomach</u>	<u>Gut</u>	<u>Resp. Tree</u>	<u>Gonads</u>	<u>Body Wall</u>	<u>Total dpm/g</u>	<u>Total dpm per animal</u>
17	55	31.2	-	<Bkgd	-	10.4	10.9	2,262
19	105	19.3	<Bkgd	33.4	<Bkgd	50.8	44.5	11,826
31	85	136	549	3,528	-	<u>m</u>	-	-
32	85	558	-	-	<Bkgd	1,033	203	18,283
37	77	1,586	16,209	1,259	-	303	2,234	407,719
38	77	788	20,308	2,290	-	301	4,233	465,642
43	56	4,271	43,013	1,326	2,868	360	5,167	1,178,075
44	56	2,724	29,190	814	1,056	581	5,408	1,251,942
47	49	1,120	2,745	1,838	2.5	46.0	256	43,453
48	49	1,208	33,252	826	422	355	1,965	335,066

Table 9. Mean Wet Weights of Organs From Sea Cucumbers
Collected at the PPO Solubility Test Site

<u>Location</u>	<u>Mean Organ Weights, grams</u>			
	<u>Mouth & Stomach</u>	<u>Gut</u>	<u>Resp. Tree</u>	<u>Body Wall</u>
Outside Cage	$\frac{7.4}{3.5-11}$	$\frac{35}{11.5-99}$	$\frac{4.0}{1-11}$	$\frac{123}{79-249}$
Cage #3	$\frac{8.2}{5.0-15}$	$\frac{11.8}{0-27}$	$\frac{3.4}{0-11}$	$\frac{143}{81-267}$
Cage #4	$\frac{9.0}{5.0-13}$	$\frac{12.7}{0-38}$	$\frac{4.9}{0-10}$	$\frac{142}{79-222}$

Table 10. Alpha Activity in Organs of Sea Cucumbers After Exposure
to Clad PPO Pellets in Sea-Water

<u>Animal Number</u>	<u>Time Period, days</u>	<u>Location</u>	<u>Mouth & Stomach</u>	<u>Gut</u>	<u>Resp. Tree</u>	<u>Gonads</u>	<u>Body Wall</u>	<u>Total dpm/g</u>	<u>Total dpm per animal</u>
49	49	Cage #1	346	5,712	14,155	51	270	1,070	128,909
50	49	Cage #1	1,436	637	49,618	30	682	1,484	287,829
51	49	Cage #2	194	-	1,186	-	826	797	223,535
52	49	Cage #2	998	135	2,442	-	537	624	82,680

FIGURE 1. Diagram of Wind Tunnel Facility.

FIGURE 2. Isokinetic Filter Paper Aerosol Samplers.

FIGURE 3. Isokinetic Cascade Impactor Nozzles.

FIGURE 4. Andersen Cascade Impactor with Multi-Speed Sample Nozzle.

FIGURE 5. Tunnel Bottom Deposition Patterns at 0.9 cm/sec Wind Speed.

FIGURE 6. PuO_2 Tunnel Bottom Deposition Patterns at 0.9 cm/sec Wind Speed.

FIGURE 7. PuO_2 Tunnel Bottom Deposition Patterns at 22.4 cm/sec Wind Speed.

FIGURE 8. Tunnel Bottom Deposition Patterns for Soil #1.

FIGURE 9. Tunnel Bottom Deposition Patterns for Soil #3.

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