289
1-22-75

52 -1142

WASH-1359

PLUTONIUM AND OTHER TRANSURANIUM ELEMENTS:

Sources, Environmental Distribution And Biomedical Effects

**U.S. ATOMIC ENERGY COMMISSION
DECEMBER 1974**

289
1-22-75

Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, VA 22161
Price - Printed Copy \$10.60, Microfilm \$2.25

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

**PLUTONIUM AND OTHER TRANSURANIUM ELEMENTS:
SOURCES, ENVIRONMENTAL DISTRIBUTION AND
BIOMEDICAL EFFECTS**

A compilation of Testimony presented before an Environmental Protection Agency hearing board, December 10-11, 1974, Washington, D.C.

Contributors

**James L. Liverman
Robert E. Yoder, Jr.
McDonald E. Wrenn
Burton G. Bennett
William J. Bair
William W. Burr, Jr.
Chester R. Richmond
Roy C. Thompson
J. Newell Stannard**

Compiled by: Bruce W. Wachholz

December 1974

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

PREFACE

This material was prepared at the request of the Division of Biomedical and Environmental Research, U. S. Atomic Energy Commission, in response to a notice appearing in the Federal Register, October 24, 1974. This notice stated the intent of the U. S. Environmental Protection Agency to hold public hearings to evaluate the environmental impact of plutonium and the other transuranium elements and to consider whether new guidelines or standards are needed to assure adequate protection of the general ambient environment and of the public health from potential contamination of the environment by radionuclides of these elements.

Necessarily the testimony was restricted in time and, consequently, in length. Therefore the various topics discussed are not thorough in-depth reviews of the subject matter, but are brief summaries of the information available. Only minor changes have been made from that material actually presented for purposes of clarification; the material presented is a matter of public record. Further information may be obtained from the references provided.

Although the data utilized was obtained from numerous individuals and laboratories, particular acknowledgement is extended to the following for their assistance:

Battelle Memorial Institute, Pacific Northwest Laboratory

Oak Ridge National Laboratory

University of Rochester

USAEC

Division of Biomedical and Environmental Research
Division of Operational Safety
Division of Military Applications
Division of Production Materials Management
Division of Reactor Research and Development
Division of Space Nuclear Systems
Division of Waste Management and Transportation
Health and Safety Laboratory

Because the information contained herein is of potential interest to persons and organizations not present at the EPA hearings, it was considered desirable to publish the AEC testimony in order to make it more easily available. Although specific subjects covered herein are discussed more extensively elsewhere, this Testimony is perhaps the most comprehensive single source of information currently available covering all of the numerous areas included in the subject material.

TABLE OF CONTENTS

Introduction	James L. Liverman	1
Potential Source Terms and Control Measures.	Robert E. Yoder, Jr..	13 ✓
Supplemental Information.		31
Environmental Levels of Plutonium and the Transplutonium Elements	McDonald E. Wrenn	89 ✓
Worldwide Distribution of Plutonium	Edward P. Hardy, Jr..	115 ✓
Environmental Pathways of Transuranic Elements.	Burton G. Bennett	131 ✓
Transuranic Elements in the Marine Environment.	Herbert L. Volchok.	155 ✓
The Biological Effects of Transuranium Elements in Experimental Animals.	William J. Bair	171 ✓
Biomedical Effects of Plutonium on Humans.	William W. Burr, Jr..	231 ✓
	Chester R. Richmond	235
Implications with Respect to Protection Criteria.	Roy C. Thompson	271 ✓
Standards for the Transuranic Elements	J. Newell Stannard.	291 ✓
Supplemental Additions to the Testimony		
Letter from Lester Rogers to Clarence C. Lushbaugh.		307
Letter from Clarence C. Lushbaugh to Lester Rogers.		308
A Critique of the Tamplin-Cochran Proposal for Revision of the Current Plutonium Exposure Standards	Roy E. Albert	311
WASH-1320		327

Introductory Testimony
by James L. Liverman
Assistant General Manager for Biomedical and
Environmental Research and Safety Programs
U. S. Atomic Energy Commission
Washington, D. C. 20545

part of the AEC presentation at the
EPA Plutonium Standards Hearings
Washington, D. C., December 10-11, 1974

My name is James L. Liverman. I am Assistant General Manager of the Atomic Energy Commission with responsibilities for biomedical and environmental research, waste management, and safety programs. I will provide introductory testimony and will be followed by members of AEC and contractor staff who will provide more specific and detailed testimony. Mr. Lester Rogers, who will represent the regulatory and licensing activities of the Atomic Energy Commission, is scheduled to testify tomorrow.

It is our understanding that these hearings are being held to gather information relevant to EPA's intention to evaluate whether new guidelines and standards are needed to assure adequate protection of the environment and public health from potential contamination of the environment by radio-nuclides of the transuranium elements. Judgments by EPA regarding new guidelines and standards for transuranic elements will influence development of nuclear energy to help meet nationwide energy needs, national security programs, and other matters of substantial importance to our society.

Public concern regarding the manufacture and use of transuranium elements is, I think, based on several facts.

1. Increasing quantities of the transuranics are being produced, and the rate of production will increase substantially in the foreseeable future

as nuclear fuels provide a growing fraction of our national energy requirements.

2. Several radioisotopes of plutonium and other transuranic elements have exceedingly long half-lives and, once released, will persist and accumulate in the environment for time periods extending over many human generations. This is, of course, also true for some naturally occurring alpha-emitting radioisotopes such as those of radium and uranium.
3. These naturally occurring alpha-emitting radioisotopes are known to produce cancer of the lung, bone, and liver in humans exposed to large concentrations.
4. Comparable concentrations of alpha-emitting transuranic elements are known to produce cancer of the lung, bone and other organs in experimental animals.

These facts make it clear that, in operations involving the production and use of transuranic elements, bioenvironmental health and safety considerations are necessarily of primary concern. This point was recognized at the very outset of the nuclear age -- when some of the first plutonium made was turned over to biomedical investigators. Since that time (in 1944) bioenvironmental and control technology programs have proceeded in parallel with and have guided engineering development of the nuclear technology. More than thirty years of research and development have produced a substantial fund of knowledge and understanding regarding the health and safety aspects of operations involving the production and use of transuranics. Through application of this knowledge, experience, and understanding, it has been possible to establish health and safety (radiation protection) procedures which have permitted many thousands of kilograms of

plutonium to be produced and processed, and we have yet to identify successfully a major health consequence attributable to its radiotoxicity. This record contrasts sharply with that for the commercial use of radium earlier in the century where manufacture and use of a few grams resulted in extensive occupational exposures and many cases of cancer.

I would like to make five points regarding the setting of standards, criteria, and guidelines for the transuranium elements and comment briefly on each.

1. Meaningful judgments on the adequacy of current standards and guidelines must be based in part on the knowledge and understanding acquired in the course of nuclear development including the research in the life sciences. This knowledge is extensive; it cannot be presented, evaluated, or even adequately summarized in a few days. It is, however, available to all, and specific measures have been taken to assure accessibility and to expedite wide dissemination of this information.

For example, Nuclear Science Abstracts (NSA) contains well over 10,000 references on all aspects of the physical, chemical, environmental, and biological properties of plutonium and other transuranic elements. RECON - a computerized bibliographic searching system -- permits rapid access to NSA and other bibliographic data bases from terminals dispersed widely through AEC, its contractors, other Federal agencies, and some universities.

There are in addition to this broad bibliographic base a number of specialized information centers relevant to the collection, dissemination, and evaluation of information on the transuranic elements. These include the data base on comparative metabolism of plutonium maintained

at the Comparative Animal Research Laboratory of the University of Tennessee and the environmental plutonium data base, the Information Center for Internal Exposure, and the Nuclear Safety Information Center, all located at Oak Ridge National Laboratory (ORNL). ORNL also maintains direct access to the "Tox-line" and "Med-line" systems and computer information systems of the National Library of Medicine.

To have another example, scientific meetings provide an important opportunity for information exchange. In 1974 alone, five meetings devoted to (i) the biological effects of internally deposited alpha-emitters, including the transuranium elements, (ii) plutonium in the environment, and (iii) radionuclide carcinogenesis have been held this year (at Los Alamos, Richland, Seattle, Alta, Utah, and Las Vegas). Proceedings of these meetings are published in the AEC Symposium series or by the sponsoring laboratory. Meetings of professional societies also provide opportunities for exchange of scientific information. Numerous international meetings on the environmental and biomedical effects of the transuranics have been sponsored by the International Atomic Energy Agency.

Finally, there are monographs on this subject, an instance of which is the recently published volume of the Handbook of Experimental Pharmacology entitled, "Uranium, Plutonium, Transplutonic Elements."

2. The next point I wish to make regarding the setting of standards relates to those organizations independent of government which have played an essential role in the analysis of much of the relevant

biomedical and environmental information and in the development of the standards, guidelines, and general procedures which are currently used for the nuclear industry. It is essential that the objectivity which these organizations represent continues to be involved in this process.

These organizations include the United Nations Scientific Committee on the Effects of Atomic Radiations, the IAEA, the International Commission on Radiological Protection (ICRP), the National Council on Radiation Protection and Measurements (NCRP), and the National Academy of Sciences (NAS) through its various committees. All of these address themselves to the analysis and evaluation of pertinent data. More so than the others, the ICRP and NCRP are involved in the development of radiation protection criteria and standards. From time to time these organizations consider special issues such as the "hot particle" issue now under consideration by a committee of the NAS.

3. Judgments regarding the adequacy of current guidelines and standards need to be based on evaluation of the results of their application to specific development and operational activities and in regulation. Particularly in the case of plutonium already in the environment the establishment of additional "generally applicable" standards may not be as effective an approach to cost-effective control of radiation hazards as can a case-by-case analysis based on current standards and guidance. Our experience suggests that in such situations specific circumstances such as physical and chemical form of the material, climate, and current and projected land use are highly variable and site specific. Each of these circumstances may exert an important influence on the

practicability, including cost-effectiveness, of alternative remedial actions. Since the number of contaminated sites is likely to remain small, it might be more effective to deal with these situations on a case-by-case basis using current standards than to attempt to develop additional standards and guidance generally applicable to all of them.

If additional guidance is developed for this purpose, it would be essential that it include the flexibility required to assure effective application in widely varying circumstances.

4. Despite the citation in Point 1 of thousands of references, information on the biomedical and environmental behavior of plutonium and other transuranics is not complete. It is not likely to ever be complete. We need to know that it is adequate to assure safety in current and future activities and we need to identify specific areas where increased understanding is likely to have the greatest impact on specific developmental, operational, and regulatory decisions so as to focus our research there.

We do wish to point out, however, that there are much greater deficiencies and uncertainties in data that would permit assessment of the environmental and health impacts of alternative energy sources leading one to the conclusion that major efforts are still required in all energy forms.

5. The AEC has a major research program on the biomedical and environmental aspects of the transuranics. This program focuses on those areas where additional information is most likely to critically influence development programs, operations, and regulations.

We continue to support research on various species of experimental animals exposed to transuranic elements in various forms. A major portion of efforts in this area focus on determining the consequences of inhaling small quantities of aerosolized transuranics and quantitating the effects of aggregation of alpha activities into "hot particles."

We continue observations on humans exposed to plutonium more than twenty-five years ago and we are expanding the Transuranium Registry of potentially exposed occupational workers.

In the environmental area we continue observations on the behavior of transuranic elements dispersed globally by weapons testing and studies of the behavior of plutonium in the quite diversified environments near weapons test areas in the U.S. and the Pacific and around operating facilities in the U.S.

The aim of this research is to reduce the need for conservative and possibly very costly assumptions by providing the information needed to make more realistic estimates of potential health and environmental hazards of transuranium elements. The results of this research are published regularly and are accessible through the various routes I mentioned earlier.

Increasingly, in anticipation of ERDA, we are integrating research activities in this area with research on the environmental behavior and potential health consequences of pollutants from alternative energy sources so that we will be better equipped to assess bioenvironmental aspects of alternative energy technologies so as to help orchestrate

their development and to provide a sound basis for operating and regulating these technologies as they are installed.

Mr. Chairman: I realize these comments have been all too brief but they will be expanded in major ways to cover in detail much of what I have alluded to during the course of the afternoon and tomorrow morning. If I could simply introduce those who will participate with me at this point and in the order in which they will appear, I could remain quiet as we proceed:

Dr. Yoder, AEC's Div. of Operational Safety will discuss
Source Terms and Control

Dr. Ed Wrenn, followed by Dr. B. Bennett, will discuss
Environmental Levels of the Transuranics

Dr. W. J. Bair, Battelle Northwest Lab, will discuss
Transuranics in Experimental Animals

Dr. W. W. Burr, Deputy Director, DBER AEC, will talk
concerning Human Exposures

Dr. Chet Richmond formerly of Los Alamos Lab but now
with Oak Ridge National Lab will discuss Biomedical
Effects in Humans

Dr. Roy Thompson, Battelle Northwest Lab, will finally
discuss Implications with Regard to Protection Criteria

AVAILABLE INFORMATION

- NUCLEAR SCIENCE ABSTRACTS
- RECON
- SCIENTIFIC MEETINGS: A FORUM
- INTERNATIONAL SCIENTIFIC MEETINGS
- MONOGRAPHS

NON-GOVERNMENTAL ORGANIZATIONS

- PARTICIPATION IN:

- ANALYSIS AND EVALUATION OF DATA
- DEVELOPMENT OF RADIATION PROTECTION STANDARDS
- ADDRESS SPECIAL ISSUES

- GROUPS INVOLVED IN ONE OR MORE OF ABOVE:

UNSCEAR

IAEA

ICRP

NCRP

NAS

- BEAR COMMITTEE
- BEIR COMMITTEE
- "HOT PARTICLE" COMMITTEE

Potential Source Terms and Control Measures *[for Transuranium materials]*

by Robert E. Yoder, Jr., Ph.D.
Assistant Director for Facilities Safety
Division of Operational Safety
U. S. Atomic Energy Commission
Washington, D. C. 20545

part of the AEC presentation at
EPA Plutonium Standards Hearings
Washington, D. C., December 10-11, 1974

Introduction

The information presented in this discussion will include the current sources of transuranium materials within the Atomic Energy Commission (AEC) operations, an indication of projected inventories, and an overview of control measures taken to reduce effluents. This is not intended to be an exhaustive review of the subject matter, but to highlight the AEC actions in managing its transuranic materials operations. Specific information regarding the location and quantity of material which has been released to the environment will be presented later.

There have been releases of plutonium and these have been well publicized. The quantities of materials involved in these instances have ranged from much less than one to a few kilograms of material, and steps have been taken to reduce the accident potential in AEC operations. A comparison of the quantities released to the quantities in use shows that a very small amount has been released in accidents. Routine emissions are now very low and still decreasing. Because the AEC is concerned about environmental discharges and any attendant buildup in the environment, it is fully implementing the "as low as practicable" concept. For example, if our routine emissions continue at the present levels by the

year 2000 less than 3 additional curies of plutonium will be discharged to the environment, compared to the kilocurie quantities already present from atmospheric weapons testing. The intensive environmental sampling program which quantifies the amount of plutonium in the environment and its specific location in identified pathways provide confirmation that our control programs are effective. Information developed in this program will be available and analyzed well before a potential problem exists and will allow ample time to take effective action.

In those instances in which environmental cleanup actions have been required, specific measures tailored to the specific site have been used. Because the number of the cases is very small, they are best handled on a case-by-case basis so that a comprehensive evaluation can be made to effectively limit the availability of these materials.

Cost-Benefit

The AEC uses plutonium or handles plutonium in three broad program categories: national defense, energy research and development, and service to other agencies and private industry. The benefit-risk analyses with regard to each of these areas is developed along separate lines of reasoning: (1) The executive and legislative branches of the government have established the benefit-risk associated with a viable national defense program which requires plutonium for its development and maintenance. The AEC produces and manufactures plutonium components for use by the Department of Defense. Also, as required by the Test Ban Treaty, the AEC maintains a viable nuclear development and test program. (2) The current energy situation has brought into focus a number of elements pertinent to the risk-benefit of plutonium reactors as sources of electric

power. The AEC conducts the research and development necessary to support the industry options to use these reactors for electric power production.

(3) Service functions are associated with the development of nuclear radioisotopic thermoelectric generators which are manufactured by the AEC for agencies who themselves have developed the risk-benefit analyses supporting their use. In this case, the AEC merely supplies the material in a form suitable for the specific program use. As an additional service function, the AEC provides the burial facilities or interim storage facilities for transuranic materials.

In June 1973 the AEC began the preparation of an environmental impact statement for the overall Liquid Metal Fast Breeder Reactor (LMFBR) program (WASH-1535) as required by the National Environmental Policy Act (NEPA). The environmental impact statement, now in the final stages of review prior to release, differs from the conventional statement because it addresses the important environmental and societal impacts from the assumed eventual commercialization of LMFBRs which are expected if the research and development goals are attained rather than the impact of the research and development activities. It was necessary, therefore, to look ahead some 40 to 50 years to foresee an LMFBR industry that would provide about 40 percent of the total installed electric generating capacity in the United States by the year 2020.

The direct costs include LMFBR development program costs and those costs normally imbedded in the price of electrical energy. Over 70 separate postulated cases were considered in estimating United States electrical power production costs over the period 1974-2020. One of the major quantifiable conclusions of the direct economic analysis was that

the introduction of a fast breeder into the United States electric power utility system will produce significant financial benefits. These benefits result largely from a reduction in uranium ore and enrichment requirements. Additionally, the fast breeder results in a nuclear power industry that will have total power costs, in constant dollars, that decrease with time. Capital requirements accumulated to the year 2020 are estimated to be about 10 percent less with a fast breeder industry due to the reduced requirement for mining, milling, and enrichment facilities.

Indirect benefits, though no less significant, come partly from the improved resource utilization. The use of plutonium as a reactor fuel for electric power production would free the finite resources of fossil fuels for their optimum use, thus assuring the domestic availability of fuel for electric power production for the long term. A mature LMFBR industry together with the nonbreeders can generate sufficient fissile material for the nuclear industry to be able to use stockpiled depleted uranium for hundreds of years as the sole source of fertile fuel material.

Other indirect benefits of note are in the cumulative health and safety effects for both occupational and public groups.

Inventories

The AEC has produced several tens of tons of plutonium-239 in its reactors, including several tons of nonweapon grade plutonium, since the mid-1940's. In addition, 200 kg of Pu-238, 4 kg of Cm-244, and 2 grams of Cf-252 have been produced in the Savannah River reactors. Additional quantities of transplutonium elements have been produced in Savannah River reactors, separated, and refabricated into targets for further

irradiation as part of the Cf-252 program.

Plutonium-238 is used principally as power sources for space research, satellites, and heart pacemakers. These applications have required about 40 kg of plutonium-238 since 1961. The requirements for specific applications through FY 1986 project a need of about 540 kg of plutonium-238. Since 1961 the United States has launched radioisotope thermoelectric generators containing 34.7 kg of plutonium-238. Of this quantity, 13 kg are on the lunar surface, 6.8 kg are in long-lived earth orbit, and 9.4 kg have been ejected out of the solar system. One kg was released to the earth's atmosphere in a reentry burnup (as designed), 2 kg of encapsulated material were recovered from the Santa Barbara Channel following a missile abort, and 2.5 kg impacted intact (as designed) in the South Pacific Ocean following an aborted lunar mission. No plutonium-238 was released from the latter two incidents.

Small quantities of plutonium-238 will be required for heart pacemakers. A potential equilibrium number of 170,000 of these devices will be needed in the United States by the year 2000. The total quantity of plutonium-238 required ranges from 25-75 kg. Several companies at present are licensed to implant these devices and an equilibrium implant rate of 10,000 per year is expected to be reached in the early 1980's.

The major increase in plutonium inventory is anticipated to be that associated with commercial nuclear power reactor activities which are projected to have "on-hand" a potential commercial plutonium inventory of 117,000 kg in the year 2000. Reactor research and development program annual plutonium requirements are projected to vary in the range of 500-2,500 kg over the next 10-year period.

The types of operations in the AEC involving plutonium include its production in reactors at Savannah River and Richland, chemical processing to separate plutonium from uranium and fission products, reduction to the metal, casting, machining, and other metallographic operations. In addition, scrap material is processed for recovery of plutonium by incineration, digestion, precipitation, and solvent extraction. Plutonium also is removed from liquid effluent streams by filtration and precipitation to reduce the volume of material which must be sent to waste handling and storage facilities, and to reduce the quantity present in waste streams. The AEC also manufactures experimental fuel elements containing plutonium and operates reactors containing plutonium fuel elements, such as the EBR II. Waste material from all AEC operations is placed in retrievable storage if the plutonium or transuranic material concentration in the waste is in excess of 10 nCi/g. A proposed rule-making would cause all commercially generated transuranic waste in excess of this limit to be sent to AEC storage sites pending the development of a final repository (Federal Register, Volume 39, September 12, 1974). In the execution of these programs the AEC engages in the transportation of radioactive materials in interstate commerce. However, this material is safeguarded as appropriate to the type and quantity of material, and the associated potential hazards.

The quantity of transuranic materials now stored or buried at AEC sites totals about 950 kg contained in approximately 1 million cubic meters of material. This material is located in burial grounds at the Idaho National Engineering Laboratory, Idaho; Richland, Washington;

Savannah River, South Carolina; and Oak Ridge, Tennessee. The characteristics of transuranium-contaminated waste will change as the breeder reactor program develops and will require the storage not only of transuranic materials, which are primarily alpha emitting materials, but also transuranic materials contaminated with gamma activity in fuel hulls which contain induced radionuclides and fission products. The assessment of projected quantities of transuranic wastes expected by the year 2000 is presented in Table 1.

Emissions

A concerted effort to accelerate the reduction of all emissions of all radioactive materials from AEC operations was initiated in 1970, including a program to quantify all past releases from AEC facilities. Figure 1 summarizes the total plutonium release data from all AEC sites having significant releases for the years 1967-1973. Figures 2 and 3 provide a breakdown of these data to show the quantities released via air and water. The important point to note is that the releases from all sites have been approximately 1.5 Ci over the 6-year period. From our operational experience and the improved control measures at all facilities, we anticipate that AEC releases will not exceed, through normal discharge systems, 0.1 Ci/yr from all operations from all sources.

In 1973, 0.053 Ci of plutonium and approximately 0.002 Ci of other transuranics were released offsite. In the same year approximately 100 Ci of plutonium and approximately 0.1 Ci of other transuranics were released onsite to treatment and disposal systems such as seepage basins. The

composition of the 1973 onsite releases is shown in Table 2. The largest component of this discharge occurred at Richland, however, this operation has been sufficiently modified so that no plutonium has been discharged to date during CY 1974. We anticipate that in CY 1974 less than 1 Ci will be discharged onsite from all sources of plutonium at all AEC sites. These reductions should be viewed with the fact in mind that much larger quantities have been discharged onsite in the past. The impact of the releases and a full assessment of their significance in the environments surrounding AEC facilities is available in WASH-1259.

Control Measures

An intensive reevaluation of the AEC handling of plutonium was initiated following the 1969 Rocky Flats Plant fire. All plutonium operations and storage facilities were surveyed and new safety criteria developed for these operations. Because the new criteria addressed in detail areas not previously highlighted, there are certain modifications which cannot be undertaken in present facilities, particularly regarding natural phenomena (tornado, earthquake) protection. However, in these cases additional safeguards and alternate protection has been provided. Plutonium operations are being conducted in glovebox and/or canyon facilities, which provide at least three barriers (e.g., glovebox, operating compartment, and outer facility walls) between the operation and the outside environment. Where possible, inert atmospheres are used to reduce the potential of fire. Fire protection is provided through the use of sprinkler systems and, in some cases, inert gas protection. Firebreaks and operation compartmentalization are used to limit the extent of any incident. At least

three high efficiency air cleaning devices are required between plutonium operations and the outside environment. This criteria is being met through the use of three or more stages of high efficiency filters or two stages of high efficiency filters in addition to a sand filter. To provide assurance for the control of plutonium in the event of a postulated accident, emergency power and protected, isolated emergency control rooms are available. A formal safety analysis report requiring AEC approval must be written for all new plutonium operations. These safety analysis reports critically review every operation, every control measure, and every interaction between operations and controls to avoid a possible loss of control. To date over \$210 million for construction has been committed to upgrade or replace existing facilities. A significant sum has been spent also in accomplishing a myriad of small projects.

The AEC has always operated under the "as low as practicable" philosophy with regard to the release of any radioactive material to the environment. However, values associated with "as low as" have continually decreased with increased operating experience and improved control technology. Every transuranium operation has been reviewed recently and additional safeguards installed to reduce effluents to the lowest level that is economically possible. At the Rocky Flats Plant, for example, a water treatment plant to clean and permit the reuse of process water is scheduled for FY 1977 in order to minimize the quantity of potentially contaminated waste water which is discharged from that site.

The plutonium-238 encapsulation operation at the Mound Laboratory is being transferred to an upgraded plutonium facility at the Savannah River Plant. By 1980 primarily only encapsulated plutonium-238 material will be handled at Mound Laboratory. This program will restrict the availability of unencapsulated material and reduce significantly the release potential of plutonium-238 oxide as well as minimize the quantity of waste material, and the number of shipments required.

Accidents

The release of plutonium and other transuranic materials in decreasing order of quantity are associated with:

1. atmospheric weapons testing;
2. weapons tests at the Nevada Test Site;
3. accidents--both from military operations and AEC plant operations; and
4. effluents from normal discharge waste streams.

The major plutonium releases associated with AEC plant operations have occurred at Oak Ridge, Rocky Flats, Richland, and Mound. The total quantity involved amounted to a few tens of curies, which represents a small fraction of that in process. In each of these areas intensive environmental survey programs are underway.

An analysis of three accidents associated with military operations is provided in the supporting documentation. The important element in the cleanup actions for these accidents is the careful analysis of the significance of any material not removed. The experience we have gained from these accidents has led to an enhancement of our capability to survey and

cleanup accident sites. Portable monitoring instrumentation useful for onsite surveys provides a sensitivity of $.1 \mu\text{Ci}/\text{m}^2$ of plutonium on the soil surface. An aerial monitoring program using sensitive radiation monitoring instruments and helicopters, allows the survey of large areas with a sensitivity of $0.3 \mu\text{Ci}/\text{m}^2$. This survey technique allows operation at 100 feet altitude above the ground surface and at an air speed of 60 knots. Analytical analyses for plutonium in environmental samples provide assessments such that we can determine plutonium at levels below $10^{-5} \mu\text{Ci}/\text{m}^2$. Now a number of AEC laboratories have a proven capability to process, specifically for plutonium, soil or vegetation samples with this sensitivity. The limit of sensitivity in these cases is limited by the quantity of material which one can manipulate in the chemical processes and the capability to analyze a large number of samples, such as may be required in an accident situation. The accidents which have occurred were in quite different environments and required very different techniques for cleanup. As a matter of course one removes all radioactive material which is practical to remove and which poses a potential significant source of exposure. The techniques which can be used include excavation, plowing, and fixation. New techniques to fix plutonium to reduce its movement prior to cleanup are under development. The guidelines which can be used for the cleanup levels must be specifically set at each site because the pathways for human exposure and ability to cleanup will vary from site to site. The situations which occurred at Palomares, Spain; Thule, Greenland; Rocky Flats, Colorado; the Nevada Test Site; and Enewetak Atoll are all unique. No two have the same problems nor do they appear at this time to be amenable to the same cleanup techniques. The quantity of plutonium involved

in any accident represents a very small fraction of the quantity of that material in the inventory, and the quantity left after cleanup is even smaller.

Summary

There have been releases of plutonium and these have been well publicized. The quantities of materials involved in these instances have ranged from much less than one to a few kilograms of material, and steps have been taken to reduce the accident potential in AEC operations. A comparison of the quantities released to the quantities in use shows that a very small amount has been released in accidents. All routine emissions are now very low and still decreasing. Because the AEC is concerned about environmental discharges and any attendant buildup in the environment, it is fully implementing the "as low as practicable" concept. For example, if our routine emissions continue at the present levels by the year 2000 less than 3 additional curies of plutonium will be discharged to the environment compared to the kilocurie quantities already present from atmospheric weapons testing. The intensive environmental sampling program which quantifies the amount of plutonium in the environment and its specific location in identified pathways provide confirmation that our control programs are effective. Information developed in this program will be available and analyzed well before a potential problem exists and will allow ample time to take effective action.

In those instances in which environmental cleanup actions have been required, specific measures tailored to the situation have been used. Because the number of the cases is very small, they are best handled on a case-by-case basis so that a comprehensive evaluation can be made to effectively limit the availability of these materials.

Figure 1

Figure 2

Figure 3

TABLE 1

TRANSURANIUM-CONTAMINATED WASTES REQUIRING REPOSITORY STORAGE
(Accumulation Thru Year-End)

<u>Year</u>	<u>Alpha Activity</u>		<u>Alpha-Beta-Gamma Activity</u>	
	<u>Cubic Meters</u>	<u>Pu & U-Kilograms</u>	<u>Cubic Meters</u>	<u>Pu & U-Kilograms</u>
1980	9,300	220	6,770	13
1985	36,200	1,160	16,700	46
1990	70,100	3,510	28,900	131
1995	125,000	8,400	48,100	302
2000	244,000	18,900	85,600	632

Reference: "Projected Shipments of Special Nuclear Material and Wastes by the Nuclear Industry,"
ORNL-TM-4631, August 1974.

TABLE 2

AEC TRANSURANIC RELEASES
CY 1973

Onsite Discharges (rounded)

Plutonium	100	Ci
Curium	0.10	Ci
Other transuranics	.001	Ci

Effluent Discharges

Plutonium	0.053	Ci
Americium	0.001	Ci
Curium	.0004	Ci
Neptunium	.0005	Ci

TABLE 3

CONVERSION FACTORS

Plutonium-238	$\frac{17 \text{ kCi}}{\text{kgm}}$
Plutonium-239	$.063 \frac{\text{kCi}}{\text{kgm}}$
Curium-244	$81.8 \frac{\text{kCi}}{\text{kgm}}$
Californium-252	$539 \frac{\text{Ci}}{\text{gm}}$

SUPPLEMENTAL INFORMATION

DIVISION OF BIOMEDICAL AND ENVIRONMENTAL RESEARCH

Potential Plutonium Source Term for Medical Uses

Radioisotope-Powered Cardiac Pacemaker

1. The purpose of the AEC program is the demonstration that technology exists which will permit the production of nuclear-powered cardiac pacemakers which will, with great confidence, substantially improve upon the reliability and lifetime capabilities of presently available chemical battery-powered pacemakers.

The 1974 pacemaker population is approximately 126,000 world-wide. The potential U.S. nuclear pacemaker market is estimated to be 10,000 per year with an equilibrium number of 170,000 in the U.S. population in the year 2000.

The primary environmental implications are from radiation during normal use and from potential exposure to fuel following a breach of the capsule and a release of the fuel to the environment.

The benefits of a long-lived nuclear pacemaker are direct functions of minimizing the number of reimplantations during the patients remaining lifetime.

2. Approximately 350 nuclear pacemakers are currently in use in the U.S. Three companies are presently licensed to implant 20 units per month each. An equilibrium implantation rate of 10,000 per year is expected to be reached in the early 1980's.

The inventory of plutonium-238 in the nuclear pacemakers ranges from 140 to 420 milligrams per unit. Thus, the total plutonium-238 inventory for a population of 170,000 in the year 2000 ranges from 24 to 71 kilograms.

3. The fuel capsule has demonstrated its capability to survive all the safety tests defined by the AEC's "Interim Safety Guide for the Design and Testing of Nuclear Power Cardiac Pacemakers" with additional substantial safety margins.
4. Fuel processing losses which are unuseable wastes amount to about 1 percent of the starting inventory. For 10,000 units per year the plutonium wastes would be in the range of 15 to 40 grams per year.

THE WASTE MANAGEMENT OF TRANSURANIC SOLID WASTE

Solid wastes of widely diverse nature and contaminated to varying degrees with heterogeneous amounts and forms of plutonium are generated in several AEC facilities. The radioactivity in such wastes has been confined within controlled areas of AEC sites by burial in shallow trenches or pits and since 1970 in easily retrievable containers. The principal burial sites include the Richland site, the Idaho Falls site, Los Alamos Scientific Laboratory, Oak Ridge National Laboratory, and the Savannah River Plant.

The waste matrices in which the transuranic isotopes are contained vary greatly in their composition. Normally, they can be segregated into those which are noncombustible and those which are combustible. Depending on the facility, the noncombustible fraction may comprise up to one-half of the total volume of waste generated, while the combustible portion includes such things as paper, rags, plastics, rubber, and discarded clothing. Table 1 shows the results of a waste composition survey taken by the Los Alamos Scientific Laboratory. This table gives an idea of the tremendous variation in composition of wastes generated by different laboratories.

Table 2 shows the projected increase in production of transuranic waste. If current process techniques continue, by the year 2000 approximately 244,000 cubic meters of alpha waste will be generated; and an additional 86,000 cubic meters of waste will be generated which has a high gamma background. Although these wastes will require much improved treatment and handling systems, our past experience indicates that release of TRU nuclides to the environment will be well within AEC guidelines based on recommendations of the International Commission on Radiological Protection. Current management programs offer little chance for environmental contamination by transuranics. Effluent treatment processes which produce some solid waste materials may lead to limited offsite radioactivity releases but these are extremely low and must always be within AEC guidelines.

Several years ago the AEC reexamined its policy regarding the management of its own transuranium-contaminated radioactive waste. Until that time, it was felt that the remoteness of the burial sites combined with favorable geological and hydrological conditions would assure the safety of the environment from TRU wastes. However, there exists the possibility that the status of sites may change as a result of new national priorities. Also, the quantities of transuranic waste generated are expected to increase substantially as a result of the generation of nuclear power. It was decided to limit the outright burial of transuranium-contaminated waste, and in March 1970, the AEC issued a directive¹ requiring its contractors to segregate transuranium-contaminated waste from nontransuranium-contaminated waste and to store the transuranics in a manner which would

¹"Policy Statement Regarding Solid Waste Burial," USAEC, IAD No. 0511-21, March 20, 1970.

T A B L E 1

ESTIMATED COMPOSITION OF LABORATORY TRASH
CONTAMINATED WITH TRANSURANIUM NUCLIDES*
(WEIGHT PERCENT)

<u>Waste Material</u>	<u>Hanford</u>	<u>LASL**</u>	<u>Mound</u>	<u>Rocky Fláts</u>	<u>Savannah River</u>
Paper & Rags	18	46	50	30	53
Rubber	15	7	25	20	25
Plastics	20	9	20	50	4
Glass	1	17	--	--	--
Metal	30	21	--	--	--
Misc.	16	--	5	--	13

* Reference: "Transuranic Waste Research and Development Program," LA-5281-MS, May 1973.

** Survey included all radioactive waste generated at LASL.

TABLE 2

**TRANSURANIUM-CONTAMINATED WASTES REQUIRING REPOSITORY STORAGE
(ACCUMULATION THRU YEAR-END)**

<u>Year</u>	<u>Alpha Activity</u>		<u>Alpha-Beta-Gamma Activity</u>	
	<u>Cubic Meters</u>	<u>Pu & U-Kilograms</u>	<u>Cubic Meters</u>	<u>Pu & U-Kilograms</u>
1980	9,300	220	6,770	13
1985	36,200	1,160	16,700	46
1990	70,100	3,510	28,900	131
1995	125,000	8,400	48,100	302
2000	244,000	18,900	85,600	632

Reference: "Projected Shipments of Special Nuclear Material and Wastes by the Nuclear Industry," ORNL-TM-4631, August 1974.

permit them to be readily retrieved in a contamination-free condition for a period of at least 20 years. This would permit time to develop a national policy which would be acceptable to the public for disposal of transuranium-contaminated waste.

The transuranic waste storage site at the Idaho National Engineering Laboratory (INEL) at Idaho Falls is a good example of current practices resulting from this directive. Here the waste is stored on an asphalt pad which has a four inch gravel base. The surface is sloped to permit moisture drainage. The waste is packaged in 55-gallon steel drums or in plywood boxes which are coated with fiberglass. The 55-gallon drums are currently lined with a 90-mil polyethylene liner to prevent contact of the waste with the wall of the drum. These packages are stacked on the pad in sections of approximately 150 feet by 80 feet. Once a section is full, the stacked waste is covered with plywood and a plastic sheet and then mounded over with earth. Normally a 3-foot space is left between each section of the pad. This 3-foot section is filled with earth and provides a fire-retarding wall.

The volume of transuranium-contaminated waste in the U.S. and the contained transuranium nuclides is expected to increase greatly in the coming years as a result of the use of plutonium in the production of nuclear power. Due to this* projected increase, the Directorate of Regulation* of the AEC has proposed a change in the Federal Regulations regarding licensee management of transuranic waste. In September 1974, a notice² was published in the Federal Register regarding a proposed rulemaking which would result in Federal management of these wastes.

According to current plans, the operating arm of the AEC** would manage commercially generated transuranic waste in the same manner as they manage AEC-generated transuranic waste. The waste would be placed into 20-year retrievable storage just as it is at INEL. The generator would pay a one-time fee which would cover the cost of all future management. An environmental impact statement³ is being prepared by the AEC which discusses alternatives for waste management in detail and is available in draft (WASH-1539).

* To become NRC about January 1, 1975.

**To become part of ERDA about January 1, 1975.

²"Transuranic Waste Disposal," Federal Register, Vol. 39, No. 178, Thursday, September 12, 1974, pp. 32921-32923.

³"Management of Commercial High Level and Transuranium-Contaminated Radioactive Waste," WASH-1539, USAEC, September 1974 (draft).

In Table 3 the total volume of radioactive waste at the major AEC sites is shown in the first column in cubic meters. The total quantity of trans-uranium nuclides contained in all of this solid waste is about 950 kilograms. Of this quantity, roughly 175 kilograms is stored in a readily retrievable fashion and the remaining 775 kg has been buried. The volume of waste retrievably stored is roughly 29,000 cubic meters, and it contains roughly one-fifth of the total quantity of transuranic nuclides generated to date.

The buried transuranium nuclides at AEC sites and the 80 kg of plutonium at licensed facilities will require continued long-term surveillance. The waste package is not considered to have any integrity after it is buried. Monitoring and surveillance programs at AEC sites have shown that migration of the buried transuranics has been negligible.⁴ However, the wastes will remain hazardous for an extremely long period of time and one cannot predict with certainty what, if any, environmental changes will take place during this time.

In consideration of the long periods of time for which these wastes must be confined, the AEC is developing hazards analysis procedures to determine the risk to man and his environment which may result from these wastes in the future. Based on these analyses, the AEC will be able to decide whether it will be necessary to remove these wastes from the burial areas.

⁴"Environmental Monitoring at Major USAEC Contractor Sites," WASH-1259, USAEC, August 1973.

TABLE 3

QUANTITIES OF SOLID WASTE AND TRANSURANIUM NUCLIDES
STORED/BURIED AT MAJOR AEC SITES THROUGH JUNE 1974^a

<u>Site</u>	<u>Total Estimated Radioactive Waste</u> ^b		<u>Readily Retrievable Waste</u>	
	<u>Cubic Meters</u>	<u>TRU-Kilograms</u>	<u>Cubic Meters</u>	<u>TRU-Kilograms</u>
LASL	222,000	14	1,317	7
NRTS	159,000	492	22,172	116
ORNL	176,000	14	879	5
RL	200,000	387	3,144	26
SRP	273,000	45	1,070	21
TOTAL	1,030,000	952	28,582	175

a - Based on information contained in July 1974 Waste Management Site Plans except Richland which is based on the July 1973 site plan data.

b - Excluding quantity of TRU nuclides disposed of to on-site cribs, ponds, and pits. (About 193 kg are contained in these areas at major AEC sites.)

SNS PLUTONIUM OPERATIONS

I. General Program Aspects

For the past 15 years, the AEC has been developing Radioisotope Thermoelectric Generators (RTG) to provide electrical power for both space and terrestrial applications. These devices convert thermal energy derived from the decay of radioactive isotopes to electrical energy utilizing the thermoelectric properties of bimetallic couples. Several isotopes have been considered as thermal sources; however, the most desirable one from an overall system point of view has proved to be Plutonium-238. The following discusses Space Nuclear Systems (SNS) operations utilizing Plutonium-238 as a fuel form.

A. Purpose

The purpose of the Plutonium RTG program is to develop long lived, unattended, reliable and light weight power system for space applications.

It is the intent of SNS to develop these systems so as not to present undue hazards to operating personnel, to the general public or to the worldwide population.

B. Advantages of Nuclear Systems

The following are advantages of using nuclear systems for space missions:

1. With the proper isotope, the system can be designed for long life, it can be compact, and it can provide a high power to weight ratio.
2. In space, it need not be dependent on sunlight or sun orientation. All outer planet exploration missions must use nuclear power because of the lack of solar energy.
3. A nuclear system is naturally radiation hardened for military applications and will withstand radiation emitted by planets.
4. It can provide both electrical and thermal power anywhere in space.

C. Specific Advantages of Plutonium

Plutonium-238 provides a specific power which enables the RTG design to meet the advantages indicated above. Because it provides its thermal power from alpha decay, little shielding is required. Also, because of its reasonably long half-life, no power flattening is required for missions as long as 10 years. The oxide of plutonium provides a reasonably high melting point which permits the design of space systems to preclude Plutonium-238 releases in thermal environments to which the system may be exposed if flight aborts should occur.

D. Safety Criteria

The general safety design objectives of a radioisotope heat source and/or its associated power system components are to contain the radioactive materials (the isotope fuel form and its radioactive products) and to limit the interactions of the radioactive materials with humans and the environment. For normal operations, containment and interaction requirements are absolute; for potential accident situations, containment requirements are based on probabilistics and are determined by a given source term (amount of radioactive material released) and/or the direct external dose to one or more random human receptors.

Containment and the limiting of interactions between the radioactive source, humans, and the environment are absolutely maintained in normal factory-to-flight and post missions operations. The exposure limits for radiation workers, individuals and the general public applicable to normal mission operations are those set forth in the Code of Federal Regulations (CFR), Atomic Energy Commission Manual Chapters (AEC-MC) and the International Commission on Radiological Protection (ICRP).

In the event of random earth impact situations, the safety objectives are to locate and recover the nuclear heat source(s).

For accident situations, the following recent probabilistic criterion is required of the system: the total probability of releasing one millicurie of fuel and/or its daughter products or exposing one or more people to a direct external radiation dose exceeding the limits set forth in the above documents should be less than 10^{-5} per flight, given the occurrence of any accident. For source terms other than one

millicurie, scaling the fuel release probability inversely-proportional to the source term should be considered. The above probabilistic criteria should be demonstrated at a reasonable confidence level ($\geq 50\%$) by analysis and/or test.

E. Safety Assessment

Before any nuclear system is used, it must be reviewed and evaluated on the bases of risk to the general public and the environment by an Interagency Nuclear Safety Review Panel (INSRP) who must submit a Safety Evaluation Report (SER) to National Security Council (NSC) for presidential approval action. This INSRP is comprised of experts in the field of nuclear system design, launch vehicle design, accident evaluations, aerodynamics, thermodynamic, meteorology, terradynamics, oceanography, astrophysics, health and safety, biology, medicine and others from EPA, NASA, DOD, AEC, NOAA, and their government agencies and their contractors.

The review covers all aspects of possible accidents from factory-to-flight including ground transportation, launch, suborbital, orbital reentry, impact and post impact situations. The panel reviews the results of safety verification tests which are generally conducted by the systems contractor to evaluate the response of the heat source in overpressure, launch-pad fires, reentry, impact and post-impact environments.

A Safety Evaluation Report prepared by the panel provides an analysis of the risk to man and the environment based on the results of the INSRP review.

Since the AEC is not the user agency of the nuclear systems, it is not responsible for benefit analyses and environmental impact statements. These requirements and the request for launch approval are the responsibility of the user agency.

II. Plutonium Applications

A. Current Applications

The following programs are currently using plutonium or contemplate using plutonium in the very near future:

1. Pioneer 10 and 11 - NASA mission of two spacecraft to Jupiter which use four RTG's per spacecraft (1200 grams each RTG). Launches in March, 1972 and April, 1973.

2. Viking - NASA mission of two Mars landing spacecraft which will carry two RTG's per spacecraft (1200 grams each RTG). Launches are in August and September, 1975.
3. LES 8/9 - DOD mission of two satellites to synchronous altitude will use two RTG's per satellite (4.2 kg per RTG). Simultaneous launch of both satellites on the same spacecraft in November, 1975.
4. Mariner/Jupiter Saturn (MJS) - NASA mission of two spacecraft to Jupiter and Saturn with three RTG's per spacecraft (4.2 kg each RTG). Launches in August and September, 1977.

B. SNS Operational Sites

The following sites are presently being utilized by SNS for their RTG activities:

1. Monsanto Research Corp. (MRC), Mound Laboratory, Miamisburg, Ohio, provide encapsulated, sealed plutonium heat sources for use in all of the programs.
2. Launches are from either Kennedy Space Center or Cape Canaveral Air Force Station, Florida.
3. The AEC's Savannah River Plant at Aiken, South Carolina, will begin providing Plutonium-238 fuel forms for the space program in the 1977 time period.

C. Current Plutonium Inventories in Space

Since 1961, the AEC has launched 34.7 kilograms of Pu-238. Of this amount, one kilogram was released to the earth's atmosphere by burn-up during reentry, two kilograms were recovered from the Santa Barbara channel after a missile abort, two and a half kilograms deposited and contained in the South Pacific Ocean near the Tonga Trench, after an aborted Apollo mission, 13 kilograms are on the lunar surface, 6.8 kilograms are in long lived earth orbit, and 9.4 kilograms have been ejected out of our solar system into deep space.

D. Forecast Inventories at Fabrication Sites

Plutonium inventories planned at fabrication sites for future SNS activities include the following:

1. Mound Laboratory - Unencapsulated fuel at Mound will be decreasing from about 40 kg at present to zero by FY 1980, in accordance with SNS plans to transfer fuel from fabrication to Savannah River. Mound Laboratory will continue assembly of heat sources in the future, using encapsulated fuel forms supplied by Savannah River. Time-average inventories of this encapsulated fuel at Mound Laboratory are expected to approximate 30 kg/yr by FY 1978.
2. Savannah River - Beginning in FY 1977, inventories will increase from zero to a time-averaged level approximating 40 kg/yr, including associated process salvage.
3. Inventories in R&D at Savannah River and Mound Laboratory will approximate 3 and 7 kg/yr, respectively. Time-average inventories in recovery operations at Savannah River may approximate 10 kg/yr.

E. Future Applications

Additional Viking missions to Mars are planned by NASA in the 1981 time period. Post 1980 missions planned by NASA include a Mariner Jupiter Uranus flyby, a Mariner Jupiter Orbiter, a Pioneer Jupiter Orbiter, a Pioneer Jupiter Probe and a Mars Sample return mission. The DOD has several planned missions in the post 1977 time period. All of these missions will utilize plutonium fuels for electrical power. The following plutonium inventories are contemplated to meet the above requirements:

1. FY 75 - 30.9 kg
2. FY 76 - 19.4 kg
3. FY 77 - 14.4 kg
4. FY 78 - 35.3 kg
5. FY 79 - 59.0 kg
6. FY 80 - 69.7 kg
7. FY 81 - FY 86 - 331.8 kg

III. Control Technology

A. Past Effluent Releases

Early isotopic space systems were designed to burn up on reentry yet provide absolute containment for all ground handling and accident situations. In 1964, SNAP 9A burned up on reentry as designed after an abort on ascent to orbit. One kg of Plutonium-238 was burned up in the stratosphere (particle size - 0.4μ).

Systems after the SNAP 9A were designed for intact reentry. Two aborts after 1964 released no radioactive material. The first, a Nimbus spacecraft was aborted in 1968 after launch at the AF Western Test Range. It was intentionally destructed at 100,000 feet after the launch vehicle went off-course. The radioisotope capsules were retrieved intact from the Pacific Ocean (contained 2 kgs). The second, an Apollo 13 aborted in 1970 and the LEM returned to earth with the radioisotope capsule. The capsule sustained reentry and impacted in the deep ocean south of the Fiji Islands in at least 20,000 feet of water with the Plutonium-238 contained in multi-containment capsules.

B. Projected Effluent Control Plans

No releases are projected in the future. Systems are designed to remain intact under all normal and all credible accident conditions. Also in the design of future systems, consideration is given to possible search and recovery of aborted systems. Plans and techniques for worldwide search and recovery exist now and are continually being updated. Salt water actuated pingers are standard items on all flights to assist in locating and recovering the Pu-238 in the event of an advertent water impact, should early launch vehicle accidents occur.

Effluent control at the Savannah River is under the Production Programs and at Mound Laboratory is under Weapons Programs, except that the SNS operations at Savannah will be a "dry process" producing no effluents.

C. Containment Test Programs

1. To verify containment design, extensive safety test programs are conducted under all simulated accident conditions: fire, over-pressure, reentry, impact, and post-impact environments.

IV. Projected Waste Quantities

No waste quantities are projected as the result of space operational activities.

Manufacturing site wastes are projected as follows:

1. Savannah River - less than 120 grams/yr, accumulating annually beginning in FY 1978.
2. Mound Laboratory - less than 600 grams in FY 1975 decreasing to less than 50 grams in FY 1980 and thereafter.
3. Transuranium wastes are sent to appropriate AEC burial sites.

DIVISION OF PRODUCTION AND MATERIALS MANAGEMENT
Overview on Plutonium and Transuranic Elements -
Source Terms and Operations

1. Production and Materials Management Program Summary

The nuclear reactors at AEC's Richland and Savannah River plants were built to produce nuclear materials for the Nation's defense program. Continued operations under AEC policies will enable the plants to continue providing protection of the population and the environment from adverse effects of radioactivity while fulfilling the function of producing plutonium and tritium for National defense. The principal product at Richland is plutonium and at Savannah River both plutonium and tritium are produced. Small quantities of other transuranic isotopes such as Np-237, Pu-238, Am-241, Am-243, Cm-244 and Cf-252 are produced at the AEC production sites. The Idaho Chemical Processing Plant, which recovers enriched uranium from test reactor and Navy irradiated fuels, is also a part of the production program.

The Richland plant was initially built during the second world war and has been in operation about 30 years. The original plant consisted of a uranium fuel fabrication facility, three graphite moderated water-cooled reactors, two chemical separations plants for plutonium recovery and decontamination and a facility for the final isolation and purification of plutonium nitrate solution. Production capacity at Richland was expanded on numerous occasions between 1948 and 1962. Six additional reactors were built including N reactor which is a dual purpose reactor producing steam for electric power production in addition to plutonium. Only N reactor is presently operated. The original batch-type plants for the separation of plutonium were shutdown by 1956 and replaced by two continuous solvent extraction plants. Neither of these plants are being operated today but one plant is being held in standby for future operation. Facilities were also provided for conversion of plutonium nitrate to oxide and metal and for the fabrication of weapons components. The fabrication plant has been shutdown for several years and is currently being dismantled. The Richland plant includes facilities for the treatment and storage of liquid radioactive wastes.

The Savannah River Plant was built in the early 1950's and included facilities for fuel and target element fabrication, five heavy water moderated and cooled production reactors, two chemical reprocessing plants for both plutonium and enriched uranium recovery, tritium separations facilities, and a heavy water production plant. Currently, only three reactors are operating. Facilities have been added for the fabrication of targets to be irradiated to produce other transuranics such as Np-237, Pu-238, Am-243, Cm-244, and Cf-252 and for the separation of these products. New facilities are currently being provided to fabricate Pu-238 oxide fuel forms for thermoelectric power sources. Facilities are also provided for the treatment and storage of liquid radioactive wastes.

The Idaho Chemical Processing Plant includes facilities for the separation and purification of irradiated enriched uranium from test reactor and Naval reactor fuels. Facilities are also provided for the temporary storage of liquid radioactive waste which contain transuranic elements. The liquid waste is converted to a calcined granular solid in a fluid bed and subsequently stored in stainless steel bins.

2. Applications of Transuranics

The principal mission of the AEC production reactors is the production of plutonium and tritium for weapon application. However, significant quantities of non-weapon plutonium has been produced in support of the civilian nuclear reactor development program and smaller quantities for foreign sales. Pu-238 is produced for use as a heat source in thermoelectric power for space application and for R&D for possible medical application (pacemakers, artificial heart, etc.). Cm-244 has been used in R&D programs and may be used as a replacement or supplement for Pu-238 in space applications. Cf-252 is used as a neutron source for neutron radiography, cancer therapy, etc.

The AEC has produced several tens of tons of plutonium in its reactors including several tons of non-weapon grade plutonium since the beginning of the production program in the mid 1940's. In addition, 200 kg of Pu-238,

4 kg Cm-244, and 2 grams of Cf-252 have been produced in Savannah River reactors. Additional quantities of transplutonium elements have been produced in Savannah River reactors, separated and refabricated into targets for further irradiation as part of the Cf-252 program.

3. Control Technology

The AEC plutonium production plants control releases of transuranic elements by the following procedures:

1. All process air passes through at least two high efficiency particulate filters (HEPA) before release. All effluents are monitored for releases.
2. Any liquid waste streams containing significant quantities of transuranic elements are stored in waste tanks. Liquids containing extremely low levels of transuranics such as evaporator overheads are released to seepage basins or other controlled facilities.

The total current releases of transuranics to controlled facilities at Richland and Savannah River are about one curie per year while stack releases from Richland and Savannah River average about 0.0013 and 0.02 curies, respectively. Releases to date from our plants are shown in the table below.

Releases of Transuranics in Curies to July 1, 1974

	<u>Richland</u> ^{1/}	<u>Savannah River</u> ^{2/}	<u>Idaho Chemical</u> ^{3/} <u>Processing Plant</u>
Stack Releases - Ci	1.36	3.64	*
Seepage Basins, Ponds, Cribs, Ditches, etc. - Ci	18,750**	20.13	0

* In the separation of enriched uranium at Idaho, fission products and plutonium are separated from enriched uranium. The high level liquid waste which contains the fission products and plutonium is subsequently calcined to a granular solid. In this operation a small

amount of radioactivity is released from the plant stack. The plutonium releases from this operation which are essentially below the level of detection are reported as less than 1 curie per year.

** The true level may be only about 66% of the reported level due to the overstatement of quantities in the Z-9 crib.

In an effort to further reduce releases, the liquid process waste from the plutonium finishing and scrap recovery operation, which was being discharged at Richland to cribs or covered ditches, is being sent to waste storage tanks. At Savannah River, a new sand filter is being built in each of the two separations areas and all air from the plutonium and transplutonium processing operations will be routed through a final sand filter in addition to at least two HEPA filters. At Idaho, a filter is being installed which will substantially reduce stack releases of radioactivity which is principally fission products but contains traces of transuranics.

The AEC has established new criteria for facilities which process or store plutonium including resistance to fires, earthquakes and tornados. Facilities meeting the new criteria are available at Richland for the storage of plutonium as metal, oxide, nitrate solution or scrap. Vaults meeting the new criteria are available at Savannah River for plutonium storage. At Savannah River a new facility is being built within an existing building for converting Pu-238 oxide powder to specific fuel forms. This building is being upgraded to meet the new plutonium criteria. The process air from this building, which will be filtered by from two to four HEPA filters, will be routed through the new F Area sand filter as further backup against potential releases.

4. Future Projections

In the future, if production levels continue at about the same level, the releases of transuranic elements can be expected to be held at about the present level or slightly reduced due to the use of new facilities which are under construction. Increases in the production of transuranic elements with short half-lives such as Pu-238, Cm-244, Cf-252 could

require additional control facilities to control releases at the current level. Such facilities would be provided in accordance with accepted AEC policies; however, at the present time there are no plans for increased production of these elements.

-
- 1/ Data taken from [ARH-2806 4Q REV], "Radioactive Liquid Wastes Discharged To Ground In The 200 Areas During 1973", and [ARH-2807 4Q], "Radioactivity In Gaseous Waste Discharged From The Separations Facilities During 1973", and updated with reports for first six months of 1974.
 - 2/ Data taken from draft WASH-1537 Environmental Statement - Waste Management Operations, Savannah River Plant and updated from SR monthly reports.
 - 3/ Data taken from draft Environmental Statement, Waste Management Operations, National Reactor Testing Station.

Overview Reactor Research and Development Program Area
Source Terms and Operations

Prepared for

EPA Public Hearings - 12/10/74

1. Reactor Research and Development Program

For over 20 years the AEC has been engaged in the development of nuclear power reactors to help meet the Nation's need for energy. To date, several reactor systems have been successfully developed and some have come into commercial use. These commercial reactors are now producing about 5 % of the Nation's electricity from the energy released from the fission of uranium, with plutonium as a by-product.

Because of limitations inherent in today's nuclear power plants, only 1 to 2% of the energy potentially available in uranium can be used. The fast breeder reactor, which is in an advanced stage of development can extract 60% or more of the energy in uranium, including depleted uranium, and also utilize the by-product plutonium for the initial fissile fuel. This use of plutonium, through the development of the fast breeder reactor to the point of large scale commercial application would mean the availability of low cost uranium sufficient to provide a large fraction of the Nation's electric energy requirement for hundreds of years, if needed.

Several promising breeder concepts have been investigated and the focus of the AEC reactor development program is now on the sodium cooled Liquid Metal Fast Breeder Reactor (LMFBR). At the same time, other breeder options are being held open by carrying forward technology efforts for breeder reactor concepts such as the Light Water Breeder Reactor (LWBR), the Molten Salt Breeder Reactor (MSBR), and the Gas-Cooled Fast Breeder Reactor (GCFR).

The LMFBR program has as its objective the timely development of technology for a breeder reactor that will offer a commercially competitive and environmentally acceptable option for helping to assure the Nation's long-term electric energy supply. The program recognizes that domestic economic uranium (and thorium) resources are finite and that of the 220 or more nuclear power plants, now in operation, under construction, or on order in the U.S., the preponderance are light water reactors which operate on the uranium-plutonium fuel cycle. The overall LMFBR program for the development of the required broad technological and engineering base, is being carried out by AEC laboratories and by industrial firms at a number of locations throughout the country. This includes many facilities which are variously used to permit testing of physics, fuel, components and instrumentation. Some of these facilities have plutonium inventories.

The total of the plutonium inventories at RRD facilities is in the range of 4000 kgs Pu. Table 1 lists the inventories of plutonium reported by each of the major field offices for RRD programs as of June 30, 1974. Table 2 shows the estimated annual Pu requirements for major RRD programs thru 1986.

August 20, 1974

Table 1

RRD Pu INVENTORY
MAJOR FIELD OFFICES BY COEI*
TOTAL KGS AS OF 6-30-74
NMIS REPORT P-114

Categories									
Field Office	110	120	130	140	150	160	170	180	Total
LASL (Los Alamos Scientific Laboratory)					6.7				6.7
Chicago		786.1		2.8	2661.5		22.6	3.9	3476.9
Oak Ridge		1.8		0.4	3.1		1.2	0.1	6.6
Richland	0.1	266.5	8.4	12.1	524.5		86.5	7.5	905.6
Idaho					0.2				0.2
San Francisco		0.1	0.7	1.4	1.8		0.3	2.4	6.7
Subtotals	0.1	1054.5	9.1	16.7	3197.8		110.6	13.9	4402.1
% of Total	-0-	24.0	-0-	0.5	72.5		2.5	0.5	100.0

*Composition of Ending Inventory

110 Stored, No definite use
120 Stored, Definite use
130 In fabrication or conversion
140 In reactor or critical
150 Misc. R&D (Not included above)
160 Irradiated and unirradiated scrap in recovery
170 Unirradiated scrap stored
180 Irradiated scrap stored

9/13/74

Table 2

MAJOR RRD PROGRAMS REQUIRING PLUTONIUM
(Annual Requirements in Kilograms Total Plutonium) (Rounded)

Program or Project <u>1/</u>	1975	1976	1977	1978	1979	1980	1981	1982	1983	1984	1985	1986
FTF Core Requirements	870	480	400	160	0	0	800	800	800	800	800	800
CRBR Plant	-	-	-	-	460	900	900	430	900	900	*	*
LMFBR - RD Program	170	160	140	200	260	240	280	350	350	350	350	350
ZPR	50	20	0	250	-	500	500	500	500	-	-	-
TOTAL	1090	660	540	610	720	1640	2480	2080	2500	2050	1150	1150

Additional Pu for Clinch River Breeder Reactor to be supplied by plant owner.

1/ FFTF - Fast Flux Test Facility

CRBR - Clinch River Breeder Reactor (Demonstration Plant)

LMFBR - Liquid Metal Fast Breeder Reactor

ZPR - Zero Power Reactor

In June 1973 the AEC began preparation of an environmental impact statement on the overall LMFBP Program (WASH-1535) as required by NEPA. The environmental impact statement which was developed, and is now in the final stages of review prior to release, differs from the conventional statement in that the important environmental and societal impacts [which are assessed] would not arise from the proposed research and development activities, but rather from the eventual commercialization of LMFBPs which would be expected if the research and development goals were attained. It was necessary therefore to look ahead some 40 to 50 years to foresee an LMFBP industry that would constitute about 40% of the total installed electric generating capacity in the U.S. by the year 2020.

Many estimates and forecasts were made for this study which could prove useful in assessing the quantities and impacts of the plutonium that would be in use. Pertinent tables from the report will be used in the following discussion. In addition, an intensive cost-benefit assessment was undertaken, considering both the direct and the indirect costs and benefits of the development of the LMFBP.

The direct costs were confined to LMFBP development program costs and to those costs normally imbedded in the price of electrical energy. Calculations for over 70 separate postulated cases were performed to estimate U.S. electrical power production costs over the period 1974 - 2020. One of the major quantifiable conclusions of the direct economic analysis was that the introduction of a fast

breeder into the U.S. electric power utility system will produce significant financial benefits. These benefits result largely from a reduction in uranium ore and enrichment requirements. Additionally, the fast breeder results in a nuclear power industry that will have total power costs, in constant dollars, that decrease with time. Capital requirements accumulated to the year 2020 are estimated to be about 10% less with a fast breeder industry due to the reduced requirement for mining, milling and enrichment facilities.

With the breeder, the nuclear industry can eventually free itself of the need to mine uranium. The advanced oxide fueled breeder, with an estimated 10 year compound doubling time, can meet the fissile fuel demand of a growing nuclear power industry with self-generated plutonium shortly after the turn of the century.

Specifically, without the LMFBR, the cumulative U_3O_8 requirement to the year 2020 is 6.3 million tons, while the cumulative U_3O_8 requirement with the LMFBR is 2.6 million tons. Without the LMFBR, U_3O_8 will be mined at an ever increasing rate, while with the LMFBR, the annual ore requirement becomes insignificant after about the year 2015, with similar trends for separative work capacity requirements.

Indirect benefits, though no less significant, come partly from the improved resource utilization. The use of plutonium as a reactor fuel for electric power production would free the finite resources of fossil fuels for their optimum use, thus assuring the domestic

availability of fuel for electric power production for the long term. A mature LMFBR industry together with the non-breeders can generate sufficient fissile material for the nuclear industry to be able to use stockpiled depleted uranium for hundreds of years as the sole source of fertile fuel material. A premium market would be established for the plutonium produced by LWRs. There would also be the potential for substituting electricity for fossil fuels in energy-intensive applications.

Other indirect benefits of note are in the cumulative health and safety effects for both occupational and public groups. Major improvements in occupational health and safety arise from the fact that the LMFBR system does not have associated with it the mining of uranium and replaces fossil plants burning coal. Timely introduction of the breeder could reduce occupational accident fatalities by 3000 persons through year 2020 (and an additional 2000 persons after 2020 due to plants then in operation). In contrast the use of coal instead of nuclear fuel would lead to an additional 51,000 occupational fatalities and 27,000 more public fatalities due to associated fuel transportation accidents.

These are but a few of the conclusions from the extensive study soon to be published, which will be available to the EPA.

2. Applications Using Plutonium

As has already been stated, plutonium is a by-product of the reactors built, under construction and planned by the electric utility industry, both LWRs and High Temperature Gas-Cooled Reactors (HTGRs). Plutonium now is being considered as a potential fuel for recycling in the LWR reactors and as a fuel for the breeder. The AEC Directorate of Licensing has prepared a draft generic environmental statement for the use of recycle plutonium in light water-cooled reactors (GESMO - WASH-1327, August 1974). Starting on page VIII-65 of "GESMO" there is a discussion of the dollar value of plutonium as a reactor fuel for recycling. Using the assumptions stated, plutonium has a near-term value of about \$11.00 per gram fissile. Since this value is tied in to the cost of enriched uranium, the value continues to rise to about \$16.50/gram fissile plutonium in 1995. As a breeder reactor fuel the value is expected to be considerably higher. These are the prospective values shown in GESMO:

Projected Plutonium Value*
(\$/gm fissile plutonium)

<u>Year</u>	<u>Pu Value</u>
1975	9.90
1980	11.75
1985	14.89
1990	15.81
1995	16.50

*if recycled promptly in LWR's.

To make the assumed value more meaningful one may look at the quantity of plutonium that will be produced by the commercial reactors and that can be

reused as reactor fuel. AEC's publication, "Nuclear Power Growth 1974-2000" (WASH-1139 (74), February 1974) projects 4 cases. Case D, one of the more conservative cases, assumes a general reduction in the growth of electricity use and some improvement over recent experiences in nuclear plant construction and regulation. Under these assumptions, Case D forecasts the following nuclear generating capacity in thousands of MW:

<u>Year</u>	<u>MW x 10³</u>	<u>Year</u>	<u>MW x 10³</u>
1975 -	47.3	1990 -	475
1980 -	102.1	1995 -	760
1985 -	250.0	2000 -	1090

Table 3 taken from WASH-1139 (74), shows the fissile plutonium recovery and utilization in the U.S. using Case D assumptions.

As shown, in Table 3, the total plutonium recovered by the year 1995 would aggregate to about .86 million Kg. Assuming a value of \$16.50/g in 1995 (according to GESMO) this inventory would be worth about \$14.2 billion if it were to be used in recycling, and potentially more as a breeder fuel. Considering the plutonium inventories and their prospective values, it would appear that we are already in a plutonium economy

3. Control Technology - Identification of Source Terms

In the LWR and HTGR fuel cycle, plutonium is present in the irradiated or spent fuel. It is stored for a cooling period and then shipped in shielded containers to a reprocessor for the recovery of plutonium and uranium.^{1/}

^{1/} Environmental Statements have been prepared by Directorates of Licensing and Regulatory Standards, USAEC.

Table 3

FISSILE PLUTONIUM RECOVERY AND UTILIZATION, KILOGRAMS
United States, Case D (Plutonium Recycle, 75% capacity factor)

-CY-	RECOVERY					UTILIZATION					YEAR END INVENTORY
	LWR	BREEDER	NATURAL AND OTHER	TOTAL ANNUAL	CUMULATED	LWR RECYCLE	BREEDER FUEL	OTHER USES	TOTAL ANNUAL	CUMULATED	
1973	0	0	0	0	0	0	0	0	0	0	0
1974	0	0	0	0	0	0	0	0	0	0	0
1975	230	0	0	230	230	0	0	16	16	16	220
1976	2400	0	0	2400	2600	380	0	680	1060	1090	1570
1977	5200	0	0	5200	7800	2400	0	1500	3900	5000	2800
1978	8600	0	0	8600	16400	6300	0	1690	8000	13000	3400
1979	10200	0	0	10200	26600	7400	0	2100	9500	22500	4200
1980	10300	0	0	10300	36900	7900	0	2400	10300	32900	4100
1981	12200	0	0	12200	49100	9700	0	2400	12000	44900	4300
1982	17000	0	0	17000	66200	14900	0	0	14900	59700	6500
1983	21300	0	0	21300	87500	18800	0	0	18800	78500	7000
1984	25300	0	0	25300	112800	23100	0	0	23100	101500	11200
1985	30400	0	0	30400	143200	28500	0	0	28500	120000	12200
1986	35600	0	0	35600	178800	31200	0	0	31200	161200	17600
1987	42800	0	0	42800	221600	35800	3400	0	39200	200400	21100
1988	48300	0	0	48300	269900	41900	820	0	42700	243100	24700
1989	54900	0	0	54900	324800	41200	7200	0	48400	291500	31300
1990	62300	860	0	63200	388000	45000	11400	0	56400	348000	4000
1991	69800	1480	0	71200	459200	46300	18000	0	64300	412300	47000
1992	77900	3000	0	80900	540100	48700	25500	0	74200	486400	53700
1993	86300	6600	0	92900	633100	49400	36200	0	85600	572000	61100
1994	94100	11800	0	105900	739000	48700	48800	0	97500	669500	69500
1995	101100	19400	0	120500	859500	47800	65400	0	113200	782700	76800
1996	108200	29400	0	137600	997100	46100	82400	0	128500	911200	89000
1997	115500	42000	0	157400	1154500	44600	103400	0	147900	1059100	96400
1998	121900	57600	0	179500	1334000	49000	123800	0	172800	1231900	104000
1999	127400	76100	0	203500	1537500	57000	137400	0	194400	1426300	111200
2000	132400	97400	0	229800	1767300	59000	164300	0	223300	1649600	117700

In the LMFBR fuel cycle, plutonium and uranium bearing materials would be combined at the fuel fabrication plant for use in the reactor. After cooling, the irradiated fuel would be shipped in specially built containers to the reprocessing plant. The separated fission products would be solidified and eventually shipped to a waste storage facility^{2/} and the plutonium would be recycled as LMFBR fuel. A summary of materials and quantities shipped for a 1000 MW LMFBR is shown in Table 4.

2/

WASH-1539, Draft Environmental Statement" Management of Commercial High Level and Transuranium Contaminated Radioactive Waste" September, 1974.

Table 4

A SUMMARY OF MATERIALS AND QUANTITIES SHIPPED FOR A 1000-Mw LMFBR REACTOR

Type of shipment	Probable mode of transport	Quantity shipped per year ^a (kg)	Quantity shipped per package ^a (kg)	Est. gross package weight (kg)	No. of packages per vehicle	Heat generated per package (W)	Est. activity per package (Ci)	Average No. of shipments per year	Est. average shipping distance ^{b,f} (miles)	Shipment destination ^j
Unirradiated material										
UF ₆	Truck	17,335	8,604	15,090	1		3.21	2.01	750	FP
UO ₂	Truck	16,048	97	245	64	2.6×10^{-3}	1.60	2.58	750	FP
PuO ₂	Truck	1,679	9	105	64	81	1.04×10^3	2.91	750	FP
Fresh fuel - core and axial blanket	Truck	16,123	235	1,820	4	218	2.81×10^3	17.15	750	PS
Fresh fuel - radial blanket	Truck	2,702	229	1,820	4	6.3×10^{-3}	3.78	2.95	750	PS
Irradiated material										
Spent fuel - core and axial blanket	Rail	15,462	667	114,000	1	1.33×10^4	3.26×10^6	22.83	750	RP
Spent fuel - radial blanket	Rail	2,669	681	114,000	1	1.83×10^3	4.54×10^3	3.92	750	RP
Type of shipment	Probable mode of transport	Quantity shipped per year (ft ³)	Quantity shipped per package (ft ³)	No. of packages per year	No. of packages per vehicle	Heat generated per shipment (W)	Est. activity per shipment (Ci)	Average No. of shipments per year	Est. average shipping distance ^{b,f} (miles)	Shipment destination ^j
Waste from fuel preparation and fabrication plants										
Alpha waste	Rail	5,000 ^b	^c	^c	^c	22.5	2.79×10^4	5	1,000	FR/BG
Low-level beta-gamma waste	Truck	21,000 ^b	7.4	2,800	64	2×10^{-3}	0.475	45	500	BG
Solid waste from power stations										
Low-level beta-gamma waste	Truck	2,000	7.4	270	64	2.85	475	4.2	500	BG
Tritium	Truck	6.12 ^d	0.16	38	64	1.13	3.2×10^6	0.60	1,000	FR/BG
Solid wastes from reprocessing plants										
Alpha waste	Rail	770 ^b	^c	^c	^c	22.5	2.79×10^4	0.77	1,000	FR/BG
Alpha-beta-gamma waste	Rail	2,000 ^b	25	80	3	1.12	500	27	1,000	FR/BG
Low-level beta-gamma waste	Truck	3,000 ^b	7.4	405	64	2×10^{-3}	0.475	6.3	500	BG
Cooling hulls	Rail	165 ^e	3.5	47	36	10.3×10^3	1.46×10^6	1.3	1,000	FR/BG
High level waste	Rail	55	6.28	9	12	2.5×10^6	7.8×10^8	0.75	1,500	RSSF
Noble gases	Truck	1.7 ^f	1.7 ^g	1.0	6	1.47×10^3	9.0×10^3	0.164	1,500	NGSP
Iodine	Truck	0.078	0.16	0.5	64	1.0×10^{-3}	1.46	0.0076	1,000	FR/BG

^aAll quantities of materials shipped are given in kilograms of heavy metal.^bCompacted a factor of 10 from original volume generated.^cAlpha waste is packaged in 55-gal (7.4-ft³) drums and large boxes; each rail car contains 1000 ft³ of waste.^dShipped as Ca(O³II)₂.^eHulls compacted to 8.8 ft³ per ton of fuel.^fCompressed gas at 2,200 psi.^gStandard gas cylinder.^hEstimated distance to one-of-a-kind repository, 1500 mi; between facilities, 750 mi; to multiple burial ground sites, 500 mi.ⁱDistance of 1000 mi is a compromise between 1500 mi to one-of-a-kind repository and 500 mi to multiple burial ground sites.^jF: fabrication plant; PS: power station; RP: reprocessing plant; FR: Federal repository; BG: burial ground; RSSF: retrievable surface storage facility; NGSP: noble gas storage facility.

The estimated releases of alpha-emitting transuranic elements from normal operations of the LMFBR fuel cycle are summarized in Table 5. The principal airborne release is from the fuel reprocessing plant, totaling 0.36 mCi per 1000 MWe-year. Other airborne releases are small by comparison. Liquid transuranic effluents are assumed to occur only for the fuel fabrication plant and are estimated at 0.05 mCi per 1000 MW-year.

The estimated population dose from alpha-emitters expected from operation of the LMFBR fuel cycle is shown in Table 6. The estimated man-rem exposure from LMFBR transuranic releases is shown in Table 7.

In addition to normal operational experience various accident situations were postulated in developing the environmental impact statement for the LMFBR program. Estimated releases of transuranic elements for major accident categories are summarized in Table 8 which shows only those quantities of material estimated to pass all containment barriers which have been designed into the system. The total plutonium released annually due to postulated transportation accidents would average less than 10^{-6} Ci per year associated with the operation of a 1000 Mw LMFBR.

Table 5

ESTIMATED RELEASES OF TRANSURANIC ELEMENTS DURING NORMAL OPERATION OF THE LMFBR FUEL CYCLE

Facility or Process	Release Mechanisms	Protective Systems	Nature of Release	Radio-nuclide	Quantity (Ci/1000 MWe Yr)
Fuel Repro- cessing	Particles entrained by gas from all parts of process	Process equipment, cells, and HEPA ^b filters (CFC = 2×10^9)	Airborne particles of PuO_2 or $\text{Pu}(\text{NO}_3)_4$ AMAD $\sim 0.3 \mu\text{m}$	Pu-238	$.18 \times 10^{-3}$
				Pu-239	$.04 \times 10^{-3}$
				Pu-240	$.05 \times 10^{-3}$
				(Pu-241) ^d	(5.4×10^{-3})
				Am-241	$.01 \times 10^{-3}$
				Cm-242	$.07 \times 10^{-3}$
				Cm-244	$.01 \times 10^{-3}$
				Total α	$.36 \times 10^{-3}$
			Liquid		zero
Fuel Fabri- cation	Particles entrained by gas, principally from PuO_2 powder grinding	Glove boxes, roughing and HEPA filters (CF = 10^{12})	Airborne particles of PuO_2 AMAD $\sim 0.3 \mu\text{m}$	Pu-238	$.36 \times 10^{-6}$
				Pu-239	$.07 \times 10^{-6}$
				Pu-240	$.10 \times 10^{-6}$
				(Pu-241) ^d	(10×10^{-6})
				Am-241	$.02 \times 10^{-6}$
				Total α	$.55 \times 10^{-6}$
			Liquid		
				Pu-238	$.030 \times 10^{-3}$
				Pu-239	$.006 \times 10^{-3}$
				Pu-240	$.008 \times 10^{-3}$
				(Pu-241) ^d	$(.91 \times 10^{-3})$
				Am-241	$.002 \times 10^{-3}$
				Total α	$.046 \times 10^{-3}$
Reactor	Failed fuel to sodium to cover gas	Cover gas purification system			negligible

Table 5 Continued

Facility or Process	Release Mechanisms	Protective Systems	Nature of Release	Radio-nuclide	Quantity (Ci/1000 MWe Yr)
Waste storage	Exhaust air from receiving and handling areas	HEPA filters			$< 1.5 \times 10^{-6}^f$
Transportation					zero

^dHEPA = high efficiency particulate.

^eAMAD = activity median aerodynamic diameter.

^fBeta emitter not included in totals. The 5.4×10^{-3} Ci of Pu-241 decays to produce an additional 0.16×10^{-3} Ci of Am-241, which is included as a part of the source term, where appropriate.

^gCF = Confinement factor = Ci processed/Ci released.

^fThe estimated normal release of radioactivity from waste storage is 1 mCi/yr (4.6.3.1); this includes all radio-isotopes, the isotopic distribution of which will depend upon a number of variables. The RSSF is forecast to hold all of the commercial high-level waste produced through the Year 2000, including all types of reactors. In order to be conservative it is assumed that the entire 1 mCi consists of α -emitting transuranics solely from the LMFBR power capacity during a given year only (1999). (This ignores cumulative power production up to 1999 and radio-isotopes other than the α -emitting transuranics.) In 1999 LMFBR generating capacity is estimated at 178 000 MWe-year [derived from the metric tons of uranium fabricated in that year (3090 - Sect. 4.1) and the metric tons of uranium required per 1000 MWe-year reactor (17.335 - Fig. 4.5-1)]. In addition, there will be 20,170 metric tons of uranium fabricated for other types of reactors (Table 4.1-1). Assuming that the quantity of high level waste is approximately the same from all types of reactors (see 9.1.2), then

$$1 \text{ mCi} + \frac{3090/23260}{178} = 1.3 \times 10^{-3} \text{ mCi, or } < 1.5 \times 10^{-6} \text{ Ci.}$$

Table 6

ALPHA-EMITTING RADIOISOTOPES IN THE POPULATION
OF THE UNITED STATES

Source	Amount of Alpha Emitters in U.S. Population (curies)	Population Dose from Alpha Emitters in U.S. Population (man-rems/ 70 year lifespan)	
		Bone	Lung
Natural Radioactivity	$> 10^{-1} \text{a}$	$> 7 \times 10^8 \text{a}$	$> 7 \times 10^8 \text{a}$
1000-MWe-year LMFBR	$< 10^{-8} \text{b}$	$< 30 \text{ b}$	$< 4 \text{ b}$

^aSource: Ionizing Radiation: Level and Effects, Vol. 1, "Levels," United Nations, New York, 1972.

^bSee Table 4.7-2

Table 7

ESTIMATED MAN-REM EXPOSURE FROM FALLOUT PLUTONIUM AND FROM
LMFBR TRANSURANIC RELEASES

Organ	Dose Equivalent To Current Generation from Fallout Pu ^a (man-rem)	Dose Equivalent to all Subsequent Generations from LMFBR Transuranic Releases (man-rem)	
		For Generating Capacity of 1000 MWe-Year ^b	For Year 2020 Generating Capacity of 2,200,000 MWe-year
Lung	3.0×10^6	4	0.9×10^4
Bone	1.4×10^6	17	$4. \times 10^4$
Liver	0.8×10^6	7	$2. \times 10^4$
Lymph nodes	100×10^6	200	40×10^4
Gonads	----	0.23	0.05×10^4

^aEstimate of Bennett, based on New York City air concentrations.⁶⁰

^bDerived in Tables II.G-10 to II.G-14.

Table 8

ESTIMATED RELEASES OF TRANSURANIC ELEMENTS DUE TO MAJOR ACCIDENTS ASSOCIATED WITH THE LMFBR FUEL CYCLE

Facility or Process	Type of Accident	Nature of Release	Quantity of Alpha-activity Released (10^{-3} Ci)	
			per Accident	per 1000 MWe-Yr ^a
Fuel Reprocessing ^b	Leakage of Highly Radioactive Material	to air	1.8	.002
	Solvent Fire	to air	0.11	.0001
Fuel Fabrication ^b	General Facility Fire	to air	3.7	.005
	Criticality	to air	0.18	.0002
Reactor ^c	Class 9 Core Melt-down with Breach of Primary Containment	to air	37	.00004
Waste Storage ^d	Rupture of Dropped Canister	to air	0.4	.002
Transportation ^e	Railcar Fire	to air	1.4	.0003

FOOTNOTES FOR TABLE 8

^b Beta emitter not included in totals.

^c Assumes one accident of the designated type, per fuel cycle facility, per 10-year interval (discussed in text).

^c The calculated release of plutonium to the environment from a hypothetical core disruption accident in a 1000 MWe-yr LMFBR is 0.1 gram (Sec. 4.2.7.8.3). Assuming that the alpha-emitting transuranic radioisotopes released in a HCDA consist of the same isotopic distribution, by weight, as that normally released by a fuel reprocessing plant (Table II.G-1), the activity released is as follows:

²³⁸ Pu	18.7 mCi
²³⁹ Pu	4.3 mCi
²⁴⁰ Pu	5.3 mCi
²⁴¹ Pu	577.4 mCi
²⁴¹ Am	1.1 mCi
²⁴² Cm	7.3 mCi
²⁴⁴ Cm	0.1 mCi
Total	614.2 mCi
Total α-emitting transuranics	36.8 mCi

The probability of a core meltdown with a release from the primary containment vessel with subsequent leakage of 0.1 gram plutonium through secondary containment for a 1000 MWe yr is less than 10^{-6} . Assuming a risk of 1 in 10^6 per year, or 1 in 10^6 reactor-years, then $36.8 \text{ mCi} \div 10^6 \text{ reactor-years}$ equals $36.8 \text{ mCi} \times 10^{-6}$, or 36.8 mCi per reactor-year. A maximum credible accident is estimated to release approximately 10 mCi of activity to the environment (Sec. 4.6.3.1.5). Of the waste inventory anticipated in the Year 2000, 4.5% is expected to consist of α-emitting transuranic elements ("Retrievable Surface Storage Facility Alternative Concepts: Engineering Studies," Atlantic Richfield Hanford Company, July, 1974, ARH-2888 REV, Table 10-II). Therefore, assuming a similar isotope distribution, of the 10 mCi released, 0.45 mCi would be α-emitting transuranic isotopes. If LMFBR power capacity (1999) is 178 1000 MWe-yr (see Table II.G.1-1), then the release per 1000 MWe-yr is 2.5 μCi. (This ignores power produced in previous years and so is conservative on a per unit basis.)

^c Of the 58 mCi Pu release given in 4.5.5.4, 2.4% consists of α-emitting radioisotopes (Appendix II.N.4). The number assumes a total of six 1000 mile alpha waste shipments required per 1000 MWe-year (4.5, Table 4.5-12) and 2.23 accidents per 10^6 railcar miles (Appendix II.C). There would be only $6 \times (2.23 \times 10^3) / 10^6 = 13 \times 10^{-3}$ accidents per year involving alpha waste shipments by rail. About 1.5% of all rail accidents involve fire (Appendix II.A). Thus, there would be about 2×10^{-4} alpha waste rail car fires per 1000 MWe-year. Alternatively, there would be one such fire for every 5×10^3 1000 MWe-years. Thus, the addition of this accidental release would add $1.4 \text{ mCi} / 5000 = 0.0003 \text{ mCi}$ to the routine release of 0.36 mCi per 1000 MWe-year. Americium-241 buildup from the decay of plutonium-241 would increase this to 0.0006 mCi per 1000 MWe yr.

The development of reactor technology that would be reliable and safe and reduce radioactive effluents including the transuranics, to levels that are as low as practicable has been one of the concerns of the Division of Reactor Research and Development from the beginning. One example is a current research and development program to further reduce the effluents expected in the reprocessing of IMFBR fuels to meet as low as practicable guides. This concern for public safety from man-made sources of radiation has been a feature of the fuels program, the engineering and components development program and is a major concern of the reactor safety activity of the division, as well as the statutory requirement of the AEC.

SOURCE TERM FOR TRANSURANIC MATERIAL OF WEAPONS ORIGIN

The principal environmental contaminant will be weapons plutonium. Initially, the major alpha source following a weapons plutonium release incident would be 239-Plutonium. After a decay-growth period, then would be added 241-Americium at a substantial level relative to the 239-Pu level.

The amounts of plutonium involved in the U. S. weapons program are necessarily classified. As an unclassified approximation it can be stated that yearly plutonium processing is on the order of 100,000 Curies.

The only reason for U. S. weapon plutonium to enter the environment would be from accidental circumstances. Historically, the AFC's accidental releases have taken place during production. Most plutonium fires are fully contained within the production facility; however, plutonium contamination has escaped in fractional curie amounts during one or possibly two fires. Contamination in amounts from 10 to 100 Curies escaped in a waste spill from rusted waste storage drums. Improved methods and facilities make the possibility of these types of release highly improbable. Based on accident history, releases of transuranie contamination from the AFC's weapon program total less than 100 Curies. Future accident release potentials from the weapons program are conservatively estimated at 1 Curie per year for five years lowered to a small fraction of a Curie per year after 1980 due to better facilities and improved performance. Future releases from routine operations of the weapons program are estimated at 0.01 Curies per year.

While the U. S. no longer conducts atmospheric testing of nuclear weapons, foreign atmospheric testing and other foreign nuclear weapons operations might continue to contribute 500 to 5,000 Curies per year until there is a complete stop to atmospheric testing.

COSTS OF PLUTONIUM FACILITY IMPROVEMENTS

Expenditures for fire safety and operating conditions improvement in plutonium facilities following the fire at Rocky Flats have been greater than \$240 million. Major items are tabulated below:

1. Rocky Flats Plant - \$155 million
 - a. New Plutonium Recovery and Waste Treatment Facility
 - b. Filter Plenum Improvements, Various Manufacturing Buildings (four buildings - two tunnels)
 - c. Liquid Plutonium Waste Treatment Facility Improvements (Building 774)
 - d. Inerting Hazardous Gloveboxes, Various Manufacturing Buildings
2. Los Alamos Scientific Laboratory - \$75 million
New plutonium processing facilities and improved fire protection.
3. Pantex Plant - \$1.7 million
Mainly fire protection for production facilities.
4. Mound Laboratory - \$1.6 million
Fire protection and fire detection in gloveboxes.

RADIOACTIVE MATERIAL SPREAD BY WEAPONS FIRE

An accident occurred at McGuire Air Force Base in 1960 in which a missile containing a nuclear weapon burned. Certain aspects of the physical situation at McGuire would probably be pertinent to many weapon fire circumstances: (1) a weapon containing plutonium and explosives was involved, (2) the weapon was housed in an enclosed structure, (3) water was used to fight the fire, and the fire occurred in the presence of quantities of jet fuel. No HE detonation took place.

The explosive burned along with the jet fuel with an intense heat for nearly an hour. The plutonium became melted and much of it puddled under the burned out missile. No criticality problems occurred. The detectable plutonium spread was mostly limited to the area covered by smoke and water from fire fighting efforts. Environmental monitoring conducted on the day after the accident showed almost no contamination scatter. Zero ground contamination was found beyond 100 feet from the accident location. Some close-in contamination was found; evidently this was associated with fire-fighting water. No air contamination was detected.

While the cost due to decontamination and loss of the weapon was high, the overall magnitude of the accident was less than that which might have resulted from a one-point detonation and caused no serious off-site effects. Under these or similar circumstances, the off-site effects of this type of accident at a weapons maintenance facility would probably be much less severe than those for a one-point detonation.

WEAPON RADIOACTIVITY SPREAD BY DETONATION IMPACT

On January 17, 1966, a B-52 aircraft carrying four nuclear bombs collided with a KC-135 tanker over Palomares, Spain, during refueling operations. Four bombs broke free in the crash as the disabled aircraft plummeted near the village. One fell in a steep bank of soft earth and did not detonate. One fell into the sea and was later recovered. However, the HE components of two weapons did detonate upon impact; one in low mountains and the other on land used for agriculture. The wind velocity at the impact site was approximately 30 knots at the time of the impact and the area contaminated by plutonium from the detonated weapons was long and narrow. The area contaminated from 3.2 to 32 $\mu\text{Ci}/\text{m}^2$ was slightly over one-half mile long and one-sixteenth of a mile wide, with plutonium detectable to a distance of approximately two miles.

As mentioned above, the high explosives in two of the weapons detonated on impact. The quantity of plutonium involved in these weapons remains classified. Evidentially, much of the plutonium involved in these detonations was converted into a fine, oxide and was dispersed by the 30 knots wind which prevailed at the time of the accident. The Air Force survey showed that the 32 $\mu\text{Ci}/\text{m}^2$ level covered something over five acres with an additional 41 acres contaminated to more than about 3.2 microcuries per square meter. An additional 500 acres were found to be contaminated to greater than 0.32 $\mu\text{Ci}/\text{m}^2$.

The Air Force did careful appraisals to learn the whereabouts of all radioactive material and radioactive contamination following the accident. Most of the aircraft wreckage was found to be uncontaminated or to have negligible contamination. All of the wreckage with slight contamination was gathered and disposed of by dumping in deep, remote Atlantic waters. More contaminated wreckage was picked up, packaged, and transported to the United States for disposal.

No plutonium or tritium contamination was found in the sea water at any time following the accident nor was any tritium found at the impact sites or anywhere in the Palomares area higher than background.

Shortly after the accident, United States experts met with officials from the Spanish Government to advise and assist Spain in handling this contaminating accident, and to decide upon criteria for disposal of contaminated soil. It was agreed that soils contaminated with more than 32 $\mu\text{Ci}/\text{m}^2$ would have the top 10 centimeters (four inches) removed for disposal in the United States. Areas with less contamination (3.2 to 32 $\mu\text{Ci}/\text{m}^2$) would be plowed to a depth of about 30 centimeters.

Based on the agreement reached with Spain on the decontamination process the top four inches of soil for the area which had been contaminated at greater than $32 \mu\text{Ci}/\text{m}^2$ were removed from approximately 5.4 acres and placed in drums for shipment to a United States storage site; approximately 550 additional acres which had been contaminated to less than $32 \mu\text{Ci}/\text{m}^2$ were plowed to a depth of about 30 centimeters.

While it had been agreed to plow areas having contamination levels greater than $32 \mu\text{Ci}/\text{m}^2$, in the actual field operation areas plowed included all down to about $3.2 \mu\text{Ci}/\text{m}^2$ where feasible. Crops grown on soil on the area which had been contaminated to more than $32 \mu\text{Ci}/\text{m}^2$ were also shipped to the United States for burial, while those grown on the deep-plowed soil in the area which had been contaminated to less than that figure were burned at the sea shore on days when the wind was blowing toward the sea.

After the decontamination was finished, lung counts for radioactivity were taken on the 100 villagers most likely affected and no positive counts were found in the group. Urine samples for plutonium were negative for 70 persons and showed only insignificant amounts for the others (0.1 to 0.2 dpm in a 24 hour sample).

In 1971, six years after the accident, there appeared to be little change in the community from the time before the accident. Farming habits have changed, but mostly due to other factors such as drought, flash flooding, and economics. Followup studies show little change in exposed persons and none is expected.

RADIOACTIVE MATERIAL SPREAD BY AIRCRAFT CRASH

An aircraft carrying four unarmed nuclear weapons crashed on ice in the Arctic while attempting an emergency landing necessitated by an on-board fire. The angle of impact of the main body of the plane was about 15°. The left wing was about 60° low, and the velocity of impact was estimated at 500 knots or greater. At impact, it appears that the high-explosive components of all four weapons detonated, dispersing the plutonium in the devices into the conflagration resulting from ignition of the jet fuel, all of which was released with high forward velocity at the instant of impact and detonation of the weapons' high-explosive components.

The gross weight of the plane was about 1.9×10^5 kg, of which 1.0×10^5 kg (35,000 gallons) was JP-4 fuel. The perpendicular momentum vector would be attenuated very fast by the inertia of the ice and water and the binding and crush strength of the ice (30 to 40 inches thick), while the parallel vector would undergo relatively slow attenuation, resulting in a great forward splash of fuel and debris. This deduction was clearly indicated by an aerial photograph of the crash site, showing a long patch of black discolored ice extending away from the aircraft's impact point (See figure).

At impact, there was a large explosion and intense fire. The fire continued to burn for at least 20 minutes. Actual measurements of the cloud showed a height of approximately 2400 feet and a length of about

Plutonium contamination levels observed.

Ice core sample locations.

2200 feet. At the earliest possible time, the crash site was monitored for occurrence of nuclear criticality. Absolutely no evidence was obtained of any nuclear yield, limiting the problem to one of contamination of the broken ice in the impact area and the surface of the packed snow by plutonium, tritium, uranium, plane debris, and jet fuel.

The plutonium in the accident was converted largely to oxides by the explosion and fire. The plutonium oxide was dispersed as fine insoluble particles (from fractions of a micron to several microns in diameter). Plutonium oxide is notoriously insoluble; its solubility in sea water at 20°C is only 20 micrograms (μg) per liter. Particles of plutonium oxide were impinged into all bomb and plane surfaces struck by the high-explosive shock wave, entrained and carried forward in the splashing fuel, blown into the crushed ice at the impact point, and carried aloft in the smoke plume along with the combustion products of the burning fuel.

At the time of the crash, there was a 6-knot wind blowing from slightly east of north to slightly west of south. There was a stable inversion at approximately 2400 feet altitude and no turbulence. This stable condition persisted for several hours after the crash. Three days after the crash, a surface wind of about 25 knots (with gusts up to 45 knots) persisted for approximately 12 hours. Four days later, a similar storm occurred which persisted for about 24 hours. These winds blew from southeast to northwest, spreading some surface contamination in the downwind direction.

TECHNICAL INFORMATION

Following the accident, certain technical information was obtained, including: amount, distribution, particle size, and mode of fixation of plutonium in the crushed and refrozen ice in the impact area; the amount and distribution of plutonium contamination remaining on the surface; particle size, depth, and degree or mode of fixation of plutonium in the surface crust; tenacity of fixation of plutonium to the metal debris of the aircraft in the event large, highly-contaminated pieces went through the ice; monitoring of the bottom end of ice core samples taken around the impact area in the event large pools of contaminated jet fuel had been trapped beneath the unbroken ice; and sampling and analyzing snow surfaces along nearby shorelines.

Surface Distribution of Plutonium

It appears certain that a large fraction of the fine debris from the disintegrating aircraft and the plutonium oxide from the weapons were entrained in the large amount of JP-4 fuel released and projected forward by the impact and detonation of the weapons' high-explosive components. When the ignited fuel dropped back to the surface of the snow, it continued to burn until oxygen and temperature dropped below combustion levels. Extinction of the fire left a large quantity of unburned fuel, particularly in the blackened crust, some of which percolated down into the white snow pack beneath. The fuel that went below the blackened zone, however, carried little or no plutonium with it. From 1 to 40 percent of jet fuel on-board may remain unburned at the crash site, depending on the porosity and other characteristics of the surface

over which the fuel is spread. Packed snow would appear to constitute a porous surface. In this incident, estimation of the jet fuel in random samples of the blackened crust suggests that about 18 percent (1.9×10^4 kg, 6500 gals.) of the fuel remained unburned.

Microscopic and autoradiographic examination of solids filtered from melted samples of blackened crust showed plutonium oxide particles frequently associated with small fragments of debris of all kinds (metal, glass, and nylon fibers, plastic, rubber, flecks of paint, etc.).

The significance of these observations, of course, has to do with the ultimate distribution of the plutonium in the event large amounts of the blackened crust were allowed to break up with the ice and melt. The fact that large amounts of jet fuel are involved, in which the plutonium associated with low specific gravity debris may float, could result in contamination of the shore line during the summer season.

Plutonium Surface Deposition from Monitoring Results

Large-area monitoring was accomplished by running two radial grids, and the immediate vicinity of the crash site was monitored according to a rectangular grid (50-foot intervals). On another occasion the crash site was monitored at 50-foot intervals along the long axis of the blackened pattern and along three diagonals (readings at 25-foot intervals), one on each end and one near the middle. The counts per minute readings along each grid line and radial were plotted and fitted with smooth lines, from which interpolations were taken for contour plotting using the combined data. The contour readings were converted to milligrams (mg) of Pu^{239} per m^2 by an instrument calibration factor (determined both in the

laboratory and the field) adjusted by x-ray absorption factors determined through correlations between field readings and laboratory analysis of crust samples representative of the principal contour areas. Integral areas within the contours were determined by planimetry. Total amounts of plutonium were then estimated by integrating the surface concentration of plutonium as a function of area. The plutonium values obtained are probably good to ± 20 percent out to the edge of the blackened crust area, which corresponds roughly with the 0.9-mg/m^2 contamination contour. This information indicates 3150 ± 630 g of plutonium on the surface, of which about 99 percent was in the blackened area and would be removed by removing the full thickness of the blackened crust. Assuming removal of crust and packed snow to an average depth of 4 inches, the volume of material removed would be $6,000 \text{ m}^3$ (1.6×10^6 gallons). Assuming further that the volume ratio of packed snow to water is approximately 5.0, this would constitute 3.1×10^5 gallons of water, which would contain between 2,500 and 3,700 g of plutonium.

Plutonium Particle Size in Crusted Area

The diameters of plutonium oxide particles and the inert particles to which the plutonium oxide particles were frequently attached were measured in two blackened crust samples using a photomicrographic-autoradiographic technique.

The average plutonium oxide particle count median diameter (CMD) was about 2 microns with a standard deviation (σ_g) of approximately 1.7. The average calculated mass median diameter (MMD) was about 4 microns. The MMD of the inert particles with which the plutonium was frequently associated appeared to be 4 to 5 times larger than the plutonium particles themselves.

Lavation of Plutonium from Metal Debris

In order to learn how contaminated metal debris might behave after it reached sea water, pieces of metal varying in weight from 10 to 120 g, which had been recovered from the crash site, were subjected to the washing action of sea water, and the removed activity was determined. Total removal after 41 hours of washing varied from 23 to 89 percent. The rates of removal were different for each sample. All lavation curves, however, tended to level off with time. One might expect the rates of lavation and total amounts of plutonium removed to vary from sample to sample, depending on the velocity at which the plutonium oxide was impinged and on the hardness and nature of the surface against which it was blown.

Amount, Distribution, and Nature of Plutonium Contamination in and Under the Ice in Impact Area

To investigate the distribution of plutonium in and below the ice, a total of 182 core samples (7.5 cm in diameter) were taken. The plutonium activity in the cores was usually segregated into one or two bands associated with blackened material. Some cores showed a single band of activity, ranging from about 5 to 30 cm in thickness. This band was usually near the top, but in a few cases (about 18 percent of the samples in the crushed ice area) it was near the bottom, indicating that an occasional ice cake had been contaminated and inverted. Some cores showed both top and bottom bands of blackened activity. The bands were usually horizontal but were occasionally tipped at a significant angle with respect to the core axis.

Nature or Form of Fixation of the Plutonium

Differential analysis of the plutonium within the cores indicated that about 85 percent (range 75 to 95 percent) was associated with large fragments of material which settled out almost immediately when the cores were melted. Microscopic examination showed the plutonium associated with fragments of fiberglass, rubber, plastic, metal, paint, etc. There was no JP-4 fuel floating on the surface but only a thin film of fine carbonized material. The remainder of the plutonium was retained on the surface associated with this carbonized film. Only about 1 percent was suspended through the water phase as very fine particles. This rapid settling of most of the plutonium decreases greatly the possibility of shoreline contamination from floating debris when the ice melts.

Amount and Spatial Distribution of the Plutonium

As would be expected from the mode of dispersal, no plutonium was found in or on the bottom of the ice except in the immediate vicinity of the primary impact point where the ice was drastically broken, displaced, and refrozen.

The only significant plutonium contamination in the impact area was confined to the vicinity of the point of primary impact where the ice had been severely crushed and broken.

The observed plutonium distribution pattern in this area was highly erratic and suggested a highly segregated pattern probably related to reorientation of blocks of ice displaced at the moment of impact and detonation of the weapons' high-explosive components.

Plutonium Contamination of Land Areas

The amount of plutonium accounted for on the surface within the contours and in the broken ice at the primary point of impact is less than the total inventory of the four nuclear weapons. The remainder of the plutonium was removed with the salvaged aircraft debris and carried up in smoke plume, which had a visible height of about 2400 feet and a length of about 2200 feet. The plutonium carried up in the cloud with fine particles of debris and combustion products of the fuel. The meteorological conditions persisting at crash time and for several hours thereafter caused wide dispersion in a southerly direction. Undoubtedly, the substance of the cloud and the accompanying plutonium traveled hundreds of miles and settled out over a vast area, producing extremely low surface plutonium levels. The amount of plutonium involved in this long-range distribution pattern and associated with salvaged aircraft debris will never be known and can never be estimated perhaps to better than an order of magnitude. However, the low-level surface contamination was measured on land masses in the near vicinity of the crash site. Plutonium analyses of these samples showed contamination levels that were insignificant with respect to producing any risk either to the inhabitants or to their ecology.

Summary

Complete plutonium accountability estimates are only approximate and some of that information must necessarily remain classified. However, it can be estimated that only a small percent of the total plutonium involved in the accident escaped as an airborne aerosol for distribution away from the local area of the accident. The plutonium was distributed

over a relatively small patch of ice about 100 meters wide by 700 meters long where it attached to the surface ice and snow. The balance of the plutonium was attached to aircraft and bomb fragments scattered about the crash location.

After the accident, independent scientists concluded that the amount and distribution of plutonium in the area after the accident was such that it could not be of significance to the health of inhabitants*(as close as about 10 kilometers).

*Even so, all the contamination that was reasonably accessible on the ice was removed so that the amount of remaining contamination was substantially reduced.

ENVIRONMENTAL LEVELS OF PLUTONIUM AND THE TRANSPLUTONIUM ELEMENTS

by McDonald E. Wrenn, Ph.D.
U. S. Atomic Energy Commission
Division of Biomedical and Environmental Research

part of the AEC presentation at the
EPA Plutonium Standards Hearings
Washington, D. C., December 10-11, 1974

Introduction

My name is McDonald E. Wrenn. I am a member of the biomedical programs staff of the Division of Biomedical and Environmental Research of the United States Atomic Energy Commission. This testimony was assembled with the assistance of the staff of the AEC Health and Safety Laboratory and the Division of Operational Safety. Supplementary written testimony from Edward P. Hardy, Jr. of the AEC Health and Safety Laboratory will also be submitted for inclusion in this presentation.

Objective

This section briefly summarizes information about the locations, amounts, origins and distributions of plutonium and transplutonium elements present in the environment, available for environmental transport, and not readily amenable for retrieval.

The summary and analysis presented here is drawn largely from AEC-generated information in the public domain and selected references are given where appropriate.

Units of measure

The total amounts of plutonium and transplutonium elements will be expressed in curies (Ci) or kilocuries (kCi), the amounts found deposited upon the earth's surface will be expressed in activity per unit area -- in millicuries per square kilometer (mCi/km^2), or activity in soil in

picocuries (10^{-12} curie) per gram (pCi/g), or in the case of air in femtocuries (10^{-15} curie) per cubic meter (fCi/m³). It will become apparent that the range of activities to which we need to refer exceed 10^{20} .

Environmental plutonium can be described in two general categories namely that which is widely or globally distributed and that which is of limited distribution and attributable to a local source.

Globally distributed levels of plutonium and the transplutonic elements.

Estimates of the amount of globally distributed plutonium are shown in Table 1. There are two sources, nuclear weapons testing and space nuclear applications. Our best estimate of the global inventory is 460,000 curies, primarily of plutonium-239 and 240, which are both alpha emitters with half lives of 24,000 years and 6,600 years, respectively. Most of the activity (about 60%) is Pu-239, but the analytical techniques commonly used to measure environmental plutonium cannot distinguish between the 239 and 240, and accordingly the reported measurements which are sometimes expressed for brevity as Pu-239 activity are generally the sum of the two alpha activities. A source of Pu-238, about 17,000 curies was released when an isotopic generator used in the space nuclear program burned up in the atmosphere in 1964.

The estimates of plutonium associated with weapons testing are essentially those made by Harley in 1971⁽¹⁾ updated through 1973. The estimate of globally distributed Pu was based on the results of a soil sampling program conducted in 1970-71 by the Health and Safety Laboratory of the AEC specifically designed to evaluate Pu. Cores of soils were collected around the world at locations selected to best represent the cumulative deposition. From the amount of Pu-239, 240 measured in each sample, the amount per unit area was

calculated. The global inventory in Table 1 was constructed by summing the products of the areas in given latitude bands by the areal density of Pu-239,240 present there.

The total inventory from weapons testing was deduced from multiplying estimates of all fission yields in testing to date by the known yield of Sr-90, and the measured ratio of Pu-239/Sr-90.⁽²⁾ This ratio has not varied greatly over the years of weapons testing. Estimated in this manner the total Pu-238,239,240 injected into the atmosphere in weapons testing is approximately 440,000 curies. Other transplutonium elements are produced in nuclear weapons testing although relative to Pu only americium (Am-241) is significant in activity, comprising about 25% (110,000 curies) of the present Pu alpha activity. This estimate of the quantity of Am-241 is based on the relative activities measured in soils. A roughly equal amount of Am-241 will build in from already extant Pu-241. An estimate of the amount of curium (Cm-245,246) produced in all weapons tests of 90 curies has also been made using the approach of Thomas and Perkins.⁽³⁾ This estimate, which is roughly a thousand fold lower than the estimates for Pu and Am, was made by applying nuclide yields determined in a single test to all tests, and is accordingly only approximate.

The great bulk of nuclear weapons testing occurred prior to 1963 and the accumulated deposition from this early testing on the earth's surface is almost complete.

Figure 1 shows the yearly deposition of Pu-239 measured in New York since 1954; 1963 was the year of peak deposition. Figure 2 shows the cumulative deposition which is now increasing slowly. 90% of the current cumulative deposition had occurred by the end of 1965.

The present stratospheric inventory is about 1% of the amount which has been deposited on the earth's surface and the concentration of Pu-239 in surface air sampled at Richland, Washington, since 1962, which is shown in Figure 3 shows that the concentration of Pu-239,240 in ground level air (expressed in disintegrations per minute per thousand standard cubic meters of air) has decreased considerably since the early 1960's.⁽³⁾ The sustained concentrations of Pu-239,240 in ground level air during the last several years result from nuclear weapons testing in the atmosphere by China and France. The increased ratio of Pu-238/Pu-239 in 1966 reflects the arrival in ground level air of Pu-238 from the SNAP-9A burnup in 1964.

Accordingly the bulk of the plutonium and transplutonium elements produced in weapons testing have already deposited on the earth's surface. At any given terrestrial location, the cumulative accumulation may differ from cumulative deposition depending on the site if erosional processes are at work which may accentuate accumulation or deplete it. In addition the cumulative deposition also may vary with location at a given latitude due to variation in depositional processes such as rainfall. Accordingly, the distribution of plutonium is not uniform on the earth's surface.

Figure 4 shows how Pu-239 was distributed between the northern and southern hemispheres, as of 1970-71. About 250 kilocuries were dispersed in the northern hemisphere and 70 kilocuries in the southern hemisphere making the total global inventory 320 kilocuries. For comparison, about 16 kCi had deposited on the conterminous United States and about 3 kCi on the Australian continent which is of comparable area. The distribution of Pu-239 with latitude should be essentially the same as for Sr-90, shown in Figure 5. The highest deposition of Pu-239 is in the mid-latitudes of the northern hemisphere and it falls off toward the north pole. There is a low in the equatorial region and then a small rise in the mid-latitudes

of the southern hemisphere, again dropping toward the south pole.⁽²⁾

The total deposit of Pu-238 is about 7 percent of the Pu-239 and the SNAP-9A debris is a major contributor, particularly in the southern hemisphere. The Pu-238 from the SNAP device almost tripled the global deposit of this plutonium isotope and we know from stratospheric measurements that it has essentially all been deposited.

Figure 6 shows the Pu-239 accumulation in mCi per km² at various locations in the United States. Generally the drier areas are lower than the wet areas indicating that precipitation scavenging is an important mechanism for bringing nuclear debris to the surface. Fallout in some western areas is higher per unit of precipitation than in sites along the Pacific coast or east of the Mississippi. Evidence seems to indicate that these are regions where stratospheric debris preferentially enters the troposphere and is deposited. Most of the values for this limited sampling program across the U.S. vary by only a factor of 3 or 4, and within a particular constant rainfall region the variability in deposition appears to be much smaller.

Much information about the distribution and variability of Pu-239,240 in soils can be obtained by comparison with Sr-90 and Cs-137 which have been studied much more extensively.

For example, the vertical distribution of plutonium in soils has been determined at a few sites; whereas the vertical distribution of Sr-90 and Cs-137 have been studied at many. Globally deposited Pu-239,240 is deposited initially as small particles which do not remain indefinitely on the surface. Figure 7 shows the vertical distribution of Pu-239, Sr-90, and Cs-137 in a sandy loam soil sample from New England. Most of the deposited plutonium is in the top 5cm (2 inches) and its distribution is

similar to that of Cs-137. Both nuclides can be found in measurable concentrations down to 20 cm (8 inches) but the amount below 5 cm is only about 20 percent of the total. Strontium-90, by comparison, is less retained in the top soil and can be found to 30 cm (12 inches) so one can conclude that it is migrating at a faster rate than Pu-239 or Cs-137.

Accordingly, the concentration of plutonium inferred from measurements in surface soils will depend on the depth of the sample and the variation of Pu-239,240 concentration with depth. The vertical distribution will most likely change with time at varying rates in different types of soils and different environments. The factors which influence the rate of vertical migration are not well understood and are the subject of active investigation.

In Table 2 the widespread plutonium alpha activity is compared with the total alpha activity from naturally occurring actinides in soil, using the continental United States as a model and assuming that soils contain about 1 pCi/g of both uranium and thorium. Approximately 4.4 million curies of natural alpha emitters are present in the top 2 centimeters of soil of which approximately 1.6 million curies are alpha emitting actinides (Uranium and Thorium). Compared in this manner the concentration and amount of alpha activity from Pu-238,239,240 in surface soils (the top two centimeters) is about 1% of the natural background actinide activity.

Representative concentrations of Pu-239 in air, soil, ocean water, and human tissue^(2,3,4,5) are shown in Table 3, all expressed in pCi/g of medium, with selected measurements chosen around 1971 and 1972.

The lowest concentration is found in surface air. This reflects the transient nature of the atmospheric content. Ocean water, soil, and human

tissue all show higher concentrations partly as a result of accumulation of material over many years. Surface ocean water was typically an order of magnitude higher than air but much lower than soil due to efficient dilution to great depths. In addition there has been some removal to the marine sediments. The concentration in human lung tissue reflects continued accumulation from air by breathing and the relatively slow removal of material from lung or lymph tissue (see Bennett's testimony)⁽⁵⁾. The highest concentrations are observed in surface soils. These relatively higher concentrations result from the cumulative nature of the deposit on the surface and the very slow downward rate of migration into the soil.

There are to my knowledge no good estimates of the total amount of globally distributed plutonium in biota. However, this has been studied in local ecosystems⁽⁶⁾.

Local Accumulations

Local accumulations of plutonium may occur in association with specific facilities or activities. Table 4 lists the estimated local cumulative releases and inventories in excess of 0.1 curie which are environmentally available for a selected number of AEC and AEC contractor sites. These estimates are drawn from cumulative measurements of liquid and gaseous releases, evaluations of non-routine releases such as accidents, annual routine environmental surveillance reports, and environmental measurement programs designed to provide information on environmental inventories. Because soil measurement programs, useful for estimating the amount in the environment from soil measurements alone, are still in progress at

most facilities, estimates of environmentally available amounts may change as more complete information becomes available. In Table 5 the maximum off-site surface soil concentrations which have been observed are listed. These values are drawn from the reported environmental surveillance program results for individual operational facilities and sites.⁽⁷⁾ The results of the environmental surveillance program for all sites are reported annually in the EPA publication of Radiological Health Data. In addition, a compendium of the results of all sites and facilities programs is assembled annually as an AEC report.⁽⁸⁾

The sources fall in five general categories, which are nuclear weapons testing, nuclear weapons accidents, major AEC production and test sites, AEC contractor industrial type facilities, and purposeful release of wastes under controlled conditions to the environment. Nuclear weapons testing can be a source of local contamination both surface and sub-surface. The U.S. test areas include the Nevada test site and the Pacific testing sites, Bikini and Enewetak. An extensive radiological survey and evaluation has been completed for Enewetak⁹ where concentrations in soil range from 30 to 3000 pCi/g. This evaluation concludes that the Pu present will not seriously limit the reinhabitation of the atoll. In fact the most limiting nuclide in terms of dose is Sr-90.

The behavior of environmental Pu has been under study at the Nevada Test site for many years. Early intensive studies were conducted during and following tests, and presently intensive studies are underway to evaluate the environmental behavior of Pu which has been in situ from one to close to two decades. Under investigation are such aspects as the vertical and horizontal migration of Pu and Am in soils, the factors which affect resuspension, and the particle size of resuspended Pu.⁽¹⁰⁾ An

inventory of material available from such testing at the Nevada Test Site is underway but not complete. Most of the material on the surface of the site is inside government site boundaries. Some of it is available for transport by wind but much of it has been either weathered or treated to minimize redistribution.

Nuclear weapons accidents near Palomares, Spain (1966), and Thule, Greenland (1968), resulted in local environmental contamination. Extensive decontamination was effected at each site. From the Thule accident^(15,16) it may be estimated that about 25 Ci are in marine sediments and surface soils. The residual plutonium in Spain is being followed by the Junta de Energia Nuclear.^(17,18) These accidents are discussed briefly in the supplementary material provided.

There has been little offsite environmental contamination at major AEC development, production and testing facilities, including Hanford, Idaho, Savannah River and Oak Ridge. The amounts of materials stored in wastes at these facilities were discussed in section B as well as releases onsite to treatment and disposal systems such as seepage basins. Only small portions of these have become environmentally available. This question is treated in environmental statements being prepared for the major AEC production and testing sites.⁽⁹⁾ The environmentally available amounts (i.e., available on the surface for transport by wind) are not well known. However, it is probable that these numbers do not exceed a few tens of curies. For example, at Savannah River about 5 Ci has been released to the atmosphere and surface soils, of which approximately 2 Ci is estimated to be outside the site boundaries. However, the cumulative amounts measured

in soil at the site boundary (1.9 mCi/km^2 for 10 cores taken at 4 locations) are not distinguishable from the background levels from global fallout (1.8 mCi/km^2 at 160 km radius, 10 cores from 3 locations).⁽¹¹⁾

Another category consists of industrial type facilities such as Mound Laboratories and Rocky Flats. Pu contamination of the environment around Rocky Flats has been reasonably well documented and is described in a series of HASL reports published in recent years.^(12,13,14) Our best estimate is that several curies are distributed off-site and on the order of ten curies in surface soils on-site.

The following are examples of purposeful release of wastes to the environment. The U.K. for many years has made it a policy to dispose at sea of some of their intermediate level wastes which contain plutonium and americium. Upwards of 3000 curies of α activities have been disposed of in this manner from a pipe at Windscale into the Irish Sea; this activity is probably half Pu and half Am. Between 1951 and 1963 the U.S. disposed at sea of about 6400 Ci of drummed wastes described as plutonium contaminated. In 1971-72, the European Nuclear Energy Association disposed of a large number of 55 gallon drums of solid wastes containing an estimated 1300 curies of alpha-emitting wastes. Local releases in foreign countries other than the examples just cited have not been summarized here, although they may influence local radionuclide concentrations.

In summary, it may be concluded that the major transuranic activity in the environment is composed of plutonium and americium from weapons testing and that this material is detectable in surface soils around the world, although their presence raises the alpha background in surface soils

generally less than 1%. The Pu and Am activity per gram near the surface will decrease slowly with time. Finally, local sources of plutonium although much smaller in quantity than that from globally distributed weapons testing fallout can result in concentrations of Pu in soil exceeding the concentrations of the global level from weapons testing.

Selected References

1. J. H. Harley, "Worldwide Plutonium Fallout from Weapons Tests," p. 13-19, Proceedings of Environmental Plutonium Symposium LA-4756, Los Alamos, New Mexico, 1971.
2. Testimony of Edward P. Hardy, Jr., "Worldwide Distribution of Plutonium"
3. C. W. Thomas and R. W. Perkins, "Transuranium Elements in the Atmosphere," BNWL-1881 UC-48, Battelle Pacific Northwest Laboratories, presented at the ANS Meeting, Washington, D.C. 1974.
4. Testimony of Herbert L. Volchok, "Transuranic Elements in the Marine Environment"
5. Testimony of Burton G. Bennett, "Plutonium Fallout Pathways to Man"
6. F. W. Whicker, C. A. Little, and T. F. Winsor, "Plutonium Behavior in the Terrestrial Environs of the Rocky Flats Installation," IAEA Symposium on Environmental Surveillance Around Nuclear Installations, IAEA/SM-180/45, Warsaw, Poland, 1973.
7. Annual Reports of Environmental Surveillance at AEC Facilities and Contractors Facilities.
8. Environmental Monitoring at Major USAEC Contractor Sites, CY 1973, WASH-1259, 1973
9. NVO-140, "Eniwetok Radiological Survey," USAEC, Nevada Operations Office, Las Vegas, Nevada, October, 1973.
10. NVO-142, "The Dynamics of Plutonium in Desert Environments," Nevada Applied Ecology Group Progress Report, July 1974.
11. H. A. McClearen, "Plutonium in Soil at the Savannah River Plant," DPSPU 74-30-14, Second AEC Environmental Protection Conference, Albuquerque, New Mexico, 1974.
12. P. W. Krey and E. P. Hardy, "Plutonium in Soil around the Rocky Flats Plant," USAEC Report HASL-235, 1970.
13. P. W. Krey and B. T. Krajewski, "Plutonium Isotopic Ratios at Rocky Flats," USAEC Report HASL-249, 1972.
14. H. L. Volchok, R. Knuth, and M. Kleinman, "Plutonium in the Neighborhood of Rocky Flats, Colorado: Airborne Respirable Particles," USAEC Report HASL-246.

15. "USAF Nuclear Safety," AFRP 122-1, Jan/Feb/March 1970, No. 1, Vol. 65 (Part 2) Special Edition.
16. A. Aarkog, "Proceedings of the International Symposium on Radioecology Applied to the Protection of Man and His Environment," pp. 1213-1218, Rome, Italy, September 1971, (CONF 710973, EUR-4800, Vol. 1, 2).
17. "First Results from the Programme of Action Following the Palomares Accident," E. Iranzo, Junta de Energia Nuclear - Madrid, Spain, Symposium on Radiation Protection of the Public in a Mass Disaster, International Radiation Protection Association (IRPA), Interlaken 1968.
18. "Experience of an Accidental Contamination by Radioactive Materials, Palomares, Spain (1966)," E. Ramos and E. Iranzo, Junta de Energia Nuclear, Madrid, Spain, Second International Civil Defense Symposium on Nuclear Radiation Hazards, Monoco, 1966.

Figure 1&2

Pu²³⁹ DEPOSITION RATE NEW YORK
CUMULATIVE DEPOSIT OF Pu²³⁹ IN NEW YORK CITY

Figure 3. ^{238}Pu AND ^{239}Pu IN SURFACE AIR, 45°N

Figure 4. DISTRIBUTION OF DEPOSITED Pu^{239}
1970 - 1971

Figure 5. AVERAGE LATITUDINAL DISTRIBUTION OF DEPOSITED Sr90
FROM ANALYSES OF SOIL COLLECTED
1965-1967

Figure 6. CUMULATIVE DEPOSIT OF Pu^{239} IN mCi PER km^2

Figure 7
DEPTH DISTRIBUTIONS OF Sr^{90} , Cs^{137} , AND $\text{Pu}^{239, 240}$
IN SANDY LOAM SOIL

NORTH EASTHAM, MASS.
OCTOBER 1972

Table 1.

GLOBALLY DISTRIBUTED AMOUNTS OF ALPHA EMITTING PLUTONIUM FROM ATMOSPHERIC INJECTIONS

SOURCES	AMOUNT (CURIES)	% ACTIVITY BY ISOTOPE		
		Pu-238	Pu-239	Pu-240
ATMOSPHERIC TESTING 1945-74	110,000	3	58	39
DEPOSITED NEAR TESTING SITE	330,000	3	58	39
DEPOSITED WORLD WIDE				
SPACE NUCLEAR	17,000	100	—	—
TOTAL	457,000			
TOTAL GLOBAL EXCLUDING NEARIN TO TESTING SITE	347,000			

Table 2.

**ACTINIDES: ALPHA EMITTING CONCENTRATIONS
AND AMOUNTS IN U.S. SOIL**

NUCLIDES	AMOUNT (kCi)	REPRESENTATIVE CONCENTRATION IN SURFACE SOIL pCi/g
Pu-239, 240, 238	16	.05
Am-241	4	.01
Th-232, 230, 228 & U-238, 234 to 2 cm. depth	1600	5

Table 3

Representative Concentrations of Pu-239
in Various Media (Circa 1971)

<u>Media</u>	<u>pCi/g</u>
Air (Richland, Washington 1971)	1×10^{-7}
Ocean water (surface)	5×10^{-7}
Human tissue (lung - U.S. 1971)	3×10^{-4}
Soil (Northeast U.S., top 5 cm.)	5×10^{-2}

Table 4

Preliminary Estimates of Local Sources of Plutonium in the Environment (Quantities > 0.1 Ci)

<u>Facility</u>	<u>Quantity</u>	<u>Location</u>
NV NTS + Pacific Thule	~ 1,000 Ci ~ 25 Ci	Pu-239 to Onsite Soil Pu-239 in Marine Sediments and Surface Soils
Rocky Flats	0.04 Ci 0.1 Ci 4.0 Ci 10.0 Ci	Pu-239 to Atmosphere U + Pu-239 to Offsite Streams Pu-239 to Offsite Soil Pu-239 to Onsite Soil
Mound Laboratory	0.4 Ci 0.5 Ci 5.0 Ci 0.5 Ci	Pu-238 to Atmosphere Pu-238 to River - Routine Releases Pu-238 to Canals and Ditches Pu-238 to Soil
Savannah River	2.9 Ci 0.6 Ci 1.5 Ci 0.1 Ci	Pu-239 to Atmosphere Pu-239 to Atmosphere Pu-239 to Onsite Soil Pu-239 to Onsite Soil
LASL	1.3 Ci 0.3 Ci	Pu-239 to Atmosphere Pu-239 to Soil

Table 5
Concentrations of Plutonium in Soil

<u>Location</u>	<u>Approximate Range (picocuries/gram)</u>
Worldwide (Fallout)	0.0 to 0.2
Utah	0.2 to 0.45
Argonne National Laboratory	0.01 to 0.1
Hanford Plant	Offsite: 0.004 to 0.02
Lawrence Livermore Laboratory	0.0006 to 0.86
Los Alamos Scientific Laboratory	Offsite: 0.003 to 50
Mound Laboratory (Pu-238)	Offsite: 0.003 to 2.2
Rocky Flats	Offsite: 0.2 to 95
New Brunswick Laboratory	0.00009 to 0.75
Central New Mexico (Trinity)	0.13 to 10
Nevada Test Site	Offsite: 0.1 to 10
Bikini Atoll	1.3 to 190
Eniwetok Atoll	34 to 3,200
Palomares	0 to 1,800

WORLDWIDE DISTRIBUTION OF PLUTONIUM

by Edward P. Hardy, Jr.

Health and Safety Laboratory
U. S. Atomic Energy Commission
New York, N. Y. 10014

part of the AEC presentation at the
EPA Plutonium Standards Hearings
Washington, D. C., Dec. 10-11, 1974

Introduction

It is certainly clear that plutonium contamination of the environment on a global basis is primarily the result of atmospheric nuclear weapons testing. There are localized areas where plutonium contamination has occurred through accidents or inadvertent releases from nuclear facilities. The total amounts released are on the order of curies as compared with hundreds of thousands of curies from nuclear tests. Contamination levels in these areas are documented and studies are being carried out to follow the movement of plutonium through the eco-system pathways to man. For the future, however, it is important to know how plutonium from nuclear tests is distributed because this so-called "background" will be the baseline against which perturbations of the environment by the escalating nuclear industry must be assessed.

Nuclear Tests

Most of the plutonium now dispersed around the world was produced by nuclear tests conducted through 1962.⁽¹⁾ The above-ground tests carried out by the People's Republic of China in the Northern Hemisphere and France in the South Pacific since 1963 have contributed an additional ten percent

to the global inventory.⁽²⁾ The principal isotopes of plutonium that have been measured are Pu-239,240 and Pu-238. These are the isotopes that have been produced in greatest abundance. With time, a daughter isotope - Am-241, will become an important contributor.

SNAP-9A

Although its contribution to the plutonium radioactivity now dispersed over the earth is relatively small it is worthwhile noting that Pu-238 was released in the high stratosphere in April 1964 when a nuclear powered satellite failed to achieve orbit and disintegrated. This Pu-238 has now settled out on the earth's surface and constitutes a substantial increment to the Pu-238 fallout from nuclear tests.⁽³⁾

Sampling Programs

The Atomic Energy Commission has traced the dispersal of plutonium from atmospheric tests and the SNAP-9A satellite through its sampling programs in the stratosphere, and at ground level. I intend to review the measured levels and show how this radioactivity has changed with time. To give some perspective to the amount of plutonium produced, the Pu-239,240 radioactivity is about 2 to 3 percent of the long-lived fission product Sr-90 radioactivity. The Pu-238 radioactivity from nuclear tests is only 2 to 3 percent of the Pu-239,240 radioactivity. From now on when I refer to Pu-239 it can be inferred that I mean Pu-239 + Pu-240 because these two isotopes cannot be distinguished by conventional alpha spectrometry.

Stratosphere

Figure 1 shows the amount of Pu-239 in the stratosphere as a function of time. The unit is kilocuries and separate curves are given for the northern and southern hemispheres. After the intensive period of testing

in 1961 and 1962, the levels of Pu-239 declined with a half residence time of 10 to 11 months. Since 1967, sporadic testing by France and the People's Republic of China has maintained relative constant or only slightly diminishing amounts of Pu-239 in the stratosphere up to the present time.

Figure 2 represents the amount of Pu-238 in the stratosphere from the satellite called SNAP-9A. This was a one-time input and it is now impossible to distinguish the level from this source against the Pu-238 from nuclear tests. The SNAP device released 17 kilocuries⁽⁴⁾ while the total amount of Pu-238 that reached the stratosphere from weapons tests was about 9 kilocuries.

Surface Air

Throughout this period of weapons testing, it is generally agreed that human exposure to plutonium is primarily through inhalation. The surface air concentrations of Pu-239 in New York City as illustrated in Figure 3 show that at peak level in 1963 the concentration was 1.7 femtocuries per m³ or about 9 percent of the most conservative concentration guide for populations.⁽⁵⁾ Recent surface air concentrations attributable primarily to Chinese tests in the northern hemisphere are about 4 percent of this peak level. To assess the inhalation hazard, the fraction of the total concentration which may deposit in the nonciliated portion of the lung, must be known. Measurements of this so-called respirable fraction indicate that 80 - 85 percent of the Pu-239 aerosol is associated with particle sizes that are respirable.⁽⁶⁾ These data refer only to plutonium from weapons tests.

Deposition

Measurements of deposited plutonium have made it possible to estimate the total amount on the earth's surface and to determine how it is distributed. A properly selected soil sample can represent the accumulated deposit, and, based on a worldwide soil program carried out in 1970-71, Figure 4 shows how Pu-239 is distributed between the northern and southern hemispheres. About 250 kilocuries is dispersed in the northern hemisphere and 70 kilocuries in the southern hemisphere making the total global inventory 320 kilocuries. For comparison, about 16 kCi has deposited on the conterminous United States and about 3 kCi on the Australian continent which is of comparable area. The highest deposition of Pu-239 is in the mid-latitudes of the northern hemisphere and it falls off toward the north pole. There is a low in the equatorial region and then a small rise in the mid-latitudes of the southern hemisphere, again dropping toward the south pole.

The total deposit of Pu-238 is about 7 percent of the Pu-239 but the SNAP-9A debris is a major contributor, particularly in the southern hemisphere (see Figure 5). The Pu-238 from the SNAP device almost tripled the global deposit of this plutonium isotope but we know from stratospheric measurements that it essentially all deposited.

Figure 6 shows how much Pu-239 in mCi per km² has deposited at various places in the United States. Generally the drier areas are lower than the wet areas indicating that precipitation scavenging is an important mechanism for bringing nuclear debris to the surface. Fallout in some western areas is higher per unit of precipitation than in sites along the Pacific coast or east of the Mississippi. Evidence seems to indicate that these are regions where stratospheric debris preferentially enters the troposphere and

is deposited.⁽⁷⁾ The extremes in Pu deposition for the stratospheric source vary by only a factor of 3 or 4 and within a particular constant rainfall region, the variability in deposition is less than 15 percent.⁽⁸⁾

The total deposit of plutonium in the region of Salt Lake City is about two times higher than expected from global fallout. The excess plutonium probably came from the high explosive detonations involving unfissioned plutonium that were carried out at the Nevada test site in the late 1950's.⁽⁹⁾

The deposition rate of Pu-239 with time can be illustrated for New York City (see Figure 7). The pattern is similar to that for surface air, as expected. By integrating these rate data, the total deposit is in good agreement with the more direct measurement in soil.

Some information on the depth distribution of plutonium in soil is available which shows that most of the deposited plutonium is in the top 5 cm (2 inches) and that its distribution is similar to that of Cs-137, the most abundant long-lived fission product generated in nuclear tests. Both nuclides can be found in measurable concentrations down to 20 cm (8 inches) but the amount below 5 cm is only about 20 percent of the total. Strontium-90, by comparison, is less retained in the top soil and can be found to 30 cm (12 inches) so one can conclude that it is migrating at a faster rate than plutonium or cesium.⁽¹⁰⁾

Americium²⁴¹

It was mentioned earlier that Am-241 is a daughter of the isotope Pu-241 which builds up with time. Knowledge of its behavior in the environment is important because its chemical properties are different from plutonium. The few measurements that have been made of this nuclide in fallout show that its activity level is about 25 percent of that from

Pu-239. Despite elemental differences, Am-241 from global fallout shows a depth distribution in soil similar to Pu-239 but further measurements are needed.

Summary

Nuclear tests conducted in the atmosphere are the major sources of plutonium contamination on a worldwide basis. About 320 kilocuries of Pu-239 have deposited and about 4 kilocuries remaining in the stratosphere will reach the earth's surface. Measurements are being made of air concentrations at ground level and the deposition rate. Inhalation is the major route of human exposure and later testimony will be presented to show the resulting body burden. Contamination levels in foods have also been measured and the comparatively smaller body burden from ingestion will also be discussed.

References

1. Harley, J. H.
Worldwide Plutonium Fallout from Weapons Tests
USAEC Report, LA-4756, Los Alamos, pp. 13-17, Dec. 1971
2. Estimated from information provided by the news
media and other unclassified sources
3. Hardy, E., P. Krey and H. Volchok
Global Inventory and Distribution of Fallout Plutonium
Nature, 241, No. 5390, pp. 444-445, Feb. 16, 1973
4. Krey, P.
Atmospheric Burn-up of a Plutonium-238 Generator
Science, 158, No. 3802, pp. 769-771, Nov. 10, 1967
5. International Commission on Radiological Protection
Report of Committee II on Permissible Dose for Internal Radiation
ICRP Publication 2 (1959)
Assuming soluble plutonium with bone as the critical organ
and based on 1/30 of the occupational level for a 168 hour week
6. Volchok, H., R. Knuth and M. Kleinman
The Respirable Fraction of Sr-90, Pu-239 and Pb in Surface Air
USAEC Report HASL-278, pp. I-36 to I-40, January (1974)
7. Volchok, H. L.
High Fallout in the Western United States - An explanation
USAEC Report HASL-257, pp. I-18 to I-32, July (1972)
8. Hardy, E.
Regional Uniformity of Cumulative Radionuclide Fallout
USAEC Report HASL-288, pp. I-2 to I-9, January (1975)
9. Hardy, E., P. Krey and H. Volchok
Plutonium Fallout in Utah
USAEC Report HASL-257, pp. I-95 to I-118, July (1972)
10. Hardy, E.
Depth Distributions of Global Fallout Sr-90, Cs-137, and
Pu-239,240 in Sandy Loam Soil
USAEC Report HASL-286, pp. I-2 to I-10, October (1974)

FIGURE 1
STRATOSPHERIC INVENTORY OF Pu-239

FIGURE 2
STRATOSPHERIC INVENTORY OF SNAP-9A Pu-238

Figure 3. Pu²³⁹ IN SURFACE AIR
New York

Figure 4

DISTRIBUTION OF DEPOSITED Pu^{239}

Figure 5. DISTRIBUTION OF DEPOSITED Pu²³⁸

Map of the United States showing the distribution of the American bison. The map displays the continental United States with state boundaries and names. Circles with numerical values are placed across the map to indicate the distribution of the American bison. The values are: 1.4 (Washington), 1.9 (Montana), 2.5 (Nebraska), 2.3 (Wyoming), 2.1 (Illinois), 2.4 (Kansas), 2.2 (Arkansas), 2.4 (North Carolina), 1.0 (Florida), 0.9 (California), 0.7 (California), 0.9 (California), 1.7 (Maine), 2.3 (New York), 2.4 (New York), 2.2 (New York), 2.4 (North Carolina), 1.0 (Florida), 0.9 (Florida). A scale of miles is provided at the bottom left, ranging from 0 to 1000 miles.

Figure 7.

Pu²³⁹ DEPOSITION RATE NEW YORK

ENVIRONMENTAL PATHWAYS OF TRANSURANIC ELEMENTS

Burton G. Bennett

Health and Safety Laboratory
U. S. Atomic Energy Commission
New York, N.Y. 10014

I. GENERAL CONSIDERATIONS

Exposure of man to transuranic element contamination may occur by the inhalation or ingestion pathways. A large number of laboratory and ecological studies have been performed or are in progress to elucidate specific aspects of these pathways, such as resuspension of deposited activity, plant uptake, and physical and biological transfers in terrestrial and aquatic environments. In addition, the measurements of fallout plutonium, tracing the course of this material in air and diet to man, provide some of the most directly appropriate data regarding the environmental pathways.

It is generally recognized that for an initially airborne release, the inhalation pathway is the dominant contributor to the body burden in man. The low solubility of the transuranic elements inhibits plant uptake and absorption from the gastrointestinal tract and minimizes the importance of the ingestion pathway. The physical mechanisms of transfer of activity are thereby emphasized instead of biological accumulation mechanisms.

Dispersion of airborne radioactivity is governed by the local and regional meteorology. For contamination which originates on the ground surface, such as leakages or spills, resuspension could be an important consideration. Radioactivity in air or in soil may reach plants by deposition on plant surfaces or by root uptake. Plant uptake is governed

by the isotope and soil chemistry and by the plant physiology. Disposition of inhaled or ingested material and the biological behavior have been studied in numerous animal study programs.

There have been a few good summaries, reviews and bibliographies of pathway considerations, as well as applications of such data in environmental statements.⁽¹⁻⁹⁾ I could not possibly elaborate here on all of the accumulated data. I can, at best, remind you of some of the considerations and then present the fallout experience.

Resuspension

Resuspension is a much discussed phenomenon. Although its importance is recognized in ground contamination situations, for initially airborne releases, such as fallout radioactivity, resuspension has not markedly influenced the measured concentrations in air.⁽⁹⁾ A very large range of possible resuspension factors is often quoted - a range of 9 or 10 orders of magnitude. It is important, however, to carefully distinguish between those values which have been suggested for hazard analyses and those values that might reflect more realistic situations.

Measurements of resuspension have been conducted at contaminated areas of the Nevada Test Site. Soon after contaminating events, resuspension factors (ratios of radioactivity per unit volume of air to radioactivity per unit surface area of soil) on the order 10^{-4} to 10^{-6} m^{-1} have been obtained, decreasing subsequently with 35 to 70 day half-times.⁽¹⁰⁻¹²⁾ We are often reminded of those results, 10^{-4} to 10^{-6} . Less often, however, are we reminded of the conditions under which they were obtained. Langham and his co-workers set out in 1956 to measure resuspension at the Nevada

Test Site. Having little initial success, they found it necessary to assist the micrometeorology. They arranged to have heavy trucks roar back and forth in front of the air sampler. The first value reported by Langham⁽¹¹⁾ $7 \times 10^{-5} \text{ m}^{-1}$ is described as obtained during "extensive vehicular traffic." A second value, $7 \times 10^{-6} \text{ m}^{-1}$, was derived for "dusty rural air." Langham⁽¹¹⁾ had a feeling that 10^{-6} might be more appropriate for general usage, and Stewart⁽¹⁰⁾ also suggested that value for "quiescent conditions." In later years, subsequent to weathering and downward movement of plutonium in soil, values of 10^{-9} to 10^{-10} m^{-1} have been reported.^(13,14) The results for the desert environment are admittedly not easily generalized to more usually inhabited areas.

An important constraint in estimating a realistically applicable general resuspension factor is the fallout experience. Values should not be so large that when applied to the fallout deposition amounts, the measured air concentrations are exceeded. For fallout plutonium and as applied to natural uranium in soil as a tracer, most reasonably assumed is a resuspension factor of 10^{-9} m^{-1} to be applied to accessible activity in soil (activity in top 1 cm or less).⁽⁹⁾

It is recognized that a number of parameters are involved in describing the resuspension process, including soil conditions, moisture, wind, and vegetative cover. There may also be perturbations due to mechanical disturbances, such as digging or traffic. Activity may also be transferred to the body or clothing which may also lead to additional intake. Healy⁽⁶⁾ has discussed many of these considerations. Research efforts under way should provide increased understanding of the resuspension phenomenon and allow better prediction of short-term effects.

Plant Uptake

A number of tracer studies have been conducted in which specific influences of plant uptake of transuranic elements have been identified. These factors include chemical form, solubility, oxidation state of the radioactive element, composition and pH of the soil, and plant species. Reviews of the various studies have recently been prepared by Price⁽³⁾ and Francis⁽⁴⁾, and Durbin⁽⁵⁾ has given detailed treatment primarily of the chemical considerations involved. In Table 1, I have made a further attempt at synthesis. Additional effort will be required to achieve completeness and uniformity of reporting.

The laboratory "pot experiments" are limited by the sometimes unusual growing conditions or spiking methods and often involve seedling plants in quite short duration studies. One may worry about the extension of these results to the edible portions of mature plants grown under field conditions. On the other hand, the controlled experiments are better able to isolate the factors which apply, and the data on relative uptake of several isotopes are quite useful. The lowest values of uptake in Table 1 may reflect the short duration of exposure.⁽²⁸⁾ The highest values, field data from Palomares, may involve some direct contamination as well as root uptake.⁽²⁶⁾ The result for peeled potatoes from soil contaminated by global fallout fits in well with the laboratory study results.⁽²²⁾

Longer term experiments have not demonstrated significant uptake changes. Neubold⁽²¹⁾ indicated a slight increase in uptake for ryegrass in a 2 year experiment, but the data are inconclusive, since the concentrations barely exceeded minimum detection levels. Romney⁽³²⁾ reported a 7-fold increase

in 5 years, due possibly to increased root development. Buchholz⁽²³⁾ found equal or greater variations in uptake measurements over a 4 year period, but there was no time trend.

There appear to be no isotope differences in plant uptake, either for plutonium or curium isotopes, though the data are not extensive.

If one were to generalize on the basis of the data on hand, one might assign plant uptake of plutonium to be 10^{-4} (pCi/g fresh wt. per pCi/g dry soil), plus or minus an order of magnitude, with uptake of americium and curium about 30 times greater.

Uptake from foliar deposition is being studied. Very little translocation of plutonium following deposition has been found in initial laboratory experiments,⁽¹⁵⁾ in agreement with fallout plutonium measurements of wheat.⁽²²⁾

Ecological Studies

Ecological studies are in progress to assess the distribution of transuranic contamination in plant and animal communities.⁽¹⁶⁻¹⁸⁾ Changes with time and any reconcentration processes will be investigated. An important contributor of plutonium contamination of plants has been shown to result from wind-borne activity deposited on plant surfaces.⁽¹⁷⁾ The amounts of activity present in small mammals have not been unusual, considering the exposure pathways which exist.^(6,32)

Aquatic Studies

The only evidence for concentrations of transuranics above surrounding background comes from studies of aquatic environments. There, it is as much the low retention of suspended activity in water as it is accumulations in plant or animal organisms. Plutonium is readily removed to sediments.

The highest activity levels in biota are found in marine plants, with concentration factors as great as 1000 (pCi/kg wet wt. per pCi/l water).

The concentration factors decrease with increasing trophic level. The general behavior of plutonium in fresh water systems is comparable.⁽¹⁹⁾

I have appended a summary of transuranics in the marine environment by Herbert L. Volchok of the Health and Safety Laboratory which includes a large literature compilation.

Biological Behavior

An excellent discussion of the entry of plutonium and other actinides into animals and man and the biological behavior is presented in ICRP Publication 19.⁽²⁰⁾ Absorption from the gastrointestinal tract and entry through intact skin and wounds are considered. The ICRP Task Group Lung Model has been formulated to be used in determining the disposition of inhaled material. Fractional deposition in the three lung regions is determined by particle size. Three classes of transfer parameters, dependent primarily on chemical form of the inhaled material, are provided to determine subsequent movement from the lung to other organs in the body or to excretion. The Lung Model provides a sound basis for considering inhalation intake.

Other Aspects

There are many other aspects of environmental pathways which can and should be considered, such as mobility in soil, chelation effects on plant uptake, wash-off from plant surfaces, and animal intake pathways with transfer to meat and milk. Much data on these topics is available and more is being accumulated. Let me go on, however, to the fallout experience.

References

1. Langham, W.H., "The Biological Implications of the Transuranium Elements for Man," Health Phys. 22, 943 (1972)
2. Bair, W.J., R.C. Thompson, "Plutonium: Biomedical Research," Science, 183, 715 (1974)
3. Price, K.R., "A Review of Transuranic Elements in Soils, Plants, and Animals," J. of Environ. Quality 2, 62 (1973)
4. Francis, C.W., "Plutonium Mobility in Soil and Uptake in Plants: A Review," J. of Environ. Quality 2, 67 (1973)
5. Durbin, P.W., "Transfer of Plutonium from Soil to Plants: A Review of the Problem," Univ. of Calif. Report UCID-3689 (1974)
6. Healy, J.W., "A Proposed Interim Standard for Plutonium in Soils," Los Alamos Scientific Laboratory Report LA-5483-MS (1974)
7. Thompson, R.C., "Biology of the Transuranium Elements - A Bibliography," Battelle Pacific Northwest Lab. Report BNWL-1782 (1973)
8. Environmental Plutonium Data Base Group, "Environmental Aspects of Plutonium and Other Elements - A Selected, Annotated Bibliography," Oak Ridge National Lab. Report ORNL-ETS-73-21 and 74-21 (1974)
9. Environmental Statement, "Environmental Impact of the LMFBR," USAEC Report WASH-1535, Sec. 4G.2 (1974)
10. Stewart, K., "The Resuspension of Particulate Material from Surfaces," in Surface Contamination (B.R.Fish, ed.) Pergamon, New York pp. 63-74 (1964)
11. Langham, W.H., "Plutonium Distribution as a Problem in Environmental Science," Proceedings of Environmental Plutonium Symposium, Los Alamos Scientific Lab. Report LA-4756 (1971)
12. Anspaugh, L.R., Phelps, P.L., Kennedy, N.C., Booth, H.G., "Wind-Driven Redistribution of Surface - Deposited Radioactivity," Proceedings of Environmental Behavior of Radionuclides Released in the Nuclear Industry, IAEA, Vienna (in press)
13. Anspaugh, L.R., et al., "Resuspension of Plutonium: A Progress Report," Lawrence Livermore Laboratory Report UCRL-75484 (1974)
14. Volchok, H.L., "Resuspension of Plutonium-239 in the Vicinity of Rocky Flats," Proceedings of Environmental Plutonium Symposium, Los Alamos Scientific Lab. Report LA-4756 (1971)

15. Klepper, B.L., D.K. Craig, "Plutonium Aerosol-Foliar Interaction Program, Summary," Battelle Pacific Northwest Lab., unpublished (1974)
16. Nevada Applied Ecology Group Progress Report, "The Dynamics of Plutonium in Desert Environments," USAEC Report NVO-142 (1974)
17. Whicker, F.W., C.A. Little, T.F. Winsor, "Plutonium Behavior in the Terrestrial Environs of the Rocky Flats Installation," Proceedings of Symposium on Environmental Surveillance Around Nuclear Installations, IAEA (1974)
18. Hakonson, T.E., L.J. Johnson, "Distribution of Environmental Plutonium in the Trinity Site Ecosystem After 27 Years," Los Alamos Scientific Lab. Report LA-UR-73-1291 (1973)
19. Radiological and Environmental Research Division Annual Report, Argonne Nat. Lab. Report ANL-8060, Part III (1973)
20. International Commission on Radiological Protection, "The Metabolism of Compounds of Plutonium and Other Actinides," ICRP Publication 19 (1972)
21. Neubold, P., "Absorption of Plutonium-239 by Plants," U.K. Agricultural Research Council Report ARCRL-10 (1963)

Literature Cited - Table 1

22. Bennett, B.G., "Fallout ^{239,240}Pu in Diet," in Fallout Program Quarterly Summary Report, USAEC Report HASL-286, October (1974)
23. Buchholz, J.R., W.H. Adams, C.W. Christenson, E.B. Fowler, "Summary of a Study of the Uptake of ²³⁹Pu by Alfalfa from Soils," Los Alamos Scientific Lab. Report LADC-12897 (1971)
24. Cline, J.F., "Uptake of ²⁴¹Am and ²³⁹Pu by Plants," Battelle Pacific Northwest Lab. Report BNWL-714 (1968)
25. Cummings, S.L., L. Bankert, "The Uptake of Cerium-144, Promethium-147, and Plutonium-238 by Oak Plants from Soils," Rad. Health Data and Reports 12, 83 (1971)
26. Fowler, E.B., et al., "Soils and Plants as Indicators of the Effectiveness of a Gross Decontamination Procedure," Los Alamos Scientific Lab. Report LA-DC-9544 (1968)
27. Hale, V.Q., A. Wallace, "Effect of Chelates on Uptake of Some Heavy Metal Radionuclides from Soil by Bush Beans," Soil Science 100, 262 (1970)

28. Jacobson, L., Overstreet, R., "The Uptake by Plants of Plutonium and Some Products of Nuclear Fission Adsorbed on Soil Colloids," Soil Science 65, 129 (1948)
29. Nishita, C., E.M. Romney, K.H. Larson, "Uptake of Radioactive Fission Products by Plants," in Radioactive Fallout, Soils, Plants, Food, Man, E.B. Fowler (ed.), Elsevier, New York (1965)
30. Price, K.R., "Uptake of ²³⁷Np, ²³⁹Pu, ²⁴¹Am, and ²⁴⁴Cm from Soil by Tumbleweed and Cheatgrass," Battelle Pacific Northwest Lab. Report BNWL-1688 (1972)
31. Rediske, J.H., J.F. Cline, A.A. Selders, "The Absorption of Fission Products by Plants," Hanford Research Report HW-36734 (1955)
32. Romney, E.M., Mark, H.M., Larson, K.H., "Persistence of Plutonium in Soil, Plants and Small Mammals," Health Physics, 19, 487 (1970)
33. Thomas, W.A., D.G. Jacobs, "Curium Behavior in Plants and Soil," Soil Science 108, 305 (1969)
34. Wallace, A., "Increased Uptake of ²⁴¹Am by Plants Caused by the Chelating Agent DTPA," Health Physics, 22, 559 (1972)
35. Wilding, R.E., T.R. Garland, "Influence of Soil Plutonium Concentration on Plutonium Uptake and Distribution in Shoots and Roots of Barley," J. Agr. Food Chem. 22, 836 (1974)
36. Wilson, D.O., J.F. Cline, "Removal of Plutonium-239, Tungsten-185 and Lead-210 from Soils," Nature 209, 941 (1966)

Table 1
PLANT UPTAKE OF TRANSURANIC ELEMENTS

Concentration Factor (pCi/g fresh weight per pCi/g dry soil) - when necessary, data adjusted on basis of ash wt. 1% and dry wt. 20% of fresh wt.

	<u>Conc. Factor ($\times 10^{-4}$)</u>	<u>Reference</u>	<u>Plant Species and Method</u>
<u>^{239}Pu</u>			
	.02 - .1	Jacobson, Overstreet	barley seedlings in clay suspension for 24 hr.
	.04 - 1	Price	cheatgrass, tumbleweed - 2 mo - radioactive layer in soil.
	.001 - .3	Buchholz, Adams, Christenson, Fowler	alfalfa, barley, beans, tomatoes, lettuce - 4 yr. - nitrate, oxide and weapons contaminated soil
	.1 - .4	Cline Wilson, Cline	barley - 18d. - beans in solution
	.1 - 1	Wildung, Garland	barley shoots - 30d - factor of 2 higher in roots
	.04 - .3	Romney, Mark, Larson Nishita, Romney, Larson	clover - 5 yr. - NTS soil
	.4 - 4	Rediske, Cline, Selders	beans, tomatoes, barley, thistles - leaves - nutrient media for 20d
	3	Bennett	potatoes, peeled - fallout background soil
	1 - 300	Fowler, et al.	tomatoes, maize, beans, alfalfa - Palomares soil - veg. samples washed but may still include external contamination.
<u>^{238}Pu</u>	.02 - 2	Cummings, Bankert	oat shoots - 3 wks. in 9 soils - radioactive layer in soil
<u>^{241}Am</u>	1 - 3	Price	(see ^{239}Pu)
	3	Buchholz, et al.	"
	4 - 6	Cline	"
	20	Hale, Wallace	bush beans - leaves
	300	Wallace	soy beans - leaves and stems - higher in roots - chelates increase uptake
<u>^{244}Cm</u>	1 - 4	Price	(see ^{239}Pu)
<u>^{242}Cm</u>	0 - 4	Thomas, Jacobs	beans - % uptake agrees with Price - forage grass - no detectable uptake
<u>^{237}Np</u>	25 - 220	Price	(see ^{239}Pu)
<u>^{233}U</u>	.5 - .7	Buchholz, et al.	(see ^{239}Pu)

Summary:

^{239}Pu	10^{-5} to 10^{-3}	^{244}Cm	same as ^{241}Am	^{233}U	10^{-4}
^{238}Pu	same as ^{239}Pu	^{242}Cm	same as ^{244}Cm		
^{241}Am	10^{-4} to 10^{-2}	^{237}Np	10^{-2}		

Relative Uptake - experiments with identical plant species and soils for each isotope

<u>^{239}Pu</u>	<u>^{233}U</u>	<u>^{241}Am</u>	<u>^{244}Cm</u>	<u>^{237}Np</u>	<u>Experiment</u>
1	6	30			Buchholz (see ^{239}Pu above)
1		20-30			Cline "
1		30	40	2000	Price "

II. FALLOUT PLUTONIUM PATHWAYS TO MAN

Fallout plutonium reaches man by the inhalation and ingestion pathways.

Direct inhalation of the initially airborne weapons-produced material has been the dominant contributor to the plutonium body burden. Plutonium in air is deposited on vegetation or on soil, contaminating food by direct deposition or by root uptake. The low plutonium concentrations occurring in food and the very low transfer across the gastrointestinal tract make the importance of the ingestion pathway very minimal. Both pathways, however have been considered in detail.

Ingestion Pathway

A complete diet sampling, consisting of representative foods from 19 separate categories, was conducted in 1972 in New York by the Health and Safety Laboratory.⁽¹⁾ The plutonium concentrations in the various foods were determined and the annual intake estimated.

Because of the low plutonium concentrations in the foods, relatively large samples were required (100 g ash). The total sampling comprised 237 kg of fresh food. Table 1 lists the analytical results. The highest concentration was found in shellfish, followed by grain products and fresh fruits and vegetables. Lower concentrations were found in meats, eggs, peeled potatoes, and canned or processed foods. No activity above the minimum detection level (.01 dpm/sample) was found in milk.

The listing indicates that external contamination is a factor in the occurrence of plutonium in foods. The difference between fresh and canned foods indicates that some of the activity is lost through washing and processing. The plutonium concentration on potato peels exceeded the concentration in the peeled potatoes by a factor of 60. The activity on

the peels could be accounted for by the soil activity.

Separate analysis of clams and shrimp, which made up the shellfish sample, showed 8 times higher plutonium concentration in the clams. Much of the activity in clams, which are filter feeders, is no doubt associated with the gastrointestinal portion. The meat portion of the fresh fish sample had a concentration 10 times less than the shellfish sample.

The absence of detectable activity in milk had been observed earlier for milk sample in 1965.⁽²⁾ A tracer study has indicated very low transfer of plutonium to milk (10^{-6} of ingested dose per liter).⁽³⁾

Analysis of the identical food samples for ^{90}Sr indicated that plutonium is deficient in all food items, relative to the deposition amounts. No unusual concentrating processes have been observed. The few plutonium analyses of food sampled some years earlier indicate that the concentrations are decreasing as the deposition rate decreases.

The concentration factor for $^{239,240}\text{Pu}$ in potatoes was determined from analysis of Long Island (New York) potatoes and representative Long Island soil.⁽¹⁾ The result was 3×10^{-4} (ratio of concentration in fresh peeled potatoes to that in dry soil). Tracer studies have indicated similar results for other plant species. This determination indicates that there should be no unexpected behavior for the uptake to the edible portion of plants at low environmental levels of plutonium.

Analysis of plutonium in New York tap water (.3 fCi/l in 1973) indicates that plutonium, as does cesium, becomes largely removed to sediments.⁽¹⁾

Ingestion intake of fallout plutonium has been determined from the concentration results and food consumption estimates. These results are listed in Table 2. The annual intake during 1972 was 1.6 pCi, due 35% to

grain products, 20% each to vegetables, fruits, and meats, and less than 4% to dairy products. The annual intake in 1965 was estimated to be 2.6 pCi.⁽²⁾

Uptake of plutonium from the gastrointestinal tract is estimated to range from 3×10^{-5} to 10^{-6} .⁽⁴⁾ On this basis, the 1.6 pCi ingestion intake during 1972 would have contributed, at most, 5×10^{-5} pCi to the body burden. While inhalation intake during 1972 (.2 pCi) was less than the ingestion intake, the contribution to body burden was greater than the ingestion contribution by a factor of 1000.

Inhalation Pathway

Inhalation intake of fallout plutonium can be determined directly from the measured air concentrations. Estimates of retention in lung, transfer to blood, and organ distributions are obtained using the ICRP Task Group Lung Model. Details of the model and results of the computations for fallout plutonium were reported recently.⁽⁵⁾

Based on the measured and inferred plutonium concentrations in air in New York and a constant inhalation rate ($20 \text{ m}^3/\text{d}$), the inhalation intake is determined. The intake, listed in Table 3, reflects weapons testing activity - a decrease in 1960 during the test moratorium, a maximum in 1963 (12.2 pCi) following the 1961-62 tests, and declining intake after the 1963 Test Ban Treaty. The cumulative inhalation intake through 1973 has been 42.2 pCi.

The lung model is shown in Figure 1. The transfer rates and fractions are the amended values accepted by ICRP Committee 2 in 1971.⁽⁴⁾ The Class Y parameters have been assumed, the form of the fallout plutonium being most likely PuO_2 . Deposition in the nasopharynx (N-P) and tracheo-bronchial (T-B)

regions is cleared rapidly and almost entirely to the G.I. tract. Elimination from the pulmonary (P) region is primarily with a 500 day half-time with 80% going to the G.I. tract (40% with a 1 day half-time), 5% to blood, and 15% to the lymph system. Some permanent retention in the lymph nodes is assumed - 10% of the amount passing through the lymph system. Equal partition (45% each) of the amount transferred from blood to bone and liver is assumed. An additional 1% transfer from blood to kidney can be assumed, the remainder going to other soft tissue and excretion. The removal half-time from bone is taken to be 100 years and from liver and kidney 40 years. Transfer of plutonium from the gastrointestinal tract to blood is quite low (10^{-6}), making negligible contribution to organ burdens from activity passing from lung to the G.I. tract.

Regional deposition in the lung depends on the particle size. From measurements of size characteristics of airborne fallout,^(6,7) it has been assumed that fallout plutonium radioactivity is attached to representative 0.4 μm aerosol particles. This size results in 32% deposition in the pulmonary region. Since 60% of the deposition receives longer term retention, about 20% of the inhalation intake contributes to the initial lung burden. The lung model transfer parameters indicate that about 6% of the intake reaches blood, and with the 45-45-1 subsequent distribution to bone, liver and kidney, 2.75% of the inhalation intake can be expected to be found in bone and liver and 0.06% in kidney.

The cumulative inhalation intake of 42 pCi of $^{239,240}\text{Pu}$ could thus have resulted in about an 8 pCi body burden (42 pCi x 20%), but since the intake occurred over several years, a maximum body burden of 4 pCi was reached in 1964 (Table 3). Little activity having been removed from bone

and liver, the burdens estimated from cumulative intake ($42 \text{ pCi} \times 2.75\% \cong 1 \text{ pCi}$) agree closely with the more detailed computations⁽⁵⁾ of the current burdens. Table 4 lists the computed organ amounts for 1973. The total body burden is 2.5 pCi.

Comparison of the results of the lung model calculations of organ burdens of fallout plutonium are available with the human tissue autopsy analysis results of the Los Alamos Scientific Lab. The comparisons have shown good general agreement. The averages observed for 50 to 75 samples obtained during 1972-73 were recently reported.⁽⁸⁾ The observed-computed comparisons are .3 - .2 pCi in lung, .2 - .5 pCi in lymph nodes, 1.6 - 1.0 pCi in bone, 1.1 - .9 pCi in liver, and .2 - .02 pCi in kidney.

Figure 2 shows the measured inhalation intake of fallout plutonium and the computed organ burdens, including extrapolated values which assume no further intake beyond 1974. The continuing air concentration and tissue sampling programs will provide further checks of our prediction capability following inhalation intake.

The doses due to the computed burdens have been determined, based on uniform distributions within the organs.⁽⁵⁾ The cumulative doses through 1973 to an individual exposed throughout the entire fallout period since 1954 have been 15 mrem to lung, 8 mrem to bone and 4 mrem to liver. Longer term retention in liver and bone eventually causes the doses to these organs to exceed the dose to lung. The cumulative doses through the year 2000 to the same exposed individual are estimated to be 34 mrem to bone, 17 mrem to liver, and 16 mrem to lung. These dose commitments are less than 10% of the total dose commitments due to all other fallout radionuclides.

References

1. Bennett, B.G.
Fallout $^{239,240}\text{Pu}$ in Diet
USAEC Report HASL-286, October (1974)
2. Magno, P.J., P.E. Kauffman, B. Shleien
Plutonium in Environmental and Biological Media
Health Physics 13, 1325 (1967)
3. Sansom, B.F.
The Transfer of ^{239}Pu from the Diet of a Cow to its Milk
British Veterinary Journal 120, 158 (1964)
4. International Commission on Radiological Protection
The Metabolism of Compounds of Plutonium and other Actinides
ICRP Publication 19 (1972)
5. Bennett, B. G.
Fallout ^{239}Pu Dose to Man
USAEC Report HASL-278, January (1974)
6. Schleien, B., N.A. Gaeta, A.G. Friend
Determination of Particle Size Characteristics of old and Fresh
Airborne Fallout by Graded Filtration
Health Physics 12, 633 (1966)
7. Lockhart, L.B., R.L. Patterson, A.W. Saunders
The Size Distribution of Radioactive Atmospheric Aerosols
Journal of Geophysical Research, 70, 6033 (1965)
8. Annual Report of the Biomedical and Environmental Research Program
of the LASL Health Division, January - December 1973
Los Alamos Scientific Lab Report LA-5633-PR, May (1974)

Class Y Parameters

Particle Size: $4\mu\text{m}$

$D_3 = .10$

$D_4 = .08$

$D_5 = .32$

The ICRP Task Group Lung and Blood Clearance Model for Plutonium

Figure 1

Inhalation Intake and Burden in Man of Fallout $^{239,240}\text{Pu}$

Figure 2

Table 1

²³⁹, ²⁴⁰Pu CONCENTRATION IN FOOD
NEW YORK 1972
(pCi/kg)

SHELL FISH	0.011	FRESH FISH	0.0016
BAKERY PRODUCTS	0.0085	RICE	0.0015
WHOLE GRAIN PRODUCTS	0.0060	POTATOES	0.0013
FRESH FRUIT	0.0051	EGGS	0.0012
DRY BEANS	0.0049	MACARONI	0.0012
FRESH VEGETABLES	0.0043	CANNED VEGETABLES	0.0009
ROOT VEGETABLES	0.0035	MILK	<0.0003
POULTRY	0.0033	FRUIT JUICE	<0.0003
FLOUR	0.0028	CANNED FRUIT	<0.0002
MEAT	0.0026		

Table 2

^{239, 240}Pu DIETARY INTAKE
NEW YORK 1972
(pCi/y)

BAKERY PRODUCTS	0.37	CANNED VEGETABLES	0.019
FRESH FRUIT	0.30	EGGS	0.019
FRESH VEGETABLES	0.21	DRY BEANS	0.015
MEAT	0.20	FRESH FISH	0.013
FLOUR	0.095	SHELL FISH	0.011
WHOLE GRAIN PRODUCTS	0.066	FRUIT JUICE	<0.0072
POULTRY	0.066	RICE	0.0046
MILK	<0.064	MACARONI	0.0036
POTATOES	0.048	CANNED FRUIT	<0.0018
ROOT VEGETALBES	0.035		
		FOOD	1.5 pCi/y
		TAP WATER	0.1
		TOTAL	1.6 pCi/y

Table 3

**FALLOUT ^{239,240} Pu INHALATION INTAKE AND
COMPUTED BODY BURDENS**

<u>YEAR</u>	<u>INHALATION INTAKE (pCi)</u>	<u>BODY BURDEN (pCi)</u>
1954	1.0	0.2
55	1.3	0.4
56	1.7	0.5
57	1.7	0.7
58	2.3	1.0
59	3.3	1.3
60	0.6	1.2
61	0.9	1.1
62	4.6	1.8
63	12.2	3.5
64	6.7	3.9
65	2.4	3.6
66	0.9	3.3
67	0.4	3.0
68	0.6	2.8
69	0.4	2.7
70	0.5	2.6
71	0.4	2.6
72	0.2	2.5
73	0.1	2.5

Table 4

FALLOUT ^{239, 240}Pu IN MAN*
1973

<u>ORGAN</u>	<u>BURDEN (pCi)</u>
LUNG	0.13
LYMPH	0.44
BONE	0.98
LIVER	0.90
KIDNEY	0.02
<hr/>	
TOTAL	2.5 pCi

*CALCULATED

TRANSURANIC ELEMENTS IN THE MARINE ENVIRONMENT

Herbert L. Volchok

Health and Safety Laboratory
U. S. Atomic Energy Commission
New York, N. Y. 10014

Following is a brief summary of current information concerning transuranics in the marine environment. Most of this material was developed during a workshop convened by the Ocean Affairs Board of the National Research Council in late 1973. The workshop subject was "Assessing Potential Ocean Pollutants"; one chapter (Volchok et al., 1974) is devoted specifically to transuranics. The report of this workshop will be published in the very near future. The entire bibliography of the workshop chapter has been included here, along with the actual references cited.

In marine environments, transuranic elements have been introduced, in dispersed form, in four ways:

1. Close-in fallout from nuclear explosives testing.
2. World-wide fallout from nuclear explosives testing.
3. Atmospheric burn-up of plutonium power supplies.
4. Fluid wastes from chemical reprocessing and reactor operations.

The environmental redistributions of plutonium have been followed, to some extent, after its introduction by each of these avenues, and a small amount of information exists about americium. Unfortunately the data now at hand do not permit us to distinguish among the distributions of plutonium following its introduction in different ways. It has been assumed by Bowen and his co-workers (Wong et al., 1970a, 1970b; Bowen et al., 1971;

Noshkin and Bowen, 1973) that both plutonium and americium from world-wide fallout do pass through soluble phases in the oceans, but this has not yet been confirmed.

Pillai et al. (1964) found that plutonium concentrations in surface water, collected while fallout rates were high, were explainable by considering the precipitation exposure of the water masses sampled. More recently, samples collected while fallout rates were low showed concentrations that were explained by sedimentation of plutonium, in contrast to the conservative behavior of Sr^{90} and Cs^{137} in the same samples (Miyake and Sugimura, 1968; Miyake et al., 1970; Bowen et al., 1971; Noshkin, 1972; Noshkin and Bowen, 1973).

Profiles of Pu concentration as a function of depth in ocean water columns have been measured, primarily by Bowen and his co-workers at Woods Hole Oceanographic Institution (WHOI) in AEC-DBER and GEOSecs programs.

Noshkin and Bowen (1973) studied the relationship between the Pu^{239} in the sediment (expressed as a fraction of the estimated delivery to the latitude sampled) and the depth of the overlying water; several shallow water cores contained 100 percent of the predicted delivery, whereas their deepest core (over 5300 m) contained, in 1971, no measurable Pu^{239} . The major marine removal pathway for this transuranic is apparently biogenous sedimenting particles.

Data are insufficient to show that this behavior can be generalized for other transuranic elements, or even for Pu^{238} that was introduced by SNAP-9A. Schell and Young (1973) suggest that Am^{241} is being removed from the water column in Bikini atoll; Sugihara and Bowen (1962) and Bowen and Sugihara (1965) argued that lanthanide sedimentation was usually on inorganic

particulates, and americium is predicted to be much more lanthanide-like than is plutonium. Unpublished data (Bowen et al., unpublished manuscript) show, in one N. Atlantic water column, a steady increase with depth, of the $\text{Am}^{241}/\text{Pu}^{239}$ ratio, from 0.16 at 200 m, to 0.38 at 3200 m; this appears to be too great an increase to be explained by generation of Am^{241} by decay of Pu^{241} , and in that case, would argue strongly for control of plutonium and americium distributions by their solution chemistry in sea water.

Aarkrog's (1971) study of the accidental release of Pu^{239} and Am^{241} near Thule, Greenland, showed that of the Pu measured, upwards of 95 percent went to the sediments; 1 percent found in the water column was fine particles (as was expected of recently formed PuO_2 particles). The biological data are not yet sufficient to show the extent of remobilization of the Pu^{239} , and no examination has yet been made of the Am^{241} . The reprocessing waste discharge into the Irish Sea from Windscale also results (Preston and Mitchell, 1973; Mitchell, 1971a, 1971b) in association of most of the Pu^{239} and Am^{241} with the sediments close to the discharge area.

Pu^{239} and Am^{241} from fallout have been found in marine organisms from a variety of places in both hemispheres (Noshkin, 1973; Cherry and Shannon, 1974). Cm^{242} has been found (Livingston, personal communication, 1973) in *Fucus* from the Irish Sea, and Np^{237} (at very low levels) in a variety of samples from Eniwetok (Noshkin, personal communication, 1973).

Sources in Polykarpov (1966) indicate concentration factors of about 1000 for Pu^{239} in marine plants; Noshkin (1973) has tabulated much recent data shown in Table 1, indicating that this level is often exceeded by marine benthos and zooplankton, and usually very greatly exceeded by the Atlantic Ocean species of pelagic *Sargassum*.

Data reported by Bojanowski et al. (1974), and by Mitchell (1971a, 1971b) show that Am^{241} , from fallout or from waste, is strongly concentrated by Sargassum or rooted algae leading to in-plant ratios of $\text{Am}^{241}/\text{Pu}^{239}$ several times higher than those in the medium. Their data indicate that Am^{241} is more concentrated in Porphyra from the Irish Sea than Fucus ($\text{Am}^{241}/\text{Pu}^{239}$ as high as 1.4 in Porphyra, versus 0.18 in Fucus).

Study by Wong et al. (1972) showed that in the great Pacific kelp the maximum Pu^{239} concentrations (about 2 pCi/kg) were confined to the thin outer layers of the plant, the inner parts showing concentrations only 1/200 of that near the outside surface.

In general it appears (Noshkin, 1972) that marine invertebrates exhibit higher concentrations of fallout Pu^{239} than do fish, this is also the trend of the data on close-in fallout Pu^{239} about the Pacific test sites. The fish Pu^{239} shows bone or liver-seeking behavior, as it does in mammals; liver-seeking may also be characteristic of Pu^{239} in molluscs and crustacea, but there are few data. The tendency shown by molluscs and lobster, for high concentrations of Pu^{239} associated with shells, can be explained by the well-known tendency for shell-periphyton to accumulate trace constituents from the medium, rather than by the invertebrate equivalent of bone-seeking.

The data show little evidence for any trophic level enhancement of Pu^{239} accumulation, although in the one case specifically studied (Wong et al., 1970) starfish showed consistently higher Pu^{239} (fallout) than did the mussels on which they were feeding.

By analogy one would expect Pu^{239} or Am^{241} , once incorporated in tissues of marine organisms, to show long residence half-times; in the one case studied, Hodge et al. (1973) suggested a half-time of 3.5 years for Pu^{239} in albacore liver.

It has been noted a number of times that even Pu^{239} accumulations from fallout represent higher radiobiological doses (in rems) to some marine organisms analyzed than do either their Sr^{90} or Cs^{137} contents. This is because of the relatively greater biological effectiveness of alpha particles versus the beta or gamma radiation of Sr^{90} or Cs^{137} .

Bibliography and

LITERATURE CITED

- Aarkrog, A. 1971. Radioecological investigations of plutonium in an arctic marine environment. Health Phys. 20:31-47.
- Adams, W.H. and E.B. Fowler. 1970. ^{238}Pu incorporated in fish living in water containing $^{238}\text{PuO}_2$. LA-DC-12899. Health Division, Los Alamos Scientific Lab., Univ. of California, Los Alamos, New Mexico. 7 pp.
- Andelman, J.B. and T.C. Rozzell. 1970. Plutonium in the water environment. I. Characteristics of aqueous plutonium. In: Radionuclides in the environment (R.F. Gould, ed.), pp. 118-37. Advances in Chemistry, Series No. 93. American Chemical Society, Washington, D.C. 529 pp.
- Avargues, M. and H.P. Jammet. 1966. Étude du site marin de La Hague en relation avec le rejet d'effluents radioactifs. In: Proceedings of conference on disposal of radioactive wastes into seas, oceans and surface waters, May 16-20, 1966, Vienna, pp. 787-95. International Atomic Energy Agency, Vienna. 898 pp.
- Aviation Week and Space Technology. 1973. Forecast and inventory issue. 98:1-186.
- Bair, W.J., J.E. Ballou, J.F. Park, and C.L. Sanders. 1973. Plutonium in soft tissues with emphasis on the respiratory tract. In: Handbook of experimental pharmacology (H.C. Hodge, J.N. Stannard, and J.B. Hursh, eds.), Vol. 36, Chapter 11, pp. 503-68. Springer-Verlag, Berlin-Heidelberg-New York. 995 pp.
- Ballou, J.E., R.C. Thomason, W.J. Clarke, and J.L. Palotay. 1967. Comparative toxicity of plutonium-238 and plutonium-239 in the rat. Health Phys. 13:1087-92.
- Bentley, G.E., W.R. Daniels, G.W. Knobelock, F.O. Lawrence, and D.C. Hoffman. 1971. Separation and analysis of plutonium in soil. In: Proceedings of environmental plutonium symposium, August 1971, Los Alamos. Symposium No. LA-4756, pp. 59-61. Los Alamos Scientific Lab., Univ. of California, Los Alamos, New Mexico. 119 pp.
- Bishop, C.T., W.E. Sheehan, R.K. Gillette, and B. Robinson. 1971. Comparison of a leaching method and a fusion method for the determination of plutonium-238 in soil. In: Proceedings of environmental plutonium symposium, August, 1971, Los Alamos. Symposium No. LA-4756, pp. 63-71. Los Alamos Scientific Lab., Univ. of California, Los Alamos, New Mexico. 119 pp.

- Bojanowski, R., H.D. Livingston, D.L. Schneider, and D.R. Mann. A procedure for analysis of americium in marine environmental samples. In: Reference methods for marine radioactivity studies. II. Ruthenium, iodine, silver and the transuranic elements. International Atomic Energy Association, Vienna. In press.
- Bowen, V.T. 1973. Woods Hole Oceanographic Institution, Woods Hole, Massachusetts 02543. Personal communication, October 1973.
- Bowen, V.T. and T.T. Sugihara. 1965. Oceanographic implications of radioactive fallout distributions in the Atlantic Ocean from 20° N to 25° S, from 1957 to 1961. J. Mar. Res. 23(2):123-46.
- Bowen, V.T., K.M. Wong, and V.E. Noshkin. 1971. Plutonium-239 in and over the Atlantic Ocean. Talanta 29:1-10.
- Bundesminister für Bildung und Wissenschaft. 1972. Umweltradioaktivität und Strahlenbelastung: Jahresbericht 1971. Der Bundesminister für Bildung und Wissenschaft, Bonn. 174 pp.
- Cherry, R.D. and L.V. Shannon. The alpha-radioactivity of marine organisms. Atomic Energy Rev. In press.
- Crandall, J.L. 1971. Applications of transplutonium elements. Presented at the third international transplutonium element symposium, October 20-22, 1971, Argonne Natl. Lab., Argonne, Ill. E.I. du Pont de Nemours and Company, Aiken, South Carolina. Unpublished ms.
- Durbin, P.W. 1973. Metabolism and biologic effects of the transplutonic elements. In: Handbook of experimental pharmacology (H.C. Hodge, J.N. Stannard, and J.B. Hursh, eds.), Vol. 36, Chapter 18, pp. 739-879. Springer-Verlag, Berlin-Heidelberg-New York. 995 pp.
- Foster, R.F. 1973. Sources and inventory of radioactivity in the aquatic environment. Presented at the Health Physics Society meeting, June 17-21, 1973, Miami Beach, Florida. Battelle-Pacific Northwest Laboratories, Richland, Washington. Unpublished ms.
- Foster, R.F. and J. Soldat. 1966. Evaluation of the exposure resulting from the disposal of radioactive wastes into the Columbia River. In: Proceedings of conference on disposal of radioactive wastes into seas, oceans and surface waters, May 16-20, 1966, Vienna. International Atomic Energy Agency, Vienna. 898 pp.
- Gillette, R. 1973. Radiation spill at Hanford: The anatomy of an accident. Science 181:728-30.

- Hakonsen, T.E. and L.J. Johnson. 1973. Distribution of environmental plutonium in the Trinity Site ecosystem after 27 years. LA-UR-73-1291, Conf. 730907-14. Los Alamos Scientific Lab., Univ. of California, Los Alamos, New Mexico. 6 pp.
- Hanford Biology Symposium, Eleventh. 1972. The biological implications of the transuranic elements. Health Phys. 22:533-954.
- Hardy, E.P., P.W. Krey, and H.L. Volchok. 1973. Global inventory and distribution of fallout plutonium. Nature 241:444-45.
- Hodge, V.F., T.R. Folsom, and D.R. Young. 1973. Retention of fallout constituents in upper layers of the Pacific Ocean as estimated from studies of a tuna population. In: Radioactive contamination of the marine environment. Proceedings of a symposium on the interaction of radioactive contaminants with the constituents of the marine environment, July 10-14, 1972, Seattle, Washington, STI/PUB/313, pp. 263-76. International Atomic Energy Association, Vienna. 786 pp.
- ICRP - See International Commission on Radiological Protection.
International Commission on Radiological Protection (ICRP). 1972. The metabolism of compounds of plutonium and other actinides. ICRP Publication Series No. 19. Pergamon Press, Oxford-New York-Toronto. 59 pp.
- Janes Weapons Systems. 1972. 1971-1972 Vol. Institute for Strategic Studies, London, U.K. 586 pp.
- Jeffries, D.F., A. Preston, and A.K. Steele. 1973. Distribution of caesium-137 in British coastal waters. Mar. Pollut. Bull. 4(18): 118-22.
- Johnson, L.J. 1972. Los Alamos land areas environmental radiation survey, 1972. LA-5097-MS. Los Alamos Scientific Lab., Univ. of California, Los Alamos, New Mexico. 27 pp.
- Joseph, A., P.F. Gustafson, I.R. Russell, E.A. Schuert, H.L. Volchok, and A. Tamplin. 1971. Sources of radioactivity and their characteristics. In: Radioactivity in the marine environment (Report of the Panel on Radioactivity in the Marine Environment, Committee on Oceanography, National Research Council), pp. 6-41. National Academy of Sciences, Washington, D.C. 272 pp.
- Krey, P.W., D. Bogen, and E. French. 1962. Plutonium in man and his environment. Nature 195:263-65.
- Krey, P.W. and B.T. Krejewski. 1972. Plutonium isotopic ratios at Rocky Flats. HASL-249, April 1, 1972, pp. I-67 - I-94. Health and Safety Lab., New York Operations Office, U.S. Atomic Energy Commission, New York, New York.

- Kubose, D.A., M.G. Lai, H.A. Goya, and H.I. Cordova. 1967. Radioactivity release from radionuclide power sources. VI. Release from plutonium dioxide to seawater in the presence of ocean-bottom material. Technical Release 67-71. U.S. Naval Radiological Defense Lab., U.S. Department of the Navy, Washington, D.C. 9 pp.
- Langham, W.R. 1971. Plutonium distribution as a problem in environmental science. In: Proceedings of environmental plutonium symposium, August, 1971, Los Alamos. Symposium No. LA-4756, pp. 3-11. Los Alamos Scientific Lab., Univ. of California, Los Alamos, New Mexico. 119 pp.
- Leitenberg, M. 1970. So far, so good. Environment 12(6):26-35.
- Livingston. 1973. Woods Hole Oceanographic Institution, Woods Hole, Massachusetts 02543. Personal communication.
- Magno, P.J., P.E. Kaufman, and B. Shleien. 1967. Plutonium in environmental and biological media. Health Phys. 13:1325-30.
- Marzocchi, A., G. Cassano, F. Gera, G. Lenzi. 1971. Lo smaltimento dei rifiuti radioattivi al centro di Ricerche Nucleari della Trisaia. Minerva Fisico Nucleare 15:38-45.
- Mitchell, N.T. 1970. Radioactivity in surface and coastal waters of the British Isles 1970. Technical Rpt. FRL 6. Ministry of Agriculture, Fisheries and Food, Fisheries Radiobiology Lab. Lowestoft, U.K. 34 pp.
- Mitchell, N.T. 1971a. Radioactivity in surface and coastal waters of the British Isles 1969. Technical Rpt. FRL 7. Ministry of Agriculture, Fisheries and Food, Fisheries Radiobiology Lab., Lowestoft, U.K. 33 pp.
- Mitchell, N.T. 1971b. Radioactivity in surface and coastal waters of the British Isles 1970. Technical Rpt. FRL 8. Ministry of Agriculture, Fisheries and Food, Fisheries Radiobiology Lab., Lowestoft, U.K. 35 pp.
- Miyake, Y. and Y. Sugimara. 1968. Plutonium contents in the western north Pacific waters. Meteorol. Res. Inst. Tokyo 19(3):481-85.
- Miyake, Y., Y. Katsuragi, and Y. Sugimara. 1970. A study on plutonium fallout. J. Geophys. Res. 75:2329-30.
- Morin, M., J.C. Nenot, and J. Lafuma. 1972. Metabolic and therapeutic study following administration to rats of Pu²³⁸ nitrate: A comparison with Pu²³⁹. Health Phys. 23:475-80.
- Mott, W.E., D.W. Cole, and W.S. Holman. 1972. The U.S. Atomic Energy Commission nuclear-powered artificial heart program. Address presented in Madrid, May 15, 1972. Unpublished ms.

- Noshkin, V.E. 1972. Ecological aspects of plutonium dissemination in aquatic environments. Health Phys. 22:537-49.
- Noshkin, V.E. 1973. Lawrence Livermore Lab., Livermore, California. Personal communication, 1973.
- Noshkin, V.E. and V.T. Bowen. 1973. Concentrations and distributions of long-lived fallout radionuclides in open ocean sediments. In: Radioactive contamination of the marine environment. Proceedings of a symposium on the interaction of radioactive contaminants with the constituents of the marine environment, July 10-14, 1972, Seattle, Washington, IAEA STI/PUB/313, pp. 671-86. International Atomic Energy Agency, Vienna. 786 pp.
- Olafson, J.H. and K.H. Larson. 1963. Plutonium, its biology and environmental persistence. In: Radioecology (V. Schultz and A.W. Klement, eds.), Proceedings of the first national symposium on radioecology, September 10-15, 1961, Colorado State Univ. Rheinhold Publishing Company, New York. 746 pp.
- Olivier, J.P. 1974. Nuclear Energy Agency/Organization for Economic Cooperation and Development, 38 Boulevard Suchet, 75016 Paris. Personal communication, January 9, 1974.
- Patin, S.A., V.A. Pechkurenov, and I.A. Shekehnova. 1971. Kinetics and mechanisms of accumulation of plutonium by Misgurnus fossilis spawn. Radiobiologia 11:742-46. Translated in AEC-Tr-7306:157-59.
- Pillai, K.C. 1973. Atomic Energy Establishment, Trombay, Bombay 73, India. Personal communication, 1973.
- Pillai, K.C., R.C. Smith, and T.R. Folsom. 1964. Plutonium in the marine environment. Nature 203:568-71.
- Pillai, K.C. and P.R. Kamath. 1971. Control of radioactive pollution in India. Mar. Pollut. Bull. 2(5): 74-75.
- Polykarpov, G.G. 1966. Radioecology of aquatic organisms. Rheinhold Publishing Company, New York.
- Polzer, W.L. 1971. Solubility of plutonium in soil-water environments. Health Services Lab. Rpt., April 16. Health Services Lab., Idaho Falls, Idaho.
- Preston, A. and N.T. Mitchell. 1973. Evaluation of public radiation exposure from the controlled marine disposal of radioactive waste (with special reference to the United Kingdom). In: Radioactive contamination of the marine environment. Proceedings of a symposium on the interaction of radioactive contaminants with the constituents of the marine environment, July 10-14, 1972, Seattle, Washington, IAEA STI/PUB/313, pp. 575-93. International Atomic Energy Agency, Vienna. 786 pp.

- Price, K.R. 1973. A review of transuranic elements in soils, plants, and animals. J. Environ. Qual. 2(1):62-66.
- Raabe, O.G., G.M. Kanapilly, and H.A. Boyd. 1973. Studies of the in vitro solubility of respirable particles of Pu²³⁸ and Pu²³⁹ oxides and an accidentally released aerosol containing Pu²³⁹. Inhalation Toxicology Research Institute Annual Report 1972-1973, LF-46, UC-48, pp. 24-30. Lovelace Foundation, Albuquerque, New Mexico.
- Richmond, C.R., J.E. London, J.S. Wilson, and J. Langham. 1968. Biological response to small, discrete, highly radioactive sources. I. Observations on gastrointestinal transit, histological change, and tissue deposition in beagles fed one-half curie ²³⁸PuO₂ for six months. Health Phys. 15:487-92.
- Richmond, C.R., J. Langham, and R.S. Stone. 1970. Biological response to small, discrete, highly radioactive sources. II. Morphogenesis of microlesions in rat lungs from intravenously injected ²³⁸PuO₂ microspheres. Health Phys. 18:401-08.
- Romney, E.M., H.M. Mork, and K.H. Larson. 1970. Persistence of plutonium in soil, plants, and small mammals. Health Phys. 19:487-91.
- Seaborg, G.T. 1963. Man-made transuranium elements. Prentice-Hall, Englewood Cliffs, New Jersey. 120 pp.
- Seaborg, G.T. and J.J. Katz. 1954. Actinide elements. National Nuclear Energy Series, Vol. 14A, McGraw-Hill, New York. 870 pp.
- Schreiber, B., L. Tassi Pelati, and M.G. Mazzadri. 1972. Radioecology research in the Gulf of Taranto: Annual report 1971. In: Studies on the radioactive contamination of the sea (M. Bernhard, ed.), pp. 148-60. Euratom Rpt. EUR 4865 3, or CNEN Rpt. RT/B10(71) 7(1972). Commission of the European Communities, Luxembourg.
- Stockholm International Peace Research Institute (SIPRI). 1972. SIPRI yearbook of world armaments and disarmament - 1972. Humanities Press, New York.
- Stannard, J.N. 1973a. Biomedical aspects of plutonium. In: Handbook of experimental pharmacology (H.C. Hodge, J.N. Stannard, and J.B. Hursh, eds.), Vol. 36, Chapter 8, pp. 309-22. Springer-Verlag, Berlin-Heidelberg-New York. 995 pp.

- Stannard, J.N. 1973b. Plutonium in the environment. In: Handbook of experimental pharmacology (H.C. Hodge, J.N. Stannard, and J.B. Hursh, eds.), Vol. 36, Chapter 15, pp. 669-88. Springer-Verlag, Berlin-Heidelberg-New York. 995 pp.
- Stover, B.J. and W.S.S. Jee, eds. 1972. The radiobiology of plutonium. Univ. of Utah Press, Salt Lake City. 539 pp.
- Sugihara, T.T. and V.T. Bowen. 1962. Radioactive rare earths from fallout for the study of particle movement in the sea. In: Radioisotopes in the physical sciences and industry, pp. 57-65. International Atomic Energy Agency, Vienna.
- U.S. Atomic Energy Commission. 1971. Siting of fuel reprocessing and waste management facilities. ORNL-4451. Oak Ridge Natl. Lab., U.S. Atomic Energy Commission, Oak Ridge, Tennessee. 415 pp.
- U.S. Atomic Energy Commission. 1972. Nuclear power: 1973-2000. WASH-1139 (72). Forecasting Branch, Office of Planning and Analysis, U.S. Atomic Energy Commission, Washington, D.C. 43 pp.
- Vaughan, J., B. Bleaney, and D.M. Taylor. 1973. Distribution, excretion, and effects of plutonium as a bone-seeker. In: Handbook of experimental pharmacology (H.C. Hodge, J.N. Stannard, and J.B. Hursh, eds.), Vol. 36, Chapter 10, pp. 349-502. Springer-Verlag; Berlin-Heidelberg-New York. 995 pp.
- Volchok, H.L. 1973. U.S. Atomic Energy Commission, Health and Safety Lab., 376 Hudson Street, New York, N.Y. 10004. Personal communication, October 1973.
- Volchok, H.L., R.H. Knuth, and M.T. Kleinman. 1972. The respirable fraction of plutonium at Rocky Flats. Health Phys. 23:395.
- Volchok, H.L., V.T. Bowen, R. Dyer, W. Forster, J. Herring, J. Perkowski, T. Rice and J. Stannard. Chapter 3 "Transuranic Elements" In: "Assessing Potential Ocean Pollutants" Ocean Affairs Board, National Academy of Sciences. In press.
- Washington Post, July 16, 1973, p. 10.
- Webb, G.A.M. and F. Morley. 1973. A model for the evaluation of the deep ocean disposal of radioactive waste. NRPB-R14, June 1973. National Radiological Protection Board, Harwell, Didcot, Berkshire, U.K.
- Wick, O.J., ed. 1967. Plutonium handbook: A guide to the technology, Vols. 1 and 2. Gordon and Breach, Science Publishers, New York. 966 pp.

- Wong, K.M., V.E. Noshkin, and V.T. Bowen. 1970a. Radiochemical procedures of Wong (now used as WHOI) for the analysis of strontium, antimony, rare earths, cesium, and plutonium in sea water samples. In: Reference methods for marine radioactivity studies, Tech. Series No. 118, pp. 119-26. International Atomic Energy Agency, Vienna. 284 pp.
- Wong, K.M., V.E. Noshkin, L. Surprenant, and V.T. Bowen. 1970b. Plutonium-239 in some organisms and sediments. In: Fallout program: Quarterly summary report, March 1, 1970-June 1, 1970 (E.P. Hardy, Jr., ed.), HASL-227, pp. I-25 - I-33. New York Operations Office, Health and Safety Lab., U.S. Atomic Energy Commission, New York, New York. 190 pp. + Appendix.
- Wong, K.M., V.F. Hodge, and T.R. Folsom. 1972. Plutonium and polonium inside giant brown algae. Nature 237:460-62.
- Zlobin, V.S. 1966. Accumulation of uranium and plutonium by marine algae. Radiobiologiya 6(4) [Translation on pp. 219-27; Russian, pp. 613-17].
- Zlobin, V.S. 1971. Active phase of assimilation of Pu-239 by the marine algae Ascophyllum nodosum. Proc. Sci. Res. Planning Inst. Sea Fisheries Oceanogr. 29:169-75. Translated in AEC-Tr-7418:207-17.
- Zlobin, V.S. and Mokanu, O.V. 1970. Mechanisms of accumulation of Pu²³⁹ and Po²¹⁰ by the brown algae Ascophyllum nodosum and marine phytoplankton. Radiobiologiya 10:584-89. Translated in AEC-Tr-7205:160-69.
- Zlobin, V.S. and M.F. Perlyuk. 1971. Photosynthesis and the mechanism of the action of cyanide on cell respiration and Pu-239 accumulation by marine algae. Proc. Sci. Res. Planning Inst. Sea Fisheries and Oceanogr. 29:159-68. Translated in AEC-Tr-7418:195-206.

TABLE 1. PLUTONIUM IN MARINE ORGANISMS

- 14 -

Organisms	Collection Date	Location	Tissue and Number Samples	Mean Pu 239 pCi/kg wet	Pu238 Pu239	Range of Conc. Factors
<u>Plants</u>						
Algae(attached)	1964	California Coast	Whole (4)	0.47	--	660-1570
	1970-71	Cape Cod, Mass.	" (6)	0.53	--	100-1600
	1971	California Coast	" (19)	0.64	--	260-3500
Sargassum(attached)	1971	California Coast	" (2)	0.28	--	325-450
Sargassum(pelagic)	1965-70	Atlantic Ocean	" (6)	20.7	--	3000-100,000
<u>Animals</u>						
Plankton(mixed)	1950	California Coast	" (1)	0.023	--	--
"	1961	Atlantic Ocean	" (1)	2.03	--	2300
"	1964	California Coast	" (1)	1.08	--	2600
Sponge	1970	Cape Cod	" (1)	1.80	0.07	2100
Annelid Worm	1970	" "	" (1)	3.50	0.06	4100
Starfish	1970	" "	" (1)	0.87	0.09	1020
Gastropods	1970	" "	Body (2)	0.35	0.05	140-660
"	1970	" "	Shell (2)	0.42	0.09	300-690
Bivalves	1964	California Coast	Body (2)	0.22	--	230-290
"	1968	Danish Coast	? (1)	7	--	8200
"	1970	Cape Cod	Body (5)	0.31	0.07	300-520
"	"	"	Shell (2)	0.47	0.08	490-600
Salpa	1958	Atlantic Ocean	Whole (3)	1.40	--	900-2400
Crustacea	1968	Danish Coast	" (5)	1.5	--	1060-4500
Fish	1964	California Coast	" (1)	0.001	--	3
"	1969	Cape Cod	Muscle(1)	0.005	--	1-5
"	"	"	Liver (1)	0.03	--	--
"	"	"	Bone (1)	0.6	--	--
"	1970	Cape Cod	Muscle(3)	0.003	--	1-2
"	"	"	Liver (3)	0.05	--	14-60
"	"	"	Gut (3)	0.68	--	40-1100
"	"	"	Bone (3)	0.11	--	50-600
"	1970	Cape Hatteras	Muscle(3)	0.003	--	4
"	"	"	Liver (3)	0.15	--	175
"	"	"	Bone (3)	0.02	--	21
"	1971	Cape Hatteras	Muscle(2)	0.006	--	10-15
"			Bone (2)	0.03	--	30-50

Blank Page

The Biological Effects of Transuranium
Elements in Experimental Animals

by W. J. Bair, Ph.D.
Director, Life Sciences Program
Battelle - Pacific Northwest Laboratories
Richland, Washington 99352

part of the AEC presentation at the
EPA Plutonium Standards Hearings
Washington, D. C., December 10-11, 1974

INTRODUCTION

The toxicity of plutonium has been of concern since milligram quantities were first produced in the Oak Ridge reactor starting in late 1943. In 1944 milligram quantities were allocated for biomedical studies. In 1947 and 1948 the first biological experiments were completed with americium and curium. Since then, biological research has been in progress at several laboratories in the United States and abroad. Most of the research effort has been directed towards the compounds of the most common isotope of plutonium, ^{239}Pu . Within the past 10 years as the concept of plutonium recycle and the fast breeder program developed, recognition of the potentially increasing abundance of the transplutonium elements has led to expanded biomedical research on neptunium, plutonium, americium, curium, californium, berkelium, and einsteinium.

The purpose of these animal experiments is to enable us to predict the health consequences of transuranium elements in man. These studies have considered two different kinds of contamination events. One of

these, occupational exposures to transuranics released into the work environment, involves relatively small numbers of people. Such exposures can be simulated reasonably well in the laboratory with the expectation of observing unmistakable effects in statistically significant numbers of animals. The second kind of contaminating event of concern is the exposure of large populations to very low levels of the transuranium elements. Such low level exposures cannot be directly simulated in the laboratory because exceedingly large numbers of experimental animals are required to yield meaningful results. Therefore, an understanding of the mechanisms of the biological action of low levels of transuranics is required to predict what cannot be measured in the laboratory.

An overriding concern of all the animal experimentation is the confidence with which the results can be extrapolated to man. While sophisticated experiments can be performed with rodents, dogs, cats, swine, and primates, questions regarding the applicability of the results to man will be settled only as data from human exposure cases become available.

The major health effects of the transuranic elements are due to their emission of alpha radiation and the nature of the effects depends upon which tissues are irradiated. This is determined by the disposition of these radioactive elements in the body which in turn is determined by their chemical and physical properties and their route of entry into the body. Thus, in this presentation, I will review in a general way the

absorption of the transuranic elements into, and the distribution within the body following ingestion, deposition on skin, and inhalation. Then I will summarize the biological effects which have been observed in experimental animals, in particular the late effects resulting from relatively low level radiation exposures. The emphasis will be on plutonium because it is the transuranic element we know most about.

Much of the information included in this presentation has appeared in several recent reviews (Hodge, Stannard, Hursh, 1973; Bair and Thompson, 1974; Bair, 1974; Thompson, 1974; Bair, Richmond, and Wachholz, 1974; Dolphin et al., 1974; Buldakov et al., 1969; Sanders et al., 1970; Healy, 1974; Thompson, 1974; Park, 1974; Stover, 1974).

DISPOSITION OF TRANSURANICS IN THE BODY

Transuranium elements released to the environment may reach man through three pathways. Transuranics which become incorporated into foodstuffs may be ingested and absorbed from the gastrointestinal tract, while those dispersed in air may be either deposited on the skin and absorbed, or inhaled and deposited in the respiratory tract. Occupational exposures may include entry through a wound.

ABSORPTION FROM THE GASTROINTESTINAL TRACT

Experiments confirm that most transuranic element compounds are not readily absorbed from the gastrointestinal tract. Table 1 gives values for the gastrointestinal tract absorption of uranium and several transuranic elements in newborn rats and adults. In rats, neptunium nitrate was most readily absorbed, nearly 1 percent. The least absorbed was PuO_2 , 0.0001

percent. Gastrointestinal tract absorption was one or two orders of magnitude greater in the newborn rat than in the adult. Americium, curium, berkelium, and einsteinium show at least a 10-fold greater absorption than plutonium.

ABSORPTION THROUGH INTACT SKIN

Although percutaneous absorption of all available transuranic elements has not been thoroughly studied, results from experiments with plutonium indicate that absorption through intact skin is a relatively minor route of entry into the body, Table 2. The highest value, 2 percent, was obtained in rat skin exposed to $\text{Pu}(\text{NO}_3)_4$ in 10 N HNO_3 for 5 days. All other experiments gave values of less than 1 percent. About 0.05 percent of einsteinium, the only other transuranium for which data are available, was absorbed over a period of 7 days through rat skin. Results from animal experiments and human contamination incidents indicate the intact skin to be an effective barrier to the entry of plutonium and einsteinium, and probably the other transuranium elements.

RETENTION AND TRANSLOCATION FROM LUNG

Airborne transuranic particles are similar to most other particles when they are inhaled in that deposition in the respiratory tract is primarily dependent upon the physical properties of the particles and the respiratory characteristics of the individual inhaling the particles.

Clearance From Lung

Animal experiments and limited human data provide a range of values for the retention half-times of several plutonium compounds. These are

summarized in Figure 1 for ^{238}Pu and ^{239}Pu . The retention half-times for organic complexes of plutonium, plutonium nitrate and fluoride range from less than 100 days to about 300 days in rats and dogs. The retention half-times for PuO_2 are substantially longer, ranging from 200 to 500 days in rats, 300 to 1000 days in dogs, and 250 to 300 days in human beings. The wide range of values for dogs is largely due to extensive experimentation with a variety of plutonium oxides with different particle size characteristics. The relatively low retention values for human beings, compared with dogs, suggests either that man clears plutonium particles from his lungs more rapidly than do dogs or that the materials inhaled in the human accident cases were more soluble than plutonium dioxide. Studies with $^{238}\text{PuO}_2$ in dogs indicate a much shorter lung retention time than is observed for $^{239}\text{PuO}_2$. This appears to be due to instability of $^{238}\text{PuO}_2$ particles, possibly caused by radiolysis in tissue fluids.

Figure 2 illustrates the effect of the physical properties of the inhaled particles on retention of plutonium in lung. Retention half-times are given in days for several plutonium oxides. Each bar represents data from one dog. Plutonium oxide prepared by calcining the oxalate at 1000°C was retained with a half-time of 650 to 950 days compared with 300 to 400 days for an oxalate calcined at 350° . Oxides prepared from metal powder at temperatures of 123° to 450° were retained in lung longer than the low fired oxalate. Particle size is also important.

The small particle size high-fired oxide was retained with a half-time of 400-500 days, compared with half-times up to 900 days for the larger particle size high-fired oxide. Retention of the small particle size $^{238}\text{PuO}_2$ was less than for the comparable $^{239}\text{PuO}_2$.

Pulmonary retentions of inhaled transuranic compounds have been compared in rats and dogs. In rats, both ^{241}Am and ^{242}Cm nitrates were cleared much more rapidly than ^{238}Pu and ^{239}Pu nitrates, Figure 3. Autoradiograms from this study indicated ^{241}Am and ^{242}Cm to be dispersed much more throughout the lung than ^{238}Pu and ^{239}Pu . In another experiment the rate of clearance of intratracheally instilled einsteinium chloride was found to be much like that reported for ^{242}Cm nitrate.

The retention rates of several inhaled transuranics in beagle dogs, Figure 4, compare favorably with the results from rat experiments. Plutonium oxide, nitrate and fluoride were retained in the lung much longer than curium and americium oxides.

Spatial Distribution of Transuranics Within Lung

From the moment transuranic elements are deposited in the respiratory tract, biological and physical forces are at work to cause their removal. Thus, it is difficult to visualize plutonium and the other transuranics remaining static throughout their residence time in lung. It is not possible to document the course of individual particles and aggregates of particles in lung. However, the temporal and spatial characteristics of radioactive particles within tissues can be inferred from autoradiographs of tissue sections prepared from animals exposed to radioactive aerosols.

The first observation is that radionuclides are nonuniformly deposited throughout lung. Further, the radionuclides may deposit unequally among the lung lobes or among portions of lung lobes. Studies of inhaled plutonium nitrate in both rats and dogs show that immediately following the inhalation exposure, plutonium is present in both particulate and nonparticulate forms, as evidenced by the presence of alpha stars and single tracks in autoradiographs, Figure 5. Einstinium nitrate, Figure 6, and einstinium hydroxide also show particulate and nonparticulate forms a few days after deposition in the lung. Autoradiographs prepared from dogs exposed to inhaled $^{239}\text{PuO}_2$ show an initial relatively diffuse distribution of particulate plutonium throughout the entire lung.

A fraction of the amount of transuranics deposited in the lung may be dissolved and absorbed into the blood. The remaining transuranic particles and aggregates may be engulfed by macrophages. This has been demonstrated in studies with plutonium. Phagocytized plutonium dioxide particles are rapidly localized in the cells, Figure 7.

The alveolar macrophage appears to be capable of transporting transuranic particles and aggregates from the alveoli to the ciliated epithelium lining the bronchioles. These phagocytic cells containing particles and aggregates can then be removed from the lung in the mucous blanket which is propelled up the respiratory passage by ciliary action. Transuranium elements removed from the lung by this route are swallowed and excreted in the feces.

Both soluble and insoluble transuranics not immediately cleared from the lung tend to become further aggregated. This mobility and aggregation of transuranics may have large effects on the temporal and spatial distribution of the alpha radiation dose. A few days after inhalation of plutonium nitrate and other relatively soluble compounds single tracks in autoradiographs decrease and after several weeks nearly all of the radioactive material appears to be aggregated. Figure 8 shows aggregation of ^{238}Pu in rat lung 100 days after inhalation of $\text{Pu}(\text{NO}_3)_4$. Curium tends to aggregate less than plutonium (LaFuma et al., in press).

Particles of transuranium elements are transported via lymphatic vessels in the lung and collected in the thoracic lymph nodes. Autoradiographs of lung tissues taken from dogs several weeks and months after inhalation of PuO_2 show alpha stars concentrated in subpleural areas, apparently in lymphatic vessels, Figure 9. Autoradiographs also suggest that radioactive particles become immobilized in scar tissue in subpleural areas. Figure 10 is an example of a $^{238}\text{PuO}_2$ particle located in scar tissue of a dog 5 years after exposure. Areas around the scar tissue appeared to be normal.

Radioactive particles transported to lymph nodes eventually appear sequestered in "hot spots" of scar tissue and do not appear to be mobile. The residence time for plutonium in lymph nodes appears to be very long.

There is ample evidence that transuranic particles deposited in lung are subjected to biological and physical forces. This argues against either particles or aggregates of transuranium elements remaining

static indefinitely, except for that which becomes immobilized in scar tissue. To the contrary, while the rates may be low, movement of transuranic particles within lung tissue by several mechanisms certainly occurs as the lung attempts to expel the radioactive particles and other foreign material which may have been inhaled. The migration of deposited radioactive particles in lung partially compensates for the nonuniformity of the radiation exposure from the particles.

Translocation From Lung to Other Tissues

The relative distribution among body tissues of the transuranics translocated from lung by the circulating blood is essentially the same for all transuranic compounds, but may differ quantitatively depending upon the chemical and physical state of the inhaled material.

In beagle dogs within several months after inhalation of relatively soluble plutonium nitrate, the fraction remaining in lung decreased to 40 percent or less of the amount deposited in the lower respiratory tract, Figure 11. Translocation of plutonium from lung resulted in bone accumulating about 30 percent, and liver about 10 percent. A small percentage was found in spleen, lymph nodes, and other soft tissues and the remainder was excreted in urine and feces.

When plutonium dioxide is inhaled, the lymphatic system accounts for a large fraction of plutonium cleared from lung, Figure 12. Data from a 11-year study with beagle dogs shows that after 5 years lung and thoracic lymph nodes each contained 30 percent of the plutonium

initially deposited in the lower respiratory tract. After 11 years the amount in the lung had decreased to about 10 percent and the thoracic lymph nodes had accumulated 40 percent. Translocation of plutonium from lung resulted in levels in liver of about 10 percent, in bone of about 5 percent, and in the abdominal lymph nodes of about 7 percent.

The average radiation doses to these tissues bear the same relationship as the plutonium concentration in the tissues, Table 3. The average concentration of plutonium was highest in the thoracic lymph nodes and next highest in the abdominal lymph nodes. Average concentrations in lung and liver were over 1000 times less than those in the thoracic lymph nodes. The concentrations in spleen and bone were about one-fifth to one-tenth those in lung. Thus, the lymph nodes received a much higher average radiation exposure than other tissues in the body.

Data were presented showing that $^{238}\text{PuO}_2$ may be cleared from lung more rapidly than $^{239}\text{PuO}_2$. It has also been found that translocation of ^{238}Pu from lung to other tissues in the body may be greater than for ^{239}Pu , Figure 13. Distributions of plutonium in tissues of beagle dogs 5 years after inhalation of $^{238}\text{PuO}_2$ and $^{239}\text{PuO}_2$ are compared. After 5 years only 10 percent of the body burden of the ^{238}Pu was in lung compared with 46 percent for ^{239}Pu . Accumulation in thoracic lymph nodes was three times greater for ^{239}Pu than for ^{238}Pu ; however, the bone burden of ^{238}Pu was 12 times that of ^{239}Pu . This illustrates that the behavior of $^{238}\text{PuO}_2$ in the body may differ significantly from that of $^{239}\text{PuO}_2$.

All of the transuranics tend to translocate from lung to bone and liver and, to a lesser extent, to spleen and kidney. However, the rate at which translocation from lung to these other tissues occurs differs considerably among the transuranics, depending mostly on the in vivo solubility of the compound inhaled. For example, $^{244}\text{CmCl}_3$, $^{244}\text{CmO}_{1.73}$, and $^{241}\text{AmO}_2$ are readily translocated to the bone, similar to plutonium nitrates, Figure 14. Translocation of plutonium occurs very slowly after inhalation of plutonium fluoride and plutonium dioxide.

In dogs exposed to aerosols of oxides of transuranium elements, ^{244}Cm was almost equally distributed among lung, liver, bone, and muscle after one month, Table 4. The translocation of ^{241}Am was predominately to the liver while translocation of a relatively soluble form of ^{238}Pu , the hydrated oxide, was mainly to bone. More stable ^{238}Pu oxides and $^{239}\text{PuO}_2$ showed little translocation from lung to other tissues, but showed greater accumulation in the thoracic lymph nodes than occurred after inhalation of $^{241}\text{AmO}_2$ and $^{244}\text{CmO}_x$.

The tissue distribution of transuranium elements absorbed through the skin, from wounds or from the gastrointestinal tract is similar to that observed after translocation from the respiratory tract. The rate of translocation may vary, however, because deposition in a wound or in the lung provides a reservoir for continuous absorption into the blood stream. Absorption from skin or the gastrointestinal tract will be of short duration, until the skin has been decontaminated or the GI tract cleared, except in cases of continuous exposure.

The quantitative differences in translocation of different transuranic elements and compounds from the lung and other deposition sites can lead to significant differences in biological effects which may occur.

DEPOSITION IN GONADS

Because of the concern for possible genetic effects of radionuclides deposited in the body, measurements have been made of the amounts of radionuclide which accumulate in the gonads of experimental animals. This is summarized for plutonium in Table 5. About 0.05 percent of the plutonium in the circulating blood may deposit in testes and only about 0.01 percent in ovaries. In the testes about half of the plutonium appears to be associated with germinal tissue. Few data are available on the deposition of the other transuranics in gonadal tissue but preliminary results suggest that the fractional uptake will be about the same as that for plutonium.

CROSS PLACENTAL TRANSFER

The transfer of transuranics and other radionuclides across the placenta have been studied in rats, Table 6. Neptunium, plutonium, and einsteinium show significantly less transfer to the fetus as well as to the placenta and the placental membranes than uranium, cesium, and cerium. Preliminary results from studies of ^{241}Am and ^{244}Cm suggest less transfer than has been observed for plutonium. These results show that the placenta is an effective barrier to transuranium elements, and

that extremely high levels of contamination would have to occur in the pregnant female before appreciable quantities of transuranium elements would occur in the fetus.

BIOLOGICAL EFFECTS

It has been shown that the distribution of the transuranium elements among the tissues in the body varies depending upon the route of entry, the chemical compound and the radioisotope. The biological effects which may occur will depend upon the radiation exposure and the relative radiation sensitivity of each tissue into which the radionuclide is deposited. These are primarily blood, bone, liver, lung, and the lymphatic system. The biological effects of greatest interest are those that might occur at low doses. Animal experiments have identified neoplasia as the most sensitive response to the long-term effects of transuranic elements deposited in the body.

BLOOD

Transuranic elements are cleared from the circulating blood within a few days after absorption from the site of deposition such as the gastrointestinal tract and lung. Therefore, the major effects seen in blood cells will be due to irradiation of the hematopoietic tissue in which the radionuclides are deposited or to irradiation of blood circulating through tissues containing deposits of the radionuclide.

Most of the hematologic effects observed after deposition of the transuranics in the body occur at relatively high doses, doses above those which have resulted in cancer, Table 7. A variety of hematologic

effects have been reported in all animal species studied. Summarized in this table are only data from dog and pig experiments. The erythrocyte levels are only reduced at the highest doses. Elements of white blood cells show transient reductions following intravenous injections of all of the transuranics studied similar to those seen after exposure to external radiation. Although the transuranics deposit in bone and lymph nodes, leukemia has not been a common finding in animal experiments. It has been reported in less than a dozen rats after intravenous injection of plutonium and americium, in a few rats after intratracheal injection of einsteinium, and in one dog (which also had osteosarcoma) after inhalation of ^{238}Pu .

The most consistent hematologic response seen after injection and inhalation of the transuranics is lymphopenia. This is well documented after inhalation of ^{238}Pu , ^{239}Pu , and ^{241}Am . In current experiments with dogs this is the most sensitive indication of a biological effect, occurring at dose levels of inhaled $^{239}\text{PuO}_2$ which have not yet shown cancer, Figure 15. The possible health consequences of a plutonium-reduced level of circulating lymphocytes are not yet known. One cannot rule out the possibility of a relationship between the reduction of circulating lymphocytes, lymph node pathology, decreased immunological competence, and the pathogenesis of transuranic-induced cancer.

BONE

Osteogenic sarcomas appear to be the most sensitive effect following the skeletal deposition of plutonium, americium, curium, and einsteinium in mice, rats, and dogs. In most of these experiments the solutions of

the transuranics were injected intravenously. However, osteogenic sarcomas have occurred after pulmonary deposition of ^{244}Cm and ^{253}Es in rats and ^{238}Pu in rats and dogs. In these experiments the ^{244}Cm , ^{253}Es , and ^{238}Pu were largely translocated to the skeleton.

The most informative experiment on the effects of transuranics in bone is a beagle dog study with plutonium at the University of Utah. This experiment began in 1952 to compare the long-term effects of intravenously injected plutonium and radium. The objective was to determine the toxicity ratio between plutonium and radium in dogs, so that the radium toxicity data available from human exposures can be used to infer the toxic response to plutonium in man. The results from the plutonium animals in this experiment are shown in Table 8. There were 9 to 13 animals per exposure group, injected at levels differing by about a factor of three, and ranging from 2.9 $\mu\text{Ci/kg}$ to 0.016 $\mu\text{Ci/kg}$. There was a substantial incidence of osteosarcoma, 31 percent at the lowest level. When it became evident that effects were occurring in the lowest exposure groups, additional groups were added to the experiment at levels down to 0.0006 $\mu\text{Ci/kg}$, which is equivalent to the occupationally permissible body burden for man.

A number of long-term studies in rodents have also pointed to osteosarcoma as the most sensitive indicator of plutonium in the skeleton, Figure 16. The incidence of osteosarcoma is plotted against cumulative radiation dose to bone. For each datum point 95 percent binomial confidence limits are shown. Each point represents a group of animals at a given dose level; each type of symbol represents a given experiment. The

open circles represent the Utah dogs and are quite clearly a separate population from the rodents. From these kinds of data, Mays and Lloyd, assuming a time-independent linear dose-response relationship, have calculated an increased incidence per rad of 0.38 percent for beagles, 0.10 percent for mice, and 0.06 percent for rats. Although few data are available, it appears that doses greater than those which caused bone cancer in dogs do not cause bone cancer in miniature swine.

Of more interest than absolute incidence figures and their uncertain extrapolation to man, is the finding in the Utah studies that plutonium-239 is 5 to 10 times more toxic than radium-226, on the basis of the same total energy delivered to bone. This difference is attributed to the more hazardous localization of plutonium on bone surfaces, whereas radium is distributed more uniformly throughout bone. The cells from which bone tumors originate are located near bone surfaces.

The inhalation of $^{239}\text{PuO}_2$ has not led to the development of osteogenic sarcomas in experimental animals. However, osteogenic sarcomas have occurred in dogs after inhalation of $^{238}\text{PuO}_2$, Table 9. These occurred as a result of the translocation of ^{238}Pu from the lung to bone, which received a higher radiation dose than the lungs. Lung cancer was a secondary finding in one of these dogs. Another dog had leukemia and fibrosarcoma as well as osteosarcoma.

LIVER

Liver is comparable to bone and lung in terms of transuranic content and radiation exposure. However, the liver appears to be less sensitive than bone and lung to the carcinogenic action of alpha radiation.

Malignant liver tumors were the primary cause of death in two of 96 plutonium dogs at risk in the Utah experiment. Small, benign bile duct tumors were incidental findings at autopsy in eight other dogs, but these were also seen in controls at a somewhat lower incidence. The liver tumors had a longer latent period than bone tumors which might explain the lower incidence. Because of this, the possibility remains that liver tumors might predominate at lower doses.

LUNG

Inhalation of relatively soluble plutonium compounds such as organic complexes, plutonium nitrate, and $^{238}\text{PuO}_2$ has resulted in primary lung cancer in rodents, rabbits, and dogs in addition to the bone cancer already mentioned. Lung cancer has also occurred in rats given $^{253}\text{EsCl}_3$ by intratracheal instillation and in rats after inhalation of $^{244}\text{CmO}_x$, $^{244}\text{Cm}(\text{NO}_3)_3$, $^{238}\text{Pu}(\text{NO}_3)_4$, $^{241}\text{Am}(\text{NO}_3)_3$, or $^{241}\text{AmO}_2$. Lung cancer has also been observed in beagle dogs, baboons, and rodents after inhalation of $^{239}\text{PuO}_2$.

The experimental data on plutonium-induced lung cancer are shown in Figure 17, with tumor incidence in percent plotted against the lung dose in rads. These are rat, mouse, and rabbit data except for the results from one dog study represented by the square symbols. The dogs seem to be more susceptible than the rodents; however, there are no dog data below about 1000 rads. Some of the low incidence data are of uncertain significance because there was a low incidence of lung tumors

in the controls of some of the experiments. For both lung and bone tumors, the lowest dose at which a clearly significant affect has been observed is about 30 rads.

In rats given $^{253}\text{EsCl}_3$ by intratracheal instillation, the malignant lung tumor incidence was 4 percent for a lung dose of 38 rads and 12.5 percent for a lung dose of 1900 rads. The incidence of osteogenic sarcomas in these animals was 42 percent at 230 rads and zero at 5 rads. In comparison with soluble forms of the 24,000 year half-life ^{239}Pu , the 20.5 day half-life ^{253}Es was less efficient in producing lung cancer and more efficient in causing bone cancer, providing some basis for speculating on the dose rate effects of alpha radiation (Ballou et al., in press).

Other experiments are in progress in the United States and abroad to determine the carcinogenic response of inhaled curium and americium relative to ^{238}Pu and ^{239}Pu . Squamous cell carcinomas and bronchiolo-alveolar carcinomas are being observed in these experiments.

Preliminary results from several laboratories indicate that the transuranics readily induce lung cancer in rats. However, these same laboratories find that Syrian hamsters tend to be much less sensitive to the carcinogenic action of the alpha-emitting transuranium elements than rats. This is in contrast to results being obtained in other laboratories with ^{210}Po introduced by intratracheal injection which show a high incidence of lung cancer.

The available data on plutonium-induced lung cancer has been analyzed to describe mathematically the relationship between cancer incidence and radiation dose. Although other models may have equal merit, a logarithmic probit curve was selected based on its long usage in toxicology. In Figure 18, arithmetic representations of the fitted function are shown by heavy lines for probit curves and linear regressions. Dotted lines show a limited extrapolation of the fitted functions.

This analysis indicates the current status of information on plutonium-induced lung cancer. Studies now in progress are adding substantially to our understanding of the dose-effect response.

LYMPH NODES

It was shown that plutonium accumulates in lymph nodes following deposition of plutonium in the respiratory tract. Months or years after the contaminating event, lymph nodes may attain concentrations of plutonium many times the average concentrations remaining at the site of deposition and consequently the accumulated radiation dose to some lymph nodes may be greater than to any other tissue. The fact that the biomedical significance of plutonium concentrations in lymph nodes is unknown is a major concern in establishing permissible limits for plutonium.

Although dogs have been studied for 11 years after inhaling $^{239}\text{PuO}_2$ and rodents have been studied in life span experiments after inhalation of a variety of plutonium and other transuranic compounds, primary cancer of lymphatic tissue has not occurred. In dogs which had primary

cancer, metastasis to mediastinal lymph nodes and lymphatics occurred, but only one dog had a possible malignant lymphoma and this was confined to the mesenteric and mandibular lymph nodes. Therefore, it can be concluded that the lymph nodes are not especially susceptible to the carcinogenic action of alpha radiation from plutonium.

RELATIVE SENSITIVITY OF DIFFERENT SEGMENTS OF THE POPULATION

An important question relevant to the establishment of exposure standards for radiation or other potentially hazardous agents is whether all segments of the population are equally sensitive. Research on this question relative to the transuranics has not been extensive; however, some information has been gained from experiments with rats.

Effect of Age on Osteogenic Sarcoma Response to Plutonium

Studies of rats given plutonium intravenously indicate that the newborn and weanling may be slightly more sensitive than the adult to plutonium-induced osteosarcoma, Figure 19. The decreased incidences at the higher doses for the weanlings and newborns are probably due to shortened life spans for these groups which did not allow the full cancer potential to be expressed. However, the high dose adult group also showed a significantly shortened life span and still had a high incidence of bone cancer (50 percent).

A similar study with ^{253}Es did not indicate a difference in the incidence of osteogenic sarcoma between the adult and weanling groups at bone doses of 100, 500, and > 2000 rads (D. D. Mahlum, personal communication).

The Effect of Iron Deficiency

Iron deficiency is common in the human being particularly in pregnant women, pre-menopausal women, and in young children. Because of the metabolic relationship between iron and the transuranics in the blood, iron deficiency could have a bearing on the distribution and subsequent biological effects of the transuranics. In an experiment with mice rendered iron deficient it was found that deposition of intravenously injected plutonium in bone of the iron deficient mice was much greater than in the controls. This suggests that persons deficient in iron could have an increased susceptibility to plutonium-induced osteosarcoma. Iron deficiency did not affect the deposition of plutonium in gonads (H. A. Ragan, personal communication).

THE "HOT PARTICLE" ISSUE

It was recognized in the early 1940's that plutonium particles deposited in the lung would irradiate cells in the immediate vicinity of the particle rather than the entire lung. This gave rise to the concern that plutonium particles might be exceptionally efficient in causing lung cancer. Research during the past nearly 30 years has provided no evidence for an enhanced effect of the localized radiation dose from plutonium. Although not conclusive, experimental results to date suggest that plutonium particles might be less hazardous than the same amount of plutonium distributed throughout the lung because many fewer cells would be exposed to alpha radiation. However, experiments to resolve this question are technically difficult because inhaled plutonium does not

distribute throughout the lung but tends to aggregate. Also, such experiments require life time studies with large numbers of animals. A few experiments which bear on the issue have been completed with several transuranic compounds, others are still in progress.

Rats exposed to relatively non-particulate soluble ^{238}Pu which was highly dispersed during its relatively short residence time in the lung developed a higher incidence of lung cancer than has been observed for more particulate ^{238}Pu and ^{239}Pu sources (Sanders, 1973). The implication is that the dispersed ^{238}Pu exposed more cells to the carcinogenic action of the alpha radiation than particulate plutonium.

In France the influence of non-uniform distribution of alpha radiation in lung is being studied in about 700 rats exposed to $^{238}\text{Pu}(\text{NO}_3)_4$, $^{239}\text{Pu}(\text{NO}_3)_4$, $^{239}\text{PuO}_2$, $^{241}\text{Am}(\text{NO}_3)_3$, $^{241}\text{AmO}_2$, or $^{244}\text{Cm}(\text{NO}_3)_3$. Of these transuranics, ^{244}Cm was the most uniformly distributed throughout the lung and was most effective in reducing survival time followed in descending order by ^{238}Pu , ^{241}Am , $^{239}\text{Pu}(\text{NO}_3)_4$, and $^{239}\text{PuO}_2$ which was the most heterogeneously distributed (LaFuma et al., in press).

The relatively uniform distribution of ^{244}Cm in lung is illustrated by the autoradiogram in Figure 20. This can be compared with the more aggregated $^{239}\text{Pu}(\text{NO}_3)_4$ in Figure 21. Although the experiment is not yet completed, nearly 200 squamous cell carcinomas and bronchiolo-alveolar carcinomas have been observed. The authors report that the results to date indicate that for lung cancer induction, ^{244}Cm , the most widely

dispersed alpha emitter, is more effective than the more particulate transuranics. Again, the implication is that more cells are exposed to the alpha radiation from the dispersed ^{244}Cm than from the other less-dispersed transuranics.

Another experiment was designed specifically to address the "hot particle" issue at the Los Alamos Scientific Laboratory (Richmond and Voelz, 1972, Anderson et al., 1974). It is impossible to distribute alpha emitting sources uniformly throughout the lung by inhalation exposure due to the tendency for such material to be mobilized and aggregated by clearance processes. Therefore, 10 μm zirconium oxide microspheres containing PuO_2 at specific activities corresponding to respirable particles were given intravenously to hamsters. The microspheres were observed to be firmly fixed in the lung vasculature and were highly dispersed throughout the lung. By varying the quantity of Pu in the microspheres the microdistribution of the radiation dose could be controlled. A total of over 2000 hamsters have been given 2000 to 1,600,000 microspheres ranging in activity from 0.07 to 59 pCi per microsphere. Total lung burdens range from 0.14 nCi to 354 nCi. This study is still in progress. However, nearly 1200 animals have lived their full life span or have been sacrificed. These animals were given a total of about 5.7×10^6 microspheres, each containing in excess of 0.07 pCi. Three malignant tumors were observed. This suggests a tumor risk of about 10^{-7} per particle. The preliminary results from this study do not suggest that particulate sources are more hazardous than equivalent less-particulate sources.

In this experiment and in other experiments with plutonium particles, the lack of significant histopathology in areas adjacent to the particles is a common finding. An example is shown in Figure 22 which is an autoradiograph of a section from a lung of a rat given $^{239}\text{PuO}_2$ by intraperitoneal injection (Sanders, in press). From 0.2 percent to 2 percent of the plutonium was phagocytized and transported to the lung where the particles lodged in the vasculature similar to the microsphere experiment with hamsters. The lungs of these rats surprisingly showed little evidence of pulmonary pathology that could be attributed to the plutonium. The radiation doses to the lungs of the several groups of a total of 151 rats were 10, 20, 40, 170, and 600 rads. Only one lung tumor occurred; this was observed after 823 days in one rat of 36 which had a lung dose of 10 rads.

Another example of Pu particles residing in lung which shows no evidence of histopathology is illustrated in Figure 23. This is an autoradiograph of a histologic section from a lung of a dog about 2 years after inhalation of $^{239}\text{PuO}_2$ showing several particles in areas of normal lung. Other sections of the lung from this dog showed evidence of plutonium-induced changes. However, normal appearing areas such as this containing plutonium particles, are not uncommon in animal experiments.

The pathogenesis of plutonium induced neoplasia is not fully understood. However, observations made during the past 10 years, suggest that phagocytosis of the plutonium particle is one of the steps leading to necrosis and a connective tissue response such as fibrosis and/or an epithelial response such as hyperplasia, metaplasia, and eventually neoplasia.

In agreement with the observations from studies of plutonium and other transuranics deposited in lung are the results from a recent study of the incidence of chromosome aberrations in the liver as a function of the size of the $^{239}\text{PuO}_2$ particle administered. For the same total quantity of plutonium administered, the more uniform dose was more effective in producing chromosome aberrations than the more localized doses (Brooks et al., 1974).

The "hot particle" issue continues to be the subject of controversy and will not be settled to everyone's satisfaction until more of these difficult and expensive experiments are completed. However, the results of relevant experiments at laboratories in the United States, France, and the United Kingdom have led the scientists conducting the experiments to believe that particulate alpha emitting transuranics in the lung do not represent a higher risk of lung cancer than the equivalent quantity of relatively non-particulate transuranium elements distributed throughout the lung (LaFuma et al., in press; Dolphin et al., 1974).

COUNTERMEASURES FOR INHALED PLUTONIUM

An important consideration in evaluating the potential health effects of plutonium is the availability of effective countermeasures for plutonium exposures. Since no acceptable therapy exists for radiation exposures, the only really effective countermeasure for inhaled transuranics is their removal from the body. Inhaled insoluble plutonium is not effectively mobilized by a wide variety of agents which have been tested.

The most effective method for removing plutonium from the lung is lavage with isotonic saline. In rats, dogs, and baboons about 50 percent of the lung burden can be removed by lavage.

A chelating agent, Diethylenetriaminepentaacetic acid (DTPA), has had wide application for treating persons occupationally exposed to plutonium. It reduces the liver burden and, to a lesser extent, the bone burden causing an increased urinary excretion of plutonium. DTPA has also been found to be effective in reducing the systemic burden of Am, Cm, and Es. However, it is not effective in removing insoluble transuranics from lung or lymph nodes.

Therefore, it must be recognized that truly effective counter-measures for transuranic contamination have not yet been found, and that for all practical purposes transuranics deposited in the body will remain there until removed by natural processes, most of which are very slow. The development of therapeutic procedures for removing transuranics from the body are high priority research projects in several laboratories.

SUMMARY AND CONCLUSIONS

STATUS OF CURRENT RESEARCH

Research relevant to the problem of low level exposures to transuranium elements has increased significantly during the past 10 years. Life span animal studies of the biological effects of the transuranium elements have greatly expanded. The current status of our knowledge of plutonium is shown in Figure 23. The dose levels at which major biological effects have been observed in experimental animals are shown relative to the

maximum permissible lung burden of 0.016 μCi for occupational exposures. Lung cancer has been observed at dose levels equivalent to about 100 times the maximum permissible lung burden. Current experiments are directed towards determining whether health effects will occur at lower levels. However, because of the cost in terms of time and money of such experiments, the most productive research may be that which is directed towards understanding the mechanisms by which alpha emitters induce cancer. Some of these studies are in progress and more are anticipated.

RESEARCH IN PROGRESS

Research is in progress in several laboratories in the United States and abroad to examine the late effects of low levels of transuranic elements. Three dog studies are in progress in the United States. At the University of Utah the late effects of transuranics are being studied after intravenous administration, Table 10. This study is primarily directed at effects in bone and liver. At the Lovelace Foundation dogs are being exposed to monodisperse aerosols of ^{238}Pu and ^{239}Pu , Table 11. A total of 360 dogs are being given single exposures to plutonium aerosols with particle sizes ranging from 0.75 to 3 μm aerodynamic diameter. Initial lung burdens range from 0.1 to 5.6 μCi .

At Battelle-Northwest 221 dogs have been exposed to polydispersed aerosols of $^{238}\text{PuO}_2$ and $^{239}\text{PuO}_2$, Table 12. Initial lung burdens ranged from 0.002 μCi to 5 μCi .

In addition to these major dog experiments, all of the available transuranics are being studied in thousands of rats and hamsters following inhalation, ingestion and intravenous injection. These long term animal

experiments are being supplemented by increasing levels of research to develop a better understanding of how alpha radiation from the transuranium elements causes cancer and other possible health effects.

No significant surprises are expected from this intensified research effort. However, the results will help us sharpen our predictions of the health consequences of the expected increased utilization and availability of the transuranium elements.

References

- Anderson, E. C., Holland, L. M., Prine, J. R., and Richmond, C. R., "Lung Irradiation with Static Plutonium Microspheres," In: Experimental Lung Cancer, Carcinogenesis and Bioassays, Springer-Verlag, Heidelberg, in press, CONF-740648-1, 1974.
- Bair, W. J., and Thompson, R. C., "Plutonium: Biomedical Research," Science Vol. 183: 715-722, 1974.
- Bair, W. J., "Toxicology of Plutonium," In: Advances in Radiation Biology, Vol. 4 (J. T. Lett, H. Adler, and M. Zelle, Eds.), Academic Press, 1974.
- Bair, W. J., Richmond, C. R., and Wachholz, B. W., "A Radiobiological Assessment of the Spatial Distribution of Radiation Dose from Inhaled Plutonium," USAEC, WASH-1320, Sept., 1974.
- Bair, W. J., "Consideration of Reactor Accident Exposure Guides for Plutonium," In: Conference on Fast Reactor Safety (CONF-740401). NTIS, Springfield, VA, 1974.
- Bair, W. J., "The Effects on Populations of Exposure to Low Levels of Ionizing Radiation." Health Phys. 26, 588-591. (book review).
- Ballou, J. E., Dagle, G. E., and Morrow, W. G., "The Long-Term Effects of Intratracheally Instilled $^{253}\text{EsCl}_3$ in Rats," Radiation Res. (in press).
- Buldakov, L. A., Lyubchansky, E. R., Moskelev, Y. I., and Nifatov, A. P., (1969), In "Problemy Toksikologii Plutoniya", Atom Publications, Moscow (translated as "Problems of Plutonium Toxicology," U.S. Atomic Energy Commission LF-tr-41, 1970).
- Brooks, A. L., Retherford, J. C., and McClellan, R. O., "Effect of $^{239}\text{PuO}_2$ Particle Number and Size on the Frequency and Distribution of Chromosome Aberrations in the Liver of the Chinese Hamster," Radiation Res. 59, No. 3, Sept., 1974.
- Dolphin, G. W., Smith, H., Popplewell, D. S., Stather, J. W., Adams, N., Spoor, N. L., Brightwell, J., and Bulman, R. A., "Radiological Problems in the Protection of Persons Exposed to Plutonium," Document NRPB-R29, National Radiological Protection Board, Harwell, England, 1974.
- Healy, J. W., (1974), "Contamination Limits for Real and Personal Property," Los Alamos Scientific Laboratory Report No. LA-5482-PR.

Hodges, H. C., Stannard, J. N., and Hursh, J. B. - Editors, Uranium, Plutonium, Transplutonic Elements Handbook of Experimental Pharmacology XXXVI, Springer-Verlag, New York, 1973.

Hungate, F. P., Ballou, J. E., Mahlum, D. D., Kashima, M., Smith, V. H., Sanders, C. L., Baxter, D. W., Sikov, M. R., and Thompson, R. C., "Preliminary Data on ^{253}Es and ^{249}Bk Metabolism in Rats," pp. 653-656. In: Biological Implications of the Transuranium Elements, R. C. Thompson and W. J. Bair (eds.), Health Physics, Vol. 22, June 1972.

LaFuma, J., Nenot, J. C., Norin, D., Masse, R., Detivier, H., Nobile, D., and Skupinski, W., "Respiratory Carcinogenesis in Rats after Inhalation of Radioactive Aerosols of Actinides and Lanthanides in Various Physico-chemical Forms," Symposium on Experimental Respiratory Carcinogenesis and Bioassays, Seattle, WA, 1974 (in press).

Mahlum, D. D., Sikov, M. R., "Influence of Age on the Late Effects of Plutonium-239 in Rats," Radiat. Res. 59, 171, (abstract), 1974.

Mahlum, D. D., and Sikov, M. R., "Distribution and Toxicity of Monomeric and Polymeric ^{239}Pu in Immature and Adult Rats," Radiat. Res. 60, 75-88, 1974.

McClanahan, B. J., Ragan, H. A., and Mahlum, D. D., "Plutonium Metabolism in Newborn and Weanling Pigs," Radiat. Res. 59, 171, (abstract), 1974.

McClellan, R. O., Boyd, H. A., Gallegos, A. F., and Thomas, R. G., "Retention and Distribution of ^{244}Cm Following Inhalation of $^{244}\text{CmCl}_3$ and $^{244}\text{CmO}_1.73$ by Beagle Dogs," Health Physics, Vol. 22, pp. 877-885, 1972.

McClellan, R. O., "Progress in Studies with Transuranic Elements' at the Lovelace Foundation, Health Physics, Vol. 22, pp. 815-822, 1972.

Park, J. F., Catt, D. L., Craig, D. K., Olson, R. J., and Smith, V. H., Solubility Changes of ^{238}Pu Oxide in Water Suspension and Effect on Biological Behavior after Inhalation by Beagle Dogs, pp. 719-724, In: Third International Congress of the International Radiation Protection Association, Vol. 1, W. S. Snyder (ed.), (CONF-730907). NTIS, Springfield, VA, 1974.

Park, J. F., Lund, J. E., Ragan, H. A., Hackett, P. L., and Frazier, M. E., "Inhaled $^{238}\text{PuO}_2$ -Induced Bone Tumors in Dogs," presented at Bone Tumor Conference, Dusseldorf Germany, 1974.

Park, J. F., "Late Effects of Inhaled Plutonium in Dogs," Fifth International Congress of Radiation Research, Seattle, WA, July 14-20, 1974 (in press).

Richmond, C. R., Langan, J., and Stone, R. S., "Biological Response to Small Discrete Highly Radioactive Sources," Health Physics Vol. 18, pp. 401-408, 1970.

Richmond, C. R. and Voelz, G. L., eds., Annual Report of the Biological and Medical Research Group (H-4) of the LASL Health Division, January through December 1971, Los Alamos Scientific Laboratory report LA-4923-PR, pp. 18-34, April, 1972.

Richmond, C. R., and Voelz, G. L., eds., Annual Report of the Biological and Medical Research Group (H-4) of the LASL Health Division, January through December 1972, Los Alamos Scientific Laboratory report LA-5227-PR, pp. 1-11, March, 1973.

Richmond, C. R., and Sullivan, E. M., eds., Annual Report of the Biomedical and Environmental Research Program of the LASL Health Division, January through December 1973, Los Alamos Scientific Laboratory report LA-5633-PR, pp. 1-9, May, 1974.

Richmond, C. R., and Thomas, R. L., "Plutonium and other Actinide Elements in Gonadal Tissue of Man and Animals." Accepted for publication in Health Physics (in press).

Sanders, C. L., and Adey, R. R., "The Ultrastructure of Mononuclear Phagocytes following Intraperitoneal Administration of $^{239}\text{PuO}_2$ Particles," J. of the Reticuloendothelial Soc. Vol. 6, No. 1, Feb., 1969.

Sanders, C. L., "Carcinogenicity of Inhaled Plutonium-238 in the Rat," Radiat. Res. 56, 540-553, 1973.

Sanders, C. L., Thompson, R. C., and Bair, W. J. (1970), "Lung Cancer: Dose Response Studies with Radionuclides," In: Inhalation Carcinogenesis, (M. G. Hanna, Jr., P. Nettesheim, and J. R. Gilbert, eds.), CONF-691001: 285, AEC Symposium Series No. 18, U.S. Atomic Energy Commission, Office of Information Services, Springfield, VA.

Sanders, C. L., "Effects of PuO_2 Particles Deposited in the Lung Following Intraperitoneal Injections," Health Physics (in press).

Sanders, C. L., and Dagle, G. E., "Studies of Pulmonary Carcinogenesis in Rodents Following Inhalation of Transuranic Compounds," Symposium on Experimental Respiratory Carcinogenesis and Bioassays, Seattle, WA, 1974, (in press).

Sikov, M. R., and Mahlum, D. D., "Influence of Age on ^{239}Pu Distribution, Dosimetry, and Effects." Trans. Amer. Nuclear Soc. 18, 389, (abstract) 1974.

Smith, V. H., "The Biological Disposition of $\text{Es}(\text{NO}_3)_3$ in Rats after Intravenous, Intramuscular and Subcutaneous Administration," pp. 725-730, In: Third International Congress of the International Radiation Protection Association, Vol. 1, W. S. Snyder (ed.), (CONF-730907). NTIS, Springfield, VA, 1974.

Stover, B. J., "Dose-Response Relationships for Beagles Injected with $^{239}\text{Pu}(\text{IV})$ or $^{241}\text{Am}(\text{III})$," Fifth International Congress of Radiation Research, Seattle, WA, July 14-20, 1974 (in press).

Thomas, R. G., McClellan, R. O., Thomas, Randi L., Chiffelle, T. L., Hobbs, C. H., Jones, R. K., Mauderly, J. L., and Pickrell, J. A., Health Physics Vol. 22, pp. 863-871, 1972.

Thompson, R. C., "Effects of Plutonium in Animals," Presentation before the ad hoc Committee of the USAEC Advisory Committee on Reactor Safeguards, Los Alamos, NM, January 4, 1974.

Thompson, R. C., Summary and Speculative Interpretation Relative to Exposure Limits, Fifth International Congress of Radiation Research, Seattle, WA, July 14-20, 1974 (in press).

Acknowledgement: The author is indebted to the staff of the Biology Department, Battelle, Pacific Northwest Laboratories; to Dr. E. C. Anderson, Health Division, Los Alamos Scientific Laboratory; and to Dr. C. H. Hobbs, Inhalation Toxicology Research Institute, Lovelace Foundation for results from current research and for future research plans at their respective laboratories.

RETENTION OF PLUTONIUM IN PULMONARY REGION OF LUNG

Figure 1

The attached tables were inadvertently
omitted from WASH-1359. They should follow page 203.

1

TABLE 1

Gastrointestinal Tract Absorption of
Transuranics in Rats

(percent of Administered Dose)

<u>Transuranic</u>	<u>Compound</u>	<u>Newborn</u>	<u>Adult</u>
^{233}U	Nitrate	7	0.2
^{237}Np	Nitrate	1	0.9
^{238}Pu	Nitrate	2	0.03
^{239}Pu	Nitrate	0.3	0.003
	Chloride	-	0.007
	Oxide	-	0.0001
^{241}Am	Nitrate	9	0.07
	Chloride	-	0.03
	Oxide	0.5	0.01
^{244}Cm	Nitrate	6	0.2
	Chloride	-	0.05
	Oxide (aged in H_2O)	2	0.1
	Oxide (fresh)	0.3	0.03
^{249}Bk	Chloride	-	0.01
^{252}Cf	Nitrate	4	0.1
^{253}Es	Nitrate	4	0.03
	Chloride	-	0.06

Information in this Table was developed from published reports and from results of current research at PNL by M. F. Sullivan.

TABLE 2

Absorption of Pu and Es through Intact Skin

<u>^{239}Pu or ^{253}Es Compound</u>	<u>Animal Species</u>	<u>Duration of Exposure</u>	<u>Percent Absorbed</u>
$\text{Pu}(\text{NO}_3)_4$ in 10 N HNO_3	rat	1 hour	0.05
Pu-tributyl phosphate in CCl_4	rat	15 min	0.04
$\text{Pu}(\text{NO}_3)_4$ in 0.1 N HNO_3	rat	5 days	0.1-0.3
$\text{Pu}(\text{NO}_3)_4$ in 10 N HNO_3	rat	5 days	1-2
$\text{Pu}(\text{NO}_3)_4$	rabbit	14 days	0.15
Pu citrate	swine	10 days	0.25
Pu in 9% HCl + EDTA	man	-	0.01
$\text{Pu}(\text{NO}_3)_4$ in 0.4 N HNO_3	man	1 hour	0.002
$\text{Es}(\text{NO}_3)_3$ in 0.01 N HNO_3	rat	7 days	0.05

TABLE 3

Relative Concentrations of Plutonium in Tissues of
Dogs 7-9 Years After Inhalation of $^{239}\text{PuO}_2$

<u>Tissue</u>	<u>Relative Concentration of Plutonium</u>
Lung	1
Thoracic Lymph Nodes	1400
Abdominal Lymph Nodes	100
Liver	0.5
Spleen	0.2
Bone	0.06

TABLE 4

Translocation in Dogs 30 Days After Inhalation
of Oxides of Transuranium Elements

<u>Transuranic Oxides</u>	<u>Particle Size</u>		<u>Tissue Content</u>					
			<u>Percent of Final Body Burden</u>					
			<u>Lung</u>	<u>Thoracic Lymph Nodes</u>	<u>Liver</u>	<u>Bone</u>	<u>Muscle</u>	<u>All Other</u>
$^{244}\text{CmOx}$	AMAD (μm)	GSD						
$^{244}\text{CmOx}$	0.5	2.1	20	0.5	25	25	22	9
$^{241}\text{AmO}_2$	1.3	2.	55	-	19	11	11	4
$^{238}\text{Pu}(\text{hydrated oxide})$	0.9	2.6	64	0.5	8	23	2	3
$^{238}\text{PuO}_2$ (750°)	2.2	2.3	96	0.8	0.3	0.5	3	0.3
$^{238}\text{PuO}_2$ (PPO)	1.9	1.7	96	1	0.2	1	1	1
$^{238}\text{PuO}_2$ (PMC)	2.	1.9	94	1	0.6	1.4	1.4	2
$^{239}\text{PuO}_2$	2.	2.	97	2	0.01	0.01	0.01	0.2

(Unpublished data provided by D. K. Craig, PNL)

TABLE 5

Deposition of Plutonium in Gonads

	<u>Percent of Pu in Blood</u>
Testes	0.05
Ovaries	0.01

From C. R. Richmond and R. L. Thomas - in press

TABLE 6

Cross-Placental Transfer of Transuranic Elements in Rats
(Percent of Injected Dose per Gram)

Time of Injection (Day of Ingestion)		^{233}U	^{237}Np	^{238}Pu	^{239}Pu	$^{239}\text{Pu}^*$	^{253}Es	^{137}Cs	^{144}Ce
15	Fetus	0.01	0.01	0.01	0.01	0.0002	0.002	0.09	0.02
	Placenta	0.01	0.04	0.22	0.40	0.01	0.02	0.16	0.36
	Membranes	0.13	0.81	3.80	1.93	0.03	0.04	0.08	1.79
19	Fetus	0.03	0.02	0.01	0.01	0.0004	0.008	0.08	0.02
	Placenta	0.05	0.18	0.51	0.91	0.02	0.08	0.14	0.39
	Membranes	0.33	1.24	4.30	3.20	0.07	0.05	0.07	1.60

*Polymeric

(Summary of published and unpublished data provided by D. D. Mahlum and M. R. Sikov, PNL)

TABLE 7

Hematologic Effects of Transuranic Elements

<u>Isotope</u>	<u>Species</u>	<u>Dose</u> <u>(μCi/kg)</u>	<u>RBC</u>	<u>Neutro</u>	<u>Lymph</u>	<u>Mono</u>	<u>EOS</u>
<u>I.V.</u>							
^{226}Ra	dog	.06-10.4	<u>+</u>	↓ ↑	↓	↓ ↑	↓ ↑
^{239}Pu	dog	.02-2.9	<u>+</u>	↓ ↑	↓	↓ ↑	↓ ↑
^{228}Th	dog	.02-2.8	<u>+</u>	↓	↓	↓ ↑	↓
^{228}Ra	dog	.05-8.5	<u>+</u>	↓	↓	↓ ↑	↓
^{241}Am	dog	.02-2.8	<u>+</u>	↓	↓	↓ ↑	↓
^{249}Cf	dog	2.8	-	↓ ↑	↓	↓ ↑	↓ ↑
^{253}Es	dog	2.9	↓ ↑	↓ ↑	↓ ↑	↓ ↑	↓ ↑
^{253}Es	pig	3.0	-	↓ ↑	-	↓ ↑	↓ ↑
^{242}Cm	dog	2.6	↓ ↑	↓ ↑	↓	↓ ↑	↓
<u>Inhaled</u>							
^{239}Pu	dog	.08-5.8*	-	-	↓	-	-
^{238}Pu	dog	.14-5.4*	-	<u>+</u>	↓	-	-
^{241}Am	dog	~ 25	-	↓	↓	↓	↓

*Initial Lung Burden (μ Ci)

+Depression at highest doses only

↓Sustained depression

↓↑Depression with evidence of recovery

-No effect

(Summary of published and unpublished data provided by H. A. Ragan, PNL)

TABLE 8

Plutonium-Induced Bone Cancers
in Utah Dog Study

Injected Dose ($\mu\text{Ci/kg}$)	Cancer Incidence	Dose to Bone of Cancer Dogs (RAD)
2.9	7/9 = 78%	4900
0.9	12/12 = 100%	1300
0.3	12/12 = 100%	600
0.1	10/12 = 83%	310
0.05	9/13 = 69%	190
0.016	4/13 = 31%	78
Controls	0	

TABLE 9

Osteosarcoma in Dogs after Inhalation of $^{238}\text{PuO}_2$

$^{238}\text{PuO}_2$	Survival Time (Months)	Terminal Body Burden (μCi)	Plutonium Distribution (% of Body Burden)				Osteo- sarcoma
			<u>Lungs</u>	<u>Thoracic Lymph Nodes</u>	<u>Liver</u>	<u>Bone</u>	
Calcined at 350°C	23-70	2.6-3.0	17	9	23	47	5/8*
Crushed microspheres	22-76	0.2-3	20	12	16	24	4/8**

* 1 lung tumor

** 1 myelogenous leukemia and 1 fibrosarcoma
(Park et al., in press)

TABLE 10

Life Span Studies of Intravenously Injected
Transuranium Elements in Dogs at University of Utah
(Numbers of Dogs*)

Nuclide	Age Group of Dogs	Injected Doses, $\mu\text{Ci/kg}$										
		<u>0</u>	<u>.0006</u>	<u>.0016</u>	<u>.005</u>	<u>.01</u>	<u>.015</u>	<u>.05</u>	<u>.1</u>	<u>.3</u>	<u>.9</u>	<u>2.8</u>
^{239}Pu	Juveniles	5					1			4		12
	Young adults	31	26	42	25	36	12					
	Aged									2		3
^{241}Am	Young adults			13	13		12	13	10	16	1	
^{249}Cf	Young adults	6	6		6		6		6	6		
^{252}Cf	Young adults	6	6		6		6		6	6		
^{253}Es	Young adults									3		4

*August, 1974 - 356 dogs

TABLE 11

Life Span Study of Inhaled Plutonium
in Dogs at Lovelace

	<u>Particle Size*</u> (μm)	<u>Initial Lung Burden</u> (μCi)	<u>Number of Dogs</u>
$^{238}\text{PuO}_2$	1.5	5.6, 3, 1.4, 0.7, 0.3, 0.1	72
$^{238}\text{PuO}_2$	3.0	"	72
$^{239}\text{PuO}_2$	0.75	"	72
$^{239}\text{PuO}_2$	1.5	"	72
$^{239}\text{PuO}_2$	3.0	"	72
			<hr/> 360 + 60 controls

*Aerodynamic diameter of monodisperse aerosols

TABLE 12

Life Span Study of Inhaled Plutonium in
Dogs at Battelle-Northwest

$^{238}\text{Pu}^{16}\text{O}_2$		$^{239}\text{PuO}_2$	
Initial Lung Burden (μCi)	No. of Dogs*	Initial Lung Burden (μCi)	No. of Dogs*
.002	20	.004	20
.02	20	.02	20
.05	20	.08	20
.35	20	.3	20
1.3	20	1.1	20
5.2	13	5.8	8
	<u>113</u>		<u>108</u>

*Half male and half female + 40 controls

PULMONARY RETENTION OF INHALED PuO_2 IN DOGS

Figure 2

Pulmonary Retention of Inhaled PuO_2 in Dogs

Figure 3

Retention of transuranium elements in rat lungs (^{238}Pu , ^{239}Pu , ^{241}Am and ^{242}Cm - Nenot et al., 1972; $^{253}\text{EsCl}_3$ - Ballou et al., submitted for publication)

Figure 4

(Redrawn from R. O. McClellan, 1972)

Figure 5

Autoradiograph showing particulate and non-particulate plutonium in lung of a rat immediately after inhalation of $^{239}\text{Pu}(\text{NO}_3)_4$
(Provided by J. E. Ballou, PNL)

Figure 6

Autoradiograph of a histologic section from a
rat lung 7 days after inhalation of $^{253}\text{Es}(\text{NO}_3)_3$
(Provided by J. E. Ballou, PNL) PNL747349-7

Figure 7

Electromicrograph of plutonium dioxide particles (inset) from an aerosol inhaled by rats and of a cell from the lung of an exposed rat. The dense appearing material in the cell is plutonium dioxide which had been engulfed by the cell.

(Provided by C. L. Sanders and R. R. Adey, PNL) PNL0673470-5

Figure 8

Autoradiograph showing aggregation of ^{238}Pu in a lung
of a rat 100 days after inhalation of $^{238}\text{Pu}(\text{NO}_3)_4$.
(Provided by J. E. Ballou, PNL) PNL747349-3

Figure 9

Autoradiograph showing $^{239}\text{PuO}_2$ particles in a subpleural
area of lung from a dog. (Provided by G. E. Dagle, PNL)
PNL747349-2

Figure 10

Autoradiograph showing ^{238}Pu particles in scar tissue in a lung of a dog 5 years after inhalation of $^{238}\text{PuO}_2$.
(Provided by J. E. Lund) PNL747349-6

DISTRIBUTION OF PLUTONIUM IN DOGS AFTER INHALATION OF $^{239}\text{Pu}(\text{NO}_3)_4$

Figure 11

DISTRIBUTION OF PLUTONIUM IN DOGS AFTER INHALATION OF $^{239}\text{PuO}_2$

Figure 12

**DISTRIBUTION OF PLUTONIUM IN TISSUES OF DOGS
5 YEARS AFTER INHALING $^{238}\text{PuO}_2$ OR $^{239}\text{PuO}_2$**

Figure 13

Figure 14

(Redrawn from R. O. McClellan, 1972)

EFFECT OF INHALED $^{239}\text{PuO}_2$ ON BLOOD LYMPHOCYTE LEVELS

Figure 15

PLUTONIUM-INDUCED OSTEOSARCOMA IN EXPERIMENTAL ANIMALS

Figure 16

Plutonium-induced Osteosarcoma in Experimental Animals. Mean incidence and radiation dose values are those reported in the literature. Binomial confidence limits were calculated from data included in the referenced literature.

- o ^{239}Pu Citrate, Monomeric - IV - Dogs (from Jee, 1972)
- Δ ^{239}Pu Citrate - Inhaled - Rats (from Buldakov and Lyubchansky, 1970)
- ∇ ^{239}Pu Plutonylpentacarbonate - Inhaled - Rats (from Buldakov and Lyubchansky, 1970)
- $\diamond$ ^{239}Pu Nitrate - Sub- and Intracutaneous - Rats (from Buldakov, et al., 1971)
- $\blacktriangledown$ ^{239}Pu Citrate - Oral (Daily) - Rats (from Buldakov et al., 1969)
- $\bullet$ ^{239}Pu Plutonyltriacetate - I.T. - Rats (from Erokhin et al., 1971)
- $\blacktriangle$ ^{239}Pu Citrate - IV - Mice (from Finkel and Biskis, 1962)
- $\square$ ^{239}Pu Citrate, Monomeric - IV - Mice (from Rosenthal and Lindenbaum, 1967)

- ^{239}Pu Citrate, Polymeric - IV - Mice (from Rosenthal and Lindenbaum, 1967)
- X $^{238}\text{PuO}_2$ - Inhaled - Rats (from C. L. Sanders, 1973)
- ◆ ^{239}Pu Nitrate - I.T. - Rats (from Erokhin et al., 1971)
- + ^{239}Pu Nitrate - I.T. - Rabbits (from Koshnurnikova et al., 1971)
- * ^{239}Pu (Pentacarbonate) - Inhaled - Rabbits (from Koshnurnikova et al., 1971)
- ◁ ^{239}Pu Citrate - Inhaled - Rats (from Koshnurnikova et al., 1971)
- ◀ ^{239}Pu Pentacarbonate - Inhaled - Rats (from Koshnurnikova et al., 1971)

(See Bair, 1974 for complete references)

Figure 17

Plutonium-induced Lung Cancer in Experimental Animals
Mean incidence and radiation dose values are those reported in the literature. Binomial confidence limits were calculated from data included in the referenced literature.

- $^{239}\text{PuO}_2$ - Dogs (from Park and Bair, 1972)
- ▽ $^{239}\text{PuO}_2$ - Mice (from Temple et al., 1959)
- △ $^{239}\text{PuO}_2$ - Mice (from Temple et al., 1959)
- ◆ $^{239}\text{PuO}_2$ - Mice (from Wager et al., 1956)
- ^{239}Pu Citrate - Rats (from Buldakov and Lyubchansky, 1970)
- ^{239}Pu - Plutonylpentacarbonate - Rats (from Buldakov and Lyubchansky, 1970)
- x ^{238}Pu - Rats (from C. L. Sanders, 1973)

- △ ^{239}Pu - Rats - $\text{Pu}(\text{NO}_3)_4$ (from Erokhin et al., 1971)
- + ^{239}Pu - Rabbits - $\text{Pu}(\text{NO}_3)_4$ (from Koshnurnikova et al., 1971)
- * ^{239}Pu - Rabbits - NH_4 Pu Pentacarbonate (from Koshnurnikova et al., 1971)

(See Bair, 1974 for complete references)

Figure 18

(J. M. Thomas and W. J. Bair, to be published)
PNL66233-1

OSTEOSARCOMA INCIDENCE IN FEMALE RATS

Figure 19

(D. D. Mahlum and M. R. Sikov, PNL, to be published in Pacific Northwest Laboratory Annual Report for 1974) PNL747510-13

Figure 20

Autoradiograph of a histologic section from a lung of a rat after inhalation of $^{244}\text{Cm}(\text{NO}_3)_3$. (Provided by J. LaFuma, Commissariat a l'Energie Atomique, Association Euratom. C.E.A. CEN. FAR. France.) PNL747596-4

Figure 21

Autoradiograph of a histologic section from a lung of a rat after inhalation of $^{239}\text{Pu}(\text{NO}_3)_4$. (Provided by J. Lafuma, Commissariat a l' Energie Atomique, Association Euratom. C.E.A. CEN. FAR. France.) PNL747596-1

Figure 22

Autoradiograph of a histologic section from a lung of a rat 356 days after intraperitoneal injection of $^{239}\text{PuO}_2$ particles. (Provided by C. L. Sanders, PNL) PNL747358-1

Figure 23

Autoradiogram of a histologic section from a lung of a dog 2 years after inhalation of $^{239}\text{PuO}_2$. (Provided by G. E. Dagle, PNL) PNL66199-1

OBSERVED BIOLOGICAL EFFECTS OF INHALED PLUTONIUM

Figure 24

Biomedical Effects of Plutonium on Humans

~~p 231 thru 269~~
p 231 thru 269

by William W. Burr, Jr., M.D.
Deputy Director, Division of Biomedical
and Environmental Research
U. S. Atomic Energy Commission
Washington, D. C. 20545

→ end see p 235

part of the AEC presentation at
EPA Plutonium Standards Hearings
Washington, D.C., December 10-11, 1974

Although considerable data exists concerning the biological effects of plutonium on experimental animals, comparatively little information is available regarding the effects of plutonium and other actinide elements on man. However, despite its limited availability, we regard the human data as highly relevant.

The human data serves at least two purposes. First, it provides a check on the metabolic behavior of the actinide elements in man as compared to experimental animals. Secondly, the human observations will, in the course of time, provide an improved basis for the guidelines and standards under which the industry operates. The available data make us confident that present exposure standards are not grossly inadequate and that the experimental animal work can be accepted as relevant to the human situation. However, the human data are far too limited at this time to permit more specific conclusions to be drawn from them with respect to exposure limits.

Those individuals who have been exposed to and/or retained plutonium for the longest periods of time are of particular interest. There are three such groups. The first of these consists of those persons who have

been occupationally exposed to plutonium at some time during their working life. While some of these exposures occurred 30 years ago, others have occurred more recently; it will be some time before these more recent exposures will contribute to our store of knowledge in a meaningful way. Industrial accidents during the Manhattan Project resulted in inhalation exposures of a number of individuals to plutonium. Some of these persons have maintained multiples of the maximum permissible body burden for nearly three decades.

In addition to the depositions that date from the mid-forties, we know of over 200 industrial exposures between 1953 and 1970 that resulted in burdens of plutonium exceeding 25% of the maximum permissible body burden. Other exposures have occurred before and since the period covered by these statistics. Although the exact levels of internal contamination are uncertain in most such cases, it is evident that these exposures constitute a valuable resource for current and future study. Some information has been obtained already from particular groups of industrially exposed persons; other studies are now being formulated and expanded to collect data from additional members of that population.

A second source of valuable information is a group of 18 people, thought to be hopelessly ill, who were injected with plutonium during and immediately after the days of the Manhattan Project to study excretion and distribution patterns in man. Data from these persons provided the basis for the excretion equations developed by Dr. Wright Langham that have been used in modified form to estimate plutonium body burdens in workers ever since then.

A third population of interest is that of the world at large. The general population has accumulated minute quantities of plutonium from the fallout debris that resulted from nuclear testing in the atmosphere and from the atmospheric burn-up of a thermoelectric generator. Study of this population will give insight into the extent to which man takes up plutonium from the biosphere.

Programs for the analysis of human tissues obtained at autopsy from exposed workers and from non-occupationally exposed persons will in time furnish much information regarding the efficiency with which man incorporates plutonium into his body and will provide data regarding the distribution of plutonium among the various body tissues. In the case of occupational exposures these studies permit us to compare estimates of body burdens based on analysis of urine specimens or external lung counting with estimates based on actual analysis of tissues obtained from the same individuals. These programs have been expanded in recent years.

In view of the fact that thousands of persons have been exposed or potentially exposed to plutonium during the course of their work, it is inevitable that some of those people who may have some lung or body burden of plutonium will die of cancer, including lung cancer. When this occurs it must be recognized that the appearance of common forms of cancer in persons with plutonium burdens does not constitute proof that the deposition is causally related to the disease. In order to establish whether or not the number of such deaths exceeds our expectation for a comparable unexposed population, our scientific resources will be taxed to the utmost. We are now entering a period when the working population that was young during the 1940's may be expected to develop a meaningful incidence of disease of all kinds. Follow-up studies attempting to establish whether any detectable

increase in relevant disease may be seen in the exposed populations will become increasingly important.

Although the clinical follow-up of persons with burdens has been reassuring, any conclusions with respect to late effects of plutonium in man must remain tentative for some time. We can, however, state with confidence that available data does not support the viewpoint that the current radiation protection standards and guidelines which have been followed for many years underestimate by many orders of magnitude the risk due to plutonium deposition in man.

Dr. Richmond, Associate Director for Biomedical and Environmental Sciences at the Oak Ridge National Laboratory, will summarize the data obtained from the various populations that I have mentioned.

Biomedical Effects of Plutonium on Humans

by C. R. Richmond
Oak Ridge National Laboratory
Oak Ridge, TN 37830

part of the AEC presentation at
EPA Plutonium Standards Hearings
Washington, D.C., December 10-11, 1974

INTRODUCTION

My name is Chester R. Richmond. I am the Associate Director for Biomedical and Environmental Sciences at the Oak Ridge National Laboratory. However, the views I express here are my own.

Plutonium was recognized as a potentially hazardous material soon after its discovery in early 1941. The urgency to conduct biological studies with plutonium was appreciated by several people, notably Dr. Seaborg, with the hope that the unfortunate problems experienced with radium earlier in the century would not be repeated. Within three years of the discovery of plutonium (^{238}Pu) in February, 1941, 0.5 g ^{239}Pu had been separated from the material produced by the Clinton pile and on 8 February 1944, Dr. J. G. Hamilton and coworkers at Berkeley received about 10 mg to begin experimental studies in rodents.

During late 1943 and early 1944, plutonium operations at Los Alamos consisted of research activities involving milligram quantities of material. During late 1944, gram quantities were processed in research activities directed mainly toward the production of pure plutonium metal and investigation of its physical and chemical properties. By mid-1945, kilogram quantities were processed as part of the effort to produce the nuclear components for the Alamogordo and Nagasaki weapons. Some of our most

relevant data as regards exposure of humans to plutonium comes from the medical follow-up of the military personnel who worked with plutonium at Los Alamos in 1944 and 1945.

Although we have accumulated a considerable amount of information on the biological effects of plutonium on experimental animals, there is little to be said of the data on effects in humans. Obviously, we should be encouraged because of the lack of data on biological effects of plutonium in man.

DEVELOPMENT OF MAXIMUM PERMISSIBLE BODY BURDEN (MPBB) FOR Pu

Throughout 1943 and the first nine months of 1944, a maximum permissible body burden of 4-5 μg was assumed to be an acceptable guide even though no reliable method of estimating personnel exposure to plutonium had been developed. The value was derived by using bone as the critical organ and making a direct comparison with the energy deposited from 0.1 μg of ^{226}Ra fixed in the body (assuming 50% radon exhalation). Later, because of apparent differences in bone deposition patterns between Pu and Ra in rodents, a safety factor of about 5 was introduced, and the maximum permissible body burden became 1 μg . This value was used until the Tripartite Permissible Dose Conference at Chalk River, Canada, in late September 1949, at which time Dr. A. Brues presented experimental chronic toxicity data from rodents that suggested ^{239}Pu was 15 times more damaging than ^{226}Ra when both were injected in equivalent microcurie quantities. The conference recommended that the MPBB be reduced to 0.1 μg .

Subsequent reexaminations of the experimental data led to a recommendation of 0.6 μg as the MPBB for ^{239}Pu . This decision was based upon the following observations related to the assumption that 0.1 μCi of fixed ^{239}Pu was

equivalent to 0.1 μCi of fixed ^{226}Ra .

(1) The Pu:Ra toxicity ratio of 15:1 was based on the injection of known amounts into rodents. Since $\sim 75\%$ of the injected Pu was retained in rodents while only $\sim 25\%$ of the Ra was retained, the ratio on the basis of retained dose could be lowered by a factor of about 3.

(2) Because radon was about 50% retained in man and only about 15-20% retained in rodents, the toxicity ratio could be lowered by another factor of at least 2 on the basis of relative energy deposited.

Thus, strictly on the basis of biological data, the MPBB for man was calculated to be:

$$(\text{MPBB})_{\text{Pu}} = 0.1 \times \frac{24,000}{1,600} \times \frac{1}{15} \times \frac{3}{1} \times \frac{2}{1} = 0.6 \mu\text{g} \text{ (0.04 } \mu\text{Ci)}$$

As a result of this information, the AEC authorized 0.5 μg (0.033 μCi) ^{239}Pu as the MPBB. In 1951, the International Commission on Radiological Protection (ICRP) at a meeting in London recommended a value of 0.04 μCi which was later endorsed at the Tripartite Conference on Permissible Dose at Harriman, New York in March 1953. In the fall of 1953, both the National Committee on Radiation Protection and Measurements (now the National Council) and the ICRP recommended a MPBB of 0.04 μCi for ^{239}Pu in their official publications; the value has remained unchanged to date although the MPBB has been discussed in more recent publications of both organizations.

MANHATTAN PROJECT EXPOSURES

Since the discovery of plutonium over three decades ago,⁽¹⁾ personnel exposures have been studied and reported on in varying degree, both during life and after death.⁽²⁻⁹⁾ One of the most interesting groups, because of

both the length of the period since exposure and the levels of exposure, is that of the Manhattan Project plutonium workers.⁽¹⁰⁾

Twenty-five male subjects, who worked with plutonium during World War II under very crude working conditions by today's standards, have been followed medically during the intervening period. Within the past several years, 21 of these men have been examined at the Los Alamos Scientific Laboratory. In addition to physical examinations and laboratory studies (complete blood count, blood chemistry profiles and urinalysis), roentgenograms were taken of the chest, pelvis, knees and teeth. Chromosomes of lymphocytes cultured from peripheral blood and pulmonary cytology were also studied. Urine specimens assayed for plutonium yielded calculated body burdens which ranged from 0.005 to 0.42 μ Ci. These estimates of body burden are generally higher than earlier estimates based on radioassay of urine samples collected in the past, perhaps reflecting uncertainties in the models used to estimate body burden from excretion data. Table 1 indicates the kinds of information obtained from the Manhattan Project plutonium workers. Most, but not all, of these examinations have been conducted every four to five years since the group has been studied.

This group of men in their early to mid-fifties had only the usual diseases encountered in this age zone. One man had a coronary occlusion but had recovered and was well compensated. Another of the original group died in 1959 of a coronary occlusion at age 38. Another had a hamartoma of the lung surgically removed without complication in 1971. A third had a melanoma of the chest wall (regional lymph nodes were negative).

A fourth had a partial gastrectomy for bleeding ulcer. Several had mild hypertension and moderate obesity, and one had gout. All were working actively. More detailed information on this particular group of workers has been published.⁽¹¹⁻¹²⁾

Blood samples were obtained from the group during the most recent medical checkups for chromosome studies using standardized established techniques. No abnormalities were found in these subjects. Except for one special case reported by Schofield and Dolphin⁽¹³⁾ chromosome aberration studies carried out on plutonium workers in the United Kingdom showed no significant increase in aberration yield.⁽¹⁴⁾

Because lung cancer has been observed experimentally in animals exposed to plutonium aerosols, cytological examinations of bronchial cells in sputum samples have been added to these studies. In a few subjects, moderate to severe dysplastic changes have been observed. The significance of these changes is not clear except in one man who was a heavy cigarette smoker (3 packages per day).

It is important to realize that these men worked under very crude working conditions as judged by today's standards. At times, the activity to which some of these personnel were exposed was orders of magnitude over the presently accepted maximum permissible air concentrations. Most of the exposures were believed to have occurred via inhalation as evidenced by a strong correlation with frequent contamination of the nasal vestibule and highly contaminating operations. The nasal swabs on one occasion yielded over 1 μg Pu from each nostril. Figure 1 shows the building in which these men worked in the early years.

Attempts have been made to estimate the number of particles inhaled by the Manhattan Project plutonium workers. By making certain assumptions with respect to the mass median diameter, geometric standard deviation of the distribution and the particle density, one can calculate the mass fraction for plutonium dioxide particles larger than any stated size. For example, the mass fraction for plutonium particles larger than 0.6 micron diameter is approximately 15%. Further calculations indicate that approximately 10^7 particles larger than 0.6 micron diameter could have been retained by the 25 subjects during their exposures in 1944 and 1945.⁽¹⁵⁾ The observed lung cancer incidence is zero almost 30 years since exposure.

Table II shows the current status of several LASL plutonium study groups. Group 1 was discussed in the preceding paragraphs. Group 2, which is now in the early stages of study, will expand the size of the original cohort by perhaps 40 people. Twenty-eight of the 40 men who have been identified have been located and have responded to questionnaires. Once again, these are extremely important subjects to study intensively as approximately three decades have elapsed since their exposures. Group 3 will comprise a broader spectrum of exposures, including more recent accidents and some exposures to ^{238}Pu . All are estimated to have systemic plutonium burdens of 4 or more nanocuries. Table III shows the

estimates of plutonium systemic body burdens on certain workers in Group 1, all of whom had estimated burdens greater than 120 nCi (3 maximum permissible body burdens) in 1972. The table also contains the 1953 and 1962 estimates for these individuals. The increase in the values for each individual with time is at least partly attributed to modifications in the method of estimation that have generally resulted in higher estimates in more recent years. The model for estimation of body burdens from urine assay values involves uncertainties that limit the accuracy of estimation.⁽¹⁶⁾ It is also true that some of the body burden estimates are based on relatively few data points. Again, the original exposures were in the early 1940's so that Case 3, who now has approximately 10 times the allowable occupational bone burden, has carried this estimated 410 nCi of plutonium for approximately three decades. The selected cases shown in Table III represent systemic plutonium burdens ranging from 0.13 to 0.42 μ Ci, which correspond to annual bone doses of approximately 2 to 6 rad.

Table IV contains information which is detailed in an earlier publication.⁽¹⁰⁾ The data are for the $^{239,240}\text{Pu}$ content for some tissues that were removed from Case 2 of Group 1, who developed a non-malignant growth (hamartoma) in the lung. Surgical removal of the hamartoma, which was found during a medical follow-up study, afforded an opportunity to obtain tissue from the hamartoma, lymph nodes, rib and normal lung for radiochemical analysis. The concentration of $^{239,240}\text{Pu}$ was approximately the same in both the tumor and normal lung tissue. The lowest plutonium concentration was found in a rib sample and the highest in the lymph node. This distribution is consistent with experimental findings in dogs exposed to plutonium dioxide by inhalation. If one assumes a total lung weight of 1000 grams, tracheo-

bronchial lymph node weight of 20 grams and a homogeneous distribution of plutonium throughout these tissues, the total plutonium burden is estimated to be 8 nCi, roughly equally divided between the lung and lymph nodes. This estimate of the burden of plutonium in the thorax based on extrapolation from the analysis of lung and lymph node tissue is in reasonable agreement (within a factor of 2) with the estimate based on chest counting procedures.

Figure 2 is a photomicrograph of an autoradiograph of a plutonium particle in a lymph node section removed from Case 2. Additional observations on histologic sections of lymph node tissue suggested a non-uniform radiation dose distribution from the plutonium particles.

PLUTONIUM ADMINISTRATION STUDIES IN HUMAN SUBJECTS

In an attempt to determine relationships between urinary excretion, total excretion and body content of plutonium, 18 persons received plutonium parenterally during 1945-1947⁽¹⁷⁻¹⁸⁾ as shown in Table V. Fifteen of the 18 were older than age 45, and all but two of the 18 were given ^{239}Pu only (one received both ^{238}Pu and ^{239}Pu and another received only ^{238}Pu). The amounts of plutonium administered ranged from about 0.1 to about 6 μCi . For comparison, the current occupational maximum permissible body burden for ^{239}Pu is 0.04 μCi .

Although these subjects were thought to be hopelessly ill, four of the group were alive in November 1973, almost three decades after receiving the plutonium. Excretion data for some of the survivors have been reported recently.⁽¹⁹⁾ These data provide a unique opportunity to verify the excretion equations that are used currently by radiation protection personnel to estimate body burdens. It is of considerable interest that much of the data used to establish the excretion equations was obtained from this group

of 18 subjects during the relatively short period (several months in most cases) during which they were studied. In addition to the data obtained from those individuals, Langham used data obtained from several Los Alamos occupational exposure cases for about 300 days and one for about 1700 days in formulating his excretion curves. These equations have been very useful although they have proven to be somewhat conservative when estimated body burdens based on urine assay are checked against estimates based on post-mortem analyses.

One of the original 18 plutonium recipients is of particular interest. He was a white 58 year old male who was believed to have a gastric carcinoma with hemorrhage when he received 5.19 μCi of ^{238}Pu and 0.12 μCi of ^{239}Pu as $\text{PuO}_2(\text{NO}_3)_2$ by intravenous injection. Gastrectomy disclosed a gastric ulcer from which the patient recovered. He did not die until some 21 years later; the cause of death was cardiovascular disease. We can obtain a very rough estimate of the bone dose by assuming 40% deposition in the skeletal tissues with no subsequent loss. Under these circumstances, the skeletal dose over the 21-year period would be approximately 900 rad. The annual dose rate to the skeletal tissues would be approximately 40 rad, a factor of approximately 70 higher than the annual skeletal dose rate of 0.6 rad delivered by the maximum occupational bone burden of 40 nCi of ^{239}Pu .

Some information can be obtained on the amount of plutonium in the gonads of some of these subjects. The fraction of administered plutonium found in the gonads at autopsy was 9×10^{-5} for one female and about 3×10^{-4} for three male subjects. These numbers agree quite well with data obtained from several species of experimental animals. (20)

U. S. TRANSURANIUM REGISTRY

During the summer of 1968, the United States Atomic Energy Commission authorized the establishment of the National Plutonium Registry which was later renamed the United States Transuranium Registry (USTR). The registry is operated by the Hanford Environmental Health Foundation in Richland, Washington, and collects information from AEC contractors and licensees regarding employees potentially exposed to transuranium elements.⁽²¹⁻²³⁾ Cooperation with the USTR is completely voluntary on an individual basis and includes release of medical and health physics data. Permission is also obtained on a voluntary basis for postmortem analyses of tissues of interest. Major AEC contractors and certain licensees handling plutonium and other transuranium elements have agreed to endorse the program and have recommended to their employees that they participate in this program.

The principal criterion used by the USTR to determine inclusion of an individual in the Registry is that the employer provide a routine surveillance program because of a reasonable likelihood that an exposure could occur. This rather broad criterion allows for the different methods used to estimate the extent of contamination, e.g., urine analysis, chest counting, or air concentration data, and for uncertainties in estimating burdens under certain conditions, e.g., chronic inhalation of insoluble plutonium. It also avoids the exclusive consideration of cases involving heavy exposures.

At autopsy, comparisons can be made between estimates of the body burden based upon tissue analyses and estimates made previously on the basis of health physics and operational data. In addition to a medical history, information may be obtained on an employee's work history, smoking habits, exposure to toxic materials, and other pertinent data.

Preliminary findings for the first fourteen autopsy cases reported by the USTR appeared in the proceedings of the 12th Hanford Biology Symposium held in 1972.⁽²⁴⁾ To date, information obtained by the USTR indicates that estimates of the plutonium systemic burden based on urine analysis have been on the conservative side, that is, they are higher than estimates based on analysis of tissues obtained at autopsy.⁽²⁴⁾ Workers in the United Kingdom have also found this to be true.⁽¹³⁾

Table VI indicates the status of the USTR as of June 1974. To date, most of the USTR activities have been confined to Hanford, Los Alamos, and Rocky Flats. The interested reader is directed to a recent USTR report for details of the level of cooperation between the USTR and the other AEC contractors shown in Table VI.⁽²⁵⁾

ACCIDENT CASES

A considerable amount of information has been obtained from accidental occupational exposures to plutonium. However, the total number of accident cases has been relatively small. Information obtained from the AEC's Division of Operational Safety as shown in Table VII indicates that during the period 1957 to 1970 about 200 contractor personnel had depositions greater than 25% of the occupational maximum permissible body burden (MPBB) or lung burden for plutonium. These data also indicate that inhalation is the major portal of entry and that more than half the exposure cases represent plutonium burdens less than 50% of the maximum permissible burden. Eighteen percent of the total exposures resulted in plutonium burdens greater than one MPBB. Table VII also shows that about 18% of the cases were treated by chelation therapy. Fifty-four percent resulted from production activities.

Operational experience at Windscale in the United Kingdom shows that 15 men have exceeded the maximum permissible body burden of 40 nCi during a time covering about 7000 man-years of plutonium production and handling. It is also possible that about half these men have considerably less plutonium in their bodies than the calculations based upon urine radiochemistry currently indicate.⁽¹³⁾ Because of the importance of human data it is important that studies of personnel involved in accidents be continued and perhaps expanded.

A case of contamination resulting from a puncture wound is extremely interesting as it has been interpreted by some as an example of cancer in man resulting from plutonium deposition. The lesion was first described in the literature more than ten years ago⁽²⁶⁾ and was included along with other information on plutonium wounds at a later time.⁽⁴⁾ The 5 nCi particle of plutonium was surgically excised from the individual's palm approximately four years after the accident. The radiation dose around the plutonium implanted in the palmar skin was estimated to be about 75 million rads. However, this kind of dose estimate is probably meaningless as we do not know which cells were exposed or for what time periods. The entire lesion was very small (estimated to be about $3 \times 10^{-5} \text{ cm}^3$). Figure 3 shows a histologic section of the lesion. The pathologist involved in the study described the cellular pattern in the lesion as having "a similarity to known precancerous epidermal cytologic changes."⁽²⁶⁾ This particular lesion appears to be the most severe demonstrable effect having a direct relationship to plutonium deposition in man.

TISSUE ANALYSIS PROGRAMS

For many years the Los Alamos Scientific Laboratory⁽²⁷⁾ and other AEC contractor laboratories⁽²⁸⁻³¹⁾ have conducted tissue analysis programs to

determine plutonium levels in tissues obtained at autopsy from both exposed occupational personnel and members of the general population who are not engaged in work with plutonium. These programs were started in the 1940's in the Hanford plant and at the Los Alamos Scientific Laboratory. A report from one program contains information on approximately 350 autopsies.⁽²⁸⁾ Additional reports from this and the other groups are available.⁽³²⁻³⁴⁾ Table VIII shows plutonium concentrations as determined for lung, liver, lymph nodes, kidney, and bone for the period 1959-1971 for non-occupationally exposed persons from several parts of the United States and for occupationally exposed persons.⁽³⁵⁾ Data for plutonium concentrations in gonadal tissue, which appeared in the original publication, are not included in Table VIII because errors associated with a change in analytical procedures were detected by the authors subsequent to the original publication. Similar data shown in Table IX have been obtained for non-occupationally exposed persons and represent analyses made during the period 1972-1973.⁽³⁵⁾ The average concentration in the lungs for the data shown in Table IX is about 0.3 pCi for a 1000 gram lung, and the lymphnode concentration is about 11 pCi/g. No unusually high concentrations of plutonium in gonadal tissue have been observed in this particular study. Recent analysis of the gonadal data suggest that the concentrations of plutonium are about 0.18 pCi/kg for non-occupationally exposed persons.

The higher plutonium concentrations in lymph node tissue of non-occupationally exposed individuals in Table IX as compared with those in Table VIII are not thought to represent real increases but rather to reflect an improvement in the technique for dissecting the lymph nodes from the lungs.

The AEC's Health and Safety Laboratory (HASL) recently has used information obtained from the International Commission on Radiological Protection to model the intake and body burden resulting from plutonium in fallout and to estimate the radiation dose to man from this source.⁽³⁶⁾ The cumulative lung and bone doses for the period 1954-2000 are estimated to be 16 and 34 mrem respectively. The HASL group has also compared body burdens based on their model with values obtained from the tissue sampling programs. The agreement is quite good between the Colorado-New Mexico tissue data and the model predictions as shown in Table X for 1970-1971. The comparison based on the New York tissue sampling data is not as good and may reflect the small sample of 25 autopsies that were included in the analysis.

Results of the tissue sampling programs for occupationally exposed plutonium workers has also given us the opportunity to compare the body burden at autopsy with that estimated during life on the basis of bioassay data. Almost without exception, workers in the USA^(25,29,35) and United Kingdom⁽¹³⁾ have found less plutonium by significant factors at autopsy as compared with the amount predicted during life. For example, the United Kingdom workers found that for 9 plutonium workers studied at autopsy, the body burden estimates based upon tissue analysis were lower by factors of 1.2 to 8.3 than those estimates made during life on the basis of urinary excretion analyses. Thus it would appear that estimates of the body burden made during life are conservative in that they predict more plutonium than is actually present in the body. Because a considerable amount of relevant data is now available, it may be appropriate for scientists in the field of radiation protection to explore this observation in more detail as regards current radiation protection practices and the guidelines followed in the nuclear energy industry.

Recently several investigators have examined the United Kingdom medical experience for workers handling plutonium. They have concluded that the information to date cannot conclusively validate or repudiate the presently accepted working levels for plutonium, but the information does allow for a certain amount of cautious optimism. They also state, "it is true to say that after 30 years' experience in the USA and 22 years' in this country, no disease attributable to plutonium toxicity has been diagnosed in any worker concerned in the production or manipulation of plutonium."⁽¹³⁾

PLUTONIUM IN MAN FROM FALLOUT

Plutonium is present in extremely small quantities in various organs of man today. Although most of the plutonium in fallout resulted from atmospheric testing of nuclear weapons by several countries prior to the 1963 limited test treaty ban, some material from contemporary atmospheric weapons testing by France and the People's Republic of China adds to the total human burden. The current lung burden as estimated for persons in the United States is about 0.3 pCi of ^{239,240}Pu, and a very rough estimate of the total amount in the body is perhaps 3.5 pCi as shown in Table XI. Estimates of the total amount of plutonium produced in the course of nuclear weapons testing vary, but a value of about 0.4 megacurie is a reasonable estimate. Of this amount, if 0.3 megacurie has returned to the biosphere, very little (about 10^{-8}) has found its way into the earth's population (3×10^9 people). Another approach to this matter is to divide the estimated average human burden (3.5×10^{-12} curies) by the estimated amount in the biosphere (0.3×10^6 curie); the average accumulation is about 10^{-17} per person.

CONCLUSION

Control of industrial hazards of ^{239}Pu processing is based upon the premise that personnel exposure should be as low as practicable not because the maximum permissible body burden is a level which would do harm but because it is sound industrial medical and health protection practice.

The lack of demonstrable biological effects of plutonium in man is reassuring and represents presumptive evidence that the standards are not grossly inadequate. My personal opinion is that those standards are adequate and that there is no compelling reason at this time to initiate changes, either upward or downward.

I would like to now quote a portion of the Rulison decision as given by Judge Arraj as I believe it is appropriate.

"The field of radiation protection is constantly changing with the appearance of new scientific knowledge on the biological effects of ionizing radiation. Careful decisions must be made in the context of contemporaneous knowledge. Such decisions cannot be indefinitely postponed if the potentials of atomic energy are to be fully realized. All that is required to establish reasonableness of the decision setting a standard under the statutory directive to protect the public health and safety is that it be made carefully in light of the best available scientific knowledge. Absolute certainty is neither required nor possible."

REFERENCES

1. G. T. Seaborg, Plutonium revisited. In Radiobiology of Plutonium, (B. J. Stover, and W.S.S. Jee, Eds.), pp. 1-21. The J. W. Press, University of Utah, Salt Lake City, 1972.
2. H. F. Anderson, W. E. Sheehan, J. R. Mann and R. W. Bistline, Evaluation of accidental personnel exposure to plutonium-238: Whole body counting and bioassay results. Health Physics 18, 631-639 (1970).
3. J. R. Mann and R. A. Kirchner, Evaluation of lung burden following acute inhalation exposures to highly insoluble PuO_2 . Health Physics 13, 877-882 (1967).
4. C. C. Lushbaugh, R. J. Cloutier, G. Humason, J. Langham and S. Guzak, Histopathologic study of intradermal plutonium metal deposits: Their conjectured fate. Ann. N. Y. Acad. Sci. 145, 791-797 (1967).
5. W. D. Norwood, J. A. Norcross, C. E. Newton, Jr., D. B. Hylton and C. Lagerquist, Preliminary autopsy findings in U. S. Transuranium Registry cases. In Radionuclide Carcinogenesis (C. L. Sanders, R. H. Busch, J. E. Ballow, and D. D. Mahlum, Eds.), pp. 465-474, AEC Symposium Series #29, USAEC Office of Information Services, June 1973.
6. C. R. Lagerquist, S. E. Hammond and D. B. Hylton, Distribution of plutonium and americium in the body 5 years after an exposure via contaminated puncture wound. Health Physics 22, 921-924 (1972).
7. W. H. Langham, J.N.P. Lawrence, Jean McClelland and L. H. Hempelmann, The Los Alamos Scientific Laboratory's experience with plutonium in man. Health Physics 8, 753-760 (1962).
8. C. E. Newton, Jr., M. V. Larson, K. R. Heid, I. C. Nelson, P. A. Fuqua, W. D. Norwood, S. Marks and T. D. Mahoney, Tissue analysis for plutonium at autopsy. In Diagnosis and Treatment of Deposited Radionuclides.

(H. A. Kornberg and W. D. Norwood, Eds.), pp. 460-468. Excerpta Medica Foundation, N.Y., N.Y. (1968).

9. H. Foreman, W. Moss and W. Langham, Plutonium accumulation from long-term occupational exposure. *Health Physics* 2, 326-333 (1960).
10. L. H. Hempelmann, W. H. Langham, C. R. Richmond, and G. L. Voelz, Manhattan Project plutonium workers: A twenty-seven year follow-up study of selected cases, *Health Physics* 25, 461-479 (1973).
11. L. H. Hempelmann, C. R. Richmond, and G. L. Voelz, A twenty-seven year study of selected Los Alamos plutonium workers, Los Alamos Scientific Laboratory report LA-5148-MS, 31 pp. (1973b).
12. L. H. Hempelmann, W. H. Langham, G. L. Voelz, and C. R. Richmond, Biomedical follow-up of the Manhattan Project plutonium workers, *In Proc. Third Congress of the Int. Radiation Protection Assoc.*, Washington, D. C. 1974 pp. (Sept. 9-14, 1973).
13. G. B. Schofield and G. W. Dolphin, U. K. experience on the medical aspects of radiological protection of workers handling plutonium, *Annals of Occupational Hygiene* (In press).
14. G. W. Dolphin, D. C. Lloyd and R. J. Purrott, Chromosome aberration analysis as a dosimetric technique in radiological protection, *Health Physics* 25, 7-15 (1973).
15. W. J. Bair, C. R. Richmond and B. W. Wachholz, A radiological assessment of the spatial distribution of radiation dose, USAEC WASH-1320, 47 pp., (Sept. 1974).
16. James N. P. Lawrence, "PUQFUA: An IBM-704 code for computing plutonium body burdens, *Health Physics* 8, 61-66 (1962). See also, Los Alamos Scientific Laboratory Report LA-2329 (April 1960).

17. W. H. Langham, S. H. Bassett, P. S. Harris, and R. E. Carter,
Distribution and excretion of plutonium administered intravenously
to man, Los Alamos Scientific Laboratory report LA-1151, p. 16
(Sept. 1950).
18. P. W. Durbin, Plutonium in man: A new look at the old data, (B. J.
Stover and W.S.S. Jee, Eds.), pp. 469-530. The J. W. Press, University
of Utah, Salt Lake City (1972).
19. J. Rundo, P. M. Starzyk and J. Sedlet, The excretion rate of plutonium
10,000 days after acquisition, Abstract of paper presented at Radiation
Res. Congr. Seattle, Washington. Radiation Res. 59, pp. 86-87, (July,
1974).
20. C. R. Richmond and R. L. Thomas, Plutonium and other actinide elements
in gonadal tissue of man and animals, Health Physics, In press.
21. H. D. Bruner, A plutonium registry, In Diagnosis and Treatment of
Deposited Radionuclides, (H. A. Kornberg and W. D. Norwood, Eds.),
pp. 661-665, Excerpta Medica Foundation, (1968).
22. W. D. Norwood, U. S. Transuranium Registry: progress and expectations,
In Radiobiology of Plutonium, (B. J. Stover and W.S.S. Jee, Eds.),
pp. 531-537, The J. W. Press, University of Utah, Salt Lake City,
(1972).
23. J. A. Norcross and C. E. Newton, Jr., U. S. Transuranium Registry: A
Progress Report, Health Physics 22, 887-890 (1972).
24. W. D. Norwood, J. A. Norcross, C. E. Newton, Jr., D. B. Hylton and
C. Lagerquist, Preliminary autopsy findings in U. S. Transuranium cas
In Radionuclide Carcinogenesis, (C. L. Sanders, et al., Eds.), AEC
Symposium Series 29, pp. 465-474 (1973).

25. W. D. Norwood and C. E. Newton, Jr., United States Transuranium Registry Summary to June 30, 1974 to USAEC Division of Biomedical and Environmental Research Hanford Environmental Health Foundation Report HEHF #22 28 pp., (1974).
26. C. C. Lushbaugh and J. Langham, A dermal lesion from implanted plutonium, Arch. Dermatol. 86, 461-464 (1962).
27. E. E. Campbell, M. F. Milligan, W. D. Moss and H. F. Schulte, History of the Plutonium Bioassay Program at the Los Alamos Scientific Laboratory, 1944-1972, USAEC Document LA-5008, 7 pp., (1972).
28. I. C. Nelson, K. R. Heid, P. A. Fuqua, and T. D. Mahony, Plutonium in autopsy tissue samples, Health Physics 22, 925-930 (1972).
29. C. R. Lagerquist, S. E. Hammond, D. L. Bokowski and D. B. Hylton, Distribution of plutonium and americium in occupationally exposed humans as found from autopsy samples, Health Physics 25, 581-584, (1973).
30. C. R. Lagerquist, D. L. Bokowski, S. E. Hammond and D. B. Hylton, Plutonium content of several internal organs following occupational exposure, Am. Ind. Hyg. Assoc. J. 30, 417-421 (1969).
31. E. E. Campbell, B. C. Eutsler, J. McClelland, and H. M. Ide, Plutonium in man, Health Physics 22, 931 (1972).
32. C. E. Newton, Jr., I. C. Nelson, K. R. Heird and H. V. Larson, Trans-uranium elements and thorium in man, assessment, applicability, of biologic models and needed research, BNWL-SA-279, (August 1965).
33. C. E. Newton, Jr., K. R. Heid, H. V. Larson and I. C. Nelson, Tissue sampling for plutonium through an autopsy program, BNWL-SA-918 (Sept. 1966).

34. E. E. Campbell, M. F. Milligan, W. D. Moss, H. F. Schulte, and J. F. McInroy, Plutonium in autopsy tissue, Los Alamos Scientific Laboratory report LA-4875, 47 pp., (January 1973).
35. Annual Report of the Biomedical and Environmental Research Program of the LASL Health Division for 1973 to the USAEC, Division of Biomedical and Environmental Research, Los Alamos Scientific Laboratory Report LA-5633-PR, (C. R. Richmond and E. M. Sullivan, Eds.), (1974).
36. B. G. Bennett, Fallout ^{239}Pu dose to man, In Fallout Program, Quarterly Summary Report, Health and Safety Laboratory report HASL-278 (January 1, 1974).

Figure 1

Figure 2

Figure 3

TABLE I

**BIOMEDICAL WORK RELATED TO MANHATTAN
PROJECT PLUTONIUM WORKERS**

**MEDICAL HISTORY AND EXAMINATION
RADIOLOGY
KARYOLOGY
PULMONARY CYTOLOGY
URINE RADIOCHEMISTRY
CHEST COUNTING (URANIUM L X-RAYS)
BLOOD CHEMISTRY PROFILES
HEMATOLOGY**

TABLE II

**LOS ALAMOS SCIENTIFIC LABORATORY
PLUTONIUM STUDY GROUPS**

GROUP 1^a

25 MEN EXPOSED DURING 1944-1945
PERIODIC BIOMEDICAL FOLLOW-UP FOR THREE DECADES
REPORTS AVAILABLE IN LITERATURE

GROUP 2

42 MEN ALSO EXPOSED DURING MANHATTAN PROJECT
28 LOCATED AND RESPONDED TO QUESTIONNAIRES

GROUP 3

190 EARLY AND CURRENT PLUTONIUM WORKERS
175 IDENTIFIED BY SOCIAL SECURITY NUMBER
2 IDENTIFIED BY MILITARY SERVICE NUMBER
29 RESPONSES

^aMANHATTAN PROJECT PLUTONIUM WORKERS

TABLE III

**PLUTONIUM SYSTEMIC BODY BURDEN ESTIMATES FOR SELECTED
MANHATTAN PROJECT PLUTONIUM WORKERS AT THREE DIFFERENT TIMES ^a**

<u>CASE CODE</u>	<u>²³⁹⁻²⁴⁰Pu (nCi)</u>		
	<u>1953</u>	<u>1962</u>	<u>1972</u>
1	30-60	10	210
3	80	130	420
4	80	140	260
5	80	140	180
6	60	70	140
7	60	80	150
17	40	90	130

^aPERSONS WITH MORE THAN 120 nCi ²³⁹⁻²⁴⁰Pu SYSTEMIC
BURDEN IN 1972.

TABLE IV

**²³⁹⁻²⁴⁰ PLUTONIUM CONTENT OF TISSUES
REMOVED FROM CASE NO. 2 IN MAY 1971**

<u>TISSUE</u>	<u>WET WEIGHT (g)</u>	<u>²³⁹⁻²⁴⁰ PLUTONIUM</u>	
		<u>dpm/g</u>	<u>pCi/g</u>
LUNG	70.85	8.48	3.85 ^a
LYMPH NODE	1.25	451.00	205.00
HAMARTOMA	0.77	7.47	3.40
RIB	20.00	3.55	1.61

^aCONTEMPORARY LEVEL FROM WEAPONS DETONATIONS IS
ABOUT 0.004 pCi/g.

TABLE V

PLUTONIUM RECIPIENTS

- EIGHTEEN PERSONS RECEIVED PLUTONIUM IN 1945 AND 1946.
- FIFTEEN OF THE 18 WERE OLDER THAN AGE 45.
- ALL BUT 2 GIVEN ^{239}Pu ; ONE RECEIVED ^{238}Pu ONLY AND ONE RECEIVED ^{238}Pu AND ^{239}Pu .
- AMOUNTS RANGED FROM ~ 0.3 TO $\sim 6 \mu\text{Ci}$ (CURRENT MAXIMUM PERMISSIBLE OCCUPATIONAL BODY BURDEN IS $0.04 \mu\text{Ci}$).
- DATA FROM GROUP PROVIDED BASIS FOR PLUTONIUM EXCRETION FUNCTIONS.
- ONE MALE RECEIVED $5.2 \mu\text{Ci } ^{238}\text{Pu}$ AND $0.12 \mu\text{Ci } ^{239}\text{Pu}$ AND $\text{PuO}_2 (\text{NO}_3)_2$. LATER HAD TOTAL GASTRECTOMY AND SPLENECTOMY. DIED 21 YEARS LATER OF CARDIOVASCULAR DISEASE.
- SOME OF ORIGINAL GROUP STILL ALIVE THREE DECADES LATER.

TABLE VI

**CURRENT STATUS OF
UNITED STATES TRANSURANIUM REGISTRY**

<u>AEC CONTRACTOR</u>	<u>WORKERS IDENTIFIED</u>	<u>RECORD RELEASES^a</u>	<u>AUTOPSY AGREEMENTS</u>	<u>AUTOPSIES PERFORMED</u>	<u>AUTOPSY REPORTS COMPLETED</u>
HANFORD	2199	2132	525	12	8
LOS ALAMOS	259	259	127	1	0
ROCKY FLATS	1504	1489	167	30	21
SAVANNAH RIVER	1559	0	0	0	0
MOUND LABORATORY	322	0	0	1	1
OAK RIDGE	0	0	0	1	1

^aHEALTH PHYSICS AND MEDICAL RECORDS

TABLE VII

**INTERNAL PLUTONIUM DEPOSITIONS EXCEEDING 25% OF THE
OCCUPATIONAL MAXIMUM PERMISSIBLE BODY BURDEN
AMONG AEC CONTRACTOR PERSONNEL DURING THE PERIOD 1957-1970**

<u>BY YEAR</u>		<u>BY ACTIVITY</u>		<u>BY % OF PERMISSIBLE BODY BURDEN</u>	
1957	12	RESEARCH	25	25 TO 50%	118
1958	6	PRODUCTION	109	50 TO 75%	35
1959	10	MAINTENANCE	17	75 TO 100%	13
1960	10	DEVELOPMENT	4	100 TO 200%	15
1961	16	HEALTH PHYSICS	18	200 TO 500%	15
1962	20	CONSTRUCTION	2	500 TO 1000%	7
1963	9	ANALYTICAL	1		
1964	29	RECOVERY	5		
1965	22	UNKNOWN	22		
1966	27				
1967	10				
1968	8	<u>BY ROUTE OF ENTRY</u>		<u>BY THERAPY EMPLOYED</u>	
1969	6	INHALATION	131	CHELATE	37
1970	3	WOUND	48	EXCISION	21
???	15	BOTH	8	BOTH	10
TOTAL	203	UNKNOWN	16	NONE	135

TABLE VIII

50TH PERCENTILE DISTRIBUTION OF PLUTONIUM IN HUMAN TISSUE (1959-1971)

	PLUTONIUM DISINTEGRATIONS PER MINUTE PER KILOGRAM									
	<u>LUNG</u>		<u>LIVER</u>		<u>LYMPH NODE</u>		<u>KIDNEY</u>		<u>BONE</u>	
<u>NONOCCUPATIONALLY EXPOSED:</u>										
LOS ALAMOS	1.3	(57) ^a	1.1	(58)	5.0	(52)	0.1	(54)	0.4	(35)
NEW MEXICO AND U. S.	1.0	(76)	0.9	(73)	4.0	(66)	0.2	(66)	0.5	(41)
COLORADO	0.5	(66)	1.7	(60)	2.0	(46)	1.4	(45)	0.9	(65)
NEW YORK	0.4	(26)	1.7	(26)	b		b		2.0	(25)
ALL POPULATIONS	0.8	(217)	1.4	(217)	3.0	(164)	0.6	(163)	0.6	(166)
<u>OCCUPATIONALLY EXPOSED:^c</u>										
LOW POTENTIAL	4.0	(44)	1.0	(41)	15.0	(42)	0.1	(42)	0.3	(25)
HIGH POTENTIAL	100.0	(15)	100.0	(15)	700.0	(14)	10.0	(13)	50.0	(11)

^aNUMBER OF SAMPLES (IN PARENTHESES)^bSAMPLES NOT REQUESTED^cDATA CANNOT BE COMPARED AS A GROUP BECAUSE OF DIFFERENCES IN TYPE AND DURATION OF EXPOSURE

TABLE IX

50TH PERCENTILE DISTRIBUTION OF PLUTONIUM IN HUMAN TISSUE (1972-1973)

	<u>PLUTONIUM DISINTEGRATIONS PER MINUTE PER KILOGRAM</u>									
	<u>LUNG</u>		<u>LIVER</u>		<u>LYMPH NODE</u>		<u>KIDNEY</u>		<u>VERTEBRAE</u>	
<u>NONOCCUPATIONALLY EXPOSED:</u>										
LOS ALAMOS	0.8	(8) ^a	1.6	(5)	35	(4)	0.2	(5)	1.6	(5)
NEW MEXICO AND U. S.	0.4	(17)	0.7	(10)	20	(15)	1.2	(10)	0.4	(16)
COLORADO	0.7	(29)	1.8	(25)	15	(22)	3.0	(25)	1.1	(25)
NEW YORK ^b	0.3	(34)	1.4	(31)	c		c		0.7	(32)
SAVANNAH RIVER	0.4	(20)	1.2	(14)	40	(6)	2.2	(11)	0.7	(12)
ALL POPULATIONS ^d	0.6	(74)	1.5	(54)	25	(47)	1.5	(51)	0.7	(58)

^a (n) NUMBER OF SAMPLES (IN PARENTHESES)

^b NEW YORK DATA FOR 1972 INCLUDE ALL DATA ANALYZED

^c SAMPLES NOT REQUESTED

^d ALL POPULATION DATA FOR 1972 DO NOT INCLUDE THE NEW YORK DATA ANALYZED DURING 1972

TABLE X

PLUTONIUM-239 IN MAN**COLORADO-NEW MEXICO (1970-1971)**

	CONCENTRATION (dpm/kg)	BURDEN (pCi)	COMPUTED BURDEN (pCi)
LUNG (1.0 kg)	0.6 (96)	0.30	0.3
LYMPH (0.015 kg)	5.0 (73)	0.03	0.6
LIVER (1.7 kg)	1.8 (88)	1.40	0.8
KIDNEY (0.3 kg)	1.1 (73)	0.10	—
BONE (5.0 kg)	0.6 (96)	1.40	0.9
		<u>3.20</u>	<u>2.6</u>

NEW YORK (1968)

LUNG (1.0 kg)	0.4 (25) ^a	0.20	0.6
LIVER (1.7 kg)	1.7 (25)	1.30	0.7
BONE (5.0 kg)	2.0 (25)	4.50	0.8
LYMPH (0.015 kg)	—	—	0.7
		<u>6.00</u>	<u>2.8</u>

^aNUMBER OF SAMPLES (IN PARENTHESES)

TABLE XI

PLUTONIUM IN MAN FROM ATMOSPHERIC NUCLEAR WEAPONS TESTS

A.	PLUTONIUM PRODUCED FROM ATMOSPHERIC WEAPONS TESTS				~ 0.4 MCi
B.	PLUTONIUM NOW ON EARTH'S SURFACE				~ 0.3 MCi
C.	PLUTONIUM IN CONTEMPORARY MAN				
		<u>dpm·kg⁻¹</u>	<u>pCi·kg⁻¹</u>	<u>kg</u>	<u>pCi-ORGAN</u>
	LUNG	0.8	0.4	1.0	0.4
	LIVER	1.4	0.6	1.7	1.0
	BONE	0.6	0.3	5.0	1.5
	OTHER	—	—	—	0.6
				TOTAL	~ 3.5pCi
D.	EACH PERSON ON EARTH ACCUMULATED ABOUT 10 ⁻¹⁷ OF B				
		<u>3.5 x 10⁻¹²Ci·PERSON⁻¹</u>			
		0.3 x 10 ⁶ Ci			
E.	ABOUT 10 ⁻⁸ OF THE EARTH'S INVENTORY FOUND ITS WAY INTO THE				
	3 x 10 ⁹ PERSONS ON EARTH				
		<u>(3 x 10⁹ PERSONS) (3.5 x 10⁻¹² Ci·PERSON⁻¹)</u>			
		0.3 x 10 ⁶ Ci

Implications with Respect to Protection Criteria [For Plutonium and other
Transuranic element in
environment],

by R. C. Thompson
Battelle, Pacific Northwest Laboratory
Richland, WA 99352

part of the AEC presentation at
EPA Plutonium Standards Hearings
Washington, D.C., December 10-11, 1974

My name is Roy Thompson. I am a staff scientist in the Biology Department
of Battelle Pacific Northwest Laboratory.

Introductory Description of Problem

Those preceding me in this presentation have tried to summarize the
facts that define the problem--present and projected--of plutonium and
other transuranic elements in our environment. It is my task to suggest
to you how these facts might be utilized in arriving at appropriate
standards for control of these transuranic elements, for the protection
of the general populace. This is a presumptuous undertaking, considering
the collective wisdom represented on the Hearing Board. I do not expect
to tell these gentlemen something they do not already know about the
philosophy or practice of radiation protection.

I've therefore chosen the opposite approach. If I cannot propose
a new concept to solve our problem, I will try, instead, to review the
problem, in its most basic aspects and hope that from this "return to
fundamentals" we may achieve some clarification of what it is we need
to do. If you feel at times that your intelligence is insulted by my

simplistic approach, I can only ask that you bear with me--I won't take very long.

In the first figure, I present what seems to me the most basic formulation of our problem. An exposure to some noxious substance results in an undesirable effect. We wish to prevent or minimize this effect. So, from our knowledge of this exposure-effect relationship, we establish a standard, which in some manner acts to control the exposure at a level that does not produce an unacceptable effect.

For the case of plutonium, we can elaborate this a bit, as shown in Figure 2. Exposure is often translated into terms of a radiation dose that is thought of as producing the effect. We know that exposure must occur via some kind of environmental pathway, and that the plutonium in the environment has its origin in some "source term." It also seems obvious that the best place for the standard to be applied for control of exposure is at the level of the source term.

Note that "dose" is enclosed in brackets, to indicate that it is really not an essential step in the process. Dose, as employed in radiation protection, is a concept in the mind of the scientist, which may or may not be useful in relating exposure to effect. Exposures, on the other hand, are real occurrences that happen to people. Effects happen to people. If a relationship between exposure and effect is known, a standard may be set to control that exposure and eliminate that effect--in total ignorance of the dose or any other knowledge of the mechanism

by which the effect is thought to be produced. I stress this point because, as will be noted later, much of the confusion in this field is due, in my opinion, to a misplaced emphasis on, and confidence in, dose. I might add that this problem does not trouble most other areas of industrial toxicology where we are too ignorant of mechanisms to be concerned with such sophistication.

Exposure

I would like to consider the individual elements of our basic problem--first exposure. I think it will help us to think in terms of three kinds, or levels, of exposure. First, there are the levels that we know can produce effects in animals. Roughly speaking, these are exposures that result in lifetime doses to bone or lung in excess of about 30 rad (1). Now, I'm speaking of dose, because it's a convenient way to lump a lot of exposure data; but, you know from Dr. Bair's presentation, that I'm lumping rats and mice and dogs and baboons--so you won't place too much confidence in my numbers. I must also specify that I am talking about average organ dose. And I must remind you that in terms of dose equivalent, as currently defined, we are talking about 300 rems to lung or 1500 rems to bone (2). These exposures, we know, have resulted in a low, but significant incidence of malignant tumors in animals.

Second, there are the exposure levels that have occurred in man. Some occupational exposures approach, or perhaps even exceed, the minimum levels that have produced effects in animals (3,4). But most occupational exposures are much lower, and the exposures from fallout plutonium, very much lower (5).

Finally, there are the exposure levels about which we should be concerned if we are to protect the general populace from plutonium effects. We don't know exactly what these levels are--or we wouldn't be here. But, I think everyone would agree that these levels must certainly be much lower than those we have considered acceptable for occupational exposure.

Effect

Now, let's try to relate these exposure categories to the effects side of our relationship. For the exposure levels studied in animals, this is relatively easy--easy because of 30 years, and many millions of dollars worth of research. We know more about the effects of plutonium in animals, I would guess, than about any other industrial pollutant.

For the human exposures, one is tempted to say that we know nothing about effects, but this is not strictly true. Effects clearly attributable to plutonium have been observed at the cellular level--histological changes (6), perhaps chromosome aberrations (7). Of course we knew before we looked that each alpha disintegration would probably kill cells. But we cannot, even qualitatively, relate these kinds of effects to health consequences. So, effectively, we know nothing about effects in humans.

But this absence of information is itself informative. It sets some upper limit to our problem. We don't quite know how to evaluate this limit, because we don't know as much about our population of plutonium-

exposed humans as we should, and because we may yet see effects, although it may be difficult to identify these effects as due to plutonium.

However, we can at least qualitatively contrast the problem with that of exposures to radium, or exposures in uranium mines, where, within much less than 25 years after these exposures commenced, lethal effects were all too obviously evident. This kind of experience we have thankfully not had with plutonium.

If we have no useful human data on plutonium effects, can we perhaps utilize other radiation effects data that are available for humans? We can certainly make such an attempt--the UNSCEAR (8) and BEIR (9) committees, among others, have done so--but we must be aware of the uncertainties involved in such an approach. In fact, we must be aware that whatever route we take to the estimation of the health consequences of plutonium in humans, we are necessarily involved in some uncertain extrapolations. We must either extrapolate from observed plutonium effects in animals to predicted effects in humans, or we must extrapolate from observed non-plutonium radiation effects in humans to predicted plutonium effects in humans. Whichever of these routes we take, we must further extrapolate from exposure levels where we have data, to the much lower exposure levels that are of primary concern in the protection of large populations.

Extrapolations

Let us look at some of the problems involved in these extrapolations. The human radiation effects data, as recently summarized in the BEIR Report, derive largely from whole-body, external exposure to penetrating

radiation, at high dose rates and high dose levels (9). We must extrapolate to a condition of very non-uniform, internal exposure of a few organs to alpha radiation, at very much lower average dose rates, but perhaps very much higher local dose rates. Such extrapolation can only be made through the medium of simplifying assumptions regarding dose and dose equivalent. These assumptions are familiar to you--I will mention only a few of the more critical ones. One must assume a relative biological effectiveness for alpha particles--a number derived from non-human studies (2). One must correct for non-uniformity of distribution--a correction presently based on animal data in the case of bone, and assumed to be insignificant in the case of lung (2) [though this latter assumption is challenged by some as grossly in error, on dosimetric grounds (10)].

For my money the extrapolation of the animal data is subject to fewer uncertainties. The whole quagmire of dose can be sidestepped. Animals can be exposed to the actual materials of concern, whether they be "hot particles" or solutions, ingested, injected, or inhaled, chronically or in a single exposure. And, effects are directly observed. They are, of course, effects in a rat or a dog--not in man. But bone and lungs of different animals are not that different, as can be demonstrated by comparative toxicity studies in several animal species. Such differences as are observed can often be explained in terms of anatomical or physiological factors and corrections can be applied for the predicted influence of these factors in man.

But one needn't choose between these two approaches. Both animal and human data should be utilized as best one can, and the encouraging fact is that the two approaches lead to similar predictions. Figure 3 shows some of these predictions as prepared for inclusion in the LMFBR Environmental Impact Statement. These cancer risk predictions are stated in terms of cancers produced per million person-rem. The range of numbers estimated from the BEIR Report data are not maximum and minimum estimates, but the range of "best" estimates derived by different procedures--all involving an assumed linear response to dose (9). The numbers derived from animal data also assume linearity of response, and are of varying quality (1). Much better animal data should be available within a few years.

The point of this comparison is not to establish a particular number, but to indicate the kind of agreement that is seen. As an indication of the agreement seen in a related area, Figure 4 shows similar numbers for radium, where we have direct data for humans to compare with experimental animal data (11). The animal data show a generally higher incidence, but there is certainly "ballpark" agreement. Recalling the numbers on Figure 3, let me say that I would have considerable confidence in that "ballpark" of plutonium numbers, if applied to individual "person" exposures in the range of a few hundred to a few thousand rem. Whether any of these numbers have any significance when applied to exposures in the range of a rem or less is far less certain, and is the major extrapolation uncertainty that we face.

Figure 5 attempts to graphically portray this problem. We are plotting average organ dose equivalent, to bone or lung, in rem, against effect in unspecified units. The heavy line at doses in excess of 100 rem represents the hard data from animal studies. The vertical lines are meant to indicate the considerable statistical uncertainty in that data. Actually, the hard data now stops at about 300 rem for lung and 1500 rem for bone, but I think that experiments currently in progress may well extend the range of observed significant effects to something approaching this level of 100 rem. Below 100 rem we have no data on plutonium toxicity nor can we expect to obtain any. We can, with confidence, assume zero effect at zero dose; and the simplest interpolation over the unknown interval is a straight line between the last data and the zero-zero point. I won't try to review the theoretical arguments that have been presented to justify such a linear interpolation, nor will I present the theoretical arguments for some kind of enhanced effectiveness at lower dose levels, or the arguments for a threshold dose below which there will be no effect. Because they are based on unsupported theory, none of these arguments are totally convincing, though some I must admit are more convincing than others.

What I do find convincing is this zero point. And, I feel quite confident that the approach to this zero point is much more likely to be asymptotic to the dose axis than to the effect axis. I said a moment ago that we can expect to obtain no experimental data in this region below a lifetime dose of about 100 rem, but we do have data of a sort in this

region that I have cross-hatched, below about 10 rem. An experiment in this dose region has been in progress for quite a few millions of years, during which time the human race has received lifetime doses averaging about 3 or 4 alpha rem per person to bone and lung, and approaching 10 rem total lifetime radiation dose (18, 20). The numbers on the viewgraph represent 70-year cumulative background doses. Though we know something about the dose, we can't evaluate the effect side of this experiment, except to say that we are the effect. What we might have become if not subjected to this radiation is a fascinating, but academic question. It is quite apparent, however, that we have not evolved in a manner to spare us from this radiation. We have not developed external shielding nor have we rejected potassium as a metabolically critical element, because of its ^{40}K content. These facts argue strongly, I think, that any precipitous change in the dose-effect relationship does not occur within the range of natural background. If there is an enhanced effect of radiation at low dose levels as represented by the upper range of curves on the viewgraph, this effect certainly cannot continue to zero dose, and I think cannot reasonably be thought to persist within the region of background dose.

Conclusions

Now, may I express just a few general conclusions. Most importantly, I would say that "As Low As Practicable" is still a very good idea-- particularly for plutonium, in view of the uncertainties we've discussed;

and particularly for population exposure standards where it may be often difficult to identify an individual benefit to balance an appreciable risk.

Exposure of total populations should be controlled at some fraction of natural background levels, because it is only in this range of exposure that I feel we have assurance of insignificant effect--an assurance based upon the survival, over past eons, of the human race.

Because the dispersal of plutonium amongst the general populace will, for many centuries at least, be quite non-uniform, I think we cannot accept limitations on person-remS as a totally adequate basis of control. While it may be expedient, and even conservative, to assume dose-effect linearity for the evaluation of risk to populations, we cannot use such an assumption as an argument for permitting individual overexposure. If 1000 person-remS are distributed evenly among 1000 persons, the probability of an eventual effect may be less than, equal to, or greater than the probability associated with the same 1000 person remS delivered to a single individual--we cannot be sure. And in no way should that uncertainty be used to justify the high individual exposure. We don't need to choose between these alternatives. We should protect the "critical individual" as well as the total population, although not necessarily to the same per-capita limits.

It seems inevitable that control must be based on environmental monitoring, since the appropriate "people-limits" will be certainly unmeasurable. This places a burden of great significance on our knowledge of food chain and inhalation pathways, which is required for translating

a people-limit to an environment-limit. Of critical importance is the assumption to be made with regard to an ultimate environmental sink for plutonium. Certainly, the longer-lived plutonium isotopes need not be assumed to remain optimally available to man for the hundreds of thousands of years before they undergo complete radioactive decay.

It may be expedient to express exposure limits for the individual in the population as some appropriate fraction of an occupational exposure limit, because the derived occupational limits will incorporate considerations of exposure pathways and the summation of dose commitments to critical organs. This "appropriate" fraction, however, cannot be an arbitrary number applicable to all radionuclides under all circumstances. It should be set primarily in relation to considerations of natural background and "practicability." Thus, the absolute value of occupational exposure limits is of little relevance to population exposure--a different set of criteria are involved on the benefit side of the risk-benefit equation.

Finally, I would just like to address, very briefly, a few specific, critical questions that have been raised with regard to the adequacy of plutonium toxicity information. Enough has probably already been said about the "hot particle" problem. This is a theoretical argument, centering around totally inadequate knowledge of the microdose-macroeffect relationship (10). Fortunately, there are experimental animal data and human experience, not dependent upon theoretical interpretation of dose, that denies the existence of any major enhanced effectiveness of such particulate exposure (13).

Concern has been expressed that the potential for genetic effects from plutonium deposited in gonads may not be adequately evaluated. Although animal experiments to prove this point have not been done, and would be very difficult, there is evidence from both animal and human data that plutonium is not concentrated in gonads, and that the dose from gonadal plutonium is therefore small--much smaller, and therefore of less concern, than the dose to lung, bone, or liver (14). Some early analyses, which seemed to indicate high levels of fallout plutonium in human gonads, have been shown to be in error (14, 15).

Lymph nodes draining the lung will probably receive the highest radiation dose from inhaled plutonium but this dose has not been considered in setting occupational exposure standards. This seems justified in light of the overwhelmingly greater incidence of cancer in bone and lung of animals that have inhaled plutonium (1).

One must be particularly concerned for the exposure of the very young members of a population, since they are usually considered to exhibit an enhanced radiosensitivity. The fact that the radiation exposure from each increment of internally deposited plutonium is spread over the whole subsequent lifetime of the exposed person, provides an automatic safety factor for the young child. Assuming uniform continuous exposure, dose rate will increase throughout the life span, and will be at its lowest value in the newborn. While gastrointestinal absorption of plutonium may be markedly increased in the infant, this condition is probably limited to a very short period following birth, a period during which the infant is relatively protected against most forms of exposure (16).

REFERENCES

1. W. J. Bair, "Toxicology of Plutonium," in Advances in Radiation Biology, Vol. 4, Academic Press, New York, pp. 255-315, 1974.
2. International Commission on Radiological Protection, "Report of Committee II on Permissible Dose for Internal Radiation," ICRP Publication 2, Pergamon Press, 1959.
3. D. M. Ross, "A Statistical Summary of United States Atomic Energy Commission Contractors' Internal Exposure Experience," in Diagnosis and Treatment of Deposited Radionuclides, Excerpta Medica Foundation, Amsterdam, pp. 427-434, 1968.
4. L. H. Hemplemann, W. H. Langham, C. R. Richmond, and G. L. Voelz, "A Twenty-seven Year Study of Selected Los Alamos Plutonium Workers," Health Physics 24, pp. 461-479, 1973.
5. B. G. Bennett, "Fallout ^{239}Pu Dose to Man," Fallout Program Quarterly Summary Report, HASL-278, 1974.
6. C. C. Lushbaugh and J. Langham, "A Dermal Lesion from Implanted Plutonium" Arch. Dermatol. 86, pp. 461-464, 1962.
7. W. F. Broman, R. W. Bistline, A. D. Bloom, and P. G. Archer, "Somatic Cell Chromosome Changes in Humans Exposed to ^{239}Pu Plutonium," Radiation Research 59, p. 206, 1974.
8. U. N. Scientific Committee on the Effects of Atomic Radiation, Ionizing Radiation; Levels and Effects, Vol. I and II, United Nations, NY, 1972.
9. U. S. National Academy of Sciences - National Research Council, The Effects on Populations of Exposure to Low Levels of Ionizing Radiation," Report of the Advisory Committee on the Biological Effects of Ionizing Radiations, Washington, DC, 1972.

10. A. R. Tamplin and T. B. Cochran, "Radiation Standards for Hot Particles," Natural Resources Defense Council, Washington, DC, 1974.
11. C. W. Mays and R. D. Lloyd, "Bone Sarcoma Incidence vs. Alpha Particle Dose," in Radiobiology of Plutonium (B. J. Stover and W. S. S. Jee, eds.) J. W. Press, Salt Lake City, pp. 409-430, 1972.
12. W. M. Lowder, J. H. Harley, and J. E. McLaughlin, "Data Needs for the Assessment of Population Dose from Background Radiation," Population Exposures (J. C. Hart, R. H. Ritchie, and B. S. Varnadore, eds.) USAEC Doc. CONF-741018, pp. 23-28 (1974).
13. W. J. Bair, C. R. Richmond, and B. W. Wachholz, "A Radiobiological Assessment of the Spatial Distribution and Radiation Dose from Inhaled Plutonium," WASH-1320, 1974.
14. C. R. Richmond and R. L. Thomas, "Plutonium and Other Actinide Elements in Gonadal Tissue of Man and Animals," Health Physics, Manuscript Submitted, 1974.
15. E. E. Campbell, M. G. Milligan, W. D. Moss, H. F. Schulte, and J. F. McInroy, "Plutonium in Autopsy Tissue," Los Alamos Scientific Lab. Doc. LA-4875, 1973.
16. M. R. Sikov and D. D. Mahlum, "Plutonium in the Developing Animals," Health Physics 22, 707-712, 1972.

FIGURE 1

FIGURE 2

PLUTONIUM-INDUCED CANCER PREDICTIONS

(cancers per million person-rem)

<u>PREDICTION BASED ON DATA FROM:</u>	<u>CANCER TYPE</u>		
	<u>LUNG</u>	<u>BONE</u>	<u>LIVER</u>
MAN (BEIR REPORT)	16-110	2-17	1-7
DOG	70	70	
RAT	60 (700)*	10	
MOUSE		20	

* ^{238}Pu

FIGURE 3

RADIUM-INDUCED BONE CANCER EXPERIENCE **(cancers per million person-rems)**

<u>DATA FROM:</u>		
MAN	$^{226,228}\text{Ra}$	5
	^{224}Ra (ADULT)	9
	^{224}Ra (JUVENILE)	15
DOG	^{226}Ra	43
	^{228}Ra	130
MICE	^{226}Ra (FEMALE)	43
	^{224}Ra (FEMALE)	120
	^{224}Ra (MALE)	7

FIGURE 4

LIFETIME BACKGROUND DOSE

FIGURE 5

Standards for the Transuranic Elements

by J. Newell Stannard
University of Rochester School of
Medicine and Dentistry
Rochester, N. Y. 14642

part of the AEC presentation at
EPA Plutonium Standards Hearings
Washington, D.C., December 10-11, 1974

The following remarks are in lieu of personal appearance for testimony which I regret previous commitments precludes. It is not heavily documented or intended to be in any way a complete summary of the many important considerations which have had or will have a significant bearing on both these important standards and their implementation. Reference will be made, however, to especially pertinent documents and summaries where needed.

While I take responsibility for this statement entirely as an individual scientist, it cannot help but be influenced by my current interests and activities for the National Council on Radiation Protection and Measurements whose Scientific Committee-34 I happen to Chair, and by interaction among its members and others in the field. Committee-34 has as its charge the recommendation to the Council of radionuclide concentration standards for both occupational and population exposure in the United States. It is actively engaged in consideration of aspects of this urgent subject but has not yet reached any conclusions on matters as detailed as standards for the transuranics.

My personal view is that some of the current recommendations regarding exposure to transuranics, now many years old, will be changed by the responsible

national and international bodies such as ICRP and NCRP and will, quickly, or even before, be modified somewhat in the Federal and probably the State codes. It is also my view that the modification will be in the direction of a reduction in derived limits, perhaps also in the primary dose standards, although the change in the latter might be relatively smaller. Decisions regarding the magnitude of any future reductions and to which nuclides they might be applied is now the prime source of debate and delay. Fortunately the extant philosophy of operating on the "as low as practicable" basis makes such delays in decision-making of less importance than they might otherwise be. However, none of the above remarks should be interpreted as implying that we have reliable scientific evidence showing significant biological damage in either workers or populations who have never been exposed above the current standards. But the future increases in numbers of potential exposees and in the quantities of transuranium nuclides potentially available makes the current emphasis on reexamination of standards a very legitimate enterprise even though the organizations concerned have and continue to maintain constant vigilance over the validity of the basic recommendations.

Basis for Present Standards

The biological basis for ICRP, NCRP and various governmental standards has been reviewed and scrutinized in depth by many, probably also at these hearings. In addition to basic documents, I especially commend the excellent historical review of maximum permissible body burdens and concentrations of plutonium by Langham and Healy⁽¹⁾, and of concentration and body burdens of the transplutonic elements by Dolphin.⁽²⁾

Yet a few points need reiteration even here. Firstly is the fact that the body burden standard for plutonium is based on a large volume of excellent

biological work first in rodents and then in the dog, and to a lesser extent primates. In addition there is a respectable amount of information on the behavior of plutonium in man which suggests no major differences in its metabolism in the human as compared to the animal species used.

It was the empirical ratio of the toxicity of plutonium relative to radium found in animal experiments which led to a reversal of proposed early standards based on energy considerations alone and to our present standards with bone as critical organ. But we have no storehouse of information on effects of plutonium in man as we do for radium (both -226 and -224). Indeed fortunately we have nothing but one or two isolated instances of effects which may be attributable to plutonium exposure. But it is frequently forgotten that we do have a storehouse of metabolic information both in man and animals and of effects in animals upon which to base a standard for plutonium.

Because of its similarities to radium the standard for plutonium in bone was therefore based on the biological information alluded to above, including the experience with radium in man. By contrast both body burden and derived standards for plutonium when organs other than bone are critical were derived quite differently viz: by calculation of that amount in the organ which will yield no more than the maximum allowable dose or dose rate to that organ. This maximum allowable dose or dose rate was derived not from experience with radium but largely from experience with external radiation sources. For most soft tissues in which the transuranics deposit (lung, liver, kidney, GI tract, et cetera) this rate is 15 rem per year for occupational exposure. Since the calculated dose to bone using the radium experience is

at least double this quantity, a factor of at least two enters into the prime standards between bone and other likely critical organs. A difference of this magnitude is well within the limits of our present knowledge.

More recently the ICRP has essentially abandoned the system of direct comparison to radium and substituted calculation of the dose to endosteal cells as the basic procedure for bone. Since this would allow a dose rate of only 15 rem per year for occupational exposure situations, the prime standard might be expected to be reduced by a factor of at least two by this change. But, as of the present writing, the complexities of calculating dose to endosteal cells from alpha emitters have made the ICRP decide to keep the old approach for alpha-emitting bone seekers.

The transplutonic elements such as Am, Cm, Cf, Es, Fm, et cetera, seemed by and large to behave enough like plutonium to be handled in much the same way, i.e., bone as critical organ and the comparison to radium retained. Or so it would seem from the official ICRP and NCRP publications on internal emitters. But Dolphin makes no mention of direct comparison to radium in deriving body burden figures for the trans-plutonics in his recent survey.⁽²⁾ Instead he cites the basic dose standards of 5, 15, and 30 rem/year, the latter presumably applying to bone and calculates therefrom. The NCRP in its most recent report on basic radiation standards⁽³⁾ does not make specific mention of any dose rate for bone, although it might be presumed to be included in "other organs...." at 15 rems per year. In fact, the NCRP Report specifically defers judgment (paragraph 202, page 77, reference 3) on bone doses to await recommendations from its committees on internal emitters; which committees have yet to issue judgements beyond the one contained in the referenced paragraph above.

Thus the official primary standards for the transuranium elements remain, despite much ferment, pretty much as they were in 1959 except for certain additions and modifications of derived figures. But let it not be forgotten that changes would have been made quickly and certainly if the growing storehouse of biological information had begun to indicate any serious flaws in the basic information used originally. The situation would not have remained static very long.

The Current Reexamination

A. Changes in Models and Metabolic Parameters

Over the years since 1959 much new biological information has been gathered and formulated. In addition to different values for some of the metabolic parameters, we have a new and more versatile lung model, a new GI tract model, much more about standard man, et cetera. Only a small fraction of this has found its way into official use, although it has been generally drawn upon for almost every other purpose. In my paper at Los Alamos⁽⁴⁾ I presented "old" and potential "new" figures for Maximum Permissible Annual Intake by ingestion or inhalation of plutonium-238 and 239 and the derived values for air concentrations. For the "new" figures I selected likely - but totally unofficial - metabolic values and models. The changes in derived values were almost all within a factor of ten, some increasing and some decreasing. While it must be admitted that other choices - especially of aerosol characteristics might have made larger differences, the prediction that the newer models and metabolic data would lead to drastic reduction in standards, has not been realized for plutonium.

The accumulated information on toxicity ratios between trans-uranics and radium in the monumental dog experiment at the University of Utah indicates rather remarkable similarities to the ratios derived from shorter-lived rodents. However, the current ratio of five (expressed as the "N" factor in ICRP and NCRP formulations) is certainly none too high. Some of the dog experiments indicate factors more like eight or ten. This combined with Marshall's indication that slower movement of the nuclides in bone structure of man might lead to a higher toxicity in man indicates a potential for some reduction in basic derived standards on this basis when the responsible bodies complete their sifting of the newer (and still accumulating) data. Newer work with the transplutronics has been ably summarized by Durbin.⁽⁵⁾ Her survey plus currently appearing papers suggest larger accumulations of some of these nuclides in soft-tissues and correspondingly less in bone than in earlier work. If the critical organ dose calculation mode is used, these changes would seem unlikely to lower standards. If the risk estimate approach is employed the situation might be different. Results so far suggest that the transplutronics may be about equally effective with plutonium (on activity basis) in inducing bone tumors. Although the experiments are far from complete, especially those with ^{249}Cf and ^{252}Cf , if this is indeed true then these nuclides by their greater mobility compared to plutonium may present greater risks to soft tissue. The net result might well be a reduction of derived standards. But current data do not suggest a large reduction on this basis if it does occur.

B. Cancer Incidence Relationship

In an important paper at the Fifth International Congress of Radiation Research, Dr. Roy C. Thompson presented two summary graphs which put together in one place most of the animal data on bone and lung cancer incidence from long-term exposure to plutonium. These present the incidence rates as a function of calculated radiation dose and are fraught with all of the pitfalls and difficulties of determining the true radiation dose which caused the cancer. These are especially serious difficulties for an internal emitter like plutonium. Also, the combining of data from many experiments employing many species into a single summary graph is a very gross way to express relationships. Yet, the broad outlines of cancer incidence rates are discernable since the data are reasonably coherent.

As Dr. Thompson will no doubt point out in his own testimony at these hearings, the composite figures for lung cancer show a small but apparently significant increase at a cumulative mean dose as low as 30 rads. The composite portrayed for plutonium-induced osteosarcoma shows several points above the abscissa (i.e., above zero increase in incidence) below 10 rads, although the mean figures are not above zero until a cumulative rad dose of slightly over 20 rads. If one considers that at 15 rem per year and 50 years of exposure, the total allowable cumulative occupational dose to the lung could be 750 rem while 30 rad is only 300 rem, one wonders if the safety factor in this standard is as large as desirable.

Fortunately the occupational limits for air concentration have been arranged so that the limiting dosage rate of 15 rem per year is reached only in the 50th year of exposure. Thus 750 rem would never be reached under this regimen. But if this annual rate were to be applied to single or a series of short exposures, the total doses might come uncomfortably close to those associated with detectable increase in cancer incidence.

A similar argument might be made for bone, although the practice of tying its limits directly to radium exposures in humans and the even greater difficulties of calculating radiation dose to bone for alpha emitters make it more likely to be specious.

Application of the dose commitment concept for single or short bursts of exposure has probably helped to prevent unacceptably large accumulations in man. But it seems reasonable to expect that the presence of finite cancer incidence at calculated doses as low as 10 rads or 100 rems will exert pressure toward lowering of the present official ICRP, NCRP, and other permissible exposures and intakes.

C. Non-uniform Distribution

It is amply clear that the transuranic nuclides show a marked tendency to form aggregates in vitro and in vivo, especially in the chemical and physical states of most likely exposure. Thus, the maximal radiation doses in some areas may far exceed the average dose to the organ conventionally calculated. A special case of this well-known non-uniform distribution phenomenon is the "hot particle"

problem emphasized in extenso by Tamplin and Cochran⁽⁶⁾ and the Natural Resources Defense Council, Inc.

While the bulk of current biological information does not support the notion of any special carcinogenic effectiveness of "hot particles" in the lung, there are many critical pieces of information which are not available. Nor can they be expected to appear until more is known of the mechanisms of carcinogenesis itself. While I disagree with the 115,000 factor by which Tamplin and Cochran suggest the standards should be lowered, I do believe the "hot particle" discussion will tend, along with other factors noted in the testimony, to drive standards down, even if the scientific basis is not clearly evident from experimental work.

There is one aspect of the "hot particle" problem, however, which I feel needs special emphasis. As discussed earlier, we are dealing with actual incidence data for plutonium, and to a lesser extent for the transplutonics. The animal exposures undoubtedly involved non-uniform distributions, even "hot particles." The tissue responses measured have thus largely resulted from non-uniform sources. If maximal doses are utilized for the incidence curves which take into account the non-uniformity of distribution, instead of the average dose to the whole tissue, then the apparent radiosensitivity, i.e., the dose to produce the effect, would be correspondingly lowered. The "effective dose" would be much higher by this convention. Then standards should be keyed to those doses

rather than to the calculated average dose. Thus, in a sense the dosage calculations have led us astray. We should be sure we do not forget that direct biological information should always take priority. The great strength of the data for radium in man and of the derivations of many of the standards for internal emitters therefrom, lies in the fact that activity and effect can be correlated without obligatorily going through the step of calculating a radiation dose. This is also true in our extensive animal data with transuranics. Thus, I personally will await a clear demonstration of a special effectiveness of hot particles in the lung before accepting a drastic reduction based on dosage calculations alone.

D. Population Exposures

The enormous impetus of the "fall-out controversy" and the subsequent UNSCEAR and BEAR committee deliberations led first the Federal Radiation Council⁽⁷⁾ then the ICRP⁽⁸⁾ and NCRP⁽³⁾ to set down specific limits for radiation exposure of the general population both on the average and to an individual. These were applied to radioisotope releases in the Federal Code of Regulations essentially by scaling down the occupational figure by factors of 1/10 or 1/30 (the iodines and particulates have received separate and much more stringent treatment). Such a scaling down process introduces many dilemmas for the transuranics as described in a paper I gave at the Los Alamos Plutonium Symposium⁽⁴⁾ so long as one continues to use the calculation of dose and critical organ convention. An alternative, taken up at length in the report of the BEIR

Committee⁽⁹⁾ and in the 1972 UNSCEAR report⁽¹⁰⁾ is to attempt to assay total or specific health effects. Estimates of risk and decisions regarding acceptable risk take the place of comparing a given exposure with a maximum allowable dose or dose rate. This approach has its attractions, but it must lean heavily on estimates of risk derived from a few special human populations.

While there are an increasing number of papers appearing in which the direct estimation of total or some specific health effect is employed in connection with aspects of the nuclear fuel cycle (e.g., Barr⁽¹¹⁾), neither ICRP or NCRP has yet officially embraced this alternative. What effect if any such a change if it does occur will have on basic and derived standards is difficult to predict, particularly since opinions are still widely divergent on details of risk assessment. Nonetheless, it seems unlikely that the figures for total health effects or even the most likely specific effects from the presently accepted dose rates or total doses will be accepted if the technology will permit lower risks without undue loss of benefits. In any event, the use of a population exposure figure for the transuranic group of nuclides based on a simple scaling factor from occupational levels is probably untenable. Thus change in these, not necessarily drastic, but definite seems very likely, and it will probably be downward.

E. The Lymph Node Problem

A final example of how dosage calculations, unmodified by biomedical information, can lead to dilemmas and claims of overexposure is seen in the deposition of inhaled nuclides in pulmonary, particularly bronchiopulmonary, lymph nodes. This was discussed at some

length in my paper at Los Alamos in May 1974.⁽⁴⁾ Every toxicologist knows that insoluble materials leaving the lung deposit, sometimes in very high concentrations, in the lymph nodes along the channels of lymphatic drainage of the lung. This is fully true for the transuranic elements and very high radiation doses can be calculated as resulting from the amounts deposited. But there has yet to be found a primary cancer associated with or resulting from these deposits. The nodes may be completely fibrosed and essentially non-functional, but no cancer. Lung and bone cancers appear in the same animals and at much lower calculated doses.

Because of the strong concentration of nuclide in lymph nodes, dosage rate alone would make them critical organ after inhalation exposure in nearly all cases of inhalation of insoluble compounds and derived standards would be automatically reduced. But this has not been done because of the apparent radioresistance of this tissue in terms of local cancer induction. Perhaps this should be recognized and the dilemma of not allowing an "overdose" solved for the health physicist by providing a more realistic maximum permissible annual dose rate for this tissue. But I would not personally wish to see such a move allow doses which would produce significant fibrosis or other definite damage, and these would probably not be so far above current dose levels that the dilemma would cease to exist. On this basis, I feel that a proper accounting should be made of the lymph node activities and biological changes associated therewith rather than simply pass the problem by as too difficult to

handle. I can envision such a move as resulting in some lowering of current standards under certain conditions.

Conclusions

This statement is much more speculative in some respects than I usually allow myself to engage in. But since my subject concerns the probable future trends of standards for the transuranic elements, I feel it necessary to indulge in many personal prognostications. These in summary predict downward alterations of present primary and derived standards but of magnitudes which would probably not strain current technology unduly. But on the other hand, it must be stressed that we still need some very important biomedical information which only time, patience, and careful scientific work can supply and standards will probably continue for some time to involve many assumptions which must rest on incomplete information.

REFERENCES

- (1) Langham, W. H. and J. Healy, Chapter 12 in Vol. 36 of the Handbook of Experimental Pharmacology series, entitled "Uranium, Plutonium, and the Transplutonic Elements." H. C. Hodge, J. N. Stannard and J. B. Hursh, Ed., Springer-Verlag, New York, Heidelberg, Berlin, 1973.
- (2) Dolphin, G. W., Ibid. Chapter 19.
- (3) National Council on Radiation Protection and Measurements, Report No. 39, "Basic Radiation Protection Criteria." NCRP Publications, Washington, D.C., 1971.
- (4) Stannard, J.N., "The Concepts of Critical Organ and Radiation Dose as Applied to Plutonium," presented at Los Alamos Biology Symposium, May 1974. To be published in Health Physics.
- (5) Durbin, P.W., Chapter 18 in Vol. 36 of the Handbook of Experiment Pharmacology series, entitled "Uranium, Plutonium, and the Transplutonic Elements." H. C. Hodge, J. N. Stannard and J. B. Hursh, Ed., Springer-Verlag, New York, Heidelberg, Berlin, 1973.
- (6) Tamplin, A. R. and Cochran, T. B., "Radiation Standards for Hot Particles, A Report on the Inadequacy of Existing Radiation Protection Standards Related to Internal Exposure of Man to Insoluble Particles of Plutonium and Other Alpha-Emitting Hot Particles," Natural Resources Defense Council, Washington, D.C., February 14, 1974.
- (7) Federal Radiation Council (FRC), Report No. 1, 1960, (U.S. Department of Commerce).

- (8) International Commission on Radiological Protection (ICRP), 1966,
Publication No. 9 (Pergamon Press, Oxford).
- (9) National Academy of Sciences-National Research Council (NAS-NRC),
Division of Medical Sciences, 1972, Report on "The Effects on
Populations of Exposure to Low Levels of Ionizing Radiation,"
(NAS-NRC, Washington, D.C.).
- (10) United Nations Scientific Committee on "The Effects of Atomic Radiation
(UNSCEAR), 1972, Vol. II: Effects, (United Nations, N.Y.).
- (11) Barr, N. F., "Quantitative Health Estimates of Transuranic Releases,"
Division of Biomedical and Environmental Research, paper presented
at American Nuclear Society meeting, Washington, D.C., October 1974.

UNITED STATES
ATOMIC ENERGY COMMISSION
WASHINGTON, D.C. 20545

AUG 16 1974

Clarence C. Lushbaugh, M.D.
109 Darwin Lane
Oak Ridge, Tennessee 37830

Dear Dr. Lushbaugh:

The Atomic Energy Commission has received from the Natural Resources Defense Council a petition to establish special standards for alpha-emitting radionuclides in insoluble, particulate form. A copy of this petition, and a supporting statement submitted with the petition, are enclosed. On pages 26 through 29 of the supporting statement a quotation from an article by C. C. Lushbaugh and J. Langham, published in the Archives of Dermatology in 1962, is used as the basis for the following conclusions:

- (1) A single Pu-239 particle is capable of inducing cancer;
- (2) The risk of cancer may be greater than 1/1000 per particle.

The Commission's standards for exposure to insoluble, airborne plutonium and other alpha-emitters are based on a permissible lung burden of 16 nCi, which could consist of many thousands of particles deposited in the lung, the actual number depending upon the size of the particle. For example, 16 nCi is equivalent to 2×10^6 particles of 0.3-micron diameter. The risk associated with such a large number of particles would obviously be unacceptable if the risk per particle is as great as concluded in the supporting statement.

The Commission is currently conducting an evaluation of its standards for airborne, alpha-emitting radionuclides in insoluble form, and great importance is attached to the risk which may be associated with relatively small numbers of alpha-emitting particles in the lung. In this connection we would appreciate receiving from you a statement as to whether your findings in the case reported in the Archives of Dermatology do in fact support the two conclusions drawn in the supporting statement, as listed above.

Sincerely yours,

A handwritten signature in dark ink, appearing to read "Lester Rogers", is written over the typed name.

Lester Rogers
Director of Regulatory Standards

September 10, 1974

Mr. Lester Rogers
Director of Regulatory
Standards
U.S. Atomic Energy Commission
Washington, D.C. 20545

Dear Mr. Rogers:

In reference to your letter of August 16, 1974, I should point out that earlier this year I worked with Dr. Bruce Wachholz of Bio-medical Programs, DEER, Germantown Headquarters, on the initial stages of a document recently numbered WASH-1320; entitled, A Radiobiological Assessment of the Spatial Distribution of Radiation Dose from Inhaled Plutonium Particles; and authored by W. Bair, C. Richmond, and B. Wachholz. Although I have not seen this paper in its final form as it is at this moment still being printed, I am certain that it contains an attempt to answer the question of whether or not Mrs. Langham's and my article in Archives of Dermatology (1962) supports the contention of Dr. Tamplin and Mr. Cochran that a single particle of Pu-239 is capable of inducing cancer and that the risk of cancer from such a particle is 1 per 1000. We believe that these conclusions cannot be derived from the histopathologic observations we reported in this case report nor in the other cases we subsequently published along with it in the Annals of the New York Academy of Science.

In the petition from the Natural Resources Defense Council to which you refer, one can see that the authors apparently do not know the difference between a precancerous cellular change and a cancer. While it is true that the term "precancerous change" contains the implication that a cancer follows it, this is not always the case because precancerous changes are reversible and reparable. In fact when a lesion showing precancerous changes is removed surgically, the surgeon knows from this diagnostic impression given him by the pathologist that the lesion he removed is not a cancer and that he does not have to worry about it further. My object in using

Mr. Lester Rogers

-2-

September 10, 1974

the term "precancerous" to describe the cytologic appearance of some of the epithelial cell nuclei around the plutonium particles in the skin of the case in Arch. Dermatol. was to point out that in spite of the amazingly huge dose of alpha radiation over a period longer than 4 years a cancer had not developed and that one could at most only call the changes pre-cancerous. In reviewing this case in the Annals of the New York Academy article, we attempted to show that the strictly localized injury caused by the plutonium particles was developing in such a fashion (like a pimple) that the particles would have been shed in time along with a small amount of pus-like material as the pimple "ripened" and drained spontaneously. Dr. Tamplin in his arguments assumes that fibrosarcomas in rat skin are equate-able with the minimal changes we described in the skin of this man. Of course, they are not. The statement that it is "clear" on the basis of this one human case that plutonium can cause skin cancer in man is false. If this case and others like it show something of radiobiologic importance, they show only that the development of cancer from plutonium exposures of human tissues must be much more difficult to obtain than cancers in rodent tissues, since no human cancers have ever been seen or reported following plutonium exposure of human beings. Logically, if there is no observed plutonium-induced human cancer case, the one per thousand per particle level of cancer risk for plutonium exposure has no basis in fact and amounts to only a conjecture on the part of the authors of the NRDC petition.

C. C. Lushbaugh, M.D., Ph.D

A Critique of the Tamplin-Cochran Proposal
for Revision of the Current Plutonium Exposure Standards

Roy E. Albert, M.D.
Professor and Vice Chairman
Institute of Environmental Medicine
New York University Medical Center

March 25, 1974

Summary

Largely on the basis of rat skin tumor experiments, Tamplin and Cochran propose that a single radioactive particle in the lung which delivers a local dose of more than 1000 rem per year will produce focal tissue damage and that this focal damage per se confers a risk of lung cancer of one in two thousand.

A review of current knowledge about the relationship of tissue damage to the induction of cancer does not support the contention that tissue damage is a proximate cause of cancer; rather that tissue damage represents a parallel toxic action of carcinogens which, to some extent, may enhance the development of tumors produced by carcinogens. Since the Tamplin-Cochran proposal is based almost wholly on radiation tumor studies of the rat skin hair follicles, the decisive argument against this proposal is the evidence that focal alpha irradiation of localized regions on the hair follicle, in a pattern similar to that from a plutonium particle, is non-tumorigenic.

The Tamplin-Cochran Proposal (1)

The authors point out that the current ICRP occupational exposure standard for insoluble plutonium in the air is

4×10^{-11} uCi/ml. This is the calculated level of atmospheric contamination that would lead to a maximum permissible lung burden (MPLB) of 0.016 uCi and would be associated with a maximum permissible lung dose of 15 rem/yr when the radiation dose is averaged over the entire lung.

They point out that the dose is not delivered uniformly to the entire lung:

"It would take 53,000 particles...(1 u in diameter, 0.28 pCi)...to reach the MPLB of 0.016 uCi which results in 15 rem/yr to the entire (1000 g) lung. However...these particles would irradiate only 3.4 g of this 1000 g to the lung, but at a dose rate of 4000 rem/yr...these particles result in an intense but highly localized irradiation. A fundamental question is, then: is this intense but localized irradiation more or less carcinogenic than uniform irradiation?" (ref. 1, pg. 17).

The Tamplin-Cochran approach to the risk assessment from hot particles is based on the Geesaman Hypothesis (2, 3) which in turn is based almost wholly on the radiation skin experiments of Albert and co-workers. The interpretation placed on these experiments by Tamplin and Cochran and the rationale for their proposed standard is described by the following excerpts from their report (1).

"A high incidence of cancer was observed after intense local doses of radiation, and the carcinogenesis was proportional to the damage or disordering of a critical architectural unit of the tissue, the hair follicles." (ref. 1, pg. 23).

"Certainly a reasonable interpretation of these experimental results is: when a critical architectural unit of a tissue (e.g., a hair follicle) is irradiated at a sufficiently high dosage, the chance of it becoming cancerous is approximately

10^{-3} to 10^{-4} . This has become known as the Geesaman hypothesis." (ref. 1, pg. 26).

"Geesaman indicates that the tissue repair time in the lung is of the order of one year. It therefore seems appropriate, but not necessarily conservative, to accept as guidance that this enhanced cancer risk occurs when particles irradiate the surrounding lung tissue at a dose rate of 1000 rem/yr or more." (ref. 1, pg. 33).

"As seen from Table IV, using Geesaman's lung model, a particle with an alpha activity between 0.02 pCi and 0.14 pCi is required to give a dose of 1000 rem/yr to irradiated lung tissue. For purposes of establishing a maximum permissible lung particle burden we will use 0.07 pCi from long half-lived (greater than one year) isotopes as the limiting alpha activity to qualify as a hot particle." (ref. 1, pg. 34).

"The existing standards for uniform radiation exposure of the whole body or lung can be used as the basis for establishing particle exposure standards by equating the risk of cancer induction between the two types of exposure (uniform vs. grossly non-uniform). The most recent assessment of the risk associated with uniform irradiation of man was performed by the NAS-NRC Advisory Committee on the Biological Effects of Radiation. Their report, published in 1972, is referred to as the BEIR Report.

The existing occupational exposure standard for uniform whole body irradiation is 5 rem/yr and for the lung, 15 rem/yr. The BEIR Report estimates that exposure of the whole body of an individual to 5 rem/yr would lead to a cancer risk between 4.5×10^{-4} and 2.3×10^{-3} /yr. Their best estimate is 10^{-3} /yr." (ref. 1, pgs. 41-42).

"It is recommended here that the best estimate of the effects of uniform exposure by the BEIR Committee be used together with a risk of cancer induction of 1/2000 per hot particle in determining the MPLPB for insoluble alpha-emitting radionuclides in hot particles. This is a somewhat arbitrary compromise and is not the most conservative value that could be recommended. Thus, the recommended MPLPB for occupational exposure from hot particles of alpha-emitting radionuclides in the deep respiratory zone is 2 particles. This corresponds to a MPLB of 0.14 pCi and represents a reduction of 115,000 in the existing MPLB." (ref. 1, pgs. 43-44).

Differences Between the Tamplin-Cochran Proposal and the Geesaman Hypothesis

Whether intentional or not there is a subtle but important difference between Geesaman's hypothesis and the Tamplin-Cochran proposal. The pertinent portion of Geesaman's conclusion is the following:

"Tissue injury and disturbance are a primary consequence of intense radiation insult, and are observed in association with carcinogenesis. Albert has exhibited a simple proportionality between skin carcinomas and atrophied hair follicles. No general description of precarcinogenic injury exists, but in a crude sense the available observations are compatible with the idea of an injury-mediated carcinogenesis. Cancer is a frequent instability of tissue. Since tissue is more than an aggregate of cells, and has a structural and functional unity of its own, it would not be surprising if some disrupted local integrity, a disturbed ordering, comprises a primary pathway of carcinogenesis. The induction of sarcomas with inert discs of Mylar, cellophane, Teflon and Millipore (Brues et al.) is indicative that such a mechanism exists." (ref. 3, pgs. 6-7).

Geesaman is saying that "...some disrupted local integrity, a disturbed ordering, comprises a primary pathway of carcinogenesis." This means that it is not the radiation but rather the tissue damage which is the proximate cause of cancer.

Tamplin and Cochran blur the issue by saying: "Certainly a reasonable interpretation of these experimental results is: when a critical architectural unit of a tissue (e.g., a hair follicle) is irradiated at a sufficiently high dosage, the chance of it becoming cancerous is approximately 10^{-3} to 10^{-4} . This has become known as the Geesaman hypothesis." Taken literally, Tamplin and Cochran do not require that tissue damage be produced, only that the "...critical architectural unit is irradiated at a sufficiently high dosage..."

The Tamplin-Cochran proposal is evaluated here from two standpoints: (1) the Geesaman hypothesis, i.e., does tissue

damage, per se cause cancer? (2) The Tamplin-Cochran interpretation of the Geesaman hypothesis, i.e., would intense irradiation of a "critical architectural unit" cause tumors, regardless of whether damage was produced?

The Theory that Tissue Damage Causes Cancer

The Geesaman hypothesis, on which the Tamplin-Cochran proposal is based, revives one of the oldest theories of cancer, namely that the cause of cancer is chronic tissue damage. This is the chronic irritation theory propounded by Virchow in 1863. As reviewed by Oberling (4), the theory was in vogue for about 50 years. It stemmed from the early clinical observations that cancer rarely appears in healthy tissue and is almost always preceded by chronic inflammatory conditions such as scars, ulcerations or fistulas. Post-mortem observations in this era suggested that the same association applies to internal organs.

Virchow pointed out that every injury of tissues is followed by a state of irritation in which the cells are stimulated to multiply in order that the damage may be repaired. If the noxious influence persists, the irritation persists with it and the proliferation grows more and more excessive and irregular. Virchow argued that if such a condition persists year after year, cancer will occur.

The Virchow theory claimed that chronic irritation was the

sole and non-specific cause of cancer, i.e., cancer was the secondary outcome of a whole series of conditions widely differing from one another and possessing no features in common except chronic damage.

As pointed out in a review by Berenblum (5), Virchow's theory was demolished by experiments beginning in 1918 which showed that cancer can be produced by very potent substances that vary widely in their capacity to cause damage whereas many agents which cause damage do not cause cancer. Furthermore, there are many conditions in humans in which tissue disorganization and damage is a characteristic feature where no association with cancer has been demonstrated, e.g., tuberculosis and silicosis of the lung and traumatic injuries associated with war wounds that have occurred by the millions during this century. The focus of cancer research long ago shifted away from tissue damage as a cause of cancer. Nevertheless, the frequent association between tissue damage and cancer remains valid for many types of human and experimental cancer but there are other types of cancer where no association exists.

The most probable reason for the association is that virtually all carcinogens are highly toxic agents. The only outstanding exceptions are the oncogenic RNA viruses. There are many examples to show that an appreciable yield of tumors

can be produced only at carcinogen doses which cause a large amount of cell death in the target tissue. This can be seen for example in relating the doses of radiation required to produce tumors in mouse skin (6) to those which cause cell death (7). The action of chemical carcinogens and ionizing radiation in producing the parallel effects of cell death and neoplastic cell transformation is also evident in tissue culture studies (8, 9).

There are various forms of damage produced by carcinogens which depends mainly on the target tissue. For example, application of a chemical carcinogen or ionizing radiation to the surface epidermis of the skin or the bronchial mucosa results in cell loss followed by a hyperplastic response in which the number of epithelial cells is much increased for long period of time. In the bronchial epithelium the hyperplasia is also accompanied by squamous metaplasia of mucosal cells. Another form of tissue damage can be produced by inhaled radioactive particles which deposit in the alveoli; such particles can produce fibrotic damage. Atrophy is still another form of tissue damage as illustrated by the damage to the hair follicles in the irradiated rat skin.

Although tissue damage cannot be assigned a primary causal role in cancer induction, there are various ways in which tissue

damage could contribute to tumor formation. One possibility is that the killing of a portion of cells in the target tissue has the consequence of stimulating the survivors to proliferate in order to restore the cell population. There is evidence that neoplastic transformation does not become fixed unless cell division occurs within a relatively short period after carcinogen exposure. This is true for ionizing radiation (10) and viruses (11). The likelihood of producing transformed cells could thus be increased by provoking cell division particularly in a tissue which normally has a low rate of proliferation.

There is evidence that neoplastically transformed cells in physical contact with normal non-transformed cells are inhibited from proliferating (12). Tissue injury could free transformed cells from this type of growth restraint.

It is possible that an area of tissue, heavily damaged by a carcinogen, particularly with scar formation, would constitute an immunologically privileged site and thus interfere with important defense mechanism against neoplastically transformed cells (13).

There are several other speculative ways in which damage could contribute to tumor formation which can be mentioned: cell damage might interfere with repair of carcinogen-induced DNA damage; dedifferentiation of surviving cells in a heavily damaged organ

could make them more susceptible to infection by oncogenic viruses, as with the irradiated thymus (14); in the case of chronic carcinogen exposure the increased cell proliferation induced by tissue damage could make cells more susceptible to the transforming action of a carcinogen.

Although all of the above mechanisms for the enhancement of carcinogen effects by various forms of tissue damage have some basis in scientific evidence, the degree of importance as contributing factors has not been established.

The Effect of a "Hot Particle" Type of Irradiation Tumor Induction in the Rat Skin

The Tamplin-Cochran proposal uses mainly the results obtained by the Albert-Burns radiation skin experiments to infer alpha-particle risks in the lung. Hence, the critical test of their hypothesis is the question of whether a hot particle pattern of alpha irradiation of the skin could produce tumors.

Two approaches were used in skin experiments. The first approach determined whether isolated areas of irradiated skin gave the same yield of tumors per unit area as large-area skin irradiations. The focal irradiation pattern was produced by use of sieve plates. Low LET radiation, such as electrons (15) and soft X-rays (16) showed pronounced suppression of tumor

formation with sieve irradiation. A higher LET radiation (protons) did not show a protective effect of sieve radiation (17).

The second approach involved the use of irradiations at different depths in skin. The results of electron irradiations with different penetrations on the induction of tumors and atrophic follicles suggests the existence of target cells at a depth of about 0.3 mm in the skin corresponding to the lower end of the resting hair follicle (18). This critical depth remains constant even when the skin is irradiated with the hair in the growing phase, i.e., when the follicles extend to a depth of 0.8 mm (19). There is a quantitative association between the incidence of tumors and atrophic follicle for various types of ionizing radiation, various spatial distributions of dose within the skin and for different phases of hair growth (20). In our view, a plausible explanation for the experimental results is that each follicle has a population of stem cells at a depth of 0.3 mm that are concerned with the production of sebaceous cells and hair. These stem cells constitute the most sensitive tumorigenic cell population to ionizing radiation in the rat skin. The tumors are mainly of hair follicle origin (21). Neoplastic transformation of a significant number of these target cells requires large radiation doses which in turn kills most of the

target cells and thus causes follicle atrophy.

Since the radiation from alphas has a range of only about 45 microns from a plutonium particle, the effect of focal irradiation at different levels of the hair follicle is a crucial test of the Tamplin-Cochran proposal. Alpha and proton irradiations that extend from the skin surface to a depth of about 0.15 mm do not produce tumors (22). This result, however, is consistent with the existence of a target cell population at a depth of about 0.3 mm. However, selective irradiation of the lower end of the hair follicle at a depth of 0.3 mm by use of the Bragg peak from an alpha beam did not produce tumors or atrophic follicles unless there was substantial irradiation of the entire follicle (22). This observation suggests that even though the critical cell population is located at 0.3 mm, that there are recovery mechanisms that block tumorigenesis when only part of the "critical architectural unit of tissue" is irradiated. What these recovery processes might be is not understood. Nevertheless, this result does not support the contention that a single plutonium particle positioned next to a "critical architectural unit" such as the hair follicle, will produce a tumorigenic risk of the magnitude assumed by Tamplin and Cochran.

It might be argued that since particles can move about in

the lung, it is appropriate to consider the effects of a single plutonium particle in the skin which moves up and down and irradiates the entire follicle. However, under these circumstances it should be necessary to consider the important factor of temporal recovery from the tumorigenic action of ionizing radiation which has been shown by split dose experiments to be very large for low LET radiation (23); preliminary data from an ongoing split dose experiment suggests that recovery from proton radiation is also very substantial (24). Data applicable to estimation of the recovery rates from exposure to a moving radioactive particle are not available. Geesaman's estimate of a one year recovery time for radiation effects on the lung is mere speculation.

References

1. Tamplin, A. R. and T. B. Cochran. Radiation Standards for Hot Particles. A Report on the Inadequacy of Existing Radiation Protection Standards Related to Internal Exposure of Man to Insoluble Particles of Plutonium and Other Alpha-Emitting Hot Particles. Natural Resources Defense Council, Washington, D.C., 1974.
2. Geesaman, D. P. An Analysis of the Carcinogenic Risk from an Insoluble Alpha-Emitting Aerosol Deposited in Deep Respiratory Tissue. Lawrence Radiation Laboratory, University of California, Livermore, TID-4500, UC-48, Feb. 1968.
3. Geesaman, D. P. An Analysis of Carcinogenic Risk from an Insoluble Alpha-Emitting Aerosol Deposited in Deep Respiratory Tissue: Addendum. Lawrence Radiation Laboratory, University of California, Livermore, TID-4500, UC-48, Oct. 1968.
4. Oberling, C. Riddle of Cancer, Yale University Press, New Haven, 1952.
5. Berenblum, I. Irritation and Carcinogenesis. Arch. Path. 38:233-244, (1944).

6. Albert, R. E., F. J. Burns and P. Bennett. Radiation-Induced Hair-Follicle Damage and Tumor Formation in Mouse and Rat Skin. J. Natl. Cancer Inst. 49:1131-1137, (1972).
7. Withers, H. R. The Dose-survival Relationship for Irradiation of Epithelial Cells of Mouse Skin. Brit. J. Radiol. 40:187-194, (1967).
8. DiPaolo, J. A., K. Takano and N. C. Popescu. Quantitation of Chemically Induced Neoplastic Transformation of BALB/3T3 Cloned Cell Lines. Cancer Res. 32:2686-2695, (1972).
9. Borek, C. In Vitro Cell Transformation by X-Rays. Radiat. Res., (1973). (Abst.)
10. Sachs, L. An Analysis of the Mechanism of Carcinogenesis by Polyoma Virus, Hydrocarbons, and X-Irradiation. In: Moleculare Biologie des Maligner Wachstums, Springer Verlag. New York, 1965.
11. Todaro, G. J. and H. Green. Cell Growth and the Initiation of Transformation by SV40. Proc. Natl. Acad. Sci., U.S. 55:302-308, (1966).

12. Sivak, A. and B. L. Van Duuren. A Cell Culture System for the Assessment of Tumor-Promoting Activity. J. Natl. Cancer Inst. 44:1091-1097, (1970).
13. Bates, R. R. and R. T. Prehn. Role of the Fibrous Capsule in Carcinogenesis by Plastic Film. Nature 205:303-304, (1965).
14. Kaplan, H. S. On the Etiology and Pathogenesis of the Leukemias: A Review. Cancer Res. 14:535-548, (1954).
15. Albert, R. E., F. J. Burns and R. D. Heimbach. Skin Damage and Tumor Formation from Grid and Sieve Patterns of Electron and Beta Radiation in the Rat. Radiat. Res. 30:525-540, (1967).
16. Unpublished data.
17. Burns, F. J., R. E. Albert, P. Bennett and I. P. Sinclair. Tumor Incidence in Rat Skin After Proton Irradiation in a Sieve Pattern. Radiat. Res. 50:181-190, (1972).
18. Albert, R. E., F. J. Burns and R. D. Heimbach. The Effect of Penetration Depth of Electron Radiation on Skin Tumor formation in the Rat. Radiat. Res. 30:515-524, (1967).

19. Unpublished data.
20. Albert, R. E., F. J. Burns and R. D. Heimbach. The Association between Chronic Radiation Damage of the Hair Follicles and Tumor Formation in the Rat. Radiat. Res. 30:590-599, (1967).
21. Albert, R. E., M. E. Phillips, P. Bennett, F. Burns and R. Heimbach. The Morphology and Growth Characteristics of Radiation-Induced Epithelial Skin Tumors in the Rat. Cancer Res. 29:658-668, (1969).
22. Heimbach, R. D., F. J. Burns and R. E. Albert. An Evaluation by Alpha-Particle Bragg Peak Radiation of the Critical Depth in the Rat Skin for Tumor Induction. Radiat. Res. 39:332-344, (1969).
23. Burns, F. J., R. E. Albert, I. P. Sinclair and P. Bennett. The Effect of Fractionation on Tumor Induction and Hair Follicle Damage in Rat Skin. Radiat. Res. 53:235-240, (1973).
24. Unpublished data.

WASH-1320

WASH-1320, "A Radiobiological Assessment of the Spatial Distribution of Radiation Dose from Inhaled Plutonium," by W. J. Bair, C. R. Richmond and B. W. Wachholz, September 1974, was entered into the Record as an integral part of the AEC Testimony. A copy accompanies this document.