CONF-801075--1

MASTER

THE PLUTONIUM CONTROVERSY

Chester R. Richmond

Oak Ridge National Laboratory

Oak Ridge, Tennessee

Research sponsored by the U.S. Department of Energy under contract W-7405-eng-26 with the Union Carbide Corporation.

THE PLUTONIUM CONTROVERSY

I. Introduction

I have been asked to speak to you on the subject of "The Plutonium Controversy. It seems fitting, therefore, to begin with how plutonium, element 94, received its name. I think this is of interest because there are numerous stories, both reliable and unreliable, as to how plutonium received its name. Plutonium is often thought of as being a man-made element which was discovered in Room 307, Gilman Hall, at Berkeley by Glenn Seaborg and colleagues in February 1941. This is a true statement but it suffers from the sin of omission. Actually, plutonium was created when the earth was formed and can be found today in several of its isotopic forms in very dilute concentrations throughout the Earth's surface. In fact, it has been found in Gabon, Africa, in association with other racionuclides as part of the geologic evidence that a natural nuclear fission reaction took place over many years about 1.8 billion years ago.

The year 1941, which marked the "discovery" of plutonium by Seaborg, was, indeed, a very interesting year. Among other things, it marked Pearl Harbor and our entry into World War II and, perhaps less widely known, the discovery by Charlotte Auerbach and J. M. Robson in this country that chemicals could induce genetic detriment in animals. This phenomenon was discovered independently for other chemicals in Germany by F. Oehlkers and in the Soviet Union by I. A. Rapoport at the same time. Auerbach et al. found that nitrogen mustard compounds were, indeed, mutagenic in animals. However, the work was not made widely known until 1946 because of wartime classification. On the other hand, perhaps a bit ironic, the first person to show mutagenic effects of any agent of living systems was Herman Muller, who published his findings of the effects of ionizing radiation on fruit flies in 1928 and received the Nobel Prize for his efforts in 1946. Please note, 1947 was the same year in which the work of Auerbach et al. was made public, some five years after its discovery because of wartime classification. So much for classification, secrecy, and the like.

Plutonium was not named until March, 1942. Perhaps the most credible story on how plutonium was named is that it was named after the planet Pluto following a precedence set for other actinide elements such as uranium (named in 1789 by Martin Klaproth after the planet Uranus) and neptunium (discovered in 1937 and named after the planet Neptune). Pluto was discovered by Claude Tombaugh in 1930. Others would have us believe that plutonium was named after the Greek underground god Pluto; thus the mythical association with hell. For example, one paper on plutonium has the following title: "The Element of the Lord of Hell." It is not clear whether the credit for the emotional title mentioned

should go to its authors, or to the editor of the New Scientist who published it.[1] Even this reliance on mythology as regards plutonium's namesake is a source of controversy. For example, Watson points out that Hades, the brother of Zeus and Poseidon, who ruled the underworld, was also called Pluto the rich one by the tactful Greeks.[1] Pluto, it seemed, was the deity who provided the Greeks with various items including their primary standard of wealth, that is, corn, from beneath the surface of the earth. Thus with Greek mythology, the two-sided nature of plutonium is foreshadowed, i.e., its use for energy and defense and its potential toxicity to man.

II. Plutonium Toxicity

I would now like to discuss with you some of the things we know about plutonium toxicity. Biological studies were started in the early 1940's, using ten milligrams of the first plutonium produced in the Clinton reactor in Tennessee. Responsible individuals within the Manhattan Project were aware of the detrimental effects resulting from the misuse of radium and other alpha particle emitters in the 1920's. Bone cancers were produced in radium dial painters and the patients given radioactive radium compounds.[2]

Within a short period of time important information on the absorption, distribution, retention, excretion, and biological effects of plutonium was produced, primarily by Dr. J. D. Hamilton and colleagues at Berkeley, California. I think it is correct to say that the toxicity of plutonium was understood and appreciated in the early days of the Manhattan Project.

The biomedical research group at Los Alamos under Wright Langham played a key role in our understanding of the biological behavior and effects of ^{239}Pu. Thus, it should come as no surprise that the Los Alamos group initiated studies on the so-called "hot particle" problem in the 1960's. Part of this renewed interest in hot particles or, more properly, non-uniform dose distribution, at Los Alamos stemmed from the use in space missions of radioisotope thermo-electric generators containing plutonium and the interest in particulate radioactive materials that might be produced from fission reactors in Project Rover.

You will note that I said renewed interest. I should state clearly that there has been a sustained interest in problems of non-uniform dose distribution among regulatory agencies, standards-setting bodies, and the international scientific community for many years. In fact, the issue of hot particles and non-uniform dose distribution has been explicitly addressed by standards-setting bodies in the past.[3]

III. Conjecture, Theory, Hypothesis, Fact, Truth

To study the plutonium controversy, the way science answers questions should be understood. Researchers use the scientific method to interrelate hypotheses, speculation, fact, and truth. The scientist often works from a base of accumulated knowledge and is sometimes guided by intuition. He then develops an hypothesis which can be tested by experiment. The development of simulation models is also a powerful means for us to construct an analytical framework for hypothesis testing. Something that is hypothetical is purely conjectural; that is, based upon an hypothesis. Hypotheses are assumptions subject to verification or proof, as a conjecture that accounts for a set of facts and then can be used as a basis for future investigation.

Theories allow us to try to "guess" an answer in advance of obtaining experimental proof. In one sense theory is a conclusion-jumping process, but it is not enough to jump to a conclusion without obtaining necessary supporting information. It has also been said that theory is a bold conjecture that sends us off to look for facts. Theories are revised as more information is obtained and occasionally they result in the establishment of a generally accepted body of knowledge or facts that are accepted as truth. However, as scientists should know, even the most solidly established facts are approximations of truth. The role of the scientist is to seek the truth, not to publicize hypotheses.

Thus we see an evolutionary process whereby theories are formed, working hypotheses are established and tested, facts and information are accumulated, and theories and hypotheses are either unsupported or more firmly established. We need to realize that a theory or an hypothesis is not an established fact or truth. The press might do well by learning, or perhaps relearning, this fundamental scientific process.

What about common sense and convictions? Here we must use caution, for Einstein once said, "Common sense is a layer of prejudice laid down in early life by inadequate training," and Friedrich Nietzsche stated, "Convictions are more dangerous enemies of truth than lies."

We must also remember that any new idea at the time of its inception is unique and stands alone. Sometimes a new idea is not easily accepted, and its ultimate acceptability by the scientific community depends on how well it withstands the scrutiny of the scientific process and peer review. New ideas are the fuel which fires the scientific process. We must, however, distinguish between acceptance of new ideas and dissention. Perhaps Eric Hofer put it best when he said, "A dissenting minority feels free only when it can impose its will on the majority; what it abominates most is the dissent of the majority."

IV. Plutonium - Major Controversies

1. Hot Particle

The revival of interest in the "hot-particle" problem, especially as regards particulate plutonium and other actinide elements in the lung, has stimulated a great deal of thought on this subject during the past several years. For many years nonuniformity of dose distribution has been of interest to standards-setting bodies and other groups, such as the National Academy of Sciences, and to health protectionists. In fact, interest in the subject as regards alpha-emitting radionuclides predates the discovery of plutonium in 1941.

The hot-particle problem was brought to the attention of several Federal agencies by the Natural Resources Defense Council (NRDC), Inc. The NRDC's original petition and supporting documentation were submitted to the U. S. Atomic Energy Commission (AEC) and the U. S. Environmental Protection Agency (EPA) on February 14, 1974.[4] Because of the Energy Reorganization Act of 1974, which created the Energy Research and Development Administration (ERDA) and the Nuclear Regulatory Commission (NRC), the Federal response to the NRDC petition became the responsibility of the EPA and the NRC. Although many organizations have considered the hot-particle problem for decades, there has been considerable reassessment of the problem since February 1974.

The NRDC petition states that in its view the present radiation standards are too high by a factor of 115,000 when applied to hot particles. In addition, the petition states that each NRDC member is a potential victim of exposure to hot particles. The document supporting the petition, prepared by A. R. Tamplin and T. B. Cochran, hypothesized that the highly localized alpha radiation from inhaled particles greater than a specified size (about 0.6 micrometer in diameter) causes greater tissue damage and is subsequently more carcinogenic than more uniformly delivered radiation. They proposed that a single radioactive particle in the lung capable of delivering a local radiation dose of 1000 or more rems per year will produce local tissue damage. The local tissue damage, in turn, produces a risk of lung cancer of 1 in 2000 (5×10^{-4}); in other words, exposure to 2000 such hot particles would produce one lung cancer.

Thus, the so-called "hot-particle" hypothesis became transformed into a legal petition and, unfortunately, was interpreted by some as having an acceptable established scientific basis. In effect, the petition to make drastic changes in radiation protection standards initiated many responses to and reviews of the petition and its underlying hypothesis.

I have given the chronology of reviews of the NRDC submission to the AEC (Table 1) and I shall discuss some but not all of these responses.

TABLE 1. CHRONOLOGY OF RADIATION PROTECTION STANDARDS

ISSUE - HOT PARTICLE PETITION

Date	Report
14 February 1974	National Resources Defense Council Petition to Atomic Energy Commission and Environmental Protection Agency
July 1974	United Kingdom - National Radiological Protection Board Report
September 1974	USAEC Report - Wash-1320
November 1974	Los Alamos Scientific Laboratory Report LA-5810-MS
February 1975	United Kingdom Medical Research Council Report
August 1975	U.S. National Council on Radiological Protection and Measurements
October 1975	U.S. Open Literature Publication - Health Physics; then others
19 February 1976	Federal Republic of Germany - Comments of Ad Hoc Committee of the Radiation Protection Committee
12 April 1976	Nuclear Regulatory Commission* rejects petition in Federal Register
September 1976	United Kingdom's Royal Commission on Environmental Pollution Report "Nuclear Power and the Environment"
October 1976	U.S. National Academy of Sciences Report
6 January 1977	Environmental Protection Agency Rejects Petition in Federal Register

*Created from AEC

The British were the first to respond with a report from its National Radiological Protection Board (NRPB) in July 1974.[5] The short critical review concludes:

> "It is noted that the basis of ICRP recommendations is the average radiation dose to an organ and not the number of radioactive particles in the organ. This dosimetric basis of radiological protection has been established for many years by observation of humans and experimental work with animals. A better evaluation than offered by Tamplin and Cochran would be needed for this system to be set aside in favor of the hot-particle concept. Their estimate that there is a risk of cancer being generated in cells surrounding a hot particle of 1 in 2000 cannot be substantiated by our present knowledge."

In September 1974, the United Kingdom's NRPB published a report[6] entitled "Radiological Problems in the Protection of Persons Exposed to Plutonium." The following quotations are taken from Section 9 of the report entitled "Hot Particles."

After discussing the issue, the section concludes:

> "In summary, there is no biological evidence available at present which suggests that 'hot spots' carry a higher risk of cancer induction. Hence there is no necessity to change from the present system of using average dose to organs or tissues. However, it would be prudent to continue research into the biological effects of non-uniform dose distributions within organs."

Another report that should be consulted by those interested in the hot-particle issue is WASH-1320, "A Radiobiological Assessment of the Spatial Distribution of Radiation Dose from Inhaled Plutonium," which was published in September 1974 by the USAEC.[7]

The report was not meant to be a direct critique of the NRDC petition on hot particles. It addressed the main generic issue of the problem, that is, the question of the biological importance of spatial distribution of radiation dose from inhaled plutonium.

The following information was taken from the November 1974 Los Alamos Scientific Laboratory report, prepared by a group of biomedical researchers with relevant plutonium research experience.[8] This report, "A Review of the Natural Resources Defense Council Petition Concerning Limits for Insoluble Alpha Emitters," concludes:

> "The preceding review has indicated that the Tamplin-Cochran conclusions are based upon a hypothesis which requires considerable extrapolation of the data upon which it is based. Later evidence, of the same nature as was used in the derivation (ie., rat skin data), does not support the assumptions of the original model.

> "The Tamplin-Cochran interpretation of the model not only fails to take into account the later evidence but appears to present the hypothesis as fact. The supporting evidence on human data which they present is based upon unsupported assumptions and distortions of the words of the authors they quote. Most importantly, they fail to use or acknowledge direct evidence on the effect of radioactive particles. Such evidence indicates that the basic damage model which they use overestimates badly the carcinogenic effects of radioactive particles.
>
> "We conclude, therefore, that the application of the average organ dose to the establishment of limits is still appropriate, although experimentation to narrow existing uncertainties on the effects of nonuniform dose distribution should continue."

Neither takes the position that the hot-particle hypothesis is correct. Note that both reviews call for additional work on the subject as a means of reducing the uncertainty of the situation. Thus the subject is not completely closed.

The EPA held hearings in December 1974 (Washington, D.C.) and January 1975 (Denver, Colorado) on the subject of plutonium standards, and the question of hot particles was addressed.[9] Proceedings from these hearings are available in a three-volume publication. A compilation of the AEC's testimony presented at these hearings was made available earlier in WASH-1359,[10] entitled "Plutonium and Other Transuranium Elements: Sources, Environmental Distribution and Biomedical Effects."

The National Council on Radiation Protection and Measurements (NCRP) released NCRP Report 46, entitled "Alpha-Emitting Particles in Lungs." The report was prepared by an ad hoc committee, reviewed and approved by the entire NCRP, which is comprised of approximately 70 individuals, and published in 1975.[11]

The NCRP report concludes that:

> "A substantial body of experimental animal data indicates that particulate plutonium in the lung is no greater hazard than the same amount of plutonium distributed more uniformly throughout the lung.
>
> "The above observation from animal data is consistent with the theoretical analysis of the microscopic distribution of energy absorption in each case.
>
> "The current NCRP practice of averaging absorbed dose over the lung is defensible when used in conjunction with appropriate dose limits.

> "More precise consideration of spatial distribution of absorbed dose cannot be profitably used to derive permissible exposure limits until we have more understanding of the relation between dose and effect."

In 1975 the United Kingdom's Medical Research Council's Committee on Protection Against Ionizing Radiations published a report[12] entitled "The Toxicity of Plutonium." In the section of the report on hot particles, the authors state rather directly and with some British humor:

> "The conclusions of Tamplin and Cochran cannot be any better founded than the hypothesis on which they are based, and that is too tenuous to be worth further discussion here. Tamplin and Cochran also put themselves in the difficult situation that the risk is considered to be decreased by a factor of 115,000 if a particle containing 0.1 pCi plutonium were to break into two equal halves."

The report concludes that there is presently no evidence to suggest that irradiation of the lung by plutonium particles is likely to be markedly more carcinogenic than for the case when the same activity is uniformly distributed.

Another publication suggesting possible reductions for permissible exposure to plutonium considered the lung and the question of hot particles.[13] The author states:

> "No one knows the answer to this question at the present time. Certainly we would like to have more information. Tamplin and Cochran suggest that because of the very large dose (thousands of rem/y) in the vicinity of a micron size particle of ^{239}Pu lodged in the lung tissue, the present q for lung ($\sim$0.015 Ci) and the corresponding values of $(MPC)_a$ for occupational exposure as well as those for members of the public should be lowered by a factor of 10^5. Perhaps they are right, but I believe that they have not made a strong case for this factor simply because adequate biological data are not available and much of that which we have seems to give contradictory information."

In a 1975 letter to the New Scientist, Tamplin conceded that their original risk estimates for hot particles were high by a factor of one hundred.[14]

In April 1976, the U.S. Nuclear Regulatory Commission denied the NRDC's petition to establish new standards for "hot particles" of plutonium and other radionuclides. In rejecting the petition in the Federal Register,[15] the NRC stated that:

- NRDC did not validly interpret the scientific data which it used in developing its position.

- o NRDC's position is not supported by the existing body of relevant knowledge on the biological effects of radiation.

- o Present radiation protection standards based on the assumption of uniformly distributed radioactive material in the lung are adequately conservative.

An ad hoc subcommittee of the National Academy of Sciences Committee on the Biological Effects of Ionizing Radiation reviewed the question of nonuniform lung irradiation from plutonium and other actinide elements. Their 1976 report, "Health Effects of Alpha Emitting Particles in Respiratory Tract," prepared for the Environmental Protection Agency, was the basis for the rejection of the NRDC petition by EPA.[16] The EPA rejection of the petition appeared in the Federal Register in 1977.[17]

An interesting analysis of the hot particle hypothesis was made by Federal Republic of Germany in early 1976.[18] It found no basis for support of the "hot particle" hypothesis. The authors, all professors, were associated with the following institutions:

University of Dusseldorf (Professor L. E. Feinendegen)
University of Munchen (Professor O. Hug)
University of Munchen-Neuherberg (Professor W. Jacobi)
University of Sarrlandes, Manburg/Saar (Professor E. Oberhausen)

Professor John T. Edsall of Harvard stated[19] that the "hot particle" hypothesis was likely to be incorrect but that it had not been "rigorously disproved." In response to comments[20] on his article,[21] Edsall stated, "As to the 'hot particle' hypothesis, my view in the published article was skeptical. In my opinion, the more recent studies, cited by Richmond, do now make the hypothesis highly improbable." However, Edsall goes on to say that he would not give the hypothesis any significant weight in making decisions about plutonium toxicity as there "is plenty of cause for concern without invoking hot particles."

The recent ICRP report on Biological Effects of Inhaled Radionuclides contains a section on Hot Particles.[22] The report concludes that lung cancer mortality in the beagle dog appears to be adequately accounted for by the conventional method of averaging the absorbed dose from plutonium over the entire lungs. Also, the results of the LASL studies discredit hypotheses that postulate an increased hazard from localized irradiation and demonstrates that the opposite is true. That is, at least for hamsters, localized irradiation is less tumorigenic than is diffuse exposure.

The summary of the section on Hot Particles continues, "The Task Group concludes that the risk of lung cancer from inhaled radiation (sic) particles will be greatly overestimated if based on hot particle concepts."

What can one conclude from the information now available on the hot-particle question? It appears that the majority of responsible researchers and others who have reviewed the question of lung irradiation from particulate plutonium have rejected the hot-particle hypothesis as put forth by Tamplin and Cochran as being unsupportable. However, this is an important radiobiological question that will continue to be of interest to researchers and radiation protection groups.

It is regrettable, however, that questions such as this require such long periods of time for response. The NRC rejected the NRDC's petition 26 months after it was filed. The same action was taken by the EPA in January 1977, some 35 months after the petition was filed. Let me restate what I wrote in 1976:[23]

> "Perhaps we could accelerate the examination process by immediately directing important issues to established organizations such as the NCRP or, on an international scale, the ICRP, to determine if the question or issue is of reasonable importance and priority to command immediate attention of the responsible regulatory agency. There must also be judgments other than technical that enter the decision-making process on a given issue, and these must not be overlooked. It is difficult, however, for the lay public to have thrust upon them complex issues that have not first been evaluated by national or international organizations established to provide guidance on the issues being questioned or examined. I believe much time, money, and frustration could be saved if we developed a better system for review. In some instances, questions might be identified as "non-issues" and set aside in deference to questions that may indeed require review and decision making by the technical and other components of society."

2. The Most Toxic Substances Known to Man

I chaired a panel discussion on "Plutonium - Implications for Man and His Environment" at the 1975 Symposium on Transuranium Nuclides in the Environment.[24] In the discussion following a presentation by R. B. Coulom (France), the following statement was made by B. R. Hookway (United Kingdom). "There is little doubt that when dealing with plutonium, we are dealing with the most dangerous substance known to man - the only substance a single microgram of which can induce mass hysteria throughout all nations . . . . "

Although many people like to state, sometimes with much apparent authority, that plutonium is the most toxic or hazardous substance known to man, it is difficult to find the scientific basis for such a statement. The National Council of Churches has declared that plutonium is "morally dubious" and Dr. Glenn Seaborg, the element's discoverer, has referred

to it as the "ornery" element.[25] Others state that plutonium is toxic beyond human experience, and Time has called it "searingly radioactive." We also find some who claim that plutonium is "relatively harmless."[26]

What then can we state about plutonium's toxicity as compared with both other radioactive nuclides and stable elements and compounds? Plutonium is a very toxic material. It is an alpha particle emitter and much information has accumulated over the years on the biological effects of alpha-emitting radionuclides. Up to the fall of 1943, the total plutonium in existence, which was cyclotron-produced, amounted to only two milligrams. As plutonium began to arrive at various sites from the Clinton Laboratories in milligram amounts, Glenn Seaborg began to worry about potential detrimental effects to workers, and on 5 January 1944, sent a memorandum to Dr. Robert S. Stone, Director of the Health Division at the Metallurgical Laboratory.[25] Part of that memo is as follows:

> "It has occurred to me that the physiological hazards of working with plutonium and its compounds may be very great. Due to its alpha radiation and long life it may be that the permanent location in the body of even very small amounts, say one milligram or less, may be very harmful. The ingestion of such extraordinarily small amounts as some few tens of micrograms might be unpleasant, if it locates itself in a permanent position. In the handling of the relatively large amounts soon to begin here and at Site Y [Los Alamos], there are many conceivable methods by which amounts of this order might be taken in unless the greatest care is exercised.
>
> "In addition to helping to set up measures in handling so as to prevent the occurrence of such accidents, I would like to suggest that a program to trace the course of plutonium in the body be initiated as soon as possible. In my opinion such a program should have the very highest priority."

To the scientific community, the experience of the radium dial painters from the 1920's was evidence that poor management of alpha emitting materials could lead to significant incorporation within the body which, in turn, could lead to detrimental biological effects. Many members of the radiobiological and health protection communities were aware that lung cancers were produced in the underground miners as far back as the 1500's. In 1932 it was reported that nine of seventeen deaths observed during 1929-1930 among miners of uranium-bearing ores in Joachimsthal, Czechoslovakia, were due to cancer of the lung.[27] These investigators thought that the most probable cause of the lung tumors was radiation in the air of the underground mines. So, for plutonium, health protection was part of early studies and production.

Some would have us believe that the adverse effects from plutonium are mysterious and different from other radionuclides since plutonium is a "man-made" element. This has been proven to be untrue by the many

st· lies of the effects of alpha particles on living organisms, including man. If I may use poetic license, an alpha is an alpha is an alpha is an alpha (except for energy differences).

Let us compare plutonium toxicity with the toxicity of some materials that are known to be extremely toxic. Table 2, based upon information from Stannard,[28] shows information on super toxic substances, some of which are familiar to this audience. There are also many chemical poisons which vary as to their acute toxicity in man, such as potassium cyanide, hydrocyanic acid, lead acetate, mercury dichloride, methyl mercury, and cadmium. Many of these are classified as Class 1, or extremely toxic, in generally accepted classifications of toxicity. In this particular scheme, the median lethal dose (that amount which will kill half the exposed individuals) is less than one milligram per kilogram of body weight. It is also a level for which the estimated lethal dose for a person is approximately one gram, and the level at which about 33 to 66% mortality would be observed following the inhalation of less than ten parts per million of the material in question.

I should emphasize that we are considering only acute toxicity, that is, where the effects are rapidly manifested and very often lethal. Table 3, based upon data from Stannard,[28] gives some acute toxicity data for ^{239}Pu. Clearly, plutonium is less toxic than some of the materials listed in Table 2. Some studies shown in Table 3 would lead us to classify plutonium as an extremely toxic material (Class 1) as identified earlier, or perhaps as highly toxic (Class 2). For Class 2 materials the LD_{50}, the median lethal dose, is one to fifty milligrams per kilogram of body weight.[29] The estimated lethal dose for persons is approximately one teaspoon and 33 to 66% mortality would result following the inhalation of ten to a hundred parts per million of the material.

We also need to consider chronic biological effects resulting from plutonium contamination. Table 4 summarizes information on the long-term cancer induction resulting from experiments in which ^{239}Pu was administered either by inhalation, intravenous injection, or by intratracheal injection.[28] These data show that Pu is very effective in producing lung cancer at some time during the life span of the experimental animals. There are no comparable data for man nor are there means of making direct comparisons with chemical carcinogens to which humans are exposed since we lack standard methodology for estimating chronic effects and chronic toxicity. It has been suggested that the inhalation of about 90 micrograms (about 30 microcuries) of plutonium would shorten life expectancy about as much as smoking one pack of cigarettes per day. I do not believe such comparisons are valid in view of the uncertainties of the information upon which extrapolations and comparisons must necessarily be made.

TABLE 2. SMALLEST QUANTITY OF SUBSTANCE REQUIRED FOR ACUTE LETHALITY IN EXPERIMENTAL ANIMALS FOLLOWING INTRAPERITONEAL (IP) OR INTRAVENOUS (IV) INJECTION

Substance	Species	Route	Quantity(g/g)
Botulin Toxin A (crystalline)	Mouse	IP	7×10^{-18}
Botulin Toxin A	Mouse	IP	3×10^{-15}
Tetanus Toxin	Mouse	IP	1×10^{-13}
Diphtheria Toxin	Mouse	IP	3×10^{-10}
Bufo Toxin	Cat	IV	4×10^{-7}
Curare	Mouse	IP	5×10^{-7}
Strychnine	Mouse	IP	5×10^{-7}

TABLE 3. ACUTE TOXICITY OF ^{239}Pu ADMINISTERED BY INTRAVENOUS ADMINISTRATION (IV) OR INHALATION (INH)

Criterion	Species	Entry Route	Quantity(g/g)
$LD_{50/30}$*	Dog	IV	3.2×10^{-7}
$LD_{50/30}$	Dog	INH	1.3×10^{-6}
MST** = 80 d	Dog	INH	5 to 8×10^{-7}
$LD_{50/30}$	Rat	INH	2×10^{-6}
MST = 7 to 10 d	Rat	INH	4×10^{-6}

* $LD_{50/30}$ = median lethal dose in 30 days.

** MST = median survival time.

TABLE 4. LONG-TERM CANCER INDUCTION BY ^{239}Pu

Chemical Form	Species	Entry Route	Time	Quantity (g/g)	Type of Cancer
Oxide	Dog	Inhalation	11.5 yr	10^{-9} (1.1 g in total lung)	Carcinoma (incidence, 100%)
Oxide	Dog	Intravenous	Years	2.6×10^{-10}	Osteosarcoma (incidence, 33.3%)
Oxide	Mouse	Intratracheal	500 d	1.3×10^{-7}	Fibrosarcoma (incidence, 5%)
Oxide	Mouse	Intratracheal	100 d	6.4×10^{-9}	Bronchiolar Carcinoma (incidence, 2.5%)
Oxide	Mouse	Inhalation	500 d	4×10^{-9}	Bronchiolar Carcinoma (incidence, 1.4%)

One can also compare the potential risk of plutonium to that of other radionuclides if one compares the values of the maximum permissible concentrations of various radionuclides in either air or water. One finds that plutonium is not the radionuclide with the most restrictive exposure levels for all routes of exposure. Following oral intake, radionuclides such as ^{131}I, ^{90}Sr, ^{233}U, and ^{241}Am present similar or greater relative risks as compared to ^{239}Pu. It is only when one considers the inhalation route that ^{239}Pu represents the largest relative risk by one to several orders of magnitude as compared to other radionuclides.

3. A Pound of Plutonium Will Kill . . .

It has been said, sometimes with much emotion, that

- a pound of plutonium will kill millions of people.
- a pound of plutonium could cause eight billion cancer cases.
- a piece of plutonium the size of an orange is sufficient to kill the population of the British Isles.

There are many variations of this theme. In many instances similar statements such as the following are attributed to "experts" and broadly disseminated:

> "The byproduct of breeder reactors, Plutonium-239, has a half-life of nearly 25,000 years, yet experts (my italics) suggest that a lethal dose for the whole human race need not be larger than an apple."[30]

I always knew it was wrong to mix apples and oranges, but I had no idea of the seriousness of doing so.

In fact, many members of the public are led to believe that any exposure to plutonium, no matter how small, will result in the development of lung cancer which in turn leads to death. Admittedly, plutonium is a very toxic material but not for which the scientific and technical community has developed a healthy respect. However, exaggerated statements such as those made above are not accurate and their use only serves to introduce more emotionalism into the subject.

The important point to remember is that many of the arguments which tend to exaggerate the potential harm from plutonium assume that a given amount is deposited within the human body - for example, in the lungs. Often there is no attempt to mention or explain the fact that all the plutonium that might be released from a given facility or incident would not end up quantitatively within the lungs of people. In addressing the

charge that a piece of plutonium about the size of an orange would kill off the population of the British Isles, Sir John Hill stated,[31]

> "That is about as sensible a statement or as relevant as saying that one road tanker of liquid chlorine has enough poison to kill everybody in Europe; or, for that matter, saying that the air in this assembly room would, if injected into the veins of the public, kill everybody in the world. All these statements would be near enough true if the population was stupid enough to allow someone to inject just the right amount of material in question into the critical organ of the body."

Some individuals have been quite specific. For example, one individual has stated that there are 7,830,000,000 "lung cancer doses" per pound of plutonium.[32] That is almost two for everyone in the world!!! Such statements often are based on the assumption that the Pu is completely deposited in the lung. Also, the estimates of the number of health effects produced per unit of radiation dose are substantially higher than those estimated by national and international bodies that provide guidance to health protection organizations within various countries.

Let us now return from speculation and hypothesis to the real world. We can approach this problem from the standpoint of the environmental inventory of plutonium. The main source of transuranium elements in the environment is fallout from nuclear weapons testing. About 4.2 tons (3,818 kilograms), or about 330,000 curies $^{239-240}Pu$ have been globally dispersed as fallout from atmospheric testing of nuclear weapons. Another 110,000 curies $^{239-240}Pu$ have been deposited locally around nuclear test sites.[33] About 60% of this mixture is ^{239}Pu. Because these nuclides of plutonium have similar alpha energies, they are often measured together and reported as ^{239}Pu.

In addition, about 120,000 curies of ^{241}Am contribute additional alpha activity equivalent to roughly 25% of that from the plutonium nuclides. The formation of a beta-emitting nuclide, ^{241}Pu, which decays to ^{241}Am with a half life of 432 years, will approximately double the amount of ^{241}Am originally produced during weapons testing. Small amounts of Pu and other transuranium radionuclides are still added to the atmosphere by several countries that did not sign the 1963 Limited Test Ban Treaty.

Air concentrations of $^{239-240}Pu$ have been measured at different times and locations worldwide. For example, peak surface air concentrations of 0.0017 picocurie per cubic meter, measured for New York City in 1963, decreased to 0.001 picocurie per cubic meter at stations in Washington State by 1971.

The accumulated deposition of globally dispersed Pu from weapons testing is virtually complete. Plutonium deposition rates have been made annually in some areas. Those in the New York area show that of the more than 300,000 curies ^{239}Pu produced by atmospheric tests up until

1962, about 90% had returned to the earth's surface by 1965.[34] About 80% of this Pu was deposited in the northern hemisphere (where most of the testing occurred) and the rest in the southern hemisphere.[35] It is estimated that by 1970-71, about 16,000 curies ^{239}Pu had been deposited in the conterminous United States and about 3,000 curies on the Australian continent which is about the same land area.

To put these quantities in perspective it may be helpful to consider the quantities of naturally occurring alpha-emitting radionuclides that also are in the soil. It has been estimated that the continental USA contains about 4,400,000 curies of natural alpha-emitting radionuclides in the upper 2-centimeter (0.8 inch) layer of soil. Radionuclides of uranium and thorium comprise 1,600,000 curies. Therefore, the 16,000 curies of alpha radioactivity from $^{239-240}$Pu in the upper 2 cm of soil in the USA is about 1% of that from naturally occurring actinide radionuclides (uranium and thorium) and about 0.36% of the total naturally occurring alpha radioactivity.[36]

Incidentally, the deposition of Pu around the world is not uniform. Analyses of soil samples in the late 1960's yielded concentrations of 2.3 $\pm$0.3 millicuries ^{239}Pu per square kilometer.[37] Samples collected around Ispra, Italy, in 1966-1967 gave a mean value of 2.1 $\pm$ 0.7 millicuries ^{239}Pu per square kilometer.[38] However, much higher or lower values have been reported for extreme environments. Total ^{239}Pu deposition in Thule, Greenland, through 1968 was 0.3 millicuries per square kilometer, while in selected areas of the Austrian Alps, ^{239}Pu deposition values of 15.3 $\pm$ 2.9 millicuries per square kilometer have been reported.[39] I noted earlier the roughly five-fold difference between total Pu deposition in northern and southern latitudes.

Now let us attempt to estimate how much of the weapons-related Pu finds its way into human beings. Estimates have been made of the radiation dose people might receive from plutonium in weapons-produced fallout. The bases of the calculations are both measured and inferred air concentrations and dietary levels of ^{239}Pu. The metabolic models used for plutonium were developed by Committee 2 of the International Commission on Radiological Protection.[40]

Table 5 gives information on intake, transfer to blood, excretion, retention, and body burden of ^{239}Pu from weapons fallout for the general population of the northern hemisphere in 1972. Plutonium from nuclear weapons accounts for virtually all that is present in the environment.

Intake data comes primarily from the Environmental Measurements Laboratory in New York City.[34] Limited data from other sources such as Italy are in general agreement.[41] Uptake of plutonium from food and water to blood is taken to be 0.01% (one part per 10,000 ingested, the remaining 99,000 are excreted) on the assumption that plutonium will be transferred to man from the food chain only in the soluble form. The amount of plutonium moving from air to blood is derived from the ICRP lung-model transfer parameters for very insoluble (Class Y) compounds.

TABLE 5. ^{239}Pu INTAKE, TRANSFER TO BLOOD, EXCRETION, INFERRED RETENTION AND ORGAN BURDENS FOR A PERSON IN THE GENERAL POPULATION OF THE NORTHERN HEMISPHERE - 1972

Intake	picocurie	millibecquerel
air	0.2	7.4
food	1.5	56
water	0.04	1.5
total	1.74	64
Portion of the intake transferred to blood		
from air[a]	0.013	0.48
from food[b]	0.00015	0.0055
from water[b]	0.000004	0.00015
total	0.013	0.48
Excretion		
urine	negligible	negligible
faeces	1.7	63
Portion of the intake retained in various organs		
lung and lymph nodes[c]	0.04	1.5
bone[d]	0.0059	0.22
liver[d]	0.0059	0.22
Actual content of various organs		
lung and lymph nodes	0.41	15
liver	0.89	33
bone	1.00	37
total	2.30	85

a 6.3% of intake from air (assumes Pu is inhaled in particles of 0.5 μm AMAD)

b 1/10,000 of intake from food and water (amended, ICRP 1979) assumes Pu is Class W (most insoluble form)

c 20% of intake from air ($T_{\frac{1}{2}}$ = 500d)

d 45% of amount entering blood ($T_{\frac{1}{2}}$ = 100y for bone, 40y for liver)

The model assumes that inhaled fallout plutonium is attached to aerosol particles of 0.5-micrometer diameter (aerodynamic). Of this material, 33% is deposited in the pulmonary region of the lung, 16% in the nasopharynx and 8% in tracheobronchial regions. About 6% of the total amount inhaled is transferred to blood.

We are most concerned about the Pu that is retained in the pulmonary region of the lung. About one-third of the total amount inhaled reaches this region. Some is lost rapidly (several days), but most is retained for longer periods. The amount retained in the pulmonary region of the lung with a 500-day half-time (the maximum allowed in the model and the best current estimate) represents about 20% of the inhaled plutonium. This plutonium irradiates the lung tissue until it leaves the lung via blood of lymphatic drainage or is swallowed and then lost from the body via the faeces.

The fractions of plutonium in blood going to bone and liver (both 45%) are those recommended by ICRP for the purposes of calculating radiation dose.[40]

The content of plutonium in human beings has been estimated for the period 1954–1974 using this approach. Table 6 gives the intake of weapons fallout ^{239}Pu through the lungs and computed body contents in man during that period. The intake reflects weapons testing activities with maxima in the late 1950's and again in 1963 with a declining intake following the 1963 Limited Test Ban Treaty. Cumulative intake up to and including 1974 was estimated to be about 43 picocuries which resulted in a maximum body content of about 4.3 picocuries in 1964. The estimated average total body content for 1972 of 2.3 picocuries is in good agreement with actual body contents derived by the Environmental Measurements Laboratory.[34] These values are similar to plutonium measurements made on human tissues obtained by autopsy at several locations.[42]

Very little of the multi-ton quantities of weapons-produced Pu has found its way into members of the public. Of that produced, only a total of about 0.25 grams of Pu probably found its way into the world's population. Approximately 5 picocuries per person and 3 billion people yields a total of 0.015 curie or 0.25 grams ^{239}Pu. (This would be 0.18 gram of $^{239-240}Pu$.) Because some 300,000 curies were released and deposited on earth, we can estimate that about 0.00000005 (0.015 curie/300,000 curies) or 5×10^{-8} of that Pu deposited world wide has found its way into the human population. Therefore, each person would have incorporated a tiny fraction (10^{-17} or 0.00000000000000001) of the total amount produced during atmospheric testing. Thus, the transfer to and uptake of plutonium by man from the environment is very low. We cannot assume that all released plutonium ends up quantitatively in the lungs of people. To do so introduces huge errors in any subsequent calculations of harm.

TABLE 6. RELATIONSHIP BETWEEN WEAPONS FALLOUT DEPOSITED ^{239}Pu LEVELS IN MAN AND NATURALLY OCCURRING ALPHA RADIOACTIVITY

Deposited Fallout Plutonium	curies*	picocuries
World	330,000	3.3×10^{17}
Northern Hemisphere	265,000	2.6×10^{17}
USA	16,000	1.6×10^{16}
Australia	5,000	5.0×10^{15}
Naturally Occurring Alpha Activity		
USA (top 5 cm)	5,940,000	5.9×10^{18}
Fallout Plutonium in Man, Northern Hemisphere		
Average Person, 1964	4.1×10^{-12}	4.1
Average Person, 1972	2.3×10^{-12}	2.3
Average Person, cumulative intake	4.3×10^{-11}	43.0
Naturally Occurring Alpha Activity in Man		
Average Person, Northern Hemisphere	5.1×10^{-10}	510

* Curie = 1 thousand billion (10^{12}) picocuries

How much plutonium would one need to release into the environment to have one pound deposited into human beings? Based upon information given above the answer is several hundred thousand tons!!!

Once we know the amount of plutonium in a person, we can estimate the radiation dose using standard methodology as, for example, developed by the ICRP. Cumulative doses up to and including 1974 to a person exposed throughout the entire fallout period are about 16 millirem (0.16 millisieverts) to lung, 9 millirem (0.09 millisieverts) to bone, and 5 millirem (0.05 millisieverts) to liver.[34] These estimated radiation doses from fallout ^{239}Pu are considerably smaller than those arising from naturally occurring alpha-emitting radionulides in the environment.

Radiation dose commitments from naturally occurring radionuclides are estimated to be about 3,000 millirem (30 millisieverts) to the lung throughout th lifetime of a person. The comparable value for bone is about the same. Thus, the contribution to the total dose from naturally occurring alpha emitters by ^{239}Pu is small.

IV. Conclusion

Let me state the following. Plutonium is a toxic material. Many materials are toxic and must be handled and treated as a potentially hazardous material. It is also a valuable material because it is a source of energy and used in nuclear weapons. Plutonium toxicity can be expressed only after it is incorporated into the body and entry is quite inefficient. The inhalation route currently appears to be the entry route of most concern. Other toxic materials may present greater overall hazards to man because they may be produced in much larger quantities and more efficiently find their way into people where their toxicity can be expressed. Perhaps cigarette smoke is a good example. Plutonium is not the most toxic material on earth, but we have learned to treat it and other materials with respect.

We need to accumulate more information on what level of health effects (including genetic) we might expect per unit of plutonium deposited in people. This, however, can be said of practically any potentially hazardous material to which human beings are exposed. Society must somehow make these important yet extremely complicated decisions about how much information is needed for a given chemical or radionuclide so that it can decide if coexistance with the material is possible or desirable. However, we must accumulate and present the scientific and technical information in traditionally established and accepted ways. Emotionalism and exaggeration instill fear, not knowledge, into the non-experts.

Let me return to our earlier discussion on early work on plutonium toxicity. Animal experiments using mice to determine the toxicity of

uranium and plutonium were performed at the Metallurgical Laboratory at the University of Chicago in 1944 by Brues, Lisco, and Finkel. In 1947, these workers published a paper in Cancer Research entitled "Carcinogenic Action of Some Substances Which May Be a Problem in Certain Future Industries."[43] These same workers published another paper titled "Carcinogenic Properties of Radioactive Fission Products and of Plutonium in Radiology."[44] This happened within six years of the discovery of plutonium and only several years after its availability for biomedical research. Perhaps there is a lesson to be learned from this information now in 1980, some 33 years later, as we try to sort out which of the many materials in the environment are potential or real health problems.

References

1. Watson, G.M. Plutonium. Atomic Energy in Australia 19(3), 7-17, July 1976.

2. Blum, T. Osteomyelitis of the mandible and maxilla. Journal of the American Dental Association 11, 802, 1924.

3. International Commission on Radiological Protection. Radiosensitivity and spatial distribution of dose, reports prepared by two task groups of Committee 1 of the International Commission on Radiological Protection, in ICRP Publication 14, Pergamon Press, Oxford, 1969.

4. Petition to Amend Radiation Protection Standards as They Apply to Hot Particles, submitted to the Environmental Protection Agency and the Atomic Energy Commission by Natural Resources Defense Council, February 1974; and 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 to Man to Insoluble Particles of Plutonium and Other Alpha-Emitting Hot Particles, Natural Resources Defense Council, Washington, 1974.

5. Dolphin, G.W. Hot particles. Radiol. Prot. Bull. 8, 8-9, July 1974.

6. Dolphin, G.W., et al. Radiological problems in the protection of persons exposed to plutonium, in National Radiological Protection Board Report NRPB-R29, September 1974.

7. Bair, W.J., C.R. Richmond and B.W. Wachholz. A radiobiological assessment of the spatial distribution of radiation dose from inhaled plutonium, in USAEC Report WASH-1320, September 1974.

8. Healy, J.W., E.C. Anderson and C.R. Richmond. A review of the Natural Resources Defense Council petition concerning limits for insoluble alpha emitters, in USAEC Report LA-5810-MS, Los Alamos Scientific Laboratory, November 1974.

9. Proceedings of Public Hearings: Plutonium and Other Transuranium Elements, U.S. Environmental Protection Agency, Office of Radiation Programs Report ORP/CSD-75-1, 1975.

10. Plutonium and Other Transuranium Elements: Sources, Environmental Distribution and Biomedical Effects, USAEC Report WASH-1359, December 1974.

11. Bair, W.J., et al. Alpha-emitting particles in lungs, in National Council on Radiation Protection and Measurements Report NCRP-46, 1975.

12. Medical Research Council (Great Britain). The Toxicity of Plutonium. Her Majesty's Stationery Office, London, 1975.

13. Morgan, K.Z. Suggested reduction of permissible exposure to plutonium and other transuranium elements. Amer. Ind. Hyg. Assoc. J. 36, 567-575, 1975.

14. Tamplin, A.R. Hot particles. New Scientist, 65(941), 738, March 1975.

15. Federal Register. Vol. 41, No. 71, 15371, April 12, 1976, Docket No. PRM-20-5, Natural Resources Defense Council -- Denial of Petition for Rule Making.

16. National Academy of Science - National Research Council. Ad Hoc Committee on "Hot Particles" of The Advisory Committee on Biological Effects of Ionizing Radiation. Report prepared for EPA on "Health Effects of Alpha Emitting Particles in Respiratory Tract." EPA 520/4-76-013, Washington, D.C., EPA, 1976.

17. U.S.E.P.A. Alpha-emitting hot particles - response to petition. Federal Register 42, 1228, 1977.

18. Feinendegen, L.E., O. Hug, W. Jacobi and E. Oberhausen. Zur toxizitat inhalierter heisser partikel insbesondere von plutonium. Comments of an ad hoc committee of the Radiation Protection Committee. Der Bundesminister Des Innern, 1-18, February 1976.

See also: Jacobi, W. Zum problem der heissen partikel Atomkernenergie (ATKE) Bd. 28, Lrf. 1, 1976.

19. Edsall, John T. Toxicity of plutonium and some other actinides. Bulletin of the Atomic Scientists 72(7), 26, September 1976.

20. Richmond, C.R. Plutonium hazards. Bulletin of the Atomic Scientists 33(4), 6-7, 1977.

21. Edsall, John T. Response to letter. Bulletin of the Atomic Scientists 33(4), 7, 1977.

22. International Commission on Radiological Protection. Biological effects of inhaled radionuclides, in ICRP Publication 31, Pergamon Press, Oxford, 1980.

23. Richmond, C.R. Current status of the plutonium hot-particle problem. Nuclear Safety 17(4), 474, August 1976.

24. Richmond, C.R. Chairman, panel discussion on plutonium - implications for man and his environment, in Transuranium Nuclides in the Environment, pp. 627-646, IAEA, Vienna, 1976.

25. Seaborg, Glenn T. Plutonium revisited, in Radiobiology of Plutonium, edited by B. J. Stover and W.S.S. Jee, pp. 1-21, The J. W. Press, Salt Lake City, Utah, 1972.

26. Cohen, B.L. Plutonium toxicity: an evaluation indicates it is relatively harmless. Nuclear Engineering International, 35-38, November 1976.

27. Pirchan, A. and H. Sikl. Cancer of the lung in the miners of Jachymor. American J. Cancer 16, 681-722, 1932.

28. Stannard, J.N. Plutonium toxicology and other toxicology, in The Health Effects of Plutonium and Radium, edited by B.J. Stover and W.S.S. Jee, pp. 363-372, The J. W. Press, Salt Lake City, Utah, 1972.

29. Hodge, H.C. and J.H. Sterner. Tabulation of toxicity classes. Amer. Ind. Hyg. Assoc. Quarterly 10, 93-96, 1943.

30. Wood, B. Interview in The Christian Science Monitor, November 30, 1973.

31. Hill, Sir John. The abuse of nuclear power. IAEA Bulletin 19(2), 42-49, April 1977.

32. Gofman, J.W. The cancer hazard from inhaled plutonium, in Committee for Nuclear Responsibility, CNR Report 1975-1-R, 1975.

33. Hardy, E.P. Worldwide distribution of plutonium, in Plutonium and Other Transuranium Elements, WASH-1359, USAEC, 1974.

34. Bennett, B.G. Transuranium element pathways to man, in Transuranium Nuclides in the Environment, IAEA, Vienna, pp. 367-383, 1976.

35. UNSCEAR. Sources and effects of ionizing radiation, in United Nations Scientific Committee on the Effects of Atomic Radiation, Report to the General Assembly, 1977.

36. USAEC. Environmental impact statement on the liquid metal fast breeder reactor, in WASH-1535, Appendix 11G, 1974.

37. Hardy, E.P. and P.W. Krey. Determining the accumulated deposit of radionuclides by soil sampling and analysis, in Environmental Plutonium, LA-4756, 1971.

38. DeBortoli, M.C. and P. Gaglione. Natural and fallout radioactivity in the soil. Health Physics 17, 701, 1969.

39. Irweck, K. Die bestimmung von plutonium in bodenproben, in Oesterreichische Studiengesellschaft fur Atomenergie GmbH, Vienna, SGAE-23810, ST-70/77.

40. International Commission on Radiological Protection. Limits for intakes of radionuclides by workers, in ICRP Publication 30, Pergamon Press, Oxford, 1979.

41. Clemente, G.F., P. Belloni, S. Di Pietro, G. Santori, and G.P. Santaroni. Tritium and plutonium content in the Italian diet and transfer to man, in Biological Implications of Radionuclides Released from Nuclear Industries, IAEA, Vienna, Vol. II, 257, 1979.

42. Richmond, C.R. Current status of information obtained from plutonium contaminate people, in Radiation Research, pp. 1248-1265, Academic Press, Inc., New [illegible] 1975.

43. Brues, A., H. Lisco and M. Finkel. Carcinogenic action of some substances which may be a problem in certain future industries. Cancer Res. 7, 48, 1947.

44. Brues, A., H. Lisco and M. Finkel. Cacinogenic properties of radioactive fission products and of plutonium. Radiology 49(3), 361-363, September 1947.