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ECOLOGICAL ASPECTS OF PLUTONIUM DISSEMINATION
IN TERRESTRIAL ENVIRONMENTS*

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Abstract—The potential benefits to be derived for mankind from continued development of peaceful applications for plutonium cannot be denied. The technology of plutonium production and processing is already established, but the realization of its peaceful applications depends largely upon the development of methods for preventing its distribution in the environment. Because of safeguards and effective control measures, no accidental plutonium contamination of the public domain has imposed serious risks to a population group. Trace amounts of plutonium from above-ground nuclear detonations are contained in world-wide fallout; however, the levels of plutonium in foodstuffs and other components of the environment are insignificant compared to the amounts known to be hazardous. There has thus been very little interest in the study of ecological aspects of plutonium contamination. The result is a paucity of information on the behavior

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of plutonium in ecosystems and its radiological effects on natural fauna and flora. The Nevada/Ecology Group is embarked upon a program at the Nevada Test Site to investigate the long-range effects of plutonium disseminated into the desert ecosystem. Emphasis has been placed upon standardization of analytical methods, delineation of contaminated areas, problems of resuspension and redistribution, food chain transport, and ecological effects.

INTRODUCTION

From an aesthetic point of view, the applications for plutonium represent some unique contrasts. Its use in nuclear weaponry strikes fear in the hearts of men and stokes the furnace of the "prophets of doom." On the other hand knowledgeable and positive thinkers cannot ignore the potential benefits to be derived for mankind from the continued development of peaceful applications for the energy resource offered by plutonium isotopes. The technology of plutonium production and processing is already established, but the realization of its peaceful applications depends largely upon the development of methods for preventing its dissemination in the environment.

Much is known about the physiological and toxicological properties of plutonium (half-life 24,400 years) because of the fine work of many investigators. It is not our task to cover these subjects inasmuch as past knowledge will be reviewed and recent findings will be reported in other papers presented at this symposium. Our task is to discuss some ecological aspects of plutonium contamination in terrestrial environments. In contrast to the extensive knowledge of plutonium health hazards acquired during the past three decades, (1-7) there is a paucity

of information on the behavior of this element in terrestrial ecosystems and its radiological effects on natural fauna and flora. In part this is because the USA and USSR, the major developers of nuclear-detonation technology, now conduct nuclear detonation tests underground which prevents the dissemination of radioactive contaminants. Immediate and effective measures also have been taken to decontaminate sites where accidents involving plutonium have occurred, so that the public has not been subjected to any serious risk from plutonium.⁽⁸⁾ In addition, the levels of contamination found in surveys of produce and components of the natural environment^(9 - 13) have been so insignificant, compared to the amounts known to be hazardous, that there simply has been very little interest in the study of ecological aspects of plutonium contamination. Some ecological guidelines fortunately are known; yet, in order to keep abreast of technical advancements involving future applications of plutonium, there is need for additional investigations of the long-range fate and effects of this element in terrestrial environments.

CONTAMINATION OF TERRESTRIAL ENVIRONMENTS

Without proper safeguards, plutonium is a hazardous material both because of its radioactivity and because it is a chemically toxic substance. Industrial processing accidents involving plutonium have occurred, but the contamination generally has been confined to in-house areas and controlled within occupational safety standards by decontamination measures.^(14 - 16) Two recent aircraft accidents involving nuclear weapons have caused relatively large-area contamination. In each case, however, the decontamination measures taken reduced the element of risk to conditions acceptable for normal occupancy of the affected area.^(8, 17)

Beginning with the first test of a nuclear fission weapon in New Mexico in 1945, terrestrial environments world-wide have since received plutonium contamination in fallout produced by nuclear weapons detonations. Harley⁽¹⁸⁾ estimates that the subsequent testing of nuclear weapons, up to the beginning of the atmospheric test ban treaty, distributed about 300 kilocuries of ^{239}Pu over the surface of the earth and that later tests by France and Communist China probably added about 15 kilocuries. An additional 17 kilocuries of ^{238}Pu was introduced into world-wide fallout when a SNAP-9A satellite power source burned up over the Indian Ocean in 1964.⁽¹⁹⁾ Langham⁽⁸⁾ estimates that the total amount of plutonium contributed to the world environment to date from all sources may amount to about 500 kilocuries. He also pointed out that on a weight basis, with other factors being the same, a ^{238}Pu contamination incident would be worse than ^{239}Pu by a factor of 270..

ACUTE AND RESIDUAL INHALATION HAZARDS

Upon analyzing potential health hazards arising from plutonium disseminated in the form of fine particulates, Langham, Harris and Shipman⁽²⁰⁾ concluded that fallout conditions having high concentrations in the air would create an immediate or acute inhalation hazard to anyone caught in or entering an area before all of the particulate matter has settled to the ground. The magnitude of this hazard depends upon the concentration of airborne plutonium, the respiratory rate of the individual, the length of time of exposure, and, to some extent, the particle size distribution of the radioactive fallout material. Langham and coworkers further concluded that the residual surface contamination would cause a potential

health hazard to persons living or working in the area, but they emphasized that any amount of plutonium deposited on the surface constitutes no hazard so long as it remains deposited. This residual plutonium hazard, therefore, is directly dependent upon that fraction of the surface contamination which subsequently is disturbed and resuspended in a manner which will enable it to gain entrance into the body, either by inhalation or ingestion.

The conditions just described primarily concern the potential inhalation hazard to man; however, the same problem applies to wild animals living in a contaminated area which would incorporate plutonium into their bodies through inhalation and ingestion. Man has the capacity to protect himself, and to some extent his domestic animals, from the fallout inhalation hazard. In contrast, the wild animal life would have to suffer its full onslaught. The residual hazard might even be magnified in some animals depending upon certain inherent living habits such as digging in soil, taking dust baths, or preening fur. In any event, it seems reasonable to predict that the acute inhalation hazard would account for most of the biological decimation that could result from the dispersal of toxic levels of plutonium in terrestrial environments. This prediction is somewhat conjectural, however, because of the paucity of results from actual field events or experimental studies upon which to draw conclusions concerning wild animal life. Some data on airborne concentrations of plutonium dispersed by high explosives during safety tests at the Nevada Test Site have been reported by Shreve.⁽²¹⁾ Wilson, Thomas and Stannard⁽²²⁾ also have reported some results with domestic animals in biomedical and aerosol studies

associated with a field release of plutonium dispersed by high explosives.

Workers from the Laboratory of Nuclear Medicine and Radiation Biology, University of California Los Angeles periodically have conducted field surveys in fallout areas downwind from these safety tests in order to investigate the biological availability of the residual plutonium contamination to native small mammals. Details of these surveys have been reported by Olafson and Larson, (23) Paglia, (24) Mork, (25) and Romney, Mork and Larson. (26) In general, these studies disclosed that the lung burdens of kangaroo rats (Dipodomys microps and D. merriami) were less than 1.0 pCi ²³⁹Pu per animal for specimens living in areas where the residual contamination level did not exceed 100 $\mu\text{g}/\text{m}^2$ (7.0 $\mu\text{Ci}/\text{m}^2$). Specimens collected from a safety test area 2.8 and 10.8 years after residual plutonium had been deposited at an initial level of 720 $\mu\text{g}/\text{m}^2$ had lung burdens varying from 5 to about 25 pCi ²³⁹Pu per animal. The lung burdens in jackrabbits (Lepus californicus) sampled from plutonium contaminated areas generally were found to be 2 to 5 times higher than those found in kangaroo rats. Hematopathologic studies disclosed no abnormalities which could be attributed to the available ²³⁹Pu burdens. Gross anatomic lesions were minimal and nonspecific and in no way incriminatory of radiogenic damage. While these findings are indicative of only three species of wild animals living in the desert ecosystem, one can hope that the inhalation problem from residual plutonium contamination also would be as insignificant in other kinds of terrestrial environments as it appears to be at the Nevada Test Site. Additional information on wild animals is needed to help interpret the significance of findings from field surveys as a means of predicting potential hazards from plutonium dispersed into the environment. (46)

DECONTAMINATION OF TERRESTRIAL ENVIRONMENTS

In addition to an evaluation of potential health hazards, certain economical and political considerations will influence any decisions concerning the decontamination of terrestrial environments. Prudence should prevail in such deliberations until the environmental factors are adequately considered, inasmuch as the disturbance from decontamination measures might cause greater decimation than would the contaminant left in place. Prudence is especially called for when considering residual plutonium contamination, because of its low-order entrance into the ecological cycle. (19, 27)

One of the best examples of effective, large-area decontamination occurred at Palomares, Spain, where plutonium had contaminated part of a small village, a hillside, and some irrigated agricultural land producing alfalfa and tomatoes. (8, 28) After contamination contours were established, all crops were stripped from fields and destroyed where readings above $5 \mu\text{g}/\text{m}^2$ were observed. Areas with readings between 5 and $500 \mu\text{g}/\text{m}^2$ were plowed to a depth of at least 10 inches to dilute the contamination and lower the probability of resuspension. Areas with readings greater than $500 \mu\text{g}/\text{m}^2$ were stripped of top soil. In all, the top soil was removed from about 2 hectares and vegetation was removed from about 250 hectares which was subsequently plowed. Contamination of $50 \mu\text{g}/\text{m}^2$ or greater was worked into the surface by hand in areas too rough to plow. Follow-up ecological studies indicate that no health hazard exists from the intake of agricultural produce grown in the areas where plutonium was dispersed. Fowler and coworkers (29) calculated that, after decontamination measures had been taken, a person would need to ingest daily 10^3 kg of fresh tomatoes or 28 kg of dry maize seed from plants grown in this soil in order to obtain an amount of

plutonium equivalent to the maximum permissible concentration in drinking water (MPC_w).

Decontamination measures of the kind used for agricultural land would not be very useful in wilderness areas because of subsequent soil erosion and water-shed control problems. Revegetation by natural means is difficult in many ecosystems, if not impossible, from a practical point of view. Re-planting is sometimes possible but not always economically feasible. Unquestionably, the denuding of a wilderness area of its natural vegetation would extract the greatest biological cost and should be considered only as a last resort. It should be remembered that, in most cases, the natural vegetation would not be destroyed even if an acute plutonium inhalation hazard destroyed some wild animal life. Plant uptake of the residual plutonium would be too low to cause any hazard, and the native vegetation cover would tend to lower the probability of resuspension. It appears then, from an ecological point of view, that low-level residual plutonium contamination dispersed into wilderness areas might prove best left alone and, if they contain contamination levels greater than would be safe for lifetime occupancy, ^{they} should be restricted from public domain. Conceivably, these would be very small areas.

RESUSPENSION AND REDISTRIBUTION OF RESIDUAL PLUTONIUM

The residual plutonium of greatest ecological significance is that fraction which becomes resuspended in a manner which will enable it to gain entrance into animal life, either by inhalation or ingestion. The meteorological factors and environmental conditions which would influence resuspension in one kind of terrestrial environment also would be active in

another kind, but the magnitude of their influence might differ markedly. For example, the effects of wind on soil particle movement would be different in the dry, sparsely vegetated desert than in the damp, densely vegetated forest. Consequently, no set of standard conditions exists from which to draw definitive conclusions applicable to different kinds of terrestrial ecosystems. It appears, however, that the meteorological and environmental conditions of the desert might represent some of the more extreme conditions one would expect to encounter.⁽⁷⁾ Resuspension factors are used to calculate the potential inhalation hazard to man from residual plutonium contamination. This factor is defined as the ratio of air concentration ($\mu\text{g}/\text{m}^3$) above the contaminated surface to the surface contamination level ($\mu\text{g}/\text{m}^2$).⁽⁸⁾ For the desert conditions of the Nevada Test Site these resuspension factors lie in the range of 10^{-5} to 10^{-7} .^(30, 31)

It is a matter for conjecture how well these resuspension factors might apply to some wild animals, because of their living habits. Most of the desert rodents, for example, dig in the soil and take dust baths which might generate respirable dust particles ($< 10\mu$ dia) within their own microcosm, quite independent of the factors required to resuspend respirable particles to heights above one meter. Nevertheless, it would appear from our scanty data,^(24 - 26) indicating low-order lung burdens of plutonium, that resuspension causes no serious residual inhalation problem for wild animals living in fallout areas at the Nevada Test Site.

Redistribution of residual plutonium onto vegetation ingested by animals appears to be an important continuing process in the desert environment, and probably in other kinds of terrestrial environments. Romney and

others^(26, 32) observed that small mammals, sampled periodically in fallout areas during a 10-year period, continued to have relatively high levels of plutonium passing through their gastrointestinal tracts. They also observed that the inhalation route of entry became less important with passing time, whereas ingestion continued to be a very important route through which ⁹⁰Sr, ¹³⁷Cs, and ¹⁴⁴Ce entered small mammals living in fallout areas downwind from nuclear detonations.

Some indirect biological concentration processes might be manifest in time from local redistribution of residual plutonium contamination. Periodic resurveys of study plots in fallout areas indicate that the initially-deposited fallout debris gradually is moved from bare ground areas by wind and water erosion and redeposited under the nearby shrub clumps. It is within these shrub clumps that most of the life processes function in the desert ecosystems, and it is here that continued observations might well disclose important steps in biological concentration processes within and between the tropic levels.

FOOD CHAIN TRANSPORT OF RESIDUAL PLUTONIUM

Availability from Soil

Any factor which may influence the availability of plutonium in soil would also help control its transfer to plants and animals. The chemical forms of plutonium most likely to be dispersed include extremely insoluble particles and complex polymers which are adsorbed strongly onto soil colloids. (33, 34) Edaphic factors such as soil structure, organic matter content, soil pH, and the amount and kind of clay mineral present, no doubt influence the degree of availability of plutonium; however, their combined effects

probably do not exceed a factor of 10. It appears unlikely that residual plutonium moves downward in the soil in solution form, because of its slow rate of diffusion and leaching by invading groundwater. (35) Fine particles, however, may move with the flow of infiltrating water or by mechanical means. (36) Romney, Mork and Larson (26) observed downward migration of plutonium in undisturbed desert soil to depths varying from 6 to 9 cm during the decade after fallout was deposited. Movement from the surface down into the soil profile results in an attenuation of the surface contamination, and reduces the probability of resuspension, but it gradually brings the residual plutonium more closely in contact with plant roots. (26) This raises the question of whether, by this process and others, the residual plutonium becomes more biologically available with passing time. One might suspect that weathering processes involving microbial activity also could have an influence; however, the extent to which this may occur awaits further investigation.

Plant Uptake

Several workers (26, 37-40) have reported that plant uptake of plutonium through the root system is very limited. The discrimination factor from soils to plants (conc. in plants to conc. in soil) is of the order of 10^{-4} to 10^{-6} , depending on plant species, soil type, and amendment additives. Plutonium seems to concentrate within and on the outside of roots; but there is no evidence of selective concentration in other plant parts. These findings are based upon work with only a small number of food crop plants. More work is needed on native plant species important in wild animal food chains; again, there is a need to determine whether the form of plutonium accumulated in plant tissue is more available than other forms

of plutonium ingested by animals.

Animal Uptake

It should be recognized that because of its low radiation energies, plutonium deposited in the environment constitutes no hazard to animal life unless it enters the body in significant amounts. Undoubtedly, inhalation is the most hazardous route of entry; however, this route is unimportant unless the air contains high concentrations of respirable particles. Laboratory experiments with domestic animals^(40 - 43) indicate that absorption from the lung may vary from less than one percent of the amount deposited in the lung to as much as 10 to 15 percent, depending upon such factors as chemical form, particle size, solubility, respiration rate, etc. The absorbed plutonium tends to concentrate in lymph nodes, lung, liver, and bone.

Ingestion is the primary route of entry involved in food chain transport of residual plutonium dispersed in the environment.^(26, 47) Laboratory studies^(44, 45) indicate that absorption from the gastrointestinal tract is only from 0.002 to 0.05 percent, and nearly all of the plutonium absorbed from the gut (90%) is ultimately deposited in bone. The major source of residual plutonium ingested by wild animals is that which is deposited on the surface of vegetation used in the diet, or that which is picked up directly from the soil while eating and from the fur while grooming.

ADDENDUM

The Plutonium Program of the Nevada Applied Ecology Group

The Nevada Applied Ecology Group of the USAEC Nevada Operations Office, Las Vegas, Nevada was established in 1970 in order to coordinate the environmental

programs necessary to support continued nuclear testing activities, and to provide the mechanism to effectively comply with requirements of the National Environmental Policy Act of 1969, Executive Order 11514, and AEC Manual Chapter 0510. The function of the Nevada Applied Ecology Group, coordinated by the Nevada Operations Office, is a cooperative effort between the Nevada Operations Office and its radiological safety and effects evaluation contractors; Lawrence Livermore Laboratory; Los Alamos Scientific Laboratory; Sandia Laboratories; and other groups such as the Western Environmental Research Laboratory of EPA, the Air Resources Laboratory of NOAA, Department of Commerce, the Laboratory of Nuclear Medicine and Radiation Biology of the University of California Los Angeles and the Battelle Memorial Institute who are responsible for bioenvironmental research and development conducted at the Nevada Test Site and other nuclear test sites.

The distribution, fate and significance of plutonium in the environment resulting from nuclear detonation tests ^{were} / selected as the first problem for study. Consequently, a Plutonium Environmental Studies Program is under development within the Nevada Applied Ecology Group as an interdisciplinary, interorganizational research team effort. The scope of the program is to:

1. Compile and evaluate information on the sources, characteristics, distribution, and subsequent behavior of plutonium that has been discharged into the environment from nuclear testing activities.
2. Inventory and document the distribution, amounts, characteristics, and behavior of measurable plutonium dispersed from testing activities at the Nevada Test Site.
3. Describe the extent and conditions under which plutonium on the

Nevada Test Site and its environs is redistributed, considering the routes and rates of movement and with emphasis on its availability to man.

4. Evaluate and document the potential impact on man from plutonium that is deposited on the Nevada Test Site and its environs.

5. Evaluate effects, if any, of plutonium contamination in the environment on native plants, animals, and ecological systems.

6. Evaluate methods for stabilizing and cleaning up plutonium from the environment and the impact of these measures on the ecology of the region.

7. Determine the need for cleanup of plutonium from the environment.

The Plutonium Environmental Studies Program will specifically function to identify and evaluate environmental and radiological health questions associated with plutonium contamination on the Nevada Test Site and its environs. Initial emphasis is placed upon standardization of analytical methods, delineation of contaminated areas, problems of resuspension and redistribution, food-chain transport and ecological effects.

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