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Contamination Limits for Real and
Personal Property
January - June 1974

by

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CONTAMINATION LIMITS FOR REAL AND PERSONAL PROPERTY

Progress Report for the Period of

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ABSTRACT

A summary is given of the work performed during this period. Studies included continuation of work on contamination transfer and the FIDLER response and a review of a petition on plutonium limits by the Natural Resources Defense Council. In addition, a summary of comments on the proposed soil standard for plutonium is included.

Although primary attention was devoted to the question of surface contamination and the transfer of such contamination to the body or other objects, several developments during the period required some effort on previous studies. The most noteworthy was the petition of the Natural Resources Defense Council to EPA and AEC¹ to reduce the limits for insoluble alpha emitters in the form of particulates by a factor of 115 000. In addition, a number of comments were obtained on the proposed interim standard for plutonium.

Summaries of the various studies conducted during this period are given in this report. As before, it is intended that detailed treatment will be in separate topical reports.

I. REVIEW OF NRDC PETITION

Shortly after issuance of the report, "A Proposed Interim Standard for Plutonium in Soils,"² the Natural Resources Defense Council (NRDC) petitioned the AEC¹ to reduce standards for "hot" alpha-emitting particles (defined by them as particles

containing insoluble alpha emitters in quantities greater than 0.07 pCi) by a factor of 115 000 and establishing a soil level for such particles of $9 \times 10^{-8} \mu\text{Ci}/\text{m}^2$ or roughly one such particle per square meter.³ This was based upon a hypothesis that single particles of this activity or greater have a chance of 10^{-3} to 10^{-4} of producing cancer when lodged in the lung. The discrepancy between this value for soil and that recommended in the earlier document led to an intensive review of the claims supporting the NRDC contention in cooperation with other Groups and individuals at LASL who have an interest in the subject. Current plans are to issue a document providing the results of this review.

While the detailed review indicated many uncertainties and inaccuracies in the evidence quoted, several of the more important conclusions are given below.

1. The Geesaman^{4,5} study, which serves as the basis for the NRDC estimate of particle effects, is based upon long extrapolations from the production of tumors in a very specific structure in the skin of the

rat (hair follicle) to the production of lung cancers in humans. Such quantitative extrapolations are extremely uncertain because of the large differences in response which have been noted in experimental studies between tissues and between species or even strains. Such comparisons between tissues and species are usually made only when all other data are lacking and, then, with full recognition and discussion of the uncertainties involved.

2. Radiation doses from insoluble alpha emitting particles involve tissue quantities on the order of tens of micrograms. The experiments referred to by the NRDC involve quantities on the order of grams. Thus, the "localized" irradiations used as a basis of the NRDC quantitative estimates involve extrapolations in the quantity of tissue irradiated of about 10^5 . Although the range of tissue volumes exposed is small, the rat skin data of Albert^{6,7} used by the NRDC does show an apparent decrease in effect with size of area (and, thus, volume of tissue) irradiated. This effect is not included, even in qualitative terms in the NRDC report. The long extrapolation in tissue volume by the NRDC would be warranted, in view of our lack of information about cell interactions in carcinogenesis, only if other data were not available.

3. There are data directly applicable to the irradiation by particles and their effect as compared to uniform irradiation of a tissue in the literature. Many of these data were cited in the previous study² and these data seem to provide direct evidence that the same amount of energy delivered as particulates is no more carcinogenic than the homogeneous distribution through the tissue. One of the more interesting newer experiments^{8,9} is the preliminary work with 480 hamsters, each receiving 2000 microspheres containing 0.07 to 59.4 pCi of plutonium per sphere lodged in the vascular of the lung. Most of these animals have now died with only two lung cancers resulting. These data, involving

irradiation conditions close to those of concern in the particle case, indicate that the predictions of the NRDC extrapolation are not correct

4. The analyses of the two human cases involve extrapolation from one tissue to another, and are superficial in nature with little real attempt to analyze the information available in any detail.

Based on this study, we believe that the presentations and conclusions in our previous study are still valid.

II. COMMENTS ON THE PROPOSED INTERIM STANDARD FOR PLUTONIUM IN SOILS

The proposal for an interim standard was issued in January 1974 and sent to about 100 individuals with a specific request for comments. In addition, some 150 copies have been sent to individuals requesting the document. A number of comments have been received, nearly all of which seemed favorable. Many pointed out **typographical errors or statements** which were not clear. Others raised detailed technical points on parts of the discussion. All will be very helpful in future presentations. Several, however, raised points which were not covered in adequate detail or were overlooked in the preparation of the document. Such points include:

1. The importance of the ingrowth of ^{241}Am to the eventual hazards of plutonium in the soils was emphasized. While this point was made in Table I and in the brief discussion on pages 3 and 4 of the report, it was largely dismissed in the calculations because of the paucity of data on the metabolic behavior and environmental behavior of the element. However, in addition to the possibly different behavior between plutonium and americium, the commenter noted that for plutonium of the composition given in Table I for plutonium recycled in a light water reactor (16.8% ^{241}Pu), the eventual ^{241}Am activity would be some 20 times the initial alpha activity of the plutonium. Thus, the environmental

consequences of ^{241}Am must be considered along with some specification of ^{241}Pu content. This, of course, implies that some method of measurement of ^{241}Pu may be required.

2. Several individuals commented on the significant soil erosion in the higher rainfall areas of the U.S. with consequent sedimentation in rivers and estuaries. This, in combination with the relatively high uptake of plutonium in the edible portions of certain marine organisms or in non-edible organs used in fish protein concentrates could rival inhalation as a pathway in these areas where resuspension is minimized by ground cover and high rainfall. Further study of this possibility is needed, probably in conjunction with studies of the effects of size of area contaminated and regional terrain and environmental features.

3. Only a few individuals commented on the specification of the top millimeter of soil. These comments were based on the difficulty of measurement and the fact that such a layer would be unstable on any long-term basis. One comment was received on the fact that one could, in theory, infer the concentration in the top layer by measuring the profile and extrapolating. This was considered during the formulation of the standard but attempts to make reasonable fits to the available data were not promising. It is surprising that more comments were not received on this feature of the standard and may indicate that serious consideration has not been given to the application or that the approximations proposed in the document may be acceptable.

4. One interesting comment was received indicating that application of the proposed limits could result in problems with control of contamination inside of nuclear energy installations. This would be due to the tracking of contamination from outside. For example, the concern is that "smearable" levels of 100 dpm per 100 cm^2 in clean zones or that hand and shoe alarm levels would be exceeded. While such considerations were specifically excluded in

the study, they certainly deserve notice and inclusion in final limits, perhaps through the "minimum practicable" approach.

5. The subject of the long-term chemistry of plutonium in the environment and the influence of this on the behavior was mentioned several times with the observation that considerably less is known about this subject than about the toxicity of plutonium. Many of the discussions on this subject are speculative with information only by extrapolations from laboratory or meager field studies. However, interest in the subject is increasing as is the number of studies, and information to provide some modification to account for the environmental "sinks" as well as increases in availability will eventually become available.

It is apparent from these comments, as well as further thinking on the problem, that some modifications of the proposed standard to account more specifically for, at least, broad differences in environmental conditions and sizes of area contaminated may be desirable. As was indicated in an earlier report,¹⁰ some factors such as rainfall, vegetative cover, and broad soil classes could be investigated through data on wind erosion and atmospheric dust loading, but until better data on resuspension and transfer to the body for different particle sizes are available, it is doubtful that all factors could be considered.

III. SURFACE CONTAMINATION LIMITS

In studying the equation formulated in the last report¹¹ for contamination transfers, it was concluded that application would be difficult unless all portions of the problem were better defined in more detail. In particular, it will be necessary to derive models, however general, for the uses of the various types of objects of interest and to relate this model to the transfers of contamination and resulting modes of contamination. It now appears

that the transfer methods of most importance will be the direct movement of contamination to the body and resuspension in the air with sub-classes combining these two. For example, direct transfer to the body or clothing followed by localized resuspension or transfer to another person. A sub-class of the direct transfer results if the object is abraded resulting in potential transfer of fixed contamination.

Radiation exposures can result through a number of different mechanisms depending upon the characteristics of the contaminant and the types of exposure to the contaminated object. Most important of the radiation exposures are believed to be:

1. Direct external radiation from objects contaminated with penetrating radiations, including exposures from close proximity to the object (such as handling).

2. External radiation to the skin from contaminants emitting penetrating radiations following transfer to the skin or clothing.

3. Inhalation of radioactive materials resuspended either from the object or secondarily from other objects which have become contaminated by contamination transfer.

4. Absorption through the skin of those radioactive elements or compounds which can pass the skin barrier to any significant degree. Wounds and absorption through other external mucous membranes must be considered for materials not normally absorbed through the skin but are not believed, at this time, to be controlling.

5. Ingestion of radioactive materials moved to the hands, usually by direct transfer, or to parts of the body where it may be transferred to the mouth on foodstuffs or other materials is a final possibility.

These modes of exposure and methods of transfer will vary widely in importance for the same type and level of contamination depending upon the use of the object and the environment of its use. These factors can vary widely, particularly since many

of the items can be used in various ways and, frequently, in ways which may be hard to foresee. (As for example, the use of certain scrap or machine parts in sculpture.) Further, the routes by which the items reach the hands of the final user will vary depending upon the methods of disposal at the various institutions. It is this wide variability in possibilities which makes any approach to the problem difficult. We are currently attempting a qualitative description of possible uses and methods of disposal for various broad classes of items to see if some generalized types of models cannot be derived which would serve as a basis for classification and calculations. We will then attempt to estimate limits for radioactive materials on the body and these, along with the MPC's for ingestion and inhalation will be applied using the transfer equations from the last report to see if an approach to realistic transfer coefficients can be derived.

IV. FIDLER CALIBRATION

Work continued on application of the equations and program for the Wang 600 calculator in describing the response of the FIDLER (Field Instrument for the Detection of Low Energy Radiation) to various configurations of plutonium and americium in soils. This study is intended to permit better evaluation of the instrument response in a given situation and to define the importance of variables such as soil composition, height of measurement, variations in source size and depth of burial in relation to the standard. This study was started to permit improved interpretation of such readings in the literature in relation to the model developed for resuspension calculations. The results have proven valuable in assessing optimum methods of using the instrument for various configurations.

During this period data were obtained on chemical composition of three soil types,

and a scheme was outlined for accounting for various moisture contents in the calculation of the absorption coefficient. These data were then applied to estimating the expected reading, and variation in reading, resulting from these variables as well as differences in the distribution of the contaminant in the soil profile. A brief study of the use of the instrument in the laboratory to measure plutonium and americium in gross soil indicated the importance of optimizing such variables as distance to sample surface, diameter of sample, and thickness of the sample. For sample sizes less than 20-30 grams and one centimeter spacing between the soil surface and the instrument face, there is an optimum sample diameter for each sample size depending upon the balance between self-absorption as the sample increases in thickness and loss of geometry as the diameter increases. Sensitivities in this application range from 4-5 disintegrations/gram of soil per count for 10-gram samples with a 5-cm radius or 20-gram samples with a 6-cm radius for ^{239}Pu . For ^{241}Am samples thickness can be greater and sensitivities of about 0.1 disintegrations/gram of soil per count are obtained with a 40-gram sample and a 4.5-cm sample radius.

REFERENCES

1. Natural Resources Defense Council, "Petition to Amend Radiation Protection Standards as they Apply to Hot Particles," submitted to AEC and EPA, February 15, 1974.
2. J. W. Healy, "A Proposed Interim Standard for Plutonium in Soils," Los Alamos Scientific Laboratory report LA-5483-MS, January 1974.
3. A. R. Tamplin 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, 1710 N Street N.W., Washington D. C., February 14, 1974.
4. D. P. Geesaman, "An Analysis of the Carcinogenic Risk from an Insoluble Alpha-Emitting Aerosol Deposited in Deep Respiratory Tissue," Lawrence Radiation Laboratory report UCRL-50387, February 9, 1968.
5. D. P. Geesaman, "An Analysis of the Carcinogenic Risk from an Insoluble Alpha-Emitting Aerosol Deposited in Deep Respiratory Tissue: Addendum," Lawrence Radiation Laboratory report UCRL-50387, Addendum, October 9, 1968.
6. R. E. Albert, 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," Rad. Res. 30:525-540, 1967.
7. R. E. Albert, F. J. Burns, and R. D. Heimbach, "The Association Between Chronic Radiation Damage of the Hair Follicles and Tumor Formation in the Rat," Rad. Res. 30:590-599, 1967.
8. C. R. Richmond and G. L. Voelz, M.D., (Editors), "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.
9. C. R. Richmond and E. M. Sullivan, "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. 1-12, May 1974.
10. J. W. Healy, "Contamination Limits for Real and Personal Property, Progress Report for the Period January to June 1973," Los Alamos Scientific Laboratory report LA-5482-PR, January 1974.
11. J. W. Healy and W. J. Smith, "Progress Report: Contamination Limits for Real and Personal Property, July-December 1973," Los Alamos Scientific Laboratory report LA-5579-PR, April 1974.