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A Proposed Interim Standard for Plutonium in Soils

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by

J. W. Healy

This work supported by the US Atomic Energy Commission's Division of Operational Safety.

A PROPOSED INTERIM STANDARD FOR PLUTONIUM IN SOILS

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ABSTRACT

Current standards for controlling health effects from plutonium in the body are discussed. Available information on possible sources of exposure of people living in an area where the soils are contaminated with plutonium is analyzed to arrive at estimates of intake. From these estimates, a recommended interim standard for the upper limit of concentration of plutonium in the soils in inhabited areas is derived. The recommendation is based upon conservative assumptions where information is lacking and further studies should result in revision. The subjects of resuspension, deposition velocity of particles and effectiveness of radioactive particulates in producing lung cancer are discussed in appendices.

I. INTRODUCTION

Plutonium has been utilized and processed in relatively large quantities (hundreds to thousands of kilograms total) in several different countries over the past three decades. It now can be found in small quantities in soils and oceans over the entire world as a result of widespread dissemination from nuclear weapons tests in the atmosphere and one burn-up of a space nuclear power generator containing ^{238}Pu. More localized distributions are found in the immediate vicinity of facilities used for processing plutonium, at the locale of several accidents involving weapons containing plutonium, and in remote areas which were used for safety tests with weapons. In each of these areas, some measure of potential hazard is necessary to enable adequate decisions as to future disposition of the area or any special considerations on habitation or land use restrictions. Thus, it is necessary to have some indication of the degree of hazard associated with various levels of plutonium so that such decisions can be adequately based.

The following study was commissioned by the Division of Operational Safety of the U. S. Atomic Energy Commission to provide an interim or provisional standard for plutonium in the soil to meet this need. It was recognized that the problem of establishing such a standard is very complex due to the many potential mechanisms of exposure from this source and that the data available for detailed definition of the problem are inadequate. However, the necessity of making decisions on acceptable levels requires that some guidance be made available for comparison with measurements made in potentially contaminated areas. To answer this need, it was decided to apply the information now available and to arrive at a standard which, while overly conservative from a hazard

standpoint, would give some guidance in making these decisions. At the same time, it was felt that such a study would permit assessment of the information available so that future research and development programs could be more effectively aimed at the areas of greatest uncertainty.

It is stressed that a conscious effort has been made to err on the conservative side in view of the many uncertainties. (In this case, the conservative side is defined as the over-estimation of exposure from the plutonium in the soils.) In addition, the depth of investigation and the conditions considered have been limited in a number of possibly important areas in an attempt to arrive at some guidance as soon as possible. For these reasons, it is urged that the numerical values derived herein be regarded as truly provisional and not be incorporated into rules and regulations which are difficult to change. It is anticipated that changes in the numbers, and perhaps the concepts, will be forthcoming from future work.

In the derivation of the numerical guidance, we have considered primarily considerations of health and hazard to man. In recent years there has been a tendency to derive such standards based upon the practicality of achievement rather than upon effects on health. While such standards have their rightful place in providing control of sources of pollution, there is a tendency to regard them as safety standards so that exceeding them becomes a matter of great concern. It is also of importance, even on the practicality basis, that an upper limit be clearly established, as based on safety, so that one can assure that the practical limits are, indeed, safe, and that the additional margin of safety attained by lower limits can be assessed in comparison to the costs in resources and manpower of achieving them. With this philosophy we have not provided two standards, one for control of sources on a continuing basis and one for the application of countermeasures in an area already contaminated, although such considerations are appropriate to any safety program and in the application of a standard such as the one derived here. Again, as better information becomes available and we reach a stage where intelligent and informed assessment of actual risks at various levels of plutonium in the soils are possible, such considerations will be included.

Supplemental information on several items not covered in detail in the literature are given in appendices. These include a model for calculating resuspension of particulates in Appendix A, a treatment of the deposition velocity for particles in Appendix B, and a discussion of the effectiveness of radioactive particles in producing lung cancer in Appendix C.

II. PLUTONIUM STANDARDS

A. Properties

Plutonium is not a simple material. It is a man-made element in which the isotopic composition, and thus, the radioactive properties, vary widely depending upon the history of its production and any subsequent neutron exposure as a reactor fuel or in a detonation. There is increasing evidence that the metabolic behavior and, in some situations, the gross chemical behavior may vary with specific activity of the isotope probably because of the influence of the energy emitted as radiation as well as mass effects. The isotopic compositions of several typical mixtures are given in Table I.

The compositions given in Table I are illustrative rather than definitive with wide variations possible, particularly in the materials used for power fuels. The low irradiation material is reasonably representative of that used in the weapons programs of the AEC which has utilized a large fraction of the plutonium produced in the past.

Since the isotopes of plutonium are primarily alpha emitters with little accompanying penetrating radiations, the hazard associated with plutonium is almost completely due to potential intake into the body. Plutonium-241 is a beta emitter but,

TABLE I

PLUTONIUM MIXTURES

Isotope	Principal[1] Emission	$T_{1/2}^{1}$ yrs	Specific Activity Ci/g	Low Irradiation wt %	Pu Recycle in LWR wt %	Heat[2] Source wt %
^{236}Pu	α	2.85	532	----	5×10^{-6}	10^{-4}
^{238}Pu	α	86.4	17.4	0.0115	2.9	80.3
^{239}Pu	α	24,390	0.0614	93.6	39.6	15.87
^{240}Pu	α	6,580	0.226	5.9	25.6	3.00
*^{241}Pu	β (0.02 MeV)	13.2	112	0.4	16.8	0.72
^{242}Pu	α	3.79×10^{5}	3.9×10^{-3}	0.013	15.0	----
^{244}Pu	α	7.6×10^{7}	1.9×10^{-5}	0.02	------	----
Specific activity (alpha) of mixture (Ci/g)				0.073	0.059	14
Specific activity with ^{241}Am ingrowth (Ci/g)				0.084	1.08	14

* Daughter is ^{241}Am, an alpha emitter, with half-life of 458 years and a specific activity of 3.24 Ci/g. This will reach a maximum from the ^{241}Pu in about 70 years with one gram of ^{241}Pu resulting in 2.91 Ci of ^{241}Am.

again, the energy of the beta particle is low enough that self-absorption and small penetration into the body makes the external dose insignificant. It is true that massive quantities of plutonium, as encountered in fuel fabrication plants or other facilities handling large quantities of plutonium, pose some problems in control of external exposures to workers, particularly as the quantities of isotopes of higher mass than 239 increase in heavily irradiated fuel materials. However, in the quantities expected in soils, these external radiations are of no significance in comparison to an internal uptake. Thus, our concern with the properties of plutonium is limited to those which will influence intake.

On an overview basis, plutonium is probably not as bad an actor in the environment as many other isotopes because of its relative insolubility. As a result, it is not taken up to any large extent in the ecosystems so that transfer by biological mechanisms is usually minimal particularly for plutonium in soils. Although there are measurements indicating some concentration in marine organisms,[3,4] none seem to indicate anything other than biological discrimination in plants and animals on contaminated soils. It must be noted, however, that experience in this regard is relatively limited and some mechanisms for biological uptake in terrestrial situations may occur, even if only in limited areas where the soil and biological conditions are proper. For example, the action of natural chelating agents in the soils may result in compounds which could be biologically more active. However, with the information now available, it appears that for purposes of this interim standard, the physical modes of transport and intake are of the most importance.

One further reservation on the behavior of a mixture of isotopes in the environment relates to the eventual buildup of ^{241}Am. This isotope begins to appear in significant quantities from ^{241}Pu mixtures within a few months to years. While the assumption is frequently made that all of the transuranic elements have similar metabolic behavior (as in ICRP 2),[5] this assumption was based primarily on the need for MPC's to be used for control purposes. The chemical properties of americium

are, indeed, different from those of plutonium particularly in the tendency of plutonium to produce insoluble polymers and one would expect the ecological behavior to be different. Some measurements have indicated a much higher uptake of ^{241}Am by plants[6] while others have shown the transfer from plasma to milk[7] to be higher than for plutonium. Since the contaminated areas now of interest have resulted from plutonium with low ^{241}Pu content, this has not been an important consideration. As information becomes available, the importance of this factor to the interim standard will be assessed.

B. Basic Limitations on Plutonium in Humans

As a basis for the potential harm to humans from the intake and deposition of plutonium we will use the current standards as recommended by the NCRP and ICRP. These were basically derived for occupational exposures and are applicable primarily to adults in reasonably good health. In application to populations they are reduced to allow for the lower risk which should be applicable to such groups and to provide a margin for children or ill individuals. A brief review of the origin of the occupational standards is given in this section to provide a basis for the application to population groups in the next section.

It will be noted that we have not based our studies on estimates of the risk to individuals in spite of the fact that this approach is advocated by many people. Such estimates, even for low LET radiation, require many assumptions and are based upon data which have a wide range of uncertainties. As a result, the estimates reflect more the individual assumptions and interpretations than they do the real risk. There is a wide difference between arriving at a value which the evidence indicates is "safe" without attempting to quantify this term and in providing a quantitative, numerical value for the risk. In the former case, the informed judgment of people who have studied the information available can be used. In the latter case, a mechanistic calculation is substituted with judgments on the assumptions compounding the uncertainties in the final number. It is true that value judgments as to "how safe is safe" are required for the non-numerical method but the general agreement among bodies as diverse as the NCRP,[8] the ICRP,[9] and the Federal Radiation Council[10] would indicate a remarkable similarity in such value judgments in spite of the differences in objectives and composition of these groups. In the case of alpha emitters, such as plutonium, we would also note that the uncertainty in the risk estimates may be greater than for low LET radiation because of the uncertain RBE to be applied and the apparent lack of repair of damage from these high LET radiations. (Note that the rem should not be applied in such risk estimates since this unit is defined for use in radiation protection and uses the Quality Factor which is arbitrarily assigned as based on a conservative estimate of all effects.)[11] The rem is intended for control of radiation exposures and not for estimates of risk.

1. Body Burden. The basic standard for plutonium absorbed into the body (i.e., outside of the lung or other site of initial deposition) is 0.04μCi for occupational exposure. This value was derived by biological comparison of the late effects when injected into animals with those of radium for which a significant body of information on the effects in humans exists. A recent review of the derivation of this value and its application to obtaining maximum permissible concentrations was made by Langham and Healy.[12] The value resulted from the work of Brues[13] at the Argonne National Laboratory, in which known quantities of both plutonium and radium were injected into animals and the comparative late damage noted. As a result of these experiments, it was determined that the relative toxicity of plutonium is about 15 times that of radium-226 on the basis of equal injected doses (microcuries). In the rodents used, the retention of plutonium was about 75% while that

of radium was about 25%. On a retained basis, this would lead to the conclusion that plutonium is about five times as toxic per microcurie; however, a large part of the energy delivered from radium results from the radon daughters. In these animals the retention of the radon was about 15-20% as compared to about 50% for man. Thus, in man, the higher radon retention would lead to expectation of increased damage for the radium in comparison to the plutonium. The relative toxicity of the plutonium per microcurie retained would be expected to be lower by about a factor of two or about 2.5 times that of radon. Thus, for man, the maximum permissible body burden is 0.04 microcuries. However, on an energy delivered basis, the energy from the plutonium alpha particles is five times as toxic as that from radium since the total energy from the radium is about twice that of the plutonium in man. The increased effectiveness of the plutonium energy has been attributed to the fact that the plutonium is not as uniformly distributed through the bone matrix as radium (although radium is not uniformly distributed) tending to concentrate on the surfaces so that a smaller portion, and perhaps more sensitive portion, of the bone receives a higher insult. Similar experiments performed at the same time with ^{89}Sr gave results similar to these and the increased effectiveness of these two materials on an energy delivered basis has been generalized to the "dose distribution factor" of five which has been applied to all bone seekers except radium.

The value of 0.04 μCi was first derived at, and immediately following, the Chalk River Conference in 1949[14] and still remains as the primary standard for plutonium in the body. Additional studies with dogs at the University of Utah[15] have essentially confirmed the number although the Utah results indicate that the relative toxicity on an energy basis may be closer to ten than five and have demonstrated that other organs may have significant uptake and retention of plutonium depending upon the path of administration. In their experiments, the plutonium was administered intravenously in a citrate solution and the liver appeared to be a major site of deposition although the majority of late effects noted seemed to be primarily involved with bone. Studies of the effects of plutonium on animals continues and it is anticipated that some revision of the 0.04 μCi value may occur in the next few years, but a major change, for reasons of health effects, is not anticipated. (The qualification on health effects is necessary since there is a growing tendency to base such standards on practicality of attainment rather than potential damage. For example, the FRC recommendations[16] for the intake of ^{226}Ra and ^{90}Sr are based on their conclusion that operations can be carried out without exceeding the recommended intake. In application of these standards it is important to recognize the basis.) There are now some human data[17] based on exposures of 27 individuals in 1944 and 1945 (28 years). Estimates of the body burden by urine analysis are uncertain, but the latest analysis of the data indicates that 60-70% of the individuals have plutonium burdens at or above the 0.04 μCi level with the maximum individual perhaps 5-10 times this value. Followup medical examinations have shown no changes which could be attributed to this plutonium. While the sample is small and the time is relatively short in comparison to the life span of man, these data are encouraging in that they indicate no gross problem such as occurred with radium.

It should be noted that this derivation is based directly on biological evidence of damage and does not utilize the concept of radiation dose except indirectly in the comparison of energy delivered by the two materials. There has been an attempt to fit the derived value into the overall framework of dose calculations with the result that the original basis for the number and the meaning of the derived numbers is not always clear. For example, in their 1959 report on internal emitters,[5] the ICRP presented the concept as follows: "The effective

RBE dose delivered to the bone from internal or external radiation during any 13 week period averaged over the entire skeleton shall not exceed the average RBE dose to the skeleton due to a body burden of 0.1 μCi of ^{226}Ra (derived from a dose rate of 0.06 rad/week, an RBE of 10 and n = 1)." In this statement, n is the so-called dose distribution factor and corresponds to the value of five on an energy basis derived from the reasoning described above for plutonium. The dose rate from 0.1 μCi of ^{226}Ra retained in the body was obtained assuming that 99% of the radium in the body was in the bone, the mineralized portion of the bone weighing 7000 grams was the appropriate organ, 30% of the radon daughters were retained in the bone and a quality factor of ten was appropriate to describe the LET effects of the alpha particles. In this calculation, 0.04 μCi of plutonium in the body with 90% in the bone would deliver an average dose rate of 0.5 rads per year to the mineralized portion of the bone or, with a quality factor of ten and a dose distribution factor of five, about 25 rems per year which, within the accuracy of the estimate, is the same as the radium value of 30 rems per year.

2. Lung Burden. The basic limitation to the lung for workers is a dose equivalent rate of 15 rems per year as derived from the experience with external radiation exposure and the application of the critical organ concept first set forth by the NCRP.[18] This translates, for a 1000 gram lung, to a lung burden of 0.016 μCi of plutonium based on the average dose to the entire lung. However, in contrast to plutonium mobilized into the body which is retained with great tenacity, the lung has elimination mechanisms which serve to remove plutonium or other materials. As a result, the total dose delivered by a given deposit is limited by the time of retention of the material in the lung. In addition, the deposition of material in the lung is strongly affected by a number of factors, the most important of which is undoubtedly the effective particle size. The ultimate fate of the material deposited in the lung must be considered in relation to the radiation dose received by other parts of the body. These questions of intake and retention will be discussed in the next section of the report.

The applicability of a dose calculated on the basis of the average dose to the lung (i.e., total energy delivered divided by the total weight of the organ without regard to the distribution of the energy within the organ) is frequently questioned on the basis that the plutonium particulates will produce "hot spots" where the local radiation dose far exceeds the average. These high doses to limited volumes of tissue are, then, presumed to constitute a high risk. A recent review of the information available on this question[19] (reproduced here as Appendix C) indicates that the experimental data available, while not completely adequate for low activity particles, strongly supports the finding that the non-homogeneous distribution of dose is probably less hazardous than the uniform dose. Nearly all of the support for the increased effect of single hot particles arises from theoretical calculations of doses to individual cells with the cell response assumed from experiments on other types of cells in different configurations of dose distribution. The evidence for cancer induction from limited volume irradiation strongly indicates that a calculation of the dose on the average organ basis is conservative if the irradiation is from particulate sources. For this reason, we will use the average organ dose throughout for the lung.

C. Application to Population Groups and Individuals

It is generally recommended that exposure of population groups or individuals in the population be limited to values below those recommended for occupational workers. However, there are some differences in the recommendations of various groups as to the exact degree of reduction to apply. A brief review is given to aid in choosing the limits to be applied in this work.

The ICRP position as of 1965[9] recommended that the annual dose limits for members of the public be one-tenth of the corresponding annual occupational limit with the exception that the thyroid dose to children under the age of 16 be limited to 1.5 rems rather than the previously used 3.0 rems. The occupational limit listed for "bone" is given as 30 rems per year and for "all other organs" as 15 rems per year. Thus, the dose limit for plutonium in the body would be 0.004 μCi (assuming the rem is calculated as given earlier) and the maximum quantity in the lung would be 0.0016 μCi. For genetic exposure, they recommend a maximum of 5 rems in 30 years or an average for a population group of 0.17 rems per year. However, for the somatic dose of concern here, they state "--it is expected that the dose limits for individuals will ensure that the number of somatic injuries that could possibly occur in a population will remain at a low level." From this, it appears that they did not feel that a specific limit for groups, based on somatic effects, was necessary.

The current recommendations of the NCRP[8] provide dose limitations based on somatic considerations for individuals and for the average population dose. These are given as: "The dose limit for the critical organs (whole body) of an individual not occupationally exposed shall be 0.5 rem in any one year---" and "The dose equivalent to the critical organs (whole body) for the population of the United States as a whole from all sources of radiation other than natural radiation and radiation from the healing arts shall not exceed a yearly average of 0.17 rem (170 mrem) per person." This establishes the population group of interest as the entire population of the country with, presumably, averaging of the dose permitted over this group as a whole. Under these conditions, it is apparent that the dose to the individual is strongly limiting for any accidental or industrial situation where the sources of exposure are relatively limited in number. The definition of the 0.5 rem and its application to plutonium is not clear. If one takes it at face value using the definition of the rem as including the "dose distribution factor" of five derived from the comparative experiments with radium, then the maximum body burden for an individual becomes $(0.5/30) \times 0.04 = 0.0007$ μCi or about one-sixth of the ICRP value for the individual. However, referring back to the statement on occupational exposure we find the words: "For the purposes of this recommendation, the critical organs are considered to be the gonads, the lens of the eye, and red bone marrow." and "--assessed in the gonads, lens of the eye or red bone marrow--." In paragraph 202, with respect to other organs, they indicate; "Detailed discussion is left to other NCRP reports." Thus, it appears that the current NCRP recommendations do not apply to the case of an organ such as the bone, where the damage of concern does not include the red bone marrow or other specifically designated critical organs. In the previous report on internal emitters,[22] the position was: "--The maximum permissible average body burden or radionuclides in persons outside of the controlled area and attributable to operations within the controlled area shall not exceed one-tenth of that for radiation workers." While it appears clear that the NCRP intends a lower average for the population than for the individual, the best interpretation of their recommendations at the moment seems to be 0.004 μCi in the body of an individual.

The Federal Radiation Council has considered the problem of population dose both with respect to external radiation[10] and the somatic dose from radium-226[16] specifically. For environmental contamination they point out that there may be conditions where the only data available may be related to average contamination or exposure levels. They then suggest the use of an arbitrary assumption that the majority of individuals do not vary from the average by a factor greater than three. From this, and their recommendations of 0.5 rem whole body radiation for individuals, they obtain a value

of 0.17 rem for yearly whole body exposure of average population groups. They also warned that the use of the average figure, as a substitute for evidence concerning the dose to individuals is permissible only when there is a probability of appreciable homogeneity concerning the distribution of dose within the population included. For radium, they rejected the use of the factor of ten between the occupational exposure limit and that for the individual in the population because of the differences in characteristics of the child, the longer time for carcinogenesis and the difference in distribution of the radium in the bone from an environmental accumulation over a number of years and the acute type of exposure from the worker exposures. They noted that the dose to the skeleton from all natural causes averaged between 0.1 and 0.15 rads per year while the quantities of radium and its daughters required to give comparable doses were about 0.003 to 0.005 μgm. They also compared the natural occurrence of radium in the skeleton which they quote to range from about 0.001 μgm to some two or three times this amount in most areas of the U.S. In considering the dose to the bone they state: "There is insufficient information on the relative biological effectiveness of the radiation from radium to attempt a realistic conversion of this dose in rads to the skeleton from radium and its decay products into rems." They, thus, specifically reject the conversion of the body burden into dose equivalent as a basis for deriving or expressing limits to the bone. In considering operations involving the release of radium to the environs, they feel that such operations can be carried out in such a manner that the average daily intake in an exposed population group will not exceed 20 pg. They also quote that data on the average intake and average body burden indicate that the quantity of radium in the adult skeleton does not exceed a value of about fifty times the daily intake. They then chose a value for the daily intake of 20 pg per day as the radiation protection guide with an alternate value for individuals in the general population of 0.003 μg in the adult skeleton. For a suitable sample of the exposed population, the average value was set at 0.001 μg in the adult skeleton. It can be seen that the factor of fifty for the body burden of the average individual as compared to the intake, when applied to the RPG of 20 pg/day intake, corresponds to the value for the average of the population group, while the value for the individual is three times this. Application of these values to the plutonium case, again selecting a factor of five for the dose distribution factor would indicate that one could permit only 0.0012 μCi in the body of the individual or about 0.0004 μCi in the adult skeletons of a suitable sample of the population. However, it is also noted that the value was selected on the basis of a finding that operations could be conducted with radium at this level but the same finding has not been made for plutonium. The direct application of this recommendation is therefore in doubt.

For the purposes of this document it appears appropriate to consider an upper limit for deposition in the body of an individual in the population of 0.004 μCi of plutonium and one-third of this value as applied to a suitable sample of the population as defined by the FRC. This will result in an average dose rate to the mineralized portion of the bone of 0.03 to 0.06 rads per year or, using a quality factor of ten for the alpha particles and a dose distribution factor of 5, a dose equivalent rate of 1.5 to 3 rems per year depending upon the fraction of the plutonium deposited in bone as compared to other organs. The lung dose will be based on a limit of 1.5 rems per year (0.15 rads/year) calculated on the basis of an average dose to the entire lung. As noted earlier, this method of calculation is believed to be conservative in control of actual damage.

III. UPTAKE AND RETENTION IN THE BODY

The application of the foregoing standards for the maximum quantity permissible in the body is usually done through "maximum permissible

concentrations" (MPC's) for air and water to be breathed or ingested. These are derived by considering the uptake and metabolic patterns of the isotope in the body. Such MPC's have been given primarily for occupational exposure and, for the values currently in use, the models used for describing the retention and elimination are outdated. For these reasons, we have chosen to review the current information and arrive at independent assessments of the proper intake levels appropriate to the population exposure rather than to rely on published MPC's. The reasons for this decision are discussed in this section of the report along with the derivation of values to be used.

A. Inhalation

The current MPC's recommended by the NCRP[20] and the ICRP[6] were calculated by the use of a simple lung model which dates conceptually back to the Chalk River Conference in 1949.[14] This model differentiates between "soluble" and "insoluble" materials without, however, any definition of the terms other than their assumed behavior in the body. For soluble materials, it was assumed that 25% of the material is retained in the lung and absorbed rapidly into the bloodstream from which it is deposited in other organs of the body, the remainder is eliminated by exhalation or ciliary action to the throat. For insoluble materials, it was assumed that 25% is exhaled with 50% deposited in the upper respiratory passages and subsequently eliminated by ciliary action and swallowed. The remaining 25% is deposited in the deep lung with one-half of this eliminated from the lung and swallowed within the first 24 hours. The remainder (12.5%) is retained in the lung with a half-life of 365 days (for plutonium) with this portion assumed to be taken up by body fluids. Thus, on this model, the inhalation of one microcurie of material will result in the deposition in the lung for long term retention about 0.125 μCi. In general, the components retained for shorter times are ignored in the dose calculations because of the relatively small dose which they will deliver over the period of elimination. In this model, a deposit of 0.016 μCi in the long term retention compartment will then deliver $15/\lambda$ or 15 x 1 year/0.693 = 22 rems over the period of elimination. However, the material taken up by body fluids remains to be accounted for. If one assumes that all of this material goes to the bloodstream and is later deposited according to the pattern for soluble material, then the uptake to the body becomes limiting and not the lung dose. On this basis, the MPC for insoluble material should be about twice that of the soluble since the uptake by the blood is considered to be only half of that of the soluble. In practice, the MPC for the soluble material is about 0.06 times that of the insoluble because the insoluble value was calculated based only on lung dose without consideration of this fraction taken up into the body.

However, it is known that not all of the material retained by the lung eventually passes into the bloodstream. Instead a major portion is taken up by the lymph nodes which drain the lungs. This has been demonstrated by autopsy on individuals[21,22] who have inhaled plutonium as well as by animal experiments.[23] In response to this, as well as to improved information on the overall deposition and retention of various materials, a Task Group working under the auspices of the ICRP[24] has described a more definitive lung model which provides in some detail the variation in retention in various parts of the lung with particle size and gives some indication of the fate of the materials deposited in various parts of the respiratory tract. Although this lung model has not, as yet, been adopted by the ICRP, and there apparently will be some changes when issued, it is useful for indicating the relative comparison between the older model used for calculating current MPC's and these more refined considerations. The model provides curves for estimating deposition in three regions of the respiratory tract depending upon the particle size.

It then provides three clearance classes depending upon the rate of pulmonary clearance: Class Y - those materials retained in the lung for long periods, perhaps years; Class W - those materials with intermediate retention on the order of weeks; and Class D - those materials rapidly cleared. Classes Y and D correspond to the "insoluble" and "soluble" materials considered in the earlier lung models. Although the Task Force, presumably because of the lack of detailed studies of the behavior of various compounds in the lung, implies that certain of the chemical compounds of plutonium may belong to Class D, the tendency of soluble compounds of plutonium to hydrolyze in body fluids and, in some forms, to produce colloidal polymers would indicate that even the more soluble compounds should be in Category W rather than D. This is at least, partially confirmed by the studies at Hanford using Beagle dogs in inhalation of the nitrate and the fluoride.[23] Here pulmonary retention times of 100 to 200 days were observed.

A summary comparison of the lung model used by the ICRP in deriving the present MPC's with the Task Force model for several particle sizes is given in Table II along with the MPC's evaluated for a worker exposed 168 hours per week. Although the Task Group chose a value for the half-life in the pulmonary region for the Class W plutonium of 38 days as based on early studies with nitrate, we have retained their general 90-day half-time for this class on the basis of the studies with dogs cited earlier. In general, there are no really striking differences apparent in this comparison, although the inclusion of the uptake in the body for the insoluble calculation eliminates the former wide discrepancy between the "soluble" and "insoluble" concept.

This discussion was presented to illustrate the uncertainties which exist in estimating the deposition and transfer of material from the lung. In general, it is concluded that the MPC for soluble compounds as calculated on the old lung model may be somewhat conservative in estimating the buildup of plutonium in the body. On the other hand, it does not fully account for the final site of deposition since both the injection experiments at Utah[15] and the inhalation experiments at Hanford[23] indicate that considerably less than 90% of the plutonium in the body is in the bone with the liver (and lymph nodes) as the major alternate sites of deposition. Since the effects on the bone still predominate in the Utah experiments, however, this partition means that less energy will be deposited in bone compared to that in the total body since the fractional bone deposition is smaller. While some concern may be felt for use of the insoluble MPC in some situations because of the lack of accounting for the movement into the body, the results from the newer lung model would indicate that the transfer from the lung to the blood may be on the order of a factor of three to ten lower than was considered on the older model so that overall buildup even at the higher MPC should not exceed the body burden limits. However, it is noted that considerable uncertainty exists with respect to the initial deposition in the lung because of the lack of data on particle sizes in the usual situation. This will be serious only in the very small particle sizes where the deposition will be increased. On the other hand, even for particles of 0.1 micron size, the pulmonary deposition is predicted by the new model to be only 50%, a factor of two higher than was used for the one micron particle. In view of the many other uncertainties, including the uncertainty in the dose limitation to the lung, such a factor is of little real significance, particularly when the conservative nature of the present MPC's is considered.

For application to the public, it is believed to be inappropriate to use two limits based on the "soluble" and "insoluble" concept without considering the interactions between the two. Possible values of the MPC for an individual in the population based on lung dose of 1.5 rems per year as

TABLE II

COMPARISON OF LUNG MODELS (INHALED BASIS)

Model	Pulmonary Deposition %	Long Term Retention %	Half-life da	To Blood %	To Lymph %	(a) MPC's Lung μCi/cc	(a) MPC's Body Burden μCi/cc
ICRP - "Soluble"	25	---	---	2.5	-----	-----	6×10^{-13}
ICRP - "Insoluble"	25	12.5	365	12.5 (?)	-----	1×10^{-11}	-----
Task Force - Class Y							
0.1 μm	50	30	500	3.34	(b) 7.5	4×10^{-12}	4×10^{-12}
1 μm	25	15	500	2.0	(b) 3.75	8×10^{-12}	6×10^{-12}
5 μm	12	7.5	500	1.61	(b) 1.8	2×10^{-11}	7×10^{-12}
Task Force - Class W							
0.1 μm	50	30	90	10.9	(c) 2.5	2×10^{-11}	1×10^{-12}
1 μm	25	15	90	8.8	(c) 1.25	4×10^{-11}	1×10^{-12}
5 μm	12	7.5	90	10.7	(c) 0.6	9×10^{-11}	1×10^{-12}

(a) For worker - 168 hour per week

(b) 10% of this transfers to blood with 500 da $T_{1/2}$ (included in blood)

(c) Transfers to blood with 90 da $T_{1/2}$ (included in blood)

recommended by the ICRP, 0.5 rems per year as recommended by the NCRP or a total deposition of 0.004μCi in the body are given in Table III as adapted from Table II.

Since it appears unlikely that there would be significant airborne concentrations of the Class W compounds in pure form from resuspension and processes of agglomeration in the soil could result in relatively large average particle sizes, an MPC in air of 3×10^{-13} μCi/ml applicable to both classes would appear to be appropriately conservative.

TABLE III

INHALATION MPC'S FOR AN INDIVIDUAL IN THE POPULATION
μCi/ml

	Lung 0.5 rems/yr	Lung 1.5 rems/yr	Uptake in Body
Class Y			
0.1 μm	1×10^{-13}	4×10^{-13}	4×10^{-13}
1 μm	3×10^{-13}	8×10^{-13}	6×10^{-13}
5 μm	6×10^{-13}	2×10^{-12}	7×10^{-13}
Class W			
0.1 μm	7×10^{-13}	2×10^{-12}	1×10^{-13}
1 μm	1×10^{-12}	4×10^{-12}	1×10^{-13}
5 μm	3×10^{-12}	9×10^{-12}	1×10^{-13}

B. Absorption from GI Tract

Plutonium is only slightly absorbed from the GI tract when ingested so that intake with foods or other materials through this path is not usually considered to be a limiting method of exposure. In rats, chronic ingestion at low mass concentrations of the nitrate resulted in an average uptake of 0.003% of that fed with 90% of the small fraction which was absorbed deposited in the skeleton.[25] It was estimated with a 90% confidence level that the retention did not exceed 0.01% in 99% of the rats. A similar absorption of 0.002% was noted in pigs following feeding of pH2 nitrate solution.[26] The MPC in drinking water of the NCRP[20] and the ICRP[5] for so-called "soluble" plutonium is based on an uptake of 0.003%.

The uptake from the GI tract can be affected by the presence of complexing agents, the valence state of the plutonium and the age of the animal. The variation with valence state and the presence of citrate is shown in Table IV as obtained from Thompson's review.[27] Thompson also reported experiments by Carritt et al in which the absorption of nitrate in rats was increased from 0.01%

TABLE IV

ABSORPTION OF PLUTONIUM FROM SEVERAL SOLUTIONS FED INTRAGASTRICALLY TO RATS

Principal Anion	pH	Type of Solution Fed: Plutonium Valence State Identified % (III)	(IV)	(VI)	Plutonium Retained 4 Days after Single Feeding, %
Nitrate	1		68		0.28
Nitrate	1			100	1.9
Nitrate	2	90	10		0.006
Nitrate	2	7	93		0.005
Nitrate	2		96		0.0013
Nitrate	4		97		0.0017
Citrate	2		99		0.03
Citrate	2		96	4	0.29
Citrate	2		85	15	0.41

in the absence of citrate to 0.3% with 5% sodium citrate.

In one day old rats, the absorption of plutonium from a pH2 nitrate solution averaged 0.25%. This absorption dropped to 0.1% at 7 days of age, to 0.02% at 21 days and to the adult value of about 0.003% at 33 days of age. [27]

Although these uptakes are low in most normal situations, they cannot automatically be dismissed in all environmental situations. Romney et al, [28] for example, report data on the plutonium content of the lung, GI tract and bone of kangaroo rats and jackrabbits at the Nevada Test Site where they had been living in areas contaminated with plutonium. Data from the animals taken from the higher contamination areas are reproduced in Table V.

TABLE V

PLUTONIUM IN ORGANS OF ANIMALS LIVING IN CONTAMINATED AREAS

Station	Bone dis/min	GI Tract Contents dis/min	Lung dis/min	[a] Transferred to Bone per Year dis/min
Kangaroo Rats 1958				
11D	7.13	2052	11.40	22.5
13-2	4.30	1255	0.12	13.7
Kangaroo Rats 1966				
11D	47.05	1050	61.28	11.5
13-3	2.72	170	5.80	1.9
Jackrabbits 1958				
13-1	128.48	5.5×10^5	57.50	6000
13-2	11.68	3.2×10^4	0.36	350
13-3	1.75	5712	0.24	63
Jackrabbits 1966				
11D1	665.40	4.1×10^4	98.25	450
11D2	88.76	1.6×10^4	8.92	175
13-3	19.27	1360	1.92	15
13-5	2.34	781	0.10	9

[a] Assuming GI content measurement represents one day feeding and 0.003% per day transferred to bone.

At first glance, the bone values appear to be high considering the low absorption of plutonium. However, the high GI tract contents indicate the possibility of ingestion of considerable amounts of soil so that a large quantity of plutonium is available for transfer. In the last column we have calculated the amount of plutonium which would be expected in the bone after one year considering that the GI tract contents represent one day's intake and 0.003% of this quantity is transferred to the bone each day. Even ignoring any absorption from the lung, it can be seen that, within the accuracy of the estimate, the apparently high bone values can be accounted for on this basis. It may also be noted that these values, even though significant, should be of little concern in a predator food chain because of the low uptake from the GI tract of the predator.

The effects of unabsorbed plutonium passing through the GI tract have been studied in acute administrations to rats. [27] A dose of 88 mCi/kg of nitrate caused death in the first day, apparently from effects other than radiation. Doses of 56 mCi/kg did not produce grossly evident damage. Oxide doses as high as 230 mCi/kg produced no gross evidence of damage while 155 mCi/kg produced transient histological changes in the cecum and colon which appeared three days post administration but not at six days. These data have indicated that the alpha radiations do not penetrate to the sensitive tissues of the GI tract with any efficiency and serve as the basis for the ICRP and NCRP assumption that only 1% of the alpha energy at the surface of the GI tract contents is effective in producing a dose to the GI tract.

The foregoing data would indicate that the 0.003% absorption from the GI tract chosen for calculation of the MPC's for occupational exposure is appropriate for this use. However, for the environmental exposure of the public in situations such as living in a contaminated area where exposure can be continuous, both the higher absorption by children and the possible effects of combination of ingestion along with foods containing various additives such as citrates, preservatives and even chelating agents must be considered. For the young rat, absorption above 0.1% was in the first week of life corresponding approximately to the age of the human baby when motility is low and the environment is relatively carefully controlled so that access to ingestion by routes other than foods is small. The high uptakes with citrate occur with high acidity and significant percentages of the plutonium in the +6 valence state both of which are unlikely to occur with any degree of regularity under normal conditions. Thus, it is concluded that an uptake about ten times larger than that used by the ICRP for occupational exposure and about one-tenth of the highest values noted for very young animals or citrate complexed plutonium would be reasonable and, at the same time, relatively conservative particularly for the relatively insoluble forms of plutonium expected to occur in the environment. This would, then, be an uptake of about 0.03% and would apply particularly to the most susceptible group, children between the ages of about one and ten years.

C. Skin Absorption

Although the intact skin serves as an excellent barrier against the passage of plutonium on its surface, a small rate of absorption through the skin can occur. Such rates are insignificant for most cases of sporadic, infrequent skin contamination but we must consider the possibility of long term accumulation from living in a contaminated area where a continued maintenance of some level of contamination on the skin can be assumed.

Data on the absorption of plutonium nitrate from 0.1 N acid solution on rat skin indicates absorption rates of $2 - 30 \times 10^{-5}$ percent per minute over periods of 15 minutes to one day.[29] When applied in a mixture of tributyl phosphate and carbon tetrachloride with traces of nitric acid, the initial rates were up to ten times higher, with indications that higher rates were maintained through at least five days. Human data are meager and may indicate somewhat lower absorption rates as could be expected from data on other materials with several species of animals as compared to humans.

In deriving skin contamination limits for control purposes, a rate of penetration of 10^{-5}% per minute was used for plutonium based upon an examination of available data.[29] This primarily relates to contamination resulting from solutions rather than the more insoluble particulates. However, the possible effects of agents such as lotions, detergents, and various household chemicals have not been examined to see if they could have a possible effect of increasing the penetration. One would expect the plutonium in soils or the environment to be initially in the form of insoluble oxide or firmly attached to other particles so that the skin absorption should be lower than for the solutions. In view of the uncertainty of possible effects of other agents, however, the absorption rate of 10^{-5}% per minute will be used as a conservative value.

If we again limit the intake by absorption to that which would result in a deposition of 0.004 μCi after 70 years (ignoring elimination) the rate of absorption is 0.35 dis/min per day or assuming a 10^{-6}% per minute rate of skin absorption, one could permit continuously over the 70-year period some 2400 dis/min on the body. The surface area of the body is about 1.85 m^2 for an average man, about 1.6 m^2 for an average woman and about

0.25 m^2 for the newborn. [30] Data are not available for the average quantity of dirt or soil carried on th the body. Treagar [31] indicates that about 1 mg/cm^2 of liquid is about as much as can be held on the human skin without forming a noticeable liquid pool. Since the skin is normally cleansed at intervals, particularly before bedtime, and it is protected over a major portion by clothing, an average quantity of environmental soil of about 0.1 mg/cm^2 is assumed to be continuously present. Note again, that the child, who is more likely to be somewhat soiled, has a smaller surface area and, thus, for the same deposit a smaller total quantity of dirt. Under these assumptions, the average man would have some 1.85 grams of dirt on his body which could contain about 1300 dis/min per gram.

This calculation assumes the dirt on the body to contain the same concentration of plutonium as the soil in the environs. Since one would expect the smaller soil particles to be preferentially deposited on the body, a mechanism for concentration or depletion of the plutonium in the soil on the body depending upon the relative particle size does exist. Normally, however, one would expect the smaller plutonium particles to be attached to soil particles, particularly after a residence in the environment of some significant period of time so that this possibility of concentration may not be as significant as it would seem, particularly with the inherent conservatism of the calculation.

The possibility of by-passing the skin barrier by deposition in an injury or damaged skin also exists. The mechanism is of particular concern in plant operations where concentrated quantities of plutonium are handled and significant amounts, in relation to the maximum permissible body burden, can be introduced into a single wound. However, at the low concentrations expected in soils at an acceptable level, the amount of plutonium associated with the soils is very small. Data on absorption through cuts indicates that uptake may be 10-100 times that noted through intact skin. [32] Thus, for this mechanism of uptake into the body to be equally effective compared to skin absorption, some 1-10% of the body must be continually abraded and contaminated to these levels. Probably of more significance in this case is the reduction of conservatism in the number derived.

IV. INTAKE IN CONTAMINATED AREAS

The problem of estimating the intake of plutonium by a heteorogeneous group of people visiting or living in a contaminated area is exceedingly complex and provides the major source of uncertainty in the derivation of a standard. Past investigations [33, 34] have used a simplified concept of the resuspension factor to provide estimates of the air concentrations and the resulting inhalation. Intake by ingestion, absorption or through ecological chains was shown to be negligible in comparison to the inhalation. While it appears that the general concepts of these prior investigations are reasonable, a more detailed study of the various methods whereby air concentrations or ingestion can occur is needed to assure that the generalized concept of the resuspension factor, for example, covers all of the cases.

It is noted at this point that the mechanisms of intake to be discussed are primarily physical in nature rather than biological as can occur in an ecological chain leading to concentration in one or more links. While the evidence is not complete that biological accumulation may not be important in some situations, particularly as the plutonium in the soil ages and is possibly recycled through biological systems, it now appears that plant uptake or uptake in higher animals is low enough that the physical methods of direct contamination will be of greatest interest in this problem. This complicates the study because of the large number of possible direct contamination transfer systems, marked variability with different situations and the lack of firm experimental data all of which limit our ability to quantify and rank these mechanisms in order of importance.

A. Mechanisms of Intake

The intake of plutonium from the soils can be by a varied series of pathways, either direct or indirect, which are dependent upon the nature of the contaminated area, the nature and distribution of the contaminant and the actions of individuals in the area. We have not attempted to formalize these pathways at the present time since they need considerable additional definition and data to quantify them. As will be seen, however, there are a few generalizations which can be used to approximate the hazard in such situations.

If one considers the situation occurring in an area where soils are contaminated and families are living, it is immediately apparent that a relatively complex description is needed. We can start with the ambient air concentrations which will result from wind pickup. This will depend upon the type of terrain and vegetative cover, the wind speeds and directions with respect to the contaminated area, the penetration of the particles into any shelter plus other variables as discussed in Appendix A. This type of exposure will be relatively constant in time and, given certain of the variables, can be generally evaluated for the average concentration. Other perturbations in the exposure conditions are both more localized and intermittent depending upon certain actions at the time. For example, mechanical disturbance of the soils by such simple actions as walking or digging can produce localized air concentrations. These, in turn, can result in contamination of the body or clothing from which additional plutonium intake can occur by ingestion, absorption through the skin, or inhalation as a result of localized actions (i. e., taking a dress or shirt off over the head). Further, such a mechanism can result in transfer of contamination to other areas, such as the home or a vehicle, where the nature of the surroundings is such that more intimate and prolonged contact could result in significant intake. A probably more important variation of the same mechanism is that of children at play in the area. This is because of their generally more active nature and more intimate contact with soils during such activities. The presence of pets in many homes provides another mechanism for transfer of contamination into the home with possible intake by individuals. Of particular interest here is the localized concentration for inhalation which could occur by fondling or hugging the pet.

Aside from living in the area there is the question of working. Agricultural pursuits (including home gardening) involve considerable effort directly with the soils and disturbance of the soils by mechanical and animal activities. It is possible that just this type of disturbance may result in mixing of the contaminant in the soil making it less available or causing redistribution over a wider area. Again the possibility of transfer to houses or vehicles with more intimate contact and exposure of other people exists. Other types of outdoor work, such as construction, is usually for a limited period of time and, while soil disturbance is large, it usually results in a high degree of mixing and, frequently, burying some portion of the contaminant in an inaccessible location.

It will be noted that we have concerned ourselves with areas in which people are living. While it is appropriate to consider the possibility of different standards for areas with only occasional visitation, the data available on contamination transfer and the long-term behavior of the plutonium are not now adequate to provide an assessment which would be applicable to conditions some years after the contaminating event when habitation of the area is possible.

Much of the effort on these mechanisms of exposure for this interim standard has been devoted to the question of resuspension and inhalation since this still seems to be the predominant mode for taking plutonium into the body. However, future studies will attempt to better define and quantify these other possibilities, and in particular the transfer mechanisms, in order to remove uncertainties

and to, perhaps, permit a range of values applicable to different situations.

B. Ingestion

Primary methods of ingestion of plutonium from the soils are considered to be casual ingestion by transfer from the hands (or other parts of the body) to the mouth or by contamination of food crops grown in the area. There is a definite possibility of deliberate ingestion of the soils by young children.

Data on the quantities likely to be ingested in this manner are not available but, for the casual ingestion, it would appear that one gram per day would be a high estimate with 0.1 gram per day a more likely value. If we limit the total intake by this mechanism so that the body burden at the end of 70 years is 0.004 μCi with an uptake of 0.03%, the 0.1 gram per day ingestion would lead to a soil concentration of 5×10^{-3} μCi/g or 11,000 dis/min per gram.

The deliberate ingestion of soil by children is limited to a relatively short period of time, say one year, and is intermittent over this period. If we assume an average of one gram per day ingested with the limitation on accumulation during this one year at 1/70th of the maximum permitted body burden, the soil concentration should not exceed 5×10^{-4} μCi/g or about 1100 dis/min per g.

In the above analysis, we have lumped several possible individual pathways of exposure into our value of 0.1 g of soil (or the plutonium contained therein) ingested per day. These include the intake with foods, casual ingestion, and intake with water which may have become contaminated from runoff from the contaminated area. Data for individual assessment of each of these mechanisms are not adequate to trace, in any detail, the intake from each of these. However, for terrestrial environments, it is noted that the root uptake of plutonium by plants is low[35,36] so that this should not comprise a major source of plutonium to humans. Data on the transfer to muscle of animals[26] or to milk[37] again indicate a low uptake from the GI tract and translocation to these sources of food. The rate of transfer to streams or lakes by normal erosion processes will vary with the terrain and climactic conditions, but the insoluble nature of most plutonium compounds and the tendency of solubilized material to undergo ion exchange means that the great majority of the material washed into bodies of water will end up in the sediments. This may well produce a mechanism of greater significance for concentration in the biological chain than for the terrestrial ecosystems since, as was noted earlier, there is evidence of concentration of plutonium in certain marine organisms. Probably of greater significance, however, is the direct contamination of plants used as food by man and animals by the direct physical mechanisms in the environs such as resuspension and impaction on the plant surface.

These potential sources of exposure all require additional study and definition before one can arrive at a final standard. However, it is believed that the assumptions made in this section are reasonable for the present, particularly when it is realized that other mechanisms of exposure, such as inhalation, appear to be more limiting by an order of magnitude.

While these are crude estimates, it is believed that they are conservative in that the chemical forms of plutonium expected in the soils (usually oxides or polymeric forms of the hydroxides) are insoluble and the uptake from the GI tract would be expected to be considerably lower than the 0.03% chosen. It may be noted at this point that the fraction of the soil involved in the casual ingestion will be heavily weighted toward the small particle fraction since these smaller particles are more likely to stick to the clothing, skin or food crops than the larger particles. In the specification of the final standard, this particle size effect must be considered.

C. Skin Absorption

In Section III C, a value of 1300 dis/min per gram or about 6×10^{-4} μCi/g was derived as a limiting concentration in the soils for the possibility of skin absorption. Again, it is believed that this value is conservative because of the relatively high absorption rate chosen, particularly for the compounds expected in the soils. It is deliberately conservative, however, in view of the uncertainty of the influence on the absorption rate of the various lotions, makeups, soaps and other chemical materials used on the skin.

As was noted, the smaller particle fraction in the soils (or of the contaminant) is again of particular interest since this fraction will stick to the skin.

D. Inhalation

In order to be inhaled, the particles must become airborne and arrive at the vicinity of the nostrils. Usually, this requires energy from an external source to dislodge them from their resting place and to keep them suspended in the air for a time period sufficient for inhalation. (Although one can visualize a direct transfer to the air stream entering the nostrils by "sniffing" or inhaling vigorously with the nostrils close to a contaminated object.) For inhalation and retention of the particles in the respiratory tract, the particle size must be relatively small, usually considered as less than 10 μm aerodynamic diameter. Larger particles will deposit in the upper respiratory tract and be eliminated from the body in a matter of hours to days through the GI tract. Because of the low absorption from the GI tract and the protective layer of mucous between the contents and the GI tract wall, this fraction is of little or no concern for the alpha radiations from plutonium. The fraction of the particles retained in the respiratory tract increases as the particle size decreases with the best estimate of this factor as given by the ICRP Task Force on Lung Dynamics.[24] This factor has been discussed and considered in the revised MPC to be used for this study in Section III A.

The need for considering particle size of the contaminant in the soil and in its transfer to the air is of considerable importance in all of the inhalation transfers. Particles of the contaminant which are larger than the "respirable" size in the soils are of little concern from a potential inhalation hazard standpoint unless reasonably efficient mechanisms for breaking the particles into smaller sizes are available. Thus, in the following considerations, primary emphasis is placed on the smaller particles and mechanisms for movement which affect the larger particles, such as saltation or surface creep, are considered to be of secondary importance.

In the transfer of particles to the atmosphere or to surfaces, the distribution of the contaminant through the soil is an important factor. One can visualize, for illustration, two theoretical limiting conditions. The first condition prevails for an indeterminate period of time following an initial deposition when the material is spread over the surfaces of the ground and other objects in a thin layer. As time passes, the erosive effects of the wind or runoff and the removal of the material from the surfaces of plants by washing, growth and decay, or from other surfaces by winds or rains, leads to the condition where the contaminant is mostly in the soils and is distributed through a layer extending to a depth dependent upon the time since deposition, the nature of the soils, the influence of physical factors acting on the soil (such as freezing, thawing, rainfall leaching, or wind or mechanical movements resulting in mixing) and even the biological factors such as microbial action, burrowing animals, etc. This is further complicated by the fact that plants and other surfaces will intercept resuspended materials, usually diluted by the accompanying soils, and these items will serve as sources for further

resuspension. However, it can be seen that the vulnerability of the material on the surface is much higher in the condition representing an initial deposit since all of the material is in a position to be affected by winds or other disturbance while in the latter condition, a portion of the material has penetrated into the soil and its availability depends upon the depth of penetration of any disturbance. In addition, its availability may also be affected by any chemical or physical reaction, such as binding of contaminant particles to soil particles, which may have occurred. In the final limiting condition, the contaminant will be essentially uniform throughout the soil profile and will behave much as the other constituents of the soil in producing airborne dust when disturbed.

The above considerations apply directly to the airborne accident case when the deposition occurs in a short period of time so that penetration into the soil and binding to the soil particles does not occur during the period of deposition. In the industrial situation of continuous, low level, airborne releases, the deposition continues over a period of time so that these mechanisms are continuously at work and only the material deposited recently is in the upper layer of highest susceptibility and undiluted with soil particles. Another situation of interest in the industrial area would be that in which the contaminant is carried in a liquid, such as the buildup of materials on sediments from low level effluents, or the situation in an area where higher level wastes are percolated through the ground to remove the contaminants by adsorption on soils. In these cases, the penetration of the contaminant into the soil layers is much greater so that, even after drying, the contaminant is diluted to a large extent with soil and the plutonium is associated with the normal particle sizes in the soil although there may be a strong tendency for association with the smaller particles because of the nature of the ion exchange process. Another special case is the area used for burial of solid wastes. Here the immediate problem is controlled by covering the contaminated material with a thick layer of clean dirt and excluding the area from use. Concern with such practices stems from the possibility of later use of the area with digging into the material, from later erosion bringing the material to the surface, or from translocation by leaching. Here, again, the effects of time and physical and biological processes will result in a much more uniform mixing of the material (particularly if the contaminated objects are biodegradable) than when they were buried. If one could, for example, postulate complete mixing then the appropriate limits could be based upon the total contaminant and the total volume of the burial area.

1. Estimate from Dust Loading. The normal dust loading of the atmosphere results, at least in part, from the resuspension of soil particles from the earth's surface to the air. Thus, the quantity of such material normally found in a given region can be considered to be a crude index of the resuspendability of the surface materials for some indeterminate distance upwind. (Note that in a dust storm the material in the air at a given location could have originated miles upwind as, for example, from a large area of plowed fields, so that the dust load must be regarded as an index to the average condition over a large area.) If we assume that the plutonium contamination is uniformly mixed with the soil particles so that the same mechanisms which result in the resuspension of the soils are equally effective in causing resuspension of the plutonium, then a limiting concentration in that part of the soil layer which is resuspended can be estimated from the standards for particulates and for radioactivity. We have earlier concluded that an MPC of 3×10^{-13} μCi/cc seemed appropriate for the exposure of an individual in the population when applied to materials most likely to be encountered. The Federal Secondary Standard for particulates in the air is expressed as a geometric mean (mean of the logarithms of the concentrations) of 60 $\mu g/m^3$.[38]

The geometric mean is smaller than the arithmetic mean by a factor depending upon the geometric standard deviation of the measurements. Since the average exposure (and thus, presumably the average amount inhaled and retained) depends upon the arithmetic mean, it is necessary to convert this standard. Equations for this purpose are given by Drinker and Hatch.[39] Experience with most airborne contaminants indicate that the most likely geometric standard deviation is about two. For this value, the arithmetic mean concentration corresponding to the standard is 76 $\mu g/m^3$. However as the standard deviation increases, the mean increases rapidly, being 116 $\mu g/m^3$ for σ_g = 3, 152 $\mu g/m^3$ for σ_g = 4, and 219 $\mu g/m^3$ for σ_g = 5. We will arbitrarily, and somewhat conservatively, consider a geometric standard deviation of three to be appropriate with a mean concentration of 120 $\mu g/m^3$ to correspond to the standard. Thus, the concentration in airborne dust, when the particulates and the plutonium both reach their appropriate standard, would be 2.5×10^{-3} $\mu Ci/g$ or about 5500 dis/min per gram of dust.

The dust in the atmosphere will result primarily from the lower particle size fraction of the soils so that the comparison here is for the smaller particles rather than the total soil. It is conceivable that the contaminant may have a particle size distribution sufficiently different from that of the soil that some fractionation will occur during the pickup and subsequent dispersion of the dust and soil. An excellent illustration of this possibility would occur during the early resuspension of a contaminant initially deposited on the surface following an accident since it is not distributed into the soil profile and could be preferentially injected into the air. In the longer time period, however, mixing with the soil will minimize this factor.

Although the uncertainty of possible fractionation exists, it is believed that the value derived by this reasoning is conservative in that the dust loading at a given location will be as the result of the pickup over a wide area, possibly much larger than most areas of contamination and it is assumed that the dust loading is continually at the standard. In practice, the dust standard may be exceeded for periods of time but the average loading should be lower, particularly for the time spent inside the buildings, and the concern, in terms of plutonium accumulation, is with the average concentration over long periods of time. It is also believed that this value is probably the least uncertain at this time since it does not involve calculations based on uncertain and poorly defined mechanisms.

2. General Resuspension. General Resuspension consists of those mechanisms which result in a relatively uniform concentration over a reasonable area. It includes wind pickup and mechanical disturbances far enough upwind that the concentration in the air is more dependent on the dispersive mechanisms of the general atmosphere than on the very local source. It is distinguished here from the localized concentrations resulting from mechanical disturbance or other means of producing concentrations in a small volume in order to permit use of different estimating techniques. A discussion of general resuspension, the equations used and current data available is given in Appendix A supplemented by Appendix B.

As can be seen from a review of Appendix A, the information available on the resuspension rate under various conditions of atmospheric stability and ground cover is very meager and is confined almost to several experiments with ZnS particles which were freshly deposited. For the wind pickup, a value of K, the resuspension rate, divided by the wind speed squared of about 2×10^{-8} appears to be reasonable for a level, poorly vegetated plain with unstable atmospheric conditions. Higher values are appropriate for localized areas upwind where disturbances, such as vehicle traffic or construction occur, but these are generally short-lived either in duration or, if continuous, will result in rapid depletion of the contaminant or mixing through the soil profile.

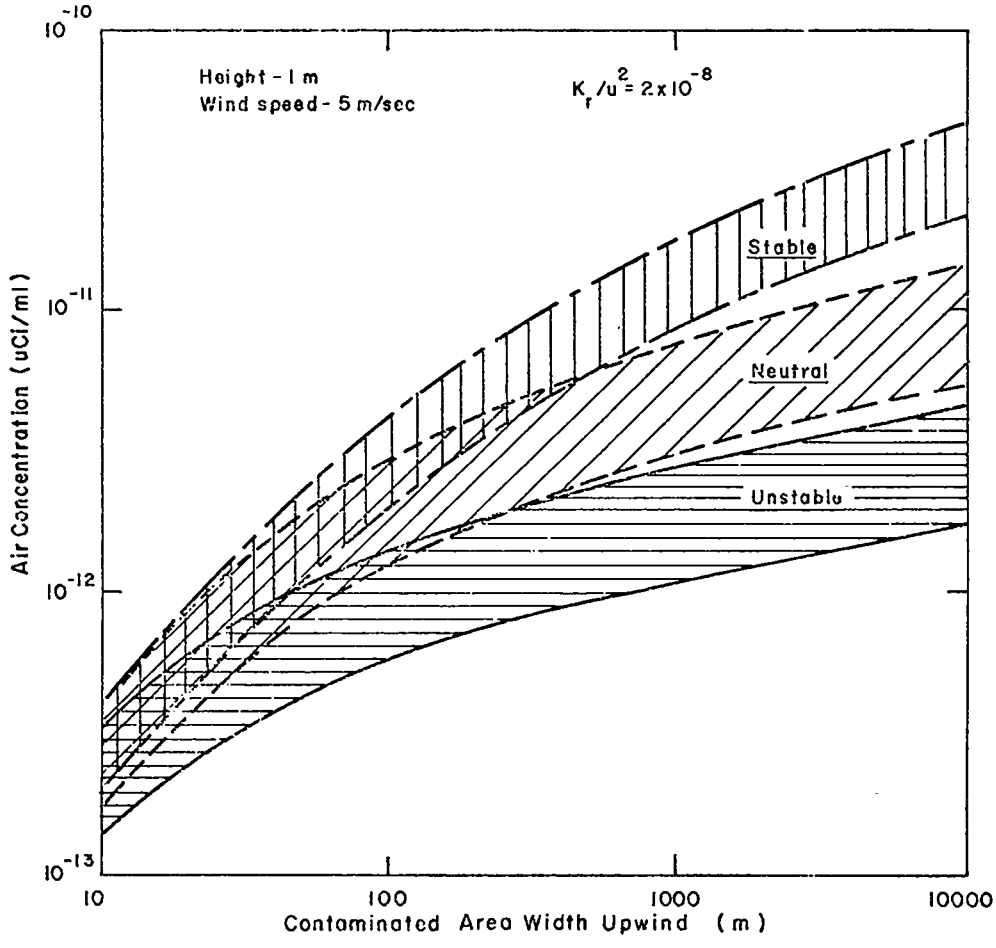

Fig. 1. Air concentrations at maximum point downwind from an infinite crosswind area uniformly contaminated to 1 μCi/m^2.

As would be expected, the concentration at a given location is highly dependent upon its location with respect to the contaminated areas and the wind directions. Thus, a precise definition of the expected air concentration could only be given for a particular location if the contamination pattern and meteorology were well defined. Some parametric calculations on the importance of this factor are in progress but are not completed to the point where they would be particularly useful in this study. However, in view of the variations possible, we have used a situation where the location of interest is at the edge of a uniformly contaminated area which extends to infinity in the crosswind direction. The wind is blowing directly over the plain toward the sampler. The results for this calculation, using the freshly deposited value of K/u^2 for zinc sulphide particles of 2×10^{-8} are given in Fig. 1. Since the wind pickup is assumed to increase as the square of the wind speed and the dispersion as the first power of the wind speed, the concentration downwind should increase directly with the wind speed. The value of 5 m/s used in the calculation is a reasonable average for many locations. The bands in Fig. 1 result from calculations using various values of deposition velocities, depending upon particle size and vegetative cover in the area.

In assessing Fig. 1, there are several uncertainties and factors of conservatism that should be borne in mind. The applicability of this calculation

to the stable case is particularly questionable since wind speeds are generally lower in this condition. The use of the same pickup rate for this stable condition as for the unstable case would seem to overestimate the concentration since one would expect the pickup to be lower because of the decreased wind turbulence. The effect of aging on the pickup rate is uncertain. While others have assumed a decrease in the air concentration[33,34] with a half-life of 35 days, examination of the data available (Appendix A) does not substantiate the continued decay at this rate. There is no doubt that the resuspension rate will decrease with time, probably rapidly at first and then at a decreasing rate. Calculations on the air concentrations at one area in Nevada (GMX) are given in Appendix A. A few measurements in this area[40] indicate the resuspension rates to be significantly lower than those calculated. This is some 20 years or so after the deposit was laid down. An additional uncertainty is in the effect of particle size on the pickup rate. The pickup constant was derived primarily from data using ZnS particles. Data are not available to assess possible changes in the rate with particle size. The question of wind variability has not been seriously considered since the formalized type of calculation used for Fig. 1 tends to average the concentration over a wide angle due to the assumption of an infinite extent in the crosswind direction. This, in turn, tends to maximize the estimate of the air concentration in the real case.

Although the uncertainties are large in this type of estimate, it would appear that a uniform concentration (or average over a large area) of about $0.1 \mu Ci/m^2$ for material freshly deposited from the atmosphere would be reasonably conservative in meeting the average MPC. One would expect the air concentration to decrease with time. It is believed that a decrease of a factor of ten would not be unreasonable over the first year.

3. Resuspension Factor. The resuspension factor approach has been widely used for estimating air concentrations from surface deposits. It does tend to give average values for the particular conditions under which it is measured and a reasonable amount of information is available for several different conditions. We tend to believe that it is more useful for describing localized concentrations resulting from various types of disturbances but, in view of the paucity of other data, its use for this problem is discussed below.

The resuspension factor is defined as the ratio of the air concentration to the quantity of material per unit area on the ground. If the air concentration is given in quantity per m^3 and the unit area on the ground is in m^2, the resuspension factor will have units of m^{-1}. Outdoor measurements of this factor have been made in arid or semiarid country following safety tests of nuclear weapons with a few studies in other areas using relatively small plots seeded with known levels of radioactive materials. Stewart[41] has concluded that a representative value for quiescent conditions outdoors is about $10^{-6} m^{-1}$ while in areas of moderate activity the value may increase to $10^{-5} m^{-1}$. A review of values accumulated from the literature by Mishima[42] indicates values ranging from 8×10^{-10} to $3 \times 10^{-4} m^{-1}$ under conditions of no mechanical disturbance and from 1.5×10^{-6} to $5 \times 10^{-4} m^{-1}$ as measured under conditions of vehicular or pedestrian traffic or in areas with people working. It is noted that the minimum values under quiescent conditions occurred from one test using ^{91}Y. If these values were excluded, the range without mechanical disturbance is reduced to 8×10^{-8} to $3 \times 10^{-4} m^{-1}$. Langham,[34] in assessing limits for a weapons accident, uses a value of $10^{-6} m^{-1}$ with, however, an exponential decrease with time with a half-life of 35 days, thereby implying a value of 7×10^{-10} one year after deposit and $5 \times 10^{-13} m^{-1}$ two years after deposit. Kathern[33] assumes a value of $10^{-4} m^{-1}$ decreasing with a half-life of 45 days. The primary evidence cited for the

decrease with time is a series of air samples taken over a period of 20 weeks in an area contaminated by plutonium following a weapons safety test. We do not believe this magnitude of decrease to be appropriate, as is discussed in Appendix A, but do believe that some decrease will occur over the first year following a deposition.

It is noted that the resuspension factor is sensitive to the methods used for estimating the quantity of plutonium on the ground, to the location of the sampler with respect to the contaminated area and to the meteorological conditions at the time of the measurement. Thus, if measurements are made under quiescent conditions, as far as mechanical disturbance is concerned, the wind speed and the depth of the plutonium in the soil would appear to be important factors for the particular locality. Measurement of the surface contamination by an alpha meter will detect to depths of a few milligrams per square centimeter (perhaps a few hundreths of a millimeter) and may underestimate the contamination. Sampling of the area to a depth of two inches will include soils which will not be affected by the surface disturbance and, if contaminated to the full depth, will result in an overestimation of the surface quantity available for resuspension. While the measured factor will indicate the probable air concentration under identical conditions of plutonium distribution, these variables plus others discussed earlier make the extrapolation of the values to different areas or different types or patterns of contamination extremely uncertain.

However, if we use the value of $10^{-6}m^{-1}$ as recommended by Stewart as an overall average value for quiescent conditions, we find that a quantity of $0.3\mu Ci/m^2$ will result in an air concentration of $3 \times 10^{-13}\mu Ci/cc$. Although mechanical disturbance of the surface will result in higher concentrations, the time period over which such disturbances will occur is usually fairly short, continued disturbance over a long period of time will result in depletion of the resuspendable material and the values are still within the uncertainty of the estimate of the overall factor.

4. Mechanical Disturbance. Mechanical disturbance of the soils by actions ranging from walking across an area to heavy vehicular traffic or even excavation of the area can result in increased dust loading downwind and, presumably, increased resuspension of contaminants contained in the soil. Such actions can also hasten the "aging" process by mixing the material in the upper layer of the soil and diluting the contaminant particles with soil particles. In extreme cases, such as the presence of heavy vehicular traffic over a given area for an extended period of time, the expected increase in resuspension rate will result in depletion of the contaminant from the particular area.

Quantitative evaluation of the potential effect of such mechanical disturbance requires a quantitative relation between the degree of disturbance for a particular area and the resuspension, a reasonable description of the disturbance expected in the contaminated area and the relation between these and the location of people. As was noted earlier, Stewart has concluded that an increase by a factor of ten for the resuspension factor for an area of moderate activity is reasonable. In Mishima's tabulation, the lower end of the range of resuspension factors is increased by about three orders of magnitude while the upper end is increased by a factor of two when areas of vehicular or pedestrian traffic are compared to undisturbed areas. However, in this tabulation it is difficult to account for degree of disturbance or for the frequencies with which measurements were taken under each condition. In a rough analysis of data obtained by Mork on air concentrations downwind from a vehicle driven across a contaminated area of the Nevada desert, (see Appendix A), it was concluded that resuspension rates up to one hundred times those caused by the winds could occur. It is noted that mechanical disturbance is a mechanism

whereby material from the ground can become airborne during conditions of maximum atmospheric stability and minimum wind speed, thereby resulting in minimum dilution downwind. At the same time, this mechanism results in the dislodging of large numbers of soil particles so that a dilution of the contaminant particles with these soil particles occurs, making the dust calculation more appropriate to this condition than the more conventional resuspension assessment.

From the data available, it appears that mechanical disturbance can result in increased air concentrations downwind over those to be expected solely from wind actions. However, if the disturbance is over a short period of time, the contribution to the average concentration will be well within the uncertainty in knowledge of the wind effect. For more intense disturbances or longer duration, the effect of mixing in the soils and/or depletion of the source will, again, minimize the contribution to the long term average. This is not to minimize the possible importance of such a factor in certain situations but, rather, it would appear that the uncertainties in the knowledge of resuspension and changes of resuspension with time will incorporate the variations due to such disturbances in most situations.

5. Personal Contamination. A possible mechanism of intake by inhalation is contamination of the skin or clothing while working or playing in a contaminated area, followed by resuspension of the material directly from the surface of the skin or clothing into the breathing zone or transfer of the contamination into the home with subsequent exposure of those living there.

Data on the transfer of contamination from the ground to the skin or clothing are very sparse so that any direct calculation of the resulting intake will produce results of limited value. However, it has been estimated from data available in the literature[29] that the inhalation from contaminated clothing or skin during normal activities could be equivalent to the inhalation of the contamination from about one cm^2 per hour. If we consider the inhalation rate to be 20 m^3 per day, the MPC of 3×10^{-13} μCi/ml would permit the inhalation of about 6×10^{-6} μCi/day. If the inhalation from clothing continues over the full 24 hours at the above rate and the clothing is continuously contaminated to the same level throughout the day, then the allowable skin or clothing contamination would be about 2.5×10^{-7} μCi/cm^2 or about 0.5 to 0.6 dis/min per cm^2.* If we consider an average of one mg/cm^2 (or about 20 grams total on an adult male) to be a reasonable value for the soil transferred to clothing and skin, the concentration would be on the order of 500-600 dis/min per gram. (Note that the 1 mg/cm^2 here includes both clothing and skin and not just skin as was used earlier.) Again, the considerations of particle size and mixing of the contaminant with the soil discussed earlier are pertinent to this evaluation. It should also be noted that the rate of intake will vary widely depending upon clothing changes, bathing, etc., and may well be lower at night because of the decreased physical activity.

The possible problems encountered from the movement of plutonium into the home on the clothing of workers was also examined in Reference 29. Here it was assumed that 30% of the material brought into the home was transferred to the home area and that it remained in resuspendable form with a half-life of one week. Resuspension rates of 5×10^{-4} per hour were used as representative of the activity in the house with two air changes per hour. Under these conditions and using an MPC for air of 2×10^{-14} μCi/ml it was concluded that 0.01 μCi could be brought in per day without exceeding the maximum permissible limits. For the revised MPC of 3×10^{-13} μCi/ml used for this study,

*This value for the allowable surface contamination is higher than that given in Reference 29 because of the reexamination of the appropriate MPC in this study.

TABLE VI

PROBABILITY OF INHALATION OF PARTICLES

Action	Probability of Inhaling One Particle	Grams of Soil Contaminated to 10^3 dis/min per gram
Changing Tire	3×10^{-5}	400
Sweeping Bus	5×10^{-4}	30
Sweeping Car	6×10^{-4}	20
Driving Car for One Hour		
No Ventilation	6×10^{-7}	20,000
High Ventilation	2×10^{-6}	7,000
	*2×10^{-5}	700

*Measured immediately after placing powder on floorboards. Other value represents the mean of two additional determinations.

it is concluded that this rate could be 0.1 to 0.2 μCi per day. As a comparative figure, if the soil contamination were 1000 dis/min per gram, this would require bringing in about 200 to 400 grams per day. While this is not a physical impossibility, particularly in muddy weather, this rate seems somewhat high for most conditions.

Schwendiman[43] has measured the probability of inhalation of particles under several conditions associated with automotive transport and cars, using ZnS particles of about 2 μm median diameter. A summary of these probabilities and the quantity of soil which must be present at a contamination level of 1000 dis/min per gram to cause the inhalation of 6×10^{-6} μCi (the amount which could be inhaled in one day at the MPC of 3×10^{-13} μCi/ml) during the given action are listed in Table VI.

These values, while for a time shorter than the full 24 hours per day, represent measured conditions in confined areas with relatively severe activity. As such, they provide some indication that the previous values estimated for the home are not unreasonable.

V. A PROPOSED INTERIM STANDARD

In the preceeding discussions, we have touched on several points which are of importance in considering the conversion of estimates of exposure to a standard for soils. To some extent these factors are interrelated and involve the questions of distribution in the soil profile, units of measurement and sizes of the particles of concern.

Previous recommendations for soil limits have been expressed in units of quantity of plutonium per unit area (i.e., μCi/m² or μg/m²) because the primary mechanism of exposure was considered to be resuspension in the atmosphere and inhalation. However, this method of designation has led to uncertainties in interpretation since the layer of soil involved and of interest was presumably that associated with the resuspension factor applied and this was not defined in the studies. Thus, there was, in these recommendations, no clear guidance as to the depth in the soil profile to which the limit should apply and varying sampling and measurement depths have been used in different studies. In the assessment of exposures in this paper, we have used both the concentration in the soil and the quantity per unit area depending upon the type of estimate made. The two methods of expressing the limit can be interrelated if such factors as the thickness of the soil profile of interest and the soil density can be defined. Thus, either method can be used as a primary unit as long as the information to permit conversion to the other is provided.

The concentration in the soil is preferred in this study because many of the potential mechanisms of exposure are more directly related to this quantity and the common methods of measurement, sampling and analysis, provide answers directly in concentration units. Even with direct measurements of external radiation, such as with the FIDLER, the quantity per unit area is applicable only when the material is in a thin layer on the surface. For a uniform depth profile with a large thickness compared to the effective range of the photons, the reading with this type of instrument is proportional to the concentration in the soil. We have, therefore, chosen to express the standard in units of plutonium concentration but, also, including a specification of the thickness of

the soil layer to be considered. This, of course, has the advantage of relating the standard to the measurements to be made in a contaminated area.

The selection of an appropriate layer must consider the mechanisms of exposure and their relative importance. For dust loading of the atmosphere and resuspension, the appropriate thickness will depend upon the type of disturbance which causes the input to the atmosphere. Similar considerations also apply to the transfers to the body since the material available for transfer must be that to which the individual is exposed in the soil layer.

We have tended to consider the material at the very surface of the ground to be limiting in the sense that it is more available for transfer or for resuspending. The definition of the "very surface of the ground" is difficult since it can change with conditions such as wind speed, turbulence or degree of mechanical disturbance. Further, the sampling and measurement of a thin layer on the ground surface is difficult even on bare ground and next to impossible in heavily vegetated areas (such as a lawn). However, in heavily vegetated areas the access of people to the soils is limited so that somewhat different considerations will apply to exposure from the soils. In view of the potential importance of wind pickup and the lack of information on the thickness of the layer actually involved in this phenomenon, we have arbitrarily chosen a layer on the order of one mm thick to serve as a standard for the low vegetated areas. The wording "on the order of" is deliberately chosen to indicate that the actual thickness cannot be specified closely because of the impossibility, in most cases, of sampling a precisely defined layer with any degree of precision. It is suggested that a reasonable interpretation of this term would be a shallow scraping of the surface layer taking into account the many imperfections and various sizes of small objects encountered in such a scraping. The use of a measured area and weighing of the sample will permit an estimate of the average thickness. For vegetated areas, where the surface is not as readily available a thickness on the order of 5 mm would seem to be appropriate. Since the specification of this thickness provides an averaging thickness over which the plutonium in the soil is measured, such a specification would permit the averaging of a thinner layer over the full depth and would permit a total of up to five times as much expressed as quantity per unit area in the vegetated area as in the barren area. The decreased exposure to people due to the smaller access to these soils and to the decreased pickup by the winds would appear to more than compensate for this.

For layers deeper in the soil profile, a thicker layer would again appear to be appropriate since exposure would result only by mechanisms which either remove the upper layers or mix the soils to a significant depth. Thus, for the soils beneath the surface, averaging over a one centimeter depth would seem to meet the intent of the limitation.

Note that in the above discussion, the limit on concentration in the soil remains constant regardless of the thickness of the layer with those mechanisms of exposure resulting from direct contact or transfer of the soil and plutonium to the body not affected. The main purpose of specifying the layer is to provide an appropriate thickness for averaging and controlling this thickness so that averaging over deeper depths will not result in samples meeting the limit but still presenting a high level at the surface for the resuspension mechanisms.

The second parameter of interest is the particle size of the contaminant. For inhalation, particles larger than about ten μm aerodynamic diameter will have a very low probability of retention in the lung and the solubility of plutonium is such that particles deposited in the upper respiratory tract will not be of significance in adding to the body burden before they are eliminated. It is noted that for plutonium oxide particles, a ten μm

aerodynamic diameter corresponds to an actual particle size of about 3 μm for spherical particles due to the density effect. In the earlier discussions it was noted that even for mechanisms involving transfer to the body, fractionation toward the smaller sizes will occur both in the dislodging of the particles from their resting place and in the consideration of the retention on the body. There is, therefore, good reason for believing that the smaller particles are of predominant importance in all mechanisms of exposure and some recognition should be given to this in the formulation of the standard.

If the overwhelming and only consideration in exposure were inhalation one could confidently use an upper limit for the size to be considered on the order of 5-10 μm based on the possibility of attachment of plutonium to particles of low density. For pure oxide particles, the size limit could be even lower. There is, however, the problem of the uncertainty in the estimates of the other mechanisms and the possibility that they may assume some importance for the somewhat larger particles if the controlling size were based only on inhalation. In addition, the possible problems of aggregation with breakup under the disturbance which transfers the material to the air must be considered. We have, therefore, chosen an arbitrary limit of particle sizes for these considerations of less than 100 μm to represent the fraction of the soil of concern. This corresponds to a screen in the Tyler series of 150 mesh (actually this is 105 μm). It is recognized that normal screening will not break up some of the aggregates which could later be broken up and serve as a source of exposure, but the choice of the 100 μm size should provide sufficient conservatism that such errors will not be important.

However, for considering the state of the contaminated area over long periods of time, one must also consider the possibility that breakup of the particles in the normal processes of soil formation will occur and will serve as a source of smaller particles continuously feeding to the fraction of interest. Although it is believed that redistribution mechanisms over the time periods of interest for soil formation will predominate in determing the soil concentrations, it is proposed that the total concentration in all particle sizes be limited to an arbitrary value of twice the concentration in the fraction below 100 μm. Since the times for soil formation from the matrix material range from decades to tens of decades, this limitation should be extremely conservative.

With this background on the application of the numbers, we are now ready to review the estimates from the exposure mechanisms to arrive at a value for the plutonium concentration in the defined layers and fractions. The estimates of soil concentration obtained earlier are summarized in Table VII for this purpose.

In assessing these values and considering the degree of conservatism relative to each, it was concluded that a value on the order of 500 d/m per gram or about 2×10^{-4} μCi per g would be appropriate. The resuspension values for fresh deposits are somewhat lower than this, but, for the long term exposure, it is expected that the values will increase by a factor of ten or more. Further, the estimates were deliberately made for an unrealistic type of area in which it would be expected that the calculations would lead to a high air concentration.

TABLE VII

ESTIMATES OF LIMITING SOIL CONCENTRATIONS FOR SEVERAL MECHANISMS OF EXPOSURE

Mechanism	Soil Concentration μCi/g	D/M per g	μCi/m²
Ingestion			
Casual	5×10^{-3}	11,000	-
Deliberate	5×10^{-4}	1,100	-
Skin Absorption	6×10^{-4}	1,300	-
Inhalation			
Dust Loading	2.5×10^{-3}	5,500	-
General Resuspension			
Fresh Deposit	*6×10^{-5}	*130	0.1
Aged Deposit	*6×10^{-4}	*1,300	1
Resuspension Factor	2×10^{-4}	*330	0.3
Clothing	3×10^{-4}	600	-

*Based on 1 mm thickness of soil with a density of 1.6 g/cm³

500 dpm/g = 227 pCi/g. TA-1 clean-up peak value - 200 pCi/g ave value - 20 pCi/g

TABLE VIII

RECOMMENDED INTERIM STANDARDS FOR PLUTONIUM IN SOILS

	In <100 μm particle Size Fraction			Total**		
	D/M per g	μCi/g	μCi/m²	D/M per g	μCi/g	μCi/m²
Top 0.1 cm*	500	2×10^{-4}	0.4	1000	4×10^{-4}	0.8
Any one cm layer	500	2×10^{-4}	4	10000	4×10^{-4}	8

* For bare soil or areas with sparse vegetation. Where area is reasonably well vegetated (greater than 50% of the area is covered with low vegetation) and a reasonable root mat exists to hold the soil, the concentration listed can be applied to a 0.5 cm layer which would permit up to 2 μCi/m² in this layer.

** With the provision that the fraction with particle sizes less than 100 μm is known not to exceed the limits given. If this is not known, the values for the <100 μm fraction should be applied to the total.

This standard for the concentration can now be combined with the previous discussion as to the limits of applicability to provide the final set of standards as given in Table VIII.

The resuspension mechanisms, which strongly influence the choice of the concentration value tend to average the pickup from wide areas so that the presence of small areas in the general vicinity which have higher concentrations are not of great importance. Since the other methods of more direct transfer from the soil give higher estimates for the limiting concentration and, in themselves, require consideration of occupancy factors and types of human activity in the contaminated area, it is tempting to specify that the above values are averages over large areas and that smaller localized depositions of several times these concentrations could be permitted. In view of the many uncertainties and the magnitude of the values, an allowance of this nature is not recommended for general use at this time. However, as additional data are obtained it is anticipated that the standard will be revised to include such a feature. In the meantime, it is possible that detailed investigation of a particular area may provide sufficient information for that area to permit the application of such a concept for that area. Such investigations aimed at a particular situation will always provide better answers than a general standard of this nature and such an approach to individual problems is entirely appropriate.

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APPENDIX A

PICKUP OF PARTICLES FROM THE GROUND AND DOWNWIND DISPERSION-GENERAL RESUSPENSION

Airborne concentrations resulting from particulate contaminants in the soil can be a possible mechanism of exposure of people and animals to the contaminant. Such airborne concentrations can be of two general types which are distinguished by their persistence and the nature of the investigations required to define their relative importance. The first type is the localized concentration where the material may be in high concentration in the breathing zone of one or a few people usually due to some mechanical disturbance of a contaminated soil or object. Such a localized concentration can result directly from the soils or by contamination of other objects which can, then, transfer the contamination to the localized breathing zone of an individual. In general, the magnitude of the concentration will be a function of the contamination level and characteristics of the contamination over a relatively small area. The second type of concentration is the more general, widespread concentration which results from the pickup of materials from the ground to the atmosphere with dispersion downwind over a large area and, possibly, involving many people. Such concentrations can result from either wind or mechanical disturbance of the soils and are a function of the contamination levels over a relatively wide area upwind for the wind disturbance and of the localized levels at the site of a disturbance for mechanical suspension. In this discussion, we are concerned with the second type of concentration which we will refer to as "general resuspension" as opposed to the local resuspension for the first type.

I. APPROACH

The general resuspension, along with the localized resuspension, has been described in terms of a resuspension factor which is defined as the ratio of the air concentration to the level of contamination on the ground at a given location.[1,2] A brief reflection on the upwind source, with a potentially large source area involved, will indicate the inapplicability of this concept to the problem of general resuspension. This is one of the chief reasons for distinguishing between the localized and general types of resuspension. Thus, the material in the air at the receptor may arise from the pickup many meters or even kilometers upwind. Further, the source will change with the wind direction or the specific area in which mechanical disturbance occurs.

In order to permit estimation of air concentrations resulting from such a contaminated area, a rough model based upon work done some years ago in estimating the importance of wind pickup and transport of larger particles with eventual impaction on the person[3,4] was revised. In this model, each element of the contaminated area is considered to be a source for airborne material with the source strength defined by the rate at which the contaminant enters the air with the specific disturbance considered. The concentration downwind is then estimated from the dispersion and deposition relations developed over the past years.

In general terms, if we consider a point source of material on the ground which is subject to resuspension, the concentration in the air at some distance downwind is given by Eq. (A-1).

$$\chi = K\Omega D'D'' \qquad \text{(A-1)}$$

In this equation, χ is the air concentration, Ω is the quantity of material on the ground in a position where it is subject to being injected into the atmosphere, K is the fraction of this material which is injected per unit time by the specific disturbance considered, D' is the dispersion which occurs

downwind as a result of turbulent diffusion, and D'' is the fraction of the material which is not deposited between the point of pickup and the receptor. In atmospheric dispersion terms, KΩ is the source term and the remainder of the equation is conventional.

The air concentration resulting from a contaminated area can then be evaluated by integrating the point source equation over the area taking into account the variations in contamination level.

$$\chi = \int_{x_1}^{x_2} \int_{y_1}^{y_2} K\Omega\, D'D''\, dy\, dx \qquad (A-2)$$

In the application of Eq. (A-2), the dispersion and deposition downwind can be evaluated from existing information resulting from micrometeorological studies, although the exact choice of parameters will affect the results and the choice is made somewhat difficult by the fact that the source is truly a ground level source. The distribution of contaminant on the ground can be measured by a number of possible techniques but, again, there is some difficulty in defining exactly the depth of importance and the other parameters, such as particle size, which will be appropriate. The primary value for which little data are available is K, the rate of resuspension under the particular conditions of interest. However, it may also be noted that Eq. (A-2) may be used in the study of the values of K in areas where the other parameters are known.

While Eq. (A-2) appears relatively simple, it describes a number of very complicated processes, many of which can be described only in semi-quantitative terms at this time. In several of the areas where existing information is adequate for other purposes, added accuracy may well be needed to permit a realistic and adequate description for this use. However, we have attempted to survey such information as is available and to apply it in a simplified fashion in order to both illustrate the application of the technique and to derive some feeling for the sensitivity of the result to some of the parameters.

II. DISPERSION AND DEPOSITION

A. Equations

The basic equations used for estimating the dispersion downwind and intervening deposition according to conventional models are well-known and have been documented elsewhere.[5] A brief presentation of these equations is included here for reference and for orientation of the user, particularly in those aspects having to do with the uncertainty.

The dispersion in the atmosphere from a continuous point source by turbulent diffusion is usually described as a Gaussian distribution of the material in the horizontal and vertical crosswind direction. Thus, the concentration at a point (x, y, z) with the origin at the source, x taken in the downwind direction and y and z the distances in the horizontal and vertical crosswind directions respectively is given by:

$$\chi_{(x,\,y,\,z)} = \frac{R\,(K\Omega)}{2\Pi\sigma_y\sigma_z\bar{u}} \exp\left[-\left(\frac{y^2}{2\sigma_y^2} + \frac{z^2}{2\sigma_z^2}\right)\right] \qquad (A-3)$$

In Eq. (A-3), u is the average wind speed, R is a reflection factor to account for the presence of the ground and σ_y and σ_z are the lengths corresponding to the standard deviation of concentration in the y and z directions. The other symbols are as given earlier.

Deposition from such a plume will result in depletion of the material originally airborne thereby reducing the quantity available at the receptor. The evaluation of the deposition rate is usually accomplished by use of a deposition velocity, V_d, defined as the ratio of the rate of deposition on a given area to the air concentration at a reference height above the area.[6] The dimensions of such a ratio are those of velocity. The product of the deposition velocity and the concentration gives the absolute rate of deposition from the atmosphere at

a given location. The quantity depositing between the source and the receptor is, then:

$$\frac{\partial (K\Omega)}{\partial x} = \int_{-\infty}^{+\infty} \chi V_d \, dy \qquad \text{(A-4)}$$

$$D'' = \exp\left[- \frac{R V_d}{\sqrt{2\Pi}\ \bar{u}} \int_o^x \frac{\partial x}{\sigma_z} \right] , \qquad \text{(A-5)}$$

In this method of accounting for deposition, a fraction of the material in the plume is assumed to deposit per unit of plume length and this fraction is removed from the plume. In essence, this correction factor reduces the source term to allow for the material which is lost. It is unsatisfactory in many ways since it implies a uniform depletion through the full height of the cloud and does not account for the concentration gradient which will exist in the profile above the ground because of the continual depletion at the ground surface. An alternate approach would be to account for the rate of change in the vertical cloud dimensions as expressed by the change in σ_z as a factor in bringing the material to the layer above the ground. However, this equation will be used in this model until further development of concepts can be made.

In order to apply these equations, relations between the values of σ_y, σ_z and the distance from the source must be used. A number of different methods of expressing these correlations have been derived by different individuals. In one of the earliest methods, Sutton[7] provides a relation between the standard deviation and distance using two additional parameters which are dependent upon the atmospheric stability and the turbulence. The standard deviation for both the horizontal and vertical growth increase downwind as a power of the distance with the power changing as the atmospheric stability changes. Pasquill[5] provides a set of curves for the growth of σ_y and σ_z based upon a classification of the stability and values of σ_θ, the standard deviation of the wind direction fluctuations which have been found to be reasonably characteristic of these conditions. Fuquay[8] uses the product of $\sigma_\theta \bar{u}$ where the σ_θ term is the same as in Pasquill's and the $\bar{u}$ is the average wind speed. In addition, Fuquay expresses the dispersion as a function of the time of travel rather than the distance. Other systems of classification are available but the above indicates some of the variations. While the experience of the author indicates that the system of Fuquay has much merit and gives about as good correlation as can be expected, it also has the disadvantage that the expressions are complicated, making integration difficult, and the information available in most situations to evaluate $\sigma_\theta \bar{u}$ is meager. The Pasquill scheme, with the data expressed as curves, again makes it difficult to perform integrations, such as those given in Eq. (A-5). In view of the uncertainties in many of the other parts of the problem, the Sutton method was chosen for the expression of the equations with, however, the reservation that the actual values of the coefficients could be chosen to provide a fit to the other schemes. In this method of expression, the standard deviation of plume width or height is given in the general form:

$$\sigma = \frac{C x^{\frac{2-n}{2}}}{\sqrt{2}} \ . \qquad \text{(A-6)}$$

The coefficient C can be different for horizontal or vertical growth. The value of n is dependent upon the stability of the atmosphere and controls the rate of growth of the plume. Using this method for expressing cloud dimensions, Eq. (A-3) for the point source corrected by Eq. (A-5) for the deposition becomes:

$$\chi = \frac{RQ}{u\,\Pi\,C_y\,C_z\,x^{(2-n)}}\ \exp\left[-\left(\frac{y^2}{C_y^2\,x^{2-n}} + \frac{z^2}{C_z^2\,x^{2-n}}\right)\right]\exp\left[-\frac{2RV_d}{\sqrt{\Pi}\,C_z\,n\bar{u}}\,x^{\frac{n}{2}}\right], \qquad \text{(A-7)}$$

From the point source equation, the contribution from other configurations of the source can be evaluated by integration. For a complex deposition pattern which cannot be expressed as an equation, this integration must be performed by numerical techniques. However, there are several simple configurations for which analytical expressions can be derived, particularly with the Sutton method of expressing the plume growth. These are given below for the convenient, dimensionless parameter $\chi\bar{u}/K\Omega$.

Infinite Line Source Upwind

$$\frac{\chi\bar{u}}{K\Omega'\sqrt{\Pi}\,C_z\,\bar{u}x^{\frac{2-n}{2}}} = \frac{R}{} \exp\left[-\left(\frac{z^2}{C_z^2x^{2-n}} + \frac{2RV_d}{\sqrt{\Pi}\,C_z\,n\bar{u}}\,x^{\frac{n}{2}}\right)\right] \qquad \text{(A-8)}$$

Ω' - Source contamination per unit length of line.

Gaussian Line Source Upwind

(Sampler directly downwind from peak concentration, Ω_p. Material along line distributed with standard deviation of A meters.)

$$\frac{\chi\bar{u}}{K\Omega'_p\sqrt{\Pi}\,C_z\,x^{\frac{2-n}{2}}\bar{u}\sqrt{C_y^2\,x^{2-n}+2A^2}} = \frac{\sqrt{2}RA}{} \exp\left[-\frac{z^2}{C_z^2x^{2-n}} + \frac{2RV_d}{\sqrt{\Pi}\,C_z\,n\bar{u}}\,x^{\frac{n}{2}}\right] \qquad \text{(A-9)}$$

Uniform Area Source - Infinite in Y

(Receptor at ground level, x_1 distance to nearest boundary; x_2 distance to further boundary of contaminated area.)

$$\frac{\chi\bar{u}}{K\Omega''} = \frac{\bar{u}}{V_d}\left[\exp\left(-\frac{2RV_d}{\sqrt{\Pi}\,C_z\,n\bar{u}}\,x_1^{\frac{n}{2}}\right) - \exp\left(-\frac{2RV_d}{\sqrt{\Pi}\,C_z\,n\bar{u}}\,x_2^{\frac{n}{2}}\right)\right] \qquad \text{(A-10)}$$

$$\frac{\chi\bar{u}}{K\Omega''} = \frac{\bar{u}}{V_d}\left(D_1'' - D_2''\right) \qquad \text{(A-10a)}$$

Ω'' - source contamination per unit area.

Equation (A-10a) is somewhat misleading in that it provides the concentration at the ground surface rather than at some height above the ground. In this situation, the small area immediately upwind contributes strongly to the final answer while, in practice, the material from this area may contribute only slightly to a receptor at some height because the growth in the vertical height of the plume may be low enough in this distance so that the material from the ground does not have a chance to reach the receptor elevation. For an elevated source, a numerical integration is needed for the initial distance where $\exp(-z^2/C_z^2x^{2-n})$ is less than one with application of Eq. (A-10a) beyond this distance.

B. Choice of Parameters

The dispersion parameters to be used in the foregoing equations can be obtained from the correlations of past experiments on turbulent diffusion.[6] These correlations are not completely satisfactory in a number of respects but they do represent a body of experience which can be applied without repeating all of the experimental work under the specific conditions of interest in this problem. However, we do emphasize the following limitations on these data. In the correlations, the data are stratified into arbitrary classifications of stability while the atmosphere in its variations acts as a continuum. Thus, some restraint is placed on the description of the variability by the categorization. Of probably greater importance, there is no agreed-upon method of defining stability for the purposes of classification so that different investigators will use different parameters (or different variations of the same parameter) in describing the classes. This also gives rise to a subjective interpretation of the meaning of the classes for experimenters working in different areas or for individuals applying the data to different areas. For example the term "strong inversion" can well have a different meaning to an individual in a flat desert country where very strong inversions can occur or to an individual in an area where temperatures are moderate with cloud cover a large portion of the time. In the following work, we will see instances where different parameters are used to describe the degree of stability for different parameters to be used in the equations. As will be noted, there is no assurance that the judgments made on these two different methods of expressing degree of stability represent the same condition of the atmosphere. While this factor is troublesome from the standpoint of logic and, to some extent scientific accuracy, this method of classifying the data is probably about as good as can be done without running into an overwhelming mass of detail and the results are undoubtedly adequate considering the remainder of the unresolved uncertainties that occur elsewhere in the problem. A more serious problem would seem to arise from the uncertain dependence of these parameters on the time of sampling or the time of interest at the receptor. Particularly for the value of σ_y the fluctuations in wind direction will increase as the time of sampling increases making the value of the cloud spread dependent upon the time. Sutton recognized this problem in his early work and specified his parameters for a relatively short sampling period.[7] Many of the differences between the present correlations and those of Sutton are undoubtedly due to the fact that most of the samples incorporated in these experiments were taken for periods of 30-60 minutes. The importance of this factor lies in the fact that the selection of a given parameter for the dispersion also implies a given fluctuation of wind direction and averaging of the downwind plume over these fluctuations which, in turn, implies a given time of sampling under the turbulent conditions existing. Such considerations are of greatest importance when attempts are made to derive values of the pickup rate from air concentration measurements around a known source of contamination on the ground. Related to both of these problems is the uncertainty of the growth of the plume in the vertical, particularly in the stable condition. While the argument can be made that wind pickup should not be of great importance under stable conditions because of generally low wind speeds and turbulence, this has not been demonstrated and the problem of dispersion from mechanical disturbances occurring under these conditions still exists. As an illustration of this problem, there are data for very stable conditions which indicate the vertical growth to be considerably lower than is predicted by any of the models normally used. The value of σ_z is of particular importance to these calculations since the deposition between the source and the receptor is strongly dependent upon this parameter and the importance of the long term average

concentration at a given elevation means that the primary dispersion mechanism over the long period of time is due to the vertical growth (i. e., the horizontal dispersion in the plume is averaged out by the changes in wind direction so that the value of σ_y is of interest only for the short sampling period.)

In view of the above considerations, it would seem that experiments designed to measure the pickup from the ground should provide a direct method of measuring the dispersion parameters and their growth during the time of sampling. This could be, for example, a smoke plume or other tracer material which would give direct evidence on the actual conditions at the time. Alternately, one could use a line source of sufficient length so that the value of σ_y is not important and concentrate on the vertical dispersion, perhaps by measuring a profile with height. This is not to say that the conventional measures of stability and wind fluctuation should be disregarded. Rather, these methods should be used to supplement the more conventional meteorological data.

For calculations in this paper, we have chosen a set of parameters reasonably representative of unstable, neutral and stable conditions. These are given in Table A-I.

The values of σ_y and σ_z resulting from these choices are compared with those of Pasquill in Figs. A-1 and A-2.

The selection of an appropriate deposition velocity is difficult because of the lack of an organized set of information on this subject. In order to provide a method of choosing the deposition velocity in relation to the particle size and the differing atmospheric conditions, a rough model to describe the

TABLE A-I

SUTTON PARAMETERS USED

	Atmospheric Condition		
	Unstable	Neutral	Stable
n	0.2	0.25	0.5
C_y	0.45	0.2	0.3
C_z	0.3	0.1	0.07

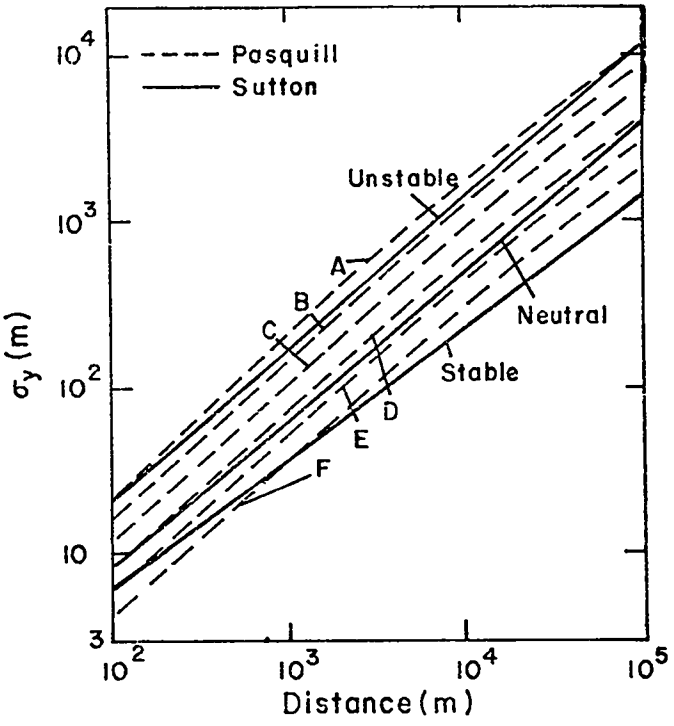

Fig. A-1. Comparison of σ_y for Pasquill's curves and Sutton using parameters of Table A-I.

deposition velocity was derived as based upon current data. This model is described in detail in Appendix B. This work predicts that the deposition velocity will vary directly with the wind speed for the particle sizes of interest and will also be dependent upon the deposition surface as measured by the surface roughness parameter z_o. It is in the choice of this value that one of the main difficulties occurs in selecting parameters which are consistent for a given stability class since the stability classification basis for the deposition velocity is the Richardson's number which can be only indirectly related to the stability used to describe the Sutton parameters chosen.

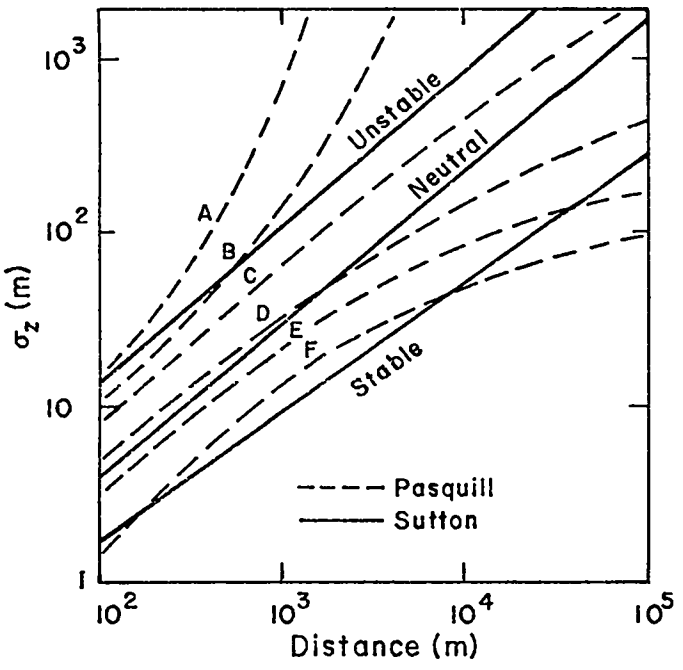

Fig. A-2. Comparison of σ_z for Pasquill's curves and Sutton using parameters of Table A-I.

In the development of the deposition velocity model, a constant rate of transfer of the particles through the boundary layer to the ground was considered due to the turbulent transfer across this boundary. The primary effect of particle size (in sizes less than those where the gravitational forces predominate) was considered to be in the retention once the particles were brought to the ground. From the data available, it appears that particles above about 1.5 μm will be strongly retained with the retention dropping off with particle size. For this reason, the reflection factor in the dispersion equations was written as (2-f) where f is the fraction of the material reaching the ground which is retained. Thus, for a material completely retained, the reflection factor becomes 1 or the concentration is simply due to the direct transport from the source.

The parameters for the deposition and reflection which have been chosen are given in Table A-II.

TABLE A-II

DEPOSITION PARAMETERS USED

	Atmospheric Condition					
	Unstable		Neutral		Stable	
z_o (cm)	0.1	2.3	0.1	2.3	0.1	2.3
>1.5 μm Particulates						
(2-f)	1	1	1	1	1	1
V_d/u	0.0093	0.017	0.0028	0.0080	0.0046	0.0029
<0.1 μm Particulates						
(2-f)	1.97	1.97	1.97	1.97	1.97	1.97
V_d/u	0.0003	0.0005	0.00008	0.0002	0.00001	0.00009

III. RESUSPENSION RATE MEASUREMENTS

The key to the application of a model such as this is, now, the definition of the rate of resuspension under the conditions of interest. Unfortunately, the information available from experiments which included the necessary meteorological data as well as the measured distribution of activity on the ground and the air concentrations resulting are extremely limited and cover only a few of the many terrain and soil possibilities of interest. However, in this section we will discuss the data available which bear on this question in order to arrive at the best possible answer at this time and to provide some illustration of the application of the method.

A. General

The movement of surface grains under the action of winds has been studied for desert sands by Bagnold[9] and for agricultural fields by Chepil[10] with only a few of Chepil's many papers referenced here. This work has outlined the mechanisms involved in producing soil erosion and has described the influence of a number of important factors. A brief outline of some of these concepts which may be important to resuspension on the scale of interest here is given below with no attempt to make this an exhaustive treatment.

Three chief methods of movement of soils are given as surface creep, saltation and suspension. In surface creep, the grains move along the surface either by the direct forces transmitted to them by the winds or by the impact of other grains.

In saltation, the grains rise into the air for a limited distance during which distance they gain momentum in the direction of the wind and fall back to the ground along a diagonal path to the horizontal. In suspension, the grains, once raised from the ground, are small enough so that the turbulence of the air stream keeps them suspended and will move them to higher altitudes by turbulent diffusion. Surface creep occurs at the ground surface while grains in saltation seldom rise more than a few feet above the ground. While these latter phenomena are of importance in spreading an initially contaminated area and in possibly eroding the size of the grains so that they become capable of suspension, the resuspension of concern in this paper is primarily associated with the fraction which these authors have categorized as the suspended fraction since this will be composed, at least partially, of grains with particle size in the inhalable range.

The soil grains in the medium size range of about 0.1 to about 0.5 mm are the ones affected by saltation with grains 1 mm or greater in diameter too large to be moved even in surface creep by ordinary erosive winds. Chepil[10] points out that the erosive action of the winds is primarily due to the saltation process since the bounding particles pick up energy from the winds during the period which they are airborne and this energy is transmitted to the soil particles upon impact to move the larger ones by surface creep, to provide energy to saltate more particles or to dislodge the smaller ones to permit the wind to carry them away in suspension. He has also observed a threshold velocity in the wind speed which will result in soil movement. This threshold velocity is least for soil particles of about 0.1 to 0.15 μm in diameter and increases for both smaller and larger particles. At this minimum, the threshold velocity is about 8 to 9 miles per hour at 6 feet above the ground. For the smaller particles, the higher threshold velocity is attributed to the smooth character of the surface attained and the nature of the turbulence in the air above the surface. However, for mixtures containing both erosive particles and fine particles the threshold wind velocity can be much lower than for the fine particles alone. Chepil also points out that the threshold velocity is not affected by surface roughness features such as ridges.

This work on the erosive properties of winds provides considerable insight into the forces and problems encountered in the resuspension problem. However, the complete applicability of the concepts and measurements to the problem at hand is questionable since the observations are necessarily of a gross nature because of the interest in the movement of large quantities of soil. Thus, considerable interest in the erosion work is attached to the condition where the mass of soil carried by the wind is sufficient to change the wind profile close to the ground because of the added momentum of the soil carried. In some of the data for mass flow over loamy soils, the suspension flow varies from about 30 mg/cm^2 at ground level to about 3 mg/cm^2 at 24 inches above the ground with wind speeds of 13 to 30 mph at a height of 12 inches. These flows convert to concentrations of tenths of grams to grams per cubic meter. This is not to say that such conditions are not of concern in resuspension work but that the mechanisms involved in resuspending the small particles of interest may be different from those observed in the gross erosion studies and the frequency of occurrence of the heavy loads in most areas is relatively small unless there is widespread disturbance of the soil surface, as in many agricultural practices.

There is a series of observations on the effect of soil condition and terrain which would seem to have some relevance to the resuspension studies. It is indicated that particles less than 0.005 mm (5 μm) do not exist, as such, in ordinary soils since they become aggregated into larger particles. However, it is also noted that large quantities of nonerosive soil are converted to erosive material by abrasion caused by the moving soil grains. Thus,

one would expect some breakup of the aggregates formed by the soil and a contaminant by a similar mechanism or by mechanical disturbances. If a surface has been undisturbed for some time, the initiation of erosive movement can require a higher velocity than for succeeding wind storms due to the formation of a surface crust which is broken by the erosion caused by the first high wind. When soil is carried by saltation, it can be sorted into dunes. This process can increase the susceptibility of the soil to later pickup and decrease the threshold velocity. Such increase in susceptibility may be of particular importance when the succeeding wind comes from a different direction and can, therefore, pick up material previously deposited in an eddy behind an obstruction. A rain storm may have an effect in increasing the threshold velocity but it has been observed that such effects will not persist after the rain since a few grains in saltation will break the surface crust.

It is obvious that no one resuspension rate will be applicable to all conditions. A listing of variables which would be expected to influence the results would include:

1. Particle size distribution in the soils
2. Particle size distribution of the contaminant
3. Distribution of contaminant through the soil profile
4. Moisture content of the soil
5. Chemical composition of the soil (cementing and compacting)
6. Type and magnitude of vegetative cover
7. Obstacles to airflow and turbulence inducers

Since all of these factors can also change with time, in particular the moisture content, vegetative cover, distribution of the contaminant in the soil profile and the particle size of the contaminant through aggregation, a thorough understanding of the mechanisms of resuspension will require characterization of many variables. Since powerful numerical techniques are now available for studying hydrodynamic problems such as this, a theoretical program to provide some insight into the importance of such variables would seem to be of great value. In addition to providing this insight, such a study could provide valuable guidance in defining the types of measurements to be required in the field.

One factor not considered above, nor in this calculational model, is that of surface redistribution through runoff of water or movement through the actions of the winds. There is no doubt that this is of considerable importance since such redistribution will affect not only the area covered but the redeposition will be in places where the susceptibility to resuspension may differ from that in the original position. However, the complexities of this problem are beyond the scope of this treatment and study of this will be deferred to a later date.

Item 3, the distribution through the soil profile, is of considerable importance in interpreting field results. For wind pickup, for example, the material which is deeply buried will not be in a position where the wind forces can act on it and should, therefore, not be included in any estimate of the source term. Thus, samples taken to a depth of several inches can be misleading if a significant part of the contaminant occurs below the surface but is included in the measurement of the inventory and is interpreted as part of the contamination of concern. It can be predicted that the critical thickness of the contaminated layer of interest will vary with the degree of disturbance which causes the material to become airborne. However, for most cases, the prediction of this thickness is not now possible. For pickup by winds we can speculate that this thickness may vary with the wind speed due, in part, to the increased size and energy of the particles carried in saltation or surface creep with higher wind speeds and their consequent ability to disturb a deeper layer of soil. Note that if this is true, the dependence of air

concentration with wind speed will have a term not usually considered if the contamination is deeper than the immediate surface. (i. e., the source term will change due to the availability of material buried in the ground at greater depths.) Thus, in measurements of resuspension, as well as in applying measured rates to other areas, it is of great importance to specify clearly the depth of burial of the contaminant and to take into account differences in the depth. Again, some theoretical studies using the numerical, hydrodynamic techniques now available could possibly be of use in better delineating this problem and some of its ramifications.

Another factor which can contribute uncertainty to the final result is the question of particle size of the contaminant (and soil) and its influence on the rate of resuspension. The work of Chepil, discussed earlier, indicates a definite dependency of the erosion rate on particle size distribution of the soils with uniform, relatively small particles requiring higher wind speeds to dislodge than heterogeneous mixtures of several hundred μm particles. Again, the mechanisms by which the particles are transferred to the air are of importance but not well defined quantitatively. For non-vegetated areas, the mechanical transfer of energy from particles in saltation or surface creep would seem to be the primary sources of such energy. Here, the energy available would seem to increase as the square of the wind speed but it is not clear that this energy would be transferred to the soil particles in such a manner that particles of all sizes would be dislodged in proportion to the energy. In other words, it is not now known whether all particle sizes will contribute to the source term in proportion to their fraction in the soil at all wind speeds or whether one would expect changes in the fraction of different sizes airborne as the wind speed changes. (Note that there will be a change in the upper end of the particle size spectrum, with the number of larger particles increasing with wind speed, simply because the energy available will dislodge larger particles at the higher speeds and the increased turbulence will result in longer times of residence in the atmosphere. The effect on the smaller, inhalable sizes, however, is not clear.)

B. Rate of Resuspension - Direct Experiments

The bulk of the information available in the literature on the resuspension factor is not adequately documented with meteorological conditions and extent of the contaminated area to permit derivation of the rate of resuspension from the measurements of the air concentrations although, as will be seen, some estimate of the order of magnitude under the conditions at the time may be derived.

In the earlier papers on resuspension,[3,4] a rate of resuspension was derived from experimental measurements downwind from a source of zinc sulphide particles spread on the ground. In this paper, an attempt was made to account for particle size by inclusion in the pickup coefficient of a term combining the particle density and area exposed to the wind, a refinement presently not believed to be completely applicable until more data on the effect of particle size are available. For this reason, the pickup coefficient in the earlier paper is not the same as the one used in Eq. (A-1). However, a conversion can be made by comparison of the equations. This technique was used since the pickup coefficient in the earlier paper was calculated using the actual wind direction in relation to the position of the sample in order to make a correction for an off-center plume and these data are no longer available. From this comparison, the MMD of the zinc sulphide particles used (7μm) and the density of ZnS (4.1 g/cc), the conversion from the coefficient in the earlier work (K') to the coefficient used in this paper (K) becomes:

$$K = \frac{K' \bar{u}^2}{\rho d} = 0.035\, K' \bar{u}^2 \qquad \text{(A-11)}$$

TABLE A-III

RATE OF WIND PICKUP OF ZnS PARTICLES

First experiment - sandy soil, sparse desert grass, and clumps of sagebrush 0.5 to 1 meter high.

$\bar{u}$ m/sec	K $Sec^{-1} \times 10^9$	$K/\bar{u}^2$ $Sec/m^2 \times 10^9$	$\bar{u}$ m/sec	K $Sec^{-1} \times 10^9$	$K/\bar{u}^2$ $Sec/m^2 \times 10^9$
2.7	90	9.5	1.8	60	17
3.1	140	15	2.7	150	20
2.7	50	6.7	2.2	60	13
0.9	130	16	3.6	160	13
2.7	70	9.5	1.8	26	8.1
2.7	40	18	1.3	40	24
1.8	10	3.9	1.3	40	24

Second experiment - prepared courses.

Course	$\bar{u}$ m/sec	K $Sec^{-1} \times 10^9$	$K/\bar{u}^2$ $Sec/m^2 \times 10^9$	Remarks
Control	5.8	120	3.5	
	10	2450	25	
	8.1	70	1.1	Damp
	6.7	310	6.7	Wet
	8.2	940	14	Wet then dry
Furrowed	5.8	350	11	
	10	700	7	
	8.1	920	14	Damp
	6.7	140	3.2	Wet
	8.2	240	3.5	Wet then dry
Rock	5.8	350	11	
	10	3500	35	
	8.1	230	3.5	Damp
	6.7	470	11	Wet
	8.2	470	7	Wet then dry
Snow fence	5.8	47	1.4	
	10	350	3.5	
	8.1	140	2.1	Damp
	6.7	310	7	Wet
	8.2	240	3.5	Wet then dry

Values of the rate of pickup for these particles as obtained from the conversion are given in Table A-III. The value of K/u^2 is included since the total energy available in the wind varies as the square of the wind speed although there are other factors, such as wind profile and turbulence which will also affect the results.

These results are representative of the particular type of particles used as a tracer and represent the pickup a relatively short period of time after the deposition has occurred. No significant change was seen in the rate of resuspension in the one week period over which measurements were made in the first experiment. It is of interest to note that positive concentrations were measured at low wind speeds, on the order of one m/s or less, or lower than the minimum threshold velocity given by Chepil. This may be due to the existence of gusts with speeds much above the average but cannot be attributed to pickup at an earlier time when winds were stronger because of the short distances involved in the experiment.

Several other experiments under field conditions were examined briefly to provide an order of magnitude estimate of the results. In most of these experiments, the published literature is inadequate to permit full evaluation since required details are not given. They are reviewed, however, with assumptions made as to the missing data.

Wilson et al[11] report on an air sampling program associated with the contamination of an area following a safety test with a nuclear device containing plutonium at the Nevada Test Site. Data on the detailed contamination patterns are not given although it is noted that samplers were located approximately northeast of ground zero at distances of about 7500 feet (at the $10 \mu g/m^3$ contour), 2500 feet at the $100 \mu g/m^3$ contour and at about 750 feet at the $1000 \mu g/m^3$ contour. It was further noted that the winds blew generally from the south during the period of the experiment (about 60% of the time) and that this wind direction missed the highest contamination areas. Because of the wide variation in the air concentrations measured, only the median concentration of the three samplers at each of the 10 and $100 \mu g/m^3$ isopleth and the two samplers at the $1000 \mu g/m^3$ isopleth are given. These were read from a plot in the reference. Sampling started about 23 days after the contamination pattern was established and continued for 20 weeks.

It was assumed, for rough estimation purposes, that the average concentration level in the area over which the wind blew was three times the level at the location of the sampler and the average distance of contamination over which the wind blew before reaching the sampler was equal to the distance from the sampler to ground zero. The average wind speed was taken to be two meters per second with neutral conditions. Since particle size analysis showed the resuspended materials to have

TABLE A-IV

ROUGH ESTIMATES OF PICKUP RATE FROM NEVADA STUDY

10 μg/m²			100 μg/m²			1000 μg/m²		
*dis/min	K x 10⁹ Sec⁻¹	K/ū² x 10⁹ m²/sec	*dis/min	K x 10⁹ Sec⁻¹	K/ū² x 10⁹ m²/sec	*dis/min	K x 10⁹ Sec⁻¹	K/ū² x 10⁹ m²/sec
630	60	20	1200	10	3	8000	9	2
---	--	--	680	7	2	1200	1	0.3
330	30	8	250	3	0.6	1200	1	0.3
330	30	8	1200	10	3	1800	2	0.5
140	10	3	130	1	0.3	230	0.3	0.06
31	3	0.7	620	6	2	950	1	0.3
55	5	1	290	3	0.8	2500	3	0.7
220	20	5	95	1	0.2	1100	1	0.3
90	8	2	120	1	0.3	2500	3	0.7
150	10	4	600	6	2	1100	1	0.3
82	8	2	230	2	0.6	1000	1	0.3
35	3	0.8	310	3	0.8	2100	2	0.6
110	10	3	70	0.7	0.2	850	0.9	0.2
68	6	2	35	0.4	0.09	140	0.2	0.04
58	5	1	150	2	0.4	650	0.7	0.2
42	4	1	200	2	0.5	490	0.5	0.1
65	6	2	23	0.2	0.06	180	0.2	0.05
39	4	0.9	130	1	0.3	200	0.2	0.06
52	5	1	85	0.9	0.2	100	0.1	0.03
22	2	0.5	95	1	0.2	180	0.2	0.05
av	10	3		3	0.8		1	0.4

*Quantity collected in one week at a flow rate of 17 liters per minute.

an average mass median diameter of 1.5 to 2.5 μm, the deposition velocity was assumed to be that corresponding to the turbulent transfer velocity. The values of the expected air concentrations for a unit deposition level were calculated from Eq. (A-10) and were corrected for the assumed two m/sec wind speed. These results are given in Table A-IV for the successive weeks of sampling.

The uncertainties in these results due to the necessary assumptions are obvious, but it is of interest that they are generally not greatly different from those measured with the zinc sulphide particles. It is not surprising that they are lower since the zinc sulphide data were obtained with short period air samples with the wind blowing from the source to the sampler while these values represent a week's sample with no correction for the fraction of time that the wind blew across the contaminated area. Further, there is no correction for periods of higher wind velocity nor for differences in stability of the atmosphere which would be expected from day to night. Soil sampling at the location of the samplers indicated the nominal isopleths to be high by a factor of about four at the 10 μg/m² location, 2.5 at the 100 μg/m² location and 2 at the 1000 μg/m² location. Use of these soil values for the calculation would increase the resuspension factors estimated by these factors. The generally lower resuspension rates for the higher contamination areas could be due to a number of different causes including a greater sensitivity in these areas to a misestimate of the effective path length of the wind over the contaminated areas and the fraction of the time that the wind blows over this path; shorter crosswind dimensions so that the area is not really infinite in the y direction as is assumed in Eq. (A-10);

differences in particle sizes of the contaminant with larger particles in the more heavily contaminated area close to ground zero; or failure of the contamination to reach the five foot height of the samplers in the shorter, more contaminated areas.

Mork[12] reports on an experiment at the Nevada Test Site in which a vehicle was driven back and forth across a stretch of ground contaminated with plutonium for one hour while air samples were taken at two points 20 feet from the vehicle path and at two points 100 feet from the vehicle path. The experiment was conducted in a region between two sampling points designated as number 62 and 63 which, in turn, were reported to have contamination levels of 6.21×10^6 dis/min per square foot and 6.63×10^6 dis/min per square foot. The vehicle path was 1320 feet long with the samplers located about one-third of the distance from each end. The experiment was conducted twice. The first in the daytime was from 1138 to 1238 and the second (labeled as at "night") was from 1725 to 1825. Since it is doubtful that the nighttime inversion would have set in by this time in the evening, both sets of data were evaluated considering the atmosphere to be unstable.

Neither the wind direction or the wind speed were given. It was, therefore, assumed that the wind was blowing across the vehicle path toward the samplers. Gummed paper deposition collectors were included which permitted a rough estimation of the deposition velocity to this surface by comparison with the air concentration. The results of this experiment as calculated under these assumptions are given in Table A-V.

The deposition velocities would indicate that larger particles were involved in most cases. The relatively lower values of resuspension in the second experiment may indicate that the course had undergone depletion or that some other factor in the conditions had changed. The value of K/u^2 is not estimated for these data since the source term is assumed to be more dependent upon the mechanical disturbance than on the energy transmitted by the

TABLE A-V

MECHANICAL DISTURBANCE RESUSPENSION

	Location			
	20'		100'	
	West	East	West	East
1138-1238 V_d (m/sec)	0.5	0.004	0.9	0.02
K (sec^{-1})	$160 \times 10^{-9}\bar{u}$	$9000 \times 10^{-9}\bar{u}$	$3000 \times 10^{-9}\bar{u}$	$2600 \times 10^{-9}\bar{u}$
1725-1825 V_d (m/sec)	0.3	2	---	0.4
K (sec^{-1})	$130 \times 10^{-9}\bar{u}$	$150 \times 10^{-9}\bar{u}$	---	$180 \times 10^{-9}\bar{u}$

winds. Although comparison with the values of K from the other experiments is difficult, it does appear that resuspension rates up to 100 times those from the natural winds can occur from this type of mechanical disturbance in this type of terrain.

In a more recent paper by Sehmel[13] the particle resuspension due to moving vehicles on an asphalt road was measured by use of ZnS particles with a mass median diameter of about 5 μm. These particles were distributed uniformly over a length of 100 feet on one lane of an asphalt highway and cars and trucks were driven at different speeds either on the adjacent lane (by-pass) or through the contaminated lane. Runs were made while the winds were perpendicular to the highway and the fraction resuspended per pass was evaluated from air samples and deposition samples downwind. For purposes of this discussion, the values for the resuspension per pass were converted to fraction resuspended per second from consideration of the wind speed of the vehicle and the 100 foot path length. This was done to enable comparison with previously derived resuspension rates. The data from this work are given in Table A-VI.

As is pointed out by Sehmel, the relative constancy of the ratio of the rate of pickup to the square of the vehicle speed indicates the primary mechanism of pickup is due to the turbulence induced by the passage of the vehicle. Again, the data indicate increased pickup rates, in this case by as much as three or four orders of magnitude over those measured from ZnS in the soils and

TABLE A-VI

RESUSPENSION RATES OF ZnS PARTICLES FROM ASPHALT ROAD BY VEHICLES

Time Since Deposition (days)	Vehicle	Vehicle Path	Vehicle Speed (m/sec)	Fraction Resuspended per pass	Fraction Resuspended per sec (K)	$\frac{K}{(\text{Vehicle Speed})^2}$
0	Car	By-pass	2.2	4.8×10^{-5}	3.5×10^{-6}	7.0×10^{-7}
			6.7	2.8×10^{-4}	6.2×10^{-5}	1.4×10^{-6}
			13.4	7.7×10^{-4}	3.4×10^{-4}	1.9×10^{-6}
			22.4	1.1×10^{-3}	8.1×10^{-4}	1.6×10^{-6}
		Through	2.2	1.9×10^{-4}	1.4×10^{-5}	2.8×10^{-6}
			13.4	6.9×10^{-3}	3.0×10^{-3}	1.7×10^{-5}
			22.4	1.1×10^{-2}	8.0×10^{-3}	1.6×10^{-5}
	*Truck	Through	2.2	2.5×10^{-3}	1.8×10^{-4}	3.7×10^{-5}
			22.4	6.7×10^{-3}	4.9×10^{-3}	9.8×10^{-6}
4	*Truck	By-pass	6.7	1.2×10^{-5}	2.6×10^{-6}	5.9×10^{-8}
			6.7	4.8×10^{-6}	1.1×10^{-6}	2.3×10^{-8}
			13.4	8.6×10^{-5}	3.8×10^{-5}	2.1×10^{-7}
			13.4	8.2×10^{-5}	3.6×10^{-5}	2.0×10^{-7}
		Through	2.2	1.3×10^{-5}	9.5×10^{-7}	1.9×10^{-5}
			6.7	5.2×10^{-4}	1.1×10^{-4}	2.5×10^{-6}
			6.7	2.1×10^{-4}	4.6×10^{-5}	1.0×10^{-6}
			13.4	1.0×10^{-3}	4.4×10^{-4}	2.5×10^{-6}
			22.4	2.3×10^{-3}	1.0×10^{-3}	2.0×10^{-6}
5	Car	Through	13.4	5.7×10^{-5}	2.5×10^{-5}	1.4×10^{-7}
30	Car	Through	13.4	5.5×10^{-6}	2.4×10^{-6}	1.4×10^{-8}
			22.4	2.6×10^{-5}	1.9×10^{-5}	3.8×10^{-8}

*3/4-Ton Pickup Truck.

natural winds. The data taken at later times, however, indicate a relatively rapid decrease whether due to fixation of the particles or prior removal is uncertain. Sehmel does provide a rough calculation, however, which indicates that the depletion from such a surface with any significant traffic would be rapid.

C. Changes With Time

It is to be expected that the susceptibility of a contaminant to resuspension will change with time due to factors such as agglomeration with the soil particles; possible chemical changes of the contaminant; migration of the particles downward in the surface through action of rainfall, alternate freezing and thawing; and redistribution, perhaps into areas protected from the winds. For materials deposited in an area with high mechanical disturbance, such as a highway, the latter factor will be of great importance in moving the material to an area where the disturbance is lower.

Wilson, et al[11] investigated the resuspension of plutonium in an area at the Nevada Test Site which was contaminated during a safety test in April 1957. Samples were taken with impactors at a height of five feet above the ground starting about one month after the area was contaminated and continuing for twenty weeks. Three samplers were placed at the nominal $10\,\mu g/m^2$ contour (~7500 feet from GZ), three at the $100\,\mu g/m^2$ contour (~2500 feet) and two at the $1000\,\mu g/m^2$ contour (~750 feet). The samples were pooled to give weekly estimates of the concentrations at these locations. The samplers were located so that the winds blew directly over the most contaminated area only about 10% of the time during the period of sampling. It was noted that: "The sampling data are too erratic to establish half-times for the 'decay' of air concentration beyond a very crude estimate." These estimates were obtained by plotting the median of the stations

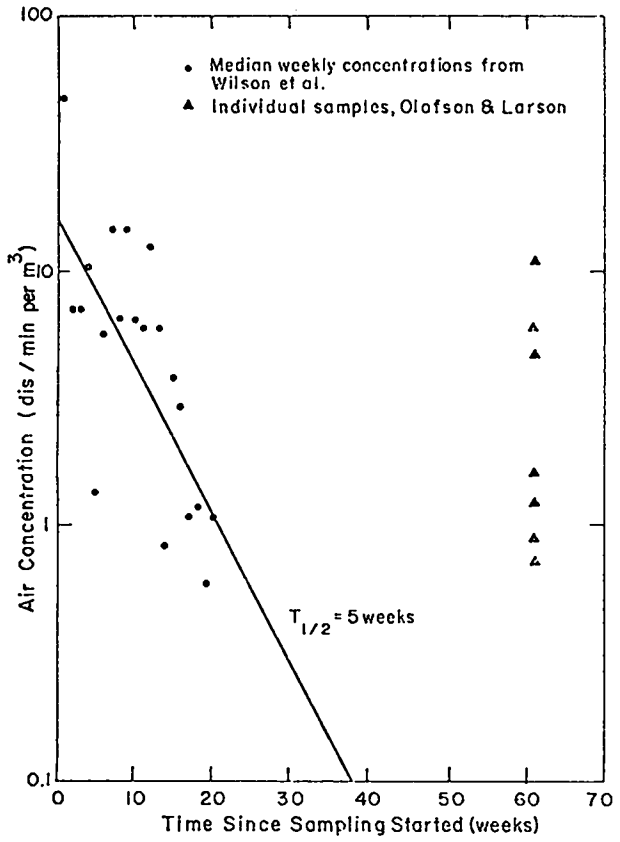

Fig. A-3. "Decay" of Air Concentrations - 1000 μg/m².

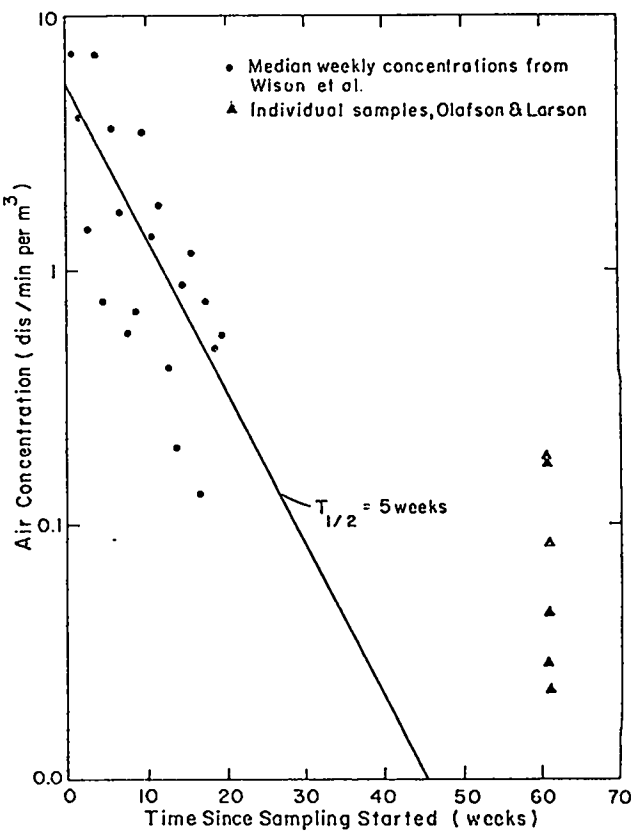

Fig. A-4. "Decay" of Air Concentrations - 100 μg/m².

on a given isopleth. This yielded an estimate of five weeks for the half-time of concentration decay.

During the summer of 1958, studies of the air concentrations were conducted at the same site.[14] The two high level locations for the location of the samplers were quoted as being at "... essentially the same location as the nominal 100 and 1000 μgm locations of Wilson et al." The data for these two locations for the median of the weekly samples from Wilson and for the individual samples reported by Olafson and Larson are plotted in Figs. A-3 and A-4. While the use of the week-long sample and the median value of several samplers tends to reduce the absolute concentration due to wind fluctuations over this time and also tends to reduce the statistical spread, the concentrations reported 40 weeks after the first series raise some questions as to the long term applicability of the 35-week half-life. It is noted that there are other factors which could produce a reduction in the air concentrations such as a seasonal shifting of the winds resulting in a lower contaminated area upwind or a seasonal change in wind speed. Both of these variables could result in a regular decrease in air concentration with time if the change in the winds occurred in a systematic manner.

In one other experiment, Mork[12] reports data taken three feet above the ground in October 1956 and in July 1958 at Station 61 at the Nevada Test Station. These samples were in an area contaminated with plutonium during December of 1955 and January of 1956. In a seven day period in 1956 series the concentrations averaged about 2×10^{-8} μg/m³ with a range of about 4×10^{-9} to 6×10^{-8} μg/m³. In a 20 day period in the 1958 series, the concentrations averaged 2×10^{-7} μg/m³ with a range

in the values from 0 to 7×10^{-7} $\mu g/m^3$. While, again the wind directions and speeds may well have differed during these two periods, there is no indication of a measurable decrease with time over a period of some twenty months or about 85 weeks.

As was indicated, one would expect a change with time, but it is believed that the data now available are not adequate to permit the assignment of a particular decay rate, particularly for areas of different characteristics from the Nevada Site. In particular, it is believed that the use of the 35-day half-life is inappropriate since this would indicate that the concentrations drop rapidly and, for long term occupancy considerations, result in exposures estimated only for the initial period of occupancy. A more reasonable estimate to describe this phenomenon would be to consider a drop by a factor of ten over the first year or two with the conditions stabilized thereafter to give relatively little decrease.

In view of the information available on the initial concentrations at specified locations at the Nevada Test Site, it would appear to be reasonable to mount a campaign to resample these locations over a period of time and to attempt to reconstruct the meteorological conditions for the initial samples. While the deposits have been disturbed by the various activities in these areas, such a series of samples could give some indication of the long term decrease.

D. Dust Concentrations

The concentrations of natural dust in the atmosphere arise, at least in many areas, from resuspension of soil grains under natural or mechanical disturbance mechanisms. As such, data on these concentrations can be used to give at least gross indications of the importance of resuspension in various terrains and locations. Again, such information must be interpreted with restraint, considering the differences which may exist between the deposited contaminant and the natural soil particles with little real data to interpret the effect of particle size or the effect of depth in the soil profile on the rate of resuspension. In addition, calculations indicate that dust in the atmosphere may well have originated a considerable distance upwind and could have been resuspended under completely different meteorological conditions.

One set of data collected by Hilst[15] at the meteorological facility at Hanford has been examined. Here the number of dust particles per cubic foot were measured in five size ranges by use of cascade impactors. In one experiment, measurements were made at five heights ranging from 1.25 feet to 400 feet while in the others, the measurements were made at three intervals from 0.9 feet to 41.3 feet. These observations show that, in general, the dust concentrations, expressed as mass per unit volume, decreased rapidly with height. However, the mass median diameter of the particles also decreased so that the change in concentration of the smaller particles with height was much less pronounced and, in several cases was not detectable.

The fractional rate of pickup from the ground cannot be determined from these data because of the lack of information on the source material. However, the source term, itself, can be determined and compared to the wind speed. These data are given in Table A-VII. Again, the ratio of the rate of pickup, as determined from these data assuming unstable conditions and a source upwind approaching infinity, to the square of the wind speed appears to be relatively constant.

E. An Example of Application

As a part of the effort of the Nevada Applied Ecology Group to define potential problems with plutonium contamination on the Nevada Test Site, an extensive effort is being made to measure contamination patterns, resuspension and redistribution, and the ecological behavior of the plutonium. We have chosen the GMX area for analysis because

TABLE A-VII

DUST CONCENTRATIONS AND ESTIMATED PICKUP RATES

Date	Height Ft	$\bar{u}$ m/S	0.9 - 5 μm $V_D/\bar{u} = 0.013$ χ #/m³	$K\Omega$ $Sec^{-1}m^{-2}$	$K\Omega/\bar{u}^2$ $Sec^{-1}m^{-3}$	5 - 20 μm $V_D/\bar{u} = 0.013$ χ #/m³	$K\Omega$ $Sec^{-1}m^{-2}$	$K\Omega/\bar{u}^2$ $Sec^{-1}m^{-3}$	20 - 60 μm $V_D = 0.2$ m/sec χ #/m²	$K\Omega$ $Sec^{-1}m^{-2}$	$K\Omega/\bar{u}^2$ $Sec^{-1}m^{-3}$	60 - 240 μm $V_D = 1$ m/sec χ #/m²	$K\Omega$ $Sec^{-1}m^{-2}$	$K\Omega/\bar{u}^2$ $Sec^{-1}m^{-3}$
8/11 p.m.	1.25	6.3	2.3×10^5	37,000	950	91,000	15,000	380	26,000	6600	170	2500	2500	64
	50.	11	1.8×10^5	30,000	770	55,000	9,000	230	9,700	2400	62	320	320	8
	100.	12	1.2×10^5	20,000	500	40,000	6,600	170	6,500	1600	42	110	110	3
	200.	12	1.4×10^5	22,000	570	41,000	6,700	170	5,200	1300	33	35	35	0.9
	400.	14	8.0×10^4	13,000	340	29,000	4,800	120	3,100	780	20	70	70	2
5/4 a.m.	0.9	1.3	7.8×10^4	2,700	1500	12,000	440	240	1,300	260	150	70	70	40
	14.7	1.8	5.3×10^4	1,900	1000	6,700	240	130	210	40	20	10	10	6
	41.3	1.8	5.6×10^4	2,000	1100	6,400	220	130	280	60	30	30	30	20
5/4 p.m.	0.9	1.3	7.8×10^4	2,700	1500	21,000	740	400	2,000	400	220	50	50	28
	14.7	1.8	6.4×10^4	2,200	1200	6,400	220	130	280	60	30	10	11	6
	41.3	2.2	5.6×10^4	2,000	1100	6,400	220	130	320	60	40	20	21	10
*5/27 a.m.	0.9	2.7	3.1×10^4	2,200	300	5,300	370	50	670	140	20	35	35	5
	14.7	3.6	2.0×10^4	1,400	200	3,900	270	40	320	70	9	35	35	5
	41.3	4.5	2.5×10^4	1,800	250	3,900	270	40	460	90	13	35	35	5
5/27 p.m.	0.9	2.2	5.6×10^4	3,600	620	24,000	1,500	260	2,700	550	96	70	70	10
	14.7	3.1	4.9×10^4	3,100	540	11,000	700	120	850	170	30	70	70	10
	41.3	3.6	4.2×10^4	2,700	470	9,500	600	110	460	90	16	35	35	6
10/5 p.m.	0.9	2.7	1.8×10^5	12,000	1700	26,000	1,900	260	2,900	600	80	280	280	40
	14.7	5.8	1.2×10^5	8,700	1200	8,600	600	80	1,200	250	40	70	70	10
	41.3	8.0	1.8×10^5	12,000	1700	8,100	570	80	1,100	220	30	70	70	10

*Ground surface damp.

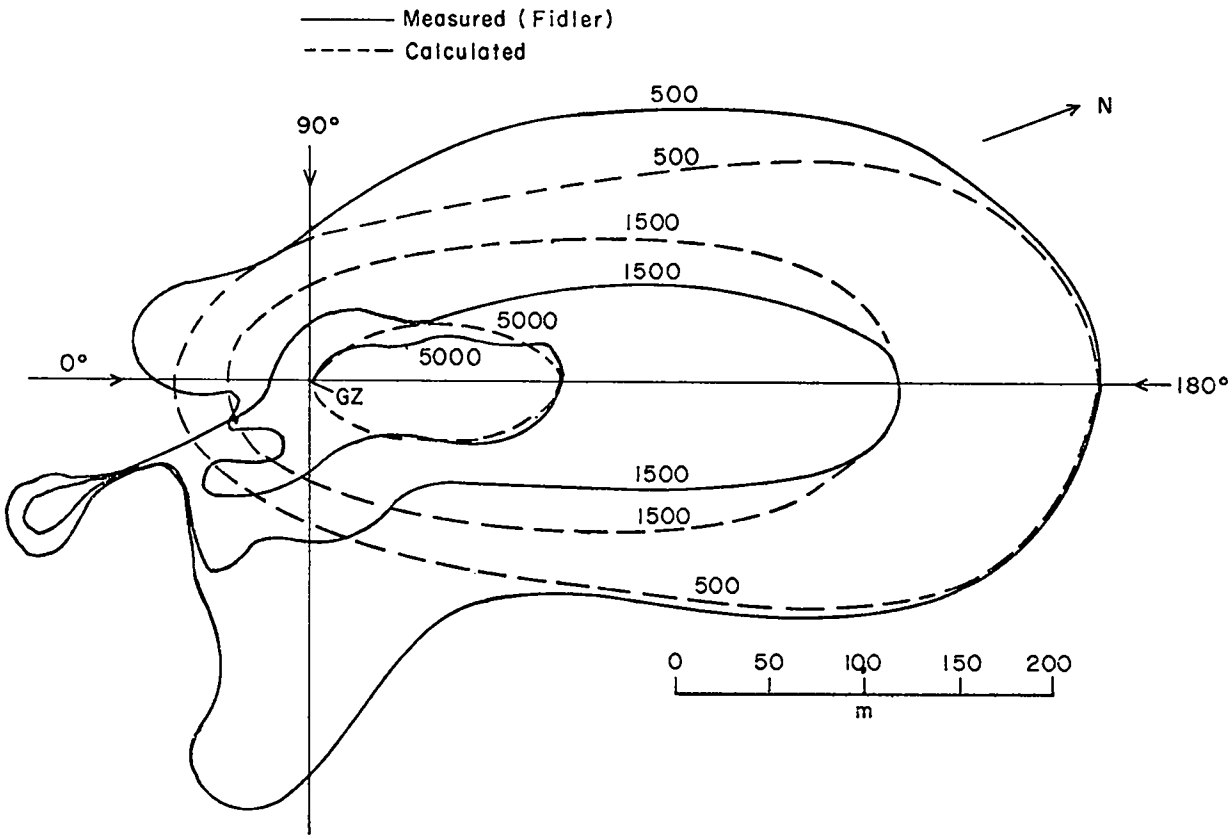

Fig. A-5. GMX Area.

initial resuspension measurements have been planned there and some preliminary data on ground deposition are available. The pattern of deposition has been measured by FIDLER surveys using the 60 kev photons from the ^{241}Am associated with the plutonium in June 1971.[16] This pattern is given by the solid isopleths in Fig. A-5. If we assume that the pattern is Gaussian in the cross-pattern direction with the standard deviation increasing exponentially with distance from ground zero (GZ) and the centerline deposition as given in Fig. A-6, the isopleths shown by the dotted lines in Fig. A-5 are obtained. These isopleths are considered to be sufficiently representative of the pattern to be usable in the calculation of expected air concentrations or the derivation of resuspension rates from measured air concentrations. Although it would be desirable to have an analytical relationship between the position on the pattern and the peak deposition at the centerline, none was found and the relation in Fig. A-6 was used. For the change in standard deviation of pattern width with distance from GZ, the following relation was used:

$$\sigma_y = 37.3 \exp(0.00335a) \qquad \text{(A-12)}$$

where σ_y is the standard deviation in meters at a distance of a meter from GZ.

Calculations were performed for locations on the centerline of the pattern to take advantage of the symmetry so produced. The basic approach was to calculate the expected concentration from Gaussian line sources at various distances from the station with the standard deviation of the pattern at that distance according to Eq. (A-12). Wind

directions were varied at 22.5 deg intervals from 0 deg (wind directly up-pattern) to 180 deg (wind directly down-pattern). These directions are indicated on Fig. A-5. The total concentration was then obtained from these values by weighting according to the centerline concentration from Fig. A-6 and multiplying by the interval represented between the successive line sources. Since the calculations were performed on a Wang 600 programmable calculator it was necessary to limit the number of upwind line sources considered to a total of 50 per calculation. The concentration resulting from a Gaussian line source with a standard deviation of A meters, a centerline deposition of Ω Ci/m^2 and a wind direction of θ to the pattern centerline can be obtained from:

$$\frac{Xu}{K\Omega p} = \frac{(2-f)}{\pi C_y C_z} \sum_{y=-4A}^{y=4A} \frac{\Delta}{(a\cos\theta + y\sin\theta)^{2-n}} \exp\left[-\left\{\frac{1}{(a\cos\theta + y\sin\theta)^{2-n}}\left(\frac{(a\sin\theta - y\cos\theta)^2}{C_y^2} + \frac{z^2}{C_z^2}\right)\right.\right.$$

$$\left.\left. + \frac{2(2-f)Vd}{\sqrt{\pi}\, C_z n\bar{u}} (a\cos\theta + y\cos\theta)^{\frac{n}{2}} + \frac{y^2}{2A^2}\right\}\right] \quad \text{(A-13)}$$

In Eq. (A-13), the angle θ is measured between the perpendicular to the line source and the wind direction. In practice, calculations were made separately for the up-pattern wind direction (0-90°) and the down-pattern (90-180°) switching the sign of the coordinate system so that a was always positive.

Values of the pickup and dispersion parameter were calculated for stations located 150 m up-pattern from GZ; at GZ; and at 75 m, 200 m, and 400 m down-pattern from GZ. The values were then normalized to a value of 1 Ci/m^2 at the peak deposition point, 6400 c/m on the FIDLER. These results are given in Figs. A-7 and A-8.

Since the deposition pattern was measured with the FIDLER, a relation between this measurement and the quantity of plutonium in the soils in a position to be picked up by the winds is needed. Eberhardt and Gilbert[16] have provided a statistical summary of the data on soil analyses in this area including some preliminary correlations between the FIDLER readings and the soil analyses. In this study, FIDLER readings were made at given locations followed by sampling in three 5-in. circles to a depth of three centimeters. The number of samples taken was limited and no correlation was found for those measurements in the lower two isopleths. However, in the >5000 c/m isopleth, six samples showed an apparent correlation with the FIDLER reading. These data indicated about 0.3 dis/min of plutonium per gram of soil for each count per minute on the FIDLER. While this correlation is rough and the authors warn against applying it to the lower contamination levels, in this preliminary study we will accept it with the reservation that as more data become available the correlation (as well as the shape of the pattern) should be revised.

There is, however, no information on the change in plutonium concentration with depth in the soil profile or on the microdistribution of plutonium in the area measured by the FIDLER. Some rough calculations indicate that the FIDLER sensitivity for ^{241}Am decreases to about 50% of the surface value for a plane source buried 8 mm and to about 10% for a plane source at a depth of 2.5 cm. The soil sampling procedure averages the total quantity of plutonium over the sampling depth of 3 cm. Thus, while the correlation would indicate that there would be about 7×10^{-9} Ci/m^2 (assuming a soil density of 1.6 g/cm^3) per c/m on the FIDLER, the actual fraction of this which is effective in

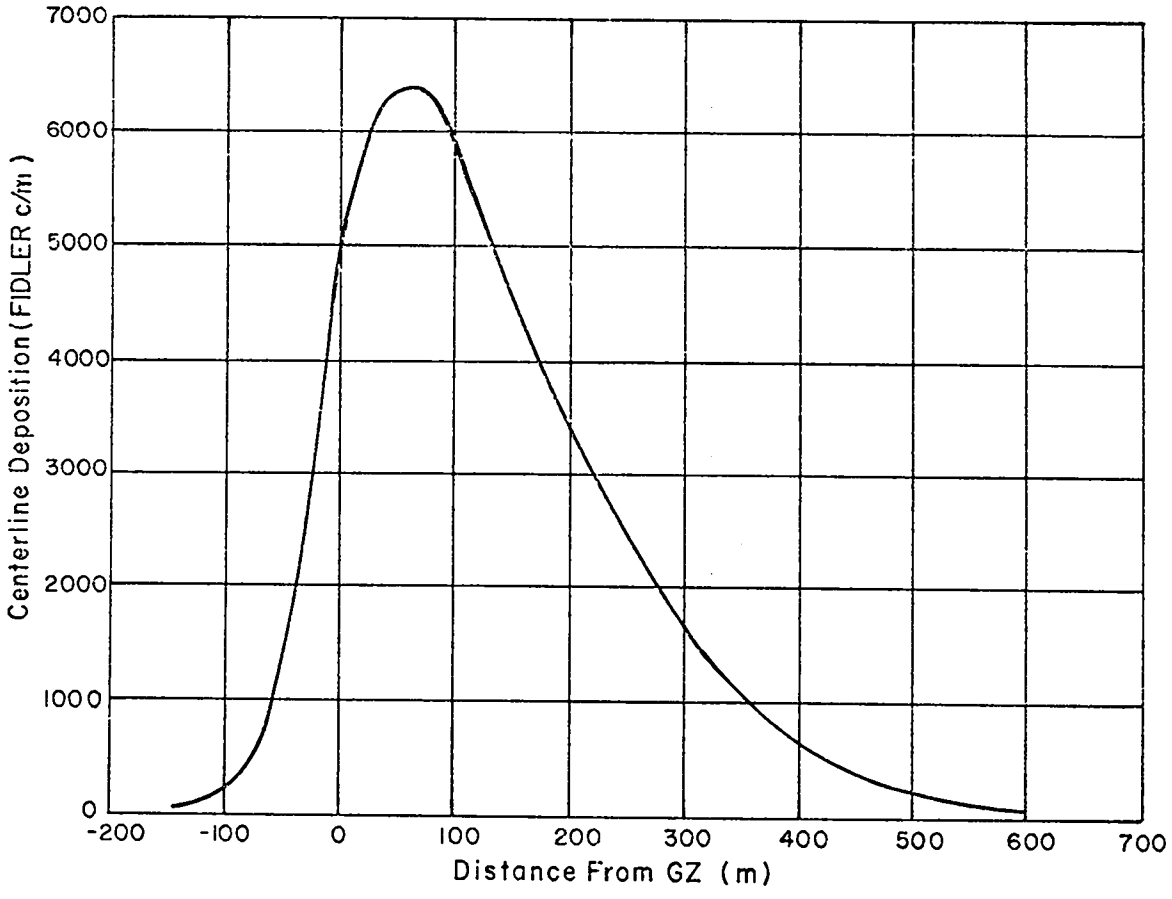

Fig. A-6. Centerline Deposition Pattern in GMX Area.

producing air concentrations is not known. For these preliminary calculations, we will use the above value but remember the reservations quoted.

Several air samples have been obtained in this area during tests of the Lawrence Livermore's high volume air sampler.[17] The sampler was located about 250 feet (76 m) north of GZ. Five samples were taken in April of 1972: two for periods of 17-19 hours and three for periods of 4-5 hours. The resuspension rates for the three shorter samples were estimated from Fig. A-7 assuming the peak deposition on the pattern to be 6400 c/m on the FIDLER or 4.3×10^{-5} Ci/m^2. These data are given in Table A-VIII.

For these calculations, it was assumed that unstable atmospheric conditions existed. This seems appropriate for the middle of the day at this time of year. For the longer samples, which were taken overnight, there was considerable variation in both wind direction and wind speeds with low wind speeds occurring during the middle of the night. In addition some of the data on wind speeds are missing. An attempt to approximate the value of K/u^2 was made by using the hourly recorded values of wind speed and direction. These results, while very crude indicated values on the order of 10^{-13} to 10^{-14} with the lower value increasing to about 10^{-13} if it were assumed that pickup occurred only during the unstable periods with higher wind speeds.

These resuspension rates have considerable uncertainty, particularly with respect to the definition of the surface deposit. If, for example, it is considered that the top millimeter of the soil is

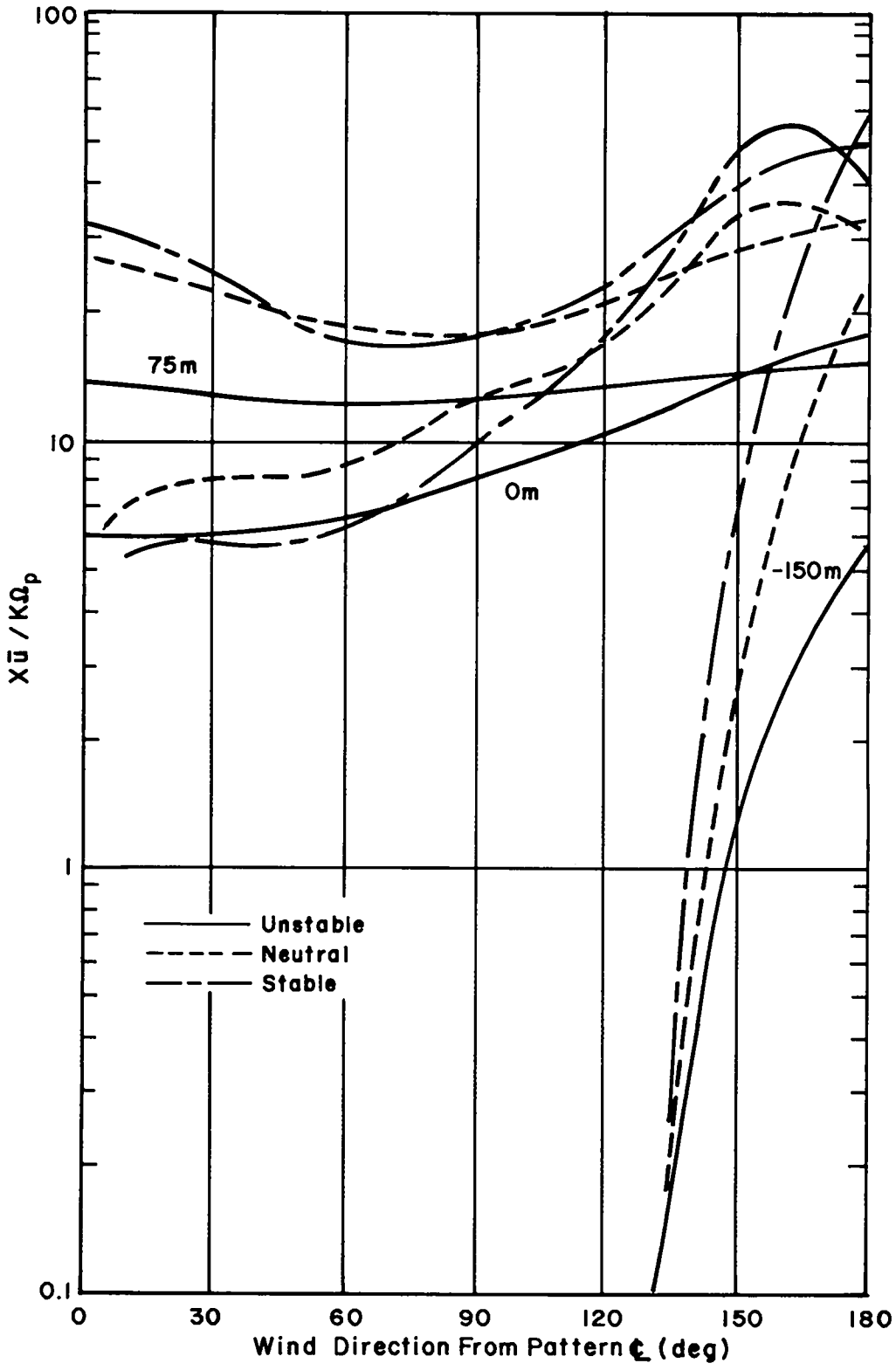

Fig. A-7. Concentration Integrals for Different Wind Directions with Respect to Pattern Centerline.

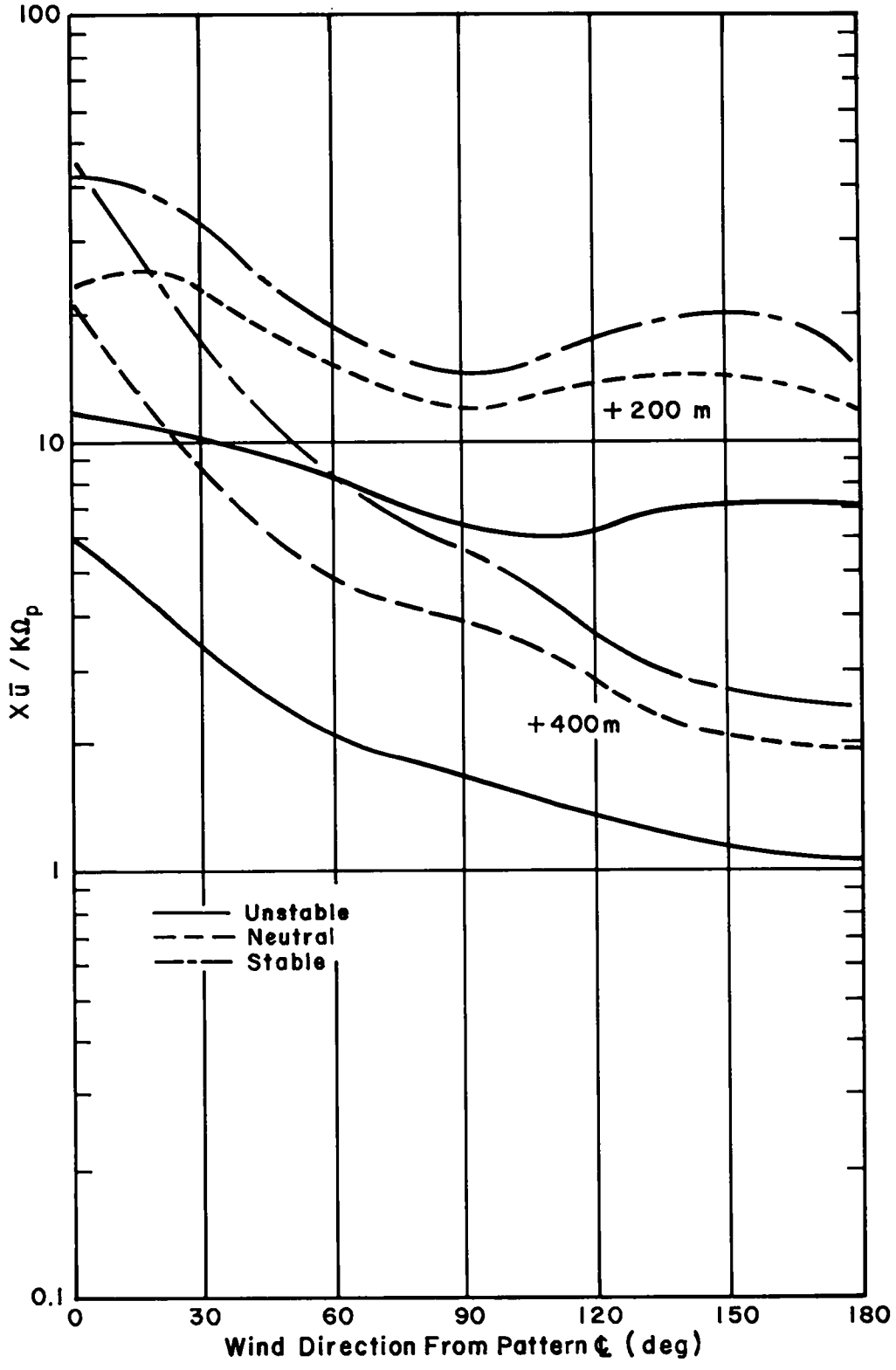

Fig. A-8. Concentration Integrals for Different Directions with Respect to Pattern Centerline.

TABLE A-VIII

ESTIMATED RESUSPENSION RATES FOR THE GMX AREA

Time of Sample	Measured Concentration (Ci/m^3)	Wind Direction True (deg)	Wind Direction *Pattern (deg)	Wind Speed (m/s)	$\frac{\chi u}{K\Omega_p}$	K	K/u^2
4/19/72 1200-1600	3.5×10^{-16}	350	155	3	15	1×10^{-12}	2×10^{-13}
4/27/72 1100-1600	1.4×10^{-14}	218	18	4	14	1×10^{-10}	5×10^{-12}
4/20/72 0930-1400	1.5×10^{-16}	220	20	5	14	1×10^{-11}	5×10^{-13}

*Pattern centerline is 20 deg east of true north. Value given here is for application to the directions used in the calculation.

the layer of importance and the plutonium is distributed uniformly through the three centimeter sampling thickness, then the effective surface deposit is only 1/30th of that used above and the resuspension rates will be increased by a factor of 30. In addition, the sampling period is relatively long in comparison to that believed appropriate to the dispersion coefficients used in the integrations and these may underestimate the actual cloud spread, again resulting in some increase in the resuspension rates. Even considering these factors, however, the value of K/u^2 appears to be considerably lower than the results quoted earlier for fresh deposits. Additional studies in the area will be needed to fully explain the results and the relatively large variation in K/u^2 from these few samples, but it can be postulated that at least a part of the reason for the lower values may be due to aging of the deposit and redistribution by particle size.

Data are not available on the influence of mechanical disturbance in this area on the resuspension rate. It is noted, however, that the calculation of the average concentration from the full area involves the derivation of values for a line source at various distances upwind with the wind in different directions. Studies of the influence of mechanical disturbance by people or animals walking across the area or a vehicle driving across could be made by sampling during such periods of disturbance. Such results would be extremely valuable in assessing the relative importance of wind pickup and such mechanical disturbance. Similarly no attempt has been made, as yet, to evaluate the long term average concentration taking into account the shifts in wind direction, speed and atmospheric stability. It is believed, however, that the above method, in conjunction with the appropriate meteorological measurements would result in a reasonable estimate.

The above example emphasizes the need for adequate meteorological support in providing the dispersion and deposition parameters to be applied during experiments to assess the pickup rate. Ideally, such measurements should be adequate to permit a more accurate estimate of the dispersion coefficients than has been used here and the actual equations used should be modified to apply the most accurate estimate of the dispersion.

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APPENDIX B

THE VELOCITY OF DEPOSITION

The concentration downwind from a source of airborne material depends upon the amount of material removed from the atmosphere by natural processes in the region between the source and the receptor as well as upon the source strength and the atmospheric mixing processes. The removal rate varies with the physical nature of the airborne material, the state of the atmosphere and the nature of the terrain involved. Two basic removal mechanisms are washout or rainout, during periods of precipitation, and dry deposition at other times. In this treatment we will be concerned with the dry deposition phenomena since the purpose of the study is to estimate the amount remaining airborne rather than the amount deposited. Information on the washout processes[1,2] can be adapted to estimate the effects of these processes on the air concentrations during periods of precipitation.

Early experience with the effluents from a radiochemical separations plant at Hanford, as well as experiments by Chamberlain indicated that ^{131}I in vapor form deposits strongly from the atmosphere onto surfaces.[3,4] Later experiments by Megaw and Chadwick[5] using solid fission products generated by arcing an irradiated wire showed that the deposition rate of these fine particles was considerably lower than that for iodine vapor. The close-in fallout from nuclear detonations consists of large particles which have a terminal velocity sufficiently great that they will not remain suspended for any length of time but will settle from whatever height they reach in the initial cloud meanwhile being carried by the winds.

This information indicates that there must be at least three separate considerations in the dry deposition of such material from the air: (1) gravity settling for large particles; (2) transfer from the air to the ground by the turbulent eddies in the atmosphere for small particles which remain suspended for considerable periods of time and for gases or vapors; and (3) retention of the receiving surface once the material is brought into contact. Thus, both the fine particles and the iodine vapor should be brought to the ground at about the same rate by the turbulence of the atmosphere, but the iodine, being in a chemically reactive form is apparently retained at the ground surface better than the small particles. Such considerations permit separation of the problem into several parts depending upon the physical nature of the atmospheric contaminant.

It has been customary to express the rate of removal of a given material by the ratio of the rate at which it deposits to the concentration in the atmosphere at the point of concern. Thus:

$$\frac{\text{curies per m}^2\text{ per sec}}{\text{curies per m}^3} = \text{m/sec}. \qquad \text{(B-1)}$$

This ratio, which can be measured directly, is referred to as the velocity of deposition since it has the units of velocity. It is the purpose of this appendix to explore the various factors which can influence the velocity of deposition for particles and to derive a simplified model incorporating the important variables so that some indication of the variation expected with these parameters can be derived. The model is not exhaustive in its treatment of the various theories and information available since the intent is to provide an overall picture which is commensurate with our knowledge of the applications, particularly in regard to the influence of this variable on the air concentrations resulting from resuspension of particles from the ground.

I. GRAVITATIONAL SETTLING

The settling of larger particles under the influence of gravity has been studied for many years. This rate of settling is characterized by a terminal

velocity in which the force exerted by gravity is exactly balanced by the aerodynamic drag from the passage through the air. As the particle size decreases, the terminal velocity decreases to a point where the turbulent eddies in the atmosphere exert sufficient force to overcome the gravitational forces and the particle remains suspended. For our purposes a large particle can be defined as one in which the terminal velocity predominates over the atmospheric turbulence and the deposition velocity is essentially equal to the terminal velocity. Note that this is not a definition of a particular particle size since the eddy forces will depend upon the degree of turbulence in the atmosphere at a particular time. Thus, one would expect larger particles to be suspended in an unstable atmosphere than in a stable one.

The simplest particle is a sphere and the terminal velocity of such spheres are given by Stokes' Law[6,7] for particles up to 50-100 μm in diameter. Above this value the drag coefficient increases so that the terminal velocity is smaller than would be calculated from Stokes' Law. However, for simplicity and since we are primarily interested in particles smaller than this, we will apply Stokes' Law throughout. This gives the settling velocity as a function of particle size and density as:

$$V_g = \frac{2 g r^3 \rho}{9 \kappa} \qquad \text{(B-2)}$$

Here V_g is the terminal velocity in cm/sec, g is the acceleration due to gravity in cm^2/sec, r is the particle radius in cm, ρ is the particle density in gm/cm^3 and κ is the viscosity of the air in poises. Actually, the density term should reflect the density difference between the particle and the air, but the air density can be neglected with respect to the density of most particles of interest. It may be noted that the viscosity of air varies from 171 micropoises at 0 deg C to 190 micropoises at 40 deg C so that a minor effect will occur because of temperature. The terminal velocity for several sizes of spherical particles with a unit density evaluated at 45 deg latitude and sea level with a temperature of 18 deg C are given in Table B-I.

TABLE B-I

TERMINAL VELOCITY OF UNIT DENSITY SPHERES IN AIR

Particle Size			Particle Size		
Radius μm	Diam μm	Velocity cm/sec	Radius μm	Diam μm	Velocity cm/sec
0.05	0.1	3.0×10^{-6}	5	10	0.30
0.1	0.2	1.2×10^{-4}	10	20	1.2
0.2	0.4	4.8×10^{-4}	20	40	4.8
0.5	1.0	3.0×10^{-3}	50	100	30
1.0	2.0	1.2×10^{-2}	100	200	120
2.0	4.0	4.8×10^{-2}			

From Stokes' Law, the terminal velocity of a given diameter of particle will vary directly as the density. Thus, particles from a material with density ten will have terminal velocities ten times those given in the table. Since the velocity also varies as the square of the particle radius, the sizes of particles having the same terminal velocity will vary as the square root of the densities. That is, a one μm particle of a material with a density of nine will have the same terminal velocity as a three μm particle of unit density. Thus, the behavior of the higher density particle in situations where the behavior is dependent upon the aerodynamic properties will be similar to a larger particle of the lower density material.

The situation with spherical particles is highly idealized for most real-life situations. Instead, the particles may be highly irregular in shape so that it is difficult to even characterize them according to any one dimension. The drag characteristics of these irregular particles also vary from that of the sphere so that even particles of the same mass will have different terminal velocities depending upon their shape. The situation is further compounded by the fact that many particles of interest may be agglomerates of other particles frequently of particles of different composition and origin. For this reason, and the variation with density, an aerodynamic diameter is frequently used in particle work.

This may be defined as the diameter of a particle of unit density which has the same terminal velocity as the particle of interest. This diameter will be used throughout this paper unless a correction for density is specifically indicated. Such an aerodynamic diameter essentially defines the inertial behavior of the particle which is of importance in many problems, such as sampling by particle size where the separation is done by inertial means, consideration of impaction on a surface, or even deposition of the material in the respiratory tract. Rough conversion factors between spherical particles and particles of known and definable shapes can be found in the literature but are not considered here because of the preliminary nature of many of the data discussed herein and the resulting uncertainties from these causes.

II. TURBULENT TRANSFER

For the turbulent transfer to the ground, theoretical treatments have been published by Stewart,[8] Owen,[9] and Chamberlain.[10] Fuquay in unpublished work[11] has considered the transfer of mass across the boundary layer of the atmosphere to be equal to the transfer of momentum and has evaluated the transfer coefficient or velocity to be:

$$V_t = \frac{(u^*)^2}{u_z} \qquad \text{(B-3)}$$

Here, the transfer velocity is designated as V_t to indicate that it is the component due to the turbulent transfer, u* is the friction velocity and u_z is the wind velocity at a reference height z. The friction velocity is the ratio of the shearing stress in the lower layers of the atmosphere to the density, with the shearing stress considered to be constant with height in the surface layers of concern. Chamberlain's expression for the resistance to transfer in the boundary layer[10] is the reciprocal of Eq. (B-3) and he has evaluated the velocity of deposition for submicron particles from a theoretical treatment by Owen to be $0.004\,u^*$. Markee[12] indicates that the deposition velocities for iodine with a one-meter reference height at the National Reactor Testing Station have shown an approximate linear relation with u* where $V_d = 0.012\,1\,u^*$. Since, as will be seen, the friction velocity for a given atmospheric condition and surface is proportional to the wind velocity at a reference height, Eq. (B-3) reduces to the same form as these observations.

The value of u* can be evaluated from the wind profile (change in wind velocity with height) and a parameter representing the nature of the surface. For a surface in which the irregularities are large enough so that a laminar layer submerging the irregularities cannot form the flow will be turbulent down to the surface.[13] Such a condition is called fully-rough flow and occurs for nearly all natural surfaces at moderate or high wind speeds. Sutton,[13] for example, indicates that for a wind speed of 5 m/sec at a height of two meters, only a surface such as smooth mud flats or a large sheet of ice would be aerodynamically smooth. A closely cut and well rolled lawn would be smooth for winds below 1 m/sec measured at two meters height but would be rough at higher wind speeds. Note that the wind must flow over a surface for some distance before the surface layer takes on the turbulence characteristics of that surface. This means that significant changes in the character of the turbulent layer occur with changes in terrain, with possible significant changes in the turbulence transfer velocity. Where artifical surfaces, such as paper of limited area, are used to sample deposition, it is probable that the transfer velocity is characteristic of the terrain immediately surrounding the sampler with the retention characteristics those of the sampling medium so that results from differing terrains or samplers may not compare.

The wind profile in a neutral atmosphere and its relation to the friction velocity has been studied more extensively and is better characterized than for stable or unstable atmospheres. For a neutral atmosphere, the wind profile is logarithmic and in

fully rough flow can be described[14] as:

$$\frac{\bar{u}_z}{u^*} = \frac{1}{k} \ln \frac{z}{z_o} \qquad \text{(B-4)}$$

In Eq. (B-4), k is the Van Karmen constant with a value of about 0.4, $\bar{u}_z$ is the wind speed at a height z and z_o is a constant characteristic of the surface. This constant arises as a constant of integration in the derivation of Eq. (B-4) and represents the height at which the flow can be extrapolated to zero. It can be measured for a given surface from the wind profile in a neutral atmosphere and is reasonably independent of wind speed, although in situations where the surface changes with wind speed, as in the development of waves on a body of water or the bending of tall grasses in the wind, the value of z_o can either increase or decrease with wind speed. Deacon[15] has given typical values for various surfaces along with an estimate of the wind speeds above which fully rough flow can be expected so that the treatment of Eq. (B-4) is applicable. His plot is reproduced in Fig. 1. It may be noted that this treatment is not strictly applicable to surfaces with higher roughness features such as a forest.

With stable or unstable conditions, the logarithmic wind profile no longer holds. For these conditions, Deacon[15] indicates that the change in wind velocity with height (du/dz) provides a reasonable fit to a power function. From this, he derives

$$\frac{\bar{u}_z}{u^*} = \frac{1}{k(1-\beta)} \left[\left(\frac{z}{z_o} \right)^{1-\beta} - 1 \right] \qquad \text{(B-5)}$$

The symbols in Eq. (B-5) are the same as those in Eq. (B-4) with the addition of β which is the exponent in the derivative of the wind profile. Beta is greater than one for an unstable atmosphere, less than one for a stable atmosphere and one for a neutral atmosphere. It is assumed that z_o is characteristic of the terrain and is the same in all stabilities so that a measure of this constant under neutral conditions will permit application to Eq. (B-5). The validity of this assumption has not been definitely shown and, particularly in very stable atmospheres, the criterion for fully rough flow may not be met and the profile may differ from the power function.

By combining Eq. (B-3) with either Eq. (B-4) or Eq. (B-5) we can derive a functional relationship between V_t and these parameters.

$$\text{Neutral} \qquad V_t = \left[\frac{k}{\ln\left(\frac{z}{z_o}\right)} \right]^2 \bar{u}_z \qquad \text{(B-6)}$$

$$\text{Other} \qquad V_t = \left[\frac{k(1-\beta)}{\left(\frac{z}{z_o}\right)^{1-\beta} - 1} \right]^2 \bar{u}_z \qquad \text{(B-7)}$$

The above derivation is not intended to be complete for all surfaces and some corrections have been omitted in the interest of simplicity. It is intended to indicate the functional form of this transfer with the meteorological variables under most conditions of interest in the field. If we accept the assumption that the transfer of mass is equal to the transfer of momentum in this situation then the transfer velocity will be directly proportional to the wind speed and should vary with the stability of the atmosphere, being greater for the unstable condition and smaller for the stable condition. This conclusion is of some importance since it indicates that the amount of material deposited from the atmosphere is independent of the wind speed for a given stability. This is because the concentration from a point source decreases inversely with the wind speed while the deposition increases directly as the wind speed so that the two terms cancel. It is also of interest that the ratio of the turbulent transfer velocity to the wind speed is equal to the "drag coefficient" as given by Priestley[14] or one half of the drag coefficient as given by Deacon.[15]

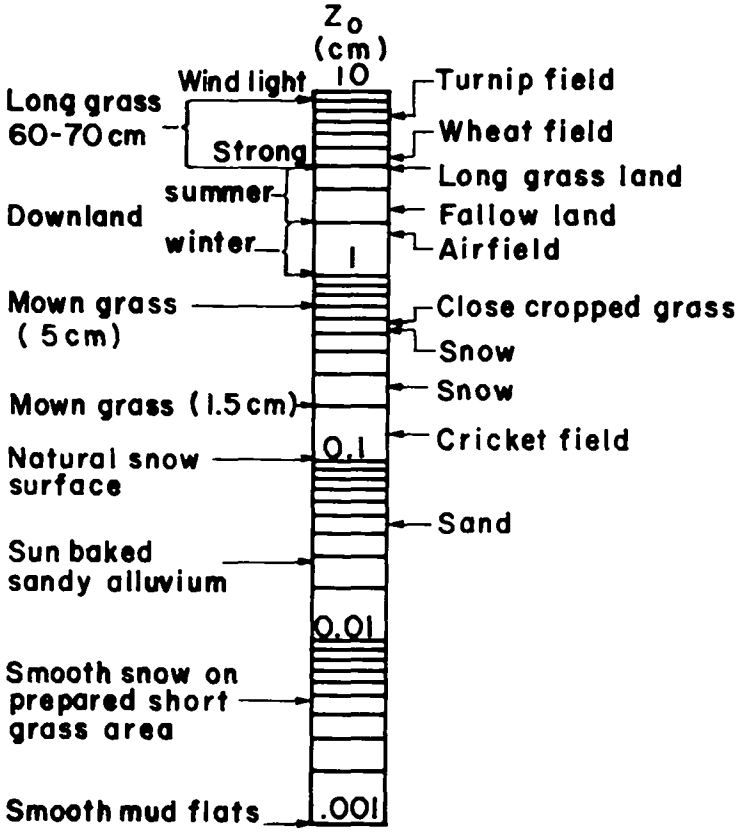

Fig. B-1. The Roughness Parameter of Various Surfaces (After Deacon.)[15]

We can obtain an estimate of the magnitude of the turbulent transfer velocity for various surfaces and the change with stability of the atmosphere by using values of z_o and β as given in the literature. The values of z_o chosen were those given by Sutton[13] as representative but to be used as general guides only. They may be compared with those of Deacon as given in Fig. B-1. Deacon[15] has plotted values of β as a function of stability (expressed as the Richardson's number) from measurements made over a short grass surface (z_o = 0.27 cm) and from observations over snow (z_o = 0.25 cm). We will assume, for purposes of illustration, that the value of β is independent of z_o and use these data to estimate the ratio of V_t to $\bar{u}$ as measured at a height of two meters ($\bar{u}_2$). (This is equivalent to calculating the transfer velocity for a wind speed of one meter per second at the reference height.) These values, along with the values of z_o are given in Table B-II.

It may be noted that the nature of the surface is more important in determining this transfer in the stable case than in the unstable. Thus, for high values of z_o, both the stable and unstable case are within a factor of two of the neutral case while for the low values of z_o the stable case transfer is lower by about a factor of twenty with the unstable case transfer higher by only a factor of six.

The variation in the turbulent transfer velocity is shown as a function of the stability expressed as the Richardson's number in Fig. B-2 for several of the values of z_o from Table B-II. The values of β for this plot were again taken from Deacon's plot and the Richardson's number is that for the layer of air between 0.5 and 4 meters.

TABLE B-II

CALCULATED VALUES OF $V_t/\bar{u}_2$

Surface	z_o (cm)	Stable $R_i = 0.08$ $\beta = 0.79$	Neutral $R_i = 0$ $\beta = 1$	Unstable $R_i = -0.2$ $\beta = 0.18$
Very smooth	0.001	0.000049	0.0011	0.0066
Grass up to 1 cm	0.1	0.00046	0.0028	0.0093
Thin grass up to 10 cm	0.7	0.0014	0.0050	0.013
Thick grass up to 10 cm	2.3	0.0029	0.0080	0.017
Thin grass up to 50 cm	5	0.0052	0.012	0.022
Thick grass up to 50 cm	9	0.0084	0.017	0.028

III. EXPERIMENTAL DATA

Data taken on an adequately controlled basis to permit checking of these concepts are scarce. In many cases the particle size or physical nature of the contaminant is not known while in others the wind speed or other meteorological variables are not given. Perhaps, the most common is the use of an isolated small area of a collection material either at ground level or at some arbitrary distance above the ground. The meaning of these results in terms of the local deposition is not known since the area is usually not large enough to establish the full turbulent layer over the test surface. For such surfaces on the ground, the final result is probably a mixture of the characteristic ground surface in the area and the retention characteristics of the test specimen.

A compilation of some of the data available on deposition are given in Table B-III separated according to stability. A brief discussion of the data identified by the letter in the source column of the table is given below.

A. These results come from experiments by Chamberlain and Chadwick and Megaw and Chadwick.[10] Elemental iodine was dissolved in CCl_4 and sprayed into the air. Measurements of the air concentration at several heights and of the deposited material were made across arcs at several distances downwind. The reference wind speed given in the table is at a height of two meters. It was assumed for the purposes of the tabulation that the measurements on a sunny day were in unstable conditions while those on a cloudy day or at dusk were in neutral conditions. Data are also given for the friction velocity. From this, the estimated turbulence transfer was calculated for a wind speed at the reference height of two

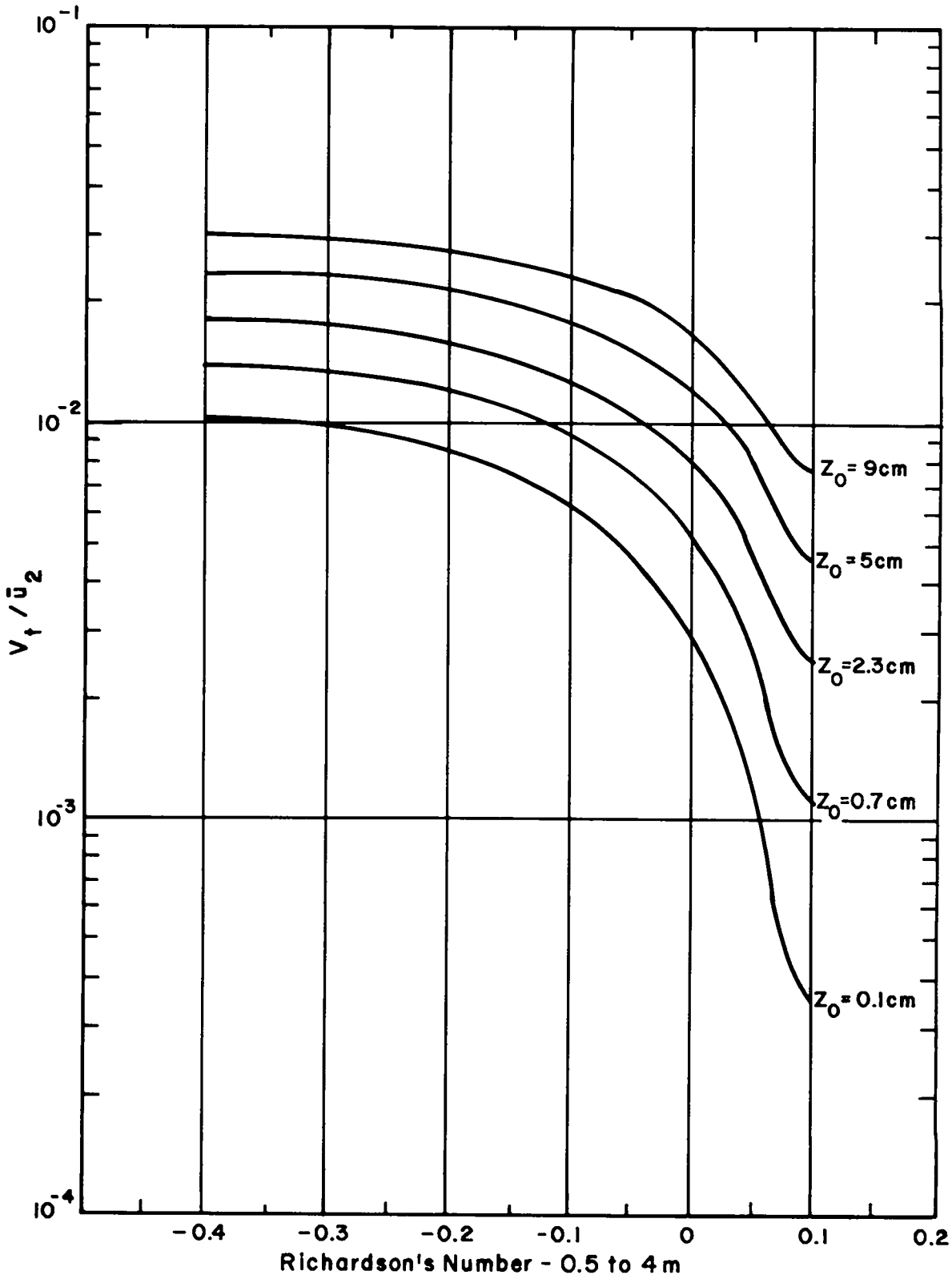

Fig. B-2. Change in Turbulent Transfer with Stability.

TABLE B-III

MEASUREMENTS OF DEPOSITION VELOCITY

Material	Source	Surface	$(V_d/\bar{u}) \times 10^2$
Stable Atmosphere			
ZnS tracer ~1 μm MMD	C	Desert	0.077, 0.88
ZnS tracer	G	Desert	0.85, 0.55, 0.78
Fission Products from Melted Fuel Element			
^{131}I	E	Sticky paper - 1 m	0.38
		Water	0.36
		Sand	0.12
^{137}Cs		Sticky paper - 1 m	0.018, 0.043, 0.029
^{103}Ru		Sticky paper - 1 m	0.64
		Water	0.86
		Sand	0.24
Zr-Nb		Sticky paper - 1 m	0.44
		Water	0.86
		Sand	0.98
Ce		Sticky paper - 1 m	0.14
^{131}I	F	Sticky paper - grd.	0.054, 0.10, 0.059 0.14, 0.16, 0.13, 0.22
		Sticky paper - 1 m	0.094, 0.052, 0.17, 0.16, 0.32
		Rye grass	0.42, 0.81
^{137}Cs		Soil	0.0091
^{103}Ru		Sticky paper - grd.	0.046, 0.14, 0.13, 0.20, 0.078, 0.078
		Sticky paper - 1 m	0.11, 0.22, 0.02, 0.15
		Grass	0.12, 0.31
		Soil	0.029
Te		Sticky paper - grd.	0.15
Unstable Atmosphere			
^{131}I vapor	A	Grass	0.37, 0.35, 0.91, 0.76
		Dandelion leaf	0.25, 0.30
		Paper leaf	0.39, 0.37
		Paper - Petri dish	0.12, 0.17, 0.15
ZnS tracer ~1 μm MMD	C	Desert	1.33, 0.94, 0.87 1.1, 1.1
Fission Products Arc	B	Grass & substrate	0.069, 0.049
		Filter paper	0.024, 0.018, 0.012
Neutral Atmosphere			
^{131}I vapor	A	Grass	0.60, 1.03, 0.28
		Dandelion leaf	0.30, 0.078
		Paper leaf	0.61, 0.16
		Paper - Petri dish	0.16, 0.18
^{131}I - melted fuel element	E	Sticky paper - 1m	0.16, 0.12
		Water	0.26, 0.34
	F	Sticky paper - 1m	0.13
		Sticky paper - grd.	0.16
		Rye grass	0.81

TABLE B-III (Continued)

Material	Source	Surface	$(V_d/\bar{u}) \times 10^2$
Rn daughters	D	Flat surface	0.01 - 0.02
ZnS tracer ~1 μm MMD	C	Desert	0.69, 0.47, 0.62, 0.56
Fission Products Arc	B	Grass & substrate Filter paper	0.023 0 023, 0.042
Fission Products from Melted Fuel Element			
^{137}Cs	E	Sticky paper - 1 m Water Sand	0.009, 0.017, 0.027, 0.055 0.01, 0.029 0.012, 0.009, 0.055
^{103}Ru		Sticky paper - 1 m Water Sand	0.20 0.25, 0.30 0.055, 0.063
Zr-Nb		Sticky paper - 1 m	0.12 0.71 0.18
Ce	F	Sticky paper - grd.	0.22
Te		Sticky paper - grd.	0.20

meters from Eq. (B-3). These values are compared to the measured deposition below.

Run No.	Calculated V_t/u	Measured V_d/u
1	0.0085	0.0037
2	0.0065	0.0060
3	0.0087	0.0035
4	0.0087	0.0091
5	0.0083	0.0103
6	0.0076	0.0074
7	0.0045	0.0028

B. Megaw and Chadwick[5] produced a fume of fission products by an arc between an irradiated wire and an electrode. Deposition was measured downwind along with the air concentration. Particles were probably submicron in size. Chamberlain[10] reports that cascade impactor samplers would indicate a particle size of 0.2 μm or less if the density of the particles was that of uranium oxide. It was noted that the deposition velocity of strontium from this experiment seemed to be less than that of the other solid fission products.

C. Islitzer and Dumbauld, as reported in Ref. 16, computed the deposition velocity for fluorescent tracer particles of one μm MMD from tracer material balance measurement at the National Reactor Test Station in Idaho over level terrain sparsely covered with sagebrush. They noted, in particular, a marked variation in the deposition velocity with stability. Measurements of the deposition velocity on flat, sand covered plates 0.1 m^2 in area were also made. In unstable conditions, these measurements indicated deposition velocities over an order of magnitude smaller than those found by the depletion technique.

D. Chamberlain[10] quotes Bullas as measuring the deposition of radon decay products onto flat surfaces. It was estimated that over 95% of the decay products would be attached to nuclei which Wilkening found to be about 0.02 μm median diameter. Bullas found the deposition velocity to depend on the wind velocity. For purposes of Table B-II it was assumed that the deposition quoted with "fresh" winds occurred in neutral atmospheres with wind speeds of about 5 meters per second. It is noted that in "calm" weather values of the deposition velocity were as low as 0.005 to 0.01 cm/sec. Measurements were also made by Bullas of the deposition

of fission products in long range fallout with deposition velocities ranging from 0.063 to 0.16 cm/sec. Similar observations by Stewart quoted in Ref. 10 gave a mean velocity of 0.07 cm/sec for the fission products. Booker is reported to have measured the ^{95}Zr component of long range fallout on filter paper with an average deposition velocity of 0.1 cm/sec. When he repeated the experiment indoors, the deposition velocity was 0.007 cm/sec although the atmospheric concentration was 80% of that outdoors. These values are not included in the table since there is no indication of the wind speed or stability.

E. A field experiment at the National Reactor Testing Station in Idaho was conducted for the Aircraft Nuclear Propulsion program.[17] Irradiated uranium fuel elements were melted in a furnace at ground level and the resulting fission products were measured downwind to a distance of about 3.2 km. Andersen samplers indicated that the bulk of the material penetrated to the backup filter. Particle size estimates for zirconium-niobium were 1 to 5 μm and for cerium 50% from 1 to 5 μm and 50% less than one μm. All others were estimated to be less than one μm. The majority of the deposition measurements were made on 13" x 13" sticky paper mounted on a support an unspecified distance (presumably about 1 meter) above the ground. Water trays with an area of 135 in.^{2} and sand trays with an area of 161 in.^{2} were placed at ground level and around sagebrush. The deposition velocity at each measurement arc was computed for the ration of the areas under the deposition profile to the area under the air concentration profile. This technique of fitting a Gaussian curve by area strongly weights the points in the middle of the profile and essentially ignores the points at the tail of the curve. If there were diffusion patterns leading to different deposition rates at the center and the tails of the curves, this technique would provide an estimate primarily of the centerline deposition. It may be noted that the points quoted in this test for unstable conditions are included in the neutral section of Table B-II. This is because the wind profiles for these tests were logarithmic and the Richardson's numbers estimated from the data available in the report were close to zero indicating a reasonably neutral atmosphere.

F. The second series of tests for the ANP project was made at the Dugway Proving Ground in Utah.[18] This is a very flat region with little vegetation or surface roughness to induce turbulence. Techniques were similar to the previous test except that the sticky paper was primarily used on the ground and patches of rye grass 8" x 6" at ground level were used for some of the tests. Most of the data were taken for stable atmospheres although one test had a small temperature differential between 4 and 16 m and was considered as neutral. It was concluded that there was some indication of a change in deposition velocity with wind speed but no change with stability. The data are variable, however, and the range in stability was not great. This report also indicated that the sticky paper used for the bulk of the measurements changes retention efficiency with the humidity of the atmosphere, thereby adding another variable to the measurements.

G. Simpson[19] has reported detailed measurements of the plume depletion and horizontal and vertical profiles of concentration following the ground level release of a zinc sulfide tracer (MMD ~2.7 μm) at Hanford. The deposition velocities were calculated from the estimated values of the exchange coefficient and the vertical concentration gradient. These runs were made under very stable conditions with Richardson's numbers at 1.5 to 3 m ranging from 0.046 to 0.223. He also reports values for the friction velocity which lead to estimates of the ratio of turbulent transfer velocity to wind velocity from 4.9 to 22 cm/sec per meter/sec.

The data from Tables B-II and B-III are compared for the neutral atmosphere in Fig. B-3. The terminal velocities for spherical particles with a density of four are included for comparison. Note that particles with a different shape or irregular

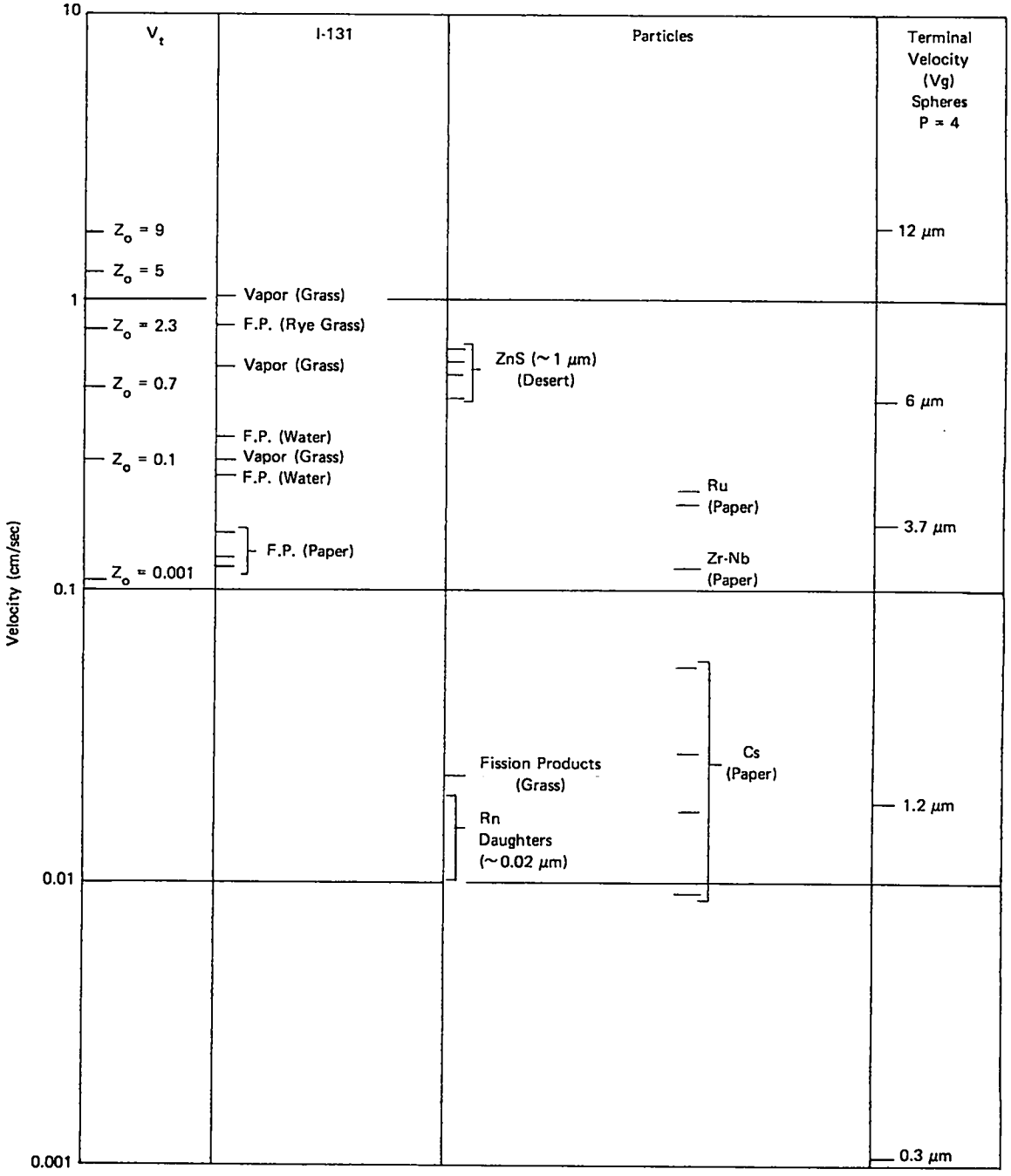

Fig. B-3. Transfer Velocities - Neutral Atmosphere.
$\bar{u}$ = 1 m/s

particles will have a lower terminal velocity.

IV. RETENTION ON SURFACES

The turbulent transfer velocity places an upper limit on the movement of the smaller particles through the boundary layer. If the material passing downward is retained on the surface with 100% efficiency, then the overall velocity of deposition must equal the turbulent transfer velocity. If, however, the particle rebounds from the surface or does not contact it because of inertial effects, then the retention can be less than 100% and the overall velocity of deposition will be smaller than the turbulent transfer velocity.

The iodine data over grass indicates that the measured deposition velocity for the vapor is close to that predicted by the turbulent transfer mechanism. The iodine from the fission product release may be somewhat low in its deposition on paper, but the form of the iodine, the retention characteristics of the sticky paper and its elevated position introduce uncertainties. The tracer material with a median diameter of about one μm similarly seems to have a high efficiency of deposition. It is of interest to note that the value of z_o, as obtained from two of the logarithmic wind profiles in the first series of release tests at NRTS, was about 1 cm, a value in close agreement with the deposition velocity measured for the same general area. Also, the measured deposition velocity for this material is much greater than the terminal velocity for gravitational settling indicating the importance of the turbulent transfer.

On the other hand, the data for radon daughters and fission products produced by an electric arc indicate deposition velocities much lower than would be indicated by turbulent transfer but still higher than would be predicted for submicron particles in gravitational settling. A similar pattern is shown for cesium in the ANP tests although the ruthenium and, perhaps, the zirconium-niobium velocities appear to be higher. These data lead to the conclusion that the finer particles, while transferred according to the theory, do not remain on the surface. Thus, the measured deposition velocity is lower than would be predicted. On the other hand, materials such as iodine vapor and μm-sized particles appear to be held with relatively high efficiency particularly on surfaces such as grass.

Possible mechanisms for retention on surfaces are varied and undoubtedly differ with the size and nature of the particle. We can speculate that absorption, adsorption, electrostatic effects, inertial effects and others may all be of importance under given conditions. In order to investigate the possible effects of one of these mechanisms, the inertial forces, a crude model was established and impaction efficiency estimated under several conditions. It is emphasized that these calculations are intentionally naive and are not intended to represent reality, but simply to illustrate one of the possible mechanisms.

Studies of the efficiency of impaction of small particles carried by an air stream have been made on a theoretical basis by Langmuir[7] and on an experimental basis by several investigators.[20,22] Figure B-4 presents the efficiency of impaction on a cylinder predicted by Langmuir with the results of several experiments for comparison. This efficiency is defined as the ratio of the quantity of material collected on the unit projected area of a cylinder to the quantity passing through the unit area normal to the direction of the flow. In Fig. B-4 this efficiency is correlated to the dimensionless parameter $V_g u/gC$ where V_g is the terminal velocity of the particle in free fall through the atmosphere, u is the velocity of the air stream, g is the acceleration due to gravity and C is the diameter of the cylinder upon which the particles are impacting.

It is noted that the experimental data are in reasonable agreement with the Langmuir prediction except for those points below the cutoff value of the parameter. The experimental values were obtained

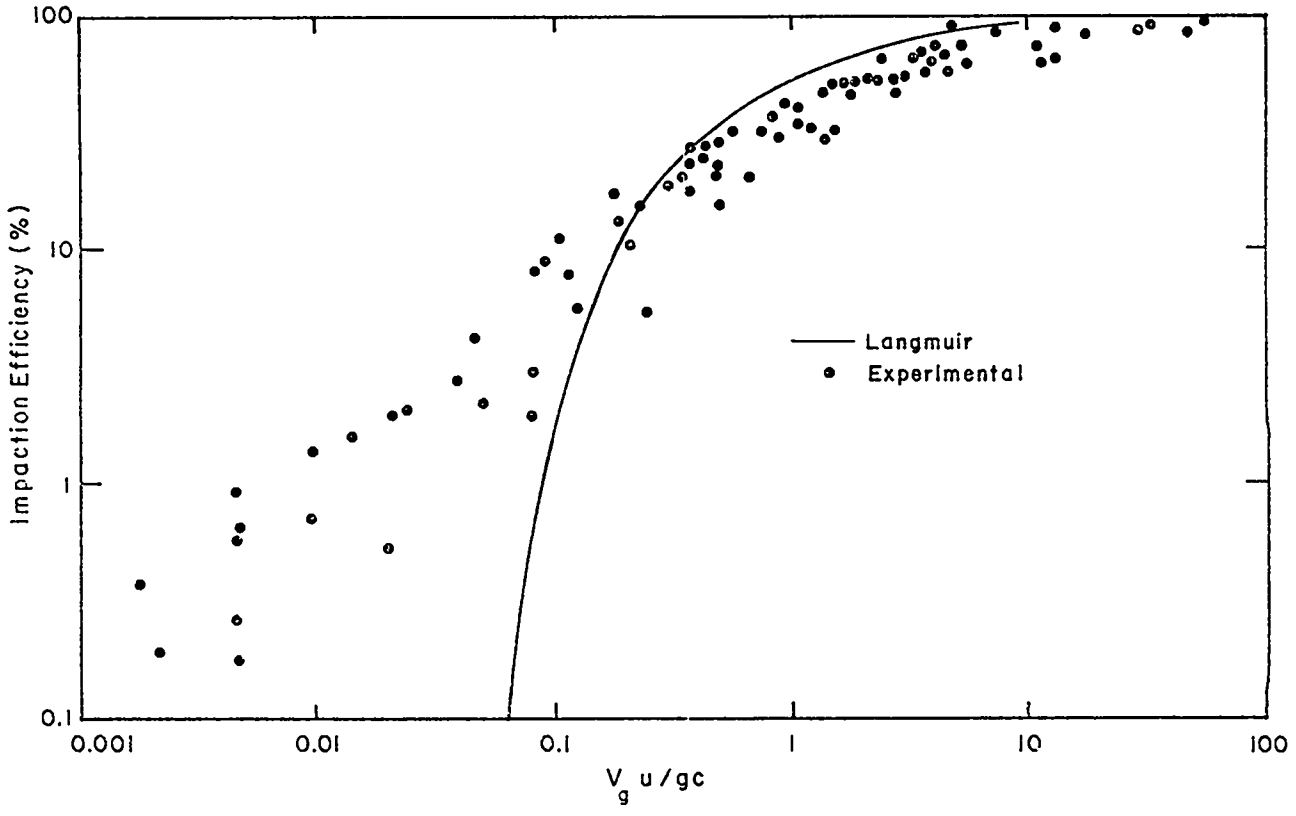

Fig. B-4. Efficiency of Impaction of Particles on Cylinders.

with aerosols of finite size distribution which means that there were particles much larger than the mean values used, and the measured collection at these small sizes was undoubtedly due to the presence of the larger particles. For the further calculations, the theoretical predictions of Langmuir will be used so that the values obtained will be characteristic of a uniform sized aerosol.

For these calculations, particles of density four were chosen since this corresponds to the zinc sulfide fluorescent tracer commonly used in meteorological experimentation. Figure B-5 presents the impaction efficiency for various values of u/C and particle sizes for spherical particles of density four. From these curves the impaction efficiency for various airstream speeds of diameters of impaction cylinders can be obtained. For example, for a one millimeter diameter cylinder, the curve of u/C = 10^4 gives the efficiencies for a ten meter/second airstream, while the curve of u/C = 10^3 gives the efficiencies for a one meter/second airstream. Conversely, for a ten meter/second airstream the curve of u/C = 10^4 represents the efficiency of impaction on a one mm cylinder while the curve of u/C = 10^3 represents the efficiency of impaction on a ten mm cylinder.

In order to illustrate the effects of this aerodynamic behavior of particles, a simple calculational model was used in which the particles are brought to the ground by atmospheric turbulence. Upon reaching the ground, they are carried past obstructions in the form of cylinders of several diameters where they are impacted according to the efficiencies given. Only the mechanism of impaction on cylinders was considered at this point so that any additional mechanisms such as electrostatic

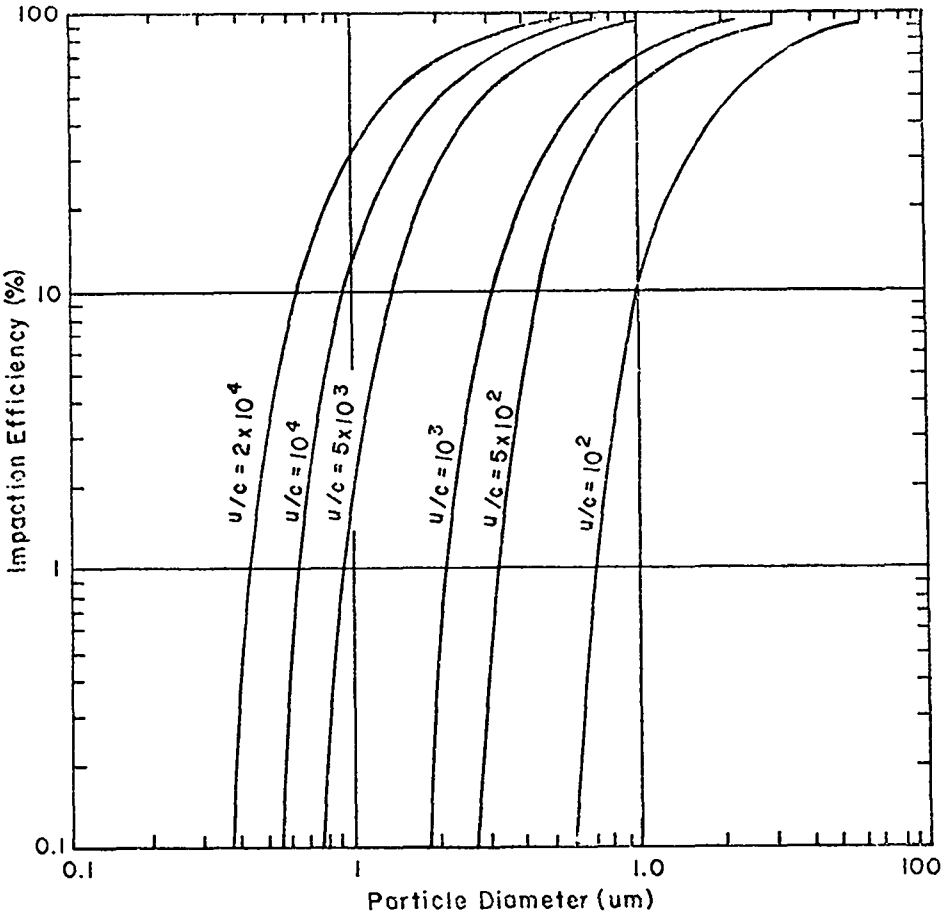

Fig. B-5. Impaction Efficiencies for Spheres of Density 4 g/cm^3

effects; impaction on surfaces other than cylinders; or, for the very small particles, diffusion to the interceptor surface will change the picture given.

It should be noted that two wind speeds are of importance in these calculations of the deposition velocity. The first is the wind speed at the boundary layer which determines the rate of mixing through the boundary layer or the limiting value of the deposition velocity. Thus, from the measurements available, the mixing phenomena can account for a deposition velocity of about 2.5 cm/sec in neutral conditions with a wind speed of 5 meters/second. At one meter/second the deposition velocity due to mixing should be about 0.5 cm/second. The other wind speed of importance is the speed of the airstream past the impacting surfaces. This is undoubtedly lower than the wind speed at the boundary layer due to surface friction effects and will probably vary with the nature of the surface (short grass, long grass, bushes, trees, etc.). In the present calculations, the speed of the airstream at the interceptor is taken to be the same as the wind speed since detailed information to choose a better value is not available.

The definition of the impaction efficiencies for Fig. B-4 must be considered in the model since they represent a fraction of the particles that are in the streamline intercepting the cylinder so that only those particles so exposed are represented. In other words, no correction is made for the relative areas of the interceptor surfaces and the total flow area. Two situations are calculated. The first uses the efficiencies directly from Fig. B-5. This assumes that the intercepting cylinders are so placed that each particle is on a streamline headed toward an interceptor once during the passage. The second

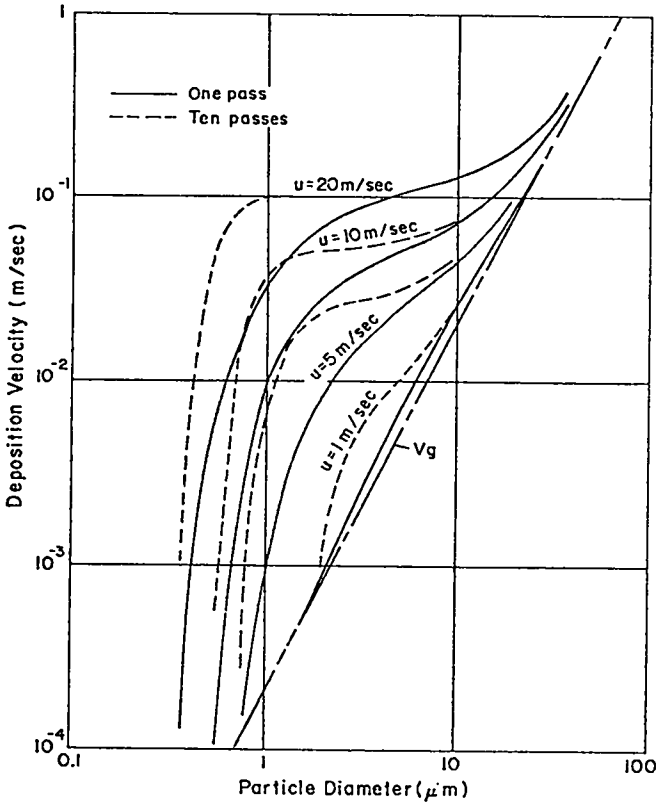

Fig. B-6. Calculated Deposition Velocity for Spheres of Density 4 Impinging on 1 mm Diameter Cylinders in a Neutral Atmosphere.

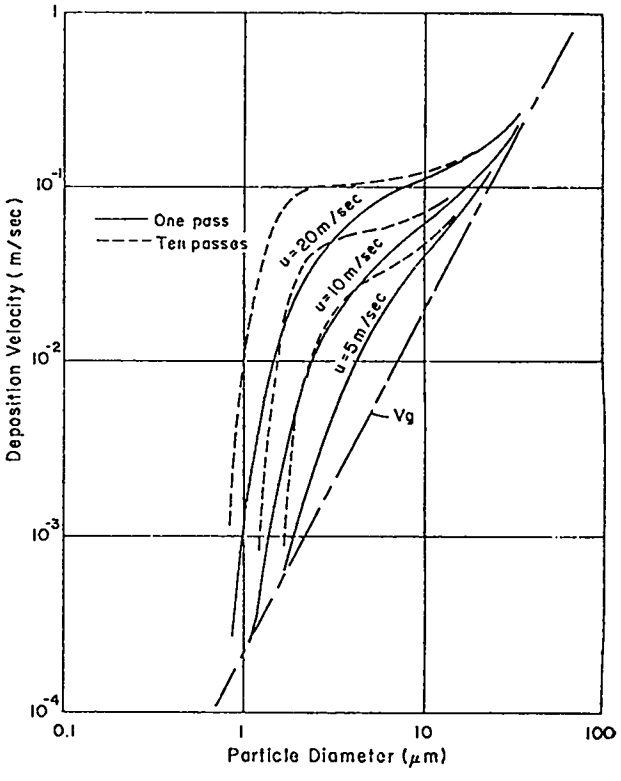

Fig. B-7. Calculated Deposition Velocity for Spheres of Density 4 Impinging on 5 mm Diameter Cylinders in a Neutral Atmosphere.

assumes a passage through a number of interceptors so that each particle is exposed to the chance of capture ten times.

The values of the deposition velocity in this idealized situation were computed from Eq. (B-8).

$$V_d = V_g + f\left(\frac{V_t}{u}\right)u \qquad \text{(B-8)}$$

Where V_d is the velocity of deposition, V_g is the terminal settling velocity of the particle, f is the efficiency of impaction, (V_t/u) is the limiting velocity of deposition from mixing across the boundary layer and u is the wind speed. Equation (B-8) is based on a simple additive process between the two mechanisms considered. It is doubtful that this is valid at low particle sizes where the energy which can be imparted to the particle by eddies in the atmosphere is much greater than the energy which can be dissipated in an equivalent time period by the simple settling of the particle.

In the case of the "multiple pass" over the interceptors, f was evaluated by:

$$f = 1 - e^{-f'n} \qquad \text{(B-9)}$$

Where f is the fraction remaining after n passes each with an efficiency of f' in removing material.

Figure B-6 presents the velocity of deposition calculated for both the "single-pass" and the "ten-pass" cases for impaction on one millimeter cylinders and wind speeds of 20 m/sec, 10 m/sec, 5 m/sec and 1 m/sec. It was assumed that the atmosphere had neutral stability so that (V_t/u) was 5×10^{-3}.

Figure B-7 presents the same calculations for impaction on five mm diameter cylinders.

V. DEPOSITION VELOCITY MODEL

From these considerations we can obtain a qualitative picture of the variation in deposition velocity with particle size. With large particles, the predominant mechanism will be gravitational settling. As the particles become smaller, they will become more likely to remain suspended by the turbulent air motions and, at some size, the velocity of settling becomes small in comparison to the turbulent transfer across the boundary layer. This transition size is a function of the stability of the atmosphere in determining the turbulence, the wind velocity, and the nature of the surface. From Fig. B-3 the transition size for a spherical particle with a density of 4 in a neutral atmosphere with a grass surface and a wind speed of 1 m/sec, would be about 6 to 10 μm. For shapes other than spherical, the transition size would be larger. Since the gravitational settling is not affected by the wind speed while the turbulent transfer velocity changes proportionately, the transition point occurs at larger particle sizes at higher wind speeds. In the previous example, but at a wind speed of 10 m/sec, the transition point would come at a particle size of 20 to 30 μm. Once the particles are brought to the ground, the probability of retention on the surface is undoubtedly a function of the particle size and the wind speed, although definitive data are not available on this. Variations in the retention with wind speed could account for some of the variability in the measured deposition velocity. One would expect that the larger particles would have greater retention by inertial impaction which would appear to be the primary force responsible for these particles. As is indicated by the data in Table B-III, the retention of zinc sulfide particles of one or a few μm appears to be high. Thus, the possibility of a plateau in the curve of measured deposition velocity versus particle size at the value of the turbulent transfer velocity seems probable. The length of this plateau and the particle size at which a significant decrease in retention occurs will probably depend upon the stability and the wind speed, but would seem to extend down to one micron or slightly less. Below this value the retention will decrease, although not as rapidly as the consideration of inertial forces would indicate, since new mechanisms, such as electrostatic attraction, will come into play for the very small particles.

Selection of numbers for this qualitative picture is difficult and uncertain because of the lack of detailed data. Using as a reference the turbulent transfer velocity for z_o = 2.3 cm, one can calculate the efficiency of retention for the measured deposition velocities in Table B-III. For the one μm MMD tracer particles at Idaho Falls, the efficiency of retention is 73% for neutral conditions, 63% for unstable conditions, and 28% for stable conditions. For the radon daughters on a flat plate the efficiency is about 2%. For the fission products produced by the arc and deposited on grass, the efficiency is about 3% in neutral conditions and 3.5% in unstable conditions. The deposition of Cs on the paper, sand, and water in the ANP tests varied from 2.5 to 3.4% in neutral conditions and from 3 to 10% in stable conditions. While the absolute magnitude of these numbers can vary with the assumption of z_o, the values are consistent with a rapid decrease in retention efficiency in the range of 0.5 to 1 or 2 microns with a relatively constant retention efficiency of about 1 to 5% at 0.1 micron. The other values in the ANP tests were not used here because of the indications of large particle size or the possible chemical reactivity of the ruthenium.

The information available obviously does not permit a detailed functional relationship between the particle size and efficiency of retention particularly when differences due to changes in stability, wind speed, and nature of the surface are included. For purposes of estimation, we will assume that the fission products produced by the electric arc in the experiments of Megaw and Chadwick[11] are about 0.1 μm with a retention of about 3% and the one μm tracer particles of Islitzer[22] have a retention of

about 70% with a linear relation between. Below 0.1 μm the efficiency is assumed to remain about 3% as based on the radon daughter deposition on the flat plate. The linear relationship was chosen as the simplest to represent the meager data, although it is probable that the actual relation is sigmoid with the steepest drop in retention somewhere between 0.1 and 1 μm. The linear relation can be approximately represented by f = 0.74d - 0.04 where f is the fractional retention and d is the particle size in μm. Extrapolation to 100% retention would indicate this to occur with particle sizes of about 1.4 μm, which is not in disagreement with the high retention implied by Simpson's data with 2 to 3 μm particles in stable atmospheres. At the upper end of the spectrum of particle sizes, it is assumed that the deposition velocity remains constant at the value for the turbulent transfer until $(V_t/\bar{u})\ \bar{u} = V_g$.

The particle density has not been included in the above considerations, again because of the lack of data on its influence on retention. Most of the experiments in Table B-III were run with particles of density ranging from about 3 to, perhaps, 10g/cm^3 so that the retentions chosen may represent reasonably realistic particles of concern.

From this crude model of retention and the turbulent transfer velocities of Table B-II, it is possible to approximate the deposition velocities for various particle sizes and limited types of terrain. Some of these approximations are given in Table B-IV.

It is again emphasized that the fractional retention values are particularly uncertain so that these deposition velocities must have wide limits of uncertainty until appropriate experimental data and study permit better estimates.

TABLE B-IV

APPROXIMATIONS OF THE RATIO OF DEPOSITION VELOCITY TO WIND SPEED AT ONE METER HEIGHT

Particle Size	f	Short Grass z_o = 0.1 cm	Thick Grass 10 cm z_o = 2.3 cm	Thick Grass 50 cm z_o = 9 cm
		Neutral, Ri = 0		
>1.5 μm	1.0	0.0028	0.0080	0.017
~1 μm	0.7	0.0020	0.0056	0.012
~0.5 μm	0.3	0.00074	0.0024	0.0051
<0.1 μm	0.03	0.00007	0.0002	0.0005
		Unstable, Ri = -0.02		
>1.5 μm	1.0	0.0093	0.017	0.028
~1 μm	0.7	0.0065	0.012	0.020
~0.5 μm	0.3	0.0028	0.0051	0.0084
<0.1 μm	0.03	0.0003	0.0005	0.0008
		Stable, Ri = 0.08		
>1.5 μm	1.0	0.00046	0.0029	0.0084
~1 μm	0.7	0.00032	0.0020	0.0059
~0.5 μm	0.3	0.00014	0.0009	0.0025
<0.1 μm	0.03	0.00001	0.00009	0.0003

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APPENDIX C

PARTICULATE LUNG DOSE EFFECTS

The following discussion appeared originally in a progress report for this study. It is reproduced here because of its importance to the subject of plutonium standards.

Current standards for limiting lung dose from internal emitters are based upon a calculation of the average dose delivered to the lung by assuming that the radiation absorption is uniform throughout the mass of the tissue. It is known that this condition does not exist for most "insoluble" radioisotopes which provide focal spots of high level radiation close to the particle decreasing with distance in a pattern dependent upon the type and energy of the radiation. Thus, a one μCi ^{32}P particle which, if the energy were averaged over the 1000 gram lung of the standard man would deliver a dose rate of 0.035 rads/day or a total dose of 0.75 rads, will deliver a dose rate of about 80,000 rads per day or a total dose approaching 2,000,000 rads to the tissue at 100 μm distance. Richmond, et al., report an alpha particle dose rate of 10 rads/hour at the surface of a 180 μm ^{238}Pu particle. The photon dose rate at the surface is about 500 rads/hour. Dose and dose rates drop off rapidly with distance from the particle so that the total volume of tissue involved is small. Table I illustrates the same point for the maximum permissible lung burden of plutonium if this lung burden is divided into uniform particles of various sizes.

From these illustrations, the physical conditions of such irradiation are vastly different from the uniform distribution and the particulate exposure results in a relatively small number of cells irradiated to widely differing doses. While one would expect differences in the outcome of irradiation of an organ by these two modes, it is not clear on a priori basis which would be the most damaging. For acute effects occurring after high levels, limiting the volume of tissue can greatly ameliorate the outcome. However, data are not available to indicate whether a similar situation exists for the late effects.

It is clear that this problem is a subclass of a more general problem in arriving at radiation protection standards--

TABLE I

RELATION BETWEEN PARTICLE DIAMETER, PARTICLE NUMBER, DISINTEGRATION RATE AND NUMBER OF CELLS IRRADIATED FOR A LUNG BURDEN OF 0.016 μCi $^{239}PuO_2$

Diameter (μm)	Number of Particles	Disintegration Rate ($d \cdot week^{-1} \cdot particle^{-1}$)	Number of Cells Irradiated[a]
0.01	5.4×10^{10}	6.7×10^{-3}	1.4×10^{13}
0.1	5.4×10^{7}	6.7	1.4×10^{10}
1.0	5.4×10^{4}	6.7×10^{3}	1.5×10^{7}

[a] For each particle size the number of cells exposed within a 40 μm alpha particle range is estimated. The total number of cells irradiated becomes the product of the number of cells irradiated per particle and the number of particles. A cell volume of 10^3 μm^3 is assumed.

that of nonhomogeneous dose in any organ. In order to focus more clearly on the important question, we have considered the current limits for uniform radiation to be acceptable and have, then, asked whether there is any evidence which indicates that the nonuniform radiation to an organ, such as occurs in extreme form in the particle problem, is more or less damaging than the homogeneous radiation. Thus, the focus is on the relative, and not the absolute effect. Further, since we assumed that no clear-cut information was available, we went to the literature to see if even a tentative conclusion could be made as to whether the preponderance of the evidence indicated which assumption should be made.

A. Review of the Literature - J. Furchner

A large number of papers and reports on radiation dose to the lung and subsequent damage was reviewed. One problem with much of the work reviewed, and particularly that having to do with individual implanted sources, was the lack of consistency in the dosimetry. Thus, some groups expressed the dose as the average to the lung while others calculated the dose at some reference distance from the source. While the original intent of this study was to attempt recalculation of the doses on a common basis, this was made difficult in some cases by the lack of data in the published article, the uncertainty of location of the source and the lack of time to complete a job of this magnitude, particularly when the initial appraisal indicated that the results would be uncertain.

For each paper of interest, or potential interest, to the present study a brief abstract emphasizing the actual data presented was prepared. Although such abstracts are of primary usefulness to those who are familar with the original article, they are presented below as orientation to the data available. In each case, comments by the abstractor are presented in parentheses.

1. IMPLANTED SOURCES

^{60}Co Warren and Gates 1960[2]

Mice 4-6 weeks old. ^{60}Co wire implanted through chest wall by trochar. Wire dimension: 2 mm by 0.5 mm. Radioactivity 170-250 µCi. Among 190 mice that survived 97 days (time of appearance of first lung cancer) 20 had carcinoma of the lung or bronchus. The last treated mouse died 315 days after implantation. The doses ranged from 90,000 to 460,000 rep. (Trauma to the lung is unavoidable; compare with Richmond et al. 1970).[8]

^{60}Co Warren and Gates 1968[3]

Mice, rats, hamsters, guinea pigs and rabbits. ^{60}Co wire, 2 mm long by 0.5 mm were implanted by trochar through the chest wall. The activity ranged from 70-636 µCi. Most sources were between 150-200 µCi.

Treatment Group (Species)	Number of Animals	Median Duration of Exposure (days)		Mean Total Dose (R x 10^3)		Malignant Cancer Incidence (%)	
		Lung	Esophagus[a]	Lung	Esophagus	Lung	Esophagus
Mice	286	180	188	262	145	20	15
Rats	20	204	202	353	121	75	30
Hamsters	25	495	368	443	178	8	28
Guinea Pigs	20	416	363	510	454	25	30
Rabbits	12	427	299	909	250	42	25

[a]Data do not include tumors at other sites or animals with no tumors. (As animal size increases the carcinogenic dose increases.)

^{106}Ru Laskin et al. 1963[4]

Rats. Hollow platinum cylinder plated with ^{106}Ru implanted in bronchus. 5 mm long, 1.2 mm diameter, wall thickness 0.2 mm. Hooks on cylinder maintained position in bronchus after implantation by trochar and tracheotomy.

Treatment Group µCi on implant	Number of Animals	Median Survival Time (Days)	Median Time to Cancer (Days)	Number Surviving 143 days[a]	Number with Lung Cancer[b]
0.008	37	340	430	27	2
0.057	56	310	400	39	6
0.59	57	320	380	40	15
5.0	67	225	325	42	25
13.6	48	190	315	29	20
Pt. control	60	330	---	--	--
Ru Control	22	320	---	--	--

[a]Two rats had cancer before this time.
[b]Squamous cell carcinoma.

The authors rearranged the groups on the basis of calculated doses for those animals surviving 143 days.

Average Dose (Rads)[a]	Number of Animals	Squamous Cell Carcinomas Numbers	Squamous Cell Carcinomas %
740	6	0	0
3,400	41	3	7.3[b]
36,000	41	9	21.9
460,000	57	33	57.9
1,600,000	32	21	65.6

[a]Dose calculated at "target" tissue taken as basal layer of the epithelium of the bronchus in which pellet was implanted - 100 µm from pellet surface.

[b] One tumor at 1400 rads. (There is no mention of the incidence of respiratory infections or causes of earlier deaths. The considerable trauma associated with implantation may be a factor).

^{106}Ru, ^{32}P Laskin et al. 1964[5]

Rats. Hollow platinum cylinder plated with ^{106}Ru implanted in bronchus. 5 mm long, 1.2 mm diameter, wall thickness, 0.2 mm. Hooks on cylinder maintained position in bronchus after implantation by trochar and tracheotomy. A single dose level of ^{106}Ru (5 µCi) was given. The animals were autopsied after spontaneous death and serial sacrifice. Phosphorus-32 pellets were also implanted in rats by this technique.

^{106}Ru

Mean Time of Death Sacrificed A	Mean Time of Death Spontaneous B	Number of Animals A	Number of Animals B	Mean Dose[a] (Rads x 10^5) A	Mean Dose[a] (Rads x 10^5) B	Cancer Incidence (%) A	Cancer Incidence (%) B
122	129	10	9	4.2	3.2	0	0
166	158	13	18	4.3	4.5	15.4	11.1
198	193	10	10	5.2	5.2	30.0	20.0
212	233	10	10	5.2	5.5	60.0	50.0
225	300	16	13	5.4	6.9	81.3	84.6
247	347	10	11	6.3	8.0	90.0	81.8
282	378	9	9	6.5	8.6	88.9	88.9
357	424	8	9	7.7	9.2	100.0	100.0

(No cancers before 158 days. Only 4 animals in 31 had lung cancers after doses of 4 x 10^5 Rads over 160 days. Again no mention of chronic respiratory infection is made).

^{32}P

Treatment Group	Number of Animals	Lung Dose[a] (Rads)	Lung Cancer %
20.0 µCi	18	4×10^5	56
2.0 µCi	15	4×10^4	33
0.2 µCi	15	4×10^3	0

(The ^{32}P dose response agrees with the ^{106}Ru data from Laskin et al. 1963, better than does the ^{106}Ru data in this paper).
[a] See note on Laskin et al. 1963 for meaning of dose calculation.

^{106}Ru Divertie, Titus and Shorter 1967[6]

Rats 150-200 g. Silicone rubber pegs (2.5 mm x 1 mm) impregnated with 50µm ceramic spheres containing ^{106}Ru were inserted into a bronchus via tracheotomy. Twelve control animals had inert pegs inserted into a bronchus. Thirteen of 16 rats receiving radioactive pegs had squamous cell carcinomas. None were found in controls. No doses are given.

ANIMALS WITH SQUAMOUS CELL CARCINOMA

Duration of Exposure (wks)	18	19	20	21	23	25	30	34	34	35	36	36	38
µCi Dose Inserted	15.6	14.0	10.0	13.4	13.4	13.4	11.3	8.1	14.8	12.6	13.4	12.6	14.1
Recovered	11.3	10.1	7.5	10.0	9.6	8.1	7.6	6.0	9.4	7.6	8.1	7.6	8.3

In two of the three experimentals no pegs were recovered. Pneumonitis was usually found distal to the pegs.

^{90}Sr Altmann, Hunstein and Stutz 1961[7]

Rats. Plexiglass capsules containing ^{90}Sr were sewed to the underside of the diaphragm. The activity range was 27-62 µCi with most values between 39 and 48 µCi.

Treatment Group Exposure Time (Months)	Number of Animals	Lung Tumors Number Carcinoma	Sarcoma	Adenoma
0 - 3	28	0	0	0
3 - 6	21	2	0	0
6 - 9	48	8	0	0
9 - 12	36	30	1	0
over 12	31	26	1	1

The last two groups also had nonpulmonary tumors. (There is no clue to dose data given).

$^{238}PuO_2$ Richmond, Langham and Stone 1970[8]

Rats, male, 325 g. Injection of PuO_2 spheres via femoral vein. Spheres 122-200 μm in diameter. The spheres are trapped in the capillary network of the lung. The animals were sacrificed serially and examined histologically. Note this method involves no surgical trauma to the lung.

Time of Sacrifice (Days)	1	3	7	14	21	30	60	90	120	152	180	211
No. of Animals	3	2	2	2	2	6	10	5	9	6	7	6

The surface dose rates were 10^8 and 10^3 rad/hr for the alpha and gamma radiations respectively. A sphere of cellular debris and collagenous tissue surrounded the spheres. A footnote, added in proof, states that animals at 600 days postinjection show histological changes qualitatively similar to those seen at 90, 120, and 211 days. No tumors were found.

^{90}Sr Cember and Watson 1958[9]

Rats, male, 286 g. Glass beads with incorporated ^{90}Sr were implanted with a hypodermic needle through the chest wall. The beads were 320± 110 μm in diameter and contained from 1.09 to 59.3 μCi. Dose data was given only for tumor bearing rats. The mortality data for the inert Sr and saline control groups was much the same as that for the experimentals and was due to injection trauma.

Dose Rate (Rad/day)	Exposure Time (Days)	Total Dose (Rads x 10^4)	Tumor
160	561	9.0	Lymphosacoma
160	487	7.8	Squamous cell carcinoma
220	561	12.0	Squamous cell carcinoma
277	169	4.7	Lymphosarcoma
277	545	15.0	Lymphoma
440	581	26.0	Squamous cell carcinoma
660	333	22.0	Squamous cell carcinoma

The first death occurred at 131 days and the last at 575 days post injection. (4 of 23 rats had squamous cell carcinomas at doses of $\sim 10^5$ Rads).

2. INHALATION OR INTRATRACHEAL INJECTION (Beta Emitters)

^{35}S Cember et al. 1955[10]

Rats, female, 125-200 g. A single intratracheal injection of $Ba^{35}SO_4$ particles (1.45 μm ± 0.40 μm).

Treatment Group	Number of Animals	Dose to Lung (rep)
4.5 μCi	23	58
45 μCi	21	3,200
4,500 μCi	38	24,000
Controls	25	-

Rats were killed serially in a 9 month period. No tumors were found in any group. (The calculated doses were delivered almost entirely during the first month). Chronic and acute inflamation were common.

^{35}S Cember and Watson 1958[11]

Rats, female, 244 g. Intratracheal insufflation of 375 μCi of $Ba^{35}SO_4$ once a week for 10 consecutive weeks. Particle size 1.45 μm ± 0.40 μm. Dose estimated to be between 12,000 and 20,000 Rads.

Treatment Group	Number of Animals	Number Surviving 10 weeks	Number Dead at 500 days	Squamous Cell Carcinoma
Colony control	14	--	2	0
Inert $BaSO_4$	24	17	10	0
3,750 μCi	24	16	24	2

The tumors were found in rats that died at 312 and 319 days.

^{144}Ce Cember et al. 1959[12]

Rats, male, 279 g. $^{144}CeF_3$ by intratracheal injection. Particle size 1.0 μm, Std dev 1.4.

Treatment Group	Number of Animals	Number of Survivors[a]	Days to First Tumor	Lung Dose (Rads)	Number with Squamous Cell Carcinoma
Colony control	20	--	--	0	0
Inert CeF_3	29	--	--	0	0
5 μCi	27	10	178	2,400	1
15 μCi	23	21	48	5,100	1
25 μCi	28	19	93	10,700	7
50 μCi	15	6	83	21,000	4

[a]To observation of first tumor. Severe, acute pneumonia appeared in the two high dose Groups within several days. The first tumor appeared in 48 days. (Mortality of the inert CeF_3 group was not given nor was the duration of the experiment).

^{144}Ce Cember 1963[13]

Rats, male, 283 g. $^{144}CeF_3$ by oral, intratracheal injection. Particle size 1.0 μm std dev 1.4.

Treatment Group	Number of Animals	Days to First Tumor Death	Lung Dose[a] (Rads)	Primary Lung Tumors	Squamous Cell Carcinoma	Undifferentiated Carcinoma	Adeno-Carcinoma	Lymphoma
Inert CeF_3	29	--	--	0	0	0	0	0
0.5 μCi	41	528	600	8	3	3	3	1
1.0 μCi	44	367	1,100	6	2	1	3	0
2.0 μCi	34	620	2,500	4	2	1	1	2
4.0 μCi	42	381	4,400	14	11	1	2	0

[a]Dose at death with first tumor. Earliest tumor at 361 days. (Experiment lasted at least 1,033 days. No mention of chronic pulmonary disease was made).

^{144}Ce Cember and Stemmer 1964[14]

Rats, male, 258 g. $^{144}CeCl_3$ solution given by intratracheal injection of 0.15 ml by mouth.

Treatment Group	Number of Animals	Number of Survivors[a]	Days to First Lung Tumor	Lung Dose (Rads)[b]	Number of Primary Lung Tumors
Inert $CeCl_3$	21	21	--	--	-
10 µCi	68	61	306	14,000	9
15 µCi	55	52	197	19,600	31
30 µCi	58	37	70	25,000	27

[a]Two month survivors

[b]Dose at death with first tumor

(Dosimetry differs somewhat from Cember et al. 1959, where the dose rate for 5 µCi $^{144}CeF_3$ is given as 59.6 rads/day in a 1.5 g lung. Here the dose rate is given as 28 rads/day for 1 µCi/g. Cember considers all the data for ^{144}Ce given in Cember et al.1959,[12] Cember 1963,[13] and Cember and Stemmer 1964[14] in Cember 1964.[15,16]

^{144}Ce Cember 1964[15,16]

Values given here are estimated from Fig. 39, Cember 1946a[15] and from Fig. 5, Cember 1964b.[16]

1964a		1964b	
Dose (Rads)	Tumor Frequency (%)	Dose (Rads)	Tumor Frequency (%)
660	2.2	650	1.5
1,300	4.0	1,200	2.5
2,500	5.6	4,500	5.0
5,500	9.5	14,000	10.0
15,500	12.5	20,000	12.5
26,000	22.2	41,000	22.5
43,500	29.8	49,500	25.0

^{144}Ce Hahn et al. 1973.[17]

Beagles. By inhalation of ^{144}Ce fused in clay particles. ~1.4-2.7 A.M.A.D., g std dev 1.5-2.3. To date 15 of 126 beagles are dead of fibrosis and pneumonitis at 143-410 days; and 5 are dead of pulmonary neoplasia at 750-1,318 days.

Initial Lung Burden		Time To Death	Dose to Lung At Death (Rads)[a]	Lung Pathology
µCi/kg	Total µCi	(Days)		
26	230	765	27,000	Hemangiosarcoma
27	190	1,185	23,000	Hemangiosarcoma
34	330	1,318	36,000	Hemangiosarcoma
35	380	916	34,000	Hemangiosarcoma + bronchiolocarcinoma
46	470	750	48,000	Hemangiosarcoma + fibrosarcoma
33	320	193		Pneumonitis +fibrosis,no tumors
41	330	185		" " "
51	440	410		" " "
53	440	279		" " "
56	520	273		" " "
65	590	234		" " "
66	470	246		" " "
66	540	257		" " "
68	600	186		" " "
96	740	189		" " "
120	890	171		" " "
180	2,000	182		" " "
190	1,500	173		" " "
190	1,700	181		" " "
210	1,700	143		" " "

[a]Calculated by Hahn et al. (It appears that more than 700 days must elapse before tumors are found).

^{144}Ce Kurshakova and Ivanov 1962[18]

Twenty rabbits 2.5 - 3.0 kg were injected with 25 μCi of ^{144}CeF by piercing the anterior wall of the trachea through the skin. The particle size was 0.025 μm. One rabbit died of bronchopneumonia on the 3rd day. Half of the rabbits died between the 60th and 238th days of sclerosis, bronchiectasis, etc. Tumors were found in 6 of the animals surviving to 238 days. The last tumor was found at 327 days. The doses to the lungs at 238 and 327 days were 51.4 and 68.9 kilorads respectively. There were 5 bronchogenic and alveolar lung cancers and one squamous cell carcinoma of the esophagus.

^{106}Ru Temple et al. 1960[19]

Mice, female. $^{106}RuO_2$ in Tween-80 was injected intratracheally.

Treatment Group	Number of Animals	Days After Administration	Adenomas %	Number of Malignant Tumors	Dose (Rads)
Colony Control	28	403-470	78	0	0
Inert RuO_2	21	335-500	9	0	0
3.0 μCi	23	350	82	1 Bronchiolar carcinoma	9,000
1.93 μCi	11	369-422	90	1 Bronchiolar carcinoma	4,000
0.15 μCi	10	340	--	1 lympho sarcoma	300

(The natural incidence of adenomas is a factor of unknown importance to radiation carcinogenesis).

$^{152-154}Eu$ Berke and Deitch 1970[20]

Rats, female, 180-200 g. Rats were made to inhale aerosols of radio-europium chloride for 7 h/day, 5 days/wk for 6 months. The particles were characterized only as "submicronic". The lung dose varied with time and was as much as 6×10^4 rads at 720 days. No animals were free of pulmonary pathology. Severe chronic inflammatory changes and lung abscesses were present in the majority of the animals. There was a complete absence of pulmonary neoplasia.

^{24}Na, ^{32}P, ^{59}Fe, ^{198}Au Kochetkova et al. 1959[21]

Rats. These isotopes were given by intratracheal injection. Particle size unspecified.

Treatment Group	μCi	Number of Animals	Beta Dose (Rads) First Day	Beta Dose (Rads) Total	Metaplasia of Bronchial Epithelium Number	Metaplasia of Bronchial Epithelium Months	Lung Cancer Number	Lung Cancer Months
$^{59}Fe_2O_3$	1-27	52	10-20	500-5000	17	6-9	8	6-9
$Cr^{32}PO_4$	40-100	76	300-700	1300-15000	20	2-12	11	6.5-18
^{198}Au	100-150	30	1400-2100	5400-8000	4	1-3	3	2.5-12

100-200 μCi of $^{24}NaCl$ in single and multiple doses produced no tumors.

^{32}P Kochetkova and Avrunina 1957[22]

Rats - Intratracheal injection of $Cr^{32}PO_4$. No particle size specified.

Treatment Group (μCi injected)	Number of Animals	Mean Lung Burden (μCi)	Lung Dose (Rep) 24 h	Lung Dose (Rep) Total	Life Span (Days)	Pathology
270	10	180	2,900	19,000-46,000	3-32	Metaplasia
100	42	81.5	830	10,000-18,000	19-65	Metaplasia
70	34	54	670	8,000-16,000	15-395	3 Squamous Cell Cancers
40	10	38	350	4,500- 7,400	60-451	3 Squamous Cell Cancers

There were no tumors after single and multiple injections of $^{24}NaCl$ (200-1,900 μCi). Of 25 rats that received 320 μCi of radiogold all died in 2.5 months. Three of these rats had squamous cell cancer. The doses were in 9,000-9,700 rep range.

3. INHALATION OR INTRATRACHEAL INJECTION (Alpha)

^{239}Pu Temple et al. 1960[19]

Mice (BAF) PuO_2 suspended in Tween-80 or Pluronics for injection. Particle size 0.6-0.06μm mean 0.5 μm.

Treatment Group	Number of Animals	Days After Administration	Dose (Rads)	Lung Tumor
Colony Control	28	400	---	22 Adenomas
0.16 μCi	41	100	4,000	1 Bronchiolar carcinoma
0.06 μCi	17	400	2,300	2 Squamous cell carcinomas
0.003 μCi	21	500	115	1 Fibro sarcoma

At 400 days 78% of the colony controls had adenomas. The fibrosarcoma at the 0.003 level was considered non-radiogenic (the use of surface active agents as vehicles for the particles is a factor of unknown importance).

^{210}Po Yuile, et al. 1967[23]

Rats, male, exposed once to an aerosol of ^{210}Po as the chloride, particle size: geometric mean 0.098μm, geometric std dev 1.81.

Treatment Group	Number of Animals	Number of Deaths	Age Range At End (wks)	Lung Dose[a] (Rads)	Primary Lung Tumors	Squamous Cell Carcinomas
NaCl Control	147	88	87-100	0	0	0
0.15 μCi	119	119	106	538	22	17
0.05 μCi	129	98	95-100	202	15	5
0.02 μCi	132	71	89-91	71	4	1

Dose accumulated at 280 days - little increase thereafter. The aerosol was a NaCl solution acidified to a pH of 1. Pulmonary infection was endemic in the colony and an epidemic of acute pneumonia occurred during the second year. The experiment was terminated when the last high-dose animal died at the 96th week.

^{239}Pu Wager et al. 1955[24]

Mice (BAF) female - Intratracheal injection with Tween 80. Particle size 0.05 to 0.6 μm. Of 10 mice that received 0.06 μCi of $^{239}PuO_2$, 3 had squamous cell carcinoma at 1 year post-injection.

^{210}Po Scott and Thomas 1957[25]

Rats. Intratracheal injection of ^{210}Po nitrate solution. Experiment terminated at 15 months when there were 5 survivors.

Treatment Group	Number of Animals	Squamous Cell Carcinomas	Time to Tumor (weeks)
10 μCi/kg	15	0	----
5 μCi/kg	15	2	5, 15

All animals exhibited varying degrees of murine pneumonia.

^{210}Po Little et al. 1970[26]

Syrian golden hamsters. ^{210}Po adsorbed on 3 mg of Fe_2O_3 particles (98% < 0.75 μm): suspended in saline given in 15 consecutive weekly intratracheal injections.

Treatment Group	Number of Animals	Number of Dead Animals	Current Week	Tumor Bearing Animals No.	%	First Tumor	Total Dose at 2 yr (Rads)
Control	63	52	93	0	0	----	----
Fe_2O_3 only	32	30	93	0	0	----	----
0.2 μCi/wk	35	35	60	32	91	15th wk	4,500
0.01 μCi/wk	34	21	59	10	30	40th wk	225

The number of animals consists of the survivors of the 15-week treatment period which were autopsied (60 animals/group at start). The doses given are maxima-carcinogenic doses which are less than 225 rads.

^{210}Po Grossman et al. 1971[27]

A later report on Little et al. 1970[26] gives the incidence of bronchogenic tumors as 91% and 43% in the high and low dose groups respectively.
Syrian golden hamsters were given intratracheal injections twice weekly for 7 weeks. The doses were given in two separate intratracheal instillations (a and b in table below).

Treatment Group a	Treatment Group b	47 wk Survivors %	Tumors at 27 wk
3 mg Fe_2O_3	0.2 μCi in saline	2	17
Saline	0.2 μCi	6	9
Saline	0.2 μCi on 3.0 mg Fe_2O_3	28	7
Saline	0.2 μCi on 0.3 mg Fe_2O_3	32	3

^{210}Po alone is said to be homogenously distributed in the lung.

^{238}U Leach et al. 1970[28]

Monkeys, dogs, and rats were exposed to UO_2 dust (M.M.D. 1.03 μm, g std dev 2.40) 5 mg/m^3 for 6 h/day, 5 days/wk. The rats, after an exposure of 1 year, showed no pathological changes in the lung apart from pigmented macrophages in the alveoli and bronchi. In dogs there were no pathological changes in the lung after 5 years of exposure and estimated radiation doses of 400 rads. Monkeys responded with a patchy hyaline fibrosis that first appeared at 3.6 years after a dose of 500 rads. No tumors were reported in any animals at the end of the 5 year exposure. Despite doses to the tracheobronchial lymph nodes of dogs and monkeys that were on the order of 10^4 rads no pathology other than an occasional necrosis and fibrosis were reported.

^{239}Pu Antonchenko et al. 1969[29]

Rats, 140-160 g, were exposed to an aerosol (90% 0.7-1.9μm, median diameter 1 μm) of Pu citrate or ammonium plutonium pentacarbonate. (pH 5 and 8, respectively) for 20 min.

Treatment Group	Number of Animals	Average Life Days	Average Life Dose (rads)	Lung Pathology (%) Carcinoma	Adenoma	Adenoma-like Structures	Epithelial Metaplasia
Controls	248	673	0	---	---	----	---
Citrate							
1.028 μCi[a]	23	64	3,820	---	---	----	9.1
0.803 μCi	12	69	3,090	---	---	----	8.3
0.511 μCi	94	124	2,370	2.2	---	8.9	73.4
Carbonate							
1.460 μCi	12	77	7,320	---	---	----	9.09
0.774 μCi	23	78	3,900	---	---	4.4	13.0
0.455 μCi	69	139	2,780	4.6	3.08	12.0	61.6

[a]Initial deposition. (Apparently the short survival time in the higher dose groups precluded the development of the characteristic pathology).

^{239}Pu Buldakov et al.[30]

Rats. Inhalation of soluble Pu compounds: citrate and ammonium pentacarbonate.

Treatment Group (μCi deposited)		Number of Animals	Mean Survival (Days)	Lung Dose (Rads)	Lung Tumors %
Citrate	0.008	157	635	47	5
	0.02	124	585	117	2.5
	0.04	203	545	234	8.4
	0.08	31	546	467	35.5
	0.15	105	464	852	23.8
	0.25	113	416	1,390	23.0
	0.36	39	221	1.740	7.7
	0.51	90	124	2,370	0
	0.80	12	63	3,090	0
	1.03	20	64	3,820	0
Ammonium-Plutonium-Penta-Carbonate	0.004	48	571	41	4.2
	0.007	101	571	80	5.0
	0.017	91	584	186	13.2
	0.045	126	582	497	36.4
	0.15	83	484	1,065	42.7
	0.25	126	361	1,615	26.4
	0.35	22	247	2,140	9.0
	0.46	65	139	2,780	0
	0.77	23	78	3,900	0
	1.46	11	77	7,320	0

The tumors were squamous cell carcinomas, adenocarcinomas and hemangiomos. Note the tumor incidence at low doses and the absence of tumors at high doses.

^{239}Pu Clark et al. 1964[31]

Dogs inhaled particles (0.5-0.65 μm). At 855 days 28 were dead. There was one lung tumor at 150 days. Six more died between 855 days and 1,446 days, of these, four had bronchiolo-alveolar tumors. The estimated doses were between 9,000 and 23,000 rads, resulting from burdens of 0.6 to 19 μCi.

^{239}Pu Park et al. 1967[32]

Dogs inhaled particles (0.5-0.65 μm) (continued from above). Of 25 dogs dying or sacrificed between 850 and 2,270 days, 12 had primary pulmonary tumors. The estimated doses to the tumor bearing animals ranged between 3,100 and 13,600 rads, resulting from terminal lung burdens of 0.5-2.7 μCi.

^{239}Pu Park et al. 1972[33]

Dogs inhaled PuO_2 particles (0.5-0.65 μm) (continued from above). Of 65 dogs exposed, 62 are dead, and 24 had pulmonary neoplasia. Between 55 and 1,600 days, 36 died of pulmonary insufficiencies (edema, fibrosis, hyperplasia etc.) Twenty of 21 dogs surviving 1,600 days had lung tumors. Estimated initial lung burdens were 0.2 to 3.3 μCi. At 11 years the average dose to the lungs of tumor-bearing animals was in the 2,000-12,000 rads range.

^{238}Pu Park et al. 1970[34]

Twelve dogs inhaled $^{238}PuO_2$ particles: CMD 0.05 μm GSD 1.9.

Terminal Burden (μCi)	Lung Burden (% Terminal Burden)	Survival Time (Days)
261	92	27
167	94	30
168	93	35
112	92	56
74	91	56
140	94	61
84[a]	90	70
88[a]	90	76
58	91	77
44[a]	91	94
17[a]	80	125
25	77	180

[a] animals with lung tumors: bronchiolo-alveolar carcinoma. Dose range, all animals, 8,000 to 26,000 rads. Almost total necrosis of tracheobronchial, mediastinal, and sternal lymph nodes.

^{238}Pu C. L. Sanders 1973[35]

Rats, female, were exposed to an aerosol of crushed $^{238}PuO_2$ microspheres: CMD 0.02, GSD 2.1. The material was considered soluble (72% ultrafilterable). Life-time study (>1,000 days).

Treatment Group	Number of Animals	Lung Dose[a] (Rads)	Lung Tumors %	Median Life Span (days)
Control	92	0	1.1	825
5 nCi[b]	30	9	6.6	~ 650
18 nCi[b]	30	32	23.3	675
230 nCi[b]	30	375	25.0	550

[a] mean dose in 2 years
[b] mean initial lung deposition

Lung tumor incidence in the 5 nCi group was not significantly different from lung tumor incidence in the control group. Of the 19 pulmonary tumors found, 14 were bronchiolo-alveolar carcinomas, 2 were mixed carcinomas and there was one epidermoid carcinoma, one undifferentiated carcinoma and one lymphosarcoma. The author concludes: " - - - that spreading the Pu dose in the lung, as compared to concentrating in PuO_2 particles, is more carcinogenic due to the greater number of epithelial cells 'hit' by alpha emissions from Pu".

^{241}Am Thomas et al. 1972[36]

Dogs were exposed to an aerosol (AMAD 0.9 μm, GSD 1.5).

Lung Burden (μCi) Initial	Lung Burden (μCi) At Sacrifice	Days to Sacrifice	Lung Dose (Rads)	Lung Pathology
31	2.3	127	3,000	Inflammation
21	1.0	256	3,200	Fibrosis
26	0.71	512	3,800	Fibrosis and mineralization
23	0.38	1,022	5,300	Fibrosis and mineralization

Doses to lung were delivered early; more than 90% of the ^{241}Am had left the lung by 127 days. The highest doses were delivered to the tracheobronchial lymph nodes (3,500-17,400 Rads), but the chief pathologies were fibrosis in the medullary areas and depletion of lymphoid elements.

^{237}Np Levdik et al. 1971[37]

Rats were injected intratracheally with nitrate and oxalate solutions of ^{237}Np (pH 2-3 and 5, respectively).

Treatment Group (μCi/kg)	Number of Animals	Average Life Span(Days)	Dose (Rads)	Lung Tumor Incidence (%) Malignant	Benign
Control	274	700	0	3.65	0.36
Nitrate					
0.017	50	660	5	16.0	2.0
0.083	44	684	28	20.5	4.65
0.41	48	685	138	12.68	8.35
2.0	49	505	2,500	14.28	2.1
Oxalate					
0.017	85	645	27	10.6	0.0
0.083	89	661	134	9.0	3.36
0.41	89	649	671	28.0	5.6
2.0	81	453	3,220	37.45	3.75

Part of the increased carcinogenicity is attributed to the chemical toxicity of Neptunium.

4. EXTERNAL IRRADIATION

X-ray Koletsky and Gustafson 1955[38]

220 kV, 15 ma, filters: 1.0 mm Al, 0.5 mm Cu, 60 R/min.

Rats, male, 200 g were exposed to a single total dose of 660 R of whole-body radiation. The 123 rats that survived 6 months or more were autopsied at death.

Treatment Group (Time of Death)	Number of Deaths Irrad.	Number of Deaths Con.	Number With Tumors Irrad.	Number With Tumors Con.
6-12 months	46	3	7	1
12-18 months	47	6	32	0
18-24 months	29	14	29	2
over 24 months	1	13	1	5

One rat had primary carcinoma of the lung, an undifferentiated carcinoma in the lower left lobe. The right lung had an adenocarcinoma. The time of appearance is not given.

X-ray Cember et al. 1956[39]

100 Kv, 4 ma, filters: 1 mm Al. 61.8 R/min.

Rats, female, 270 g; equal doses on 5 consecutive days. Only the thoracic region was exposed. The rats were rotated 4 times during each exposure.

Treatment Group	Number of Animals	First Death (wks)	Median Lethal Time	Last Death	Tumors
Control	20	No losses	---	---	---
5,750	20	6	12 months	6 Sac.@ 15 months	2 lymphoma
11,500	20	3	166 days	11 months	1 lymphoma
17,250	15	3	37 days	6 months	1 lymphoma

The primary loci of the tumors is uncertain because of metastases. Broncho-pneumonia was the most common finding.

X-ray Maisin et al. 1958[40]

250 kV, filters: 1.0 mm Al, 0.25 mm Cu. 90 R/min.

Rats, 145-165 g were exposed to single doses of whole or partial body irradiation. Only animals alive at 6 months post exposure were considered. These survivors were autopsied at death.

Treatment Group	Number Autopsied	Total Number of Cancers	Leuco-Sarcomas	Epithelio-mas	Sarcomas	Epithelio-Sarcomas
Control	460	13	10	2	1	---
300-2,000 R	1,237	100	24	52	21	3
2,000 R left lung	10	5	--	3	2	---

Only one rat (cervico-sternal shield - 600 R) receiving less than 2,000 R had a pulmonary cancer (at 15 months post exposure). Of the 5 cancers in the 2,000 R group, which had the entire body, save the left lung region, shielded, 3 were bronchial epitheliomas appearing at 11, 15, and 23 months. There were 2 sarcomas of the left thoracic wall. No other pulmonary pathologies were reported.

X-ray Castanera et al. 1968[41]

200 kV, 15 ma, filters: 1.0 mm Al, 0.5 mm Cu. 27 R/min.

Rats, male, free of epidemic respiratory infection were exposed to single, whole-body irradiation with x-rays or fast neutrons (12 Mev H^+ on Be).

Treatment Group (Rads)	Number of Animals	Time of Last Death (Days)	Adenomas (%)	Adenocar-carcinomas (%)
X-ray				
430	88	700-800	16	3
680	107	600-700	11	4
Neutrons				
230	41	500-600	17	7
320	73	500-600	10	6
Control	129	800-900	1	0

Tumor frequency was estimated from chart 5. Operable tumors were removed surgically. There were no other primary pulmonary tumors.

Castanera et al. 1971[42]

Male rats were given a single whole-body exposure to fast neutrons (12 Mev H^+ on Be). The rats were free of epidemic respiratory infections.

Treatment Group Age (months)	Dose (Rads)	Number of Animals	Median Survival Time (Days)	Primary Lung Tumors Benign	Malignant
1	215	79	433	13	3
1	0	40	699	5	0
3	230	41	436	17	7
3	0	41	601	0	0
21	215	53	167	8	2
21	0	24	158	4	0

All tumors were bronchiolar in origin. Multiple tumors were found in other organs.

X-ray DeVilliers and Gross 1966[43]

135 kV, 4 ma, filters: 2.43 mm Al. ~100 R/min.

Male Syrian golden hamsters and male rats were exposed to 5 equal doses of x-rays delivered on 5 consecutive days. A collimated beam was directed at the chest region. Four portals were varied through 90° per day. Hamsters received 4,000 R, rats 3,570 R.

	Time of Sacrifice (Months)	Number of Animals	Tumors Adenomas	Malignant
Rats	4	12	1	---
	8	11	---	---
	12	12	2	1 Adenocarcinoma
	24	12	2	2 - Reticulum cell sarcoma Squamous cell carcinoma

	Post Irradiation Time (weeks)											
Hamsters	6	7	10	11	12	13	14	15	16	17	18	19
Spontaneous Deaths	1	1	2	3	3	13	6	4	2	2	2	3
Squamous Cell Cancer	-	-	-	-	-	2	-	2	1	-	-	-

Fifty-seven hamsters were irradiated, all died spontaneously. Only those dying between 6 and 20 weeks (42) are listed. Pulmonary cancers were not found later than 3 1/2 months, although 4 at 6 months and at 12 months and 13 at 24 months, whereas 7 of the eight tumors found in rats were found at 12 or 24 months.

X-ray Gross et al. 1969[44]

3,000 R, 110 kV, 6.4 ma, filters: 1.83 mm Al. 75 R/min and 4,000 R, 110 kV, 8.6 ma, filters: 1.83 mm Al. 100 R/min.

Rats and hamsters were exposed to a collimated beam of x-rays directed at the chest region. The animals were rotated axially at 7 rpm during exposure which was given in 5 equal doses on 5 consecutive days. Eight weeks after exposure some animals were treated with dimethyl benzanthracene (DMBA) and/or jewelers rouge (Fe_2O_3).

Treatment Group (Rats)	Number of Animals	9 Month Survivors	Adeno-carcinoma	Squamous Cell Carcinoma	Fibro-sarcoma	Undiffer-entiated
4,000 R						
DMBA + Fe_2O_3	40	37	15	3	1	1
Fe_2O_3	40	29	7	0	0	1
Radiation only	43	42	18	4	1	1
3,000 R						
DMBA + Fe_2O_3	40	39	14	2	1	0
Fe_2O_3	40	39	16	0	0	0

Data for the appropriate controls are not tabulated. In the unirradiated rats Fe_2O_3 with or without DMBA caused no tumors. DMBA did not increase the prevalence of cancer in irradiated rats.

Treatment Group (Hamsters)	Number of Animals	2.5 Months Survivors	Number of Cancers	Time To Tumor(Months)
4,000 R				
DMBA + Fe_2O_3	40	37	2	2.5, 7.5
Fe_2O_3	38	36	0	---
Radiation only	46	46	1	11
3,000 R				
DMBA + Fe_2O_3	38	38	0	---
Fe_2O_3	38	37	1	14
Radiation only	8	8	0	---
Control				
DMBA + Fe_2O_3	40	35	2	9.7, 18.5
Fe_2O_3	20	19	0	---
No treatment	10	10	0	---

The first appearance of a cancer was the base for the selected survival times (column 3). Only frank malignancies are included, microlesions were not considered. Chronic bronchitis was prevalent. (Note the decreased incidence of cancer among hamsters compared with that of DeVilliers and Gross[43]1966).

B. Discussion - J. W. Healy

Prior to World War II and the Manhattan Project, radiation exposure limits had been derived for X-rays or radium gamma rays, for radon in the air and for radium as an internal emitter.* The external limits were based on the radiation field to which the individual was exposed with little or no consideration of the distribution of radiation through the body or of the exposure of specific organs.** During the Manhattan Project, the need for considering radiations other than X- or gamma, the presence of varying energies of radiations and the availability of a wide variety of radioactive chemical species resulted in the extrapolation of these limits to the new conditions through the derivation of new concepts (such as the rem) and an increased sophistication in dosimetry as applied to individual organs.

Following the war, considerable attention was given to formalizing these concepts in a manner which could be used by those responsible for guiding radiation protection practices in the vastly increased uses of radiation and radioactive materials resulting from nuclear energy. This work was carried out by the NCRP in consultation with foreign scientists through conferences and informal discussions. In 1954, the NCRP subcommittee on Permissible Internal Emitters published their report[45] that first expounded on the critical organ concept which has served as the basis for the majority of the internal emitter limitations.* Here the critical organs were defined on the basis of experience with external radiation. The skin was chosen as one organ because of the production of skin cancers, usually on the hands from the greater exposure which they received. The increased incidence of leukemia in radiologists led to the designation of the blood-forming organs as one of the more important critical organs, while cataracts produced by high LET radiations resulted in the lens of the eye receiving special designation. Since leukemia was the primary outcome from whole body radiation in the experience available, it was considered "- - - safe to assume at present that the blood-forming organs constitute the most critical organs".** Exposure to the more deeply seated organs was then limited to that of the blood-forming organs.

In the 1954 NCRP document, the limits for the blood-forming organs and other organs were established at 0.3 rems per week (if received every week this would be essentially 15 rems per year). This is the limitation used by the Internal Dose Committee[46] in obtaining their values for organs other than bone. In 1957, the NCRP again revised their recommendations for

*For an excellent review of the information available on the effects of internal radiation on humans at the time of World War II, the reader is referred to "The Tolerance Dose" MDDC 1100 by S. T. Cantril and H. M. Parker.

**In this statement we are referring to the official limitations adopted by the NCRP and the ICRP. Individuals did concern themselves with these matters in reviewing the data available and in applying the limits.

*Subcommittee 2 on Permissible Internal Dose published its report in 1953[46] listing MPC's and maximum permissible body burdens based on the critical organ concept. The dose limitations were those given in the later report of the external dose subcommittee and seem to reflect the NCRP decisions arrived at in the later report.

**Genetic considerations are not pertinent to this review but they were not ignored. "From the point of view of genetic damage manifestable in future generations, the gonads, of course, constitute the critical tissues 'par excellence'." However, the contribution of occupational exposures to the dose to the population as a whole was not considered limiting.

***Bone limits were based upon a biological comparison with radium.

workers to lower the radiation doses to the whole body, head and trunk, active blood-forming organs and gonads to an average of 5 rems per year over the working years beyond age 18.[47] However, the recommended limits for internal organs other than thyroid, skin, and gonads remained at 15 rems per year. In the same document the NCRP recommended levels of one-tenth of those for workers for individuals outside of the controlled area. The latest report of the NCRP[48] continues the use of 15 rems per year for organs other than red bone marrow, skin, and gonads for occupational workers, but recommends a limitation of 0.5 rems per year to individual organs for members of the general public.

Thus, it can be seen that the current limitation of 15 rems per year for the lung of workers can be traced to the original critical organ concept and the dose limitations derived from early experience with external radiation. The recent lowering of the recommended limit for the lung of individual members of the public by the NCRP is by a factor of three and is expressly indicated as being "- - - based primarily on the desire for numerical simplicity in the standards and not on an established biomedical need." At the same time, the 1971 NCRP recommendations include a concept of "significant volume" over which the dose should be averaged. The implication being that any redistribution of a given dose within this volume would not significantly affect the outcome. The 1971 NCRP report continues, "It is usually assumed that the 'significant volume' should be of the order of one cubic centimeter. This will be grossly conservative under most circumstances, and in special estimations, use of a larger volume is justified."*

Although the original decision to use the average dose to the lung (or other organs) was made in the early period of the derivation of dose limitations, it should not be inferred that those bodies responsible for such recommendations have ignored the subject. In the Chalk River Tri-Partite Conference with scientists from the U.S., U.K. and Canada,[49] the statement is made: "In relation to the possible pathological effects of radioactive particulates in the lungs, Dr. Hamilton pointed out that the cells in the immediate neighborhood of a dust particle containing 1 or 2% of plutonium would be subjected to a dose of about 400 r/day. The general opinion which emerged from the discussion was that the carcinogenic effect per unit volume is probably considerably less for the irradiation of small masses of tissue than for large." The ICRP has addressed this general question of non-uniform dose periodically, usually by special groups commissioned by the ICRP to study the question. In its Publication 9 (1966),[50] the ICRP stated:

"In the case of non-homogeneous distribution of absorbed dose in the lung, an estimate of the Dose Equivalent to the whole lung, determined merely by the product of QF and the mean absorbed dose, may be greatly in error, but our full understanding of this problem must await further experimental evidence. In the meantime there is no clear evidence to show whether,

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*The foregoing review has been greatly shortened to indicate the salient points in the derivation of the current lung limitations. At the same time, it has focused on the NCRP recommendations because of their importance in the early days when the present limits were first derived. The ICRP recommendations differ in detail but follow the same general pattern. The reader with interest in this subject is urged to review these documents for further detail.

with a given mean absorbed dose, the biological risk associated with a non-homogeneous distribution is greater or less than the risk resulting from a more diffuse distribution of that dose in the lung." In Publication 14 (1969)[51] prepared by two Task Groups of ICRP Committee 1, the irradiation from radioactive particles was considered specifically. Here, it is stated: "The problems of high local concentration of dose are at their most severe with radioactive particulate material in the tissue, especially with alpha emitters. Here the local dose can reach very high values even though the mean tissue dose may be very low. Certainly it cannot be assumed that linearity of dose and effect will hold at these high doses and dose rates. On the other hand, there may be a great deal of cell death, and particularly with alpha emission, with its short and well-defined range, the number of affected but viable cells may be small compared with the number of killed cells. However, this ratio will depend on the size and activity of the particles, the extent to which they aggregate, and their movement within the tissue, and the movement of the cells past them.

"On the basis of general considerations and some experimental data and clinical experience the Task Group were of the opinion that, for late effects, the same radiation energy absorption might well be less effective when distributed as a series of "hot spots" than when uniformly distributed. Thus, with particulate radioactive sources within a tissue, a mean tissue dose would probably introduce a factor of safety. However, a severe practical problem has now been recognized in connection with the inhalation of plutonium particulates, and is now being considered in detail by a Task Group of Committee 1 of ICRP."

The Task groups also considered the problem of translocation of plutonium to lymph tissue and concluded:

""In the meantime, the Task Group are of the opinion that any immediate change in the dose limit for plutonium on the basis of risk of lymphoid tissue is not warranted."

The potential outcome of an inhalation of radioactive materials can be changed by a number of factors. If, for example, the material is readily translocated from the lung to other organs, the eventual damage to these other organs may well appear earlier than, and overwhelm any lung damage.* Thus, in considering lung dose we are focusing primarily on those materials which will be retained in the lung for reasonably long periods of time. If the quantity in the lung is large enough, death will result at early times due to pulmonary insufficiency resulting from an adema or destruction of functional living tissue. In practice, we are interested in low dose effects which will occur late in life and carcinogenesis would seem to present the end point of greatest interest. Life shortening has been noted in many experiments, particularly at higher levels, and is used as a criterion of damage. The statistical uncertainties in most experiments occasioned by the limited numbers of animals and the variation in death times make this a relatively nonsensitive indicator, even though the argument can be made that a finding of no significant life shortening is of importance since a death is a death, regardless of whether it is caused by a heart attack or a cancer. However, in many experiments in which life-shortening was not significant, the incidence of cancer at the end of life was significant, indicating that radiation effects did occur. As a result, the present studies focused primarily on cancer incidence as being the appropriate end point.

*Of particular interest in this respect is the recent work at Battelle Northwest[52] which indicates that certain forms of $^{238}PuO_2$ are rapidly translocated from the lung to the bone when inhaled resulting in the production of bone tumors.

In most of the experiments there appears to be a relation between the radiation dose and the time of occurrence of malignancies in animals: In general, the higher the dose (or in case of internal emitters, the dose rate) the shorter the time required for cancer production. This phenomenon is frequently used to invoke the possibility of an "effective threshold" since the time required to permit cancer formation following a low dose will be so great that it exceeds the normal life span even if the induction follows a linear relation with dose. However, in interpreting data, it must be borne in mind that the opposite phenomenon will occur when the dose or dose rate becomes too high. That is, the animal will die from other causes before there is time to induce cancer. This was seen in the results from the dogs at Hanford[31,33,34] where the early deaths were due to pulmonary insufficiency with cancers eventually appearing only in the animals with lower lung burdens and which had lived most of their life span. Thus, if radiation dose is used as a primary parameter in investigating incidence, it is important not only that the animals live out their normal life span so that the full cancer incidence develops, but that the total dose is not so high that deaths occur from other causes before the cancer can develop. These conflicting trends in causes of death can result in an apparent optimal dose for the production of malignancies. However, even at this optimal dose, the full expression of the malignancies possible per unit dose at lower values will not occur.

Akin to this concept is that of "overkill" of single cells close to the particle. In the case discussed above, the production of early death by causes other than cancer can be regarded as a result of "wasted radiation" in interpretations based upon the narrow concept of carcinogenesis as an end point.* From this standpoint, doses which lead to death before cancer appears can be considered to be overkill of the organism since the full expression of the carcinogenic effects is not attained. For a single particle in the lung (or other tissue) the dose rates at close approaches to the particle can be high enough so that even a relatively limited time of residence in the tissue will result in the death of cells within a given radius depending upon the activity of the particle and the type of radiation. Such cells will not be able to later reproduce and, regardless of the degree of damage, will not lead to cancer.** From this standpoint, therefore, one would expect that particles which lead to such overkill would be less hazardous than uniform radiation to the overall organ since not all of the radiation is used in attaining the final end point, cancer. In fact, such a concept would lead immediately to the conclusion that the larger the particle (in terms of activity) the less effective it would be in producing cancer since the dose rates close to the particle would increase as the activity increased thereby leading to a greater fraction of radiation wasted on dead cells. One clear cut experiment possibly showing this effect was done by Passonneau[1,53] using Sr-90 beads on rat skin. Here the same amount of activity was used for the same area of skin but the activity was distributed either as a uniform flat plate, in 50 beads, in 20 beads or in 10 beads. The results given in Table II indicate clearly a decrease in the tumor production efficiency as the activity was

*We have already mentioned that this is an appropriate end-point for consideration of dose limitation since it appears to be the latest effect in time to occur even when other effects are relatively ineffective in shortening the life span.

**However, the presence of dead cells, cellular products or fibrosis may be required before a cellular transformation can express itself as a cancer. This is an interesting possibility which needs more study.

TABLE II

TUMOR PRODUCTION IN RAT SKIN

UPON EXPOSURE TO FLAT PLATE AND POINT SOURCES

Source	Activity	No. of Animals	No. of Tumors	Tumors per μc	Relative Efficiency
Flat Plate 1000	28.6 μc/cm^2	71	89	4.94×10^{-4}	1.59
Flat Plate 1500	42.9 μc/cm^2	73			
50 beads	30 μc/bead	58	27	3.10×10^{-4}	1.00
20 beads	75 μc/bead	77	24	2.08×10^{-4}	0.671
10 beads	150 μc/bead	74	16	1.44×10^{-4}	0.464

subdivided into more active particles. Gamertsfelder, in an analysis of these data,[1] assumed a mid-lethal dose for cells of either 4635 or 9300 rads and a probability of tumor production increasing as the nth power of the dose to the cell. He then calculated the ratio of the number of tumors expected relative to those produced by the 30 bead configuration. The range of the experimental data is not great enough to permit distinguishing between the curves represented by different values of n but within this limited range, the calculations fit the observed trend. It is of interest to note that these calculations indicate a maximum in the relative efficiency of tumor production if n is greater than 1 while if n is equal to one, the curve approaches an asymtote as the activity per particle gets smaller. The value of this asymtote for the assumed median lethal dose of 4650 rads is 3.2 and for 9300 rads is 2.42. Since the condition where the activity per particle becomes very small is essentially that of a uniform, plane source, the comparison between this value and the value of 2.4 noted in the experiment (corrected linearly from the 1000 μCi flat plate source data) may be of significance. A somewhat similar calculation by Langham and Dean[54] but on an absolute basis, to predict the probability of tumor production from various sizes of plutonium particles, used data derived by Albert[55] on the production of tumors in rat skin versus dose to the cell. The results of this calculation show a very high probability of tumor production from most particle sizes. However, as the authors indicate, the paper was published to illustrate the method rather than to provide results. The results of this work can be questioned on many grounds including the use of the data on tumors in rat skin for lung tissue, the finding of Albert that the sensitive cells are at the base of the follicle in the rat skin and the fact that the assumed efficiency of production of lung cancer per cell does not conform to the experience with humans in the production of lung tumors from external radiations.

The results of wasted radiation in the production of lung fibrosis at high levels of administration of radioisotopes or the induction of other causes of death before cancer can develop raises the question of the possible effects of such wasted radiation in the particle case. Richmond, et al.[8] investigated the effects of Pu-238

dioxide particles lodged in the lung vascular following IV injection. These particles averaged about 180 μm in diameter and gave average dose rates to the entire lung of about 3.5 rems per hour with the dose rate in the vicinity of the particle on the order of 10^9 rads per hour. The longest exposure until sacrifice was a group of 6 rats which lived to 600 days. Examination of the lung following these exposures indicated the presence of a microleison with complete degeneration of the cells close to the particle. However, the evidence indicated that this was not simply a stable type of scar tissue but rather that the lesion was in a dynamic state in which the collagen was renewed constantly with subsequent liquification. Within this time period there was no indication of effects which would be deleterious to the animal's overall well being. It is noteworthy that the energy delivered to the lung, if averaged over the full lung would be on the order of 2,000,000 rads,* well in excess of those doses which have been shown to produce deaths in relatively short times when more uniformly distributed and considerably above the doses required to produce lung cancers.

One of the uncertainties with such an analysis of overkill of cells is, of course, the possibility of movement of the particles within the lung tissue so that the number of cells at risk becomes much greater and the doses delivered become smaller. In the experiment of Richmond, et al.[8] quoted above, the particles were relatively firmly held in the blood vessels and, therefore, were not representative of particles actually deposited in the alevoli. Movement of such particles is known to occur through ejection with mucus and movement by the cilia and by engulfment by macrophages. Thus, quantitative estimates of the degree of overkill of cells and the fraction of radiation wasted would be uncertain since such movement is difficult to model. However, it would seem that such arguments would be of more interest in the actual quantitative sense than in the conceptual sense. If the particles are large enough so that very high dose rates are encountered in the near vicinity, there still, will be a degree of overkill and wasted radiation although it may be considerably lower than would be estimated by the static model.

Additional uncertainty is added by the possible reactions of the cells located at the periphery of the zone of destruction caused by the radiation. This would involve cells receiving radiation doses ranging from just sublethal to essentially zero. If there is attempted repopulation of the volumes of destruction, this could result in rapid proliferation of these cells which have already been damaged. This situation would appear to be the most serious contender for the production of cancer and also one which would be the most difficult to investigate experimentally without an understanding of the basic mechanism of cancer production and the response of individual cells to these conditions in an otherwise normal environment and surrounded by otherwise normal cells. Information on this possibility is limited, but some indication that it is not a predominant problem can be obtained from the experiments of Passonneau[53] and Richmond[8] which did involve just such conditions in several types of tissue.

The outstanding example of increased carcinogenity of a deposited radioactive material due to localization and nonuniform dose distribution is plutonium in bone.

*Richmond, et al.[8] indicates that Halley has estimated the average dose to a human lung for the same size of particle to be 3.5 rems per hour. Using an RBE of 10 for alpha particles and considering the rat lung to be on the order of 1/500th the mass of the human lung, the dose in 600 days becomes:

$$\frac{3.5}{10} \times 500 \times 24 \times 600 = 2{,}500{,}000 \text{ rads.}$$

Here, the classical work of Brues[56] led to the conclusion that plutonium is about five times as effective for the same energy deposition as is radium, which is, in itself, nonuniformly distributed. Studies of the comparative deposition in bone of these two isotopes have indicated that the radium, being chemically similar to calcium, tends to deposit in the mineralized portions of the bone and eventually is distributed through the bone mineral by remodeling or is covered by new layers of calcified materials. By contrast, the plutonium is deposited on the bone surface in locations where it is adjacent to the regenerative cells and, in remodeling of the bone tends to redeposit on these surfaces. Thus, this represents the case of a very nonhomogeneous organ where the comparative isotope (radium), while not uniformly distributed, is more uniformly distributed than the plutonium. Further, the plutonium is preferentially deposited in the vicinity of the regenerative cells which are presumably more sensitive to the induction of cancer than the mineralized bone. This situation would seem to represent a localization of the radiation dose at cells which present a more sensitive target and therefore, eliminates some of the wasted radiation which occurs with radium in the mineralized portion of the bone. In essence, the bone can be regarded as composed of three regions of differing criticality: the marrow, the proliferating cells on the bone surfaces and the mineralized portion which has minimal metabolic activity and serves primarily as a structural supporting member for the body. In this case, the sensitive tissues are the marrow and the regenerating cells with the regenerating cells of most interest for plutonium as the average dose to the marrow from the poorly penetrating radiations from plutonium is comparatively low. Again, however, some significant dose rates to the marrow on a localized basis can be calculated. These are to a small fraction of the marrow falling within a few tens of micrometers of the deposited plutonium. The fact that leukemia is a relatively rare outcome in experimental animals given plutonium may serve as an indicator that irradiation of a small portion of an organ (the marrow) to a high dose is not particularly troublesome as long as the average dose is low.

A similar situation may, of course, occur in any organ as a number of different cell types can be present in the same organ and any mechanism which results in preferential irradiation of the more sensitive cell types could, theoretically lead to the same type of result. The high incidence of lung tumors in uranium miners from radon in mine atmospheres is attributed to the deposition of the particulate daughters of radon on the bronchi, particularly at points of division where the turbulence in the air stream produces increased impaction and deposition.

The estimation of the radiation dose to the assumed critical tissue, the bronchial epithelium, is complicated by the uncertainties in the areas of deposition and the thickness of the mucus layer which serves to absorb some of the energy of the radon daughters deposited on the surface. However, in a review of the dosimetry for the Federal Radiation Council[57] Parker considers, with important reservations, that one working level month corresponds to a dose to the bronchial epithelium of 2.8 rads. The working level for exposure to radon daughters is defined as any combination of radon daughters in one liter of air that will result in the ultimate emission of 1.3×10^5 MeV of potential alpha energy. One working level month, then, is the total exposure resulting from working in such an atmosphere for 170 hours. If we assume that all of the alpha energy associated with the daughter products is released in the lung (i.e. all of the daughters are deposited and none are eliminated before

they decay) the average dose to a 1000 gram lung would be 0.44 rads. This is undoubtedly a maximum estimate since some of the daughters will be exhaled and a portion will be eliminated by ciliary action. However, much of the activity is associated with small particles which are deposited in the bronchi and lower pulmonary regions with relatively high efficiency. The linear velocity of particles moving up the bronchi is 0.25 to 1 cm/min while in the trachea rates can increase to 3 cm/min.[1] Because the longest half-life of the radon daughters of interest is 26.8 minutes, it would appear that a sizeable fraction of the material deposited in the bronchi would decay before elimination and that all of the material deposited below the ciliated region would contribute their full energy. If we apply this estimate of the average lung dose to the estimated exposures of the uranium miners in those exposure ranges where the incidence of lung cancer is high, we find that the dose to the total lung calculated on an average organ basis is, indeed, significant and in the range where animal data would indicate such an outcome to be expected.* Since there is uncertainty about the actual significance of the increase in lung cancer at the lower exposure levels, we will not discuss this phase. However, the dose levels corresponding to the exposure ranges used in the epidemiological study[57,58] assuming an average dose to the lung of 0.44 rads per WLM are listed in Table III.

TABLE III

AVERAGE LUNG DOSES CORRESPONDING TO LEVELS OF EXPOSURE USED IN THE URANIUM MINER EPIDEMIOLOGICAL STUDY

Exposure WLM	Average Lung Dose-Rads
< 120	< 53
120 - 359	53 - 158
360 - 839	158 - 370
840 - 1799	370 - 792
1800 - 3719	792 - 1636
> 3720	> 1636

An additional argument concerning the present bases for radiation protection standards should be included in this discussion. As a basis for dose limitations, it is normally assumed that the response to a given dose is proportional to the dose received and that there is no threshold. While there is considerable evidence to support the use of this assumption, there is also evidence that the dose rate is an important factor, at least for low LET radiations, with the response decreasing as the dose is protracted, presumably due to the repair of the damage in the intervening time before the full dose is accumulated.* Acceptance of this assumption would indicate that the result of a dose to a small portion of a given tissue would be the same

*In order to permit a rapid appraisal of the data presented in the abstracts on the incidence of lung cancer at various dose levels, Fig. 1 presents a crude plot of the data for the alpha emitters. No attempt was made in this plot to reevaluate the dose estimates or to correct for experiments in which the incidence was measured before the full life-span of the animals. The five points at the lowest doses were the results of the ^{237}Np and the ^{210}Po administrations. The human data are estimates of doses received by a group of 37 individuals exposed during work with plutonium and represent periods of time ranging from 4 to 24 years after exposure.[59]

*We note that the same argument cannot be made for alpha emitters since current evidence indicates that the damage from high LET radiations is not repaired. Thus, the assumption of linearity with dose, regardless of dose rate, would seem to be more appropriate for these materials than for the gamma or x-rays. As an aside, we also note that the amount of repair for gamma radiations appears to be on the order of 90%. If we assume no repair for the alpha radiations, the late result (after repair is over) would be about ten times as great for the alpha radiations as for the gamma. This appears to be about the same as the commonly accepted RBE or Quality Factor for alpha radiations.

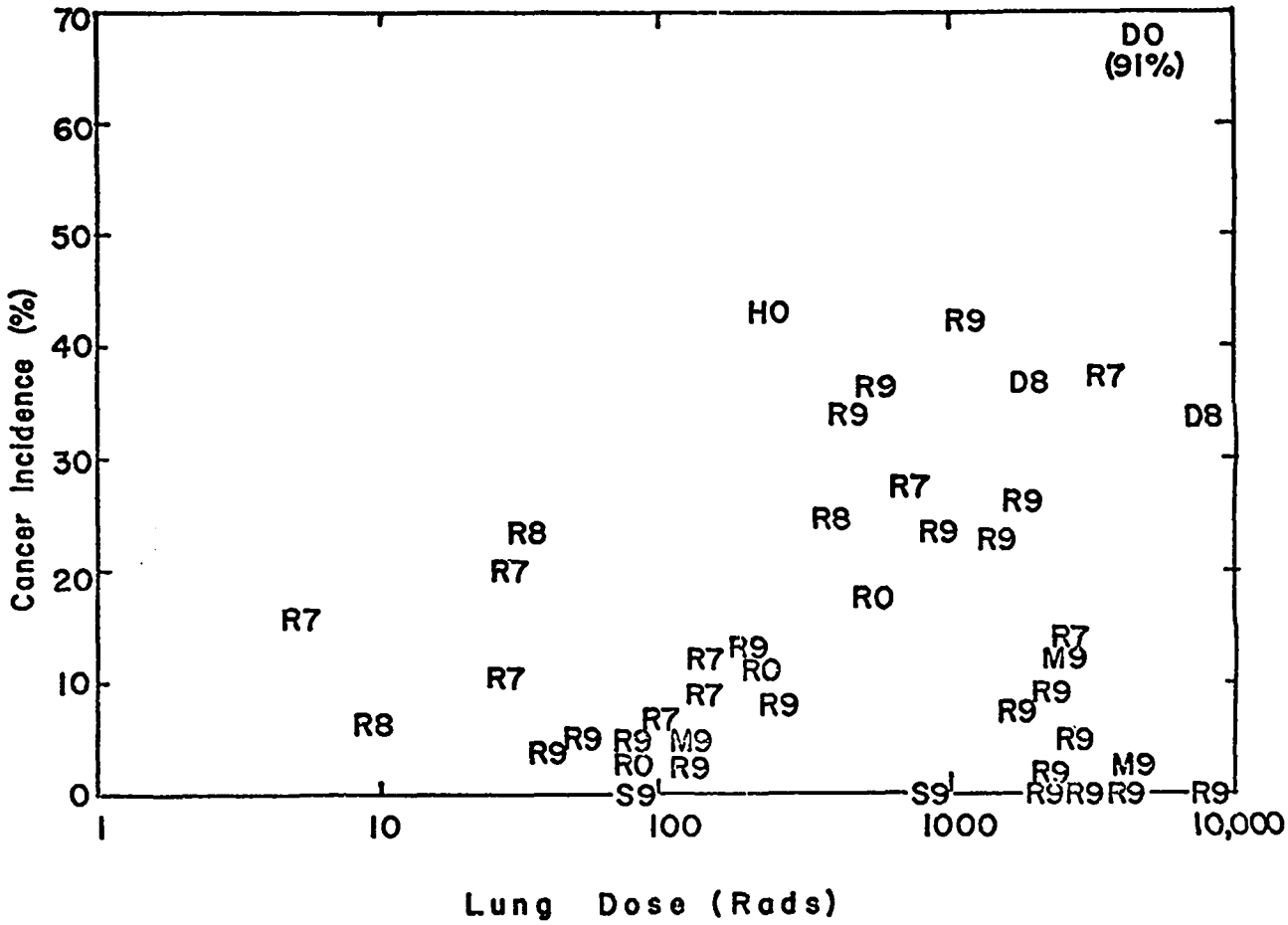

Fig. 1 A plot of the crude data for lung tumor incidence versus lung dose. Data have not been screened for length of exposure or accuracy of dose calculations. All doses are expressed as the average to the lung.
Legend: M-mice; R-rats; D-dogs; H-hamsters; S-humans.
0-^{210}Po; 7-^{237}Np; 8-^{238}Pu; 9-^{239}Pu.

as if the same amount of energy were distributed over the entire tissue. We have seen that this is not the case in some extreme situations such as in the overkill of cells close to a particle or the induction of more lethal effects at high dose rates. However, acceptance of the assumption of linearity at the more moderate conditions would lead to the conclusion that there should be no difference in outcome regardless of the distribution of the dose throughout the tissue, unless a critical portion of the organ is more sensitive. This would lead to the conclusion that nonuniform distribution of dose could have no greater effect than a uniform dose. Because this is based upon an assumption which is made in an effort to be conservative and is based upon effects at relatively low doses, we do not believe that this argument is very strong. However, a conclusion of nonlinearity of effect could have a major impact upon current radiations standard setting practices unless it is shown that such nonlinearity occurs only at very high cell doses.

No clear cut, overall picture of the relative effects of uniform versus focal dose can be drawn from the present data. It appears, from the $^{238}PuO_2$ microsphere data and the skin experiments with ^{90}Sr that, in the extreme situation of a single, very active particle, the focal radiation is considerably less damaging. Cember[15] concludes

that the focal source is less damaging for beta emitters than is the uniformly distributed source. The data of Grossman, et al.[27] for ^{210}Po on iron oxide particles indicates a seeming decrease in the tumor incidence as well as increased survival for the focal sources. Saunders,[35] as a result of his studies with soluble ^{238}Pu derived from crushed microspheres arrives at a conclusion that spreading the dose more uniformly results in an increased cancer incidence due to the greater number of epithelial cells involved. This conclusion was based on the observation of "- - a significant incidence of tumors in the lung and in other tissues at radiation doses that have not previously been shown to be carcinogenic in animals". In Figure 1, it is of interest to note that two of these data points are included in the five lowest dose points with the other points being the results of ^{237}Np administration. In both cases, significant numbers of tumors were also noted in locations other than the lung indicating a more general insult to the entire body.

Most of the support for particulates being more hazardous than a uniformly distributed material seems to arise from calculations based upon dose distribution around the particles and an assumed response of individual cells to this dose. In an overall appraisal of the information available, it does not appear that the majority of the data support the hypothesis that the particles are more hazardous than the uniform dose. A reasonable case can be made that they are less hazardous. The conclusion of this work to date, therefore, is that the preponderance of the evidence indicates that the use of an average lung dose is appropriate in limiting exposures and may well be conservative.

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