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An Examination of the Pathways from Soil to Man for Plutonium

by

J. W. Healy

For Reference

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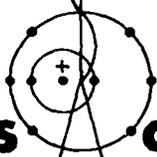
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ABSTRACT

The data available on resuspension and ingestion as pathways of plutonium from soil to man were reviewed and a recommended limit based upon a conservative interpretation of the National Council on Radiation Protection and Measurements (NCRP) recommendations for limiting values was derived. Wind resuspension appeared to be the least limiting value with mechanical resuspension and pica in children among the more important. Ingestion of foodstuffs could also be an important pathway if it is assumed that all food is produced in the contaminated area.

I. INTRODUCTION

The derivation of a limit for plutonium in soils is difficult because of the widely differing conditions in which it may be applied. Thus, when the vegetated areas of the East with relatively heavy rainfall are contrasted with the farm lands of the middle of the country or with the arid and semiarid regions of the West, the conditions are so different that it is difficult to find a common ground. If, then, the differing uses of the land, which range from pasture through agriculture or forests to the building of town and cities, are included, the situation becomes almost hopeless. It is for this reason that many people feel that one should not attempt to derive a specific guide for soils but should examine each case on an ad

hoc basis.   There is considerable merit to this viewpoint but
this means that the battle for acceptance and concensus must be
fought anew for each of these cases and it may be difficult to
explain to a public not used to this type of an approach the
details of why different values are acceptable at different
locations.

For these reasons, we have tried to derive a numerical value
for plutonium in soils which is believed to be safe at any loca-
tion.   It would be ideal to believe that this could be used as a
"decision level,"[1] or a level above which further investigation
and decision is needed taking into account the existing
conditions, thereby combining some of the advantages of a fixed
value with the advantages of an ad hoc determination.   It is ap-
parent to the author, however, that the forces of society are
such that this type of flexibility will not be accepted and we
must resign ourselves to the fact that any value accepted will
be used throughout.   However, we must also recognize that the
approach used here is not unique in its problems or the method
of handling although this may be an extreme example.   For exam-
ple, the present limits for radioactive materials in air or
water involve the reduction of a large number of variables to
single numbers that represent a "safe" limit.

This analysis was prepared for a particular purpose: to give
guidance as to levels on properties to be returned to the public
after potential contamination from nuclear work.   This limits
our consideration to areas where the contamination has been
present for some time since it is unlikely that an area would be
disposed of immediately following an accident that resulted in
contamination.   While many of the same considerations may be of
importance in a limit to be used following an airborne deposi-
tion, the primary factor of possible mobility of the fresh de-
posit will not be present here.

It is assumed that the property, once sold, is subject to no
further control so that it could be used for any purpose: homes,
farms, pasture, a factory, or any similar use.   It can be seen
that this eliminates the variable of use, since the limitation

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must be gauged to the use with highest probability of intake. It is possible that restrictions could be placed in the deed at the time of sale so that certain uses are forbidden. Such a practice may be the wisest choice when the damage to the environment is such as to destroy valuable resources. It must be remembered that cleaning of soil contamination requires removal of the contaminated soil and replacing with clean soil from another location. A less drastic measure is plowing but even this can cause damage in some cases. Since the soil is the basic support for all terrestrial ecosystems such "decontamination" can severely affect an area. Areas with endangered species or those which have fragile ecosystems that may not recover for centuries deserve special attention with the wisest course, possibly, to not decontaminate so that the consequent disruption does not occur. Such a decision must be made at the time by responsible officials.

Since the considerations in this report were primarily based upon the population being in close proximity to the contaminated area, the values derived here should not be considered as applicable to areas still under control of access. Such limitations should be based primarily on airborne transport (or in some areas water transport) to the surrounding population.

In accordance with good radiation protection practice and as recommended by the NCRP, the value given should be regarded as the maximum allowed with values reduced to as far below this value as is practicable before disposal. While this requires a value judgment on the part of both the disposer and the recipient, this principle, in general, can be defined as removal of contamination to a level where further reductions would be exorbitantly expensive or would result in unacceptable damage to the property or the environment.

The limitation is not intended to be translated into an "acceptable" value for the release of plutonium as based on land contamination. With present controls on the design and operation of plutonium-handling facilities, it is believed that land

contamination in the future will occur only in the event of an accident. In such cases, where the land is under the control of an operator, it is considered good practice to clean up any contamination to the extent practicable and, if possible, to below the limitation given, always with respect for the practical degree to which cleanup can be accomplished without completely destroying what one is trying to protect.

II. SOIL CHARACTERISTICS PERTINENT TO PLUTONIUM TRANSFER

The properties of plutonium in soils and the possibilities of movement are related to the characteristics of the soil as well as to the method whereby the plutonium was originally deposited. While considerable attention has been paid to the presence of oxide particles as a result of accidents or safety tests, it must be realized that other methods of soil contamination, such as adsorption from liquids, do occur. In the case of the oxide particles, they will have an initial particle size characteristic of the method of formation and deposition. They will undoubtedly be degraded in size with time by the normal method of soil formation. However, for the area contaminated with plutonium in solution, the absorption will be chiefly on the smaller particles in the soil, the clays. These particles are normally present in the soils as aggregates or fastened to the larger sand particles although the degree of aggregation depends upon the soil type, humus content, and moisture content.

Since the soil and its properties play an important part in the interactions with plutonium, we have reproduced in the remainder of Section II, a discussion on soils and wind erosion by J. Nyhan from a previous report.[2]

A. Wind Erosion in the U.S.

Although few regions in North America are entirely free from wind erosion, several regions exhibit high activity. Figure 1

shows that the erosion of soil by wind has been the worst in the Great Plains, as well as the Columbia River plains, and some parts of the Pacific Southwest, and the Colorado Basin. Other general regions of interest include the muck and sandy areas in the Great Lakes region and the sands of the Gulf and Atlantic seaboards.[3] It is useful to consider three factors influencing the mechanics of wind erosion: wind, soil, and the nature of the eroding surface.

Regions in the U.S. with relatively high average surface wind velocities (Fig. 2) correspond to areas of high soil erosion by wind. This is not surprising since the intensity of wind erosion apparently varies as the cube of wind velocity.[4] The wind factor is centered around the wind velocity pattern near the soil surface, its effect on the initiation of soil movement, and the transportation and deposition of soil particles.

The soil factor involves the influence of soil moisture and soil structure on the severity of wind erosion. Periods of drought, wind erosion, and dust storms have occurred simultaneously in the Great Plains, and the intensity of wind erosion varies inversely as the square of either moisture at the soil
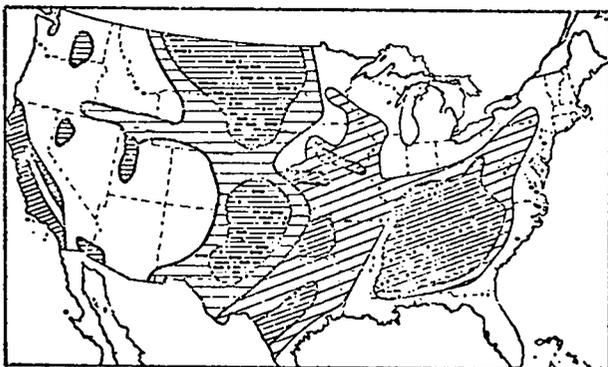

Fig. 1. A generalized map (101) showing the chief large areas of high soil erosion by wind (horizontal shading) or runoff (slanting shading).
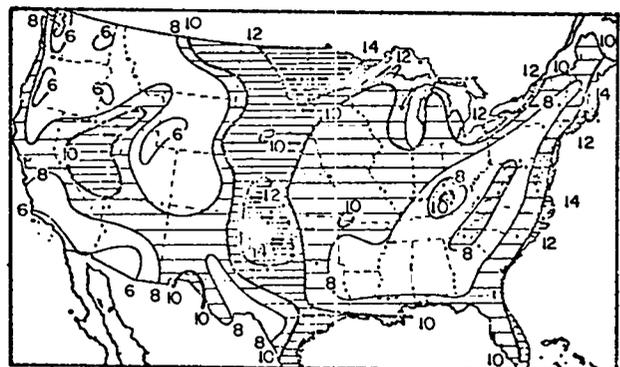
Fig. 2. Generalized map (101) of average surface wind velocity (mph).

surface or effective precipitation.[4] Again, a good correlation exists between geographic regions of intense wind erosion (Fig. 1) and areas where evaporation is usually in excess of precipitation (Fig. 3). These observations are related to the fact that there are very few winds that have sufficient velocities to overcome the very large cohesive forces of moisture at a tension of 15 atmospheres in an air-dried soil.[5,6] Soil structure, or the arrangement of individual sand, silt, and clay particles with respect to each other, also influences soil resuspension. Soil structure is in turn affected by the particle size distribution of the soil, as well as the organic matter and polyvalent cation content of the soil components important in binding the particles together. Although enormous forces are necessary to break the covalent bonds involved in these soil-cation-organic molecule complexes, winds can move groups of these aggregates in the form of clods less than 840 μm in diameter.[7] The stability of these structural units is decreased by tillage operations and by freeze-thaw and wet-ting-drying cycles at the soil surface.

The nature of the eroding surface is another factor which influences the mechanics of wind erosion. Surface conditions which affect wind erosion are the surface roughness and the de-gree of protection by surface cover. The erosion decreases with increasing surface roughness, when this roughness is due to non-erodible structural units, mainly because of the diminished wind velocity at the soil surface. Vegetation not only adds to sur-face roughness but also provides cover for the soil surface. Regions of the U.S.A. where the vegetation is minimal (grassland or desert) also exhibit a potential for significant wind erosion (Fig. 1) compared with the minimal wind erosion in prairie-for-est or forest zones (Fig. 4). Vegetation effectiveness as a wind barrier depends primarily upon its density, width in the wind direction, porosity, and orientation with respect to the prevailing winds.[7]
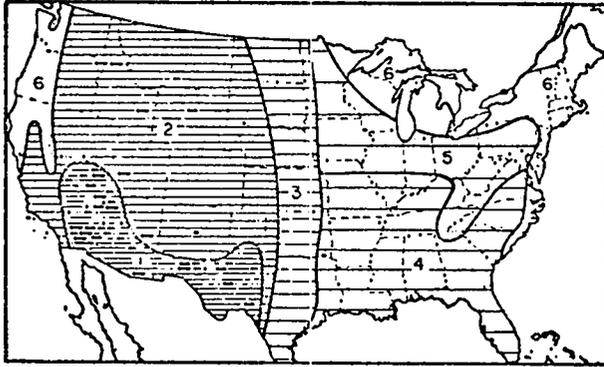
Fig. 3. Evaporation regions:
1. evaporation much in excess of precipitation
2. evaporation considerably in excess of precipitation
3. evaporation usually in excess of precipitation
4. evaporation in excess of precipitation in warm months
5. precipitation in excess of evaporation in cooler months
6. precipitation generally in excess of evaporation (101).
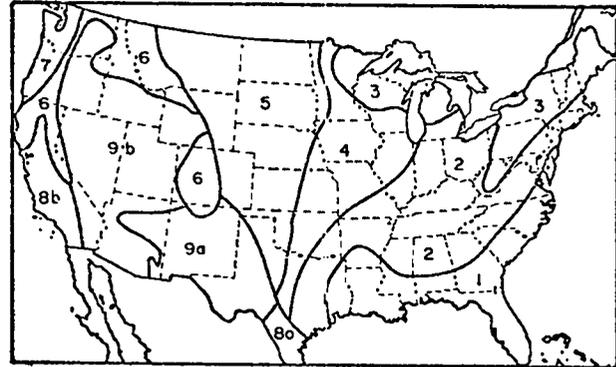
Fig. 4. Generalized vegetation map (101).
1. southeastern forest
2. deciduous forest
3. northern evergreen forest
4. prairie-forest transition
5. grassland
6. western conifer forest
7. northwestern conifer forest
8a. desert shrubs
8b. semidesert shrubs
9a. semidesert
9b. desert

B. Variations in Soil Properties Which Influence Erosion

In order to understand the natural variability of soil properties affecting wind erosion, an understanding of some factors in soil formation is helpful. The five factors of soil formation that influence, have influenced, or may influence soil genesis are as follows: parent material, organisms, climate, relief (topography), and time (Fig. 5). When considering soil variations throughout an area as large as the Great Plains, it is useful to relate the scale of action of these five factors to soil variability within this large region. Climate and organisms tend to be relatively uniform across distances of tens or hundreds of miles (kilometers), whereas parent materials are fairly uniform over miles of distance. Almost all of the short-distance changes in soil type are related to the topographic factor.

The five soil formation factors are responsible for general patterns of soils occupying positions on contemporary landscapes within the wind-eroded areas in Fig. 1. Transects across the
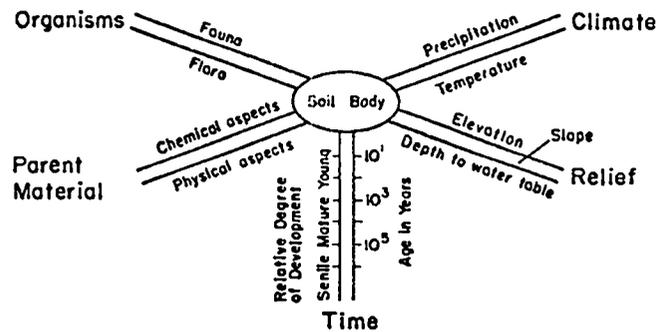
Fig. 5. The five general factors of soil formation.

northern and southern Great Plains exhibit large areas of entisols* and aridisols** in the western regions, grading into vast areas of mollisols*** to the east. These soils are also the major soils in the areas of Washington, Nevada, California, and Arizona where high wind erosion activity exists. Soils belonging to the Entisol order of the U.S. Comprehensive Soil Classification System[8-10] represent the simpler end of the soil continuum relative to soil development and usually occur in mountainous, desert, and alluvial floodplain regions. These soils are morphologically similar to their parent materials, partially due to the immobilization of clay illuviation by inert materials, highly siliceous sediments, and carbonates. The entisols are similar to soils of the Aridisol order in that they both contain light-colored topsoils with less than 1% organic matter. However, the aridisols, the soils of the arid regions, are more weathered than the entisols and exhibit minimal clay illuviation, maximum carbonate accumulation, and many times a surface "pavement" of gravel. The mollisols, the darker colored grassland soils of the prairies, contain greater than 1% organic matter in the top 7 in. (18 cm) of their profiles. Several

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*Newly formed soils.
**Soils of arid regions.
***Grassland soils of steppes and prairies.

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pedogenetic processes are involved in mollisol profile development that are minimized in entisol-aridisol profiles: (1) the process of darkening of soil by organic matter additions, (2) accelerated eluviation and illuviation of clays, and (3) loess additions to the topsoils of some mollisols.

Jenny[11] has advocated a scheme for comparing soils or individual soil properties. This scheme is based on the principle of comparing a given soil property among soils when one soil formation factor is dominant and the other four are ineffective. Jenny took soil samples along an 11° C isotherm in the arid-humid transition belt extending from Colorado through Kansas to western Missouri, and correlated natural variations in soil properties with annual rainfall.[12] The results (Fig. 6) indicated that the depth of the carbonate horizon and hydrogen-ion concentration depend most sensitively on rainfall, with nitrogen content (related to organic matter content) and clay concentrations less influential. Since these soil properties influence soil structure directly, wind erodibility will be inversely related to this climatic factor.

The particle size distribution of a soil is an important factor contributing to soil aggregate formation. The U.S.D.A. has developed a system of classifying soils relative to their texture, i.e., the relative proportions of sand (50- to 2000-μm-diam particles), silt (2- to 50-μm-diam particles) and clay (particles with diameters less than 2 μm). These 12 classes are usually presented as a soil-texture triangle (Fig. 7). These 12 classes can be further grouped into coarse-, medium-, and fine-textured soils as in Table I.

Natural variations in particle size distributions of surface soils [i.e., top inch (2.5 cm) of soil] have not received much attention, mainly because the top 15-30 cm of soil is usually the important unit for agricultural and soil classification reasons. However, it is known that climatic factors have a large influence on the particle size distribution of surface
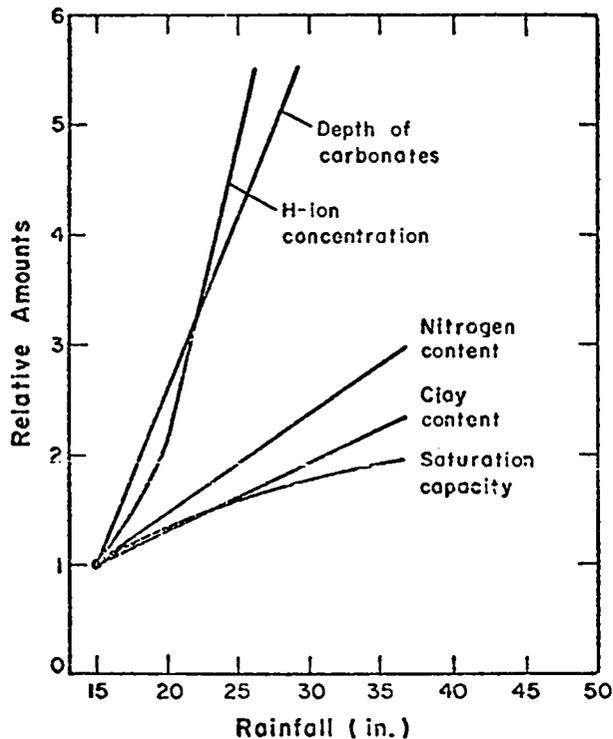
Fig. 6. Relative soil property–rainfall curves for some soils in the Great Plains (69).
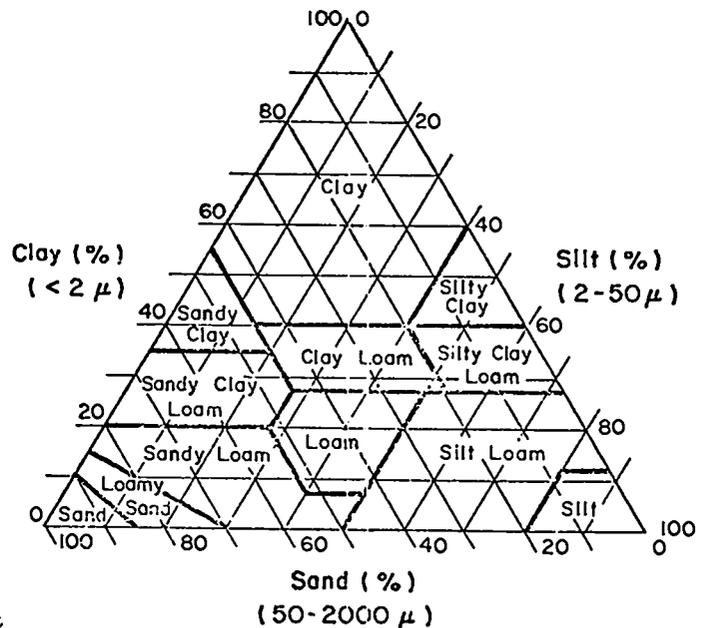
Fig. 7. The soil texture triangle (USDA).

soils, a factor involved in aggregate formation. With increasing aridity, physical processes assume relatively greater significance than chemical forces in the disintegration of rocks. Thus, aridisols bear a closer relation to their parent material than mollisols. However, reduction in size of sand and silt particles is performed largely by physical weathering via differential thermal expansion of minerals, frost action, and by erosion and deposition by water, ice, and wind. In contrast, clay particles have relatively large surface areas that are able to react relatively fast via chemical reactions, all of which require the presence of water.

In order to observe the net result of physical and chemical weathering processes on parent materials, the particle size distributions of selected textural types of soils is presented in Figs. 8-13 and Table II. This compilation of results for over

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TABLE I

SOIL-TEXTURE CLASSIFICATION

Coarse-textured	1. Sands and loamy sands	Sand Loamy sand
	2. Sandy loams	Sandy loam Fine sandy loam
Medium-textured soils	3. Loamy soils	Very fine sandy loam Loam Silt loam Silt
	4. Moderately heavy soils	Sandy clay loam Clay loam Silty clay loam
Fine-textured soils	5. Clays and silty clays	Sandy clay Silty clay Clay

40 soil types[13-27] is presented as graphs of percentages of particles (by weight) smaller than a specified particle size. Whenever the data are available, the mechanical composition of "typical soils," as given by one author[28] is also plotted for comparison (solid lines).

Figures 8-13 indicate that very few soils have appreciable amounts of sand particles greater than 500 μm in diameter. This seems to substantiate the observation that many soil materials, notably those in glaciated areas where much of agricultural science developed, tend to have a minimum in particle size distribution around 1000 μm.[29] At the other end of the spectrum, many soils may consist of over 30% by weight of particles smaller than 0.2 μm in diameter; this represents a documented, 5
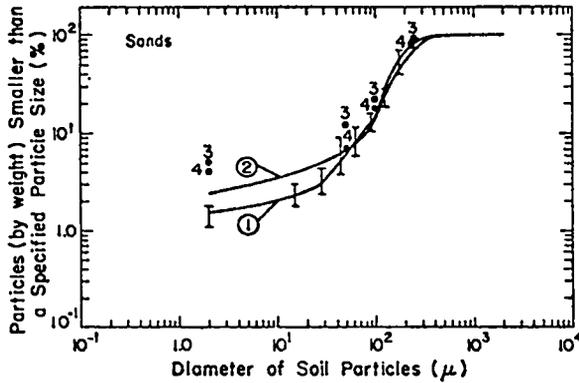
Fig. 8. Summation percent curves for sands (numbers in figure refer to soil sample numbers in Table II.
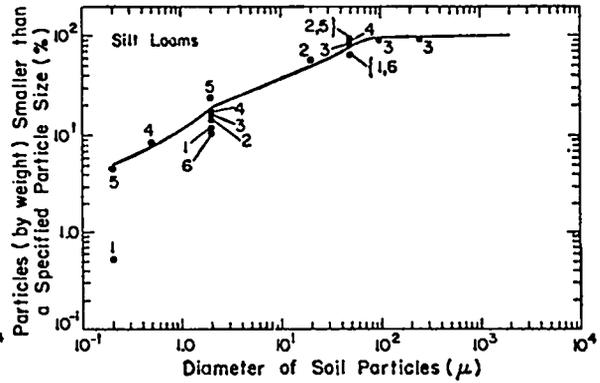
Fig. 11. Summation percent curves for silt loams (numbers in figure refer to soil sample numbers in Table II.
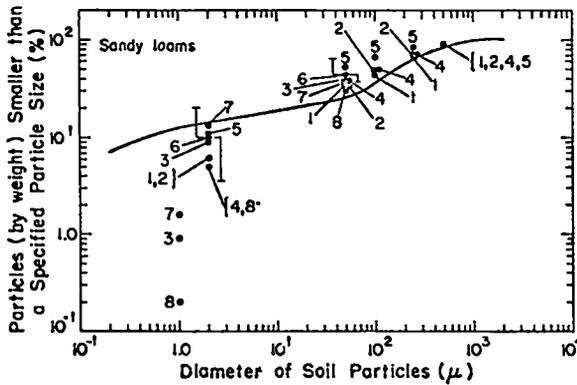
Fig. 9. Summation percent curves for sandy loams (numbers in figure refer to soil sample numbers in Table II.
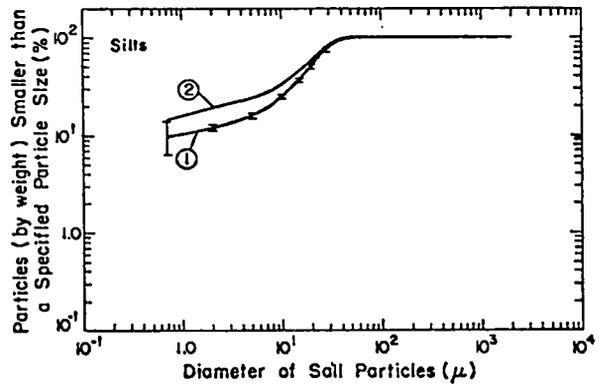
Fig. 12. Summation percent curves for silts (number in figure refer to soil sample numbers in Table II.
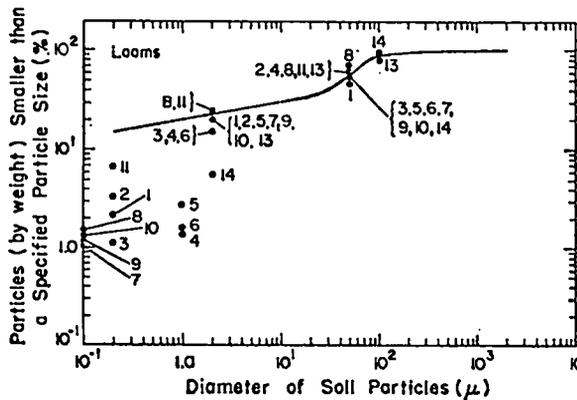
Fig. 10. Summation percent curves for loams (numbers in figure refer to soil sample numbers in Table II.
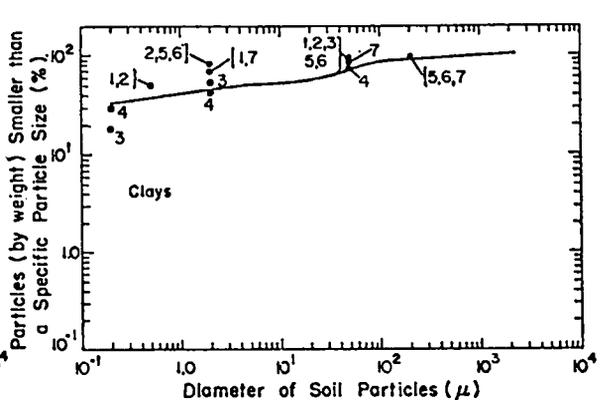
Fig. 13. Summation percent curves for clays (numbers in figure refer to soil sample numbers in Table II.

TABLE II

IDENTIFICATION OF SOILS IN FIGURES 8-13 BY TEXTURAL CLASSES

Textural Class	Sample Number In Figure	Soil Type	Sampling Depth (inches)	Location	Comments	Reference
			A. Coarse-textured soils			
Sand	1	Average of 5 soil samples from the Bloomfield and Princeton soil series	Varied from 40-200	Indiana	Samples from C horizons of Wabash River transect	13
Sand	2	Average of 7 soil samples from the Bloomfield and Princeton soil series	Varied from 52-136	Indiana	Samples from C horizons of a White River transect	13
Sand	3	Not classified	0-2	Northern Alaska	Samples taken from a tundra soil adjacent to the Meade River	14
Sand	4	Not classified	0-5	Northern Alaska	Samples taken in sand dunes adjacent to the Meade River	14
			B. Medium-textured soils			
Sandy loam	1	Scituate sandy loam.	0-9	New Hampshire	Sample taken at crest of a drumlin	15
Sandy loam	2	Scituate fine sandy loam	0-7	New Hampshire	Sample taken in middle slope region of drumlin	15
Sandy loam	3	Greenfield fine sandy loam	0-10	California		16
Sandy loam	4	Timpahute sandy loam	0.2	Utah	Samples from arid region	17
Sandy loam	5	Escalante fine sandy loam	1-12	Utah	Samples from arid region	17
Sandy loam	6	Average of 7 Ptarmigan sandy loams	0-8	Montana	Located on steep-gentle mountain slopes	18
Sandy loam	7	Musick sandy loam	0-6	California	Podzolic soil	19
Sandy loam	8	Caspar sandy loam	0-6	California	Podzolic soil	19
Loam	1	Unclassified but in the Oxbow soil association area	0-7	Saskatchewan	Orthic Black Soil	20
Loam	2	"	0-7	Saskatchewan	Podzolic Orthic Dark Grey Soil	20
Loam	3	Unclassified, but in Whitewood Association	0-4	Saskatchewan	Podzolic Dark Grey Wooded soil	21
Loam	4	Tehama loam	0-8	California	-	16
Loam	5	Kimball loam	0-4	California	-	16
Loam	6	San Joaquin loam	0-6	California	-	16

TABLE II (cont)

IDENTIFICATION OF SOILS IN FIGURES 8-13 BY TEXTURAL CLASSES

Textural Class	Sample Number In Figure	Soil Type	Sampling Depth (inches)	Location	Comments	Reference
Loam	7	Barnes loam	0-6	North Dakota	Publication soil # S53ND-2-1 from central North Dakota	21
Loam	8	Barnes loam	0-2	North Dakota	Publication soil # S53ND-11-1 from southeastern North Dakota	21
Loam	9	Barnes loam	0-5	North Dakota	Publication soil #S54ND-10-2 from northeastern North Dakota	21
Loam	10	Aastad loam	0-6	North Dakota	Grassland soil from central-eastern North Dakota	21
Loam	11	Nappanee loam	0-8	Ohio	lake plain sample	22
Loam	12	Nappanee loam	0-8	Michigan	glacial till plain sample	22
Loam	13	Redfield loam	0-6	Utah	alluvial soil in an arid region	17
Loam	14	Uvada loam	0-1	Utah	-	17
Loam	15	Melbourne loam	0-7	California	lateritic soil	19
Silt loam	1	Not classified	0-1	Saskatchewan	Orthic Grey Wooded soil	20
Silt loam	2	Hosmer silt loam	0-8	Illinois	loess-derived podzolic soil	23
Silt loam	3	Antelope Springs silt loam	0-3	Utah	-	17
Silt loam	4	Coropina silt loam	0-6	British Guiana	podzolic coastal soil	24
Silt loam	5	Fairbridge silt loam	1-3	British Columbia	-	25
Silt loam	6	Hopleys silt loam	0-6	Montana	alpine soil	18
Silt	1	Average of 5 soil samples from the Alford, Ragsdale and Sylvan series	varied from 40-140	Indiana	Samples from C horizons of a Wabash River transect	13
Silt	2	Average of 4 soil samples from the Alford, Ragsdale, and Hosmer series	varied from 52-76	Indiana	Samples from C horizons of a White River transect	13

C. Fine-textured soils

Textural Class	Sample Number In Figure	Soil Type	Sampling Depth (inches)	Location	Comments	Reference
Clay	1	Frontland clay	0-6	British Guiana	Coastal soil, unleached phase	24
Clay	2	Frontland clay	0-6	British Guiana	leached and cultivated phase	24
Clay	3	Alberni clay	0-2	British Columbia	-	25
Clay	4	Average of 3 Thoeny clays	Unspecified	Montana	Samples from B horizons	26
Clay	5	Princes town clay	0-6	West Indies	-	27
Clay	6	Carnon Hall clay	0-6	West Indies	-	27
Clay	7	Fitches clay	0-6	West Indies	-	27

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orders of magnitude difference in soil particle sizes. The results of summation curves for 560 German soils[30] indicated that: (1) particle size distributions were normally distributed when the log of the particle size was plotted on the abscissa, (2) every particle size range has one maximum size distribution, and (3) certain distributions did not occur naturally. Examples of soils that were not found were soils with less than 20% silt and greater than 30% clay and soils with less than 65% sand, less than 5% clay and more than 30% silt.

C. Classification of Soils Into Wind Erodibility Groups

Eolian materials were described in a recent review[31] by their pathway of transportation in the atmosphere, persistence in the atmosphere, and particle size (Table III). More specifically, we are interested here in the air concentrations from the soil surface to about 2 m above the soil surface. Chepil[32-34]

TABLE III

CLASSIFICATION OF EOLIAN MATERIALS

Type of Material	Dominant Particle Size (μm)	Transport		Persistence in Atmosphere
		Pathway	Range (km)	
Dune sand	400-200	At or near ground level	10^{-3} to 10^{0}	Highly transient phase
Loess	50-10	Lower troposphere	10^{-1} to 10^{2}	Transient phase
Aerosolic dust	10-1	Middle and upper troposphere	10^{4} to 10^{6}	Residence time depends on precipitation frequency but is thought to be days to weeks
Stratospheric dust	<1	Stratosphere	$\sim 10^{9}$	Residence time, years
Extraterrestrial dust	40-0.01	Interplanetary space	$\sim 10^{13}$	Undetermined

has documented the dynamics of wind movement of arable soils in this zone via surface creep, saltation, and suspension, involving progressively smaller soil particles.

After the violent wind erosion in the Great Plains in the 1930s and 1950s, criteria were needed to determine the capability classification of soils in these areas--criteria based on more than land use experience. In 1957 Coover and Moldenhauer[35] used a formula, which expressed soil loss rates as a function of a soil "erodibility index," plant surface residue, and surface roughness, to quantitatively evaluate the wind erosion in capability classifications for Texas soils. In 1960 Chepil[36] developed the more generalized relationship between actual soil loss rates and his soil erodibility index, based on many wind tunnel and field measurements, for Garden City, Kansas. Chepil later developed a wind erosion climatic index for application of the Kansas experiments to many geographical regions.[4] The final accomplishment came in 1963 when Chepil and Woodruff[37] published the wind erosion equation, which expressed the many factors involved in soil losses from erosive wind effects:

$$E = f(I \ C \ K \ L \ V) .$$

This equation expresses the annual soil loss in tons per acre (E) in terms of the soil erodibility index (I), the climatic index (C), soil surface roughness (K), the unsheltered field width (L), and the equivalent quantity of vegetative cover (V).

The Soil Conservation Service (S.C.S.) has recently developed criteria for grouping soils into "wind erodibility groups" as described in Tables IV and V.[38] The procedure is to collect about 2 kg of dry soil from the surface inch of the field, sieve the soil through an 0.84-mm square-opening sieve in the field, and relate the percentage of nonerodible soil aggregates greater than 0.84 mm to the wind erodibility group and soil erodibility index (the I factor in the wind erosion equation). When large areas of land must be classified and sieving is not a feasible

TABLE IV

RELATIONSHIP OF DRY SOIL AGGREGATES >0.84 mm
TO SOIL ERODIBILITY INDEX (I) AND WIND ERODIBILITY GROUP

Wind Erodibility Group (WEG)	Dry Soil Aggregates > 0.84 mm-percent	Soil Erodibility Index tons/acre/year
1	0.5-1.5	340-280
2	5.0-15.0	180-117
3	16.0-34.0	113-67
4	15.0-35.0	117-65
4L	15.0-35.0	117-65
5	28.0-52.0	79-33
6	36.0-54.0	63-29
7	36.0-64.0	63-17
8	---	---

procedure, the wind erodibility group can be roughly estimated by the texture by feel method. Once the wind erodibility group has been estimated, site-specific information on the other four factors of the wind erosion equation can be collected to estimate the potential amount of wind erosion for a given field under local climatic conditions.

This work has many direct applications to the problems of concern, especially because the S.C.S. has already collected these data for many soils. The total amount of soil loss per year, as predicted by the wind erosion equation, does consist of about 90% soil loss via saltation (and minimal surface creep) and about 10% soil loss in suspension - on the average.[39] Thus, many large soil particles would move via saltation close to the soil surface (90% below a height of one foot),[3] and would initiate suspension of smaller soil particles (silt, clay, and aggregates) in the respirable size range. This represents no small amount of soil - estimates of up to 277 million tons per year were found in suspension over the Great Plains in the 1950s.[39]

TABLE V

GUIDE FOR ESTIMATING WIND ERODIBILITY GROUP
FROM APPROXIMATE SOIL TEXTURES

Texture of Surface Inch	Wind Erodibility Group
Very fine sand, fine sand,and medium sand	1
Loamy sand, loamy fine sand	2
Very fine sandy loam, fine sandy loam, sandy loam	3
Clay, silty clay, noncalcareous clay loam and silty clay loam with more than 35% clay content	4
Calcareous loam and silt loam; calcareous clay loam and silty clay loam with less than 35% clay content	4L
Noncalcareous loam and silt loam with less than 20% clay content; sandy clay loam, sandy clay	5
Noncalcareous loam and silt loam with more than 20% clay content; noncalcareous clay loam with less than 35% clay content	6
Silt; noncalcareous silty clay loam with less than 35% clay content	7
Very wet or stony; not subject to wind erosion	8

III. MECHANISMS OF EXPOSURE

The radiations emitted by plutonium are nonpenetrating so that any potential health hazard from plutonium in the soils must result from transfer from the soils into the body. Material transferred to the surface of the body is also of little importance except that this may serve as an intermediate step in the intake to the body. In a previous report,[40] we considered the following mechanisms of transfer.

1. Absorption through the skin or wounds. The absorption factors derived were based primarily on the plutonium being applied as a solution rather than in the form of insoluble

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oxide or soluble plutonium absorbed on soil particles. Even then, it was estimated that the amount of plutonium-bearing soil on the skin could be as high as 1300 dis/min/g (590 pCi/g) continually carried throughout life. This is believed to be a very conservative estimate, perhaps by several orders of magnitude when the form of the plutonium on the soils and occupancy factors for the contaminated area are considered. Since there have been no additional data on skin absorption obtained in the interim, we will not consider this mechanism further in this report.

2. <u>Inhalation as a result of general resuspension</u>. General resuspension is considered to be that due to either winds or mechanical disturbance that results in relatively widespread air contamination with the meteorological dispersion characteristics of the area an important factor.

3. <u>Inhalation as a result of· local resuspension</u>. Local resuspension is considered to be that due to mechanical disturbance that produces a high local concentration in the breathing zone of the individual. It also includes the production of local resuspension following transfer to another medium, for example, the production of high concentrations during the removal of clothing that was contaminated in the area. The latter mechanism could, perhaps, be better labeled as a "transfer-local resuspension" but we will continue to include it in the local resuspension category.

4. <u>Ingestion as a result of eating plant material raised in the contaminated area or animal products from the contaminated area</u>. In general, this mechanism was earlier judged to be relatively minor, but it will be reviewed again in light of the more recent data.

5. <u>Casual ingestion of soil by individuals working in the area or as a result of dust derived from the area</u>.

6. Deliberate ingestion of soil by children living on the
   contaminated area.

In general, the pathways explored in this report are similar
to those in the previous report. Little additional data are a-
vailable on the local resuspension problem but there have been a
relatively large number of experiments on wind resuspension
which have led to increased understanding of the problem
although there still remains a paucity of data with which to
really provide a quantitative estimate for areas different from
those investigated.

IV. INTAKE LIMITS

In recent years, particularly since the issuance of the BEIR
report,[41] there has been a trend toward the derivation of
estimated numbers of health effects under assumed conditions to
arrive at the consequences of a given action, such as establish-
ing a new program or a new limit. The uncertainties of such a
procedure in converting an estimated dose to numbers of cancers
or genetic defects are well known and were emphasized in the
body of the BEIR report. In fact, the NCRP in a review of the
current state of radiation protection policy stated:[42] "The
NCRP wishes to caution governmental policy-making agencies of
the unreasonableness of interpreting or assuming upper limit
estimates of carcinogenic risks at low radiation levels, derived
by linear extrapolation from data obtained at high doses and
dose rates, as actual risks, and of basing unduly restrictive
policies on such an interpretation or assumption. The NCRP has
always endeavored to insure public awareness of the hazards of
ionizing radiation, but it has been equally determined to insure
that such hazards are not greatly overestimated. Undue concern,
as well as carelessness with regard to radiation hazards, is
considered detrimental to the public interest."

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In the present study, the uncertainties in estimating the intake and resulting doses far outweigh the uncertainties of even the calculations referred to above. Thus, measurements from which resuspension rates can be derived are relatively few and only in selected areas. The behavior patterns of children with respect to playing in the soil are poorly defined and even the latest data on foodstuff contamination tends to be indicating higher transport than was previously believed.

We could have proceeded by producing a mathematical "model" including known methods of intake and metabolism and used best guesses as to the transfers. However, this would also involve choosing one or two model locations so that the conditions could be related to the location. Our experience indicates that such an approach tends to mask uncertainties and produces numerical values that are commonly regarded as having validity far beyond their true uncertainty. For this reason we have analyzed each of the potential pathways of exposure separately, attempting in each case to carefully delineate uncertainties and to explain assumptions but, at the same time, using conservative assumptions so that it is extremely probable that accumulations in the body, even in extreme conditions, will not reach the limit chosen.

The NCRP has recommended that for individual members of the public not occupationally exposed that "the dose limit for the critical organs (whole body) of the individual...shall be 0.5 rems in any one year, in addition to natural radiation and medical and dental exposures."[43] This limit is based on possible somatic effects. For genetic considerations they recommend that: "The dose equivalent to the gonads 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...per person." (emphasis added) In considering this limit they point out that: "With an intended limit of 0.5 rem per year for most, if not all situations, the average incremental dose to population groups

from radiation plants and installations would probably easily stay below 0.1 rems per year, comfortably within the limit that consideration of genetic effects sets for the whole population." We will, therefore, in general, base our considerations on a dose to the critical organs of the maximum individual of 0.5 rems/year. This, of course, is lower by a factor of 3 than is recommended by the ICRP[44] for most critical organs of an individual but does correspond to their limit for the average of a population group.

The basic strategy in the examination of the individual pathways was to estimate the dose to a maximum individual in any environment. Thus, the mechanical resuspension was considered for a farmer tilling his fields and this pathway may be of much lower importance in other situations. Some other pathway may be of greater importance in other environments. The dose limits from the NCRP are to cover all sources of man-made radiation aside from medical. This, of course, can change from year to year and from place to place depending upon fallout conditions as well as the use of devices producing x rays. The degree of conservatism in the derivation of the plutonium limits as well as the focus on the maximum individual, leads to the conclusion that any allowance for other exposure is not necessary unless such exposure reaches a major fraction of the 500 mrem per year.

The primary sources of intake and subsequent exposure are ingestion and inhalation. Absorption through the skin was considered in the earlier report[40] and appeared to be of little consequence even with assumed absorption rates higher than appear reasonable. Since no new information has been obtained, this potential pathway will be ignored in this study.

Following ingestion the ICRP[45] summarizes the absorption as follows: "...following ingestion of relatively insoluble material, such as PuO_2, the absorbed fraction may be taken to be about 10^{-4}%. For more soluble compounds (e.g., plutonium nitrate) the absorption is also low, due to hydrolysis; the absorbed fraction may be assumed to be about 3×10^{-3}%. This fraction may be increased substantially if the ingested material is administered

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to immature animals or is in a strongly acid solution. Absorption of chemically stable, soluble complexes (e.g., Pu-DTPA chelates) from the gastrointestinal tract may be as high as 2% of the amount administered." However, in reference to the high absorption with complexed plutonium they point out that if the complex is highly stable and the complexing liquid is of high stability, as is DTPA, disassociation of the complex in the body will be minimal and excretion of the absorbed complex will be rapid and over 99% will be excreted in 2 days. Weaker complexes, such as the citrate, however, will be metabolized and the retained fraction may be considerably higher than following a soluble salt, such as the nitrate, by perhaps one or two orders of magnitude. It must be remembered, however, that these studies on complexing agents are done with plutonium already complexed and usually with an excess of complexing agents so that behavior in environmental situations may be different.

Both the ICRP[46] and the NCRP[47] in derivation of drinking water MPCs have used an absorption from the G.I. tract typical of a soluble salt, such as the nitrate, of 0.003%. While this is appropriate for water, there is some question whether this value should be used for foods. Adequate studies have not been done on the absorption of plutonium which has been metabolized in plants or animals, and in prepared foods there are many additives (although in small quantities) which could affect the uptake from the G.I. tract. For these reasons, we have chosen an uptake value ten times that used in setting MPCs for drinking water, or 0.03%, for use in ingestion of foodstuffs. However, for ingestion of soils we have used the value for the soluble salt or 0.003%. If the plutonium in the soil is in the form of plutonium dioxide, as would occur from most atmospheric dispersals, this value is undoubtedly high, perhaps by one or two orders of magnitude depending upon particle size. If the plutonium is absorbed on the clay fraction and if desorption in the G.I. tract occurs it may behave as the soluble salt. No allowance was made over and above these considerations for increased

absorption by children because the age period during which this is important is small and any additional accumulation during this period will be within the range of uncertainty when the full lifetime accumulation is considered.

In assessing the permissible ingestion rate we consider the critical organs of concern as the bone and the liver. The ICRP, for inhalation, recommends that the quantity of plutonium in these two organs be partitioned equally since, in the extreme case, this will not result in an error of more than a factor of 2 to either organ.[45] In the ingestion case it appears likely that the quantity going to the bone will be greater than that to the liver. However, we will consider the ICRP recommendation on inhalation to apply for ingestion since this is well within the other uncertainties in the study. Thus, of that absorbed to the bloodstream, it is assumed that 45% deposits in bone, 45% in liver, and the remaining 10% is distributed to the remainder of the body. Again following the ICRP,[45] the half-life in the skeleton is taken as 100 years and in the liver as 40 years. The permissible intake was based upon accumulating a quantity in the body which would result in 0.5 rems per year to the critical organ at the end of a 70-year exposure period. For a liver weight of 1700 g and a G.I. tract absorption of 0.03%, this per-mits an ingestion of 440 pCi/day to result in a final liver con-tent of 880 pCi after 70 years. For an absorption of 0.003%, the intake would be 4400 pCi/day. For a 7000-g skeleton, the amount to give 0.5 rems per year is 730 pCi and the correspond-ing intakes for 70 years would be 270 pCi/day for 0.03% absorp-tion or 2700 for 0.003% absorption when the "dose distribution factor" of 5 is included. It is noted that the bone limit is different in origin from the liver limit in that it was derived by biological comparison with radium and these data were used to derive the dose distribution factor.[48] The NCRP[43] did not address bone as a critical organ except for a footnote "For most alpha particle emitters deposited in bone, the bone itself may be the overriding critical organ with respect to bone cancer."

The detailed study of the question of the application to bone was deferred to a study of internal emitters.

Inhalation is normally considered the most probable method of intake of a relatively insoluble material which is poorly absorbed through the G.I. tract. The ICRP Task Group on Lung Dynamics[49] has provided a detailed review of information on deposition of particulates in the lung and the subsequent elimination and transfer to other organs. A Task Group of Committee 2 of the ICRP has proposed revisions to the behavior of plutonium following deposition in the lung.[45] To estimate the intake which would produce 0.5 rems per year at the end of a continuous exposure for 70 years, we have calculated the dose to the lung, skeleton, and liver for various particle sizes of both the Class Y (retained in the lung for long times) and Class W (retained in the lung for intermediate times) plutonium. The more soluble material represented by Class D is not believed to be representative of plutonium compounds. The lymph nodes were not included since the evidence with animals indicates that they are insensitive to the long term effect of cancer production and dose limits have not been derived for these organs.

The deposition in the three parts of the respiratory tract considered were read from the curves of the ICRP Task Group[49] and are given in Table VI.

The relatively high deposition in the pulmonary region (3-9%) for particles above 10 μm AMAD is presumable due to the presence of small particles in the distribution although the Task Group does not discuss this point. It may be noted that the values for the larger particle sizes total more than 100% deposition. These were the values estimated from the curves in the Task Group report and were retained rather than attempting adjustment. For the smaller particle sizes the difference between the deposition and 100% represents the fraction exhaled without depositing.

While we have used the model by the ICRP[45] for movement into the bloodstream, it should be noted that there is some question

TABLE VI

DEPOSITION IN RESPIRATORY TRACT

Particle Size AMAD (μm)	Deposition		
	Nasopharnyx (%)	Tracheobronchial (%)	Pulmonary (%)
0.1	1	8	50
0.2	4	8	40
0.5	15	8	30
1	30	8	25
2	50	8	20
5	75	8	12
10	83	8	9
20	87	8	5
50	99.5	8	3

about the values of transfer from the upper respiratory tract to the blood. These were included in the model to account for the relatively rapid initial transfer to the body noted in many experiments. However, the absorption appears to be very high as compared with, for example, the absorption from the G.I. tract. This, as will be seen, produces high quantities absorbed to the body from large particles deposited in the upper respiratory tract. If this initial high transfer were due to a small fraction of higher solubility material deposited in the deep lung, then the absorption from the upper tract would be an artifact and not representative of actual absorption. We have, however, used the model as given rather than attempting a revision. Table VII gives the results of these calculations in terms of the quantity that could be inhaled each day to limit the final dose rate after continuous exposure for 70 years to 0.5 rem per year.

The relatively small change with particle size for the material transferred to the skeleton and liver reflects the deposition and absorption to the blood from the upper respiratory tract. This model would say that the particle size is relatively

TABLE VII

QUANTITY OF PLUTONIUM INHALED PER DAY

TO RESULT IN 0.5 REMS/YR AFTER 70 YEARS (pCi)

Particle Size–AMAD (μm)	Class Y			Class W		
	Skeleton	Liver (pCi/day)	Lung	Skeleton	Liver (pCi/day)	Lung
0.1	0.9	1.5	2.4	0.57	0.94	24
0.2	1.1	1.8	3.0	0.65	1.1	30
0.5	1.5	2.4	4.0	0.70	1.2	40
1	1.7	2.7	4.8	0.67	1.1	41
2	2.0	3.2	6.0	0.61	1.0	60
5	2.7	4.5	10	0.57	1.0	100
10	3.2	5.3	13	0.57	0.94	130
20	4.4	7.2	24	0.58	0.97	240
50	5.0	8.2	40	0.55	0.91	400

unimportant for materials which can be transferred to the bloodstream. Thus, the choice of a general number to represent the intake by inhalation depends upon an assessment of the form of the plutonium in the soil and its probability of transfer to the bloodstream following inhalation. Certainly when high-fired $^{239}PuO_2$ is present, the class Y is most appropriate for, at least, the immediate problem. However, possible changes in the form of the plutonium with time have not been defined. Heat source plutonium (^{238}Pu) appears to be more soluble than ^{239}Pu because of its high specific activity. For these reasons, we have chosen to use a limiting intake of 0.7 pCi/day to represent the general situation. This will provide 0.5 rems per year to the skeleton after 70 years of continuous exposure and a sizable safety factor for the lung in the event the actual material is more insoluble than is defined by the 500-day half-life of the class Y material. This amounts to a continuous air concentration of 3.5×10^{-14} μCi/ml over this time period.

V. GENERAL RESUSPENSION

General resuspension is defined as the process or processes
by which concentrations are produced over a reasonably large
area and are strongly affected by the existing meteorological
conditions. The driving force for this type of resuspension can
be either winds or mechanical disturbances of the soils with the
resulting dust carried and dispersed by the winds.

A. Wind Resuspension

The movement of soils from agricultural fields has been
studied for many years. The processes responsible have been
classed as surface creep (for larger particles), saltation (for
intermediate size particles), and suspension (for smaller parti-
cles that can be carried by winds, often to long distances). The
suspended fraction consists of the smaller particles that are
respirable and usually of most interest. However, if Table VII
is correct, some interest must be attached to the larger parti-
cles, particularly if the plutonium is somewhat soluble in body
fluids. The surface creep and saltation are usually dominant in
terms of the mass of soil moved (at least under erosion condi-
tions) and can be an important element in moving soil (and con-
taminant) from one area to an adjacent one. However, under the
assumptions of this study, such movement is not an important
element. If the levels are low enough to meet the criteria in
the original area, movement to another area with resulting di-
lution should result in the new area meeting the criteria.
In our previous report[40] we assumed, on the basis of very
meager evidence, that the rate of pickup of contaminant parti-
cles increased as the square of the wind speed. Since this
time, important studies have been done that have shown this as-
sumption to be incorrect and have further illustrated the im-
portance of such factors as aggregation in the soil and corre-
sponding sizes of dust particles in the atmosphere. We also as-
sumed that a surface layer was the source of the wind-blown

material and that the rate of pickup could be characterized by that of freshly deposited zinc sulfide particles. While there may still be merit in the idea of a surface layer, the more recent data on weathering and consequent agglomeration, as well as the apparent stabilization of an area by particles moving into a position where they are sheltered from the wind, indicate that the situation is extremely complex and any useful definition of a surface layer must await further studies. In this report we will review only a few of the more pertinent aspects which are directly applicable to the estimation of general resuspension by winds.

Gillette[50,51] has calculated the vertical flux of 0.5- to 10- μm dust particles during erosion episodes in agricultural fields from the vertical profiles of dust and wind speeds. In two soils, having a silt and clay fraction of 4-4.5%, he found that the vertical flux increased as the 5.14 power of the friction velocity above a threshold while, in another having a sand and silt content of about 19%, the vertical flux increased as the 9.67 power of the friction velocity. This increase of vertical flux with wind velocity was greater than the increase of the horizontal flux of sand. He attributed these observations to the production of small particles in the breakup of aggregates under "sandblasting" from the particles in saltation. Shinn et al.[52] by measurements of dust profiles and a simplified and generalized model similar to Gillette's, have measured the dependency on wind speed of the resuspension rate at the GMX area in Nevada and at a recently cultivated agricultural field in Texas. Characteristics of the area and fluxes derived from the measurements are given in Table VIII.

Since the saltation in the undisturbed soils at the Nevada site is low while that in the disturbed soil in Texas was high, these results appear to be in good agreement with those of Gillette and confirm the conclusion that under conditions of erosion the resuspension rate increases rapidly with wind speed. One can also speculate from the results of Gillette that the

TABLE VIII

SHINN'S VALUES FOR TWO SITES

SITE	Z_o cm	u_x/U_2 [a]	Flux mg/$(m^2 \cdot s)$
GMX	2.0	0.1	0.73 $u_*^{3.09}$
Texas	0.044	0.05	62.6 $u_*^{7.38}$

[a] u_x is friction velocity and U_2 is the wind speed at 2 m height.

actual rate of increase may be a function of the soil composition. In fact, Shinn[52] has proposed a tentative relationship between the soil erodibility index[53] and the rate of increase with wind speed.

The resuspension rate from several contaminated areas or from tracers has also been measured. Ansbaugh et al.[54] have measured the resuspension rate from the plutonium in the GMX area using an ultrahigh volume sampler. In this area, the plutonium was deposited about 20 years ago following chemical explosions with plutonium present. His results showed values of the resuspension rate which varied from 2×10^{-12} to $6.5 \times 10^{-11} s^{-1}$ with one outlier at about $4 \times 10^{-9} s^{-1}$. When these results were normalized by dividing by the cube of the friction velocity, the values ranged from 2×10^{-11} to about $10^{-10} s^2/m^3$. Sehmel,[55] in a correlation of air sampler results from a station near Rocky Flats, found that the measured concentration at one station varied as the 2.1 power of the wind speed, which implies that the pickup rate varies as the 3.1 power of the wind speed since the dilution increases at the first power of the wind speed. Michels,[56] by integrating the quantity of plutonium in a line of samplers downwind from a contaminated area at Rocky Flats, estimated that about 7.3×10^{-4} of the plutonium on the area was carried by winds in a 14-month period. This amounts to an average pickup rate of 2×10^{-11} per second, a value within the

short-term measurements by Ansbaugh at the GMX area. Sehmel[57] measured resuspension rates using a submicrometer calcium molybdate sprayed in a 29 m radius circle in a lightly vegetated area at Hanford. The average resuspension rates were 10^{-10} s^{-1} over a period of 4 months with no apparent decrease. In a more recent report[58] he indicates that the rates varied from 10^{-10} to 10^{-8} s^{-1} over an 8-month period with little sign of decrease with time. In one experiment the samplers were turned on and off as a function of wind speed at 2.1 m above the ground. Using the midpoints of the three wind-speed ranges for calculation, the rate of pickup increased with wind speed to a power ranging from 2.5 to 3.1. If the third power of the wind speed is used, the rate of pickup can be expressed as 1×10^{-11} u^3 with the constant, evaluated from this one experiment, ranging from 9.2×10^{-12} to 1.3×10^{-11}. These values appear to be somewhat lower than those measured elsewhere for freshly deposited sources. This may well be due to the method of application or to the small particle size, either of which could promote aggregation.

The data from the zinc sulphide experiments used earlier[40] were re-examined with the conclusion that they indicate an increase in resuspension rate as the square of the wind velocity. However, in view of the evidence that a cube relation may be more viable they were fitted to this with the result that they indicate a resuspension rate of 5×10^{-9} u^3.

The various measurements have been drawn together in Table IX with some liberties taken with the data. Where the values were given in units of the friction velocity they were converted to wind speed using a ratio of $\bar{u}/u_*$ of 0.1 in the GMX area and 0.05 in the Texas experiment. The final value marked as "extrapolated" was obtained by assuming that the ratio of resuspension of soils in the GMX and Texas areas was the same as the ratio of resuspension of the plutonium in these two areas.

These values illustrate well the wide variations possible. However, we will explore the meaning of some of these variations in a practical situation in later discussion.

TABLE IX

WIND SPEED.DEPENDENCY OF RESUSPENSION RATE

Source	Based on Friction Velocity	Wind Speed	Units	Material
Gillette	$K\,u_*^{5.14}$	-	$mg/(m^2 \cdot s)$	Disturbed soil - 4% fines[a]
Gillette	$K\,u_*^{9.67}$	-	$mg/(m^2 \cdot s)$	Disturbed soil - 19% fines
Shinn	$0.73\,u_*^{3.09}$	$6 \times 10^{-4} u^{3.09}$	$mg/(m^2 \cdot s)$	Soil - GMX
Shinn	$62.6\,u_*^{7.38}$	$1.6 \times 10^{-8} u_*^{3.09}$	$mg/(m^2 \cdot s)$	Disturbed soil - Texas
Ansbaugh	$10^{-10} u_*^3$	$10^{-13} u^3$	s^{-1}	Pu - GMX
Sehmel	-	$K\,u^{3.1}$	s^{-1}	Pu - Rocky Flats
Sehmel	-	$10^{-11} u^3$	s^{-1}	$CaMoO_4$ - Hanford
Healy	-	$5 \times 10^{-9} u^3$	s^{-1}	ZnS - Hanford
Extrapolated	$10^{-8} u_*^{7.38}$	$2 \times 10^{-18} u^{7.38}$	s^{-1}	Pu - disturbed soil - Texas

(a) Fines are silt plus clay .

The extrapolated value for plutonium in disturbed soil was obtained by the ratio of Shinn's two coefficients for GMX and Texas and multiplying by the coefficient for plutonium found by Ansbaugh at GMX.

As can be seen, the values available are for a few limited areas characterized by either undisturbed, semiarid conditions or by disturbed agricultural lands. However, it is precisely in these types of areas that wind-driven resuspension could be of importance so that the use of these values should describe the areas of most interest for this pathway.

The question of the particle size of the plutonium contaminant in the soil and its influence on the resuspension rate or the relative concentration of dust in the air as compared to soil is one discussed in the previous report. Gillette[50,59] has studied the clay fraction in the soil and its existence under

several conditions. In one case he noted that the size distribution for particles <1 μm in the resuspended fraction implied a fraction of <0.1% of the soil mass in the range $0.3 \leq r \leq 1$ μm while the analysis of the soil indicated 13.7% by mass of the soil in the clay fraction (r <1 μm). However, the clay fraction was determined by use of a detergent to obtain an ultimate separation of the clay particles from the larger soil particles. The comparison of the aerosol size fraction with the soil size distribution indicated that less than 0.8% of the submicron soil particles were available to produce the submicron portion of the aerosol. Photomicrographs showed the larger soil particles to have a coating of clay particles which may be several layers thick. From these studies he concluded that aerosol derived from soil has a size distribution which seemingly follows the size distribution of the soil <u>except for r < 1 μm</u> where soil agglomeration is effective.[59] Thus, for particles of PuO_2 in the soil one would expect that those <1 μm in radius (aerodynamic diameter if spherical ∿6 μm) would be associated with the larger soil particles and would behave in an aerosol similar to these particles. As the PuO_2 particles decrease in size from 1 μm, the agglomeration forces become much stronger and the probability of disassociation by "sandblasting" in saltation or otherwise separating into discrete particles becomes much lower. In the case where the plutonium enters the soils from solutions the absorption occurs chiefly on the smaller soil particles (clays) which in turn are subject to agglomeration. Thus, while one tends to think of discrete particles of PuO_2 in the soil and air, this is probably true only in the size range at the upper end of the respirable range and above. Such agglomeration of the smaller particles and association with the larger soil particles is undobutedly an important part of the weathering process, although the speed with which it occurs, as compared to other actions which tend to shelter the particle from the wind, is unknown.

One of the concerns expressed by many is the possibility of large air concentrations and subsequent large intake of plutonium during periods of high winds and resulting dust storms. Certainly the concentrations of soil in the air are high under these conditions. Orgill,[60,61] by correlations of visibility measurements with dust concentration, has estimated the frequency of dusty hours from 343 weather observations in the continental U.S. His results indicate that a large area in the south-central U.S. exceeds 1% of the time in which the dust concentration is in excess of 3-5 mg/cm^2. On the east coast the frequency is low ranging from 0.005% to about 0.1%. The west coast, west of the mountains, is similar to the east coast.

Information is not available to fully relate these findings to potential plutonium concentrations, particularly from relatively small contaminated areas. In general, one would expect that as the wind speed increases, to produce these dusty conditions, larger and larger particles would be carried by the winds in suspension. That this may be so is indicated by data of Chepil and Woodruff[62] in which the "average equivalent diameter" (determined by the settling rate) was measured from some dust storms in Kansas and Colorado. Here the average equivalent diameter at 20 ft (∿6 m) above the ground was about 0.05 mm (50 μm) increasing to about 65 μm at 2 ft (∿0.6 m) above the ground. Porch,[63] while measuring the short-term dust concentration fluctuations in a desert area with a light scattering instrument, had two dust devils pass directly over the instrument. On neither occasion was a detectable reading observed. This he tentatively ascribed to the fact that the sampling lines would effectively eliminate particles larger than 5 μm and that it was possible that the sand transported by the dust devils was too large to get into the instrument. Sinclair[64] measured the particle size distribution from a small dust devil by use of petri dishes coated with vaseline. His results indicated a mass median diameter of about 80 μm with 70% of the mass as particles greater than 50 μm. Some 10-12% of the mass was associated with

particles less than 10 μm. He indicated that the results suggest that dust devils remove essentially the same number of small particles (<10-μm diam) that are removed by larger scale resuspension mechanisms and that small-to medium-size dust devils are capable of removing and vertically transporting sand particles as large as approximately 350 m from a wide range of desert surfaces. He further estimated that during a dust devil season of 45 days, for a typical southwestern desert area, some 157 tons per day are transported over an area of 285 km^2. However, due to the area ratios and duration times, the dust storm mean daily dust transport differs from this by a factor of 10^7.

General information on the concentrations in the air near contaminated areas is available from several current studies. In each of these cases, the plutonium has been in place for one to two decades so that they satisfy the criterion for aged sources.

At the GMX site at the Nevada Test Station, the contamination occurred through dispersion of plutonium which was intimately associated with explosive charges. Size distribution analyses of the soil particles in two samples indicated that only 1-2% of the plutonium is associated with soil particles with diameter less than 5 μm while about 90% was associated with particle sizes of 20 to 250 μm.[65] Profile samples have shown that greater than 90%, probably 95%, of the plutonium is in the first 5 cm depth from the surface.[66] The area of study is about 240 000 m^2 with an estimated total depositon of 2.5 Ci of plutonium.[66,67] Thus, the average contamination is about 10 μCi/m^2 or about 0.25 nCi/g in the top 2.5 cm. The contamination over the area is decidedly nonuniform, however, with one area of 1400 m^2 surrounding ground zero having an average level of 530 μCi/m^2.

Air samples were taken at the fence around the area in a NE and SW direction for 17 months.[68] If one estimates an average resuspension factor using the average deposition in the area, the sample to the NE showed a value of 6x10^{-10} m^{-1} while the one

to the SW gave a value of 5.8×10^{-11} m^{-1}. These results were essentially confirmed by shorter time period samples in the SE and NW directions. The higher value to the NE correlates well with the direction of the higher wind speeds even though the total frequency of winds in both directions was about the same. If we express these results in a different manner, the higher value corresponds to 2.4×10^{-5} g/m^3 (ratio of the air concentration to the average quantity per unit weight in the soil). Five cascade impactors were also operated for a period of 36 days in the summer of 1972.[68] These impactors were located 250 ft from ground zero on an arc to the NW (the direction of maximum wind speeds). The contaminated area upwind of this point was much smaller but contained the bulk of the plutonium. A rough estimate from Ref. 67 indicated the area to be about 17 500 m^2 with a total of about 1.8 Ci of plutonium. This would make the average about 100 $\mu Ci/m^2$ or, in the first 2.5 cm depth in the soil, some 2.5 nCi/g. However, it was only about 150 ft to an area of 1400 m^2 that averaged 530 $\mu Ci/m^2$. The impactor samples averaged between 1×10^{-14} $\mu Ci/ml$ and 3.9×10^{-4} $\mu Ci/ml$ with an average of 2.3×10^{-14} $\mu Ci/ml$. If we use the average concentration upwind this amounts to a resuspension factor for this smaller area of 2.3×10^{-10} m^{-1}. On the mass basis, the average value was about 10^{-5} g/m^3 with the maximum about 1.5 times greater. Of course, if one used the average for the entire area (much of which was downwind during periods of high winds) then the resuspension factors would be ten times greater because of the smaller average deposition. The average soil particle size in the air from the study was 1.6 μm with a σ_g of 15 while the AMAD for the airborne plutonium was 3.0 μm. It was noted that this latter distribution indicated that only about 20% of the plutonium was of a size that would undergo pulmonary deposition according to the formulations of the ICRP Task Group on Lung Dynamics.[49]

The contamination at the Rocky Flats Plant occurred when drums containing oil, previously contaminated with plutonium in

machining operations, leaked onto the ground. The contamination was then, presumably, spread to neighboring areas during a period of cleanup since downwind air samples showed high readings during this period. The actual area of drum storage was later covered with an asphalt pad to immobilize the highest levels of plutonium in the soil. Michels[56] indicates that an on-plant test section (including the area around the pad) of 172 000 m^2 contained 0.63 Ci of plutonium for an average of 3.7 $\mu Ci/m^2$. Krey and Hardy[69] estimate the total plutonium off the plant land and extending downwind to the 3-mCi/km^2 contour (about 7-8 miles) to be about 2.6 Ci. In this survey, samples were taken to a depth of 8 in. ($\sim$20 cm) and eight profile samples indicated that about 50-60% of the plutonium is in the top 5 cm (range 39 to 91%). In a more recent study, Krey[70] reports the plutonium off-site to be 3.4±0.9 Ci. HASL[71] also estimates the ^{241}Am to grow in from the decay of ^{241}Pu to a maximum in the year 2033 of about 18% of the ^{239}Pu activity.

Johnson,[72] of the Jefferson County Health Department, took a series of samples by sweeping dust from the ground surface. These samples were size separated by water elutriation and the <5-μm portion analyzed for plutonium. The results, in comparison with those from the Colorado Health Department who sampled to a depth of 1/8 in. ($\sim$0.3 cm) and did not do particle size separation, were higher at similar locations by factors ranging from 4 to 285 indicating that a significant portion of the plutonium is associated with the smaller particles of the soil. Nathans and Holland[73] measured the size distribution, by the use of fission track autoradiography, of the "equivalent PuO_2 diameter particle" from two air samples taken in the area. They noted that there were a large number of single and double tracks in the sample indicating the possible presence of particles smaller than 0.1 μm or the shielding of the alpha tracks by the sand grains. The size distributions of the particles with activity equivalent to PuO_2, measured by the number of tracks radiating from a point, were characterized by median diameters

of 0.15 and 0.13 μm with σ_g's of 1.7 and 1.6. The size distributions of the soil particles on the filters were characterized by median diameters of 10 and 8.7 μm with σ_g's of 2 and 2.1. Volchok,[74] using both a horizontal elutriator and a cyclone to pass particles proportional to the respirable fraction, estimated that about 25% of the airborne plutonium was respirable. This is in agreement with the size distribution measurements of the airborne soil particles, but much lower than would be expected from the "PuO_2 equivalent" particles. However, the sample size on the autoradiography study was small so that the results may represent only a portion of the population of particle sizes. Sehmel,[75] from the ratio of plutonium collected on an impactor and a high-volume air sampler, concluded that the material collected on the air sampler contained nonrespirable particles with the respirable fraction at 2 m height about 20%. This fraction increased with height, but his studies showed that this was due to an extraneous source not associated with the resuspension. From these data, it appears that the plutonium in the soils at Rocky Flats is primarily present in or on the smaller particle size of the soil, possible as aggregates with the soil particles.

A number of air sampling stations are located downwind from this contaminated area. For this comparison we have used the values of the Colorado State Health Department for Station 0-3, which is downwind from the test area described by Michels and about at the downwind edge. This station for the period 1970 to 1974[76] showed a concentration, adjusted for the background from fallout, of 6×10^{-9} μCi/m^3. Other data indicate this to be close to the maximum station at this distance. If we compare this with the average deposition found by Michels[56] for the test area, we find a resuspension factor of about 2×10^{-9} m^{-1}. If we further assume that about 40% of the plutonium is in the top 2.5 cm, the resuspension factor based on quantity per unit weight of the soil becomes about 1.6×10^{-4} g/m^3.

The data from these contaminated areas indicate the concentrations from wind resuspension to be relatively small even for areas such as the GMX area where the quantity of plutonium in the soil is significant. However, the very rapid increase in resuspension rate with wind speed noted in agricultural soils raises some doubt as to what would happen if these areas were contaminated with plutonium.

It must be remembered that the concern with plutonium in the air (as with other long-lived radioactive materials) is with the average concentration over a long period of time, such as one year, and not with the instantaneous concentration, which is represented by the measurements giving the rapid increase with wind speed. To determine the effect of wind speed on the average concentration, we calculated the concentration at the edge of a 10 000-m-wide strip infinite in the crosswind direction assuming that the wind is blowing continually across the field directly at the receptor. However, to assess the importance of frequency of various wind speeds, we used the frequencies given by wind roses in three locations.

The concentration at the edge of such a source, uniformly contaminated, was given in our previous report[40] as

$$\chi = \frac{k\Omega''}{Vu} \left[\exp\left(-\frac{4V}{\sqrt{\pi}\ C_z n}\ x_1^{\frac{n}{2}}\right) - \exp\left(-\frac{4V}{\sqrt{n}\ C_z n}\ x_2^{\frac{n}{2}}\right) \right] \quad . \tag{1}$$

When K is the rate of resuspension, Ω'' is the quantity of plutonium per unit area, $\bar{u}$ is the wind speed, C_z is the diffusion coefficient in Sutton's formulation, n is Sutton's stability parameter, x_1 is the distance to the nearest boundary of the area and x_2 is the distance to the farthest boundary. We have changed the equation slightly by defining V as equal to V_d/u where V_d is the deposition velocity. The rationale for expressing the deposition velocity as a function of the wind velocity

was discussed in the previous report.[40]   Equation (1) was derived for a height above ground of zero and provides an unrealistically high answer if the near boundary of the area is close to the receptor.   To provide a more reasonable value, the contribution of the near area was solved by numerical integration using the equation for an infinite line source upwind and a height of 1 m.

$$\chi = \frac{2K\Omega'}{\sqrt{\pi}\ c_z \bar{u}\ x^{\frac{2-n}{2}}} \left[\exp - \left(\frac{z^2}{c_z^2 x^{2-n}} + \frac{4v}{\sqrt{\pi}\ c_z n}\ x^{\frac{n}{2}} \right) \right] , \qquad (2)$$

where Ω' is the plutonium deposition per unit length of line. Since the wind roses available were broken down into frequency by stability classes, we chose to do the calculations for three stability classes: stable, unstable, and near neutral.   Where the data were broken into Pasquill categories, we combined A and B as unstable, C and D as near neutral, and E and F as unstable. One would predict that the rate of pickup for a given wind speed would differ depending upon the stability and turbulence of the atmosphere. Data on this factor are not available. However, as an approximation we assumed that the pickup coefficients were measured under neutral conditions (true in Texas) and applied a correction factor, s, based upon the change in deposition velocity with stability estimated in the previous report.[40]   Intuitively one would believe these factors to be similar since they depend upon much the same factors. Coefficients used are given in Table X.

The results of the calculations are given in Table XI. As can be seen, there is little overall difference in the average concentration although if one compares the contribution from each wind speed it is apparent that the importance of the higher increase with wind speed is important in the $\bar{u}^7$ column since the

TABLE X

COEFFICIENTS FOR CALCULATION

	Stable	Neutral	Unstable
c_z	0.07	0.1	0.3
n	0.5	0.25	0.2
s	0.33	1.0	2.3

largest contribution is in the higher wind speeds. It is probable that the $\bar{u}^7$ values at the lower wind speeds are low since Gillette[51] has used a threshold value of $\bar{u}$ of 0.18 m/s which, assuming a z_o of 0.04, would correspond to a wind speed of 3.6 m/s. Calculations assuming the GMX relationship to apply below this threshold indicate little difference in the average. This type of calculation, on the average, tends to minimize differences in extreme values, such as for wind speeds well in excess of the "greater than" values. We have tried to minimize this by choosing a relatively high value for the wind speed in the category above the top range, but this factor may result in a somewhat higher answer for the higher wind speeds. It is doubtful that it would be as much as a factor of 2.

The absolute values are of interest since this type of calculation, assuming the wind blows in the same direction continuously, would be a maximum. If we assume, as in the GMX area where the coefficients were.measured, that the plutonium is in the top 2.5 cm, then 1 $\mu Ci/m^2$ amounts to a level of about 20 pCi/g. The soil concentration to give an MPC in air of 3.5 x 10^{-14} $\mu Ci/cm^3$ would be between 2400 and 4700 pCi/g for the LASL winds, 1300 and 1800 pCi/g for the Hanford winds, and 960 and 2400 pCi/g for the Pittsburgh winds. It should be noted that the Hanford wind rose, measured at 200 ft ($\sim$61 m) represents wind speeds much higher than those at the ground which will tend

TABLE XI

APPLICATION OF POWER LAW INCREASE IN RESUSPENSION RATE

TO SOURCE INFINITE IN Y AND 10 000 M WIDE IN X.   SOURCE 1μCi/m^2

LASL Winds[77]

ū	Freq	X	
m/s	%	$10^{-13}u^3$ [a] μCi/cc	$7\times10^{-18}u^7$ [a] μCi/cc
0-1.3	45	4.2x10^{-18}	5.9x10^{-23}
1.3-2.7	18.4	1.7x10^{-17}	1.9x10^{-20}
2.7-5.4	23.2	9.0x10^{-17}	1.6x10^{-18}
5.4-8	8.7	9.1x10^{-17}	1.2x10^{-17}
8.0-10.7	2.43	4.9x10^{-17}	2.7x10^{-17}
10.7-13.4	0.60	2.4x10^{-17}	3.4x10^{-17}
>13.4(16)	0.21	1.6x10^{-17}	7.4x10^{-17}
Total	-	2.9x10^{-16}	1.5x10^{-16}

Hanford - 200' (M α AE)[78]

ū	Freq	X	
m/s	%	$10^{-13}u^3$ [a] μCi/cc	$7\times10^{-18}u^7$ [a] μCi/cc
0-1.3	14.3	1.2x10^{-18}	1.6x10^{-23}
0.3-3.1	24.4	1.8x10^{-17}	4.7x10^{-20}
3.1-5.4	22.3	5.0x10^{-17}	1.4x10^{-18}
5.4-8	17.8	9.7x10^{-17}	1.6x10^{-17}
8.0-10.7	8.33	8.9x10^{-17}	5.3x10^{-17}
>10.7(15)	4.86	1.4x10^{-16}	4.9x10^{-16}

		3.9x10^{-16}	5.6x10^{-16}

Pittsburgh[79]

ū	Freq	X	
m/s	%	$10^{-13}u^3$ [a] Ci/cc	$7\times10^{-18}u^7$ [a] Ci/cc
0-1.5	11.3	8.5x10^{-19}	2.1x10^{-23}
1.5-3.1	28.8	3.4x10^{-18}	3.1x10^{-20}
3.1-5.2	33.6	1.8x10^{-16}	4.7x10^{-18}
5.2-8.2	22.2	3.5x10^{-16}	5.8x10^{-17}
8.2-10.8	3.5	1.1x10^{-16}	7.2x10^{-17}
>10.8(15)	0.6	4.4x10^{-17}	1.6x10^{-16}
	-	7.3x10^{-16}	2.9x10^{-16}

[a] Relation of pickup rate to wind speed from Table IX.

to make the estimates of resuspension much higher. The LASL wind rose was also measured at the top of a building about 50 ft ($\sim$15 m) above the ground.

A final factor that must be considered is the size of the contaminated area. The above calculations were made for a very large area which does not seem credible for most conditions. If we assume the "infinite" dimension in the cross-wind direction to be 10 000 m, this area represents the deposition of 100 Ci of Pu. We are currently deriving a method of calculating the average concentration downwind from the area source by use of the wind rose for an area, averaging over the appropriate sector and integrating by numerical means over the area. The results from use of the wind rose from M&AE and the resuspension factors from the GMX area ($K = 10^{-13}u^3$) for square areas are given in Fig. 14. As can be seen, the maximum air concentration at the edge of the area is higher by a factor of about 7 for a 1000-m-square area than for a 20-m-square area. For the 1000-m square, the soil concentration corresponding to the 3.5×10^{-14} uCi/ml limit, using a soil density of two and a 5-cm sampling depth (thereby averaging the material on the surface through this depth) is 7600 pCi/g.

B. Mechanical Disturbance

The mechanical disturbance of soils can provide forces which will cause resuspension of dust and associated contaminants during the period of the disturbance. Such disturbances can range from the relatively mild, such as animals grazing or an individual walking through the area, to the very severe, such as plowing or other agricultural operations or the use of earth moving equipment. In the latter case, the increased suspension can be noted by the dust cloud produced.

Suspension by mechanical means differs from wind resuspension in several respects. First, the driving force is a function
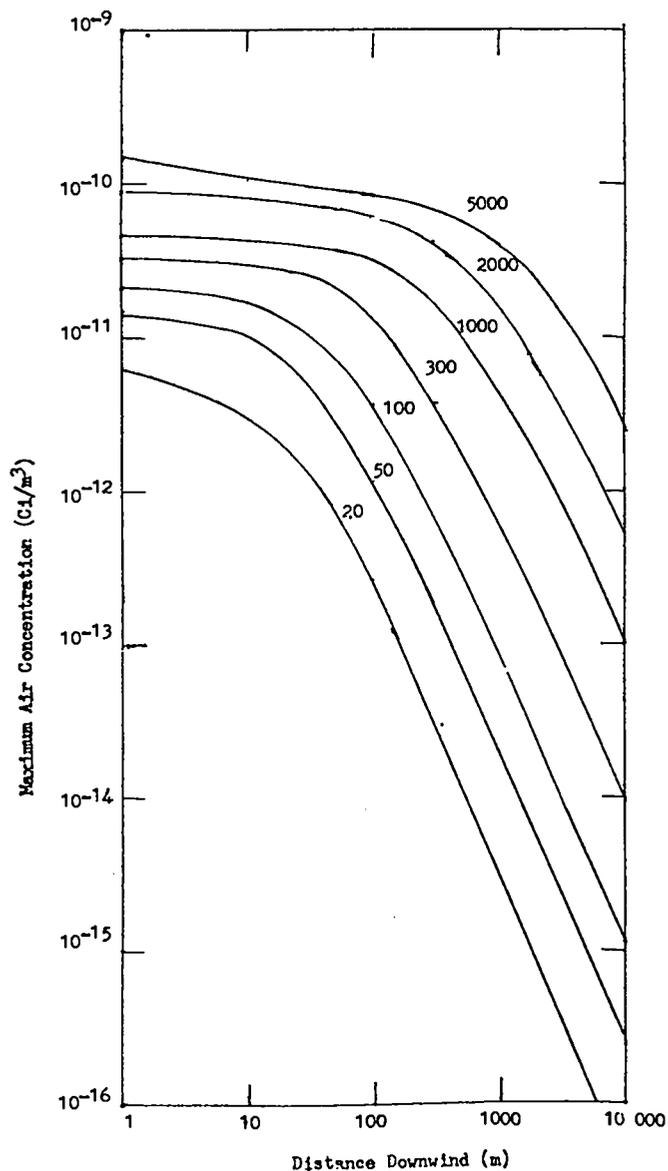
Fig: 14.

Air concentrations downwind from square areas of dimensions given having a uniform level of one Ci/m^2. Pickup rate is given by $10^{-13}u^3$.

of the action causing the suspension and can vary widely depending upon the magnitude of this action. In many cases, the disturbance is greater than that caused by the winds and is probably different in character since the action can also disturb the

subsurface and result in suspension of material from some distance below the surface, with this distance dependent upon the nature and magnitude of the disturbance. Note that we are here considering only the suspension at the time of the action; possible increases by winds following such a disturbance due to the change in the nature of the surface is covered by the considerations in the previous section where the ultimate in disturbed surfaces, a plowed field, was considered. Since the suspending force is divorced from the wind speed, it should be a constant depending only on the action. The concentration downwind will vary with the wind speed, being lower at the higher wind speeds because of the increased dilution.

The second difference arises from the localized nature of the mechanical disturbance and the resulting resuspension. In other words, the dust arises only from the very localized area or areas of disturbance rather than from the entire area and occurs only when the disturbance is active. Thus, a moving automobile or tractor will generate a dust cloud as it moves with resuspension dependent upon the speed of the vehicle as well as its size and the character and condition of the soil. This moving point source will average to a line source with the average resuspension rate and resulting average concentration downwind dependent upon the time of averaging. A farmer plowing in a field will produce a point dust cloud which will average to an area source with the average resuspension rate from the area source again dependent upon the time of averaging.

A third difference is in the increased rate of mixing of contaminant in the surface layers with the soils underneath, at least for the more active disturbances. Thus, if excavating is in progress in the area, the contaminant in the top of the soil is mixed with the soil underneath. In the case where the contaminant is chiefly in the top layers, this will result in a dilution of the contaminant. This point is of interest even for aged contamination areas, since weathering usually results in a gradient with decreasing concentrations as one moves deeper in the soil.

Some information is available on the effects of mechanical disturbance from trucks moving through contaminated areas in arid country following dispersal of plutonium in safety tests. However, these experiments were done in freshly contaminated areas rather than the aged areas of interest here, and the results were expressed as resuspension factors rather than in a form which can be used for analysis. It is of interest to note that the data from wind resuspension at Rocky Flats include a certain undefined component of mechanical disturbance. This may be one factor in causing the somewhat higher resuspensions noted earlier in this area as compared to the GMX area in Nevada.

In our previous report,[40] data from Mork[80] resulting from a vehicle driving through a freshly contaminated desert area were analyzed. If a wind speed of 2 m/s is assumed for this experiment, then out of seven values, four ranged between 2.6×10^{-7} s^{-1} to 3.6×10^{-7} s^{-1} while the other two indicated 2×10^{-5} to 6×10^{-5} s^{-1}. The difference has not been resolved. It was also noted that the particle sizes resuspended must have been very large since the deposition velocities measured with sticky paper at the point of sampling ranged from 0.004 to 2 m/s with only two of the seven values less than 0.1 m/s.

The study of Sehmel[81] of resuspension from an asphalt road due to passage of a car and the three-quarter ton pickup truck was also included in the previous report.[40] Here the pickup rate varied from 10^{-8} s^{-1} to 8×10^{-3} s^{-1} depending primarily on the speed of the vehicle since the resuspension rate was shown by Sehmel to be proportional to the square of the speed of the vehicle. Sehmel also indicated that the depletion of the resuspendable contaminant with any significant traffic would be significant. While these results are not directly applicable to the soil problem, since they represent fresh deposition on an asphalt surface, they do indicate some maximum orders of magnitude for a hard surface and also indicate that such surfaces when heavily disturbed will not greatly affect the outcome of a contaminating event since they will serve only to

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provide a slight delay for the contaminant deposited on the surface before the contaminant continues downwind.

Sehmel[82] also measured the resuspension of ZnS particles from the passage of a 3/4-ton truck through an area covered with cheat grass at speeds of 8.05 km/h, 24.14 km/h, 48.28 km/h, and 64.37 km/h. The area was seeded with the particles, wind resuspension measured, and the truck passed through at 8.05 km/h. The other runs were made the following day. Results from the truck passage experiment are given in Table XII. The wind resuspension during the first day was 3.4×10^{-8} s^{-1} for a 4.7 m/s wind speed. Sehmel notes that this is a high value for the resuspension and probably reflects an increase due to resuspension of the particles from the upper portions of the cheat grass. The decrease after the first truck pass was attributed to the prior removal of the most readily resuspendible particles from the grass itself. Following this, there was an increase with truck speed although the resuspension rates at the higher speeds are one to two orders of magnitude lower than for the asphalt road.

In a recent paper, Milham et al.[83] described the results of air sampling during the agricultural preparation of two fields

TABLE XII

MECHANICAL RESUSPENSION FROM TRUCKS

IN CHEAT GRASS AREA

Truck Speed (m/s)	Resuspension	
	per pass	per second
2.2	2.5×10^{-4}	3.7×10^{-5}
6.7	9×10^{-6}	4.0×10^{-6}
13.4	9×10^{-6}	8.0×10^{-6}
17.9	8×10^{-5}	9.4×10^{-5}

having low levels of plutonium accumulated 25-30 years earlier as a result of an airborne release from a nearby stack. The measurements were made downwind 7.6 and 30.5 m from the edge of the field with six samples at face height; one located on the center line of the field width and the other two 7.6 m on either side. Results of this experiment are summarized in Table XIII with the air concentrations being the average of the three samplers at a given distance.

A somewhat similar experiment was done by Myers[84] at Livermore. The plutonium was applied to a small field (10m x 17m) in the form of a digested sludge from a sewage plant. The sludge contained 2.8×10^{-6} uCi/g of Pu and was applied to a depth of 7.5 to 10 cm. The sludge was allowed to dry for 4 weeks during a period of no rain and the area was rototilled. The rototiller was 2 m wide and was pulled behind a tractor with the dust cover open in order to maximize the escape of dust. Air sampling was done at one end of the plot with an Anderson cascade impactor. The soil after rototilling contained 4.3×10^{-7} µCi/g and the air sampler indicated an average concentration of 2.4×10^{-8} µCi/m^3.

TABLE XIII

MECHANICAL DISTURBANCE RESULTS

FROM SAVANNAH RIVER

	North Field	South Field	
Soil Conc (pCi/g)	0.9	4.6	
Wind Speed (m/s)	2.7	5	
Field Length (m)	105	30	
Field Width (m)	30	145	

	Distance from Field Edge			
	7.6m	30.5m	7.6m	30.5m
Air Conc (pCi/m^3)				
Bush Hogging	0.046	0.028	0.88	0.033
Disking	0.02	0.011		
Subsoiling	0.34	0.0052	0.029	0.0014
Fertilizing	0.01	0.0058	0.0089	0.002
Planting	0.536	0.0062	0.014	0.012

About 50% of the plutonium in the air sampler was on the filter indicating a particle size in this fraction of <1.1 μm AMAD.

A resuspension rate was estimated from these data by integrating over the area using Sutton dispersion parameters of $C_y=0.2$, $C_z=0.1$, $n=0.25$ and ratio of deposition velocity to wind speed of 0.005. Since the wind speed was slightly off the axis of the field in each case, a correction was applied for this. These resuspension rates from both experiments are given in Table XIV.

From these data, a value of 10^{-7} s^{-1} to represent mechanical disturbance was chosen. Ideally one would like to have a sufficient number of values with different disturbances so that a reasonable pattern could be chosen and calculations made for each in assumed conditions. However, the paucity of data does not allow this. While some values in the table are higher than 10^{-7} s^{-1}, many with vigorous disturbance are lower. Since we are trying to represent an average situation over a period of time, the choice appears justified and may be conservative when the full range of possible activities in an area is considered.

For application we considered the case where mechanical disturbance occurs repetitively year after year such as a farmer

TABLE XIV

ESTIMATED RESUSPENSION RATES (s^{-1})

SAVANNAH RIVER

	North Field		South Field	
	7.6 m	30.5 m	7.6 m	30.5 m
Bushhogging	9×10^{-8}	2×10^{-7}	1×10^{-6}	8×10^{-8}
Disking	4×10^{-8}	6×10^{-8}	–	–
Subsoiling	7×10^{-7}	3×10^{-8}	3×10^{-8}	3×10^{-9}
Fertilizing	2×10^{-8}	3×10^{-8}	1×10^{-8}	1×10^{-8}
Planting	1×10^{-6}	4×10^{-8}	6×10^{-7}	2×10^{-8}

LIVERMORE

| Rototilling | 9×10^{-8} | | | |

plowing a field with the farm house or other occupancy on the edge of the field. A series of square areas was considered upon which mechanical disturbance appropriate to the resuspension rate of 10^{-7} s^{-1} was carried out for 40 h/yr. The resulting air concentrations at a height of one meter at the downwind edge of the area was calculated by numerical integration using the dispersion coefficients previously employed for the wind resuspension calculation and the wind directions and frequencies given for the 200-ft (61-m) level at Hanford in Meteorology and Atomic Energy, 1968.[78] The hemisphere with the maximum frequency of high wind speeds was chosen. These results are given in Table XV.

The values for different depths are shown to indicate the difference resulting from depth of disturbance if the soil is uniformly contaminated to this depth and the disturbance is such that equal probability of resuspension occurs to all depths or the effect of different sampling depths if the contamination were all in the first centimeter.

TABLE XV

LIMITING SOIL CONCENTRATIONS AT EDGE OF VARIOUS

SIZED AREAS UNDER MECHANICAL DISTURBANCE

Area Size (m^2)	DEPOSITION			
	$\mu Ci/m^2$	pCi/g [a]		
		1 cm depth	2.5 cm depth	5 cm depth
20	140	7000	2800	1400
50	60	2900	1200	580
100	35	1800	700	350
300	20	1000	400	200
1000	14	680	270	140

[a]Assuming soil density of two .

VI. LOCAL RESUSPENSION

The concept of local resuspension was defined to permit the assessment of inhalation from activities which can produce concentrations over a small volume in the vicinity of an individual or his nostrils and which is not measured by air samples taken in the immediate vicinity. It includes both inhalation from disturbance of the soil by the individual resulting in local concentrations and the more casual activities such as removing clothing contaminated in the area or potential direct inhalation from contaminated skin or objects. The "resuspension factor" measured as the ratio of the air concentration to the quantity on the ground is a valid technique for estimating the mechanical disturbance since it relates the local air concentration to the local ground conditions. However, data on casual inhalation are meager, particularly if one must consider the transfers of contamination from the soil to the skin, clothing, or another contaminated object which is the agent carrying the contamination to the point of exposure.

For local resuspension, while an individual is outside working or playing in the soil, the important soil fraction would appear to be the smaller particle sizes which could remain suspended for a period of time in the absence of winds. If a wind strong enough and turbulent enough to hold larger particles were blowing it would rapidly remove the material from the vicinity of the individual. Since the soils in the area of interest are unknown, we have considered several soils of varying clay content (<2 µm as determined by sedimentation following breakup of the aggregates) and assumed that all of the plutonium in the soil was on these fine particles. This is undoubtedly a worst case since the clays are normally strongly adsorbed on the sand particles or agglomerated. It is considered that an individual spends five 8-h days in vigorous activity on the contaminated area each year during periods when the soil is bare of vegetation and dry enough to produce dust.

Data to fully justify a choice of resuspension factor for various activities are few. Experiments at Savannah River[83] demonstrated resuspension factors in the cab of a tractor during various agricultural operations of 4×10^{-9} m^{-1} and 2×10^{-7} m^{-1}. At Livermore[84] rototilling under maximum dust conditions with the plutonium in a dry sewage sludge on the surface gave a value of 6×10^{-7} m^{-1}. Stewart[85] has recommended a value of 10^{-5} m^{-1} for moderate activity in an area of fresh deposition. For over-all resuspension factors others have considered the value in an aged area to be 10^{-4} times that in a freshly deposited area.[86] Considering the wide variance in activities in such an area, we will use a value of 5×10^{-7} m^{-1} for estimating the average expo-sure during periods of mechanical disturbance.

If f is the fraction of small particles in the soil (clays plus some fraction of the silts), d is the depth of the distur-bance in the soil, and ρ is the soil density, for a resuspension factor of 5×10^{-7} m^{-1}, the average concentration of soil in the air (χ_s) over a one-year period for the 40 h of activity is

$$\chi_s = 2.3\times10^{-5}\rho f d \qquad (3)$$

The coefficient takes into account the resuspension factor, the conversion to m^2, and the averaging of the 40-h exposure over a one-year period. It may be noted that this equation assumes that only the small particle sizes are resuspendable. Thus, the re-suspension factor does not correspond to the measured values where the total mass of contaminant or soil in the 1-m^2 area is used in the denominator. In fact, this resuspension factor corresponds to the measured factor as 1/f. Thus, in an area where only 10% of the particles are available to be resuspended, the measured value would be 10 times the value used or 5×10^{-6} m^{-1}. Since data on the relation between the resuspension factor and soil composition are not available to either justify or deny this assumption, this method was used as one technique to il-lustrate the possible effect of particle size.

If we assume that all of the plutonium in the soil is in the fine particles, the concentration in the small particles to limit the air concentration to 3.5×10^{-14} μCi/ml (0.035 pCi/m^3) is

$$pCi/g = \frac{0.035}{2.3 \times 10^{-5} \, \rho fd} = \frac{1500}{\rho fd} \qquad (4)$$

Values for the limiting soil concentration for several values of these parameters are given in Table XVI.

TABLE XVI

SOIL CONCENTRATION TO GIVE ANNUAL INHALATION

FOR VARIOUS DEPTHS OF DISTURBANCE OR SAMPLING

AND SOIL PARTICLES (ρ = 2 g/cm^3)

Soil Depth (cm)	% Fines	Fines (pCi/g)	Soil Concentration in Total Soil (pCi/g)	Air Conc (µg/m^3)[a]
1	1	75 000	750	100
2.5	1	30 000	300	250
5	1	15 000	150	500
1	10	7 500	750	1 000
2.5	10	3 000	300	2 500
5	10	1 500	150	5 000
1	30	2 500	750	3 000
2.5	30	1 000	300	7 500
5	30	500	150	15 000

[a] During disturbance.

For the results in this table we have ignored aggregation and have considered all of the clays and small silt particles to be equally susceptible to resuspension. As was noted earlier, this is an unrealistic assumption since the smaller particles are absorbed onto sand particles or are aggregated with each other.

It is of interest that the value based on total soil is independent of the fraction in small particles although the concentrations in the fines, themselves, will vary widely depending upon the fraction present. If one considers the top one centimeter to be the layer of importance because of its susceptibility and there is no activity in lower layers, then the value given for the 2.5- and 5- cm depth as a sampling depth will be appropriate. Of course, if the activity has penetrated below 1 cm, application of the value for the deeper sample will be adequate. It is noted that this type of calculation indicates one possible difference in the derived limit between a sampling based on total soil and on only the respirable fraction. It does, however, assume that the forces acting at 5 cm depth are as likely to resuspend the small particles as those at the surface. This appears to be unlikely and is probably a conservative assumption for this sampling depth.

We attempted to derive a value for indirect transfer based upon dust in the home. However, no useful information on the amount of dust in the home that was derived from outdoor soil (as opposed to dust generated from other sources) was found to permit relating to a soil concentration. In a literature review of comparative concentrations indoors and outdoors, Benson et al.[87] concluded that indoor atmospheric particulate concentrations appear to be generally lower than outdoor concentrations, especially at high outdoor levels, and the particulates indoors contain more organic material than those outdoors. The later finding would seem to indicate that a significant fraction of the indoor dust may originate from sources other than the soil. In two studies out of those reviewed, the conclusions were that

the indoor activities were as important as the outdoor dust load and in one case a correlation was found between the number of people in a class room and the dust loading.

This potential pathway must remain unsolved until better data are available. However, the comparison between atmospheric dust indoors and dust outdoors would indicate that this pathway may not be limiting.

Dunster[88] in a review of studies by others indicates that a recommended "safe" value of 0.1% of the material present on the hands inhaled during smoking of a cigarette is high compared to the measured value by a factor of about 50. Thus, a reasonable fraction transferred would be 2×10^{-5}. Lepow[89] has measured the quantity of dust on the hands of children as 0.011 g. Since the hands of an adult are about four times as large as a two-year-old, this would imply 0.05 g on the adult assuming similar actions. If an individual smokes one pack per day the quantity of dust inhaled would be 2×10^{-5} g. For one pCi/day inhaled this would give a dust concentration of 50 000 pCi/g or a concentration for the surrounding soil of 5000 pCi/g. If we consider the actions of children, or other actions of adults, which bring the hands in proximity to the face, the increased proximity to the nostrils and the frequency of these actions would indicate that the value should be lower, perhaps by a factor of ten or twenty.

There are data on the concentration in the breathing zone as a result of wearing contaminated clothing. During the changing of cotton coveralls, Healy[1] estimated from the data of Brunskill[90] that the quantity inhaled would be equivalent to that on 1-1/4 cm^2 of the coverall. Butterworth and Donoghue[91] studied the concentration resulting from contaminated coveralls while the individuals were carrying boxes, walking, and filing a metal rod. This resulted in a transfer equivalent to that on 0.3 cm^2/h for 50% terylene - 50% cotton and 0.5 cm^2/h for cotton coveralls. If we consider a dust quantity on the clothing equivalent to that on the children's hands (0.06 mg/cm^2) over a 12-h period about 0.44 mg would be inhaled. For an inhalation rate of 0.7

pCi/day this would correspond to 1600 pCi/day of dust or 160 pCi/g of the surrounding soil.

VII. INGESTION

Ingestion of plutonium is usually considered to be minimal hazard as compared to inhalation because of the low uptake from the G.I. tract. The G.I. tract itself is of even smaller concern since only a small fraction, if any, of the alpha radiation from the contents can reach the critical cells. However, it is necessary to reconsider the uptake for people living continually in a contaminated area since there will be mechanisms different from those encountered in occupational situations of less intimate contact.

In this section we consider the normal pathway of intake with food and elaborate somewhat on questions as to intake by children.

A. Foodstuffs

The uptake by plants from soils has been investigated in some detail. We will not, here, detail the results of these investigations since there are several reviews available.[92,93] In normal soils, the uptake is low with the concentration factor (ratio of quantity per unit weight of plant material to quantity per unit weight of soil) of 10^{-4} or less. The application of chelating agents, such as FeDTPA, to the soil will increase this uptake greatly. Values of the concentration factor as high as 1, or somewhat greater, have been obtained in pot culture by application of chelating agents in quantities well above those used for normal agriculture. There is no doubt, however, that the use of such agents in agriculture will increase plant uptakes.

One of the concerns with such measurements is that they may not be representative of the plant uptake after the plutonium

has resided in the soil for some period of time. Here it is hypothesized that agents such as soil bacteria or natural chelating compounds will change the form of the plutonium to increase the uptake. There has been increased uptake noted in perennial plants cropped over a period of three to five years. However, Romney et al.[94] have pointed out that this may be due to increased root development so that a conclusive answer cannot be given. Schulz et al.[95] grew wheat plant seedlings to maturity in pots containing a high-yielding forest soil, an agriculture soil from near Sacramento, and a fertile calcareous soil from the San Joaquin Valley. Plutonium, as nitrate or chloride, was intimately mixed with the soil. In this experiment it was anticipated that the uptake from the acid forest soil might be greater than from a neutral valley soil with low organic matter. However, with either the chloride or nitrate, the uptake in the forest soil was about one-fifth that of the valley soil.

However, the root uptake is not the only method of accumulation of plutonium in vegetation. Romney[96] has shown that the vegetation in the contaminated areas of the Nevada Test Site has dry weight concentration factors on the order of 0.05-0.2 due to the direct accumulation of soil on the surface of the plant. McLendon et al.[97] at the Savannah River plant indicate that the average concentration factors between soil and camphor weed and honeysuckle (native plants) ranged from 0.07 to 0.22 on a dry weight basis with the lower values obtained offsite where the soil concentrations were lower. Since these were on a dry weight basis, they should be about a factor of 5 lower, or 0.014 to 0.044, to represent the more applicable wet weight basis representative of the diet. On the same wet weight basis Romney's values would be 0.01 to 0.04 giving excellent agreement between these two widely different types of areas. Meyer et al.,[84] at Livermore, grew vegetables in garden plots 10 x 17 m fertilized with sewage treatment sludge containing small amounts of plutonium. They measured concentration factors, after preparing the vegetables, ranging from 3×10^{-5} to 2×10^{-4} for corn, turnips,

broccoli, and tomatoes. Interestingly, the crops grown on control plots with only fallout indicated higher uptakes ranging from 9×10^{-3} to 1×10^{-2}. In each case, broccoli was highest. These values were again on a dry weight basis indicating that they should be reduced to 6×10^{-8} to 4×10^{-5} for the sludge enriched plots and to 2×10^{-3} for the control plots.

The most pertinent data arise from a study of plutonium from fallout in the New York diet[98] as discussed in the LMFBR Environmental Impact Statement[86] since this includes a wide variety of foods grown in different conditions and a representative indication of the quantities of each eaten by the population. In this study, the cumulative plutonium fallout of 2.65 mCi/km^2 in 1972 at New York City was assumed to be representative of the fallout where the foods were produced. This plutonium was further assumed to be distributed through a 20-cm depth of soil of a density 2, again in the areas where the food was produced. These results were compared with the concentration in the diet for 1972 to arrive at concentration factors for each component of the diet. The fresh fish and shellfood were removed since our concern is with soil. These results are given in Table XVII.

These data indicate that the overall quantity of plutonium in plants used for food will be above that predicted from laboratory experiments on plant uptake. The data on fallout in the New York data are the most convincing since the results are for plants and plant products in a table-ready condition. The data on the native plants at Nevada and Savannah River give higher values than the New York City data, but these values are for plants directly from the area and not in a prepared form. It must be remembered that in the assessment of these data, we are interested in the probable average uptake considering the full diet of plant material. That is, it is perfectly acceptable to have a portion of the plant-derived food 10 times the value chosen as long as it is balanced by another food in equal quantity 10 times as low. In this consideration, however, the

TABLE XVII

PLUTONIUM IN NEW YORK FOODS - 1972

Food Item[a]	Plutonium Concentration (10^{-12} Ci/kg)	Conc Factor	Per Capita Annual Intake	
			Food (kg/yr)	Pu(10^{-12} Ci/yr)
Plant-Derived				
Bakery Products	0.0085	1.3×10^{-3}	44	0.37
Whole Grain Products	0.0060	9.1×10^{-4}	11	0.066
Fresh Fruit	0.0051	7.7×10^{-4}	59	0.30
Dry Beans	0.0049	7.4×10^{-4}	3	0.015
Fresh Vegetables	0.0043	6.5×10^{-4}	48	0.21
Root Vegetables	0.0035	5.3×10^{-4}	10	0.035
Flour	0.0028	4.2×10^{-4}	34	0.095
Rice	0.0015	2.3×10^{-4}	3	0.004
Potatoes	0.0013	2.0×10^{-4}	38	0.048
Macaroni	0.0012	1.8×10^{-4}	3	0.004
Canned Vegetables	0.0009	1.4×10^{-4}	22	0.019
Fruit Juice	<0.0003	$<5 \times 10^{-5}$	28	<0.007
Canned Fruit	<0.0002	$<5 \times 10^{-5}$	11	<0.002
Total Plant-Derived Diet	0.0037[b]	5.6×10^{-4}	314	1.17 - 1.18
Animal-Derived				
Poultry	0.0033	5×10^{-4}	20	0.066
Meat	0.0026	3.9×10^{-4}	79	0.20
Eggs	0.0012	1.8×10^{-4}	15	0.019
Milk	<0.0003	$<5 \times 10^{-4}$	200	0.064
Total Animal-Derived Diet	0.0010[b]	1.5×10^{-4}	314	0.29-0.35
Total Diet				1.5

[a] Food categories and quantities are based on Department of Agriculture food purchase surveys.
[b] Values are calculated from total per capita annual intake of food and of plutonium.

question of variation in the makeup of the diet among individuals must play a prominent part.

From these data, overall concentration factors for foodstuffs as eaten (wet weight) over the concentration in soil are taken as 6×10^{-4} for plants and 2×10^{-4} for meat. With an allowable intake of 270 pCi/day based on 0.03% absorption from the

G.I. tract, the soil concentration corresponding to these concentration factors is 390 pCi/g for the total New York diet. If one considers that vegetarians eat the combined weight of meat and plant materials, the soil concentration would be 260 pCi/g. Note that these values are based upon distribution of the plutonium through the cultivated layer of the soil and they further assume that <u>all</u> of the food consumed by the individual is obtained from the contaminated area. The home gardeners who grow their own fruit, vegetables, and potatoes are a more frequent occurrence. If we use these items from Table XVII, the quantity consumed is 158 kg/yr and the soil limit becomes 1100 pCi/g.

B. Direct Ingestion

The question of direct ingestion of the soil was introduced in the previous report[40] as a potentially important pathway of exposure. This does not include the soil ingested with fruits or vegetables because this was included in the enhanced uptake used in the previous section. Here we consider direct ingestion as the actual consumption of soil either accidentally or deliberately.

There is considerable evidence available from the studies of lead accumulations and poisoning in people that children are the critical group for this element for several reasons including greater absorption into the body than adults, their ingestion of foreign materials such as paint chips, and of interest to this study, their greater contact with soil and household dust. While it is not possible to make direct comparisons of these data with plutonium intake because of the multiple sources of lead in the environment of children, the lack of a firm correlation at various ages between intake and blood levels, and the difference in metabolism of the lead and plutonium, there is considerable information of value for assessing the potential plutonium problems in these studies. A brief review of the results applicable to casual ingestion by children will be followed by an application to plutonium before addressing the deliberate ingestion question.

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Sayre et al.[99] noted that the data on ingestion of paint flakes could not account for the high fraction of inner-city children that had high blood levels of lead as compared to adults in the same region or suburban children of the same age group. He tested for the amount of lead on children's hands and on windowsills and floors of the play area. He indicated that a clear relationship was evident between individual hand and household levels. He also noted that blood levels are not commonly elevated until one year of age while children do not crawl until about eight months. The blood levels were fairly constant in children between 2 and 4 years of age (40-50 ug/100 ml) and they then fell to 35 µg/100 ml in children 5 years of age. He concluded from this that, although intensive mouthing decreases after 2 or 3 years of age, dirty hands are possibly more common and hand-to-mouth activities do persist to some degree. While they do not definitely implicate soil and dust, in contrast to others of the multiple sources of contamination, the authors do indicate that the results suggest dust ingestion as a possible mechanism of exposure.

Antopol et al.[100] measured the reservoirs of lead in dust collected from the outer ledge of the southeast facing windows in Beth Israel Hospital as a result of automobile emissions. Concentrations ranging from 2000 to 50 000 ppm were found. They contrast this with the air concentrations of 3-5 $\mu g/m^3$ in light traffic and 6-9 $\mu g/m^3$ in heavy traffic and indicate that an individual might inhale 1 mg or more of lead from the dust during several deep inspirations. They also note the observation that brain lesions are more frequent in preschool children and occur most often from late spring into early autumn. From this they suggest that this corresponds to the season when windows are open and the preschool child's mouth and nose reach the level of the windowsill or just above it.

Lepow et al.[89] report that many of the urban children with 40-60 µg/100 ml blood levels attending the University of Connecticut clinics had no history of pica (a craving for nonfood

substances) for paint nor was leaded paint (>1% lead) found in the dwellings. There was no rural counterpart for this group. They studied 10 children with chronically elevated lead levels (40-120 µg/ml of blood). When a source of leaded paint had been found it was removed, and yet blood lead levels remained significantly elevated. Soil samples taken outside of 13 dwellings gave a mean of 1200 µg/g. The levels were highest near the street and next to buildings. The mean level in household dust was 11 000 µg/g with the highest concentrations on windowsills and in floor dust. The mean lead concentration of hand samples from 22 children was 2400 µg/g with the mean weight of hand sample 11 mg.

Landrigan et al.[101] in a study of a smelter in El Paso, Texas found lead concentrations in the soil within 200 m of the smelter of 560-11 450 ppm with a mean of 3457 ppm. The lead content was consistently higher at the surface. Household dust in Smeltertown, a village with a population of 500 adjacent to the smelter gave a geometric mean of 22 191 with a range in 53 samples of 2800-103 750 ppm. The distribution of individuals with blood levels above a given value are given in Table XVIII.

This table clearly indicates the increased susceptibility for intake to children. It is difficult to ascribe the entire lead intake by the children to a given source since air concentrations were elevated and paint samples indicated the presence

TABLE XVIII

BLOOD LEVELS BY AGE GROUP IN SMELTERTOWN

Age Group (Yr)	No. Measured	% 49-59 µg/100 mℓ	% >60 µg/mℓ
1-4	49	55	14
5-9	101	34	11
10-19	109	25	6
20+	98	16	0

of paint with a lead content greater than 1% in 30% of the homes. In a separate survey to better measure the sources of exposure, the geometric mean lead content of household dust was found to be 4022 ppm and a highly significant relation was found between the lead content of household dust and lead levels in the blood of children 1 to 19 years old. Children with blood levels $\geq$40 µg/100 ml (approximately the upper limit of "normal" values) lived in homes where the geometric mean lead content in dust was 6447 ppm while those with levels less than 40 µg/100 ml lived in homes with 2067 ppm in dust.

Roberts et al.[87] in a study of two smelters in Toronto noted that the blood lead concentrations of children increased with proximity to the smelter although there was no significant trend for adults. In this case the soil concentrations near the smelters were 40 000 and 16 000 µg Pb/g of soil trailing to an urban background of 100 to 500 µg/g at distances of about 400 m. However, only 30% and 8% of the variation in blood lead could be attributed to this change with distance. Examination of head hair indicated the lead content did not vary significantly with distance along the hair from the head indicating that the rate of absorption of lead was fairly constant. While the air concentration was higher closer to the smelters than at a distance, 70% of this increase was attributed to large, nonrespirable particles and indoor concentrations during the winter months were only 20-30% of those outside. From this, they conclude that poor personal hygiene and play habits of children living in the areas where the lead content of house dust, outdoor dust, and soil is elevated can result in excessive lead absorption. However, Randall et al.[102] in studies with baboons has shown that the blood lead levels from inhalation of "coarse" particles (mean diameter 1.6 µm) is greater than from "fine" particles (mean diameter 0.8 µm) presumably due to lesser expiration of the coarse particles and subsequent absorption from the gut. The data are not adequate to indicate whether the increased particle size near the smelters could explain a portion of the increased uptake.

Vostal et al.[103] measured the lead on floors and windowsills of homes along with the lead on the hands of children in the house by a smear technique. There was a definite correlation between the two in an inner city area where the levels were high enough to give reliable measurements. In one home where four measurements were made on one child by different investigators over a period of several months, the ratio of lead on the hands to the average of lead per ft^2 on the floor ranged from 0.23 to 0.84 with an average of about 0.5. They also measured the distribution of dust on a child's hands with the results shown in Table XIX.

Lepow et al.[104] investigated the environmental and behavioral factors in 10 preschool children having high blood levels over the previous 6-25 month period. Readily accessible leaded paint had been eliminated as a significant source for at least three months before the study. The mean age of the children was 52 months. All but one had a previous history of pica and current pica was reported for four. They reported outdoor dirt with concentrations having 95% confidence limits of 700-1750 µg Pb/g (mean 1200) while house dust had a 95% confidence limit range of 4900-17 000 µg/g (mean 11 000). In each case samples were taken with preweighed, self-adhesive labels pressed on the

TABLE XIX

DISTRIBUTION OF DUST ON THE HAND

	Right Hand % of total	Left Hand % of total
Thumb	14.9	11.9
Index finger	23.3	18.0
Middle finger	12.6	17.0
Ring finger	15.8	19.2
Small finger	66.3	12.5
Palm	17.8	21.3

surface. The concentration in the dirt on children's hands had 95% limits of 650-4100 µg/g (mean 2400), a value only twice that of the soil although the house dust was about 10 times. It was also noted that the difference between lead content on the hand samples obtained when children were playing outdoors was not significantly different from that obtained when they were playing inside of the dwelling. They point out that the higher lead concentration in house dust than outside may be due to a number of mechanisms including the presence of old pulverized paint indoors. Following observation of the 10 children it was concluded that a theoretical mouthing frequency of 10 times a day was an extremely reasonable, if not conservative, estimate of the frequency of hands or nonfood items being placed in the mouths of these children.

Ter Haar et al.[105] measured the natural ^{210}Pb in various materials to use as a tracer as to the origin of lead in the feces and urine of children having x rays showing opaque materials in the gut, a history of pica, elevated blood lead, or x-rays showing lead lines in the long bone. The ^{210}Pb content of various materials was determined with the results given in Table XX.

TABLE XX

^{210}PB CONTENT OF NONFOOD MATERIALS

Material	^{210}Pb (pCi/g)
Paint chips	0.005 - 0.07
Urban airborne particles	60-150
Fallout dust	3-30
Vacuum cleaner sweepings	0.4-1
Yard dirt	0.3-2
Street dirt	0.4-4

The ^{210}Pb content of the excreta from children with signs of exposure and that of the children with low exposure was identical. Chelation treatment of one child with very high excretion of normal lead increased the stable lead output by a factor of 10-30 but the ^{210}Pb did not increase. From these data the author concludes that children with pica (and other evidence of high lead intake) and normal children excreted identical amounts of ^{210}Pb. Thus, dust and air-suspended particulate were not the sources of lead in these urban children.

Baltrop et al.[106] measured the lead concentration in soils, in blood of children and mothers, and in feces of children in two towns where the lead content of the soil differed by a factor of about 2, in an attempt to determine the importance of soil content in lead intake. Data from the soil sampling are given in Table XXI.

The similarity of the deep sample and surface sample in the grasslands at Matlock indicates that the lead may be of natural origin. The mean suspended particulate was 0.61 g/m^3 in Matlock and 0.29 g/m^2 in Buxton for a one-year period. This is low enough that the airborne lead was not contributing significantly to the intake. The results of the sampling of child and mother are given in Table XXII.

TABLE XXI

SOIL SAMPLING RESULTS - MATLOCK AND BUXTON

| | | Matlock | | | Buxton | | |
	Depth	# Samples	Mean	Range	#	Mean	Range
Front[a]	0-5	44	1263	164-7400	86	817	118-3300
Back[a]	0-5	36	1367	305-5080	53	557	160-3300
Back[a]	0-5	50	1121	230-8000	59	423	82-2640
Back[a]	30-45	51	913	92-4400	68	295	46-1100
Garden[a]	0-15	8	980	270-1830	13	365	119-530
Grasslands[b]	0-15	5	1184	660-1575	6	224	141-285
Grasslands[b]	30-45	5	1289	555-2695	6	82	36-193

[a] Of homes.
[b] In and around towns.

TABLE XXII

BLOOD AND FECAL RESULTS

	Spring Blood lead, µg/100 mℓ Child	Spring Mother	Feces, µg/sample	Summer Blood lead, µg/100 mℓ Child	Summer Mother	Feces, µg/sample	Surface soil, µg/g dry soil
Matlock							
All	20.1 (47)[a]	17.0 (47)	67.8 (47)	24.7 (28)	21.0 (24)	65.8 (27)	909 (47)
No or past pica	19.0 (19)	17.0 (19)	75.6 (19)	24.2 (13)	22.9 (11)	73.1 (12)	904 (19)
Present pica .	20.9 (28)	16.9 (28)	63.0 (28)	25.2 (15)	19.5 (13)	60.4 (15)	912 (28)
Present pica for soil	20.4 (14)	15.3 (14)	64.0 (14)	25.2 (8)	18.4 (7)	54.0 (8)	709 (14)
Buxton							
All	22.1 (69)	18.1 (70)	69.4 (68)	28.1 (36)	22.6 (34)	67.8 (39)	398 (72)
No or past pica	21.5 (15)	16.8 (15)	60.2 (15)	22.5 (7)	19.9 (7)	56.6 (7)	380 (15)
Present pica	22.2 (54)	18.5 (55)	72.3 (53)	29.8 (29)	23.4 (27)	70.5 (32)	403 (57)
Present pica for soil	22.6 (37)	18.9 (37)	81.0 (36)	31.5 (16)	22.4 (14)	78.0 (19)	406 (38)

[a] Number of samples.

Both children and mothers had higher blood lead concentrations in the summer than in the spring and the children, both with and without pica, had higher blood lead concentrations than their mothers. The definition of pica used in this study included those children who habitually put fingers or toys in their mouths. Of the 119 children in both towns, only 11 of the 51 classified as having pica were definitely known to have swallowed soil. No statistically significant correlations were found between the blood, fecal, or hair lead values and the soil lead levels in the immediate environment of the child. In a further experiment, for which preliminary results are available, they compared two other villages where the mean soil lead concentrations are about 10 000 ppm and 500 ppm. The higher levels are in villages near old mine workings with some of the houses built on old waste material. The preliminary results of this study are given in Table XXIII.

TABLE XXIII

1973 SURVEY: PRELIMINARY BLOOD AND HAIR RESULTS

	Geometric means[a]		
	Blood (μg Pb/100 mℓ)		Hair (μg Pb/g)
	Child	Mother	
High soil lead area			
All	25.0 (48)	18.0 (44)	12.8 (48)
No current pica	23.6 (27)	18.1 (26)	10.8 (27)
Present pica	26.8 (21)	17.6 (20)	15.8 (21)
Present pica for soil	26.4 (16)	17.5 (16)	21.1 (16)
Low soil lead area			
All	20.9 (34)	14.7 (30)	7.5 (34)
No current pica	19.9 (17)	14.6 (16)	5.7 (17)
Present pica	21.9 (17)	14.6 (15)	9.8 (17)
Present pica for soil	22.1 (16)	14.5 (14)	9.0 (16)

[a]Numbers of individuals in parentheses.

The blood values in the high soil lead area are higher than in the low soil lead area but are within the accepted normal range. Barltrop et al. concluded that the results to date suggest that local soil lead concentrations on the order of 10 000 ppm are without major significance and, on present evidence, the concern with regard to contaminated soils in cities is not well founded.

We have not attempted to provide a complete review of the literature on lead or, even, to completely review the papers used. Instead, we have concentrated on the question of the hazard posed, particularly to children, by living in an area where the soil is contaminated with a toxic agent. As can be seen, there are different opinions. It appears that those who have made measurements on uptake in children along with environmental measurements feel that this pathway is not of major importance while those who have only made environmental measurements, and deduce the intake from these data, feel that it is of importance.

In applying these data to plutonium, it must be remembered that there are major differences between lead and plutonium in its behavior in the environment and its uptake in the body. The absorption of lead from the G.I. tract in adults is on the order of 5-10% but for children can be considerably higher as is shown by studies with rats, Kostial et al.[107] using ^{203}Pb in rats 5-7 days old showed absorption (and retention for 80 h) of about 55%. Forbes and Reina[108] using carrier-free ^{212}Pb by gavage showed absorption and retention 8 h after administration of about 83% at 16 days of age dropping to about 15-16% at 32 days of age. These results would indicate that the greater susceptibility of children to lead poisoning as compared to adults is probably a function of the lower discrimination against lead as well as increased opportunity for intake.

There are two methods of assessing the hazard to children of a contaminant in the soil: (1) by measurements of the environmental conditions and extrapolating to intake by using assumed

or observed behavioral patterns, and (2) by measuring directly the resulting conditions in children actually living in a contaminated area. We will use both techniques in order to allow comparison.

It would appear that the household dust (or other dustfall) provides the chief pathway of concern since it appears to be elevated in concentration about a factor of 10 above the soils. (Note that this conclusion is open to some question since many of the measurements were made largely in areas where the soil contamination resulted from atmospheric deposition in the vicinity of smelters and heavy highway traffic. It is possible, therefore, that sizable atmospheric concentrations still existed which would have served to increase household dust concentrations.) We will assume, following Lepow et al.,[103] that the child places his hands in his mouth 10 times per day and ingests the full quantity of dirt which was removed by the label technique (11 mg). Thus the dust ingestion would be 110 mg/day. At a permissible intake of 2700 pCi/day (0.003% absorption), the house dust concentration could be about 24 000 pCi/g. Given the factor of 10 between the soil and house dust, the limit for soil would be about 2400 pCi/g. This calculation can be criticized on the grounds that such behavior only occurs for a short time in the life span and the accumulation during this period is small. However, if we assume a 5-yr period of mouthing, the accumulation is 8% of the liver burden and 9% of the bone burden for an adult. In relation to the organ size of a child, these produce larger doses. Since the limit is high compared to other pathways, we have not tried to refine it further.

From the results of Baltrop et al.[105] in the two English villages, one can arrive at an approximate value by several assumptions on the measured uptake. If we assume that the uptake came only from the soils and the feces to be a reasonable measure of the total intake, then the children without pica in the village of Buxton with mean soil content of 400 µg/g had a daily intake of 60.2 µg/g in April and 56.6 µg in July. Those in

Buxton, where the soil concentration was about 900 µg/g had corresponding intakes of 75.6 µg and 73.1 µg. Thus, for a difference of 500 µg/g in the soil, the differences in intakes were 15.4 µg and 16.5 µg. These reduce to a daily intake of 0.03 and 0.033 of the quantity in one gram of soil. Again the permissible intake of 2700 pCi/day would result from about 90 nCi/g in the soil. It should be noted that the values used in this calculation were not significantly different so that one cannot be assured that at conditions where an actual significant difference occurs the soil value may not be even higher. An additional uncertainty occurs in the assumption that the fecal content represents the full intake since the absorption of lead by children is not well known but is believed to be much higher than adults. However, even if the absorption is 50%, one arrives at a value of 45 nCi/g. The uncertainties in this calculation are such that the result should be viewed with caution. However, it does illustrate a potentially fruitful field for future investigation.

In addition to casual ingestion by mouthing of hands and other objects, we must be concerned with the deliberate ingestion of nonfood items, in this case soil, by a subgroup of children. The condition of craving such materials to the extent of going out of the way (often far out of the way), to obtain the nonfood item is called "pica." In many studies it includes any ingestion of foreign materials and even such acts as mouthing articles. However, unless it results in a health problem, such as the lead toxicity problems from ingesting chips of leaded paint or plaster, it often goes unnoticed. Robischon[109] notes several characteristics of pica upon which there is general agreement.

1. It is normal for a child to mouth and sometimes swallow nonedible substances in the first year and the beginning of the second year of life.

2. Although pica occurs in mentally defective children, it is found in the mentally normal as well.

3. Pica tends to be self-limiting in many instances. In these cases it has disappeared by the end of the preschool period.

4. Pica is commonly of a general nature with many types of substances ingested.

5. Pica is about evenly distributed between the sexes.

In general, pica which continues beyond the age of about two years is unusual and is regarded as abnormal. It is difficult to fully describe the incidence of pica in populations since the studies have generally been done with particular groups of individuals, often those known to have problems, and most frequently with children in run-down urban areas. Barltrop[110] indicates that soil and dirt consists of 23-44% of all nonfood objects ingested. Obviously it is difficult to estimate the quantity of soil ingested but at least one study reports the ingestion of "handfuls" of soil every day in a severe case.[111] Layman et al.[112] discuss another form of pica, the eating of clay, which is common practice in parts of the southeastern U.S., Indonesia, Australia, Africa, the West Indies, Central America, and some parts of South America. Different types of clay are eaten with the most popular being the red clay and the white clay. However, we have tended to discount this practice as a problem with plutonium because of the nonpermeable nature of clay banks and the consequent low probability of the contaminant penetrating to any depth into the clay. This does remain a potential problem under very unusual circumstances, however, and the health professional should be alert to this unusual condition.

There are numerous additional studies but none which give any quantitative information on the overall frequency or seriousness of the problem. Obviously, it could be, and in some circumstances is, a serious problem in modern society, particularly when the items ingested contain a high percentage of harmful substances, such as lead or bacteria, as from fecal matter. However, it is of interest that the studies of Barltrop[105] and Ter

Haar[104] quoted earlier do not indicate a serious problem with soil as a source among the children they studied.

To give some dimension to the problem, we will assume a child with a reasonably severe condition who eats 10 handfuls of soil per day with each handful containing 2 g of soil. The condition lasts over a period of 4 years from the age of one to five. During this period the plutonium will accumulate in the bone but will approach only a small fraction of equilibrium. According to the ICRP,[113] the skeletal weight of a 5-year-old child is about 2.4 kg so that a dose of 0.5 rems per year would correspond to a burden of 250 pCi. Calculations indicate the skeleton to be more critical than the liver with an intake over the period of some 13 000 pCi/day required to reach this level at the end of 4 years. This calculation assumes adult parameters but the difference between children and adults are within the uncertainty of the other estimates. For an intake of 20 g/day over this period, the limit would be on the order of 650 pCi/g. The total body burden would be 250 pCi, which, without considering elimination, would mean that the dose from this source would be only 0.15-0.2 rems/yr for the remainder of his life.

VIII. DISCUSSION

A. A Reasonable Limit

A listing of the limiting values for the individual pathways is given in Table XXIV. It must be recognized that the relative importance of each of these pathways will differ depending upon the particular area, its climate and habitation. In addition, the numerical values can have real meaning only when the source of the information and its use in deriving the value is understood. In the next part of this section we will attempt to indicate primary uncertainties, and where possible, degrees of conservatism or nonconservatism in relation to the chosen value so that the reader may better understand these important factors.

TABLE XXIV

SUMMARY OF LIMITING VALUES

Pathway	Soil Conc (pCi/g)
Wind Resuspension	
Infinite area upwind	960-4700
1000 m square area	7600
Mechanical Resuspension	
1000 m square area	
Depth 1 cm	680
2.5 cm	270
5 cm	140
Local Resuspension	
Depth 1 cm	750
2.5 cm	300
5 cm	150
Transfer to objects or clothing	160-500
Ingestion	
Foodstuffs - All grown on area	260-390
Foodstuffs - Home gardener	1100
Casual	2400
Deliberate (pica)	650

A useful limit must consider the following factors:

1) A numerical limitation;

2) the area over which it applies;

3) the depth in the soil to be considered.

These factors are, to a large extent, interdependent and can vary with the relative importance of the individual pathways in a given area. However, numerical values can be chosen for several pathways to be dependent on the depth chosen for sampling as is shown by the variation described for resuspension. For

others, the depth is more indeterminate and depends upon the availability of the soil to the pathway. For agricultural crops one would consider the zone of cultivation to be appropriate. However, for pasture land it would be the depth of the roots of the forage crops. For casual or deliberate ingestion, the depth would be that available for the pathway under the ambient conditions.

In arriving at a single number to represent a limit of concern we have considered the values for individual pathways in Table XXIV along with a qualitative estimate of the probability of several pathways combining to increase the dose to the individual. This was not done on a formal basis, since as was noted above, this probability could vary depending upon the area use and habitation. However, a numerical value of 100 pCi/g to a depth of 5 cm would appear to provide an adequate margin for the multiple pathway exposing the infrequent maximum individual in the region. We make no specific recommendation for deeper contamination although there is good reason to believe that the limit should be higher than above because of the low probability of exposure of people and the dilution that will occur when the material is exposed either by natural or artificial means. A study on appropriate limits for burial grounds currently in progress will shed some light on this question.

The appropriate area again differs with the pathway considered. Wind resuspension requires very large areas to allow the concentration to reach the values given. These are on the order of 1 km^2 (250 acres) or greater. Smaller areas will make the wind resuspension less critical. Mechanical resuspension again requires a relatively large area primarily so that the very vigorous activities used as the basis will be carried out on the area year after year. Agriculture, in order to allow an area sufficient to provide food for the full year, requires a significant area. The limiting factor here would be a vegetarian maintaining a home garden or small farm and growing the large bulk of his food. The greatest limitation on an area would be

the direct ingestion and possibly the local resuspension, although it is unlikely that activities appropriate to the resuspension factors used would occur year after year in a very small area.

For these reasons, we have chosen an area over which the values can be averaged as equivalent to a play area or yard. While these can vary in size, an area of 100 m^2 or about 10 000 ft^2 was chosen as an appropriate averaging area. However, to assure that excessive exposure does not occur from single hot spots, it is further recommended that the maximum value in any single sample not exceed 1000 pCi/g. The problems of sampling to give reasonable assurance of meeting such a level will depend upon the form of the contamination (particulate or well dispersed). Simple techniques of direct alpha counting of soil samples usable in areas of moderate natural alpha activity in the soil are available to provide the ability to measure to this level in a large number of samples in a short time so that reasonable assurance is possible.[114]

At the outset we addressed the question of the advisability of preparing recommendations for a limit such as this because of the sparse knowledge of many of the factors involved and the variations possible depending upon the nature and the use of the individual site. The following discussion of factors of conservatism as well as the analyses indicate that a general limit, such as recommended herein, will indeed provide great conservatism for individual sites. We recommend, therefore, that this limit be used for situations where knowledge is insufficient to provide better analysis of the individual situation or where the cost of procuring such knowledge is excessive compared to the cost of remedy. In particular, we urge that decisions to destroy sensitive, unique, or important ecosystems be taken with considerable forethought and not be based solely on this limit.

B. Major Uncertainties and Degree of Conservatism

The uncertainties in the data are large, which is the reason for attempting to use reasonably conservative parameters. One of the largest uncertainties, which is difficult to assess, is in the wide variability of land use and actions on that land. We believe that it is obvious that levels in an uninhabited area with only occasional use, say forest land, could be considerably higher than we recommend. Our basis has been to limit the maximum individual for any of the uses to which the land may be placed.

The data on wind resuspension is only in the past few years coming to the point of allowing coherent patterns to be derived. We have attempted to provide such a pattern for the areas which have been studied but have not tried to extrapolate to unknown areas. Fortunately the studies that have been done are in areas where the problems with this pathway would be expected. The data on air concentrations around individual contaminated sites indicate that wind resuspension is probably a minor problem. Anspaugh[115] has come to a similar conclusion by use of the mass loading approach in the GMX area since his limiting value is on the order of nanocuries per gram.

The dust storm as a mechanism for producing large concentrations in some areas was not well evaluated in this study because of the evidence that indicates the particle sizes to be relatively large. If, however, one believes that the plutonium so suspended could have the metabolic transfer properties of the Class W in the ICRP models, then the particle size is unimportant as was shown earlier and, in areas where this can occur, this may be limiting. It must be remembered, however, that large areas are required for a dust storm to develop and the contribution of dust from small contaminated areas will be small compared to the total dust from the entire contributory area.

The use of the total air concentration rather than the respirable fraction in deriving resuspension rates could add a factor of 2 to 5 conservatism to this pathway based on usual

concepts. This will be discussed later in connection with the limits chosen. However, the value chosen as a limit based on other pathways is lower than the value we would choose for wind resuspension by an order of magnitude or so, so that uncertainties in these values are not important.

The data for mechanical resuspension are poor and the times chosen for their prevalence are arbitrary although we believe that the combination of resuspension rate and time spent per year would occur only in unusual circumstances. Again, the resuspension rate chosen was representative of a violent action and did not fully account for the mixing action of the first few years of the disturbance presumably resulting in lower concentrations in the future years. More study definitely needs doing for this pathway since, for the parameters used herein, it may be one of the most limiting.

We have chosen a value for the local resuspension factor based upon the upper end of the disturbance level. In theory, there is a series of disturbances, or actions each of which can be characterized by resuspension factors in a given soil condition and by a time in which they occur. The proper value to use, then, would be a summation of the product of these. Until such values are available for the conditions appropriate to this study we cannot estimate any degree of conservatism or nonconservatism although we believe that the analysis given here represents a reasonable worst case. A factor in applying the result is, again, the size of the area contaminated since this will have a bearing on the time that a disturbance can occur in the area.

For the transfers to the person or the home we have used an earlier analysis since there appears to have been little, if any, study of the factors defining this important pathway in the intervening years. We have, however, added a factor of 10 between dust concentration in the home and surrounding soil concentration. This may well be a factor of conservatism since it arose from the lead data and not all studies showed this

factor to exist. It was also noted that in many of the studies the source of the lead was from automotive traffic which results in the lead being released to the atmosphere. Under these conditions it could adsorb directly on the particulates in the air resulting in high concentrations in airborne particles and dust. That is, under these conditions there may be some doubt as to whether the dust originated from the soil. In other studies, it was noted that the lead in the dust could have originated from crumbling paint in the home.

The ingestion, according to this analysis, provides a surprisingly strong component to the accumulation. For foods, this is due to the use of a reasonable uptake factor in plants and animals, and the use of an uptake from the G.I. tract of the human of a value of 10 times the one normally used. We believe these to be appropriate considering the unknown behavior of plutonium in many soil types with time of residence and the unknown behavior of plutonium that has been metabolized in a plant or animal or processed with the preservatives and condiments now used. The values chosen are for a situation where all of the food for 70 years is grown in a contaminated area. While not an impossible situation it is unusual in this day of mass marketing of foodstuffs.

The pica question arises from somewhat unusual circumstances but not so unusual that it can be ignored. The history of lead poisoning from ingestion of paint chips or plaster indicates the potential problem. The choice of 20 g of soil as a daily quantity for ingestion appears reasonable for a relatively bad case but it could be exceeded in a very bad case. The absorption from the G.I. tract was taken to be the same as for a nitrate solution. Certainly for oxide particles the uptake would be lower by one or two orders of magnitude. The uptake from plutonium adsorbed on soil has not been determined but could, perhaps, be lower than the nitrate.

We have used the NCRP recommendation of 0.5 rems/yr to the most exposed organ of the exposed individual, in our case the

skeleton. It may also be noted that the 0.5 rems/yr was calcu-
lated to occur at the end of the 70-yr occupancy time so that
the average dose over this period would be lower. We have also
based our calculations on a contamination spread uniformly over
the area. In actual situations there will be wide variability
so that control to the limit stated will usually result in av-
erages for the area lower than the estimated limit. One of the
more conservative assumptions is that the individual occupies
this area for 70 yr without leaving.

The conversion of the 0.5 rems/yr to the organ to the quan-
tity inhaled was based upon the ICRP lung model as modified by
the Task Group of Committee II on the metabolism of plutonium.
This modification adds considerable emphasis to the absorption
from the upper respiratory tract. The value chosen for intake
was based largely on the more soluble plutonium (Class W), which
we do not know will exist in soils.

Currently, there is a movement toward reducing radiation
limits on ad hoc basis using the principle of "minimum prac-
ticable." Thus, the EPA[116] proposed limits for the nuclear fuel
cycle of 25 millirems per year to all organs except the thyroid,
skin, or cornea. For an individual at age 70, our limit was
based on an exposure level 20 times that proposed by the EPA.
We have attempted to base our limit on prevention of damage to
health and not on the minimum practicable. We feel that such a
limit is required before minimum practicable decisions are made
since it sets an upper limit. We would also urge that the pres-
ent limit not be converted to a lower dose level by simple ra-
tioing. Instead, a new analysis should be made properly fac-
toring in the degrees of conservatism required for such an as-
sessment.

The primary basis in this derivation is the dose to the max-
imum individual which is a different basis than that used by
most modelers who prefer to attempt to derive the average dose
to a population or a segment of a population. It should be

clearly recognized that, although we have not attempted to derive an average dose to the population, it will be considerably lower than for the individual. The factor depends upon the relative importance of the various pathways in individual areas but is believed to be 1% or less of the maximum individual in many areas.

C. Sampling for "Respirable Fraction" in Soil

In our previous report on this subject,[40] we introduced the concept of the "very surface layer" and recommended limitations based upon the smaller particle sizes. This was done in an attempt to apply the few experiments available to determine a resuspension rate. Recently, as a result of the finding that the plutonium concentration in the small soil particles at the ground surface in the vicinity of Rocky Flats is higher than the concentration in the total soil,[72] there have been recommendations to lower the existing Colorado standard for developers by applying the numerical value to only the fraction smaller than 5 μm. This recommendation was not accompanied by any analysis showing an increase in the probability of intake. Instead, it appears to be tacitly assumed that if the plutonium was present on or in the small particles, they would be inhaled in proportion to this concentration rather than that in the total soil.

We have not chosen such an approach but have based our proposed limit on the total concentration in the soil although we did examine the question in several of the appropriate pathways. There are a number of reasons for the lack of acceptance. These are discussed briefly below.

1. The data available for the resuspension mechanisms are all based upon total content in the soil. If one assumes, as is implicit in the "respirable fraction" sampling technique, that only the smaller particles are resuspended, then these resuspension rates and resuspension factors must be recalculated based upon the fraction of the plutonium (or other tracer) in the smaller particles. This is done essentially by multiplying the

rate or factor by this fraction. When applied, the answer should be about the same since we are using a smaller factor to convert to air concentration with a higher soil value. In other words, if one is to make this change for the sampling procedure it should also be made for the initial data upon which the standard is based. Tables XIV and XV illustrate this change.

It is true that there can be differences in this factor in different soil types and it would be advisable to start building a data base as to possible effect by making more detailed measurements of the soil characteristics in areas where measurements are being made so that more complete understanding of the phenomenon can be obtained. However, the data currently available do not indicate such an approach to be warranted.

2. The resuspension by winds is a complex phenomenon. Data on erosion indicate that the particle size undergoing saltation is a controlling factor. Such factors as the aggregation of the smaller particles and adsorption on the larger particles tend to minimize the importance of the small particle fraction in becoming airborne. This is well illustrated by those soils having a high fraction of clays where the agglomeration forces are such that they harden into rock-like structures when dry. Only the fraction that is abraded or otherwise removed is effective. It is probable that there is a soil composition which is most subject to erosion and resuspension but this must be based on relative composition of various sizes of particles as well as humus content and moisture content. In other words, we cannot relate the presence of only one fraction to the probability of becoming airborne.

3. The use of a thin layer at the surface raises many questions as to its meaning. Such a sample is difficult to reproduce and represents only a temporary situation because of the ephemeral nature of such a surface layer in most areas.

4. The use of only the small particle fraction in the soil provides only one component of the total answer required. The smaller particles in the soil originated by the breakdown of

larger particles. We can expect this to happen for larger plutonium particles as well. Thus, even if we assume that the small particles are representative of the hazard, this will represent only present conditions and totally ignores potential future hazards.

5. The use of only the small particles and a surface sample is based on the assumption that inhalation is the only pathway. As we have seen, there are others, involving ingestion, that must be taken seriously. Thus, even if the "respirable fraction" at the surface is measured, additional samples to measure the total plutonium content are required. This will cause the number of samples and cost to be doubled.

It may be noted that the respirable fraction has not been neglected in this study. Instead we have incorporated it into the total soil limit. If the small fraction is to be used as a criterion for resuspension, then the limit should be increased by the ratio of the total soil to the small fraction.

D. Preliminary Application to ^{241}Pu and ^{241}Am

Plutonium-241 is a beta-emitting isotope of plutonium that occurs to the extent of only about 0.5% in weapons plutonium but will be considerably higher, perhaps 15-20% in plutonium from high burnup plutonium. While it has a half-life of only 13.2 yr, it decays to produce ^{241}Am, an alpha emitter with a half-life of 458 yr. The buildup of this daughter is such that a maximum is reached about 70 yr after the purification of the plutonium.

The low-energy betas from the ^{241}Pu add little to the alpha dose so that this material can be ignored. However, the alpha particles from the Am can produce additional doses to the lung, bone, and liver. Thus, any limit for ^{241}Pu must be related to a limit for the ^{241}Am that it produces. About 0.025 Ci of ^{241}Am will result from the decay of 1.0 Ci of ^{241}Pu.

The potential hazards from ^{241}Am have not been as well explored as have those of plutonium. The following summary is

taken from Durbin.[117] It has a long retention time in the body and the liver and bone are the main sites of deposition once it reaches the blood stream. Studies of the carcinogenecity are in progress at Utah but have not continued long enough to give any comparison with radium or plutonium. However, the deposition in the bone is similar to that of plutonium so that the dose-equivlent calculation for the absorbed material should be similar to that for plutonium. The absorption from the lung in compounds where the americium is not bound with plutonium may be somewhat higher than plutonium.

It is concluded that the calculations of permissible intake for plutonium by inhalation are reasonably indicative of americium. The possibly higher absorption from the lung may, in fact, indicate that ICRP model used may apply better to americium than to plutonium. We would, therefore, conclude that the inhalation pathways, as calculated for plutonium, are appropriate for americium.

However, for the ingestion pathway there is a different story. While data are considerably less for americium than for plutonium, there are sufficient available to indicate greater plant uptake and greater absorption from the gut. In a critical review of data available, Thomas et al.[118] found values of the concentration factor for dry plant tissue ranging from about 10^{-5} in the grain of wheat to about 0.2 in the leaves of bush beans. An average of 12 measurements indicated a concentration factor of 0.07. However, the values were obtained from widely differing experimental conditions ranging from layering of the contamination in the pot to use of vermiculite to improve drainage to uniform contamination in the soil. It is difficult to assess a reasonable uptake value from these data because of the different experimental conditions, use of different soils, and use of different parts of the plant for analysis with only a few measurements on the portion used for food. We will use, as a preliminary basis until more returns are in, a value of 10^{-3} for

the concentration factor on a wet weight basis. This corresponds to a value of perhaps 5×10^{-3} to 10^{-2} on a dry weight basis.

The uptake from the G.I. tract also appears to be higher for americium than for plutonium. Durbin[117] indicates the best estimate of the uptake of trivalent actinides to be 10^{-4} to 6×10^{-4}. Thompson[119] provided a conservative estimate of 10^{-3}. For our purposes a value of 5×10^{-4} to 10^{-3} appears appropriate without being overly conservative. We will use 10^{-3} for estimates that will give some allowance for direct accumulation on plant surfaces.

There is one other factor that will influence the potential dose from ^{241}Am in the environs. This is its greater mobility in soils that may make its long-term behavior in the environment different from plutonium. Martell[120] has noted that this greater mobility may result in increased movement away from the root zone of plants, thereby partially offsetting the increased uptake.

In estimating a limit for ingestion we have not increased the uptake from the G.I. tract from food as was done for plutonium because of the uncertainty in value and the conservative value we have used for the uptake.

On the assumption that americium taken up into the body is distributed similarly to plutonium, the increased uptake will limit the daily intake, either for foods or pica, to 80 pCi/day. Assuming the uptake to the meat is increased in the same proportion as the increased uptake in the human G.I. tract, the food limits for the diet of Table XXIV become 70 pCi/g while a vegetarian living completely off the land would limit to 50 pCi/g. The home gardener limit would be about 200 pCi/g. The pica case, calculated for the ages of 1 to 5 in a manner similar to that used for plutonium but with an uptake of 10^{-3} leads to a limit of about 20 pCi/day.

We will accept the 20 pCi/g as a recommended limit based upon the possible occurrence of pica in children and to provide

some factor of safety for the uncertain values used in the up-take calculation. However, it should be noted that the pathways for this isotope are limited with inhalation values about the same as plutonium. In this case, the average dose to the population will be a much smaller fraction of the maximum dose than for plutonium.

In application to ^{241}Pu, the limit for this beta emitter could be increased by a factor of 40 to allow for the decreased quantity of ^{241}Am remaining after decay. This would place the ^{241}Pu limit at 800 pCi/g.

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