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September 18, 1984

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Dr. William Bair & Roy Thompson Pacific Northwest Laboratories Batelle Avenue Richland, Wa. 99352

Dear Roy,

Enclosed is the report on the resuspension study at Bikini. You will note that Table 7 on page 19 of this report is the table that was included in the Bikini dose assessment report UCRL-53225. The problem with the units in Table 1, page 8 of UCRL-53225 is that the micro (μ) symbol was omitted from the units of both the dust aerosol column and the soil Pu activity column. Thus, the units for the dust aerosol should be μ g/m³ not g/m³. Similarly, the units for the soil Pu activity should be aCi/ μ g (which is proportional to pCi/g) and not aCi/g. This was done to make a direct calculation possible for pulmonary deposition in aCi/h; sorry for the confusion.

I have also enclosed (Enclosure 1) a comparison of the dose methodology for bone marrow and endosteal cells for surface deposited transuranics for the Spiers and ICRP models. The values we use for the parameters in the lung model are those listed in ICRP 30, Part 3, and attached as enclosure 2.

Sincerely,

William L. Robison Section Leader Terrestrial & Atmospheric Sciences

WLR/mt enclosures

Enclosure 1 Outline

- 1. Ratio of Bone Marrow to Endosteal Cell Dose
 - A. Spiers
 - B. ICRP
 - C. ORNL
 - D. UNSCEAR
 - E. Comparison of endosteal cell to red marrow dose ratio for the various methods.
- 2. Absolute Magnitude of the Bone Marrow Dose Conversion
 - A. Spiers
 - B. ICRP
 - C. ORNL
 - D. UNSCEAR
- 3. Overall Comparison
- 4. Our Method of Calculating the Marrow and Surface Cell Dose for Surface Deposited Pu.
- 5. Areas of Difference Between Methods
- 6. References

Enclosure 1

Comparison of Spiers and ICRP Models for Dose from Pu to Red Marrow and Endosteal Cells

1. Ratio of Bone Marrow to Endosteal Cell Dose

A. Spiers (References 1,2,3).

In Spiers method the dose is calculated to a tissue filled cavity and is referred to as Do. The dose in bone, D_B , is then related to D_0 by the ratio of the mass stopping powers of bone and tissue.

Thus D₀ S_B/S_T

From geometry consideration and deposition and energy loss characteristic for alpha emitters, the doses to red marrow, D_M , and endosteal cells, D_S , may be calculated by applying appropriate factors to D_B . The factors for 239 Pu are:

$$D_{M} = 0.26 D_{B}$$

 $D_{S} = 3.11 D_{B}$

Thus, the ratio between the red marrow dose and endosteal cell dose is: $\frac{3.11}{0.26} = \sim 12$.

B. ICRP (ICRP 30, Supplement to Part 1)

From ICRP-30, Supplement to Part 1 page 414 and 415, are listed the committed Dose Equivalent (C.D.E.) in target organs or tissues per intake of unit activity. The ratio of these C.D.E. for bone surface cells and bone marrow provides the necessary comparison for the ICRP model to Spiers model. Thus, for $f_1 = 1.0 \times 10^{-4}$, the red marrow CDE is 1.6 x 10^{-7} Sv/Bq and the bone surface CDE is 2.1 x 10^{-6} Sv/Ba. The ratio between the red marrow dose and endosteal cell dose is: $\frac{2.1 \times 10^{-6}}{1.6 \times 10^{-7}} = 13$

C. ORNL

From NUREG/CR - 0150 Vol. 2, 1979 Estimates of Internal Dose Equivalent to 22 Target Organs for Radionuclides Occurring in Routine Releases from Nuclear Fuel-Cycle Facilities, Vol.11; D.E. Dunning, Jr., S.R. Bernard, P.J. Walsh, G.G Killough, and J.C. Pleasant.

This analysis is essentially that of ICRP. It is based on the same data from Marshall, Thorne, ICRP etc. In this report they give "S-Factors" for various target organs in rem/µCi-d. For a quality factor of 20 the "S Factor" for red marrow is 0.90 rem/µCi-d and for endosteal cells in 12.58 rem/µCi-d. The ratio of endosteal cells to red marrow is, therefore, $\frac{12.58}{0.90} = 14$.

D. UNSCEAR

In the 1982 UNSCEAR report the committed dose per unit intake of Pu is listed as 0.008 μ Gy/Bq for red marrow and 0.1 μ Gy/Bq for bone living cells. Thus the ratio of endosteal cells to red marrow is $\frac{0.1}{0.008} = 12.5$

E. Comparison of the endosteal cell to red marrow dose ratio for the various methods.

The ratio of bone surface to bone marrow dose from Spiers is 12, from ICRP 13, from ORNL 14 and from UNCSEAR 12.5. Thus, from these comparisons, there is very little difference in the ratio of endosteal cell to bone marrow dose for the various options one could select for the calculation.

2. Absolute Magnitude of the Bone Marrow Dose Conversion.

A. Spiers

Spiers lists the dose conversion for <u>bone marrow</u> in reference 1 as 16.9 rad/y- μ Ci for a 1 μ Ci skeletal content (mineralized skeleton 5000g; endosteal surface 16m²). This converts to 338 rem/ μ Ci-y for a QF = 20.

equivalent (CDE) per unit of activity intake for bone marrow is given as 1.6×10^{-7} Sv/Bq for a gut transfer of 10^{-4} . Based on these numbers the equivalent dose rate is 315 rem/µCi-y.

C. ORNL

The value for bone marrow for a QF=20 of 0.90 rem/ μ Ci-d is equivalent to 329 rem/ μ Ci-y.

D. UNSCEAR 1982

The UNSCEAR listed value of 0.008 μ Gy/Bq is identical to the ICRP value of 1.6 x 10⁷ Sv/Bq which I have already shown is equivalent to 315 rem/ μ Ci-y.

3. Overall Comparison

Spiers dose conversion for bone marrow of 338 rem/ μ Ci-y is 7% higher than ICRP value of 315 rem/ μ Ci-y. When these values for Spiers and ICRP are multiplied by the endosteal to marrow ratio of 12 and 13 respectively the resulting endosteal dose for ICRP numbers is 1% higher than that of Spiers.

Thus, the two methods appear to be quite comparable for estimating the bone marrow and endosteal cell dose for Pu.

4. Our Method of Calculating the Marrow and Surface Cell Dose for Surface Deposited Pu.

Starting with a pCi/d intake derived from the Pu concentration in foods times an average dietary intake for each food item, we calculate, using a gut transfer factor of 10^{-4} and 45% deposition in bone, the pCi/g of Pu in bone. This number is then multiplied by 338 rem/uCi-y to develop the bone marrow dose. The endosteal cell dose is then calculated by multiplying the bone marrow dose by 12, the ratio of endosteal to marrow dose.

could reflect a difference in the distribution of the endosteal mass between trabecular and cortical bone. Spiers was the absorbed fraction for Pu rather than an average value for alpha emitters as used by ICRP and this would make some difference. Spiers does make the point that cortical endosteal tissue may not be at as high a risk per unit dose as the trabecular endosteal tissue.

I think that the dose rates Spiers lists in reference 1 of 129 rad/ μ Ci-y for trabecular endosteal cells and 193 rad/ μ Ci-y for cortical endosteal cells (compared with 16.9 rad/ μ Ci-y for bone marrow) are wrong. Based on these numbers, the trabecular endosteal cell ratio to bone marrow is 7.6 while that for cortical endosteal cells is 11.4. This is inconsistent with his published dose factor ratios of 0.26 for marrow, 3.11 for trabecular surface cells and 4.7 for cortical surface cells which give ratios of surface cells to marrow of 12 and 18 respectively.

However, as I mentioned before, we use a dose conversion of 338 rem/uCi-y to develop the bone marrow dose and multiply by a factor of 12 to get the surface cell dose.

6. References

- 1.) F.W. Spiers and Janet Vaughn, "Hazards of plutonium with special reference to the skeleton." Nature Vol. 259, 531-534 (1976).
- 2.) F.W. Spiers, J.R. Whitwell, and A.H. Beddoe, "Calculated dose factors for the radiosensitive tissues in bone irradiated by surface deposited radionuclides." Phys. Med. Biol. Vol. 223, 481-494 (1978).
- 3.) F.W. Spiers and J.R. Whitwell, "Dosimetry of ²³⁹Pu and ²²⁶Ra in man and animals" in The Health Effects of Plutonium and Radium, ed. by Webster Jee, J.W. Press, Salt Lake City, Utah (1976).



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·	×)	v	V	Y	
Region	Compart- ment	T day	F	T day	F	T day	F
$N-P$ $(D_{N-P} = 0.30)$	a b	0.01 0.01	0.5 0.5	0.01 0.40	0.1 0.9	0.01 0.40	0.01 0.99
$\begin{array}{l} \mathbf{T} - \mathbf{B} \\ (D_{\mathbf{T} - \mathbf{B}} = 0.08) \end{array}$	c d	0.01 0.2	0.95 0.05	0.01 0.2	0.5 0.5	0.01 0.2	0.01 0.99
$P_{D_{\rm P}} = 0.25$	e f g	0.5 n.a. n.a.	0.8 n.a. n.a.	50 1.0 50	0.15 0.4 0.4	500 1.0 500	0.05 0.4 0.4
U_p = 0.257	h	0.5	0.2	50	0.05	500	0.15
-	i j	0.5 n.a.	1.0 п.а.	50 n.a.	1.0 n.a.	1 000 ∞	0.9 0.1

Fig. 5.2. Mathematical model used to describe clearance from the respiratory system. The values for the removal half-times, T_{s-i} and compartmental fractions, F_{s-i} are given in the tabular portion of the figure for each of the three classes of retained materials. The values given for D_{N-P} , D_{T-B} and D_P (left column) are the regional depositions for an aerosol with an AMAD of 1 μ m. The schematic drawing identifies the various clearance pathways from compartments a-i in the four respiratory regions, N-P, T-B, P and L. n.a. = not applicable.

Page 76 of Part 1 and pages 54 and 55 of its Supplement: As a consequence of the Commission's decision to reduce its recommended dose equivalent limit for the lens of the eye from 0.3 Sv to 0.15 Sv in a year, values of DAC for 81 Kr and 83m Kr are amended as follows.

Radionuclide	Semi-infinite cloud	1 000 m ³ room	500 m ³ room	100 m ³ room
⁸¹ Kr	2×10^{7}	1×10^{8} (5 × 10 ⁸)	1×10^8 (6 × 10 ⁸)	1×10^8 (9 × 10 ⁸)
^{8 3m} Kr	4×10^{8} (7 × 10 ⁹)	Lens 4×10^8 (7×10^9)	Lens 4×10^8 (7×10^9)	Lens 4×10^{8} (8×10^{9})
	Lens	Lens	Lens	Lens

aerosol which is namic Diameter with AMADs intates of depowith an AMAD to aerosols with

Page 78, The values of ALI and DAC for ⁸⁰Sr are incorrect because no allowance was made for the contribution to committed dose equivalent of ⁸⁰Rb, the daughter of ⁸⁰Sr. The corrected dosimetric data are given in Supplement B of Part 3. The correct values of ALI and DAC are given below:

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opinions and conclusions stated are those of the author and may or may not be those of the Laboratory.

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RESUSPENSION STUDIES AT BIKINI ATOLL

Joseph H. Shinn, Donald N. Homan, and William L. Robison Environmental Sciences Division Lawrence Livermore Laboratory Livermore, California

ABSTRACT

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The following experiments were conducted on Bikini Atoll to provide key parameters for an assessment of inhalation exposure from plutonium-contaminated dust aerosols: (1) a characterization of background (plutonium activity, dust, plutonium, sea spray, and organic aerosol concentrations), (2) a study of plutonium resuspension from a bare field, (3) a study of plutonium resuspension by traffic, and (4) a study of personal inhalation exposure. Studies similar to (1) and (2) have been previously performed at Enewetak Atoll.

Dust concentrations of 21 μ g m⁻³ and sea spray of 34 μ g m⁻³ were the background throughout Bikini Island except within 50 m of the windward beach. Background concentrations of ²³⁹⁺²⁴⁰Pu were 60 aCi m⁻³ in the coconut grove and 264 aCi m⁻³ over rain-stabilized bare soil. The ratio of plutonium activity in aerosols (pCi q^{-1}) relative to the activity in underlying soil (pCi q^{-1}), defined as the enhancement factor, EF, was typically less than one apparently due to dilution by non-contaminated soil aerosols at a ratio of 1.8:1. Enhancement factors increased about 3.8 as a result of tilling on both Bikini and Enewetak. Plutonium resuspension flux was estimated at 0.49 pCi m⁻² year⁻¹ over most of Bikini Island. Aerosol size distributions associated with mass and with plutonium activity were typically log-normal with median aerodynamic diameter 2.44 µm, which decreased to 2.0 µm above freshly tilled soil. The Pu concentration in aerosols collected over disturbed soil increased by a factor of 19.1 due to increased dust aerosol concentrations and increased enhancement factors. Vehicular traffic produced dust pulses typically of 10 s duration, 28 μ g m⁻³ average concentration, and plutonium enhancement factor 2.5. Personal dosimetry showed that enhancement of dust by a worker stirring his own immediate environment was a factor of 2.64 when doing heavy work outdoors near the ground and 1.86 when doing light work in and around houses.

Pulmonary deposition of plutonium was calculated for various exposure conditions as determined by inhalation rate, aerosol dust concentration, plutonium activity in soil, plutonium enhancement factor, personal dosimeter enhancement factor and the pulmonary respirable-fraction. The pulmonary deposition ranged from 1476 aCi h^{-1} to 12 aCi h^{-1} with intermediate values for heavy outdoor work (139 aCi h^{-1}) and for light work in and around houses (78 aCi h^{-1}) plus a value of 1.58 aCi h^{-1} at roadside when passed by a typical vehicle once per hour.

INTRODUCTION

A study of inhalation exposure from plutonium-contaminated soil was conducted to provide the parameters for a rigorous assessment of the inhalation dose to man. The study was needed because in previous dose assessments only a minimum amount of information was available about total mass-loading of suspended aerosol $(\mu g/m^3)$,

aerosol size, and total radioactivity per unit mass. Although necessary, these parameters are insufficient to describe the fraction of suspended radioactivity within the respirable size range and how either the concentration of radioactivity or the respirable fraction vary with surface conditions and local resuspending (dust-lifting) mechanisms.

Our investigations recently conducted on Bikini and Enewetak Atolls in the Marshall Islands provided new data which have implications not only for the local dose assessment concerning rehabitation of those sites, but which are important for understanding low-level inhalation exposure to toxic radionuclides in general.

BACKGROUND

This study conducted on Bikini Island in May 1978 provided a more complete set of data, following our preliminary studies on Engebi Island of Enewetak Atoll in February 1977. The Bikini Island study utilized extensive soil sampling and in situ gamma spectroscopy to determine isotope concentrations in soil and vegetation. Also various air sampling devices were used to determine particle size distribution and mass loading, and micrometeorological techniques were used to determine aerosol fluxes. Subsequent wet chemistry analysis provided radionuclide and elemental concentrations in collected aerosols. Four simultaneous experiments were conducted: (1) a characterization of the normal (background) suspended aerosols and the contributions from sea spray off the windward beach leeward across the island, (2) a study of resuspension of radionuclides from a field purposely laid bare by bulldozers as a worst-case condition, (3) a study of resuspension of radioactive particles by vehicular and foot traffic, and (4) a study of personal inhalation exposure using small dosimeters carried by volunteers during their daily routines. Less complete studies similar to (1) and (2) had been performed previously on Engebi Island at Enewetak Atoll and background studies similar to (1) were performed later on Eneu Island at Bikini Atoll.

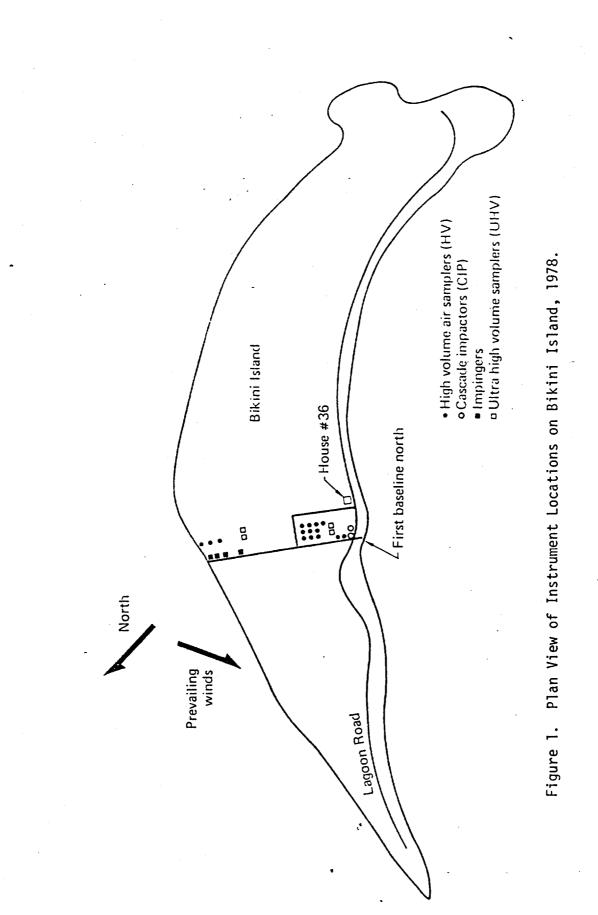
METHODS

Soil and vegetation samples were collected for analysis of radionuclide concentrations. 238 Pu, $^{239+240}$ Pu, and 241 Am concentrations were determined by isotope dilution and alpha spectrometry and 90 Sr concentrations by yttrium-90 separation and beta counting. These analyses were performed by LFE Corporation. Gamma spectroscopy using Ge(Li) diode detector was used to determine 137 Cs concentrations. Also, because the ratios $(^{241}$ Am)/ $(^{239+240}$ Pu) and $(^{238}$ Pu)/ $(^{239+240}$ Pu) are constant on Bikini, it was possible to estimate plutonium soil concentrations by measuring 241 Am soil concentrations using a gamma spectroscopy system consisting of a planar, high-purity germanium diode which was cryogenically cooled to achieve a minimum detectability for 241 Am less than 1.0 pCi g⁻¹ (1). The detector was mounted facing downward on a tripod so that the volume of soil integrated was contained in a circle of probable detection of nominally 3 m radius and 5 cm depth. Because the nuclear events causing the original contamination of Bikini Island were far removed, the fallout was relatively evenly dispersed across the Island.

Impingers were used to collect soluble sea spray aerosols in a 250 ml distilled water trap similar to the method of Hsu and Whelan (2). Air flow rates were $0.36 \text{ m}^3 \text{ h}^{-1}$ (6 l min⁻¹) through the water trap and measured amounts of water were added each day to replace evaporated water (nominally 40 cm³ day⁻¹). Impingers

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It was found that blank memorane fifters inexpircably gained weight with time but that standard deviations within 10% of mass could be achieved where filter blanks from the same lot were monitored for weight gains over the time period of the experiment. The membrane filters were used as a substrate for a scanning electron microscope (SEM) study of particle characteristics. The SEM operated by the LLL Particle Characterization Facility has a large chamber for specimens (90 mm) and has a resolution of 0.015 µm. In the SEM mount, microprobe chemistry of individual aerosol particles on the membrane filters was accomplished by X-ray fluorescence with a resolution of 160 eV, which provided quantitation of particles containing elements with atomic numbers equal or greater than sodium.



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	high-tide waterline (Table 1). The leeward decrease was verified by HV measurements

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measurements show that the background sea spray aerosol calculated from Na and Mg concentrations was remarkably uniform throughout the remainder of the island $(\bar{x} = 34 \ \mu g \ m^{-3}, S = 8.7, n = 27)$. The HV results are summarized in Table 2.

Background is here defined as the aerosol concentrations at the 1.1 m height over surfaces which are relatively stabilized and under normal wind conditions. After a week, even the bare soil tended to reach the same average level of dust aerosol concentrations $(21 \ \mu g \ m^{-3})$ as the coconut grove (Table 2). An analysis of the personal dosimeter data (discussed later) showed that about 10% of the background dust aerosol was organic. $^{239+240}$ Pu concentration (aCi/m³) was a factor of 258/60 = 4.3 greater over bare soil than in the coconut grove. (Soil activity was 15.3 and 8.01 pCi/g respectively, which is not significantly different within the normal variation encountered.)

If we examine the vertical fluxes of plutonium (aCi $m^{-2} s^{-1}$), the ratio of fluxes from the two sites will be proportional to the ratio of their wind friction velocities, u_* , where:

 $u_{\star} = C_{D} U_{1}$ [1]

and CD is a drag coefficient equal to 0.106 in the coconut grove and 0.077 in the bare field as determined by our wind profile measurements and U_1 is the wind speed at the 1 m height, which was 4.1 x greater for the bare field than the coconut grove. By Equation 1, the ratio of friction velocities is 3 x greater in the bare field than in the coconut grove. The ratio of their plutonium fluxes is also proportional to the ratio of their concentrations; hence, the plutonium flux is a factor of 4.3 x 3 = 12.9 greater in the stabilized bare field than in

DATE 1978	DISTANCE TO MINDWARD SHORE (m)	GROUND COVER	SEA SPRAY (µg m ⁻³)	DUST AEROSOL (µg m ⁻³)	PLUTONIUN CONCENTRATION (aCi m ⁻³)*	TYPE (number) OF INSTRUMAENTS	WINC SPEED D) (m s ⁻¹)	WIND ** SPEED DIRECTION (m s ⁻¹) (deg)
5/6-8	780	Bare Soil	1	167	7295	CIP (2)	4.7	53
5/9-16	730	Bare Soil	1	б	246	CIP (2)	4.6	43
5/6-8	600-700	Bare Soil	34	136	6466	(S) VH	4.7	53
5/10-11	600-700	Bare Soil	34	23	338	(01) VH	4.1	52
5/12-16	600-700	Bare Soil	35	18	189	HV (10)	4.6	33
5/8-16	20-160	Coconut Trees	34	21	65	HV (2)	4.6	-7- -7-
5/8-16	820	Road	33	41	421	(L) NH	4.6	45
5/8-16	IJ	Shrubs	40	8	29	(L) NH	4.6	45
5/10-11	370	Coconut Trees	1	3	51	(L) AHN	4.1	52
5/12-16	760	Bare Soil	1	1	212 -	UHV (2)	4.6	33
Background ***	* >50	Bare Soil	34	21	258	CIP(2),HV(20),UHV(2)	(2)	
Background ***	* >50	Coconut Trees	34	21	60	HV(2), UHV(1)		

** Wind measurements recorded for the 4.5 m height at a station in the open field.

*** Averaged over the surfaces that are stabilized.

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SUMMARY OF MASS AND PLUTONIUM AEROSOL CONCENTRATIONS ON BIKINI ISLAND

TABLE 2

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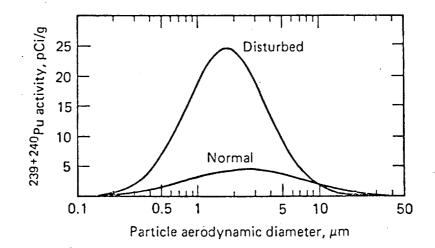
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(MAD) and geometric standard deviations (GSD) obtained by fitting cascade impactor data (Table 3). The aerosol size distributions for plutonium activity determined by CIP and the total mass loading (sea spray plus dust) determined by optical particle analyzer were satisfactorily approximated by a log normal distribution with the given GSD values in Table 3. All other MAD values of Table 3 were determined by cascade impactor, but more data would be required to determine if a log normal distribution exists for the other aerosol definitions.

Two typical cases of number density $(dN/d \ln D)$ and volume density distributions $(dV/d \ln D)$ determined by the optical particle analyzer over the stabilized bare soil surface on May 9 and May 11 are shown in Figure 3. It should be noted that the optical particle analyzer sees all liquid and solid aerosols including



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Figure 2. Plutonium activity (pCi g^{-1}) versus particle aerodynamic diameter D (µm) at the 1.1 m height over bare soil on Bikini Island (wind speed 4.5 m s⁻¹, surface contamination 15.3 pCi g^{-1}).

Median Aerodynamic Diameters (µm)	Disturbed Bare Soil	Stabilized Bare Soil
Pu Activity (pCi g ⁻¹)	1.73	2.46
Pu Concentration (aCi m ⁻³)	2.05	2.43
Mass Loading (µg m ⁻³)	2.03	2.46
Mass Loading-Optical ($\mu g m^{-3}$)		2.40 (.11)*
Sea Spray - Mg (µg m ⁻³)		2.59
Geometric Standard Deviation		
Pu Activity (pCi g ⁻¹)	2.16	3.09
Nass Loading - Optical (µg m ⁻³)	 .	2.82 (.25)*

TABLE 3.	Aerosol size characteristics on Bikini Island determined by cascade	
	impactors and the optical particle analyzer at a height of 1.1 m.	

* Optical particle analyzer data with standard deviations in parentheses.

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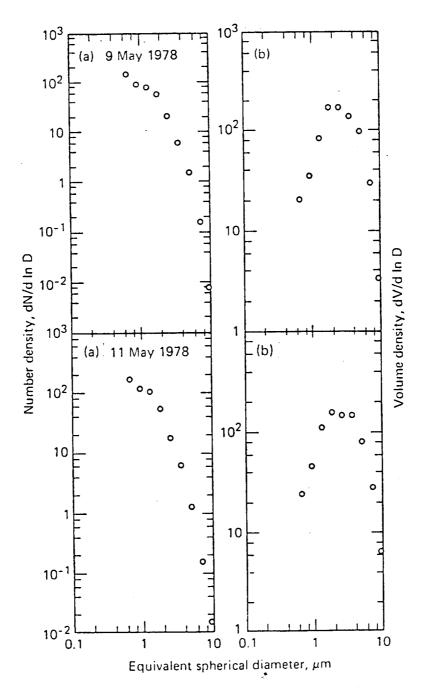
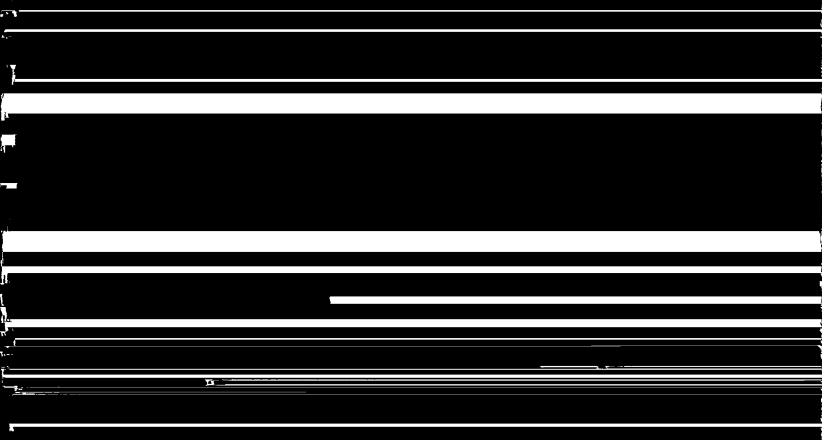


Figure 3. Typical particle size distributions over the stabilized bare soil surface. Number density (a) has units of particles cm⁻³; volume density (b) has units µm³ cm⁻³.

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ratio values should be greater than one, but gives no evidence (8). The ratio of organic particles to calcareous soil particles remained about constant (10%) as determined by X-ray fluorescence on the PD filters exposed at this site during the same period. We know from previous studies that one component of organic matter, plant leaves, had a ratio of 10^{-3} plutonium concentration relative to soil and could serve to dilute the inorganic aerosol.)

Site	Date	Soil Activity (pCi g ⁻¹)	Aerosol Activity (pCi g ⁻¹)	Enhancement Factor	
Disturbed bare soil	5/6-8	15.3	47.5	3.10	
Stabilized bare soil	5/10-11	15.3	14.7	0.96	
Stabilized bare soil	5/12-16	15.3	10.5	0.69	
Coconut grove	5/8-16	8.01	3.29	0.41	6
Road with traffic	5/8-16	4.10	10.3	2.5	

TABLE 4. Enhancement factors for plutonium activity of aerosols on Bikini Island (HV data).

Under dusty conditions, EF values exceed one such as in the cases of the disturbed bare soil (3.1) and the road with traffic (2.5). So there are two different factors producing increased plutonium aerosol concentrations (aCi m⁻³) during unusual resuspension. The aerosol dust concentration increases, but also the plutonium activity increases. For example, averaged over 10 HV instruments,

the ratio of plutonium concentration over bare soil on May 6-8 compared to May 10-11 is caused by a 5.91 x increase in dust aerosol concentrations (Table 2) and 3.23 x increase in enhancement factor (Table 4) for a combined effect on aerosol plutonium concentration (6466 aCi $m^{-3}/338$ aCi m^{-3}) of 19.1.

Resuspension of Radioactive Particles by Vehicular and Foot Traffic

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The integrating nephelometer was installed with intake at 1.2 m height and 2 m leeward from the position of average tire tracks on a frequently-traveled, one-lane dirt road on Bikini Island. Even though the traditional vehicular traffic of light trucks at low speeds was increased in frequency by our experimental activity, we were interested in characterizing the resuspension of plutonium and inhalation exposure per vehicle pass. The nephelometer provided details on magnitude, duration, and frequency of dust concentrations, while plutonium and dust aerosol concentrations (Table 2), and plutonium activity and enhancement factors (Table 4) were obtained by a co-located HV.

Dust concentrations above background rose in a pulse exceeding 10 s duration where the peak was obtained in a period about 4.5 s after the passage of the vehicle (Figure 4). This characteristic time to arrival of the peak, regardless of concentration, was determined by X/σ_{U} where the travel distance X is 2 m and the RMS turbulent velocity $\sigma_{\rm U}$ is about one-tenth the local wind speed of 4.5 m s⁻¹. Hence the dust pulse was traveling by diffusion and not characterized by translation in the wake of the passing vehicle. The dust pulse example of Figure 4 represents an extreme case (more than 90% of occurrences had lower concentrations), but demonstrates the characteristic peak to mean ratio of 3.6 and the slow return to background on the tail of the pulse. The amplitude and frequency of dust pulses due to motor vehicle, bicycle, and foot traffic were recorded during May 11-15. The sixty-eight cases of motor vehicle passes observed showed an approximate log-normal frequency distribution with median peak concentration (above background) of 100 μ g m⁻³ and geometric standard deviation of 3.4, Figure 5. Bicycle traffic could not be distinguished from foot traffic. In the seven observed cases of foot traffic, we found an approximate median peak concentration above background of 26 μ g m⁻³.

It should be emphasized that the log-normal concentration implies a fairly high chance (5%) of an exposure to a vehicular-induced peak concentration of 760 μ g m⁻³ having a mean concentration 760/3.6 = 211 μ g m⁻³ for about 10 seconds. The plutonium enhancement factor was estimated at 2.5 in this study (Table 4).

Personal Inhalation Exposure and Dosimetry

Until now, the discussion has centered on the (combined) isotope $^{239^+240}$ Pu, since in fact this is the most important component of inhalation exposure. Extensive soil sampling on Bikini Island has established that a relatively homogeneous mixture of isotopes exists in the soil (Table 5). In the aerosols, some of the isotopes become significantly enhanced (238 Pu and 137 Cs) but they remain of lesser inhalation-hazard. The <u>in situ</u> gamma (ISG) spectroscopy system which measured 241 Am in the soil at Bikini was highly correlated to that measured in surface soil samples by special chemistry methods ($r^2 = 0.910$), which gives confidence in both methods. However, data from the ISG system was consistently lower than the soil sampling method by a factor of 0.7 because it integrated a

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11:05 a.m., May 15, 1978 (wind speed 4.5 m s⁻¹).

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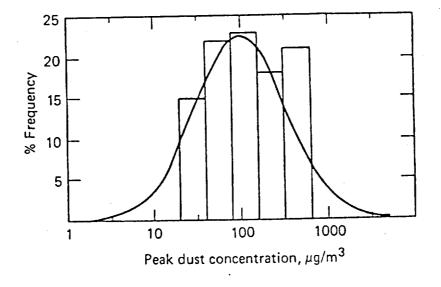


Figure 5. Frequency of peak dust concentrations on the downwind side of a one-lane dirt road following passage of light motor vehicle.

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	²³⁸ Pu	^{2 4 1} Am	¹³⁷ Cs	⁹⁰ Sr	239+240pu	
	239+240Pu	239+240pu	239+240Pu	239+240Pu	(pCi g ⁻¹)	
Soil	0.0013	0.556	9.80	9.15	15.3	
FSD*	0.56	0.55	0.48	0.52	0.57	٠
Aerosols (HV)	0.050	0.439	61.4	12.8	12.6	
- FSD*	0.64	0.34	0.32	0.51	0.35	

TABLE 5.	Radioactive isotope ratios in soil and aerosols at a stabilized bare	
	field on Bikini Island	_

Fractional Standard Deviations (s/\overline{x})

view volume about 5 cm depth and the exponential decrease of isotope concentration with depth gives lower mean values. After correction by a factor 1.44, the ISG method was the primary method for mapping 241 Am as a tracer for the source of suspended plutonium isotopes. The horizontal variations in soil isotope concentrations (ISG data) were small enough so that one could justify mean values as local regional values for soil. For example, on the one-hectare, bare field, it was determined that plutonium in surface soil had the mean value 15.3 pCi g⁻¹ with an observed range of 2.3 to 28, and a fractional standard deviation (s/\overline{x}) of 0.57. There was no apparent pattern to the soil concentrations and they exhibit approximately normal, random variation perhaps due to previous tilling and blading locally. (The data did not fit a log-normal distribution any better.)

In the context of this surface soil contamination, personal dosimeters (PD) provided information about inhalation exposure of individuals relative to the reference HV monitors. It was found that the fraction of dust in the total aerosol collected by the PD was greatest for workers exposed during heavy tilling but was also high for workers exposed in and around houses (Table 6) partly because of a lowered fraction of sea spray in both cases. In this and prior studies, we found that the ratio of PD Dust/HV Dust has a value of approximately 0.5 where both PD and HV are sampling the same aerosol cloud of this size particles (2.5 µm MAD) because of the cyclone particle-discriminator on the PD. Therefore, the enhancement of inhalation exposure by a worker's own actions where the PD and HV are <u>not</u> sampling the same cloud can be estimated by a <u>personal</u> dosimeter enhancement (PDE):

$$PDE \simeq 2x (PD Dust/HV Dust)$$
 [4]

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Values so computed show significant enhancement (PDE) of inhalation exposure (2.64) during heavy work outdoors by persons sitting or kneeling while digging or using tools on the ground (Table 6). The second highest enhancement (1.86) came from persons with duties in and around the houses. Other work, including

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Comparative Analysis of Personal Dosimeters (PD) Worn by Volunteers in Various Work Assignments

				Tarida and Autrida
Activity	Outdoors Over Disturbed Bare Field	Uutdoors Uver Vegetated	Outdoors Over stablilzed and Vegetated Surfaces	Around Houses
	During Tilling	Light Work	Heavy Work	Light Work
Number of Volunteers	2	4	3	e
PD Dust Fraction *	94%	56%	56%	89%
PD Dust (µg m ⁻³)	62	12	28	20
PD Dust/HV Dust	.46	.55	1.32	.93
PD Enhancement **	.92	1.10	2.64	1.86

Personal Dosimeter dust is corrected for sea spray but contains about 10% organic matter, both estimated by x-ray fluorescence. ¥

** PD Enhancement = 2x (PD Dust/HV Dust).

heavy tilling, produced inhalation exposures satisfactorily monitored by HV and thus, their PDE values were close to unity. The main limitation of the PD data and the derived enhancement values (PDE) is that no information is obtained about the plutonium enhancement factors expressed by Equation [3]. It should be recalled that plutonium enhancement factors (EF) of the same magnitude as these PDE were detected by HV (Table 4).

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Pulmonary deposition is the penetration and retention of respirable size particles into the deep, alveolar regions of the lung and constitutes the major vector of inhalation dose. The efficiency of pulmonary deposition varies with particle size and is conventionally estimated by the ICRP Task Group on Lung Dynamics' deposition model (9). For example, the observed change in size distribution of plutonium activity from the stabilized soil case to the disturbed soil case (Fig. 1), increased the calculated pulmonary respirable-fraction (RESP) from 19% to 24%. Pulmonary deposition (attocuries per hour) using an inhalation rate (IN. RATE) and previously defined terms may be estimated as follows:

DEPOSITION = IN. RATE x HV DUST x SOIL ACTIVITY x EF x PDE x RESP [5] (aCi h⁻¹) (m³ h⁻¹) (μ g m⁻³) (aCi μ g⁻¹)

Using Equation [5], data from Tables 2, 4, and 6, and the best estimates for the enhancement factors, inhalation rate, and respirable fraction extrapolated from our measurements, we calculated pulmonary deposition of $^{239+240}$ Pu for four cases on Bikini (Table 7). Under the worst case condition (during tilling in a disturbed bare field), the pulmonary deposition was 1476 aCi h⁻¹, and in the best case (light work in a coconut grove), the pulmonary deposition was 12 aCi h⁻¹. Intermediate values were 139 aCi h⁻¹ for heavy work in a bare field, and 78 aCi h⁻¹ for light work in around houses. (In the latter case, we had to use an enhancement factor measured in the nearby field rather than in and around the houses.)

Walking along the road with one vehicular passage per hour produced an estimated 50% chance of additional pulmonary deposition of 1.58 aCi h^{-1} (above background) but the soil plutonium activity on the road was notably lower (4.1 pCi g⁻¹) compared to the field (15.3 pCi g⁻¹); see Table 7.

To put these estimated pulmonary deposition values in perspective, we have estimated the lung and bone doses from inhalation of ²³⁹⁺²⁴⁰Pu and ²⁴¹Am using the ICRP lung model (8). The scenario we adopted is arbitrary; we assume that a person is in high activity conditions (1500 aCi/h pulmonary deposition) for 5 hours per day and in a situation averaging 80 aCi/h for the other 19 hours. The daily average is therefore 376 aCi/h pulmonary deposition. If it is assumed that people are exposed to this level everyday throughout their life, then the maximum bone dose rate is 5.5 mrem/y and the maximum lung dose rate is 2.4 mrem/y; the 30 year integral doses are 36 mrem and 70 mrem for bone and lung respectively. These doses are well below the Federal Guidelines for bone and lung of 500 mrem/y and 5 rem in 30 years. It is also quite possible that the selected scenario of 5 hours at the high activity deposition rate (1500 aCi/hr) is on the average a very high estimate of the annual time spent under such conditions.

<pre>nary Deposition of Plutonium (239⁺²⁴⁰Pu) for Worst Inhalation Dust Soil Pu Enh Rate Rate Aerosol Activity [m³ h-1) (ug m⁻³) (aCi ug⁻¹) 1.04 136 15.3 1.04 21 15.3 Houses, 0.83 21 15.3 0.83 21 8.0</pre>	Pulmonary Deposition of Pluton Inhalation Rate (m ³ h ⁻¹)	ıium (239+2 Dust Aerosol (µg m ⁻³)	⁴⁰ Pu) for Wo Soil Pu Activity (aCi µg ⁻¹)	rst Case and B		
$ \begin{array}{c cccc} Inhalation & Dust & Soil Pu & Enhancement \\ Rate & Aerosol & Activity & Factor \\ (m^3 h-1) & (\mu g m^{-3}) & (aci \mu g^{-1}) & (Ff) \\ 1.04 & 1.04 & 136 & 15.3 & 3.10 \\ 1.04 & 21 & 15.3 & 0.83 \\ Houses & 0.83 & 21 & 15.3 & 0.83 \\ 0.83 & 21 & 15.3 & 0.41 \\ 0.83 & 21 & 8.0 & 0.41 \\ \end{array} $	Inhalation Rate (m ³ h-1)	Dust Aerosol (µg m ⁻³)	Soil Pu Activity (aCi µg ⁻¹)		est C	_
ld, 1.04 136 15.3 1d, 1.04 21 15.3 Houses, 0.83 21 15.3 0.83 21 8.0				Enhancement Factor (EF)	Enh	
ield, 1.04 21 15.3 d Houses, 0.83 21 15.3 e, 0.83 21 8.0		136	15.3	3.10		
d Houses, 0.83 21 15.3 e, 0.83 21 8.0	Field,	21	15.3	0.83		
e, 0.83 21 8.0	und Houses,	21	15.3	0.83		
		21	8.0	0.41		
0.023 28 4.1	Roadside, Vehicle/Hr * 0.023	28	4.1	2.50		

SUMMARY AND CONCLUSIONS

Mass loading (all aerosols) on a HV filter was 55 μ g m⁻³ on Bikini Island over stabilized and vegetated surfaces (e.g., in a bare field following rain and in a coconut grove). This compares to 56 μ g m⁻³ measured at a vegetated site on Engebi Island of Enewetak Atoll in February 1977, and a 42 μ g m⁻³ weekly average for 10 weeks in a coconut grove on Eneu Island of Bikini Atoll May-August, 1978. (Wind speeds were comparable, 4-5 m s⁻¹, in all cases.) The more detailed studies at Bikini revealed that 34 μ g m⁻³ of the mass loading was salt from sea spray, and that this sea spray contribution remained constant across Bikini Island beyond 20-50 m from the windward beach.

The "background" concentrations of aerosol plutonium on Bikini are comparable to those on Engebi Island, Enewetak, when one considers the surface soil plutonium activity (Table 8). And, by assuming that Engebi Island had the same aerosol sea spray background ($34 \ \mu g \ m^{-3}$) as Bikini (which has not been verified by actual measurement) we found that the enhancement factors agree reasonably well. The normal enhancement factor is 0.56, if one assumes that values less than one (Table 8) represent normal variations about the mean of 0.56. Apparently, the process of resuspension is preferentially selective to non-contaminated particles on these atolls to the extent that an aerosol plutonium dilution of 1.8:1 normally occurs.

Location		Plutonium Aerosol oncentration (aCi m ⁻³)	Surface Soil Plutonium Activity (pCi q ⁻¹)	Estimated Enhancement Factor*
Normal "	Background"			
Bikini	Coconut Grove	60	8.01	0.41
Bikini	Stabilized Bare Soil	264	15.3	0.82
Engebi	Vegetated Field	240	24.2	0.45
Unusual	Conditions			
Bikini	Field, freshly tilled	6466	15.3	3.10
Engebi	Garden, freshly tilled	7420	24.2	4.41
Engebi	Garden, 1 wk after til	led 3060	24.2	2.55
Bikini	Road with traffic	421	4.10	2.50
Engebi	Downwind of road	1090	35.2	0.56

TABLE 8. Plutonium Aerosol Concentrations on Bikini and Enewetak Atolls Compared (Winds 4-5 m s⁻¹).

*Calculated by assuming 34 μg m^3 sea spray which has been verified by measurement on Bikini.

bare field stabilized by light rain. Since fields do not remain unvegetated for more than a few months, the coconut grove resuspension flux is probably representative of the Island as a whole, even though the wind speeds are one-fourth as high in the coconut grove canopy as in the open.

Particle size distributions measured by both optical and cascade impactor methods show that over the rain-stabilized bare field, the total aerosol size distribution is log-normal with median aerodynamic diameter of 2.44 μ m and geometric standard deviation of 3.0, but there is no significant size difference between aerosol plutonium activity and aerosol mass concentration. During the unusual condition of tilling, the size distribution significantly shifts from a median aerodynamic diameter of 2.44 μ m to about 2.0 μ m with a concurrent increase in plutonium enhancement factor from less than one to 3.1 on Bikini (4.4 on Engebi) and an increase in the pulmonary respirable-fraction from 0.19 to 0.24. In the case of a soil disturbed by tilling on Bikini, the plutonium concentration increased by a factor of 19.1 due to a 3.23 x increase in enhancement factor and a 5.91 x increase in dust aerosol concentrations.

Vehicular traffic produced dust pulses of nominal 10 s duration in a 4.5 m s⁻¹ wind, which were log-normally distributed having time-averaged concentrations above background of 28 μ g m⁻³ less than 50% of the time and 211 μ g m⁻³ less than 5% of the time. (Peak concentrations were a factor of 3.6 higher.) The plutonium enhancement factors for vehicular traffic was 2.5. Foot and bicycle traffic produced dust pulses about one-fourth as large as vehicular traffic.

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Personal dosimetry showed that under various exposure conditions, workers inhaled different fractions of inorganic dust and salt, while the organic fraction remained constant at about 10%. Consequently, a personal dosimeter enhancement factor was defined to express the effect a worker has by stirring up dust in his own immediate environment. As a result, pulmonary deposition of plutonium could be calculated for various exposure conditions as determined by inhalation rate, aerosol dust concentration, plutonium activity in soil, plutonium enhancement factor, personal dosimeter enhancement factor, and the pulmonary respirable-fraction. Under the worst case (during tilling), pulmonary deposition was 1476 aCi h⁻¹, and in the best case (light work in a coconut grove), it was 12 aCi h⁻¹. Intermediate cases were for heavy outdoor work (139 aCi h⁻¹) and for light work in and around houses (78 aCi h⁻¹). Walking along a road produced an exposure of 1.58 aCi h⁻¹ above background for one vehicular pass per hour. But even with a worst case exposure scenario for 30 years, the lung dose and bone dose due to inhalation on Bikini are well below present Federal Radiation Guidelines.

In conclusion, this study has been the most comprehensive to date, in providing the key parameters for inhalation dose assessment of exposure to plutonium contaminated aerosols. Preliminary dose assessments have been verified now by aerosol measurement methods and at different locations. There remain several unexplained and untested results. It is not yet clear why the aerosol dust concentration is apparently uniform for different surface cover and wind conditions (e.g., coconut grove versus bare field). It is also not known why the plutonium enhancement factor is less than unity in the normal case, while at the same time, the aerosol plutonium activity and the aerosol mass size distributions are not significantly different. Long-term monitoring on these remote atolls is not yet very practical, but, some attempts will be made in the near future to monitor meteorological and aerosol levels by the use of satellite telemetry from remote data acquisition systems.

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REFERENCES

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- J.A. Kirby, L.R. Anspaugh, P.L. Phelos, and G.W. Huckaby, "A Comparison of <u>In Situ</u> Gamma Soil Analysis and Soil Sampling for Mapping ²⁴¹Am and ²³⁹Pu Soil Concentrations at the Nevada Test Site," <u>IEEE Transactions on Nuclear</u> Science, Vol. NS-24, No. 1, 587-590 (1977).
- S.A. Hsu and T. Whelan, "Transport of Atmospheric Sea Salt in Coastal Zone," Env. Sci. and Tech., 10(3), 281-283 (1976).
- 3. R.A. Horne, "Bubbles and Chemical Mass Transport from the Sea into the Atmosphere," p 346-360 in <u>Marine Chemistry</u>, Wiley-Interscience Press, New York (1969).
- 4. E.D. Goldberg, W.S. Broecker, M.G. Gross, and K.K. Turekian, Ch. 5, "Marine Chemistry," p 139-140 in <u>Radioactivity in the Marine Environment</u>, Nat. Acad. Sciences (1971).
- 5. D.H. Yaalon and J. Lomas, "Factors Controlling the Supply and the Chemical Composition of Aerosols in a Near Shore and Coastal Environment," <u>Agr.</u> Meteorol. 7, 443-454 (1970).
- G.F. Rosskneckt, W.P. Elliott, and F.L. Ramsey, "The Size Distribution and Inland Penetration of Sea Salt Particles," J. Appl. Meteorol. 12(5), 825-830 (1973).
- L.R. Anspaugh, J.H. Shinn, P.L. Phelps, and N.C. Kennedy, "Resuspension and Redistribution of Plutonium in Soils," <u>Health Physics</u> 29, 571-582 (1975).
- 8. E.A. Martell, <u>Actinides in the Environment and Their Uptake by Man</u>, Report NCAR-TN/STR-110, National Center for Atmospheric Research, Boulder, Colorado, Department of Commerce, NTIS PB-245727 (1975).
- 9. Task Group on Lung Dynamics (International Commission for Radiation Protection), "Deposition and Retention Models for Internal Dosimetry of the Human Respiratory Tract," Health Physics 12, 173-207 (1966).

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