DP-MS-75-29
IAEA-SM-199/83
Conf-751105--12

MEASURED PLUTONIUM RESUSPENSION AND RESULTING DOSE FROM AGRICULTURAL OPERATIONS ON AN OLD FIELD AT THE SAVANNAH RIVER PLANT IN THE SOUTHEASTERN UNITED STATES

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MASTER

A paper proposed for presentation at the *International Symposium on Transuranium Nuclides in the Environment* in San Francisco, California on November 17-21, 1975 and for publication in the Proceedings

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Abstract

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Plutonium resuspensions, based on air-sample activity, were measured during field preparation and planting of winter wheat at the Savannah River Plant (SRP). Plutonium was originally deposited in the field during the previous twenty years as a result of atmospheric releases from SRP's nuclear fuel processing facility located adjacent to the field, and from global fallout following nuclear weapons tests conducted at other sites. During soil cultivation when the prevailing wind speed ranged from 0 to 6.3 m/sec, airborne plutonium upwind of the field consistently averaged 1.7 fCi**/m^3. Airborne plutonium concentrations during the various activities associated with site preparation and planting averaged 210 fCi/m^3 at 7.6 m and 10 fCi/m^3 at 30.5 m downwind from the edge of the field. The air at face level on the tractor contained 49 fCi/m^3. The average concentration of plutonium in the 0-5 cm soil layer following cultivation was 3100 fCi/g. Dose calculations based on measurements of resuspended plutonium indicated that the 70-year bone-dose commitment to an agricultural worker after one hundred 8-hour days would be 49 mrem, 210 mrem, or 1.8 mrem, depending on whether the worker remained in the tractor seat, 7.6 m downwind of the edge of the field, or 7.6 m upwind of the edge of the field. Resuspension factors for the agricultural activities were $103 \times 10^{-8} m^{-1}$ at 7.6 m downwind, $5 \times 10^{-8} m^{-1}$ at 30.5 m downwind from the edge of the field, and $24 \times 10^{-8} m^{-1}$ at the tractor location.

* The information contained in this article was developed during the course of work under Contract No. AT(07-2)-1 with the U. S. Energy Research and Development Administration.

** 1 fCi = 1 femtocurie = 1×10^{-15} curie.

INTRODUCTION

A comprehensive environmental studies program of plutonium behavior under humid conditions is being conducted at the U. S. Energy Research and Development Administration's Savannah River Plant (SRP) to evaluate the effects of twenty years of reprocessing of irradiated nuclear fuel on the distribution of plutonium in the nearby water, air, soil, and vegetation. Results of studies of the resuspension of plutonium from an old field that was converted to agricultural production for research purposes are discussed in this paper. The field, which is located adjacent to SRP's nuclear fuel reprocessing facility, covers approximately 0.75 hectares and is split into two sections, hereafter referred to as the north field and the south field.

The field was selected using information obtained from the environmental monitoring program [1] that indicated that the soil from this field has plutonium concentrations that are as high or higher than soils from any other field on SRP. The plutonium concentration of the soil on this field is approximately two hundred times higher than the concentration of offplant soils. Offplant soils have plutonium concentrations that are similar to the concentrations expected in the coastal plain region from fallout of plutonium from atmospheric testing of nuclear devices.

Plutonium was deposited in the field following atmospheric releases from the adjacent reprocessing facility and also by global fallout following nuclear weapons tests conducted at other sites. Atmospheric releases of plutonium from the reprocessing facilities have totaled 3.6 curies since operations began in 1955. Most of the plutonium was released either in 1955 or in 1969. The 1955 releases ($\sim$2 curies) occurred prior to the installation of backup high efficiency filters on exhaust air systems. The 1969 release ($\sim$0.8 curies) was the result of a partial breakthrough of the sand filter bed in April as a consequence of corrosion of the filter media support system. The plutonium atmospheric releases for the calendar year 1974 were 8 mCi. With current technology and operating procedures, releases are expected to remain at or below the 1974 level in future years.

Total plutonium inventory in the field is approximately 4 mCi. Resuspension of plutonium was measured during the various operations required to prepare and plant a winter wheat crop.

The sections that follow include an outline of the method of preparing soil and planting the wheat crop, methods of sampling air and soil before, during, and after field preparation, and a discussion of the estimated 70-year bone-dose exposure that would be received by persons in or near the field under the conditions of the plutonium resuspension test.

METHOD

During November 1974, the area beside the irradiated-fuel reprocessing facility was cleared of volunteer pine trees, plum bushes, and other vegetation in preparation for planting agricultural crops. On November 22, the north field was mowed, disked, subsoiled, fertilized, and planted. On November 25, these same operations were performed on the south field. Measurements of resuspended plutonium were made during each operation.

The field was mowed by a large tractor-drawn rotary mower that chopped the old field vegetation into small pieces, thereby facilitating incorporation of the vegetation into the soil during subsequent disking and subsoiling operations. Disking loosened and mixed the surface soil; the subsoil was then broken up to assist root growth. Fertilizer and lime were applied to the fields with a tractor-mounted rotary spreader; grain was broadcast by hand, then covered with soil by light disking. Water content of the soil was approximately 0.11 g per g of soil during field preparation and planting.

To determine plutonium resuspension caused by field preparation, airborne particles were collected by high volume (2 m^3/min) air samplers during the entire agricultural operation. The air samplers were arranged (Fig. 1) downwind, upwind, and on the tractor all at face height. The downwind array included two rows; the first row consisted of three samplers 7.6 m out from the edge of the field and 15.2 m apart, the second row consisted of three samplers 30.4 m out from the edge of the field and 15.2 m apart. The upwind sampler was 7.6 m beyond the edge of the field. Airborne particles were retained in the sampler on 15 cm × 20 cm × 0.045 cm sheets of Hollingsworth-Vose HV-70 filter media. The filters were changed between each agricultural operation and analyzed for plutonium.

To determine plutonium distribution in soil depth prior to and following tillage operations, soil samples were taken at 30 locations on the field. Samples from each location included a vacuumed sample and a soil core that was segmented into 0 to 5 and 5 to 15 cm portions. The purpose of the vacuumed sample was to obtain an estimate of the potential resuspendible plutonium. A portable vacuum cleaner and suction head were modified to pick up the potential resuspendible material at a velocity of 6 m/sec. The suspended materials were collected in a filter bag and the entire sample was used for analysis.

Meteorological data during land preparation were obtained using an instrumented, portable 10-m tower. Anemometers were placed at 1, 2, 4, 5, and 10 meters above the ground. Wind vanes were at the 5- and 10-m levels. Data were recorded on strip charts.

RESULTS

The wind velocity during the agricultural operations varied as shown in Fig. 2. The average wind speed on November 22, 1974 was approximately 2.5 m/sec at the 2-m height on the north field during the cultivation and planting, but the wind direction was highly variable after 1400 hr. The average wind speed on November 25, 1974 was approximately 5 m/sec at the 2-m height on the south field and was consistent in direction.

The resuspended plutonium content upwind (Table I) averaged a consistent 1.7 fCi/m^3. The average airborne plutonium concentration at face height on the tractor was 49 fCi/m^3. Downwind from the field, plutonium concentrations (assumes same time period for each operation) averaged 210 fCi/m^3 at 7.6 m and 10 fCi/m^3 at 30.5 m. The large means and standard deviations shown in Table I for the sampler locations 7.6 m downwind from the edge of the field were caused by 3 high measurements out of 27. Individual sampler values at this distance from the north field ranged from 1.6 fCi/m^3 during fertilizing, to 1020 fCi/m^3 during subsoiling, and 1580 fCi/m^3 during planting and light disking. Similarly the south field values ranged from 3.6 fCi/m^3 during planting and light disking to 2580 fCi/m^3 during mowing.

Table II shows the plutonium concentration in the resuspendible layer and soil layers as well as the ^{238}Pu and $^{239-240}Pu$ relationship. Before cultivation, there was a relatively high ^{238}Pu concentration gradient between the 0 to 15-cm layer of soil (25α% ^{238}Pu)* and the resuspendible layer (50α% ^{238}Pu).

The steep ^{238}Pu concentration gradient provided an excellent tracer for determining transport modes. The ^{238}Pu observed in the air samples downwind from the cultivation operations generally reflect the average value observed in the soil of 25α% following cultivation and not that in the resuspendible fraction of 50α% prior to cultivation.

Calculated atmospheric particulate loadings at the various locations on the field are listed in Table III. If a plutonium concentration of 3120 fCi/g (the average concentration in the top 5 cm following cultivation) is assumed, the particulate loadings ranged from 560 μg of $soil/m^3$ to 69,000 μg of $soil/m^3$. Loadings in rural areas of various southeastern states average 30-45 $\mu g/m^3$ [2].

* α% ^{238}Pu = (^{238}Pu α activity/total Pu α activity) × 100.

DISCUSSION

Resuspension Measurements

Erodibility and resuspension of soil particles under natural environmental conditions have received considerable attention [3]. These studies have indicated that resuspension is dependent upon particle size, particle size distribution, the area covered by nonerodible particles, surface cover, cohesiveness of surface particles, density and shape of individual particles, soil water content, antecedent weather effects on soil surface, and current meteorological conditions. Extensive resuspension studies have been conducted in relation to plutonium resuspension at the Nevada Test Site [4,5,6], Rocky Flats [7,8,9], and in Spain [10] in order to obtain experimental information on the plutonium content of the atmosphere in the vicinity of increased levels of plutonium in the soil. Resuspension studies from agricultural operations under humid climatic conditions have not been conducted to the authors' knowledge.

Resuspension Factor

The term resuspension factor is frequently used in a discussion of airborne plutonium from sources on the ground surface. Resuspension factor is defined as the

$$R_f = \frac{\text{Concentration of plutonium in air (Ci/m}^3\text{)}}{\text{Ground deposition (Ci of plutonium/m}^2\text{)}}$$

Engelman [11] states that this factor has physical validity only as one approximates an infinite plane of uniformly distributed contamination and the measured soil contamination should be the resuspendible fraction. For the agricultural activities of this report, the need for an infinite plane is removed because the contribution from the soil preparation is much greater than the contribution from tho upwind area. For discussion purposes, one can assume that all the plutonium present in the top 5 cm (2.03×10^{-7} Ci/m^2) is resuspendible and obtain resuspension factors of $24 \times 10^{-8} m^{-1}$, $103 \times 10^{-8} m^{-1}$, and $5 \times 10^{-8} m^{-1}$ at the tractor, at 7.6 m, and at 30.5 m from the edge of the field respectively. The assumption of 5 cm is reasonable because agricultural activities generally disturb the soil to this depth. These resuspension factors are lower than the 10^{-5} resuspension factor chosen in the Liquid Metal Fast Breeder Reactor Environmental Impact Statement [12] as adequately representing the initial conditions for the average population in the relatively well vegetated area of interest in that report.

No attempt is made in this paper to test existing models [12,13,14,15] for predicting resuspension or particulate behavior. The variation between samplers during a particular operation indicated that larger arrays as well as measurements at greater distances are necessary before detailed testing of the models is possible. The data, however, can be used to calculate the dose that would be received by farmers working routinely in similar soils with similar plutonium concentrations.

Bases of Dose Calculations

The radiation dose from inhalation of particulates is a function of the particle concentration, size distribution, and solubility in pulmonary fluid. The dose is delivered to the lungs from direct deposition by inhalation, to the gastrointestinal (GI) tract from clearance by the lungs and ingestion, and to internal organs from dissolution of particles and subsequent transport via body fluids. A secondary pathway to internal organs occurs from uptake in the GI tract followed by circulatory transport. For plutonium inhalation, the highest concentration occurs in lymphatic tissue. However, the rare occurrence of neoplasia has prompted recommendation that lymphatic tissue not be considered limiting for dose calculation of inhalation exposure to plutonium [16]. Consequently dose calculations were not made for this tissue.

The Task Group on Lung Dynamics proposed a deposition-and-retention model for inhalation of particulates [17]. The model consists of three compartments; nasopharynx (N-P), tracheabronchial (T-B), and pulmonary (P). The pulmonary compartment is the functional area (exchange space) of the respiratory system and the compartment for which radiation dose is greatest because of residence time. Material in the N-P and T-B compartments is cleared more rapidly by ciliary action, resulting in lower doses to these compartments.

Particulates are classed according to their pulmonary clearance times as D (days), W (weeks), or Y (years). Most common plutonium compounds are Class Y cxccpt for nitrates and citrates, which are Class W.

The primary consideration for dose calculation from inhalation of plutonium particulates is particle size. Particles of plutonium dioxide larger than 2 μm in diameter are not transported to the pulmonary compartment, but are stopped in the N-P or T-B compartment [18]. Particles larger than 4 μm are retained totally in the N-P compartment. Maximum pulmonary deposition occurs at 0.5 to 0.7 μm and below 0.02 μm. Radiation dose to lung and bone is calculated for the particle size range of 0.01 μm to 10 μm.

The radiation dose was calculated for the pulmonary compartment of the respiratory systems, the bone, the liver and the GI tract. (The GI tract dose is minimal and will not be considered further.) Calculations were made for both Class Y and Class W clearance rates. All dose calculations are integral 70-year dose commitments, i.e., the integral dose delivered to the organ in the 70 years immediately following the uptake. Dose calculations are based upon methods given in International Commission on Radiological Protection (ICRP) Publications 2 [19], 19 [16], and 20 [20]. Anatomical and physiological data are from ICRP Publication 23 [21]. Lung doses were calculated from clearance rates given in ICRP Publication 19 [16]. The difference between the dose commitments for different clearance classes is as expected. A Class W clearance gives a lower dose to the lung than a Class Y but gives a greater bone dose because a larger portion enters the circulatory system.

For dose calculations, it is generally assumed that plutonium that reaches the circulatory system is deposited equally in two primary sites, viz., bone and liver. The distribution factor term of the effective energy causes the bone dose to be 2.5 times that of the liver. i.e., the bone is the "critical organ." The dose to the bone is via two pathways. The direct route is from lung via blood to bone. A second pathway results from transport of material from lung to GI tract with subsequent transfer to the circulatory system. This route is minimal because the plutonium absorbed is 3×10^{-5} of the amount present in the GI tract.

Seventy-Year Bone-Dose Commitment for Inhaled Plutonium

A nomograph for dose commitment calculation for ^{239}Pu is shown in Fig. 3. The nomograph dose assumes a log normal particle size distribution with a median equivalent aerodynamic diameter of 2 μm [22,23] because cascade impactor information was not available. Dose commitments in mrem to the bone can be obtained for combinations of radioactivity concentrations and exposure times. Dose commitments to the lungs are lower than those to the bone by factors of 5 and 130 for Class Y and Class W clearance rates, respectively. The dose from ^{238}Pu is 81% of that from ^{239}Pu.

Fig. 3 was used to calculate the 70-year bone dose commitment an individual would receive from spending a selected number of 8-hr days in various environments. The environments selected (Table IV) include on the tractor, 7.6 m upwind, 7.6 m downwind, and surface air averaging 0.050 fCi/m^3 [23]. For illustrative purposes the time period of exposure chosen was 100 days, a conservative time period for routine cultivation activities in the course of one year. The calculated dose received by the tractor driver, 49 mrem, compares with a nominal 7000 mrem from natural background during the 70-year time period. The downwind locations are not realistic for considering inhalation dose because normally individuals would not be in these locations for the exposure times used in the calculations.

ACKNOWLEDGMENTS

Our thanks to Helen Pfuderer and Cathy Sanders of the Ecological Sciences Information Center, Oak Ridge National Laboratory for conducting the literature search, and to Barry Gilbert of U. S. Environmental Protection Agency for suspended-particulate information in the southeastern United States.

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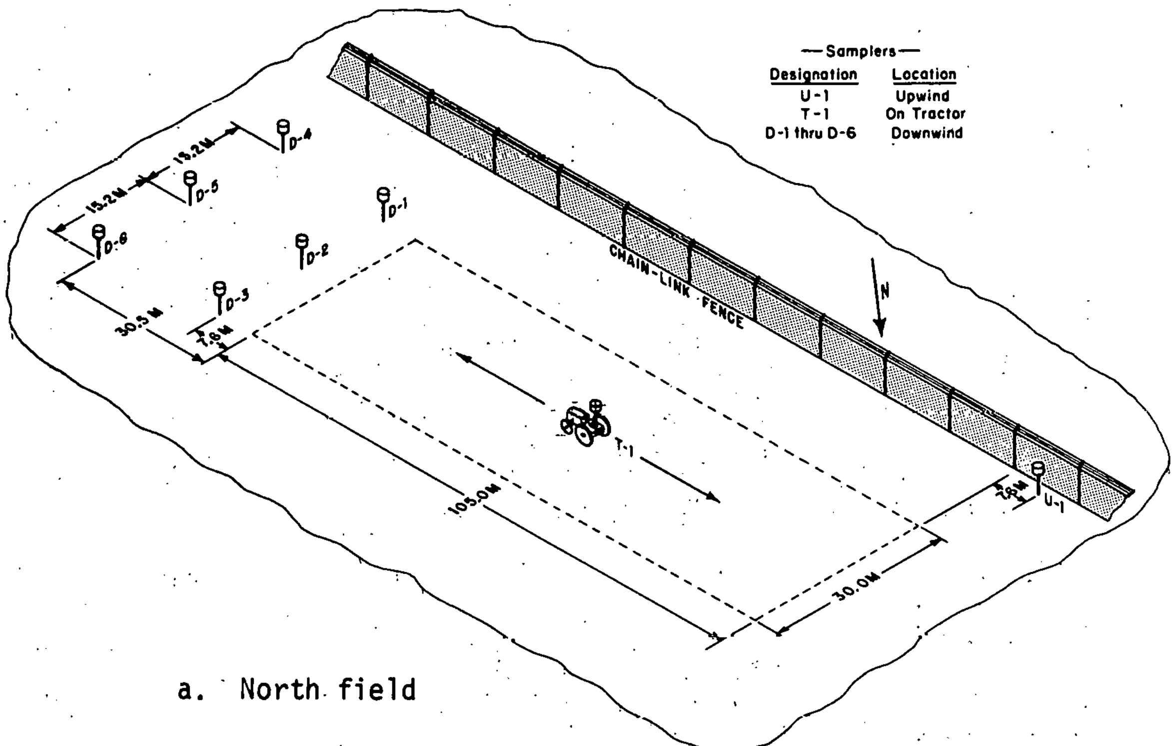
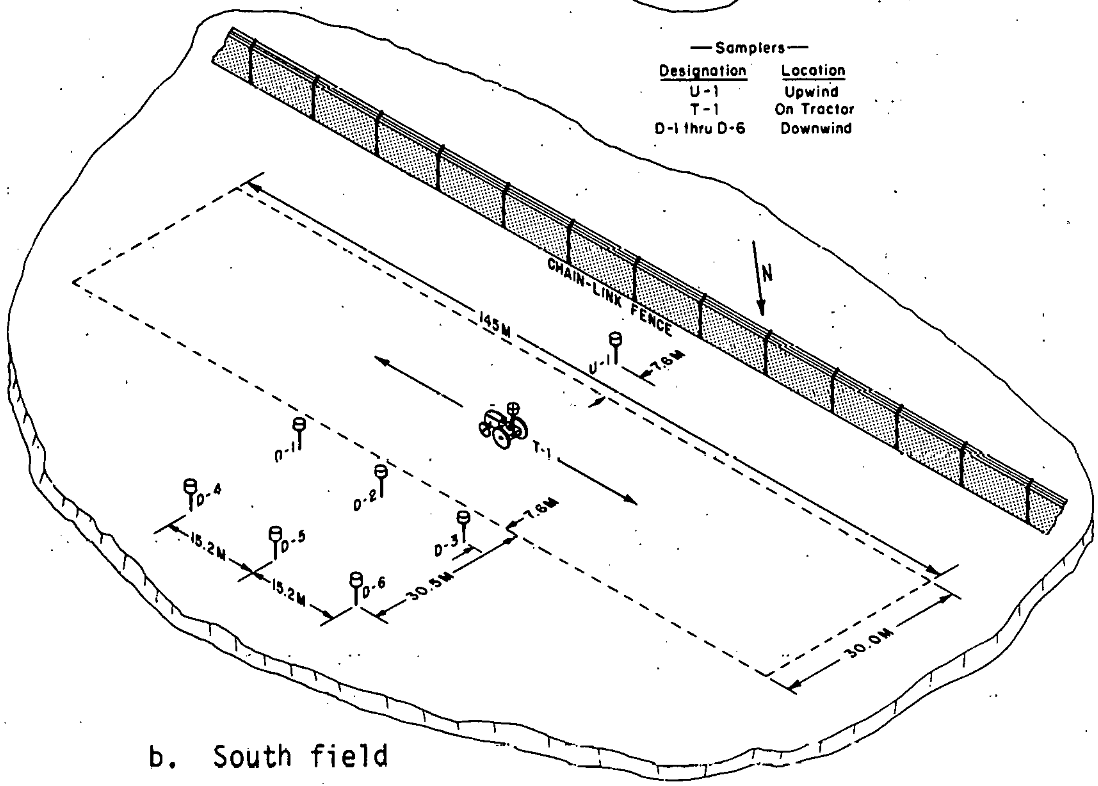
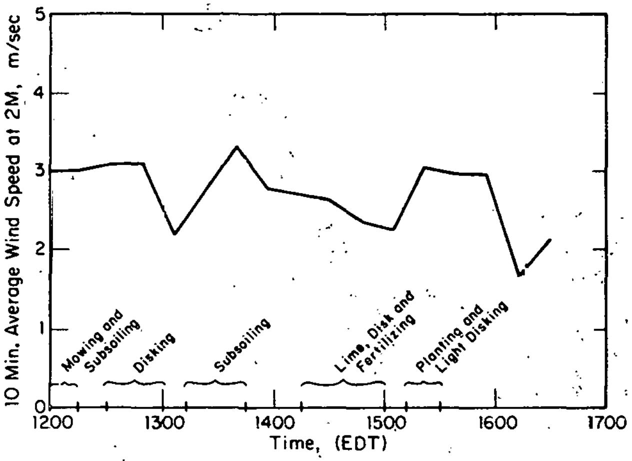


a. North field

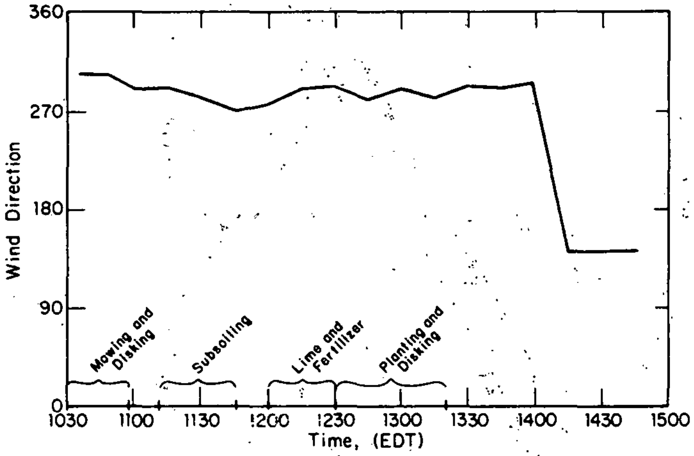


b. South field

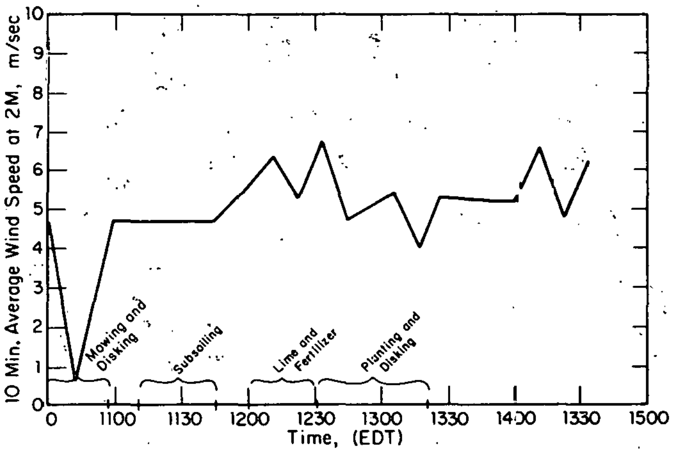
FIG. 1. Locations of air samplers



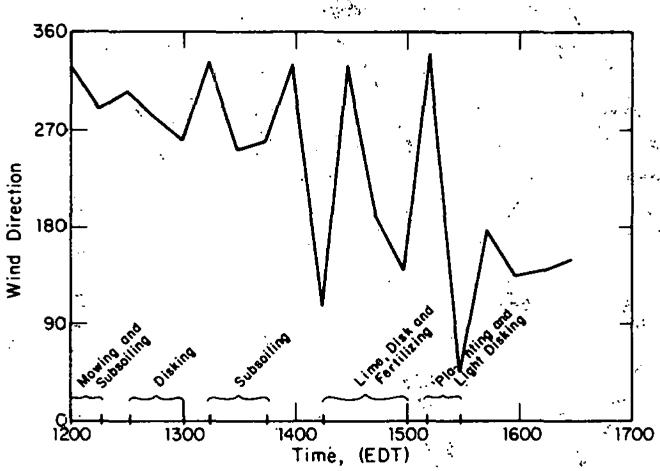
a. Wind speed in north field.



b. Wind direction in north field.



c. Wind speed in south field.



d. Wind direction in south field.

FIG. 2. Variation in wind velocity during agricultural operations.

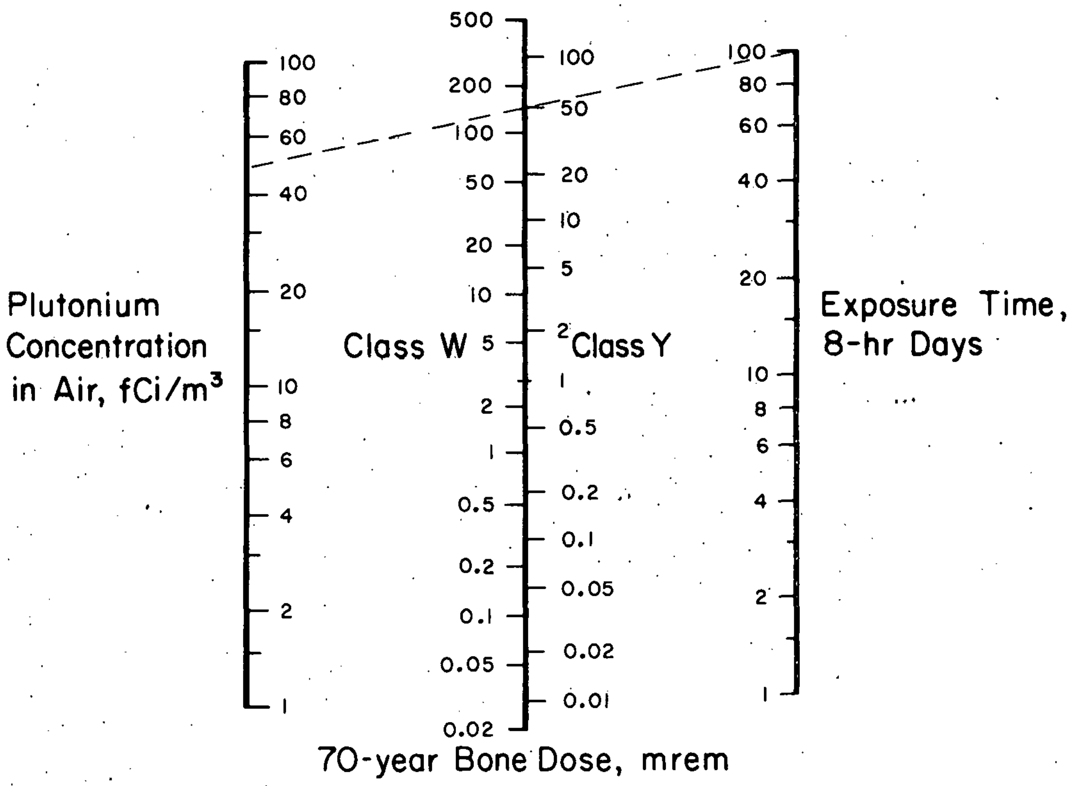


FIG. 3. Inhalation nomograph for bone dose calculations.

TABLE I. SUMMARY OF PLUTONIUM AIR CONCENTRATIONS AND ALPHA PERCENT Pu MEASURED DURING AGRICULTURAL OPERATIONS

Sample Location	*Number of Filters Analyzed*	*Sampling Distance from Edge of Field, m*	*Average Concentration,[a] fCi/m³*		
			North Field	*South Field*	*Both Fields*
Upwind Background	7	7.6	1.4 ±0.3	2.0 ±0.6	1.7 ±0.6
Tractor	8	0	7.4 ±8.6	91 ±81	49 ±69
Agricultural Operations (Downwind)	27	7.6	192 ±463	233 ±740	210 ±590
	25	30.5	11 ±18	9.1 ±13	10 ±16
			Average α% ^{238}Pu[a,b]		
			North Field	*South Field*	*Both Fields*
Upwind Background	7	7.6	12 ±4	18 ±3	15 ±4
Tractor	8	0	27 ±15	25 ±27	26 ±20
Agricultural Operations (Downwind)	27	7.6	15 ±14	18 ±11	17 ±13
	25	30.5	32 ±19	38 ±20	35 ±19

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a. Mean value and standard deviation

b. $\frac{^{238}\text{Pu } \alpha \text{ activity}}{\text{Total Pu } \alpha \text{ activity}} \times 100$

TABLE II. PLUTONIUM ANALYSIS OF SOIL PRIOR TO AND FOLLOWING CULTIVATION

	α% ^{238}Pu[a]			*Total Pu Concentration, fCi/g*		
	North Field	*South Field*	*Both Fields*	*North Field*	*South Field*	*Both Fields*
Soil prior to cultivation						
Resuspendibles (0 to 0.1 cm)	49 ±9	51 ±9	50 ±9	1600 ±900	13000 ±16000	8500 ±14000
0.1 to 5-cm cores	20 ±9	28 ±8	25 ±9	600 ±500	1600 ±1500	1200 ±1500
5 to 15-cm cores	20 ±8	30 ±10	26 ±11	90 ±60	120 ±120	110 ±100
Soil following initial cultivation						
Resuspendibles (0 to 0.1 cm)	27 ±8	28 ±5	28 ±6	900 ±400	4800 ±2400	5500 ±2700
0.1 to 5-cm cores	26 ±8	23 ±8	24 ±8	900 ±700	4600 ±2500	5100 ±2700
5 to 15-cm cores	25 ±13	22 ±9	23 ±11	200 ±100	900 ±1500	600 ±1200

a. $\frac{^{238}\text{Pu } \alpha \text{ activity}}{\text{Total Pu } \alpha \text{ activity}} \times 100$

TABLE III. CALCULATED PARTICULATE LOADINGS AT VARIOUS LOCATIONS ON THE FIELD

Location	*Concentration,[a]* $\mu g/m^3$
On Tractor	16,000
7.6-m Upwind	580
7.6-m Downwind	67,000
30.5-m Downwind	300
Average for Rural Southeastern U.S.[b]	30-45

a. Assumes a plutonium concentration in particulate of 3120 fCi/g.

b. Ref. [2].

TABLE IV. DOSE FROM SELECTED ENVIRONMENTS[a]

Location	α% ^{238}*Pu*[b]	*Pu Concentration,* fCi/m^3	*70-Year Dose Commitment for 100 Day Exposure, mrem* *Bone*	*Liver*	*Lung*
On Tractor	26	49	49	20	7.8
7.6-m Upwind	15	1.8	1.8	0.7	0.4
7.6-m Downwind	17	210	210	87	44
Average Environmental Plutonium Concentration[c]	15	0.05	0.05	0.02	0.01

a. 70-year dose commitment from inhalation of measured plutonium concentrations in air during agricultural activities, assuming Class Y clearance rate and assuming 100 eight-hour work days.

b. $\frac{^{238}\text{Pu } \alpha \text{ activity}}{\text{Total Pu } \alpha \text{ activity}} \times 100$

c. Reference [2].