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BIOMEDICAL DIVISION

RESUSPENSION OF PLUTONIUM: A PROGRESS REPORT

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February 19, 1974

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Lawrence Livermore Laboratory

Bio-Medical Division

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Resuspension of Plutonium: A Progress Report

Compiled by: L. R. Anspaugh
P. L. Phelps

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Resuspension Element Status Report

I. Introduction

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The resuspension element of the Nevada Applied Ecology Group (NAEG) was formed to undertake the task of studying at the Nevada Test Site (NTS) the movement of plutonium by wind driven forces and assess the potential biological hazards associated with airborne plutonium particles. It was also to provide input to the Nevada Operations Office of the Atomic Energy Commission regarding cleanup of plutonium contaminated areas. This involvement gave the LLL Bio-Medical Division an opportunity to study fundamental processes involved in the resuspension of plutonium particles from a soil surface and develop a time and spatial dependent mathematical model describing average concentration of airborne plutonium as a function of the source (geometrical configuration, soil surface characteristics) and driving forces. It is intended that the model will also provide the necessary input to models of lung dynamics of plutonium behavior in the human respiratory tract. The NAEG resuspension elements plan for meeting the above objectives is diagramed in Fig. 1.

Prior to our involvement in the NAEG program we had carried out resuspension studies related to nuclear cratering experiments and accidental releases from nuclear device tests. One of these studies was in conjunction with the Schooner cratering experiment.¹⁻³

We had conducted radioactivity measurements on an arc approximately 50 miles from the Schooner surface ground zero (SGZ). Some of the highest readings were obtained in the vicinity of the Queen City Summit outside the northern boundaries of NTS. Based upon conjecture that the reason for this was unique interactions between wind flow and topography at the Queen City Summit region which had existed during the Schooner cloud passage, we hypothesized that similar conditions could have existed during atmospheric device testings in previous years at NTS which could have resulted in unusually high plutonium depositions at the same site.

Consequently we analyzed soil samples for plutonium and obtained data in support of our hypothesis. Plutonium levels in soil at Queen City Summit were found to be 100 times over the average background levels in U.S. soil. Subsequent measurements were made by the Environmental Protection Agency's National Environmental Research Laboratory, Las Vegas (formerly the U.S. Public Health Service), which confirmed our earlier findings. These findings added impetus to carry out resuspension studies at NTS.

Subsequent to starting our plutonium resuspension field studies, the opportunity arose to conduct a long term experiment using the radioactivity deposited on the soil surface during the Baneberry accidental venting.³ The incident occurred in December 1970. We carried out the study over approximately a one year period. The study was directed at observing the resuspension of radioactivity as a function of time.

Following the Baneberry study, efforts were directed toward designing a series of experiments at plutonium contaminated areas on NTS. A joint proposal from LLL and NOAA/ARL outlining the experimental plan was submitted for AEC review. Early in 1972 research, development, and procurement were started on special apparatus needed to carry out the experiment. This included developing ultra-high volume air samplers, in-situ particle spectrometers,

light-scattering type particle counters and establishing a micrometeorology field laboratory. A plutonium contaminated site was selected and surveyed for layout of the experiment.

Resuspension Experiment: GMX-Area 5

The GMX Site, in Area 5, was selected for conducting the first plutonium resuspension experiment. This area had previously been used for carrying out a series of experiments involving plutonium and high explosives. At no time were nuclear detonations involved. These plutonium-high explosive tests started in late 1954 and ended in early 1956. Thus, as of this date, the mean life of the plutonium source at GMX is 18 years. Following the 1954-1956 series of tests, the highly contaminated area was fenced off and remained essentially undisturbed.

Various groups within the NAEG program have gathered data which support our studies. These data include the areal distribution of plutonium, the distribution and concentration of plutonium within the soil, and a population census of vegetation covering the GMX study area.

The initial experiment was designed with the goal of developing a model to predict air concentrations of plutonium given the source characteristics and relevant meteorological parameters. Our objective was then to conduct a series of measurements within and downwind of the plutonium source. This was to be done during a period of time when the winds were consistent. Examination of several years of wind data showed that this was possible for a 2-4 hour period. The experiment was laid out as shown in Fig. 2. The line showing the sampling stations is located at 020° true, which corresponds to the mean direction of winds over 10 mph from March 24 to August 30, 1971 at the GMX site.

Micrometeorological measurements were directed at characterizing the driving forces, i.e., winds, turbulence and related parameters that are

considered to be most likely related to moving or picking up particles from the desert pavement. These measurements are discussed in detail in Part II. The main micrometeorology station was established 1200 feet downwind from GZ, at R-1 as shown in Fig. 2. Air sampling stations were established at GZ, 1200 feet, 2400 feet and 4800 feet from GZ. In addition to the downwind sampling stations, control stations were established upwind from GZ, at Mercury and in Livermore, California. These background stations utilized the identical type of ultra-high-volume air sampler.

During a field experiment, which usually lasted from 1-4 hours, the various micrometeorological parameters were recorded and the ultra-high-volume air samplers operated. All the samplers were synchronized to start and stop together. Concurrent with these measurements, particle spectrum analyses at various heights above the ground were made using the Climet particle analyzer (Part V). Also overlapping in time with these measurements, background air samples were collected 900 feet upwind from GZ, at Mercury, Nevada and in Livermore, California.

Saltation/creep samplers have been installed for collecting particles that roll or bounce on the soil surface (Part IV). These samplers as yet have not been integrated into the field experiments. It is planned to finish evaluation studies in 1974 and use the instruments shortly afterwards.

Cascade impactors were located on an arc 250 feet from GZ and were run continuously for approximately 30 day periods. These instruments gave results on particle size fractions for plutonium and total airborne particulates. Results are given in Part VI.

A time-lapse camera on an 80 ft tower was located up-wind from GZ such that the entire GMX experimental site was in its field of view. The camera has been operated during the daylight hours with a frame exposure every 5 seconds. During the times of high dust devil activities (June-September)

the camera was used to record dust devils traversing the GMX experimental site. This was done to qualitatively assess the impact of dust devils on transporting surface material into the atmosphere. The data which were available for the GMX experiment are presented in detail in Part VI of this report. A preliminary analysis of the air concentration data collected by REECO prior to the initiation of this study is also presented in Part VI.

Development of Specialized Instruments and Techniques

A variety of specialized instruments had to be designed and fabricated before the plutonium redistribution experiments could be conducted at the Nevada Test Site. One of the major undertakings was the development of an air sampler capable of collecting adequate amounts of airborne plutonium particulates during a 2-4 hour sampling period. This time period corresponded to the time "window" that consistent meteorology could be expected. Calculations indicated that if the most sensitive radiochemical procedures were used in analyzing the air filters for plutonium, an air sampler with a throughput at 1500-2000 m³/hr would be required. Samplers with this required flow rates were not available; therefore, we developed an ultra-high-volume air sampler meeting the desired specifications. Tests on the prototype sampler were very encouraging and indicated that even at global background levels surface air could be sampled for a time period of one hour and accurate determinations of plutonium air concentrations made. Fourteen ultra-high-volume air samplers were eventually fabricated. They are deployed in Area 13, GMX and Mercury. One unit is in operation at Livermore.

Micrometeorological forces that may cause resuspension of particles from the soil surface have significant transient behavior over very short periods of time, probably less than a second to a few minutes. The airborne particles resulting from resuspension normally are collected by use of a filter

media-airpump combination. The collected particles are then analyzed for plutonium and the concentration of radioactivity in air is determined from the ratio of plutonium to the amount of air sampled. Therefore, since sampling must be done over a minimum of one hour, which corresponds to the least time possible with the ultra-high-volume air sampler previously mentioned, mechanisms related to pick-up of particles from the soil surface cannot be studied in real time and with high resolution.

To overcome these limitations, two light scattering instruments were adapted to measure the mass loading of particles in air and the number distribution as a function of particle size over very short periods of time. Both instruments give data in real time, such that it is possible to correlate micrometeorological events of short duration with the concentration of airborne particles. These instruments examine the particle spectrum in the region of 0.1 to 10 µm, and do not differentiate between radioactive and non-radioactive particles. Nevertheless, valuable data has been obtained on the mass loading of particles in air as a function of the forces causing pick-up from the soil surface.

The first instrument employed was a light scattering type particle analyzer. This instrument, when used with a pulse height analyzer, will give particle size histograms over a range of 0.5 to 10.5 µm. Sufficient data can be collected in two minute or less periods. Particle sizes as measured by this technique are related to their physical diameters. If the physical properties of the particles are known, aerodynamic or "effective" particle sizes can be determined. This is necessary for modelling of particle deposition in the human respiratory tract. Details of the instrument and its application to the GMX experiment are covered in Part V. Another light scattering device we are currently employing is the nephelometer. It has extremely fast response (< 1 sec), however, it does not give particle size distribution data but only

relative particle concentration. It is an exceptional instrument for correlating very short time duration meteorological events with mass loading of particles in the air.

A recently introduced high-volume (20 cfm) cascade impactor was adapted for use in the GMX experiment. This instrument has 5 stages which correspond to 1.1 μm , 1.1-2.0 μm , 2.0-3.3 μm , 3.3-7.0 μm and greater than 7.0 μm . The filter papers may be analyzed for total plutonium per stage and total mass for all particles on each stage. Thus the particles picked up from the soil surface may be characterized according to their aerodynamic sizes and the relationship between plutonium and total mass may be established. Further refinements are being made on this instrument which will reduce re-entertainment of particles and improve its particle intake characteristics.

Particles that roll or bounce along the soil surface (creep/saltation) may be responsible for the most significant net movement of surface mass as compared to other processes. Existing instruments that had been used in agricultural erosion studies and for measuring the movement of sand in windstorms were evaluated. A series of laboratory studies were conducted at LLL on borrowed or copied instruments. Out of this work we developed a multistage sampler capable of collecting the particle fraction moving by creep and particles moving in saltation at various levels above the surface. Details on the design and preliminary results are covered in Part IV.

Initial dust-devil observations were made with a time lapse camera adapted for use on an 80-foot tower. The camera was programmed to expose one frame every 5 seconds. Experimentation with optimum camera location and lens selection resulted in being able to view the entire GMX experimental site. This system was developed as a survey tool in order to get an approximate idea of the dust devil population. A visual count in the GMX and surrounding area during a one-hour period in July 1973 showed a dust devil population of

approximately one dust devil formation per minute, with an average life-time of about two minutes.

An elaborate meteorological data gathering system capable of measuring wind and temperature profiles, fluctuations of the horizontal and vertical wind structure, soil moisture, soil temperature, humidity, rainfall and other miscellaneous meteorological measurements was set up. This was under the direction of the Air Resources Laboratory-Las Vegas and is covered in detail in Part II.

Future Studies

A number of problems dealing with understanding redistribution of radioactivity at NTS still remain. These problems are related to possible long-term health and safety issues, future land uses of NTS, and other AEC nuclear programs. So far, our studies have involved the movement of plutonium at the GMX site and redistribution of gamma-emitters released from the Schooner cratering event and the Baneberry device test. To date the GMX studies have focused on plutonium particles carried into suspension by the force of the wind. Emphasis has been on collecting samples of airborne plutonium particles and measuring a number of micrometeorological parameters such that the relationship between air concentrations of plutonium and the important driving forces may be understood and a mathematical model constructed. The wind forces studied so far have been diurnal in nature and occasionally those associated with dust storms.

a. Dust Devils. There is, however, another driving force: dust devils, which might be a significant mechanism under certain circumstances for transporting surface radioactive material into the atmosphere.

Because of the high wind speeds generated close to the ground and the large vertical structure, dust devils are capable of removing and transporting vertically a significant portion of loose surface material. Dust

devil measurements by Sinclair^{4,5} indicate that on the average a typical dust devil source region of 285 km² provides a total mean seasonal vertical dust transport of over 7×10^3 tons of desert sand. This is one to two orders of magnitude larger, for the same surface area, than pre-dust storm conditions⁶ and smaller by approximately the same amount within the most intense dust storms. Consequently, the vertical and horizontal transport of surface material by dust devils may in itself be an important component in the overall resuspension budget.

However, of more importance is the question of transport and ultimate fate of radioactive materials by dust devils and in general by atmospheric convective elements that originate in the surface boundary layer. We plan to make direct quantitative measurements of the amount, the distance and the direction of dust transported from a known dust devil source region at NTS. The analysis of these data will provide estimates of the possible radioactive substances such as plutonium by dust devils.

b. Variation in Site Characteristics. The availability of plutonium for resuspension is related to a significant degree to the soil-plant surface characteristics of the plutonium contaminated area. This is particularly true for aged sources that are found at the Nevada Test Site. A number of plutonium contaminated sites exist at the Nevada Test Site. Each of the sites have more-or-less different and unique characteristics that influence the availability and movement of plutonium. The properties of importance are the initial physical-chemical form of the source, source age, source strength (surface related), source area and shape, physical-chemical makeup of the soil, soil surface structure, plant coverage, animal population, climatology and land utilization. These properties are to be intensively studied.

An example of an area not yet studied is the Tonopah Test Range. This area is suitable for cattle grazing. A native species of grass, not found in the GMX area, is in abundance. Also, the physical "desert pavement" characteristics are significantly different than those so far studied. The Tonopah area affords an opportunity to study a plutonium contaminated area with different surface characteristics not heretofore studied.

c. Man-made Effects on Redistribution. Displacement of surface soil by mechanical means can increase the rate of redistribution. The impact from future uses of the Nevada Test Site is of particular interest. In addition to continued device testing, future use may include radioactive fuel processing, radioactive waste storage, solar energy research and agricultural uses. Coupled with these uses are a variety of ways that sufficient mechanical disturbances to contaminated sites can result. These include construction of facilities and roads, vehicular traffic over desert pavement, and grazing by cattle. A number of the areas contaminated from past device testing and nuclear cratering events should be studied. A natural follow-up experiment in the GMX area would be to introduce a variety of man-made disturbances to the soil surface and quantitate the change in resuspension of plutonium.

d. Source Term Analysis and Redistribution of Radioactivity at NTS

An evaluation of the total Nevada Test Site contribution of dose to man is limited by the lack of accurate mapping of the resuspendable radioactive source material distributed throughout the site. However, elements within the NAEG program are compiling information on several radionuclides and their distribution in soil which if extended to all radionuclides and areas could be the basis for input to such a mapping.

Assuming that a data bank on radionuclide distribution at NTS was established, the other necessary inputs would be a comprehensive modelling of the meso-scale meteorology related to wind flow at NTS, and an understanding of resuspension for the variety of source characteristics present through-out the site.

This study would provide information on the relationship between source and the dose to man from existing radioactivity and possible future uses.

ACKNOWLEDGEMENTS

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Figure Captions

- Fig. 1. This diagram illustrates the inter-relationship between the field experiments (data gathering) and the various objectives of the NAEG redistribution element.
- Fig. 2. Layout of the resuspension experiment at GMX, Area 5. The distance between R0 and R4 is approximately 5700 ft. The line shown at 020° True corresponds to the mean direction of winds above 10 mph from March 24 to August 30, 1971. The ultra-high-volume air samplers are operated during periods when the wind is toward the 20° radial.

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II. Measurements of Meteorological Parameters

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A part of the Air Resources Laboratory-Las Vegas support to the Resuspension Element has been the recording of meteorological information at air-sampling locations. Early in the program it was recognized that some, even if rather gross, measurements of wind direction, speed, and precipitation were desirable and these have been made at REECo and LLL air sampling-sites. These data are being used in the analysis of sample variation.

In conjunction with the more detailed GMX area resuspension experiments, we have established a more elaborate meteorological data gathering system. A brief outline of the wind, temperature, moisture, and other miscellaneous meteorological measurements made during sampling periods follows:

WIND

Mean wind direction is measured using standard vanes. A bivane is used to measure fluctuations of the horizontal and vertical directions with electronic computation of the resulting standard deviation of the azimuth and vertical angles.

Horizontal wind speed is measured at six levels (0.5, 1.0, 2.0, 4.0, 8.0 10.0 meters) using cup anemometers. Wind tunnel calibrations of the 3-cup

anemometers are made periodically. Cup revolutions of the lowest 5 sensors are counted and converted to mean horizontal speed for the period of interest, usually a 10 minute to 6 hour period. Analog recordings of speed and direction are made for the 10 meter sensor to give a real time indication of wind characteristics and a record of gust speeds at that level.

TEMPERATURE

Temperature is sensed at and recorded for five levels. Two levels are at 1 and 3 cm below the soil surface and three levels are at 0.5, 2.0 and 8.0 meters. Above ground sensors are in aspirated shields.

MOISTURE

Relative humidity is measured using a hygrothermograph supplemented during experiments with observations using a sling psychrometer.

Soil moisture is obtained for representative times using the average of five approximately 100 gm samples of the top 0.5 cm layer of the soil. Samples are weighed, then dried and reweighed.

Precipitation is measured using a recording rain gage.

SOLAR RADIATION

Incoming and outgoing solar radiation are measured using a pyranometer and two radiometers. These measurements will be used primarily for dust devil studies.

OTHER MEASUREMENTS

Atmospheric pressure is measured with an aneroid barometer at specific intervals.

Clouds are observed visually as well as sensed by the pyranometer.

Visible weather phenomena, such as showers in the area, blowing dust over the dry lake, and dust devil activity are recorded.

An upper air sounding (radiosonde) is available twice a day at standard observation times from a station about 10 miles to the northwest.

We have operated the system about thirty days over the past year for system checkout, during ultra-high volume air sample runs and during particle analyzer runs.

Figure 1 shows the wind and temperature profiles for four high-volume air sampler runs. One profile is for light winds, one of strong winds and the other two for the first two runs for which we have received the ^{239}Pu activity data.

The wind speed profile is log height/linear speed diagram using mean wind speed for the period the sampler was operating. The temperature diagram is similar but uses temperature differences between levels. Soil temperatures were about 10°C higher.

Certain recorded and derived parameters are being observed in order to characterize the meteorology in ways that will relate it to the variation in the measurements of suspended material.

Wind direction is the mean direction for the sample period. The sampler array is on a bearing of 200 degrees which is close to the climatologically most frequently observed downwind direction. Mean gust is the mean of the peak gusts recorded for each 10 minute period during the sample period. Soil moisture has varied from about 10% to 0.1%. Cloud cover is the mean of hourly values of low and middle cloud sky coverage.

The wind speed profile give a measurement of the vertical shear of the wind and an indirect estimate of eddy momentum flux (or stress) through computation of the friction velocity (U^*). This is done with reservations during periods of extreme thermal instability.

The temperature profile is a measure of vertical stability and in combination with the wind speed profile gives a parameter which is a measure of the ratio of buoyant to inertia forces (Ri).

The mean and standard deviation of the horizontal component of the wind direction are indicators of the direction of travel and horizontal dispersion. The standard deviation of the vertical component of wind direction is an indicator of the vertical dispersion and also can be used to estimate eddy momentum flux.

Figure 2 shows similar information for ten minute periods that correspond to particle analyzer run times.

In general, we are attempting to measure some of the differing values for meteorological parameters that we feel will assist us in understanding variations in the air samples. We hope to be able to relate our measures of moisture to particle availability, wind pressure forces to initial particle motion, eddy momentum flux to suspension, and wind direction variance to dispersion.

FIGURE CAPTIONS

Figure 1. Meteorological data for four ultra high volume sampler runs at the GMX site.

Figure 2. Meteorological data for four optical particle analyzer runs at the GMX site.

Meteorological Data for Four UHV Air Sampler Runs at GMX

DATE	TIME PST	WIND DIR. DEG.	MEAN GUST m sec ⁻¹	SOIL MOISTURE %	CLOUD COVER	Ri	σ _w (1 m) m sec ⁻¹
730316	1300 1700	180	4.0	0.3	0	0.958	.21
730613	1220-1830	210	9.8	0.3	.8	0.025	.39
770531	1150-1400	209	13.9	0.3	.8	0.033	.52
730417	1130-1620	216	13.9	0.4	.2	0.030	.44

Fig. 1
Kennedy, N. C.
and
Booth, H. G.

Meteorological Data for Four Particle Analyzer Runs at GMX

DATE	TIME PST	WIND DIR. DEG.	MAX GUST m sec ⁻¹	SOIL MOISTURE %	CLOUD COVER	RI	σ _w m sec ⁻¹
730515	1250	200	3.6	0.4	.4	-1.20	.25
730531	0920	155	4.0	0.3	.1	-0.675	.30
730510	1240	225	8.0	0.3	0	-0.132	.29
730802	1400	185	17.4	m	.6	-0.0217	.75

Fig. 2
Kennedy, N. C.
and
Booth, H. G.

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III. Ultra High Volume Air Sampler

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General Description of Air Sampler

An air sampler with a nominal volumetric flow rate of 1500 m³/hr was developed so that a measureable amount of ²³⁹Pu could be collected in as short a time span as two hours. A photograph of the air sampler is shown in Fig. 1. The design of the unit is patterned after an air sampler built by Asikainen and Blomqvist¹ which had a flow rate of 1000 m³/hr. The air flow through the sampler is maintained by a centrifugal blower whose inlet is attached to one side of a large plenum. The filter, which measures 0.6 m x 1.7 m, is mounted horizontally on top of the plenum at a height of 1.3 m above ground level. The blower exhausts through a 0.3 m diameter by 3 m long sheet metal duct which is inclined at an angle of 10° with respect to the ground. The length of the duct precludes any interference of the exhaust flow stream with the air intake to the sampler. The duct is inclined so that the exhaust flow stream dissipates without directly striking the ground which would artificially introduce aerosol into the air. The centrifugal blower is a 0.4 m diameter wheel with radial blades, and is driven by a 7.5 hp electric motor.

Filter Material

The filter material used is Delbag 99/98 Microsorban* which is made of mats of polystyrene fibers with diameters of $1\mu\text{m}$ and less. This material was chosen because of its low flow resistance, high particle collection efficiency, and ease of analysis for radionuclides. At the nominal flow rate of $1500\text{ m}^3/\text{hr}$ the pressure drop across the filter is approximately 3000 N/m^2 . For a face velocity of 0.42 m/sec , corresponding to the above nominal flow rate, the penetration of a $0.3\mu\text{m}$ diameter particle is 0.25% . At all other face velocities, the penetration is less than $0.25\%^2$.

Due to its poor mechanical strength, a backing material must be used beneath the Microsorban. This combination is held in a hinged filter holder made of stainless steel. The filter material is supported on a 12.5 mm mesh screen with a 1.5 mm diameter wire. The filter holder is taped to the air sampler to eliminate any air leakage around its edges. A hinged metal frame on which a sponge rubber gasket is mounted is then used to firmly clamp the filter holder to the air sampler.

Flow Calibration

The volumetric flow rate of air through the filter is determined during each test run by the rate of rotation of an unloaded fan blade located in the exhaust duct. The unit consists of a modified cup type anemometer with the cup rotor replaced with a 0.25 m diameter six blade aluminum fan. An electromechanical counter records one count for every 60 revolutions of the

* Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U. S. Atomic Energy Commission to the exclusion of others that may be suitable.

fan blade. Given the number of counts per minute, the air flow rate can be determined from a calibration curve such as that shown in Fig. 2. Notice that the rate of revolution of the fan blade is a linear function of flow rate. An independent measurement of the flow rate is obtained from the pitot tube located 1.2 m upstream from the fan blade.

Prior to deployment in the field, each air sampler is calibrated with a Meriam laminar flow element which is a primary flow measuring element. A schematic of the calibration setup is shown in Fig. 3. The laminar flow element is connected to the sampler by a straight duct 10 diameters long as recommended by the manufacturer and by a transition piece above the filter which is not present when the air sampler is in field use. The flow rate is determined by measuring the pressure drop across the laminar flow element which is accurate to within 0.5% . The overall accuracy of the calibration technique is estimated to be within 5% . Both the counts/minute from the modified anemometer and the pitot tube reading of the velocity pressure are calibrated against the laminar flow meter over a range of flow rates as shown in Fig. 2. Note that the velocity pressure as measured by the pitot tube is a function of the air density as well as volumetric flow rate.

Experimental Procedure

Prior to each test run the Microsorban filter material is placed into a decontaminated filter holder. The filter holder is then placed into a sealed plastic bag for transportation to the site of the air sampler. The air sampler has previously been decontaminated and sealed by taping an aluminum cover in place. The aluminum cover is shown in the foreground of Fig. 1 in its storage location on the side of the sampler. Several hours before a test run, the filter holder is removed from its plastic bag and placed in the

air sampler by raising the hinged framework on which the aluminum cover is taped. After all of the edges of the filter holder have been taped in place, the hinged framework is lowered and secured. A thermal radiation shield is attached 80 mm above the aluminum cover to ensure that the Microsorban filter material does not exceed its maximum operating temperature of 60° while installed in the sampler.

At the start of the test run, the aluminum cover is removed and the centrifugal blower is turned on. The counts from the modified anemometer, as well as the pitot tube reading, along with the atmospheric pressure and temperature are recorded at the beginning and end of the test run which normally lasts for a period of 4-5 hours.

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FIGURE CAPTIONS

- Figure 1. Photograph of the LLL ultra high volume (nominal $1500 \text{ m}^3/\text{hr}$) air sampler.
- Figure 2. Typical calibration curve for the ultra high volume air sampler. A fan blade in the exhaust duct produces the count/min and sec/30 counts data. Both the fan and the pitot tube are used for field measurements of air flow.
- Figure 3. Schematic diagram of the ultra high volume air sampler and primary calibration apparatus showing typical pressure drops throughout the combined system.

Fig. 1
Goluba, R. W.

Fig. 2
Goluba, R. W.

SCHEMATIC OF CALIBRATION SETUP FOR THE ULTRA HIGH VOLUME AIR SAMPLER

Fig. 3
Goluba, R. W.

Resuspension Element Status Report

IV. Saltation and Creep Sampler

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Introduction

The motion of windblown sand close to the ground can occur by two mechanisms. The first of these mechanisms is creep which is the rolling motion of sand along the ground. The second mechanism occurs when the windspeed picks up and sand "hops" along the ground in short, arching paths; this is saltation. The measurement of these mechanisms of sand movement, along with the resuspended sand, is necessary for the understanding of the total flux of wind-borne soil. In previous studies^{1,2,3} various devices have been used to measure saltation and creep. In order to develop an efficient sampling unit several of the above devices were evaluated to determine the most efficient, and then modifications were made to further increase efficiency. The testing of the units was conducted under controlled conditions at Livermore so that more reproducible tests could be conducted than would be possible at NTS.

Discussion

Two types of samplers were tested, one was of the funnel type, developed by Sehmel¹, which was built at Livermore and is shown in Fig. 1. The second (Fig. 2) was of a type developed by Bagnold², and was borrowed from Dale Gillette of the National Center for Atmospheric Research. The two samplers were of equal heights, but the funnels had a cross sectional width of approximately 7 cm while the Bagnold collector had a 1 cm width. Saltation was experimentally simulated by the use of a $\frac{1}{2}$ hp blower with its nozzle angled at 45° to the ground. The blower was used to lift the soil into the air where it was then carried by its own momentum, and the momentum of the prevailing wind towards the two samplers. The samplers were spaced six feet apart, with the distance measured perpendicular to the wind, so that the wakes would not interfere with the flow pattern around each sampler. The blower was also situated in such a manner that each sampler was exposed to the same two-phase flow. This experimental situation did not perfectly duplicate the actual physical conditions of saltation, but it was capable of giving quantitative results about the relative effectiveness of each sampler.

Tests were conducted with collection times varying from 45 minutes to 90 minutes. Since these tests were aimed at strictly comparing the two samplers, the ratio of mass collected by each sampler was the only information required; however, because the funnel sampler results could conveniently be broken down into mass vs height data, this was also done. In Table 1 the mass collected vs height for the funnel collector is presented along with

the total mass for each sampler, for a typical run. For our tests, the mass collected by the Bagnold collector was consistently .85 that collected by the funnels. However, because of the larger collection area of the funnel sampler, it appears to be approximately only 20% as efficient as the Bagnold collector. Tests conducted by Gillette³ on the Bagnold sampler show the collection efficiency to be 60% while Horikawa and Shen⁴ found an efficiency of 53%. Efficiency is defined as the actual mass of sand collected/mass of sand that should have passed through the collection area. The above results imply that the funnel sampler would be only 12% - 15% efficient; this is in contradiction to the initial assumption made about the funnel sampler¹. The results, however, agree with recent findings by Sehmel⁵.

Other conclusions about the samplers can be drawn from our experimental results which can be applied in the evaluation of new sampler designs. Our experiments show that a difficulty arises when a circular sampler is used, since there is a degree of uncertainty involved in determining the width of the collection area. This complicates the actual determination of the collection efficiency for the unit. It is also evident from our observations that a detailed analysis of the particle paths would have to be performed to fully understand the funnel type sampler. The experiments further point to a very practical consideration in designing a sampler, and that is the ease of sample retrieval in the field. In the case of the Bagnold collector, the sample was collected in a box below the unit; this box could conveniently be removed. In the case of the funnel collector, each of the fourteen funnels had to be removed and sealed in a clean plastic bag; during field operations this proved to be a time consuming process.

Both of the above mentioned units proved to have the same disadvantage. Both units are more appropriate for soil erosion studies and as a result are most suitable when at least 50 g of sand are collected. For a typical test conducted at Livermore with our experimental apparatus, on the order of 3 g of sand was collected by each sampler. For the present resuspension study being conducted at NTS, it appears that even smaller quantities of sand than those found at Livermore would be moved by saltation and creep. The above conclusion is based on the type of ground cover at the GMX site. At this site there is an abundance of large pebbles on the desert pavement as well as a profusion of desert plants; from personal observation and comparison to Chepil's results⁶ it appears the GMX has a stable soil. It, therefore, was appropriate to develop a modified sampler more suitable for collection of small amounts of sand.

A modified saltation sampler was designed and built at Livermore. The sampler, shown in Fig. 3, is basically a Bagnold type of sampler. Modifications have been made which enhance the particle flow through the collector and make the particle retrieval easier. The unit is described in more detail below.

The modified sampler developed for use at NTS is 30.5 cm high compared to a height of 83 cm for the Bagnold unit. The shorter height was chosen because it has been found that even in strong dust storms over 90% of the sand moved by saltation is collected below 30 cm². In order to increase collection efficiency the modified unit has vented collection passages for the sand. By allowing the air to flow through the unit the pressure in the unit more nearly approximates the ambient air so particle paths are less affected. The vents are shown in Fig. 3. Each vent is covered with two

layers of 300 mesh screen so no particles greater than 10 μ m can escape through the unit. The sand upon entering the unit is collected in containers with one container for each collection slot and a container for the creep slot. Upon termination of each run the containers are capped and numbered; this collection process is simple and convenient even on strong wind days and there is a minimum chance of losing the sample. Five of these units have now been built and are deployed in the GMX area at NTS.

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TABLE I

Funnel Height (cm)	Mass (g)
3.81 cm	1.30
7.62	.56
11.43	.29
15.24	.37
19.05	.12
22.86	.11
26.67	.04
30.48	.03
34.29	.02
38.1	.01
41.91	.03
45.72	.01
49.53	.04
53.34	.04
1) Total	2.97 g (funnel collector)
2) Total	2.50 g (Bagnold collector)

Length of run 60 minutes

FIGURE CAPTIONS

- Figure 1. Photograph of the funnel type sampler fabricated at LLL based upon the design of Sehmel¹. The "funnels" are individually removable.
- Figure 2. Photograph of the Bagnold type of sampler used for comparison tests. This sampler was borrowed from Gillette of the National Center for Atmospheric Research. The vane on top of the sampler keeps the entrance pointed into the wind.
- Figure 3. Schematic diagram of the LLL saltation and creep sampler. The saltation collection paths are vented to avoid stagnation. Five such samplers are now deployed at NTS.

Fig. 1
Reichman, J. M.

Fig. 2
Reichman, J. M.

Fig. 3
Reichman, J. M.

Resuspension Element Status Report

V. In Situ Optical Particle Size Analysis of Ambient Aerosol

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DESCRIPTION

The Climet Particle Analyzer* is a light-scattering instrument capable of sizing and counting particles in the range 0.5 to 10 μ m in diameter. Fig. 1 shows a simplified drawing.

The small, carbon-vane pump draws particle-laden air into the unit at a fixed rate of 0.25 cfm. Clean "purge air," which has been filtered to remove particles above 0.1 μ m, is introduced as a sheath around the incoming stream.

Particles next enter the view volume, defined by the cross section of the particle air stream and vertical extent of an intense light beam. The Climet view volume occupies approximately 6×10^{-4} cm^3 and is located at one focus of an elliptical mirror. For concentrations below about 35 per cm^3 , particles can be assumed to enter the view volume individually.

*Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U. S. Atomic Energy Commission to the exclusion of others that may be suitable.

As it passes through the view volume, the particle scatters light in all directions. A portion of the light scattered in the near-forward direction is reflected by the mirror to its other focus, where a photomultiplier tube is positioned. A light-absorbing cone blocks the direct beam and allows virtually no light to reach the photomultiplier when particles are not present in the view volume.

The intensity of each light pulse collected by the tube is a function of particle size, shape and refractive index. Each pulse from the photomultiplier is amplified and applied to a network which linearizes the relationship between pulse height and particle size (assuming uniform spherical shape and constant index of refraction).

For particle counting, the Climet pulse output is fed into a multi-channel pulse height analyzer. This instrument classifies each pulse according to amplitude and maintains a running total in 200 channels which represents the size range 0.5 to 10.5 μm . Fig. 2 represents a typical display as seen on the height analyzer's CRT. In addition to the CRT display, a small line printer provides hard-copy output of the channel-by-channel particle count.

CALIBRATION

Although not shown on Fig. 1, the Climet Particle Analyzer includes a built-in system for field calibration. Light pulses of known magnitude are fed via light-pipe to the photomultiplier tube. In the calibration mode, output from the tube is displayed on a front-panel meter. Photomultiplier high-voltage is adjusted to obtain the correct output.

Since the field calibration is only a relative one and serves to compensate for component aging, some absolute calibration is also required. For this purpose various sizes of polystyrene latex spheres were employed in conjunction with a Royco aerosol generator. Mono-disperse aerosols in seven sizes were produced and individually fed to the Climet Analyzer. Each of these aerosols were displayed on the CRT as a nearly Gaussian distribution of particle sizes. The center of each distribution was chosen as the representative size for that aerosol. A plot of the calibration curve derived from this procedure is shown in Fig. 3. The straight-line fit is reasonably good and use of the linear relationship simplifies data reduction.

FIELD EXPERIMENTS

The Climet Particle Analyzer is configured as shown in Fig. 4 for data collection at the Nevada Test Site. The optical components, lamp and photomultiplier are mounted separately from the air pump, flow regulators and electronics. The two enclosures are joined by approximately 15 feet of air tubing and electrical cable. Separation of the two units allows sampling at heights up to 20 feet off the ground while still permitting convenient flow adjustment and calibration checks.

The optical unit is fitted with a wind vane coupled to the air inlet tube. Although the inlet is kept pointing into the wind by this vane, there is no simple way to maintain the inlet velocity equal to the wind speed. Maintaining equal velocities (isokinetic sampling) insures that the particles which enter the inlet tube are a true representation of

ambient concentrations. If the inlet velocity is significantly lower than the wind speed, a greater proportion of large particles than are actually present per unit volume of air will be collected. Similarly, fewer large particles are collected when the inlet speed exceeds that of the wind.

Since the Climet flow rate is only slightly variable about a fixed value, the intake velocity cannot be adjusted by varying the flow rate. A varying flow rate would also make it difficult to determine the volume of air sampled.

The other possibility for changing the inlet velocity involves changing the inlet diameter while holding flow rate constant. Unfortunately, attempting to accomplish this on a real-time basis as the wind speed changes is a complex problem. The present setup, described below, makes only limited use of this principle.

The wind vane is constructed to accept three different-sized copper tubing inlets. At the normal flow rate, the tubes provide intake velocities of approximately 2.5, 4, and 7 m/sec. During field operation, an attempt is made to fit the vane with the tube which has an intake velocity most closely approximating the mean wind speed. If a significant wind change occurs, the inlet tube is changed accordingly. Inlet size is recorded for each data run.

When sampling at the Nevada Test Site, the two Climet units are placed in an open area usually with the main chassis on the ground and the sensor unit on a tripod. About 75 feet of coaxial cable connect the Climet to the pulse height analyzer, CRT and printer mentioned above. These latter components are housed in a small trailer.

Following a 30 minute warm-up for all equipment, the Climet field calibration and flow rate are checked and adjusted. At this time the applicable inlet tube is installed. After all pulse height analyzer channels are cleared, pulse counting is initiated. Normally particle counts are collected for 120 seconds — timed automatically by the equipment. At the completion of the run cumulative counts in each of the 200 channels are printed on paper tape by the line printer. The channels are again cleared and the equipment is ready for the next run. Runs are usually scheduled each 10 minutes to correspond with the 10-minute intervals during which meteorological data is being collected. Pertinent data such as time, sampling height and inlet size are recorded manually on the paper tape for each run.

DATA REDUCTION

During the time between 18 April and 2 August 1973, data from 132 Climet Particle Analyzer runs were collected at the Nevada Test Site. Since the paper tape output of the line printer does not allow any direct means of automated processing, data from all tapes were keypunched onto punch cards. Each run is now characterized by 30 cards — 29 for the channel-by-channel counts and one which contains date, time, sampling height, inlet size and any appropriate comments included by the operator.

To date, only a very preliminary data reduction effort has been completed. Computer programs are being written to operate on the large volume of particle data. Several areas of investigation are planned using the Climet data alone. These include log-normal distribution fits and analysis of

concentration change with height. By far the most important phase of the data analysis, however, is the study of correlation between the particle data and the meteorological measurements made at the same time.

Significant meteorological parameters which correspond in time with each Climet run have also been put on computer cards. Storing both the particle and meteorological data bases in the computer will allow fast, efficient manipulation of the data. The primary goal of this data analysis will be detection of correlations between particle concentration and one or several meteorological parameters. As yet, the programs required for this effort have not been completed. We have, however, manually assembled some of the data.

Fig. 5 displays data from three Climet runs which represent the full range of particle concentrations encountered thus far. The meteorological parameters listed are typical of those to be used in the correlation studies. While quantitative conclusions cannot be drawn from these limited data, it is apparent from the figure that, at least in a general sense, there exists a correlation between various wind parameters and particle concentration. It is our intent to identify and model the quantitative relationships which exist between particle loading and micrometeorology.

FIGURE CAPTIONS

- Fig. 1. Simplified schematic of the Climet Particle Analyzer and associated equipment. A particle present in the view volume at one focus of the elliptical mirror scatters light to the photomultiplier at the other focus.
- Fig. 2. Drawing of a typical CRT display after a Climet Particle Analyzer run. The actual CRT display contains 200 vertical bars, one for each channel. Height of each bar designates the number of counts in that channel. Data is also recorded by digital line printer.
- Fig. 3. Calibration curve derived from measurements made in the laboratory with polystyrene latex spheres. Seven sizes with diameters between 0.48 and 7.3 μ were used.
- Fig. 4. Typical field setup at NTS for the Climet Particle Analyzer. The wind vane atop the sensor unit keeps the inlet tube pointed into the wind.
- Fig. 5. Concentration data from three Climet runs representing the range encountered between April 18 and August 2, 1973. As expected, wind parameters calculated for each run exhibit the same general trend as the particle data.

Fig. 1
Koval, J. S.

Fig. 2
Koval, J. S.

Fig. 3
Koval, J. S.

Fig. 4
Koval, J. S.

Fig. 5
Koval, J. S.

Resuspension Element Status Report

VI. Results and Data Analysis

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The long-range goal of the resuspension studies is to produce a set of equations which can be used to predict the time-dependent average concentration of resuspended material downwind from a source of any geometrical configuration and soil surface characteristics. A great deal of work remains to be done before this goal can be accomplished. Results which are currently available are limited and/or have not been fully analyzed; they are presented in the nature of a status report and as representative of the types of measurements being made. (Preliminary results of the *in situ* optical size measurements are presented separately in Section V.)

These preliminary results have already found practical application in another AEC program which has the goal of evaluating the potential hazards associated with the reoccupation of the Enewetak Atoll. The use of the NTS data and expertise to predict the levels of resuspended air activity arising from the aged Enewetak source is treated in Appendix A of this report.

REECO Air Sampling Measurements

One of the short-range goals of the total NAEI program was to determine if resuspension created an occupational hazard at the fence boundaries of the GMX area. To accomplish this goal, Reynolds Electrical and Engineering Company (REECO) collected and analyzed for ^{239}Pu 503 air filter samples collected from February, 1971 to July, 1972; 1 200 control samples were also taken at Mercury at Building 214 during the same period. At GMX, two stations were maintained throughout this time period. Both were located on the fence perimeter, one to the northeast and one to the southwest of ground zero.

* Although results are frequently quoted here and elsewhere for ^{239}Pu , the usual methods of analysis cannot distinguish between ^{239}Pu and ^{240}Pu .

Two additional stations, located on the fence perimeter to the southeast and northwest of ground zero, were also operated from February through April, 1971. All samples were approximately 1m above ground. The filter face was vertical and at each location was oriented toward ground zero. The sample collection period was generally 48 hr.

The measurement results are summarized in Table 1 and the cumulative distributions for the GMX NE, GMX SW, and Building 214 stations are plotted in Figure 1. The data range over four orders of magnitude and are obviously not normally distributed. If the data were lognormally distributed, they should fall on a straight line when plotted as in Figure 1. The measurements at GMX NE approach this condition, but data from the other two stations do not.

Unfortunately, most well-known statistical analysis procedures are not applicable to data that are not normally distributed, and it has been our experience that the use of such tests when the data do not meet this criterion frequently leads to erroneous conclusions. We have, therefore, used in our analyses only nonparametric, or distribution-free statistical tests.³

The groups of samples which had detectable quantities of ²³⁹Pu from the three major stations were tested to determine the statistical probability that the three groups of samples were drawn from the same population. The Kruskal-Wallis one-way analysis of variance was used and the result was $p = < 10^{-5}$. The same statistical test was also applied to the three combinations of sample groups taken two at a time. The results are given in Table 2, and indicate that the probability is negligible that any of the sample groups were drawn from the same population.

The conclusion is, therefore, drawn that the ²³⁹Pu deposited at the GMX site more than 15 years prior to the measurements still represents a significant resuspension source. At the perimeter of the fence surrounding the exclusion area, however, the average air concentration of resuspended ²³⁹Pu is only a small fraction of the presently accepted maximum permissible concentration for occupational exposure (Table 1).

The difference between the results for the GMX NE and GMX SW stations is of particular interest. A gross summary of the continuous meteorological data⁴ taken during the same time period is shown in Figure 2. The wind direction was most frequently toward the GMX SW station; however, the strong

winds (> 10 mph) had a markedly different distribution with direction and most frequently were toward the GMX NE station. This gross correlation of air concentration with wind speed is predicted by theory,^{5,6} and has been reported for a single station in the vicinity of the ²³⁹Pu contamination at the Rocky Flats Plant.^{7,8} It was not demonstrable, however, during our studies at NTS following the accidental venting of the Baneberry underground nuclear explosion.⁹ More detailed studies using the wind velocity data and the individual measurements at the GMX NE station will be done to examine further the relationship between wind speed and air concentration of ²³⁹Pu.

An additional question of considerable interest is whether the measurements at the control station (Building 214) are representative of world-wide background values, or are influenced by local NTS sources. Data for background surface air concentrations from several sources are tabulated in Table 3, and cover essentially the same time period as the Building 214 measurements. It is somewhat difficult to compare these measurements as the sampling periods range from 48 hr to one month to one quarter, and the data spread for a single station appears to be inversely related to the sampling period. A statistical test which is sensitive to any difference in the distribution of samples is therefore undesirable. We can ask the more narrow question of whether the different groups differ in central tendency by applying the median test.

Four sets of data were considered: 52 measurements for 9 stations of the EPA Radiation Alert Network, 96 measurements for 6 stations of the HASL Surface Air Sampling Program, 36 measurements for 2 sites of the LLL Environmental Monitoring Program, and 200 measurements for the REECO Building 214 station. For the latter set, all non-detectable values were set equal to zero. The median test was applied to the three sets of "background" data (EPA, HASL, LLL); the resultant probability that all three sets were drawn from the same population is > .90. For all four sets of data, the resultant probability is $< 10^{-4}$ that all four were drawn from the same population. Therefore, although there must be some reservations about the application of any statistical test to such disparate sets of data, the tentative conclusion is drawn that the air concentrations of ²³⁹Pu in Mercury are influenced by the local NTS sources.

Distributions of Mass and Radioactivity with Particle Size

The distribution of mass and radioactivity as a function of particle size of the soil surface, and materials moving in saltation and suspension is an important aspect of the resuspension research program. Such measurements are expected to help define the erodibility of different soil surfaces,¹⁴ to aid in the development of resuspension models,¹⁵ and to provide input to models of lung dynamics.¹⁶ Particle size distribution data for the GMX area soil are given elsewhere in this report by Tamura as part of the Soil Element; progress in developing a method to collect saltation particles for analysis is given by Reichman in Part IV of the Resuspension Element.

This section is concerned primarily with particle size distribution data for aerosols at the GMX site. Samples were collected by deploying five Andersen 2000* high-volume cascade impactors at the GMX in the location shown in Fig. 2 of Part I of this report. The samplers were on an arc 250 ft. from the GMX bunker with a spacing of 50 ft.

The Andersen high-volume cascade impactor has four stages plus an after filter. At the design flow rate of 20 ft³/min, the effective cutoff diameters of the four stages are 7.0, 3.3, 2.0, and 1.1 μm .¹⁷ Each cascade impactor was mounted in a covered housing of the type recommended by the Environmental Protection Agency¹⁸ for the determination of suspended particulates in the atmosphere. Air intake into the housing was at a height of 1 m.

The first set of samples was collected by running the samplers continuously over the 36-day period from July 7, 1972 to August 12, 1972. This sampling period is longer than desirable, but was chosen to ensure that analyzable quantities of ²³⁹Pu would be collected on each stage of the impactor. The pressure drop (corrected for altitude) across each impactor head was initially set to achieve a flow rate of 20 ft³/min according to the manufacturer's calibration curve for each individual impactor unit. The pressure drop across each unit was periodically recorded during the run but was not readjusted.

* Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Atomic Energy Commission to the exclusion of others that may be suitable.

The 25 samples were weighed at Livermore to determine the concentration of particulates. Other analyses were done by Myers of the McClellan Central Laboratory;¹⁹ these included the radiochemical determinations of ²³⁸Pu, ^{239,240}Pu, and ²⁴¹Am and the mass spectroscopic analysis of the separated plutonium from two of the after filters.

The mass and radiochemical data for all stages of each sampler were combined in order to calculate the total air concentration for each species. These data are given in Table 4. The mean result for the concentration of ^{239,240}Pu is considerably higher than the REECO results which is to be expected due to the proximity of the impactors to areas of greater ground deposition. A gross summary of the continuous wind data⁴ taken during the high-volume cascade impactor run is shown in Fig. 3. The distribution of winds with direction is quite similar to that shown in Fig. 2, but the most frequent direction of strong winds was rotated about 10° clockwise. This is consistent with the higher radionuclide concentrations indicated by samplers number 4 and 5.

The mass and radionuclide activities on each stage of one of the samplers are tabulated in Table 5. Data for the individual stages of each cascade impactor were used to determine the distributions of mass and activity as a function of particle size. This was accomplished by transforming the data with the dependent variable the cumulative per cent of the mass or activity associated with particle sizes less than the effective cutoff diameter of the preceeding stage. The logarithm of the effective cutoff diameter of the preceeding stage was the independent variable. Because the data were to be fit to a lognormal distribution and 100% has no meaning in such a distribution, and because the effective cutoff diameter or largest size particle which enters the total sampling system is unknown, only four data points per sampler were available for fitting. These four data points were fit to a lognormal distribution rising Bevington's non-linear least-squares fit routine, CURFIT.²⁰ The cumulative probability of a normal distribution needed for input to CURFIT was supplied by Bevington's AGAUSS routine modified to calculate the probability from -∞ to x rather than from -x to x. Data were weighted by 1/σ² of the gravimetric or radiochemical determination error. Two parameters are derived from this fitting process, the geometric mean particle size, $\bar{x}_g$ and the geometric standard deviation, σ_g. Sixty-eight per cent of the total mass or activity is therefore between the particle sizes of $\bar{x}_g/\sigma_g$ and $\bar{x}_g\sigma_g$. If the data points are indeed well fit

by a lognormal distribution, then $\bar{x}_g$ is also equal to the median diameter. Because the cascade impactor separates particles by inertial impaction and is calibrated for unit density spheres,¹⁷ $\bar{x}_g$ is also equal to the mass median aerodynamic diameter (MMAD) or activity median aerodynamic diameter (AMAD). These are the standard parameters needed for input to models of lung dynamics.¹⁶

The results of the particle size distribution fits are presented in Table 6. The data were generally well fit by a lognormal distribution as measured by the reduced chi-square of the fit. Sampler 5, however, was an exception. In this case, the data for radionuclides were not well fit by a lognormal distribution because of a large amount of activity present on the fourth stage (1.1 to 2.0 μ m). The difference between the five impactor units was most pronounced for the values of σ_g . Some of this difference is probably due to fluctuations in the flow rate which were recorded during the run; fluctuations were most pronounced for sampler number one.

In all cases the distribution with particle size was so broad as indicated by the large values of σ_g that the fit for any individual impactor was very insensitive to σ_g as indicated by the high standard deviation of the determination of σ_g . With the goal of obtaining a better fit, the data from all five impactors were combined in two ways: 1) the total mass or activity on each stage for all five impactors was used to derive a new cumulative distribution and 2) the cumulative per cent distributions for the five impactors were averaged. The results of fitting a lognormal distribution to these data are also shown in Table 6; these fits were more sensitive to σ_g .

The fits to the combined data, and generally the fits for any individual impactor, indicate that there is no difference in the distribution of activity with particle size for the three species ²³⁸Pu, ^{239,240}Pu, and ²⁴¹Am. The total mass, however, is distributed distinctly differently with a smaller median aerodynamic diameter and a larger geometric sigma. The mean results for mass and ^{239,240}Pu are plotted in Fig. 4.

The values of σ_g derived from these data are larger than those usually reported for aerosols of uranium and transuranium compounds,²¹⁻²⁴ and are also larger than those explicitly considered in the ICRP Task Group on Lung Dynamics report.¹⁶ There are several possible explanations for this difference. First, we must maintain some reservations about the performance of the Andersen 2000 cascade impactor itself. This is a relatively new product and has received only limited field evaluation;^{17,25,26} Sehmel²⁶ has indicated that there may

be significant wall losses and reentrainment problems. We are presently conducting laboratory and field studies to evaluate further these effects and their influence upon results derived from this impactor.

There are, however, very few data which are directly comparable to those reported in Table 6 for plutonium and americium. Nearly all results reported in the literature pertain to laboratory-generated aerosols or "field" aerosols. The word field is somewhat of a misnomer, however, as it has been used to describe aerosols in laboratories where plutonium is processed. A few results are available for more nearly equivalent situations from measurements made at NTS. Cascade impactors were used to study resuspended plutonium aerosols during Project 57.²⁷ A median diameter of about 1.5 μ m was reported. No specific data were given for σ_g , but it was stated that the data were strongly biased because many samples with a σ_g of 10 or higher were discarded. Results of particulate studies were also reported for the initial cloud of plutonium aerosol produced by the non-nuclear Double Tracks and Clean Slate 1 events of the Roller Coaster Operation.²⁸ Much of the plutonium in these initial, explosion-produced clouds was associated with particles larger than could be sampled by cascade impactors, but it was stated that in the region where sizing by impactors is possible, a value for σ_g of 6 was obtained.

Even higher values of σ_g are given in Table 6 for mass of suspended particulates. Considerable data for comparison are available from studies conducted for the National Air Surveillance Networks (NASN)^{29,30} and by Lundgren.³¹ The results of these studies are that values of σ_g as large as 15 are not unusual in either polluted or non-polluted atmospheres even over time periods of a few hours. Distributions other than lognormal have been proposed for natural aerosols.^{32,33} The well-known Junge distribution³², for example, predicts an essentially flat volume distribution as a function of particle size between 0.1 and 10 μ m. Such a distribution is therefore compatible with large values of σ_g derived from a fit to a lognormal mass distribution.

The most immediate application of the results in Table 6 is the determination of the fraction of the resuspended plutonium aerosol at GMX which would be expected to undergo pulmonary deposition. This fraction is approximately 0.2 based upon the ICRP Task Group on Lung Dynamics model.¹⁶

The possible effect of wall losses and reentrainment in the cascade impactor used would be to overestimate the relative amount of material in the smaller size ranges separated by the impactor.²⁶ The possible bias in the respirable fraction is therefore conservative; the correct value may be lower, but it is very unlikely to be higher. Results have also been reported for the respirable fraction of the resuspended plutonium aerosol in the vicinity of the Rocky Flats Plant.³⁴ In these studies either an horizontal elutriator or a cyclone was used to remove all but the respirable fraction which was then collected on an after-filter. Results for the respirable fraction were 0.25 ± 0.09 .

A question of considerable interest is whether the resuspended plutonium aerosols show a marked difference in specific activity as a function of particle size. This can be examined in an approximate way by calculating the specific activity (activity of $^{239,240}\text{Pu}$ per gram of total particulates collected) for each stage of the impactor. Such data for all five impactors are given in Table 7. There is no obvious correlation of specific activity with particle size. Considerable caution must be exercised in interpreting the data, however, because the resuspended aerosols are diluted by natural, inert aerosols transported into the area and which undoubtedly have a different distribution of mass with particle size. The average specific activity, 890 dpm/g, is about one-third of that found in the soil in close proximity to the cascade impactors. The specific activity concept has been expanded further in Appendix A of this report to derive an interim, approximate resuspension model applicable to aged plutonium depositions.

The $^{239,240}\text{Pu}$ specific activities in this environment are very low relative to that of pure $^{239}\text{PuO}_2$. The specific activity of $^{239}\text{PuO}_2$ is 1.2×10^{11} dpm/g. The total activity present on all five stages of sampler number 4 (Table 5) could have been contained in a single $^{239}\text{PuO}_2$ particle of $15\mu\text{m}$ actual diameter, or 4 $10\mu\text{m}$ particles, or 30 $5\mu\text{m}$ particles, or 3500 $1\mu\text{m}$ particles. The aerodynamic diameter of such particles would be approximately 3.4 times larger.

One of the 25 results tabulated in Table 7 is anomalously high. This is for the 1.1 to $2.0\mu\text{m}$ stage of sampler 5. The relative excess of $^{239,240}\text{Pu}$ activity on this stage is about 250 dpm. In order for a single, pure $^{239}\text{PuO}_2$ particle to produce this effect, it would require an actual diameter of $7\mu\text{m}$, which is equivalent to an aerodynamic diameter of about $24\mu\text{m}$. Theoretically, such a large particle should not be present on this stage, but may have been

reentrained from a previous stage.²⁶

Mass spectroscopy analyses were also performed by Myers¹⁹ on the plutonium separated from two of the cascade impactor back-up filters. The results are given in Table 8. The weighted mean of the mass ratio of ^{240}Pu to ^{239}Pu is 0.0545. The equivalent activity ratio is 0.202. A more useful way of expressing the latter information is that 17% of the total $^{239,240}\text{Pu}$ activity at GMX, which is frequently referred to as only ^{239}Pu , is actually due to ^{240}Pu . The mass ratios in Table 8 plus the ^{238}Pu and $^{239,240}\text{Pu}$ activity measurements can be used to deduce the complete isotopic composition of the smaller airborne particles at the GMX site. These calculations were made and the results given in Table 9 where they are also compared to the typical plutonium values published by Del Pizzo, Owen, and Putzier.³⁵ The calculated values for GMX plutonium agree quite well with the typical plutonium values, except for a deficiency of ^{238}Pu . The apparent deficiency of ^{241}Pu is partly due to radioactive decay of the aged GMX plutonium.

The absolute mass of the ^{241}Pu on the back-up filters can be used to calculate the ^{241}Pu activity. This can be combined with the separately determined activity of ^{241}Am , which is the daughter product of ^{241}Pu , to calculate the age of the plutonium. We do in fact know, of course, how long the plutonium has been present at GMX, but such a calculation serves a useful purpose. It provides a check on the various types of analyses performed; and, if we assume that the analyses have no systematic errors, it provides a sensitive way of testing for the differential behavior of plutonium and americium in a natural environment.

The relevant equation is

$$\frac{A_1}{A_2} = \frac{\lambda_2 - \lambda_1}{\lambda_2 \left[1 - e^{-(\lambda_2 - \lambda_1)T} \right]}$$

where A is activity, λ is the radiological decay constant, T is the age of the plutonium since purification, and the subscripts 1 and 2 denote the parent and daughter radionuclides, respectively. Such a calculation is highly dependent upon the half-lives of ^{241}Pu and ^{241}Am ; published values of both have changed substantially in recent years. For ^{241}Am , the value used was 433 ± 2 years³⁶, and for ^{241}Pu 15.16 ± 0.19 years.³⁷ The calculated activity ratios and ages are given in Table 10. The actual weighted average

time since deposition of the GMX plutonium is 16.7 years. The result for sampler 3 is statistically different from this at the 98% confidence level. As both calculated values are shorter than the true time since deposition, a possible interpretation is that americium has left that fraction of the upper soil layer which is resuspendible at a faster rate than has plutonium, or is preferentially associated with larger particles. The relative deficiency of americium is about 20%.

Considerable attention has been devoted elsewhere in this volume to the determination of the ratio of $^{239,240}\text{Pu}$ activity to ^{241}Am activity in soil samples, and the correlation between the two measurements. The 25 data points for the cascade impactor samples were also examined in a manner similar to that used for soil samples. The correlation between the two measurements was calculated using the nonparametric Spearman rank method. The resulting value of r_s is 0.94 ($p = < 10^{-5}$). The data are shown in Fig. 5; the line is the least-squares fit to the data. The average value of the ratio is 10 ± 2 , which is statistically indistinguishable from that reported for the GMX soil samples. The $^{239,240}\text{Pu}$ to ^{241}Am ratios for the cascade impactor samples were tested to determine if there is a significant difference in the ratio for the five particle-size classes. The data were considered as five groups of related samples and tested by the nonparametric Friedman two-way analysis of variance.³ The result is that the null hypothesis cannot be rejected at the 0.05 level of significance.

A second set of samples was collected using the same five cascade impactors at the same positions. They were run continuously from April 24 to May 23, 1973, a period of 29 days. Complete results are not yet available for the activities of plutonium and americium; the gravimetric results are given in Table 11. The mass concentration during this sampling period was nearly a factor of two lower than the previous one, and the mass median aerodynamic diameter (MMAD) was also lower. Both observations are consistent with less material being resuspended from the soil surface; we anticipate that the concentration of resuspended plutonium was also lower. The values of σ_g derived from fits to a lognormal distribution are also a factor of two lower and imply a narrower distribution of mass with particle size. The mean data for the two runs are plotted in Fig. 6.

Additional cascade impactor samples are presently being collected. The sample collection periods have been shortened due to favorable experience in collecting more than adequate plutonium for analysis on every stage. One cascade impactor has been modified according to Sehmel's design;²⁶ this incorporates a rotating cowl which always faces the inlet into the wind. Results with this design will be compared with those using the EPA-recommended housing.

Ultra-High-Volume Air Sampling Results

The primary method of studying the resuspension of plutonium is the utilization of ultra-high-volume air samplers. The development of this sampler is described in Part III of this report. The purpose in developing this sampler was the ability to collect an analyzable sample of airborne plutonium within a few hours. This is highly desirable because, in order to relate resuspended airborne plutonium to meteorological parameters, it is almost essential to collect samples during time periods which are shorter than significant changes in meteorological regimes.

Ten ultra-high-volume air samplers have been installed in the GMX area. Two samplers were located at each of the five positions indicated in Fig. 2 of Part I of this report. All five positions are on a single radial which was selected because it corresponds to the most frequent direction of high-speed winds. One location is upwind, one is at the approximate center of the highest concentration of plutonium ground deposition, and the other three are downwind. An additional control sampler is located on the outskirts of Mercury.

The filter face of all samplers is parallel to the ground surface and is at a height of 1.3 m. Duplicate samplers were initially installed in order to examine the reproducibility of results obtained over short time periods; they are about 5 m apart perpendicular to the radial. If reproducibility proves to be satisfactory, the duplicate samplers will be deployed at other radials and/or heights to obtain additional information about concentration gradients.

The long-range goal in making such measurements is the derivation of a source term in terms of the fractional amount of activity injected into the air per unit area per unit time as a function of meteorological variables. This also requires the use of a suitable meteorological transport model which can be used to derive such a source term given the measured air concentrations

and the geometrical configuration of the source, plus the relevant meteorological parameters. Measurements of mass loading alone, which can be made over periods as short as one second, should greatly aid in deriving the relationships between the injected source term and meteorological parameters. It is, however, essential to demonstrate that any such relationships are consistent with measurements of resuspended plutonium concentration as well.

Several sample series were collecting during the past year. Because of the large number of samples involved and the higher priority given by the AEC community to the analysis of samples generated by the Enewetak survey, complete results for only two series were available for inclusion in this report.

Results for the two available series are shown in Figs. 7 and 8 where the average of the duplicate samples is superimposed upon the sample locations and the ground deposition isopleths. Data for the individual samplers are tabulated in Table 12. The average wind speed at 8m during the first run was 7.6 m sec^{-1} ; during the second it was 6 m sec^{-1} . Other basic and derived meteorological parameters for these runs are given in Fig. 1 of Part II of this report. There is a gross correlation between many of the wind speed related parameters and the concentration of resuspended plutonium. This is encouraging, but more data are obviously needed before a meaningful analysis can be attempted.

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Table 1. Results of the REECO measurements for ^{239}Pu air concentration at the GMX area and the control location at Mercury (Bldg. 214). Results are expressed in aCi/m^3 ; one aCi/m^3 equals $10^{-18} \mu\text{Ci/cm}^3$

Location	All samples			Samples with detectable ^{239}Pu			
	N	Arith. Mean ^a	Median ^a	N	Arith. Mean	Geo. Mean	Median
GMX NE	254	6100	560	236	6600	790	620
GMX SE	31	870	200	23	1200	400	320
GMX SW	186	580	120	156	690	190	150
GMX NW	32	310	180	29	340	230	200
Bldg. 214	200	230	87	147	310	140	130

Occupational MPC^b for 168-hr week (aCi/m^3):

Insoluble 10^7
Soluble 6×10^5

^a Non-detectable results set equal to zero.

^b Reference 2.

Table 2. Probability that the samples paired below were drawn from the same population. The statistical test used was the nonparametric Kruskal-Wallis one-way analysis of variance.

	GMX NE	GMX SW	BLDG 214
GMX NE	1.0		
GMX SW	$< 10^{-19}$	1.0	
BLDG 214	$< 10^{-27}$	0.009	1.0

Table 3. Results of measurements of ^{239}Pu in surface air at other locations during January, 1971 to June, 1972. With the exception of Rocky Flats, Colorado these results should reflect the "background" levels of ^{239}Pu due to the injection of debris into the stratosphere by nuclear weapon tests.

Location	^{239}Pu concentration, $\mu\text{Ci}/\text{m}^3$			
	Minimum	Maximum	Arith. Mean	Geo. Mean
<u>Radiation Alert Network, 48 contiguous states (EPA)^{a,b}</u>				
Phoenix, Arizona	16	110	63	55
Denver, Colorado	17	125	65	56
New Orleans, Louisiana	19	75	40	36
Baltimore, Maryland	14	82	40	32
Buffalo, New York	16	104	48	37
Gastonia, North Carolina	20	120	59	51
Pierre, South Dakota	15	104	51	41
Austin, Texas	c	c	37	36
Seattle, Washington	10	131	41	29
All 9 stations			50	40
<u>Surface Air Sampling Program, Latitude 51° to 25°N (USAEC, HASL)^{d,e}</u>				
Moosonee, Ontario	c	c	54	44
New York, New York	14	135	51	40
Salt Lake City, Utah	33	315	98	76
Rocky Flats #1, Colorado	c	c	4600	4000
Sterling, Virginia	18	118	45	37
Miami, Florida	c	c	57	45
Bimini, Bahamas	c	c	76	62
Six stations (Rocky Flats #1 excluded)			63	49
<u>Lawrence Livermore Laboratory (LLL)^{e,f}</u>				
Main Laboratory	5	340	58	37
Site 300	14	110	52	44
Both sites			55	40

^a Ref. 10.

^b Quarterly composite samples.

^c One or more results missing.

^d Ref. 11.

^e Monthly composite samples.

^f Refs. 12 and 13.

Table 4. Results of analyses for air concentrations summed over all particle sizes for the first high-volume cascade impactor samples. Five impactors were run at the GMX site continuously for 36 days from July 7, 1972, to August 12, 1972. Radiochemical analyses were done by Myers¹⁹.

Sampler ^a	Mass $\mu\text{g}/\text{m}^3$	^{238}Pu $\mu\text{Ci}/\text{cm}^3$	$^{239,240}\text{Pu}$ $\mu\text{Ci}/\text{cm}^3$	^{241}Am $\mu\text{Ci}/\text{cm}^3$
1	69	1.9×10^{-16}	1.0×10^{-14}	1.0×10^{-15}
2	59	2.5×10^{-16}	1.2×10^{-14}	1.1×10^{-15}
3	71	3.8×10^{-16}	2.0×10^{-14}	2.4×10^{-15}
4	74	7.2×10^{-16}	3.9×10^{-14}	4.1×10^{-15}
5	81	9.5×10^{-16}	3.6×10^{-14}	3.5×10^{-15}
Mean	71	5.0×10^{-16}	2.3×10^{-14}	2.4×10^{-15}

^a Samplers are numbered consecutively in a clockwise direction.

Table 5. Mass and radionuclide activities on each stage of high-volume cascade impactor number 4. Values below are the totals accumulated during a continuous sampling from July 7, 1972 to August 12, 1972. Radiochemical analyses were done by Myers¹⁹.

Stage	Mass mg	²³⁸ Pu dpm	^{239,240} Pu dpm	²⁴¹ Am dpm
> 7 μ m	580	18. $\pm$ 2.	990 $\pm$ 70	100 $\pm$ 10
3.3 to 7.0 μ m	350	5.9 $\pm$ 0.5	310 $\pm$ 20	31 $\pm$ 4
2.0 to 3.3 μ m	140	5.6 $\pm$ 0.6	280 $\pm$ 20	31 $\pm$ 3
1.1 to 2.0 μ m	120	4.5 $\pm$ 0.4	230 $\pm$ 20	26 $\pm$ 3
Filter	990	13.0 $\pm$ 0.9	740 $\pm$ 40	77 $\pm$ 7
Total	2180	47 $\pm$ 2	2550 $\pm$ 90	260 $\pm$ 20

Table 6. Results of analysis of the distribution of airborne mass and radioactivity with particle size at the GMX area. Data were obtained by running five Anderson 2000 high-volume cascade impactors continuously from July 7, 1972 to August 12, 1972. The $\pm$ values are the standard deviations of the fit.

Sampler	Total Mass		²³⁹ Pu		^{239,240} Pu		²⁴¹ Am	
	MMAD	σ g	AMAD	σ g	AMAD	σ g	AMAD	σ g
1	1.5 $\pm$ 0.3 μ m	30 $\pm$ 40	4. $\pm$ 1.4 μ m	30 $\pm$ 30	3.5 $\pm$ 0.6 μ m	30 $\pm$ 20	4. $\pm$ 1.4 μ m	30 $\pm$ 30
2	1.8 $\pm$ 0.4	20 $\pm$ 20	2.2 $\pm$ 0.3	8 $\pm$ 4	2.3 $\pm$ 0.3	9 $\pm$ 4	2.8 $\pm$ 0.4	9 $\pm$ 5
3	1.5 $\pm$ 0.2	6 $\pm$ 2	2.7 $\pm$ 0.3	6 $\pm$ 2	2.4 $\pm$ 0.2	7 $\pm$ 2	3.6 $\pm$ 0.3	4.1 $\pm$ 0.7
4	1.6 $\pm$ 0.3	20 $\pm$ 20	3.7 $\pm$ 0.4	8 $\pm$ 2	3.7 $\pm$ 0.3	9 $\pm$ 2	3.5 $\pm$ 0.5	8 $\pm$ 3
5	1.7 $\pm$ 0.3	20 $\pm$ 20	3.2 a	3 a	3.3 a	5 a	2.9 a	3 a
Total ^b	1.6 $\pm$ 0.1	15 $\pm$ 5	3.1 $\pm$ 0.2	5.4 $\pm$ 0.7	3.1 $\pm$ 0.1	7.4 $\pm$ 0.9	3.2 $\pm$ 0.2	5.4 $\pm$ 0.6
Mean ^c	1.6 $\pm$ 0.1	15 $\pm$ 5	3.0 $\pm$ 0.2	7 $\pm$ 1	3.0 $\pm$ 0.1	8 $\pm$ 1	3.2 $\pm$ 0.2	6.4 $\pm$ 0.9

^aThe reduced chi-square of the fit indicates that the data are not well fit by a lognormal distribution.

^bThe materials on each stage were summed for the five impactors, and the cumulative per cent distribution calculated.

^cThe cumulative per cent distributions were averaged for the five impactors.

Table 7. The specific activity of $^{239,240}\text{Pu}$, dpm/g, on the stages of the five high-volume cascade impactors. The samplers were run continuously from July 7, 1972 to August 12, 1972.

Stage	S A M P L E R					Mean
	1	2	3	4	5	
> $7\mu\text{m}$	480	420	940	1700	1200	950
3.3 to $7\mu\text{m}$	210	500	710	880	1200	700
2.0 to $3.3\mu\text{m}$	420	1300	570	2000	880	1000
1.1 to $2.0\mu\text{m}$	540	300	430	1900	3300	1300
After Filter	250	410	510	750	500	480
Mean	380	590	630	1400	1400	890

Table 8. Mass ratios of the plutonium separated from two of the Cascade impactor back-up filters. The impactors were run continuously from July 7 to August 12, 1972. Analyses were performed by Myers.¹⁹

Sampler	Plutonium Mass Ratios		
	240/239	241/239	242/239
2	0.0547 $\pm 0.5\%$	0.00189 $\pm 2.5\%$	0.000161 $\pm 7.5\%$
3	0.0520 $\pm 2\%$	0.00195 $\pm 8\%$	0.000479 $\pm 33\%$
Weighted Mean ^a	0.0545	0.00190	0.000163

^aWeighted by $1/\sigma^2$

Table 9. Isotopic composition of the plutonium separated from two of the Cascade impactor back-up filters. The impactors were run continuously from July 7 to August 12, 1972. Results are calculated for the composition at time of collection.

Isotope	Per Cent by Weight		
	Sampler 2	Sampler 3	Typical Plutonium ^a
²³⁸ Pu	0.0085 ± 0.0009	0.0073 ± 0.0006	0.04 ± 0.01
²³⁹ Pu	95. ± 8.	95. ± 6.	93.34 ± 0.5
²⁴⁰ Pu	5.2 ± 0.4	4.9 ± 0.3	6. ± 0.5
²⁴¹ Pu ^b	0.18 ± 0.02	0.18 ± 0.02	0.58
²⁴² Pu	0.015 ± 0.002	0.05 ± 0.02	0.04

^aRef. 35

^bAbundance is significantly influenced by radioactive decay during the time since deposition at the GMX site.

Table 10. Results of the calculated activity ratio of ²⁴¹Pu to ²⁴¹Am, and the calculated age of the plutonium present on two of the cascade impactor back-up filters.

Sampler	$\frac{{}^{241}\text{Pu Activity}}{{}^{241}\text{Am Activity}}$	Age, years
2	30 ± 4	14.6 ± 1.4
3	35 ± 5	13.2 ± 1.5

Table 11. Concentration of total suspended particulates and distribution with particle size for the second Cascade impactor run at GMX. The five impactors were run continuously from April 24 to May 23, 1973. Radiochemical results are incomplete for this set of samples.

Sampler	Concentration $\mu\text{g}/\text{m}^3$	MMAD μm	σ_g
1	38	1.21 ± 0.05	7.3 ± 0.6
2	37	1.13 ± 0.01	6.8 ± 0.1
3	47	1.28 ^a	7.0 ^a
4	47	1.06 ± 0.05	7.9 ± 0.8
5	37	1.17 ± 0.03	6.3 ± 0.4
Total ^b	41	1.17 ± 0.05	7.0 ± 0.6
Mean ^c	41	1.17 ± 0.05	7.0 ± 0.6

^a The reduced chi-square of the fit indicates that the data are not well fit by a lognormal distribution.

^b The mass on each stage was summed for the five impactors, and the cumulative per cent distribution calculated.

^c The cumulative per cent distributions were averaged for the five impactors.

Table 12. ^{239,240}Pu concentration derived from two sample sets collected with the ultra-high-volume air samplers. The samplers were run between the hours of 1150 and 1400 PST on May 31, 1973 and 1200 and 1830 PST on June 13, 1973. Some of the meteorological data recorded during these periods is given in Fig. 1 of Part II of this report.

Location	^{239,240} Pu Air Concentration, $\mu\text{Ci}/\text{cm}^3$			
	May 31, 1973		June 13, 1973	
	West	East	West	East
800 ft, Upwind	4.1×10^{-15}	8.0×10^{-15}	1.8×10^{-15}	2.2×10^{-15}
Ground Zero	5.6×10^{-13}	1.1×10^{-12}	8.2×10^{-14}	2.2×10^{-13}
1200 ft, Downwind	4.7×10^{-14}	5.3×10^{-14}	9.7×10^{-15}	2.3×10^{-14}
2400 ft, Downwind	3.1×10^{-15}	3.1×10^{-15}	5.1×10^{-15}	2.6×10^{-15}
4800 ft, Downwind	1.4×10^{-14}	3.1×10^{-15}	7.1×10^{-16}	^a
Control	1.3×10^{-15}		^a	

^a Analysis not yet available

FIGURE CAPTIONS

- Fig. 1. Cumulative distributions of $^{239,240}\text{Pu}$ air concentration measured by REECO from February, 1971, to July, 1972, at three sites. Two sites were located at the northeast and southwest points of the perimeter fence surrounding the GMX site in Area 5. The control location (Bldg. 214) is located in Mercury.
- Fig. 2. Wind frequency distribution with direction at the GMX site from February, 1971, to July, 1972. This time period corresponds to the collection period of the REECO data shown in Fig. 1. The scale is per cent occurrence within a 15° sector.
- Fig. 3. Wind frequency distribution with direction at the GMX site from July 7, 1972, to August 12, 1972. This time period corresponds to the collection period of the first high-volume cascade impactor series. The scale is per cent occurrence within a 10° sector.
- Fig. 4. The average cumulative distributions of mass and $^{239,240}\text{Pu}$ as a function of particle size as determined from the first high-volume cascade impactor run at the GMX site. The lines represent least-squares fits to lognormal distributions. Values of the derived parameters are given in Table 6.
- Fig. 5. The relationship between the total $^{239,240}\text{Pu}$ and ^{241}Am on individual stages of the five high-volume cascade impactor samplers. Data are for the first run at GMX on July 7 to August 12, 1972. The line represents a least-squares fit of the data.
- Fig. 6. The average cumulative distributions with mass as a function of particle size as determined from two high-volume cascade impactor runs at the GMX site. The lines represent least-squares fits to lognormal distributions. Values of the derived parameters are given in Tables 6 and 11.

- Fig. 7. $^{239,240}\text{Pu}$ air concentration at the GMX site derived from a set of ultra-high-volume filter samples. The samplers were run from 1150 to 1400 PST on May 31, 1973. The numbers are averages of duplicate samples.
- Fig. 8. $^{239,240}\text{Pu}$ air concentration at the GMX site derived from a set of ultra-high-volume filter samples. The samplers were run from 1220 to 1830 PST on June 13, 1973. The numbers are averages of duplicate samples.

Fig. 1.
Anspaugh, L. R.
and
Phelps, P. L.

Fig. 2.
Anspaugh, L. R.
and
Phelps, P. L.

Fig. 3.
Anspaugh, L. R.
and
Phelps, P. L.

Fig. 4.
Anspaugh, L. R.
and
Phelps, P. L.

Fig. 5.
Anspaugh, L. R.
and
Phelps, P. L.

Fig. 6.
Anspaugh, L. R.
and
Phelps, P. L.

Run 7, $\mu\text{Ci}/\text{cm}^3$
MAY 31, 1973

-92-

Fig. 7.
Anspaugh, L. R.
and
Phelps, P. L.

Run 9, $\mu\text{Ci}/\text{cm}^3$
JUNE 13, 1973

-93-

Fig. 8.
Anspaugh, L. R.
and
Phelps, P. L.

RESUSPENSION ELEMENT STATUS REPORT

Appendix A. The Use of NTS Data and Experience to Predict
Air Concentrations of Plutonium due to Resuspension
on the Enewetak Atoll*

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There are no general models that may be used with confidence to predict the resuspended air activity in the vicinity of an area contaminated with Pu. However, two approximate methods may be used - the resuspension factor approach and an argument based upon ambient air particulate concentrations with the assumption that the particulates are derived from the contaminated surface. The former method has been frequently used, but almost always in the context of a fresh surface deposit. The latter method is inappropriate to the fresh deposit situation, but should be reasonably valid after enough time has elapsed for the surface deposited material to become fairly well mixed with a few centimeters of the soil surface.

Resuspension Factor Approach

The resuspension factor, K, is defined as

$$K = \frac{\text{Air Concentration (Ci/m}^3\text{)}}{\text{Surface Deposition (Ci/m}^2\text{)}}$$

and thus has units of m^{-1} . It is almost always implied that both measurements are made at the same location. The difficulties with this approach are fairly obvious - no allowance is made for the geometrical configuration of the source, the particle size distributions of the contaminant and the soil

* This also appears in Enewetak Radiological Survey, USAEC, Nevada Operations Las Vegas, Rept. NVO-140 (1974).

surface, vegetation cover, etc. Stewart¹ and Mishima² have tabulated values of K from many experiments including those involving laboratory floors as well as native soils. As would be expected, the tabulated values cover an enormous range and vary from 10^{-2} to $10^{-13}/\text{m}$. Most of the high values, however, are derived from experiments with laboratory floor surfaces and/or with artificial disturbance.

For outdoor situations, Stewart¹ suggests as a guide for planning purposes that a value for K of $10^{-6}/\text{m}$ be used "under quiescent conditions, or after administrative control has been established in the case of an accident." A value of $10^{-5}/\text{m}$ is suggested under conditions of moderate activity. Stewart states, however, that exceptionally higher values (mean of $10^{-5}/\text{m}$) were observed during the Hurricane Trial (Monte Bello Islands) and credited this to the nature of the small islands exposed to sea breezes. Values approaching $10^{-3}/\text{m}$ when dust is raised by pedestrians and vehicles are also reported by Stewart.

Kathren³ has also considered the resuspension factor approach and has recommended the use of $10^{-4}/\text{m}$ as a conservative, but appropriate value for setting standards for PuO_2 surface contamination.

Langham^{4, 5} has suggested that a value of $10^{-6}/\text{m}$ is a reasonable average value to use in estimating the potential hazard of occupancy of a plutonium contaminated area. At the same time, however, Langham notes that many measured values lie in the range of 10^{-5} to $10^{-7}/\text{m}$ and reports that his own measurements in 1956 produced a value of $7 \times 10^{-5}/\text{m}$.

These recommended values, however, are all intended for application during the time period immediately following deposition. Numerous

studies^{1, 5-8} have shown that air concentrations of resuspended materials decrease with time. With the assumption that this decrease can be represented by a single exponential function, half-times of 35 to 70 days have been reported^{5,7,8}. This decrease in air activity is not explainable by the relatively minor loss of material from the initial site of deposition^{1,6}, but is presumably caused by the migration of the initial surface deposited material into the soil.

Attempts to use the resuspension factor approach to derive acceptable levels of soil surface contamination have included this "attenuation factor" as a simple exponential function with half-times of 35⁴ or 45³ days. There are major uncertainties in such a formulation, however. The longest study of this decrease with time extended to only eleven months following the initial deposition⁸, which is extremely short compared to the half-life of a radionuclide such as ²³⁹Pu. There are also published reports which indicate on experimental and theoretical bases that the decrease with time will not be adequately represented by a single exponential function, but that the rate of decrease itself will also decrease with time^{1,6}. Fortunately, the exact nature of this time dependence is not critical in determining the integrated exposure from the time of initial deposition due to the fairly well documented rapid decrease at early times. However, it is obviously the controlling factor for questions concerning the reoccupation of areas many years after the contaminating event.

As an illustration, the most conservative published model (Kathren³) may be used to calculate a resuspension rate for material 15 years

after deposition:

$$K = \frac{10^{-4}}{m} \exp \left[\frac{-0.693 \times 15 \text{ y} \times 365 \text{ d}}{45 \text{ d} \times y} \right] \\ \approx 10^{-41}/m$$

If, however, the resuspension rate asymptotically approached some finite value 10⁻⁶ of the original, then the resuspension rate 15 years later would obviously be 10⁻¹⁰/m; but the total integrated air activity (from t = 0 to ∞) for ²³⁹Pu would be changed only by

$$A 10^{-4} \int_0^{\infty} \exp (-0.693 t/45 \text{ d}) dt + A 10^{-10} \int_0^{\infty} \exp (-0.693 t/24,400 \text{ y}) dt \\ = 6.5 A \times 10^{-3} + 1.3 A \times 10^{-3}$$

which is an increase of 20%, and more importantly, cannot be accumulated during an individual's life span.

Because the functional nature of the decrease in resuspension rate with time cannot be confidently extrapolated, previously published models should not be applied to the reoccupation of areas many years after the contaminating event.

The resuspension factor approach can be applied in an approximate way, however, if resuspension factors are used which were derived from measurements over aged sources. Perhaps the most relevant data are unpublished results from current resuspension experiments at the GMX site in Area 5 of the Nevada Test Site. The ²³⁹Pu at this location was deposited following 22 high-explosive detonations during the time period from December, 1954, to February 1956. Measurements of resuspended air activity levels at this site

during 1971 - 1973 appear to be the only available data concerning resuspension of ^{239}Pu from a source of this age.

Data from two types of measurements are available and can be used to derive average resuspension factors. The first type of measurement⁹ was accomplished by placing five high-volume cascade impactors¹⁰ within the most highly contaminated area, and running them for 36 days, from July 7 to August 12, 1972. The collected $^{239}, ^{240}\text{Pu}$ activity was distributed lognormally with particle size with an activity median aerodynamic diameter (AMAD) of 3.0 μm and a geometric standard deviation of 8.2. The $^{239}, ^{240}\text{Pu}$ concentration varied from 1.0 to $3.9 \times 10^{-14} \mu\text{Ci}/\text{cm}^3$ with an average of $2.3 \times 10^{-14} \mu\text{Ci}/\text{cm}^3$ for the five samplers. At the present time only limited data are available regarding the soil activity in the area. Four soil samples of depth 0 - 3 cm from approximately the same location have been analyzed with results¹¹ of 2060 to 3550 dpm/g with a mean of 2700 dpm/g. Profile data from other locations at the same general site indicate that about 90% of the total deposition is contained within the top 2.5 cm of the soil¹². Measurements of soil density in the area average $1.8 \text{ g}/\text{cm}^3$ ⁹. The resuspension factor is therefore

$$\frac{2.3 \times 10^{-14} \mu\text{Ci}}{\text{cm}^3} \times \frac{\text{g}}{2700 \text{ dpm}} \times \frac{\text{cm}^3}{1.8 \text{ g}} \times \frac{0.9}{3 \text{ cm}} \times \frac{10^2 \text{ cm}}{\text{m}} \times \frac{2.22 \times 10^6 \text{ dpm}}{\mu\text{Ci}}$$

$$= 3 \times 10^{-10}/\text{m}$$

Additional air samples were taken by the Reynolds Electrical and Engineering Co. (REECo) on the edge of the contaminated area during the period of February, 1971, to July, 1972, with a sampling time of approximately 48 hours¹³. Measurements were made at four locations, but the most pertinent

is the one which was most frequently in the direction of strong winds from the strongly contaminated area and where the highest air activities were recorded. Here, 254 individual air filter samples were collected and detectable results reported for 236. $^{239}, ^{240}\text{Pu}$ concentrations ranged from 3.5×10^{-17} to $6.3 \times 10^{-13} \mu\text{Ci}/\text{cm}^3$, with arithmetic and geometric means of 6.6×10^{-15} and $7.9 \times 10^{-16} \mu\text{Ci}/\text{cm}^3$ respectively. Results for four soil samples taken from approximately the same location range from 128 to 202 dpm/g with a mean of 160 dpm/g¹¹. Because the arithmetic mean is a better representation of average lung exposure, it is used to derive a resuspension factor at this site:

$$\frac{6.6 \times 10^{-15} \mu\text{Ci}}{\text{cm}^3} \times \frac{\text{g}}{160 \text{ dpm}} \times \frac{\text{cm}^3}{1.8 \text{ g}} \times \frac{0.9}{3 \text{ cm}} \times \frac{10^2 \text{ cm}}{\text{m}} \times \frac{2.22 \times 10^6 \text{ dpm}}{\mu\text{Ci}}$$

$$= 2 \times 10^{-9}/\text{m}$$

This value is nearly an order of magnitude higher than the one previously calculated, and reflects some of the inherent difficulties in the resuspension factor approach, i.e., that no allowance is made for the geometrical configuration of the source and that higher ground activities may be present upwind.

It is obvious that this approach is subject to major uncertainties, but does serve as an order of magnitude indication of the resuspended air activities that may arise from a $^{239}, ^{240}\text{Pu}$ contaminated area which has weathered for 15 to 20 years. The data discussed above also demonstrate unequivocally that resuspension of $^{239}, ^{240}\text{Pu}$ does in fact occur from such aged deposits and at levels many orders of magnitude higher than would be

expected if the often noted decrease with time were represented by a single exponential function with a half-time of 35 to 70 days.

Mass Loading Approach

The other approximate prediction method is based upon measured or assumed levels of particulate matter in ambient air with the assumption that this material is derived from the contaminated soil. For fresh deposits this approach is not valid because the freshly deposited debris is much more likely to be resuspended than the remainder of the weathered soil surface. After many years of weathering since the initial deposition, however, the contaminating material should be reasonably well mixed with a centimeter or two of soil such that the contaminant activity per gram of airborne particulate should approximate that in the upper soil. A major difficulty could arise, however, if for example $^{239}, ^{240}\text{Pu}$ were preferentially associated with the smaller particle sizes more likely to become airborne. For the Nevada Test Site, such is not the case as determined by soil analyses¹⁴

and by the high-volume cascade impactor study. The latter study found an AMAD of $3.0\text{ }\mu\text{m}$ for $^{239}, ^{240}\text{Pu}$ whereas the total mass median aerodynamic diameter was $1.7\text{ }\mu\text{m}$. The specific activity of the material collected on each stage can also be examined for a preferential association of Pu with particle size. Average data from all five samplers are:

<u>Size (μm)</u>	<u>$^{239}, ^{240}\text{Pu}$ (dpm/g)</u>
> 7	950
3.3 to 7	700
2.0 to 3.3	1030
1.1 to 2.0	1300
.01 to 1.1	480
All Stages	890
(Soil)	(2700)

Although there is considerable spread in these data, there is no indication of a preferential association of $^{239}, ^{240}\text{Pu}$ with a particular particle size; and as would be expected due to dilution by inert aerosol, the specific activity is lower than that of the soil.

If we assume that this is generally true, a general and conservative method of predicting resuspended air concentrations of contaminants would be to simply multiply the ambient air mass loading by the contaminant concentration in soil. A factor of some uncertainty for a specific calculation is what value to use for the ambient air mass loading in the absence of specific data. This becomes even more uncertain by the possibility that the people involved may be highly correlated with the source in the

sense that children playing in sand, adults cultivating crops, etc., may generate their own "ambient air" which contains much more mass than would be recorded by a stationary sampler.

The lower and upper bounds of ambient air mass loading can be fixed rather easily for any site. There has been considerable interest in establishing a "background level" of mass loading and this is generally believed to be on the order of $10 \mu\text{g}/\text{m}^3$ ¹⁵. The upper bound can be established in a reasonable way by the levels found in mine atmospheres which have led to a considerable prevalence of pneumoconiosis in the affected workers¹⁶. Examination of these data indicate that current standards for occupational dust exposure ($\sim 1 - 10 \text{ mg}/\text{m}^3$) have a very small, or perhaps no margin of safety such that a reasonable upper bound can be taken as $1 \text{ mg}/\text{m}^3$. (British data¹⁷ indicate that if the general public were exposed to dust levels in excess of $1 \text{ mg}/\text{m}^3$, the public health problem from the dust alone might be enormous. The presumption is therefore made that if ^{239}Pu , ^{240}Pu were also in the dust at levels sufficient to achieve the maximum permissible concentration, the additional insult would be insignificant.) The reasonableness of the upper limit value of $1 \text{ mg}/\text{m}^3$ is also demonstrated by data which indicate that nonurban ambient air mass concentrations this high are usually associated with conditions described as dust storms^{18, 19}.

Measurements of ambient air mass loading can be used to further define a reasonable estimate for predictive purposes. The National Air Surveillance Network (NASN) has reported such results for several years. Data for 1966²⁰ show that there were 217 urban and 30 nonurban stations reporting. The

annual arithmetic average for the urban stations ranged from 33 (St. Petersburg, Florida) to $254 \mu\text{g}/\text{m}^3$ (Steubenville, Ohio), with a mean arithmetic average for all 217 stations of $102 \mu\text{g}/\text{m}^3$. For the nonurban stations, the range was from 9 (White Pine County, Nevada) to $79 \mu\text{g}/\text{m}^3$ (Curry County, Oregon) with a mean arithmetic average for all 30 stations of $38 \mu\text{g}/\text{m}^3$. No data in this report are available for nonurban locations on small islands similar to the Enewetak group; perhaps the closest analogue is the urban station at Honolulu, Hawaii, which had an annual arithmetic average of $35 \mu\text{g}/\text{m}^3$.

More pertinent, but limited, data have recently been published for the island of Hawaii^{21,22}. Data are given for three locations: Mauna Loa Observatory located at a height of 3400 m, Cape Kumukahi, and the City of Hilo. NASN data for Hilo (for an unspecified period) are given as $18 \mu\text{g}/\text{m}^3$, and nephelometer measurements varied from $18 \mu\text{g}/\text{m}^3$ during the day to $26 \mu\text{g}/\text{m}^3$ at night. At Cape Kumukahi the nephelometer measurement was $9.2 \mu\text{g}/\text{m}^3$. The greatest amount of data is available for Mauna Loa Observatory. Here, the NASN measurement was $3 \mu\text{g}/\text{m}^3$, and the nephelometer measurements varied from $1.7 \mu\text{g}/\text{m}^3$ at night to $6.5 \mu\text{g}/\text{m}^3$ during the day. Additional measurements made by the USAEC Health and Safety Laboratory (HASL) were $3 \mu\text{g}/\text{m}^3$. It is of interest in the present context that Simpson²² made the following comment concerning the HASL measurements: "The HASL filter samples contain substantial dust ($3 - 5 \mu\text{g}/\text{m}^3$ of air sampled) because of the fact that the filter was located less than one meter above the ground surface near areas with substantial personnel activity at the observatory site." Thus, while this method of measurement may not have

coincided with Simpson's interest, it does indicate that ambient air mass loadings may be very low on such remote islands even when considerable human activity is occurring nearby.

On the basis of the above data, it would appear reasonable to use a value of $100 \mu\text{g}/\text{m}^3$ as an average ambient air mass loading for predictive purposes. Indications are that this value should be quite conservative for the Enewetak Islands, and therefore allows room for the uncertainty involved because the people themselves may generate a significant fraction of the total aerosol. They may therefore be exposed to higher particulate concentrations than would be measured by a stationary sampler.

Supporting evidence that $100 \mu\text{g}/\text{m}^3$ is a reasonable value to use for predictive purposes is provided by the National Ambient Air Quality Standards²³. Here ambient air is defined as "...that portion of the atmosphere, external to buildings, to which the general public has access." The primary ambient air standards define "levels which... are necessary, with an adequate margin of safety, to protect the public health." The secondary standards define "levels which...(are) necessary to protect the public welfare from any known or anticipated adverse effects of a pollutant." These standards for particulate matter are given below.

National Ambient Air Quality Standards

for Particulate Matter

(in $\mu\text{g}/\text{m}^3$)

	<u>Annual Geometric Mean</u>	<u>Max. 24-hr Concentration not to be exceeded more than once/yr</u>
Primary	75	260
Secondary	60	150

Data to support these standards in terms of health effects, visibility restrictions, etc. have been provided²⁴.

An arithmetic mean would be more desirable for predictive purposes. Data from 1966²⁰ for nonurban locations indicate that the annual arithmetic mean is (on the average) 120% of the annual geometric mean.

Representative Calculations

Because one of the primary objects is to derive an acceptable soil level for the Enewetak Islands, the approaches developed above were used to derive such levels for both soluble and insoluble ²³⁹Pu. The derived values are given in Table 1. The two methods agree within a factor of two, at least for soil distributions like those found at the Nevada Test Site. (The ambient air mass loading at NTS during the cascade impactor run was measured to be $70 \mu\text{g}/\text{m}^3$.)

Such derived values must, of course, be used with a great deal of discretion. They are based on simple model systems which are believed to be generally conservative, but individual situations can be imagined which could exceed the predictions.

Other Considerations

The above calculations relate only to the resuspended air activity in ambient air, and do not consider the additional problems of resuspension of material from contaminated clothing or the resuspension of material which has been transferred to homes.

Healy²⁶ has considered these and other problems, and has provided tables of "decision levels" for surface contamination levels and home

transfer levels. A decision level is based upon National Council on Radiation Protection and Measurements (NCRP) recommended dose limitations. Because the derivations are rather tenuous, Healy has used the phrase decision level and states that its use is to serve as a signal that further careful investigation is warranted.

Healy's decision levels for soluble ^{239}Pu are given in column 1 of Table 2. The values in column 2 are derived from these and an acceptable soil concentration of 1 nCi/g from Table 1 to give equivalent dirt (soil) contamination and transfer levels. The results are interpreted as indicating that the potential exists for greater dose contributions from these infrequently considered pathways than from the usually considered pathway of resuspension as calculated for ambient air. (This conclusion would be the same for insoluble ^{239}Pu .) Therefore, if dose calculations based on the usual resuspension pathway should appear limiting compared to other pathways such as food-chain transfer, these pathways considered by Healy need to be carefully evaluated for the specific Enewetak situation.

Table 1

Acceptable soil levels of ^{239}Pu for a source which has weathered for several years. Values are approximate and are subject to uncertainty. The reference standard used in the derivations is 10% of the Maximum Permissible Concentration in Air for 168 hr occupational exposure (MPC_a)²⁵.

	<u>Insoluble</u>	<u>Soluble</u>
Acceptable Air Concentration	$10^{-12} \mu\text{Ci}/\text{cm}^3$	$6 \times 10^{-14} \mu\text{Ci}/\text{cm}^3$
<u>Resuspension Factor Approach</u>		
Assumed Resuspension Factor	$10^{-9}/\text{m}$	$10^{-9}/\text{m}$
Acceptable Soil Deposition ^a	$10^3 \mu\text{Ci}/\text{m}^2$	$60 \mu\text{Ci}/\text{m}^2$
Acceptable Soil Concentration ^b	20 nCi/g	1 nCi/g
<u>Mass Loading Approach</u>		
Assumed Mass Loading	$10^2 \mu\text{g}/\text{m}^3$	$10^2 \mu\text{g}/\text{m}^3$
Acceptable Soil Concentration	10 nCi/g	0.6 nCi/g

^aEquivalent to approximately $10^4 \mu\text{g}$ of insoluble $^{239}\text{Pu}/\text{m}^2$.

^bAssumes same distribution of ^{239}Pu with depth and soil density as measured at the Nevada Test Site.

Table 2

Decision levels²⁶ for soluble ²³⁹Pu, and their equivalent in soil mass based upon the "acceptable soil concentration" from Table 1.

<u>Pathway</u>	<u>Decision Level</u>	<u>Mass Equivalent</u>
A. Direct Personal Contamination		
Direct Inhalation ^a	2×10^{-5} nCi/cm ²	1×10^{-5} g/cm ²
Direct Ingestion ^b	0.2 nCi/cm ²	0.2 g/cm ²
Skin Absorption ^c	8×10^{-4} μ Ci	0.8 g
B. Transfer (to Homes) Levels		
Resuspension ^d	0.01 μ Ci/day	10 g/day
Direct Inhalation	0.01 μ Ci/day	10 g/day
Direct Ingestion	100 μ Ci/day	10^5 g/day
Skin Absorption	0.03 μ Ci/day	30 g/day

^a"The contamination level on clothing and skin that could result in inhalation of air at the MPC_a for the public."²⁶

^b"The contamination level on skin or clothing that could result in ingestion of a quantity of radioactive material equivalent to the ingestion of water at the MPC_w for an individual in the public."²⁶

^c"The total quantity of radioactive material maintained on the skin for 24 h/day that could result in absorption of a quantity equal to that which would be absorbed from the GI tract if water at the MPC_w for "soluble" isotopes for an individual in the public were ingested."²⁶

^d"The amount transferred per day that could result in air concentrations due to resuspension in a medium-sized home averaging at the MPC_a for an individual in the public."²⁶

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