

DETERMINATION OF RADIOACTIVE FALLOUT

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J. J. FITZGERALD

KNOLLS ATOMIC POWER LABORATORY
SCHENECTADY, N.Y.

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ABSTRACT

The original, the quality, and the quantity of radioactive materials that may be discharged to the atmosphere are discussed. A distinction is made between deposition of activity on the ground by surface adsorption, fall-out, wash-out, and rain-out. The rate of deposition from wash-out is a factor of 4 to 20 times the rate of deposition during fall-out under similar circumstances. Rain-out deposition at a distance of one mile from the point where the activity is dispersed is a factor of 1000 greater than wash-out deposition. A formula for determining the accumulated external dose from the maximum deposition is given and is in good agreement with experimental data.

INTRODUCTION

The rate of radioactive deposition on the earth from the atmosphere and the effects of these depositions on humans have been discussed in many technical papers. (1,2,3,4,5,6) This phenomenon of deposition is commonly known as "fall-out." In a strict sense, fall-out refers only to deposition of radioactivity on the earth during non-precipitant weather. In the broad sense, fall-out includes deposition during precipitant weather. In this discussion, fall-out shall be referred to in the strict sense of the word to distinguish it from rain-out, wash-out, and snow-out.

Although it is difficult to predict the precise amount of radioactivity that will be deposited on the earth, it is possible to determine the maximum rate of deposition. In this paper, a method for determining the maximum deposition of activity on the earth from the atmosphere will be discussed. In addition, the rate at which radioactive particles are cleansed from the atmosphere under a heavy rainfall will also be given as a function of particle size. The total integrated dose from long-lived and short-lived radioactive materials deposited on the ground will be estimated.

Before discussing the rate of deposition of radioactivity on the ground and the subsequent doses received from this radioactivity, it is appropriate that we discuss the origin, the quality, and the quantity of radioactivity that may be present in the atmosphere.

ORIGIN, QUANTITY, AND QUALITY OF RADIOACTIVITY IN THE ATMOSPHERE

Radioactivity in the atmosphere may arise from "natural" causes, A-bomb or thermonuclear explosions and induced radioactivities and fission products emanating from medical, industrial, and/or atomic energy operations.

Most of the natural radioactivity in the atmosphere is a result of the gaseous diffusion of radon (Rn-222) and thoron (Rn-220) from the earth's surface. These gaseous radioactive isotopes are the decay products of natural uranium and thorium found in the earth. Since radon has a longer lifetime than thoron, it escapes in larger quantities into the atmosphere before it decays to a non-gaseous product. Thoron concentrations in air are only a few percent of the radon concentrations. These concentrations vary considerably and are significantly affected by the concentrations of uranium and thorium in the earth and the meteorological

conditions prevailing in the atmosphere. Radon concentrations in air are of the order of 10^{-10} μC (Rn-222)/cc (of air) which are approximately 1/100 of the maximum permissible lifetime breathing concentrations.(7)

Radioactivity may be dispersed in the atmosphere as a result of an A-bomb explosion. The process wherein these radioactive isotopes are formed is called the fission process. Absorption of a neutron by a heavy metal such as uranium may result in the formation of two new radioisotopes. In this process, 30 to 40 new elements may be formed. Each of these elements or radioisotopes may decay to another element which is also radioactive. In this manner, 90 to 100 new isotopes are created by the fission process. Each of these radioisotopes may release beta particles which may also be accompanied by gamma radiation. The frequency at which the new isotopes are created during the fission process varies with the mass of the newly formed isotope. Fission yields vary from a fraction of a percent up to six percent. For example, Kr-85, Sr-90, and I-131 have fission yields of 0.32, 5.50, and 2.55%, respectively. The rate at which a radioisotope decays or diminishes in radioactivity is characteristic of the radioisotope. The decay rate of the radioisotopes formed in the fission process varies from half lives of seconds to half lives of years. The decay rate is somewhat complex since an isotope can be formed directly in the fission process or it can result from the decay of another isotope formed directly or indirectly in the fission process. Therefore, radioactive isotopes are being created and destroyed at the same time even after the fission process has been completed.

The total beta and gamma radioactivities from the dissipation of fission products energies are given by K. Way⁽⁸⁾ as $1.40t^{-1.2}$ and $1.26t^{-1.2}$ in Mev/sec/fission, respectively. The value of t , (the decay time or time after the fission process ended) is expressed in seconds. Therefore, the fissioning of one gram of uranium will result in the formation of $1.3 \times 10^{11} t^{-1.2}$ curies of gamma activity which has an average energy of 0.7 Mev/disintegration. In the detonation of a "nominal" type A-bomb (20 kiloton equivalents of TNT), approximately 1000 grams of uranium⁽³⁾ will be fissioned. Therefore, although a 20 kiloton bomb would only involve the fissioning of approximately two pounds of fissionable material, it would result in the dispersion of $1.3 \times 10^{14} t^{-1.2}$ curies of gamma activity into the atmosphere. This activity, however, decays rapidly. It decays by a factor of ten with each increase in time by a factor of seven. Of the long-lived fission products, only 7.95×10^3 curies of Sr-90 would be formed during the

fissioning of a 20 kiloton equivalent TNT bomb. This activity, however, would only decay by a factor of two in a period of 25 years.

A thermonuclear bomb⁽⁹⁾ would, of course, release much more gamma and Sr-90 radioactivity.

The final sources of radioactivity which could be dispersed in the atmosphere are emanations from the reactor operations and separation process stack effluents. The history of the Atomic Energy Program shows clearly that the control and containment of radioactive materials is not only possible but has, in fact, been very successfully achieved. A discussion of the quantities of radioactivity that could be discharged from atomic energy operations is presented here to complete the possible sources of radioactivity in the atmosphere. The first two sources of radioactivity in the atmosphere resulted from "natural" causes and well-designed experimental tests. The probability of a large fraction of the third source entering into the atmosphere is very small. The radioactivity developed in a nuclear reactor is approximately 9.1×10^6 curies/megawatt of power⁽¹⁰⁾ when the reactor has been operating for a period of at least one day. For a power reactor operated at 1000 megawatts, the total stored radioactivity in the reactor could be approximately 10^{10} curies. This activity will decay at a much slower rate than the A-bomb or thermonuclear bomb radioactivities. For each 100-fold increase in time, the activity would decay only by a factor of 0.4. As illustrated in Figure (1), although the initial activity (at one second) from a 20 kiloton bomb is approximately 14,000 times greater than the activity from a 1000 megawatt reactor, the rapid decay from the bomb test radioactivity is such that the activities are approximately equal in a period of four hours. It takes the thermonuclear type bomb many days to reduce its activity to the level of the 1000 megawatt reactor in a corresponding decay period. Thermonuclear explosions are of such a magnitude that the radioactive cloud is carried up into the stratosphere.⁽¹¹⁾ The deposition of activity on the ground from such explosions is relatively slower than from A-bomb explosion where the cloud is carried to heights of 40,000 feet. The rate of deposition of activity from an A-bomb explosion may be at the rate of 50% in 22 days whereas the rate of deposition from the stratosphere and the thermonuclear cloud is of the order of ten to 20% per year.⁽¹¹⁾ Therefore, although the intensity of the thermonuclear bomb may be greater than that of the A-bomb, additional decay before the activity reaches the earth's surface is obtained by virtue of the height to which the cloud is initially carried. A reactor which has operated at a power level of 1000 megawatts for a period of one year would have stored in the reactor core 1.6×10^6 curies of Sr-90. That quantity of Sr-90 is 200 times the amount in a 20 kiloton bomb.

The concentration of relatively large quantities of long lived fission products in the reactor core of a power reactor is the basic reason that the Atomic Energy Commission has insisted on and has achieved a relatively high degree of containment of these fission products.⁽¹²⁾

PROCESSES OF DEPOSITION

In evaluating the quantity of radioactivity falling out of the cloud one may assume that the radioactivity is deposited on the earth by electrical attraction, gravitational force and impaction on raindrops or snowflakes. One must also consider the initial size of the particles in the radioactive

cloud and their tendency to grow. In addition, the diffusion of the cloud in the atmosphere under a series of meteorological conditions must be evaluated. Ultimately when these assumptions are made, a specific quantity of activity is computed to be deposited on the ground as a function of time or unit distance from the origin at which the activity is dispersed.

Deposition from the atmosphere may result from (1) ground level adsorption of radioactivity, (2) dry weather deposition, (3) wash-out of radioactivity and, (4) rain-out or snow-out of radioactivity.

GROUND LEVEL ADSORPTION

If the radioactive cloud is emitted at ground level and the meteorological conditions are such that the cloud does not rise, ground level adsorption and deposition will be important. This classification of fall-out would refer chiefly to a set of conditions that could be achieved only under a reactor incident. In this instance, the reaction and the associated quantity of heat may not be sufficient to elevate the cloud. Low level inversion meteorological conditions would tend to keep the radioactivity at ground level and enhance the adsorption of radioactivity on grass surfaces, trees, buildings, etc. Ground level adsorption could result from gravitational settling, impaction on vertical and horizontal surfaces, electrostatic attraction to these surfaces, direct interception, and diffusion. An excellent summary of these factors is given in an AEC handbook, "Meteorology and Atomic Energy."⁽¹³⁾

DRY WEATHER DEPOSITION

Deposition of radioactivity in non-precipitant weather is significantly less than the quantity deposited during precipitant weather. Chamberlain's equations⁽¹³⁾ indicate that the maximum deposition in non-precipitant weather is $n/2$ times the maximum deposition in precipitant weather. In his equation, n is the stability parameter which has a value of one-quarter for average meteorological conditions and one-half for a moderate inversion. Stewart, Crooks, and Fisher⁽¹¹⁾ have reported deposition rates four times greater during periods of rainfall following atomic bomb explosions than during periods of non-precipitant weather. Following thermonuclear explosions, rain deposition was a factor 20 times greater than deposition during non-precipitant weather. The increase in deposition during rainfall in the case of thermonuclear explosions may be attributable to the slower rate of dry deposition as previously stated and to its relatively higher percentage of soluble material. Stewart, et al, report thermonuclear material contains 55% of soluble material as opposed to 20 or 30% of soluble material in atomic bomb fission products material.

WASH-OUT DEPOSITION

Wash-out deposition refers to the quantity of radioactivity deposited on the earth by the scrubbing action of raindrops or snowflakes passing through the radioactive cloud. The rate at which particles are washed out of the atmosphere has been discussed in detail by Ranz and Johnstone.⁽¹⁴⁾ The percentage wash-out as a function of the particle size upon which the radioactive material is embedded initially is given in Figure (2) utilizing the formula of Ranz and Johnstone. In these calculations it was assumed that the rainfall was heavy (8×10^{-4} drops/cm³) and the average size of the raindrops was 737 microns.

Since the geometric mean size and the mass mean size of particles in the atmosphere are approximately 0.05⁽¹⁵⁾ and 0.3 micron, respectively, the deposition rate in a heavy rainfall should not exceed 0.5%/hour.

RAIN-OUT DEPOSITION

Rain-out deposition is defined as the deposition of radioactivity from the atmosphere to the earth's surface when the radioactive particles are the nuclei for the formation of rain droplets. This method of deposition will produce the maximum rate at which particles are deposited on the ground. The rate of maximum deposition has been discussed in detail by Fitzgerald, Hurwitz, and Tonks.⁽¹⁶⁾ As previously noted, Chamberlain⁽¹³⁾ also has calculated the maximum rate of deposition. The final equations of the two methods are identical.

The maximum rate of deposition at a distance x_0 meters from the origin of the nuclear incident is given in the following equation:

$$C_A = \frac{Q_{t_0}}{ex_0(\pi^{1/2}r_0)} \quad (1)$$

Where C_A is the maximum rate of deposition in curies/meter².

Q_{t_0} is the quantity of activity in the cloud in curies at time, t_0

t_0 is the time in seconds when the cloud arrives at the distance x_0 meters where deposition occurs

r_0 is the cloud radius in meters and is given by the following expression:

$$r_0 = C_d x_0 \frac{2^{-n}}{2}$$

C_d is the average coefficient of diffusion in meters^{n/2}

n is the stability parameter in non-dimensional units

Assuming average meteorological conditions in which $C_d = 0.12$ meters^{3/4} and $n = 1/4$, the maximum fraction of activity deposited per meter² is given in Figure (3) as a function of distance from origin of the incident.

INTEGRATED EXTERNAL DOSE FROM RAIN-OUT

The dose rate from the maximum deposition of radioactivity on the ground (assuming a uniform deposition of activity on the ground) has been determined by Fitzgerald, Hurwitz, and Tonks.⁽¹⁶⁾

$$D_R = \frac{2\pi\alpha}{3.6 \times 10^3} \left[-E_i\left(\frac{h}{\lambda}\right) \right] C_A t_0^{-n} \quad (2)$$

Where D_R is the dose rate in r/sec.

α^* is the conversion factor for the unit dose rate per unit deposition in $[(r/hr)/(curie/meter^2)]$ and is equal 0.4 for an average gamma energy of 0.7 Mev/disintegration

$$-E_i\left(\frac{h}{\lambda}\right) = \int_{\frac{h}{\lambda}}^{\infty} \frac{e^{-t}}{t} dt$$

h is the distance above ground at which the dose rate is computed in meters

λ is the mean free path of the gamma photon in meters⁻¹

C_A is the maximum rate of rain-out in curies/meter²

t_0 is the decay time in seconds

n has a value of 1.2 for short lived fission products and 0.2 for long lived fission products

*For an average gamma energy of 0.7 Mev/disintegration from a uniformly distributed ground concentration of radioactivity of one curie/meter, the dose rate at one meter above the ground will be 12.56 r/hr.

The integrated dose at one meter above the ground from the maximum deposition of radioactivity on the ground for the case of short lived fission products can, therefore, be computed by the following formula:

$$D_{S.L.} = 1.76 \times 10^{-3} C_A t_0^{-0.2}$$

Where $D_{S.L.}$ is the cumulative dose in roentgens when values of C_A and t_0 are applied in the terms previously stated.

This formula can be used for estimating the accumulative dose from the maximum deposition of radioactivity from an atomic bomb explosion. For example, the integrated dose from the maximum deposition of radioactivity on the ground at a distance of 4000 miles from a bomb site at which a 20 kiloton bomb is exploded is 87 mr if the radioactivity is deposited in that area after a period of two days. Clark⁽¹⁷⁾ reported that the accumulative dose from fall-out in the Troy, New York area in April 1953 (under similar circumstances to those stated above) was approximately 100 mr. Since the actual size of the bomb is not known, more precise calculations cannot be made. The agreement of the theoretical and experimental calculations, however, indicate that the formula can be useful in estimating the upper limit to the hazard of rain-out. In the case of thermonuclear activity, however, the maximum deposition may not be obtained since a large portion of the activity could still be above the rain clouds.

In the case of deposition of radioactivity following the dispersion of long lived fission products, the activity would not be expected to be dispersed in a uniform manner on the ground. The initial fission products cloud of the long lived material would be relatively small and deposition would be over a relatively small area in comparison to the area covered by the A-bomb and the thermonuclear clouds. The distribution would be a Gaussian distribution and the dose rate is given in the following equation:

$$D_r = \frac{\pi}{2} \alpha K_0 (Z) e^Z C_A t_0^{-0.2}$$

Where D_r is the dose rate in r/hr.

α is the unit dose rate per unit deposition, in $(r/hr)/(curie/meter^2)$ and is equal to 0.4 for an average gamma energy of 0.7 Mev/photon

$K_0(Z)$ is a Bessel Function of the second kind where

$$Z = \frac{h^2}{2r_0^2}$$

h is the distance above ground at which the dose rate is computed, in meters

r_0 is the cloud radius in meters as previously given

C_A is the maximum rate of deposition in curies/meter²

t_0 is the decay time, in seconds

The maximum integrated or accumulative dose from the long lived fission products at a distance one meter above the ground for an exposure period of t seconds after the deposition is given by the following expression:

$$D = 2.18 \times 10^{-4} K_0\left(\frac{1}{2r_0^2}\right) e^{\left(\frac{1}{2r_0^2}\right)} C_A \left[(t + t_0)^{0.8} - t_0^{0.8} \right] \quad (5)$$

Where D is in roentgens when the values of r_0 , C_A , t , and t_0 are given in meters, curies/meter², seconds and seconds, respectively

It is more realistic in the above case, however, to replace the quantity of rain-out by the fraction of activity being washed out. In general, the release of long lived fission products would not penetrate the upper atmosphere wherein the particles would be the nuclei for raindrops. In general, long lived fission products would be wash-out off the lower atmosphere. In addition, the above formula is valid for small values of r_0 , or in the case in which the activity is distributed as a Gaussian function on the ground.

At a distance of one mile from the origin of a nuclear incident, the maximum fraction of activity deposited at that point would be 1.7×10^{-6} meters⁻². If 10^8 curies of long lived activity were in the rain cloud, the maximum ground concentration would be 1.7×10^2 curies/meter². The fraction of activity washed out is considerably less. Assuming an average coefficient of diffusion of 0.12 meters^{1/8} and the value of r_0 at a distance one mile from the incident is 75 meters, the concentration at a distance of 114 meters from the center of the cloud will be 10% of the center concentration. Assuming a heavy rainfall with the radioactivity deposited on atmospheric dust (mass mean size of 0.3 micron), the rate of wash-out is 0.5% /hour. The radioactive deposition at a distance of x_0 meters from the origin is given by the following relationships:

$$W_A = 2.78 \times 10^{-4} \frac{F_w Q}{v \sqrt{\pi r_0^2}}$$

Where W_A is the deposition from wash-out in curies/meter²

F_w is the fractional wash-out in hr⁻¹

v is the cloud velocity in meter/sec

Q is the quantity of activity in curies in the cloud when the cloud is over the position x_0

r_0 is the cloud radius in meters

Assuming the values given above, the rate of deposition of 0.5% /hour at a distance of one mile from the origin would produce a fractional deposition of approximately 2.1×10^{-9} meter⁻². This deposition is approximately $1/1000$ of the maximum deposition obtained under the condition of rain-out. The ground concentration at a distance of one mile from a reactor incident containing 10^8 curies in the cloud could result in a ground concentration of 0.21 curies/meter².

CONCLUSIONS

Radioactive deposition depends on (1) the source and decay rate of activity, (2) the initial thrust of activity into the troposphere or the stratosphere, (3) the solubility and the particle size of the material containing the radioactivity and, (4) the prevailing meteorological conditions.

Equation (3) is in good agreement with the maximum experimental accumulative external dose received under similar circumstances following deposition on the ground by rain-out. Therefore, maximum deposition appears to be achieved as a result of rain-out. Wash-out deposition is a factor of 4 to 20 greater than dry deposition (fall-out). Wash-out deposition at a distance of one mile from the point where the activity is released is $1/1000$ of the rain-out deposition.

The actual doses received from fall-out will, however, be at least one order of magnitude less than those predicted by measurements or calculations since no account for shielding is taken into consideration. Absorption of the radioactivity into the soil and deposition into water systems will also reduce the actual total dose received. The effective dose from short lived fission products will be significantly reduced by taking immediate shelter in the basement of a wooden frame or brick house. Such action may reduce the total dose⁽¹⁸⁾ by factors of 10 to 50 . Absorption of radioactivity in soil will be more important in reducing the dose from long lived fission products.

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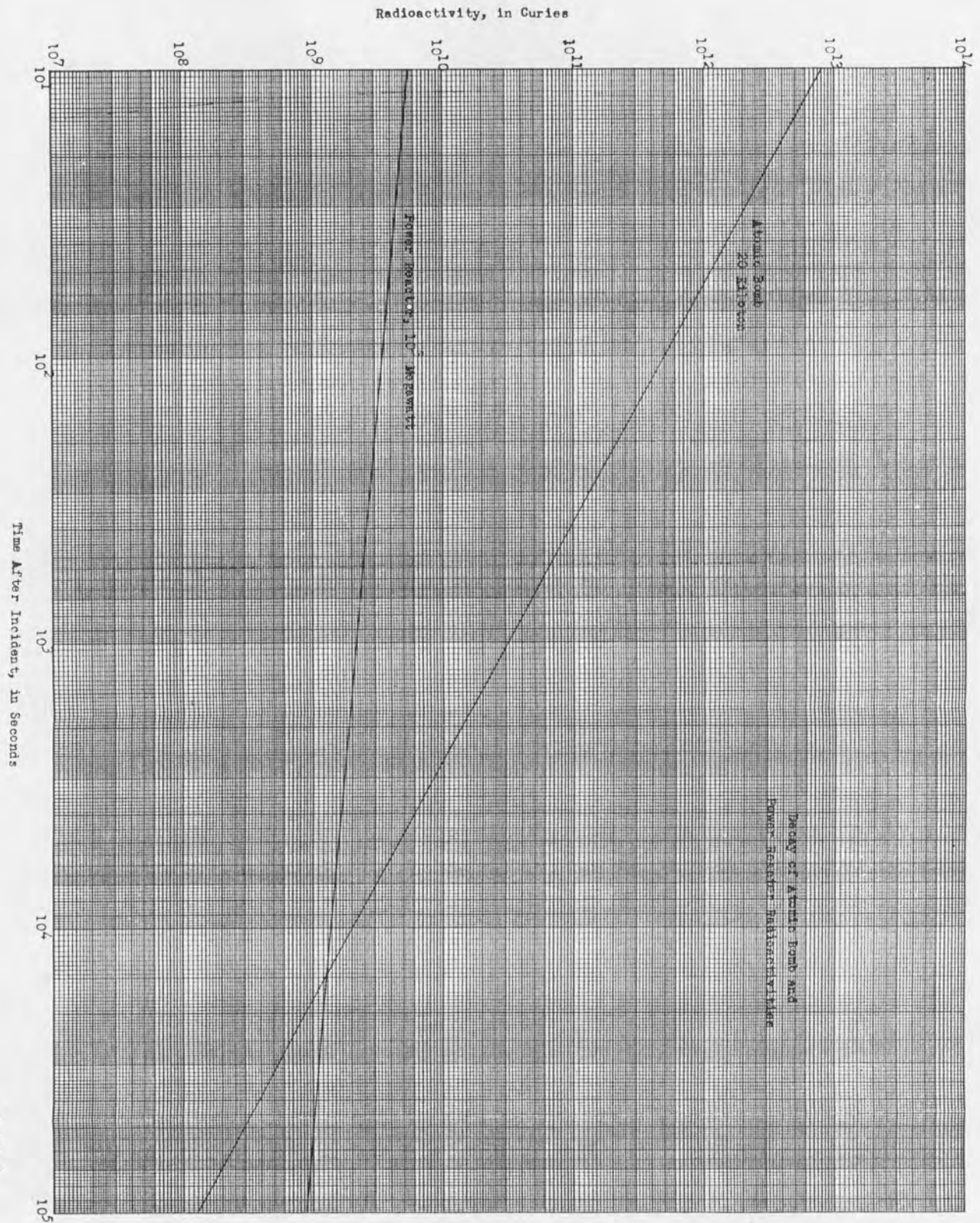
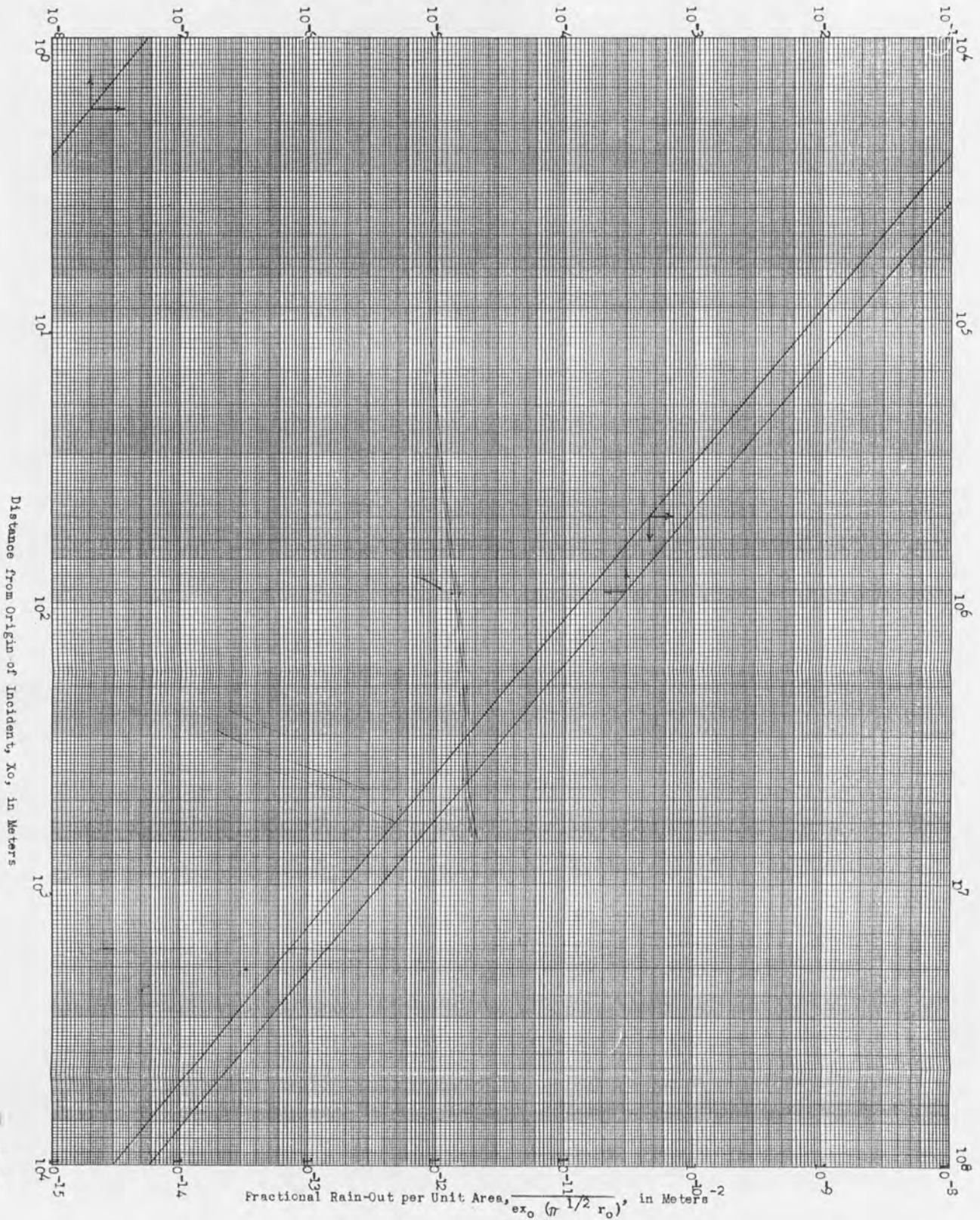


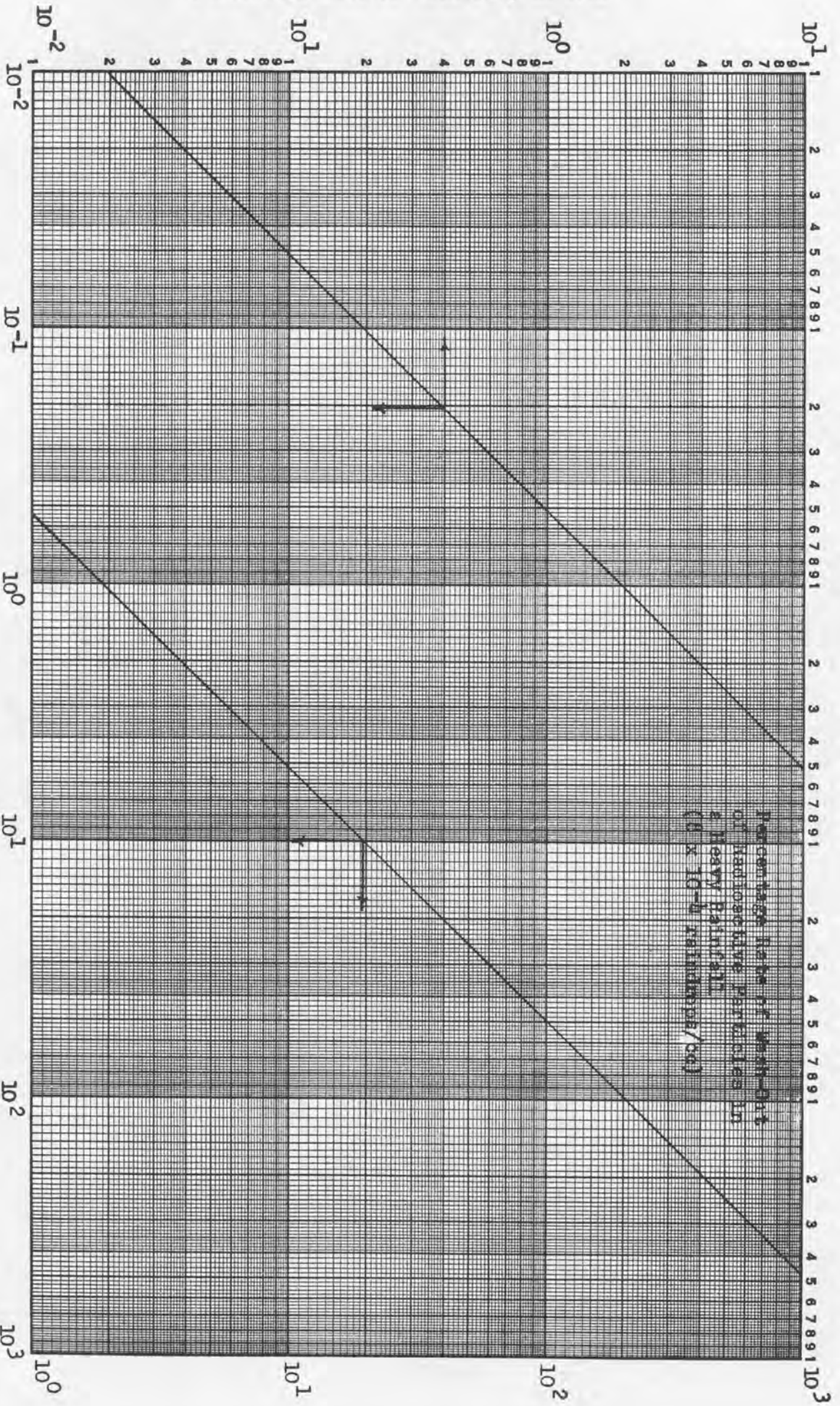
Figure 1

Fractional Rain Out per Unit Area, $\frac{1}{ex_0 (\pi^{1/2} r_0)}$, in Meters⁻²



KS-(3)

Percentage Rate of Wash-Out, in %/hr



Particle Size, in micron

Figure (2)