Some quantitative properties of a fallout model

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(Manuscript received April 4, 1963; revised version August 31, 1963)

ABSTRACT

According to a suggested model of the vertical transport of small particles in the upper air, concentration and the accumulation on the ground after an instantaneous injection have been calculated for the case of particles with a density of 5 g/cm³ and with sizes up to 0.005 mm. Equations and parameters are presented in detail.

1. Introduction

The vertical transport of small particles in the free atmosphere is essentially due to gravitational settling, eddy diffusion and vertical motion of the air. In addition the particles may be captured by rain and snow and brought to the ground. However, the material balance requires the long time global mean effects of the vertical wind components to vanish. A simplified model with special attention to high latitude conditions based on these processes was suggested to the author in 1959. Together with data from the literature and with some further assumptions this model can be used to give a quantitative picture of the vertical propagation of air-borne particles. Such a picture is the world-wide horizontal integral of the particle mass and has no local significance, because the velocities and space available for horizontal motion are several orders of magnitude bigger than the vertical ones. Therefore, it shows the relative importance of the particleconnected gravitation forces and the environmental turbulence and precipitation effects rather than observable phenomena.

In this work the flux and concentration of particles in the air after an instantaneous injection and their accumulation on the ground have been studied. As these properties depend not only on the atmospheric conditions but also on the shape and weight of the particles, a treatment of all combinations of parameters would not be practical. Therefore, approaching the case of radioactive fallout from bomb tests the calculations have been restricted to spherical particles with a density of 5 g/cm³ and with diameters up to 5μ. No assumption concerning the size spectrum of the particles has been made, but six single sizes have been studied separately. Data regarding the air were taken from the ARDC Model Atmosphere (1959).

Particles larger than 5 μ in diameter have very large settling velocities compared with the diffusion rates, making their downward transport a nearly trivial straight-line fall, a case in which the concept of concentration is not useful.

Further, the following assumptions have been made:

Above 30 km the diffusion is negligible making this altitude a roof of the distribution volume for particles injected below.

Between ten and eleven kilometers altitude there is a transition layer including the tropopause between the troposphere and the stratosphere.

At the earth's surface very complex conditions prevail, as the ground is a reflector for the air and an absorber for the particles. However, these phenomena can be treated in terms of a measured ratio between the deposition rate and the surface air concentration, the socalled "deposition velocity".

List of symbols

The following symbols will be used below.

Φ particle flux (t^{-1})

c particle concentration (l^{-1})

M particle contents of the atmosphere

F	particle deposition on the ground
R	wet particle deposition on the ground
DD	dry particle deposition on the ground
z	altitude above the ground (sea-level), upwards taken as positive (l)
r	"roof"-altitude (l)
v	gravitational settling velocity of the particles ($l\,t^{-1}$)
ϱ	density of particles ($m\,l^{-3}$)
d	diameter of particles (l)
g	gravitational acceleration ($l\,t^{-2}$)
μ	viscosity of the air ($m\,l^{-1}t^{-1}$)
λ	mean free path of air molecules (l)
v'	deposition velocity at the ground, due to other causes than gravitation ($l\,t^{-1}$)
n	number density of air molecules (l^{-3})
D	diffusion coefficient (l^2t^{-1})
p	air pressure ($m\,l^{-1}t^{-2}$)
T	absolute temperature
t	time (t)
k	coefficient for absorption of particles by rain and snow (t^{-1})
h	height of altitude intervals (l)
N	number of altitude intervals

2. Mathematical formulation of the model

The dry transport of the particles is given by the equation (LETTAU, 1951)

$$\Phi = -vc - nD\frac{\partial}{\partial z}\left(\frac{c}{n}\right) \tag{1}$$

or, assuming the air to be an ideal gas,

$$\Phi = vc - D\frac{\partial c}{\partial z} - \frac{D}{H}c, \tag{2}$$

where $$H = \left[\frac{\partial \ln T}{\partial z} - \frac{\partial \ln p}{\partial z}\right]^{-1}$$

The time-dependence of the concentration after an instantaneous injection at $t = 0$ follows the partial differential equation

$$\frac{\partial c}{\partial t} = -\frac{\partial \Phi}{\partial z} - kc \tag{3}$$

with the boundary conditions

$$\Phi = -c(v+v') \text{ at } z = 0$$

$$\Phi = 0 \quad \text{at } z = r$$

$$c = c(z,0) \quad \text{when } t = 0; \quad c(z,0) \geqslant 0$$

or in different terms

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial z^2} + \frac{\partial c}{\partial z}\left(v + \frac{\partial D}{\partial z} + \frac{D}{H}\right) + c\left(\frac{\partial v}{\partial z} + \frac{1}{H}\frac{\partial D}{\partial z} - \frac{D}{H^2}\frac{\partial H}{\partial z}\right) - kc. \tag{4}$$

The dry flux to the ground is

$$\frac{\partial DD}{\partial t} = (v+v')c(0,t), \tag{5}$$

the wet flux via rain and snow, the rain-out rate, is

$$\frac{\partial R}{\partial t} = \int_0^r k(z)c(z,t)\,dz \tag{6}$$

and the total fallout rate is the sum of these expressions.

The deposition on the ground, the fallout, is the time-integral of the fallout rate

$$F(t) = R(t) + DD(t) = (v+v')\int_0^t c(0,t)\,dt + \int_0^t\int_0^r k(z)c(z,t)\,dz\,dt \tag{7}$$

or it can be deduced according to the rule of conservation of matter

$$F(t) = \int_0^r [c(z,0) - c(z,t)]\,dz = M(0) - M(t), \tag{8}$$

if $F(0)$ is assumed to be zero. As $c \geqslant 0$, F is a non-decreasing and M a non-increasing function.

3. Numerical treatment

The numerical solution of these equations was performed on a PACE 15-131 R analogue computer (PRAWITZ, 1962). However, because the machine operates with one independent variable only and equation (4) has two, time and altitude, the derivatives in z were replaced by finite differences, which means that the altitude range was divided into layers of equal height. The layers are numbered from the ground upwards. Thus, layer one has the ground as its lower boundary. The concentration in the nth layer is called C_n and follows the equation deduced from equation (3)

$$\frac{\partial C_n}{\partial t} = \dot{C}_n = -\frac{1}{h}\left(\Phi_{n\to n+1} + \Phi_{n-1\to n}\right) - k_n C_n, \quad (9)$$

where $k_n = 1/h \int_{(n-1)h}^{nh} k(z)dz$ and $\Phi_{n\to n+1}$ is the flux between the layers n and $n+1$, or from equation (4)

$$\dot{C}_n = \frac{1}{h^2}[D_+(C_{n+1} - C_n) + D_-(C_{n-1} - C_n)]$$
$$+ \frac{1}{h}\left[C_{n+1}\left(v_+ + \frac{D_+}{H_+}\right) - C_n\left(v_- + \frac{D_-}{H_-}\right)\right] - k_n C_n, \quad (10)$$

where the suffixes + and − are applied to the constants for the upper and lower boundary of the layer, respectively.

For the roof-layer we get

$$\dot{C}_N = \frac{1}{h^2}D_-(C_{N-1} - C_N) - \frac{1}{h}C_N\left(v_- + \frac{D_-}{H_-}\right) - k_N C_N \quad (11)$$

and for the ground layer

$$\dot{C}_1 = \frac{1}{h^2}D_+(C_2 - C_1) + \frac{1}{h}\left[C_2\left(v_+ + \frac{D_+}{H_+}\right) - C_1(v_- + v')\right] - k_1 C_1. \quad (12)$$

The particle content of the air is

$$M = h\sum_{i=1}^{N} C_i. \quad (13)$$

The fallout is obtained from the wet and dry fluxes to the ground, equations (5) and (6),

$$\dot{F} = \dot{R} + D\dot{D} = C_1(v(0) + v') + h\sum_{i=1}^{N} k_i C_i. \quad (14)$$

Combining equations (10–13) we have

$$h\sum_{i=1}^{N}\dot{C}_i = -\dot{F} = \dot{M} \quad (15)$$

and after integrating and assuming $F(0) = 0$

$$M(0) - M(t) = F(t) \quad (16)$$

and because no v and k value is zero,

$$F(\infty) = R(\infty) + DD(\infty) = M(0). \quad (17$$

The flux between any two layers $n+1$ and n is

$$\Phi_{n\to n+1} = -\frac{1}{h}D(C_{n+1} - C_n) - C_{n+1}\left(v + \frac{D}{H}\right). \quad (18)$$

The error ε_n introduced by solving the equations (10–12) instead of equation (4) is given by

$$\frac{\partial \varepsilon_n}{\partial t} = \frac{\partial}{\partial t}\left(C_n - \frac{1}{h}\int_{z_n - \frac{1}{2}h}^{z_n + \frac{1}{2}h} c(z,t)dz\right)$$
$$\approx \frac{h}{2}\frac{\partial}{\partial z}\left[U(z_n)\frac{\partial}{\partial z}\left(1 - \frac{h}{4}\frac{\partial}{\partial z}\right)c(z_n,t)\right], \quad (19)$$

where z_n is the midpoint in layer n and $U = v + \partial D/\partial z + D/H$.

4. Radioactive properties

Atomic bomb debris contains radioactive fission products, which after their creation decay with time according to the approximate relation

$$A = A_1 t^{-1.2} \quad (t > 1 \text{ hour}),$$

where A_1 is the radioactivity at unit time. For $t = 1$ day $A(t) = 70$ Megacuries per kiloton fission but for the sake of normalization we assume $A_1 = 1$, so the activity on the ground A_F becomes

$$A_F(t) = F(t)t^{-1.2}. \quad (20)$$

As $F(0) = 0$ surpressing the behaviour of $t^{-1.2}$ at zero time $A_F(0) = 0$.

The accumulated dose on the ground D_F is

$$D_F(t) = \int_0^t A_F(\tau)d\tau. \quad (21)$$

The radioactive properties can be included in the analogue computer set-up (Prawitz, 1962). However, in this work the digital computer (FACIT-EDB) results of Bergquist (1963) have been reproduced.

5. Values of the parameters

The height of the layers, h, was chosen to be 2 km. As the roof-altitude r is 30 km, the number of layers N is 15. For instance layer 7 extends from 12 to 14 km altitude. The layers 1–5 belong to the troposphere, the lower half of layer 6 is the tropopause, and the upper half of layer 6 and the layers 7–15 are in the stratosphere.

The gravitational settling velocities, v, were calculated according to Stokes' law with Cunningham's correction applied

$$v = \frac{\varrho g d^2}{18\mu}\left[1+\frac{2\lambda}{d}(A+Be^{-Cd/2\lambda})\right],$$

where A, B, and C are constants, by GREEN & LANE (1957) reported to be $A = 1.25$, $B = 0.44$, and $C = 1.09$. Values of the constants g, μ and λ were taken from the ARDC Model Atmosphere.

The deposition velocity, v', is not very easily extracted from the literature. Many different values are reported. A value of 0.33 km/day, has been used in this work. It is calculated from measurements of fallout between 1 Sept. 1956 and 30 Sept. 1959 reported by SMALL (1959) and is the dry day mean for periods with very low total deposition velocity $(v+v')$ a long time after bomb-tests, when very small particles with a negligible gravitational settling velocity are assumed to be alone in the surface air. Monthly means of v' are scattered within a factor 2. The work of PEIRSON & KEANE (1962) shows the same scattering but a lower mean value 0.22 km/day. ERIKSSON (1960) has reported deposition velocities of sea salt particles between 0.18 and 3.03 km/day with the mean 1.24 km/day over land in Sweden and estimates a value 0.60 km/day over the sea. However, these higher values refer also to bigger particles with significant settling velocities. The uncertainty in v' justifies the approximation using C_1 instead of $c(0)$ in the equations (12, 14). The ratio $c(\frac{1}{2}h)/c(0)$ has been measured by PEIRSON, CROOKS & FISHER (1960) in Great Britain and found to be ~ 2.

The diffusion coefficient is taken from MACHTA & LIST (1959) and is 4 km²/day in the troposphere and 0.2 km²/day in the stratosphere below 30 km altitude and zero above this. In the tropopause layer there has been used a weighted mean of these two values, 0.4 km²/day, estimated with data given by NORDSTRÖM (1960).

The H-values were calculated according to the ARDC Model Atmosphere.

The rain-out constant, k, could not be directly extracted from the literature. Relative rain-intensities were taken from ATLAS & KESSLER (1957). Relative absorption constants for different particle sizes were taken from GREENFIELD (1957). This information gives the k_n-values except for a common scale-factor. The mean rain-out for periods a long time after bomb tests with a very low total deposition rate when the smallest particles only are assumed to remain in the air, is extracted from SMALL (1959) to be 88.3 % of the total fallout. The unknown scaling-factor was found by varying it until the solution of the equations (10–12, 14) for $v = 0$ gave the proper value of the rain-out.

All values of v, H and k actually used in this work are tabulated in an earlier report (PRAWITZ, 1961).

Results

Among the parameters in the system of equations (10–12, 14, 18) D, H, and v' depend only on the properties of the atmosphere, while k and v also depend on the size of the particles and v in addition on their density. Thus, there is one system for every pair of particle density and diameter. Moreover, the initial conditions may be chosen arbitrarily. Thus it seems as the possible combinations of input data are unlimited. Of these a few have been selected with the intention to give an over all view without too many details.

In all the calculations a particle density of 5 g/cm³ was used. For the results shown in Tables 1–5 and Figs. 1–7 the initial conditions are $C_9 = C_{10} = 1$ and all other $C_n = 0$, which means that the particles are injected with equal density into the altitude-range 16–20 km, approximately the case of a 500 kton air burst.

TABLE 1. *Asymptotic percentage of wet and dry accumulation on the ground.*

Kind of accumulation	Particle diameter in μ					
	~ 0	0.2	0.5	1	2	5
Wet	88.3[a]	88.6	88.2	87.5	95.5	99.8
Dry	11.7	11.4	11.8	12.5	4.5	0.2

[a] According to definition.

TABLE 2. *Time in days required for 10, 50, and 90 % of the particles to accumulate on the ground.*

Per cent particles on the ground	Particle diameter in μ: ~0	0.2	0.5	1	2	5	Per cent particles in the air
10	47	45	42	36	24	8	90
50	174	140	121	93	56	17	50
90	750	460	355	244	122	32	10

TABLE 3. *Maximum fluxes in per cent/day and associated times in days for different particle sizes.*

Particle diameter μ	Fallout rate	Rainout rate	Dry deposition rate	Flux from the tropopause into the troposphere
~0	0.481 %/day	0.424	0.056	0.487
	58.3 days	59.0	58.6	50.2
0.2	0.520	0.461	0.060	0.527
	56.2	56.3	56.3	47.7
0.5	0.593	0.523	0.070	0.601
	55.0	55.8	56.1	46.9
1	0.766	0.675	0.091	0.778
	50.9	51.2	51.6	41.7
2	1.352	1.282	0.065	1.371
	37.6	38.9	38.4	30.0
5	5.004	4.995	0.008	5.142
	13.8	13.6	13.2	11.5

TABLE 4. *Times when fallout rate definitely decreases below 1 % of its maximum value.*

Particle diameter in microns	~0	0.2	0.5	1	2	5
Time in days	1250	1050	760	600	260	60

TABLE 5. *Residence time in the troposphere for different particle sizes.*

Particle diameter in μ	<1	2	5
Time in days	7.5	4.7	2.1

The particle diameters studied are 0(< 0.1), 0.2, 0.5, 1, 2, and 5 μ.

The functions primarily obtained from the analogue computer are the C_n, $\Phi_{n\to n+1}$, $\dot{F}$, $\dot{R}$, $\dot{DD}$, F, R, DD, and also the tropospheric, stratospheric, and total air inventory. A couple of such functions have been reported earlier (PRAWITZ, 1961). These raw computer out-puts have been further investigated to give the results in the present work.

The development with time of the vertical concentration distribution of the different particle sizes is shown in Figs. 1–3.

The ratios of the concentration above and below the tropopause, here defined as C_7/C_5, are given in Fig. 4.

The time dependence of the deposition on the ground, F, is given in Fig. 5.

The radioactivity from particles on the ground and the accumulated dose, as defined by equations (20, 21), are reproduced here from the work of BERGQUIST (1963).

The asymptotic values of the wet and dry deposition on the ground as defined by equation (17) are given in Table 1.

Times when 10, 50, and 90 per cent of the

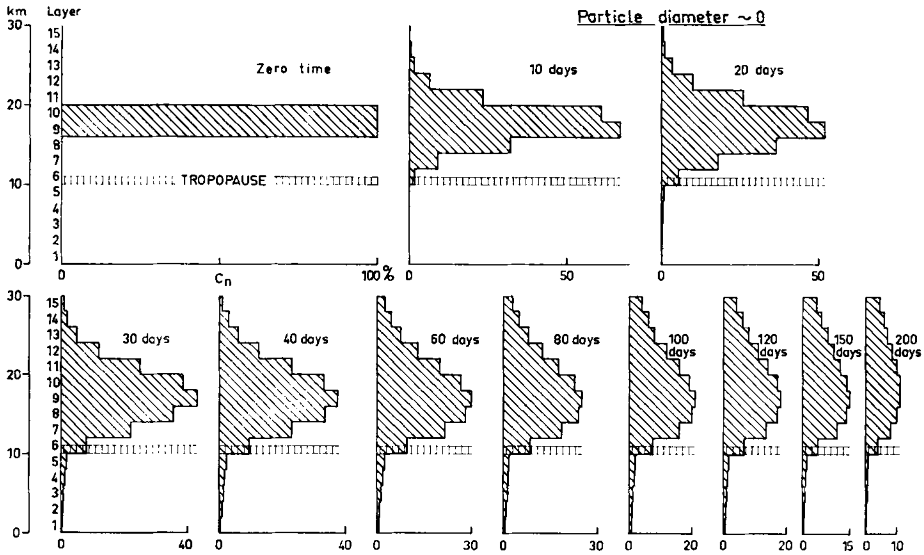

Fig. 1.

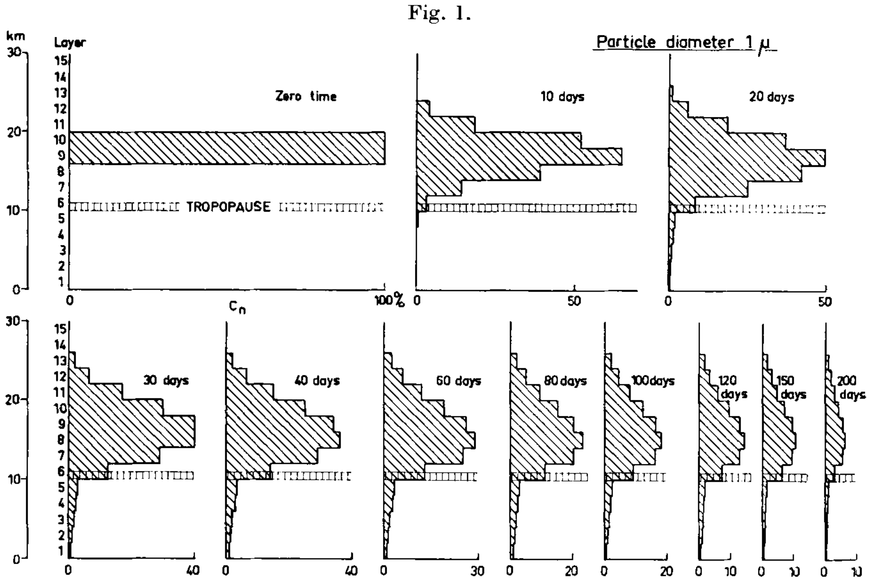

Fig. 2.

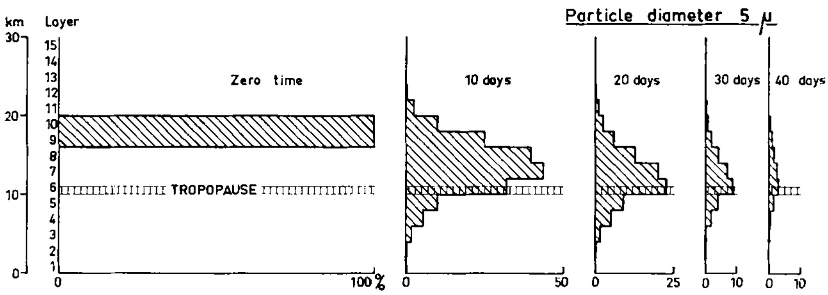

Fig. 3.

FIGS. 1–3. Vertical distribution of particle concentrations at different times.

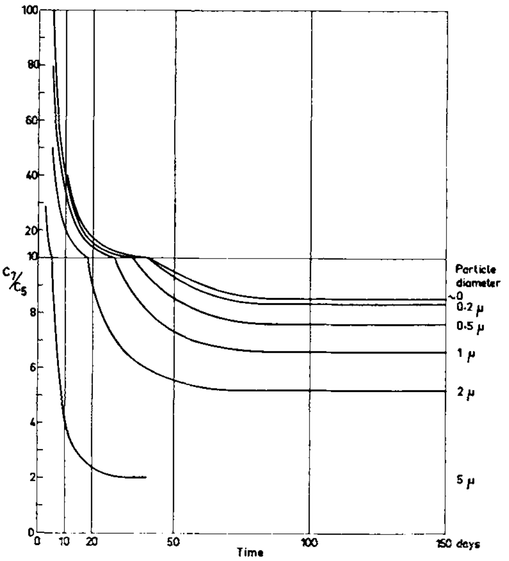

FIG. 4. Ratio of the concentration above and below the tropopause, C_7/C_5, as a function of time for different particle sizes.

particles are present in the air (and on the ground) are given in Table 2.

The coordinates of maximum points of the fallout rate, the rain-out rate, the dry deposition rate, and the flux from the tropopause into the troposphere, defined as $\dot{F}$, $\dot{R}$, $D^{\cdot}D$, and Φ_{6-5} respectively are listed in Table 3.

From equation (17) it is obvious that $\dot{F}(\infty) = 0$. In order to indicate the approach to this value, times when $\dot{F}(t)$ definitely has decreased below one per cent of its maximum value are given in Table 4.

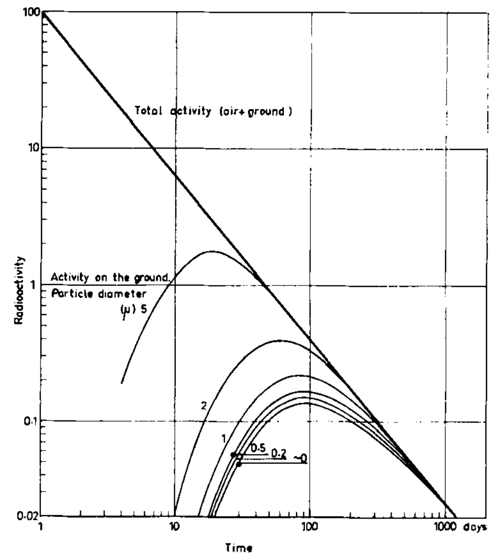

FIG. 6. Relative radioactivity on the ground as a function of time for different particle sizes (Bergquist, 1963).

It happens that the functions $\dot{F}$ and $\Phi_{6\to5}$ are congruent from a relatively short time after passing their maximum values. The time delay between the "tails" of these functions, which may be regarded as a "residence time" in the troposphere, is given in Table 5.

Finally the influence of different injection altitudes on the accumulation on the ground for 1 μ particles is shown in Fig. 8. Every $F(t)$-function has been calculated in response to an injection in one layer only, all others being empty. They are numbered according

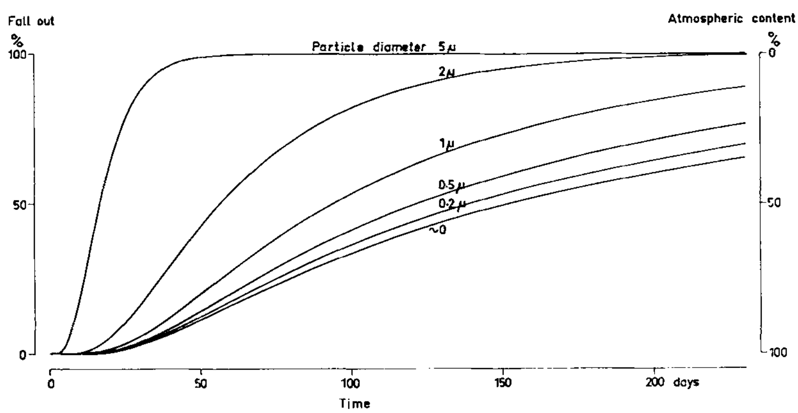

FIG. 5. The fallout, F, and atmospheric content, M, in per cent injected mass as a function of time for different particle sizes.

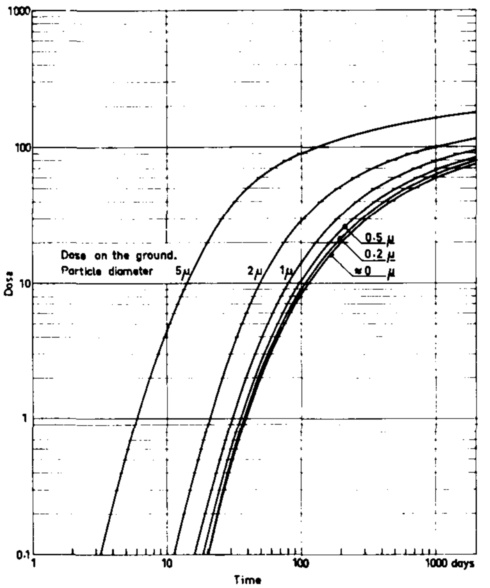

FIG. 7. Relative radioactive doses accumulated on the ground as a function of time for different particle sizes (Bergquist, 1963).

to the rule: $F_i(t)$ has the initial condition $C_i = 1$, $C_n = 0$, $n \neq i$, $i = 1, 2, \dots N$. The functions F_1 to F_5 are given for formal reasons only. They develop so fast that global mean value considerations may not be valid.

The numerical errors introduced have been described earlier (PRAWITZ, 1962). They are of the order of one per cent and seem to be negligible compared with the effect of the errors associated with the numerical values of the parameters, primarily D and k_n.

Acknowledgement

The author is extremely indebted to Dr. K. Edvarson, who in 1959 suggested the basic ideas behind the studied model, for most valuable discussions and comments during the course of the work. He also wishes to express his thanks to Mr. G. Bergquist for his skilful extracting the figures from the computer output data, and the analogue computer group at FOA 2 for their valuable support.

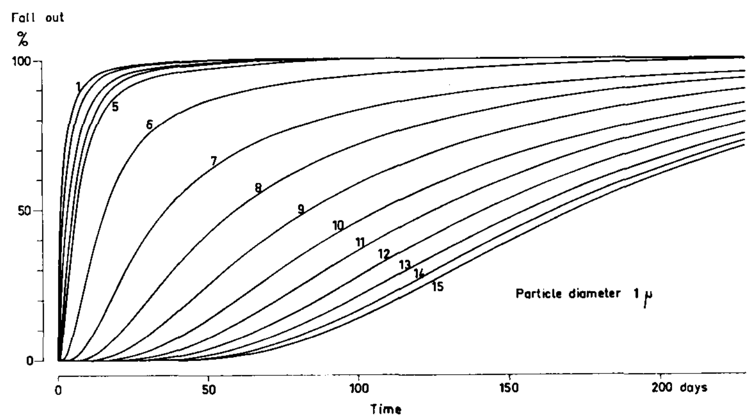

FIG. 8. Fallout in per cent injected mass as a function of time for different injection altitudes. The numbers on the curves refer to the injection layer, i.e. 8 means that $C_8 = 100\,\%$ at zero time, the other layers being empty.

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