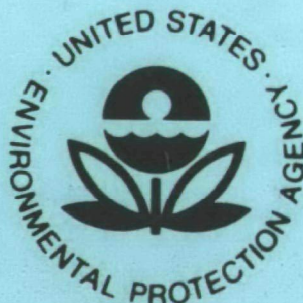


GENERALIZED MODEL OF THE TIME-DEPENDENT WEATHERING HALF-LIFE OF THE RESUSPENSION FACTOR

FEBRUARY 1977



**U.S. ENVIRONMENTAL PROTECTION AGENCY
OFFICE OF RADIATION PROGRAMS
LAS VEGAS FACILITY
LAS VEGAS, NEVADA 89114**

Technical Note
ORP/LV-77-4

GENERALIZED MODEL OF
THE TIME-DEPENDENT WEATHERING HALF-LIFE
OF THE RESUSPENSION FACTOR

GEORGE V. OKSZA-CHOCIMOWSKI

FEBRUARY 1977

Office of Radiation Programs-Las Vegas Facility
U.S. Environmental Protection Agency
Las Vegas, Nevada 89114

PREFACE

The Office of Radiation Programs of the U.S. Environmental Protection Agency carries out a national program designed to evaluate population exposure to ionizing and non-ionizing radiation, and to promote development of controls necessary to protect the public health and safety.

Radioactive contaminants discharged from facilities in the nuclear fuel cycle may deposit on the ground surface, to be subsequently resuspended and, possibly, inhaled by members of the general public. This report introduces time-dependent models of empirical parameters commonly used in characterizing the extent and duration of the significant hazards posed by resuspension. Readers of this report are encouraged to inform the Office of Radiation Programs of any errors or omissions. Comments or requests for further information are invited.

A handwritten signature in black ink, reading "Donald W. Hendricks". The signature is written in a cursive, slightly slanted style.

Donald W. Hendricks
Director, Office of
Radiation Programs, LVF

TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	iv
LIST OF FIGURES	v
LIST OF TABLES	viii
LIST OF SYMBOLS	ix
ACKNOWLEDGMENT	xi
INTRODUCTION	1
RESUSPENSION FACTOR MODELS	5
TIME-DEPENDENT HALF-TIME MODEL BASED ON ANSPAUGH'S MODEL OF THE RESUSPENSION FACTOR	12
BASES OF PROPOSED INTERIM MODEL OF THE TIME-DEPENDENT HALF-TIME	17
Initial Values of $T_{1/2}(t, R_f(0)/R_f(\infty))$	17
Values of $T_{1/2}(t, R_f(0)/R_f(\infty))$ Within One Year After Deposition	18
Values of $T_{1/2}(t, R_f(0)/R_f(\infty))$ After One Year From Deposition	22
Final Values of $T_{1/2}(t, R_f(0)/R_f(\infty))$	25
PROPOSED INTERIM MODEL OF TIME-DEPENDENT HALF-TIME	27
APPLICATION	33
SUMMARY AND COMMENTS	40
REFERENCES	41

ABSTRACT

A generalized model has been developed to predict the changes with time in the weathering half-life of the resuspension factor for plutonium 239 and other long-lived radioactive contaminants. The model is largely based on assumptions and empirical data presented by other authors, and is applicable to a wide range of average conditions akin to those for which data is available. These conditions are parametrically described as ratios of initial and final resuspension factors, valid for a given locality.

As a direct application of the above model of time-dependent half-life, the corresponding general model of time-dependent resuspension factor, also by the present author, is introduced in the report. Included in the latter are graphs of both models for a range of conditions, as well as graphic comparisons of specific cases of these general models with models proposed by others.

LIST OF FIGURES

<u>Number</u>		<u>Page</u>
1	Half-Time $T_{1/2}$ as function of time and local conditions (expressed as ratios of initial-to-final resuspension factors, $R_f(0)/R_f(\infty)$), as derived (Equation (14)) from Anspaugh's model of $R_f(t)$.	14
2	Tungsten-181 air activity as a function of time at five downwind stations within six miles that were closest to the line of maximum deposition from Project Schooner [From Anspaugh et al. (1973)].	18
3	Median weekly air concentrations, corresponding to three isopleths, from Operation Plumbbob [From Wilson et al. (1960)].	20
4	Least-square fit to gross-gamma air activity levels three to eleven months following Baneberry venting [From Anspaugh et al. (1973)].	21
5	Contour representation of plutonium-239 distribution at Rocky Flats [From Volchok (1971)].	23
6	Airborne plutonium at Sampling Station S-8. Adapted from Sehmel and Orgill (1974).	24
7	Half-time $T_{1/2}$ as function of time and local conditions (expressed as ratios of initial-to-final resuspension factors $R_f(0)/R_f(\infty)$) according to proposed model and "Case 2" constants (Table 2). Curves numbered according to ratio $R_f(0)/R_f(\infty)$, in orders of magnitude, i.e., "2" corresponds to $R_f(0)/R_f(\infty) = 10^2$, etc.	29

- 8 Half-time $T_{1/2}$ as function of time and local conditions (expressed as ratios of initial-to-final resuspension factors $R_f(0)/R_f(\infty)$) according to proposed model (Equation (33)) and "Case 1" constants (Table 2). Curves shown correspond to upper and lower limits of range of values presumed possible for $R_f(0)/R_f(\infty)$. 31
- 9 Resuspension factors as functions of time based on the proposed model of time-dependent half-time $T_{1/2}$, assuming various initial values but the same final resuspension factors. The first captioned ordinate represents $R_f(t)$ values presumed possible based on Table 1. Some of the higher values do not apply to outdoors pollutant resuspension by wind, under normal conditions, but have been included for the purpose of demonstrating applications of the general model. The second captioned ordinate pertains to the generalized case. 34
- 10 Resuspension factors as functions of time based on the proposed model of time-dependent half-time $T_{1/2}$, assuming the same initial value but various final resuspension factors. The first captioned ordinate shows values presumed possible based on Table 1. The initial value of 10^{-2} m^{-1} is highly improbable for outdoor pollutant resuspension by wind, under normal conditions, but serves to illustrate applications of the model for the full range of values postulated in the model development. The second captioned ordinate pertains to the generalized case. 35

NumberPage

- 11 Three expressions of the time-dependent resuspension factor based on the proposed model of the time-dependent half-time $T_{1/2}$, with initial and final values as assumed in previous models with which these expressions are compared. See also Table 3. 37
- 12 Two forms of the proposed model of weathering half-time $T_{1/2}$ as function of time and local conditions (expressed) as ratios of initial-to-final resuspension factors $R_f(o)/R_f(\infty)$, compared ① with Anspaugh's model and ② with Langham's and Kathren's models. Values of $R_f(o)$ and $R_f(\infty)$ employed in the proposed model correspond to those used in the comparison models. 38

LIST OF TABLES

<u>Number</u>		<u>Page</u>
1	Resuspension factors for plutonium and other radioisotopes [From Mishima (1964)]	3
2	Examples of constants used in general model	28
3	Comparison of proposed model (Case 1) of the time-dependent weathering half-time $T_{1/2}$ with previous models, implicit or derived, and corresponding models of the time-dependent resuspension factor, including numerical values	36

LIST OF SYMBOLS

The numbers in parentheses following the description of each symbol refer to the equations in which the symbols are first used or defined. The symbols have been separated into these groups, according to their primary function in the context of this report, as follows: constant parameters, time-dependent parameters or variables, and special values of time. Some explanations are included.

- A = constant coefficient, days, in (31)
- B = constant, dimensionless, in (31)
- C = constant coefficient, day^{-D} , in (31)
- D = constant exponent, dimensionless, in (31)
- K = constant coefficient, $(\text{days})^{-1/2}$, in (11)
- R_f = constant resuspension factor, m^{-1} , in (1), (4)
- $R_f(0)/R_f(\infty)$ = ratio of initial-to-final resuspension factors,
determined or predicted for a given site, in
(14), (24)
- S_A = total contamination per unit area, regardless of depth
at which contaminant is found, constant, $\mu\text{Ci}/\text{m}^2$, in (8)
- $T_{1/2}$ = weathering half-life, also "half-time", days, in (2)
- λ_A = constant attenuation factor, days^{-1} , in (2)

- $C(t)$ = time-dependent airborne concentration, $\mu\text{Ci}/\text{m}^3$, in (3)
- $R_f(t)$ = time-dependent resuspension factor, m^{-1} , in (8)
- $S_a(t)$ = time-dependent surface contamination, $\mu\text{Ci}/\text{m}^2$, in (4)
- $T_{1/2}(t)$ = time-dependent weathering half-life, days, in (12)
- $T_{1/2}(t_i)$ = empirically obtained weathering half-times, days,
corresponding to indexed values of time t_i , where
 $i = 1, 2, 3$, in (25), (26), 27)
- $T_{1/2}(t_{\min})$ = minimum value of weathering half-life, for values
of $t > t_{as}$ (see infra), days, in (18). Literally,
the expression signifies "the value of $T_{1/2}$ at a
time t_{\min} (see infra) corresponding to a minimum
value of $T_{1/2}$ ". Such awkward notation was adapted
to maintain continuity of exposition in the text
- $\lambda(t)$ = time-dependent attenuation factor, days^{-1} , in (12)

t = time, days

t_{as} = time at which values of $T_{1/2}$ become infinitely large, days,
in (16), (17)

t_i = indexed values of time corresponding to empirically
observed weathering half-times, days, in (25), (26),
(27)

t_{min} = time, days for $t > t_{as}$, at which the weathering half-time
 $T_{1/2}$ reaches a minimum, in (18)

ACKNOWLEDGMENT

The author gratefully acknowledges the assistance and advice of numerous individuals in the preparation of this report. Special recognition is extended to Dr. W. Wood, of the Office of Radiation Programs, Criteria and Standards Division, of the Environmental Protection Agency, and to Mr. V. E. Andrews, Dr. P. N. Lem and Mr. H. K. Maunu of the Office of Research and Development, Environmental Monitoring and Support Laboratory (EMSL) - Las Vegas, of the same agency.

The author, although recognizing the assistance of others, accepts full responsibility for the contents of this report.

INTRODUCTION

Particulate matter deposited on a surface can be resuspended by mechanical action or wind forces, phenomena familiar to anyone who has erased a blackboard, or complained about dust on a windy day. When material thus prone to resuspension includes radioactive particles, as in localities contaminated, in some cases, by nearby nuclear facilities, continuous occupancy of such areas may present a potential health hazard, ascribed to the possible inhalation of radioactive particulates of respirable size.

The extent to which radioactive contaminants can be resuspended from the ground or other environmental surfaces is often characterized by an empirically determined quantity, the "resuspension factor". This is defined as the ratio of volumetric airborne pollutant concentration at some given height to the areal concentration of the pollutant on the ground. The height of interest may be specified to be within the "breathing zone" of the average individual, from which respirable aerosols may penetrate into the individual's lungs. Coupling thusly obtained resuspension factors with some realistic assumptions regarding the fraction of respirable-size particles present in the total concentration of breathing height, these resuspension factors may provide a rough index of potential inhalation concentration due to a given ground concentration of contaminant.

Some criticism can be leveled at the concept of resuspension factor in general. In the first place, it assumes that the air concentration above a contaminated surface is directly proportional to this surface contamination level, rather than on the extent of ground contamination upwind of the sampling site, as would be more logical and, in fact, was found to be true (Stewart, 1967; Mishima, 1964). In the second place, the resuspension factor, as a constant of proportionality relating air and ground concentrations, is meant to include the effects of a myriad parameters, such as wind velocity, surface roughness, physical and chemical characteristics of both pollutant and soil surface, vegetation cover, etc. Whereas some of these factors may change little at a particular site, some others, such as wind velocity, tend to be less constant. Consequently, the resuspension factor must be

considered to be, in a strict sense, an empirically determined value applying only to specific or prevailing conditions at a given site, at a given time or for a specified time period, respectively. Thus, it is not surprising that the measured values of R_f vary from $10^{-2}m^{-1}$ to $10^{-13}m^{-1}$, as shown in Table 1 (Mishima, 1964).

In view of the described inaccuracies, continued use of resuspension factors may be justifiable only under such conditions as would tend to minimize the liabilities of the concept, or allow to compensate for these adverse characteristics. Ideally, these conditions would include uniform distribution, unchanging with time, of the contaminant over a large ("infinite") area, in the absence of additionally contributing "sources" of significance (inadequately operating nuclear facilities) or "sinks" (rivers, lakes, other large bodies of water) and of unpredictable or extreme climatic conditions. To simplify discussion, such optional conditions are assumed to exist for all cases considered in this report, and to have existed whenever actual numerical values used in the report were originally obtained.

Evaluation of potential health effects from continuous occupancy of a contaminated region involves both estimations of the amount of material apt to be resuspended and the duration of the hazard. The latter necessitates establishing some reliable or credible models of the decrease of airborne concentration with time, which may be effected by describing correspondingly varying time-dependent resuspension factors. The present report cites a number of observations indicating that concentrations of resuspended materials do, indeed, decrease with time, and several models based on such observations.

The models mentioned above had to be, to considerable degree, site-specific (corresponding to conditions an exact replica of which would not be found at any other site) and also "date-specific" (produced by circumstances that would never be exactly repeated, at a given site, at any later time). The relative abundance of independent observations, when contrasted with the paucity of reliable data on which accurate models could be based, suggested the advantage of developing a general model that would partake of the features and assumptions of previous models, while retaining enough flexibility to allow incorporation of new data.

**TABLE 1 RESUSPENSION FACTORS FOR PLUTONIUM AND OTHER RADIOISOTOPES
[FROM MISHIMA (1964)]**

Conditions of Resuspension	Resuspension Factor
Average Resuspension Factor in Accidents Involving Plutonium	$4 \times 10^{-6} \text{ m}^{-1}$
Vehicular Traffic (Nevada)	7×10^{-8}
People Working or Active in a Closed Area	4×10^{-5}
Dirty Rural, Suburban, or Metropolitan Areas	7×10^{-6}
People Working or Active in an Open Area	2×10^{-6}
Isolated Area	$\sim 7 \times 10^{-7}$
Resuspension of Aged Plutonium Deposit (0.74 to 752 $\mu\text{Ci}/\text{m}^2$) from "Plumbob"	6.2×10^{-10} to 10^{-13}
Plutonium Oxide, No Movement	2×10^{-8}
Plutonium Oxide, 14 steps/min	10^{-5}
Plutonium Oxide, 36 steps/min	5×10^{-5}
Plutonium Nitrate, No Movement	2×10^{-8}
Plutonium Nitrate, 14 steps/min	10^{-6}
Plutonium Nitrate, 36 steps/min	5×10^{-6}
Plutonium Oxide, Change Room ($>3000 \text{ ft}^2$), 9 air changes/hr, 0.01 $\mu\text{Ci}/\text{m}^2$, 4 to 6 persons active in area	
"Loose" Contamination (estimated by smears)	10^{-3}
"Loose" Contamination (estimated by water-detergent wash)	2×10^{-4}
Changing Coveralls, Static Sampler, No Ventilation	2.8×10^{-3}
Changing Coveralls, Personal Sampler, No Ventilation	6.4×10^{-3}
Personnel Traffic in a Small Unventilated Room	4×10^{-3}
Proposed Resuspension Factors for Plutonium Oxide:	
Outdoors (quiescent conditions)	10^{-6}
Outdoors (moderate activity)	10^{-5}
Indoors (quiescent conditions)	10^{-6}
Indoors (moderate activity)	10^{-4} to 10^{-5}
From Crater of Tower Shot, No Artificial Disturbance	8×10^{-8} *
Survey of Road, No Artificial Disturbance	8×10^{-8} *
Survey of Road, Landrover, D-Day + 4	1.4×10^{-5}
Survey of Road, Landrover, D-Day + 7	1.5×10^{-6}
Survey of Road, Tailboard of Landrover, D-Day + 7	2×10^{-5}
Survey of Road, D-Day + 1 and 2	4×10^{-7}
Sample Collection in Cab of Landrover, H-Hour + 5	6.4×10^{-5} **
Sample Collection in Cab of Landrover, H-Hour + 8	2.5×10^{-5} †
Uranium Sample Downwind of Crater, Sample Height, 1 ft Above Ground	3×10^{-4} ††
Uranium Sample Downwind of Crater, Dust Stirred Up, Sample Height: 1 ft	10^{-3} ††
Uranium Sample Downwind of Crater, Sample Height, 2 ft	10^{-5} ††
Plutonium Sampled 1 ft Above Ground, Vehicular Dust	5×10^{-4} ‡
Pedestrian Dust	1.5×10^{-6} ‡
Iodine-131, Enclosed (Chamberlain & Stanbury)	2×10^{-4} to 10^{-5}
Iodine-131, Open (Chamberlain & Stanbury)	2×10^{-6}
Yttrium-91, 0-8 μ Particles, Natural Turbulence, Sampled 1 ft Above Ground:	
Ground Contamination Level 1.8 $\mu\text{Ci}/\text{m}^2$	1.8×10^{-7}
Ground Contamination Level 6.8 $\mu\text{Ci}/\text{m}^2$	10^{-8}
Ground Contamination Level 24.6 $\mu\text{Ci}/\text{m}^2$	3×10^{-10}
Polonium-210, 0-8 μ Particles, Natural Turbulence, Sampled 1 ft Above Ground:	
Ground Contamination Level 0.6 $\mu\text{Ci}/\text{m}^2$	2×10^{-6}
Ground Contamination Level 3 $\mu\text{Ci}/\text{m}^2$	2×10^{-6}
U₃O₈, 0-4 μ Particles, Natural Turbulence, Sampled 1 ft Above Ground:	
Ground Contamination Level 112 g/m^2	2.4×10^{-6}
Grass Contamination Level 70 g/m^2	8×10^{-6}
Concrete Contamination Level 180 g/m^2	2×10^{-6}
Plutonium Oxide, Sampling Height: 5 ft	
Floor Level 0.1 $\mu\text{Ci}/\text{m}^2$, No Circulation	1.6×10^{-6}
Floor Level 24.6 $\mu\text{Ci}/\text{m}^2$, No Circulation	4.4×10^{-7}
Floor Level 0.1 $\mu\text{Ci}/\text{m}^2$, Fan	1.3×10^{-3}
Floor Level 0.91 $\mu\text{Ci}/\text{m}^2$, Fan	1.4×10^{-3}
Floor Level 0.086 $\mu\text{Ci}/\text{m}^2$, Fan and Dolly	1.0×10^{-2}
Floor Level 1.3 $\mu\text{Ci}/\text{m}^2$, Fan and Dolly	9.4×10^{-3}
Floor Level 1.3 $\mu\text{Ci}/\text{m}^2$, After Test	9×10^{-4}
Floor Level 1.1 $\mu\text{Ci}/\text{m}^2$, After Test	9×10^{-4}
Uranium, Sampling Height: 5 ft	
Floor Level 0.086 $\mu\text{Ci}/\text{m}^2$, No Circulation	1.8×10^{-6}
Floor Level 0.75 $\mu\text{Ci}/\text{m}^2$, No Circulation	2.2×10^{-7}
Floor Level 0.015 $\mu\text{Ci}/\text{m}^2$, Fan	1.5×10^{-4}
Floor Level 1.1 $\mu\text{Ci}/\text{m}^2$, Fan	1.1×10^{-4}
Floor Level 0.11 $\mu\text{Ci}/\text{m}^2$, Dolly	1.6×10^{-4}
Floor Level 1.3 $\mu\text{Ci}/\text{m}^2$, Dolly	1.3×10^{-4}
Floor Level 0.91 $\mu\text{Ci}/\text{m}^2$, Fan and Dolly	4.6×10^{-4}
Floor Level 1.0 $\mu\text{Ci}/\text{m}^2$, Fan and Dolly	1.9×10^{-4}

* One high value excluded

† Only ~20% of particles $< 6 \mu$

‡ Particles primarily in 20 to 60 μ size range, $< 1\%$ $< 6 \mu$

** Only ~10% of particles $< 6 \mu$

†† Only ~5% of particles $< 6 \mu$

The present report describes what may well be called a "back-door" approach in developing a general model of the time-dependent resuspension factor. It is rooted initially in a collation of known or accepted facts about rates of decrease of airborne contamination, as characterized by the "decay half-life" or "half-time" of such decrease, originally assumed to be exponential. Based on these facts, a general model of the "time-dependent weathering half-life of the resuspension factor" is developed, followed by the corresponding general model of the resuspension factor itself. Conditions representative of the various sites from which data is available are operationally described by "ratios of initial-to-final resuspension factors", as observed or expected at each of the given sites.

RESUSPENSION FACTOR MODELS

One of the earliest attempts to devise a simple means of predicting the extent of resuspension of pollutant from a previously contaminated surface dates back to 1956. P. S. Harris and W. H. Langham correlated surface deposition and air concentration measurements of plutonium at the Nevada Test Site (NTS) by defining a quantity known as the "resuspension factor", R_F (Langham, 1971)

$$R_F = \frac{\text{Air Concentration } (\mu\text{Ci plutonium}/\text{m}^3)}{\text{Surface Deposition } (\mu\text{Ci plutonium}/\text{m}^2)} \quad (1)$$

From measurements made at two different times following a contaminating event, under circumstances involving "extensive vehicular traffic", it was concluded that $R_F \approx 7 \times 10^{-5} \text{m}^{-1}$ applied to "disturbed Nevada desert conditions".

In addition, an "attenuation factor", λ_A , was calculated to describe the exponential decline in air concentration with time, due to progressive reduction of the amount of contaminant available for resuspension. For the conditions previously described, λ_A corresponded to a "half-time", $T_{1/2}$, of 35 days (Langham, 1969, 1971)

$$\lambda_A = \frac{\ln 2}{T_{1/2}} = \frac{0.693}{35 \text{ days}} = 0.0198 \text{ days}^{-1} \quad (2)$$

Other values of $T_{1/2}$ have been estimated or proposed, such as $T_{1/2} \approx 40$ days, for prevailing conditions at NTS (Langham, 1971), and $T_{1/2} = 45$ days, by Kathren (1968), in his lung dose model.

Applying the "attenuation factor" concept, the time-dependent concentration of resuspended contaminant may be represented by the following expression:

$$C(t) = C(o) e^{-\lambda_A t} \quad (3)$$

where $C(t)$ = airborne contaminant concentration at time t , $\mu\text{Ci}/\text{m}^3$
 $C(o)$ = initial airborne contaminant concentration, at some
arbitrarily assigned time $t = o$, $\mu\text{Ci}/\text{m}^3$
 λ_A = attenuation factor, days^{-1}
= 0.0198 days^{-1} , disturbed NTS conditions (Langham, 1971)
= 0.0173 days^{-1} , prevailing NTS conditions (Langham, 1971)
= 0.0154 days^{-1} , proposed by Kathren (1968)
 t = time, days

From the definition of resuspension factor, the air concentration $C(t)$ at a given time, t may be related to the existant surface concentration of contaminant, $S_a(t)$, by the following version of Equation (1).

$$C(t) = R_F S_a(t) \quad (4)$$

where R_F = resuspension factor, constant at a given location, for
prevailing conditions, m^{-1}
 $S_a(t)$ = surface concentration of contaminant at time t , $\mu\text{Ci}/\text{m}^2$

For some initial time $t = 0$, Equation (4) becomes

$$C(o) = R_F S_a(o) \quad (5)$$

where $S_a(o)$ = initial surface concentration of contaminant, at time
 $t = o$, $\mu\text{Ci}/\text{m}^2$

Combining Equations (3) and (5), the change with time in the airborne pollutant concentration can be seen to be proportional to an exponentially decreasing surface concentration.

$$C(t) = C(o) e^{-\lambda_A t} = R_F S_a(o) e^{-\lambda_A t} \quad (6)$$

This expression serves to emphasize an important assumption, implicit in the concept of a "constant resuspension factor", as expressed by Equations (1) and (4). That is, the amount of contaminant found in resuspension above some

given surface will represent a constant fraction of the amount of contaminant available on that surface. Consequently, all other factors remaining equal, any reduction in the concentration of airborne pollutant must be merely due to a corresponding reduction in the concentration of contaminant present on the surface. From Equations (4) and (6), this progressive reduction in the surface concentration of pollutant must clearly be

$$S_a(t) = S_a(0) e^{-\lambda_A t} \quad (7)$$

In reality, such behavior of a surface contaminant would seldom, if ever, result in maintaining the relationship described by Equation (4), namely that a time-invariant fraction of pollutant on a surface is resuspended. Such assumed relationship takes into account only one of the processes whereby a pollutant becomes unavailable for resuspension, which are commonly grouped under the general term "weathering" (Anspaugh, 1975). These include not only the transport of small contaminant-bearing particles downward into the soil by "percolation", but also the "cementing" of such particles into or onto larger ones, on the surface, by the forces of adhesion or cohesion. Whereas the first of the processes mentioned would undoubtedly result in a decrease in surface contamination, it is equally clear that the second would not. Consequently, the surface concentration of contaminant $S_a(t)$ must consist both of "uncemented" and "cemented" particles, the latter being unavailable for resuspension.

To justify the continued use of Equation (6), it would be necessary to postulate that the "uncemented" particles would constitute a constant fraction "k" (less than 1.0) of the "unpercolated" particles remaining on the surface. This constant fraction could then be incorporated, conceptually, into the empirically determined constant resuspension factor, and the relationship indicated by Equation (6) maintained.

Such a postulate, unfortunately, would imply that a constant fraction "1 - k" of the "unpercolated" particles on the surface must consist of "cemented" particles. For this fraction to remain, indeed, constant, the surface concentration of "cemented" particles would have to decrease in

proportion to the reduction in "uncemented" particles surface concentration. However, this entails a contradiction since, by definition of the processes involved, a "cemented" particle cannot "percolate" into the ground.

The inconsistency described in the preceding discussion can be avoided by defining a "time-dependent fraction ' $k(t)$ ', available for resuspension, of the surface concentration of pollutant $S_a(t)$ ". As direct consequence of such definition, the functions $C(t)$ and $S_a(t)$ would no longer parallel each other, as expressed by Equations (3) and (7); the behavior of $C(t)$ would depend on the product of two time-dependent functions, $k(t)$ and $S_a(t)$. Furthermore, resuspension factors determined empirically as ratios of airborne to surface concentrations would include, implicitly, the time-dependent fraction $k(t)$. For most practical purposes, this would mean that the general relationship expressed by Equation (4) is inaccurate, and should be replaced by a formulation describing the decaying airborne concentration $C(t)$ as the product of a time-dependent resuspension factor and a time-dependent surface concentration.

The added complexity of such formulation justifies seeking simpler expressions, one of which may be introduced by considering an alternative to the use of "surface concentration" as the contaminant source-term. Note that the extent to which a given "surface" has been contaminated is determined by means of samples taken to some depth, which varies according to technique (Bernhardt, 1976). However, regardless of technique, such sampling is limited to the top layer of soil, containing that portion of the total contamination which is presumed to be, at least in part, available for resuspension. Thus, "surface concentration" of contaminant should be differentiated from TOTAL activity or contamination present in the soil - regardless of depth - per unit surface area, since a fraction of the latter may be present at depths greater than that of the easily erodible surface layer.

The results of such differentiation should be examined in the framework of conditions best suited to the application of the resuspension factor concept, i.e. uniform deposition of pollutant over a large - ideally "infinite" - area. Under such conditions, the net effects of redistribution and losses of

contaminant material by saltation, creep and resuspension would be negligible. Ignoring, for purposes of this discussion, the effects of radioactive decay, the only remaining factor of importance is that of "percolation" of contaminant into the soil. As already discussed, this would have significant effect on the "surface concentration" $S_a(t)$. However, by definition, penetration of pollutant to greater depths into the soil would not affect TOTAL CONTAMINATION PER UNIT AREA S_A . Thus, once deposition of contaminant has concluded, S_A may be assumed to remain constant.

This suggests an alternative definition for " $k(t)$ ", as that "time-dependent" fraction, available for resuspension, of the total contamination per unit area S_A . The obvious advantage of determining such a new "resuspendible fraction" is that it would permit expressing the decay with time of the airborne pollutant concentration as the product of a constant resuspension factor and a constant "total contamination per unit area" times a time-dependent resuspendible fraction of the latter.

Anspaugh et al. (1974, 1975) were of the opinion that such time-dependent fraction could not be realistically determined, and that it would be more advantageous to define a time-dependent resuspension factor $R_f(t)$ assuming a constant value for the soil concentration, equal to the total deposition per unit area S_A , regardless of distribution with depth. Expressing (4) in this format, it becomes

$$C(t) = R_f(t) S_A \quad (8)$$

$$\text{where } R_f(t) = R_f(0) e^{-\lambda_A t} \quad (9)$$

with $C(t)$ = air concentration at time t , $\mu\text{Ci}/\text{m}^3$
 $R_f(t)$ = time-dependent resuspension factor, m^{-1}
 $R_f(0)$ = initial resuspension factor, at time $t = 0$
(deposition time), m^{-1}
 λ_A = attenuation constant, days^{-1}
 S_A = total soil activity per unit area, constant, $\mu\text{Ci}/\text{m}^2$
 t = time, days

Using (8) and (9) with values of λ_A corresponding to half-times of 35 days (Langham, 1971) or 45 days (Kathren, 1968), relationships observed for up to several weeks after a contaminating event may be approximated reasonably well. However, Anspaugh et al. (1975) quoted a number of observations indicating that such models are less accurate for longer periods of time. These include a half-time of 10 weeks determined by Anspaugh et al. (1973) from observations made 12-40 weeks after accidental venting of an underground explosion, a half-time of about 9 months obtained by Sehmel and Orgill, (1974) at Rocky Flats, and a resuspension factor of 10^{-9} m^{-1} at a location contaminated 17 years previously. The significance of this last observation can be best demonstrated by rewriting (9) as follows:

$$R_f(0) = R_f(t) e^{\lambda_A t} \quad (10)$$

where $R_f(t) = 10^{-9} \text{ m}^{-1}$

with $t = 17 \text{ years} \times \frac{365 \text{ days}}{\text{year}} = 6205 \text{ days}$

and $\lambda_A = \frac{0.693}{T_{1/2}}$

Solving (10) for $R_f(0)$ with 35 days $< T_{1/2} < 45$ days results in values of $3.16 \times 10^{32} \text{ m}^{-1} \leq R_f(0) \leq 2.275 \times 10^{44} \text{ m}^{-1}$, which are clearly impossible, since it is highly unlikely that $R_f(0)$ should exceed 1.0 m^{-1} , with 10^{-2} m^{-1} being the largest value reported to date (Mishima, 1964).

The obvious implication of the preceding discussion is that half-time $T_{1/2}$ must increase with time, and be considerably greater than 35 or 45 days at 17 years post deposition. The direct observations of Anspaugh and Sehmel and Orgill mentioned above, also support this conclusion. Based on these observations, and on the assumption that $T_{1/2} \approx 35$ days is valid during the first 10 weeks after deposition, Anspaugh et al. (1974) developed a "time-dependent" model of the resuspension factor conforming to the following constraints: "1) The apparent half-time of decrease during the first 10 weeks should approximate a value of 5 weeks and should approximately double over

the next 30 weeks; 2) The initial resuspension factor should be 10^{-4} m^{-1} ; and 3) The resuspension factor 17 years after the contaminating event should approximate 10^{-9} m^{-1} ." This model can be represented by (11).

$$R_f(t) = R_f(0) e^{-K \sqrt{t}} + R_f(\infty) \quad (11)$$

where $R_f(t)$ = time-dependent resuspension factor, at time t , m^{-1}
 $R_f(0)$ = initial resuspension factor, at time $t = 0$, m^{-1}
 $= 10^{-4} \text{ m}^{-1}$, as specified by Anspaugh et al. (1974)
 $R_f(\infty)$ = final resuspension factor, at time " $t = \infty$ ", m^{-1}
 $= 10^{-9} \text{ m}^{-1}$, as specified by Anspaugh et al. (1974)
 t = time from deposition, days
 K = constant coefficient = $0.15 (\text{days})^{-1/2}$

The "final resuspension factor" $R_f(\infty) = 10^{-9} \text{ m}^{-1}$, in the above expression, reflects the expectation that there would be no further measurable decrease in the resuspension process after 17 years, which, in 1974, was "the longest period post deposition for which measurements (had) been reported". At such time, the mechanical behavior of the aged pollutant deposit would not differ, presumably, from that of the native soil itself, by virtue of the two having become intimately associated.

As Anspaugh pointed out, this model "was derived from a composite of numerous experiments," and "contains no fundamental understanding of the resuspension process," but intends merely to describe it. Nevertheless, two basic assumptions are implicit in the model formulation, as follows:

1) The half time $T_{1/2}$ is time-dependent,

2) The resuspension factor $R_f(t)$ reaches a limiting value, $R_f(\infty)$, at a long ("infinite") time after deposition ($t = \infty$).

Should these two assumptions be accepted as valid, some generality may be attached to Anspaugh's model as expressed by (11), derived as it was from "numerous experiments", and thus, at many locations; "there have not been measurements at any individual source over such long time-periods" (Anspaugh et al. 1974, 1975).

TIME-DEPENDENT HALF-TIME MODEL BASED ON ANSPAUGH'S RESUSPENSION FACTOR

There is evident need to model "weathering" processes on a fundamental basis, particularly, as regards resuspension, in view of the conceptual connection between "weathering" and the time-dependent behavior of the half-time, $T_{1/2}(t)$. The present author makes no pretense of providing such a solid foundation, but merely attempts to produce a general empirical model of half-time as function of time and local conditions, represented by "initial" and "final" resuspension factors, $R_f(0)$ and $R_f(\infty)$, respectively. This model is based on observations and models reported by previous authors, with liberal use of the assumptions inherent in their development.

An obvious starting point for a tentative model is provided by Equations (9) and (11). Incorporating into the former the postulated time-dependence of the half-time and hence that of the attenuation factor $\lambda(t)$, previously denoted as λ_A , it may be formally rewritten as

$$R_f(t) = R_f(0) e^{-\lambda(t) t} = R_f(0) e^{-\frac{0.693}{T_{1/2}(t)} t} \quad (12)$$

Equation (12) may be interpreted as being mainly a definition of $T_{1/2}(t)$. With some rearrangement, it becomes

$$T_{1/2}(t) = -\frac{0.693 t}{\ln \frac{R_f(t)}{R_f(0)}} \quad (13)$$

Replacing $R_f(t)$ in the above equation with Anspaugh's model as expressed by (11) results in equation (14)

$$T_{1/2} \left(t, \frac{R_f(0)}{R_f(\infty)} \right) = \frac{-0.693 t}{\ln \left[e^{-K\sqrt{t}} + \frac{R_f(\infty)}{R_f(0)} \right]} \quad (14)$$

Note that, in a strict sense, $T_{1/2}$ is no longer a function solely of time but also depends on local conditions, parameterized by initial and final resuspension factors as obtained or extrapolated from actual measurements in a given area. While Anspaugh and his coworkers assigned specifically values of 10^{-4} m^{-1} and 10^{-9} m^{-1} , respectively, to these factors, both higher and lower values have been observed. Table 1 (Mishima, 1964) presents resuspension factors ranging over 11 orders of magnitude, from 10^{-2} m^{-1} (interiors) and 10^{-3} m^{-1} (disturbed exterior conditions) to 10^{-13} m^{-1} (aged deposit). Although some of these values would be normally quite inappropriate in a study concerning specifically wind resuspension, their use may be allowed for the purposes of developing a general model. In particular, the ratio of the lowest to the highest values in Table 1 can be assumed to represent a credible lower limit of the ratio $R_f(\infty)/R_f(0)$ in Equation (14). A tentative upper limit for this ratio may then be provided via the assumption that $R_f(0)$ and $R_f(\infty)$ must differ by at least one order of magnitude. Consequently, the value of $R_f(\infty)/R_f(0)$ in Equation (14), applied generally, should vary over 10 orders of magnitude, provided constraints are met.

$$10^{-11} \leq \frac{R_f(\infty)}{R_f(0)} \leq 10^{-1}, \quad (15)$$

where $R_f(0) \geq 10 R_f(\infty)$ (assumption)

Using Anspaugh's value for "K" of $0.15 \text{ day}^{-1/2}$, the constraints imposed on $T_{1/2}$ (a value of 35 days during the first 10 weeks, double the value over the next 30 weeks) are clearly met, regardless of the value of $R_f(\infty)/R_f(0)$, as seen in Figure 1. In fact, any ratio $R_f(\infty)/R_f(0) \leq 10^{-1}$ should satisfy this requirement.

Nevertheless, Equation (14) has one serious drawback, which becomes apparent upon examination of the denominator in the right-hand side of this equation. For certain specific values of time t , this denominator will approach zero and consequently the half-time will tend to $+$ or $-$ infinity (∞) along a vertical asymptote (Figure 1).

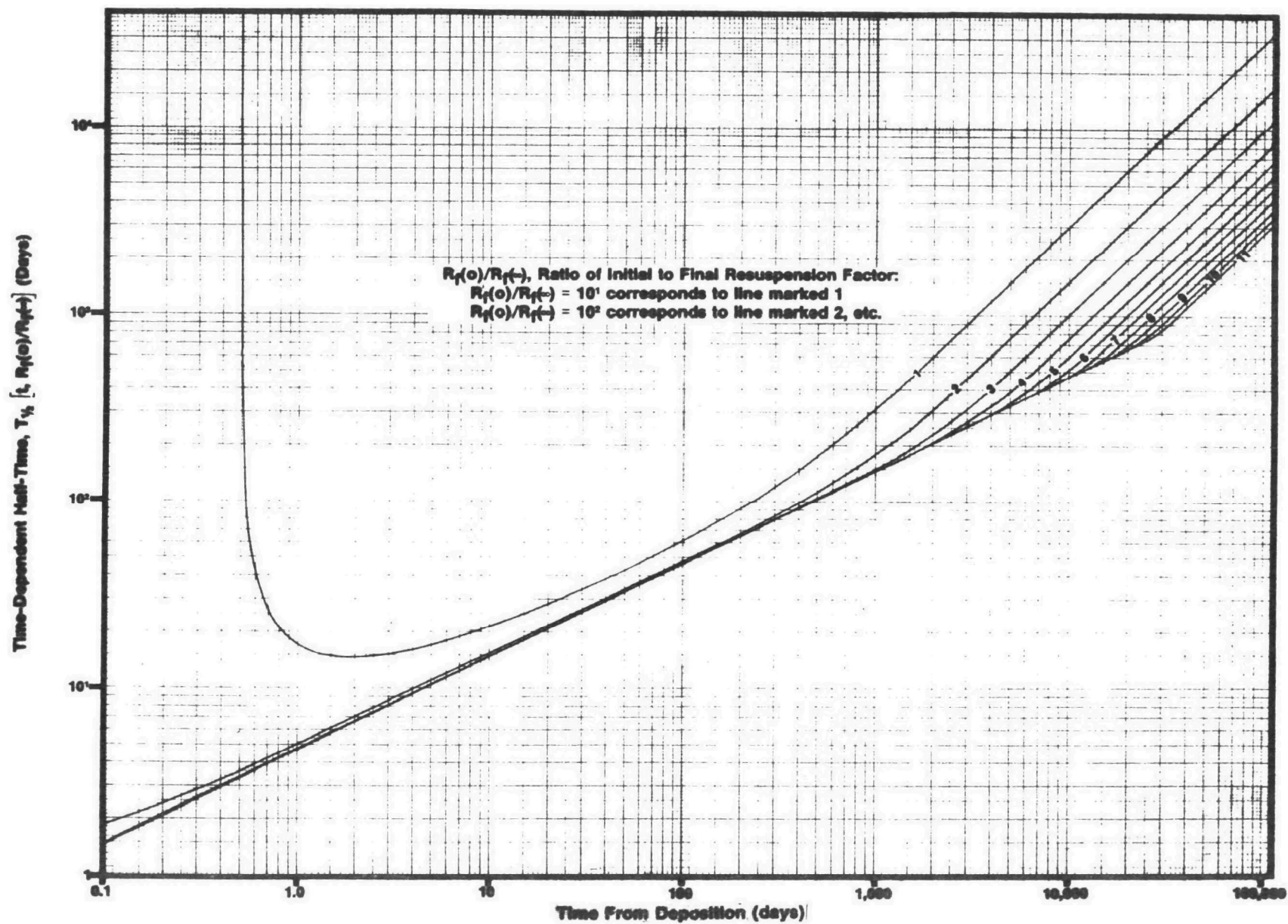


FIGURE 1. Half-time $T_{1/2}$ as function of time and local conditions (expressed as ratios of initial-to-final resuspension factors, $R_f(0)/R_f(\infty)$), as derived (Equation (14)) from Anspaugh's model of $R_f(t)$.

$$T_{1/2} \rightarrow +\infty \quad \text{as } t \rightarrow t_{as}^+ \quad (16)$$

$$T_{1/2} \rightarrow -\infty \quad \text{as } t \rightarrow t_{as}^- \quad (17)$$

$$\text{where } t_{as} = \frac{1}{K} \ln 1 - \frac{R_f(\infty)}{R_f(0)}^2$$

For $t < t_{as}$, $T_{1/2}$ will be negative. For $t > t_{as}$, $T_{1/2}$ will have a minimum at some specific time t_{min} , which implies that $T_{1/2}$ will decrease with time for values exceeding t_{as} but less than t_{min} , and increase thereafter. Thus, every positive value of $T_{1/2}$ will occur twice, with exception of $T_{1/2}(t_{min})$. These observations are summarized below.

$$\frac{d T_{1/2}}{dt} < 0 \quad \text{for } t < t_{min} \quad (18)$$

$$\frac{d T_{1/2}}{dt} > 0 \quad \text{for } t > t_{min} \quad (19)$$

$$0 > T_{1/2} > -\infty \quad \text{for } 0 > t > t_{as} \quad (20)$$

$$+\infty > T_{1/2} \geq T_{1/2}(t_{min}) \quad \text{for } t_{as} < t \leq t_{min} \quad (21)$$

$$T_{1/2}(t_{min}) \leq T_{1/2} < +\infty \quad \text{for } t_{min} \leq t < +\infty \quad (22)$$

Equations (16) through (22) illustrate the deficiencies of Equation (14), formally described as "the time-dependent half-time model derived from Anspaugh's model of the resuspension factor". These may be itemized as follows:

- 1) The discontinuity at $t = t_{as}$, described by Equations (16) and (17), has no physical parallel.
- 2) The decrease with time of $T_{1/2}$ indicated by Equation (18) is contrary to empirical evidence.
- 3) The minimum value of $T_{1/2}$ at t_{min} , implied by Equations (18), (19), (21), and (22), contradicts the expectation of there being only one (absolute) minimum $T_{1/2}$ at $t = 0$.

- 4) The negative values of $T_{\frac{1}{2}}$ given by Equation (20) are clearly impossible.
- 5) The behavior of $T_{\frac{1}{2}}$ expressed by Equation (21), beginning with a value "close to infinity", at some short time after deposition, and decreasing thereafter, is highly unlikely.

In summa, only Equations (19) and (21) conform to physical reality, thus limiting the applicability of the model to times t greater than t_{\min} . Note that the value of t_{\min} depends both on the ratio $R_f(\infty)/R_f(0)$ and the constant coefficient "K". For a value $K = 0.15 / \sqrt{\text{days}}$, the model would be generally satisfactory with $t_{\min} \lesssim 2$ days, for all ratios $R_f(\infty)/R_f(0) \leq 10^{-1}$. However, smaller values of "K" would result in higher values of t_{\min} , and a further decrease in the domain of applicability of the model. Such limitations suggest the need for alternative formulations of a general model. One such expression is developed in the next section.

ERRATUM: The expression describing t_{as} , following Equation (17) on page 15, has been printed Incompletely. The correct expression is as follows

$$t_{as} = \left\{ \frac{1}{a} \ln \left[1 - \frac{R_f(\infty)}{R_f(0)} \right] \right\}^2$$

BASES OF PROPOSED INTERIM MODEL OF TIME-DEPENDENT HALF-TIME

Two basic assumptions are implicit in Anspaugh's formulation of the time-dependent resuspension factor (11). They are: 1) that the half-time $T_{1/2}$ is time-dependent, $T_{1/2}(t)$; and 2) that the resuspension factor $R_f(t)$ approaches a limiting value $R_f(\infty)$, applicable to "aged" deposits such that " $t \rightarrow \infty$ ". The present author proposes to formulate an interim generalized model of $T_{1/2}$ as a function of time and local conditions (as reflected by $R_f(0)/R_f(\infty)$) by first reviewing the facts at his disposal in the light of these two assumptions.

Initial Values of $T_{1/2}(t, R_f(0)/R_f(\infty))$. Measurements of half-time $T_{1/2}$ at several locations and at various times after a contaminating event suggest that the half-time increases with time post deposition (Anspaugh et al. 1973, 1974, 1975). Since conceptually "weathering" is the only phenomenon (or group of phenomena) affecting the resuspension factor, this time-dependent behavior of the half-time must reflect a dependence on weathering as well. Furthermore, since both weathering and half-time increase with time after deposition, an initial time following a contaminating event, when the extent of weathering is small, the half-time is correspondingly short. For the case of "just deposited pollutant", at time $t = 0$, when the process of weathering begins, the half-time could be extremely small; it would be finite, however, since forces of adhesion between the contaminant and the native soil become effective upon contact. Thus,

$$T_{1/2}(0) > 0 \quad (23)$$

Values of $T_{1/2}(t)$ measured within days to several months after deposition appear to be roughly similar, in spite of the different local conditions under which these measurements were made (Anspaugh et al., 1973, 1974; Wilson et al. 1960). However, as the related resuspension factors approach final values reflecting these and other conditions, the half-times at the corresponding locales may differ from each other by one order of magnitude, as will be seen in the discussion of "Final Values of $T_{1/2}$ ". This would indicate a convergence of half-time values as time approaches $t = 0$. Lacking other data, one single initial half-time at $t = 0$, valid at all locations under all average conditions (excluding severe disturbances) may be postulated.

$$T_{1/2} \left\{ 0, \left[R_f(0)/R_f(\infty) \right]_i \right\} \approx T_{1/2}(0) \text{ for all } i = 1, 2, 3, \dots \quad (24)$$

where $\left[R_f(0)/R_f(\infty) \right]_i$ = conditions at location i

i = generalized location index

$T_{1/2}(0)$ = initial half-time, independent of location

Values of $T_{1/2}(t, R_f(0)/R_f(\infty))$ Within One Year After Deposition. Several values of half-time have been reported. Anspaugh et al. (1973) determined a half-time $T_{1/2} = 38$ days from a least squares fit to averaged measurements made from 3 days to roughly 8 weeks after a nuclear cratering event, Project Schooner (Figure 2).

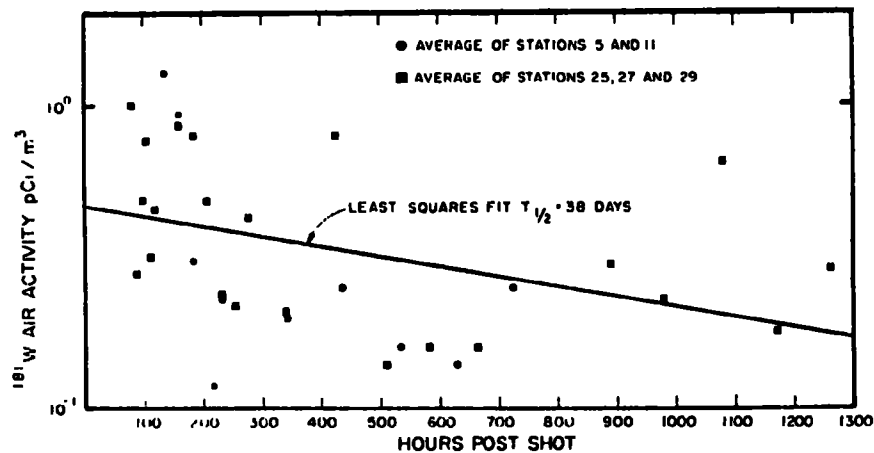


FIGURE 2. Tungsten-181 air activity as a function of time at five downwind stations within six miles that were closest to the line of maximum deposition from Project Schooner. Data were normalized to the first sample taken after 70 hours had elapsed following the detonation [From Anspaugh et al. (1973)].

Wilson et al. (1960) obtained a half-time value of 35 days as a "best apparent fit" to median data obtained between approximately 3 and 23 weeks after a non-critical high explosive detonation involving plutonium, Operation Plumbbob (Figure 3). Olafson and Larson (1961) reported the measurement period as beginning 18 days after the event and continuing to 160 days. Stewart (1967) described the decay in the average airborne concentration as having a half-life of about 37 days. Following other experiments, Langham (1971) reported "attenuation factors" of 35 and 40 days estimated at the Nevada Test Site, although the time periods to which these are strictly applicable are uncertain.

Interpreting this data somewhat loosely, it would appear that a half-time of 35 to 38 days, applicable to various conditions, should be expected between 3 and 160 days after deposition.

$$35 \text{ days} \leq T_{\frac{1}{2}}(t_1) \leq 38 \text{ days} \quad (25)$$

where $3 \text{ days} \leq t_1 \leq 160 \text{ days}$, time after deposition

and $T_{\frac{1}{2}}(t_1)$ = empirically determined half-time, days, presumed to apply to a contaminant deposit present in the soil for a time t_1

t_1 = time post deposition, days

In addition, Anspaugh et al. (1973) calculated a half-time of 76 days over a period extending from roughly 11 to 45 weeks after the accidental venting of an underground nuclear explosion (Baneberry venting, Figure 4). When corrected for background variation, the half-time was found to be 66 days.

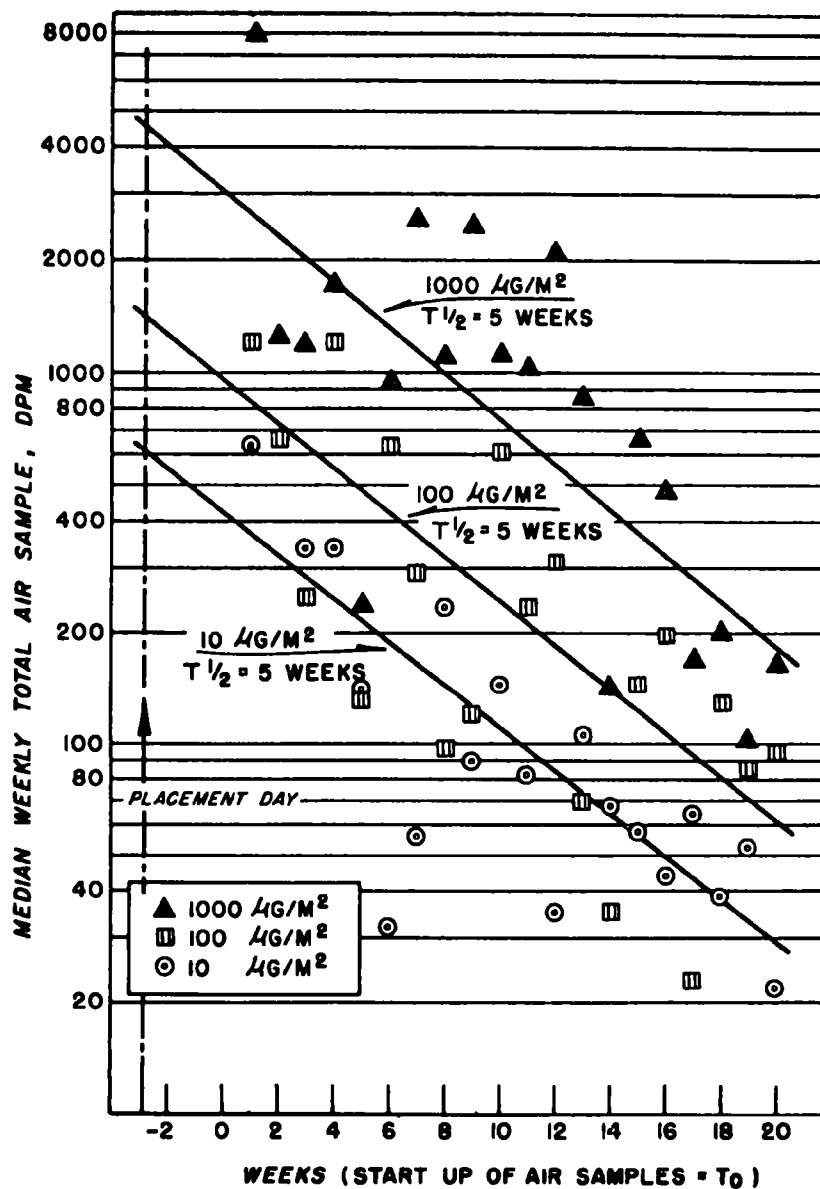


FIGURE 3. Median weekly air concentrations, corresponding to three isopleths, from Project Plumbbob [From Wilson et al. (1960)].

$$T_{\frac{1}{2}}(t_2) = 66 \text{ days} \quad (26)$$

where $77 \text{ days} \leq t_2 \leq 315 \text{ days}$, time after deposition

and $T_{\frac{1}{2}}(t_2)$ = empirically determined half-time, days, presumed to apply to a contaminant deposit present in the soil for a time t_2

t_2 = time post deposition, days

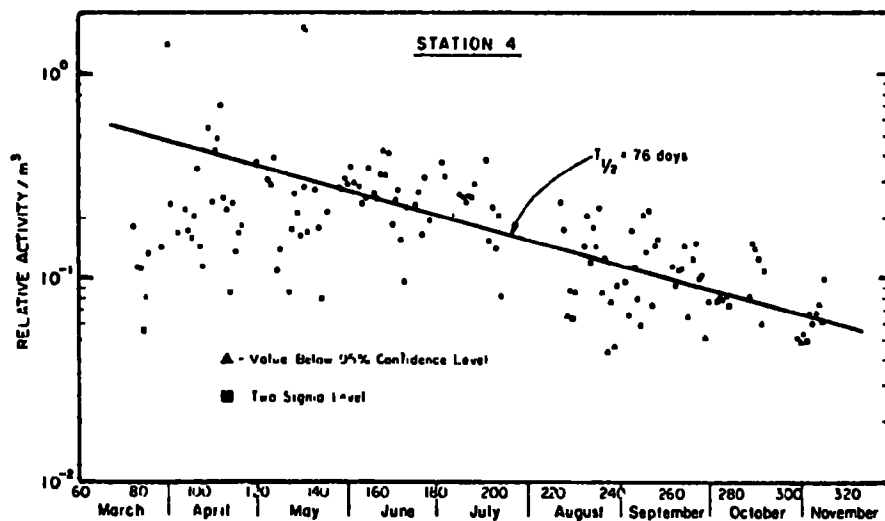


FIGURE 4. Least-squares fit to gross-gamma air activity levels 3 to 11 months following Baneberry venting [From Anspaugh et al. (1973)].

For the purposes of the interim model, the values expressed in (25) and (26) will be considered to apply generally. Additional data, when available, will serve to corroborate, extend or even reduce the time periods over which these half-times are expected to apply.

Values of $T_{1/2}$ (t , $R_f(0)/R_f(\infty)$) After One Year From Deposition. Sehmel and Orgill (1974) reported an "average weathering half-life...from April 1971 to October 1972... of about 9 months" from measurements made at sampling station S-8, Rocky Flats, near an area contaminated with plutonium from leaking drums in previous years. The age of the deposit, however, is uncertain. It can only be determined to be within a very broad range of time values, as the following review of the pertinent facts should indicate:

1) Drums containing cutting oil contaminated with plutonium "were placed in outside storage from 1958 until 1968. Initial leakage was detected in 1964" (Krey and Hardy, 1970). Apparently the drums contaminated the adjacent soil for a period of four years, from 1964 to 1968. However, the possibility that leaks had developed earlier and gone undetected should not be excluded.

2) A contour representation of plutonium-239 distribution at Rocky Flats shows a "hot spot as defined by the contours, just adjacent to the area where the leaking drums had been stored" (Volchok, 1971). Thus, there is little doubt that the plutonium concentrations measured by Sehmel and Orgill "near the original oil storage area" have the leaking drums as the original, though not necessarily immediate, source (Figure 5).

3) As implied by 2) and Figure 5, the leaking plutonium was subsequently dispersed. According to Martell (1970), it was "redistributed by winds, mainly in the period between spring 1967, when Dow started to move the drums for reprocessing, and September 1969, when a four-inch thick asphalt slab was placed over the contaminated area." Therefore, the 1971-1972 observations of Sehmel and Orgill cannot be related directly to the original contaminated area in the immediate vicinity of the drums, since this source was no longer in existence as of 1969. By elimination, the immediate source of the plutonium concentrations measured by these researchers was that contaminant redistributed by wind in the preceding years. Martell states that most of this redistribution took place in 1967-1969, 19 months to 4 years prior to the first measurement of Sehmel and Orgill. However, as mentioned in 1), leakage and possibly redistribution started much earlier, at least as early as 1964.

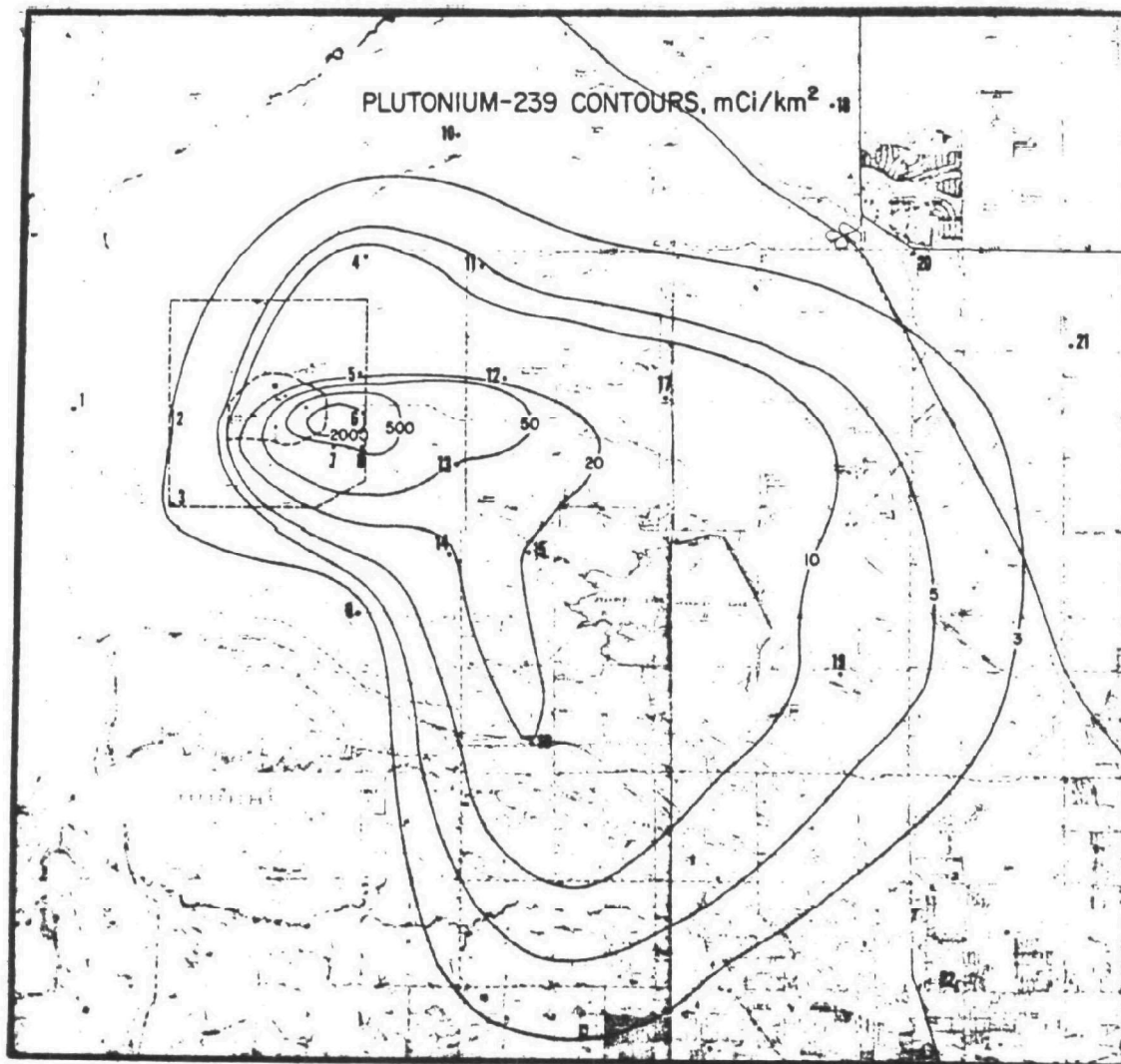


FIGURE 5. Contour representation of plutonium - 239 distribution at Rocky Flats. [From Volchok (1971)]

The difficulty of assigning a proper "weathering age" to material redistributed continuously from a source, no longer in existence, which had been subject to continuous deposition of pollutant over an uncertain period of years should be obvious. The task increases in complexity considering that, in all probability, deposition did not proceed uniformly with time, and that redistribution most certainly did not. However, one additional fact further complicates the issue, as described below.

4) Sehmel and Orgill began their data collection in April 1971. However, "In mid-March 1971, a ditch was dug east of the original oil storage area and west of sampling station S-8" (in the area of heaviest contamination) following which they observed "increased airborne activity" (Figure 6), due to the fact that "ditch digging significantly increased the availability of plutonium for resuspension" (Sehmel and Orgill, 1974). The resulting "resuspension source change" may be equated to a "source freshening" (present author's expression), suggesting that the measured half-life of nine months could, in fact, correspond to a much fresher source than what the as yet undetermined chronological age of the deposit would indicate.

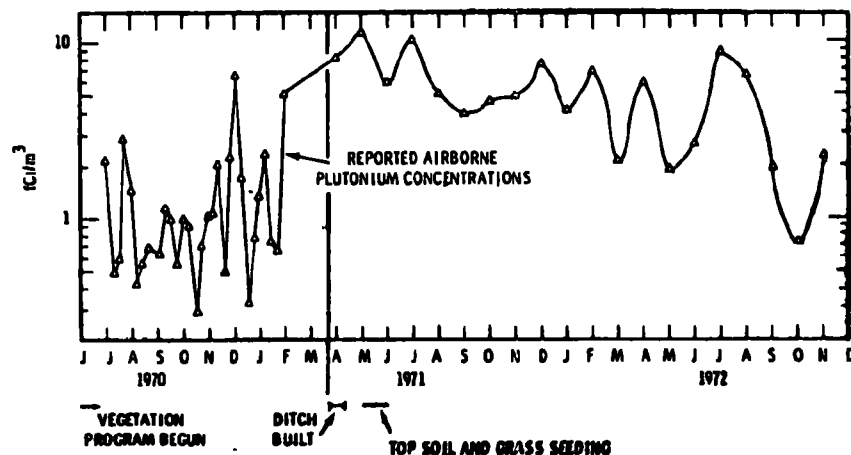


FIGURE 6. Airborne Plutonium at Sampling Station S-8. Adapted from Sehmel and Orgill (1974).

In conclusion, very limited use may be made of the 9-months half-time subject of this discussion. To include all possibilities (even the most pessimistic ones), the age of the deposit corresponding to this half-life must be tentatively assumed to be between 19 months or less and an admittedly extreme 14 years or more from deposition.

$$T_{1/2}(t_3) \approx 270 \text{ days} \quad (27)$$

where $19 \text{ months (or less)} \leq t_3 \leq 14 \text{ years (or more)}$

and $T_{1/2}(t_3)$ = empirically determined half-time, days,
presumed to apply to a contaminant deposit present in the soil for a time t_3

t_3 = time post deposition

Such a wide range of weathering ages does not permit using (27) as a constraint for the interim model. For modeling purposes, the observations of Sehmel and Orgill may be construed as furnishing proof that half-lives of 9-months do, indeed, exist, and are assumed to apply generally (given sufficient time) under all conditions $R_f(0)/R_f(\infty)$.

"Final" Values of $T_{1/2}(t, R_f(0)/R_f(\infty))$. Postulating that the function $R_f(t) = R_f(0) \exp \{-[0.693/T_{1/2}(t)] t\}$ approaches a limiting value $R_f(\infty)$ at "sufficiently large" values of time t is equivalent to assuming that, as t "approaches infinity", the time-dependent weathering half-life $T_{1/2}$ also approaches infinity, but along an oblique asymptote defined by Equation (28).

$$T_{1/2} [t, R_f(0)/R_f(\infty)] = \frac{0.693 t}{\ln \left[\frac{R_f(0)}{R_f(\infty)} \right]} \quad (28)$$

In a strict sense, this implies that $T_{1/2}$ is not solely a function of time, but depends also on average local conditions, parameterized by the ratio of initial and final resuspension factors, $R_f(0)/R_f(\infty)$. Referring to Table 1, it

may be assumed that $R_f(0)$ can be as high as 10^{-2} m^{-1*} , and that values as low as 10^{-13} m^{-1} are possible for $R_f(\infty)$. Furthermore, regardless of the actual values of $R_f(0)$ and $R_f(\infty)$, it would not seem unreasonable to assume that, for a specific locality, the final resuspension factor should be at least one order of magnitude smaller than the initial resuspension factor - $R_f(\infty) \leq 10^{-1} R_f(0)$. Thus, the presently applicable range of values $R_f(0)/R_f(\infty)$ can be adequately expressed by (29).

$$10^1 \leq \frac{R_f(0)}{R_f(\infty)} \leq 10^{11} \quad (29)$$

Applying this expression to (28) produces a range of values of $T_{1/2}$ for long times after deposition.

$$0.02736 \text{ } t \leq \lim_{t \rightarrow \infty} T_{1/2} \leq 0.30097 \text{ } t \quad (30)$$

That is, at some given time t , "long after deposition", half-times at various localities may differ from each other by as much as one order of magnitude, or more.

* It should be emphasized that such a high resuspension factor applied to disturbed interior conditions, and may be extremely unrealistic for the case of outdoors pollutant resuspension by wind. Nevertheless, the use of such an unlikely high value may be justified in developing a general model, as intended by the present author.

PROPOSED INTERIM MODEL OF TIME-DEPENDENT HALF-TIME

A simple generalized model of the weathering half-life of the resuspension factor, as a function of time (t) and of average local conditions ($R_f(0)/R_f(\infty)$), is presented. The model is based on the facts and assumptions discussed in the previous section, using them as constraints, when applicable. It is of the general form

$$T_{1/2}(t, R_f(0)/R_f(\infty)) = A \ln(1 + B + Ct^D) + \frac{0.693 t}{\ln \frac{R_f(0)}{R_f(\infty)}} \quad (31)$$

Where A = constant coefficient, days
B = constant, dimensionless
C = constant coefficient, day^{-D}
D = constant exponent, dimensionless
 $R_f(0)$ = initial resuspension factor, m⁻¹, at a given location
 $R_f(\infty)$ = final resuspension factor, m⁻¹, at the same location
t = time, days

The second term of the expression serves to meet the requirement that $R_f(t)$ approach the expected $R_f(\infty)$ at long times after initial contamination. The first term of the equation forces $T_{1/2}(t)$ to the observed or expected values at deposition and shortly thereafter. The specific choice of constants A, C, B, and D, will obviously determine what these values are and/or when they are attained, according to the model. Referring to (23), (25), (26) for explanation of the symbols used, a group of relationships may be roughly sketched.

$$T_{1/2}(0) \approx f_0(A, B)$$

$$T_{1/2}(t_1) \approx f_1(A, C, D)$$

$$T_{1/2}(t_2) \approx f_2(A, C, D)$$

Note that the constant B has no role other than determining $T_{1/2}(0)$ since, as $T_{1/2}(0)$ is assumed to be small, it is expected that $B \ll 1$.

By making judicious choices of constants, a range of initial values $T_{1/2}(0)$ may be postulated while satisfying the model constraints. Two sets of such constants are presented in Table 2 as examples of applications of the general model (31).

TABLE 2. EXAMPLES OF CONSTANTS USED IN GENERAL MODEL

	Case 1	Case 2
A (days)	28	36
B (dimensionless)	4×10^{-2}	4×10^{-3}
C (days ^{-D})	1	1
D (dimensionless)	1/3	1/4

Using "Case 2" constants in (31) with a range of ratios $R_f(0)/R_f(\infty)$ from 10^1 to 10^{11} results in the family of curves shown in Figure 7. The relevant features of the model in the present application are summarized below.

$$T_{1/2}(0) \approx 3.5 \text{ hours at } t = 0 \quad (32)$$

$$T_{1/2}(1) \approx 25 \text{ days at } t = 1 \text{ day} \quad (33)$$

$$\begin{aligned} & 35 \text{ days} \leq T_{1/2}(t_1) \leq 38 \text{ days} \\ \text{Where } & 5 \text{ days} \leq t_1 \leq 12 \text{ days} \end{aligned} \quad (34)$$

$$\begin{aligned} & T_{1/2}(t_2) = 66 \text{ days} \\ \text{Where } & 60 \text{ days} \leq t_2 \leq 276 \text{ days} \end{aligned} \quad (35)$$

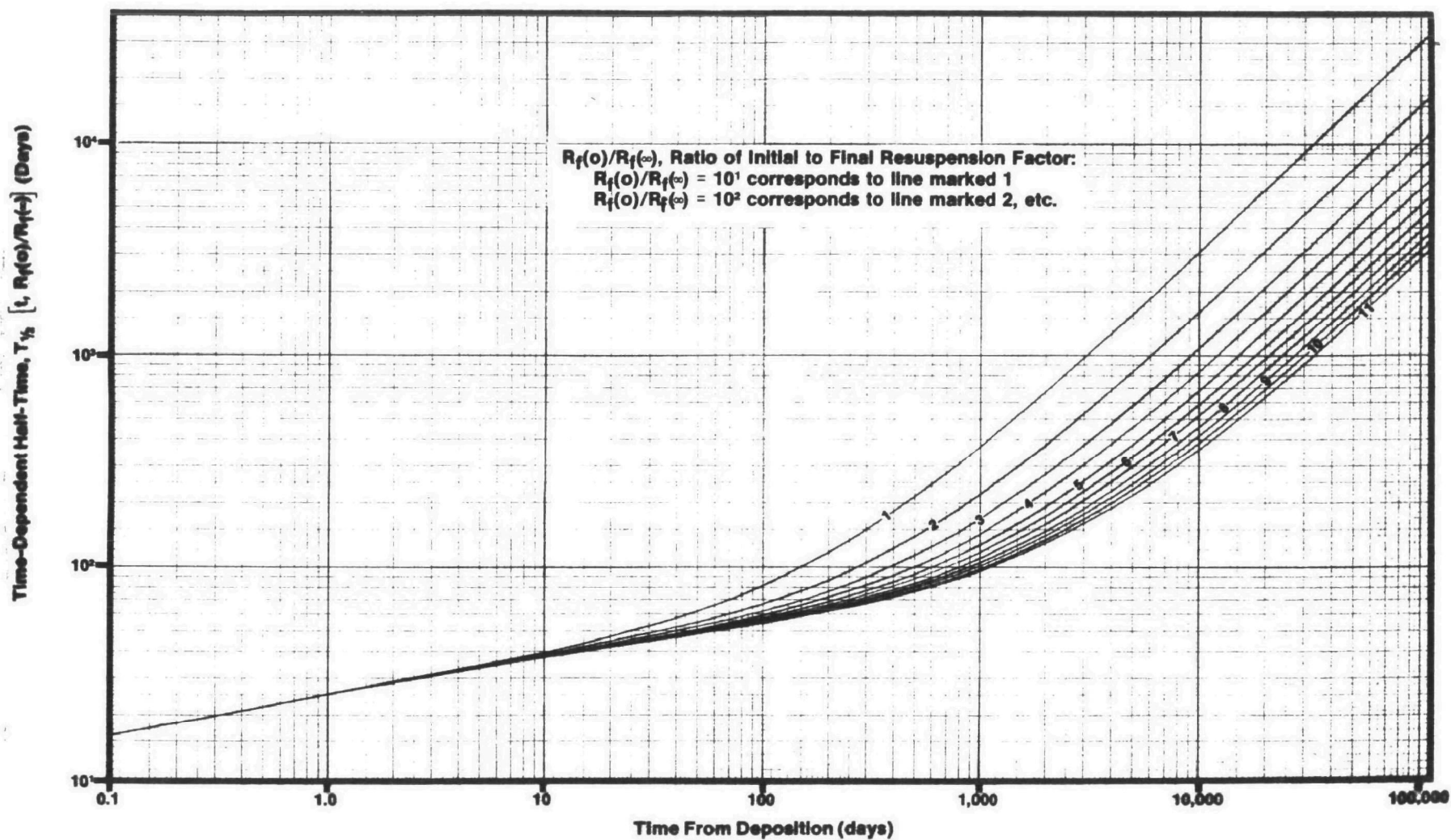


FIGURE 7. Half-time $T_{1/2}$ as function of time and local conditions (expressed as ratios of initial-to-final resuspension factors $R_f(0)/R_f(\infty)$) according to proposed model and "Case 2" constants (Table 2). Curves numbered according to ratio $R_f(0)/R_f(\infty)$, in orders of magnitude, i.e. "2" corresponds to $R_f(0)/R_f(\infty) = 10^2$ etc.

Comparing (32), (34), (35) with Equations (23), (25), and (26), it is seen that the constraints imposed by the latter three expressions are essentially met. The same applies for the conditions of "Case 1", for which the model produces the following results:

$$T_{1/2}(0) \approx 1 \text{ day at } t = 0 \quad (36)$$

$$T_{1/2}(1) \approx 20 \text{ days at } t = 1 \text{ day} \quad (37)$$

$$\begin{aligned} & 35 \text{ days} \leq T_{1/2}(t_1) \leq 21 \text{ days} \\ \text{Where } & 10 \text{ days} \leq t_1 \leq 21 \text{ days} \end{aligned} \quad (38)$$

$$\begin{aligned} & T_{1/2}(t_2) = 66 \text{ days} \\ \text{Where } & 67 \text{ days} \leq t_2 \leq 309 \text{ days} \end{aligned} \quad (39)$$

A family of curves corresponding to various ratios $R_f(0)/R_f(\infty)$ can be obtained, analogous to those of Figure 7. Figure 8 shows two of these curves, for the upper and lower limits of the range of values presumed possible for $R_f(0)/R_f(\infty)$, 10^{11} and 10^1 , respectively.

The extent to which the model conforms to the requirements set by (28) can be gaged by first examining the latter. Clearly, this equation is equivalent to

$$\lim_{t \rightarrow \infty} \frac{\partial T_{1/2}(t, R_f(0)/R_f(\infty))}{\partial t} = \frac{0.693}{\ln \frac{R_f(0)}{R_f(\infty)}} \quad (40)$$

Taking the partial derivative of the general model (31) with respect to time t , results in

$$\frac{\partial T_{1/2}(t, R_f(0)/R_f(\infty))}{\partial t} = \frac{ACDt^{D-1}}{1+B+Ct^D} + \frac{0.693}{\ln \frac{R_f(0)}{R_f(\infty)}} \quad (41)$$

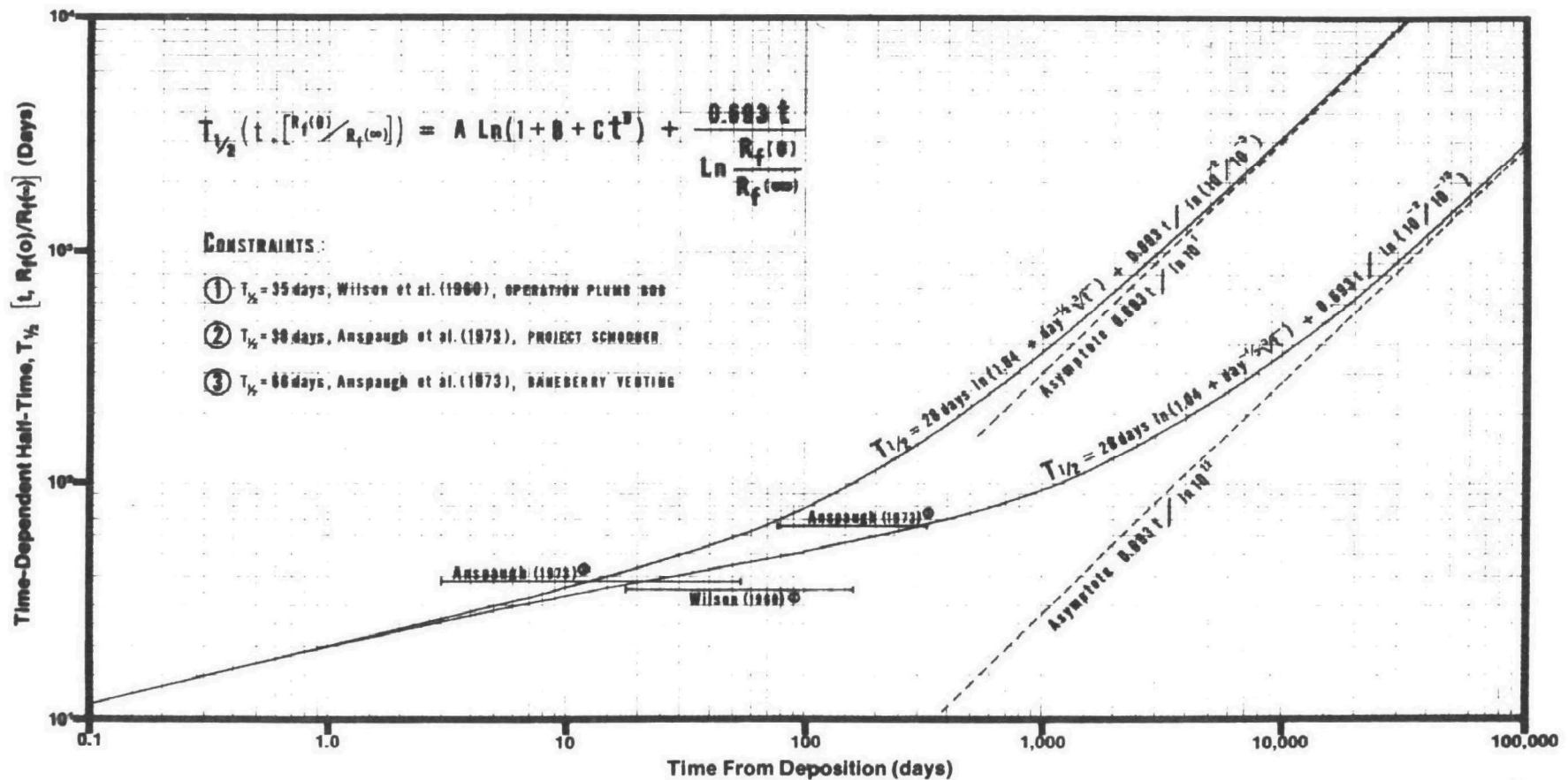


FIGURE 8. Half-time $T_{1/2}$ as function of time and local conditions (expressed as ratios of initial-to-final resuspension factors $R(0)/R_f(\infty)$) according to proposed model (Equation (33)) and "Case 1" constants (Table 2). Curves shown correspond to upper and lower limits of range of values presumed possible for $R_f(0)/R_f(\infty)$.

As t increases, the first term in (41) grows progressively smaller, vanishing entirely for values of t "approaching infinity".

$$\lim_{t \rightarrow \infty} \frac{\partial T_{1/2}}{\partial t} \quad (41) \quad = \quad \frac{0.693}{\ln \frac{R_f(0)}{R_f(\infty)}} \quad (42)$$

Clearly, the constraint expressed by (40) is met. Therefore, as time increases, the half-life $T_{1/2}$ approaches one of the oblique asymptotes indicated by (28). Two such asymptotes are shown in Figure 8.

APPLICATION

The primary application of the model is in describing time-effected changes in the magnitude of airborne contamination through the dependence of the rate of change of the resuspension factor on the modelled variable, the time-dependent weathering half-life. Figures 9 and 10 depict the behavior of the resuspension factor as a function of time for different ratios $R_f(0)/R_f(\infty)$, as predicted by Equation (12) and the present model (31) of $T_{1/2}(t, R_f(0)/R_f(\infty))$ (Case 2).

The intent in modelling the time-dependent behavior of the weathering half-life was to provide a simple predictive tool of reasonable accuracy and flexibility enabling it to meet a wide range of average local conditions. However, the present author lacks the necessary data to determine the degree to which the predictive ability and thus usefulness of the tool may have been marred by the inevitable "trade-offs" between flexibility and accuracy. For this reason, examples of the model applicability are limited to a comparison of the behavior of the half-time and resuspension factor as functions of time as predicted by the model with those predicted by or derived from other models (Figures 11 and 12). Table 3 describes the salient features of these models, specifically Langham's (1956-1971), Kathren's (1968) and Anspaugh's (1974), as well as those of the present model (Case 1), including constants used in the latter for comparison purposes.

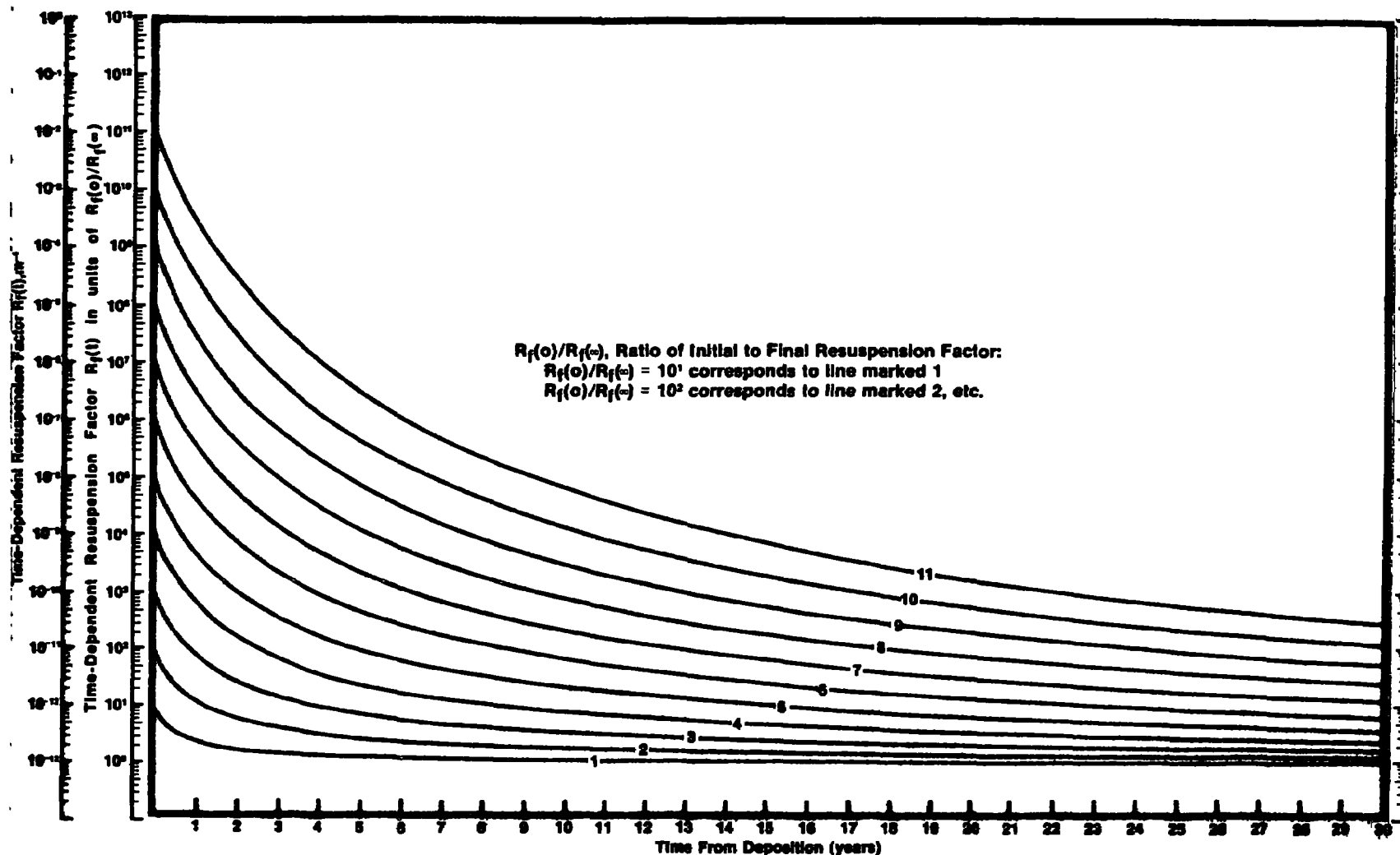


FIGURE 9. Resuspension factors as function of time based on proposed model of time-dependent half-time $T_{1/2}$ assuming various initial values but the same final resuspension factor. First captioned ordinate represents $R_f(t)$ values presumed possible based on Table 1. Some of the higher values do not apply to outdoor pollutant resuspension by wind under normal conditions, but have been included for the purpose of demonstrating applications of the general model. Second captioned ordinate pertains to generalized case.

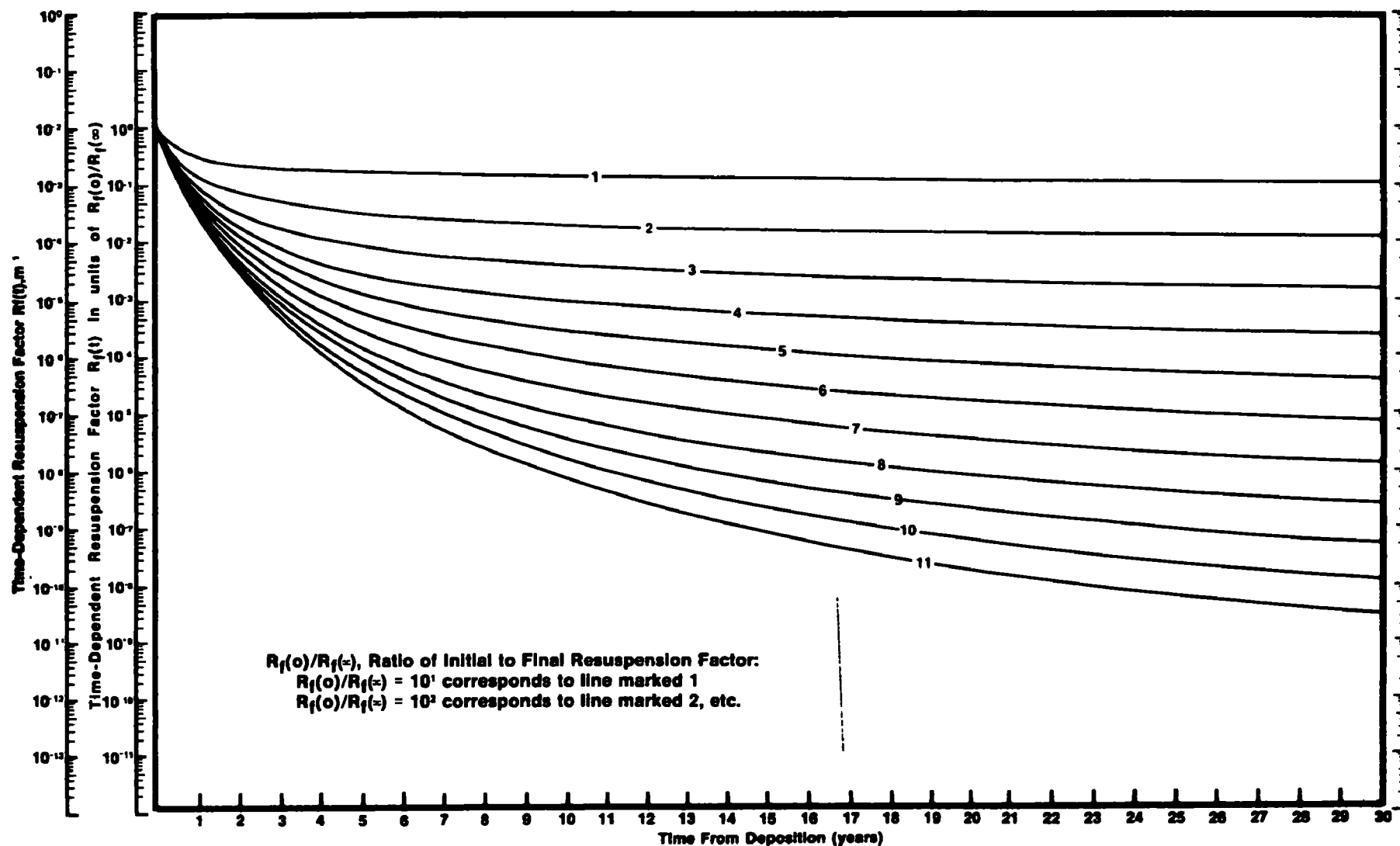


FIGURE 10. Resuspension factors as functions of time based on proposed model of time-dependent half-time $T_{1/2}$ assuming the same initial value but various final resuspension factors. First captioned ordinate shows values presumed possible based on Table 1. The initial value of 10^{-2} m^{-1} is highly improbable for outdoors pollutant resuspension by wind under normal conditions, but serves to illustrate applications of the model for the full range of values postulated in the model development. Second captioned ordinate pertains to generalized case.

TABLE 3. COMPARISON OF PROPOSED MODEL (CASE 1) OF THE TIME - DEPENDENT WEATHERING HALF - TIME $T_{1/2}$ WITH PREVIOUS MODELS, IMPLICIT OR DERIVED, AND CORRESPONDING MODELS OF THE TIME - DEPENDENT RESUSPENSION FACTOR, INCLUDING NUMERICAL VALUES

	LANGHAM'S MODEL	KATHREN'S MODEL	ANSPAUGH'S MODEL	PRESENT PROPOSED MODEL (CASE 1)
RESUSPENSION FACTOR $R_f(t)$	$R_f(o)e^{-\frac{0.693 t}{T_{1/2}}}$	$R_f(o)e^{-\frac{0.693 t}{T_{1/2}}}$	$R_f(o)e^{-\frac{0.15}{\text{days}^{1/2}} \sqrt{t}} + R_f(\infty)$	$R_f(o)e^{-\left\{ \frac{0.693 t}{28 \ln \left(1.04 + \frac{t^{1/3}}{\text{days}^{1/3}} \right) + \frac{0.693 t}{\ln \left[\frac{R_f(o)}{R_f(\infty)} \right]}} \right\}}$
HALF-TIME $T_{1/2}(t)$	35 days (constant)	45 days (constant)	$\frac{-0.693 t}{\ln \left[e^{-\frac{0.15}{\text{days}^{1/2}} \sqrt{t}} + \frac{R_f(\infty)}{R_f(o)} \right]}$ (derived)	$28 \text{ days } \ln \left(1.04 + \frac{t^{1/3}}{\text{days}^{1/3}} \right) + \frac{0.693 t}{\ln \left[\frac{R_f(o)}{R_f(\infty)} \right]}$
INITIAL VALUE $R_f(o)$	10^{-6}m^{-1}	10^{-4}m^{-1}	10^{-4}m^{-1}	Values employed in present proposed model for purposes of comparison with each of the preceding models. With these values, the model acquires the following forms:
FINAL VALUE $R_f(\infty)$	0	0	10^{-9}m^{-1}	
RESUSPENSION FACTOR MODELS, WITH SPECIFIC VALUES EMPLOYED BY THEIR AUTHORS. SEE FIGURE 11.	③ $10^{-6} \text{m}^{-1} e^{-0.0198 \text{ day}^{-1} t}$			③ $10^{-6} \text{m}^{-1} e^{-\left[\frac{0.693 t}{28 \text{ days } \ln (1.04 + \text{day}^{-1/3} t^{1/3})} \right]}$
		② $10^{-4} \text{m}^{-1} e^{-0.0154 \text{ day}^{-1} t}$		② $10^{-4} \text{m}^{-1} e^{-\left[\frac{0.693 t}{28 \text{ days } \ln (1.04 + \text{day}^{-1/3} t^{1/3})} \right]}$
			① $10^{-4} \text{m}^{-1} e^{-0.15 \text{ day}^{-1/2} t^{1/2}} + 10^{-9} \text{m}^{-1}$	① $10^{-4} \text{m}^{-1} e^{-\left[\frac{0.693 t}{28 \text{ days } \ln (1.04 + \text{day}^{-1/3} t^{1/3})} + 0.0602 t \right]}$

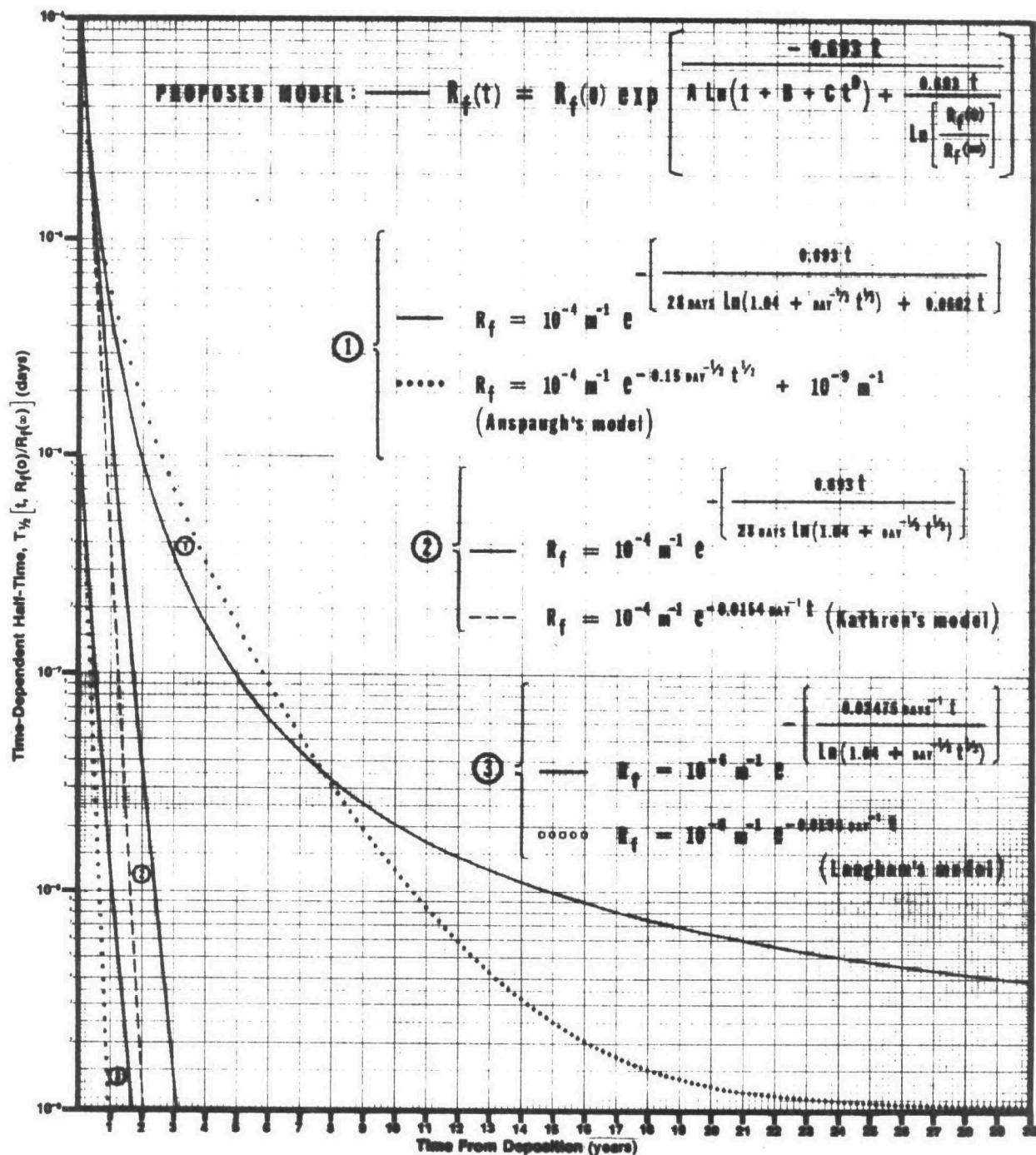


FIGURE 11. Three expressions of the time-dependent resuspension factor based on the proposed model of the time-dependent half-time $T_{1/2}$, with initial and final values as assumed in previous models with which these expressions are compared. See also Table 3.

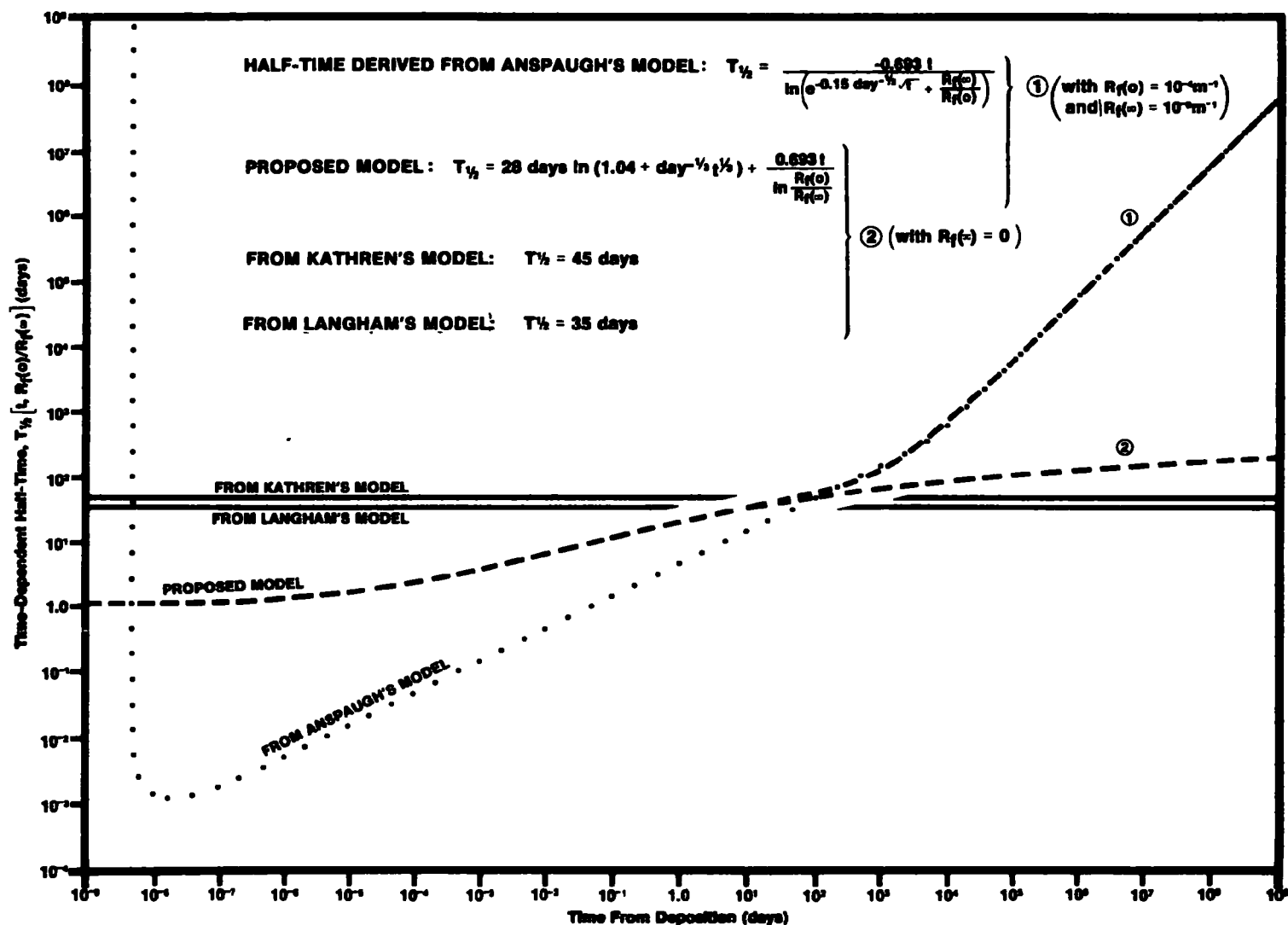


FIGURE 12. Two forms of the proposed model of weathering half-time $T_{1/2}$ as function of time and local conditions (expressed as ratios of initial-to-final resuspension factors $R_f(0)/R_f(\infty)$), compared ① with Anspaugh's model and ② with Langham's and Kathren's models. Values of $R_f(0)$ and $R_f(\infty)$ employed in the proposed model correspond to those used in the comparison models.

As clearly shown in Figure 11, the proposed model (Case 1) produces a time-dependent resuspension factor that behaves similarly to Anspaugh's model, for the same conditions $R_f(0)$ and $R_f(\infty)$. One obvious difference is in the rates at which both models approach the limiting value $R_f(\infty)$. Anspaugh's model includes the "assumption that there may be no further measurable decrease in the resuspension process after 17 years which is the longest period past deposition for which measurements have been reported". No such assumption is made in the present proposed general model, according to which the limit value $R_f(\infty)$ may be approximated before or after 17 years, depending functionally on the ratio $R_f(0)/R_f(\infty)$ (Figure 9).

The versatility of the proposed model is further illustrated in Figure 11 by comparing it with Kathren's and Langham's model. With $R_f(\infty) \approx 0$, as implied in the latter two models, the proposed model matches their behavior fairly closely.

Figure 12 shows the time-dependent behavior of the model (Case 1), comparing it with that of the half-time derived from Anspaugh's model and the values used by Langham and Kathren. Again, the values $R_f(0)$ and $R_f(\infty)$ were chosen to be those of the models with which the comparisons are made, respectively. Besides having certain advantages, evident upon examining the graph, the model has one obvious drawback, that of having inflection points at $t < 1.0$ day and at $t > 100$ days. These were not intended to represent any fundamental notions of the time-dependent behavior of the weathering half-life, but reflect merely the limitations of the model, when shown logarithmically. The first inflection point is due to the requirement that $T_{1/2}$ be other than 0 at time $t = 0$. The second inflection point occurs when both terms of (31) approach comparable values and the second term begins to exert dominance.

SUMMARY AND COMMENTS

The proposed generalized model is based on several simple assumptions and limited empirical data, obtained at various locations and under different conditions, as reported by a number of researchers, some of whom interpreted the data differently. The model is intended to be general, that is, able to conform to different average conditions, when these conditions are expressed as initial and final resuspension factors. This generality is achieved primarily through the second term of the Equation (31), which includes the ratio $R_f(0)/R_f(\infty)$. Additional flexibility is provided by the first term of the expression, which can be adapted to fit a number of empirical observations without materially altering the general model. At present, Equation (31) used in conjunction with the constants of "Case 1" appears to best accommodate the existing data. Additional data, when available, may serve to further refine the model.

The model has limitations and deficiencies, but the greatest drawbacks to be encountered in using it are those of the resuspension factor concept per se, it being best suited for large (ideally "infinite") areas, uniformly contaminated, where average local conditions are maintained or vary uniformly with time, unaltered by unscheduled severe disturbances. The degree to which any model of the resuspension factor (or of the weathering half-life) is successful is clearly linked to the extent to which the above requirements are met.

REFERENCES

- Anspaugh et al., 1973 L. R. Anspaugh, P. L. Phelps, N. C. Kennedy, and H. G. Booth, "Wind Driven Redistribution of Surface Deposited Radioactivity" Environmental Behaviour of Radionuclides Released in the Nuclear Industry Proceedings of a Symposium Organized by the International Atomic Energy Agency, the OECD Nuclear Energy Agency and the World Health Organization and held in Aix-en-Provence, 14-18 May 1973, IAEA, Vienna, 1973
- Anspaugh et al., 1974 L. R. Anspaugh, J. H. Shinn, and D. W. Wilson "Evaluation of the Resuspension Pathway Toward Protective Guidelines for Soil Contamination with Radioactivity" Lawrence Livermore Laboratory, Biomedical Division, IAEA-SM-184/13, UCRL-75250 Preprint/Proceedings of the IAEA/WHO Symposium on Radiological Safety Evaluation of Population Doses and Application of Radiological Safety Standards to Man and the Environment, Portoroz, Yugoslavia, 1974
- Anspaugh et al., 1975 L. R. Anspaugh, J. H. Shinn, and P. L. Phelps "Resuspension and Redistribution of Plutonium in Soils" Lawrence Livermore Laboratory, UCRL-76419 Preprint/Proceedings, Second Annual Life Sciences Symposium, Plutonium-Health Implications for Man, Los Alamos, New Mexico, 1975
- Bernhardt, 1976 David E. Bernhardt "Evaluation of Sample Collection and Analysis Techniques for Environmental Plutonium" Technical Note ORP/LV-76-5. U.S. Environmental Protection Agency, Office of Radiation Programs-Las Vegas Facility, Las Vegas, Nevada, April 1976
- Kathren, 1968 R. L. Kathren "Towards Interim Acceptable Surface Contamination Levels for Environmental PuO_2 " Battelle Pacific Northwest Laboratory, BNWL-SA-1510, in Proceeding of Symposium on Radiological Protection of the Public in a Nuclear Mass Disaster (Strahlenschutz der Bevölkerung bei Einer Nuklear katostroph) pp. 460-470, Interlaken, Switzerland, 1968
- Krey and Hardy, 1970 P. W. Krey and E. P. Hardy "Plutonium in Soil Around the Rocky Flats Plant" U.S. Atomic Energy Commission, Health and Safety Laboratory, HASL-235 Health and Safety (TID-4500), New York, New York, 1970

- Langham, 1971 W. H. Langham "Plutonium Distribution as a Problem in Environmental Science" Los Alamos Scientific Laboratory, LA-4756, UC-41, Los Alamos, New Mexico, December 1971
- Martell et al., 1970 E. A. Martell, P. A. Goldan, J. J. Kranshaar, D. W. Shea and R. H. Williams "Fire Damage" Environment, Vol. 12, No. 4, May 1970
- Mishima, 1964 J. Mishima "A Review of Research on Plutonium Releases During Overheating and Fires" U.S. Atomic Energy Commission, HW-83668 UC-41, Health and Safety (TID-4500, 37th Ed.), Richland, Washington, 1964
- Olafson and Larson, 1961 J. H. Olafson and K. H. Larson "Plutonium, Its Biology and Environmental Persistence" University of California, Los Angeles, School of Medicine, Dept. of Biophysics and Nuclear Medicine, (TIO-4300, 16th Ed.) Los Angeles, California, 1961
- Sehmel and Orgill, 1974 G. A. Sehmel and M. M. Orgill "Resuspension Source Change at Rocky Flats" Battelle Pacific Northwest Laboratories Annual Report for 1973 to the U.S.A.E.C. Division of Biomedical and Environmental Research, Part 3, Atmospheric Sciences -BNWL-1850- Pt. 3, UC-11, pp. 212-214, Richland, Washington, 1974
- Stewart, 1967 K. Stewart "The Resuspension of Particulate Material from Surfaces" Surface Contamination, pp. 63-74, B. R. Fish, Editor, Pergamon Press, 1967
- Volchok, 1971 H. L. Volchok "Resuspension of Plutonium-239 in the Vicinity of Rocky Flats" Proceedings of Environmental Plutonium Symposium. Eric B. Fowler, Richard W. Henderson, Morris F. Milligan, Editors. Los Alamos Scientific Laboratory, Los Alamos, New Mexico, 1971
- Wilson et al., 1961 R. H. Wilson, R. G. Thomas and J. N. Stannard "Biomedical and Aerosol Studies Associated with a Field Release of Plutonium" University of Rochester Atomic Energy Project, WT-1511. Operation Plumbbob - Test Group 57, Program 72, Rochester, New York, November 1960

TECHNICAL REPORT DATA <i>(Please read Instructions on the reverse before completing)</i>		
1. REPORT NO ORP/LV-77-4	2.	3. RECIPIENT'S ACCESSION NO.
4. TITLE AND SUBTITLE Generalized Model of the Time-Dependent Weathering Half-Life of the Resuspension Factor		5. REPORT DATE
		6. PERFORMING ORGANIZATION CODE
7 AUTHOR George V. Oksza-Chocimowski		8. PERFORMING ORGANIZATION REPORT NO.
9 PERFORMING ORGANIZATION NAME AND ADDRESS Office of Radiation Programs-Las Vegas Facility U.S. Environmental Protection Agency P.O. Box 15027 Las Vegas, Nevada 89114		10. PROGRAM ELEMENT NO.
		11. CONTRACT/GRANT NO.
12. SPONSORING AGENCY NAME AND ADDRESS Same as above		13. TYPE OF REPORT AND PERIOD COVERED
		14. SPONSORING AGENCY CODE
15. SUPPLEMENTARY NOTES		
16. ABSTRACT A generalized model has been developed to predict the changes with time in the weathering half-life of the resuspension factor for plutonium 239 and other long-lived radioactive contaminants. The model is largely based on assumptions and empirical data presented by other authors, and is applicable to a wide range of average conditions. These conditions are parametrically described as ratios of initial and final resuspension factors, valid for a given locality. Based on the above model of time-dependent half-life, the corresponding general model of time-dependent resuspension factor is developed and presented in the report. Graphs of both models for a range of conditions and graphic comparisons of specific cases of these models with existant models are included in the report.		
17. KEY WORDS AND DOCUMENT ANALYSIS		
a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Plutonium Isotopes Radioactivity Airborne Contaminants Resuspension Models	Plutonium 239 Alpha particles Resuspension Resuspension factor Weathering half-life	1802 1808 1302 1201
18. DISTRIBUTION STATEMENT Release to public	19. SECURITY CLASS (<i>This Report</i>) Unclassified	21. NO. OF PAGES 53
	20. SECURITY CLASS (<i>This page</i>) Unclassified	22. PRICE