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January 1967

RADIOLOGICAL SAFETY STUDIES
OF SPACE NUCLEAR SYSTEMS
QUARTERLY REPORT NO. 1
September 13 to December 13, 1966

Prepared by
J. R. Coleman and R. S. Denham
NUS CORPORATION

SANDIA CORPORATION

PRIME CONTRACTOR TO THE U.S. ATOMIC ENERGY COMMISSION | ALBUQUERQUE NEW MEXICO, LIVERMORE CALIFORNIA, TONOPAH, NEVADA

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for
Sandia Corporation
under
Contract 48-4310

ABSTRACT

This report describes progress on the development and application of dose models for re-entry of radioisotope-fueled space power units. Ad hoc studies requested by SEPO or Sandia Corporation are also described.

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SUMMARY

This is the first quarterly report under Contract 48-4310 for the development and application of radiation dose and interaction probability models for dispersed and intact re-entry modes of isotopic SNAP units, comparison of the re-entry modes and their radiological consequences, and such other assistance as may be required on special questions which arise during safety evaluations of SNAP systems.

The purpose of Task I of this contract was primarily to define and consider those portions of the overall problem which will require the greatest modification of the Rover Flight Safety Analysis. Concern in this early stage was directed primarily at movement of materials through the environment to man. Portions of the food chain including vegetation and meat were considered and the new ICRP lung model was generalized as much as possible for application to this project.

Efforts under Task II were limited to collection and review of input information on SNAP-19 and SNAP-29. Possible problem areas with the safety analysis of SNAP-19 were identified.

Work presently in progress under Task IV includes development of generalized safety criteria for aerospace systems and calculation of radiation dose to tissue from spherical particles of isotopic fuel materials.

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INTRODUCTION

This is the first quarterly progress report under Contract 48-4310 for the development and application of radiation dose and interaction probability models for dispersed and intact re-entry modes of isotopic SNAP units, comparison of the re-entry modes and their radiological consequences, and other work which may be required on special questions which arise during safety evaluations of SNAP systems.

The work carried out under Task I was primarily to define and consider those portions of the overall problem which will require the greatest modification of the Rover Flight Safety Analysis. Concern in this early stage was directed primarily at movement of materials through the environment to man. Portions of the food chain including vegetation and meat were considered and the new ICRP lung model was generalized as much as possible for application to this project. It is intended that the next efforts will be directed at quantitizing the problem of re-suspension of particulate materials, the GI tract dose model (on the basis of recent ICRP dosimetry) and integrating the various detailed dose models into the overall program. The feasibility of utilizing the Model B dispersion code developed by TRC will be evaluated and atmospheric burnup and transport models will be integrated with the dose models.

Efforts under Task II during the report period were limited to collection and review of input information on SNAP-19 and -29. Possible problem areas with the safety analysis of SNAP-19 were identified. During the second

quarter these efforts will be continued and the developed dose models will be applied to both SNAP-19 and -29 to evaluate the safety of the systems and missions as presently defined. Comparison of re-entry modes (Task III) may also be initiated during this report period.

Work presently in progress under Task IV includes development of generalized safety criteria for aerospace systems and calculation of radiation dose to tissue from spherical particles of isotopic fuel materials. Informal memoranda covering these projects will be issued as the work is completed, and the results will be incorporated on the quarterly report for the period 13 December 1966 - 12 March 1967.

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1.0 TASK I DOSE MODEL DEVELOPMENT

As a first step in the development of dose models under Task I, the types of situations that might occur have been characterized and the qualitative inputs for each situation defined. These qualitative input requirements are the basic framework around which the detailed dose models are to be constructed. The overall mission might be visualized as shown below with essentially these resulting situations:

Types of Resulting Situations

1: Localized situation from intact sealed capsule; relatively few individuals involved. Requirements for evaluation:

- a. probability of occurrence;
- b. populations or individuals in the areas of concern;
- c. dose parameters for external exposure model.

2: Localized dispersion problem; intermediate numbers of individuals involved; possibility of relatively high internal exposures. Requirements for evaluation:

- a. probability of occurrence and probability of specific quantities released;
- b. population densities and distributions in areas of concern;
- c. meteorologic data - environmental transport model;
- d. dose parameters for internal and external exposure model.

3: Large-scale dispersion, large numbers of individuals may be involved; doses (on the average) may be lower. Requirements for evaluation:

- a. probability of occurrence;
- b. dispersion and deposition model;
- c. population densities and distributions;
- d. environmental transport model;
- e. dose parameters for internal and external exposure models.

In relation to the situations described above, the initial interest has been focused on those areas in which it was felt that the most modification

of the Rover Flight Safety Analysis work would be required. This effort has been directed at two general problems thus far: (1) defining and detailing the routes of internal exposure and (2) adapting as much as possible the new ICRP lung model to lung dose calculations. This work will be discussed in detail in the following paragraphs.

The magnitude of the internal radiation exposure to man as a result of radioactive materials dispersed in the environment is dependent on the complex interrelationships of the physical, chemical and biological characteristics of man, the material dispersed and the total environment. A schematic diagram depicting the general routes by which material may reach man is shown below. Those routes shown by dashed lines would be of primary concern only for relatively soluble materials.

In the following paragraphs, internal exposure has been divided into the considerations of inhalation and the considerations of ingestion.

1.2 Inhalation (ICRP Model)

Not all of the particulate material that is taken in through the respiratory process will reach the lung. Many of the particles will be filtered out in the nose, mouth, or trachia-bronchial regions. The most recent attempt at devising a model that mathematically describes the fate of inhaled particles is that of the ICRP Task Group on Lung Dynamics⁽¹⁾. A schematic diagram showing the several deposition sites or compartments and the various clearance pathways proposed in the ICRP model is shown in Figure 1. This model will be used as the basis for estimating lung exposure. The discussion of this model is divided into three sections: (1) Deposition Model, (2) Clearance Model, and (3) Application to Exposure Following a Single Environmental Release.

FIGURE 1

In Figure 1, the following are represented

- N-P The nasopharynx; begins with the anterior nares and extends through the anterior pharynx back and down through the posterior pharynx to the level of the larynx or epiglottis.
- T-B The trachea and the bronchial tree down to and including the terminal bronchioles (Note: compartment N-P and T-B taken together can be considered to represent the entire epithelial area of the respiratory tract which is ciliated and covered with mucus).
- P Respiratory bronchiolus, alveolar ducts, atria, alveoli, and alveolar sacs. Consists of non-ciliated moist epithelium and regarded as the functional area or exchange space of the lungs.
- D_1 Total dust inhaled
- D_2 Dust in exhaled air
- D_3 Dust deposited in the N-P compartment
- D_4 Dust deposited in the T-B compartment
- D_5 Dust deposited in the P compartment
- a Uptake of material deposited in the N-P compartment directly to the blood

- b Clearance of dusts from the N-P compartment by ciliary-mucus transport
- c Uptake of material deposited in the T-B compartment directly to blood
- d Clearance of dusts from the T-B compartment by ciliary-mucus transport
- e Uptake of material deposited in P compartment directly to the blood
- f Clearance of dusts from the P compartment by recruitable macrophages coupled with ciliary-mucus transport.
- g Secondary pulmonary clearance processes, typically slower than "f" but still dependent on endocytosis and ciliary-mucus transport
- h Slow removal of dusts from the P compartment via the lymphatic system. Similar to "g" except that lymph transport replaces the ciliary-mucus transport
- i Secondary pathway in which dust is cleared by the lymphatic system. This implies dissolution of the dust particles partially or completely, but the turnover of lymphocytes may contribute.

1.2.1 Deposition Model

The proposed model provides for the evaluation of the deposition in the several respiratory compartments on the basis of the mass median aerodynamic diameter (MMAD) of the aerosol. The deposition is expressed as the mass percent (or fraction) of the inspired dust deposited as a function of the MMAD. Although three respiratory states were considered (i.e., minute volumes of 11, 20, and 32 liters), the 20-liter minute volume was taken as representative of all three states. Figures 2 and 3⁽¹⁾ illustrate the relationship between percent deposition on a mass basis and MMAD for the N-P and P compartments, respectively. It is assumed that the deposition in the T-B compartment is adequately approximated by assuming a constant fraction deposition for all particle sizes (i.e., approximately 8 percent).

Fig. 2 The deposition estimate for the nasopharyngeal compartment while breathing at a moderate work rate. Different aerosol distributions are represented by the mass or activity median aerodynamic diameters (0.01 to ~100).

Fig. 3 The deposition estimate for the pulmonary compartment while breathing at a moderate work rate. Different aerosol distributions are represented by the mass or activity median aerodynamic diameters (0.01 to ~100).

The model is based on representing aerosols as distributions of particle sizes rather than limiting the treatment to single particle sizes. The log-normal distribution related to particle diameters was assumed. The application of the model as presented is restricted to MMAD ranging from 0.01 to 100 microns but may be reasonably modified for larger particles. Calculations were carried out for geometric standard deviations (σ_g) ranging from 1.2 to 4. (In this case a σ_g of 1 implies a single particle size.) The assumption of the log-normal distribution was not intended to limit the model but to illustrate the broad applicability to both distributions and single particle sizes⁽⁵⁾. The deposition model is most applicable to intact re-entry followed by capsule rupture and release of significant quantities of particulate material, but may also be applied to those cases of widely dispersed materials where the probability of deposition of a single particle may be of interest. For simplicity of handling, the relationships shown in Figures 2 and 3 have been reduced to mathematical expressions and within the restrictions given above, the mass fraction deposited $(MFD)_i$ for each of the compartments can be summarized as follows:

Nasopharynx (N-P)

$$(MFD)_3 = 1/2 \left\{ 1 + \operatorname{erf} (X_3) \right\}$$

$$\text{where } X_3 = 0.561 \ln \left\{ \frac{MMAD}{1.96} \right\}$$

Tracheo-Bronchial (T-B)

$$(MFD)_4 = 0.08$$

Pulmonary (P)

$$(MFD)_5 = 1/2 \left\{ 1 - \operatorname{erf} (X_5) \right\}$$

$$\text{where } X_5 = 0.227 \ln \left\{ \frac{MMAD}{0.106} \right\}$$

1.2.2 Clearance Model

Referring to the schematic representation of the proposed respiratory system model (Figure 1), it is apparent that two parameters must be substituted for each of the clearance pathways "a" through "i" if the model is to be applied. Both the relative quantity of material in any compartment which follows a given pathway (regional fraction = F) and the rate at which this fraction of dust is cleared (biological half-time = $T/2$) must be specified. In some cases, these kinetic parameters are physiologically controlled and remain relatively constant irrespective of the dust material considered. In other cases, however, these values are dependent on the nature of the dust. Because of this latter problem, it is important that the physiochemical nature of the different materials be considered in arriving at suitable kinetic parameters when specific data are lacking.

The ICRP Task Group attempted to organize all of the quantitative clearance information which has been published. These data, presented in Appendix I of the task group report⁽¹⁾, are recommended for use with the respiratory model whenever appropriate to the specific material being considered. In the absence of such information, it has been recommended that the kinetic parameters be estimated by using the classification scheme shown in Table I, together with the clearance half-time and regional fractions presented in Table 2.

TABLE I
PULMONARY CLEARANCE CLASSIFICATION OF INORGANIC COMPOUNDS

- Class Y -- Avid retention: cleared slowly (years)
 Carbides -- actinides, lanthanides, Zr, Y, Mn
 Sulfides -- none
 Sulfates -- none
 Carbonates -- none
 Phosphates -- none
 Oxides and hydroxides -- lanthanides, actinides Groups 8(V and VI), 1b, 2b (IV and V), 3b except Sc^{3+} , and 6b
 Halides -- lanthanide fluorides
 Nitrates -- none
- Class W -- Moderate retention: intermediate clearance rates (weeks)
 Carbides -- Cations of all Class W hydroxides except those listed as Class Y carbides
 Sulfides -- Groups 2a (V+VI), 4a (IV-VI), 5a (IV-VI), 1b, 2b, and 6b (V+VI).
 Sulfates -- Groups 2a (IV-VII), and 5a (IV-VI)
 Carbonates -- lanthanides, Bi^{3+} and Group 2a (IV - VII)
 Phosphates -- Zn^{2+} , Sn^{3+} , Mg^{2+} , Fe^{3+} , Bi^{3+} and lanthanides
 Oxides and hydroxides -- Groups 2a (II-VII), 3a (III-VI), 4a (III-VI), 5a (IV-VI), 6a (IV-VI), 8, 2b (VI), 4b, 5b, and 7b Sc^{3+}
 Halides -- lanthanides (except fluorides), Groups 2a, 3a (III-VI), 4a (IV-VI) 5a (IV-VI), 8, 1b, 2b, 3b (IV-V), 4b, 5b, 6b and 7b
 Nitrates -- all cations whose hydroxides are Class Y and W
- Class D -- Minimal retention: rapid clearance (days)
 Carbides -- see hydroxides
 Sulfides -- all except Class W
 Sulfates -- all except Class W
 Carbonates -- all except Class W
 Phosphates -- all except Class W
 Oxides and Hydroxides -- Groups 1a, 3a (II), 4a (II), 5a (II, III), 6a (III).
 Halides -- Groups 1a and 7a
 Nitrates -- all except Class W
 Noble Gases -- Group 0

Note: where reference is made from one chemical form to another, it implies that an in vivo conversion occurs, e.g. hydrolysis reaction.

The following period table of the elements is used with the foregoing classification

Group																	
Period	1a	2a	3b	4b	5b	6b	7b	8		1b	2b	3a	4a	5a	6a	7a	0
I	H																He
II	Li	Be										B	C	N	O	F	Ne
III	Na	Mg										Al	Si	P	S	Cl	Ar
IV	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Kr
V	Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	Xe
VI	Cs	Ba	La*	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	Rn
VII	Fr	Ra	Ac↑														
* Lanthanides			Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy		Ho	Er	Tm	Yb	Lu
↑ Actinides			Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf		Es	Fm	Md	No	Lw

TABLE 2
CONSTANTS FOR USE WITH CLEARANCE MODEL

		Class (D)	Class (W)	Class (Y)
N-P	(a)	4 min/0.50	4 min/0.10	4 min/0.01
	(b)	4 min/0.50	4 min/0.90	4 min/0.99
T-B	(c)	10 min/0.50	10 min/0.10	10 min/0.01
	(d)	10 min/0.50	10 min/0.90	10 min/0.99
P	(e)	30 min/0.80	90 days/0.15	360 days/0.05
	(f)	n. a.	24 hour/0.40	24 hour/0.40
	(g)	n. a.	90 days/0.40	360 days/0.40
	(h)	30 min/0.20	90 days/0.05	360 days/0.15
Lymph	(i)	30 min/1.00	90 days/1.00	360 days/0.10

Note: The first value is the biological half-time; the second is the regional fraction. The lymphatic clearance for Class Y compounds indicates that a 10 per cent regional fraction follows a 360-day biological half-time. The remaining 90 percent is presumed to be permanently retained in the nodes and is subject only to radioactive decay.

Pulmonary clearance half-time ($T_{1/2}$) for specific materials are to be used whenever available in place of the fixed values suggested for e, g, h, and i in Table 2. The regional fractions to be used in conjunction with clearance half-time for specific materials are determined by the dust classification D, W, or Y which is most consistent with the pulmonary clearance value. In this case, the classification will be determined on the basis of the following:

Class D	$T_{1/2} \leq 10$ days
Class W	$10 < T_{1/2} \leq 100$ days
Class Y	$100 < T_{1/2}$ days

1.2.3 Time Relationship of Deposited Material for Dose Calculation

In order to consider the time relationship in a general sense, we can define Table 2 in terms of a position matrix as follows:

n p	Class D	Class W	Class Y
a	1, 1	1, 2	1, 3
b	2, 1	2, 2	2, 3
c			
.		p, n	
.			
i	9, 1	9, 2	9, 3

To further simplify the presentation define the biological turnover constant as:

$$\lambda = \frac{0.693}{T_{1/2}}$$

Thus we have:

$F_{n,p}$ = The regional fraction for the n^{th} class of material by the p^{th} pathway

$\lambda_{n,p}$ = Biological turnover constant for the n^{th} class of material by the p^{th} pathway

For example, from Table 2, we would have the following:

$$F_{3,4} = 0.99 \text{ (pathway d, Class Y)}$$

$$\lambda_{2,3} = \frac{0.693}{10} \text{ min}^{-1} \text{ (pathway c, Class W)}$$

With these definitions we can express the time dependent quantities of material in each respiratory compartment. However, in terms of radiological

concern, the pulmonary compartment (D_5) and the transfer from all compartments to the GI tract and the blood are of prime interest. If we consider a single exposure, then the time related quantity of material in the pulmonary compartment is:

$$D_{5,n}(t) = D_5(0) \sum_p F_{n,p} e^{-\lambda_{n,p} t} \quad p = 5, 6, 7, \text{ and } 8$$

where:

$D_{5,n}(t)$ = Grams of material of class n in the pulmonary compartment at time, t

$D_5(0)$ = Grams of material in pulmonary compartment initially deposited.

and:

$\lambda_{n,p}$'s are in consistent units with t

Insofar as transfer to the GI tract and the blood is concerned, only the relatively rapid transfer need be considered since if the material is retained in the lung for appreciable time periods, the dose to the lung would be of most concern. Thus, we can consider the transfer to the GI tract and blood on the basis of the total integrated quantities of materials transferred without considering the relatively slow pathways. The total transfer to the GI tract would thus be:

$$G_n = D_3(0) F_{2,n} + D_4(0) F_{4,n} + D_5(0) F_{6,n}$$

where:

G_n = Total grams of material transferred through GI tract following a single exposure.

The total integrated transfer to the blood is:

$$B_n = D_3(0) F_{1,n} + D_4(0) F_{3,n} + D_5(0) [F_{5,n} + F_{8,n}]$$

where:

B_n = Total grams of material transferred to the blood following a single exposure

with the restriction that $F_{5,n}$ and $F_{8,n}$ are considered only if $n = 1$ (that is the very soluble class of material).

1.2.4 Application of Model to Exposure Following a Single Environmental Release

Mass Deposited in Respiratory Compartments

Assuming spherical particles, the total mass for the j^{th} particle size group per cubic meter of air is:

$$M_j = \frac{1}{6} \pi \gamma_j^3 \eta_j \rho$$

where:

M_j = Total mass per cubic meter of air for j^{th} particle size group (gm/m³)

γ_j = Diameter of the average mass for the j^{th} particle size group (cm)

η_j = Number of particles of the j^{th} size group per cubic meter of air (m⁻³)

ρ = Density of the j^{th} particle size group (gm/cm³)

The quantity of material deposited in the i^{th} respiratory compartment is then:

$$D_{ij} = (\text{MFD})_{ij} B \int_t M_j dt = (\text{MFD})_{ij} B \frac{1}{6} \pi \gamma_j^3 \rho \int_t \eta_j dt$$

where:

D_{ij} = Total mass of j^{th} particle size group deposited in respiratory compartment i (gm)

$(MFD)_{1j}$ = Mass fraction deposited in i^{th} respiratory compartment for j particle size (defined in section 1.2.1)

B = Breathing rate; taken as 20 liters per minute, $(3.33 \times 10^{-4} \text{ m}^3/\text{sec})$

where the integration is performed over the time of exposure to particulates in air. For simplicity we will define the mean time-integrated particle concentration as N_j or:

$$N_j = \int_t \gamma_j dt \text{ (particle-sec/m}^3\text{)}$$

Thus for the quantity of material deposited in each respiratory compartment we have:

$$D_{1j} = 1.75 \times 10^{-4} (MFD)_{1j} \gamma_j^3 \rho N_j$$

The mass deposited in each respiratory compartment may now be summarized using the relationships for (MFD) given previously. These are the values for $D_1(0)$ required in the equations presented in section 1.2.3 and are so designated.

Nasopharynx (N-P)

$$D_{3,j}(0) = 0.87 \times 10^{-4} \left\{ 1 + \text{erf}(x_{3,j}) \right\} \gamma_j^3 \rho N_j$$

$$\text{where } X_{3,j} = 0.561 \ln \left\{ \frac{MMAD_j}{1.96} \right\}$$

Trachea-Bronchial (T-B)

$$D_{4,j}(0) = 1.4 \times 10^{-5} \gamma_j^3 \rho N_j$$

Pulmonary (P)

$$D_{5,j}(0) = 0.87 \times 10^{-4} \left\{ 1 - \text{erf}(X_{5,j}) \right\} \gamma_j^3 \rho N_j$$

$$\text{where: } X_{5,j} = 0.227 \ln \left\{ \frac{\text{MMAD}_j}{0.106} \right\}$$

Dose to the Pulmonary Compartment

The prime interest is the total dose delivered to the pulmonary compartment. Following the inhalation of particles of the j^{th} size group, the total dose is given by:

$$[\text{TD}]_{5,j} = \int_0^{\infty} D_{5,j}(t) S_j(t) R_{5,m} dt$$

where:

$[\text{TD}]_{5,j}$ = Total dose delivered to the pulmonary compartment following inhalation of the j^{th} particle size group (rads)

$S_j(t)$ = Specific activity of the j^{th} particle size group as a function of time (activity per gram)

$R_{5,m}$ = Dose conversion factor for the pulmonary compartment for the radioactive material (m) being considered (rad per day per unit activity)

Noting that $S_j(t)$ can be expressed as $S_j(0) e^{-\lambda_m t}$

where:

λ_m = Decay constant of material m (days⁻¹)

$S_j(0)$ = Specific activity at time of inhalation

and using $D_{5,j}(t)$ as previously given, the total dose can now be written as:

$$[\text{TD}]_{5,j} = 0.87 \times 10^{-4} \left\{ 1 - \text{erf}(X_{5,j}) \right\} \gamma_j^3 \rho N_j S_j(0) R_{5,m} \sum_p F_{n,p} \left[\frac{1}{\lambda_{n,p} + \lambda_m} \right]$$

where as before $p = 5, 6, 7$, and 8 and where $\lambda_{n,p}$ are in days⁻¹.

The total integrated dose from all particle sizes would then be:

$$\begin{aligned} TD_5 &= \sum_j TD_{5,j} \\ &= \sum_j 0.87 \times 10^{-4} \left\{ 1 - \text{erf} (X_{5,j}) \right\} \gamma^3 \rho N_j S_j(0) R_{5,m} \sum_p F_{n,p} \\ &\quad \left[\frac{1}{\lambda_{n,p} + \lambda_m} \right] \end{aligned}$$

where for summary the subscripts are designated:

- j = refers to particle size group
- 5 = refers to pulmonary compartment
- m = refers to radioactive material of interest
- n = refers to ICRP material classification
- p = refers to pathway out of pulmonary compartment

1.3 Ingestion Exposure

The ingestion of radioactive materials released into the environment can result from eating foods, drinking water or by swallowing particles trapped in the upper respiratory tract as a result of inhalation. The contribution from swallowing inhaled particles has already been considered. In this section the intake resulting from contaminated vegetation and meats will be considered.

The method of approach will be to devise equations for the concentration (either particle or activity) on a per weight basis in individual food types. These concentrations together with food consumption figures will yield the particle or activity intake from an individual food type.

Additionally, since the impact of any given exposure level is dependent on the numbers of people involved, relationships must be developed not only for exposure levels, but also for the number of people at any given exposure level.

1.3.1 Contamination of Vegetation

From section 1.1 it is apparent that the contamination of vegetation can result from three pathways: (1) that of direct deposition on plants (2) that of re-suspension and secondary deposition and (3) that of soil uptake.

This section is concerned with the direct deposition on plants and the subsequent transfer to man. The purpose of this section is to present a model applicable to the particle or activity concentration in food crops. In this regard, the time of deposition relative to the various stages in the plant growth cycle is a major factor affecting the projected intake to man resulting from a given deposition. Much less of the deposited material will enter the food chain if the deposition occurs during a period when there is little vegetation available for contamination than if the deposition occurs immediately prior to harvesting of the crop. With increasing time between harvest and deposition, the transmission of the deposited materials through the food chain will diminish as a result of dilution by new plant growth and removal by weathering. In general, this dilution and removal of deposited material (following a single deposition) on vegetation has been characterized by a half-residence time of the deposited material related to a simple exponential function. That is if we define the following:

T_p = Physical half-life of the radionuclide being considered

T_b = Half-residence time on vegetation

T_e = Effective half-life of deposition on crops $\frac{T_p T_b}{T_p + T_b}$

Then the time dependent concentration of the deposited material on the crops can be expressed as:

$$C_t = C_o e^{-\lambda t}$$

where:

C_o = Concentration immediately following the deposition

C_t = Concentration at some time, t , following the deposition

λ = Effective removal constant $0.693/T_e$

Although for the most part studies of the removal of radioactive particulate material by weathering processes have been related to grasses or grass type vegetation, studies have included such vegetation as cabbage, potatoes and desert shrubs. ^(2, 3) A surprising similarity in the half-residence time on various plants has been observed for a variety of radionuclides. Thompson ⁽⁴⁾ has suggested that this indicates the common behavior of particulate materials on plants and suggests that the most probable effective half-life of any radionuclide on plants can be calculated by:

$$T_e = \frac{13 T_p}{13 + T_p} \text{ (days)}$$

The assumption of a simple exponential model may not apply to all crops without exception. Thus, in the following paragraphs relationships will be developed both for an exponential type model and a general case and the conditions under which the general model reverts to the simple exponential will be defined. The exponential model is directly applicable to grains and leafy vegetables.

1.3.1.1 Concentration following a Single Deposition

In order to facilitate further discussion the exponential model shown above will be re-written as:

$$C_{j,k,n} = C_{j,k,1} e^{-\lambda_{j,k} (n-1)}$$

where:

j refers to the jth particle size group

k refers to the kth crop

1 refers to the time of deposition

n refers to the time of measurement or harvest

The above meanings will be attached to the subscripts for the remainder of this section. In order to develop the general case, the following will be defined:

$C_{j,k,n}$ = Concentration of the jth particle size group in the crop designated as k on the nth day following a deposition at 1 (particles/kilogram)

$d_{j,1}$ = Deposition of the jth particle size group on the 1th day (particles/m² of ground surface)

$f_{k,1}$ = Fractional ground coverage for the kth crop on the 1th day (m² effective plant surface per m² ground surface)
Consider sticking factor to be 1.0, this may be modified for various particle sizes.

$M_{k,1}$ = Mass dilution provided by the kth crop on the 1th day (kilograms of crop per m² of ground surface)

$u_{k,1}$ = Ratio of crop mass on day 1 to day 1 + 1 That is
 $M_{k,1}/M_{k,1+1}$

Note that $u_{k,i} = [1 - g_{k,i}]$

where:

$$g_{k,i} = \frac{M_{k,i+1} - M_{k,i}}{M_{k,i+1}}$$

$\delta_{j,k,i}$ = Fractional loss (due to wind rain or other cause) of the j^{th} particle size group from the k^{th} crop on the i^{th} day

Considering the above we see that:

$$C_{j,k,i} = d_{j,i} \frac{f_{k,i}}{M_{k,i}}$$

and:

$$C_{j,k,n} = d_{j,i} \frac{f_{k,i}}{M_{k,i}} \prod_{i=1}^{n-i} u_{k,i} \prod_{i=1}^{n-i} (1 - \delta_{j,k,i}) \quad n > i$$

At this point we can investigate under what conditions the more simple exponential model might be used. Consider that for a single deposition the equation may be written as shown below in terms of what has already been defined:

$$C_{j,k,n} = C_{j,k,i} \prod_{i=1}^{n-i} (1 - g_{k,i}) \prod_{i=1}^{n-i} (1 - \delta_{j,k,i}) \quad n > i$$

If for a particle crop $g_{k,i}$ and $\delta_{j,k,i}$ are not functions of time, i.e., constants over the period of interest, then the equation may be written:

$$C_{j,k,n} = C_{j,k,i} [1 - g_{k,i}]^{n-i} [1 - \delta_{j,k,i}]^{n-i}$$

This can be expressed as exponentials as follows:

$$C_{j,k,n} = C_{j,k,i} e^{\{\ln [1 - g_{k,i}]\}^{n-i}} e^{\{\ln [1 - \delta_{j,k,i}]\}^{n-i}}$$

Thus for constant fractional growth rate and constant fraction loss of particulate material, the simple exponential model would be applicable.

Additionally, if $g_{k,i}$ and $\delta_{j,k,i}$ are very small in comparison to 1.0, we can express the above equation as:

$$C_{j,k,n} = C_{j,k,i} e^{-g_{k,i}(n-i)} e^{-\delta_{j,k,i}(n-i)} = C_{j,k,i} e^{-\lambda_{j,k}(n-i)}$$

For the single depositions we therefore have two equations that might be applied (depending on the crop) in order to estimate the particle concentration following deposition.

- (B.1) The case of constant growth rate and constant fractional loss of particulates:

$$C_{j,k,n} = d_{j,i} \frac{f_{k,i}}{M_{k,i}} e^{-\lambda_{j,k}(n-i)}$$

- (B.2) The general case

$$C_{j,k,n} = d_{j,i} \frac{f_{k,i}}{M_{k,n}} \prod_{i=1}^{n-i} (1 - \delta_{j,k,i})$$

where the equation (B.2) has been simplified by noting that:

$$\frac{1}{M_{k,i}} \prod_{i=1}^{n-i} u_{k,i} = \frac{1}{M_{k,n}}$$

The importance of the time of deposition as related to the time of harvest can be illustrated simply by using the exponential model and the most probable effective half-time suggested by Thompson. Consider a radionuclide with a long physical half-life deposited on a crop near the beginning of a 100-day growing season. By the time of harvest the concentration of activity would be reduced by a factor of approximately 200, disregarding any loss by decay.

1.3.1.2 Multiple or Continuous Depositions

In general the continuous deposition can be treated as a series of single depositions and the combined effect obtained by summing the individual contributions. For the two cases considered, the particle concentration for the j^{th} particle size group on the k^{th} crop at some time, n , following a series of depositions that recurred over the period $n \rightarrow i$ would be:

(B.3) Exponential case

$$C_{j,k,n} = \sum_{i=1}^n d_{j,i} \frac{f_{k,i}}{M_{k,i}} e^{-\lambda_{j,k}(n-i)}$$

(B.4) General case

$$C_{j,k,n} = \frac{1}{M_{k,n}} \sum_{i=1}^n d_{j,i} f_{k,i} \bar{\delta}_{j,k,i}$$

where for simplicity the following has been defined:

$$\bar{\delta}_{j,k,i} = \prod_{i=1}^{n-i} (1 - \delta_{j,k,i})$$

1.3.1.3 Human Intake

To determine the human intake, we must consider the reduction due to processing the food crop and the quantity of food eaten per day. Considering these factors, the daily intake of particles of the j^{th} size group from the k^{th} crop harvested on the n^{th} day following depositions that occurred over $n \rightarrow i$ would be:

$$(B.5) \quad I_{j,k,n} = K_k F_{jk} C_{j,k,n}$$

where:

- K_k = Kilograms of the k^{th} crop eaten per day
 $F_{j,k}$ = Fraction of particles of the j^{th} size group remaining after processing or preparation (i.e., peeling, washing, etc.)
 $I_{j,k,n}$ = Number of particles of the j^{th} size group ingested per day from the k^{th} crop harvested at time, n . (Note if, $I_{j,k,n}$ is less than 1.0 it can be treated at the probability of ingesting a single particle.)

As treated here, for any given crop once harvested and processed, $I_{j,k,n}$ is a constant over the period of intake. However, the activity of the particles is not constant, if time periods are such that decay is significant. Thus, in order to apply the above equation to dose calculations we must take into account the decay of the radioactive material present. The total effective intake for dose calculations may thus be defined as:

$$(TI)_{j,k,n} = I_{m,k,n} \int_{n+\Delta}^{n+\Delta+\text{one year}} S_j e^{-\lambda t} dt$$

where:

- S_j = Specific activity of the j^{th} particle size at time of reference $t = 0$
 n = Time after reference time "0" that harvest occurs
 Δ = Delay due to processing or storage until available for intake
 λ = Decay constant for radionuclide being considered

For a radionuclide for which the decay during the one year intake period is insignificant, the total intake would be simply:

$$(TI)_{j,k,n} = I_{j,k,n} S_j \quad (365)$$

The proposed approach is not limited to just the concept of concentration in food. If individual intake figures are lacking, it is possible to use the concept of the area required to feed one person. This can be seen by observing that the number of particles retained in the crop per m^2 of contaminated area is simply:

$$C_{j,k,n} \cdot M_{r,n} = \sum_{i=1}^n d_{j,i} f_{k,i} \bar{\delta}_{j,k,i}$$

And using the area required to feed one person, the individual intake would be

$$I_{j,k,n} = \frac{\gamma_r}{365} F_{j,k} \sum_{i=1}^n d_{j,i} f_{k,i} \bar{\delta}_{j,k,i}$$

where:

γ_k = The area planted in crop, k, needed to feed one person (m^2 /person) per year

This is essentially the same equation as used in the Rover Flight Safety Report. However, in the above equation, both the deposition and the retention on plants are a function of the time of harvest as related to the deposition.

The total effective intake when the decay of the nuclide of interest is of significance would be obtained by the same time integration as shown above.

The number of people involved at any given level of intake can be estimated by using either the total quantity of crop contaminated at a given level together with the average annual intake of the crop, or the total area

contaminated at a given level together with the area required to feed one person for one year. Thus,

$$N_{j,k,l} = \frac{A_{j,k,l}}{\gamma_k} = \frac{Q_{j,k,l}}{K_k(365)}$$

where:

$N_{j,k,l}$ = Number of people exposed to a level of intake, l , from the j^{th} size group on the k^{th} crop

$A_{j,k,l}$ = Total area of the k^{th} crop contaminated to a level of l (m^2)

$Q_{j,k,l}$ = Total quantity of food crop, k , contaminated to a level of l (kilograms)

γ_k and K_k have previously been defined.

1.3.2 Contamination of Meats

Two general pathways must be considered in relation to animals in the human food chain: (1) The pathway related to ingestion of meat, and (2) the pathway related to the ingestion of animal products such as milk, cheese, eggs, etc. A model for the first of these pathways is described below. Inhalation intake by animals as related to the transfer to man will not be considered since if levels are such as to result in a significant transfer through the food chain, the inhalation intake by man himself would be far more critical. This entire pathway is important only for those elements and chemical forms generally metabolized by animals.

1.3.2.1 Concentration in Meats

For the transfer from animal feeds to meat as a route of human exposure, we will assume a simple single compartment exponential model for the animal. Although more complex systems including consideration of the interchange dynamics associated with a blood or other fluid compartment

might be envisioned, the effects of such a compartment are generally quite rapid and not of importance in considering the longer term buildup of materials in the flesh of the animal. This assumption can be considered as a parallel to the assumption of a single compartment exponential model for humans such as used by the ICRP.

The concentration of any radioactive material in the flesh of an animal (as a function of time and disregarding decay) is thus given by:

$$(CM) = e^{-\lambda_b t} \int_0^t \frac{kf'}{M'} (CF) e^{-\lambda_b t} dt$$

where:

(CM) = Concentration of material in flesh (activity per kilogram)

k = Daily intake of feed (kilograms)

f' = Fraction of intake (either particle or activity) absorbed and deposited in the flesh

M' = Mass of material in which nuclide is deposited (kilograms)

(CF) = Concentration of material in feed (either particles or activity per kilogram)

λ_b = Effective biological turnover rate for organ of interest (days⁻¹)

t = Time from initial intake to time of interest (days)

If we consider primarily those animals which are herbivorous the above equation can be restated in terms of what has previously been said regarding the particulate or activity concentration in plants. We need consider only two general cases: (1) Intake at a relatively constant concentration in the feed resulting from the feeding of harvested crops that have previously been contaminated, and (2) continuous intake with a changing concentration in the feed as a result of feeding off of growing contaminated plants.

These cases are shown below:

Continuous Intake - Constant Concentration in Feed

$$(CM)_{j,k,m,t} = \frac{k_{k,m} f_{j,m}}{M'_{m,t}} S_j \frac{C_{j,k,n}}{\lambda_b} \left[1 - e^{-\lambda_b t} \right]$$

Continuous Intake Changing Concentration in Feed

(Each day intake treated separately and total concentration is just the sum of the resultant time dependent contribution from each days' intake)

$$(CM)_{j,k,m,t} = \frac{k_{k,m} f'_{j,m}}{M'_{m,t}} S_j \sum_{n=1}^t C_{j,k,n} e^{-\lambda_b (t-n)}$$

where:

t = refers to the time of slaughter or measurement in the animal

m = refers to the m^{th} type of animal used for human consumption

$(CM)_{j,k,m,t}$ = The concentration of activity (not corrected for decay) in the meat of the m^{th} type of animal at some time, t , as a result of the contamination of the k^{th} type of feed crop by particles of the j^{th} size group (activity per kilogram)

$k_{k,m}$ = Daily intake of the k^{th} type of crop by the m^{th} type of animal. (kilograms/day)

$f'_{j,m}$ = Fraction of the activity of the j^{th} size particle absorbed from the gut and deposited in the flesh of the m^{th} type animal (for insoluble material this is a product of the small fraction that is dissolved in the gut and the fraction of this soluble portion that is eventually deposited in the flesh)

$M_{m,t}$ = Mass of the flesh of the animal in which the radioactive material deposits. Kilograms measured at time, t .

S_j = Previously defined

$C_{j,k,n}$ = Previously defined for contamination of vegetation.

1.3.2.2 Human Intake

The intake by individuals and the numbers of people involved at a given intake level can be estimated in the same manner as described for contamination vegetable crops.

2.0 TASK II - DOSE MODEL APPLICATIONS

The primary intent of this task is to utilize the dose models developed under Task I to calculate the probability and magnitude of doses for the SNAP-19 and SNAP-29 systems. Performance of this task therefore requires the dose models, which are still in process of development, and the specific input data on the SNAP-19 and SNAP-29 fuel form properties, mission profile, and failure modes. Some of these factors have not been firmly fixed for either the SNAP-19 or -29 systems. Further, the TRC dispersion code, Model B, required as part of the basic input to the dose model has not been established as a reliable working entity over the range of particle sizes and densities specifically of concern to human exposure. Consequently, efforts under this task during the first quarter of the contract period were limited to gathering and review of input information on the pertinent SNAP projects and identification of principle areas of uncertainty.

NUS representatives met with SNAP-19 personnel of the Martin Company on November 29, 1966, to discuss dose models and techniques of safety analysis being used by Martin in preparation of the SNAP-19 Safety

Analysis Report. The meeting was also attended by representatives of the AEC - SEPO and Sandia Corporation. Further discussions with both the SNAP-19 and SNAP-29 project organizations at Martin are planned the second quarter of the report period.

One area of uncertainty critical to safety analysis of the SNAP-19 is the question of particle size distribution, particularly with respect to the fine particles, less than 10 microns in diameter. These particles, which may be present both as a result of the production of the fuel form and produced in varying quantities at various stages of the mission from shipment through launch to re-entry as a result of shock vibration and thermal effects, represent the most direct route of internal exposure to man. The weight fraction of readily respirable particles at the time of capsule re-entry has been variously estimated at from 10^{-6} to 10^{-3} . In addition, fine particles are created by the re-entry heat pulse--"pitch out" fines of unknown size distribution and total mass, and ablated molecules with a particle size in the range of 10^{-3} to 5×10^{-2} microns and representing perhaps 2% of the total mass. Finally, if the capsule survives atmospheric re-entry heating but fails on impact, it has been reported that up to 10% of the total mass may be reduced to fines and available for atmospheric re-suspension and dispersion. It is clear that studies of distribution and number of fines must be continued to provide meaningful input to the dose model studies.

Another question which will require further study is the potential exposure to man through contamination of cultivated agricultural crops. The potential release of significant quantities of fine particles can, in certain locations and seasons, result in the contamination of leafy vegetables being grown for consumption by man. Although in general the solubility of fuel material is negligible, the potential exists for exposure of the GI tract to whole particles and the creation of absorbable fines by crushing of the

fuel form between the teeth. Information on the probability of such occurrences, the effect of washing of vegetables, data on passage of particles through the GI tract and other factors affecting the dose to man require exploration. At the time the discussions were held with the Martin Company SNAP-19 project group, these questions were not being explored to any significant extent, although extensive studies of the marine branch of the food chain were apparently being made.

Some questions definitely exist on the calculation and application of lung dose resulting from inhalation of one or more particles. It has been generally agreed that particle dose should be normalized over some fraction of the entire lung rather than the immediate vicinity of the particle or particles. With the distribution of radioactive material in the form of discrete fuel particles, the probability of inhalation of more than one particle by any one individual may be very small. Under these circumstances the question exists as to whether dose from a particle should be averaged over the entire mass of the lung, or only the mass of the largest single lobe (225 grams). Once this question is raised the problem of how to treat 1, 2, 3, 4, or any other number of particles must also be answered. As discussed in the Task I section of this report, the ICRP dose model is most applicable for statistical numbers of particles. The clearance section of this model can be adjusted for use with single particles, but the questions of the mass of the lung over which the dose should be averaged and the standard against which this dose should be compared remain open. However, since the only standards presently available are those based on averaging over the entire lung; it would appear reasonable to use this method of averaging in the general approach, but allowing for modifications on the basis of any new information that might become available.

Generalized criteria for space nuclear systems are being developed and studied under Task IV. With the SNAP-19 project, the prime contractor is utilizing the "PdN" concept for determining the acceptability of a system from a radiological safety standpoint. This concept compares (probability of exposure) X (number of persons exposed) to a constant divided by the dose. The present formulation used by Martin for whole body exposures is

$PdN \leq .05$ r. This is not overly restrictive on the high end of the dose scale (permitting a probability of 10^{-4} of one person receiving a lethal dose in the absence of any other exposures), but results in a very tight restriction of dose if the probability and the number of persons exposed is great, i.e., in worldwide fallout resulting from planned re-entry and burnup. In such a case the probability of exposure approaches 1, the number of persons can approach 3×10^9 and the dose is, therefore, restricted to 1.6×10^{-11} r, approximately 10 orders of magnitude less than the annual average dose from natural back-ground radiation. The basic problem of the PdN approach is that it recognize no threshold for radiation injury. In the genetic sense this may be correct, but valid questions may be raised on placing a limit on the probable genetic pool dose as low as .05 r per mission, per program, or per year.

3.0 TASK III - INTACT/DISPERSED REENTRY INTERACTION COMPARISON

No work under this task was initiated during this report period.

4.0 TASK IV - AD HOC ASSISTANCE

Work was started, at the request of the Safety Officer-SEPO, on the establishment of proposed exposure guidelines for aerospace nuclear applications. During the report period the proposed guideline for external whole body gamma exposure resulting from accident conditions was completed. Work is progressing on establishment of internal exposure guidelines for accident conditions and for both internal and whole body guidelines for normal operations.

A study of potential tissue doses from plutonium-238 and thulium-170 and -171 was initiated at the request of the Sandia Radiological Safety Division. This study is exploring the range-energy relationships for the ^{238}Pu alpha in plutonium dioxide, the quantity of tissue irradiated and the dose, and the average dose to the lung as a function of particle

size. With the thulium isotopes, the dose rate at the surface of the Tm_2O_3 particles, the average dose rate to one square centimeter area adjacent to the particle, the integrated dose to the one square centimeter area, and the dose rate as a function of tissue depth are being studied.

Also under this task, a visit was made to the office of the Safety Officer-SEPO at his request for discussion of safety criteria for the NANS program.

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