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High Altitude Burnup

Volume II

Martin Marietta Corporation

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FOREWORD

This topical report, the final requirement under Sub-subtask 7.3.4, has been prepared by Martin Marietta Corporation, Nuclear Division, in compliance with the requirements of Atomic Energy Commission Contract AT(30-3)-217.

Volume I has been prepared to permit ready dissemination of the results of this study. It presents a brief description of the problem, method of attack, conclusions and recommendations.

Volume II contains in detail the work which forms the basis for these recommendations.

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SUMMARY

The principal objective of The Upper Atmosphere Re-Entry Study has been to evaluate methods of determining the fate of radioactive fuels (in space power units) burned up on re-entry into the earth's atmosphere. This report broadly reviews several such methods. That method which best produces the essential data on the radioactive particulate, and which is investigated in detail, envisions experimental programs in two distinct areas of study:

- (1) Particulate characteristics.
- (2) Particulate fate.

The particulate characteristics experiments derive from an analytical study on the ablation of plutonium fuel. This study revealed that the particulate size distribution resulting on re-entry burnup is a function of:

- (1) The fuel form in use.
- (2) The material velocity and atmospheric density at the time the fuel is exposed.

The particulate fate experiments derive essentially from studies of the effects of thermal and turbulent diffusion, and wind and gravity dispersion, on the particulate cloud. These studies revealed a lack of information about these parameters at altitudes between 100,000 and 200,000 feet. Without such information, precise definition of the time of storage and locale of deposition for a radioactive particulate is impossible.

A subsidiary objective of the program has been to recommend experimental programs for determining:

- (1) The size distribution of particulate resulting on re-entry burnup of the fuel.
- (2) The atmospheric dispersion of the particulate.

The particulate characteristics experiments recommended are:

- (1) Tests to verify the analytical technique for prediction of final particle size of fuel as a function of initial particle size, velocity and atmospheric density.

- (2) A series of tests on other fuel forms to determine the physical constants (specific heat, heats of vaporization, combustion, etc.) required to predict analytically the particle size distribution which will result on their re-entry to the earth's atmosphere.
- (3) Nucleation and agglomeration tests to obtain empirical evidence of the fate of fuel vaporized in the presence of the vapor created on ablation of the heterogeneous generator package. A fuel bearing coagulum should result; its fate is of interest.

The particulate fate experiments recommended will produce data to permit analytical prediction of residence time, transport, dilution and area of deposition of particulate as functions of particle size, density, shape, spatial origin and season of injection. It is proposed to:

- (1) Determine turbulence coefficients and wind velocity and direction data as functions of altitude, latitude and time. When properly coupled with data on particulate characteristics this information will permit prediction of the history of a particulate cloud from generation to deposition. Turbulence is the principal mechanism governing cloud growth (dilution); wind is the principal transport agent. The necessary data on these phenomena will be derived from either of the following:
 - (a) Observation of rate of growth and movement of vapor trails deployed from rockets.
 - (b) Reduction of data obtained from accelerometer-instrumented constant pressure balloons.
 - (c) Observation of the movement of free floating balloons.
- (2) Test the predictive accuracy of the analytical model by comparing predicted with the observed results of an experiment. In this experiment it is proposed to:
 - (a) Inject into the atmosphere, at a specific altitude, latitude and time, a known quantity of readily identifiable material of known particle size distribution.
 - (b) Subsequently, collect samples and compare the observed concentrations at the sampling sites with those predicted for them.
 - (c) Correct the analytical model as indicated by discrepancies revealed. (Concentration data from this test will also provide an alternate means of determining diffusion coefficients and transport vectors.)

- (3) In the event that no adequate analytical model is derived, to proceed with the following alternate experiment:
- (a) Injection into the atmosphere at three altitudes, at each of three latitudes, in each of four seasons, known quantities of readily identifiable, different materials, in known particle size distributions.
 - (b) Frequent collection of samples of these materials at many locations and throughout the feasible sampling altitude range.
 - (c) Analysis of collected samples to determine concentrations of the different test materials as a function of particle size.
 - (d) Correlation of particle size, spatial origin, injected quantity and injection time with concentration measured at time and point of collection.
 - (e) Development of an empirical model predictive of the fate of burnup debris.

Examination of these test programs has led to the conclusion that they are feasible. This conclusion is supported by the documented engineering effort contributed, without cost to the program, by several industrial concerns competent in the following fields: test material preparation, sample collector design and manufacture, sample collection via ballooning and sample analysis.

INTRODUCTION

The Upper Atmosphere Re-entry Study was conceived for the following purposes:

- (1) To evaluate methods of determining the fate of radioactive fuel contained in space auxiliary power packages.
- (2) Recommend an experimental program to determine the size of particulate resulting on re-entry burnup of radioactive fuel.
- (3) Recommend an experimental program to determine the dispersion in the atmosphere of the by-products of nuclear fuel burnup.

This report presents in detail the work carried out in all phases of the study. The bases for, and implications of, a study of this nature have been discussed in Volume I. The paragraphs which follow are so ordered that they describe in brief the information to be found in Chapters II through IV of Volume II.

The feasibilities of alternate methods of determining resultant particle size after burnup could not be established without some knowledge of the particulate expected. To provide information about the radioisotope fuel particulate, the effects of the ablation process on plutonium were analytically evaluated. Results of this analysis, parametric in initial particle size and re-entry conditions, appear in Chapter II.

Present knowledge of atmospheric motion--bulk air transport, mean winds, turbulence, transequatorial exchange, etc.--is somewhat limited. Before experiments to ascertain the fate of a particulate cloud could be defined, the restrictions imposed on such experiments by the lack of knowledge had to be evaluated. Chapter III presents the study which explored the applicability of available information.

Chapter IV delineates the tests essential to acquiring the data necessary to meet Study Objectives (2) and (3) and reports on additional studies performed to specify test procedures, facilities and hardware required. Test feasibility investigations are documented, including supportive evidence solicited from industrial organizations in such specialty fields as materials preparation, sample collector manufacture, sample collection by balloon, and sample analysis.

Martin Marietta Corporation acknowledges in Appendix A the assistance rendered by representatives of nonprofit institutions, industry and Government in the preparation of this report.

II. PARTICULATE CHARACTERISTICS STUDIES

The goal of these studies was identification of the experimental programs necessary to determining the particulate characteristics of radioactive fuel residue resulting from re-entry burnup of radioisotope fueled generators. The factors pertinent to these experiments were revealed by the effort spent in predicting analytically the consequences of actual re-entry to a fuel. Initial effort was directed to the fuel ablation process; parameteric results were sought. This effort was followed by analysis of the immediate post-re-entry history of fuel vapors.

A. THE PARTICLE ABLATION STUDY

The scope of this study is set prior to reporting the results of the ablation process analysis. The analysis is then presented in two sections: first, a section which treats of a fuel particle initially in free molecular flow, and a second section dealing with those particles in the slip or transition flow regime. Finally, the analytical techniques used are compared critically with other methods which might have been applied.

1. Scope of the Study

In theory, there was an option for selection of the fuel which would be subjected to the re-entry heating analysis. Among fuels proposed for space system use are:

- (1) Plutonium-238 metal (Pu-238).
- (2) Curium-242 oxide ($\text{Cm}_2^{242}\text{O}_3$).
- (3) Polonium-210 metal (Po-210).
- (4) Strontium-90 metatitanate ($\text{Sr}^{90}\text{TiO}_3$).
- (5) Cerium oxide with silicon carbide ($\text{Ce}^{144}\text{O}_2 + \text{SiC}$).

Lack of high temperature thermodynamic property data for Cm_2O_3 , SrTiO_3 and $\text{CeO}_2 + \text{SiC}$ eliminated these fuels from consideration.

Since Po would be a gas under conditions which would release it to the atmosphere, its particle size was obvious. Plutonium, the only choice remaining, was the fuel selected for analysis. Reference 3 indicates that it will be a liquid, at approximately 3000° R, at the time of fuel capsule rupture by re-entry heating.

Altitudes selected as parameters for the study were 100,000, 150,000 and 200,000 feet. A sufficient number of actual re-entries has been observed, and trajectory analyses have been run, to show that if the only mechanism of fuel release is re-entry heating, the altitude range in which it can occur (i.e., in which sufficient heating can be realized) extends from 100,000 to 200,000 feet.

Since re-entry heating is manifest in a change in the kinetic energy of the re-entering fuel, the velocity of the fuel at the instant of exposure must also be a determinant of final particle size. Velocities selected as parameters for the study were 3000, 5000 and 8000 meters per second (mps). The last, 8000 mps, is high, near orbital speed, but it sets a certain upper limit to heating rates. The lowest velocity, 3000 mps, was derived by an argument similar to that employed for the lowest altitude; i.e., if the fuel element has not been penetrated by the time velocity has been reduced to this value, peak heating will have been passed and the fuel will never be exposed.

Setting limits on the range of initial particle sizes to be examined presented a more difficult problem. Qualitatively, it can be stated that if the fuel element is penetrated locally, spewing of liquid plutonium will occur because of the pressure differential existing. Depending upon the size of the penetration, the spewing will be characterized by particles of different sizes. A minimum size of 0.01 micron was assumed. The maximum size was predicated on an instantaneous general rupture of the fuel element, which would expose a large molten mass of plutonium. The maximum particle size resulting was determined by the method of Öpik (Ref. 4). He states that:

$$r_b = 1.9 r_s = 7.6 S/P_s \quad (1)$$

where

r_b = droplet radius above which breakup must occur because drag force exceeds surface tension

r_s = stable radius (cm)

S = surface tension (dynes/cm, 500 for plutonium)

P_s = drag per unit cross section (dynes/cm²) = ρv^2

ρ = air density at exposure altitude ($\text{gm-sec}^2/\text{cm}^4$)

v = velocity at exposure (cm/sec).

When the radius of the liquid droplet exceeds r_s , but is less than $1.9 r_s$, the particle flattens under aerodynamic drag into an oblate spheroid. When the radius exceeds $1.9 r_s$, the droplet breaks up into fragments of radius $1.2 r_s$. For plutonium, the maximum possible drop size is thus 1370 microns radius. The range of particle radii to be examined was therefore set as 0.01 to 1370 microns.

The span of particle sizes to be studied led to investigation of the flow regimes in which heating would take place, since mutually different heating rates are evidenced in free molecular flow, transition and slip flow, and continuum (laminar and turbulent) flow. In Ref. 5, free molecular flow is defined to exist for Knudsen numbers greater than 10; however, a Knudsen number of one was assumed to divide free molecular flow from the slip flow regime. Little difference was evidenced in the mass loss calculated by the methods applicable to the respective regimes for particle sizes so defined. Knudsen number is defined as

$$K_n = \frac{\text{molecular mean free path}}{\text{body diameter}}$$

In Fig. 1, molecular mean free path is shown as a function of altitude. This relation indicates that particles up to 253 microns in diameter must be analyzed under free molecular flow conditions. Machine solutions were actually determined for particle radii of 0.01, 0.1, 1.0, 2.0, 10.0, 20.0 and 100.0 microns.

Transition to the slip flow regime occurs at $K_n \approx 1$; slip flow exists to $K_n \approx 0.01$. This range includes the largest plutonium particle size to be examined, 1370 micron radius. Because of molecular free path dependency on altitude, it was necessary to encompass in the slip flow study a particle range of 2 to 1370 microns.

2. Plutonium Ablation in Free Molecular Flow

This section presents the derivation of equations used, the evaluation of constants required in their solution, and the parametric results obtained.

Fig. 1. Molecular Mean Free Path Versus Altitude

a. Derivation of equations

The complete energy balance which determines the effects of re-entry on a particle is

$$\frac{dE}{dt} = \frac{dE_a}{dt} + \frac{dE_c}{dt} - \frac{dE_r}{dt} + \frac{dE_d}{dt} \quad (2)$$

where

E = total energy of the particle at time t

E_a = energy input resulting from the effects of aerodynamic drag

E_c = energy input resulting from oxidation (combustion) of the fuel

E_r = energy loss resulting from radiation of heat into space

E_d = energy input resulting from decay of the fuel.

The equations used, descriptive of the energy input due to aerodynamic drag, E_a , were those developed by Whipple and Öpik (Refs. 4, 6 and 44) in their studies of meteorite ablation. It was later shown that the results obtained by this method were comparable with results which would have been obtained if the methods of Detra, Kemp and Riddel (Ref. 7) had been adopted.

Whipple and Öpik give the following equations for determining mass loss caused by heating in free molecular flow re-entry:

$$\text{Drag force} = \frac{mdv}{dt} = -\rho_a \sigma v^2 \quad (3)$$

$$\text{Rate of mass loss} = \frac{dm}{dt} = -\gamma \sigma \rho_a v^3 / 2h \quad (4)$$

where

ρ_a = atmospheric density, (kgm/m³)

σ = effective cross section (m²)

v = particle velocity (mps)

γ = fraction of kinetic energy transferred to an air molecule in unshielded impact

h = heat input required to vaporize a unit mass of material (joules/kgm).

These were further developed to the forms

$$\frac{dv}{dt} = - \frac{\rho_a \pi r^2 v^2}{\frac{4}{3} \pi r^3 \rho_s} \quad (5)$$

and

$$\begin{aligned} \frac{d\left(\frac{4}{3} \pi r^3 \rho_s\right)}{dt} &= - \frac{\gamma \sigma \rho_a v^3}{2h} \\ (4\pi r^2 \rho_s) \frac{dr}{dt} &= - \frac{\gamma \pi r^2 \rho_a v^3}{2h} \\ \frac{dr}{dt} &= - \frac{\gamma \rho_a v^3}{8\rho_s h} \end{aligned} \quad (6)$$

where

r = spherical particle of radius (meters)

ρ_s = particle density (kgm/m³).

Equations (5) and (6) are nonlinear differential equations to be solved by the Runge-Kutta method. Equation (6) gives the particle radius reduction caused by the transfer of kinetic energy to heat, the rate of which is defined by the velocity change reflected in Eq (5).

Equation (1) can be rewritten to describe the mass loss effected by all energy sources acting at re-entry:

$$\frac{dm}{dt} = - \frac{1}{h} \left[\frac{\gamma \sigma \rho_a v^3}{2} + k_1 k_2 \rho_a \sigma v - k_3 S + k_4 m \right] \quad (7)$$

where

k_1 = heat of combustion (joules/kgm)

k_2 = percent mass of oxygen per unit mass of air multiplied by the fraction of oxygen molecules reacting with the fuel

$k_3 = \epsilon \beta (T_s^4 - T_a^4)$ (watts/m²)

ϵ = emissivity

β = Stefan-Boltzmann constant ($\text{watts/m}^2 - ^\circ\text{K}^4$)

T_s = temperature of particle ($^\circ\text{K}$)

T_a = ambient temperature ($^\circ\text{K}$)

S = particle surface area (m^2)

k_4 = decay heat generated per unit mass (watts/kgm)

m = particle mass (kgm).

Particle radius reduction effected by mass loss can be expressed by rewriting Eq (7):

$$\frac{dr}{dt} = -\frac{1}{h} \left[\frac{\gamma \rho_a v^3}{8 \rho_s} + \frac{k_1 k_2 \rho_a v}{4 \rho_s} - \frac{k_3}{\rho_s} + \frac{k_4 r}{3} \right] \quad (8)$$

Equations (5) and (8) were the basic differential equations programmed for parametric solution; the parameters varied and values used were those indicated in Chapter I.

b. Evaluation of constants

The fraction of the dissipated re-entry kinetic energy that is transferred into the fuel particle as heat (γ) was assumed equal to one. The fuel particle was postulated stationary, the air molecules impacting it at re-entry velocity. To estimate the value $\gamma = 1$ required the assumption of a completely inelastic collision; i.e., all impacting air molecules are brought to rest, and their total kinetic energy is absorbed by the fuel, then evidenced by vapor formation. Öpik terms this phenomenon the "energy trap" process. Öpik and other researchers, realizing that a completely inelastic collision will not always occur, suggest a wide range of values for γ , all less than one. The value appears dependent on the material impacted, each material being susceptible to a different degree to surface penetration and subsequent bringing to rest of the air molecule impinging upon it. Since the correct value for plutonium, the fuel under investigation, was not known, a value of $\gamma = 1$ was selected to reflect the maximum heating possible.

The heat to vaporize a unit mass of plutonium, h , was determined as follows:

$$h = c_p (T_v - T_r) + h_v = 1.749 \times 10^6 \text{ joules/kgm}$$

where

c_p = specific heat at constant pressure = 183 joules/kgm - °C

T_r = plutonium temperature at rupture of the fuel element = 1394° C

T_v = plutonium vaporization temperature = 3235° C

h_v = plutonium heat of vaporization = 1.413×10^6 joules/kgm

The plutonium heat of combustion, k_1 , was not available as a measured experimental value. It was estimated by two independent methods utilizing the behavior of materials with similar properties. In the first method, the heats of combustion for lead and bismuth were extrapolated to that of plutonium as a function of their respective atomic weights. A value of 4.81×10^6 joules/kgm was estimated by this method. In the second instance, Pauling's formula for approximate heat of formation of a gas molecule containing a single bond was used. Plutonium electronegativity for use in the equation was inferred from that of materials of similar period in the periodic tables. A value of 2.14×10^6 joules/kgm was obtained. Since the reliability of both values was in doubt, the estimate resulting in the higher heating was used, consistent with the policy followed in estimating the fraction of kinetic energy transferred into the fuel particle (γ).

Since, on the basis of weight, air is composed of 23.2 percent oxygen (Ref. 51), k_2 (defined under Eq (7)) was assumed equal to 0.232.

The line of reasoning is as follows: if $\gamma = 1$, all molecules are brought to rest in the plutonium; therefore, all oxygen is brought to rest. Plutonium is pyrophoric; it is reasonable to assume that all oxygen reacts.

The radiative heat loss coefficient, k_3 , is defined as follows:

$$k_3 = \epsilon \beta (T_s^4 - T_a^4)$$

where

$$\epsilon = 0.8 \text{ (assumed)}$$

$$\beta = 5.67 \times 10^{-8} \text{ watts/m}^2 \cdot \text{°K}^4$$

$$T_s = 1667 \text{ °K}$$

$$T_a = 278 \text{ °K}$$

$$k_3 = 3.55 \times 10^5 \text{ watts/m}^2$$

The radioactive decay heat, k_4 , is a function of the isotope in use; for plutonium, it is 5.8×10^2 watts/kgm.

c. Analysis and results

Equations (5) and (8) were programmed, supplied with the constants derived in Topic 2, Paragraph b, Section A and solved for the various parameter combinations given in Table 1. Figure 1 was used to determine the maximum particle radius to be considered at each altitude. From Fig. 1:

<u>Altitude (ft)</u>	<u>Mean Free Path (microns)</u>	<u>Maximum Particle Radius Studied (microns)</u>
200,000	253	100
150,000	38	20
100,000	4	2

TABLE 1

Combinations of Parameters Used for Programmed Solution

<u>Altitude of Fuel Exposure (ft)</u>	<u>Initial Fuel Velocity, v (mps)</u>	<u>Initial Particle Radius, r_0^* (microns)</u>
200,000	8000	1.0
		2.0
		10.0
		20.0
		100.0
	5000	1.0
		2.0
		10.0
		20.0
		100.0

*Mass reduction results obtained for 1-micron particles (see Table 2) indicated no need for presentation of heating effects on 0.01- and 0.1-micron particles as had been intended (see Topic 1 of Section A).

TABLE 1 (continued)

<u>Altitude of Fuel Exposure (ft)</u>	<u>Initial Fuel Velocity, v (mps)</u>	<u>Initial Particle Radius, r_0^* (microns)</u>
200,000	3000	1.0
		2.0
		10.0
		20.0
		100.0
150,000	8000	1.0
		2.0
		10.0
		20.0
		100.0
	5000	1.0
		2.0
		10.0
		20.0
		100.0
100,000	3000	1.0
		2.0
		10.0
		20.0
		100.0
	8000	1.0
		2.0
		10.0
		20.0
		100.0
	5000	1.0
		2.0
		10.0
		20.0
		100.0
	3000	1.0
		2.0
		10.0
		20.0
		100.0

*Mass reduction results obtained for 1-micron particles (see Table 2) indicated no need for presentation of heating effects on 0.01- and 0.1-micron particles as had been intended (see Topic 1 of Section A).

The results of several of the machine runs are shown in Figs. 2 through 5 and Table 2. Significant among them are these:

- (1) Plutonium particles in free molecular flow at 100,000 feet or less are almost totally consumed if traveling at a velocity of 3000 mps or greater.
- (2) Plutonium particles, regardless of initial size or exposure altitude, are almost totally consumed (99.98% or more) if exposure velocity is 8000 mps or greater.
- (3) For plutonium, a heat balance on re-entry is generally achieved in less than one second; i.e., the ablation process is completed about one second after initial exposure, except for that contribution made to the process by oxidation.

Table 2 presents the percent mass reduction occurring in each situation examined, calculated from the following equation:

$$\text{Percent reduction in mass} = \left(1 - \frac{r_f^3}{r_0^3} \right) 100$$

where

r_0 = initial particle radius

r_f = final particle radius from IBM 7090 run.

Figures 2 through 5 show particle radius as a function of time after initial exposure, parametric in initial velocity, initial particle radius and initial altitude. Contrary to the actual mechanics of re-entry, the particles were assumed to be in horizontal--i.e., constant altitude--flight. Varying altitude by assuming varied re-entry angles would have contributed little information about the particle ablation process, while making more complex the equations to be solved. The ablation process was found to be complete within about 200 meters of the exposure point. The variation in atmospheric density over such a distance is small, regardless of re-entry angle. Figure 2 exhibits the effects of including and of excluding the oxidation (combustion) term in the ablation equation; Figs. 3 through 5 show the results with the combustion term included.

Fig. 2. Ablation of Plutonium Particle

Fig. 3. Ablation of Plutonium Particle, Including Combustion Heating

Fig. 4. Ablation of Plutonium Particle, Including Combustion Heating

LEGEND:
 ----- 200,000-ft altitude
 ————— 150,000-ft altitude

NOTE:
 Free molecular flow particle ablation--2 microns
 v_0 = initial velocity
 v_t = velocity when drag heating is negligible

Fig. 5. Ablation of Plutonium Particle, Including Combustion Heating

TABLE 2
Plutonium Particle Mass Reduction in Free Molecular Flow

<u>Altitude of Fuel Exposure (ft)</u>	<u>Initial Fuel Velocity, v (mps)</u>	<u>Initial Particle Radius, r_0 (microns)</u>	<u>Mass Reduction (%)</u>
200,000	8000	1.0	100
		2.0	100
		10.0	99.99
		20.0	99.99
		100.0	99.98
	5000	1.0	93.8
		2.0	95.89
		10.0	94.51
		20.0	94.29
		100.0	93.85
	3000	1.0	28.3
		2.0	48.8
		10.0	44.86
		20.0	39.66
		100.0	41.57
150,000	8000	1.0	100
		2.0	100
		10.0	100
		20.0	100
	5000	1.0	100
		2.0	100
		10.0	99.07
		20.0	98.86
	3000	1.0	74.6
		2.0	87.5
		10.0	82.44
		20.0	81.96

TABLE 2 (continued)

Altitude of Fuel Exposure (ft)	Initial Fuel Velocity, v (mps)	Initial Particle Radius, r_0 (microns)	Mass Reduction (%)
100,000	8000	1.0	100
		2.0	100
	5000	1.0	100
		2.0	100
	3000	1.0	100
		2.0	100

3. Plutonium Ablation in Transition and Slip Flow

Probstein, in Ref. 8, states that the heat transfer rate in the near-free molecular flow regime can be expressed as a fraction of that occurring in the free molecular flow regime:

$$\frac{\dot{q}}{\dot{q}_{fm}} < 1 .$$

Öpik's theory for giving an exact value to this expression is presented in the succeeding paragraphs. It was used so that the equations developed for free molecular flow would retain their usefulness into the transition and slip flow regimes. The results obtained by the method of Öpik are compared with those which would be obtained if the methods of other investigators were adopted.

a. Analysis and results of the Öpik theory

Öpik's treatment of the transition and slip flow regimes is encompassed in his analysis of the formation of an air cap in front of a spherical particle. Again, it is predicated on existence of an "energy trap"; however, in this instance, the air molecule is incident on the air cap rather than on the fuel particle and is reflected with negligible velocity; Öpik terms this phenomenon an air cap-air molecule accommodation of one. This permits treatment of the kinetic thickness of the air cap as an insulation layer acting to reduce γ , the energy transferred per unshielded air molecule impact, to an effective γ_c for shielded impact. Then γ_c , rather than γ , is used in the free molecular flow equations to determine the percent mass loss in transition and slip flow. The function γ_c is essentially a $\dot{q}/\dot{q}_{fm}$ for the kinetic energy transfer term of Eq (8).

It is dependent on the air cap kinetic thickness, d_c , evidenced by a particle in slip flow. This thickness is defined as:

$$d_c = \frac{0.75 r_0 \rho_a}{\lambda \rho} \quad (9)$$

where

r_0 = initial particle radius under varying re-entry conditions (cm)

ρ_a = air density at altitude of re-entry (gm/cm³)

$\lambda \rho$ = constant derived from Fig. 6 at 1/2 the particle velocity.

It can be seen that d_c is dependent on r_0 ; the values of interest for r_0 are:

- (1) The maximum stable liquid particle radius that can exist on re-entry.
- (2) The particle sizes into which a single particle slightly larger than that described in Item (1) will divide. The maximum stable sizes are defined by Eq (1), rewritten below:

$$r_b = 1.9 r_s = 7.6 S/P_s \quad (1)$$

The particle size resultant on breakup of a particle slightly larger than r_b is

$$r_d = 1.2 r_s \quad (10)$$

Table 3 calculates and presents in Columns 11 and 12 r_0 as r_b , and r_d as defined by Eqs (1) and (10). Mean free path is noted in Column 13 in order that the values presented in Columns 11 and 12 can be compared and eliminated from consideration if they are less than the mean free path--i.e., if they are in free molecular rather than slip flow. The values of r_0 are used in Table 4 to calculate d_c , the kinetic thickness of the air cap. Columns 10 and 15 of Table 4 present the values of γ_c derived from Fig. 7 as a function of d_c . A tabular presentation of particulate mass loss resulting from transition and slip flow re-entry appears in Table 5. Significant results are these:

Fig. 6. Air Cap Kinetic Thickness Evaluation Factor ($\lambda \rho$) Slip Flow (Ref. 4)

TABLE 3
Particle Sizes of Interest in Transition and Slip Flow
Calculated from Eqs (1) and (10)

Material (1)	Surface Tension, S (dynes/cm) (2)	4(2) (dynes/cm) (3)	Altitude, h (ft x 10 ³) (4)	Density, ρ (gm/cm ³) (5)	Velocity, v (mps) (6)	(6) ² (cm ² /sec ²) (7)	(5)(7) (dynes/cm ²) (8)	(3)/(8) r _s (cm) (9)	(9)10 ⁴ r _s (microns) (10)	1.9 (10) r _b (microns) (11)	1.2 (10) r _d (microns) (12)	Mean Free Path (microns) (13)
<div>↑ Plutonium ↓</div>	500	2000	200	3.09 x 10 ⁻⁷	3000	9 x 10 ¹⁰	2.78 x 10 ⁴	7.2 x 10 ⁻²	720	1370	863	253
	500	2000	200	3.09 x 10 ⁻⁷	5000	2.5 x 10 ¹¹	7.72 x 10 ⁴	2.59 x 10 ⁻²	259	492	311	253
	500	2000	200	3.09 x 10 ⁻⁷	8000	6.4 x 10 ¹¹	19.8 x 10 ⁴	1.01 x 10 ⁻²	101	192	121	253
	500	2000	150	1.81 x 10 ⁻⁶	3000	9 x 10 ¹⁰	1.63 x 10 ⁵	12.3 x 10 ⁻³	123	234	148	38
	500	2000	150	1.81 x 10 ⁻⁶	5000	2.5 x 10 ¹¹	4.52 x 10 ⁵	4.42 x 10 ⁻³	44	84	53	38
	500	2000	150	1.81 x 10 ⁻⁶	8000	6.4 x 10 ¹¹	11.6 x 10 ⁵	1.72 x 10 ⁻³	17	32	20	38
	500	2000	100	1.64 x 10 ⁻⁶	3000	9 x 10 ¹⁰	1.48 x 10 ⁶	13.5 x 10 ⁻⁴	14	27	17	4
	500	2000	100	1.64 x 10 ⁻⁶	5000	2.5 x 10 ¹¹	4.1 x 10 ⁶	4.88 x 10 ⁻⁴	5	10	6	4
	500	2000	100	1.64 x 10 ⁻⁶	8000	6.4 x 10 ¹¹	10.5 x 10 ⁶	1.9 x 10 ⁻⁴	2	4	2	4

TABLE 4

Determination of Kinetic Thickness of Air Cap (d_c) from Eq. (9) and Fig. 6;
Tabulation of Kinetic Energy Transfer Coefficient, Shielded Impact, from Fig. 7

Material (1)	Altitude, h (ft x 10 ³) (2)	Density, ρ (gm/cm ³) (3)	Velocity, v (mps) (4)	λρ at (4) / 2 (Fig. 6) (gm/cm ²) (5)	r _o = r _b (Table 3) (cm) (6)	0.75 (6) (cm) (7)	(7) (3) (gm/cm ²) (8)	(8) / (5) d _c (9)	γ _c (10)	r _o = r _d (Table 3) (cm) (11)	0.75 x (11) (cm) (12)	(12) x (3) (gm/cm ²) (13)	(13) / (5) d _c (14)	γ _c (15)
Plutonium	200	3.09 x 10 ⁻⁷	3000	2.4 x 10 ⁻⁸	0.137	0.103	3.18 x 10 ⁻⁸	1.325	0.703	0.0863	0.0647	2.0 x 10 ⁻⁸	0.833	0.802
	200	3.09 x 10 ⁻⁷	5000	3.3 x 10 ⁻⁸	0.0492	0.0369	1.14 x 10 ⁻⁸	0.346	0.915	0.0311	0.0233	0.72 x 10 ⁻⁸	0.218	0.945
	200	3.09 x 10 ⁻⁷	8000	4.37 x 10 ⁻⁸	0.0192	0.0144	0.445 x 10 ⁻⁸	0.102	FMF*	0.0121	0.0091	0.281 x 10 ⁻⁸	0.064	FMF*
	150	1.81 x 10 ⁻⁶	3000	2.4 x 10 ⁻⁸	0.0234	0.0176	3.18 x 10 ⁻⁸	1.325	0.703	0.0148	0.0111	2.01 x 10 ⁻⁸	0.837	0.802
	150	1.81 x 10 ⁻⁶	5000	3.3 x 10 ⁻⁸	0.0084	0.0063	1.14 x 10 ⁻⁸	0.346	0.915	0.0053	0.0040	0.724 x 10 ⁻⁸	0.219	0.945
	150	1.81 x 10 ⁻⁶	8000	4.37 x 10 ⁻⁸	0.0032	0.0024	0.445 x 10 ⁻⁸	0.102	FMF*	0.0020	0.0015	0.272 x 10 ⁻⁸	0.062	FMF*
	100	1.64 x 10 ⁻⁵	3000	2.4 x 10 ⁻⁸	0.0027	0.0020	3.18 x 10 ⁻⁸	1.325	0.697	0.0017	0.0013	2.13 x 10 ⁻⁸	0.888	0.79
	100	1.64 x 10 ⁻⁵	5000	3.3 x 10 ⁻⁸	0.0010	0.00075	1.14 x 10 ⁻⁸	0.346	0.907	0.0006	0.00045	0.737 x 10 ⁻⁸	0.223	0.945
	100	1.64 x 10 ⁻⁵	8000	4.37 x 10 ⁻⁸	0.0004	0.0003	0.445 x 10 ⁻⁸	0.102	0.975	0.0002	0.00015	0.243 x 10 ⁻⁸	0.056	FMF*

*FMF = Free molecular flow.

Fig. 7. Kinetic Energy Transfer Coefficient, Slip Flow (Ref. 4)

TABLE 5

Plutonium Particle Mass Reduction in Transition and Slip Flow

Altitude of Fuel Exposure (ft)	Initial Fuel Velocity, v (mps)	Initial Particle Radius, r_0 (microns)	Mass Reduction (%)
200,000	8000	At this velocity and altitude, all liquid plutonium is divided by drag forces into droplets which re-enter in free molecular flow.	
200,000	5000	311	75.00
200,000	5000	492	59.17
200,000	3000	863	7.88
200,000	3000	1370	4.74
150,000	8000	At this velocity and altitude, all liquid plutonium is divided by drag forces into droplets which re-enter in free molecular flow.	
150,000	5000	53	98.53
150,000	5000	84	98.20
150,000	3000	148	74.40
150,000	3000	234	66.20
100,000	8000	4	~100.00
100,000	5000	6	~100.00
100,000	5000	10	~100.00
100,000	3000	17	95.60
100,000	3000	27	93.24

- (1) Plutonium, when exposed as a liquid to re-entry at 8000 mps, will break up into droplets which proceed to re-enter in free molecular flow; at this velocity, such breakup will occur at all altitudes above 100,000 feet.
- (2) All plutonium, regardless of exposure altitude, is 100% consumed if the re-entry velocity is 8000 mps or greater.
- (3) If the plutonium is exposed at 100,000 feet or less, at a velocity of 5000 mps or greater, it will be 100% vaporized.

Care has been taken to specify plutonium in each instance above, because the results differ widely from those that would be evidenced by another fuel.

Some particulate initially in transition or slip flow will, after some mass loss, enter the free molecular flow regime and be subject to its characteristic heating rates. No attempt has been made here to reflect this although the IBM 7090 could have been programmed to change from γ_c to γ when a predetermined particle radius was reached. In this instance, however, it was assumed that velocity decay due to drag had been such as to make insignificant the change in mass reduction to be reflected.

b. Applicability of alternate theories

Three additional approaches to approximating the aerodynamic heating of a body in the near-free molecular flow regime were investigated. The first two, those of Probstein-Willis and Hamerling-Kivel, considered only the so-called first collisions, those of an air molecule striking, in this instance, a fuel molecule in the vapor shield of the re-entering fuel mass. The third approach studied utilized the empirical relations, derived by Reeves and Van Camp, of the flow parameters Mach, Nusselt, Reynolds and Prandtl numbers.

Method 1. Probstein, Ref. 8, interpreted the work of Willis, Ref. 9, for the heating resulting from flow of a rarefied gas past a highly cooled sphere. This system, of itself, violates the re-entry condition of hot liquid plutonium; nevertheless, a determination of $\dot{q}/\dot{q}_{fm}$ was carried out as follows:

$$\frac{\dot{q}}{\dot{q}_{fm}} = 1 - \left(\frac{R_b}{\lambda_\sigma} \right) \left[0.53 + 3.2 \frac{\bar{c}_b}{u} \right] \quad (11)$$

where

$$\lambda_{\sigma} = \bar{c}_b / n_{\infty} u \pi \sigma^2 = \text{mean free path at body}$$

$$\bar{c}_b = (8kT/\pi m)^{1/2} = \text{mean molecular velocity}$$

$$\dot{q} = \text{stagnation point heat transfer rate}$$

$$R_b = \text{radius of sphere (fuel droplet)}$$

$$u = \text{macroscopic free stream velocity}$$

$$n = \text{number of incident molecules per unit volume of gas}$$

$$\sigma = \text{effective molecular diameter}$$

$$k = \text{Boltzmann constant} = 1.38 \times 10^{-6} \text{ gm-cm}^2/\text{°K-sec}^2$$

$$T = \text{absolute temperature (°K)}$$

$$m = \text{mass of molecule}$$

and subscripts

$$b \quad \text{refers to conditions at body surface}$$

$$fm \quad \text{refers to free molecular flow value}$$

$$\infty \quad \text{refers to free stream value.}$$

Applicability was limited by the following restrictions:

$$\frac{u}{\bar{c}_b} > 2 \quad \text{and} \quad \frac{R_b}{\lambda_{\sigma}} < \frac{1}{2} \text{ or } \frac{1}{3}$$

The trial evaluation of $\dot{q}/\dot{q}_{fm}$ was conducted at an altitude of 200,000 feet and a re-entry velocity of 3000 mps. The values of $\bar{c}_b$ and λ_{σ} were determined with R_b selected to conform to $\frac{R_b}{\lambda_{\sigma}} = \frac{1}{3}$.

$$\bar{c}_b = \left[\frac{8 (1.38 \times 10^{-16} \text{ gm-cm}^2/\text{°K-sec}^2) 1667^\circ \text{ K}}{\pi (28.96 \text{ gm/mole air}) (1/6.025 \times 10^{23} \text{ molecules/mole})} \right]^{1/2}$$

$$= 1100 \text{ mps}$$

$$\therefore u/\bar{c}_b = 3000/1100 = 2.72 > 2$$

and

$$\lambda_\sigma = \frac{1100 \text{ mps}}{(6.43 \times 10^{15} \text{ moles/cm}^3) (3000 \text{ mps}) \pi (3.72 \times 10^{-8})^2 \text{ cm}^2}$$

$$= 131 \text{ microns}$$

$$\therefore R_b = 43 \text{ microns, since } R_b/\lambda_\sigma = 1/3$$

Substituting in Eq (12) gives

$$\dot{q}/\dot{q}_{fm} = 1 - 0.33 [0.53 + 3.2 (1100)/3000] = 0.44$$

This value is decidedly below the value predicted by Öpfik, where $\dot{q}/\dot{q}_{fm} \approx 1$; it would appear that this results from violation of the highly cooled body restraint on use of the method; a lower temperature would have lowered $\bar{c}_b$, therefore lowering λ_σ . This would have forced selection of a smaller R_b to meet the conditions that $\frac{R_b}{\lambda_\sigma} < \frac{1}{3}$ or $\frac{1}{2}$.

Reducing R_b would move the conditions to be evaluated very close to, if not into, the free molecular flow regime where $K_n \geq 10$. An inherent weakness of the Probstein approach is that it is applicable only in the very near free flow regime, not in the slip flow heating regime.

Method 2. Hamerling-Kivel, Ref. 10, present the following expression for evaluation of aerodynamic heating energy in the transition and slip flow regimes:

$$E/E_{fm} = [1 - (\bar{v}_e/v_0)^2] [1 - 0.2 (v_0/v_e) R_b \rho_0 S_{e0}] \quad (12)$$

where

v_0 = macroscopic free stream velocity = u of Method 1

$\bar{v}_e$ = mean emitted particle velocity = $\bar{c}_b$ of Method 1

ρ_0 = ambient number density = n_∞ of Method 1

S_{e0} = collision cross section = $\pi \sigma^2$ of Method 1.

Substituting into Eq (12) similar re-entry conditions to those used in Method 1 results in the expression

$$\frac{E}{E_{fm}} = \left[1 - \left(\frac{1100 \text{ mps}}{3000 \text{ mps}} \right)^2 \right] \left[1 - 0.2 \frac{(3000 \text{ mps}) (43 \times 10^{-4} \text{ cm})}{(1100 \text{ mps})} \right. \\ \left. (6.43 \times 10^{15} \text{ molecules/cm}^3) \pi (3.72 \times 10^{-8})^2 \text{ cm}^2 \right] = 0.86$$

This result appears reasonable. If R_b were increased, reflecting a further movement from the transition to the slip flow regime, E/E_{fm} , as expected, would decrease. A computation for the conditions presented in Table 4, Column 13, Row 1, would give a heating rate of $0.75 \dot{q}_{fm}$, as opposed to Öpik's value of $0.802 \dot{q}_{fm}$. This is considered good correlation.

Method 3. Reeves and Van Camp in Ref. 11 used the following relations of flow parameters in determining the aerodynamic heating due to moderately rarefied gas flow over a sphere:

$$\frac{Nu}{\sqrt{Re_2}} = \frac{0.672 M^{0.2} + 0.049 \alpha \sqrt{\gamma} \left(\frac{M^{3/2}}{\sqrt{Re_2}} \right) \left(\frac{\gamma+1}{\gamma} \right) Pr}{1 + 1.252 \sqrt{\gamma} \left(\frac{M}{\sqrt{Re_2}} \right)} \quad (13)$$

where

Nu = Nusselt number

Re_2 = Reynolds number evaluated behind the normal shock

Pr = Prandtl number

γ = ratio for air of specific heat at constant pressure to that at constant volume

α = accommodation coefficient = 0.9

M = free stream Mach number.

An expression for the heating rate, derived from Eq (13), is

$$\dot{q} = Nu (h_z - h_w) \bar{\mu} / DPr$$

where

h_z = enthalpy behind the normal shock

h_w = enthalpy at wall of re-entry body

$\bar{\mu}$ = mean dynamic viscosity

D = diameter of re-entering body.

Evaluated for conditions similar to those used in Methods 1 and 2,

$$\dot{q} = 39.8 \text{ Btu/ft}^2\text{-sec}$$

$$\dot{q}_{fm} = 376 \text{ Btu/ft}^2\text{-sec}$$

$$\frac{\dot{q}}{\dot{q}_{fm}} = 0.106; \text{ this is considered far too low a heating ratio}$$

for a particle only slightly removed from the free molecular flow regime.

4. Conclusions on the Particle Ablation Study

It was revealed that the analytical procedures are available for prediction of the particle sizes which will result on re-entry burnup. Further, the resultant particle sizes were demonstrated to be dependent on the following:

- (1) Altitude (atmospheric density) at which fuel exposure occurs.
- (2) Velocity of fuel at exposure to re-entry heating.
- (3) Initial particle size of fuel.
- (4) Physical and chemical properties of the fuel.

The re-entry altitude and velocity at fuel exposure, as well as the initial particle size of the fuel, are predetermined, to a degree, by the design practices followed in manufacture of the fuel, fuel element, generator and powered source. The physical and chemical properties of the fuel are a function of the isotope and fuel form selected for the mission. With this number of parameters affecting final particle size, sampling of the wake of re-entering test specimens appears an illogical if not impossible means of deriving sufficient experimental data to predict, on an empirical basis, the particulate characteristics to be expected on re-entry of a specific space system. Laboratory testing to improve the predictive quality of the analytical expressions seems more reasonable. Also, the considerations which dictated that plutonium be used in the development of the analytical approach indicate a need for additional laboratory tests to determine certain properties of prospective fuels.

The conclusions reached as a result of the ablation study are these:

- (1) The predictive quality of the analytical expression for free molecular flow heating should be tested and improved as necessary.
- (2) The predictive quality of the analytical expression for transition and slip flow heating should be tested and improved as necessary.
- (3) The expression for liquid particle breakup should be tested and improved as necessary.
- (4) The physical and chemical properties required in solution of the analytical expressions should be determined experimentally for all prospective isotopes and fuel forms.
- (5) Consideration should be given the variables affecting residue characteristics when a space system using nuclear fuel is designed.
- (6) Where burnup rather than intact re-entry is an objective in design of the system, every effort should be made to have the fuel exposed to re-entry heating effects at the highest altitude and velocity possible, and in particle sizes which ensure that initial heating occurs in the free molecular flow regime; i. e., fuel particle sizes at initial exposure should be approximately one-tenth or less of the mean free path at exposure altitude.

B. STUDY OF ABLATED MATERIAL (VAPOR)

Final particle size as a function of initial particle size has been investigated in Section A. Particle size changes because of mass loss due to vaporization. This section presents a study of the fate of this vapor as it pertains to the characteristics of the particulate cloud as finally constituted. The study is presented in the sequence of events of interest: vapor re-entry and nucleation, condensation, and agglomeration to a state of size distribution equilibrium.

1. Vapor Re-entry, Nucleation-Condensation

No experimental data, and very little theory, are available in the literature on the behavior of vapor under re-entry conditions. This is true specifically as regards particulate studies. Some data are available on vapor trail radar cross sections, ionization, etc., but are not pertinent to this study. An attempt was therefore made to describe the history of vapor re-entry to uncover and identify phenomena determinant of the characteristics of particulate of vapor origin.

Two factors of importance characterize the vapor at the instant of its creation: first, its high kinetic energy relative to the local atmospheric thermal energy; and, second, its high drag relative to that of its source. The high drag, caused by increased cross-sectional area, results in formation of a vapor trail behind the vapor source. High kinetic energy is responsible for the early history of the vapor trail. (The kinetic energy of sodium vapor per unit mass at 5000 mps is equivalent to the energy represented by a 20,000° K temperature rise of the same unit mass.) As re-entry velocity of the vapor trail is dissipated, the vapor is, in effect, heated and kinetic energy assumes a thermal distribution. This high heat initially inhibits nucleation of the vapor; however, it is rapidly dissipated by radiation and expansion cooling. Presumably, conditions are soon reached where nucleation and condensation can proceed. At this point, however, several factors, at present unevaluated, may intervene and become controlling of the rate at which the processes can proceed. These factors are:

- (1) Ionization--Ions, by their number, are known to govern the rate of condensation through provision of condensation nuclei. The magnitude of the effect is not clearly understood. In addition, there are several sources of ions. They occur naturally in varying amounts in the stratosphere; they are created as a result of the radioactivity of the fuel; they are created by impact of air molecules with fuel particles and vapor; and an ionized atmospheric sheath is created on re-entry of bodies in slip or continuum flow.

- (2) Expansion--It is conceivable that expansion of the vapor cloud is so rapid that a supercooled center section will provide condensation nuclei.
- (3) Nonfuel vapors--Materials other than fuel will be vaporized on re-entry. They may condense at higher temperatures than the fuel and form condensation nuclei for it; they may also condense at lower temperatures than the fuel.

Once nuclei are present to precipitate condensation, the particle size formed is of interest. Reference 12 presents classical theory as it is applied to vapor created by atomic bomb tests. Equations for particle size distribution and average particle size are also presented in Ref. 13. In both instances, a wide variety of assumptions is required to obtain numerical solutions. They are in general agreement in predicting that most particles formed will be less than 1 micron in diameter. It is not stated whether the droplets formed solidify before significant coalescence occurs; this should be predictable from statistics of droplet concentration.

2. Agglomeration-Sedimentation, Particle Size Distribution Equilibrium

After completion of the initial processes of particle formation, the resulting debris will be dispersed in the atmosphere along the re-entry trajectory and subsequently diluted by atmospheric turbulence, thermal motion, etc. The final particle size distribution, however, is not yet known, agglomeration and sedimentation processes remaining to be accounted for. The work of S. K. Freidlander, Ref. 14, is particularly pertinent here. He states that an equilibrium state for the particle size distribution will be reached. The mechanism of creating and maintaining the equilibrium state is the balancing of sedimentation rates of larger particles with their rate of formation through agglomeration of smaller particles. (It should be noted that naturally occurring aerosols contribute to the process.) This theory is supported by experimental determinations of particulate size distributions at similar altitudes at other latitudes. It appears that a single distribution function applicable to prediction of the particle size distribution over a wide range of altitudes can be derived. This distribution function, treating particles with radii initially larger than 0.1 micron, has been derived by Freidlander through use of Smoluchowski's agglomeration rate equation (Ref. 15) and the Stokes-Cunningham sedimentation rate equation (see Chapter III, Section B). Dimensional analysis and similarity transformations produced the following solution for the upper particle size range:

$$\frac{n}{l^2} = \frac{0.2}{n_1^2} + 2n_1^4 \quad (14)$$

where

$$n = \frac{dN}{dr} \quad (15)$$

$$n_1 = r/l_1 \quad (16)$$

r = particle radius (microns)

n = concentration density per increment of particle radius (particles/cm³)

l_1 = a length scale factor, altitude dependent (microns)

N = number concentration (particles/cm³)

For use of the equations, the length scale factor, l_1 , must be evaluated without prior knowledge of the distribution. By trial and error, Freidlander was able to determine a length scale factor which made the equation fit experimental data obtained at altitudes of 23.5, 27.5 and 30.2 kilometers (Ref. 16). It is shown below that Freidlander's equations can be further manipulated to determine a value for l_1 , if N is known. Substituting Eqs (15) and (16) into (14) leads to

$$\frac{dN}{dr} = \frac{0.2 l_1^6}{(l_1^2 r^2 + 2r^4)} \quad (17)$$

A straightforward integration over the interval of particle radii represented by N gives

$$N = 0.2 l_1^4 \left[-\frac{1}{r} - \frac{\sqrt{2}}{r} \arctan \frac{\sqrt{2} r}{l_1} \right]_{r_1}^{r_2} \quad (18)$$

Solution for l_1 must yield a positive real number; if an ambiguity exists, values of l_1 for adjacent regions of different N may serve to indicate the correct root.

3. Conclusions of the Vapor Re-entry Study

As a consequence of the vapor re-entry study, three general observations are in order; conclusions in regard to necessary experimental programs are based on them. The study has revealed:

- (1) That the vapor cloud formed in the process of re-entry burnup has peculiarities of behavior not associated with relatively stationary vapors. These result from an unspecified ion concentration and the very rapid change of vapor kinetic energy to thermal energy.
- (2) That the present level of nucleation-condensation theory is inadequate to handle the peculiarities described in Item (1); further, that, at present, the influences of radioactivity and the presence of other vaporized materials in the vapor cloud cannot be properly accounted for.
- (3) That the equilibrium distribution function for particle size as a function of altitude is in a preliminary stage of development.

The conclusions based on these general observations follow:

- (1) The effects on nucleation and condensation rate of ion concentration should be experimentally determined.
- (2) The effect on nucleation and condensation rate of radioactivity should be experimentally determined.
- (3) Empirical data on the nucleation-condensation process of mixed material vapors must be generated.
- (4) The extent to which coalescence of droplets occurs prior to droplet solidification should be investigated analytically.
- (5) The physical characteristics of the agglomerate resulting from mixed material vapors should be experimentally determined; the possibility of preferential ordering of material layers should be investigated. Findings could provide an additional criterion for generator material selection.
- (6) The predictive quality of Freidlander's equilibrium size distribution function should be checked against additional experimental data; time to equilibrium should be estimated analytically under dynamic, rather than quiescent, vapor cloud conditions. (If time to equilibrium is infinite, the significance of Freidlander's findings is reduced; the fate of the particulate could be little affected by the equilibrium size distribution.)

C. RECOMMENDATIONS--PARTICULATE CHARACTERISTICS STUDIES

The programs recommended below are divided into those resulting, first, from the particle ablation study, and second, from the vapor re-entry study. Coupled with each recommended experimental program is a brief description of the testing involved.

1. Particle Ablation Study Recommendations

a. Ablation testing--droplets and small particles

This testing and analytical effort is recommended to provide and/or prove an analytical technique for prediction of final burnup particle size. Testing in a facility where re-entry velocity and atmospheric conditions are simulated is recommended. Velocity measurements and photogrammetric data should be obtained while the test particle is in flight. When flight is terminated by drag and/or sample collectors, particulate must be recovered and examined, measured, etc. Gaseous byproducts should be recovered and analyzed qualitatively.

The materials used should simulate, and cover the range of, physical properties of nuclear fuel (heats of vaporization and combustion, thermal conductivity and critical temperatures). It will not be necessary to use actual fuels in testing. Test conditions should include:

- (1) Materials--droplets and particles in size ranges 0.1 to 1.0 micron, 1.0 to 10 microns, 10 to 100 microns, 100 to 1000 microns, and 1000 to 10,000 microns.
- (2) Atmosphere--densities corresponding to 100,000, 150,000 and 200,000 feet of altitude.
- (3) Velocity--3000, 5000 and 8000 mps.

b. Droplet breakup tests

This testing and analytical effort is recommended to verify and/or provide an analytical technique for prediction of droplet size on breakup of liquid fuel. Also required is the time to breakup after initial exposure to re-entry conditions. Testing can be conducted in a hypersonic facility; it may also be possible in a shock tube. Many of the essentials of this test may be covered incidental to the droplet ablation study; additional physical properties to be simulated in the test materials would be viscosity, surface tension and density. Test conditions should include:

- (1) Droplet size--100, 1000 and 10,000 microns.

- (2) Velocity--3000, 5000 and 8000 mps.
- (3) Atmosphere--densities corresponding to 100,000, 150,000 and 200,000 feet of altitude.

c. Fuel property tests

This testing is recommended to provide data on physical properties essential to use of an analytical procedure for prediction of final particle size. The most essential data are heat of combustion of plutonium and heats of vaporization of $\text{Cm}_2^{242}\text{O}_3$, $\text{Sr}^{90}\text{TiO}_3$ and $\text{Ce}^{144}\text{O}_2 + \text{SiC}$. However, Table 6 is included to indicate the additional information desired and the scope of testing to which this program might profitably be extended.

2. Vapor Re-entry Study Recommendations

a. Nucleation-condensation-agglomeration tests

Testing is recommended to:

- (1) Determine which materials, if any, in a mixed vapor cloud, preferentially form the nucleus of condensed particles.
- (2) Determine whether, on condensation of a mixed vapor cloud, materials coalesce in any desirable ordering of layers.
- (3) Provide empirical data on the nucleation-condensation-agglomeration rates and particulate properties as functions of the atmospheric density existing when these processes occur and when the several materials exist in various proportions of the whole. As far as possible cooling rates for the vapors should simulate those experienced at three altitudes: 100,000, 150,000 and 200,000 feet.

b. Agglomeration and nucleation rate testing

This testing is recommended to:

- (1) Quantify the effect on agglomeration rate of the radioactivity of the several fuel isotopes suitable for space use. This test should be conducted first, separate from those mentioned in Item (1) of Paragraph a in order that contamination be avoided in that test and in order that confusion as to cause and effect be reduced. Final testing is recommended as in Items (1) and (2), however, in order that the degree of fuel shielding be determined.

TABLE 6
Physical Properties of Radioisotope Fuel Forms

<u>Property</u>	<u>Materials</u>						
	<u>Po-210</u>	<u>Cm²⁴²O₃</u>	<u>Pu-238</u>	<u>UO₂</u>	<u>Ce¹⁴⁴O₂+SiC</u>	<u>Cs-137</u>	<u>Sr⁹⁰TiO₃</u>
Heat of combustion	1*	4	1	4	4	3	4
Heat of fusion	2	1	3	2	3	3	3
Heat of vaporization	2	1	3	1	3	3	3
Melting point	2	2	2	2	2	2	2
Boiling point	2	1	2	1	2	2	3
Heat capacity	1	1	3	2	3	3	2
Thermal conductivity	1	1	2	2	1	1	2
Emissivity	1	1	1	1	3	1	1
Viscosity	4	1	1	1	1	1	1
Surface tension	4	1	3	1	1	1	1
Density	2	2	2	2	2	2	2

*Key: 1 = unknown; 2 = known; 3 = uncertain; 4 = not applicable for particle or vapor re-entry study.

- (2) Quantify the part played by ion concentration in provision of condensation nuclei. The design of this test has not been considered in this study.

c. Vapor analytical studies

Additional study should be directed to:

- (1) The significance of Freidlander's findings in regard to an equilibrium state for aerosol particle size distribution in the atmosphere. The significance should be investigated as it relates the fate of radioactive particulate biologically to man.
- (2) Statistical determination of the amount of coalescing of condensed vapor occurring prior to solidification of droplets. Some related experimental data may be derived from paragraphs a and b.

III. PARTICULATE FATE STUDIES

In Chapter II, the formation of the particulate cloud and characteristics of the particulate were investigated; this chapter recounts the studies pertaining to the subsequent history of that particulate. The goal of these studies was to define the experimental programs necessary to precise prediction of the area and time of deposition of particulate, as a function of particulate cloud origin (time, altitude, latitude) and particle characteristics (density, size, shape).

A. SCOPE

The present state of knowledge concerning the mechanisms acting to disperse and deposit debris of stratospheric origin is reviewed in this chapter; these mechanisms are gravity (sedimentation rates), diffusion (molecular and turbulent) and transport phenomena (wind and temperature). A generally applicable equation is evolved, accounting for these mechanisms in predicting the fate of the particulate. The predictive quality of the equation is examined in the light of the limitations imposed by the present state of knowledge. Conclusions are drawn with respect to the additional information required, and definitive tests are designed. A minimal, but adequate, program of analysis and experimentation is recommended.

B. ANALYSIS OF MECHANISMS OF CLOUD GROWTH AND TRANSPORT

Sedimentation and diffusion are examined as the mechanisms of particulate cloud growth; bulk transport of the particulate cloud is then treated under a separate subheading.

1. Particulate Cloud Growth

Sedimentation and turbulent diffusion are the main processes controlling the growth of the cloud; the first, vertically, and the latter, about all three axes.

a. Sedimentation

Nuclear powered generators re-entering from orbit are expected to experience burnup beginning at an altitude of about 250,000 feet; fuel elements will not be penetrated until they have descended to the range 200,000 to 100,000 feet. Particulate radii from submicron to greater than 500 microns can be expected. The gravitational settling rates of these particles are dependent on particle size and density, and the

density and viscosity of the atmosphere. This gravitational settling will give a vertical profile to a particulate cloud in a quiescent atmosphere. The fall velocities experienced are defined by Stokes-Cunningham (Ref. 17) as follows:

$$V_f = \frac{2 r_s^2 g (\rho_s - \rho_a) (1 + k l / 2 r_s)}{9 \mu} \quad (19)$$

where

r_s = particle radius (cm)

ρ_s = particle density (gm/cm³)

ρ_a = ambient air density (gm/cm³)

g = gravitational acceleration (cm/sec²)

μ = viscosity of ambient air (gm/cm-sec)

l = molecular mean free path (cm)

$k \approx 0.86$ for air.

Figure 8 shows fall velocities computed from Eq 19 as a function of particle size and density, and altitude.

But the normal atmosphere is not quiescent. Winds and turbulence affect significantly the fall velocity and trajectory of all but the heaviest bodies. Where the wind profile is known, the drag forces induced by the relative wind can be computed and added vectorially to the gravitational force. Through a series of numerical integrations, the particle trajectory can be found. The usual method consists of averaging wind speed and direction over altitude intervals of about 10,000 feet, then adding this perturbation to the fall velocity computed at the median altitude. The resulting trajectory is a chain of straight-line segments from the release altitude to the earth's surface. Such approximations have been found fairly adequate for prediction of ground contamination patterns in the vicinity of nuclear weapon tests. Where the particulate in question is small--radius less than 10 microns, a similar accounting for turbulence is required. A technique applicable to this problem is discussed in paragraph b.

b. Diffusion

From Fig. 8 it is evident that particles smaller than 10 microns fall fairly slowly, so that the principal agents influencing their ultimate point of impact will be wind and turbulence. The initial cloud

Fig. 8. Terminal Velocity of Small Spherical Particles as a Function of Particle Density, Particle Radius and Altitude

growth will result from change of kinetic energy to thermal energy, with subsequent expansion. This thermal energy, however, will be rapidly dissipated; continued growth will be dependent on molecular and turbulent diffusion. Comparison of the computed molecular diffusion from 200,000 feet to ground level with observed rates of cloud growth indicates that molecular diffusion, as a mechanism of cloud growth, is of such small scale, in relation to turbulent diffusion, as to be negligible. Here, therefore, only the ultimate effects of turbulent diffusion are studied.

(1) Diffusion theories

When a cloud is considered to have developed from an instantaneous point source in infinite volume, the concentration at a general point (x, y, z) within the cloud is:

$$C(x, y, z, t) = \frac{M}{(4\pi t)^{3/2} (K_x K_y K_z)^{1/2}} \exp \left[-\frac{1}{4t} \left(\frac{x^2}{K_x} + \frac{y^2}{K_y} + \frac{z^2}{K_z} \right) \right] \quad (20)$$

where

- $C(x, y, z, t)$ = concentration of material at point (x, y, z) at time t (gm/cm³)
- t = time of this specific concentration, measured from time of material release (sec)
- M = amount of material released (gm)
- K_x, K_y, K_z = coefficients of diffusion (eddy diffusivities) in the three coordinate directions (cm²/sec)
- (x, y, z) = coordinates of point, with point of release as origin (cm).

Equation (20) is the Roberts solution (Ref. 18) to the differential equation describing molecular (Fickian) diffusion. It can be used to describe the turbulent diffusion process in an infinite homogeneous atmosphere. (G. I. Taylor, in Ref. 19, advanced the first statistical theory for treatment of diffusion where the turbulence is statistically homogeneous and isotropic.) It has given good results where the turbulent forces were accurately known. In Eq (20), homogeneity has been assumed to be analogous to accurate knowledge of the turbulence coefficients; further, the equation assumes that they are unchanging. This can be true only if a very large cloud is assumed or if activity is considered over a short time period.

That Eq (20) is applicable to only very large clouds or specific small clouds for short times is clearly shown by Richardson in Ref. 20. Richardson derives the following one-dimensional equation:

$$\frac{\partial n}{\partial t} = \frac{\partial}{\partial l} \left[K(l) \frac{\partial n}{\partial l} \right] \quad (21)$$

where

n = number of particles per unit length at distance l from a given particle

l = diameter of the largest turbulent eddy acting to spread the particles (cm)

$K(l)$ = diffusion coefficient (cm^2/sec).

This expression indicates that cloud growth rate is dependent on cloud size. Richardson was able to estimate, from experimental data, that $K(l)$ is approximately equal to $0.2l^{4/3}$. Although his theory has not yielded a directly applicable diffusion formula, it has thus put limits on the applicability of Eq (20); it is applicable, approximately, to a specific cloud size or, specifically, to the case where K_x , K_y , K_z are constant (very large clouds).

Kolmogoroff (Ref. 21) and Obukhov (Ref. 22) have been able to deduce Richardson's formula, Eq (21), from purely theoretical considerations. They obtain

$$K(l) = c \epsilon^{1/3} l^{1/3}$$

where

c = a pure number

ϵ = rate of turbulent energy dissipation

l = diameter of largest eddy causing spread of particulate.

Obtaining a value for the rate of turbulent energy dissipation poses the greatest difficulty. Taylor, in Ref. 23, suggests this might be done by correlating two-particle motion. An experimental approach to this is given in Section E of this chapter. When particular values for K_x , K_y and K_z are known, Eq 20 will be applicable to the cloud at various stages of growth. The numerical integration of particular solutions would give rise to the total solution at some time t from

which the cloud can be treated as affected by homogeneous turbulence-- i.e., constant K_x , K_y and K_z .

(2) Diffusion coefficients

The paucity of measurements or estimates of turbulent diffusion coefficients above 100,000 feet is notable. Lettau, in Refs. 24 and 47, gives estimates to altitudes in excess of 260,000 feet; they are somewhat crude and, at best, applicable to the particulate cloud only when it is very large. Reference 25, HASP program data, corroborates certain elements of Lettau's work to approximately 115,000 feet. Values for the latitudinal (crosswind) coefficient, K_y , between 5×10^8 and 5×10^9 cm^2/sec are representative of the stratosphere. Because of the normally greater stability of the tropical stratosphere (30° South to 30° North latitude) as compared to the polar stratosphere, it is felt that the lower value should be applied to a source originating over the tropics; the higher value would apply to a cloud of origin in the polar stratosphere. Values for the vertical coefficient, K_z , between 10^3 and 10^4 cm^2/sec are quoted in both Ref. 24 and Ref. 25. However, the HASP data assigns the lower value, again, to the tropical stratosphere and the higher value to the polar stratosphere; Lettau assigns the lower value to the altitude range 82,000 to 115,000 feet, and the higher value to the range 115,000 to 145,000 feet. From 145,000 to 260,000 feet, Lettau estimates K_z equal to 10^6 cm^2/sec . The meridional (downwind) coefficient, K_x , is estimated by Lettau as 10^{11} cm^2/sec . Reference 25 does not evaluate K_x from the HASP data. It does, however, state that bomb debris clouds are distributed in a band around the earth in a "few" months, and further, that they have by then lost their identity. The latter statement can be interpreted to mean that the "hot spot" is indistinguishable. These statements form the basis for an independent determination of effective K_x .

To perform this independent determination of K_x , "indistinguishable" was stipulated to mean that the maximum concentration along the X-axis of the cloud does not exceed twice the minimum concentration. The origin, $x = 0$, was taken to be the point of injection into the atmosphere and also the point of maximum concentration, $C_{\max}$; a point in the opposite hemisphere, diametrically opposed to $C_{\max}$, was termed $C_{\min}$. For this analysis, a "few" months was assumed to be 90 days (7.68×10^6 sec), the time necessary for K_x to act and cause $C_{\max}$ to equal $2C_{\min}$. The positive X-direction was taken as

toward the East. Unique values of x , P , range from 0 to S , where S is the circumference of the earth along the parallel of latitude. Again, for this analysis, 30° North was selected as the parallel of interest; therefore, $S = 3.48 \times 10^9$ centimeters.

Equation (20) was used to evaluate K_x ; since concentration on the X -axis only was of interest, $y = z = 0$ and Eq (20) becomes

$$C(x, t) = \frac{M}{(4\pi t)^{3/2} (K_x K_y K_z)^{1/2}} \exp \left(-\frac{x^2}{4tK_x} \right)$$

The form of the equation is such that a finite value of $C(x, t)$ exists for any t greater than zero. The east and west diffusing tails of the cloud will eventually overlap; the concentration at any unique point, P , where $0 < P < S$, is a function of time and the number of overlaps, n . The overlaps of $x = 0$ form the series

$$x_n = P + nS$$

where

$$n = 0, 1, 2, 3.$$

The contribution to the concentration at the point P of an overlap proceeding west to east is:

$$C_{w-e}(x_n, t) = \frac{M}{(4\pi t)^{3/2} (K_x K_y K_z)^{1/2}} \exp \left[-\frac{(P + nS)^2}{4tK_x} \right]$$

and the contribution from an east to west overlap is:

$$C_{e-w}(x_n, t) = \frac{M}{(4\pi t)^{3/2} (K_x K_y K_z)^{1/2}} \exp \left[-\frac{(S - P + nS)^2}{4tK_x} \right]$$

The sum of the C_{w-e} and C_{e-w} contributions at $x = P$ for the n th lap is:

$$C_n(x_n, t) = \frac{M}{(4\pi t)^{3/2} (K_x K_y K_z)^{1/2}} \left\{ \exp \left[-\frac{(P + nS)^2}{4tK_x} \right] + \exp \left[-\frac{(S - P + nS)^2}{4tK_x} \right] \right\} \quad (22)$$

The concentration effected by all such overlaps is:

$$C(P, t) = \sum_{n=0}^n C_n(x_n, t)$$

At $P = x = 0$, the point of $C_{\max}$, Eq (22) can be better written

$$C_n(0, t) = \frac{M}{(4\pi t)^{3/2} (K_x K_y K_z)^{1/2}} \left[1 + 2 \exp \left(-\frac{n^2 S^2}{4t K_x} \right) \right] \quad (23)$$

$$\text{and } C(0, t) = \sum_{n=0}^n C_n(0, t)$$

The use of values of n greater than one does not appear reasonable. When $n = 1$, the twin tails of the diffusing cloud have met for the first time at the origin. It seems reasonable to assume that the relative concentrations, point-to-point on the X-axis, will not be changed appreciably for $n = 2, 3, 4$. By recalling that

$$C_{\max} = 2C_{\min} \quad (24)$$

Eqs (22) for $C_{\min}$, (23) for $C_{\max}$ and (24) were solved simultaneously for K_x ($n = 1$). A value of $0.7 \times 10^{11} \text{ cm}^2/\text{sec}$ was obtained; this compared favorably with Lettau's estimate, $10^{11} \text{ cm}^2/\text{sec}$. Use of the raw HASP data in a similar manner would better evaluate this method of estimating K_x .

It has been shown that only limited data, poorly confirmed, is available on diffusion coefficients. If a random re-entry of the burn-up material of interest is assumed (and it must be), then specific coefficient data as a function of altitude, latitude and cloud size is required if the fate of the particulate is to be predicted analytically. The latitudes where these need most to be better defined as functions of season and altitude are:

- (1) Near the equator (transequatorial exchange).
- (2) In the vicinity of the break in the tropopause, the higher midlatitudes.

2. Particulate Cloud Transport

Short term fallout of burnup debris is principally associated with particulate greater than 10 microns in diameter (see Section B). For debris of this nature, the transport mechanism of major interest is wind; a fairly accurate vertical profile of wind velocity and direction in the latitude of re-entry must be known to develop the deposition pattern. A second transport mechanism should be considered for particulate which is, in general, smaller than 10 microns (certainly, for that less than 1 micron).

Two general models describing this second mechanism retain popular support. One, that proposed by Brewer and Dobson (Refs. 26 and 27) and supported by Machta and List (Refs. 28 and 50), postulates that the particulate cloud, while growing by diffusion, is carried poleward from the point of injection by a continuous slow poleward circulation of stratospheric air. In the region of the poles, debris enters the troposphere and is subsequently deposited. The spring high of debris deposition can be accounted for thus: the polar tropopause lowers at the onset of the winter, increasing the available stratospheric storage volume. In the spring the polar tropopause rises in height, placing in the troposphere debris formerly stored in the stratosphere; in about 30 days this debris is deposited.

The second model, that of Feely-Spar (Refs. 25 and 30), denies the existence of a general poleward circulation in the stratosphere. It substitutes for this a theory which states that the particulate cloud diffuses preferentially to the pole, the diffusion proceeding most rapidly at the onset of, and during, the winter. Coincident with the beginning of the winter, a powerful circumpolar vortex is formed. It is thought to become unstable and break up during the winter. A vigorous dumping of stratospheric air in the northern temperate zone is believed to be associated with this breakup; this phenomenon accounts for the measured spring high of debris deposition.

These two models predict the same type of qualitative behavior, particularly in confirming the spring high. However, disagreement between them is evidenced by the influence each predicts that altitude, latitude and season of particulate injection will have on ultimate disposition of the particulate. In apparent support of Feely-Spar, Martel and Drevinsky (Refs. 31, 45 and 46) predict strong effects of all three on residence time; further, they support a picture of a relatively poorly mixed stratospheric reservoir, implying a strong influence of altitude, latitude and season of injection on deposition pattern. Conversely, Machta (Ref. 32) and Machta and List (Ref. 28) appear to favor Brewer-Dobson and a more uniformly mixed stratospheric reservoir. Such a model reduces the influence of spatial origin on deposition pattern and residence time.

The disagreement mentioned above can be resolved only by obtaining good experimental data on poleward circulation rates and diffusion coefficients. Data from sampling of nuclear weapon debris apparently will not resolve the discrepancy; in fact, differing interpretations of the meaning of those data created the existing diversity of models. The principal reason for the dissimilar interpretations is the lack of precision with which portions of a gross sample of bomb debris can be assigned to a specific weapons test.

The many factors precluding precision comprise:

- (1) Bodies of fission debris reaching the stratosphere are consistently similar, regardless of their sources.
- (2) Tracer elements have served to reduce this similarity but have been used on relatively few tests.
- (3) The isotope ratio method of dating used by HASP (Ref. 25) and others is reduced markedly in effectiveness with increasing time after test because of the decay of short-lived isotopes.
- (4) Fractionation occurring at the source and later in the atmosphere is not accurately predicted.
- (5) The total amounts of aerosol material injected into the atmosphere can be only roughly estimated, based on weapon size, fission yield, altitude of burst, nature and amount of entrained surface materials, etc.
- (6) The percent of total aerosol yield reaching and remaining at specific altitudes can be only roughly approximated.

Particulate cloud transport tests which circumvent the shortcomings in available data are outlined in Section E of this chapter.

3. An Analytical Model for Cloud Growth and Transport

The following equations predictive of the fate of a particulate cloud are derived and presented as the rough framework within which refinements and improvements can be made as good experimental data are obtained. The equations are derived to determine the particle concentration at a fixed point due to diffusion, sedimentation and gross motion of an initially small cloud injected at the origin of the coordinates. The parameters assumed constant in this study do not invalidate results that would be obtained over sufficiently small intervals of time; hence, the equations are readily adaptable to machine computations in which the variability of parameters would be accounted for by successive approximations and numerical integration.

The situation analyzed is presented schematically in Fig. 9. For an instantaneous point cloud in an infinite medium, the Fickian diffusion equation (Ref. 18) expresses the concentration at a general point at time t :

$$C(x, y, z, t) = \frac{M}{(4\pi t)^{3/2} (K_x K_y K_z)^{1/2}} \exp \left[-\frac{1}{4t} \left(\frac{x^2}{K_x} + \frac{y^2}{K_y} + \frac{z^2}{K_z} \right) \right] \quad (25)$$

where

- M = amount of material injected
- (x, y, z) = coordinates of concentration point with respect to origin
- t = time at which concentration is taken
- K_x, K_y, K_z = diffusion coefficients about respective axes.

This expression must be modified to account for wind, poleward drift and sedimentation. For the interval of time selected, the wind plus drift portion is assumed to have a constant mean velocity, $\bar{u}$. The vector equation is

$$\bar{u} = a\bar{i} + b\bar{j} + c\bar{k} \quad (26)$$

where

- a, b, c = components of motion in the x, y, z directions, respectively
- $\bar{i}, \bar{j}, \bar{k}$ = unit vectors in the x, y, z directions, respectively.

Characteristics of the particulate is one of the parameters varied and summed by numerical integration. In a particular solution, a single particle size is treated and the terminal velocity v_m is vectorially represented

$$\bar{v}_m = -\bar{k} v_m$$

From Fig. 9

$$\bar{r}' = \bar{r} - \bar{R} \quad (27)$$

Fig. 9. Diffusion-Dispersion Model

and

$$\dot{\bar{\mathbf{R}}} = \bar{\mathbf{u}} + \bar{\mathbf{v}}_m = a\bar{\mathbf{i}} + b\bar{\mathbf{j}} + (c - v_m)\bar{\mathbf{k}}$$

$$\bar{\mathbf{R}} = \left[a\bar{\mathbf{i}} + b\bar{\mathbf{j}} + (c - v_m)\bar{\mathbf{k}} \right] t + C_1$$

$$C_1 \rightarrow 0, \text{ since } \bar{\mathbf{R}} = 0 \text{ at } t = 0$$

therefore,

$$\bar{\mathbf{R}} = a t \bar{\mathbf{i}} + b t \bar{\mathbf{j}} + (c - v_m) t \bar{\mathbf{k}} \quad (28)$$

also

$$\bar{\mathbf{r}} = x\bar{\mathbf{i}} + y\bar{\mathbf{j}} + z\bar{\mathbf{k}} \quad (29)$$

Putting Eqs (28) and (29) into Eq (27) gives

$$\bar{\mathbf{r}}' = (x - at)\bar{\mathbf{i}} + (y - bt)\bar{\mathbf{j}} + (z - ct + v_m t)\bar{\mathbf{k}} \quad (30)$$

The scalar product of Eq (30) with respect to itself is

$$(\mathbf{r}')^2 = (x - at)^2 + (y - bt)^2 + (z - ct + v_m t)^2 \quad (31)$$

Substituting into Eq (25) gives

$$C(\mathbf{r}', t) = \frac{M}{(4\pi t)^{3/2} (K_x K_y K_z)^{1/2}} \exp \left\{ -\frac{1}{4t} \left[\frac{(x - at)^2}{K_x} + \frac{(y - bt)^2}{K_y} + \frac{(z - ct + v_m t)^2}{K_z} \right] \right\} \quad (32)$$

With the assumption of spherical particles, Eq (32) is expressed in particles/ft³ as:

$$C(\mathbf{r}', t) = \frac{1.214 \times 10^3 M}{\rho d^3 t^{3/2} (K_x K_y K_z)^{1/2}} \exp \left\{ -\frac{1}{4t} \left[\frac{(x - at)^2}{K_x} + \frac{(y - bt)^2}{K_y} + \frac{(z - ct + v_m t)^2}{K_z} \right] \right\} \quad (33)$$

where

d = diameter of particle (cm)

ρ = density of particle (gm/cm³).

To find the time at which maximum concentration is reached at the general point (x,y,z), the derivative of Eq (33) is set equal to zero and solved to give:

$$t_{C_{\max}} = \frac{-\beta \pm \sqrt{\beta^2 - 4\alpha\gamma}}{2\alpha} \quad (34)$$

where

$$\alpha = \left[K_y K_z a^2 + K_x K_z b^2 + K_x K_y c^2 - 2 K_x K_y c v_m + K_x K_y v_m^2 \right]$$

$$\beta = 6 K_x K_y K_z$$

$$\gamma = - \left[K_y K_z x^2 + K_x K_z y^2 + K_x K_y z^2 \right]$$

Experimental data inputs to the equations would include

- (1) Concentration at the point (x,y,z).
- (2) Time after injection.
- (3) Particle characteristics and altitude into Eq (20) for v_m .
- (4) Distances x, y and z from point of origin.

Values to be solved for in the computer program would be

- (1) Diffusion coefficients, K_x , K_y , K_z .
- (2) Gross drift rate $\bar{u}$ as represented by a, b, c.

Obviously, an almost infinite combination of the six unknowns could be found, giving a correct solution to a single experimental concentration measurement. However, as different points are sampled at different times after injection, the possible combinations of variables fulfilling all the data input conditions will be narrowed to a single effective value for each unknown. Data derived would then be correlated with altitude, latitude and season of injection.

C. CONCLUSIONS AND RECOMMENDATIONS

Resulting from the comprehensive study of the mechanisms determinant of the fate of re-entry debris were the following conclusions and recommendations.

1. Conclusions

- (1) The primary processes influencing the final disposition of particulate are:
 - (a) Sedimentation rates.
 - (b) Turbulent diffusion.
 - (c) Gross atmospheric motions, including large-scale winds and, perhaps, very marked poleward diffusion of stratospheric air masses.
- (2) Insufficient data are available on diffusion coefficients and bulk air transport to permit accurate analytical prediction of the fate of the particulate.
- (3) The quantitative effects of altitude, latitude and season of injection on particulate disposition are poorly defined.
- (4) Weapon debris sampling is unlikely to quantify the dependencies of Item (3) or provide accurate information regarding Item (2).

2. Recommendations

To fulfill the needs implied by Conclusions (2) and (3) and to circumvent the weaknesses of weapon debris sampling, the following recommendation is made:

That a minimal, but adequate, two-part experimental program combined with an analytical correlation effort be undertaken to determine:

- (1) Residence time of high altitude particulate as a function of altitude, latitude and season of injection.
- (2) Diffusion coefficients and gross atmospheric motions as functions of the same variables.

The first recommended program would be directed to determination of diffusion coefficients and gross motion estimates for use directly in a predictive model similar to that of Section B, Topic 3. The second recommended program would serve to test the predictive model and provide sufficient concentration data for use as a secondary means of determining diffusion coefficients and gross motion vector quantities.

The objectives of the first experiment can be met through either of two approaches. Each would be conducted at a minimum of three altitudes in each of four seasons at three or more latitudes. The approaches are:

- (1) Observation, photographically, of vapor trails laid down in the stratosphere by rocket mounted smoke generators.
- (2) Recording of turbulence power spectra data by accelerometer equipped, constant pressure altitude balloons.

The objectives of the second experiment can be met by injection of a known quantity of readily identifiable test material of predetermined particulate characteristics into the stratosphere at a known position and time; subsequent sampling above, at and below the injection altitude at various times after injection, will provide isoconcentration profiles for the particulate cloud. Should it be revealed that an adequate analytical predictive model has not been obtained as a result of the first experiment, then this second experiment would be expanded to world-wide scale (i.e., injections at representative altitudes at all seasons and many latitudes). Sufficient samples of injected materials would be collected and correlated with injection variables to provide a purely empirical basis for prediction of the fate of radioactive debris. As will be pointed out in Chapter IV, use of nonradioactive test materials in these tests is feasible.

Section D, following the conclusions and recommendations of this chapter, expands on the design of the experiments recommended above.

D. DESIGN OF RECOMMENDED EXPERIMENTAL PROGRAMS

The feasibility of the experimental program outlined below is examined in Chapter IV.

1. Determination of Diffusion Coefficients and Cloud Transport Vectors

Individual small scale experiments. Two possible approaches to solution of this problem were mentioned in Section C: observation of high altitude vapor trails, and collection of balloon acceleration data. These are discussed under their individual subheadings below.

a. Vapor trail observation

In recent years the Geophysics Corporation of America (GCA) under contract to the National Aeronautics and Space Administration has conducted a series of sodium vapor trail experiments above an altitude of 260,000 feet (Refs. 48 and 49). The trail is deployed from a vapor generator carried aloft by a sounding rocket. The deployed trail extends in a continuous filament over most of the trajectory above 260,000 feet. Motion of the vapor trail is recorded from triangulated camera observation stations. Reduction of photographic data yields precise data on wind direction and velocity, small scale turbulence and molecular diffusion (a measurable factor at these altitudes).

For the purposes of this program, the altitudes studied by GCA are too high; further, the turbulence scale investigated is quite small. However, the techniques used appear applicable to determination of diffusion coefficients and gross cloud transport in the altitude range of interest--100,000 to 200,000 feet. Reduction in altitude may require use of some substance other than an oxidizing test material such as sodium; GCA, under subcontract to Martin Marietta on another phase of this Task 7 program, investigated day and night atmospheric dye and flare markers. Use of aluminum oxide powder appears feasible. Reference 33, in addition, indicates that foamed plastic particulate smoke may be suitable. Extension of the techniques to yield data on large scale turbulence is also believed possible. Observation of radial expansion of the vapor trail yields small scale turbulence data. For large scale turbulence, trail length can be used. Ballistic camera techniques (Ref. 34) will provide an accurate set of space statistics on wind velocity along the trail length. This can be treated statistically to give the variance of wind velocity over trail length as a function of altitude; diffusion coefficients can be derived from variance data (see ballooning experiments following this section).

There is another useful technique for determining large scale turbulence, requiring that the cloud generator disperse the particulate in a chain of puffs. Vertical motions will become more evident than in continuous filament trail experiments. A set of statistics on puff separation rates as a function of average separation distances would yield important information on the relationship of turbulent diffusivity to eddy size (see Richardson, Kolmogoroff and Obukhov as mentioned in Section B of this chapter).

b. Ballooning experiments

Two possible methods exist for using balloons in determination of diffusion coefficients and drift. The first requires that a constant pressure altitude balloon be instrumented with accelerometers; the

second, that the motion of a free floating balloon or chaff be recorded photographically or by radar.

In the first method a constant pressure altitude balloon is equipped with three-axis accelerometers and a transmitter for relaying data or a recorder for storing it. The balloon is deployed at the altitude, latitude and longitude of interest. It will experience a random velocity $v(t)$ which has components $v_x(t)$, $v_y(t)$ and $v_z(t)$. For the random process, $v_x(t)$, the autocorrelation function, is defined by:

$$R_{v_x v_x}(\tau) = E \left[v_x(t) v_x(t + \tau) \right] \quad (35)$$

where

E denotes expectation, and the process is assumed stationary.

The variance of the displacement in the x direction,

$$E x^2 = \bar{x}^2 = \sigma_x^2,$$

is, according to Taylor (Ref. 19),

$$\sigma_x^2(t_1) = \int_0^{t_1} \int_0^{\xi} R_{v_x v_x}(\tau) d\xi d\tau \quad (36)$$

Values for $\sigma_y^2(t)$ and $\sigma_z^2(t)$ are defined in a similar fashion. The usefulness of this information is apparent when related to injection of a particulate cloud. The variance of the dispersion in the respective x , y and z directions is determined from Eq (36). This means that 68% of the particles will be within σ_x , σ_y and σ_z of the starting point, and 96% within 2σ . Diffusion coefficients can be deduced from knowledge of the degree to which particulate has spread in a given time.

The problem in translating theory into practice is to define the three correlation functions. They are determined from measurement of balloon motion. (It is assumed that the velocity correlation function of the balloon will be that of the particulate; however, this is not entirely true. Several effects occur in the actual experiment that prevent this:

- (1) Lag in balloon response to temperature variations induced by solar heating, night cooling and ambient temperature change.

- (2) Inertial differences between the balloon and particulate.
- (3) Large scale motions too slow to permit proper measurement.

It is believed most of such effects can be reduced by good design and/or measured and corrected for prior to data analysis. Radar tracking and optical sightings might augment the accelerometers to account for large scale motion.)

To determine the velocity correlation functions, the measured and recorded balloon accelerations $a_x(t)$, $a_y(t)$ and $a_z(t)$ must be processed. This can be mechanized for analog computer solution. Recorded data are played back at a rate commensurate with the frequency response of the computer. Integration yields $v_x(t)$, $v_y(t)$ and $v_z(t)$, which are again stored on tape. $v_x(t)$ and $v_x(t + \tau)$ are obtained by using two pickup heads on the $v_x(t)$ tape. $v_y(t)$ and $v_z(t)$ are handled similarly. Only the handling of $v_x(t)$ will be considered henceforth. $v_x(t)$ and $v_x(t + \tau)$ are multiplied together and passed through an averaging device (low pass filter) from which the time-averaged product $v_x(t) v_x(t + \tau)$ is obtained. Under ergodic hypothesis this is taken to be

$$\langle v_x(t) v_x(t + \tau) \rangle = E [v_x(t) v_x(t + \tau)] \quad (37)$$

which is the desired correlation function $R_{v_x v_x}(\tau)$. Other values of $R_{v_x v_x}(\tau)$ can be determined in a similar manner for additional values of τ .

The correlation function $R_{v_x v_x}(\tau)$ is now used to evaluate Eq (36) for σ_x^2 . At least three possible methods are available for solution of Eq (36):

- (1) After plotting $R_{v_x v_x}(\tau)$ versus τ , the integration indicated in Eq (36) can be performed graphically.
- (2) After plotting $R_{v_x v_x}(\tau)$ versus τ , an expression can be written to fit the data. This expression can then be integrated to obtain σ_x^2 via Eq (36).

- (3) $R_{v_x v_x}(\tau)$ can be set up on a function generator and the integrations indicated in Eq (36) performed by computer.

The second method of using balloons or chaff to deduce diffusion coefficients requires recording of position with time. Movement can be reduced to $v_x(t)$, $v_y(t)$ and $v_z(t)$ and various values of τ used for obtaining time averaged products of $\langle v_x(t) v_x(t + \tau) \rangle$. The data is handled in a similar fashion, then, to that of the first method discussed. Ballistic camera pairs can, by triangulation, give a bias-free accuracy of one part in 10^5 parts of range (Ref. 34). A velocity component as low as 2 ft/min would give a position change of 30 feet in 15 minutes, or an average error in determination of the net velocity component of less than 10%. Such accuracy would be considered good in light of the data now available.

2. The Particulate Sampling Experiment

As noted in Section C of this chapter, this experiment is designed, first, to test the predictive quality of the analytical model describing the fate of the particulate, and, second, to generate isoconcentration profiles at several times after injection, this data to be used as an independent determination of diffusion coefficients and bulk cloud transport. The requirements for this test are suitably spelled out in Section C. Attention is here directed to the extension of that program which could be necessitated by failure of the experiments of Section D, Topic 1, to provide an adequate analytical predictive model; i.e., the experiments required to obtain an adequate empirical model are defined.

Test sample injections defining the extreme limits of residence time might be as follows:

- (1) Upper limit
 - (a) Altitude--200,000 feet
 - (b) Latitude--10° north (south)
 - (c) Season--winter (probably not too significant)
- (2) Lower limit
 - (a) Altitude--100,000 feet
 - (b) Latitude--60° north (south)
 - (c) Season--midwinter to early spring.

Logically, a good intermediate value should be determined.

(3) Intermediate value

- (a) Altitude--200,000 feet
- (b) Latitude--35° north (south)
- (c) Season--summer.

A program limited solely to these three injections would reveal little as regards deposition dependency on altitude, latitude and season. To obtain data of empirical value, the following schedule of additional injections, with subsequent sampling, is recommended.

<u>Latitude</u>	<u>Altitude (ft)</u>	<u>Seasons</u>
10° N or S	100,000; 150,000; 200,000	Summer
	100,000; 150,000	Winter
35° N or S	100,000; 150,000; 200,000	Winter
	100,000; 150,000	Summer
60° N or S	100,000	Spring, summer, fall
	150,000; 200,000	Winter, spring, summer, fall

A greater number of injections is scheduled at 60 degrees than at the other latitudes to define precisely the time of onset, magnitude and time of cessation of stratospheric dumping in this region. At 10 and 35 degrees, a comparison of sampling results for summer and winter injections will establish the need or lack thereof for spring and fall injections. Injection at 10 degrees rather than the equator is suggested as a means of defining transequatorial exchange as a function of altitude and latitude of injection. Presumably, injections at the equator are equally distributed north and south; this premise, coupled with data from 10, 35 and 60 degrees, should fully develop this dependency in the altitude range of interest.

3. Analytical Correlation of Experimental Data

In Section C, the principal recommendation alluded to a need for correlation of experimental data. This is required to give utility to the experimental data, purpose to the tests, and perhaps reduce the number of tests required.

Data received from the sampling program would consist of average particle concentration existing at a particular place and time. These data must be correlated with the various known source points of injection. Based on the point of injection, the point particle concentration will be a function of diffusion, sedimentation and gross movement of the source cloud.

Residence times can be computed directly from an average of point concentration data. Correlation of data to develop fundamental knowledge of the effects of diffusion and gross movement may serve to reduce the size of the experimental program required. Since the cloud injected at one point and season will still be present, in diluted form, in future seasons and positions, continued sampling, coupled with appropriate data handling, might also serve to reduce the number of injections required.

IV. FEASIBILITY STUDIES ON RECOMMENDED PROGRAMS

In the foregoing chapters, areas requiring experimental data were delineated. The nature of these data and experimental requirements were also explored. The purpose of the following series of studies has been to determine the feasibility of the recommended tests, and the availability of equipments and facilities required. Again, the material is divided into that investigating, first, particulate characteristics, and, second, the ultimate fate of that particulate.

A. FEASIBILITY OF PARTICULATE CHARACTERISTICS EXPERIMENTS

The essentials of the particulate characteristics experiments recommended in Chapter II, Section C, reduced to the following, each of which is treated in this section:

- (1) Materials that simulate the fuel properties most important to the ablation process.
- (2) Test facilities and instrumentation.

1. Materials for Simulation of Nuclear Fuels

It was deemed desirable, where possible, to use nonradioactive materials in testing. Satisfactory simulators were found for all the fuels studied; however, it was found that a material which closely simulates the chemical properties of a fuel does not necessarily simulate the thermal properties of the fuel. The general fuel properties which it would be most desirable to simulate in ablation testing are:

- (1) Chemical properties
 - (a) Affinity for oxygen
 - (b) Reactions at high temperature
 - (c) Solubility
- (2) Thermal properties
 - (a) Heat of fusion
 - (b) Heat of vaporization
 - (c) Melting point

- (d) Boiling point
- (e) Heat capacity
- (f) Emissivity

The chemical properties of a fuel can usually be simulated by an element in the same group of the periodic table and with similar numbers of electrons in the outer shells of the atom. If the fuel is a compound, the simulating material should be a compound. Thermal properties of elements and compounds vary widely. Simulating materials with melting points and heats of fusion similar to those of the fuel were selected. The boiling points and heats of vaporization are also very important, but it was impossible to find a simulating material possessing all four of the above properties for a given fuel.

a. Polonium simulation

The properties of polonium are given in the listing below; selection of materials simulating its thermal and mechanical properties follows.

(1) Atomic number	84
(2) Density	9.4 gm/cm ³
(3) Melting point	254° C
(4) Boiling point	962° C
(5) Emissions (mev)	$\alpha = 5.303$; $\gamma = 0.803$
(6) Half life	138 days (0.378 year)
(7) Heat of fusion at the melting point	3000 calories per gram formula weight (cal/gfw)
(8) Heat of vaporization at 1 atm pressure	14,400 cal/gfw
(9) Valence	4
(10) Outer shell configuration	

$$\text{Shell O } \left\{ \begin{array}{c} s \quad p \quad d \quad f \\ 2 \quad 6 \quad 10 \quad - \end{array} \right. \quad \text{Shell P } \left\{ \begin{array}{c} s \quad p \quad d \quad f \\ 2 \quad 4 \quad \quad \quad \end{array} \right.$$

Chemical simulator. Tellurium was found to have similar chemical properties to polonium. The isotope Te-130, the most abundant isotope, is stable. Pertinent data follow:

(1) Outer shell configuration

$$\text{Shell N } \left\{ \begin{array}{c} s \quad p \quad d \quad f \\ 2 \quad 6 \quad 10 \end{array} \right. \quad \text{Shell O } \left\{ \begin{array}{c} s \quad p \quad d \quad f \\ 2 \quad 4 \end{array} \right.$$

(2) Valence 2, 4, 6

Thermal simulator. Bismuth was found similar in thermal properties to polonium. A comparison is shown below:

	<u>Po-210</u>	<u>Bi-209</u>
(1) Atomic number	84	83
(2) Density (gm/cm ³)	9.4	9.8
(3) Melting point (°C)	254	271
(4) Boiling point (°C)	962	1560
(5) Heat of fusion (cal/gfw)	3000	2600
(6) Heat of vaporization (cal/gfw)	14,400	36,200
(7) Specific heat (gm-cal/gm atom)	6.3 at 25° C 7.35 at 254° C (mp) 7.50 at 962° C (bp)	6.11 at 25° C 7.41 at 271° C (mp) 7.50 at 1560° C (bp)
(8) Surface tension (dyne/cm)	--	376 at 300° C
(9) Entropy (cal/deg-gfw)	15.0 at 25° C 18.7 at 254° C (mp) 31.0 at 962° C (bp)	13.58 at 25° C 17.1 at 271° C (mp) 31.4 at 1560° C (bp)

b. Curium simulation

Curium-242 is the isotope used; Cm₂O₃ is the fuel form. Properties of the oxide are as follows:

(1) Atomic number (Cm-242)	96
(2) Density (gm/cm ³)	11.75
(3) Melting point (°C)	1500
(4) Boiling point	--
(5) Emissions (mev)	$\alpha = 6.08, \gamma = 0.045$
(6) Half life (days)	162
(7) Heat of fusion* (cal/gfw)	$10,600 \pm 1800$ at mp
(8) Heat of vaporization	--
(9) Valence (of curium)	3
(10) Outer shell configuration (of curium)	

$$\text{Shell P } \left\{ \begin{array}{cccc} s & p & d & f \\ 2 & 6 & 1 & - \end{array} \right. \quad \text{Shell Q } \left\{ \begin{array}{cccc} s & p & d & f \\ 2 & & & \end{array} \right.$$

*Heat of fusion estimated from $\lambda_f/T_f = 5$ to 7

where

λ_f = heat of fusion (cal/gfw)

T_f = melting point (°K) (Ref. 35).

Chemical simulator. Gadolinium oxide was found to have similar chemical properties; Gd-158, the most abundant isotope, is stable. Pertinent data follow:

(1) Outer shell configuration

$$\text{Shell O } \left\{ \begin{array}{cccc} s & p & d & f \\ 2 & 6 & 1 & - \end{array} \right. \quad \text{Shell P } \left\{ \begin{array}{cccc} s & p & d & f \\ 2 & & & \end{array} \right.$$

(2) Valence

3

Thermal simulator. The best thermal property simulators of Cm_2O_3 were found to be FeO_3 and Ce_2O_3 . A comparison of properties follows:

	<u>Cm₂O₃</u>	<u>Fe⁵⁶₂O₃</u>	<u>Ce₂O₃</u>
Melting point (°C)	1500	1565	1690
Boilint point	--	--	--
Heat of fusion* (cal/gfw)	10,600 ± 1800	9400 ± 1600	10,150 ± 1700
Heat of vapori- zation	--	--	--
Specific heat (gm-cal/gm)	--	0.148 at 0° C 0.182 at 100° C 0.263 at 400° C	--
Surface tension	--	--	--
Entropy (cal/°C-gfw)	--	21.5 at 25° C	--
Density (gm/cm ³)	11.75	5.12	6.95

*Calculated as per Ref. 35.

c. Plutonium simulation

The fuel form is Pu-238 metal. Its properties follow:

(1) Atomic number	94
(2) Density (gm/cm ³)	16.5
(3) Melting point (°C)	640
(4) Boiling point (°C)	3235
(5) Emissions (mev)	$\alpha = 5.49$, $\gamma = 0.04$
(6) Half life (yr)	92
(7) Heat of fusion (cal/gfw)	760 ± 115 at mp
(8) Heat of vaporization	80,460 ± 340 at 1 atm
(9) Valence	3, 4 (usually 3)

(10) Outer shell configuration

$$\text{Shell P } \left\{ \begin{array}{cccc} s & p & d & f \\ 2 & 6 & - & - \end{array} \right. \quad \text{Shell Q } \left\{ \begin{array}{cccc} s & p & d & f \\ 2 & - & - & - \end{array} \right.$$

Chemical simulator. Samarium was found to be the best chemical simulator of plutonium; Sm-152 is stable and the most abundant isotope. Pertinent data are:

(1) Outer shell configuration

$$\text{Shell P } \left\{ \begin{array}{cccc} s & p & d & f \\ 2 & 6 & - & - \end{array} \right. \quad \text{Shell Q } \left\{ \begin{array}{cccc} s & p & d & f \\ 2 & - & - & - \end{array} \right.$$

(2) Valence

2, 3 (usually 3)

Thermal simulator. The thermal properties of plutonium are poorly simulated by other materials. Those compared below approximate it roughly and are particularly good for droplet breakup studies since their surface tensions are similar to that of plutonium.

	<u>Pu</u>	<u>Ce-140</u>	<u>Al</u>	<u>Mg</u>
Density (gm/cm ³)	16.5	6.768	2.7.	1.74
Melting point (°C)	640	804	660	650
Boiling point (°C)	3235	2927	2450	1117
Heat of fusion (cal/gfw)	760 ± 115	2200	2550	2140
Heat of vaporiza- tion (cal/gfw)	80460 ± 340	75,000	70,200	30,750
Specific heat (gm/cal-gm atom)	8.1 at 25° C	6.89 at 25° C	5.817 at 25° C	5.71 at 25° C
	10.4 at 500° C	8 at 804° C	7.4 at 660° C	7.88 at 727° C
Surface tension (dyne/cm)	500 at 640° C	--	520 at 750° C	563 at 681° C
Entropy (cal/°C-gfw)	--	16.64 at 25° C	6.77 at 25° C	7.81 at 25° C

	<u>Pu</u>	<u>Ce-140</u>	<u>Al</u>	<u>Mg</u>
Entropy, (continued)	--	28.76 at 804° C	14.1 at 660° C	15.2 at 650° C
	--	37.51 at 2727° C	24.5 at 2450° C	38.0 at 1107° C

d. Uranium simulation

Simulators for only the oxide, UO_2 , were sought. Uranium carbide is already under investigation for similar use--i.e., safety in space. Properties of UO_2 follow:

(1) Atomic number (U-235)	92
(2) Density (gm/cm^3)	10.9
(3) Melting point ($^{\circ}\text{C}$)	2176
(4) Boiling point	--
(5) Emissions (mev)	$\alpha = 4.40; \gamma = 4.58$
(6) Half life (yr)	7.1×10^8
(7) Heat of fusion* (cal/gfw)	$13,050 \pm 2176$ at mp
(8) Heat of vaporization	--
(9) Valence (of uranium)	4, 6
(10) Outer shell configuration
(of uranium)	

$$\text{Shell P } \left\{ \begin{array}{c} s \quad p \quad d \quad f \\ 2 \quad 6 \quad 1 \quad - \end{array} \right. \quad \text{Shell Q } \left\{ \begin{array}{c} s \quad p \quad d \quad f \\ 2 \quad \quad \quad \quad \end{array} \right.$$

*Calculated per Ref. 35.

Chemical simulator. Although no other element has the same outer shell configuration as uranium, the element hafnium vaguely approximates the chemical properties of uranium and thorium. The rare earth element praseodymium has a closer outer shell configuration to uranium but its chemical properties are different. Hafnium oxide (Hf-178) is a fairly good simulator for uranium oxide. Pertinent properties are:

(1) Outer shell configuration

(2) Valence

Thermal simulator. Lack of extensive thermal data on UO_2 prevented a full comparison with other materials. The compounds CeO_2 and HfO_2 might be good thermal simulators, as indicated below:

	<u>UO_2</u>	<u>CeO_2</u>	<u>HfO_2</u>
Density (gm/cm^3)	10.9	7.3	9.68
Melting point ($^{\circ}\text{C}$)	2176	1950	2812
Heat of fusion (cal/gfw)	$13,500 \pm 2176$	$11,700 \pm 1950$	$16,870 \pm 2812$

Heat of vaporization, specific heat, boiling point, heat of fusion, surface tension and entropy could not be compared due to lack of data.

e. Cerium simulation

The oxide Ce_2O_3 is the fuel form; Ce-144 is the isotope. Another fuel form is $\text{Ce}_2\text{O}_3 + \text{SiC}$. Substitution of the stable isotope Ce-140 for Ce-144 provides the ideal simulator.

f. Cesium simulation

The isotope Cs-137 is the fuel; the stable isotope Cs-133 is an ideal simulator.

g. Availability and cost

Companies where fuel simulator materials can be obtained include those in the following list; costs of materials, as of 1961, appear in a separate listing.

<u>Fuel</u>	<u>Simulating Material</u>	<u>Companies</u>
Po-210	Te-130	American Metal Company American Smelting & Refining Company

<u>Fuel</u>	<u>Simulating Material</u>	<u>Companies</u>
		Anaconda Copper Mining Company
	Bi-209	American Smelting & Refining Company
		Eagle Picher Company
		Cerro de Pasco Copper Corporation
Cm_2O_3	Gd_2O_3	Davidson Chemical Company Division of W. R. Grace & Company
		Lindsay Chemical Division American Potash & Chemical Corporation
		Union Carbide Metals Company Division of Union Carbide Corporation
Cm_2O_3	Fe_2O_3 (Fe-56)	U. S. Steel
	Ce_2O_3 (Ce-140)	Same as for Gd_2O_3
Pu-238	Sm-152	Same as for Gd_2O_3
	Al	Aluminum Company of America
	Mg	Dow Chemical Company
	Sb	American Smelting & Refining Company
UO_2	HfO_2 (Hf-178)	Chemical Metallurgy Branch Metallurgy Division Naval Research Laboratory
Cs	Cs (Cs-133)	Dow Chemical Company Midland, Michigan
Ce_2O_3	Ce_2O_3 (Ce-140)	Same as for Gd_2O_3

Cost of Materials (1961)

<u>Element</u>	<u>Cost (\$/lb)</u>
Al	0.26
Sb	0.29
Bi	2.25
Ce-144	100.00
Cs-137	100.00
Gd	275.00
Au	510.00
Hf	30.00
Fe	0.062
Pb	0.118
Mg	0.35
Mn	0.45
Pu-238	12,700.00
Sm	275.00
Ta	35.00
Te	3.50
Ti	7.65
U-238	18.18

2. Facilities and Instrumentation for Ablation Testing

Two broad methods for obtaining particulate characteristics on burnup may be considered; each requires different equipments and facilities:

- (1) To conduct actual atmospheric re-entry burnup tests; subsequently, to sample and analyze debris. (This method was studied in compliance with a contractual obligation to evaluate

possible means for acquiring data on burnup debris characteristics; the method was rejected in favor of the approach in the following item.)

- (2) To conduct simulated re-entry experiments, using the best laboratory facilities and instrumentation available.

- a. Atmospheric re-entry testing

In an actual re-entry there is every certainty that the free flow and environmental conditions will be met and that the debris products will be representative. The difficulty lies in obtaining valid data on the process and the final products. Droplet breakup cannot be observed, nor can the velocity profile of individual small particles be obtained. No equipment exists capable of obtaining such refined data at such distances. The best that can be hoped for is to obtain a sample of the final products, a long range photographic record of the event, and perhaps an estimate of the ion density, in the debris trail, from radar cross-section measurements. However, the following means for obtaining a meaningful sample were investigated:

Case 1: Sending a sample-collecting vehicle along the same trajectory as the re-entry.

Case 2: Having the re-entry rocket vehicle carry a sampler which it ejects behind the fuel core.

Case 3: Collection of re-entry debris when it has descended to altitudes where conventional sampling is feasible.

- (1) Case 1.

There is only one operational system, capable of sampling above 70,000 feet, which can also be directed along a predetermined trajectory. This is the rocket-borne cryogenic sampler developed by the Air Force Cambridge Research Center. To date, this device has suffered two failures of the launch vehicle and has not been flown successfully. The accuracy with which the rocket can be expected to deliver the sampler to an intercept with the re-entry burnup cloud is, of course, not defined. An alternative to this approach does exist but is futuristic at present. When steerable vehicles become available for sampling in the mesosphere, placement accuracy problems probably can be overcome. The X-15 rocket plane has recently demonstrated limited capabilities in this respect.

- (2) Case 2.

It may be possible to send a collector along in the rocket, later

ejecting it behind the re-entry body (test specimens). The problem then becomes one of avoiding re-entry destruction of the sampler, yet having it follow the high energy trajectory of the ablated re-entry body. There remains yet another problem of more subtle nature; even though the sampler itself may be cooled or otherwise designed to withstand re-entry, all the material that it collects will have to pass through the sampler's shock wave and be exposed to its stagnation temperatures and pressures. If this problem is overcome, there remains the question of data interpretation; there will be confusion as to stage in the ablation process at which the material was collected. It is concluded that the design of a really effective collector of this type, capable of obtaining worthwhile data, is beyond the scope of this report. Discussions with Goodyear Aircraft Corporation did, however, elicit interest in investigation of such a device.

(3) Case 3.

This technique would have considerable merit, provided that:

- (1) The diffusing, sedimenting mass of re-entry debris can be intercepted and sampled in the lower altitudes.
- (2) Enough material can be carried aloft to produce a sufficient concentration of debris at the lower altitudes. Determination or prediction of these factors would require use of a computer program, such as discussed in Ref. 36, for prediction of fallout patterns. A very rough approximation, based on the simple equations developed in Section B, Topic 3, of this chapter, indicates that about 4.2 pounds of gold would be required if the particulate resulting on burnup were of 0.1-micron diameter or less: 33 pounds would be required if the particulate diameter were 1.0 micron.

It was concluded that actual re-entry experiments cannot be used at present to define debris characteristics with sufficient accuracy.

b. Laboratory testing

Excellent ground facilities exist for simulation of re-entry free stream conditions. These are the hypervelocity ranges developed for study of hypersonic aerodynamics, re-entry heating, ionization trails, etc. One of the more advanced facilities is that of the Canadian Armaments Research and Development Establishment (CARDE, Ref. 37). This facility uses a light-gas gun to accelerate objects into a controlled environment range. Required particle velocities and altitudes can be simulated. Much of the photographic and velocity measurement equipment required is available.

Other facilities possessing in whole, or part, these same test capabilities include:

- (1) Aberdeen Proving Ground, Maryland.
- (2) Ames Laboratories, California.
- (3) Cornell Aeronautical Laboratory, Inc., New York.
- (4) AVCO Corporation, Massachusetts.

The one disadvantage of facility testing in comparison with actual re-entry conditions is the diameter and length of the environmental range: at CARDE, the largest, it is 2 to 3 feet in diameter and 50 feet long. The study of small particle ablation has shown that a 100-micron plutonium droplet would require about 185 meters of range to complete ablation at an initial velocity of 8 km/sec at an altitude of 200,000 feet. A 2-micron droplet requires only 68 meters under the same conditions. Hence, it will be impossible to complete particle ablation in the 50-foot range. This may not be a serious disadvantage. The process of ablation is revealed by measurement of the ablation rate; coupled with the velocity profile, it can be correlated to the analytical model for extrapolation to end conditions.

The size of the range will prevent the free expansion of debris to the degree that would occur in the atmosphere. At present, it is not intended that data be collected on nucleation and agglomeration of vapor. Tests for this type of data will be run in controlled-atmosphere boxes in which test materials in wire form are vaporized by a large pulse of electrical energy (resistance heating).

B. FEASIBILITY OF PARTICULATE FATE EXPERIMENTS

These experiments are those pertaining to observation of vapor trails deployed from rockets, and the gross sampling program involving particulate injection and sample collection and analysis. Investigations are reported in the following sequence:

- (1) A materials study made to determine the feasibility of producing a visible vapor trail for diffusivity measurements.
- (2) Sample analysis techniques.
- (3) Test materials.
- (4) Collection devices.

- (5) Capability of collection device delivery systems, including rockets, balloons and aircraft.

The feasibility of the gross sampling program is a function of mutual interdependence of Items (2), (3) and (4); the compatibility of test materials, collection devices and sample analysis techniques must be adequate to the needs of the test.

1. Materials for Diffusivity Experiments

Particulate materials which, when released at high altitude, will disperse and be made visible by reflected sunlight are needed for the diffusivity experiments. Such clouds are observed by optical and photographic methods; hence, an analysis of the visibility of various particulate materials was necessary. The materials selected for evaluation were:

<u>Material</u>	<u>Color</u>	
Aluminum oxide	White	
Iron oxide	Red	
Chromium oxide	Green	
Tungsten oxide	Yellow	
Cobalt oxide	Blue	
Pemco stain GS 941	Red	} Industrial ceramic pigments produced by The Pemco Corporation.
Pemco stain GS 931	Yellow	
Pemco stain GS 205	Green	

The visibility analysis of these materials was performed by The Geophysics Corporation of America (Ref. 38) under subcontract to Martin Marietta on a task requiring similar information. Selection of one material over another was based principally on albedo (light reflection efficiency) and the degree to which scattered light contrasted with background sky light. Sky light is predominately blue, and, because solar intensity is greatest at a wavelength of about 0.55 micron, the best contrast is obtained with light scattered in the yellow, red and near-infrared portion of the spectrum. Presumably, any of the materials listed could produce a cloud detectable from the ground

if released in sufficient quantity. However, as a practical matter, the weight of material to be injected must be considered. On the basis of weight, albedo and contrast with background, the following were selected as most appropriate for use:

<u>Material</u>	<u>Color</u>	<u>Average Particulate Size (microns)</u>	<u>Theoretical Density (gm/cm³)</u>	<u>Minimum Amounts Required for Viewing (gm)</u>
Al ₂ O ₃	White	0.5 to 1	3.9	200
Pemco red stain CS 941	Red	1 to 2	4.25	600

Another material not covered in GCA's study is believed to be applicable. The material is foamed plastic smoke originally developed by B. L. Raskin of Johns Hopkins University Radiation Laboratory. A report on this material is found in Ref. 33. Dow Chemical Company has exclusive rights to its commercial development. The advantages of this material for use as a visible diffusion tracer are:

- (1) Extremely low particle density--0.002 gm/cm³, average (low payloads for large clouds).
- (2) High surface area per unit weight.
- (3) High albedo (for certain plastics).

2. Sample Analysis--Gross Sampling Experiment

Before a direct examination of materials could be attempted, it was necessary to study the various available techniques of quantitative fractionation of gross aerosol samples. It is necessary to choose the most sensitive method available to reduce the amount of material required for injection, yet permit quantification of extremely low concentrations of test materials.

a. Trace analysis techniques

Use of radioactive isotopes as test materials was considered inadvisable and impractical. Recourse to sample analysis techniques applicable to nonradioactive materials was therefore necessary. The available literature on the subject was surveyed, and opinions and technical data were solicited from various organizations having experience in analysis of aerosol samples. It was concluded that activation analysis is the most appropriate technique. This conclusion was motivated principally by:

- (1) Reference 39, where analysis techniques are rated by sensitivity as follows:

<u>Method</u>	<u>Detectable Tracer Concentration by Weight (%)*</u>
Gravimetric-volumetric	10^{-2}
Colorimetric	10^{-5}
Spectrographic	10^{-5}
Mass spectrographic	10^{-5}
Activation analysis	10^{-8}

- (2) The technical contribution of Tracerlab, Incorporated, recommending neutron activation analysis augmented by isotopic dilution mass spectrometry as the most sensitive technique applicable to the program. This document is included as Appendix B of this report.

b. Neutron activation analysis

Activation analysis sensitivity is not the same for all materials but depends on several factors:

- (1) The neutron activation cross section of the element.
- (2) The flux level and energy spectrum of the neutron source.
- (3) The half lives of the isotopes formed upon irradiation.
- (4) The length of time material is subjected to the neutron flux.
- (5) The time lapse between completion of activation and measurement.
- (6) The nature of the radiations produced by the primary isotope.
- (7) The sensitivity of the measuring (counting) equipment.

*Order of magnitude, material dependent.

- (8) The method of standardization and calibration of the counting equipment.
- (9) The amount of background materials in the sample.
- (10) The amount of interfering radiations produced by activation of materials in the gross sample.

Under the best conditions, it is possible to detect quantities of the order of 10^{-14} gram. This would apply to gold irradiated to saturation in a neutron flux of 2×10^{14} nv. The resulting one-day-old activity of the 10^{-14} gram would be about 10 counts per minute. Computations of minimum detectable quantities of most of the elements can be found in Ref. 40.

Sources of activation neutrons and their flux levels, as given in Ref. 39, are as follows:

<u>Neutron Source</u>	<u>Thermal Neutron Flux (nv)</u>
Reactor	10^{10} to 10^{14}
Cyclotron	10^8 to 10^{11}
Van der Graaff	10^6 to 10^{10}
Ra - Be, 1 gm	10^4 to 10^5
Sb - Be, 1 curie	10^3 to 10^4
Ra - Be, 25 mgm	10^2

A reactor provides the greatest flux obtainable and is the recommended source. The MTR is reportedly capable of about 5.5×10^{14} nv. Test material selection should, in part, be predicated on travel time from the reactor site to the analysis facility.

3. Selection and Preparation of Materials for Gross Sampling Program

Material requirements may be listed as follows:

- (1) The material must be detectable in small quantities in order to reduce the amount of material that need be injected into the atmosphere.

- (2) Each material must be quantitatively distinguishable from background particulates such as those present in dust from earth and meteors, sea salt spray, fission products, etc., and from each other (in all likelihood more than one tracer will be used).

a. Material selection

The most sensitive method found for aerosol quantification in a gross sample was that of neutron activation analysis. Table 5 lists elements detectable in quantities of the order of 10^{-7} gram.

Materials selected from Table 7 should be low in natural occurrence and in the yield of fission weapons. Table 8 and Fig. 10 show the natural occurrence and the fission yield of the elements. Table 9 lists those elements found in Table 8 that are low in both natural occurrence and fission yield.

Gold, tantalum, thulium, holmium and terbium appear to be very desirable materials for the program. Tracerlab, Incorporated, was asked to submit a list of 20 possible tracers; these are listed in Table 10 and Appendix B, not necessarily in the order of preference. Considerations such as those attached to Table 9 and also of cost, availability, density, etc., will afford a basis for final selection.

TABLE 7

Detectability of Elements

<u>Element</u>	<u>Minimum Amount Detectable* (μgm)</u>
Sodium (Na)	1.6×10^{-7}
Lithium (Li)	2.4×10^{-8}
Phosphorus (P)	2.4×10^{-7}
Scandium (Sc)	2.6×10^{-8}

TABLE 7 (continued)

<u>Element</u>	<u>Minimum Amount Detectable* (μgm)</u>
Cobalt (Co)	5.3×10^{-7}
Copper (Cu)	2.4×10^{-7}
Gallium (Ga)	1.5×10^{-7}
Palladium (Pd)	1.6×10^{-7}
Cadmium (Cd)	6.9×10^{-7}
Indium (In)	1.5×10^{-7}
Antimony (Sb)	5.2×10^{-8}
Cesium (Cs)	4.1×10^{-8}
Lanthanum (La)	3.3×10^{-8}
Praseodymium (Pr)	3.3×10^{-8}
Promethium (Pm)	5.6×10^{-9}
Samarium (Sm)	9.9×10^{-9}
Europium (Eu)	2.4×10^{-9}
Gadolinium (Gd)	6.1×10^{-7}
Terbium (Tb)	2.9×10^{-8}
Dysprosium (Dy)	4.8×10^{-7}
Holmium (Ho)	5.4×10^{-9}
Erbium (Er)	1.2×10^{-8}
Thulium (Tm)	1.3×10^{-8}
Ytterbium (Yb)	3.0×10^{-8}
Hafnium (Hf)	2.5×10^{-7}

TABLE 7 (continued)

<u>Element</u>	<u>Minimum Amount Detectable* (μgm)</u>
Tantalum (Ta)	2.0×10^{-8}
Tungsten (W)	5.5×10^{-8}
Gold (Au)	1.2×10^{-8}
Mercury (Hg)	6.4×10^{-7}
Thorium (Th)	1.1×10^{-7}
Uranium (U)	1.2×10^{-8}
Neptunium (Np)	4.2×10^{-9}
Plutonium (Pu)	9.8×10^{-9}
Manganese (Mn)	5.6×10^{-8}

*All samples irradiated to saturation or 30 days in neutron flux of 4×10^{14} nv; minimum count rate 10 cpm at 1 day (Ref. 40).

TABLE 8
Element Abundance in Earth's Crust

<u>Element</u>	<u>Part of Total (%)</u>	<u>Element</u>	<u>Part of Total (%)</u>
O	46.43	Ce	0.00461
Si	27.77	Sn	0.004
Al	8.13	Li	0.003
Fe	5.12	Y	0.00281
Ca	3.63	Nd	0.00239
Na	2.85	Co	0.0023
K	2.60	Hf	0.002

TABLE 8 (continued)

<u>Element</u>	<u>Part of Total (%)</u>	<u>Element</u>	<u>Part of Total (%)</u>
Mg	2.09	La	0.00183
Ti	0.629	Pb	0.0016
P	0.130	Th	0.00115
H	0.127	Be	0.001
Mn	0.096	Mo	7.2×10^{-4}
F	0.077	Sm	6.47×10^{-4}
Cl	0.055	Gd	6.36×10^{-4}
S	0.052	Pr	5.53×10^{-4}
Ba	0.048	As	5.0×10^{-4}
Cr	0.037	Ga	5.0×10^{-4}
Zr	0.028	Sc	5.0×10^{-4}
C	0.027	W	5×10^{-4}
V	0.021	Dy	4.47×10^{-4}
Sr	0.018	U	4×10^{-4}
Ni	0.008	B	3×10^{-4}
Cu	0.007	Rb	3×10^{-4}
N	0.00463	Yb	2.66×10^{-4}
Er	2.47×10^{-4}	Ir	1.0×10^{-6}
Br	1.62×10^{-4}	Rh	1.0×10^{-6}
Ho	1.15×10^{-4}	Te	1.0×10^{-6}
Eu	1.06×10^{-4}	Au	5×10^{-7}
Ge	1.0×10^{-4}	Re	1.0×10^{-7}

TABLE 8 (continued)

<u>Element</u>	<u>Part of Total (%)</u>	<u>Element</u>	<u>Part of Total (%)</u>
Sb	1.0×10^{-4}	Ra	1.0×10^{-9}
Zn	1.0×10^{-4}	Po	7×10^{-13}
Tb	9.1×10^{-5}	Rn	2×10^{-14}
Se	8×10^{-5}	Pm	3×10^{-19}
Lu	7.5×10^{-5}	Hg	5×10^{-5}
I	3×10^{-5}	Bi	2×10^{-5}
Tm	2×10^{-5}	Pt	2×10^{-5}
Cd	1.5×10^{-5}	In	1.0×10^{-5}
Tl	1.0×10^{-5}	Os	5×10^{-6}
Pd	5×10^{-6}	Ru	5×10^{-6}
Ag	4×10^{-6}	Nb	4×10^{-6}
Ta	1.2×10^{-6}		

TABLE 9

Elements Low in Fission Yield and Natural Occurrence

<u>Element</u>	<u>Fission Yield (%)</u>	<u>Natural Occurrence</u>	<u>Amount Detectable (μ gm)</u>
Scandium	Not found	5×10^{-4}	2.6×10^{-8}
Cadmium	0.05	1.5×10^{-5}	6.9×10^{-7}
Indium	0.01	1×10^{-5}	1.5×10^{-7}
Antimony	0.01	1×10^{-4}	5.2×10^{-8}
Europium	0.01	1.06×10^{-4}	2.4×10^{-9}

TABLE 9 (continued)

<u>Element</u>	<u>Fission Yield (%)</u>	<u>Natural Occurrence (%)</u>	<u>Amount Detectable (μgm)</u>
Gadolinium	0.01 or less	6.36×10^{-4}	6.1×10^{-7}
Terbium	0.0005	9.1×10^{-5}	2.9×10^{-8}
Dysprosium	Negligible	4.47×10^{-4}	4.8×10^{-7}
Holmium	Negligible	1.15×10^{-4}	5.4×10^{-9}
Erbium	Negligible	2.47×10^{-4}	1.2×10^{-8}
Thulium	Negligible	2×10^{-5}	1.3×10^{-8}
Ytterbium	Negligible	2.66×10^{-4}	3.0×10^{-8}
Hafnium	Negligible	2×10^{-3}	2.5×10^{-7}
Tantalum	Negligible	1.2×10^{-6}	2.0×10^{-8}
Tungsten	Negligible	5×10^{-4}	5.5×10^{-8}
Gold	Negligible	5×10^{-7}	1.2×10^{-8}

TABLE 10

Twenty Possible Tracers (See Appendix B)

<u>Tracer Element</u>	<u>Chemical Form*</u>
Manganese	MnO_2
Cobalt	CoO
Cadmium	CdO
Antimony	Sb_2O_3
Lanthanum	La_2O_3
Praseodymium	Pr_2O_3

TABLE 10 (continued)

<u>Tracer Element</u>	<u>Chemical Form*</u>
Samarium	Sm_2O_3
Hafnium	HfO_2
Tantalum	Ta_2O_3
Tungsten	WO_3
Thorium	ThO_2
Indium	In_2O_3
Gadolinium	Gd_2O_3
Mercury	HgS
Gold	$\text{Au}_2\text{S}_3, \text{Au}$
Palladium	Pd
Europium	Eu_2O_3
Holmium	Ho_2O_3
Gallium	Ga_2O_3
Uranium-238	UO_2

*Analytical sensitivity based on following:

- (1) Thermal neutron flux 4.3×10^{14} nv.
- (2) Count rate of isolated nuclide is 10 cpm at optimum counting time, using optimum equipment and measured efficiencies.
- (3) Irradiation to saturation or 30 days, whichever is first.
- (4) Quantitative recovery in chemical separations.

Fig. 10. Fission Yield as a Function of Mass Number

b. Quantity of pure element tracer required

If a cloud of expanding-diffusing particles is sampled, it will be necessary to measure the naturally occurring background in advance. Samples taken at constant altitude to define the extent of cloud growth will, of necessity, include several taken where the cloud has not yet mixed; this minimum can be taken as the background level. Therefore, the main consideration is that the quantities injected be sufficient to create a measurable increase over background until diffused to world-wide proportions. Where possible, the naturally occurring background should be matched world-wide; where this is not possible, the highest amount above the material's minimum detectable increment over background (compatible with an economic injection vehicle) should be used.

Sample calculation for gold. The following assumptions should yield conservative results:

- (1) The material is spread uniformly, in terms of volume, through the atmosphere in the altitude region 0 to 200,000 feet.
- (2) The sampling device samples 1000 standard cubic feet of air at sampling altitude per flight.
- (3) The minimum detectable quantity of material is given by the activation analysis sensitivity appearing in Table 7.
- (4) One million tons of particulate material reside in the total atmosphere, in which the occurrence of the elements is the same as in the earth's crust, as given by Table 8.

The number of sample aliquots lying between 0 and 200,000 feet is given approximately by

$$\text{Number of aliquots} = \frac{4 \pi r^2 \Delta h \bar{\rho}}{1000 \text{ scf } \rho_0}$$

where

$$r = 2.11 \times 10^7 \text{ ft}$$

$$\Delta h = \text{altitude range}$$

$$\bar{\rho} = \text{mean density in that altitude range}$$

$$\rho_0 = \text{density of air at sea level.}$$

Altitude Range (ft)	Number of Aliquots ($\times 10^{16}$)
0 to 10,000	4.8
10,000 to 20,000	3.5
20,000 to 30,000	2.5
30,000 to 50,000	2.8
50,000 to 100,000	1.3
100,000 to 200,000	1.0
Total	1.59×10^{17}

The activation analysis sensitivity for gold is assumed to be 1.2×10^{-14} gram. Hence, if every sample aliquot contains this much material, the total quantity is $(1.59 \times 10^{17})(1.2 \times 10^{-14}) = 1.91 \times 10^3$ grams, or 1.91 kgm. Assuming each sample aliquot to contain at least one particle gives a maximum particle mass of 1.2×10^{-14} gram. Such a gold particle would have a maximum diameter of 0.1 micron. If the total atmosphere contains 10^6 tons (approximately 9×10^{11} grams), and the natural occurrence fraction of gold in this mass is 5×10^{-9} , then the total mass of gold in the atmosphere is 4.54 kgm. Most of this naturally occurring gold resides in the troposphere; therefore, an injection at burnup altitudes of an additional 1.91 kgm of 0.1-micron gold should prove ample.

With the same assumptions and analysis, the injection recommended for tantalum is 15.9 kgm. If thulium were used and the background matched, 260 kgm would be required. However, the sensitivity of activation analysis for this element is almost the same as for gold; hence, the minimum detectable increment of mass is approximately 1.9 kgm. The largest amount above 1.9 kgm compatible with cost, vehicle capacity, etc., should be used.

c. Nonpure tracer elements

Nonpure tracer elements offer a statistical advantage in discriminating against background.

(1) Alloys

The advantage of the alloy is that not one, but two or more elements are present to give positive identification to the particle during gross

sample analysis. The alloy is assumed perfectly homogeneous for all particles. A disadvantage is that more alloy material would be needed per injection to obtain analytical sensitivity equal to that obtained with pure tracers. In addition, due to the limited number of good nonradioactive tracer materials available, it is probable that the requirements for numerous injections of different tracers will quickly exhaust the alloys available.

(2) Coated particles

If the tracer element particles can be individually coated with a protective material, it may be possible to remove the background component of the element from gross aerosol samples without removing the tracer particles. This could be done through use of a solvent that does not attack the coating, but would dissolve and allow separation of the naturally occurring compounds of the tracer element. Opinions were solicited from various companies engaged in production of colloids and protective coatings as to the feasibility of providing such coatings for batches of particles in the 0.1- to 10-micron size range. Their responses follow:

- (1) Union Carbide Chemicals Company--"The most promising approach would seem to us to be the spray drying of a suspension of the powdered metal in a dilute solution of a resin chosen for its chemical resistance and with sufficient hardness to avoid subsequent tendency to agglomerate. By varying resins and solvents, the technique should be applicable to a variety of metal particles." (Ref. 41)
- (2) Acheson Colloids Company--"We think you have an interesting project in attempting to obtain large quantities of 0.1- to 25-micron coated metallic particles. Especially, the lower size range would be difficult to coat. The upper range might well be made by running a slurry of fine particles in a binder solution through a spray-dry setup. However the droplet size of such a piece of equipment would make impossible the obtaining of any particles much below 10 microns in size. Coacervation techniques might also be employed.

"The ideal way to make the finer ones would seem to be to take particles of this known size and to coat them by solution chemical deposition with a protective, plated layer. Thus it might be possible to take a common metal and to plate it with gold or silver from solution. Such coatings would be extremely durable and chemically resistant. If we assume then that the particles are one metal plated with another for protection, there certainly might be some tendency for the

coated particles to agglomerate. They could be dispersed into the atmosphere by using suitable nebulizer techniques such as have been employed in the seeding of clouds and in the dusting process using insecticides.

"The only other type of coating which could possibly be considered would be organic-type protective films such as lacquers and resin systems. However, we feel that these would be practically impossible to handle in the small particle size range because of the limitation of methods of coating. Also, they would definitely lead to further problems in dispersing of the individual particles." (Ref. 42)

- (3) Kahlenberg Laboratories--"We find your letter of interest because we have made small particles for many years, particularly gold although we can also make other noble metals in colloidal form. We have made gold particles where the over-all dimension was 0.017 microns and have been able to do this consistently. Such particles would be small enough to be coated with a layer resistant to ultra violet light and ozone." (Ref. 43)

d. Preparation of submicron-sized particulate

Methods for reduction of bulk materials to powder form include:

- (1) Grinding--Grinding in ball or rod mill is applicable to all brittle and friable materials; it is suited particularly to metal oxides and other compounds. The particles resulting from grinding are extremely rough and irregular in shape and size, and require a separation technique for production of narrow particle size ranges.
- (2) Filing--This method would be applicable, in general, to both metals and oxides. The particles resulting (filings) will have a size distribution somewhat controlled by the coarseness of the file used; particle shapes tend to be very rough and irregular. Production of narrow or controlled particle size distribution requires careful selection of file roughness and probably an additional fractionation and separation process.
- (3) Condensation of vapor--A common method of forming metal colloidal particles is through use of an underwater arc whose electrodes are composed of the solid material. The arc vapors condense in the water and form very small spherical particles ranging from about 0.1 micron downward. A fractionation-separation process is required to produce controlled particle size ranges.

- (4) Spheroidation of small particles--Irregularly shaped particles can be made to assume a spherical shape by slight heating above their melting points. During the heating they are suspended as an aerosol in, or falling through, a gas. The surface tension of the melt pulls the particle into the spherical form which is retained upon cooling.

e. Preparation of particulate in known size distribution

Several fraction-separation techniques for preparation of large samples of controlled particle sizes in the 0.01- to 10-micron range are outlined:

- (1) Sedimentation methods--Particulate of different sizes suspended in a viscous media will, when subjected to accelerating forces, tend to separate according to mass (size). Many variations in application of this principle are found in the literature. All rely on drag force per unit mass to distribute particulate according to size. The method is well suited for separation of small particles into discrete size ranges. Commercial particle size classifiers operating on this principle are capable of providing closely controlled particle size fractions to less than 10 microns, average particle size. The limiting average particle size is about 1 micron. Slightly improved techniques could probably reduce this limit, especially under laboratory conditions. The materials-handling capacity of one of the more advanced commercial machines (the Sharples K-8 Super Classifier) ranges from 10 to 250 pounds of raw material per hour, depending on particle size and density.
- (2) Cascade filtration--Further fractionation of small particle size materials can be accomplished by passing the material through a succession of filter stages of decreasing pore size. The particle size of material trapped by a given stage would be less than the pore size of the preceding stage and greater, on the average, than the pore size of the stage that retains it. Due to the ease with which small pore size filters are clogged, this method can be used only for preparation of very small samples.
- (3) Air blast suspension--This is a variant of the sedimentation technique. The gross sample containing many different particle sizes is placed in a vertical air jet; depending on the flow velocity used, the smaller particles will be supported by the air stream while the larger particles fall from it. Velocity is varied to separate the gross sample into samples of specific size ranges.

4. Sample Collectors for Gross Sampling Experiments--Applicability and Use

This section reports the results of a literature search performed on particulate samplers, operational, developmental and theoretical-experimental. Conclusions are drawn on each sampler as regards applicability to this experiment. In addition, a brief description is given of a sampling technique believed applicable, but not disclosed by the literature.

The means of sampler delivery to the collection point was also investigated; data pertaining to use of the sampler are reported.

a. Operational particulate collection devices

Sample collectors reviewed are only crudely separable into operational, developmental and theoretical-experimental categories. However, for classification purposes, they are reported in this manner.

(1) The Goetz aerosol spectrometer

This sample collector is a power-consuming device dependent upon centrifugal force to separate and deposit particulate, preferentially, according to mass. There are conflicting opinions as to its usefulness in high altitude sampling. Atlantic Research Corporation, predicating operation on placement of the sampler (at high altitudes) by rocket, with subsequent sampling during descent by parachute, concluded that the Goetz instrument samples too small a volume of air for meaningful results. Western Precipitation Corporation concluded, however, that the device was suitable for use to 100,000 feet if balloon-carried on flights of about 100 minutes. Power requirements, to obtain high rotational speeds, are relatively large. This is, however, a design variable depending on particle size to be sampled, concentration (flow rate required), and desired collection efficiency. Atlantic Research Corporation calculations show that a 26,000-rpm device absorbing approximately 20 hp will sample 0.1-micron particles with 100% efficiency.

Conclusion. A specific unit for sampling a predetermined tracer particle size with 100% efficiency, designed for balloon operation, could possibly be developed for use in this experiment. Whether a sufficiently large air volume (1000 SCF) can be sampled remains in doubt.

(2) Rectangular cascade-type jet impactor

This balloon-borne particle collector has been fabricated, calibrated and experimentally tested by General Mills, Inc. It operates as follows:

- (1) The system consists of four collection units, each of which is designed to operate at one of four altitude ranges: 30,000

to 50,000 feet; 50,000 to 70,000 feet; 70,000 to 90,000 feet; or 90,000 to 100,000 feet.

- (2) Each collection unit consists of three stages in series--two impaction stages in which particles are collected on coated slides, and a third stage consisting of a backup filter. The ambient air flow rate is $22 \text{ ft}^3/\text{min}$; an instrument pack is carried to record variations in the rate.
- (3) A 3-hp direct-current motor drives a rotary pump connected to the four collection units. It is air-cooled and rated for use at 100,000 feet and has been tested to 95,000 feet.
- (4) Operating performance at 65,000 feet for particulate of $4.4\text{-gm}/\text{cm}^3$ density is as follows:

Collection Efficiency (%)	Diameter of Particle Collected (microns)	
	First Stage	Second Stage
0	0.0945	0.0022
25	0.157	0.0046
50	0.179	0.0068
75	0.202	0.0092
100	0.30	0.0146

Conclusion. With some redesign, this device should be applicable to this program. Increased flow rates are required as it is believed a necessity that 1000 SCF be sampled. General Mills believes this might be possible at altitudes exceeding 100,000 feet. The constant-volume pump can be replaced by multistage turbines, producing high flow rates. This is made possible because impaction efficiency increases at low ambient density even though jet velocities are lower. However, blower motors themselves give problems at these altitudes unless pressurized and sealed; this, in turn, leaves a heat dissipation problem to be solved.

(3) Submicron aerosol collector (SMAC)

A balloon-borne filter sampling unit developed for the Air Force Cambridge Research Laboratory (AFCRL) by General Mills. A single-stage Farrington blower is driven by a 0.5-hp direct current motor at a constant 13,000 rpm. It delivers approximately $500 \text{ ft}^3/\text{min}$

at 80,000 feet, and $300 \text{ ft}^3/\text{min}$ at 90,000 feet. Linear flow velocities in the AFCRL polystyrene filter must be quite low; therefore, a 10-ft^2 filter mat is used.

Conclusion. This device will be useful to perhaps 115,000 feet, where flow rates are reduced and blower motor problems could occur. Payload weight is high (140 to 150 pounds) and would be increased to give adequate flow rates much above 100,000 feet. Ballooning costs increase rapidly with payload and altitude.

(4) Direct flow air sampling unit

This sampler, a balloon-borne filter sampling unit, was developed for the Atomic Energy Commission by General Mills, Inc. It differs from the SMAC unit only in the filter used; it is more porous and is dependent on high linear velocities ($\sim 500 \text{ ft/min}$) to develop high efficiencies ($\sim 100\%$). For this reason, the cross-sectional area of the filter is only 1 ft^2 . The weight of the unit is 140 pounds.

Conclusions. Those with respect to SMAC apply.

(5) The Ash can sampler

A balloon-borne, filter-paper type device developed by General Mills, Inc., this unit is used by the Atomic Energy Commission. Since it is completely operational, its design and performance are not reported here.

Conclusion. This device could prove useful in this experiment. One disadvantage is the large filter area over which the collected sample is distributed. The disadvantage creates a problem in sample recovery for analysis. The sample, ideally, should be recovered without changing the physical characteristics of the tracer. If the filter is ashed or dissolved, it must be without harm to the tracer, and the filter must contain no background material which can act to confuse results of the trace analysis.

(6) Bag whole-air sampler

This sampler is a balloon-borne device developed and operated by General Mills, Inc. It has four units: a lift balloon, a collection bag, an armored vessel to which the air is transferred from the collection bag, and the necessary control system. This unit, too, is used by the AEC and is therefore not reported in detail.

Conclusion. This appears to be the best operational device available for sample collection between aircraft altitude and approximately 100,000 feet. The particulate sampled is not changed in physical characteristics,

and the air volume sampled can be made to equal or exceed 1000 SCF.

(7) Chemical absorbent particle sampler

A balloon-borne device, this sample collector is used by the Geophysics Research Directorate of the Air Force Cambridge Research Center. The apparatus consists of a motor-driven rotary air pump drawing air through a series of tubes containing selective absorbent material to remove the desired component from the air stream. As many as eight components are sampled. Power requirements are low--0.01 hp, and sampling rate is low--0.5 ft³/min.

Conclusion. The device does not appear attractive for this experiment. The tracer elements or compounds used do not appear suitable for the device. The flow rate of the collector is so low as to require unreasonable sampling times.

(8) Crystalline zeolite absorbent collector

This balloon-borne device was developed and tested by General Mills, Inc. The Zeolite absorbs 100 SCF of ambient air at a rate of 1 SCF/min, at 130,000 feet, if the zeolite bed is maintained at -196° C. Eighty-five pounds of zeolite are used; the total payload is 200 pounds.

Conclusion. This is the only device operational to 140,000 feet. It will be useful in this experiment for sampling close to the injection point for a short period after the injection. It is not applicable after diffusion of the injected cloud necessitates that a sample larger than 100 SCF be obtained for good statistical data.

b. Developmental particulate collection devices

(1) The cryogenic air-sampling rocket

This developmental device is a rocket-borne and incorporated whole air sampler used by the Geophysics Research Directorate of Air Force Cambridge Research Center. At prescribed points in the trajectory, the collector opens, samples, is sealed and is returned to earth by parachute. The volume sampled is dependent on the trajectory flown, the altitudes of opening and sealing of the collector, and the flow characteristics of the collector inlet and internal volume (dependent upon ice accretion and entrainment of noncondensable gases).

Conclusions. Test results are necessary. The launch vehicle, an Aerobee 100, has failed on the first two attempts. When fully developed, this collection device could provide data in the altitude range 130,000

to 270,000 feet and be useful to the program. One apparent disadvantage is that the tracer particulate collected will be along a trajectory, rather than at a point in space. This could complicate data reduction and reduce reliability of results.

(2) The electrostatic stratospheric dust sampler

This is a balloon-borne device most advanced in design and testing by Del Electronics Corporation. Appendix C is a technical statement on the status and capabilities of this device. It was solicited from Del Electronics as feasibility support of this study.

Conclusions. The device, when operational at the level of performance quoted in Appendix C, may be useful in this program. The limitations on tracer particle size could prove troublesome. Efficiency is dependent on particle size and appears to drop off at diameters above 0.1 micron. Blower motor problems, similar to those of other systems operating much in excess of 100,000 feet, can be expected. The effect of corona discharge on tracer physical characteristics must be determined. The results of sample analysis could be confused if the tracer is altered.

(3) A filter collector with corona discharge

This collection device was designed for balloon operation by General Mills, Inc. The air sample is drawn through the corona discharge, then through two layers of filter paper. Agglomeration of particulate due to particle charging in the corona increased efficiency of filtration by approximately 30% at flow rates of 400 ft/min.

Conclusions. Applicability to this experiment is subject to all the limitations of paragraph (2) above, except perhaps particle size. If the shortcomings derived of flow rate limitations could be avoided, it is likely that no requirement for corona discharge would exist. A pure filter system would then be satisfactory.

c. Theoretical-experimental particulate collection devices

(1) The focusing array impactor (rotorchute)

This rocket-launched device was developed in theory, and laboratory-tested in part, by the Atlantic Research Corporation. The focusing array of impactors is mounted on five rotating arms 14 feet in diameter. The rotational velocity developed in free fall and the radius of mounting develop the impact velocity. The unit is designed to sample 1000 SCF between 120,000 and 100,000 feet. A device of similar operating theory is under development at General Mills, Inc.; it uses a motor-driven, high speed fan about one foot in diameter. The fan blades form the impactor array.

This unit would be balloon-borne to obtain an adequate sampling volume. In both devices, efficiency of collection for a given design is dependent on altitude of operation; it increases with particle size and rotational velocity. Values to 80% have been measured.

Conclusions. The rotorchute has the disadvantage of collecting across a span of altitude rather than at a single point; this will reduce reliability of reduced data. However, the particle characteristics are retained, and the sample can be cleanly recovered for analysis. Furthermore, at least 1000 SCF can always be sampled by adjusting the altitude range of fall. This device should prove useful to this experiment when developed. The General Mills device suffers the motor problems and sampling volume limitations of all powered designs. No future is seen for this design in this program.

(2) The thermal precipitator collector

This device was studied by Western Precipitation Corporation and is currently under evaluation by Georgia Institute of Technology. It operates because particulate will move where a thermal gradient exists. The air is moved into the device past a hot surface; the particulate migrates to a cooled collecting surface. Efficiency increases with increased altitude (reduced operating pressure) and increased thermal gradient.

Conclusion. Little future is seen for this device in this program; power requirements are high and might profitably be used to move air rather than to heat plates. This conclusion could prove invalid if the motor problem is not solved and if the hot and cold surfaces can be so arranged as to induce adequate flow, yet retain acceptable precipitating efficiency. One further drawback--the high temperature could conceivably alter the structure of the tracer material.

(3) Rotating filter sample collector

A rocket-launched device studied by Sandia Corporation, this sample collector couples the rotorchute technique of impaction collection with diffusive filtration collection. The rotor blades are at zero lift and are solid-propellant driven at 10 rps, thus admitting flow to the filter. Use of the filter increases collection efficiency in the smaller particle sizes (bordering on molecular) while reduction in rotational velocity reduces impaction collection efficiency. However, this effect might be countered by filter design.

Conclusions. Fully developed and calibrated, the device could prove useful to the program. Again, the fact that it samples a column of air rather than a point will reduce the reliability of reduced data.

(4) A cascade filtration collector

This is a device studied at the U. S. Naval Radiological Defense Laboratory. By staging filters in decreasing pore size and drawing the sampled air through them, it is possible not only to collect particulate, but also to classify it according to size.

Conclusions. Such a device suffers the drawback of motor requirements; it will prove useful to this program when all such powered devices are useful.

d. A point source sampler for use to 150,000 feet

This device is designed to circumvent the major limitations of available samplers. Those limitations repeatedly emphasized in the literature surveyed were power required, volume sampled, and, in some instances, area sampled (column or trajectory sampling, rather than point sampling). The device will obtain for analysis both whole air samples and samples of air-entrained particulate. The principle of operation is inducement of air flow by creating a differential pressure. The principal components required are a supply of high pressure gas, a nonporous, light, flexible bag of essentially double-wall construction (the inner wall tacked to the outer at intervals), and a particulate filter mounted in the inlet throat to the bag. The double-wall bag (collapsed), high pressure gas supply, and filter are launched to the prescribed altitude for sampling by balloon or rocket. (If balloon launched, payload is such as to limit point source sampling to approximately 150,000 feet. If rocket launched, sampling over the entire altitude range of interest to this program is possible, but in a column, rather than at a point.) At altitude, the inlet to the bag and filter is opened; high pressure gas is admitted to the interspace between bag walls. The high pressure gas forces the bag to attempt to assume the inflated shape of a balloon. A partial vacuum is created in the internal volume of the bag; air is induced to flow into this volume by atmospheric pressure. Entrained particulate is removed at the inlet filter. As the differential pressure (internal to external) is reduced by in-flowing air, more high pressure gas is admitted to the inner space between walls; further expanding the bag and inducing additional influx of filtered air. The process continues until the bag is fully expanded. At this point, the filter is sealed to prevent contamination of the sample. If the intent has been to obtain both particulate and whole air samples, then the filter and bag are recovered; if not, the filter is separated from the bag, and it alone is recovered.

To obtain a 1000-SCF sample at 150,000 feet, a bag 113 feet in diameter must be inflated. An internal wall pressure of about 0.1 psi would be sufficient, and about 1 pound of helium would be required. The helium container would weigh about 43 pounds; the filter, another 10 pounds. A double-wall polyethylene bag would weigh 210 pounds. This

total payload of approximately 264 pounds can be carried to 150,000 feet by a 12-million ft³ balloon of 0.6-mil wall thickness, weighing approximately 900 pounds. Since balloons of 13.4 million ft³ have been flown, the technique appears feasible.

e. Conclusions on particle samplers

On the basis of the literature surveyed, the following sampling techniques are recommended for this program:

- (1) Below 65,000 feet--direct flow filter and impaction collectors mounted in aircraft of the B57-D and U-2 types.
- (2) 65,000 to 100,000 feet--balloon-borne, direct air flow samplers such as the submicron aerosol collector (SMAC) or the bag whole-air sampler used by the AEC.
- (3) 100,000 to 150,000 feet*--balloon-borne, crystalline zeolite absorbent collector in special cases where tracer concentration is high; the focusing array impactor (rotorchute), when developed. This suggestion would be negated if a breakthrough on sampled volume for direct flow filter or impaction collectors should precede the use of these units.
- (4) 150,000 to 200,000 feet*--the cryogenic air sampling rocket, in special cases where tracer concentrations are commensurate with its air volume sampling rate. This is yet to be defined. The focusing array impactor (rotorchute), when it is developed.

5. Sampling Vehicles

Vehicles used will include aircraft to 65,000 feet, balloons to 150,000 feet, and rockets at higher altitudes. Some overlap of aircraft and balloon altitude and of balloon and rocket altitude coverage will occur.

a. Aircraft

The advantages of using aircraft are obvious. The sample is collected precisely where desired; the sampling is conducted for that length of time required to sample an adequate volume; the sampler is always recovered; the sample is preserved neatly for analysis. Aircraft would be used in preference to other vehicles if higher altitude operation were feasible.

* It is recommended that the double-wall, bag whole-air sampler of Topic d be further investigated. If practical, it would be the recommended system for use from 100,000 to 150,000 feet.

b. Rockets

Rockets would be used with the cryogenic sampler and the focusing array impactor. Each sampler is designed around a specific rocket: the former, around the Aerobee 100, and the latter, presumably around the Archer, a high performance vehicle which has replaced the Arcon as a production item at Atlantic Research Corporation. Since the sampler is recovered, the approximate cost per launch will be 5 to 8000 dollars.

Rockets would also be the probable choice for injection of test materials. Some found applicable are:

<u>Rocket</u>	<u>Payload to 200,000 Feet (lb)</u>
Arcas	15
Arcon	40
Archer	~ 100
Aerobee 100	~ 150

c. Balloons

Although balloon launches are at least as costly a means of sampling as rockets, they are preferred in their altitude range of applicability because they offer point-source sampling coupled with a greater likelihood of sampling an adequate volume. Their altitude capability for sampling purposes should shortly be increased to 160,000 feet. Raven Industries, Inc., has supplied technical and cost data for this report; it is presented in Appendix D. Performance capabilities and hardware characteristics as provided by Raven are presented in Fig. 11. Figure 12 is a typical test flight performance record.

Balloons may also be useful for test sample injection, particularly if the particulate is difficult to disperse. This situation can exist if the particulate has a strong tendency to agglomerate or remain in a batch if released in that manner by a rocket. A balloon release would permit injection rate control, which could prove advantageous.

Fig. 11. Load Versus Altitude for Raven Extreme Altitude Balloons
(Courtesy RAVEN Industries, Incorporated)

Fig. 12. Typical Test Flight Performance Record, Raven Industries Balloon
(Courtesy RAVEN Industries, Incorporated)

V. REFERENCES

1. Program Plan--Radiological Safety Studies for Space Power, Contract AT(30-3)-217, Task 7, Martin Marietta Corporation, MND-P-2469, January 9, 1961.
2. Statement of Work--Upper Atmosphere Re-entry Studies, Contract AT(30-3)-217, Subtask 7.3, Martin Marietta Corporation, MN-SW-1007, May 18, 1961.
3. "Final Safety Analysis of the Transit Generators," Martin Marietta Corporation, MND-P-2479-I, May 1961.
4. Öpik, E. J., "Physics of Meteor Flight in the Atmosphere," Inter-science Tracts on Physics and Astronomy, No. 6, 1958.
5. Schaaf, S. A., and Talbot, L., "Handbook of Supersonic Aerodynamics--Mechanics of Rarefied Gases," NAVORD 1488, Vol. 5, February 1959.
6. Whipple, F. L., "Meteors and the Earth's Upper Atmosphere," Review of Modern Physics, Vol. 15, No. 4, October 1943.
7. Detra, R. W., Kemp, W. H., and Riddell, F. R., "Addendum to Heat Transfer to Satellite Vehicles Re-entering the Atmosphere," Jet Propulsion, Vol. 27, No. 12, December 1957.
8. Probst, R. J., "Shock Wave and Flow Field Development in Hypersonic Re-entry," ARS Journal, 1961.
9. Willis, D. R., "Methods of Analysis of Nearly Free Molecular Flow for a Satellite or Other Space Vehicle," The General Electric Company (MSVD), August 1960.
10. Hammerling, P., and Kivel, B., "Heat Transfer to a Sphere at the Transition from Free Flow," Physics of Fluids, Vol. 1, No. 4, June 1958.
11. Reeves, B. L., and Van Camp, W. M., "Aerodynamic Heating of Simple Geometries in the Slip and Intermediate Flow Regimes," JAS, December 1959.
12. Stewart, K., "The Condensation of a Vapor to an Assembly of Droplets or Particles," Transactions of the Faraday Society, Vol. 52, 1956.

27. Dobson, G. M. B., Proceedings of the Royal Society (London), Vol. 236A, 1956 (as referred to in Refs. 29 and 38).
28. Machta, L., and List, R. J., "Meteorological Interpretation at Strontium-90 Fallout," USAEC, HASL-42, 1958.
29. Machta, L., "Discussion of Meteorological Factors and Fallout Distribution," USAEC, HASL-42, 1958.
30. Stebbins, A. K., III, "Second Special Report on the High Altitude Sampling Program," DASA 539B, August 1961.
31. Martell, E. A., and Drevinsky, P. J., "Atmospheric Transport of Radioactivity," Science, Vol. 132, November 25, 1960.
32. Machta, L., Letter, Science, Vol. 133, March 31, 1961.
33. Raskin, B. L., "Foamed Plastic Particles--Versatile New Material," SPE Journal, Vol. 17, No. 3, March 1961.
34. Cooper, F., "Accuracy of Measuring Ballistic Missile Trajectories," ARS Journal, Vol. 31, No. 4, April 1961.
35. Hougen and Watson, "Chemical Process Principles," John Wiley and Sons, 1953.
36. Richter, A., "Fallout Predictor," AD 208066, Quarterly Report No. 1, Ford Instrument Company, September 1958.
37. "The C. A. R. D. E. Hypersonic Range ONE Facility and Associated Instrumentation," TM AB-49, November 1959.
38. Mannring, E. R., "Optical Tracers for Atmosphere Soundings," Aerospace Engineering, Vol. 20, No. 9, Part I, September 1961.
39. "Morrison and Radioactivation," ASTM-261, 62nd Annual Meeting Papers, June 1959.
40. Smith, R. A., and Menker, H. E., "Neutron Activation Analysis Calculation," TLW 1051, Tracerlab, Inc., December 1958.
41. Private Communication, Union Carbide Chemicals Company, October 23, 1961.
42. Private Communication, Acheson Colloids Company, October 12, 1961.

13. "World Wide Effects of Atomic Weapons," Project Sunshine, Rand Corporation, AECU-3488, August 1953.
14. Friedlander, S. K., "Theoretical Considerations for the Particle Size Spectrum of the Stratospheric Aerosol," Private Communication, October 1961.
15. Chandrasekhar, S., "Stochastic Problems in Physics and Astronomy," Review of Modern Physics, Vol. 15, 1943 (as listed in Ref. 14), SCR-420.
16. Junge, C. E., Chagnon, C. W., and Manson, J. E., "Stratospheric Aerosols," Journal of Meteorology, Vol. 18, February 1961.
17. Davis, C. N., "Definitive Equations for Fluid Resistance of Spheres," Proceedings of Physics Society, Vol. 57, 1945.
18. "Meteorology and Atomic Energy," prepared by the U. S. Weather Bureau for the USAEC, AECU-3066, July 1955.
19. Taylor, G. I., "Diffusion by Continuous Movements," Proceedings of London Mathematical Society, Vol. 20, 1920 (as given in Refs. 18 and 23).
20. Richardson, L. F., "Atmospheric Diffusion on a Distance-Neighbor Graph," Proceedings of the Royal Society (London), Vol. 107A, 1926 (as given in Refs. 18 and 23).
21. Kolmogoroff, A. N., C. R. Academy of Science, USSR, Vol. 30, 1941 (as used in Ref. 23).
22. Obukhov, A. M., Bulletin of the Academy of Science, USSR, Series on Geography and Geophysics, Nos. 4 and 5, 1941 (as used in Ref. 23).
23. Taylor, G. I., "The Present Position in the Theory of Turbulent Diffusion," Advances in Geophysics, International Symposium on Atmospheric Diffusion and Air Pollution, Academic Press, 1959.
24. Lettau, H., "Diffusion in the Upper Atmosphere," Compendium of Meteorology, American Meteorological Society, 1951 (as given in Refs. 18 and 47).
25. Stebbings, A. K., III, "Special Report on High Altitude Sampling Program," DASA 5 31-B, June 1960.
26. Brewer, A. W., Quarterly Journal of Royal Meteorological Society (London), Vol. 75, 1949 (as referred to in Refs. 29 and 38).

43. Private Communication, Kahlenberg Laboratories, October 21, 1961.
44. Whipple, F. L., "The Theory of Micrometeorites," Part I, Proceedings of the National Academy of Sciences of the U. S. A., Vol. 36, No. 12, December 1950.
45. Martell, E. A., and Drevinsky, P. J., "Fallout from Nuclear Weapons Tests, Hearings Before the Special Subcommittee on Radiation of the Joint Committee on Atomic Energy," Science, Vol. 133, Vol. I, 86th Congress, March 31, 1961.
46. Martell, E. A., "Evidence for High Stratospheric Holdup of Nuclear Bomb Debris," Proceedings at the Upper Atmosphere Sampling Symposium, Part I, April 1961.
47. Sutton, O. G., "Micrometeorology," McGraw-Hill Book Company, Inc., 1953.
48. Mannring, E., and Knafllich, H., "Some Measurements of the Diffusion Coefficients in the Upper Atmosphere," TN-61-3-N, Geophysics Corporation of America, March 1961.
49. Mannring, E., Bedinger, J., Knafllich, H., and Lynch, R., "Upper Atmospheric Wind Profiles Determined from Three Rocket Experiments," TN-61-1-N, Geophysics Corporation of America, February 1961.
50. Machta, L., "Transport in the Stratosphere and Through the Ionosphere," Advances in Geophysics, International Symposium on Atmospheric Diffusion and Air Pollution, Academic Press, 1959.

APPENDIX A

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Del Electronics Corporation Mount Vernon, New York	This contribution is included intact in Appendix C.
Raven Industries, Incorporated Sioux Falls, South Dakota	This contribution is included intact in Appendix D.
Atlantic Research Corporation Shirley Highway and Edsall Road Alexandria, Virginia	Private communication--sounding rockets and the focusing array impactor collector.
Union Carbide Chemicals Corporation Saw Mill River Road Tarrytown, New York	Private communication--test materials
Kahlenberg Laboratories P. O. Box 1660 Sarasota, Florida	Private communication--test materials
Acheson Colloids Company 4614 Washington Avenue Port Huron, Michigan	Private communication--test materials
Geophysics Corporation of America Bedford, Massachusetts	Private communication--vapor trail observations and upper altitude winds

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Dr. S. K. Friedlander	Faculty, Chemistry Department, The Johns Hopkins University
Miss Betty Lou Raskin	Research Associate, The Johns Hopkins Radiation Laboratories
Col. I. R. Russell, USAF	Nuclear Safety Directorate, Air Force Special Weapons Center

APPENDIX B
TECHNICAL CONTRIBUTION OF TRACERLAB, INC.

Proposal P-14061
to
THE MARTIN COMPANY
for

ATMOSPHERIC RE-ENTRY
BURNUP AND DISPERSION
STUDIES - FOR
SNAP DEVICES

submitted by Tracerlab, Inc.
2030 Wright Avenue
Richmond 3, California

October 20, 1961

TRACING AIR PARTICULATES

I. General Considerations

The nuclear fallout analysis programs have produced considerable data on residence time and distribution patterns of particulate materials in the atmosphere. The sampling techniques that have been developed are of general utility in similar problems, although the radioactive nature of the particulate fallout makes it much simpler to analyze the samples than is the case for non-radioactive materials.

The uncontrolled nature of the injection of particulate matter into the atmosphere by nuclear detonations makes it difficult to predict accurately the effect of physical and atmospheric parameters on the rate of deposition of particulates. The fallout studies have not shed any light on the particulate nature of the collected samples, since the point of interest in this program has been restricted to the radioisotopic content of the total sample. Several intensive programs have been conducted, however, on the particulate nuclear debris collected at various altitudes and times which shed some light on the size distribution of these particulates. Since these particulates were radioactive, it was possible to develop techniques for isolating these uniquely derived particles from the total sample and perform the appropriate measurements. Tracerlab was a leading organization in the development of these isolation techniques and has performed extensive measurements on air particulates.

II. Selection of Tracer Materials

A. Requirements of Tracer Materials

The requirements of a suitable tracer material must be considered before selecting the actual materials to be used and the quantity necessary for release into the atmosphere to attain a specified accuracy in subsequent analyses. Several requirements are obvious, others depend upon the analytical sensitivity, sampling procedures and similar experimental limitations.

The criteria for selection of the materials for use as a particulate meteorological tracer are reviewed in the succeeding paragraphs. In every case the conditions of atmospheric injection, i.e., 100,000 to 200,000 feet and the size range of 0.01 to 10 μ have been considered with regard to the other parameters. Consideration has also been given to the fact that these particles might be used to predict the fate of particles resulting from the burnup of a SNAP reactor or isotope device.

1. Density

Since the size range is fixed, the density will determine each particle's rate of fall. It would be preferable, although not necessary, that the density chosen be close to that of the materials used in odd and even SNAP devices. These materials would simulate the re-entry vehicle, vehicle and radiator, the reflector, the shield, reactor vessel and the fuel elements for reactor-type SNAP systems, and the re-entry vehicle, shield, isotopes and miscellaneous containment equipment for isotopic SNAP systems.

In the case of the SNAP reactor the principal hazard will be from the meltdown of the fuel elements. A good assumption is that the particles will consist primarily of fused oxides of uranium, cladding, support, and moderating material. As a result, the density of the final particle in g/cc, assuming no fissures or blowholes, should be between 7.31 that of U_3O_8 , 4.6, that of ZrO_2 and 3.025, that of BeO_2 .

For the case of the isotopic SNAP, the simplest assumption is to assume the density of the oxidized or most stable form of fuel alone. These are tabulated below.

<u>Nuclide</u>	<u>Fuel Form</u>	<u>Density (g/cc)</u>
Po ²¹⁰	Po	9.3
Cm ²⁴²	Cm ₂ O ₃	11.75
Pu ²³⁸	PuC	12.5
Ce ¹⁴⁴	CeO ₂	6.4
Pm ¹⁴⁷	Pm ₂ O ₃	6.6
Cs ¹³⁷	CsCl	3.9
Sr ⁹⁰	SrTiO ₃	4.8

The other components such as the reflector shield, etc., which will form oxides, fall in this density range.

As a result, a particle simulant for both the reactor and the isotopic SNAP, should have a density between 4 and 10 g/cc. Because of the variety of materials of construction, it is recommended that only a few densities be chosen.

The rate of fall of a 10μ 2 g/cc particle from 40 Km has been calculated to be 2.5 cm/sec and for a similar 0.1μ particle 3.5×10^{-4} cm/sec. Thus it would take 364 years for 0.1μ particle to fall 40 Km. The velocity of fall is proportional to the density so it is further recommended that the particle density should be closer to 10 than to 4 g/cc for purposes of accelerating the experiment.

If only a single density is chosen, it is worth reviewing if information would be lost. In air, if one assumes a fused spherical particle, two parameters determine the rate of fall or horizontal movement, the density and particle size. The particle size is predetermined by Martin Company so that the only variable is density. Stokes' and Cunningham's sedimentation equations for small particles in general, are applicable within the range of densities considered. If it is assumed that the information obtained from a single density can be scaled up or down by existing empirical equations, then a single density experiment will suffice.

Since the rate of fall and horizontal movement is dependent on the point angle of injection and initial component of velocity imparted in any direction in addition to the acceleration of gravity, it is expected that knowledge of the initial velocity vector will be necessary to predict and calculate the fate of such an injection.

2. Chemical Uniqueness

The particle simulant should be easily detectable from other atmospheric debris, otherwise, a serious blank problem will exist. This debris is variable according to altitude and latitude. The sources of particles in atmospheric debris at the present time are:

- a. Natural terrestrial debris
- b. Natural extra-terrestrial debris
- c. Nuclear test debris
- d. Ablation and burnup re-entry products from space vehicles
- e. Fuel residues from rockets
- f. Special high altitude experiments

At the present time, the 100 to 200,000-foot altitude is not too dirty. Some nuclear debris, particularly below the limit of optical microscopy, is present, the larger particles having descended to lower

altitudes. The quantity and composition of d, e and f is subject to speculation. However, this material is now on the increase.

At the lower altitudes, particularly in the troposphere, dust from natural debris becomes abundant, although volcanic dust in the size range 0.01 to 0.1 μ has been observed up to 132,000 feet. Therefore, the critical material, as well as the most abundant, is natural dust at lower altitudes. It will be necessary to be able to differentiate the particle simulant from this dust.

Particles of continental origin consisting of small fragments of mineral rocks, carbonates, and quartz are most common. Second are oxides which incorporate Fe, Al, Ti, Mn, Cu, Mg, K and Na. Particles originating from sea spray are salt crystal particles composed of NaCl.

Maritime air masses, especially near sea level, contain large NaCl crystal particles 1.6 to 16 μ in diameter with some NaNO_3 . As a result, it is important to choose a particle which may be differentiated from atmospheric dust and sea spray, particularly for collections at lower altitudes.

3. Stability and Integrity of Particles

Some information on the fallout and dispersion of particles from the 100- to 200,000-foot altitude, based on transport of nuclear debris from atomic bomb tests, is available in the literature. Fallout from the stratosphere is now considered to have a half life of 5 to 10 years. As a result, any simulant in which it is expected to do long-term tracing should be stable to the elements. The other limitation in time occurs when the eventual dispersion of the original injection has either mostly fallen out or become so dilute that it cannot be sampled. In either case an element or compound, insoluble in atmospheric moisture and unaffected by atmospheric gases, prolonged exposure to light, ultraviolet and similar radiation, would be the most suitable.

An additional consideration in the choice of particulates is stability in formation, storage, and injection. These factors were considered in the choice of particle material.

4. Economics

The particle materials chosen have been considered from three standpoints--the cost of the initial material, the cost of commutating and sizing the particles, and the economics of subsequent analysis and handling.

The weight of particles will be most important from the standpoint of initial cost and payload of injecting device. A sample calculation for gold (an excellent candidate, and among the most expensive materials) has been made. The following assumptions have been made on the worst case, balloon collection in the terminal part of the experiment.

- a. Injection point 200,000 feet
- b. Volume of atmosphere beneath this altitude = 3.14×10^{25} cc
- c. Balloon Direct Flow Air Sampler at 65,000 feet will collect 1.41×10^9 cc
- d. One hundred one micron particles per sample
- e. Fraction of atmosphere dispersed by end of experiment ~5%.
- f. Typical particle is 1μ .

$$\text{Total mass} = \frac{(3.14 \times 10^{25})(100)(0.05)(1.01 \times 10^{-11})}{1.41 \times 10^9} = 1.11 \times 10^6 \text{ g}$$

This is approximately a ton. This amount could easily be cut by a factor of 10^2 to 10^4 by adjusting the altitude of injection, size range of particles or sampling device. For example, in the latter parts of the experiment, aircraft collection could be used which could easily sample a hundred times the air volume of balloon sample.

5. Practical Sampling Procedures

The tracers chosen must be amenable to collection by practical sampling procedures. In general, two sampling procedures have been used extensively, impaction and filtration. The impaction technique can also be used to fractionate the particulates in 3 or more stages by using appropriately designed jets and impactor plates. Considerable design data is available for fabricating appropriate devices. The chief drawback of the impactor devices is the relatively limited volume of air that can be sampled in comparison with filtration techniques. The air filtration technique collects all particulates above a

specified size and has a high collection efficiency. Some degree of particle size fractionation can be obtained by using filters with selected cutoff sizes but the fractionation obtained by this method is not as sharp as that obtainable by jet impactors.

Atmospheric precipitation can also be used in many cases as a collection device. This technique is crude in comparison with the other techniques but it can furnish a useful estimate of total fallout. Gummed papers have also been used in the nuclear debris fallout program to estimate normal atmospheric settling processes. Here again, the data obtained is rather crude but useful in overall deposition studies.

A potential collection technique that has not been used extensively is electrostatic precipitation. This technique is most effective for fine particulates, where jet impactors and filtration are least efficient. By proper design of the electrostatic precipitator, a reasonable degree of particulate fractionation might be obtained.

6. Sample Identification and Separation

Once the particles are collected on a filter or an impactor, it will be necessary to separate the particles from the other debris and the filter media in order to identify, enumerate and measure the particles. Thus the final handling procedures should determine the initial choice of particulate material.

These particles should be easily separable from the filter media and other debris. If low temperature ashing techniques are employed to remove the filter media, the particles should be stable under ashing temperatures. If chemical dissolution techniques are employed to dissolve the filter media and debris, then the particles should be inert to the reagents chosen. If neutron activation techniques are employed as a means of detecting particles, the material chosen should be unique enough to be differentiated from other debris and filter residues.

7. Analytical Limitations

The analytical limits of sensitivity of detection and reliable measurement constitute one of the major limitations on the tagged materials to be selected for this program. Obviously, radioisotopically labeled compounds would be the ideal choice, since the detectability and accuracy of measurement would be the optimum available of all presently known techniques. The nuclear debris fallout program has proven this to be an undisputable fact. For instance, 1 micromicrocurie of Sr-90 is equivalent to 5×10^{-15} g of mass. A more favorable isotope with a shorter half life, such as Pm-147, has a sensitivity 5 times that of Sr-90. It is difficult to approach this sensitivity by other analytical techniques.

Two approaches to the analytical problem can be taken. First, the individual particles can be isolated and the mass estimated by physical measurements, such as diameter of the particles and density. This is practical if all the particles sought can be isolated from the mixed bag of materials captured by the sampling device, a rather difficult and tedious operation at best but in some cases, a distinct possibility. The second approach would be to fractionate the particulates and conduct an analysis on each fraction for the total tracer content, using the most sensitive techniques available. Third, the entire particulate collection can be subjected to analysis to determine the tracer element concentration and the collection treated as an integrated process with no resolution of the particle sizes involved.

The most sensitive analytical techniques applicable to this study appear to be neutron activation analysis and isotopic dilution mass spectrometry. Both techniques merit consideration although neutron activation is inherently more sensitive in many cases and also reduces the blank problem to manageable proportions; this is one of the most difficult problems in trace analysis. The sensitivity of various elements to neutron activation analysis varies considerably, so that each element must be considered specifically. Isotopic dilution mass spectrometry has a sensitivity of approximately 10^{-9} g, although certain easily ionized elements, such as the alkali metals, have higher sensitivities. Mass spectrometry is a precise technique that has much to recommend it. The sensitivity can probably be increased considerably, although a separate program would be required to accomplish this. The blank problem, however, is present in mass spectrometry and this constitutes a considerable drawback in the use of this technique for ultratrace analysis. In general, neutron activation techniques appear to be the most useful for this program, and this technique will be used as the basis of material selection.

8. Toxicological or Biological Hazard

Although it is not expected that the particulates will be concentrated enough when reaching the ground to be a toxicological or biological hazard, this point should be considered in choosing the simulant.

9. Shape

If it is assumed that the vaporized materials from the SNAP systems will coalesce into spheres; it is recommended that the simulant particles be spherical in shape. Experiments to determine the effect of shapes, other than spherical, can be performed on the ground and an appropriate correction can be made.

III. Material Selection

In general, metals and metallic oxides, carbonates, fluorides and similar stable inorganic compounds are recommended. Since the metals and oxides are all generally available at the lowest cost, this narrows the form of the tracer element to be selected. The cost of potential materials must also be considered since fairly large quantities of tracer material will be required. Listed below are 20 possible tracers. The choice of tracer is based on choice of experiment design and analytical methods.

<u>Tracer Element</u>	<u>Chemical Form</u>	<u>Analytical Sensitivity*</u> <u>μg of Element</u>
Manganese	MnO ₂	5.6 x 10 ⁻⁹
Cobalt	CoO	5.3 x 10 ⁻⁷
Cadmium	CdO	6.9 x 10 ⁻⁷
Antimony	Sb ₂ O ₃	5.2 x 10 ⁻⁸
Lanthanum	La ₂ O ₃	3.3 x 10 ⁻⁸
Praseodymium	Pr ₂ O ₃	3.3 x 10 ⁻⁸
Samarium	Sm ₂ O ₃	7.1 x 10 ⁻⁹
Hafnium	HfO ₂	2.5 x 10 ⁻⁷
Tantalum	Ta ₂ O ₃	1.8 x 10 ⁻⁸
Tungsten	WO ₃	2.8 x 10 ⁻⁸
Thorium	ThO ₂	1 x 10 ⁻⁷
Indium	In ₂ O ₃	1.5 x 10 ⁻⁷
Gadolinium	Gd ₂ O ₃	2.5 x 10 ⁻⁷
Mercury	HgS	3.3 x 10 ⁻⁷
Gold	Au ₂ S ₃ , Au	9.7 x 10 ⁻⁹
Palladium	Pd	5.1 x 10 ⁻⁸
Europium	Eu ₂ O ₃	4.1 x 10 ⁻¹⁰

<u>Tracer Element</u>	<u>Chemical Form</u>	<u>Analytical Sensitivity*</u> <u>μg of Element</u>
Holmium	Ho_2O_3	5.4×10^{-9}
Gallium	Ga_2O_3	5.1×10^{-8}
Uranium	UO_2	9.4×10^{-9}

*Analytical sensitivity based on following conditions.

- (a) Thermal neutron flux of 4.3×10^{14} n/cm²/sec.
- (b) Count rate of isolated nuclide is 10 cpm at optimum counting time using optimum counting equipment and measured efficiencies.
- (c) Irradiation to saturation or 30 days, whichever comes first.
- (d) Quantitative recovery in chemical separations.

IV. Analytical Techniques

A. Isolation of Particulates

The first stage in the analysis is isolation of the particulates from the filter medium. In the case of simple filters, the particles can be isolated by ashing the filter at low temperature with activated oxygen; this eliminates the considerable blank problem which might arise from the use of chemical reagents. Tracerlab has used this procedure routinely in reducing filters containing particulate matter and it is far superior to wet chemical and combustion ashing procedures. The residue of the ashing can then be transferred to a quartz or polyethylene tube for sealing and neutron irradiation.

B. Analytical Procedures

Neutron activation analysis is an extremely sensitive procedure in the hands of experienced radiochemists. Irradiation time will be selected to obtain maximum efficiency for each element or group of elements. Short half life product radionuclides formed on neutron irradiation can be separated readily from longer half life radionuclides to limit possible contamination and increase the efficiency of recovery. In general, two irradiations per sample collection would be required; a short time irradiation of 1 to 3 days for elements producing short half life radionuclides and a 2 to 4 week irradiation for elements producing

long half life radionuclides. Standards will be run for each irradiation to relate the activity to total quantity of the element sought. Appropriate chemical separation schemes will be devised as soon as material selection has been decided and laboratory runs made on synthetic mixtures to verify the separation schemes and calculated sensitivities.

Irradiation of the particles could also be used to locate the particles before or after separation by radioautography. The radioautograph could also be used to size invisible particles. This is accomplished by visual observation of spots on the X-ray film. The activated particles may then be identified and confirmed as the simulant by its radiation characteristics such as beta or gamma energy and decay rate.

Calculation with gold indicates that a 5-day irradiation at 4×10^{14} n/cm²/sec will allow the detection of 10 cpm in 1.2×10^{-14} g. Under these conditions a minimum particle size of 0.11μ may be detected by counting techniques. Since radioautography is more sensitive than counting, individual irradiated gold particles, an order of magnitude smaller, may be detected.

Other promising analytical techniques can be suggested, such as mass spectrometry, microprobe analysis on isolated particles and density separation, but all of these procedures would require some study to determine their feasibility for this specific application. The microprobe analyzer, in particular, might be applied effectively by concentrating the collection or solution in a 10- to 20-micron depression in a beryllium plate and analyzing the solid deposit by its X-ray fluorescence spectrum. The high resolution of X-ray spectroscopy makes this procedure attractive. Microprobe analysis is not sensitive from the point of view of concentration but can analyze very small volumes of material. If the concentration of tracer element is 0.1% or higher in the mixture, then this technique becomes feasible. The question here is how much normal particulate matter is picked up during atmospheric sampling in relation to the tracer elements. Salt particles have been collected up into the stratosphere so that one can expect a contribution to the total particulate collection from this source. The possibility of using the microprobe technique is an interesting one for this application. It would simplify the analysis considerably and make available a larger number of potential tracer elements since the nuclear properties would not be a consideration.

C. Sample Collection

If separation of particle size is required in this program, the most effective procedure would be by jet impaction. This type of device has been used extensively to size particulates during collection, and practical designs for applications similar to the present study are available. The practical limitations of particle size that can be collected

by jet impaction is approximately 0.1μ , depending upon the properties of the particulates and available jet velocities. The lowest collection efficiencies are obtained with the smallest particulates, but this can be overcome to a large extent by using an efficient filter at the exit end of the impactor. Fractionation of the particulates is accomplished by using varying jets in cascade to vary the velocity of the fluid stream and to collect a range of particle sizes on each impactor plate. The impactor plates are coated with a soft or sticky material to retain the impacted particles so that they are not blown off the collection surface by the air stream.

If we assume a limiting jet velocity of sonic speeds, the smallest diameter collectable will be approximately 0.1 micron. At lower jet velocities, the maximum particle diameter collectable will increase to approximately 0.5 micron at 150 ft/sec. In all cases, jet impaction will produce a distribution of particle sizes on each impactor plate so that no sharp fractionation is practical with this device unless the individual particles are widely separated in size and fragmentation of the particles does not occur on impaction. The particle size distribution on an impaction plate in a properly designed system will cover a 7:1 ratio, so that sizing the particles in steps of 10:1 will produce a reasonably sharp distribution.

Filtration collection is much simpler but size fractionation is not practical with this technique. If the particulates are sized for each individual tracer element, then particle size fractionation per se would not be necessary, since each particle would be uniquely defined by its chemical composition. The disadvantage of this approach is that only 4 combinations of 3 particle sizes are available from 12 materials. Actually, the table of materials above lists 20 possible tracer elements, so that 6 combinations may be available, if the background is clean for 18 of these elements.

D. Optimum Number of Simultaneous Samples

A minimum of 2 simultaneous samples should be taken at a specific site to permit neutron activation at 2 irradiation times. A third sample for retention in the event the results must be rechecked would be desirable although not strictly necessary.

E. Filter Media Blank

The background associated with filter media should be determined for the filter itself prior to adding the particle simulant.

Tracerlab has had considerable experience in the determination of trace materials in filters. A list of projects in trace analysis is attached.

V. Comments on Program Design *

There are several assumptions implicit in the proposed program that are open to question, in view of the rather meager data available on re-entry burnup. These assumptions are that a large mass re-entering the atmosphere at burnup velocities will produce particles of material essentially within the particle size range of 0.01 to 10 microns specified and that these particles will retain the discrete sizes determined at the time of burnup. Ablative burnup may produce larger particles since the mechanism is quite different than simple thermal vaporization. Particles larger than 10 microns may easily be produced by ablative action, although their fate after separation may be presumed to include an unknown degree of thermal vaporization since the ablated particles will still retain the velocity imparted by the re-entering mass.

The discreteness of very small particles in the atmosphere is another point to be considered. There is some evidence that particles much smaller than 1 micron tend to agglomerate on larger particles in the neighborhood. Some investigations at the Air Force Cambridge Research Center indicate a maximum in the upper atmosphere particle size distribution of approximately 1 micron and this is thought to be the minimum size of discrete stability. Probably, the nature of the material has much to do with this characteristic since we have often found particles smaller than 0.7 micron in nuclear debris collections obtained from the stratosphere which could be identified by their radioactivity. The fractionation pattern of the particulate collection by physical, i. e., impactor, devices or chemical methods would answer this particular question for the tracer materials selected, with the chemical methods furnishing the least ambiguous answer in this case. Microprobe analysis of individual particles would also furnish useful information on small particle stability.

The extrapolation of particle injection test results to burnup simulation is, as has been mentioned, open to question. A more closely simulated procedure would be to eject spheres of varying diameters or other geometric shapes with layered tracer materials at test altitudes and sample the particulate mass to determine how far in depth re-entry burnup will actually occur under selected re-entry conditions. For instance, if a multilayered sphere of several tracers were ejected under the proper conditions, the absence of one or more of the inner layer tracer elements would indicate that burnup was incomplete under these conditions. In addition, particle size fractionation would establish the expected size distribution of the particulates as a function of distance from the outer layers of the actual device. The sample ejection system for this procedure could be quite simple and the use of solid metals rather than powdered oxides would simulate actual re-entry conditions more realistically by taking into account the contribution of oxygen re-action heat to burnup.

* Martin Marietta Note--Tracerlab has misinterpreted the Martin objectives for atmospheric injections. See Chapter III.

ESTIMATED COST PER SAMPLE

Flown Filters

Based on considerable past experience in neutron activation analysis, we estimate the cost for separation and analysis would be \$50 per nuclide. In addition to these costs, a fixed charge for sample preparation, neutron irradiation and handling services would be approximately \$411 per loading of 16 samples for a 4-day irradiation and \$1,050 per loading of 12 samples for a 35-day irradiation. The irradiation charges may be reduced by contracting for a large number of samples but this will have to be determined when the magnitude of the requirement is known.

Laboratory setup time, and verification of the chemical separation schemes would represent an estimated non-recurring cost of approximately \$6,000.

The price schedule for the determination of individual particle diameter, color, activity and specific activity plus a graphic presentation of results as frequency distribution of size and specific activity is as follows. (Approximate prices to be used for budget purposes only.)

<u>Number of Particles</u>	<u>Price</u>
50 - 99	2.50 each
100 - 199	2.25 each
200 up	2.20 each

The price schedule for irradiation is as listed above. A sample will consist of either the separated particles from entire collection matrix or separated fraction.

It will be necessary to calibrate the particle size versus radioautograph spot size by material chosen. Charges for setup time and calibration would represent an estimated cost of approximately \$8,000.

Blank Filters

The cost of determining the background in filter media will depend on the filter media employed and the particle simulant selected.

The costs estimated for determining the blank are the same as that for flown filters.

PREVIOUS PROJECTS

Some of the highlights of Tracerlab's experience in the field applicable to the analytical techniques for the determination of minute impurities in various matrices are briefly summarized below.

1. Determination of Trace Impurities in Crystals and Semi-Conductors by Neutron Activation Analysis.

This is a current study contract sponsored by the Air Force Systems Command. Samples of highly purified materials are prepared for irradiation and then irradiated in a nuclear reactor. The individual trace impurities are then determined quantitatively by instrumental and radiochemical techniques.

2. Mass Spectrometric Analysis.

Tracerlab employs two surface ionization mass spectrometers for routine contract analysis. These instruments have a sensitivity for the least abundant isotope of uranium of 5×10^{-13} grams. Analyses have been performed for trace quantities of elements ranging from boron to plutonium. Highly purified materials have been scanned and semiquantitative determinations of trace contaminants made. Current routine work with these instruments includes analysis of zircalloy containing 0.01% boron, uranium isotopic analysis (10×10^{-9} g) and plutonium (at 10^{-10} g).

3. Fluorimetric Analysis for Beryllium and Uranium.

Tracerlab has offered, as a commercial and contract service for many years, trace analysis of Be and U in inorganic or biological matrices. A Jarrell-Ash and a U. V. Turner Fluorimeter are used in this work. Current analytical contracts include Pacific Gas and Electric Company, Sandia Corporation, and Air Force Special Weapons Command. A brochure describing some aspects of this analytical service is enclosed.

4. Reactor Effluent Analysis.

Under contract to the Air Force Special Weapons Command, methods were developed to isolate and analyze (from various collection matrices) particulates containing nuclear and ablated debris from the Tory reactor tests in Nevada. Techniques were developed to analyze short- and long-lived fission products and radioactive heavy elements. Separation techniques for the analysis of Be in 10^{-9} gram quantities were

developed. Two techniques were employed - fluorimetric analysis and electron microprobe analysis.

5. Nuclear Submarine Analysis.

Under contract to Westinghouse (BAPL), Tracerlab has performed startup and followup studies on a number of nuclear submarines. This work is performed at the construction site by a field team and laboratory. Cooling waters containing traces of fission and corrosion products (activated to produce measurable radioactive species) are rapidly determined in the field. Trace quantities of non-radioactive products such as iron, nickel, manganese, chromium, cobalt and zirconium were analyzed spectrophotometrically.

6. Radiochemical Techniques for Investigation of Purity of Food and Drug Packaging.

Two analytical techniques were employed on this project: radioactive tracers and neutron activation. As a result, a highly sensitive method of determining trace element contamination in food and drugs was developed.

7. Precision Radiochemical and Activation Analysis.

Under past and current contracts to the U. S. Air Force, Tracerlab has developed methods for precision analysis of 56 radionuclides and 120 radioisotopes. This involved separating individual isotopes from complex mixtures rapidly, with great purity and in high yield. Nuclear counting methods were devised to determine the true disintegration rates. Stable isotopes in 10^{-9} gram quantities or less have been determined by nuclear activation techniques. These analytical contracts have been renewed annually for the past 13 years, and the total value has been in excess of ten million dollars.

8. Electron Microprobe Analysis.

Tracerlab has, under contract with the U. S. Air Force, determined various elemental concentrations as low as 0.1% in individual particles having sizes on the order of two microns. This method is a non-destructive technique involving the impinging of high-energy electrons upon the target and subsequent ejection of orbital electrons from the K or L shell of the atoms. The energy and intensity of the characteristic x-radiation emitted by the elements in the target material is analyzed by an X-ray spectrometer.

9. Emission Spectroscopy.

Tracerlab has developed emission spectroscopic analytical techniques under contract to the U. S. Air Force Technical Applications Center and the Air Force Special Weapons Command. Examples of the limits of detection of four selected elements are:

Beryllium	$2 \times 10^{-12} \text{ g}$
Lithium	$1 \times 10^{-9} \text{ g}$
Aluminum	$5 \times 10^{-10} \text{ g}$
Iron	$5 \times 10^{-9} \text{ g}$

10. Trace Elements in Organic Moderated Reactor Coolant.

Tracerlab has performed neutron activation analysis for trace elements in virgin and recycled terphenyl and ashed residues for Atomics International. The following elements were determined: S, P, Cl, Mn, Na and Cu. The sensitivity for sulfur is 1 ppm and the others, 19 ppb. Similar studies have been performed on the U. S. Navy nuclear submarine program, wherein low-level impurities were quantitatively measured in reactor coolant water by use of the neutron activation technique.

Filter Media Testing

Tracerlab has designed, constructed and operated a large diameter, testing duct for use in determining flow and retention characteristics of various filter media. These ducts were designed for use with radioactive materials, and included electrostatic precipitators, pressure and flow measuring equipment, pumps, dust feeders and sampling ports.

Rainwater Sampling

Tracerlab has designed and built systems for removing radioactive particulates from rainwater including tanks, filter, circulating pumps and the system for adding coagulating material to tanks.

Continuous Monitors

Tracerlab has designed, constructed and tested a series of continuous monitors for radioactive particulates in hot gases incorporating wet cyclone collectors and providing instrumentation for measuring the amount of radioactivity associated with the particulate matter and the sampled gas. Auxiliary equipment including routing valves, diaphragm pumps, pressure regulators, solenoid valves, level controllers and pumps.

Mobile Monitoring Stations

Tracerlab has designed and manufactured a series of mobile radioactivity monitoring stations incorporating an electrical power generation system, high volume particulate air sampler, and background gamma and beta-gamma detector system.

Stack Gas Studies

Tracerlab has made complete theoretical studies of the gaseous and particulate activities from stack effluents for ORNL. This has included efficiency calculations, air flow and sampling design.

High Altitude Air Samplers

Tracerlab has designed air samplers to be incorporated in the wing tips of B-57A airplanes for the Los Alamos Scientific Laboratory. This has included wind tunnel testing and construction of the air sampler. Tracerlab has also designed and constructed air samplers for mounting on the T-33A aircraft and the F-84G aircraft.

APPENDIX C

TECHNICAL CONTRIBUTION OF DEL
ELECTRONICS CORPORATION

PROPOSAL FOR PARTICIPATION IN THE
MARTIN PROGRAM IN THE DETERMINATION
OF THE DISTRIBUTION OF PARTICLES ON
RE-ENTRY BURNUP OF SNAP DEVICES

Submitted by: DEL ELECTRONICS CORP.
MOUNT VERNON, N. Y.

To: THE MARTIN COMPANY

Dated: 20 OCTOBER 1961

COLLECTORS

1. With regard to specific Collectors for use at various altitudes, it is recommended that Filter Paper Samplers be used in the altitude range from 0 to 80,000 or 90,000 ft. In the range of altitude from approximately 80,000 to 140,000 ft., Electrostatic Precipitators are recommended. In the range of above 150,000 ft. to 200,000 ft., Impactors are recommended.

The filter paper type collectors have been used and are found to be feasible at altitudes up to 70,000 or 80,000 ft. Above these altitudes, the pressure drop across the filter makes operation of filter paper devices of questionable value. The electrostatic precipitation, which is undergoing tests at the present time, appears to have good collection characteristics and other features listed below, which make it desirable for use in the range of 80,000 to 140,000 ft. It is expected that future developments can extend the range of this device to higher altitudes. The exact altitude limit is questionable at the present time. At altitudes up to 200,000 ft. impaction by means of rotating devices have been proposed. Although to our knowledge none of these devices have yet been tested under actual flight conditions, they should theoretically give good collection.

2. Del Electronics Corp. has developed and is in the process of flight testing electrostatic type precipitators for use in the range of 90,000 to 140,000 ft. altitude. Five (5) of these units have already been built.
3. The electrostatic precipitator listed above was designed for collecting particulate matter in the range of .01 to .1 microns in diameter in the stratosphere at altitudes from 80,000 to 140,000 ft. The device has been tested in the laboratory with particle sizes having a mass median size of .02 to .16 microns at the rate of 100 cfm with efficiencies exceeding 95%. The aerosol used in these tests was a fluorescent dye generated in an aerosol generator of the *Whitby type. Units of this type have been flown at altitudes up to 110,000 feet by the General Mills Corporation together with their filter paper type sampler and apparently successful results have been achieved. Continued flight evaluation of these units is still under way.

*Described by Prof. K. T. Whitby, University of Minnesota

THEORY OF OPERATION

The Electrostatic Precipitator consists of a cylindrical tube in which a fine wire is axially suspended. A blower mounted at one end of the cylinder draws air through the cylinder. A potential is applied between the wire and the cylinder, creating corona at the wire. The ions so produced traverse the cylinder radially and in doing so charge the particulate matter which is airborne. These charged particles are then subjected to the usual electrostatic force due to the voltage gradients within the cylinder and are thus precipitated on a lining which is placed on the inside of the cylindrical tube. The lining consists of a 3 mil aluminum sheet which has been coated with a carbon deposit (aquadag). Valves are placed at each end of the cylinder to prevent foreign unwanted material from entering the precipitator except at altitude. Auxiliary equipment includes the high voltage power supply, provision for measuring the blower speed, corona current recorder, batteries, etc. At the present time, we are not supplying any telemetering equipment or other means of determination of altitude, temperature, etc. The unique features of this device are:

- (a) Extremely small pressure drop through the unit at reasonably high air velocities. This is a requirement in a collecting device which must operate at ambient pressures which may be as low as a fraction of a millimeter.
- (b) The ability of precipitating particles directly on the electron microscope grids.
- (c) The simplicity of removing the sample for counting and radiochemical analysis. This consists of the removal of the aquadag film, resulting in little solid material included with the sample. This avoids the treatment of large quantities of cellulose materials such as would be obtained in conventional filters.
- (d) An extremely low level of background contamination if this sampler is to be used to collect radioactive materials.
- (e) The unit is light in weight compared to other devices.
- (f) No special requirements except care in not contaminating the collection surface are required.

1. This unit has operationally been flown at altitudes of 116,000 ft. with a sample volume of air of 200 cfm ambient for approximately four (4) hours at an altitude in the range of 105,000 to 115,000 ft. The efficiency has been determined in the laboratory at flow rates of 100 cfm with particle sizes having median diameters of .02 to .16 and found to be in excess of 95%. On a theoretical basis, the efficiency of this type of unit will decrease as the size of particle increases beyond these sizes, although no actual efficiency test with the larger size particles has been conducted. The reason for this is that particles of larger sizes than those indicated are not expected to be resident at these altitudes for any great length of time. Insofar as we know there is no discrimination with regard to the physical or chemical characteristics of the particle (note, that at atmospheric pressures, there is discrimination due to dielectric constant; however, at the altitudes in question we rely upon a single charge precipitating the particle and consequently dielectric constant should not affect the efficiency).
2. It has been our experience that contamination of the collected sample has been negligible since entry and exit ports of the precipitator are closed, except during actual sampling operations. Present procedure involves the preparation of the liners within sections of the tube within the laboratory where contamination can be controlled. Upon recovery, sections of the tube containing the collecting lining are removed and sent to the laboratory for removal under laboratory conditions. This procedure avoids handling of the collecting surface by field personnel with the resulting elimination of chances of field contamination. The collectors are reusable (except for the liners) for many tests. The liners are inexpensive 3 mil aluminum sheets of standard variety, spray coated with an aquadag solution.
- 3a. Our estimates of cost on an electrostatic precipitator, consisting of the precipitator tube with blower, mounted in shock absorbing frame, a high voltage power supply designed for operation from 12 volts, recording equipment to measure the corona current, but excluding telemetering equipment and batteries for operation and no auxiliary balloon flying equipment for determination of altitude, ambient temperature, etc. are estimated as follows:

<u>Quantity</u>	<u>Unit Price</u>
10 units	\$3,000.00
100 units	~ 2,300.00
1000 units	~ 1,900.00

These costs are based on the present construction, and if 1000 units were to be procured, it is quite possible that economies could be made by re-design. Delivery of the presently designed unit is estimated to be as follows:

<u>Quantity</u>	<u>Delivery</u>
10 units	8 weeks A. R. O.
100 units	16 weeks A. R. O.
1000 units	~ 9 months A. R. O.

Note that the above deliveries are deliveries for the completion of order, and that deliveries on a scheduled basis can be made before the times specified.

- 3b. The cost estimate for performance testing of two (2) units is estimated at approximately \$20,000.00, including the cost of two (2) units and provided that equipment constructed under the Atomic Energy Commission contract can be utilized for these tests. The anticipated tests include efficiency tests at various particle sizes within the specified range of .01 to 10 microns at various altitudes from 80,000 to 140,000 ft.

MATERIALS

1. There are no restrictions placed on the selection of test materials for the electrostatic precipitator except carbon. The reason for this is that the material is collected on a surface of aquadag "colloidal carbon." If radioactive carbon, such as Carbon 14, is used in some elemental state, separation of this from the colloidal carbon will be impossible, resulting in considerable amounts of self-absorption, if a counting technique is used.
2. Although electrostatic precipitators can in theory be made to preferentially deposit discrete particle sizes, this is impractical in a unit which must sample large volumes of air. The reason for this is that particles enter over the entire diameter of the cylinder and as such differential deposition is masked by the poor resolution of such a geometry.
3. Our recommendation of test materials is as follows:
 - (1) Artificially produced isotopes of reasonable half life not normally found in bomb debris. The selection of the specific material is best left to others more familiar with the biological aspects involved in such use and in the possible confusion between the trace material and actual bomb debris.
 - (2) The use of fluorescent materials which are insoluble in water offers an excellent means, since these are generally harmless and

subject to analysis in minute quantities. Our present testing technique utilizes a fluorescent dye which is then analyzed on a mass basis by means of a photofluorimeter of unique design which we are capable of manufacturing.

SAMPLE ANALYSIS

This collector does not require any special techniques in analysis of the samples. Obviously the collection surface must be removed from the aluminum and concentrated. The procedure to be followed in analysis will of course depend entirely on the selection of the aerosol material.

ADDITIONAL COMMENTS

Del Electronics Corp. is presently starting the development of a simpler, more compact and lighter weight electrostatic precipitator which offers the promise of a very small collecting area, small input power requirements and possible operation at considerably higher altitudes. At the present time, details of this new development are not concrete; however, at such time as the proposed program is ready for action, this new precipitator may be ready for use.

APPENDIX D

TECHNICAL CONTRIBUTION OF RAVEN INDUSTRIES, INC.

October 18, 1961

The Martin Company
Baltimore 3, Maryland

Dear Sir:

You have invited us to discuss the potential of plastic balloons as platforms from which to sample the atmosphere at altitudes above 70,000 ft. We are very pleased that Raven Industries has been included in your invitation. Our company was founded with a specific interest in the design, manufacture and field use of plastic balloons. An experimental program, such as the Martin Company is considering, is therefore of extreme interest to Raven Industries.

As you know, the collection of particulate and gaseous samples from the atmosphere by balloon has been carried out in the past. The AEC, Air Force Cambridge Research Laboratories, U. S. Weather Bureau, university groups and others have conducted such programs.

A number of Raven Industries personnel have participated in these programs. This experience has included negotiation and establishment of operations on foreign bases, the design of special equipment for remote use, the management and operation of field test sites, and the organization of logistic support. Recently a program of collection and analysis was given consideration by a team including Raven Industries (balloons and flights), Acton Laboratories (samples), Dudley Observatory, Isotopes, Inc., and Fullam Laboratories (analysis). Some of these groups might also be able to contribute to the Martin program.

The natural buoyancy which is a basic characteristic of plastic balloons permits long periods of operation at a selected level. While a balloon cannot rise as high as rockets or rocket-powered aircraft, it can far surpass the altitude of conventional planes--and stay aloft for many hours. Operating limitations must be recognized; the balloon is not an all weather vehicle. In fact, even after 14 years of development, the balloon is not yet 100% reliable. A small number (less than 10%) of balloons flown on research programs fail for various reasons. When launching conditions can be selected, and balloons are not experimental in nature, the incidence of failure should be low (perhaps 5%), but it would be wrong to represent the balloon vehicle as absolutely failure-free.

A number of specific questions have been raised.

- 1.a. Materials: For flights of the character under consideration, the most suitable balloon material would be polyethylene, either 3/4 or 1/2 mil in thickness. Other films, notably Mylar, have been used and Mylar must be considered for long duration flights with little or no payload (superpressure balloons). Also, heavy load balloons, with limited altitudes, might require balloons made from fabric and plastic laminates, scrimms, etc.

Since 1957, following the successful initial use of 1/2 and 3/4 mil polyethylene, such material has been used regularly in balloons where high altitude is demanded. For such flights with gross loads of more than a few hundred pounds, the thin film is reinforced with load tapes. Heavier films, 1 to 3 mils thick, are still used for low level flights with medium and heavy payloads.

All balloon grade polyethylene comes from a single extruder, the Visking Company. Chemical compositions, strength, and physical dimensions are carefully controlled. For some thicknesses of film, MIL specs have been established but these are only partially complete for the thinner films now in use.

Inflation Gases: Most balloons flown for United States research programs use helium. Even overseas operations have been mounted with helium transported in high pressure cylinders. Two additional possibilities should be considered. If remote operations are planned, use of cryostatic (liquid) helium might be possible. (This is of doubtful economic value.) Likewise, hydrogen can be used in remote areas. Hundreds of thousands of small balloons carrying many tons of leaflets were flown from West Germany under the Free Europe Committee balloon program. Hydrogen gas, locally procured, was used throughout this program without serious accident in some five years of operations. This is a hazardous gas, but it is not impossible to work with. From the operational viewpoint, the additional 7% of lift (by volume) which hydrogen provides is not important, and it is opposed by the greater instability of this diatomic gas (helium is monatomic) which may result in an increased ballast requirement.

Manufacture: Since about 1951, nearly all plastic balloons are designed according to a computer formula developed by the Physics Department of the University of Minnesota. All balloon makers follow this design, and all makers use much

the same procedure in manufacture. Individual companies may vary somewhat in the procedures used for material inspection, quality control, in-process test and seam analysis. At Raven Industries, we have established what we believe is the most comprehensive quality control procedure in the industry.

Launching: Because large plastic balloons are liable to damage in winds of more than about 10 knots, special launching equipment and techniques have been developed. These vary slightly from user to user, but generally include a device to hold down most of the balloon while the upper portion or "bubble" is being inflated. Since less than 5% of the full volume is inflated on the ground, the remainder of the balloon skin would act like a huge sail if it were not held under restraint. Once inflation is complete, the payload must be brought beneath the vertically extended balloon and released. When the inflation is accomplished in a sheltered area, the balloon might be slowly erected. With the large balloons required to reach 100,000 ft or more, this "vertical inflation" would demand an open pit mine or the equivalent. This still is not too satisfactory as the escape from such a pit may be hazardous. Launchings from ships steaming downwind are nearly ideal, provided only that ships with reasonable speed and a clean top side area are available. Small aircraft carriers are technically ideal, but even smaller craft with helicopter decks would be very useful.

By far the most flights are made from hard surfaced land areas. Here the "dynamic" launching technique is generally used. A truck carries the payload. The balloon "bubble" is constrained during inflation. When the inflation is completed, the bubble is released. The entire balloon rises, moving with the wind. The truck must be driven underneath the balloon, and when the full erection is completed, the payload is released. This sounds simple enough, but a small shift in the wind, or any of an almost unlimited number of "emergency" situations will try the skill of even a highly skilled and thoroughly experienced crew. Because winds are often lightest at dawn, launchings frequently occur at that hour. The amount of gas introduced into the balloon is carefully calculated from temperature, pressure and volume data. A 10% error in calculations will very likely be fatal. Most flight trains have little more than 10% free lift, and would not rise at all if the error were on the side of too little gas. Too much free lift will result in an increased rate of rise to ceiling altitude. Normally, a rate of 1000 feet per minute is considered

the safe limit for light loads. When the balloon system, including the balloon, weighs over several hundred pounds the safe rate-of-rise value is generally reduced and in some cases may be as little as 400 ft per minute.

In-flight Tracking: An escort aircraft normally tracks each balloon flight. Position is determined visually and with a radio compass homing on a balloon-borne transmitter. This aircraft also acts as an in-flight control center, receiving altitude and performance data by radio from the balloon. Normally, the flight pattern is predetermined and controlled by instruments carried in the flight train. In some cases, a command override of these scheduled functions is desired. This can be accomplished from the escort aircraft using radio commands sent to a balloon-borne command receiver. Such an override might be necessary to effect flight termination at an earlier time than that set into the flight timers. If the original schedule would result in losing the payload in the ocean, or parachute impact in an area of high aircraft traffic density, it would be the escort plane commander's position to use the radio command and terminate earlier.

Recovery: Payloads are recovered by being released from the balloon on a parachute. This may be effected by timer, pressure switch or radio command. The procedure is straightforward up to altitudes of about 120,000 feet. Above this altitude, there is some uncertainty as to the parachute deployment. By using guide surface parachutes the problem of fouled parachutes (occasionally experienced in earlier years) has been overcome, but still there may exist a long period of ineffective parachute action. This can result in high velocity openings after the system has fallen into regions of higher density. Under extreme circumstances, shock loadings of 40 times gravity have been observed; however, 3 to 4 G is normal. Proper design of equipment is essential to prevent damage under such conditions. After the payload parachutes to earth, it is spotted down by the escort aircraft, and recovered by a ground crew.

A new technique has been proposed and is now being tested at Raven Industries. This consists of aerial retrieval of the parachute and payload, using specially equipped aircraft. The current tests involve payloads of 150 pounds, but it is expected that similar techniques will prove usable on loads of 500 pounds or more.

- 1.b. Rate of Ascent: Controlled by free lift, this may vary from 400 to 1000 feet per minute. With gross loads of less than 800 lb, the 1000 fpm figure has proven satisfactory.

Altitude Maintenance: Altitude sensors commonly use bellows to measure balloon altitude. Above about 120,000 feet these bellows have a sensitivity of only several thousand feet. The control of balloon height which relies on a servo driven by such a bellows is thus limited to perhaps 2000 ft at 100,000 ft, 5000 ft at 130,000 ft, and 10,000 ft at 150,000 ft. A recently developed Raven Industries technique effects altitude maintenance without reliance on such sensors. These controls keep the balloon at an altitude so constant that the sensors do not show any variation. It might be expected that variations of one or two thousand feet do occur, nevertheless.

Diurnal Effects: Once a balloon system is at ceiling, it remains floating until the internal gas volume is reduced, as by cooling, or the gas purity is decreased. Normal diffusion and leakage will reduce the lift after about four hours, and the unbalanced condition will result in descent until action is taken to restore equilibrium. Although special, poorly understood radiation field conditions can stop descent, it is generally necessary to lighten the load by dropping disposable ballast (replenishment of lifting gas from liquid stores is of theoretical interest on long flights). By the release of ballast amounting to a few percent of the total system weight, gas losses can be accommodated unless a serious "leaker" is involved.

One special circumstance requires additional compensation if constant altitude is to be maintained. At sunset (beginning in midafternoon, but largely concentrated in a period of about 1/2 hour) the temperature of the balloon film and gas is reduced from its previous, slightly superheated, condition. As the gas shrinks, the balloon becomes unbalanced. A ballast drop of not more than 5% of the gross system weight will compensate for this "sunset effect." At sunrise, a balloon which is already floating will gain lift, but the expansion of the gas will cause overflow through the ports which must be provided to prevent internal pressure buildup. The system will be lighter by the amount of weight represented by this overflow gas. Accordingly, an almost negligible increase in altitude will result at sunrise.

- 1.c. Limiting Conditions: Among the problems of ballooning which should be identified are these:

Winds at Launching: A surface wind of 10 knots could probably be tolerated, but most experienced balloon crews will not attempt launchings with winds over 5 knots if a regular or routine program is involved. On special flights, coordinated with satellite firings, solar flares and other uncontrollable events, launchings have been carried out in winds of 20 knots. This is definitely not to be recommended. Assuming a long range schedule of flights is to be carried out, climatologists can help locate areas where good surface winds can be expected. Natural features, such as mountains, modify the ambient wind, and experienced meteorologists can make good use of such features to improve the probabilities. A ship-borne operation offers the best means of operating with little concern for surface winds.

Winds Aloft: The balloon system will drift with the winds at floating level. An operational site must therefore be chosen with due concern for the prevailing winds and conditions which will affect recovery. In winter, winds aloft are generally westerly, blowing to the east with speeds which, on the average, increase with height and may exceed 100 or even 150 mph on rare occasions. For summer, the west winds only reach up to about 60,000 ft, in midlatitudes, and are overlain by easterlies. These easterlies also increase in speed with height, but do not exceed about 50 knots at 150,000 ft.

FAA Restrictions: For balloons with payloads of more than 12 pounds FAA flight restrictions apply. Launchings must be made from approved sites, under good weather conditions. A forecast of trajectory must be prepared, and regular in-flight position determinations and reports must be made. These regulations are designed to promote air safety and prevent the research balloon from becoming a hazard to air traffic. Current regulations are workable, and no special problems are expected to beset a valid scientific program.

- 1.d. What Distinguishes the Ford from the Cadillac in Balloons? Virtually all balloons built today are designed to satisfy a particular requirement. The distinguishing feature between the plush and the plain lies mainly in attention to small details. The Ford balloon may be characterized as an off-the-shelf design with a minimum of alteration or modification for the application prescribed. By contrast, the Cadillac will be designed from scratch with each facet of the construction being tailored to the application. The basic shape and dimensions may well be identical in both cases, but the film thickness, load tapes (if appropriate), appendix, end fittings and other accessories will be carefully matched to the purpose intended.

While most of our production consists of specifically designed balloons, we do produce from standard designs when a customer desires, as cost savings can be significant. In either case (as opposed to the simile) quality control and inspection are at the same high level of thoroughness. We believe that our quality control procedures, including inspection of all seals and all film through crossed polaroids, is unmatched in the industry.

As a practical matter, I suppose a balloon buyer might do well to select a manufacturer, and work on a cost-plus-a-fixed fee arrangement. This would ensure that rather more than the minimum possible time would be spent on manufacturing and inspection. When balloons are bought on the standard competitive bid basis, there is always a possibility that somebody in the industry might try to buy the job on a bid too low to provide for adequate time and end up producing a sub-Ford balloon. This is not a strong likelihood since I believe all balloon makers recognize that quality is all-important.

1.e. Standard Controls and Equipment:

Parachute.

FAA Safety Switch, usually combined with cutoff device.

Termination Timer.

Ballast control for constant-level flights of over 3 or 4 hours.

Altitude telemeter, usually combined with beacon for in-flight tracking.

Photobarograph (elective).

Radio command, usually for termination override (elective, except for Air Force).

Flashing light for night flights.

- 2.a. Site Selection and Features: The general area for an experiment will probably be defined by the subject. In the case of atmospheric sampling, a latitude effect may dictate sites at 0°, 30° and 60° N. For all-year operations at 30°, a central U. S. site might serve. Zero and 60° sites must be related to recovery. If aerial retrieval is available, there should be no problem in selecting bases from which recovery aircraft can operate. For payloads too large to be picked up in mid-air, a water recovery would permit operations from several sites. Land-to-land operations at the equator would probably demand a North African site, and 60° north operations might have to rely on ski planes in winter, float planes in summer, and for several weeks in fall and spring recoveries might be entirely precluded.

Once the general region has been selected, considerations such as air traffic will influence FAA approval. From the operations standpoint, a large hard surfaced ramp or parking area at a little used airport is to be desired. Weather, FAA and airport operations offices are also needed, although these might be at a reasonable distance from the actual launch site.

Inside working areas for storage and instrument preparation are required. Local accommodations for crew members and transportation must also be available. Most of these facilities can be provided on a military base, but unless a completely autonomous organization can be established, it is hazardous to be in any way dependent on military support. Even a military team devoted to the specific project suffers somewhat from rotation, training and crew motivation. From most geographic locations, including overseas sites, civilian support will be adequate and free from possibly obstructive military procedures.

In the event that a ship is made available for launching, its utility would more than offset these objections to military support.

Most of the ground launching facilities which are required at a balloon base are special. General purpose trucks and personnel transportation must be supplemented by helium tank cars, helium transport trailers, aircraft equipped with radio command transmitters, radio compass and other equipment to match the balloon instrumentation. Launching and inflation devices are designed to meet field use conditions.

- 3.a. Balloons Recommended for an Eight Hour Sampling Flight with a Payload of 240 Lb at Altitudes of 70,000 to 120,000 ft: Table 1 presents design data for a number of balloons which are suggested for this altitude range. All of the sizes given are "standard" and have been used many times. No special problems are to be expected in making similar specific designs for this program or in the use of such balloons.

It will be noted that the larger balloons are made from thinner material. The weight penalty would be extreme if 1.5 mil or heavier films were used. The thinner films will have a slightly greater diffusion rate, but this will not be significant in flights of less than 12 hours. Likewise, the increased toughness of the thicker films must theoretically make them less subject to damage during handling and launching. This advantage is so slight as to be offset by the larger sizes needed for signifi-

cantly heavier gross loads, brought about by the greater balloon weight. Since thin polyethylene has been in use, balloon volumes have increased from 3,000,000 cu ft in 1956 to 6,000,000 in 1958 and 9,000,000 in 1960. Such balloons have performed with an outstanding success ratio which belies the fragile nature of 1/2 and 3/4 mil films. With the possible exception of the 13,400,000 cu ft balloon mentioned elsewhere, no failures have occurred simply as a result of going to larger and larger sizes with thin films. There is no reason to believe that any limit has been approached.

In the cases where the balloon itself is small by comparison with the total load, as at 70,000 ft, heavier materials do not impose a penalty and have in fact been chosen.

None of the balloons identified in Table 1 have reinforcing load tape construction. They would have standard top-filling inflation tubes. Cylindrical end sections are commensurate with the total load to be carried, the "natural shape" design derived from University of Minnesota formula and short duct appendices for valving the free lift gas used to drive the balloon to the ceiling altitude.

The calculations assume that sunset will not be encountered and therefore no allowance for sunset ballast has been made.

Table 1 -- Balloon specifications for 240 lb scientific load

Altitude (ft)	Balloon Data				Inst and Ballast (lb)	Gross Load (lb)
	Diameter (ft)	Nom Vol (million cu ft)	Film Thickness (mil)	Weight (lb)		
70,000	69	0.135	1.5	110	100	450
85,000	97	0.36	1.5	210	110	560
100,000	128	0.8	1	227	110	577
110,000	157	1.5	1	340	120	700
120,000	187	2.4	3/4	350	125	715

- 3.b. Balloons Recommended for an Eight Hour Sampling Flight, with a Payload of 250 lb at Altitude of 120,000 to 160,000 ft: The current altitude record for a balloon of any kind is just over 150,000 ft. This was established in 1959 with a Raven balloon carrying a scientific payload of 100 lb (total payload, 137 lb). In order to meet the suggested requirements of extreme altitude, larger balloons must be built than have heretofore been essayed. Because of the weight of the balloons themselves, load-bearing tape reinforcements would be necessary in each case except the first. The design features would be similar to those of the balloons discussed in the preceding section.

Table 2 gives the specifications of balloons recommended for these altitudes. It should be noted that ballast requirements for an eight hour floating period are reduced as altitude increases. This is based on extrapolations of theoretical data with limited corroborative experience.

The only 13.4 million cu ft balloon built to date failed after launching in very unfavorable conditions. The two larger balloons have not been built to date. There is no reason to believe that such mammoth balloons could not be built and flown successfully, but it would be misleading to represent that any have been tried to date.

Table 2--Balloon specifications for 250 lb scientific load

Altitude (ft)	Balloon Data				Inst and Ballast (lb)	Gross Load (lb)
	Diameter (ft)	Nom Vol (million cu ft)	Film Thickness (mil)	Weight (lb)		
120,000	187	2.4	3/4	350	125	725
130,000	239	5.1	3/4	580	130	960
140,000	291	9.0	3/4	905	135	1280
150,000	332	13.4	1/2	940	100	1320
155,000	380	20.0	1/2	1250	100	1620
160,000	436	30.0	1/2	1660	60	1920

Because of the extreme dependence of balloon size on altitude, the significance of the payload is not as great as might be expected. Nevertheless, in Table 3, we show what would result if the scientific load were limited to 50 lb and the control instruments and ballast reduced to an absolute minimum.

Table 3--Balloon characteristics for 50 lb scientific load

Altitude (ft)	Balloon Data				Inst and Ballast (lb)	Gross Load (lb)
	Diameter (ft)	Nom Vol (million cu ft)	Film Thickness (mil)	Weight (lb)		
120,000	157	1.5	1	340	80	470
130,000	187	2.4	3/4	350	80	480
140,000	253	6.0	3/4	645	80	775
150,000	253	6.0	1/2	460	65	575
160,000	380	20.0	1/2	1250	40	1340

4. Payload Altitude Graphs
Please refer to Fig. 11, Chapter IV.

5. Cost and Delivery Data for Balloons
The prices of balloons, in various quantities, are given in Table 4. For any of these balloons, the initial delivery could be made in 45 days. Additional deliveries could be made at the rate of 10 per week for balloons up to 12 million cu ft in volume, and at the rate of one per week in sizes up to 13.4 million cu ft without special additional facilities. In the case of Raven Industries, and presumably in the case of other suppliers, 20 and 30 million cu ft balloons might require special fabrication facilities or extended delivery schedules--up to one balloon per 30 days.

All prices are based on design data described in earlier sections of this report. Prices are for estimating purposes only and should not be considered binding without confirmation.

Table 4--Balloon Costs

Balloon Sizes		Prices in lots of:		
Diameter (ft)	Volume (million cu ft)	10	100	1000
69	0.135	\$ 390	\$ 330	\$ 300
97	0.360	800	700	600
128	0.8	1,200	1,000	900
157	1.5	1,800	1,400	1,200
187	2.4	2,500	2,000	1,800
239	5.1	5,200	4,300	3,900
253	6.0	6,000	5,000	4,500
291	9.0	9,000	7,500	--
332	13.4	13,000	11,000	--
380	20.0	20,000	17,000	--
436	30.0	30,000	25,000	--

6. Cost Data for Balloon Flight Services

The major cost variable for balloon flights is the balloon itself. Other costs are related to flight duration, instrumentation, crew maintenance and frequency of flights. If we assume a program of continuous operations from a base in the United States, with aircraft tracking and ground recovery provided by a balloon contractor, the following costs might apply.

Assumptions: 5 man crew, 8 flights per month, 8 hours duration at altitude. Helium, containers and its transportation provided at no cost to the contract.

Instrumentation (assuming recovery, repair and reuse)	\$1,100 per flight
Tracking aircraft	400
Recovery expenses	200
Crew labor and overhead	850
Field expenses including subsistence for 2 crew members	150
Cost per flight	\$2,700 plus balloon, plus helium.

This could be reduced to around \$2,300 if instrumentation were built in quantities of 50 or more. From past experience it appears that a crew of five can completely handle eight flights per month, with a prospect of making seven completely successful tests including equipment retrieved.

If we move the operations to a remote site, assuming transport costs are not included in the expenses under the contract, the following extra expenses should be expected.

U. S. A. Cost	\$2,700
Additional crew expense	200
Crew replacement and rotation	150
Operating expenses	<u>550</u>
	\$3,400 per flight, plus balloons plus helium, plus transportation

This has presumed an operation from a land area, such as Australia. For operations with aerial recovery, using contractor aircraft, the estimated cost per flight might be about 50% higher, say \$5,000 per flight. Additional ramifications such as water recovery, GFE support, ski planes, etc., would lead to corresponding variation in expense. It is believed that we have satisfied the intent by detailing these specific examples. One possible expense item is that of lifting gas. If helium is not supplied GFE it must be purchased at \$1.00 per pound of lift, plus containers and transportation. The gross lift needed would be about 25% more than the gross load given in Tables 1, 2 and 3. This allows for free lift, loss in handling and leakage. Overseas procurement of hydrogen might be similarly expensive.

7. General Comments

1. Balloons are well suited for sampling vehicles, and have already demonstrated the reliability and economy by which a large research project of this sort is made feasible.
2. Present day balloon techniques begin to lose efficiency and rise in cost at altitude over 140,000 ft. The estimates of cost given in Table 4 show that a balloon to operate at 160,000 ft might be 75 times as expensive as one designed for flight at 70,000 ft.
3. Raven Industries personnel have successfully flown balloons from Alaska; Brazil; the Caribbean; Ft. Churchill, Canada; Greenland; Germany; Guam; Hawaii; Kwajalein; Panama

and the Arctic Ocean as well as many domestic sites. The establishment and operation of multiple sites can indeed be done, with or without the active support of U. S. military units. Generally, we believe civilian operations are more effective.

We will be happy to augment these data with answers to any specific questions which may still remain. Please feel free to contact J. A. Winker, Chief Engineer, P. E. Yost, Manager of Balloon Operations, or the undersigned for additional information. Thank you for inviting Raven Industries to submit this report.

Very truly yours,

RAVEN INDUSTRIES, INC.