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WORLDWIDE EFFECTS OF ATOMIC WEAPONS

PROJECT SUNSHINE

August 6, 1953

R-251-AEC
(AMENDED*)

*Amended to remove classified data; otherwise the report remains unchanged and represents the 1953 estimate of the fallout problem.

The RAND Corporation
1700 MAIN ST. • SANTA MONICA • CALIFORNIA

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U.S. ATOMIC ENERGY COMMISSION
Contract AT (11-1)-135

U.S. AIR FORCE PROJECT RAND
Contract AF 33(038)-6413

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PREFACE

At the conclusion of a conference on the long-term effects of atomic weapons, held at The RAND Corporation, Santa Monica, California, in 1953, a classified report, R-251-AEC, was prepared. Along the lines suggested by this conference a concerted effort, under the name of Project SUNSHINE, developed to evaluate the long-term effects. Since R-251-AEC has formed the basis of many subsequent studies, it was felt desirable to issue an unclassified version of this report. As presented here, the report has some deletions and changes, but differs little from the original 1953 version.

This report should be read in the context of the state of knowledge of the SUNSHINE *problem as it existed in* 1953. *It is presented as an historical document.*

The changes in some of the physical constants should be particularly noted. When the calculations in Chapter 5 and other sections of the report were made, it was believed that the half-life of strontium 90 (Sr^{90}) was 19.9 years. A more recent and better value seems to be 27.7 years.*

This value combined with a new value for the fission yield of Sr^{90} gives the basic result that approximately *two* megatons of fission will produce 1 millicurie (mc) of Sr^{90}/mi^2, if the fission products are uniformly distributed over the earth's surface.† This should be contrasted with the early (1953) correspondence of one megaton resulting in 1 mc of Sr^{90}/mi^2.

To preserve the perspective of this report, the original data stand uncorrected. It is a simple matter for the reader to apply the corrections regarding half-life and fission yield should he so desire. It should be noted that the effect of a greater half-life and a smaller fission yield will be to

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*D. M. Wiles and R. H. Tomlinson, "Half-Life of Strontium 90," *Can. J. Phys.*, Vol. 33, 1955, pp. 133–137.

†W. F. Libby, "Radioactive Strontium Fallout," *Proc. Nat. Acad. Sci.*, Vol. 42, June, 1956, pp. 365–390.

increase the megaton "limits" (i.e., decrease the apparent hazard) as calculated herein.

On the other hand, certain natural factors and concepts regarding the mechanism of the Sr^{90} path into the human body have also been revised. For example, the original assumption regarding the amount of available natural strontium in the soil is evidently too high; the effect of lowering this value is to decrease the megaton "limits." In fact, the SUNSHINE "limit," as recently estimated by W. F. Libby,* is lower than that given in this report. It is emphasized that absolute values, both biological and physical, given in this report must stand corrected in the light of investigations conducted since 1953.

Several of the speculative aspects of the report were recorded without the advantage of the data that have accumulated since the inception of the SUNSHINE experimental program. Some of the conclusions have withstood the test of time; others undoubtedly require modification on the basis of the SUNSHINE experience of the past 3 years.

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*Ibid.

ACKNOWLEDGMENTS

Project SUNSHINE was undertaken at the request of the U.S. Atomic Energy Commission; it was jointly sponsored by the AEC (Contract AT (11-1)-135) and USAF Project RAND (Contract AF 33(038)-6413).

The initial, intensive, summer effort was led by Willard F. Libby of the Institute for Nuclear Studies, University of Chicago.* A large part of this preliminary report was developed from Dr. Libby's suggestions. We gratefully acknowledge his assistance.

The summer study was climaxed by a RAND–SUNSHINE conference held July 21–23, 1953. At the conference, data and conclusions were discussed with a number of experts who represented a variety of professional interests; we have used their comments where applicable in this report. The conference attendees were:

Adams, C. E.	Naval Radiological Defense Laboratory
Bacher, R. F.	California Institute of Technology
Bell, C.	Harvard University
Bethe, H. A.	Cornell University
Bugher, J. C., M.D.	Atomic Energy Commission
Cadle, R. D.	Stanford Research Institute
Claus, W. D.	Atomic Energy Commission
Comar, C. L.	University of Tennessee
Coons, Maj. R. D.	Air Research and Development Command
Crowson, Col. D. L.	Air Research and Development Command
Edinger, J. G.	University of California
Eisenbud, M.	Atomic Energy Commission
English, S. G.	Atomic Energy Commission
Gilmore, F. R.	California Institute of Technology
Griggs, D. T.	University of California
Grossman, B.	Air Force Cambridge Research Center
Gwynn, Lt. Col. P. S.	Armed Forces Special Weapons Project

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*Now Commissioner, U.S. Atomic Energy Commission.

Healy, J. W.	Nucleonics Division, General Electric, Hanford Works
Holden, F.	Stanford Research Institute
Holzer, R. E.	University of California
Holzman, Col. B. G.	Kirtland Air Force Base
Hooks, Col. D. E.	Kirtland Air Force Base
Humphrey, P. A.	U.S. Weather Bureau
James, R. A.	University of California
Kulp, J. L.	Columbia University
Larson, K.	University of California
Lauritsen, C. C.	California Institute of Technology
Libby, W. F.	University of Chicago
Long, F. A.	Cornell University
Lulejian, Lt. Col. N. M.	Air Research and Development Command
Magee, J. L.	University of Notre Dame
Marshall, C.	Atomic Energy Commission
Morse, P. M.	Massachusetts Institute of Technology
Newgarden, Maj. G. J.	Air Force 4th Weather Squadron
Plank, H. F.	Los Alamos Scientific Laboratory
Plesset, M. S.	California Institute of Technology
Salisbury, M.	Atomic Energy Commission
Scoville, H.	Armed Forces Special Weapons Project
Smith, N. M.	Johns Hopkins University
Solomon, A. K.	Harvard Medical School
Spence, R. W.	Los Alamos Scientific Laboratory
Taylor, Col. G. J.	Air Force 4th Weather Squadron
Teller, E.	University of California
Theis, C. V.	U.S. Geological Survey
Western, F.	Atomic Energy Commission
Wexler, H.	U.S. Weather Bureau
Woodward, Col. L. L.	Kirtland Air Force Base
Wyatt, Lt. Col. W. H.	Air Force 4th Weather Squadron
Wyckoff, P.	Air Force Cambridge Research Center

RAND also used the services of the following consultants who were unable to attend the July conference:

Anderson, C. E.	Air Force Cambridge Research Center
Barad, M. L.	Air Force Cambridge Research Center
Beckerley, J. G.	Atomic Energy Commission
Dodson, R. W.	Atomic Energy Commission
Gibson, Maj. T. A.	Armed Forces Special Weapons Project
Goldberg, E. D.	Scripps Institution of Oceanography
LaRiviere, P. D.	Naval Radiological Defense Laboratory
Lieberman, J.	Atomic Energy Commission
Machta, L.	U.S. Weather Bureau
Maynard, Capt. R., USN	Armed Forces Special Weapons Project
Oppenheimer, J. R.	Institute for Advanced Study
Peacock, W. C.	Tracerlab, Inc.
Rabi, I. I.	Columbia University
Revelle, R. R. D.	Scripps Institution of Oceanography
Shearer, J. W.	Tracerlab, Inc.
Thomas, H. A.	Harvard University
Turkevich, J.	Princeton University
Von Neumann, J. W.	Institute for Advanced Study

To all of these consultants RAND expresses its appreciation for their interest and contributions.

The members of the RAND staff contributing to the conference were:

DeHaven, J. C.
Greenfield, S. M.
Hill, J. E.
Kellogg, W. W.
Kramish, A.
Krieger, F. J.
Marshall, A. W.
Mitchell, H. H., M.D.
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Marshall, A. W.
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Plesset, M. S.
Rapp, R. R.

RAND accepts the responsibility for the content and conclusions of this preliminary report.

A. Kramish
Editor, SUNSHINE Report

SUMMARY

This preliminary report discusses the various aspects of long-range contamination due to the detonation of large numbers of nuclear devices. An improved methodology for assessing the human hazard is developed, and an extensive experimental program is proposed.

CONTENTS

CHAPTER

APPENDIX

CHAPTER 1

INTRODUCTION AND SUMMARY OF CONCLUSIONS

CHRONICLE

George Bernard Shaw once opined "Much of your space and time is being wasted on the subject of atomic warfare. The disuse of poison gas in the 1939–45 war, because it was as dangerous to its users as to their targets, makes it very unlikely that atomic bombs will be used again. If they are, they will promptly make an end of all our discussions by making an end of ourselves. . . . Still, give me space for another cry in the wilderness, that my unquiet spirit, wandering among the ruins of empires, may have at least the mean and melancholy satisfaction of saying: 'I told you so.' "(1)*

Project SUNSHINE was born of kindred unquiet spirits, most of which, however, are not as grimly pessimistic as Mr. Shaw's. Its purpose is to inquire into the nature of the various large-scale disasters that conceivably might result from the detonation of large numbers of nuclear or thermonuclear weapons. By "large scale" we imply areas many magnitudes larger than the immediate destruction area, and thereby also connote an expanded temporal span.

The first reasonably comprehensive study of this problem, by Nicholas M. Smith, was submitted to the U.S. Atomic Energy Commission in the Spring of 1949 and was resubmitted in final form at the end of that year under the code name of Project GABRIEL.† A far less thorough but similar study was made available in unclassified form in a section of *The Effects of Atomic Weapons*.(2)

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*References appear at the end of each chapter.

†The original Project GABRIEL report, having been superseded by several others, including the present report, is no longer available.

Previous estimates of the number of bombs or of the total yield necessary to produce such large-scale and long-term effects have contained inherent uncertainties because of the physical and biological unknowns in the complicated history of radioactive fission products and their effects on man. These uncertainties did not seem to be of critical importance in the past because the minimum estimates of yield required comfortably exceeded the total yield of nuclear weapons detonated to date. Now, with the advent of high-yield bombs and because of the world's increasing stockpile, the margin over the early estimates is still large but is less clearly definable. It is most desirable to lessen these uncertainties by a concerted attack on the problem. Project SUNSHINE* represents an effort by RAND with the support of many of the leaders in the appropriate fields of science to formulate the problem in such a way that an explicit experimental program may be undertaken to provide the facts necessary for a more reliable estimate of this large-scale hazard.

THE LONG-RANGE RISKS

Man, in his development of more "efficient" means of waging war, has often increased the incidence of casualties to noncombatants. Now, in atomic warfare, there exist means for inflicting noncombative-type casualties long after the detonation of a number of weapons, i.e., through radiative damage.

The hazards existing near ground zero following the immediate effects of a nuclear detonation consist mainly in exposure to external radiation; these hazards are covered in detail in many weapon reports. The main concern of SUNSHINE is to examine the hazards of radioactive debris that, by one means or another, finds a way into a human being and thereby becomes a source of *internal radiation damage*; this latter hazard may be present in relatively local areas of the earth or may be spread more extensively by natural means or through the commerce of man.

The radioactive debris resulting from the burst of a nuclear device can be classified as follows:

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*Project SUNSHINE is to be distinguished from the separate "short-range" evaluations of close-in, short time-scale fallout effects.

(*a*) Products dependent on the composition and nature of the device itself.

(*b*) Induced activities dependent on burst environment.

Project GABRIEL concluded that the most dangerous radioactive product is strontium 90 (Sr^{90}), one of the most abundantly formed products of a fission reaction. Later studies by the Atomic Energy Commission's Division of Biology and Medicine corroborate the identification of Sr^{90} as the most important long-range contaminant. These conclusions were established mainly as a result of the following properties of Sr^{90}:

1. A nuclear detonation will produce 1 gram (gm) of Sr^{90} per kiloton (KT) of yield energy, or 20 gm per 20-KT "nominal" bomb. This is a considerable quantity.
2. Its physical half-life is long—20 years.
3. It possesses a long biological half-life because of its bone-seeking property.
4. Body ingestion is high.

Project SUNSHINE has further concluded that the *manner of production peculiar to this contaminant suggests that it will be readily available for incorporation into the biosphere.*

Consequently, at the present writing, we concur that Sr^{90} is the principal long-range, possibly worldwide, contaminant. Studies are being continued to determine whether or not any other products under both (*a*) and (*b*) above may present a biological hazard comparable to or larger than that of Sr^{90}. Possible substances may be certain isotopes that are formed in fairly large quantities in thermonuclear bursts.

For lack of data, several problems are not discussed in this preliminary report but are the subject of current study. They include the problem of what role the hydrosphere may have in the disposal of radioactive contaminants, the effects of the contaminant on life in the hydrosphere, and human dependence on its products. The proposed sampling program is intended, in part, to give some values necessary for the consideration of this aspect of the problem.

Another problem is ecological in nature. It asks the question, What is the effect on other forms of life as a result of the decrease in population of a given biological form through the action of Sr^{90}?

This particular report is concerned with the radioactive-biological hazard. Other possible long-range effects of nuclear bursts were briefly discussed at the SUNSHINE conference, including:

1. Loading of the atmosphere with particulate matter—thus causing a decrease in insolation—which may affect the weather of the earth. It was suggested that the efficiency of ejection of material by large-yield bombs be compared with natural eruptions such as Krakatoa and the injection of material such as zodiacal dust.
2. Large increases in upper atmosphere ionization by radioactive debris, which may affect communications systems.*

Additional long-range effects conceivably may exist; to date, however, those suggested above appear to be the most important. The popular "hazard"—the effect of nuclear detonation on the weather—has been discussed in detail elsewhere (see, for example, Ref. 3).

CONCLUSIONS

In assessing the hazard to a large population, it is necessary to ask who or what is at risk as well as what is the nature of the risk. The risk is simply this: The bone-retentive and radioactive properties of Sr^{90} endow it with a high carcinogenic capability; a given amount above threshold (which may be zero) fixed in the bone will cause a certain average percentage of the population to die of bone cancer comparable with that observed in victims of radium poisoning.

Young and growing tissue is most susceptible to radiation damage; bone formation in an individual is complete by the time he is 20 years of age, although mineral exchange occurs for the rest of his life. In our model, therefore, we have taken as the individual most at risk the one who accumulates Sr^{90} from the age of 0 to 20 years in a population having a severely contaminated environment.

The SUNSHINE model, while containing, at the present writing, some of the uncertainties of earlier models regarding fallout, availability in soil, etc., bypasses a number of intermediary biologically unknown factors by a simple assumption:

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*Suggestion by Edward Teller.

The bones of an individual who grows up in an environment of a maintained, given ratio of Sr^{90} *to natural strontium will contain* Sr^{90} *and natural strontium in the same ratio.*

This assumption, in effect, is simply the assumption used in all biological tracer experiments; i.e., the body cannot distinguish between the natural and the radioactive isotopes of an element.

We have not attempted in this preliminary report to define a "threshold"-damaging dosage, a "mean lethal" dosage, etc. The terms are misleading and the magnitude of the dosages is unknown. Instead, we have normalized our studies to the Maximum Permissible Concentration (MPC) set by the International Commission on Radiological Protection.(4) This is the amount that, it is believed, may be retained safely in the body without causing minimal damage.

The MPC for Sr^{90} is 1 microcurie (μc) (i.e., one two-hundred-millionths of a gram). *This is an industrial standard for small numbers of people.* It may be necessary to reduce the MPC values for large populations.

It is with some trepidation that we present in a preliminary report of this nature an estimate of the number of nuclear detonations that will contaminate the world. First, we fear that the concept of uniform worldwide contamination has little meaning and that the necessary assumptions for such a calculation are unrealistically simple. The contamination undoubtedly will occur unevenly—in "blobs" over large areas—mainly because of large differences in localized fallout concentrations. Nuclear detonations occurring on a worldwide scale and possibly with a long-term "atmospheric storage" may smooth out the distribution somewhat. Secondly, differences in eating habits, disease, and environmental natural strontium content of soil will render certain populations more vulnerable to the contaminant than others. We believe the strontium ratio model to be applicable to problems of localized fallout, ethnic, and environmental differences, but the number of parameters still unknown prohibit such a calculation at the present time. We are thus forced to submit, for the present, an idealized calculation on a worldwide scale.

Neglecting the question of biologically effective dosages, the parameters necessary for assessing the hazard on a worldwide scale are

1. The fraction of Sr^{90} available for distribution as a function of type of weapon, condition of burst, and meteorology.

We assume here high-altitude bursts, with the immediate area of ground zero receiving no more than its proportional share of the fallout debris. Exchange of debris between the earth's hemispheres is also neglected. The over-all assumption is *pessimistic.*

2. Atmospheric or other natural storage mechanisms that might allow appreciable Sr^{90} decay before it becomes available to humans.

Whether Sr^{90} is stored in the atmosphere or in the biosphere, this consideration is not likely to increase our estimate by more than a factor of two.

3. Availability of Sr^{90} in debris for transfer to the biosphere.

We believe that the bulk of the Sr^{90} is plated out on the surface of the debris particles and that it is also scavenged out in solution by rainfall. Thus it should be readily available for take-up by the biosphere. If our reasoning is incorrect and the Sr^{90} is contained inside insoluble particles, the calculation given below should then be regarded as highly pessimistic.

4. Availability of natural strontium in soils.

The parameter used here is 60 lb of agriculturally available strontium per acre. We feel that over a period of time such as we are considering, more fixed strontium in the soil will become available. The better value lies somewhere between one and twenty times this amount. Having used the lower limit, our estimate in this respect is also pessimistic.

5. Redistribution of Sr^{90} by plowing, fertilizer, etc.

Fallout debris deposited on untilled soil is not leached down very effectively by rainfall. In agricultural areas (the areas of interest), however, the soil is constantly well mixed to an effective depth by the efforts of man. We also assume wash-off as relatively low. These considerations, in themselves, make our calculation pessimistic.

6. Content of natural strontium in bone.

The average U.S. adult, normalized to the "Standard Man," contains 0.7 gm total strontium in his bones. This figure is probably rather accurate.

On the basis of the above assumptions and other physical parameters,

the preliminary SUNSHINE *estimate of the nuclear bomb yield required to bring the population of the world up to Maximum Permissible Concentration is*

***larger than* 2.5 × 10^4 *megatons* (MT).**

We have taken the liberty of normalizing the GABRIEL models to an MPC of 1 μc and have corrected the Sr^{90} yield factor. For comparison with the preliminary SUNSHINE estimate, the GABRIEL gauge lies between **20 MT** and **9 × 10^4 MT.**

An estimate developed on a calcium-strontium model has indicated an **800 MT** limit; the uncertainty of this limit is unknown, but it is probably pessimistic, since parameters similar to the strontium-ratio type of calculation are involved.

Thus, the SUNSHINE *methodology achieves minima that lie close to the maxima of all previous models.*

RECOMMENDATIONS

It is seen that a number of physical and biological parameters remain to be determined to greater exactitude; many are unknown to several magnitudes. Until comparatively recently it would have been extremely difficult, if not impossible, to obtain a measure of a number of the parameters. Today we are afforded the opportunity of doing a radioactive-tracer chemistry experiment on a worldwide scale.

The release in the world of several kilograms (kg) of Sr^{90} within less than a decade has probably disseminated enough of the contaminant to provide amounts that are probably now detectable in samples of inert and biological materials throughout the world. An analysis of these materials for Sr^{90} will provide us with much of the information that is now lacking.

For our model we will also require an analysis of the ordinary strontium content of soils, waters, and biological materials. Other requirements may appear as the program develops.

It is rather striking that the last comprehensive measurements of soils for natural strontium were made in 1914–1917.(5) The strontium cycle in the biosphere is almost completely unknown, and unknown meteorological factors abound. *We submit, therefore, that the adoption of a sampling* *program similar to that proposed in this report may add considerable knowledge to many scientific endeavors, exclusive of its contribution to the problem of* Project SUNSHINE.

In Chapter 5 we propose a pilot sampling program. At present a "pre-pilot" program is being conducted on a very small scale through the combined efforts of several individuals who have been contacted personally. The pre-pilot program is more for the purpose of getting a feel for the problem rather than an attempt to find good parameter values. *We consider it urgent that the program progress to an organized pilot stage as rapidly as possible.*

We have made a worldwide prognosis that seems comfortably large, remembering that it is normalized to a maximum, presumably not harmful, permissible dosage of 1 μc of Sr^{90} fixed in the bone. We re-emphasize, however, the possibility that there may be significant variations of MPC levels for different cultural populations; this factor, together with the large variations in local fallout, may well result in localized dangerous levels. *We consider it essential that a parallel biomedical effort on carcinogenic action of ingested and inhaled radioactive materials be closely coordinated with the sampling program.*

In conclusion we should like to offer the possibility of prophylactic control of Sr^{90} should the world or a local area ever reach what might at that time be considered a dangerous level. This would be through dietary control.

The megaton limit is directly proportional to the amount of available strontium in the soil. The addition of amounts of nonradioactive strontium to the soil or to materials ingested by the individual in a given area would, in effect, dilute the Sr^{90} he would be taking up in his diet. This action should reduce the bone retention of Sr^{90} proportionately.

The limit is also inversely proportionate to the normal strontium content of the bone. Individuals who tend to accumulate high amounts of normal strontium are more susceptible to the action of the contaminant. It is believed that such individuals accumulate strontium, at least partially, in lieu of the calcium which is deficient in their diet. The suggested prophylactic action is obvious—more calcium in the diet.

There undoubtedly will be found other means of protection. Control of contaminants, be they infective or radiological, does not appear to be beyond the capability of man.

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3. HOLZMAN, B., "The Effects of Atomic Bomb Explosions on Weather," *Weatherwise*, Vol. 4, No. 1, February, 1951.
4. National Bureau of Standards *Handbook* 47, June 29, 1951.
5. ROBINSON, W. O., L. A. STEINKOENIG, AND C. F. MILLER, "The Relation of Some of the Rarer Elements in Soils and Plants," *U.S. Department of Agriculture Bulletin* 600, December 10, 1917.

CHAPTER 2

ORIGIN AND NATURE OF RADIOACTIVE DEBRIS

The chemical processes that follow the explosion of a nuclear device could be of importance in determining the chemical and physical form of the radioactive cloud and hence of the particles that ultimately constitute the fallout. For this reason an attempt has been made to describe the explosion from a chemical point of view. There are obvious differences between high bursts and those in which the fireball touches the earth, so the high bursts are considered first and then an attempt is made to modify these conclusions to include various other conditions.

AIR BURST

In the early history of the explosion the temperatures are so high that chemistry is unimportant because all the materials exist as single ions and atoms. We will consider first the time scale for a 20-KT bomb. Up to about 0.5 millisecond (ms) the fireball may be considered to be an isothermal sphere, and the surface temperature corresponds to the temperature of the bomb materials. After 0.5 ms the surface is cooler than the interior until after "breakaway," when the surface temperature again increases to the temperature of the interior (see Fig. 1 on page 12).

The temperature at the second maximum is about 7000°K and this temperature is reached in approximately 0.3 sec. The fireball has in that time almost reached its maximum radius and has engulfed enough air so that oxygen may be considered to be present in large excess.

The various elements, as atoms, begin to combine with oxygen atoms to form gaseous diatomic molecules at temperatures of the order of 20,000°K for the most stable oxides and at correspondingly lower temperatures for less stable oxides. Many of these gaseous oxide molecules are sufficiently stable to form before any appreciable amount of O_2

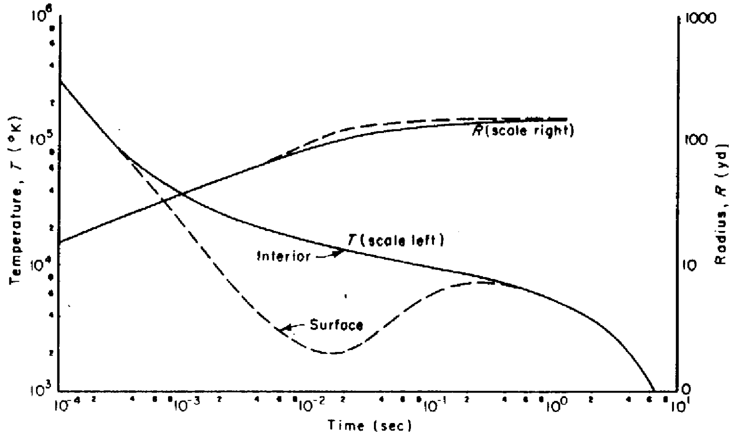

Fig. 1—Time-temperature relations

molecules is formed. (Oxygen gas at 1 atm pressure is 50 per cent dissociated at 3800°K.)

The relative stability of gaseous oxide molecules is given roughly by the D_0 value, which is the heat of dissociation of the molecule into the component atoms in the ground state at 0°K. Values of D_0 have been summarized by Brewer.(1) The following are some gaseous oxides in the order of decreasing stability: UO, ZrO_2, SiO, AlO, SrO, and FeO.

Although most of the fission products exist as oxides, some are present in other forms: iodine and bromine exist as halide ions, probably combined with very electropositive elements such as cesium and rubidium from the fireball and sodium from the dust in the air; silver, palladium, and rhodium are very likely to be in metallic form; whereas xenon and krypton remain in the atomic gaseous state.

As the materials cool, condensed phases will be formed from the gaseous oxide molecules. The order of condensation of the various oxides will depend, under idealized conditions of equilibrium (i.e., slow cooling), on the relative volatility of the oxides. The volatility of the oxides is not necessarily related directly to the stability of the gaseous molecules, but may best be related to the temperature at which the vapor pressure has a specified value.

From the data summarized by Brewer, we may arrange the oxides in the order of increasing volatility as follows:

Oxide	Vaporization Temperature (°K) (Pressure = 1 atm)
ThO_2	4670
ZrO_2	4570
(PuO)	(~4500)
(UO)	(~4500)
La_2O_3	4470
BeO	4300 (decomposes)
Al_2O_3	3800 (decomposes)
CaO	3800
SrO	3500
MgO	3350
FeO	3400 (decomposes)
BaO	3000
SiO_2	~2800 (decomposes)
Fe_3O_4	2060 (decomposes)

Condensed phases will form at some time when the temperature is between 5000°K and 2000°K, which on a time scale for a 20-KT bomb means between 1 and 5 sec after detonation. The actual condensation may occur in a very short time. At 3.5 sec the temperature will have reached 2500°K and the fireball will have attained its maximum radius and will contain approximately 2000 tons of air. If we assume that it contains 100 lb of unused plutonium or uranium,(2) a total of 1 kg of fission products, and 1 ton of bomb structure, the material in the fireball will then have the following composition:

Air	2×10^9 gm (99.9%)
Bomb structure	10^6 gm (0.1%)
Fission products	10^3 gm (0.00005%)
Pu or U	5×10^4 gm (0.0025%)

With this composition and the relative volatilities of the oxides in mind, let us now consider which oxides are likely to condense first and the probable form of the condensed particles. It should be noted that the condensable material is extremely dilute compared with that used in the laboratory conditions under which most condensation experiments are carried out.

If we consider ZrO_2, which has the highest vaporization temperature of any of the oxides of the fission products, then, assuming a maximum fission yield of 5 per cent and a total pressure of 1 atm, the partial pressure of $ZrO_2(g)$ would be 10^{-8} atm. We may estimate the vaporization temperature at 10^{-8} atm pressure to be 1300°K. In the case of FeO (assuming the whole bomb structure to be Fe), the partial pressure of $FeO(g)$ would be 10^{-3} atm. At this partial pressure a condensed phase of FeO would start to form at 2300°K. For aluminum at a partial pressure of 10^{-3} atm a condensed phase might be expected at 2700°K. Uranium oxide would have a partial pressure of 3×10^{-6} atm and would be expected to condense at 1600°K. From these considerations we see that, even assuming equilibrium (i.e., no supersaturation), the first condensed phase expected is Al_2O_3 in the case of an aluminum bomb structure and FeO for an iron structure.

Cooling through the temperature range where condensation takes place occurs at the rate of about 10^3 deg/sec for a 20-KT bomb, so that considerable supersaturation is possible, particularly in view of the very low partial pressures of most of the oxides. This supersaturation may tend to favor the initial condensation of those materials present in larger amounts, thus again indicating that the major components, such as aluminum or iron, probably condense first. Supersaturation may also lead to the simultaneous formation of a larger number of condensation nuclei, since fewer molecules are needed to form a nucleus, with the result that very small particles would be formed. Several species condense simultaneously, so that each small sample contains a mixture of the condensable materials present at the instant of condensation.

Some general statements may be made about the nucleation process:

1. The solid material present in the air engulfed by the fireball cannot be an important source of nuclei, since all of this material would be vaporized and mixed with the gaseous bomb materials.

2. The ions produced by the very intense radioactivity may serve to collect a few gaseous molecules and thus form condensation nuclei. It seems likely that this mechanism would be very important in the later stages of the condensation when the degree of supercooling is large, so that two or three molecules would be enough to form a stable condensation nucleus.

3. The material may be self-nucleating by statistical fluctuations in the number of many-body collisions.

From the experimental data that exist one can conclude that nucleation and growth must be proceeding simultaneously to give the observed range of particle sizes. If nucleation were slow and the subsequent growth process very fast, the particles would have an extremely narrow distribution in size about the mean. If nucleation were fast compared with growth, then essentially all the material would go into such nuclei and the particles would be very small and all very nearly the same size.

The experimental data on particle-size distribution have one thing in common: they all indicate in the region where the measurements are best that the number of particles of a given diameter decreases exponentially as the diameter increases. The experimental data also show a decrease in the number of particles at low diameters. The maximum number, however, occurs at a diameter that is very dependent on the method of sampling and sizing. For example, optical microscope results indicate a maximum in the number distribution at a particle diameter of 1 micron (μ) or greater, while electron microscope results indicate that this maximum occurs below 0.1 μ. It seems quite clear that this decrease in the number of particles having small diameters is not real, but is a result of the sample collection efficiency and of the resolution of the microscope. In the absence of better measurements the most reasonable assumption seems to be that the exponential increase actually continues down to very small particles of 0.01 μ or less in diameter.

With the assumption that the number distribution is given by $N_D = N_0 \exp(-D/b)$, the fraction of the particles of diameter less than a given diameter D' is given by $f_N = 1 - \exp(-D'/b)$. The fraction of the mass that resides in particles of diameter less than D' is given by

$$f_M = 1 - \exp(-D'/b)\left[\frac{1}{6}\left(\frac{D'}{b}\right)^3 + \frac{1}{2}\left(\frac{D'}{b}\right)^2 + \frac{D'}{b} + 1\right].$$

Figure 2 shows a plot of these two functions. It may be pointed out, for example, that 50 per cent of the particles has D'/b less than 0.693 and that this 50 per cent of the particles contains only 2 per cent of the mass and hence (assuming constant specific activity) only 2 per cent of the activity. On a mass basis, 50 per cent of the mass is contained in particles with $D'/b \geqq 4$ and these particles constitute only 2 per cent or 3 per cent of the total number of particles.

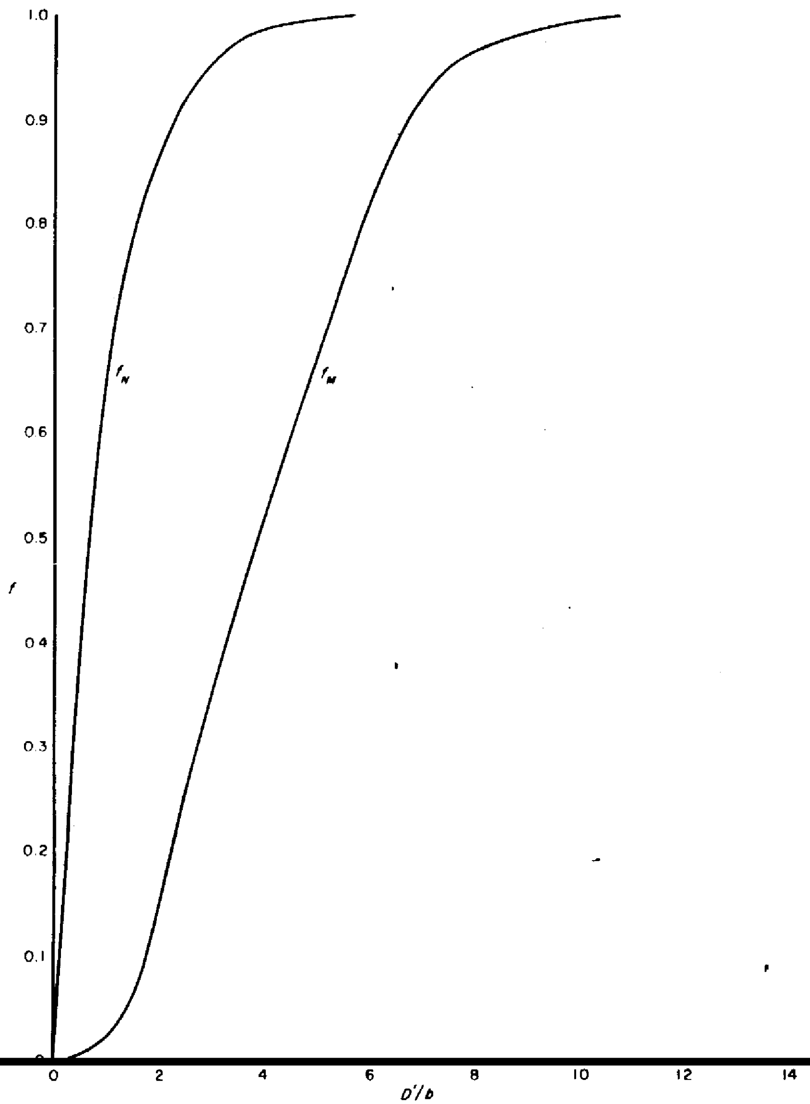

Fig. 2—Number fraction and mass fraction with diameters less than a given diameter D'

It must be emphasized, however, that these considerations assume specific activity independent of size. Since some of the activity comes from gaseous precursors and is presumably deposited on the surface of particles, the specific activity may vary with size. It is not practical to attempt an exact analysis of all the effects, but some general conclusions may be drawn.

1. The activity present in all particles having a diameter less than about one-fifth of the median diameter is entirely negligible compared with that in particles having diameters four times larger than the median diameter. In the RANGER and GREENHOUSE shots, from which the data seem to be fairly reliable, the median diameter in airborne material is 1.2 to 2.2 μ; thus the particle sizes of most interest from high air bursts (as far as over-all ground contamination from fallout or rainout is concerned) are those in the diameter range of 0.8 to 15 μ.

2. If the exponential number distribution of particle sizes is correct, the small fraction of the activity in extremely small particles may still be very important for some consideration. For example, inhalation of these small particles may be a serious hazard.

A consideration of the form of the fission products at the time of condensation is important, since this is the controlling factor in determining whether a given fission product will be distributed more or less uniformly in particles or will form a surface layer on existing particles at a later time. Even though some uncertainties exist in the half-lives of the short-lived members of the fission-product decay chains, the fraction of the total activity present in the various elements in the first 5 sec after detonation can be estimated. At 10 sec and longer, the calculation can be made with some certainty; such calculations have been presented by Hunter and Ballou,(3) the results of which are shown in Table 1 on page 18.

During the interval of 1 to 5 sec, it appears that the percentages given in Table 1 would not be vastly different from those at 10 sec. The striking fact about these considerations is that during the time when the temperature is right for condensation of solid materials, a very large fraction (approximately 80 per cent) of the radioactivity resides in the more volatile elements, such as noble gases, halogens, and alkali metals, which do not condense until low temperatures are reached or until decay to another less volatile element has occurred. An important example of this type is

Table 1

CONTRIBUTION OF ELEMENTS AS PERCENTAGE OF TOTAL ACTIVITY AT VARIOUS TIMES[a]

Element	10 sec	20 sec	1 min	1 hr	1 day	1 week
Kr	18	15	10	4.5	—	—
Xe	14	14	13	3.5	18	15
I	20	15	9	6	19	17
Rb	13	12	12	5	—	—
Br	8	8	6	1	—	—
Cs	7	12	17	9	—	—
Sb	5	6	7	—	—	—
Te	4.5	6	6	12	4	8
La	3.5	5	7	12	1.5	9
Sr	2.0	3.5	6	4.5	6	2
Mo	1	1.5	2.5	3.5	4	9
Nd	1	1	—	1	—	4
Y	—	1	2	13	19	2
Ba	—	—	—	10	1	—
Pr	—	—	—	6	3	8
Ce	—	—	—	5	6	8
Zr	—	—	—	—	9	3
Nb	—	—	—	—	9	—
Rh	—	—	—	—	—	3
Ru	—	—	—	—	—	2

[a] Those elements contributing less than 1 per cent of the total are indicated by dashes.

the mass-90 chain. Most of the material that eventually ends up as Sr^{90} is produced in the primary fission as Kr^{90} (lesser amounts of Br^{90} and Rb^{90} are formed and a very little Sr^{90}) and must go through the decay chain:

$$(Br^{90}) \xrightarrow{\text{short}} Kr^{90} \xrightarrow{\substack{33\\ \text{sec}}} Rb^{90} \xrightarrow{\substack{2.74\\ \text{min}}} Sr^{90} \xrightarrow{\substack{19.9\\ \text{yr}}} Y^{90} \xrightarrow{\substack{61\\ \text{hr}}} Zr^{90}.$$

Most of the material, therefore, spends the first few seconds after explosion as Kr^{90} and hence has no chance to enter into the growth of solid particles as Rb^{90} or Sr^{90} until after the solid particles have been formed and cooled.

These facts have many ramifications in the consideration of the effects of Sr^{90}. Particularly important are the following:

1. A large fraction of the mass-90 chain is condensed minutes after detonation, so that the solid particles formed from the fireball have become very dilute and are mixed with inactive solid particles of the air. Thus Sr^{90} may be fairly well separated from most of the other fission products and its detection by counting total activity may be more difficult.
2. The Sr^{90} produced in this manner is much more available in the biosphere, since it will be in very soluble form on the surface of the particles or actually dissolved in the water droplets of the cloud.

As indicated by the large percentage of the total fission products present as rare gases or other volatile elements at the time of initial condensation, the mass-90 chain is only one example of substances that will condense late in the history of an explosion. Other mass-number chains of possible importance are the 89, 91, 137, 139, and 140 chains.

The time scale and condensation times discussed above refer to 20-KT bombs. When larger or smaller bombs are involved, the time to reach a given temperature can be scaled approximately by the factor $(E/20)^{1/4}$ to obtain the time for a bomb of yield E.*

Thus if we take 2500°K as the condensation temperature, the time of condensation for a 1-MT bomb is about 25 sec. The rate of cooling is also much slower for a higher-yield bomb, so that condensation can occur under circumstances corresponding more nearly to equilibrium conditions tending to give larger particles. On the other hand, since the density of condensable material is inversely proportional to E, large values of E tend to increase the supersaturation and to counteract the effect of slower cooling on the particle size. The experimental data on particle size are not sufficiently reliable to determine how the particle size varies with yield. One

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*Various scaling laws have been suggested, but this law seems to correspond most nearly with the observed facts.

would expect that the degree of fractionation of the fission products would change markedly with yield, but one could not say that the over-all fractionation would be either increased or decreased. We must take individual chains and study the effects of longer time before condensation. In the case of Sr^{90} we would expect less fractionation with respect to materials such as Ce^{141} and Zr^{95} as the yield is increased, but perhaps more fractionation of Sr^{90} with respect to Sr^{89}.

SURFACE OR UNDERGROUND BURST

In the case of bursts where the fireball touches the earth, large quantities of the surface materials are swept into the resulting cloud. (This includes the tower in tower shots.) When the region is composed mostly of silicates, molten silica (SiO_2) will form, which is then swept around through the cloud in the typical toroidal type of motion. Molten silica is an extremely good solvent for the metallic oxides and hence these materials are dissolved and eventually solidified in the glass as the silica cools. These large particles appear to concentrate in the upper stem and the lower part of the mushroom.(4) A large percentage of the total activity is apparently captured by these silicates but falls out within a very short time. A small fraction remains as very small particles, perhaps even in molecular form for those substances that have gaseous precursors, and is airborne for long periods of time. This fine material represents a large fraction of the total fallout on the East Coast from bombs exploded in Nevada. Since silica glass remains soft down to 1500°K, it may collect small particles very efficiently for a long time after the explosion. This property, together with the tendency of the silica to react chemically with the metal oxides, explains the principal difference between the Nevada fallout and that from the Pacific Islands, where calcium carbonate predominates.

The carbonate materials are converted at relatively low temperatures to calcium oxide (CaO) and this may or may not melt or decompose, depending on the time at which it enters the fireball or resulting cloud. (Melting point, 2873°K; vaporization temperature, 3800°K; decomposition, 4000°K.) In any event, CaO is a much poorer solvent for metallic oxides than silica and hence would be less efficient in collecting small particles of condensed matter. In the larger particles formed by the tests in the Pacific, the activity

tends to concentrate on or near the surface of the particles as if the radioactive material had been picked up after a solid particle of CaO had been formed. Particles of this kind probably represent large CaO fragments that were not in the fireball at the time of the initial condensation but were swept through the cloud later in solid form. The CaO that was vaporized and condensed along with the fission fragments is probably much too finely divided to be sectioned and examined in the manner used in the study of the Pacific tests. This hypothesis could be tested by measuring the specific activity of the calcium in such particles and comparing it with the specific activity of calcium in fine particles or even in total samples taken from the cloud after a few hours. The particles in the fireball should have large amounts of induced calcium activity, whereas those swept into the cloud later should have much less.

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CHAPTER 3

DISTRIBUTION OF RADIOACTIVE DEBRIS

METEOROLOGY

A nuclear detonation creates a large hot bubble of air containing most of the radioactive debris. The buoyance of this bubble causes it to rise rapidly, and in the process of rising, it is cooled by adiabatic expansion and by mixing with large quantities of ambient air. The resultant rapid cooling of the rising mass brings it to equilibrium with the atmosphere in about 5 to 10 min, and it stops rising at this time. The particles in the bubble fall, under the influence of gravity; very large particles rapidly reach the ground; very small particles are so mixed by the atmospheric turbulence that they may be considered to act as a gas. The large mass of air containing the small particulate debris is subjected to the same atmospheric motions as any other large mass of air. The first step, therefore, in considering the spread of atomic debris is to inquire into the motions of the atmosphere.

The picture that one gets of the motion of the atmosphere is very much a function of the sensitivity of the measuring instruments and of the frequency and spacing of the measurements. The established networks indicate that in mid-latitudes there is a mean motion of the air from west to east and that superimposed on this motion there are cyclones and anticyclones ranging from 100 to 1000 km in diameter. Special studies on a very small scale give ample evidence that there are turbulent elements of the order of a few meters, but the evidence for motion in the range scale from 1 to 100 km is not adequate.

These different scales of motion all have their effect on the atomic cloud. The mean westerly winds and the large scale motions are, at least in the early history of the cloud, much larger than the cloud, so that the effect of these motions is to transport the cloud. In general, then, the centroid of the cloud will move toward the east and will be shifted to the north or south by the large-scale disturbances. The scale of motion of the order of magnitude of the cloud will tend to distort the cloud, stretching and twisting it into wisps and clumps. Scales of motion smaller than the cloud will stir it and expand it in an eddy diffusion process.

This is a very much simplified version of what happens, and it neglects the disparity between the horizontal and vertical motions. Horizontal motions in the atmosphere are several orders of magnitude greater than vertical motions, but the change in horizontal motion in the vertical, i.e., the vertical shear, is several orders of magnitude greater than the change of horizontal motion in the horizontal. Thus, the portions of the cloud at various heights will be moved with different winds. If the cloud extends from 30,000 to 40,000 ft, the top of the cloud will, on the average, be about 300 km away from the bottom of the cloud after 24 hr.

One additional type of atmospheric motion is convection. The most dramatic display of atmospheric convection is, of course, the thunderstorm, but many less spectacular forms of convection act to stir the entire troposphere rather thoroughly. If the cloud enters an area of moderate-to-violent convection, it can spread the cloud through the entire depth of the troposphere in a matter of a few hours.

The extreme variability and lack of adequate observations on some scales of motion of the atmosphere make it difficult to assign numbers to these atmospheric effects, but from the limited knowledge available an attempt will be made to give order-of-magnitude estimates. Consider the history of an atomic cloud in the middle latitudes in winter, with the cloud stabilizing between 30,000 and 40,000 ft and not encountering a convective area. The cloud will move eastward about 1000 mi in 24 hr and will be about 700 mi north or south of its original latitude. The top of the cloud will be about 300 mi away from the bottom. The cloud will have spread, at each altitude, to a radius of about 40 mi and may have moved vertically a few thousand feet. Thus at the end of 24 hr the cloud will be contained in a ribbon about 300 mi long, 4 mi wide, 15,000 ft thick, and about 1200 mi from its original position. The debris within this ribbon will not be distributed in any regular way, but will consist of clumps and wisps of high concentration with areas of low concentration in between them. If convection affects the cloud, it will be mixed throughout the troposphere and will be extended even more in the horizontal by the shears in the lower layers of the air. (A discussion of the settling of particles in a standard atmosphere, neglecting convection effects, is given in Appendix III.)

A cloud as large as the one described above can be acted on by the entire spectrum of atmospheric motions. In this event all the motions of the atmosphere, except the mean west-east flow, can be thought of as turbulent eddies. Defant(1) has computed an eddy diffusion coefficient for this motion, and by using his value of 10^{11} cm^2/sec, it was estimated that the spread of the cloud after 1 week would occupy an area of about 5000 mi in diameter.

The processes, other than fall, that can bring the debris to the ground are scavenging by rain and mass transport of air downward. Data gathered by the AEC New York Operations Office (NYOO) indicate that by far the most important of these is the rain scavenging. The data analyzed to date do not provide a means of measuring the scale of the mass transport. They do show, because small particles are brought down in dry air, that mass transport must, at times, be operative.

During the time that the cloud is diffusing, it is subjected to the scavenging action of rain and to the removal of debris to the ground by mass transport and fall. For detonations resulting in a large percentage of activity on large particles, the maximum deposition will result from the physical fall of the particles close to the point of detonation. For detonation resulting in a majority of the activity on small particles, the fall will be less important, and the dispersive processes of the atmosphere will spread the debris over large areas. If the debris is initially contained in the troposphere, in mid-latitudes, most of it will tend to remain in the troposphere and be carried primarily in the westerly circulation of mid-latitudes. There are, however, several ways for the material to be carried into the southern hemisphere in the troposphere: one well-organized transport in the Indian Monsoon and many small-scale motions can carry the debris south at almost any longitude.

Debris thrown into the stratosphere can easily be brought down either through breaks in the tropopause or through a process of eddy diffusion through the tropopause. In either event, however, such transport is expected to be slower than transport in the troposphere.

It cannot be emphasized too strongly that atmospheric motions are extremely variable and erratic processes. In any given situation it is possible to have a type of motion vastly different from the case given above. If it were desired to get representative statistical samples of the effect of the atmosphere on the bomb debris, it would be necessary to have many more tests than have been made and to have them in many points of the world. It would be surprising if the number of bombs exploded to date gave a statistically reliable distribution of the debris. It appears that the only way a useful purpose could be served by analyzing concentrations of debris from the existing tests is to relate them to atmospheric motions and then to extrapolate the results by the use of atmospheric data.

FALLOUT PATTERNS

While the quantity of materials produced in an atomic explosion can be calculated for any following time period, if the yield is known, the biological hazards to be expected will depend strongly on the distribution of the materials over the earth—as a result of fallout, scavenging by precipitation, etc.—and the subsequent chain of events by which such materials may get fixed in the human body. Also, reservoirs for the radioactive materials may exist in the oceans and in the upper atmosphere, which may reduce greatly the probability that such materials become incorporated in the biochemistry of living organisms, including man.

From the fallout measurements taken after TUMBLER/SNAPPER and IVY, it is possible to get some rough ideas concerning the distribution of fallout in the United States and worldwide from explosions in Nevada and at Eniwetok. These data are summarized in Table. 2. If these results are taken as representative of distributions from the two test sites, and if the TUMBLER/SNAPPER measurements are used as a calibration for the average fraction of the actual fallout retained by gummed papers,(2) a very crude estimate of the current United States and worldwide contamination due to Sr^{90} can be made.

Also, from the study of available data, important gaps in our knowledge are indicated that require additional study and experimentation for their clarification.

Table 2

SUMMARY OF FALLOUT DATA FROM TUMBLER/SNAPPER AND IVY TESTS

(All activity figures are extrapolated to January 1, 1953)

Test	Number of Stations	Maximum Fallout (d/min/ft²)	Minimum Fallout (d/min/ft²)	Average Fallout (d/min/ft²)	Average β curies/mi²	Area Sampled (mi²)	Measurement Period
TUMBLER/SNAPPER, United States	92	14,000	86	1,750	2.2×10^{-2}	3×10^6	April 1 to June 18, 1952
TUMBLER/SNAPPER, outside the United States	15	610	~8	212	0.27×10^{-2}	37.4×10^6	April 1 to June 18, 1952
TOTAL	107	. . . .	. . .	. . . . .	. . . .	40.4×10^6	. . . .
IVY, United States only	44	4,900	420	1.609	2.0×10^{-2}	3×10^6	M to M + 61 days
IVY, worldwide including the United States	49	330,000	20	4.037	5.1×10^{-2}	111×10^6	M to M + 61 days
	Estimated	. . . .	. . .	371	0.47×10^{-2}	86×10^6	
TOTAL	93	. . . .	. . .	. . . . .	. . . .	197×10^6	M to M + 61 days

Fallout in the United States from TUMBLER/SNAPPER

The TUMBLER/SNAPPER fallout was measured during the period from April 1 to June 18, 1952, at ninety-two stations spread over the United States. The last test of the TUMBLER/SNAPPER series was on June 5, 1952. The fallout measurements for each 24-hr period were extrapolated to January 1, 1953, and were totaled. The maximum total for any station was 14,000 d/min/ft^2* at Elko, Nevada, and the minimum was 86 d/min/ft^2 at North Head, Washington. The average obtained by adding the totals for all stations and dividing by the number of stations was 1511 d/min/ft^2.

Since the stations were by no means uniformly distributed, a better average was obtained by averaging the fallout by states, weighting the average for each state by its area, and computing the weighted average for the United States. This weighted average was 1750 d/min/ft^2. Where there were no stations in a state, or only one or two, the average for the state was obtained by averaging in the fallout from the nearest stations circumscribing the state boundaries. The smallest average was 270 d/min/ft^2 for both Louisiana and Mississippi. The greatest was 6585 d/min/ft^2 for Idaho, and Nevada and Utah were second and third with 6333 and 6167, respectively. Since

$$1\ \mathrm{d/min/ft^2} = \frac{(5280)^2}{3.7 \times 10^{10} \times 60} = 1.256 \times 10^{-5}\ \text{curies/mi}^2,$$

the average fallout from TUMBLER/SNAPPER extrapolated to January 1, 1953, was $1750 \times 1.256 \times 10^{-5}$, or 2.20×10^{-2} curies/mi^2.

The total β activity, as of January 1, 1953, which fell out over the United States (area $= 3 \times 10^6$ mi^2) is then

$$\frac{2.2 \times 10^{-2}}{10^6} \times 3 \times 10^6,$$

or 6.6×10^{-2} β megacuries (Mc).

Worldwide Fallout from TUMBLER/SNAPPER

Fallout from TUMBLER/SNAPPER has been extrapolated to January 1, 1953, for fifteen stations outside the continental United States. Six of these stations reported fallout for the entire TUMBLER/SNAPPER period. The other nine were activated too late to record fallout from the first 3 TUMBLER/SNAPPER shots but did report for shots 5 through 8. The area covered is roughly from 80° W to 15° E longitude and from 60° N to 35° S latitude.

The average of the six stations reporting for the full period is 294 d/min/ft^2. For the nine stations reporting a portion of the shots, the average is 110. The weighted average for the fifteen stations is 212 d/min/ft^2, or $212 \times 1.256 \times 10^{-5} = 2.66 \times 10^{-3}$ β curies/mi^2.

The area between 80° W and 15° E longitude and 60° N and 35° S latitude is

$$\begin{aligned}\frac{95}{360} \times 197 \times 10^6 \left(\frac{\cos 30^\circ + \cos 55^\circ}{2}\right) &= 52.0 \times 10^6 \left(\frac{0.8660 + 0.5736}{2}\right)\\ &= 52.0 \times 10^6 \times 0.7198\\ &\approx 37.4 \times 10^6\ \text{mi}^2\end{aligned}$$

and $2.66 \times 10^{-3} \times 37.4 \times 10^6 = 9.95 \times 10^{-2}$ β Mc. The total for the $(37.4 + 3) \times 10^6\ \text{mi}^2 = 40.4 \times 10^6\ \text{mi}^2$ outside the immediate fallout area is $0.066 + 0.0995 = 0.165$ Mc.

The close-in fallout for each shot of the TUMBLER/SNAPPER series must be estimated on the basis of yield and height of burst. The *total* activity from each shot is given from the following derivation:

The β-ray activity from a 20-KT bomb is

$$\frac{8.2 \times 10^{24}}{t^{1.2}}\ \beta\ \text{particles/sec},$$

at time t seconds after the explosion.(3) The number of β megacuries per kiloton (KT) at a time t days after an A-bomb explosion is then

$$\frac{8.2 \times 10^{24}}{20 \times 3.7 \times 10^{16}} \times t^{-1.2},$$

or $1.108 \times 10^7 \times t^{-1.2}$ Mc/KT at t seconds after the explosion; or, for t in days, the number of megacuries per kiloton is

$$\frac{1.108 \times 10^7}{(86400t)^{1.2}} = 13.2t^{-1.2}.$$

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*Disintegrations per minute per square foot.

Using this relationship, and extrapolating to January 1, 1953, it is found that after the close-in fallout is subtracted, a total of 1.68 β Mc of activity remains to be accounted for.

If this remainder of 1.68 β Mc were distributed evenly over the remaining 157×10^6 mi² of the earth, the average fallout for the 157×10^6 mi² would be

$$\frac{1.68 \times 10^6}{157 \times 10^6} = 1.07 \times 10^{-2} \text{ curies/mi}^2,$$

or

$$\frac{1.07 \times 10^{-2}}{1.256 \times 10^{-5}} = 852 \text{ d/min/ft}^2.$$

This is clearly higher than would be expected in view of the average of 212 d/min/ft² measured for the 37.4×10^6 mi² just to the east of the United States, over which the westerly winds must carry the radioactive clouds before they spread out over the rest of the world.

Since the gummed paper may not retain 100 per cent of the activity falling onto it, particularly if rainfall containing the radioactive material falls on the paper, it is of interest to assume some reasonable value for the average fallout that might be expected over the portion of the world not sampled and to calculate the factor by which the gummed-paper readings would need to be multiplied if all the TUMBLER/SNAPPER activity were to be accounted for.

It would appear that unless the efficiency of collection of the gummed paper was as low as 10 to 15 per cent, 80 per cent or more of the TUMBLER/SNAPPER activity remained suspended in the atmosphere for periods after June 18, 1952, or that large quantities of activity fell in areas not sampled.

Fallout from Operation IVY

Fallout data from Operation IVY, extrapolated to January 1, 1953, were reported for forty-four stations in the United States and for forty-nine worldwide stations. The worldwide sampling covers a much larger fraction of the earth's surface than was the case for TUMBLER/SNAPPER, although there are still large areas in the polar regions and south of the equator, and, of course, behind the Iron Curtain, that were not sampled. In all, the samples cover some 111×10^6 mi². For purposes of accounting for the activity from IVY, the sampled area was divided into four parts and the average fallout for each area was computed. From these averages the total measured fallout was determined. These data are summarized in Table 3 on page 32.

The range of fallout was from 330,000 d/min/ft² at Iwo Jima to 20 d/min/ft² at Lagos, Nigeria. In Area 3, the large reading at Iwo Jima was assigned to an area of 100 degrees squared by noting the relative position of the nearest surrounding stations. A simple numerical average of the fourteen stations in this area would give 35,111 d/min/ft², which would certainly be too high a weight for this exceptionally high reading. If the 330,000 d/min/ft² had been omitted entirely, the average for Area 3 would have been 12,428 d/min/ft².

The average fallout for the 111×10^6 mi² sampled was 5.07×10^{-2} curies/mi², or 4037 d/min/ft². The average for the forty-six United States stations was 1609 d/min/ft².

The average of six stations south of the equator and two stations north of latitude 60° N was 371 d/min/ft². If this last figure is taken as representative of the 86×10^6 mi² not included in Table 3, the worldwide total activity measured during the 61 days following MIKE can be calculated. The total for the 86×10^6 mi² not accounted for in Table 3 is 0.40 β Mc and the total IVY fallout for the world, measured during the 61 days following the test and extrapolated to January 1, 1953, is 6.03 β Mc. This is a very small fraction of the total β activity produced in IVY.

To account for the low measured activity during IVY, by means of the unknown efficiency of collection by the gummed papers, would require that in TUMBLER/SNAPPER more than ten times the activity actually produced would have been measured by the fallout experiments.

Only two possibilities remain to explain the result: (1) either most of the activity remained suspended in the upper atmosphere for periods exceeding 2 months, or (2) large quantities fell out in areas not adequately covered by the sampling network. Since the stations nearest to the test site reporting fallout for the entire period were more than 500 mi from Eniwetok, a considerable quantity of activity could have fallen in

Table 3

IVY FALLOUT

Area	Latitude Range	Longitude Range	Area (mi²)	d/min/ft² as of January 1, 1953	β Mc/mi² on January 1, 1953	Total β Mc as of January 1, 1953
1	80° N–20° S	0°–90° W	32.8×10^6	796	1.00×10^{-2}	0.328
2	60° N–10° S	90° W–180°	25.6×10^6	1,800	2.26×10^{-2}	0.579
3	50° N–0°	90° E–180°	18.9×10^6	19,485	24.5×10^{-2}	4.63
4	60° N–30° S	0°–90° E	33.7×10^6	223	0.28×10^{-2}	0.094
TOTAL	....	....	111.0×10^6	.....	....	5.631

this area. No information is available on the nearby fallout, except from verbal communications indicating that the fallout on other islands in the atoll was not abnormally high and that the islands were safe for occupation soon after the shot. Also, large areas of ocean and the whole area within the Iron Curtain were not sampled. Likewise, the polar regions and much of the area below the equator had no sampling stations. In fact, the ratio of the number of stations to the area outside the continental United States was one station per approximately 4×10^6 mi². The possibility cannot be ruled out that a large part of the activity may have fallen in a number of areas that were not sampled. Such "hot spots" have been observed in the United States from tests in Nevada.

On the other hand, since the cloud from MIKE went through the tropopause into the stratosphere, and there is evidence that a large portion of the activity may condense out in particles of 0.1 μ or less in diameter, it may be that such material will stay suspended in the atmosphere for long periods of time. In support of this hypothesis, it is known that large quantities of dust from the explosion of the volcano Krakatoa stayed suspended in the atmosphere for 2 or 3 years. In this case it is probable that the dust particles were larger than those from an atomic explosion.

IVY Fallout in the United States

As mentioned above, the average fallout from IVY in the United States was 1609 d/min/ft², or a total of 0.0606 Mc, as of January 1, 1953. This represents less than 1 per cent of the total IVY fallout for the world, and it is about equal to the total long-range fallout measured for the TUMBLER/SNAPPER series (0.066 β Mc). The range of variation in the IVY fallout on the United States was from 4900 d/min/ft² in Fort Worth, Texas, to 420 for both San Francisco and Los Angeles, although measurements at a second station in each of these cities gave 680 and 1000, respectively. The fallout was first observed in the United States 6 days after M-day at Los Angeles. The last United States station to record the arrival of the IVY fallout was Atlanta, Georgia, at M plus 23 days.

SPECULATIONS AS TO THE EXISTING HAZARDS

The opinion of nearly everyone who has studied the problem of the

SUNSHINE hazard is that the radioactive isotope Sr^{90} in nuclear weapons is the most likely to constitute the long-term hazard for man. Consequently, it is of interest to examine the quantities of Sr^{90} that may be produced by the detonation of nuclear weapons and the range of concentrations that may be encountered at various places on earth.

Extension of the data given in Appendix V of this report indicates that a megaton of fission will produce 1 mc of Sr^{90}/mi^2, if the fission products from a nuclear detonation are evenly distributed over the earth's surface.*

However, as we have seen from the TUMBLER/SNAPPER and IVY fallout measurements, the fallout from individual explosions varies by a factor of 200, either way, from the average for stations outside the test sites. Consequently, we can expect that even if all the Sr^{90} from the A-bomb explosions has fallen out, the concentration of Sr^{90} will vary over the earth's surface within this factor. *Because of this possible extreme variation of* Sr^{90} *deposition—localized, but long-range—*Sr^{90} *contamination may well be the most important aspect of the* SUNSHINE *project.*

It was also seen that for high-yield, air burst, weapons the greater percentage of the activity may be suspended in the upper atmosphere for long periods of time. Also, it is known† that Sr^{89} has a tendency to fractionate in the cloud, so that its concentration becomes higher as the cloud becomes older. The Sr^{90} should exhibit a similar behavior because of its gaseous precursor Kr^{90}. This would have two effects; namely, the fallout of Sr^{90} would tend to be spread out over greater areas than the total fission-product activity, and the Sr^{90} would have a greater tendency to stay in the upper atmosphere than the total activity fallout measurements have indicated.

Because of the uncertainties concerning the collection efficiency of the gummed paper and the lack of information concerning the fraction of Sr^{90} that may stay suspended in the atmosphere and for what lengths of time, all we can say, at present, is that the range of concentration of Sr^{90} may be of the order of a thousandth of the "limiting" concentration (as defined in the next chapter), but with a large margin of uncertainty.

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*These figures are derived from data as known in 1953. See the Preface to this report for more recent (1956) values.

† Verbal communication, R. W. Spence, Los Alamos Scientific Laboratory.

This concentration is probably reduced even more on land because of the rainfall carrying the Sr^{90} into the ocean or into deep underground waters.

RECOMMENDATIONS FOR FUTURE STUDY

From the above considerations it is clear that additional data are required in order to determine more exactly the quantities of Sr^{90} that may be available for human consumption. If it is practicable to do so, the following additional data should be obtained in future tests.

1. A thorough investigation should be made of the collection efficiency of the NYOO program's gummed paper for radioactive debris from A-bombs. This should include both the dry fallout and the material brought down with rainfall. If possible, it should also be determined if this efficiency changes with distance of travel and with the age of the debris being collected, since the average particle size may be a function of these parameters.

2. While it is obviously difficult or impossible to sample large areas of the earth's surface for the fallout of radioactive debris, more samples covering areas in the southern hemisphere and islands in the oceans should be obtained. Since the polar regions have not been sampled, advantage should be taken of any polar expeditions to obtain such samples. Also, samples should be collected near the test sites, or at least some method of survey should be used, such as that used in TUMBLER/SNAPPER, to make possible an estimate of the total fallout in these important areas.

3. After CASTLE (the next series of Pacific proving-ground tests) an effort should be made to continue the sampling during the whole period before the next test series to see if fallout from material suspended in the atmosphere will continue to be observed for longer periods than the duration of past sampling programs.* There are obvious difficulties involved because of the fission-product decay and the large number of samples that would have to be analyzed. Perhaps increasing the sampling periods and increasing the number of square feet of paper exposed at each station would solve these difficulties.

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*The NYOO is collecting fallout data at some stations during the period between UPSHOT/KNOTHOLE and CASTLE.

4. Since Sr^{90} very probably fractionates, the Sr^{90} content of fallout samples for various representative stations and for varying periods of time after the test should be measured.*

5. If possible an attempt should be made to determine the chemical and physical form of the Sr^{90} in the fallout material.

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*The NYOO is making assays on the combined Sr^{89}, Sr^{90} content of past samples collected in the TUMBLER/SNAPPER and IVY operations. Preliminary results are given in Chapter 5.

CHAPTER 4

BIOMEDICAL CONSIDERATIONS

DEFINING THE MEDICAL PROBLEM

From the biomedical point of view, any attempt to define the lower limit of hazard (exclusive of immediate bomb effects) from the effects of atomic bombing or other atomic energy releasing devices, must be concerned with the problem of radioactivity. The effects of such things as climatologic changes, dust, etc., do not seem to enter into the picture at levels where radioactivity is already assuming considerable importance.

Radioactivity has many effects on the human body, but a quantitative search for the lower limit of hazard rapidly eliminates the short-term acute effects and even the chronic gross-tissue effects, such as necrosis. The problem of a lower limit of hazard, then, enters the realm of carcinogenic and genetic effects. At the present time the data available do not permit a serious evaluation of the genetic problem. It is probable, however, that even when a reasonable estimate of genetic effects can be made, carcinogenic activity will still remain the limiting factor.

The present study, therefore, narrows down to the hazard of carcinogenic activity due to the fission products released. The prevailing opinion of workers in this field indicates that Sr^{90} is the most likely limiting factor by several orders of magnitude when one considers the products released by atomic bombs. Although we have made no quantitative comparison between Sr^{90} and other fission products, we may indicate the importance of the selected isotope because of the following:

1. It is produced in considerable quantity by the atomic bomb (e.g., 1 KT produces 1 gm of Sr^{90}).
2. It is produced in a manner that suggests that it will be in a soluble form and therefore readily available for incorporation into the biosphere.

3. It has a long physical half-life.
4. It is a bone-seeking element in animal metabolism and therefore will have a long biological half-life.

CALCIUM AND STRONTIUM METABOLISM

It is proper to point out that very little is known about the strontium pathway in the human. However, there are certain relationships established between calcium and strontium metabolism that will enable us to circumvent the constriction of "strontium ignorance" by paying attention to certain quantitative aspects of calcium metabolism.

Another important point to consider is the fact that, unlike radium, Sr^{90} will find its way into the human economy in mixture with its stable isotope.* We are thus confronted with a crude tracer-type experiment in which the radioactive material of interest will be quantitatively modified by its relationship to the inert carrier, and that the inert carrier must, inferentially, be evaluated via an assumed relation to calcium metabolism.

Calcium plays an important role in several body functions, but quantitatively about 99 per cent of the element is found in the bones; we may therefore restrict ourselves to bone metabolism. As strontium follows a similar metabolic pathway, our chief items of concern will be deposition and accumulation of radioactivity in bone.

The strontium content of human bones, which forms the basis for subsequent quantitave considerations, is given in Table 4.

The values given in Table 4 are of considerable interest. The relatively constant amount of strontium present holds for all age groups and also for the cadavers preserved since 1914. This would indicate that the variations existing in diet, etc., between the people dying in 1914 (these were all adults) and those dying in 1949 did not appreciably affect the strontium content of the bones. Also, the data indicate a fairly uniform distribution of the strontium throughout the skeleton.

If we take 0.02 per cent to be the average strontium content of bone ash and 7000 gm to be the average weight of the skeleton (50 per cent of this is ash), we arrive at 0.7 gm as the average content of strontium in the adult skeleton.

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*Except possibly through direct inhalation of atomic bomb debris; but see Appendix IV.

Table 4
STRONTIUM CONTENT OF HUMAN BONES

	Strontium in Human Bone Ash (in %)			
	Parietal	Vertebra	Rib	Femur
Fetus	0.016	0.016	0.017	0.017
All ages	0.023	0.022	0.022	0.022
"1914" cadavers	0.027	.....	0.027	0.025

SOURCE: R. M. Hodges, "The Strontium Content of Human Bones," *J. Biol. Chem.*, Vol. 185, 1950, p. 519.

NOTE:
1. The fetal bones showed a total range of 0.015 per cent to 0.019 per cent.
2. The "All-age" group showed no significant increase with age when analyses were compared in five age groups.
3. Except for a 5-month-old female with hydrocephalus and with values of 0.053 per cent to 0.055 per cent, almost all values were in the range of 0.017 per cent to 0.029 per cent.

Figure 3 is a plot of calcium accumulation versus age.(1) These are experimental data taken from growth studies on boys. Assuming that strontium is deposited in bone at the same relative rate as calcium, we can use this curve to arrive at the strontium content for any age. The right ordinate indicates the strontium values plotted for a final Sr/Ca weight ratio of 7×10^{-4}.

Figure 4 indicates the daily accretion of calcium and strontium at various ages. Here again the calcium values are experimental and the strontium values are added, so that the above ratio applies. It is obvious from this graph that the growth years are the ones that will define the tolerance levels allowable in the environment.

Figure 5 plots data regarding calcium deposition in the fetus and is included as a guide for interpreting the results of fetal samples should this study be undertaken.

Some aspects of bone formation and structure are important with regard to the Sr^{90} problem. The most active site of bone formation, and therefore the place of greatest deposition of calcium and strontium, is in the region of the epiphyses. It is here that the long bones grow in length by the

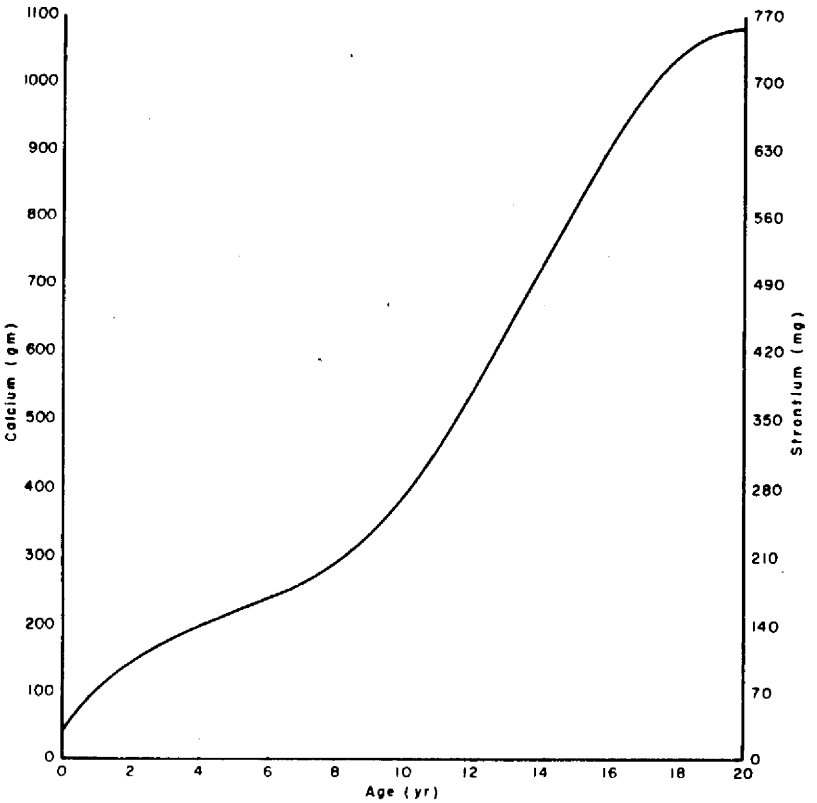

Fig. 3—Accumulation of calcium and strontium in humans

transformation of cartilage to bony structure. As the bone grows in length the deposited minerals will be found farther away from the ends of the bone.

Another important aspect concerns the dynamic equilibrium of calcium and strontium deposits in the bones. It is well known that the calcium demands of the body are fulfilled by withdrawal from the bones, if the diet is inadequate. All evidence indicates, however, that this turnover and dynamic equilibrium is not uniform for the whole bone structure. The chief source of mobilizable calcium is in the organic matrix and the bone trabeculae. The older mineralized bone, which forms the bulk of the bone

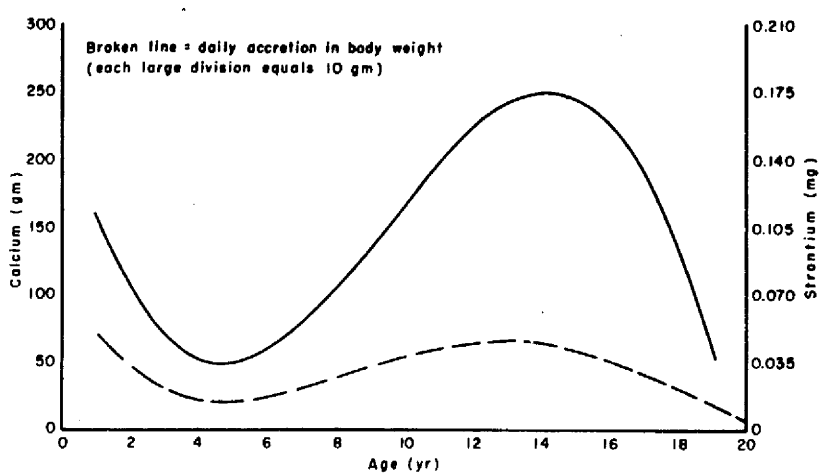

Fig. 4—Daily accretion of calcium and strontium in humans

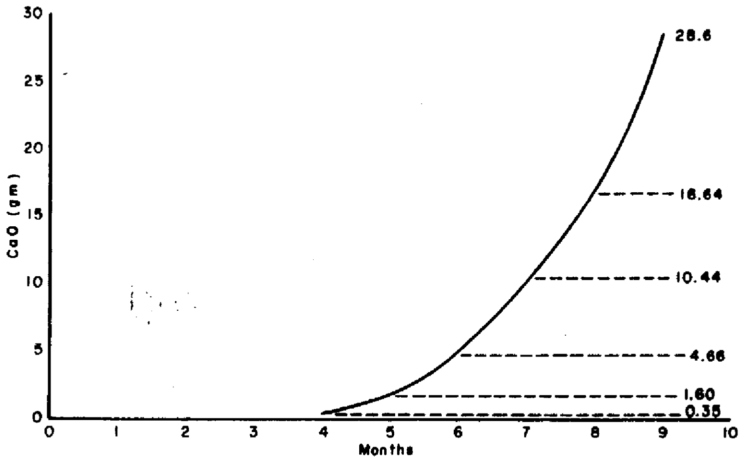

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Fig. 5—Increase in calcium of human fetus (from Hess after Schmitz) (Ref. 2)

cortex, is probably quite inactive and the calcium and strontium that reside there are probably turned over in negligible quantities. *It is this aspect of bone metabolism that renders the quantitative considerations of most recent tracer work inapplicable to the problem we are considering.* The tracer experiments that have been reported are dealing with a relatively high concentration of strontium deposited in the bone and are followed for short periods of time. Thus, in absolute terms, the bulk of the material is followed while it is present in the "high turnover" structures and is not a true quantitative picture for considering the same amount of material deposited largely in the "low turnover" portions of mineralized bone. An experiment reported by Pecher(3) supports this contention. He injected radioactive calcium and radioactive strontium intravenously into mice and after 24 hr he recovered 58 per cent of the calcium and 33 per cent of the strontium. However, when he reduced the amount of strontium injected, the recovery percentage more closely approximated the calcium values. Thus, we are probably further supported when we insist that at the low levels of natural strontium intake we assume that calcium and strontium will behave quantitatively in the same way. This supports the use of the calcium and strontium deposition graphs described earlier.

This quantitative behavioral difference at different ranges of concentration again points out the inapplicability of the tracer experiments being reported in the literature.

This brief discussion of the deposition of calcium and strontium in bone also brings to mind an important quantitative aspect relative to the mass of tissue at risk. If we are discussing the gradual accumulation of a permissible body concentration over the growth years, we can safely assume that the total mass of the skeleton will be uniformly exposed. However, if we are discussing the accumulation of radioactivity in adults, a smaller mass of tissue is involved, probably on the order of one-tenth of the total skeleton. Other factors also enter into consideration. The 10 per cent of the skeleton involved in the adult is in the region of high turnover with regard to dynamic equilibrium, and retention may be quantitatively different over a period of years when compared with that in a young growing person. On a comparative basis we must also bear in mind that young growing tissue is more radiosensitive than adult tissue having lower rates of cell division. In fact, a direct comparison between the effects on adults and the effects on growing children presents a high degree of quantitative uncertainty at this time.

MAXIMUM PERMISSIBLE BODY BURDEN FOR STRONTIUM 90

This study is attempting to define the hazard involved relative to the production of Sr^{90} by atomic bombs. In order to provide a reference standard for quantitative comparisons and evaluations of risk, it is suggested that the maximum permissible body concentration of 1 μc (i.e., 5×10^{-3} μg) of Sr^{90} as established by the International Commission on Radiological Protection (1950) be used.

Any calculations referring to the number of bombs exploded, the fallout problem, etc., may then be extrapolated to this reference point. In this manner we may then estimate, for a given set of conditions, the number of people who have been brought to this level. Or, alternatively, we can estimate the percentage of this level to which all people at risk have been brought.

This figure of 1 μc, or 5×10^{-3} μg, of Sr^{90} (as the maximum permissible body burden) has important implications relative to the amount of Sr^{90} one can calculate as being nonhazardous when released. The dependability of this standard in predicting what it is supposed to predict is a matter of vital concern in any quantitative consideration regarding the release of Sr^{90} into the earth's atmosphere. A few remarks relating to how this standard came into being and to some of the limitations involved would seem to be in order.

There is no direct experimental evidence that Sr^{90} is a carcinogenic agent in man. It has, therefore, been necessary to compare the effects of Sr^{90} in animals with those of radium and then to estimate the equivalent response in man from the known radium effects in man. The generally accepted maximum permissible amount of radium fixed in the human skeleton is 0.1 μg, and the Sr^{90} value given above is supposed to be equivalent to this. (This method of approach was accepted for the bone-seeking elements in preference to the calculation in terms of a permissible amount of radiation equivalent to 0.3 r/week to the critical tissues. Because of the distribution problem in a tissue such as the skeleton, the estimation of the concentration, and so the radiation dose, was considered too uncertain.)

The published data on radium exposure in man indicate that an osteogenic sarcoma has been found with a radium burden as low as 1.2 μg. Thus, the maximum permissible body burden has been set at one-tenth of the lowest amount known to be carcinogenic. However, skeletal changes have appeared in roentgenograms of people with body burdens as low as 0.4 μg. It appears probable that the radium safety level will have to be reduced as more cases are followed and evaluated. It is also important to remember that the International Commission on Radiological Protection was considering occupational exposure. This means that they were interested in evaluating the risk of a limited number of adults. In considering a worldwide population exposure we must remember that children are at risk, that the total years of exposure are also greater, and that a safety factor that is acceptable for, say, one hundred thousand people might not be acceptable for three billion people. In summary, then, it would seem that, from the medical point of view, there are enough uncertainties in the picture to indicate that the presently accepted maximum permissible body burden may have to be lowered. (For another point of view, see "Remarks on Calculation of Worldwide Contamination," Appendix I, page 71.)

SOME CALCULATIONS

If we accept the maximum permissible concentration of Sr^{90} as being 1 μc (i.e., 5×10^{-3} μg) and the amount of strontium in the adult skeleton as being 0.7 gm, then a ratio of

$$\left(\frac{Sr^{90}}{Sr}\right)_{\text{soil}} = \left(\frac{Sr^{90}}{Sr}\right)_{\text{body}} = 7 \times 10^{-9}$$

in the ingested material (each in a form of equal availability) will bring a person to the maximum permissible level over the growth period of 0 to 20 years.

It is also possible to calculate the amount of Sr^{90} that, when deposited in the *agricultural soil*, will bring people living off the products of that soil to the level of the MPC. In view of the fact that the strontium soil-analysis figures have been questioned, this calculation is included as an illustration and any quantitative implications should be deferred until an acceptable average amount for available strontium in the soil is at hand. If we consider 27,000 gm (60 lb) of strontium to be available per acre of land, then approximately 200 μg of Sr^{90} maintained will define an acre's contribution to the diet as one that eventually will result in the maximum permissible body-burden level.

On the basis of uniform distribution, the maintenance of the earth, including *arable land*, at this level will require 2.5×10^{7} gm of Sr^{90}. This amount of Sr^{90} is created by nuclear detonation totaling 2.5×10^{4} MT.

SUMMARY AND CONCLUSIONS

1. The importance of Sr^{90} as a hazard to human beings has been discussed.
2. The reasons why Sr^{90} is the limiting factor relative to human hazard from nonimmediate effects of the atomic bomb have been indicated.
3. The relationship between calcium, inert strontium, and Sr^{90} has been presented and a method for quantitatively assessing the hazard problem has been suggested.
4. The uncertainties regarding safety standards have been pointed out.
5. The lack of firm quantitative chemical analyses with regard to strontium in the soil, etc., is indicated, and it is strongly suggested that these data be obtained.
6. In view of all the uncertainties involved, it is suggested that a food and water monitoring system be set up to assay the Sr^{90} essentially at the point of ingestion.

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CHAPTER 5

SAMPLING AND RESEARCH PROGRAM

THE SAMPLING PROGRAM

From the foregoing description of the present knowledge of the distribution and movement of inactive strontium throughout the various cycles, and from the information available on the amounts and locations of Sr^{90} that have been or will be produced by nuclear detonations, it is quite apparent that insufficient data are presently at hand to design a full-scale investigation of the possible hazard. It is considered essential to develop a better feel for certain critical elements of the problem through a pilot query before launching a major investigation to obtain definite answers to all the important questions that can be posed.

In broad terms, the final study should answer the following needs:

1. To determine if the radioactive fallout that has occurred from the bombs detonated so far constitutes a direct or indirect menace to the health or well-being of humans.
2. If no hazard has yet occurred to man or to his economy from the present amounts of fallout, to determine the number of bombs, with proper consideration to location, rate, and mode of detonation, that may be tolerated to keep the fallout below a safe maximum.
3. To determine the mechanisms whereby undesirably high amounts of radioactive materials may be concentrated in man or products essential for his existence.

The pilot study, then, should be designed (1) to assist with the intelligent planning of a full-scale investigation to meet the objectives listed above and (2) to obtain some data to provide the basis for a quick first-order estimation of answers to some of the most important questions. The sampling program, which will provide material for the experimental portions of the pilot study, should be flexible and should be allowed to change during the study, as the first results obtained will serve as a useful guide in further sampling. Thus, experience will tell whether the sampling program outlined in this section is adequate.

An important part of the sampling program is the "mechanistic" sampling, i.e., sampling aimed at determining the physical form of the contaminant and the methods of nonlocal dissemination. This sampling program should be carried out in conjunction with what might be termed the "biological" sampling. The latter is carried out to determine how and to what extent the material is absorbed by the biosphere. We shall describe the former program only briefly; it is the most difficult and will take the longest time to develop. If data are to be obtained on an urgent basis (partially because of greater contamination after the CASTLE test series), it is believed that the results of the biological sampling program will be the most significant.

The first "mechanistic" problem is to determine the particle size and nature of particles. (See Chapter 2 and Appendix II.) There are strong indications that much of the particulate matter important to long-range considerations is in the form of ultra-finely-divided glasslike particles, possibly chemically inert. Sampling of the atmosphere requires the handling of matter in this new form.

There is so little experience with aerosols involving particles of the ranges probably occurring that the problem of sampling the atmosphere is one of no minor difficulty. There does, fortunately, seem to be two possible solutions to it—electrostatic precipitation and diffusion barriers. It appears that the electrostatic precipitation process, being extremely efficient for the collection of very-small-diameter particles, will be adequate for the task. Failing this means, the use of a diffusion barrier does seem to be promising (see "Analysis for Strontium 90" on page 58).

Because of the fact that Sr^{90} is formed through the decay of the precursor, the chemically nonactive isotope Kr^{90}, there is a strong possibility that the contaminant will be available in nearly atomic form to plate out on the newly formed particles and atomic debris. If so, the Sr^{90} should be easily available for solution. This hypothesis could be tested by various experiments designed to determine rate of "weathering" of debris after deposit. How rapidly plants take up Sr^{90} may also provide a measure of the degree of plating.

Another important aspect of the "mechanistic" program is to determine the scavenging and other methods of bringing down the contaminant from the lower and upper atmosphere.

Up to altitudes obtainable by aircraft the atmospheric sampling program appears relatively simple. The great question of what to do for samples at heights higher than those obtainable by aircraft has not been answered completely. There are possibilities of using balloons for indicating the general radioactive levels by telemetering or by actual collection of a sample. The need for these observations stems from the lack of knowledge of atmospheric processes. It has been suggested, and the suggestion appears reasonable, that fine particulate matter may be held for long periods in the stable regions of the stratosphere. Such a storage would lead to a slow, relatively steady deposition as the debris was admixed with the weather layers and brought down by the processes of eddy diffusion, fall, and rain scavenging. Some evidence of a small but measurable "drip" of debris has been reported by Merril Eisenbud of the AEC New York Operations Office. It should be noted, however, that there is very little knowledge as to how the troposphere is cleaned of the debris or of how much deposited debris is raised from the ground by wind and redeposited. Until an adequate series of samples is taken from the atmosphere, it will be impossible to show, in an unambiguous fashion, how much debris is retained in the atmosphere and how rapidly it is removed.

A first requirement of the "biological" sampling program is to establish the usefulness and sensitivity of the analytical procedures for Sr^{90} that are described later in this chapter. Concurrent with this step it is necessary to determine the general levels of Sr^{90} that may be anticipated in the samples that will be used. This is needed both to set the sensitivity for the analytical procedures and to determine the size of samples to be collected. Such preliminary information can probably be obtained from several types of samples that have already been collected by other investigators.

The first of these samples are some of the fallout specimens collected by the AEC monitoring system. (Do they contain Sr^{90}, and how much? A recent preliminary analysis showed that the samples do contain radioactive strontium, but no distinction between the 89 and 90 isotopes was made.*) Several of the more active of the older samples from TRINITY and the active rain water collected in eastern Massachusetts will be useful for the calibration mentioned. Inasmuch as human samples are important in all phases of the investigation, it is desirable to have at least one sample available during the early stages of the experimental program. (Arrangements for such human bone samples from the Boston area have already been made by A. K. Solomon of the Harvard Medical School.)

The discussion of the factors involved in this problem as presented in the earlier sections of this report serves as a guide to the type of sample and the location from which samples should be taken. It is fairly obvious that the samples should be obtained both from locations that are near to detonation sites and that represent exposure to high fallouts and from locations downstream from the detonations representing areas of lesser exposure. The areas so chosen might also represent a variation in other than distance and exposure. All such variables as dietary habits, source and types of food, water and milk sources, amount of rainfall, and local

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*Private communication, Merril Eisenbud, "Strontium Analysis of NYOO Fallout Samples," July 23, 1953:

> "The method of analysis consisted of dissolving in acid the ashed fallout samples. The alkaline metals were precipitated as oxalate and were followed by two separations of strontium as the nitrate. Collected samples spiked with strontium-90 yield an 80% to 85% recovery.
>
> "The confirmatory yttrium milking procedure has not at this writing been completed. The strontium assays follow:

Place	Date	Counts per Minute Total	Strontium	% Strontium
T/S 1 Boise	6/5	6600	760	11.3%
2 Boise	6/5	7500	250	3.3%
3 Gooding	6/5	2600	82	3.2%
IVY 4 Iwo	11/5	8800	218	2.5%
5	11/4	26900	750	2.8%
6 Manila	11/6	1700	39	2.3%

	Theoretical	
T/S 5%	3–89 2–90	Reference Hunter and Ballou."
IVY 7½%	6½–89 1–90	

soil types may influence the amounts of Sr^{90} reaching humans or affecting other important bionomic cycles.

With these factors in mind, the following six areas are suggested for investigation in the pilot program: (1) northern Utah or southwestern Idaho, (2) Kansas or Iowa, (3) New England (Boston), (4) South America, (5) England, and (6) Japan.

There are fallout data already available from most of these areas. Area 1 is selected as representing an area of maximum fallout. Area 2 represents lesser fallout, but is important as the center of the grain-growing region. Area 3 has still less fallout, but is an area where considerable information is already available on radioactive contamination of rain, streams, and reservoirs. South America, Area 4, provides an area in the southern hemisphere; and England, Area 5, is in a northern-hemisphere area where exposure should be very low, but samples readily obtainable. Japan, Area 6, is included because excellent arrangements are presently in effect there for obtaining human specimens from all age groups. Then, too, the Japanese specimens will represent a much longer exposure time and will also provide an opportunity to observe any effects of different diets and living habits on the concentration of Sr^{90} in humans.

In each of these six areas, the following types and numbers of samples should be considered:

Human material, 12 samples	0 to 10 years old, epiphysial end of rib, pooled—2 samples, 2 teeth 10 to 20 years old, epiphysial end of rib, pooled—2 samples, 2 teeth Over 20 years old, epiphysial end of rib, pooled—2 samples, 2 teeth
Livestock, 10 samples	Young animal, rib—3 samples Mature animal, rib—3 samples Bone meal—3 samples Teeth—1 sample
Foodstuffs, 8 samples	Cow milk, or milk solids—3 samples Feces, human, pooled—1 sample Local truck farms—1 grain, 1 root, 1 leaf Local egg shells—1 sample
Waters, 3 samples	
Soils, 3 samples	

The total number of samples from each area is thus 36, giving a worldwide total of 216.

The human specimens are essential for determining the actual levels of Sr^{90} that may now be present in the human body. As demonstrated in the earlier sections of this report, present knowledge of the distribution of Sr^{90} and the manner it may reach humans is inadequate for predicting under what circumstances and in what amounts it may reach the body. Previous discussion provides good argument that bone is an excellent collector and integrator of strontium. Thus the suggestion that bone specimens be used. Tooth specimens are also suggested, primarily to determine if they may not also provide concentration of strontium. The literature does not agree on this. If teeth are proved useful indicators, they will be more readily available for future studies than human bone.

The three human age groups are suggested because of the postulated difference in the uptake of strontium in these ages. It will be necessary to have specific information on this before any definite statements can be made regarding the relation between concentration of Sr^{90} in an environment and the maximum safe body burden. This requirement for human specimens suggests that only fairly large metropolitan areas be selected in the locations states. This will ensure the availability of sufficient quantities of human specimens and the presence of large hospitals with adequate staffs who can properly collect and segregate the specimens desired.

The other specimens listed as of interest for analysis have several important functions:

1. They will provide a check on fallout in the area as obtained from present sources, or give an idea of the amount of fallout where other data are not available.
2. They will present a cross section of the diet of the human population and thereby provide a means of correlating concentration of Sr^{90} in diet with the concentration in the human body.
3. They will represent samples of the hydrosphere, lithosphere, and a few bionomic cycles of importance to man, so that any concentrations of Sr^{90} that may build up in any of these prior to entry into man may be detected.

In addition to the 216 samples listed above, the following several samples of a general nature may give useful information about the value of other types of samples for possible use in the subsequent investigation:

1. Whole human skeletons. These are necessary in order to compare bone or tooth samples to whole body samples. They will be difficult to obtain and to assay, but such samples are essential.
2. Plasma, or calcium from plasma. Information about this must be secured from the American Red Cross or from the National Blood Committee. If plasma is routinely collected over resin columns, the discarded resin columns will contain almost all of the plasma calcium and can easily be leached to provide a large, pooled sample.
3. Additional hydrosphere samples. These should consist of one or more of the following: tuna fish bones from a cannery, oyster shells, and brown seaweed, which, it is believed, concentrates strontium.

For the reasons given in the biomedical section of this report, analysis for inactive strontium and calcium in all of the samples collected and used is considered most essential.

It is difficult at this time to guess at the cost for the analysis of each sample. Quick estimates ranging from $50 to $1000 a sample have been made. If the pilot analyses, as outlined here, prove to be too expensive, it may be necessary to decrease the number of samples. The list of samples in this chapter was prepared in the order of descending desirability. Consequently, the first economy would be to cut down the number of soil and water samples from 3 to 2. The second economy would be to cut down the number of foodstuff analyses. Beyond this point, it seems desirable to cut down the number of places studied, removing first, Area 4 (South America), second, Area 5 (England), and third, Area 6 (Japan). It is hoped that these stringent economies will not prove necessary, since the value of the study will be considerably impaired by cutting down the number of areas surveyed. On the other hand, if more samples could be obtained in the pilot program, one could increase the number of samples taken at each place to make the statistics more representative. Each set of 36 samples is regarded as a minimum quantity; more than one quantum might give results of greater significance.

The collection of samples will undoubtedly be expedited and the quality of sampling much improved if an individual associated with the study travels to the foreign place to supervise the sample collection. Such personal contact will undoubtedly improve the efficiency of sample collection for the domestic cases, too.

Nothing has yet been said about the probable size of the specimens required, statistical factors that relate to the sampling program outlined here, or the laboratory techniques. These subjects are discussed in the following sections of this chapter.

Statistical Considerations

The general area of study and the outline of a pilot sample survey have been presented in the previous section. The carrying out of this pilot sample survey poses several statistical problems that will have to be solved before satisfactory results can be obtained by the survey. Basically the science of statistics has two contributions to make to sampling problems:

1. To enjoin randomness in the selection of individual observations at various points in the sampling process so as to ensure against bias in the estimates obtained;
2. To increase the accuracy (minimize the standard error) of the estimates of the required quantities (e.g., average lifetime dose given some steady-state situation) by taking advantage of the structure of the underlying process in the design of the collection and in the analysis of the sample data.

The first of these contributions will come into play when the detailed prescriptions are written out for the collection of the various samples described in the preceding section. In fact the description of the samples themselves will have to be greatly expanded before any sampling can be done. Since the proposed sample survey is a pilot-type program, the second contribution of statistics cannot come much into prominence. Pilot programs are usually designed to gather information so that a subsequent, larger sampling program can be more judiciously designed, using the considerations that are subsumed under (2) above. At present we know very little about the processes we are studying, so that there is not much structure in the problem to be taken advantage of now. Something, of course, can be done, and an example of the kind of analysis one might attempt in the case of the human samples is described in the following paragraphs.

A Sample Analysis of Human Data

All hazard calculations will have to be specific to some geographic distribution. The food supply of a given city, however, comes from various regions, some of them quite distant from the city itself, but in most cases the water, milk, and fresh vegetables are locally supplied. In any case different cities have different food-supply areas and any analysis will have to take this into account. The only question is how this is to be done.

The following is a crude model of the sort of scheme on which an analysis may be built. The model has two elements: an exposure function $E(t)$ and an ingestion function $\alpha(t)$, in which, for any person, the age and all chronological times for the community are normalized to it by translation in the time axis. By exposure is meant the amount of Sr^{90} available in the total environment of the individual. The environment of the individual no matter where he lives in this country includes Kansas, for example, if the wheat in his diet comes from Kansas. The dose of an individual at age T is

$$D(T) = \int_0^T E(t)\alpha(t)\,dt.$$

The real problem for even the crudest analysis is what variables are to enter into $E(t)$ and $\alpha(t)$ and how are we to measure them. The following is suggested as a first attempt to relate the Sr^{90} deposited in the bones of human beings to some calculated exposure. Let $E(t)$, the exposure function, be determined by (1) the geographic and time distribution of fallout of Sr^{90}, (2) the food sources of the area (city) under study, and (3) the gross scavenging process of nature by means of which the fallout is made nonavailable. Denote by $X(\tau)$ the fallout of Sr^{90} at time τ per unit area over an appropriate area; the exposure $E(t)$ might then be assumed to be equal to

$$E(t) = \int_0^t X(\tau) \exp\left[-\beta(t-\tau)\right] d\tau,$$

where β represents a generalized decay and scavenging parameter. As a practical suggestion for computing $X(\tau)$, the activity level of Sr^{90} deposited, it seems sensible to weight equally the average activity level in areas within 150 mi of the city and the United States general average of time τ. The results of the soil samples and Eisenbud's data, corrected to Sr^{90} activity in whatever way seems appropriate, will have to form the basis of the estimation of exposure.

The ingestion function $\alpha(t)$ is a function of (1) age (metabolism of strontium and diet as influenced by age), (2) the general efficiency of the whole biological and plant process whereby Sr^{90} goes from the lithosphere into human beings, and (3) the availability of Sr^{90} to plants, as it is laid down, due to incasement in glass beads, soil fixation, etc.

In this general context the sampling should be roughly as follows:

1. In the 6 sample areas (probably large cities within the areas), collect rib bone samples from autopsies by age groups 0 to 10, 10 to 20, 20 and over.
2. In each age group, for each city, collect roughly 200 gm of bone; randomly sort this into two 100-gm samples. This size of sample will allow one to detect 10^{-4} of a lifetime tolerance dose, assuming that this lifetime dose is 1μc of Sr^{90} and that five counts per minute will be required to obtain a good determination of the Sr^{90} activity level (two counts per minute assumed as background).
3. In these cities one will, after the analysis of these samples, have a picture of the current levels of Sr^{90} activity in human bone material as a function of age. (The 100 gm per assay may have to be increased if the current level in human beings is less than 10^{-4} of the tolerance dose. If so, 1000 gm will detect 10^{-5} of the tolerance dose, etc. It has been suggested that a few preliminary assays be done to determine just what level we are dealing with. If it is very low, say 10^{-6} or 10^{-8} of the tolerance dose, the size of the sampling program may have to be revised to save money.)
4. For each grouped sample of 100 gm, one must compute the function $X(\tau)$ for the city and age group under analysis. The ages of the persons in the sample will be used, and in the pooled dose the separate ages should be taken into account, or at least the average age of the bone in the pooled sample. In general it will be best to have complete case histories of the various people in the pooled samples. These should be collected with the samples. The ingestion function $\alpha(t)$ is to be approximated by a step function α_i $(i = 1, 2, 3)$, a series of constants, one for each of the six age groups. The parameters to be estimated are the α_i's and β. In the absence of any detailed structure in the model, it appears reasonable to use a least-squares fit to estimate the parameters; accordingly we are to minimize:

$$\text{Min} \sum_{i=1}^{3} \sum_{j=1}^{2} [\alpha_i E_{ij}(\beta) - Y_{ij}]^2 \qquad (i = 1, 2, 3),$$

where $E_{ij}(\beta)$ is the calculated exposure for the ijth assay sample and Y_{ij} is the observed amount of Sr^{90} per gram of bone in the 100-gm sample. The exact method for computing $E_{ij}(\beta)$ will have to be worked out.

The Problem of Obtaining Individual Samples

The problem of collecting individual samples has been mentioned earlier. The experience from other sample surveys indicates that great care must be taken in writing out the detailed instructions for the collection of individual samples so that they will be properly collected, labeled, and described. Usually, sampling programs are more successful if everything except the actual collection of the sample item is done at one central headquarters. For example, in the case of the soil samples it seems reasonable that the following types of things be done:

1. For each city the surrounding area must be delineated and a map of the area obtained; on this map the food-producing soil areas must be delineated (presuming these soils to be the only ones we are interested in because only the Sr^{90} in these soils represents a human ingestion hazard).
2. From these areas the three samples must be selected in some way. Suppose each sample is to consist of the soil in a volume 1 yd^2 by 7 in. deep. We have the choice for each sample of pooling n smaller samples or of having one sample of the given volume. It would appear better to have at least some pooling of smaller-sample volumes, but it is not clear what n should be. There are certain costs of collection involved in making N very large, and these must be balanced off against the increased information obtained about the average Sr^{90} level of soil in the area. In addition there are schemes of sampling intermediately between complete random sampling for all n subsamples and having only one sample 36 in. by 36 in.

by 7 in. One of these involves first drawing *k* random subareas for sampling, and then within these subareas drawing *m* samples of soil, where $km = N$, the total sample number, each of which is equal to $1/N$th of the volume of soil required. The most advantageous sampling procedure will depend on the variability of the Sr^{90} level in the soil within the area and on the relative cost of going to the *k* subareas as compared with the cost of taking each sample of $1/N$th of the required volume of soil.

3. The actual analysis of the data, once collected, is so straightforward (unless analyses other than the sample determination of the Sr^{90} level and inert strontium level are undertaken, e.g., analyses of the soil composition and its effect upon the availability to plant life) that there is no need to mention it. The primary problem is the collection problem and it is here, on matters of detail that cannot be foreseen now, that the help of a statistician is most needed.

Similar consideration will enter into the design of the collection program for the other types of materials, e.g., animal bone, etc.

ANALYSIS FOR STRONTIUM 90

Sampling

The general principles of sample treatment and measurement are fairly obvious. One must, in the first instance, collect a fair sample of the realm being investigated. We must take a fair portion, bearing in mind the vast mixing problem, and then collect from this portion all the radioactive fission products present. From remarks made previously it is clear that the collection of all the radioactive fission products present may not be a simple task, particularly in the case of the atmosphere, for we have the possibility that a considerable portion of the radioactive particles involved are of a very small size—i.e., less than 0.1 μ in diameter and possibly of the order of 0.01 μ. Particles of this dimension are extremely difficult to isolate. The method of electrostatic precipitation is presently being exploited by the Stanford Research Institute and by the New York Operations Office of the AEC. The results are encouraging.

Additionally, we recommend that a study be made of possibilities of the diffusion barrier, since it is clear from its physical characteristics that it, indeed, must remove particles however small. This latter point is susceptible to direct test, however, by use of several barrier series and observance of the distribution of the radioactivity. That is, if the barriers are completely effective, the first in the series will absorb all the radioactivity. If it is true that diffusion barriers can be utilized to make a complete radiochemical assay on air, one might well develop a technique of producing other filters less security sensitive, but which will perform as well for the problem of removal of the colloidal aerosol fission-product matter from the atmosphere. This could be done by using bomb debris as test material, i.e., by purposely selecting fallout material as test material, or possibly by synthesizing similar material in the laboratory and conducting tests on various filters. This work could be done in the Carbide and Carbon Corporation laboratories in the diffusion plant at Oak Ridge. Professor John Turkevich of Princeton University has observed that the particles produced by an electric arc between aluminum electrodes in air have particle sizes in the range of 0.01 μ and smaller. It is conceivable, therefore, that the use of radioactive material for the electrodes would allow one to synthesize an aerosol similar to that produced by the atomic bombs. Then this might be used in developing adequate filters.

In sampling the hydrosphere one must remember the propensity of the radioactive material to pass through filters. It seems, therefore, that evaporation is necessary. The evaporation of considerable volumes of water is an unsavory task, but in the absence of information as to how to be certain of the removal of the fine colloid, no alternative seems to be available. Following evaporation, the salts left can be treated for the dissolution of such inert materials as silicon dioxide, aluminum oxide, etc., so that the radioactivity will be in true solution. The procedure for this is well known to be the treatment with about 50 per cent hydrofluoric acid solution in platinum vessels. This procedure will suffice. It may be unnecessarily drastic, however, as only direct test can show. The carrier elements, such as strontium, should then be added and an ordinary radiochemical analysis made. In the absence of an effective dissolution procedure, however, misleadingly low assays could be obtained. It is difficult to overemphasize the importance of guarding against the lost part of the radioactivity in ultra-fine colloidal form.

Lithosphere sampling is quite difficult in that the treatment of any considerable amount of dirt with hydrofluoric acid to guarantee the dissolving of the colloidal fission particles is a most unpleasant task. There does not seem to be any obvious alternative at the moment, however, to the complete dissolution of a dirt sample followed by a treatment with hydrofluoric acid solution.

The sampling principles for the biosphere are fortunately less than annoying. If the strontium has been incorporated in the biosphere, it certainly is in chemically active form. Therefore, the direct ignition of the biological material followed by the dissolving of the ash in a slightly acid solution and the radiochemical processing of the solution is a perfectly allowable procedure.

Chemical Processing of Samples

After the samples from the atmosphere, the hydrosphere, the lithosphere, and the biosphere have been properly dissolved, the subsequent processing should be accomplished approximately as follows:

1. The addition of a comfortable amount of nonradioactive strontium salt to serve as a carrier, together with other salts to serve as holdback carriers for the other fission products.
2. The separation of a pure salt of strontium from the solution. This might, in the case of the majority of samples, consist in the precipitation of carbonates by the use of sodium carbonate solution, followed by appropriate purification, such as by ion exchange or by any of the well-known precipitation methods.

We are fortunate in the nature of the decay scheme of radioactive strontium, which proceeds as follows:

$$Sr^{90} \rightarrow Y^{90} \rightarrow Zr^{90}.$$

The strontium activity corresponds to a pure β-ray spectrum of 0.54 Mev (upper limit) and a half-life of 20 years, while the Y^{90} daughter transformation involves an emission of 2.18 Mev of β rays with a half-life of 61 hours. No γ radiation is involved in either. Following the precipitation of the carbonate mixture, one could either purify the strontium as indicated above or, without further purification, redissolve the carbonate precipitate with dilute nitric acid, add a small amount of ferric nitrate, and neutralize with ammonium hydroxide. This would produce a precipitate of ferric hydroxide that should scavenge all yttrium and rare-earth activities very efficiently. To make certain of the purification of the calcium-strontium carbonate precipitate for yttrium, one might have to add a little yttrium before, or together with, the ferric nitrate. Actually lanthanum nitrate would serve as well, and it is cheaper. After purification of the strontium-calcium solution for yttrium, one should reacidify with dilute nitric acid and allow the system to stand for regrowth of the 61-hour yttrium activity. At this point a few milligrams of yttrium could be added and the solution could be treated again with ammonium hydroxide until alkaline; the yttrium precipitate could then be removed by filtration and counted.

The only other fission-product activity likely to be present in this yttrium precipitate is La^{140}, which arises from the 12.8-day Ba^{140}. This, therefore, would be important only in relatively young samples.

The possibility that Ra^{226} and Pb^{210} (RaD) would also be present in the calcium-strontium precipitate should be considered. The RaE growing from RaD might appear in the yttrium sample and interfere with the counting of Y^{90}. In this eventuality it would be necessary to separate the strontium from lead before growth of the Y^{90}.

Counting Procedures

It is clear from the procedure outlined above that the yttrium milking technique allows one to concentrate the yttrium daughter activity in equilibrium with any given amount of strontium in very small amounts of solid yttrium sample. Therefore, one can envisage the use of Geiger counters with the lowest backgrounds and the maximum sensitivity. The penetrating nature of the radiation of Y^{90} allows one to use samples as thick as several milligrams per square centimeter without serious self-absorption, and in fact to employ the known enhancement of counter sensitivity by backscattering from supports made of elements of the largest atomic numbers, such as gold or platinum. In other words, mounting thin layers of yttrium salt on gold or platinum should increase the chance of recording disintegrations occurring in the solid yttrium samples to something around

75 per cent. It should be possible to do this on counters small enough to ensure the detection of the few disintegrations per minute expected in the average sample for the worldwide assay. Typical counters that have been used in other connections have sensitive areas of 200 to 400 cm² with wall thicknesses of 2 or 3 mg/cm² and background counting rates of six or eight counts per minute. This means that up to 1 or 2 gm of yttrium sample can be mounted in thin layers under conditions of maximum common sensitivity, thereby making it possible to measure disintegration rates of one or two, or to be conservative, five, disintegrations per minute with some degree of accuracy. The techniques employ the principle of anticoincident shielding for the reduction of the counter background. The construction of counters of clean materials and the general techniques employed in other low-level counting applications should suffice. The particular apparatus has been described by several workers—e.g., the apparatus used in natural radiocarbon assay.(1)

The problem of detection and measurement of weak radioactive substances is an ancient one, around which considerable lore and artistry have been built. In the case of radiostrontium at the levels likely to be found, one needs to use some of the more sensitive techniques known. On the other hand, it is essential that the procedure be as simple and as reliable as possible. There are two obvious ways in which to proceed in the low-level strontium-measurement problem. One is to measure purified strontium, as such, in equilibrium with its yttrium daughter—the equilibrium being ensured by allowing the purified strontium to stand for at least a week before measurement. The other is to separate the yttrium daughter and to measure it alone. The first technique has the advantage of giving more radiation for measurement, but is obviously inapplicable to samples that contain a large bulk of strontium and are thus more subject to radioactive contamination errors. The second technique, though it involves sacrificing half of the radiation, affords the opportunity of measuring a large bulk of material by treating the yttrium daughter, which in itself [redacted] provided the sample has previously been purified for bulk yttrium and other rare earths. In order to demonstrate the desirability of the second procedure in these instances, we will give some calculations below that indicate the regions of applicability of the two procedures.

Since both strontium and its yttrium daughter are pure β-ray emitters, one immediately chooses a Geiger counter as the detection instrument to be used, since it is the only instrument that will detect a single thermal energy electron with apparently 100 per cent efficiency. It is, therefore, clearly the most sensitive of all instruments for the detection of ionization as such. Having selected the Geiger counter, one seeks to mount the strontium or yttrium sample in the position that will ensure the maximum ratio of sample activity to background activity. It is clear that the counting geometry of the Geiger counter should be as high as practicable. Several different types of counters might be considered, such as (1) the 4π type, (2) the conventional thin-window bell type, (3) the windowless flow-type, and (4) the thin-walled cylindrical type.(2) The choice of a particular type of counter is somewhat arbitrary and will depend on the samples to be analyzed and on the availability of the equipment.

It is further clear that one should not interpose between the counter gas and sample any more solid material than is necessary, though both the strontium and yttrium radiations are quite penetrating (Sr^{90} range, 180 mg/cm²; Y^{90} range, 1065 mg/cm²; half-thicknesses, 14 and 134 mg/cm², respectively). The solid material in the sample itself must also be considered. For an isotropic source, if X represents the thickness of the sample in mean free paths ($X = 0.693\ T/\tau$, where T is the thickness and τ is the half-thickness in milligrams per square centimeter), then the flux through the surface of the sample is given by

$$F(X) = \frac{N}{2\mu}\left[1 - e^{-X} + \int_0^X zEi(-z)\,dz\right], \tag{1}$$

where N is the number of particles originating in unit time in a unit volume of the sample, μ is the absorption coefficient, $Ei(-z)$ is the exponential integral $\int_z^0 (e^x/x)\,dx$, x is the depth of the volume element below the surface, and $z = \mu x$. The activity from a sample of thickness T mg/cm² is then

$$A = A_0\left(\frac{\tau}{0.693}\right)\left[1 - \exp\left(-\frac{0.693T}{\tau}\right) + I\left(\frac{0.693T}{\tau}\right)\right], \tag{2}$$

where $A_0 = ½$ times specific activity in d/m times area in square centimeters, and I is the integral in Eq. (1). (For an anisotropic source the integral I is deleted.)

The activity from the combined Sr^{90} and Y^{90} in a sample of thickness T is given by

$$A_{Sr,Y} = A_0 \left\{ \frac{\tau_{Sr}}{0.693} \left[1 - \exp\left(-\frac{0.693T}{\tau_{Sr}} \right) + I\left(\frac{0.693T}{\tau_{Sr}} \right) \right] + \frac{\tau_Y}{0.693} \left[1 - \exp\left(-\frac{0.693T}{\tau_Y} \right) + I\left(\frac{0.693T}{\tau_Y} \right) \right] \right\}. \quad (3)$$

If the yttrium were separated from this same sample with a small amount of carrier, its activity would be

$$A_Y = A_0 T .$$

Figure 6 shows a plot of these two functions using 14 mg/cm² and 134 mg/cm² as the half-thicknesses for Sr^{90} and Y^{90}, respectively. The intersection of the two curves occurs at 90 mg/cm².

Thus, with a given sample of material to be analyzed and a particular counting arrangement, one could decide whether to simply isolate strontium and count Sr^{90} and Y^{90} together or to separate and count the Y^{90} alone. If the sample thickness would be much greater than 38 mg/cm² when both were together, then it would be advantageous to separate the yttrium. For very thin samples of, say 20 mg/cm² or less, strontium and yttrium should be counted together. In the intermediate range of 20 to 55 mg/cm² it would make little difference as far as counting rate were concerned and the choice would be a matter of convenience.

The principal task in measurement of extremely small amounts of radioactivity remaining after one has supplied a sufficiently sensitive counter is the reduction of the extraneous background rate due to cosmic radiation and the ubiquitous radioactive products of uranium and thorium that exist in all laboratories in such amounts as to give very appreciable count rates. The latter component of the background is best removed by selecting solid material that is free of uranium and thorium and of their disintegration products such as radon. Consideration of this problem, together with the problem of cost, has led to the belief that steel should be an excellent material for a low-level counter shield.

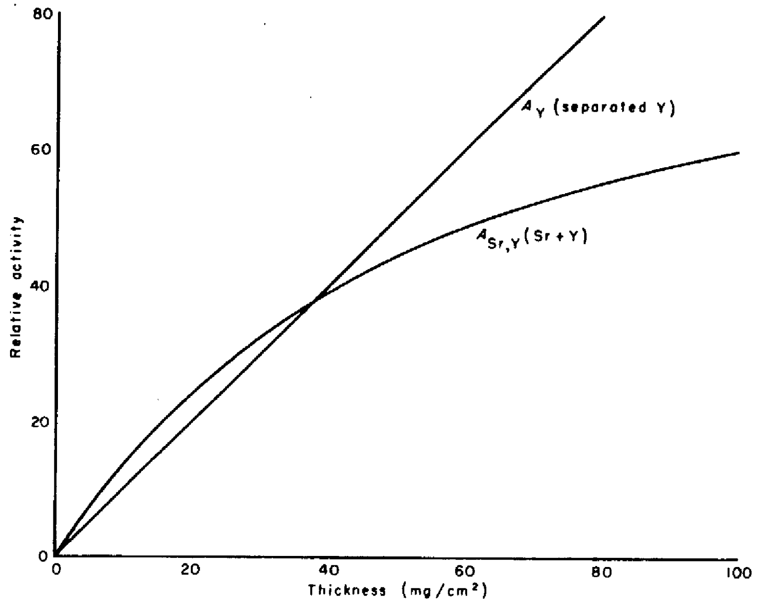

Fig. 6—Relationship between the activity and sample thickness for Sr^{90} and Y^{90} in equilibrium and for separated Y^{90}

The method used to eliminate the cosmic-ray component of the background consists in surrounding the counter and sample with a complete layer of Geiger counters that are in tangential contact. With these "anti-coincidence counters in place the background is reduced to a very small value. It can be reduced further by incorporating a device developed by Kulp(3) who introduced a 1.5-in.-thick shield of distilled mercury between the anti-coincidence shielding counters and the central measuring counter. By this means he was able to reduce the background count from five to two counts per minute.

The apparatus as described involves no extraordinarily complicated parts. In particular, since the electronics is very simple, few difficulties develop during operation. Occasionally one of the vacuum tubes in the apparatus may need replacing or some one of the anti-coincidence shielding counters may cease to function properly and require replacement, but, generally speaking, the apparatus is rugged and trustworthy and can be depended on for continuous operation over long periods of time. The total cost would be in the vicinity of $3000, depending very largely on whether the electronics is purchased or constructed in the laboratory. The electronic components, as well as the anti-coincidence counter tubes, are available from commercial companies. The shield would require special construction.

Sensitivity of Strontium Assay Procedure

One could expect to detect some six counts per minute on a background of six with reasonable time and reasonable accuracy of measurement. This would correspond to a specific activity of $6/W$ μμc/gm of sample, where W is the total weight of the strontium in the sample in grams. Thus a sample large enough to contain 10^4 μμc/gm is measurable. Correspondingly smaller samples would suffice for materials of higher specific activity.

With this result in mind one can speculate a little about the probable difficulties to be encountered in the worldwide Sr^{90} assay being recommended in this report. For a nominal atomic bomb of 20-KT energy that releases 1 kg of fission products, assuming a 5 per cent fission yield, 1.3×10^{23} atoms of strontium are produced. Uniform dissemination of this material over the 5×10^{18} cm^2 area of the earth would yield a superficial Sr^{90} density of 7.5×10^{-4} μμc/cm^2.

It seems very unlikely that the fallout strontium would have been mixed with more than 1 gm of strontium per square centimeter of the earth's surface, thus giving a specific Sr^{90} activity of 7.5×10^{-4} μμc/gm, which, as seen above, is measurable. We can *probably* conclude, therefore, that the Alamogordo bomb can now be detected in distant places of the earth if worldwide contamination is a characteristic of explosions on relatively short towers.

The specific Sr^{90} activity in soil today, assuming worldwide distribution, undoubtedly is considerably greater than that for the Alamogordo detonation alone, in view of the fact that many bombs have been fired. Considerations, similar to those above, show that it would probably be relatively easy to measure Sr^{90} in marine sediments, but somewhat more difficult to measure it in sea water itself.

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APPENDIX I

HAZARD CALCULATIONS[*]

PROJECT GABRIEL

The following is a summary of the assumptions of the earlier GABRIEL studies.

GABRIEL: November 12, 1949

The number, N, of nominal (20-KT) bombs required to reach threshold lethality over a uniform "settling area" is given by

$$N = \frac{2D}{Gad} (\times \text{ or } \div) 8,$$

D = "threshold lethality" in μg,
G = plant uptake and ingestion factor in grams per person per bomb,
a = accumulation (and absorption) factor,
d = bone-deposition factor.

It is believed that the basis for the factors 2 and 8 is to allow the answer to be too small by a factor of 16, or too great by a factor of 4 (an assumption that is not too clear in the text).

The various assumed values were:

D = 10 μg (estimated error: factor of 4),
$G = SH/AF$, where S = μg yield per bomb,
H = proportion of strontium in the soil that goes to human consumption per year (or per crop?),
A = area of spread of debris,
F = average number of people who derive their food per square mile of arable land.

The earlier GABRIEL reports made the following assumptions:

1. S was assumed to equal 55 gm of Sr^{90} per nominal (20-KT) bomb. (Actually the yield is about 20 gm.)
2. H was assumed to be 10^{-3} on the basis that edible plants take up 1 per

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[*] For corrections to some of these data, refer to the Preface of this report.

cent of the strontium in the root volume of soil and, on the average, 10 per cent of the plant is consumed.

3. *A* may represent a local or worldwide area, depending on the assumption of extent of uniform spread of debris.
4. *F* was assumed to be 200 people supported per square mile of arable land.

Other biological factors are *a* and *d*; *a* was assumed to range from 0.05 for individuals having low-calcium metabolism to 0.60 for those having high-calcium metabolism, and *d* was assumed to be 0.70.

GABRIEL: November 8, 1951, and December 3, 1951

The later GABRIEL calculations omitted considerations of oral absorption and bone deposition by calculating on the basis of

$$N = \frac{D}{S/A \times H/F} = \frac{D}{G}.$$

Note also the absence of the "2(× or ÷)8" uncertainty factor. The error assumption here was that the estimation "may be 100 too low—or approximately 10 times too high." The assumed values of the parameters were the same as those in the earlier GABRIEL study.

Using GABRIEL assumptions (but not values), it may be interesting to derive the megaton limits (using the proper conversion factor from nominal bombs) for a worldwide uniform distribution, unrealistic a figure as it may be. These are given in Table 5. *We have corrected S/A = 0.1 μg/mi²/nominal bomb.*

Table 5

GABRIEL THRESHOLD LETHALITY

(In terms of megaton yields)

Range: 4×10^4 MT to 1.8×10^8 MT

	Megaton Limits for GABRIEL Threshold Lethality (10 μg)	
	Larger than	Less than
GABRIEL, 1949, low-calcium metabolism	2.9×10^6	1.8×10^8
GABRIEL, 1949, high-calcium metabolism	2.4×10^5	1.5×10^7
GABRIEL, 1951	4×10^4	4×10^7

For standard MPC (1 μc), the values given in Table 5 are to be reduced by a factor of $\frac{1}{2000}$, which gives a range of 20 MT to 9×10^4 MT for a worldwide MPC on GABRIEL assumptions and results in an effective spread factor of 4×10^3.

REMARKS ON CALCULATION OF WORLDWIDE CONTAMINATION*

"1. Important progress has been made by the RAND GABRIEL on the basis of the calculations. This new basis was reported by Dr. Mitchell. It consists in considering the ratio of Sr^{90} to total Sr or total Ca in human bone and in the biosphere. This type of consideration is likely to give much more reliable and much more precise values for the tolerance contamination than the previous basis in which it was assumed that a certain arbitrarily chosen fraction of the Sr^{90} would find its way into human bodies. The exact assumptions about the ratio of Sr^{90} to the inert substances matter less than the adoption of the general principle of Dr. Mitchell.

"2. The calculation which I presented on the last day of the conference was predicated on a number of assumptions, four of which are definitely pessimistic. These are

"(*a*) All the Sr^{90} which is produced will fall out before appreciable radioactive decay.

"(*b*) All the Sr^{90} will be in an exchangeable form and will remain exchangeable in the soil for all its radioactive lifetime.

"(*c*) The ratio of Sr to Ca in the human bone is the same as in the soil from which the food is derived.

"(*d*) The tolerance dose for Sr^{90} is one microcurie distributed uniformly through the bone.

"In addition to these, two assumptions were made of which I do not know whether they are optimistic or pessimistic, namely,

"(*e*) The amount of exchangeable calcium in soil is one part per thousand by weight.

"(*f*) The amount of Sr^{90} taken up is related to the amount of calcium taken up independently of the ratio of inert Sr to Ca.

"In addition, an assumption is made which is probably optimistic, namely,

"(*g*) The redistribution of Sr^{90} by fertilizer, etc., does not change the ratio of Sr^{90} to Ca in the soil from which human food is derived.

3. With the assumptions made under 2, the tolerance dose is reached with a total fission yield of about 1000 megatons.

"4. Assumption (*a*) under paragraph 2 is clearly pessimistic. It would seem

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*The following remarks are the substance of a memorandum from Hans A. Bethe, Cornell University, dated June 24, 1953.

both simple and possible to calculate from the available data of Eisenbud what fraction of the Sr^{90} from MIKE will have fallen out after 30 years if the present rate of fallout continues unchanged. This would still seem to be a pessimistic assumption, and it is likely to give a number considerably less than 100 per cent. For low-yield bombs, the 100 per cent assumption is probably not unduly pessimistic.

"5. Concerning assumption (*b*), experiments such as that of Dr. Larson, but conducted with Sr^{90} rather than Sr^{89}, are clearly desirable. General opinion seems to be that Sr^{90} would not remain exchangeable even if it is so originally.

"6. Concerning (*c*), it seemed to me that we should disregard the old data indicating about 30 parts of Sr per 1000 parts of Ca in soil. But that we might reasonably use the new data which give about three atoms of Sr per 1000 of Ca. This would still indicate an advantage factor of 10 for the incorporation of Sr into the bone as compared to Ca, and thus would increase the allowable fission yield by a factor of 10.

"7. Concerning (*d*), I believe that the international tolerance of one microcurie of Sr^{90} is really inconsistent both with the accepted tolerance of Ra and with that of uniform body radiation of gamma-rays. I believe that the accepted principle is correct according to which neutrons are much more dangerous than gamma-rays per unit energy. If this is true, then also beta-rays must be much less dangerous than alpha-rays. I further believe that it is really correct to make a direct comparison of beta-rays with gamma-rays provided the beta-emitter is really uniformly distributed over the bone. With these assumptions, one gets a tolerance dose of 10 microcuries of Sr^{90} when distributed uniformly over the bone. This follows both from the accepted Ra tolerance and from the accepted gamma-ray tolerance. Of course, this number is entirely consistent with an assumed one microcurie if the latter is concentrated near the more sensitive parts of the bone, as it would probably be for an adult who has been exposed to Sr^{90} only for a few months or years. However, in this case the average bone of this adult will contain a lower concentration of Sr^{90} compared to total Ca than the food which the person has consumed recently. This factor of uncertainty is eliminated when we talk about the level of Sr^{90} in the food and in those parts of the bone which have been recently laid down. It seems to me entirely reasonable and safe to accept the 10 microcurie tolerance in connection with the calculations of GABRIEL.

"8. Concerning (*e*), nothing but renewed investigation of the chemical composition of soils can help. If there is less Ca, the allowable number of megatons will be reduced proportionally. Concerning (*f*), Larson's experiments seem to indicate just what I assume.

"9. If the ratio of Sr^{90} to Ca in animal bones, etc., is equal to that of the soil from which the food is derived, or less, as it seems to be, then the application of bone meal fertilizer will not change the ratio unfavorably.

"10. I think it is very essential to go through with the investigation proposed by you, namely, to assay the Sr^{90} content in bones of various animals including humans, and particularly in sewage and feces, at the present time to get more reliable data."

CALCULATIONS ON ASSUMPTIONS OF H. A. BETHE

The basic assumption is

$$\left(\frac{\mathrm{Sr}}{\mathrm{Ca}}\right)_{\mathrm{body}} = \left(\frac{\mathrm{Sr}}{\mathrm{Ca}}\right)_{\mathrm{soil}},$$

or

$$\left(\frac{\mathrm{Sr}^{90}}{\mathrm{Ca}}\right)_{\mathrm{body}} = \left(\frac{\mathrm{Sr}^{90}}{\mathrm{Ca}}\right)_{\mathrm{soil}},$$

or

$$(\mathrm{Sr}^{90})_{\mathrm{soil}} = (\mathrm{Ca})_{\mathrm{soil}}\left(\frac{\mathrm{Sr}^{90}}{\mathrm{Ca}}\right)_{\mathrm{body}}.$$

With 1 μc of Sr^{90} fixed in the bone, we have

$$\left(\frac{\mathrm{Sr}^{90}}{\mathrm{Ca}}\right)_{\mathrm{body}} = \frac{5 \times 10^{-3}}{10^{9}} = 5 \times 10^{-12},$$

assuming that 1 cm^2 of topsoil weighs $\overset{3.0}{30}$ gm, giving a content of $\overset{3.0}{30}$ mg of calcium, and assuming 2(*e*) in the previous section.

The required amount of Sr^{90} in soil to bring that soil to tolerance capability is 1.5×10^{-13} gm Sr^{90}/cm^2 = 4 mg Sr^{90}/mi^2. The area of the earth being 2×10^8 mi^2, the total requirement for tolerance is then 800 kg of Sr^{90}, which represents a fission energy of **800 MT.** ?

PROJECT SUNSHINE FORMULA

The energy release in megatons from fission bombs that would be required to bring the population of the earth to any given level *T* of Sr^{90} deposited in the skeleton, if the individuals grew from birth to maturity in an environment contaminated uniformly with the Sr^{90} produced, can be expressed by the following. (No decay of Sr^{90} is assumed and the Sr^{90} is assumed to be 100 per cent available for incorporation in the biological sequence from soil to man.)

Let

MT = the number of megatons of fission energy released,

W_{sr} = the number of grams of available natural strontium per square mile of area,
ω_B = the number of grams of natural strontium fixed in the human skeleton at maturity,
T = the number of grams of Sr^{90} fixed in the skeleton that is considered to be the level of interest (MPC or any other standard),
m = the number of grams of Sr^{90} produced by the release of 1 MT of fission energy,
A = the area of the earth in square miles,

then

$$\mathrm{MT} = \frac{1}{m}\left(\frac{T}{\omega_B}\right) \times W_{sr} \times A.$$

Since there is little chance that m will be changed appreciably by future measurements and A is fixed, this relation can be simplified as follows (taking $m = 1000$ gm and $A = 3 \times 10^8$ mi^2):

$$\mathrm{MT} = 2 \times 10^5 \left(\frac{T}{\omega_B}\right) \times W_{sr}.$$

Taking T to be 1 μc (the international MPC), $\omega_B = 0.7$ gm, and $W_{sr} = 1.7 \times 10^7$ gm, one finds (as in Chapter 4) MT to be 2.5×10^4 on an idealized worldwide basis. The validity of the assumption underlying this calculation is discussed in Chapter 1.

1.
bone
"10. .

APPENDIX II

PARTICLE SIZE OF DEBRIS FROM THE ATOMIC BOMB*

A theoretical consideration that may apply to the condensation process of the solid material in the fireball of an atomic bomb is presented here. The well-established nucleation theory of Becker and Döring, and others,(1) is used. Only a treatment of the condensation by self-nucleation of a homogeneous gaseous system cooled at a uniform rate is treated here. Application to the atomic bomb involves complications that have not as yet been considered in detail. However, a rough check on the magnitudes of the physical quantities seems to indicate that the mechanism is reasonable.

The slow process in the condensation is assumed to be the formation of the solid oxides from gaseous oxide molecules. The gaseous oxide molecules should form on a much shorter time scale and at much higher temperatures than those involved in the condensation.

CONDENSATION MODEL: SIMPLE ANALYTICAL FORM

In a homogeneous supersaturated gas at constant temperature, nuclei are forming at a constant rate. Once formed, a nucleus grows by condensing single molecules on its surface. The growth rate of a particle having radius R is given by

$$\frac{d}{dt}\left(\frac{4\pi R^3}{3}\right) = 4\pi R^2 F v_0, \tag{1}$$

where F is the flux of single molecules† and v_0 is the volume of a single molecule. This equation simplifies to

$$\frac{dR}{dt} = F v_0 = \beta, \tag{2}$$

where β is defined by the equation.

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[*] Communicated by John L. Magee, University of Notre Dame.

[†] An accommodation coefficient should be used, too, since all molecules that strike the surface may not remain.

The radius of any particle increases linearly with time:

$$R(t, t') = \beta(t - t'), \tag{3}$$

where t' is the time the nucleus was formed. The total volume of precipitate present at the time t is

$$V(t) = \int_0^t \frac{4\pi}{3} R^3(t, t') I(t, t')\, dt', \tag{4}$$

where $I(t')\, dt'$ is the number of nuclei that were formed in the time interval dt'.

If a gas is chilled at a constant rate, the rate of nucleation also increases with time. We shall see below that a reasonable first approximation is

$$I(t') = I_0 e^{\alpha t} \qquad (-\infty \leq t' \leq t), \tag{5}$$

where I_0 is the nucleation rate at a time that is set equal to zero, and α is a constant.

Substitution of Eqs. (3) and (5) into (4) allows an analytical integration (from $t' = -\infty$ to $t' = t$):

$$V(t) = \frac{4\pi}{3} \frac{\beta^3}{\alpha^4} I_0 (6e^{\alpha t}). \tag{6}$$

In this integration, β was taken to be a constant, which means that this formula can only be used when there is no serious depletion of gaseous single molecules of the condensate. At such a time the particle-size distribution has a particularly simple form. The number of particles having a value of R in the interval R to $R + dR$ were those formed at the time $t - R/\beta$ to $(t - R/\beta) + dR/\beta$. Thus

$$N(R)\, dR = I\left(t - \frac{R}{\beta}\right)\frac{dR}{\beta} = I_0 \exp\left(\alpha t - \frac{\alpha}{\beta} R\right)\frac{dR}{\beta}, \tag{7}$$

or, more simply,

$$N(R) \propto \exp\left[-(\alpha/\beta)R\right]. \tag{8}$$

The big parti- -fore, have the distribution given by (8). Since the rate of nu- -al in time, it can be argued that the entire distribution same function. The development of a distribution func- depletion has not been considered as yet.

FREE ENERGY OF

bon- -ster of x condensable molecules with respect to the "10. i.e.,

$$W(x) = (\mu_{\text{solid}} - \mu_{\text{gas}})x + \text{surface free energy}, \tag{9}$$

where $\mu_{\text{solid}} - \mu_{\text{gas}}$ is the difference in free energy for one molecule between a large solid and the gaseous state in question. If we can approximate the vapor pressure as a function of temperature* as

$$p = \text{const} \exp\left[-(\Delta H/RT)\right], \tag{10}$$

it can be shown that

$$\frac{1}{RT}(\mu_{\text{solid}} - \mu_{\text{gas}}) = \frac{\Delta H}{R}\left(\frac{1}{T_s} - \frac{1}{T}\right), \tag{11}$$

where ΔH is the heat of vaporization and T_s is the saturation temperature for the system; R is the gas constant in this formula (it was used as particle radius above).

The work required to form a cluster of x molecules is divided into two terms. The first term,

$$\frac{\Delta H}{R}\left(\frac{1}{T_s} - \frac{1}{T}\right)x,$$

is negative for all temperatures below the saturation temperature and varies directly as the cluster size; the second term,

$$\text{surface free energy} \propto x^{2/3},$$

is always positive and varies as the surface area of the cluster. For increasing surface area, the sum of these terms goes through a maximum. In the condensation theory it is assumed that all clusters having positive free energy and that are smaller than the cluster of maximum free energy are in equilibrium. It can be shown that

$$N(x) = N(1) \exp\left[-\frac{W(x)}{RT}\right], \tag{12}$$

where $N(x)$ is the number of clusters of size (x) per unit volume and $N(1)$ is the number of uncombined condensable molecules per unit volume.

The maximum value of the function

$$\frac{W(x)}{RT} = \frac{\Delta H}{R}\left(\frac{1}{T_s} - \frac{1}{T}\right)x + Cx^{2/3} = -Bx + Cx^{2/3} \tag{13}$$

occurs at

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*The vaporization does not have to be a simple process, but may be a decomposition into a variety of products. For example, Al_2O_3 yields the gaseous products AlO, Al_2O, and O_2. This method is also applicable in such cases.

$$x^* = \left(\frac{2C}{3B}\right)^3. \tag{14}$$

Here B and C are constants.

The concentration of clusters of this size is

$$N(x^*) = N(1)\exp\left(-\frac{4}{27}\frac{C^3}{B^2}\right). \tag{15}$$

The rate of nucleation (see below) is directly proportional to this quantity.

The constant B is given as $-(\Delta H/R)(1/T_s - 1/T)$, of course. The value of C is not so easy to obtain, but we can estimate its magnitude by several methods. If surface-tension data are known, then

$$\text{surface free energy} = 4\pi\sigma\left(\frac{3v_0}{4\pi}\right)^{2/3} x^{2/3}, \tag{16}$$

where σ is the surface tension and v_0 is the volume of a single molecule of the cluster.

Another method for the estimation of C may be helpful. Consider a small cluster having a simple symmetry. A convenient number of molecules to take is thirteen in hexagonal close packing. Thirteen molecules make a cluster with one central molecule surrounded by its twelve nearest neighbors. This collection of molecules has 12 bonds to the central molecule and 5 each to the others. The total number of bonds is $\frac{1}{2}(12 + 12 \times 5) = 36$ bonds. In the interior of a large solid, these thirteen molecules would have $\frac{1}{2} \times 13 \times 12 = 78$ bonds. The missing heat of binding, therefore, is approximately $(42/78)\sigma H$ per molecule. If we set

$$Cx^{2/3} = C(13^{2/3}) - \left(\frac{42}{78}\right)13\,\frac{\Delta H}{RT},$$

$$C = 1.25\,\frac{\Delta H}{RT}. \tag{17}$$

In this consideration we have neglected entropy considerations and so C is actually smaller, by a factor that may be as small as one-half. However, the magnitude must be

$$C \approx \frac{\Delta H}{RT}. \tag{17a}$$

Use of this value for C gives for Eq. (15):

$$N(x^*) = N(1)\exp\left[-\frac{4}{27}\left(\frac{T_s}{T - T_s}\right)^2 \frac{\Delta H}{RT}\right]. \tag{18}$$

NUCLEATION RATE

Clusters that grow beyond the size x^* have free energy that decreases with the addition of single molecules. The rate at which nuclei are formed is the rate at which clusters with $x > x^*$ are formed. This rate is(2)

$$I = FS\sqrt{\frac{C}{9}}\,N(x^*). \tag{19}$$

Here S is the area of a cluster divided by $x^{2/3}$, i.e., the area of a cluster is given by $Sx^{2/3}$; F is the flux of single molecules as used above.

Using (18), we have

$$I = FS\sqrt{\frac{\Delta H}{9RT}}\,N(1)\exp\left[-\frac{4}{27}\left(\frac{T_s}{T - T_s}\right)^2\frac{\Delta H}{RT}\right]. \tag{20}$$

If the temperature is changing linearly with time, say, $T_s - T = at$, then

$$I(t) = A\exp\left[-\frac{4}{27}\left(\frac{T_s^2}{a^2t^2}\right)\frac{\Delta H}{RT_s}\,\frac{1}{(1 - at/T_s)}\right] \approx A\exp\left(-\frac{\tau^2}{t^2}\right), \tag{21}$$

where A and τ are constants defined by the equation. In the last expression, at/T_s has been neglected.

If we assume that the precipitation takes place rather rapidly after a time t_0 has elapsed and define

$$t' = t - t_0,$$

we can write for the nucleation rate

$$I(t') = A\exp\left[-\frac{\tau^2}{(t_0 + t')^2}\right] \approx A\exp\left[-\frac{\tau^2}{t_0^2}\left(1 - \frac{2t'}{t_0}\right)\right], \tag{22}$$

which is the simple exponential approximation we proposed earlier, i.e.,

$$I(t') \propto e^{\alpha t'},$$

and we see that

$$\alpha = 2\,\frac{\tau^2}{t_0^3} = \frac{4}{27}\,\frac{T_s^2}{a^2t_0^2}\,\frac{\Delta H}{RT_s}\,\frac{1}{t_0}. \tag{23}$$

or, with substitution,

$$at_0 = T_s - T_0, \tag{24}$$

where T_0 is the temperature of condensation, and

$$\alpha = \frac{4}{27}\left(\frac{T_s}{T_s - T_0}\right)^2 \frac{a}{T_s - T_0}\,\frac{\Delta H}{KT_s}. \tag{25}$$

Here we note that one can use Eq. (21) to get the particle-size distribution instead of using Eq. (5). If this is done, Eq. (8) has a slightly more complicated form.

ESTIMATION OF CONSTANTS

The purpose of this development is the correlation of the properties of the precipitate with known properties of the oxides (or other molecules) that condense to form the precipitate. Because of lack of time and data, essentially no start has as yet been made in this direction. In this section we shall examine a few magnitudes to show that a consistent description seems to be within the realm of possibility.

Examination of particle-size data suggests a particle-size distribution for particles in the range from 2 to 10 μ to be

$$N(R) \propto \exp\left[-(\alpha/\beta)R\right],$$

with a value of the constant $\alpha/\beta \approx 10^4$ cm^{-1}. We shall estimate the magnitude to be expected for α/β from physical consideration.

The magnitude of β is easy to obtain, e.g.,

$$\beta = Fv_0 \approx \frac{1}{4} \times 10^{15} \times 10^5 \times 2 \times 10^{-23} = 5 \times 10^{-4}.$$

Here we have assumed that there are 10^{15} oxide molecules (say, AlO) per cubic centimeter at the time of condensation; their velocity is 10^5 cm/sec; the volume each adds to a particle is 2×10^{-23} cm^3.

The magnitude of α is somewhat more difficult to obtain because of the arbitrariness of the quantity t_0, i.e., the time that is set as zero in Eq. (5). Clearly this time must be near to that at which the process is complete. The total volume of precipitate will finally be $\approx 2 \times 10^{-8}$ cm^3. If we use Eq. (6),

$$V(t_0) = 2 \times 10^{-8} = \frac{4\pi}{3}\frac{\beta^3}{\alpha^4}\left[6I(t_0)\right], \tag{26}$$

$$I(t_0) = 6.4\alpha^4, \tag{27}$$

in which $\beta = 5 \times 10^{-4}$ has been used.

With the use of Eq. (20) we find that if

$$I(t_0) \approx \frac{1}{4} \times 10^{15} \times 10^5 \times 4 \times 10^{-15} \sqrt{\frac{\Delta H}{9RT}}\, 10^{15} \exp\left[-\frac{4}{27}\left(\frac{T_s}{T_s - T_0}\right)^2 \frac{\Delta H}{RT_s}\right],$$

where $S = 4\pi(3v_0/4\pi)^{2/3} = 4 \times 10^{-15}$ has been used, then

$$I(t)_0 \approx 10^{20} \exp\left[-\frac{4}{27}\left(\frac{T_s}{T_s - T_0}\right)^2 \frac{\Delta H}{RT_s}\right]. \tag{28}$$

The heat of vaporization to be taken should be about 50,000 cal/mole. (This would give more than 100,000 cal/mole for Al_2O_3, since the unit we are taking is only part of a molecule.) Thus $(\Delta H/9RT)^{1/2} \approx 1$ for $T \approx 2500$.

Substitution of α from Eq. (25) into Eqs. (27) and (28) gives

$$I(t_0) = 10^{20} \exp\left[-\left(\frac{4}{27}\frac{\Delta H}{RT_s}\right)^{1/3}\left(\frac{T_s}{a}\right)^{2/3} \alpha^{2/3}\right] = 6.4\alpha^4. \tag{29}$$

If we take $\Delta H/RT_s = 10$, $T_s = 2500$, $a = 500$, the solution is $\alpha = 25$.

This pair of solutions gives $\alpha/\beta = 5 \times 10^4$, which is somewhat larger than the probable value, although about the correct magnitude.

It is certain that for values of all the physical constants that would seem reasonable, the value 10^4 for α/β can be obtained. One of the most uncertain quantities of this calculation is the value of a, the cooling rate in degrees per second. If instead of 500 we take 100, the corresponding value of α is 7 and

$$\frac{\alpha}{\beta} \approx \frac{7}{5} \times 10^4 \approx 10^4.$$

This calculation suggests that the extreme diminution in the cooling rate, as the radiative cooling becomes ineffective at about 2000°K, may be important in the precipitation process.

As a final check on the consistency of this last calculation, we obtain the value of x^* at $t = t_0$. Using Eqs. (14) and (25), we can write

$$x^* = 2\alpha\left(\frac{T_s}{a}\right)\left(\frac{RT_s}{\Delta H}\right). \tag{30}$$

For $\alpha = 7$, $T_s/a = 25$, and $RT_s/\Delta H = 1/10$,

$$x^* = 35,$$

which shows that the nucleation mechanism would still be operating. For sufficiently low temperatures, x^* approaches unity and nucleation is no longer the slow process.

DISCUSSION

The mechanism proposed here for condensation by means of self-nucleation of oxide molecules appears to be promising enough to warrant further investigation. If such a mechanism can be established as being responsible for the particle formation, it will yield valuable information regarding scaling of particle sizes with bomb yield and type of shot (high air burst, tower, etc.), the form in which fission products are likely to be found, fractionation, and other such problems.

For a more careful study, thermodynamic data of the oxides will be necessary as well as time, temperature, and volume data on fireballs.

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APPENDIX III

SETTLING OF PARTICLES IN A STANDARD ATMOSPHERE*

In this appendix known formulas for viscous drag are applied to the problem of the settling of particles in the standard atmosphere. The atmosphere will be taken as stationary, and so convection effects are not considered.

The results of fluid mechanics can be applied without modification to the motion of a solid particle through the air provided the particle dimensions are much greater than the mean free path of the air molecules. In this case, the type of flow is determined by the Reynolds number, R, which is defined to be

$$R = \frac{u\rho d}{\sigma}, \tag{1}$$

where u is the velocity of the particle, d is a characteristic dimension of the particle, and ρ and σ are the air density and viscosity, respectively. For a spherical particle of diameter d, we have Stokes' flow with a drag force given by

$$F_D = 3\pi\sigma u d, \tag{2}$$

provided the Reynolds number does not exceed approximately 0.5. For larger Reynolds numbers, Eq. (2) underestimates the drag force, but the error is less than 20 per cent at $R = 1$; Stokes' law will be used in the present calculations up to $R = 1$.

The terminal velocity of fall, u, of a spherical particle of density ρ' is obtained by equating its weight to the drag force of Eq. (2); the result is

$$u = \frac{g(\rho' - \rho)d^2}{18\sigma},$$

$$u \approx \frac{g\rho' d^2}{18\sigma}, \tag{3}$$

where g is the acceleration of gravity, and the approximation follows from the fact that $\rho < < \rho'$.

Observations have been made on the Stokes' fall of particles with nonspherical shape.(1) An ellipsoidal shape gives a reasonable approximation for the drag force

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*Written by F. R. Gilmore and M. S. Plesset, California Institute of Technology.

for a variety of nonspherical particles. The ratio of the terminal velocity of fall for ellipsoids to the terminal velocity of spheres of the same volume is presented in one of the graphs to follow. These curves(1) have been computed from the theoretical formulas of Oberbeck,(2) which agree with the limited number of experimental measurements available. In general, particles of irregular shape have smaller terminal velocities than spheres of the same volume.

Equations (2) and (3) are not applicable to the motion of spheres in a gas if the diameter of the spherical particle is not considerably greater than the mean free path, λ, of the air molecules. For $d/\lambda = 10$, the terminal velocity as predicted by Stokes' law is low by about 15 per cent, with the error increasing with decreasing d/λ. A rough theoretical analysis of the drag for the range $2 \lesssim d/\lambda \lesssim 100$ was made by Cunningham;(3) a more complete theory was developed by Epstein.(4) This range of flow is frequently referred to as the Stokes-Cunningham range. A study of the flow has also been made for $d/\lambda << 1$, first by Cunningham and subsequently by several other investigators. The definitive analysis for this range is due again to Epstein.(4) Epstein's formulas for the two ranges, $d/\lambda << 1$ and $2 \lesssim d/\lambda \lesssim 100$, depend on the ratio of the specular to the diffuse reflection in the collisions between the particles and the air molecules. This ratio is not measured directly, but is chosen to fit observed drag values on small spheres settling in gases. For the intermediate range, $d \sim \lambda$, no theoretical formula is available. Knudsen and Weber(5) have proposed a general empirical formula that fits the data not only in this range, but also in the ranges $d < \lambda$ and $d > \lambda$. In the latter two ranges, the empirical formula agrees with the theoretical expressions. The empirical formula,(6) as used here, has constants adjusted to fit Millikan's oil-drop data, and gives the following expression for the terminal velocity in air:

$$u = \frac{g\rho' d^2}{18\sigma}\left[1 + (1.644 + 0.552 e^{-0.656 d/\lambda})\lambda/d\right]. \qquad (4)$$

Equation (4) also reduces to Eq. (3) for $d/\lambda >> 1$. The computations made here are based on Eq. (4).

Figures 7 through 10 give the results of the computations that have been prepared for this report. These figures are based on spherical particles of density, $\rho' = 2$ gm/cm^3, and on the NACA standard atmosphere.(7)

Figure 7 depicts the boundaries of the various flow regimes for different altitudes and different sphere diameters. These curves are determined by the value of the ratio d/λ except for the upper limit to the Stokes' range, which is determined by the Reynolds number ($R = 1$).

Figure 8 gives the terminal velocity as a function of particle diameter at various altitudes. Since the terminal velocity is proportional to the particle density ρ', the values on the graphs may be readily adjusted to other values of ρ'.

Figure 9 shows the times required for spherical particles to fall from a given initial altitude to sea level as a function of particle diameter.

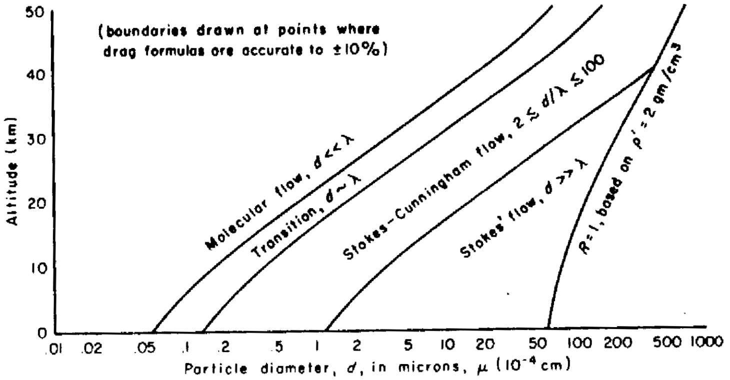

Fig. 7—Various flow regimes for the free fall of spherical particles as a function of diameter and altitude

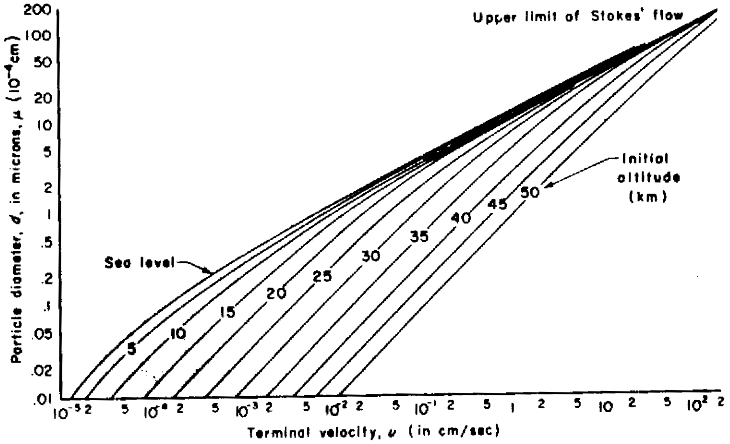

Fig. 8—Terminal velocity of spherical particles of density $\rho' = 2$ gm/cm^3, as a function of diameter, at various altitudes

Figures 8 and 9 neglect the Brownian motion component for small particles. The effect of this neglect is unimportant for the long settling times and large distances of present interest, since the root-mean-square Brownian displacement varies as the square root of time and the gravity drift displacement varies linearly with time.

Figure 10 indicates the effect of particle shape on settling velocity; it gives the

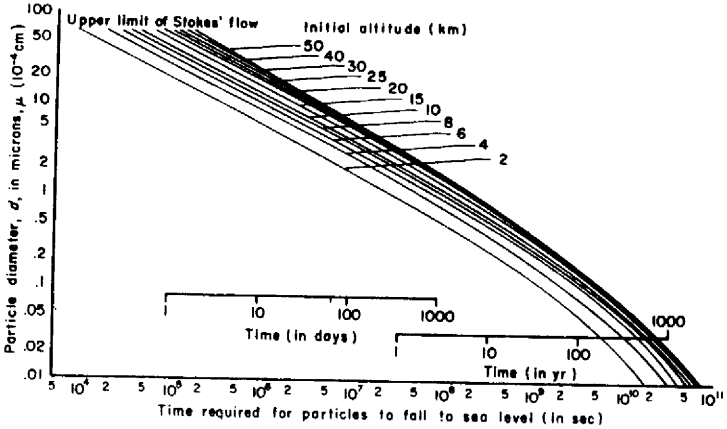

Fig. 9—Time required for spherical particles of density $\rho' = 2$ gm/cm^3 to fall from a given initial altitude to sea level as a function of diameter

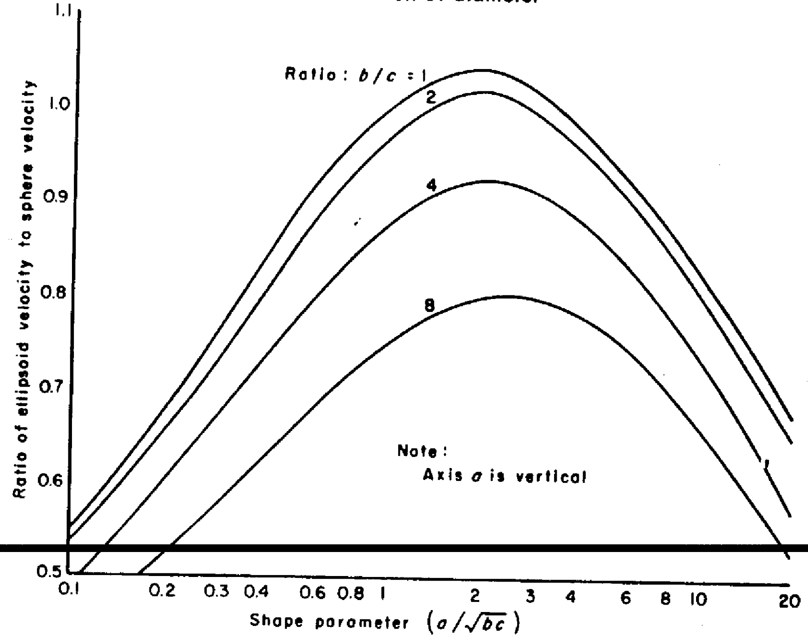

Fig. 10—Ratio of terminal velocity of ellipsoid with axes *a*, *b*, and *c* to terminal velocity of sphere of same volume

ratio of the terminal velocity of fall for ellipsoids to the terminal velocity of a sphere of the same volume. The values given are accurate only in the Stokes' range of flow but may be used as an approximation even when $d/\lambda \lesssim 10$.

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APPENDIX IV

INHALATION OF STRONTIUM 90

Aluminum oxide has been suggested as a possible trap for Sr^{90} during formation of particles in an atomic explosion. Consideration is therefore being given to the effect of this type of conglomerate material upon inhalation into the lung.

Aluminum oxide has been implicated as the causative agent of a pneumoconiotic disease (Shaver's Disease) among workers inhaling fumes of calcined bauxite. The microscopic pathological picture has been described as a diffuse fibrotic process. The alveolar walls were thickened and in places the alveoli were replaced by fibrous tissue. Hyalinization was considerable. The fibrosis was not nodular in type.

This mechanism may result in somewhat greater trapping than the conventional rule of thumb regarding the retention of particulate matter in the lungs.* However, for SUNSHINE we shall use the standard MPC of 2×10^{-10} $\mu c/ml$ of air.

It may be of some interest to calculate the inhalation-limiting world dose of Sr^{90}. This calculation is even more idealized than those given in Appendix I, especially since it assumes that all the Sr^{90} created is available in aerosol form uniformly distributed throughout the earth's atmosphere.

We take as the volume of the earth's atmosphere, 5×10^{24} cc. The required amount of Sr^{90} aerosol to bring the atmosphere up to tolerance is then $(2 \times 10^{-10})(5 \times 10^{24}) = 10^{15}\ \mu c = 10^{9}$ curies. This activity of Sr^{90} is created by a fission energy equivalent to 5000 MT.

Pending better evaluation of the inhalation hazard, this figure is to be taken *cum grano salis.*

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*International Commission on Radiological Protection; NBS Handbook 47.

APPENDIX V

PHYSICAL CONSTANTS*

FISSION ENERGY

For the purposes of SUNSHINE, we shall use the standard Los Alamos radiochemical scale.

1. U^{235} fission: 1.42×10^{23} fission gives 1 KT TNT equivalent (1 KTE), or 1 kg $U^{235} \rightarrow 18$ KTE.
2. Pu^{239} fission: 1.35×10^{23} fission gives 1 KTE or 1 kg $Pu^{239} \rightarrow 18.7$ KTE.

STRONTIUM 90 AND YTTRIUM 90

$$T_{1/2}(\mathrm{Sr}^{90}) = 19.9 \pm 0.3 \text{ year,}^{(1)}$$

$$E_{\beta}(\mathrm{Sr}^{90}) = 0.54 \text{ Mev,}^{(2)}$$

$$T_{1/2}(\mathrm{Y}^{90}) = 61 \text{ hr,}^{(3)}$$

$$E_{\beta}(\mathrm{Y}^{90}) = 2.24 \text{ Mev.}^{(2)}$$

Most, if not all, of the Sr^{90} originates in this manner in fission. The yield of Sr^{90} in thermal fission of U^{235} is

$$4.62,\ +15\%,\ -6\%.^{\dagger}$$

Thus 1 KTE energy release of U^{235} results in the production of

$$\frac{(4.62)}{(100)}\frac{(1.42 \times 10^{23})(90)}{(6.02 \times 10^{23})} = 0.98 \text{ gm Sr}^{90}.$$

Now,

$$\lambda(\mathrm{Sr}^{90}) = \frac{0.693}{19.9(3.16 \times 10^{7})} = 1.10 \times 10^{-9} \text{ sec}^{-1},$$

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*For corrections to some of these data refer to the Preface of this report.

†Private communication from Anthony Turkevich, Institute for Nuclear Studies, University of Chicago, July 17, 1953.

or 1 gm of Sr^{90} has a strength of

$$\frac{(1.10 \times 10^{-9})}{3.7 \times 10^{10})} \frac{(6.02 \times 10^{23})}{90} = 199 \text{ curies}.$$

Since the yields of Sr^{90} for various bomb neutron spectra are unknown, and for the time being no distinction between uranium and plutonium weapons is being made, we shall adopt the following yield standard for Sr^{90}:

One KT *energy equivalent yields* 1 gm = 200 curies *of* Sr^{90}. *All* Sr^{90} *yields are to be scaled proportionately to the* KTE *of a particular bomb.*

This figure is probably the least inaccurate of all the parameters involved in the SUNSHINE study. It is to be noted that this value is several times smaller than the value used in the early GABRIEL studies: 55 gm/20 KT or 2.75 gm/KT. (The basis for this figure was not given.)

GLOBAL CONSTANTS

1. *Oceans and seas*:

$$\text{Area} = 3.6 \times 10^8 \text{ km}^2 = 1.4 \times 10^8 \text{ mi}^2,$$
$$\text{Volume} = 1.4 \times 10^9 \text{ km}^3 = 3.3 \times 10^8 \text{ mi}^3,$$
$$\text{Mean Depth} = 3.8 \text{ km} = 2.4 \text{ mi}.$$

2. *Land*:

$$\text{Area} = 1.5 \times 10^8 \text{ km}^2 = 0.6 \times 10^8 \text{ mi}^2.$$

BIOMEDICAL CONSTANTS: THE STANDARD MAN

Mass of Organs

Organs	Grams
Muscles	30,000
Skeleton, bones	7,000
Red marrow	1,500
Yellow marrow	1,500
Blood	5,000
Gastrointestinal tract	2,000
Lungs	1,000
Liver	1,700
Kidney	300
Spleen	150
Pancreas	70
Thyroid	20

Mass of Organs—continued

Organs	Grams
Testes	40
Heart	300
Lymphoid Tissues	700
Brain	1,500
Spinal cord	30
Bladder	150
Salivary glands	50
Eyes	30
Teeth	20
Prostate	20
Adrenals	20
Thymus	10
Skin and subcutaneous tissues	8,500
Other tissues and organs not separately defined	8,390
TOTAL BODY WEIGHT	70,000

Chemical Composition

Element	Proportion (%)	Approximate Mass in the Body (in gm)
Oxygen	65.0	45,500
Carbon	18.0	12,600
Hydrogen	10.0	7,000
Nitrogen	3.0	2,100
Calcium	1.5	1,050
Phosphorus	1.0	700
Potassium	0.35	245
Sulphur	0.25	175
Sodium	0.15	105
Chlorine	0.15	105
Magnesium	0.05	35
Iron	0.004	3
Manganese	0.0003	0.2
Copper	0.0002	0.1
Iodine	0.00004	0.03

The figures for a given organ may differ considerably from these averages for the whole body. For example, the nitrogen content of the dividing cells of the basal layer of skin is probably nearer 6 per cent than 3 per cent.

Applied Physiology

The average data for normal activity in a temperate zone are given below.

1. *Water balance*:

DAILY WATER INTAKE

(In liters)

In food, including water of oxidation	1.0
As fluids	1.5
TOTAL	2.5

Calculations of maximum permissible levels for radioactive isotopes in water have been based on the total intake figure of 2.5 liters per day.

DAILY WATER OUTPUT

(In liters)

Sweat	0.5
From lungs	0.1
In feces	0.1
Urine	1.5
TOTAL	2.5
TOTAL WATER CONTENT OF BODY	50

2. *Respiration*:

AREA OF RESPIRATORY TRACT

(In square meters)

Respiratory interchange area	50
Nonrespiratory area (upper tract and trachea to bronchioles)	20
TOTAL	70

RESPIRATORY EXCHANGE

Physical Activity	Hours per Day	Tidal Air (liters)	Respirations per Minute	Volume per 8 Hours (cu m)	Volume per Day (cu m)
At work	8	1.0	20	10	20
Not at work	16	0.5	20	5	

CARBON DIOXIDE CONTENT OF AIR

(By volume)

	Percent
Inhaled air (dry, at sea level)	0.03
Alveolar air	5.5
Exhaled air	4.0

Duration of Exposure

1. *Duration of Occupational Exposure*: The following figures have been adopted in calculations appertaining to occupational exposure:

8 hr/day, 40 hr/week, 50 weeks/year.

2. *Duration of "Lifetime" for Nonoccupational Exposure*: A conventional figure of 70 years has been adopted.

Maximum Permissible Levels for Occupational Exposure to Certain Isotopes

Isotope	Maximum Permissible Amount in Body (μc)	Maximum Permissible Concentration in Air (24-hr day) ($\mu c/cc$)	Maximum Permissible Concentration in Drinking Water ($\mu c/cc$)
H^3	1×10^{-4}	5×10^{-5}	4×10^{-1}
C^{14} (as CO_2) (in air)	....	1×10^{-6}	....
Na^{24}	15	3×10^{-6}	8×10^{-3}
P^{32}	10	1×10^{-7}	2×10^{-4}
S^{35}	100	....	....
I^{131}	0.18 (thyroid) 0.3 (body)	3×10^{-9}	3×10^{-5}
Co^{60}	1	7×10^{-9}	1×10^{-5}
A^{41}	....	1×10^{-6}	....
Xe^{133}	....	1×10^{-5}	....
Xe^{135}	....	3×10^{-6}	....
Ra^{226}	0.1	8×10^{-12}	4×10^{-8}
Pu[illegible]	0.04	2×10^{-2}	1.5×10^{-6}
U^{233} (soluble)	0.4	8×10^{-10}	1.5×10^{-4}
U^{233} (insoluble)	0.008	1.6×10^{-11}	....
$Sr^{90} + Y^{90}$ (in equilibrium)	1.	2×10^{-10}	8×10^{-7}
Sr^{89}	2	....	....
Po^{210}	0.005	....	....

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