Radioactive Fallout,
Soils, Plants,
Food, Man

Edited by

ERIC B. FOWLER

# Radioactive Fallout, Soils, Plants, Foods, Man

EDITED BY

ERIC B. FOWLER

UNIVERSITY OF CALIFORNIA

LOS ALAMOS SCIENTIFIC LABORATORY

(CONTRACT W-7405-ENG-36)

LOS ALAMOS, NEW MEXICO



ELSEVIER PUBLISHING COMPANY
335 JAN VAN GALENSTRAAT, P.O. BOX 211, AMSTERDAM

AMERICAN ELSEVIER PUBLISHING COMPANY, INC. 52 VANDERBILT AVENUE, NEW YORK N.Y. 10017

ELSEVIER PUBLISHING COMPANY LIMITED
RIPPLESIDE COMMERCIAL ESTATE, BARKING, ESSEX

LIBRARY OF CONGRESS CATALOG CARD NUMBER 65-13896

WITH 72 ILLUSTRATIONS AND 83 TABLES

ALL RIGHTS RESERVED
THIS BOOK OR ANY PART THEREOF MAY NOT BE REPRODUCED IN ANY FORM,
INCLUDING PHOTOSTATIC OR MICROFILM FORM,
WITHOUT WRITTEN PERMISSION FROM THE PUBLISHERS

PRINTED IN THE NETHERLANDS

An editor spends considerable time attempting to collect manuscripts and at long last discovers that he is surrounded by papers with a wide variety of ideas and thoughts, many of which are conflicting, and is faced with the problem of welding the heterogenous material into a unit which can be placed between covers and called a book.

In this instance, a serious attempt has been made not to edit out differences, thus not to steriotype. The individuability of the respective author has remained sacred in his own chapter, differences lead to logical discussions and in turn to newer experimental design in order to obtain more nearly precise data.

A further aim of this work has been to gather in one place reviews by competent contemporaries in the associated fields of present research and philosophies concerning fallout, its relation to soils, plants, foods, animals, and man; the arrangement of the chapters is based on such a sequence.

It is apparent from a brief review of the Table of Contents that in certain chapters information is time-dependent. The lag between event and literature and the further lag between literature and publication of its review inevitably results in "dated" data.

Reports relative to the effect of the 1961–1962 test series are being documented. The reader is referred to U.S. Public Health Reports, especially Vols. 76–79; Radiological Health Data, Vols. III-V; and Nuclear Science Abstracts, Quarterly Indices for Vols. 17 and 18, for the most recent information.

There is an obvious repetition of certain referenced work, indicating its importance in a variety of fields of endeavor and revealing the interrelationship of problems, however, the five subsections remain discrete. The sixth subsection, Methods, applies equally well to those preceding; it might well be placed at any point. However, it has been assigned as a final chapter, a warning so to speak that data are method-dependent and must be

interpreted in this light. Such a warning is especially valuable to the newer worker in the area.

Some of the data presented in the following pages were presented at the ACS Division of Agricultural and Food Chemistry, 137th Meeting, "Symposium on Radioactive Fallout in Relation to Foods", Cleveland, Ohio, April 1960; appreciation is expressed to the ACS for its use.

Appreciation is also expressed to the many individuals who assisted in the preparation of the final manuscript, especially to those members of the Los Alamos Scientific Laboratory, the Health Division, and Group H-7, who reviewed the various aspects of the separate papers. Their patience and that of the typist was especially helpful.

Los Alamos, New Mexico August, 1964

Research work reported in certain of the following papers was supported in part by the U.S. Atomic Energy Commission. Appreciation is extended to the U.S. Public Health Service for data presented in Chapter VIII.

# TABLE OF CONTENTS

# Preface

FALLO	UT		1
Chapter	1	Considerations of Biospheric Contamination by Radio- active Fallout WRIGHT H. LANGHAM	3
SOILS			19
Chapter	2	Availability of Exchangeable and Non-Exchangeable Strontium-90 to Plants HOWARD ROBERTS, JR. AND RONALD G. MENZEL	21
Chapter	3	Relationship of Soil, Plant and Radionuclide: Soil Nutrients, Depth of Feeding and Site of Deposition within the Plant WARREN H. ADAMS, C. W. CHRISTENSON AND ERIC B. FOWLER	30
PLANTS			53
Chapter	4	Uptake of Radioactive Fission Products by Plants H. NISHITA, E. M. ROMNEY AND K. H. LARSON	55
Chapter	5	Above Ground Plant Parts as a Pathway for Entry of Fission Products into the Food Chain with Special Reference to \$9-90 Sr and \$137 Cs M. J. BUKOVAC, S. H. WITTWER AND H. B. TUKEY	82

Chapter	6	Natural Radionuclides in Foods and Food Source Materials ALFRED W. KLEMENT, JR.	113
Chapter	7	Radionuclides in Milk J. E. Campbell, G. K. Murthy, K. H. Lewis and C. P. Straub	156
Chapter	8	Transport Mechanisms Influencing 90Sr Levels in Milk Robert H. Shumway	180
Chapter	9	Transfer of Fallout Radionuclides from Diet to Man R. H. Wasserman, F. W. Lengemann, J. C. Thompson, Jr. and C. L. Comar	204
MAN			245
Chapter 10		Radionuclides in Man from Nuclear Tests J. LAURENCE KULP	
МЕТНО	DS	S	285
Chapter	11	Methods of Assessing Fallout GERALD H. HAMADA AND PAUL KRUGER	287
Glossary			304
Index			313

# **FALLOUT**

### Chapter 1

# CONSIDERATIONS OF BIOSPHERIC CONTAMINATION BY RADIOACTIVE FALLOUT

### WRIGHT H. LANGHAM

Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico (U.S.A.)

#### INTRODUCTION

Testing of nuclear weapons releases radioactive debris into the atmosphere, and its deposition over the earth's surface is termed "fallout". Fallout may be divided into three categories (local, intermedicate or tropospheric, and stratospheric or worldwide) on the basis of time and place of deposition.

Local fallout is deposited within the first 24 hours and confined to the immediate environs of the detonation. Since it is not widely distributed and much of it is deposited in the ocean, such deposition has not contributed significantly to general contamination of the biosphere.

Tropospheric fallout takes place in 20 to 30 days after a weapon test and occurs essentially in a poorly defined band around the world in the general latitude of the detonation. Small weapons like those shown in Fig.1 contribute all their fission products to local and tropospheric fallout. Although tropospheric fallout is widely distributed, it has not contributed greatly to general long-range contamination.

Weapon detonations equivalent in energy release to a million tons (megaton) or more of high explosive (Fig.2) carry their fission products into the stratosphere, where they are widely dispersed due to stratospheric diffusion and mixing. This material returns slowly to earth (average return time about 6 months to 3 years) and is referred to as stratospheric fallout. Stratospheric fallout has made the greatest contribution to long-range or worldwide contamination, since over 90 per cent of all the fission products produced by nuclear weapon tests has been from weapons with yields greater than one megaton.

The moratorium on large-scale atmospheric nuclear weapon tests makes it appropriate at this time to summarize the status of <sup>90</sup>Sr and <sup>137</sup>Cs biospheric contamination. These radionuclides are considered the most im-



Fig.1. A small nuclear detonation that contributes all its fission products to local and tropospheric fallout.

portant in long-range fallout because of their long physical half-lives and their chemical similarities to natural body constituents (respectively, calcium for skeletal formation and potassium for cellular function), which result in relatively high uptake in man and animals.

### WORLD INVENTORY OF 90Sr AND 137Cs CONTAMINATION

Weapon tests by all nations (1962) have produced fission products from about 90 megatons of fission energy release<sup>1</sup>. Total energy release from fission plus fusion (thermonuclear energy does not produce fission products,

<sup>&</sup>lt;sup>1</sup> The French tests were small and their contribution to the world inventory was insignificant.



Fig.2. A large nuclear weapon detonation that carries fission products into the stratosphere.

but it contributes energy to carry the debris into the stratosphere) has been (1962) about 170 megaton equivalents [1]. One megaton of fission energy produces about 100,000 curies of <sup>90</sup>Sr (0.1 MC, 0.1 megacurie) and about 160,000 curies (0.16 MC) of <sup>137</sup>Cs. Total production of these two potentially

6 W. H. LANGHAM

harmful radionuclides has been about 9 and 14 MC, respectively. If distributed instantaneously and uniformly over the entire earth's surface (area:  $2 \times 10^8 \,\mathrm{mi^2}$ ), the  $^{90}$ Sr contamination level would be about 45 millicuries (mC) per square mile of the earth's surface and  $^{137}$ Cs contamination would be about 70 mC/mi². These numbers, however, are misleading. The debris has not returned to earth instantaneously and, since both radionuclides have radiological decay half-times of about 28 years, they have been disappearing at the rate of about 2 per cent per year since their times of production. About 30 per cent of their total production was deposited as local fallout much of it in the ocean), where it does not contribute to general biospheric contamination. Furthermore, fallout is not uniform, but occurs preferentially in the temperate latitudes and predominantly in that hemisphere in which the tests are held. This, of course, means that the highest deposition levels occur in the north temperate latitudes, where, ironically, about 80 per cent of the world's population lives.

### DISTRIBUTION OF 90Sr AND 137Cs CONTAMINATION

As mentioned above, distribution of fallout from nuclear weapon tests is not only dependent on total yield of the detonation but also on the conditions and location of testing, and its potential hazards to the world population are strongly dependent on where it is deposited. The over-all distribution of fission products (as of November 1958, just after the U.S.S.R. and U.S. test series) was summarized in the 1959 Congressional Hearings [1]. General distribution of <sup>90</sup>Sr and <sup>137</sup>Cs contamination at that time (assuming no fractionation of the two) is shown in Table I.

TABLE I estimated over-all  $^{90}\mathrm{Sr}$  and  $^{137}\mathrm{Cs}$  distribution, november  $1958^*$ 

Location	<sup>90</sup> Sr (MC)	<sup>137</sup> Cs (MC)
Still in the atmosphere	3	4.8
On ground, world-wide	3	4.8
At or near testing sites	3	4.8

<sup>\*</sup> Derived from record of 1959 Congressional Hearings [1].

These estimations showed that about one-third of the total production of <sup>90</sup>Sr and <sup>137</sup>Cs was still in the atmosphere (November 1958), yet to be deposited; one-third had already been deposited as general or world-wide

tropospheric and stratospheric fallout; and one-third was deposited near the test sites, where it can be ignored as a general world-wide potential hazard.

Before we can understand the detailed distribution pattern of worldwide contamination, it is necessary that we consider the mechanism of stratospheric fallout. Above the earth's surface is a region of more or less constant air temperature. This region is the tropopause; the atmosphere below it is the troposphere, and that above is the stratosphere. Near the equator the height of the tropopause is about 50,000 to 55,000 feet and over the polar regions its height is 30,000 to 35,000 feet. In the temperate latitudes (40 to 50°N and S), the tropopause is ill defined or discontinuous. Masses of warm air rising into the tropopause near the equator may diffuse poleward, where they contact areas of intense cold in the stratosphere above the winter pole and upon cooling descend again into the troposphere. Return to the troposphere is preferential in the temperate latitudes because of the poorly defined or discontinuous troposphere. There is, in essence, a troposphericstratospheric air circulation system which results in fission products injected into the stratosphere being returned to the troposphere preferentially in the regions of 40 to 50°N and S latitudes. Once they are returned to the troposphere, they are deposited on the earth's surface in about 30 days, more or less in relation to area rainfall. Since most weapon tests have been held north of the equator, fallout has been much higher in the north temperate latitudes than in comparable regions of the southern hemisphere. Material injected at far northerly latitudes (as in the case of the large U.S.S.R. test series, October 1958) returns to the earth very rapidly (half-time 4 to 12 months) and all in the northern hemisphere. This mechanism of stratospheric fallout and the influence of U.S.S.R. and U.S. test site locations on world-wide distribution of radioactive contamination are illustrated in Fig.3.

On the basis of the above model of stratospheric deposition, one would expect maximum fallout levels to occur at about 40 to 50°N latitude, minimum levels near the equator, and a small peak in the south temperate latitudes. Figure 4 shows the 90Sr content of soil samples collected during 1958 as a function of 10° latitudinal bands [1]. Normalization of these data to November 1958 and integration with regard to world surface area show that indeed approximately one-third (3 MC) of the total 90Sr produced was already on the earth's surface as of that date. Aside from the 1961–62 tests, the information given in Table I and Fig.4 and estimates of stratospheric fallout rate may be used to make crude predictions of present 90Sr and 137Cs surface contamination levels and the maximum level that will be reached when the rate of fallout from the stratosphere is just enough to compensate

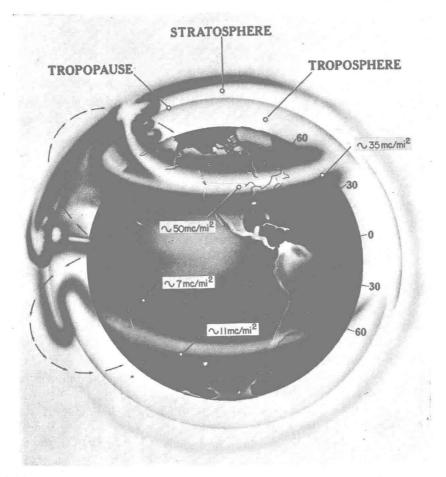


Fig.3. Mechanism of stratospheric fallout in relation to U.S. and U.S.S.R. test site locations [1]. Surface deposition levels are for \*90Sr as of November 1958.

for radioactive decay of that already deposited. Such crude predictions of <sup>90</sup>Sr surface deposition levels are shown in Fig.5. Assuming no fractionation of <sup>90</sup>Sr and <sup>137</sup>Cs during production and fallout, the <sup>137</sup>Cs levels would be about 1.6 times the <sup>90</sup>Sr values for the same latitude. These data show that the maximum <sup>90</sup>Sr surface deposition level in November 1958 in the north temperate population belt (about 40°N) was about 40 mC/mi². In 1960 the maximum may have reached about 65 to 70, and in 1962 (at the time the level should have begun to decrease) about 70 to 75 mC/mi².

The disproportionate increase between November 1958 and the present

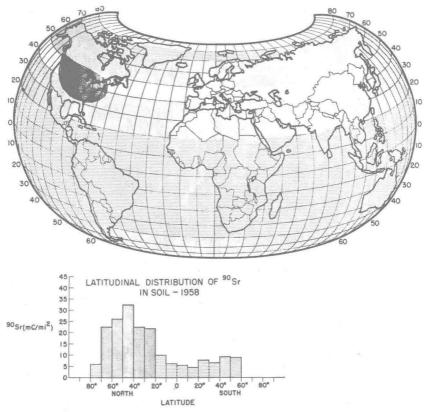


Fig.4. Latitudinal distribution of long-range fallout as of November 1958 (from 90Sr soil data [1]).

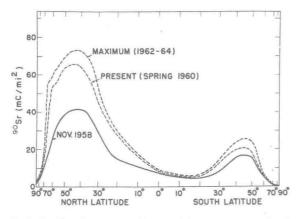


Fig.5. Predicted present and future 90Sr surface deposition levels as a function of latitude.

10 W. H. LANGHAM

at the far northerly latitudes is a result of the large U.S.S.R. test series at 73°N latitude in October 1958.

Incorporation of  $^{90}\mathrm{Sr}$  and  $^{137}\mathrm{Cs}$  into man and his ecological environment

Strontium is chemically similar to calcium, an essential constituent of bone, and cesium is similar to potassium, an essential element to living cells and tissues (especially muscle). If 90Sr and 137Cs are widely distributed over the earth, they will enter the ecological cycle from which man gets his food and be taken into his body as radioactive contaminants of calcium and potassium, respectively, thereby increasing his radiation exposure. The problem of world-wide fallout then becomes one of transport of these substances from soils to plants to animals and eventually to man. Much effort has been expended on working out and postulating ecological transport of 90Sr and 137Cs. One would expect discrimination against 90Sr with respect to calcium and 137Cs with respect to potassium at the various steps along the ecological cycle. Such discrimination is frequently expressed as the decrease in the radioisotope concentration of the required element as a result of passing through a particular step in the ecological cycle. For example, the concentration of 90Sr in the calcium of milk is only about 13 per cent of its concentration of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of milk is only about 13 per cent of its concentration of mi

tration of or solution of the cow's diet, or  $\frac{(90 \text{Sr/Ca}) \text{ milk}}{(90 \text{Sr/Ca}) \text{ forage}} = 0.13$ . From

such discrimination factors, the soil deposition levels, and average diets of the population, one can compose ecological models which can be used to make crude postulations of the 90Sr and 137Cs levels in man when he and his ecological environment are in equilibrium with the soil contamination level. Such an ecological model is shown in Fig.6 for 90Sr, and in Fig.7 for 137Cs. These projections indicate that the 90Sr concentration in the bone calcium of the U.S. population (at ecological equilibrium) should be about 7 per cent of its concentration in the available soil calcium, and that the 137Cs concentration in body potassium should be about 3 per cent of its concentration in the available soil potassium.

So many uncertainties exist in these projections that their principal value lies with their qualitative features in pointing out the routes of ecological transport. Only where entry of 90Sr and 137Cs into plants is predominantly through root absorption will their levels in man and his foodstuffs bear a consistent relationship to integrated fallout and conform to a true ecological model. Even in this case, a direct relationship between integrated fallout and