AEC ATMOSPHERIC RADIOACTIVITY STUDIES

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I. Introduction:

Studies of radioactivity in the atmosphere are conducted by the U. S. Atomic Energy Commission primarily for the purpose of evaluating hazards to man arising from the uses of atomic energy. For this reason the major part of this work is under the supervision of the Division of Biology and Medicine.

In its simplest terms, the problem is to determine the dosages to which people are likely to be exposed as the result of various uses of atomic energy, and to compare such dosages with either maximum permissible or natural "background" levels. The atmospheric monitoring activities of the AEC range from local programs associated with industrial operations to world-wide monitoring projects associated with the testing of nuclear weapons.

Although these studies have been motivated by practical necessity, their findings hold considerable interest for meteorologists. In turn, assistance of meteorologists is required in the design of the sampling networks, the interpretation and generalization of the results, and the development of models for predicting the distribution of radioactivity due to hypothetical sources.

The biomedically motivated studies are not the only studies of atmospheric radioactivity sponsored by the AEC. The Division of Research, for example, supports work on the geochemistry of uranium and on high energy physics including cosmic rays. Both of these physical science programs are generating information on the origins and distributions of naturally occurring radioactive nuclides in the atmosphere. The remainder of this discussion, however, will be limited to

those studies under the direction of the Division of Biology and Medicine, and to those of more than local interest.

The program dealing with the large-scale distribution of radioactivity has developed to its present state during a period dominated by the need to assess the world-wide hazard due to strontium-90 released in weapons tests. So much so, that the AEC program dealing with atmospheric radioactivity has been almost completely identified with the so-called "Sunshine" or strontium program. The scope of these studies has been gradually broadened, however, to include an increasing number of radionuclides and to include basic studies of mechanisms and models of global transport and deposition.

Small scale atmospheric distribution and transport of radioactive wastes due to the industrial applications of atomic energy have, of course, also been receiving steady attention. However, this part of the AEC program has become familiar to students of atmospheric turbulence and diffusion, and will be left out of the present discussion.

2. Fallout monitoring networks

The fallout sampling networks summarized in Table I provide the bulk of the AEC-sponsored measurements of radioactivity in the atmosphere and in precipitation (Table I). Where AEC is listed, this indicates that the analysis of samples is under the supervision of the Health and Safety Laboratory.

The gummed film collector (Fig. 1) consists of a square foot of cellulose acetate film coated with rubber-base cement (Ref. 1). Two are exposed at each station per day. At the end of the collection period the films are folded and mailed in a pre-addressed envelope to the AEC Health and Safety Laboratory, New York, where they are simply ashed and counted. Sampling stations are operated by the U.S. Weather Bureau and by many cooperating

TABLE I - AEC FALLOUT MONITORING NETWORKS

Type of Sample	Operating Agency	Number of Stations	Sampling Interval	Geographical Extent	Analysis
Gummed Film	AEC	112	Daily	World-wide	Total β
Precipitation:					
Pots	AEC	29	Monthly	World-wide	Sr^{90}, Sr^{89}, W^{185}, total β
Washtubs	AEC	5	Each rain	U.S. and N.Z.	Sr^{90}, Sr^{89} (U.S.), Cs^{137} (N.Z.), Ba^{140}, W^{185}, total β
Other	AFCRC*	6	Biweekly	76 N to 41 S	Sr^{90}, Ba^{140}, W^{185}, Pb^{210}, H^3
	PHS	44	Daily	U.S.	Total β
	WB-GS*	12	Biweekly	N. America	H^3
Soil	DA-AEC	87	2 yrs.	World-wide	Sr^{90}
	AEC	17	1 yr.	U.S.	Sr^{90}
Surface Air	NRL*	28	Daily	World-wide	Daily: total β Monthly: Sr^{90}, Sr^{89}, Y^{91}, Cs^{137}, Ce^{141}, Ce^{144}, Pb^{210}, W^{185}
	PHS	44	Daily	U.S.	Total β
Upper Air	AF-AEC	4	Monthly	45N to 23S	Sr^{90}, Sr^{89}, Zr^{95}, Cs^{137}, Ba^{140}, Ce^{144}, W^{185}, Rh^{102}, Total β

* Partial AEC Support

governments and institutions in foreign countries. This is by far the most inexpensive method of sampling fallout, but peculiarities of the collecting surface (Ref. 2) render the samples unsuitable for many desirable quantitative studies. However, the total beta counts, together with certain additional information, have been used in making estimates of Sr^{90} deposition (Ref. 3). Fig. 2 shows the latest world-wide estimates.

Fallout collectors which retain precipitation as well as particulates, while not providing such convenient samples for counting, have been found to collect more radioactivity per unit area than does the gummed film. In fact, it has been found that the bulk of the strontium-90 fallout is brought down by rain and is not retained by gummed film. Thus for direct measurement of the rate of deposition of Sr-90 on a monthly sampling interval high-walled stainless steel pots of about 1 foot diameter have been used (Table I). The samples are transferred to polyethylene bottles and mailed to the Health and Safety Laboratory for analysis. In addition to Sr^{90} they are analyzed for the shorter-lived isotope Sr^{89} to show whether the deposited material is of recent origin. These stations are operated through cooperative arrangements with various institutions and governments. The pots are now being replaced with a new collector consisting of a funnel and a simple ion exchange column (Fig. 3) which will simplify and standardize the sample handling procedure (Ref. 4).

The so-called "washtub" stations have been established in order to obtain adequate samples over shorter collecting periods for finer-scale studies of fallout rate (Fig. 4), and to obtain more information as to the origin of the precipitated debris by analysis of barium-140, an even shorter-lived radioisotope (Table I). At these stations, collectors of about 5 square feet area are exposed continuously and emptied after each rain. Fig. 4 shows the

detailed plot of cumulative Sr^{90} fallout vs. time at our Pittsburgh "washtub" station. The meteorological reasons for the variations in slope are still only slightly understood.

The Public Health Service program (Table I) provides rapid notification and documentation of the arrival and presence of fresh test debris in air and rain. The main use of the data is thus qualitative, but they also would indicate when gross levels of radioactivity are sufficiently high to warrant more detailed investigation.

The program of the Air Force Cambridge Research Center, recently begun and partially supported by the AEC, provides large-area rain samples for detailed analysis at selected stations of the Naval Research Laboratory air filter network. The Air Force Cambridge Research Center and the Weather Bureau are also collecting samples for tritium (H^3) analysis.

Soil samples, properly selected, provide the best measure of the total Sr-90 which has fallen per unit area of earth's surface. The analysis of bulky samples of soil for the minute traces of Sr-90 is far more difficult than the analysis of the more concentrated types of samples. The collecting and retaining properties of soils together with their flora and fauna are not well known and may be highly variable. It has been possible by extremely careful and laborious methods of soil sampling and analysis to obtain reliable measurements of total fallout representative of many land areas of the earth (Fig. 5) and to obtain useful estimates of increments over intervals of about 1 to 2 years. Plotted against latitude (Fig. 6), the 1956 data clearly show the northern hemisphere middle latitude maximum of Sr^{90} fallout. The 1958 soils, still in process of analysis, reaffirm this pattern.

Air filter samplers have been used since long before the days of atomic energy, so that at the time when the difficult problem of sampling deposition arose, efficient collectors for airborne dust were already available. Fig. 7 shows one of the Public Health Service units. Such samplers have been an essential part of local industrial radiation protection programs.

On the global scale, airborne radioactivity has been of less direct concern as a possible hazard than has the entry of deposited radioactivity into the food supply.

It has not been possible so far to relate the air concentrations to fallout in a simple, quantitative way. Air filters ~~samples~~ are unsurpassed, however, for providing concentrated samples on which counts can be taken quickly to give rapid early indications of the presence of fresh debris. The air concentrations also seem less sensitive to peculiarities of instrument exposure than is the fallout. Thus the patterns of variation in space and time obtained from the NRL 80th Meridian Network are qualitatively simple and interpretable. The NRL network is shown in Fig. 7 (Ref. 5). The monthly profiles of daily average air activity plotted against latitude for the period July 1957-June 1958 show clearly the U.S. Plumbob Series, the USSR spring tests and Operation Hardtack.

In the study of transport and deposition processes there is no doubt that the air concentration data are of fundamental quantitative importance. It is possible that they will be of more value in meteorological research than in evaluating hazards.

The stratospheric sampling program will be of special interest to meteorologists. Because of very slow vertical mixing and the lack of efficient cleansing mechanisms such as precipitation, the stratosphere is a storage reservoir for finely divided radioactive debris injected into it. At first, attempts were

made to estimate the amount of radioactivity in storage and the rate of transfer from this reservoir by subtracting the total observed fallout from the total amount released in tests. Fig. 10 shows an example (Ref. 6). This method led to estimates varying from 1 to 3 megacuries of Sr^{90} in the stratosphere in the period late 1957 to early 1958 (Ref. 7, 8, 9). Uncertainties in the estimates of total yields of tests by all nations and in the estimates of integrated fallout over the earth prevent any greater precision in estimates by the difference method. A large part of the uncertainty is in the estimate of the fraction of "local fallout". This is the fraction contained in the region of maximum deposition within a few hundred miles from the test site. In Pacific tests, for example, it is virtually impossible to obtain a sufficiently complete map of the local fallout to permit accurate integration.

In order to obtain direct measurements of the concentrations of strontium-90 in the stratosphere, a series of balloon sampling flights was made by the New York Operations Office of the AEC in 1953 and 1954 at about 80,000 to 100,000 ft. over Holloman Air Force Base, New Mexico with a simple electrostatic precipitator (Ref. 10). Fig. 11 shows a pair of sampling units ready for flight. This gave qualitative indications of the presence of Operation Castle debris in the summer of 1954 (Fig. 12). Quantitative radiochemical analysis was impractical, however, because the samples collected were too small and the efficiency of collection was unknown.

In the fall of 1956, after extensive operational tests, a program of routine stratospheric balloon sampling flights ~~developed by General Mills, Inc.~~ was undertaken with the cooperation of the Air Force using a filter developed by General Mills, Inc. paper system. This program has had the nickname "Project Ash Can" because of the shape of the sampling device (Fig. 13). Samples are collected at

50,000, 65,000, 80,000 and 90,000 ft at each of four stations once a month. These stations were originally Minneapolis, Minn.; San Angelo, Texas; France Air Force Base, Canal Zone; and Sao Paulo, Brazil. The Minneapolis station has recently been replaced by Sioux City, Iowa. The Brazil and Panama stations are being closed down after two years of operation.

The filters are analyzed for Sr^{90}, five other fission products and two tracers (Table I). Development of reliable analytical procedures has been very difficult due to the extremely minute concentrations of the radioisotopes in the air, and to the relatively low collection efficiency of the filter system. Although reporting of results by commercial analytical laboratories initially lagged more than a year behind the first routine sampling flights, this lag has been reduced to a few months. Nearly two years' data are now available in an unclassified AEC document (Ref. 11). Cross sections of the average Sr^{90} concentrations for two 6-month periods are shown in Fig. 14, prepared by R. J. List.

There are still considerable uncertainties in the collection efficiency and radiochemical analysis, but these are being narrowed. The very small number of sampling points introduces even greater uncertainties in the estimate of global inventory. However, the total amount of Sr^{90} in the stratosphere estimated on the basis of these measurements up to last spring would appear to be closer to 1 than to 3 megacuries. Studies are underway to attempt to reconcile all the available data.

3. Smaller-scale research projects

The projects discussed so far are all of an operational monitoring nature, aimed at obtaining large-scale distributions of the radioactivity in the atmosphere or in fallout by direct measurement. This is the "brute force" approach.

Attempts to interpret the data obtained by these projects have raised a steady crop of questions regarding mechanisms. The planning of the operational programs has always been strongly influenced by current ideas about these mechanisms.

An increasing effort is being supported by the AEC in the area of basic mechanism studies. These studies make use of radioisotopes as tracers, non-radioactive tracers such as ozone and water vapor, or the methods of classical synoptic and dynamic meteorology.

The mechanisms being investigated can be grouped under:

(1) global atmospheric transport and mixing processes

(2) aerosol behavior including scavenging by precipitation.

A list of those projects involving sampling and analysis of atmospheric radioactivity follows (Table II).

Table II. Research Projects Involving Measurements of Atmospheric Radioactivi

Strontium-90 and other fission products

Scripps Institution of Oceanography (rain, air)
Mt. Washington Observatory - AEC (cloud, rime, precipitation)
Armour Research Foundation - AEC (particle size fractions)

Carbon-14

Los Alamos Scientific Laboratory
Scripps Institution of Oceanography
U.S. Weather Bureau - Argonne National Laboratory
Lamont Geological Observatory

Tritium

University of Chicago
Weizmann Institute (Israel)
Scripps Institution of Oceanography

Radon

Argonne National Laboratory
U.S. Geological Survey (sponsored by Div. of Research)

Both carbon-14 and tritium (H^3) occur naturally, are also produced in weapons tests, and are radioisotopes of elements which are of importance in meteorology and ecology. Thus the global transport cycles of bomb-produced carbon-14 and tritium are inextricably associated with the carbon dioxide and water cycles in nature (Ref. 12, 13, 14, 15). It seems inescapable that these tracers should become important tools in certain branches of meteorology

4. Development of Sampling Devices

Considerable trial-and-error experimentation has gone into the design of ground-level collecting devices for airborne particulates, precipitation and deposited dust. Each system in use has been subjected to more or less thoroug calibration tests, and various more sophisticated new approaches have been suggested. However, by and large it has seemed preferable to use the simplest methods available except for special research purposes.

The upper-air sampling problem, however, has not been susceptible to such simple solutions. A substantial part of the cost of the stratospheric monitor ing program has gone into attempts to calibrate the Ash Can system and to develop better ones. The following requirements will show the nature of the difficulties.

(1) The particles to be collected are believed to be mainly in the diameter range from .01 to .1 micron.

(2) The minimum air volume containing sufficient radioactivity for analysis is of the order of 300 standard cubic feet, that is, about 3,000 cubic feet at an altitude of 50,000 ft and 20,000 cubic feet at 90,000 ft. It has been estimated that the total mass of dust collected is of the order of .001 to .1 microgram.

(3) The sampling rate must be such that the balloon flight time at altitude is less than about 2 hours since the difficulty of recovery increases with increasing distance of the impact point from the release point. This time limitation implies volume flow rates of the order of 500 cubic feet per minute assuming the collection efficiency is 25% or better.

(4) The weight must be kept as low as possible, preferably less than 100 lbs to keep the balloon size within reasonable limits.

(5) The collection efficiency must be definitely known. It should also be high so as to reduce volume flow requirements to a minimum, and should be independent of particle size, since the particle size distribution is unknown

The Ash Can system has suitable weight and volume flow rate and is relati inexpensive to manufacture and simple to operate, but it has a low filtration efficiency (preliminary calibration data indicate about 20 to 25% at the lowes sampling altitude). Furthermore, exhaustive laboratory tests have shown tha the collection efficiency varies strongly with particle size and air density. This system has the further disadvantage for the radiochemist that the tenth of a microgram or less of stratospheric dust which is collected, containing less than a billion atoms of Sr-90, is spread over 5 square feet of thick, loose-textured filter paper. One may feel some anxiety lest some of them be evaporated or otherwise lost under the rough treatment, such as ashing or dissolving in quantities of strong acid, which is necessary in order to prepa the bulky filter for chemical analysis. Some desirable methods of study such as autoradiography or electron micrography are rendered virtually impossible by the bulk and dilution of the sample.

The AEC is sponsoring development work on several more advanced concepts: an improved filter system, an improved electrostatic precipitator and a rotating impactor. The advanced filter system, designed for higher face

velocity, higher collection efficiency, smaller collection area, more reproducible performance and higher altitude capability, has been built and flown several times by General Mills, Inc. The others, still in the preliminary design stage, promise high efficiency with lower resistance to air flow, low weight, and still greater concentration of the sample.

Several flights have been made with a stratospheric air liquefaction system designed by the Chicago Midway Laboratories to collect whole air samples for calibrating the routine collectors. Fig. 15 shows the collector. The total weight of a typical flight train is close to a ton. Results of analysis of the samples collected by this liquid air system are now being evaluated.

A device is also being developed to collect separate particle size fractions in the stratosphere for radiochemical analysis.

It is hoped that the development of high-altitude particulate sampling devices will contribute not only to the solution of our own practical problems but also to a wide range of basic studies on such subjects as global exchange processes, meteoritic accretion and others.

5. Hardtack Tracer Experiment

In order to help in distinguishing among various possible routes of transport of debris from test detonations, the AEC has recently added the isotopes tungsten-185 and rhodium-102 to the list of analyses performed. These isotopes resulted from neutron activation of materials present in certain nuclear devices. As far as is known, the proportions of these radioactivities to the fission products in the debris resulting from the Hardtack test series conducted in the Pacific Proving Ground last spring and summer were appreciably higher than from other sources. These isotopes can therefore serve as tracers for the Hardtack debris.

Tungsten-185, with a half-life of 74 days, was produced in a number of detonations over silica sand.

By studying this isotope it is hoped that the origin of the middle northern hemisphere maximum of strontium-90 fallout can be clarified.

Three main models of world-wide transport have been proposed, each giving a different interpretation of this observed phenomenon. Reduced to their simplest terms, they are:

(1) A model, described by Libby (Ref. 8), based on uniform distribution of radioactive debris in the stratosphere. In this model the non-uniformity of fallout is due to non-uniform rainfall and to the contribution from low-yield tests which deposit debris in the troposphere. The low-yield tests have been mostly conducted in the middle and high latitudes of the northern hemisphere. This model, then, attributes a major part of the strontium-90 fallout which had occurred in the United States by late 1957 to Nevada tests and USSR low-yield tests.

(2) The second model, described by Machta (Ref. 6), is based on a very slow mixing rate, both horizontally and vertically in the lower stratosphere, together with a slow poleward circulation in both hemispheres. In this model material from both equatorial Pacific tests and high-latitude USSR tests tends to drift towards higher latitudes and to enter the troposphere non-uniformly with a maximum in the middle latitudes of both hemispheres in late winter and spring. If this model is correct, the major part of the strontium-90 fallout in the United States up to early 1958 had come from tests in the equatorial Pacific.

(3) A third model, recently proposed by Martell (Ref. 16) is based on a shorter stratospheric residence time for debris injected at high latitudes than at equatorial latitudes. This model attributes a large part of the

strontium-90 fallout which has occurred in the United States to USSR high-yield tests.

Underlying the study of tungsten-185 is the hope that by determining the contribution of a single equatorial test series to the fallout in all parts of the world, it will be possible to distinguish among these different models. It may be that each is partially correct or that the actual situation is considerably different from any of them. In any case it seems quite clear that any gain in understanding the route and origin of the observed fallout must be accompanied by a gain in understanding the circulation of the stratosphere.

The rhodium-102, having a half-life of 210 days, is being used to study the spread of debris from a high altitude . From available estimates of turbulent diffusion coefficients (Ref. 17) the time required for the rhodium-102 to spread down to the tropopause would be many years. By that time it would have decayed away completely for practical purposes. The main motivation for this experiment is to find out whether there is some unknown transfer process which would return such fine particulate matter to the lower stratosphere more rapidly than would be expected on the basis of known mixing processes. If such a mechanism exist with a time scale of weeks or months, there is a chance that the tracer would be detectable in some of our samples by sensitive analytical techniques, although far below the concentration of other radioactivity already present.

6. Summary

The radionuclides being studied in the various projects which have been discussed are listed in Table III. Many others, both natural and artificial, are present in the atmosphere and could undoubtedly be studied with profit. However, at the present stage much more must be learned before we can feel assured that we are making full use of the considerable number now being measured.

Table III. List of Radionuclides

(1) Fission products of direct biomedical interest.
Sr^{90}(28 y) chemically like Ca, daughter of 33 sec Kr^{90}
Cs^{137}(30 y) chemically like K, weak gamma

(2) Other fission products
Sr^{89}, Y^{91}, Zr^{95} (half lives about 2 months), used to indicate age of debris and fractionation effects due to chemistry, solubility or gaseous precursors; Zr^{95} can be detected by gamma spectroscopy.
Ba^{140} (12.8d) used to indicate very fresh debris
Ce^{141}, Ce^{144}, a pair of beta-gamma emitters of 1 and 10 months half life, used for age and fractionation determination.

(3) Other artificial activities
Rh^{102} (210 d) and
W^{185} (74 d) associated with Hardtack series.

(4) Cosmic-ray and bomb produced activities
H^3 (tritium) (12.5 y), used in studying H_2O cycle
C^{14} (5700 y) used in studying CO_2 cycle, global transport

(5) Natural terrestrial activities
Rn^{222} and Pb^{210} (RaD), 3.8 d gas and its 19.4 y solid daughter for studying micrometeorology, air masses, global exchange, scavenging.

While practical problems are central to this program, and basic meteorological research peripheral, the sampling cannot lift itself by its own bootstraps. When a body of data, such as the stratosphere or rain data, exhibits considerably greater variability or complexity than is consistent with the simple model on which the choices of sampling grids and intervals were based, it becomes doubtful whether an adequate sampling plan can be developed by studying the internal characteristics of this body of data alone. It is then pertinent to ask such questions as the following:

(1) Are the variations due to organized phenomena on a smaller scale?

(2) Are representative ranges of the controlling variables being sampled?

(3) What is the likelihood of these variables combining to produce concentrations of radioactivity at some time and place much greater than any that have been observed?

(4) Are there large scale variations or trends which might to some extent invalidate material budget estimates or estimates of hazard based on the available data?

The controlling variables are, of course, the same ones which control the global transport of heat, momentum, moisture and ozone. The AEC is supporting some basic research on stratospheric meteorology at MIT and at the Weather Bureau, employing these more familiar elements of synoptic and dynamic meteorology.

For the foreseeable future, the AEC will continue to need quantitative estimates of atmospheric contamination and fallout under various actual or hypothetical conditions. Such estimates can seldom be made by simply extrapolating from data obtained by analysis of environmental samples.

It may well be that the main practical value of the radioactivity sampling studies reviewed in this paper will be through their contribution to basic meteorology.

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