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United States Atomic Energy Commission, Washington 25, D.C.

## Radioactive Fallout

Remarks Prepared by Dr. Willard F. Libby,
Commissioner United States Atomic Energy Commission

### 1. Introduction

The whole world is concerned over the question of radioactive fallout, particularly that from the testing of nuclear weapons. This has focused world-wide attention on the problem of the effects of radiation, whether it be from atomic fallout or medical X-rays, and a field of knowledge formerly known to only a limited group of scientists is becoming a matter of general concern, thought about and discussed by millions of people. The widespread concern may be due to the general fear of the unknown which has always been a basic human instinct. If the knowledge of the effects of radiation and the magnitude of the doses from fallout were more widely known, this would considerably allay the apprehension. So the first problem is the dissemination of the knowledge of fallout and radiation effects which has been gained over the last several years, and it is for this reason that this paper is presented. In June, 1957, the Congress of the United States held extensive hearings on radioactive fallout and radiation, and the minutes of these hearings are one of the best sources of information about the whole subject. In addition, a considerable number of articles have been published since then which present more recent data and considerations. I hope to refer to some of these in the present paper.

Since there is every reason for the information on radioactive fallout and radiation to be known to any interested person, the United States Atomic Energy Commission has the policy of publishing promptly and completely on this subject, and this paper serves this function also. Before beginning a main subject of "Radioactive Fallout", I would like to mention a new development which though related is not entirely germane.

During the recent test operations of the U.S. Atomic Energy Com-

mission and the U.S. Department of Defense, in Nevada, Operation Plumbbob, a bomb was fired underground which had no radioactive fallout because its fireball was sealed in molten rock. The fireball consisted largely of vaporized rock which congealed and totally contained the radioactivity. Essentially no radioactivity, even that belonging to such a volatile material as radioactive krypton, escaped to any considerable degree.

The entirety of the radioactive material was found in some 700 tons of rock which had been fused and then cooled and crushed. Apparently the bomb, which had the power of 1,700 tons of ordinary chemical explosive, blew itself a bubble of vaporized rock about 55 feet in radius, which had a skin about 3 or 4 inches thick. The shock wave crushed rock out to about 130 feet so the weight of the crushed rock overhead crushed the thin eggshell when it cooled and broke it into fragments. These fragments contain the bomb debris essentially in its entirety. This means it is possible, at least in the small yield range, to contain and eliminate radioactive fallout in certain types of weapons tests. Of course, effects tests where such materials as structures and military equipment are being checked against atomic blast cannot be conducted in this manner, but these tests could conceivably be done with the special type of bomb with reduced radioactivity. Thus, it is likely that a technique has been developed which will make possible test operations which contribute much less fallout.

In addition, the non-military applications of atomic explosives, which the underground shot on September 19 last year disclosed, appear to be so promising that for them alone we must continue certain tests in order that these benefits may be available to the human race. For example, in the underground shot, just mentioned, we produced an earth shock which was very revealing to the seismologists in its clarity and sharpness within a considerable distance from the Nevada test site and it is certain now that from atomic detonations we will be able to determine the internal structure and character of the earth with a clarity and detail never possible with earthquake shocks because of their diffuseness in both time and location.

A second possibility is the applicability of nuclear explosions to moving earth, if the fallout hazard can be controlled. Craters produced in the Pacific islands are convincing testimony of the possibility of making harbors in regions where the local fallout hazard is tolerable. Perhaps with the devices of reduced fallout which are now being developed such applications will be possible in more populated regions.

A third most intriguing possibility is that of shaking and breaking

subterranean structures by nuclear shock. The underground detonation, despite its small 1.7 kiloton yield, is estimated to have crushed about 0.4 million tons of rock. It happened that the mountain selected consisted of rather soft rock, but nevertheless it was consolidated and supported its own weight. After the explosion, a sphere 260 feet in diameter was crushed so it could easily be mined. It was not rendered radioactive because the radioactivity was contained in thin rock shell mentioned earlier, which weighed only 700 tons and which was visually distinguishable from the ordinary rock and thus can be separated easily. It is clear that this type of application has great promise. A fourth example is the containment of the heat generated from large atomic explosions in rock structures which are dry and therefore free of the pervasive thermal conductive characteristics of steam and water. This affords a definite possibility for generating atomic power; if detonations, which are large enough to make such power economical, are practical and if the subsequent drilling and removal of the heat by injection of water to produce steam prove to be practical. A fifth example is the possibility of making radioactive isotopes by surrounding the explosive devices with appropriate materials so that the neutrons which always escape in atomic explosions can be utilized at least in part. A sixth example is the potential utilization of the radiation and heat of the bomb to cause chemical reactions. These six possible non-military applications show that nuclear explosions may have peaceful applications of real importance and that the understanding of the phenomena of radioactive fallout is useful not only in conducting a weapons test, but in the promotion of important peaceful applications.

The radioactivity produced by the detonation of nuclear weapons has been extensively studied and reported upon (1–17). From this work we have learned about the amount of radioactive fallout which occurs, and the mechanisms for its dissemination in a broad and general way. Let us consider a few of these general points.

1. The stratosphere plays an extremely important role for the fallout from megaton yield weapons, and the troposphere is the medium which disseminates the fallout from kiloton detonations; thus, speaking broadly, stratospheric debris is from megaton yield detonations and the tropospheric fallout is from those of lower yield. It is not that the yield of the detonation is determinative, but rather that the altitude to which the fireball rises before its average density is equalized with that of the surrounding air determines the fallout rates. The megaton yield fireballs are so enormous that they stabilize at levels only above the tropopause—the imaginary boundary layer dividing the upper part of the atmosphere,

the stratosphere, from the lower part, the troposphere—while the kiloton yield fireballs stabilize below the tropopause. The tropopause normally occurs at something like 40 to 50 thousand feet altitude, although it depends on season and location. In other words, low-yield bombs fired in the stratosphere would be expected to give the same slow fallout rates as high-yield weapons do when fired in the troposphere—or on the surface if attention is focused on the part of the fallout which does not come down locally to form the oval-shaped pattern pointed in the downwind direction.

2. The stratospheric debris descends very slowly, unless, of course, it is so large as to fall in the first few hours. This paper is concerned only with the world-wide fallout—that is, the fallout which does not occur in the first few hours, and excludes the local fallout which constitutes the famous elliptical pattern which is so hazardous because of its radiation intensity, but which in test operations is carefully restricted to test areas. It is worth mentioning in passing that the local fallout may be the principal hazard in the case of nuclear war. Most serious attention should be paid to it in civilian defense programs.

The world-wide fallout from the stratosphere is literally world-wide in that the rate of descent of the tiny particles produced by the detonations is so small that something like 10 years or somewhat less probably is the average time they spend before descending to the ground, corresponding to an average annual rate of about 10 per cent of the amount in the stratosphere at any given time. It is not clear as to just how they do finally descend. It seems probably that general mixing of the stratospheric air with the tropospheric air which occurs as the tropopause shifts with season and as is brought about by the jet streams constitutes the main mechanism, and that the descent of the stratospheric fallout is never mainly due to gravity; but rather the bulk mixing of stratospheric air with tropospheric air brings the radioactive fallout particles down from the stratosphere into the troposphere where tropospheric weather finally takes over. This mechanism makes the percentage fallout rate the same for all particles too small to fall of their own weight—and the same as would be expected for gases providing some means of rapidly removing the gases from the troposphere exists, so the reverse process of troposphere to stratosphere transfer does not confuse the issue.

3. World-wide radioactive fallout in the troposphere is restricted to the general latitude of the detonations for the reason that the residence time in the troposphere is about 30 days (18–21). The lifetime of fine particulates in the troposphere appears to be determined by the cleansing action of the water droplets in the clouds. For those particulates which

are below 1 micron in diameter, Greenfield (20) calculates that the mean residence time of a 1-micron particle in a typical cloud of water droplets of 20 microns diameter may vary between 50 and 300 hours, but that a particle of 0.04 micron diameter will last only 30 to 60 hours, and that a particle of 0.01 micron diameter will last only 15 to 20 hours. The theory calculates the diffusion due to Brownian motion and says that it is just this motion induced by the collisions with the air molecules which makes possible the contact between the fallout particles and the cloud drops. Since this theory is based on first principles with the single assumption that the fallout particle sticks to the water droplet on impact—an assumption so plausible as to be almost beyond doubt—it is no surprise to learn experimentally that the Greenfield theory appears to be correct.

There is essentially no world-wide fallout in the absence of rainfall; i.e., in desert regions—except for a little that sticks to tree leaves, blades of grass, and general surfaces, by the same type of mechanism Greenfield describes in the case of clouds. Thus we see that it is the moisture in the troposphere which assures the short lifetime of the world-wide fallout particles, and that when the stratospheric air which contains essentially no moisture and therefore has no cleansing mechanism descends into the troposphere, the tropospheric moisture proceeds to clean it up. On this model, we see that for submicron fallout particles, weather phenomena are controlling, and that the bombs which have insufficient energy to push their fireballs above the troposphere will have their world-wide fallout brought down in raindrops in a matter of about a month, in extreme contrast with the stratospheric material which apparently stays aloft for something like 10 years on the average. The contrast between these two lifetimes means that the concentration of radioactive fallout in the stratospheric air in terms of equal densities of air is always much higher than in tropospheric air. This has been experimentally observed to be true (22). In fact, the stratospheric content is about one hundredfold higher than that of the troposphere corresponding to the much longer stratospheric residence time. Later in this paper new data on the fallout content of the stratosphere are given.

It is inherent in the Greenfield mechanism that the total world-wide fallout will be proportional to rainfall if other factors are not allowed to vary. Thus we find that the Mediterranean basin (2) affords a good example of the truth of this principle. Other regions are the Northeastern United States, the Southeastern United States, the Northwest United States and the Southwest United States (23). It is now well established that desert areas have very little fallout.

4. After falling to the ground in the form of rain or being picked up on the surface of the leaves of grass or trees by the same type of Brownian motion accretion mechanism causing cloud drop pick up, the radioactive fallout may enter the biosphere by normal biological processes. Radioactive strontium-90 (Sr<sup>90</sup>) and radioactive cesium-137 (Cs<sup>137</sup>) are the two principal isotopes which have this facility and are produced in high yield by the fission reaction and are of long enough lifetimes to be disseminated world-wide particulary by the stratospheric mechanism about 28 years half-life for each. Sr<sup>90</sup> is produced at a level equivalent to about 1 millicurie of Sr<sup>90</sup> per square mile of the earth's surface for every two megatons of fission energy, and radiocesium is produced at about 50 per cent higher yield. Of the two isotopes, Sr<sup>90</sup>, because of its chemical similarity to calcium, collects in human bone, where it is held for years and where its radiations might then cause deleterious effects to the health of the individual, such as leukemia or bone cancer. It is interesting that Sr<sup>90</sup> constitutes a relatively less important genetic hazard because of the short range of its radioactive radiation and the fact that it is not held in the reproductive organs. Radiocesium stays in the human body only 6 or 8 months on the average, because it has no permanent structure like the bone for which it has a natural affinity. As a result, the amount of radiation occurring from internally ingested radiocesium is much less and most likely is subject to palliative measures calculated to reduce its time in the body. Sr<sup>90</sup> taken into the bone, however, appears to be stored for many years, the exact time not being known very well (24).

Radiostrontium is taken into the body because of its similarity to calcium, but there is a definite difference in chemical behavior which causes animal organisms to prefer calcium. Thus the radiostrontium content of newly deposited bone calcium is less than that for food calcium. In many countries, the principal source of calcium is milk products, so the fact that cow's milk has only one-seventh the strontium in it per gram of calcium that the cow's food has, and that milk taken into the human body similarly deposits calcium in the bones with only half the Sr<sup>90</sup> content of the milk itself means that human beings naturally have a lower  $Sr^{90}$  to calcium ratio for new bone than for the food source by something like a factor of 15 for dairy products. On the other hand, vegetation containing Sr<sup>90</sup> also deposits its strontium relatively inefficiently with a factor of something like 4 less strontium in the bone from these sources than is carried in the vegetable food itself-all relative to calcium. In some countries where calcium in the human diet comes principally from vegetables other sources of calcium contribute, some

of which contain essentially no Sr<sup>90</sup>—namely sea food. Because fallout is diluted so quickly by the action of the waves in the ocean, the concentration of the radioactive strontium in the sea calcium is very much lower than it is in the soil of the land in which the grass and vegetable crops grow. This difference becomes even larger when the effects of direct leaf and stem base pick up are considered. This perhaps accounts for the high values reported by *Ogawa* (25) for rice in Japan. So, fish from the sea are naturally at the lowest level in radiostrontium and sea food should be the lowest source of calcium among ordinary human foods. With all of these factors taken together, the world populations assimilate calcium at a much lower radiostrontium content than is exhibited by land plants to a very considerable degree. *Eckelmann*, *Kulp* and *Schulert* (10b) have given a detailed sample calculation recently, based on their extensive measurements on human bone.

5. The biological hazard from the radioactive fallout from weapons testing is not well known, and like many biological problems the determination of the hazard in any exact way seems to be almost impossibly difficult. Fortunately, however, it is possible to compare the radiation from radioactive fallout with the intensities of natural radiation to which we are always exposed. For example, it is clear that the present level of the radiostrontium in the bones of young children which are, of course, closest to being in equilibrium with the fallout, since adults have had their bones some time even before there was any radioactive fallout, is about 2 milliroentgens (mr) per year as compared to an average natural dosage of 150 to 200 mr per year, about 1 to 2 per cent of the dosage from natural sources to the bones depending upon location. Natural radioactivity present in the ground, building materials and even in our own bodies gives us an average total dose at sea level of about 150 mr per year, and medical X-rays add something like another 150 mr. The radiocesium taken into the body and the penetrating radiations from non-assimilable radioactive fallout contribute perhaps another 3 or 4 per cent to the whole body dosage. Thus the total dosage to freshly formed human body is at most 5 per cent of the natural dosage. Furthermore, we do know that the variations in natural background dosages from place to place are enormous in magnitude as compared to the average value, and of course as compared to the fallout dosage. For example, it has been found (26) that exposure rates from external radiation rise from a value of about 110 mr per year at sea level to something like 230 mr per year at 5000 to 6000 feet altitude in the United States. These numbers are considerably larger than those expected on the basis of earlier calculations and measurements (7, 27, 28), the increase apparently being due to the cosmic rays and their increase with altitudes (29). In addition, the effects of radioactivity in the soil and in building materials made of stone or soil are considerable, amounting in some instances to 50 or 100 per cent of the average natural background dose at sea level, and the magnitude of the medical exposures to X-rays approximates on the average those due to all natural sources (30).

We see, therefore, that whatever the extent of our ignorance of the biological effects of radiation, we do know that these effects are not unexperienced by the human species, even from the genetic point of view, since it is clear now that persons living at high altitudes on granitic rocks always have received extra radiation many times greater than is contained in the radioactive fallout from the testing of nuclear weapons, and that even those living on certain sedimentary rocks at sea level always have received about 10 to 20 times the present fallout dose.

Of course, this does not mean that any of the effects from radioactive fallout are in any way negligible and it does not mean that certain numbers of people will not be injured by radioactive fallout radiations, even though these numbers be very small relative to the total population of the world. However, the problem is bounded, and common sense and good judgment can be brought to bear on the extent of the biological hazards even though they are not now known exactly, and probably will not be well understood for many years. Researches to increase this understanding are being done, especially in the United States and United Kingdom and other countries. Information on radioactive fallout and all of its aspects, both physical and biological, is collected and collated by the United Nations Scientific Committee on the Effects of Atomic Radiation, which is drafting its first report at the present time.

- 6. From our study of radioactive fallout from testing, we have learned much of value about the circulation of the atmosphere of the world, and we have much more to learn as the study continues, particularly in the stratosphere by balloon and aircraft sampling techniques being carried out principally in the United States at the present time. As we undertake the problem of locating the fallout in the oceans, we undoubtedly will learn much of interest to oceanographers about the circulation of the water in the seas.
- 7. From our understanding of radioactive fallout from tests, we are the better able to devise methods of civilian defense against fallout in the case of nuclear war, and wide-spread popular interest in the potential possible hazards from radioactive fallout from nuclear tests has led to a considerable understanding on the part of the general public of these

strange phenomena. From this debate and study may come the protection for millions in the case nuclear war should occur.

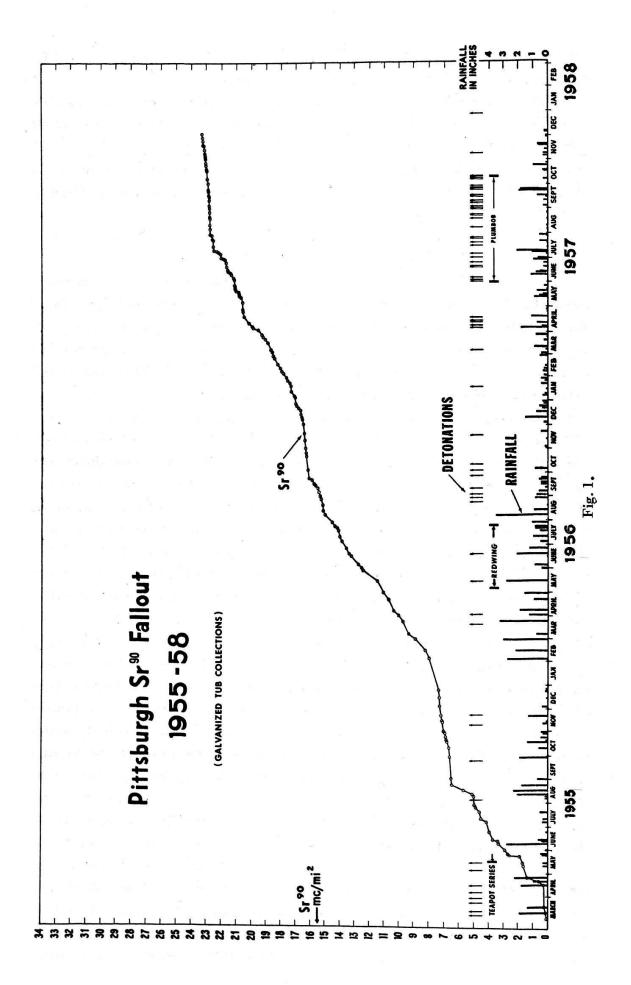
Understanding of the nature of the mechanism by which radioactive fallout is disseminated has led to the reduction of the offsite fallout from testing. We know now that bombs placed upon the ground produce relatively more local fallout and therefore less world-wide fallout. It seems likely that firing on the surface of the sea has a similar, though probably considerably less marked effect.

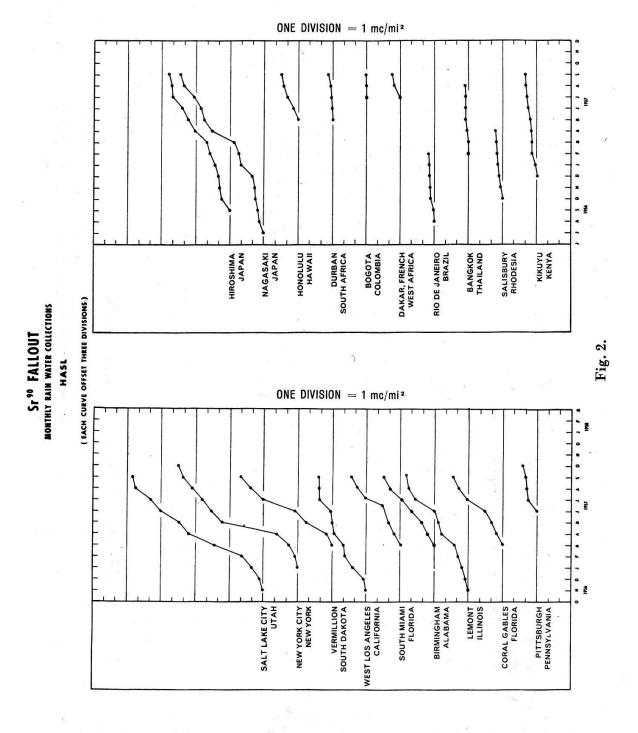
## 2. Recent Data and Their Implications

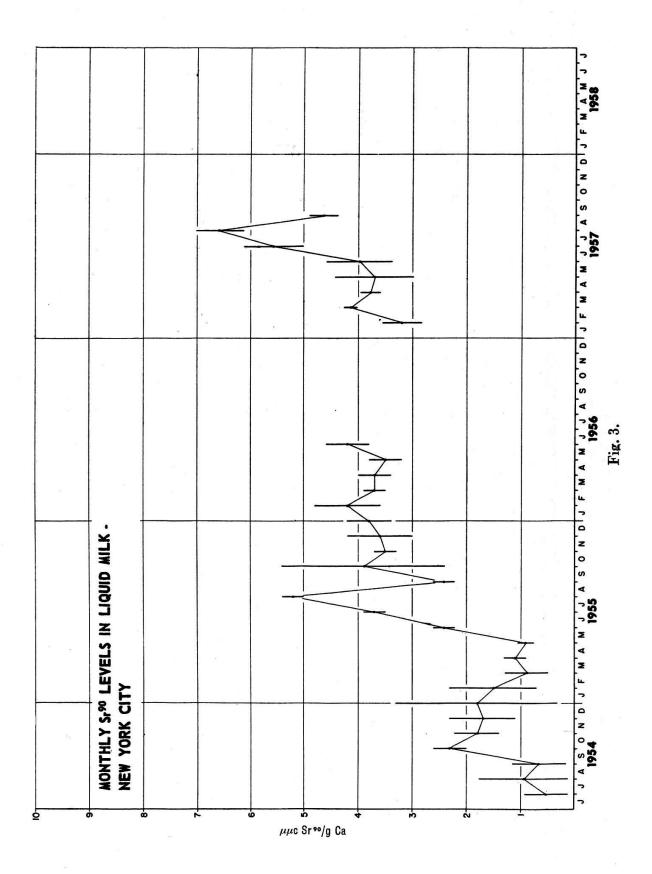
Figs. 1, 2, 3, 4 and 5 and Tables 1, 2 and 3, which are up-to-date versions of earlier publications, give the most recent results for the fallout observed for rainfall collections, for the Sr<sup>90</sup> content of milk (fresh and dry), for human bone, and for animal bone. It is particularly interesting to note that the data continue to show the principal features noted previously and that little new in principle has appeared.

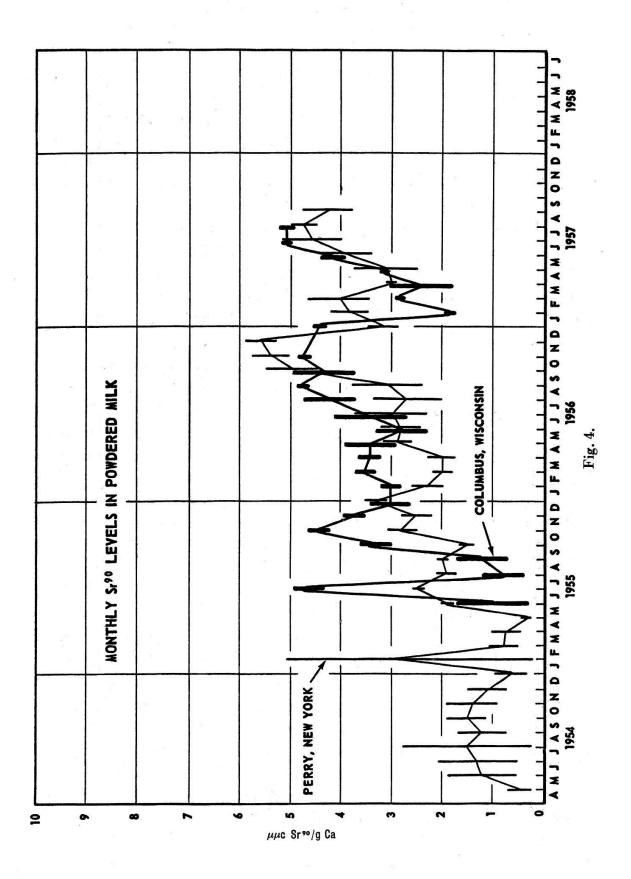
Fig. 6 shows preliminary data on the stratospheric content of Sr<sup>90</sup>. The data are preliminary for the reason that the air filter efficiencies are unknown at the present, although estimated to be something like 25 per cent. The samples are taken by pumping stratospheric air through filters which are then analyzed. It is clear that even though an enormous scatter is present for reasons of time and experiment, it also is clear that there is no large variation in the stratospheric content of Sr<sup>90</sup> between the latitude of 30° S and the Northern Hemisphere. Since most of the megaton yield explosions have occurred in the northern latitudes, though the Pacific testing grounds are only 11° north of the equator, it appears that this evidence argues for rapid north and south mixing in the stratosphere. As we shall see later, other evidence in the dissemination of non-radioactive carbon dioxide derived from the combustion of fossil fuels (31, 32, 33, 34, 35) and of the dissemination of bombderived radioactive C<sup>14</sup> seems to confirm this (36, 37, 38). It is interesting to note also that the actual content of the stratosphere is not in disagreement with the estimates given earlier (4, 5, 6), although the value of the filter efficiencies remains to be settled, and it is estimated at the efficiency of about 25 per cent on evidence assuming homogeneity of the particle size. Experiments are now underway to settle the point.

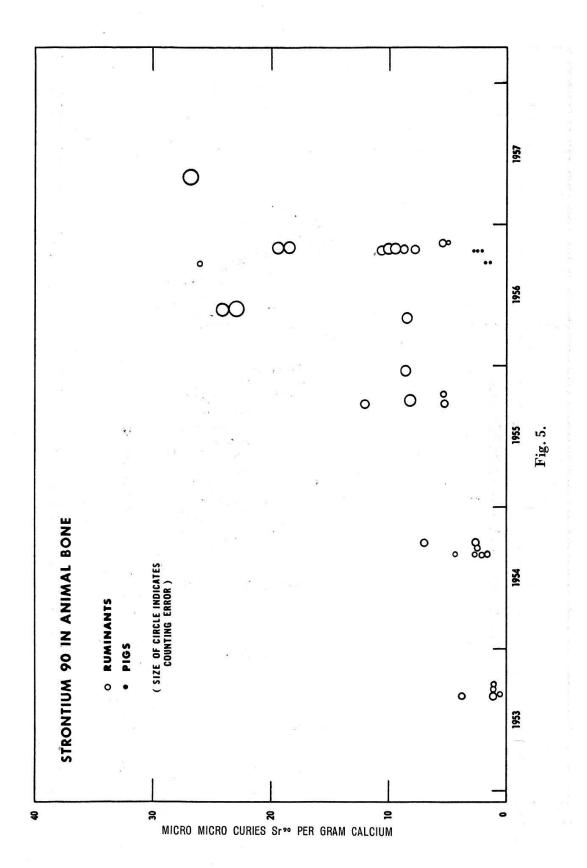
In the model previously advanced (4, 5, 6), it is proposed that material introduced into the stratosphere is mixed immediately horizontally to a uniform concentration and has a residence time of 10 years. Further, it is assumed that the latitudinal spread of tropospheric bomb clouds is only 10° with a sharp step function rather than a normal error curve











distribution. The bomb debris is arbitrarily assigned to the stratosphere except for 1 per cent tropospheric in the case of megaton yields. Local fallout is assumed to be 80 per cent for land surface shots, 20 per cent for surface water shots, and 0 per cent for air shots. All kilo-yield shots are assigned to the troposphere. On these very simple bases we are then, from classified data about the magnitudes and nature of the explosions, able to estimate the total fallout for any place on earth if the deposition from the troposphere is assumed to be proportional to the rain content at a given location. Fig. 7 gives such a theoretical latitudinal fallout profile for world-wide fallout as of December 1957, neglecting rainfall variation, and Fig. 8 is the corresponding world map. Fig. 9 gives the corresponding timewise variations in the northern latitudes and compraes them with the rainfall fallout curves for Milford Haven in England (39). Fig. 10 gives a similar comparison for Chicago and Pittsburgh. Curves for other latitudes are given in Figs. 11 and 12. Fig. 13 gives the estimated stratospheric reservoir and the expected composition in Sr<sup>89</sup> versus time. If a further assumption is made, namely that the proportion of the fallout in a given location is given by the ratio of the rainfall to the world-wide average, 0.77 m (40), it is possible to compare the detailed fallout observed by the pot collection programs in various localities with the theoretical predicted values, and these are given in Table 2.

On the basis of these comparisons and in the absence of conclusive evidence as to the age of radioactive fallout, it appears that the simple theory outlined explains the known information within the experimental error. It may develop when more reliable data are available on the age of fallout through the use of short-lived, 12.8 day half-life barium 140 (Ba<sup>140</sup>) fission product, that a mechanism by which a sort of concentrated leaking from the stratosphere occurs at a latitude of about 40° more may be proved or disproved. At the present time the observed extreme concentration may be explained as being due to coincidence of the tropospheric fallout from the U.S. and Russian tests. If this theory be correct, the Ba<sup>140</sup> content in periods of high fallout will show that the fallout is young. It is to be hoped that these data will be forthcoming soon.

Machta (1, 41) and Stewart, Osmond, Crooks and Fisher (39) have stated that meteorological considerations and likely stratospheric wind patterns, together with evidence that the Sr<sup>89</sup>/Sr<sup>90</sup> ratio of the fallout shows the fallout to be old, have led them to the conclusion that the heavier fallout observed in the 40° to 50° N latitude band is stratospheric and not tropospheric in origin as proposed here. The issue still seems to be unsettled since the radiochemical difficulties of the determination of the Sr<sup>89</sup>/Sr<sup>90</sup> ratio are large and may well have introduced sizeable errors

Table 1
Health and Safety Laboratory Pasture Program

Location		Strontium-90 in hay	00 in hay			Strontium-90 in bone	pone
Location	Site	Sampling date	% Ca ash	μμc/g Ca	Animal	Sampling date	μμc/g Ca
Tifton.	Unimproved	17	3.8	30 + 2	Calves	Fall 1953	3.8
Georgia	Improved	Sept. '54	5.2	3.9 + 0.8	Calves	Sept. '54	7.0 1+0.3
	Improved		7.5	$34 \pm 2$	Calves	Sept. '54	$2.7 \pm 0.2$
	Improved		6.9	$21 \hspace{0.1cm} \pm 1$	Calves	Oct. '55	$12 \pm 0.3$
	,		1	$89.9\pm2.3$	Calves	10-24-56	$19.45 \pm 0.47$
×	Improved	May '56	2.6	$90.8\pm2.3$			$18.36 \pm 0.44$
9	Improved	Sept. '56	5.8	$109.9{\pm}1.7\\129.5{\pm}2.7$			$9.95\pm0.35$ $9.53\pm0.34$
New Brunswick,		9-19-54	0.9	$9.1\pm0.4$	Sheep	Fall 1953	1:1
New Jersey		7-4-55	7.1	85 ±2	Sheep	9-9-54	$2.7\!\pm\!0.2$
		Oct. '55	6.9	77 $\pm 2$	Sheep	10-14-55	$4.1{\pm}0.2$
4 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0		7-3-56	6.3	$88.8{\pm}2.1$	Sheep	10-11-56	$5.63 \pm 0.05$
				$86.9\pm2.1$	*1		$5.5 \pm 0.3$
		10–13–56	0.6	$56.8\pm1.4\ 55.0\pm1.4$	Sheep	10–11–56	7.5 ±0.3
Raleigh,		9-16-54	8.4	26 +0.5	Sheep	Sept. '54	2.1 + 0.2
North Carolina		9-1-55	3.5	± 69	Sheep	- 1	8.6 + 0.4
1000	Pig pasture	7-20-56	12.5	$38.6 \pm 0.3$	Sheep	9-19-56	$26.2\ \pm0.1$
	Pig pasture	8-4-56	10.3	$24.8\!\pm\!0.3$	Pig	9-24-56	$1.87 \pm 0.03$
					Pig	9-24-56	$1.61{\pm}0.04$

2.6 ± 0.2 5.4 ± 0.3 8.8 ± 0.4 7.8 ± 0.3		1.2 4.4 4.4 1.7 1.7 1.7 1.7 1.7 1.7 1.7 1.7		
	10-20-56 10-20-56 10-20-56 10-20-56	Fall 1953 Fall 1953 Sept. '54 Sept. '54 Oct. '55 May '56	11-13-56 3-27-56 May '57	
Sheep Sheep Sheep Lamb	Lamb Hog Hog Hog	Sheep Sheep Sheep Sheep Sheep Sheep	Sheep Calf Sheep	
$0.15\pm0.07 \ 19 \pm0.8 \ 20 \pm1 \ 38.15\pm0.24 \ 15.08\pm0.27$		$10 \pm 0.8$ $6.3 \pm 0.7$ $19 \pm 1$ $8.27 \pm 0.50$ $7.89 \pm 0.53$	$\begin{array}{c} 39 & \pm 1 \\ 27 & \pm 3 \end{array}$	
33 13 12 8.1 10.3		7.0 8.2 7.5 16.2	15 3.7	
9-10-54 6-15-55 9-14-55 6-7-56 8-25-56		9-18-54 9-18-54 7-18-55 6-10-56	June '56 (sileage)	
		Robinson Farm College Farm College Farm College Farm		
Ithaca, New York		Logan, Utah	Mandan, North Dakota	

 $\mathcal{D}^{(1)}(\mathcal{Q}_{\mathcal{G}})^{(2)} = \mathcal{D}^{(1)}(\mathcal{D}^{(2)}(\mathcal{G})) = \mathcal{D}^{(2)}(\mathcal{G}) = \mathcal{D}^{(2)}(\mathcal{G})$ 

Table 2
Sr<sup>90</sup> in Fallout at Monitoring Sites outside Continental United States (High-walled stainless steel pot collections)

	Precipi- tation in inches	Observed mc Sr <sup>90</sup> /mi <sup>2</sup>	from theory mc Sr <sup>90</sup> /mi
Bangkok, Thailand (14° N)	30 5		
March 1957	1.95	0.05	0.085
April 1957	5.85	0.13	0.25
May 1957	1.56	0.037	0.068
June 1957	9.36	0.016	0.41
July 1957	6.63	0.022	0.29
August 1957	11.70	0.039	0.051
Nagasaki, Japan (35° N)			
August 1956	17.43	0.34	0.76
September 1956	16.1	0.17	0.71
October 1956	3.59	0.28	0.16
November 1956	1.44	0.08	0.063
December 1956	1.37	0.22	0.069
January 1957	3.94	1.01	0.172
February 1957	3.28	0.17	0.143
March 1957	1.40	0.38	0.061
April 1957	11.27	1.98	2.4
May 1957	6.44	0.72	1.3
June 1957	10.18	0.27	2.1
July 1957	28.67	1.07	6.0
August 1957	11.35	0.457	2.4
September 1957	14.74	0.260	3.1
Iiroshima, Japan (35° N)			
August 1956	11.93	0.50	0.52
September 1956	9.83		-
October 1956	3.51	0.27	0.15
November 1956	1.64	0.11	0.072
December 1956	0.23	0.06	0.010
January 1957	2.15	0.29	0.097
February 1957	2.26	0.53	0.098
March 1957	1.29	0.23	0.056
April 1957	11.00	1.12	2.3
May 1957	6.44	0.567	1.35
June 1957	10.22	0.493	2.2
July 1957	21.10	0.817	4.4
August 1957	4.48	0.047	0.94
September 1957	10.92	0.277	2.3
Rio de Janeiro, Brazil (23° S)			
September 1956	1.95	0.12	0.085
October 1956	3.12	0.21	0.135
November 1956	3.51	0.06	0.155
December 1956	3.51	0.02	0.155
January 1957	2.73	0.04	0.12
February 1957	5.07	0.05	0.22

Table 2 (Continued)

			Precipi tation in inch		Observed nc Sr <sup>90</sup> /mi <sup>2</sup>	Calculated from theory mc Sr <sup>90</sup> /mi <sup>2</sup>
Salisbury, South Rho	desia (20°	S)				S' 1.00.00
November 1956.			7.41	6)	0.18	0.32
December 1956.			7.80	1	0.12	0.34
			5.85	A125 II S	0.11	0.26
February 1957 .			8.97		0.08	0.04
March 1957			5.46	İ	0.05	0.24
April 1957			1.17		0.04	0.05
Kikuyu, Kenya (0°)				in the second		
January 1957 .	ing Te		9.75		0.14	0.43
February 1957 .			2.34		0.26	0.10
March 1957			3.12		0.03	0.13
April 1957			7.02		0.03	0.31
May 1957	• • • •		14.82		0.138	0.64
June 1957			1.56		0.187	0.068
July 1957			0.08	.	0.148	0.0035
August 1957			0.20		0.020	0.0087
September 1957			2.34	*	0.038	0.10
Dakar, French West			2.01	* - 1		0.20
August 1957	(85)		<b>.</b>		0.532	0.23
			5.2	- 1	0.332	0.25
September 1957			10.44		0.244	0.45
Durban, Union of Sou				areta y nati	0.000	0.015
June 1957			0.39		0.080	0.017
			0.39		0.012	0.017
August 1957			0.78		0.096	0.034
September 1957			4.64		0.230	0.21
Pretoria, Union of Sou	ıth Africa (	(30° S)				
			4.29		0.061	0.187
August 1957			1.56		0.074	0.068
Vienna, Austria (47° I	N)			8		
June 1957			0.78		0.45	0.29
July 1957			5.07		1.95	1.85
August 1957		• . • . • .	2.73	3 301	0.79	1.0
Klagenfurt, Austria (4	17° N)		10 a 15	20.		
August 1957		lete <sub>e</sub> :	3.51	31 3	1.17	1.3
	Precipi-	Obse	rved	Preci	pi- Observ	Calcu-
	tation	AEC	Weather	tatio	- 1 0 111 1	llated tran
	in	Lab.	station	in	Hawa	t hoory
* 1, 11	inches	mc Sr	$\frac{ }{^{90}/\mathrm{mi^2}}$	inch	es	$c Sr^{90}/mi^2$
	η	Inc Di	la ma		m	c or /mi
Oahu, Hawaii (20° N)				V. 2 300		
June 1957	0.32	0.72	_	0.8		ACCES TO THE PROPERTY OF THE PARTY OF THE PA
July 1957	2.10	1.36	0.477	1.6		Country of the Countr
August 1957	1.57	0.303	0.156	3.0	The second second	Maria de la compania del compania del compania de la compania del la compania de
September 1957	1.54	0.274	0.188	0.6	2 0.15	9   0.027

Table 2 (Continued)
Sr<sup>90</sup> in Fallout at other United States Monitoring Sites (High-walled stainless steel pot collections)

Figure of the second	Precipi-		AND THE RESIDENCE OF THE PROPERTY OF THE PROPE	from theory r <sup>90</sup> /mi <sup>2</sup>
Lemont, Illinois (44° N)	tation in inches	mc S <sup>90</sup> /mi <sup>2</sup>	With Russian component	Without Russian component
December 1956	1.26	0.14	0.51	0.053
January 1957	2.06	0.30	0.38	0.088
February 1957	1.77	0.27	0.72	0.076
March 1957	1.98	0.47	0.80	0.085
April 1957	6.09	1.15	2.5	1.3
May 1957	3.21	0.27	1.5	0.68
June 1957	5.94	0.48	1.95	1.0
July 1957	8.98	1.57	3.6	1.9
August 1957	5.36	0.69	2.1	1.1
September 1957	1.08	0.12	0.41	0.21
Birmingham, Alabama (33° N)			With U.S. component	Without U.S component
April 1957	5.41	0.83	1.1	0.23
May 1957	2.96	0.39	0.62	0.13
June 1957	7.70	0.95	1.6	0.33
July 1957	2.62	0.80	0.55	0.11
August 1957	4.19	1.10	0.87	0.37
September 1957	9.59	0.42	2.0	0.41
Salt Lake City, Utah			With	Without
(38° N)	8	850	Russian component	Russian component
December 1956	1.67	0.31	0.66	0.071
January 1957	1.37	0.8	0.54	0.058
February 1957	0.72	0.83	0.29	0.031
March 1957	2.18	2.39	0.87	0.093
April 1957	3.24	2.30	1.4	0.65
May 1957	3.37	0.81	1.5	0.70
June 1957	1.47	1.61	0.66	0.31
July 1957	0.31	0.94	0.13	0.06
West Los Angeles, California	(34° N)	Precipi- tation in inches	Observed* mc Sr <sup>90</sup> /mi <sup>2</sup>	Calculated from theory mc Sr <sup>90</sup> /mi <sup>2</sup>
December 1956		0.49	0.15	0.02
January 1957		3.88	0.99	0.16
February 1957		1.94	0.76	0.08
March 1957		0.95	0.09	0.041

<sup>\*</sup> Some local fallout from Nevada.

Table 2 (Continued)

2		Precipi-	The state of the s	from theory r <sup>90</sup> /mi <sup>2</sup>
West Los Angeles, Califor	rnia (34° N)	tation in inches	Observed* mc Sr <sup>90</sup> /mi <sup>2</sup>	Calculated from theory mc Sr <sup>90</sup> /mi <sup>2</sup>
April 1957	Ţ,	1.33	0.84	0.28
May 1957	Te 1	0.27	0.24	0.056
June 1957	8 5 000	0.06	0.12	0.012
July 1957		0.03	0.92	0.006
South Miami, Florida (26° N)	Precipitation in inches	Observed mc Sr <sup>90</sup> /mi <sup>2</sup>	With U.S. component	Without U.S. component
April 1957	5.04	0.53	1.07	0.22
May 1957	10.11	0.50	2.10	0.44
June 1957	5.82	0.56	1.28	0.27
July 1957 .	8.5	1.51	1.7	0.35
August 1957	13.6	0.75	2.8	0.58
September 1957	6.27	0.52	1.3	0.27

<sup>\*</sup> Some local fallout from Nevada.

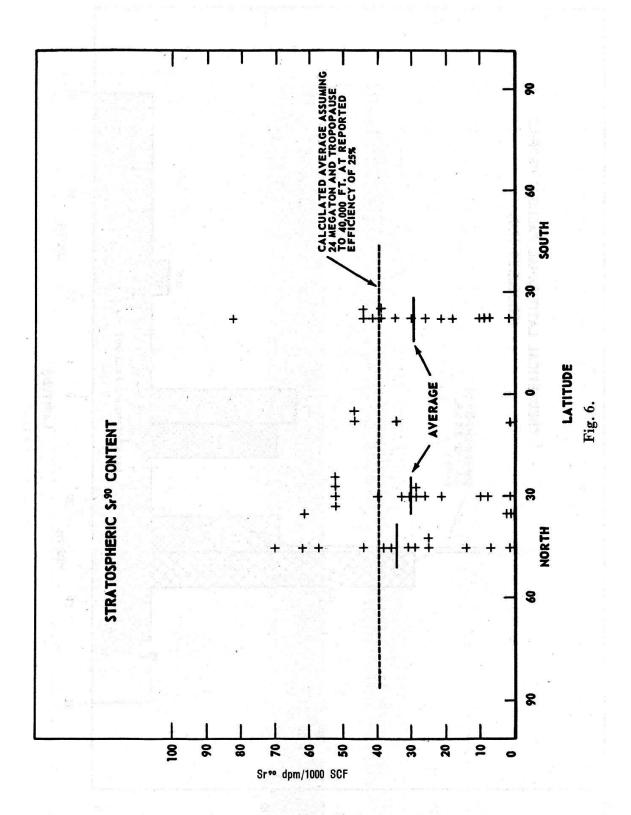
into some of the reported values for this number and since it apparently is possible to account reasonably well for the observed fallout distribution on the present uniform stratospheric fallout theory as shown in the present paper. The critical difference between the two theories is in the matter of the age of the fallout. Better and more significant results probably will be available soon using the Ba<sup>140</sup>/Sr<sup>90</sup> ratio which for both radiochemical and lifetime reasons is more suitable than Sr89/Sr90. Ba140 has a half-life of 12.8 days which is more appropriate to distinguishing between an expected fallout age of perhaps 30 days on the one hand and of about 1 to 2 years on the other, than is the Sr89 half-life of 51 days. The radiochemical procedure for Ba<sup>140</sup> is very similar to that for Sr<sup>90</sup> and both are more sensitive and reliable than the Sr<sup>89</sup> procedure which is particularly susceptible to errors from radioactive impurities such as other fission products which may have been imperfectly separated. Both Ba<sup>140</sup> and Sr<sup>90</sup> are measured by short-lived radioactive daughters of characteristic half-life and which can be repeatedly removed and measured since a new supply is grown into equilibrium each time a separation has been made.

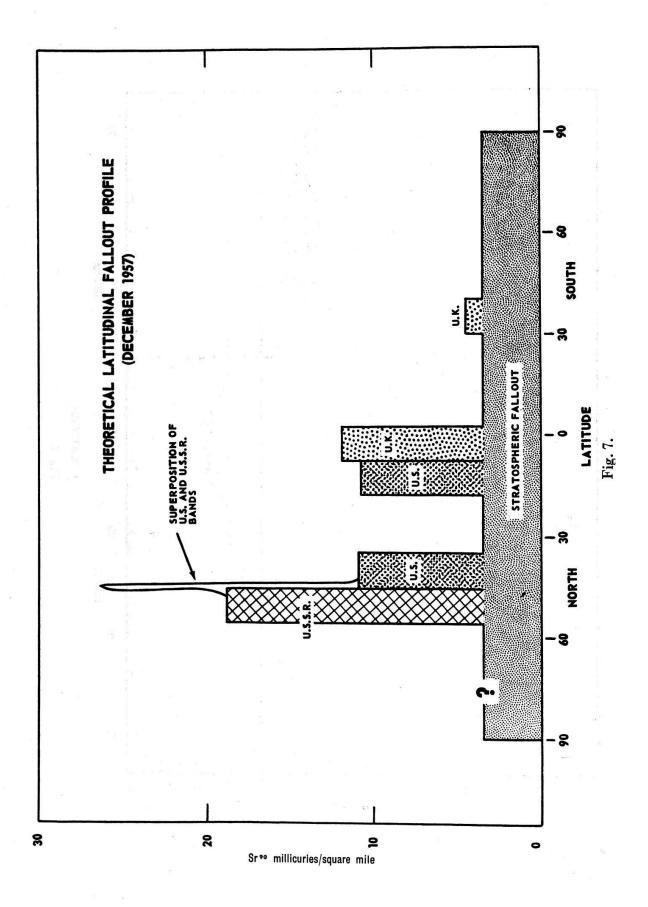
The importance of settling this point is obviously considerable for both meteorology and geophysics and certainly for the understanding of the mechanism of radioactive fallout. Perhaps the Ba<sup>140</sup> data will show the truth to lie somewhere between the two mechanisms.

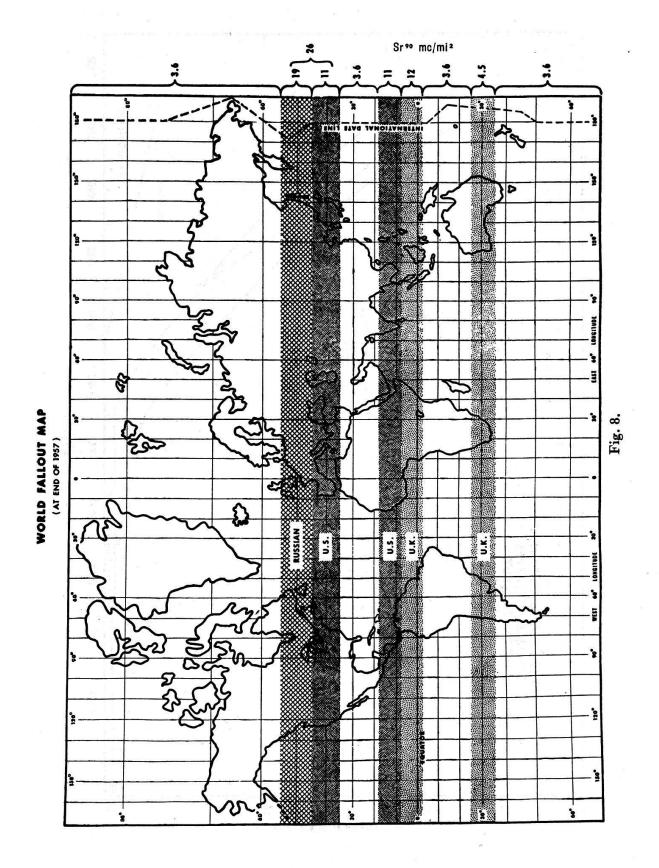
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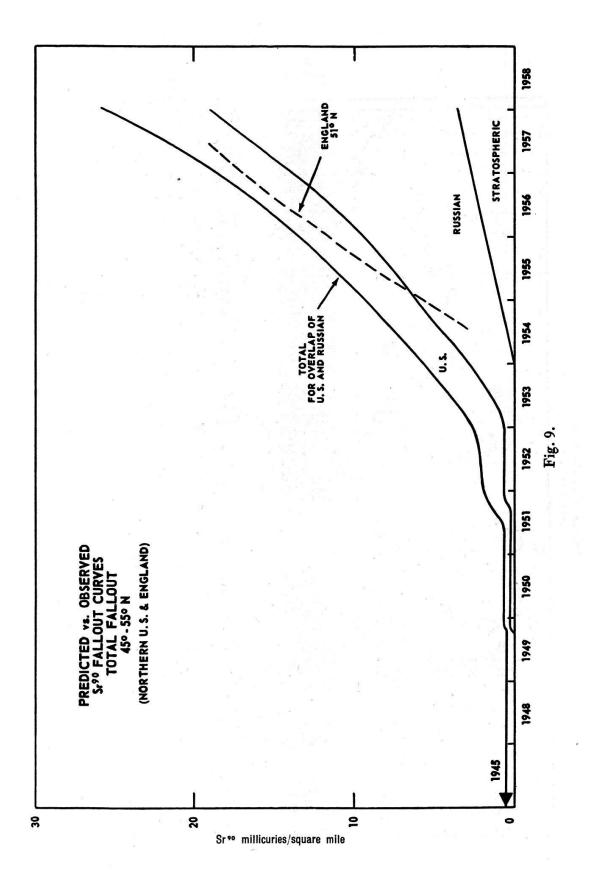
Average Strontium 90 Content in Man – July 1, 1956 to June 30, 1957 Values are in  $\mu\mu c$  Sr/g of Ca, normalized to the whole skeleton (Figures in parentheses give the number of samples in the category)

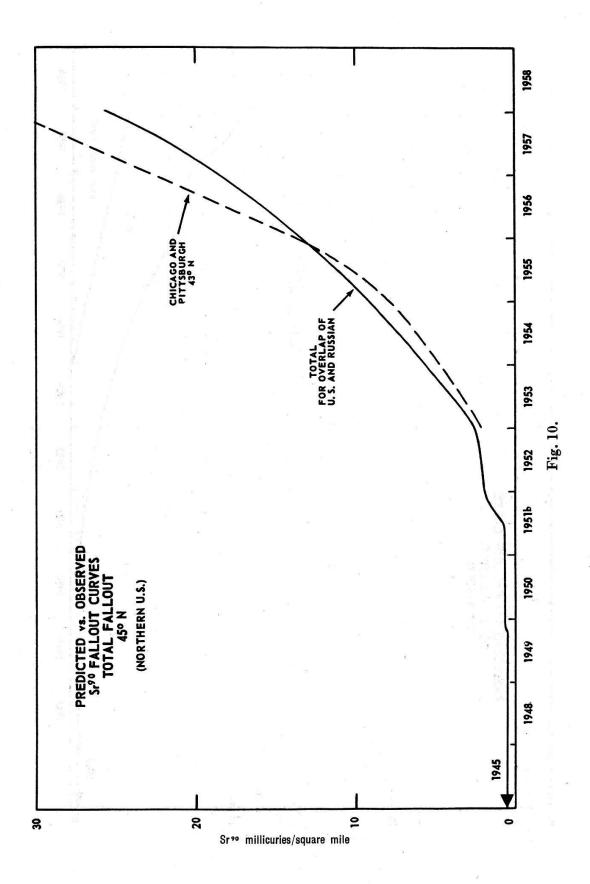
				Age	Age at death (years)	ars)			
Location	0-4	5-9	10–19	20–29	30–39	40-49	50–59	08-09	20-80 (average)
North America	0.67 (30)	0.69 (17)	0.38 (15)	0.07 (14)	0.06 (9)	0.08 (16)	0.05 (5)		0.070 (62)
South America	0.16 (3)	0.20 (1)	0.19 (5)	0.03 (5)	5559	0.03 (2)	0.06 (3)	0.01 (1)	0.034 (13)
Europe	0.65 (2)	0.34 (4)	0.34 (9)	0.06 (20)	0.07	0.04 (6)	0.06 (1)		
Africa			0.06 (2)	0.03 (2)		0.04 (4)			0.035 (9)
Asia	0.93 (1)	0.12 (2)	0.32 (2)	0.06 (8)	0.04	0.12 (8)	0.06 (5)	0.05 (5)	
Australia	0.75 (3)	0.60 (2)			0.03 (3)	0.03 (4)	0.03 (3)		0.030 (10)
Entire world	0.64 (39)	0.57 (26)	0.30 (33)	0.059(49)	0.047(27)	0.070(40)	0.052(17)	0.065(26)	0.060 (159)

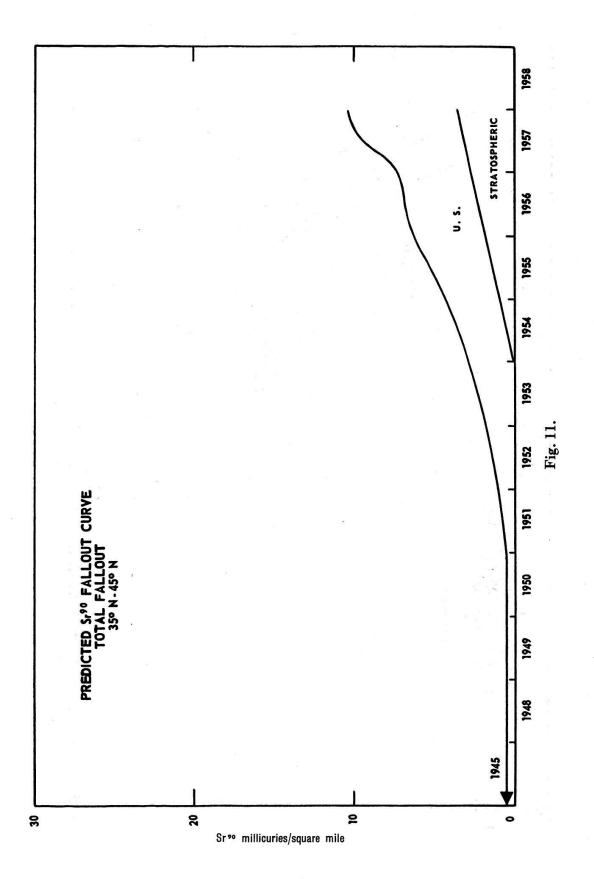


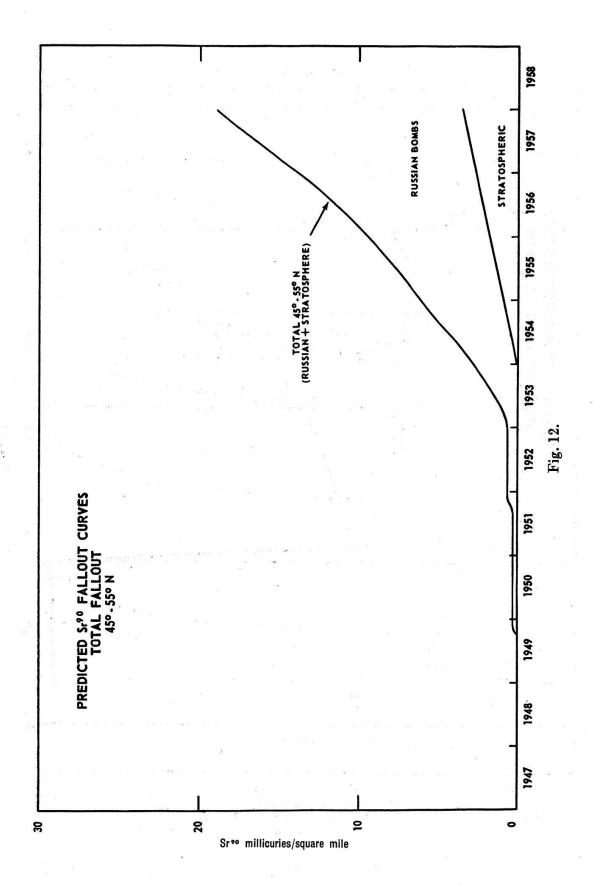


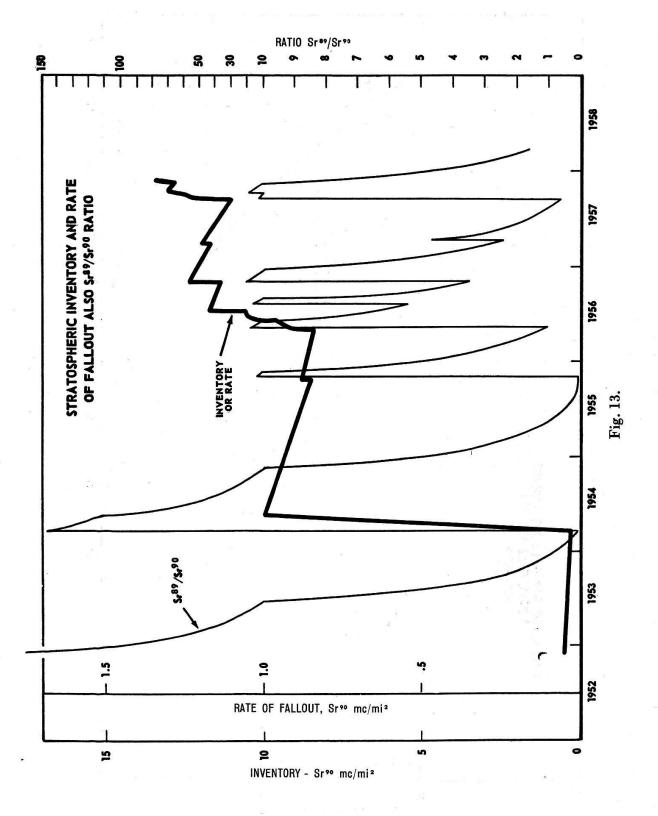












Bomb Carbon 14 (C14)

Rafter (37) and Rafter and Fergusson (36) have shown C<sup>14</sup> increases in surface air at Makara in New Zealand and in New Zealand woods and ocean carbonate as shown in Fig. 14. This additional C<sup>14</sup> is due to bomb generated neutrons which react with air nitrogen to produce it. They find about 2.1 per cent increase per year.

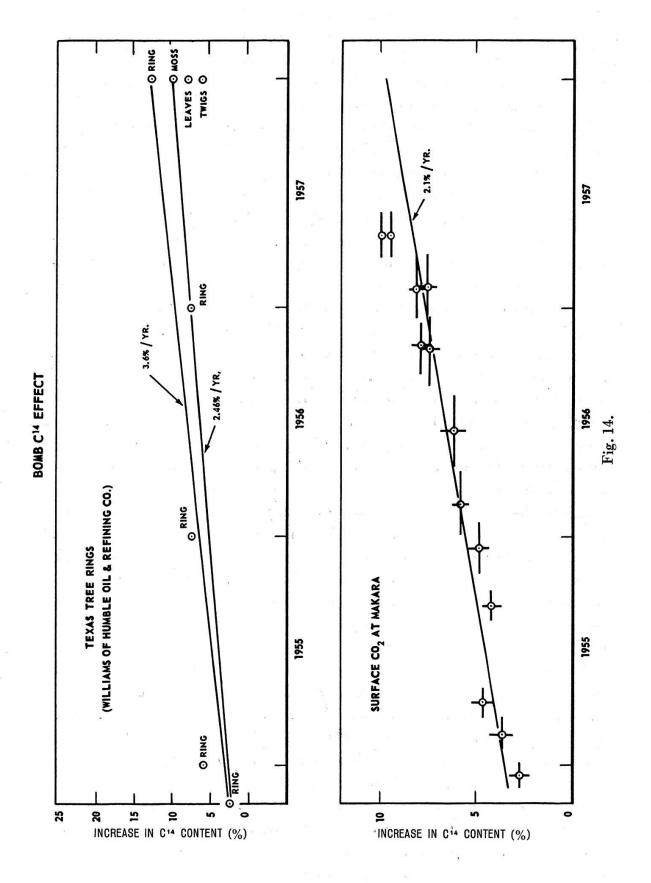
Williams (38) of Humble Oil and Refining Company, finds  $3.0 \pm 9.5$  per cent per year in Texas tree rings (Fig. 14), and de Vries (42) in Holland, and Munnich (43) in Heidelberg, Germany, both report increases. The  $C^{14}$  increase in the flesh of the land snail, helix pomatia, amounted to 4.3 per cent between November 1953 and June 1957 in Holland, while an increase of about 10 per cent during 1955 and 1956 occurred in Heidelberg in various biosphere samples. Patterson and Blifford also find increases (43a).

At a rate of 2.5 neutrons per 200 Mev of energy release, 1 megaton would generate  $3.2 \times 10^{26}$  C<sup>14</sup> atoms. The best estimate, keeping in mind that a substantial amount falls back as calcium carbonate, would be that about  $10^{28}$  C<sup>14</sup> atoms have been introduced into the atmosphere, mostly into the stratosphere. The estimate of 2.5 neutrons per 200 Mev energy released is higher than an earlier estimate based on an assumed 15 per cent escape efficiency (44), the later value being based on firmer information. It also attempts to weigh fusion and fission as they have actually occurred.

About  $9.4 \times 10^{27}$  C<sup>14</sup> atoms are normally present in the stratosphere due to cosmic ray production (45). This figure assumes 22 per cent of the atmosphere to be in the stratosphere. Therefore, with worldwide stratospheric circulation, the rise in the stratosphere should be about 100 per cent as was found in a few measurements made on samples collected in October 1956. Further measurements are in progress.

In the troposphere in the 3 years since the 1954 Castle test at the 10 per cent per year figure used for fallout, about  $3 \times 10^{27}$  C<sup>14</sup> atoms should have descended, or about  $1 \times 10^{27}$  C<sup>14</sup> atoms per year. The average C<sup>14</sup> inventory in the troposphere is  $3.3 \times 10^{28}$  without including the ocean or biosphere, so the observed C<sup>14</sup> rise might be as high as 3 per cent per year as appears to have been observed.

If mixing with the biosphere and top ocean above the thermocline occurred immediately, according to Arnold and Anderson (35) who gave 0.2 g/cm<sup>2</sup> in the top 100 m of the ocean, the total tropospheric reservoir would be  $7.5 \times 10^{28}$  giving an expected rate of increase due to the bombs of 1.3 per cent per year which is in fair agreement with the ob-



servations if we assume the mixing with the ocean and the biosphere, particularly the former, is not quite instantaneous.

The main points are that the ratio of the Northern to Southern Hemisphere effect here is not enormous and fits fairly well with the notion that stratospheric gases have a residence time not too different from that of the ultra-fine world-wide fallout particles.

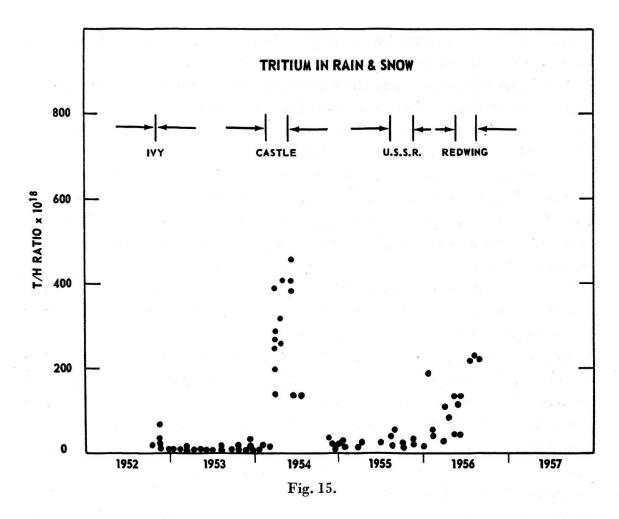
In addition, Fergusson (31) has recently found in studying fossil  $CO_2$  and its effect on reducing the  $C^{14}$  content of the biosphere that the mean life of a  $CO_2$  molecule before being absorbed from the tropospheric air into the oceans and biospheres is perhaps 2 years and that north to south mixing of the fossil  $CO_2$  occurs in less than 2 years.

Consequently, it seems clear that the 10-year residence time for stratospheric gases before descent into the troposphere seems to fit data for C<sup>14</sup> from bombs as well as the Sr<sup>90</sup> and Cs<sup>137</sup> fallout data.

Fig. 15 gives up-to-date data on the occurrence of tritium in rainwater in the Chicago area (21, 46, 47). It is clear that whereas Sr<sup>90</sup> and probably C<sup>14</sup> remain in the stratosphere for years, the tritium from highyield thermonuclear detonations does not, but descends in a matter of 1 or 2 months. This most probably is due to the enormous mass of water carried into the stratosphere by the fireballs of detonations in the moist tropospheric air. The characteristic white mushroom cloud is evidence of the formation of ice crystals in the cold stratospheric air, which if large enough to be seen in this way must certainly be large enough to fall into the troposphere where they melt and join in the ordinary phenomena; i.e. fall out as rain or snow. Thus a large fractionation relative to fission products and radioactive carbon dioxide occurs. Of course, there probably is some entrainment of fission products on the surfaces of the falling ice crystals by the Greenfield-Brownian motion accretion mechanism. In fact, it is known that about 1 per cent of megaton yield offsite fallout occurs in the early banded tropospheric manner. This may be due to this entrainment and thus one would expect that the latitudinal distributions of early tropospheric fallout of both fission products and tritium water from megaton yield bombs fired in the troposphere (11) should be identical. No satisfactory data are now available to check this point. In the calculations in this paper the Figure of 1 per cent for tropospheric contribution from megaton yields has been used.

### 3. Conclusion

The more recent data, particularly on bomb C<sup>14</sup>, when taken together with the earlier data on bomb fission products and tritium, give us some confidence in our present understanding of the fallout mechanism. All



of these observations and considerations afford unprecedented opportunities for the study of meteorology and geophysics, particularly in an international cooperative effort such as the International Geophysical Year.

### Summary

The whole world is concerned over the question of radioactive fallout, particularly that from the testing of weapons. Attention has been focused on the problems associated with the effects of radiation, and this field of knowledge, formerly known only to a limited group of scientists, is becoming a matter of general concern discussed by millions of people. If the effects of radiation and the magnitude of the doses from fallout were more widely known and understood, there would be considerably less apprehension. This paper is intended as a contribution to the general understanding of the subject.

During the recent Nevada test operation of the U.S. Atomic Energy Commission and the U.S. Department of Defense, Operation Plumbbob, a bomb was fired underground and completely contained. Essentially no radioactive fallout escaped. This is a very important result because it means not only that bombs can be tested without fallout, but that since nuclear explosions can be contained new peaceful applications become possible. Several peaceful applications are discussed briefly. Incidentally, by studying the shock waves from nuclear explosions, seismologists are able to learn about the structure of the earth with a clarity not previously possible.

The theory of the transport and deposition of fallout is reviewed. It is shown that the simple model discussed permits reasonably good calculations of the fallout to be expected in either hemisphere. Account is taken of the effect of annual rainfall. A brief review is given of the biological hazard from radioactive fallout.

Data presented in earlier publications are brought up to date so that one may see the most recent results for rainfall collections, for the strontium-90 content of milk, for animal bone, and for human bone. It is particularly interesting to note that the data continue to show the principal features noted previously and that little that is new in principle has appeared.

Information with respect to the carbon-14 present as carbon dioxide in the atmosphere and tritium present as moisture in the atmosphere is presented and it is shown that this information corroborates the theory of the transport and deposition of strontium-90. From the study of fall-out and of tritium and carbon-14 in the air, scientific information regarding the circulation of the atmosphere can be obtained, and this information is of great interest to meteorologists.

All of these observations and considerations afford unprecedented opportunities for the study of meteorology and geophysics, particularly in an international cooperative effort such as the International Geophysical Year.

# Zusammenfassung

Die ganze Welt ist über die Frage des radioaktiven Niederschlages besorgt, insbesondere über denjenigen der Kernwaffenversuche. Die Aufmerksamkeit vieler Leute wurde auf Probleme gelenkt, die mit den Auswirkungen von Strahlen zusammenhängen. Dieses Gebiet der Wissenschaften, mit dem früher nur wenige Forscher vertraut waren, wird mehr und mehr eine Angelegenheit von allgemeinem Interesse, über die Millionen diskutieren. Die Furcht könnte beträchtlich vermindert werden, wenn die Wirkungen der Strahlung und die Größen der Dosen des radioaktiven Niederschlages in weiteren Kreisen bekannt und verstanden würden. Diese Mitteilung soll einen Beitrag zum allgemeinen Verständnis des Problems leisten.

Im Rahmen eines Versuches der US Atomic Energy Commission und des US Department of Defense «Operation Plumbbob» in Nevada wurde eine Bombe unterirdisch zur Explosion gebracht und vollständig eingeschlossen. Kein wesentlicher Niederschlag konnte entweichen. Dies ist ein wichtiges Resultat; es bedeutet nicht nur, daß Bomben ohne radioaktiven Niederschlag getestet werden können, sondern auch, daß sich dabei neue Möglichkeiten friedlicher Anwendungen eröffnen. Verschiedene Beispiele werden kurz diskutiert. So können Seismologen durch das Studium der Schockwellen der Explosion Auskünfte über das Erdinnere in bis jetzt unbekanntem Ausmaß erhalten.

Die Theorie des Transportes und der Ablagerung von Spaltprodukten wird dargelegt. Es wird gezeigt, daß mit einem einfachen Modell einigermaßen exakte Berechnungen des zu erwartenden Niederschlages in beiden Hemisphären möglich sind. Die Wirkung des jährlichen Regenfalls wird einbezogen. Eine kurze Übersicht über die biologischen Auswirkungen des radioaktiven Niederschlages wird gegeben.

Angaben aus früheren Publikationen werden auf den heutigen Stand gebracht, so daß die Resultate über die Aktivität des Regens, des Strontium-90-Gehaltes der Milch und von Tier und Menschenknochen ersichtlich sind. Es ist sehr interessant festzustellen, daß diese Angaben die schon früher bemerkten Eigenschaften weiterhin zeigen und daß nur wenig prinzipiell Neues dazugekommen ist.

Auskünfte über den Gehalt an C<sup>14</sup> in der Kohlensäure der Atmosphäre und über den Tritiumgehalt der Luftfeuchtigkeit werden gegeben. Es wird gezeigt, daß diese Angaben die Theorie des Transportes und der Ablagerung von Strontium-90 bestätigen. Aus Studien über den Niederschlag von Fissionsprodukten und von Tritium und Kohlenstoff-14 in der Luft können Feststellungen über die Bewegungen in der Atmosphäre gemacht werden, die für Meteorologen von großem Interesse sind.

All diese Beobachtungen und Überlegungen stellen bis jetzt nicht dagewesene Gelegenheiten zum Studium der Meteorologie und der Geophysik dar, speziell im Rahmen eines internationalen Gemeinschaftswerkes wie dasjenige des Geophysikalischen Jahres.

#### Résumé

Le monde entier s'occupe aujourd'hui de la question des retombées radioactives, en particulier de celles qui suivent l'expérimentation d'engins nucléaires militaires. L'attention générale s'est concentrée sur les problèmes concernant l'effet des radiations, et ces questions qui autrefois ne préoccupaient qu'un nombre restreint d'hommes de science sont discutées aujourd'hui par des millions de personnes. Toutefois, si les

effets des radiations et la grandeur des doses des précipitations radioactives étaient mieux connus et compris, l'appréhension générale serait beaucoup moins grande. Le but de cette publication est de faire connaître à un public plus étendu la valeur de ces problèmes.

Au cours des essais faits au Nevada dernièrement par la Commission de l'Energie Atomique des Etats-Unis d'Amérique et par le Département de la Défense des Etats-Unis, sous le nom d'Opération Plumbbob, une bombe a été mise à feu sous terre et a pu être complètement retenue sans qu'on ait pu retrouver l'échappement d'une précipitation radioactive. Ce fait est très important, car il montre non seulement que des bombes radioactives peuvent être expérimentées sans retombée radioactive subséquente, mais encore que de nouvelles applications pacifiques sont possibles, puisque les explosions nucléaires peuvent être complètement retenues sous terre. L'auteur passe en revue toute une série d'applications pacifiques possibles, entr'autres, par l'étude des ondes de choc dues à l'explosion nucléaire, la possibilité pour les séismologistes d'étudier la structure du globe terrestre avec une clarté jusque là inégalée.

L'auteur expose ensuite les données théoriques du transport des poussières radioactives et de leur sédimentation. A l'aide d'un modèle très simple, il est possible de calculer avec une précision satisfaisante les retombées radioactives probables dans les deux hémisphères, en tenant compte également des effets des pluies annuelles. Suit une brève revue des altérations biologiques dues aux poussières radioactives. Les faits présentés dans des publications antérieures sont mis à jour, et il est possible de lire dans ce travail les renseignements les plus récents sur les pluies radioactives, la teneur en strontium-90 du lait, des os chez les animaux et chez l'homme. Il est très intéressant de constater que les données précédemment connues ont pu être vérifiées et peu de nouvelles données ont été mises à jour.

L'auteur présente ensuite les connaissances actuelles concernant le carbone-14, présent sous forme de dioxyde de carbone dans l'atmosphère, et le tritium, sous forme d'humidité de l'air, et les faits dévoilés confirment ce que l'on savait déjà du transport et de la fixation du strontium-90. En examinant les retombées radioactives et la présence de tritium et de carbone-14 dans l'atmosphère, l'on peut approfondir nos connaissances sur la circulation atmosphérique, ce qui est d'une grande importance pour les météorologistes. Toute cette étude donne des occasions inespérées de compléter nos connaissances en météorologie et en géophysique, et témoigne de l'effort de coopération internationale entrepris dans l'Année Géophysique Internationale.

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