Fallout from 1957 and 1958 Nuclear Test Series

New York City data show contributions from short-lived nuclides for as long as 14 months after testing.

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The unique production of radiotungsten in Operation Hardtack and the moratorium on testing after the autumn of 1958 have made fallout measurements during the past few years very useful in assessing some of the more perplexing aspects of offsite fallout distribution. Several comprehensive summaries and interpretive articles on ground-level contamination during this period (*1–5*) and new observations on the atmospheric behavior of nuclear debris have been published (*6–8*). As a result, many questions pertaining to fallout are now answerable. However, the emphasis historically placed on strontium-90 and cesium-137 and the practical difficulties involved in large-scale surveillance of large numbers of nuclides have resulted in a serious lack of detailed information on many of the shorter-lived contaminants that are dispersed by nuclear testing.

This article deals with some of the causes and effects of high concentrations of shorter-lived fission products in fallout in New York City during 1958 and 1959. Data previously reported (*9*) are supplemented with data on concentrations of strontium-90, cesium-137, ruthenium-106, cerium-144, strontium-89, zirconium-95, and wolfram-185, measured in monthly fallout collections during 1959. Casual examination shows that the shorter-lived fission products predominated over Sr^{90} and Cs^{137} from the beginning of 1958 through the middle of 1959. This was due primarily to the heavy rate of testing that prevailed during 1957 and 1958, but further interpretation of the measurements indicates that the conditions under which individual test series were conducted during this period also had an effect. Through analysis of isotope ratios, W^{185} concentrations, and monthly rainfall volumes, it has been established (i) that more fallout arrived in New York City from the Soviet series in October 1958 than from earlier series, and (ii) that the Soviet debris was richer in short-lived nuclides because it was deposited sooner after its production.

The New York City measurements also provide a means of investigating external doses delivered to the population from photon-emitting fission products. Theoretical gamma-radiation dose rates and integral doses are computed from the reported amounts of Zr^{95}, Ce^{144}, Ru^{106}, and Cs^{137} that accumulated on the ground during 1958 and 1959. These calculations show that fallout made substantial contributions to open-field dose rates, and that the shorter-lived nuclides, particularly Zr^{95}, produced doses comparable to doses of Cs^{137}. Since the New York City observations probably are applicable to other sites in the Northern Hemisphere, a more thorough evaluation of the world-wide effects of the shorter-lived nuclides is indicated.

Methods

Throughout 1959, replicate monthly fallout samples were taken on the roof of the Atomic Energy Commission's Health and Safety Laboratory in New York City with funnel-shaped-ion-exchange collectors (*9, 10*). At the end of each exposure the paper pulp and resin were removed, ashed at 450°C, and separated into aliquots for determination of gross beta activity. Tungsten, cesium, strontium, cerium, and zirconium fractions were then sequentially separated, purified, and counted for beta radiation. Disintegration rates were calculated by correcting the observed counts for counter efficiency and background, recovery, self-absorption, build-up, and decay (*11*). The counting factors for the separated activities were obtained by counting the beta radiation of known quantities of the individual nuclides under the counting conditions for the sample. Self-absorption and efficiency factors for the mixed beta activities were approximated by using potassium chloride as a secondary standard.

When sufficiently high levels of activity were indicated by the initial gross beta-radiation assay, at least two samples from the month's collections were analyzed for Ru^{106} by gamma spectrometry. The determination was based on the intensity of the Rh^{106} emission peak at 0.51 Mev. The detection efficiency of the scintillator was calculated from the combined Zr^{95}–Nb^{95} peak at 0.76 Mev, with the radiochemically determined Zr^{95} concentration as a standard. Niobium-95 was assumed to be in transient equilibrium with Zr^{95}, at a daughter-to-parent activity ratio of 2.4. The correction factor was then related to the rhodium measurement through the data of Heath (*12*) on peak- to total-emission ratios and total absolute crystal efficiencies. All final determinations were made after the samples had been stored for at least 120 days to minimize interference from Ru^{103} emissions at 0.49 Mev.

The ruthenium estimates were confirmed by beta-absorption analyses in which the 3.53-Mev Rh^{106} beta component was resolved from the total counting rate of the sample and corrected for counter efficiency (*13*). In addition, periodic beta-decay measurements were made, from immediately after the sampling period through the end of 1959. Mixed longer-lived nuclides were identified by the decay slope apparent from the later counts and extrapolated back to the original counting date (*14*). Approximate Ru^{106} levels were then obtained by subtracting the measured concentrations of Ce^{144}, Ce^{137}, and Sr^{90}.

Gamma-radiation doses delivered to a point in air 3 feet above the ground were derived by adapting the method of Hallden and Harley for mixed fission products (*15*) to specific

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Table 1. Activity levels measured in New York City during 1959. Isotopic activities are reported as of the end of the sampling month. Total beta activity estimates are reported as of the counting date (in parentheses).

Sampling month	Activity levels (mc/mi²)								
	Sr^{90}	Cs^{137}	Ru^{106}	Ce^{144}	Zr^{95}	Sr^{89}	W^{185}	Gross activity	
Jan.	1.00	1.60		30.9	81.7	36.3	22.4	241	(6 Feb. 59)
Feb.	1.81	3.36		49.0	88.6	31.3	16.9	229	(23 Mar. 59)
Mar.	4.45	6.58	48	86.0	90.0	61.1	18.4	356	(29 Apr. 59)
Apr.	4.16	6.93	56	83.1	79.4	39.7	9.84	413	(5 May 59)
May	1.52	1.90	13	19.1	11.3	8.03	4.69	75.7	(9 June 59)
June	2.43	3.46		31.3	15.7	8.01	3.05	97.8	(13 July 59)
July	0.52	0.88		5.69	2.56	≤0.78	0.94	21.0	(10 Aug. 59)
Aug.	0.65	0.88		7.24	2.23	≤0.49	≤0.50	23.3	(8 Sept. 59)
Sept.	0.11	0.20		1.19	1.01	≤0.21	≤0.20	4.06	(12 Oct. 59)
Oct.	0.39	0.61		2.70	≤0.33	≤0.36	≤0.14	8.30	(24 Nov. 59)
Nov.	0.34	0.51		1.64	≤0.24	≤0.17	≤0.12	5.96	(15 Dec. 59)
Dec.	0.52	0.43		2.53	≤0.17	≤0.16	≤0.068	7.54	(13 Jan. 60)
Total deposition	17.9	27.3		320	419	221	83.8	1480	
Activity level on 31 Dec. 1959	36.1	48.5		275	28.1	5.63	9.18	542	

nuclides. Emission energies, decay constants, and other pertinent decay-scheme characteristics were taken from the listings of Strominger, Hollander, and Seaborg (*16*). The radiation sources are assumed to be evenly distributed over an infinite plane, and no corrections are made for weathering, shielding, Compton scattering, or the effects of the ground-to-air interface.

The beta-radiation counters used were standard end-window Geiger-Müller tubes surrounded by anti-coincidence rings and shielded with mercury. The gama-radiation instrument was a 3- by 2-inch sodium iodide scintillator equipped with a transistorized single-channel analyzer (*17*).

Monthly fallout activity levels measured during 1959 are listed in Table 1. Each value is the mean of at least two determinations. The Zr^{95}, Sr^{89}, and W^{185} concentrations were considered undetectable after August, June, and July, respectively, when the counting error, expressed as one standard deviation, exceeded the apparent counting rate. In other cases, the relative percentage of uncertainty due to counting factors averaged 3 percent and ranged from 2.1 to 12 percent of the activities reported. The total depositions were obtained by summing the monthly levels and correcting for decay during the sampling period (*9*). The activity levels shown to exist at the end of 1959 include totals previously reported for the end of 1958, corrected for decay through the end of 1959. The cumulative gross activity estimate for 1958 was obtained by assuming a mean production date of 30 June 1958 and correcting for decay by the $T^{-1.2}$ law (*18*). The decay corrections for the 1959 monthly increments are the results of actual measurements described in the ruthenium analyses.

Since the counters used were not sensitive enough to detect the less energetic beta emitters, the gross activity estimates are probably low, and more representative of activities with energies in excess of 0.3 Mev. However, these levels do show that at least 5.1 (*9*) and 1.5 curies, respectively, of beta activity per square mile were deposited in New York City during 1958 and 1959. The predominance of shorter-lived nuclides noted in 1958 continued through the first half of 1959, with the result that the effects of radioactive decay reduced the two years' total deposition by a factor of 10 by the end of 1959.

Age and Origin of Debris

It is generally conceded that delayed fallout is primarily of stratospheric origin and that the mean atmospheric residence time of weapon-test debris is from one to three years, depending on testing conditions (*5, 6*). It follows that the possible major sources of the New York City fallout reported in Table 1 are limited to Soviet, United Kingdom, and United States test series conducted during 1957 and 1958. Although the sensitivity of debris dating methods is lessened by consideration of monthly collections rather than individual rainfalls, the production of W^{185} in the United States Hardtack series and the cessation of testing after the Soviet series of October 1958 make it possible to identify contributions to total fallout from these sources with some degree of accuracy.

From estimated radiotungsten yields for the Hardtack series (*6, 8*) and measured fission-product yields (atoms per fission) for thermonuclear weapons in general (*19*), the Sr^{90}/W^{185} and Sr^{89}/W^{105} ratios at an assumed mean production date of 1 June 1958 are estimated to be 0.00380 and 0.436, respectively. Monthly depositions of Sr^{90} in New York City from Hardtack are calculated directly from measured W^{185} levels by extrapolation of the Sr^{90}/W^{185} production ratio to the reporting dates for the fallout measurements. Levels of Sr^{90} from other sources are obtained by subtracting the Hardtack Sr^{90} and similarly determined Sr^{89} fractions from the total isotopic concentrations and analyzing the Sr^{89}/Sr^{90} ratios calculated for the non-Hardtack debris.

The interpretation of these ratios is illustrated in Fig. 1. The clearest indication of debris age occurs in the 1959 ratios, which uniformly show the Soviet series of October 1958 to be the predominant source. A second trend indicates contributions from late 1957 and early 1958 testing in samples taken before June 1958. The increases in the ratio observed during the summer months of 1958 are attributed to the arrival in New York City of mixed debris. The compositions of these mixtures are determined by extrapolating the ratio curve for the earlier measurements through the middle of 1959 and

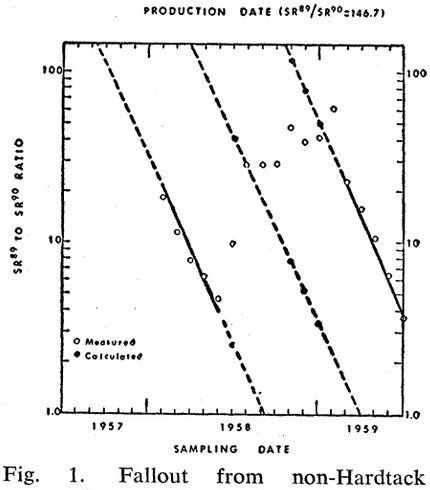


Fig. 1. Fallout from non-Hardtack sources in New York City during 1958 and 1959; Sr^{89}/Sr^{90} ratios, corrected for calculated Hardtack concentrations, are used. Most of the 1958 data are taken from the report of Welford and Collins (*9*). Strontium-89 values for the first three months of 1958 are taken from the data summaries of Hardy *et al.* (*2*).

the 1959 curve back to October 1958. Points falling in between are then resolved algebraically into two values (black dots in Fig. 1) that coincide with the extended curve.

Calculated contributions to total Sr^{90} fallout from Soviet testing in the fall of 1958, from Operation Hardtack, and from earlier series are listed in Table 2. Since the W^{185} levels fell below the detection limits of the collection system used (*10*), it is not possible to trace Hardtack activities in samples taken after July 1959. An additional difficulty for this period is the possibility that there was fallout from U.S. high-altitude devices detonated during the summer of 1958; this has been inferred by Gustafson (*1*) and others from the detection of Rh^{102} in air and ground measurements. However, it is clear that, prior to July, rates of fallout from the Soviet series were as much as ten times as high as rates of fallout from Hardtack, and that the Soviet series delivered a total of at least four times more Sr^{90} to the New York City area.

Air Activity Concentrations

The rough proportionality that exists between fallout level and amount of rainfall permits the use of the specific activity of rainfall as a relative index of concentrations of activity in air. Thus, with knowledge of the sources of debris and monthly precipitation totals, it is theoretically possible to follow the fluctuations in air levels that resulted in the Sr^{90} depositions listed in Table 2. With the additional assumption that the atmospheric dispersion of other fission products is not radically different from that of Sr^{90}, the build-up and depletion of concentrations of total Hardtack and Soviet debris in the air over New York City during 1958 and 1959 may be traced. Moreover, it is not unreasonable to expect material from possible future polar and equatorial detonations, carried out under the same conditions, to arrive in similar fashion.

Table 2. Sources of Sr^{90} fallout in New York City during 1958 and 1959.

Sampling month	Sr^{90} activity level (mc/mi²)		
	Oct. 1958–Apr. 1959	Operation Hardtack	Soviet, Oct. 1958
May 1958	3.44	0.006	
June 1958	1.23	0.039	
July 1958	1.25	0.224	
Aug. 1958	0.358	0.195	
Sept. 1958	0.378	0.197	
Oct. 1958	0.596	0.227	0.347
Nov. 1958	0.450	0.320	0.420
Dec. 1958	0.119	0.179	0.474
Jan. 1959		0.521	0.479
Feb. 1959		0.512	1.30
Mar. 1959		0.707	3.74
Apr. 1959		0.492	3.67
May 1959		0.313	1.21
June 1959		0.260	2.17
July 1959		0.104	0.416

Figure 2 compares the fractions of total fission-product yields from Hardtack and total yields from the Soviet October 1958 series found in unit monthly rainfall volumes in New York City after May 1958. The values are derived from the Sr^{90} data in Table 2; an average Sr^{90} yield of 4.0 atoms per 100 fissions for the two series was used (*19*). Total TNT megaton equivalents for the two events are taken from the reports of Libby (*8*) and Martell (*6*). A mean production date of 18 October 1958 for the Soviet series is calculated from the Sr^{89}/Sr^{90} ratios illustrated in Fig. 1. Rainfall volumes are taken from monthly weather summaries distributed by the United States Department of Commerce (*20*).

The curves show that both series produced significant levels of specific activity and, by inference, significant concentrations of activity in air in New York City as early as 30 days and as late as a year after their mean detonation dates. Although it is not possible to follow the long-term effects, it appears that the major contributions from the Soviet series occurred over a span of about 150 days, while the dissipation of concentrations from Hardtack required about 250 days. In addition, the Soviet activity build-up was higher by roughly a factor of 10 and occurred about 100 days sooner relative to the date of production than did the Hardtack build-up. The break in the Hardtack curve suggests a decline in tropospheric activities after 90 days, followed by the delayed arrival of stratospheric debris after 150 days. As a result of the difference in timing, there was appreciably more shorter-lived material in the fission fragments deposited from the Soviet series.

Table 3. Integrated gamma-radiation doses delivered by principal gamma-emitting pairs in fallout in New York City during 1958 and 1959. The 30-year dose from the estimated total Ru^{106}–Rh^{106} deposition during 1958 is given in parentheses.

Sampling quarter	30-year gamma dose (millirads)			
	Cs^{137}–Ba^{137}	Zr^{95}–Nb^{95}	Ru^{106}–Rh^{106}	Ce^{144}–Pr^{144}
Jan.–Mar. 1958	3.21	4.10		0.105
Apr.–June 1958	4.36	4.42		0.205
Jul.–Sept. 1958	2.93	2.08	(1.17)	0.0862
Oct.–Dec. 1958	3.23	7.39		0.166
Jan.–Mar. 1959	7.20	6.24	1.20	0.284
Apr.–June 1959	7.67	2.05	1.24	0.217
July–Sept. 1959	1.22	0.0925	0.178	0.0230
Oct.–Dec. 1959	0.967	0.0	0.125	0.0115
Total 30-year dose	30.8	26.4	3.91	1.10
Total 70-year dose	47.6	26.4	3.91	1.10
Total infinity dose	56.7	26.4	3.91	1.10

Gamma Dose

From theoretical gamma- to beta-fission-product ratios (*15*), the total deposition of fallout in New York City during 1958 and 1959 is estimated to have produced from one-half to one-third as many gamma photons as beta particles. Calculated contributions of cumulative levels of representative photon-emitting pairs to total infinite-plane gamma dose rates from fallout are illustrated in Fig. 3. All values are corrected for monthly decay. The Ru^{106} levels for January and February and for June through December 1959 were obtained from the Ce^{144} measurements by extrapolating the decrease observed in the Ce^{144}/Ru^{106} ratio during March, April, and May. The estimate is believed to be fairly consistent with actual ruthenium levels, since the slope of the decrease agrees with that theoretically calculated from the decay constants of the two nuclides. The estimate for gross 1958 ruthenium deposition is obtained by assuming a Ru^{106}/Ce^{144} activity production ratio of 0.71 (*19*) and a mean production date of 30 June 1958, as in the approximation for total beta activity.

Although the dose rates shown for the shorter-lived photon-emitting systems are sustained through only a few half-lives of the parent activity, significant integral doses are produced. Table 3 lists the genetically important 30-year gamma doses calculated for

quarterly isotopic depositions during 1958 and 1959. These contributions are summed to give the total dose that would be delivered if this debris were undisturbed until 1989. These levels are in turn compared with lifetime or 70-year doses and infinity doses.

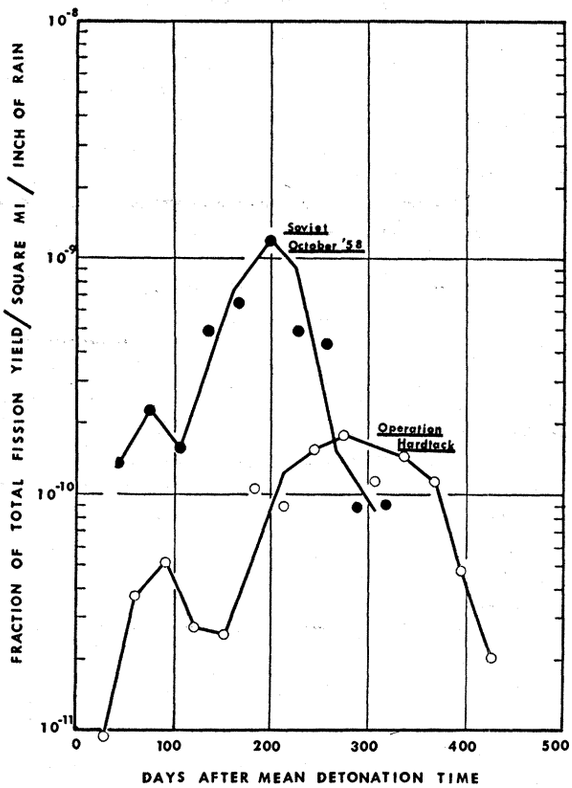
In relation to ionization-chamber measurements of dose rates from cosmic and natural terrestrial sources made by Solon and his associates (*21*), the infinite-plane dose rates from fallout in New York City reached as high as 50 percent of total background rates. Suggested correction factors for absorption produced by vertical displacement of fallout activity, Compton scattering of gamma radiation, and partial shielding by topological and architectural structures range from one-fifth to one-tenth of the uncorrected calculated doses (*3, 5*). Even with the maximum correction, however, dose rates for Zr^{95}–Nb^{95} fallout alone were between 0.2 and 0.4 microrad per hour for most of the 2-year observation period. Moreover, both the instantaneous and the long-term doses produced by the shorter-lived nuclides were of at least the same order of magnitude as those produced by the long-lived Cs^{137}–Ba^{137} chain.

Discussion

As is well known, the use of measured nuclide ratios to study meteorological factors controlling fallout is subject to serious error. In addition, the effects of rapidly changing factors, such as the air mass trajectories and the Ba^{140} concentrations which Martell (*6*) has studied, are largely obscured in monthly sampling. Nevertheless, the calculations for Hardtack and for Soviet contributions to total fallout in New York City are in fair agreement with Martell's observations on New England rains, with Gustafson's Cs^{137} analyses in Chicago (*1*), and with Lockhart's earlier measurements of Sr^{90} concentrations in air over Washington, D.C. (*7*). The extension of isotopic data to total fallout is admittedly limited by such unknown factors as fractionation of volatile fission products in the fission process, resuspension of deposited debris, variation of actual yields from theoretical values, and the assignment of single apparent production dates to series of tests. These difficulties are illustrated by the anomalous Sr^{89}/Sr^{90} ratios shown for the August and September 1958 samples in Fig. 1 and the uniform divergence of Ce^{144}/Sr^{90} and Zr^{95}/Sr^{90} ratio curves calculated for 1959 from those obtained from theoretical decay constants. Precise knowledge undoubtedly awaits more exhaustive study, but that there is more sustained and more diffuse fallout from the equatorial tests as compared with the polar series appears to be established.

Generalization of these findings to future testing is limited because the peak depositions of Soviet debris are attributable to seasonal effects as well as to latitudinal considerations. Absolute assay of either factor could only be achieved if one factor were held constant—that is, if Hardtack and the Soviet series had both occurred near the poles but at different times, or if they had been conducted simultaneously at different latitudes.

The significance of the data in terms of radiation dose levels is more clearly discernible. The calculated contributions of the fallout that accumulated on the ground in New York City during 1958 and 1959 to external-gamma-radiation dose rates and to long-term doses are illustrated in Fig. 3 and Table 3, respectively. The values are in agreement with the results of gamma-radiation spectrometric analyses of soils



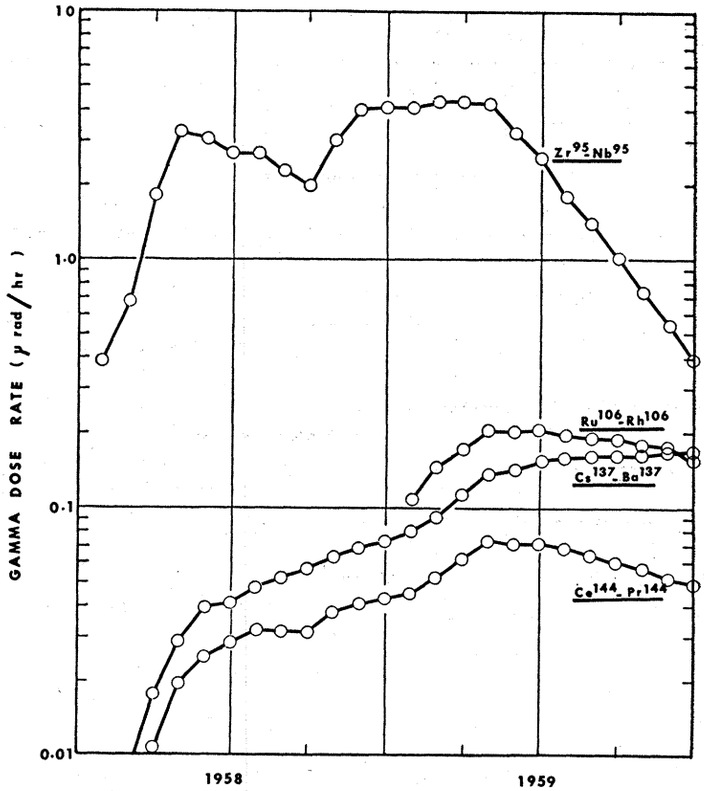


Fig. 2 (left). Comparison of the arrival patterns in New York City of debris from polar (Soviet) and from equatorial (Hardtack) test series conducted during 1958. Fig. 3 (right). Calculated infinite-plane dose rates from cumulative levels of gamma activities measured in fallout in New York City during 1958 and 1959.

from similar latitudes in both the United States (*1*, *4*) and the United Kingdom (*3*). The soil levels of Zr^{95}–Nb^{95}, which are less affected by the 1957 and earlier fallout than are levels of the longer-lived nuclides, show a maximum variation of 20 percent from the New York City cumulative fallout levels. In addition, the results of systematic ionization-chamber measurements of open-field dose rates performed by Vennart (*3*) in Belmont, Surrey, England, closely parallel the Zr^{95}–Nb^{95} dose rate curve of Fig. 3. These observations imply (i) that weathering and roughness of terrain had little effect on doses from fallout activities during the two-year period, and (ii) that the most significant contribution to total-radiation dose rates was made by radioactive zirconium, or possibly by other fission products of comparably short half-life.

Although the beta emitters in fallout are not sufficiently energetic to cause significant external doses except through direct deposition on body surfaces, internal doses do occur through inhalation and ingestion of debris particles (*5*). In addition, the chemical similarity of some of the radionuclides to elements normally assimilated by the body results in concentrated doses to specific tissues. The over-all effect of radiation on the population cannot be known, therefore, until many complex meteorological, physical, chemical, and biological factors are thoroughly understood. It is clear, however, that any valid assessment of the effect of radiation, past or future, must include consideration of fallout and more realistic treatment of the short-lived fission products.

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Conferences on Science and World Affairs

Statements by participants at the seventh and eighth conferences, which met at Stowe, Vermont, in September.

Seventh Conference

The Seventh Conference on Science and World Affairs was held at Stowe, Vermont, 5–9 September 1961. Forty-one scientists from 12 countries attended (see box, page 987).

This conference had as its theme "International Cooperation in Pure and Applied Science." Our previous conferences have been chiefly concerned with ways of preventing the misuse of science in the wholesale destruction of mankind. In this conference at Stowe, we have turned to the discussion of constructive international cooperation in science, because it is a way to create trust between nations, a trust which develops from common interests and from experience in working together.

Science misused by nations to foster their competitive interests as world powers makes possible the destruction of mankind. Science used cooperatively by all nations for the increase of human knowledge and the improvement of man's productive capacity can give all men on earth a satisfactory and worthwhile life. Scientists bear a responsibility both to foster the constructive use of science and to help in preventing its destructive use.

The deliberations of the conference were carried out in plenary sessions and in meetings of working groups. These groups were six in number, as follows: (i) Cooperation in the Earth Sciences; (ii) Cooperation in Space Research; (iii) Cooperation in the Life Sciences; (iv) Cooperation in the Physical Sciences; (v) Cooperation in Assistance to Developing Nations; (vi) Exchange of Scientists and Scientific Information.

Similar suggestions for cooperative research activities arose independently from different working groups. This is reflected in several places in this statement. This is a welcome indication of the essential unity in science. The discussions were carried on in a spirit of friendly cooperation, and full agreement was reached by the entire conference on the suggestions that will be enumerated in the following paragraphs.