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CURRENT PROBLEMS IN RESEARCH

Movement of Radiostrontium in Soils

Knowledge of climatic and soil factors is important in determining the redistribution of strontium fallout.

C. W. Thornthwaite, J. R. Mather, J. K. Nakamura

The presence of radioactive strontium-90 in the atmosphere and in the soil and waters of the earth is one of the most compelling facts faced by our present generation. Injected into the atmosphere through the testing of nuclear weapons, dispersed to all parts of the globe through the latitudinal movement of air, and returned to our earth mainly by precipitation, strontium-90 is one of the most hazardous of all of the radioactive isotopes because of its long half-life and its similarity to calcium as a bone seeker in the human body. Probably the greatest danger from this radioisotope results from our own lack of information concerning how it reacts in the soil and waters of the earth, and in the food cycle, what the maximum permissible tolerance levels are, and what remedial measures may be taken to prevent the accumulation of dangerous concentrations in soil, air, plants, animals, and human beings. As part of a general effort to learn more about radioactive strontium, the Laboratory of Climatology in Centerton, New Jersey, has begun a long-term research study, supported by the United States Air Force, relating to the redistribution of strontium in the surface layers in the soil (*1*).

Water plays a key role in the continuing relationship between strontium-90 and man, and thus it is through an understanding of the influence of water on the redistribution of strontium that we can begin to determine the nature of this relationship. There are two main ways in which water influences the movement of strontium; precipitation is the principal agent in the removal of strontium from the atmosphere, and the gravitational or surplus water in the soil is the principal agent in the redistribution of strontium with depth in the soil. While the first of these influences is of importance in determining the geographic pattern of fallout, its study is beyond the scope of the present article. This article serves, rather, to sum up some of the results of our work on the second part of the water-strontium problem. Because of our own special study of the moisture factor in climate, our approach to the problem has been through determination of the quantity and seasonal distribution of water surplus at a given place by the climatic water-balance bookkeeping technique presented by Thornthwaite in 1948 (*2*) and revised by Thornthwaite and Mather in 1955 (*3*).

Climatic Water Balance

The relative moistness or aridity of a given place can be determined from a comparison of the moisture supply (the precipitation) with the climatic need for water (the potential evapotranspiration). Potential evapotranspiration is the water flux into the atmosphere from a vegetation-covered surface whose soil is supplied at all times with sufficient moisture for all the needs of the vegetation. When precipitation exactly corresponds to the climatic need for water there is no moisture surplus or deficiency and the climate is neither moist nor dry. Such correspondence between precipitation and potential evapotranspiration occurs only infrequently in nature and then only for short periods of time. Normally, the precipitation is either greater or less than the need for water. When the precipitation is greater than the need for water and the soil is at its water-holding capacity, there will be a water surplus equal to the excess of precipitation over the water need. When water need exceeds precipitation, there is a water deficit which equals the unfulfilled moisture requirement of the vegetation—that part not supplied by precipitation or supplied from storage of moisture in the soil. The water surplus and water deficit at any time and place can be determined from a simple water-balance bookkeeping procedure. These elements of the water balance and their timing are basic factors in pedogenesis. The water surplus moves mainly downward through the soil and is the leaching solution. Thus, it is the water surplus which is responsible for the redistribution of strontium in the soil.

Mathematical Model of Strontium Movement

A number of investigators (*4*, *5*) have made actual physical measurements of the rate of movement of strontium and other radioisotopes through soils. The recent report by Miller and Reitemeier (*4*) provides a great wealth of new material on the problem of movement of strontium in soil. The authors, working with tubes of soil in a laboratory, used five different types of soils (Norfolk fine sandy loam, Hagerstown silt loam, Miami silt loam, Fort Collins silt loam, and Huntley clay loam), three different types of leaching

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solution (0.005*N* $CaCl_2$, 0.005*N* NaCl, and deionized water), and two rates of application (30 inches and 300 inches) in an effort to determine the movement of strontium-89. The source of the radioactivity was a ½-inch layer (70 grams) of soil spiked with 5 microcuries of strontium-89. This soil was placed in a layer on top of 5-inch columns of the soil in tubes and covered by another ½-inch layer of uncontaminated soil. The results showed clearly that some downward movement of strontium occurred and that there were differences from soil to soil. For a given amount of leach water added to the surface of the soil in the tubes, there was a greater amount of movement of strontium in the sandy soil than in the silt loams and the clay loam. This result was to be expected on the basis of the ion-exchange capacities of the different soils.

The experiments of Miller and Reitemeier and others show that there is an organized movement of strontium in the soil responding to the various soil and water factors in such a way that a certain percentage of the original concentration of strontium moves downward to the next lower layer in the soil with every unit of water added. The amount of the downward movement depends on the cation-exchange capacity and *p*H of the soil and the leaching efficiency of the water solution.

In order to describe the measured distributions found by Miller and Reitemeier, a mathematical model was set up. Paralleling the experimental procedure, the soil was divided into ½-inch layers or zones, and the strontium concentrated in the first or source layer was followed downward to the ½-inch zones below. The movement of strontium was conveniently divided into cycles, each cycle being complete when one-tenth of the strontium in each zone had moved downward to the next zone. Other fractions could have been used just as well. A cycle is completed when the selected percentage of the distribution of strontium has moved downward from each zone into the next lower zone.

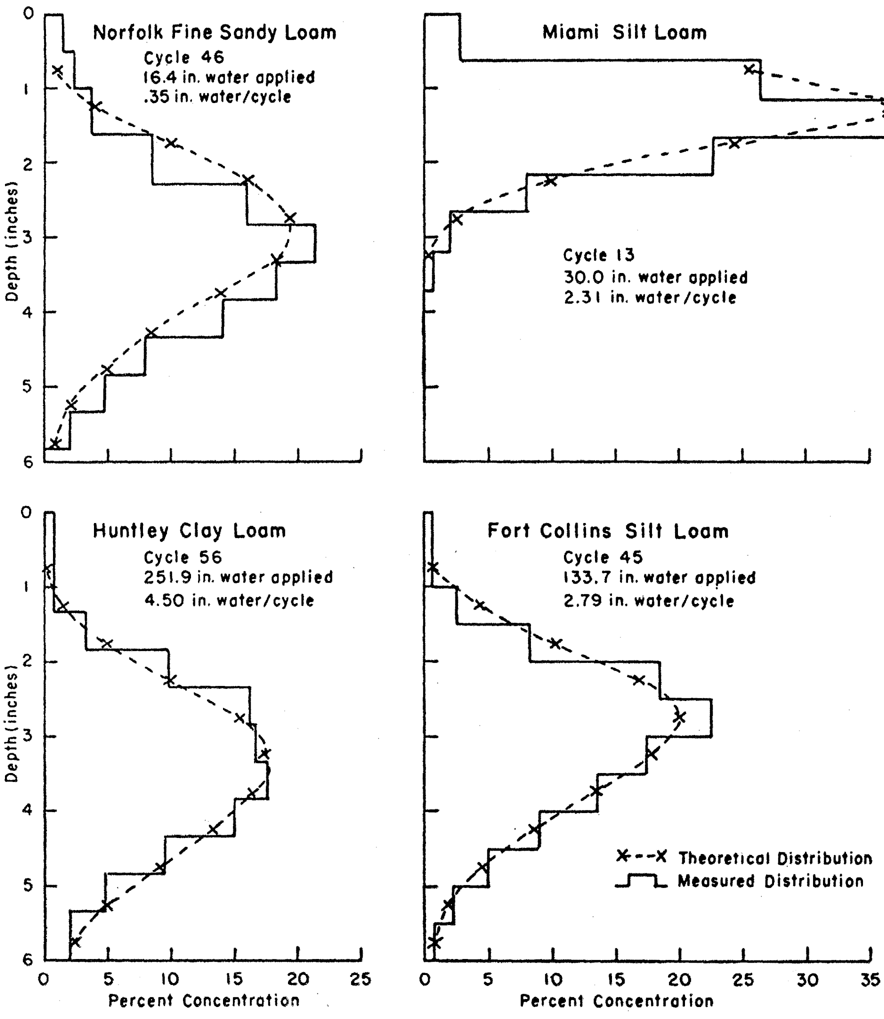

Fig. 1. Comparison between measured and theoretical strontium concentration with depth after leaching. [Miller and Reitemeier (*4*)]

Expressed mathematically, with an initial concentration X_0, the concentration of material with depth in the soil after *t* complete cycles of movement is:

$$X_n = \frac{t!}{(t-n)!\,n!}\,k^n(1-k)^{(t-n)}\,X_0$$

where X_n is the concentration in the *n*th layer, *n* is the receiving layer, *t* is the cycle of leaching, *k* is the fractional loss per cycle, and X_0 is the initial concentration of strontium-90.

The concentration in the source layer after *t* cycles of leaching (X^{n}_{0}) is expressed simply as

$$X^{n}_{0} = (1-k)^t\,X_0.$$

The number of cycles required for X_n to reach a maximum in the *n*th layer is given by

$$t = n/k.$$

There is a definite maximum concentration of the quantity X_n in any given layer. This value is reached when the amount of movement downward out of the given layer just equals the amount received from overlaying layers.

In their recent paper Miller and Reitemeier measured the concentration of strontium with depth after leaching of different soils with three different leaching solutions—deionized water, 0.005*N* sodium chloride, and 0.005*N* calcium chloride. The results for four soils leached with 0.005*N* calcium chloride are given in Fig. 1. The vertical distributions (in percentages) of an initial concentration of strontium, computed by use of the above equation, are also shown in Fig. 1. The number of leaching cycles, the quantity of water applied, and the ratio between these two quantities are given in Fig. 1. As can be seen, there is close agreement between the measured distribution of strontium with depth and the calculated distribution. In the four examples in Fig. 1, the soil type, the cation-exchange capacity of the soil, and the amount of water used for leaching differed. Similar comparisons were made of the results for the four soils after leaching with deionized water and 0.005*N* sodium chloride. Without exception there was good agreement between the distribution given by the mathematical model and that actually found by Miller and Reitemeier.

The mathematical model has, in effect, established particular leaching patterns for the various solutions. The time necessary for the establishment of such a pattern for the solutions used is dependent upon the following factors: (i) the cation-exchange capacity (CEC) of the soil; (ii) the *p*H of the soil; (iii) the leaching efficiency of the solution; and (iv) the volume of the leaching solution applied.

The general relation showing the influence of the cation-exchange capacity and the *p*H of the soil on the leaching efficiency of the solution is given in the equation

$$L = L_1 \left(\frac{7.0}{p\mathrm{H}}\right)^{-b} C^{-a},$$

in which L is the leaching efficiency in cycles per unit of applied water; L_1 is the leaching efficiency at unit cation-exchange capacity (at *p*H 7.0); C is the cation-exchange capacity in milliequivalents per 100 grams; a is the exponential cation-exchange capacity correction factor; and b is the exponential *p*H correction factor.

Since

$$L = t/P,$$

where t is the number of cycles of leaching and P is the amount of water applied, then

$$t = PL_1 \left(\frac{7.0}{p\mathrm{H}}\right)^{-b} C^{-a}$$

The following is a summary of the values of the elements in the above equations.

	L_1	a	b
$CaCl_2$	11.3	1.128	0
NaCl	0.13	0.35	0
H_2O	0.124	0.53	1.6

The results are shown in Fig. 2. Since the *p*H of the soil exerts no measurable influence on the leaching efficiency of calcium chloride and sodium chloride solutions, the graph merely shows the relation between leaching efficiency and cation-exchange capacity. The rather large effect of *p*H on the leaching efficiency of water is indicated by a displacement of the observed points on the graph, shown by the arrows. A comparison of the measured and computed values of leaching efficiency is given in Table 1.

The curves for calcium chloride and sodium chloride intersect at a cation-exchange capacity of approximately 300. It is entirely reasonable to assume that for equivalent concentrations of the various chlorides found between Na^+ and Ca^{++} in the activity series, proportional curves could be drawn between the two shown, with the value of cation-exchange capacity of 300 the focus of the chloride curves. It is also not unreasonable to theorize that the same cations with another anion, for example SO_4^{--} or OH^-, would intersect at another specific cation-exchange capacity.

Applicability to Natural Soils

The mathematical model of movement downward through the soil represents the distribution of strontium in a column of soil in the laboratory. This does not answer the practical question of whether the model will also represent the distribution with depth and time of strontium in different natural soils *in situ* after leaching with natural rain water. Because of the influence of factors of the natural environment—the presence of a natural vegetation cover and the resulting uptake of strontium, the alternate wetting and drying of natural soils due to variable daily weather conditions, the presence of earthworms, the occurrence of natural weathering, and the chemical composition of natural precipitation—it is important to test the mathematical model against actual field measurements of strontium concentration with depth.

The Health and Safety Laboratory of the New York Operations Office of the Atomic Energy Commission has undertaken a soil-sampling program in an effort to determine the amount of fallout which occurs (*6*). In addition, it has measured monthly and daily fallout by means of pot and gummed-film collectors. While these data are not sufficient to provide a complete understanding of the movement of strontium with depth, they do provide a rough check on the applicability of the mathematical model.

The distribution of strontium in the soil in New York City as determined by soil sampling for the period 1955 through 1958 was compared with the computed values. The results are summarized in Table 2.

The observed values for strontium were obtained by radiochemical analysis of soil samples taken at La Guardia Airport in October of 1955, 1956, and 1957 and in Central Park in 1958, while the computed values were obtained from

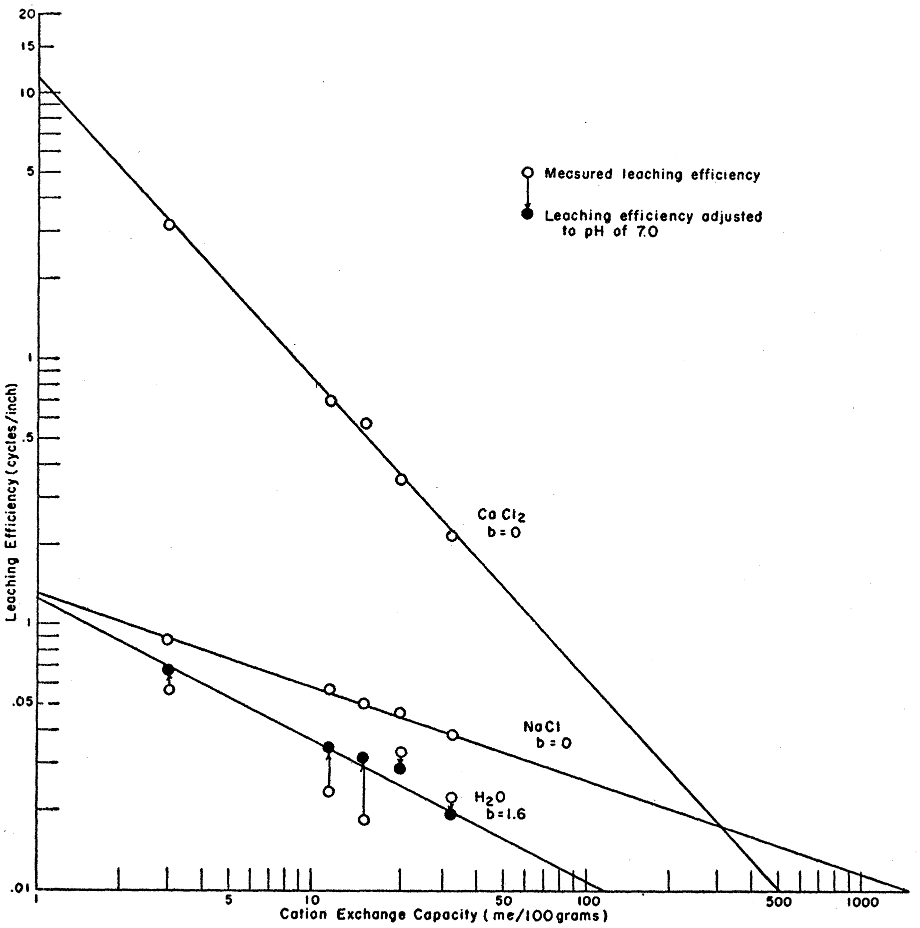

Fig. 2. Relation between various solutions and cation-exchange capacity.

Table 1. Comparison of the measured and computed values of leaching efficiency for the three leaching solutions.

Soil	*p*H	CEC (meq/100 gm)	H_2O *L* (cycle/in.)		NaCl *L* (cycle/in.)		$CaCl_2$ *L* (cycle/in.)	
			Meas.	Comp.	Meas.	Comp.	Meas.	Comp.
Norfolk	6.3	3.0	0.057	0.058	0.087	0.088	3.125	3.271
Hagerstown	5.5	11.5	.023	.023	.057	.055	0.690	0.718
Miami	5.0	15.7	.018	.017	.050	.050	.568	.505
Fort Collins	7.7	20.1	.033	.029	.047	.047	.351	.382
Huntley	7.8	32.6	.023	.023	.038	.038	.213	.222

Table 2. Observed and computed distributions of strontium-90 in the soil in New York City (in millicuries per square mile).

Soil depth (in.)	1955		1956		1957		1958	
	Obs.	Comp.	Obs.	Comp.	Obs.	Comp.	Obs.	Comp.
0 to 2	5.75	8.63	11.15	12.35	18.92	16.29	16.25	15.43
2 to 6			13.00	11.75	7.33	14.14		
2 to 9							20.30	27.68
6 to 12					7.60	10.72		
Below 9								15.24
Below 12						2.97		
Total	5.75	8.63	24.15	24.10	33.85	44.12	36.55	58.35

the monthly values of fallout by applying the mathematical model. In these computations, a leaching efficiency of five cycles of leaching for each 100 millimeters of water was found to apply for movement in the top 2-inch layer, while a leaching efficiency of one cycle for each 12 millimeters of surplus water worked well on the soil layers below 2 inches. For the period 1952 to 1954 the fallout was estimated from gummed-film data, and between 1954 and 1957 it was measured by means of a pot collector. The estimated and measured values of fallout and the computed values of water surplus for each year since 1952 are given in Table 3.

Many difficulties are encountered in trying to utilize the mathematical model to compute the distribution of strontium with depth in the soil: Measured soil values of strontium content do not agree well with accumulated values of fallout, there are errors in radiochemical analyses of the soil samples, and soil samples themselves are not always representative of the local area. These and other sources of error exist and can contribute to a lack of close agreement between the measured and the computed values of distribution with depth. An error in sampling or analysis is seen in the value for observed strontium concentration in the 2-to-6-inch layer in New York City (Table 2). While the concentration in the 0- to 2-inch layer increases from 5.75 to 11.15 and then to 18.92 millicuries per square mile, the value for strontium concentration in the layer below goes up from 0 (unmeasured but assumed to be close to 0) to 13.00 and then down to 7.33 millicuries per square mile. With the continued rapid rise in the concentration in the 0-to-2-inch layer, the decrease in concentration of almost 50 percent in the 2-to-6-inch layer from 1956 to 1957 can only be explained on the basis of error in sampling or analysis.

It is significant that the measured strontium-90 detention in the surface layer, which had been increasing steadily up to 1957, decreased in 1958. The leaching of strontium out of the upper 2-inch layer of soil must have exceeded the annual total of fallout during 1958. The large amount of leaching in 1958 is closely tied to the fluctuations in water surplus during the past 3 years, given in Table 3. Because of the weather in 1956 and 1957, with comparatively little water surplus available in 1957, the downward movement of strontium was reduced, and there was a somewhat greater detention in the upper 2 inches of the soil. Thus, in 1958, when a larger water surplus occurred, the rate of the flux increased markedly because of the increased supply of water for leaching and the already existing large detention of strontium in the upper soil layers.

Of the two main factors which influence the movement of strontium-90 through the soil, the cation-exchange capacity and the water surplus, it is clear that the water surplus is by far the more variable quantity from month to month and from season to season. Thus, on a long-term basis, the water surplus is the principal agent affecting the flux or the detention of strontium in the soil at a given place.

The annual water surplus (Table 3) shows considerable variation from 1952 to 1958. The number of leaching cycles corresponding to the water surplus varies correspondingly from year to year. The year 1955, for instance, had almost twice as many leaching cycles as the year 1956. The year 1958 had half again as many cycles as 1957. Because of the importance of leaching cycles in the movement of strontium, it is easier to think of the influence of the climate of a year in terms of leaching cycles than in terms of precipitation, water surplus, or other more common climatic parameters.

Table 3. Strontium fallout, water surplus, and leaching cycles in New York City, 1952–58.

Year	Estimated (*E*) and measured (*M*) fallout (mc/mi^2)	Computed water surplus (mm)	Leaching cycles (0 to 2 in.)
1952	1.91 *E*	612	31
1953	3.54 *E*	924	46
1954	6.56 *E*	637	32
1955	9.22 *M*	955	48
1956	10.92 *M*	589	27
1957	11.97 *M*	556	28
1958	14.26 *M*	797	40

Summary and Conclusions

From the results of laboratory experiments it was possible to develop a mathematical model of movement in the soil which showed that a certain percentage of the strontium moved downward to the next lower layer of the soil for each unit volume of surplus water which is added to the layer. The model was verified by laboratory experiments and by actual field observations of strontium content in the soil.

During the development of the mathematical model it became clear that a number of factors would be important in any attempt to apply the model to variable field conditions. Among these factors are (i) the cation-exchange capacity of the soil both areally and with depth; (ii) the quantity of surplus water available for leaching; (iii) the initial concentration of strontium in the soil and the increments to the soil due to fallout; (iv) the chemical composition of the rain water and the soil solution; and (v) the leaching efficiency of various solutions containing cations and anions.

Before the mathematical model can actually be applied to the problem of determining the distribution of strontium with depth at a given place, precise information on these factors is needed. While information on the water surplus at any time or place is relatively easy to obtain from the climatic water balance, and information on fallout is available from those stations at which pot collectors are being operated, and in highly generalized form elsewhere, information on some of the other quantities is not as readily available. For instance, even though soil samples are taken by the Health and Safety Laboratory of the New York Operations Office of the AEC at 17 different places in the United States, these are limited to one sample a year at a site and generally to only two layers (0 to 2 inches and 2 to 9 inches) in the soil. We do not know much about the seasonal or monthly variation in soil concentration of strontium or in its real variation with depth.

Information on the cation-exchange capacity of many different soils is available in the literature, but these data have never been assembled in a map, while information on the variation of cation-exchange capacity with depth is not at all readily available. Detailed information on the chemical composition of rain water and of soil solutions is practically nonexistent.

The future course of this study and its possible practical application will depend on the course of world events in the next several years. If weapon testing should be discontinued, either through international agreement or through unilateral decision, and if no further strontium-90 burden is added to the present stratospheric reservoir, fallout will soon reach very small levels and contamination of the soil will end. The strontium-90 in the soil will then serve as a tracer which will be of unparalleled value in studies of leaching, soil development, and ground-water hydrology.

If weapon testing should be resumed, or if a nuclear war should break out and soils all over the earth should be seriously contaminated, the natural processes of soil purification through leaching would proceed at different rates in different areas. After only a few years some soils would be decontaminated to a point where they would produce food suitable for human consumption. Soils in other areas would remain contaminated for many years and presumably would have to remain under quarantine. New sets of land values would develop, and a wholesale redistribution of any remaining population would occur. As the work begun under this research study continues, it will yield a body of principles which will apply regardless of the direction of future events.

References and Notes

1. This research was supported in whole or in part by the U.S. Air Force under contract No. AF49(638)-409, monitored by the Air Force Office of Scientific Research of the Air Research and Development Command. A more complete treatment of certain sections of the study will be found in C. W. Thornthwaite and J. R. Mather, "Investigations of the climatic and hydrologic factors affecting the redistribution of strontium-90 in the soil," in *Publications in Climatology* (Laboratory of Climatology, Centerton, N.J., 1959), vol. 12, No. 2, pp. 49–91.
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Science and Foreign Affairs

Recent scientific and technical advances have had a dramatic effect on international relations.

G. B. Kistiakowsky

My theme is the impact of science and technology on foreign policy. Let me try to single out, if I can, what I believe to be the important aspects of this relation between science and world affairs.

I need not devote much space here to demonstrating the proposition that the advances of science gradually, sometimes suddenly, are altering the relations between nations and peoples. Of course, it is the technology which is the carrier of change, but it is basic science, the acquisition of knowledge, that constitutes the seed from which man makes technology grow. The advances of science, in this sense, made our political isolation from the rest of the world impossible after World War II, just as they earlier made economic isolation impossible. In military affairs, perhaps, were visible the most dramatic and fast-moving changes, as technological developments took us from TNT to H-bombs, from artillery to bombers to ballistic missiles, from cavalry patrols to early-warning radar—all changes that tended to shrink the world and increase the mutual dependence and vulnerability of nations. If, for a moment, I may revert to the language of my chemist days, humanity but a century ago was in the condition of a steady-state reaction, whereas now it is in the midst of a nonsteady, branching, chain process and science is the chain carrier. Public policy, whether domestic or foreign,

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The author is Special Assistant to the President for Science and Technology. This article is adapted from an address delivered before the American Physical Society and the American Association of Physics Teachers in New York City, 29 January 1960.