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Land Surface Erosion and Rainfall as Sources of Strontium-90 in Streams[1]

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ABSTRACT

Strontium-90 concentrations in streams from 1958 to 1967 reflected the changing concentrations in rainfall and accumulation on the land surface. Correlation analysis of data from nationwide sampling networks shows that the ^{90}Sr concentration in streams was accounted for, on the average, by 1.7% of the rainout 2 months earlier, and annual erosion of 0.58% of the accumulated ^{90}Sr on the land surface. Direct runoff of ^{90}Sr in preceding rainfall was highest, 2.0 to 2.2%, in the north central and eastern United States, ranging down to no measurable direct runoff in the southwestern United States. Annual erosion of ^{90}Sr from the land surface ranged from 0.75% in the Ohio River Basin to 0.17% in the Missouri River Basin. If one allows for differences in time and area of application, these results for land surface erosion indicate the potential movement of persistent, strongly adsorbed pesticides from large land areas.

Additional Index Words: radioactive fallout, regression analysis, runoff.

Contributions of dispersed sources to water quality are largely unevaluated. The debates as to whether or not agricultural chemicals significantly affect water quality, and in what situations their use should be limited, cannot be settled until data involving many chemicals and many soil and climatic areas have been studied.

The radionuclide, ^{90}Sr, behaves similarly to many agricultural chemicals in the soil and water environments. It has been rather uniformly distributed over the entire United States in radioactive fallout since 1954. Deposition occurs predominantly in rainfall (2). Upon reaching the soil surface, the strontium ion is only slightly leached (1), and moves principally with eroded surface soil (9). Strontium-90 is a long-lived radionuclide, decaying at the rate of only 2.5% annually. Its movement from the soil surface to streams should, therefore, be similar to that of persistent, strongly adsorbed chemicals applied to agricultural lands.

Strontium-90 in streams essentially is derived either directly from rainfall or by erosion of the accumulated soil deposit. If the fractions of the ^{90}Sr entering streams from both sources are relatively constant and independent of each other, a multiple linear regression equation should describe the relationship. The fraction of ^{90}Sr in rainfall entering directly into streams would be determined by one regression coefficient. The fraction of accumulated ^{90}Sr in the surface soil that is eroded would be determined by the other regression coefficient.

These relationships were studied for the conterminous United States by the use of data collected in national sampling programs from 1958 to 1967. Climatic and geographic effects were studied by examining separately the data for eight major drainage regions (Fig. 1). Regional divisions were as follows: (i) North Atlantic, (ii) South Atlantic and Eastern Gulf of Mexico, (iii) Ohio and Tennessee rivers, (iv) Great Lakes, Hudson Bay, and Upper Mississippi River, (v) Missouri River, (vi) Lower Mississippi and Western Gulf of Mexico, (vii) Colorado River, Great Basin, and California, and (viii) Snake and Columbia rivers. Regional boundaries were those used in the U. S. Geological Survey Water Supply papers (20, 21, 22).

[1] Contribution from the Water Quality Management Laboratory, Agricultural Research Service, U. S. Department of Agriculture, Durant, Okla. 74701. Received 27 August 1973.
[2] Laboratory Director.

NUCLEAR REGULATORY COMMISSION

Docket No. 50-352/353 Official Exh. No. 164

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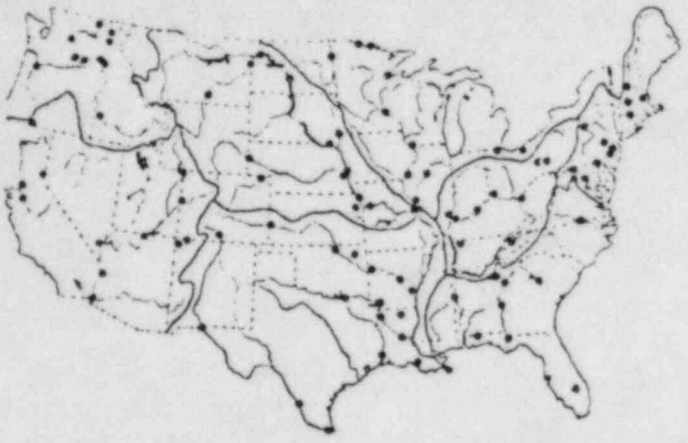

Fig. 1—Drainage regions of the United States showing stream sampling locations where ^{90}Sr concentrations were determined by the U. S. Public Health Service.

SOURCES AND TREATMENT OF DATA

The relationship $Y = ax_1 + bx_2 + c$ was fitted by least squares analysis where Y is flow-weighted average ^{90}Sr concentration in streams for a particular region and calendar quarter, x_1 is the average deposit of ^{90}Sr per unit area on soils in the region at the beginning of the same quarter, and x_2 is the precipitation-weighted average concentration in rainfall in the same region and quarter, or in the quarter preceding by 1, 2, or 3 months.

Stream Data

Strontium-90 concentrations in streams were determined by the U. S. Public Health Service from Oct. 1958, through Sept. 1967, (4, 12, 13, 18, 19). The sampling network expanded from about 50 stations in 1959, mainly on major streams, to about 130 stations in 1963, many of them on smaller streams. The expanded network operated until Sept. 1967, but only half the stations collected samples during a given quarter. Weekly samples of river water were collected manually and composited by quarters before analysis for total ^{90}Sr.

All stations affected by major discharges from nuclear reactors were excluded from the regression analysis. The stations excluded were the Hudson River at Poughkeepsie, N. Y.; the Savannah River at Port Wentsworth, Ga.; all stations below Oak Ridge, Tenn., on the Clinch, Tennessee, Ohio, and Mississippi rivers; and all stations below Hanford, Wash., on the Columbia River. In addition, stations on the Great Lakes or on rivers draining from them were excluded because of the large dilution and long residence time of water in the lakes.

Average ^{90}Sr concentrations for each region, and for the conterminous United States, were weighted by flow. Average quarterly flows were calculated from data published by the U. S. Geological Survey (20, 21, 22) and the International Boundary and Water Commission (7). In most cases, the stream gaging and ^{90}Sr sampling stations coincided. Where they did not, flow from the ungaged area between the two stations was estimated. The ungaged area was usually a small part of the total watershed, exceeding 10% of the gaged area at only 12 stations and never exceeding half of the gaged area.

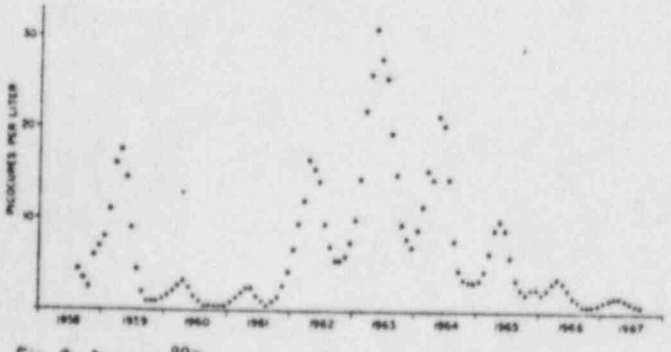

Fig. 2—Average ^{90}Sr concentrations in United States rainfall (running 3-month averages).

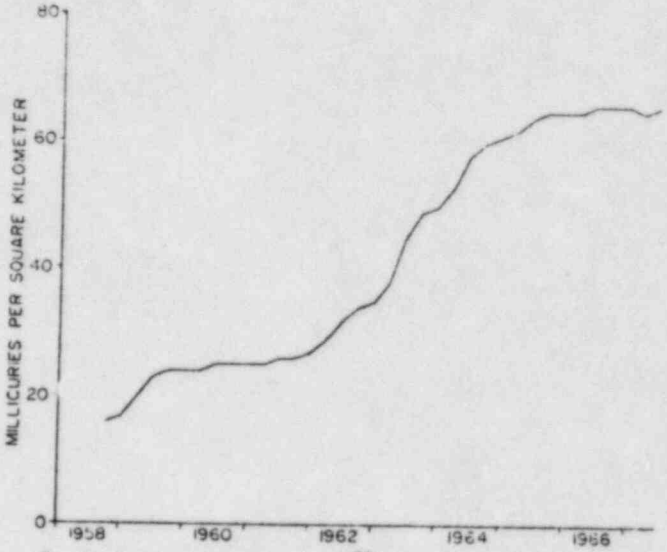

Fig. 3—Average accumulation of ^{90}Sr in United States soils.

Rainfall Data

Strontium-90 concentrations in rainfall have been determined by the U. S. Atomic Energy Commission from 1954 to the present (15). From 1959 to 1967, the sampling network consisted of about 25 widely distributed stations in the conterminous United States. The sample collectors were open-topped pots, or funnels draining into columns of ion exchange resins. The samples were collected and analyzed monthly except during 1960 and 1961 when numerous samples were analyzed by 2-month periods. Because the concentrations of ^{90}Sr were low during this period, it was possible to estimate the monthly concentrations with little error.

Average concentrations of ^{90}Sr in rainfall were computed for each of the eight regions separately as well as for all stations combined. To be consistent with the averaging method for stream water, the averages were weighted by amount of rainfall. Also to be consistent with the quarterly analysis of stream water, the monthly rainfall averages were combined to give running 3-month averages (Fig. 2). The use of 3-month averages helped to even out sample variations for those times (17% of the total number of cases) when a region had fewer than three sampling stations during a month.

Soil Data

Strontium-90 concentrations in soils were determined by the U. S. Atomic Energy Commission on samples collected by the U. S. Soil Conservation Service (2, 5, 10). Extensive sample collections were made annually from 1958 to 1965, except in 1961. The sampling sites were carefully selected on grass-covered areas that were not subject to erosion or flooding. Surface vegetation and soils were included in the sample, with repeated checks to be certain that samples were taken deep enough to include at least 95% of the ^{90}Sr in the soil profile.

The ^{90}Sr concentrations in the soils within each of the eight drainage regions were plotted against time of sampling. Cumulative deposition curves were fitted by eye for each region. Interpolation between sampling dates was based on data from rainfall sampling network. Regional averages were weighted by area to compute the national average (Fig. 3). The regional and national averages for

e 1—Multiple regression correlations of ^{90}Sr concentrations in streams (Y) with soil deposition (X_1) and concentration in rainfall (X_2) 2 months previously: $Y = ax_1 + bx_2 + c$ (±) s.d., R = multiple correlation coefficient

Region	Regression coefficients a	b	c	SD*	R
United States	0.0272	0.0627	-0.1305	0.331	0.8972
Northeast	0.0105	0.0338	0.2070	0.294	0.8091
Southeast	0.0150	0.0680	0.0800	0.513	0.6513
Ohio Valley	0.0167	0.0566	0.0308	0.304	0.9033
North Central	0.0355	0.0870	-0.2370	0.453	0.9252
Missouri Valley	0.0403	0.0725	-0.7699	0.697	0.8559
Southern Plains	0.0567	0.1415	-0.9727	0.522	0.9074
Southwest	0.0327	-0.0041	0.5211	0.738	0.5240
Northwest	0.0120	0.0279	0.0750	0.417	0.6170

* Standard deviation.

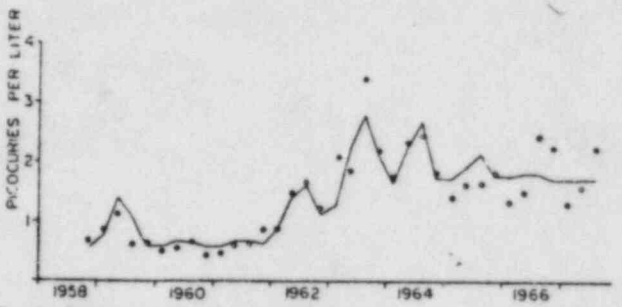

Fig. 4—Computed (line) and measured (points) average concentrations of ^{90}Sr in United States streams.

1966 were within (±) 7% of values obtained by planimetry of the cumulative deposition maps of Meyer et al. (10), with one exception. The soil sample average for the Snake and Columbia river basins was 16% less than that obtained from the deposition map, indicating possible bias in location of the sampling sites in this region. The cumulative deposition curves were adjusted to agree with the deposition map before they were used in the regression analysis.

The soil and rainfall analyses give independent measures of ^{90}Sr deposition. The two measures agreed perfectly for the North Atlantic region. In the other regions, the rainfall analyses showed consistently less deposition, ranging from 63 to 92% of that measured by soil analysis. The poor efficiency of the resin columns may account for much of the discrepancy. On the average, these resin columns caught 88% as much ^{90}Sr as the pot collectors at seven locations, where both types of collectors were used (23). Column collectors were used at more than half of the rainfall sampling stations. Those regions with the greatest discrepancy between soil and rainfall analyses had the greatest number of column collectors. Unfortunately, there is no way of correcting the discrepancy, but it is thought to be due to improper maintenance of the collectors and would vary accordingly.

RESULTS

Multiple regression coefficients and correlation coefficients are listed in Table 1 for each of the eight regions and for the combined regions. The coefficients listed were obtained for rainfall concentrations observed 2 months earlier than the stream concentrations. Different lead times for the rainfall concentrations made little difference in the correlation coefficients, but 2- or 3-month lead times usually gave the highest correlation.

In all cases except one, both X variables were highly correlated ($P > 0.99$) with Y. The exception occurred in the southwest region, where there was no significant effect of rainfall concentration. The X variables showed little correlation with each other ($P < 0.48$), as may be appreciated by comparing Fig. 2 and 3.

Figure 4 shows the concentrations of ^{90}Sr in United States streams as computed from the regression equation in comparison with observed concentrations. The variations in observed concentrations are well-predicted until the last 2 years of observation. Then observed concentrations varied more than computed concentrations, with observed concentrations being high in the summer and fall quarters and low in the winter and spring quarters.

Regression coefficients were also computed separately for each quarter of the year. In some cases, this gave significantly better agreement between computed and observed concentrations than did the use of single coefficients, "a" and "b", for all quarters. Since the improvement was slight, occurred in only half the drainage regions, and complicated interpretation of the data, it is not reported here.

The coefficients in the regression equation give measures of the average contributions of soil erosion and direct runoff of rainfall to the ^{90}Sr concentrations in streams. The first coefficient, "a", indicates the pCi of ^{90}Sr/liter of stream water that are derived from 1 mCi of $^{90}Sr/km^2$ of land surface. Since 1 cm of runoff produces 10^7 liters/km^2, and 1 mCi equals 10^9 pCi, it is only necessary to multiply coefficient "a" times the average annual runoff in centimeters to obtain the annual percentage of accumulated ^{90}Sr being eroded into streams. The second coefficient, "b" indicates the ratio of ^{90}Sr concentrations in streams and associated rainfall. Thus, multiplying coefficient "b" times the percentage of rainfall that runs off gives the percentage of ^{90}Sr in the rainfall that ran off.

Regional values for annual average ^{90}Sr runoff and erosion are listed in Table 2. These values were calculated after adjusting the multiple regression planes to intercept the origin without changing the ratios of coefficients "a" and "b." Obviously, if there were no ^{90}Sr in rainfall and none on the land surface, there should be none in the stream water, since stream locations that were affected by nuclear reactors were excluded from this analysis. The "c" coefficients in Table 1 seem to vary randomly at about zero, and are usually smaller than the residual standard deviations.

The direct runoff of ^{90}Sr ranged from slightly above 2% in the north central and eastern United States, to no measurable direct runoff in the southwestern United States. If the true concentrations of ^{90}Sr in rainfall were higher than the measured concentrations, because of poor

Table 2—Regional values for rainfall, runoff, sediment yield, and percentages of ^{90}Sr runoff and annual erosion of ^{90}Sr from the land surface

Region	Annual rainfall	Annual runoff	Sediment* yield	Annual erosion of accumulated strontium-90	Strontium-90 runoff
	cm		tons/km²	%	
United States	76	23	86	0.58	1.72
Northeast	104	51	69	0.69	2.11
Southeast	135	38	49	0.62	2.17
Ohio Valley	118	44	69	0.75	2.15
North Central	76	20	70	0.63	2.02
Missouri Valley	51	6	159	0.17	0.59
Southern Plains	71	13	101	0.51	1.79
Southwest	39	8	390	0.44	+0.14
Northwest	67	41	44	0.54	1.87

* From references (5) and (8). Ohio Valley includes only the drainage area above Cincinnati. North Central region only the Upper Mississippi Valley.

collection efficiency, the percentage of direct runoff would be lower. Annual erosion of ^{90}Sr from the land surface ranged from 0.75% in the Ohio River Basin to 0.17% in the Missouri River Basin.

DISCUSSION

The poor agreement between observed and computed concentrations in 1966 and 1967 is not readily explained. Apparently, some factor affecting ^{90}Sr concentrations in streams has not been recognized. Two possibilities are that ^{90}Sr accumulates in streambed deposits, and that the regression coefficients change with time.

Streambed deposits of ^{90}Sr would become resuspended during high flows, thus increasing the measured concentrations. However, the deviations from computed values were not correlated with stream flow. Likewise, streamflow data did not improve the multiple correlation when added as a third independent variable to those shown in Table 1.

When the regression coefficients were analyzed by season, some possibly significant differences were found. For the combined regions, the soil erosion coefficient was 1.3 times as high in the summer and fall quarters as it was in the spring quarter. The runoff coefficient was 2.5 times as high in the winter as in the other quarters, but this was counterbalanced by a lower erosion coefficient. Statistically significant improvements in data fit were found using seasonal coefficients in the Northeast, North Central, and Ohio Valley regions. In each of these regions, the runoff coefficient was higher in the winter than in other quarters. This effect may be related to the occurrence of rainfall predominantly on saturated or frozen soils. The erosion coefficient in the Northeast and Ohio Valley regions was higher in the fall than in other quarters. In the North Central region it was lower in the winter than in other quarters. Interpretation of these changes is speculative.

Erosion of the soil deposit might decrease with time because of the slow movement of ^{90}Sr from the surface to deeper soil layers. This hypothesis was tested with recently published data for ^{90}Sr concentrations in 1971 in streams east of the Continental Divide (16, 17). Strontium-90 concentrations computed from regression equations for the appropriate regions averaged 42% higher than measured concentrations. Erosion of the soil deposit decreased by about this percentage since rainfall contributed a minor portion of the ^{90}Sr in streams in 1971. However, variations in stream concentration were similar in magnitude to those observed in 1966 and 1967.

Sediment yields from the major river basins (Table 2) in the humid region rank, in order, similar to the annual erosion of ^{90}Sr. Agreement would be better if more of the drainage areas in the Ohio Valley and North Central regions were included in the sediment yield data. However, in those regions with less than 20 cm of annual runoff, erosion of ^{90}Sr is much less than might be expected on the basis of sediment yields. This is explained by the tendency toward gully and channel erosion in semi arid or arid climates (14). Gully and channel erosion would provide sediment mostly uncontaminated by ^{90}Sr.

Studies on small plots show great variability in movement of applied chemicals during runoff events. Some of the important factors contributing variability include size of plot, amount of runoff, type and amount of soil cover, distribution of applied chemical with depth in the soil, and adsorption characteristics of the chemical. Simulated rainfall, applied soon after the chemical, washed up to 40% of applied 2,4-D esters from cultivated plots (3). More commonly, about 1% of the chemicals was washed off over study periods lasting 1 or 2 years (11).

Pesticides are applied mainly on cropland, whereas fallout of ^{90}Sr occurs equally on cropland, grassland, forest, and other areas. Since runoff and erosion are usually greater from cropland than from grassland or forest, movement of pesticides to streams is probably greater than indicated for ^{90}Sr. The difference is apparently not extreme, for the percentage of cropland in the humid regions ranges from 11% in the Northeast and Southeast to 37% in the North Central, yet the annual average runoff and erosion of ^{90}Sr is about the same in these regions. Of course, tillage of cropland reduces the amount of ^{90}Sr on the soil surface where it is most subject to erosion. Tillage would also reduce the susceptibility of some pesticide applications to erosion.

The 2% of ^{90}Sr that runs off directly to streams simulates movement of pesticides that might occur if they were applied during or immediately before rainstorms. Thus, 2% is probably the maximum movement into streams, averaged over large areas and all types of land use. This degree of movement could occur with pesticides that are strongly adsorbed on soils and not mixed into the soil soon after application.

CONCLUSIONS

Strontium-90 concentrations in United States streams are strongly related to those in rainfall and on the land surface. The best correlation is obtained when stream concentrations are measured 2 months after rainfall concentrations. Erosion of ^{90}Sr from the land surface is related to sediment yield in regions where sheet erosion predominates but not in regions where gully or rill erosion predominates.

Direct runoff of ^{90}Sr amounted to 2% of that in the rainfall over wide areas of the United States. Average annual erosion of ^{90}Sr from the land surface was less than 0.75% and appears to decrease with time.

Persistent pesticides with adsorption characteristics similar to ^{90}Sr would enter streams in similar proportion.

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Columns Representing Mound-Type Disposal Systems for Septic Tank Effluent: I. Soil-water and Gas Relations[1]

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ABSTRACT

Columns were designed to represent the vertical dimensions of a mound-type disposal system for receiving septic tank effluent on problem soils. The columns were filled with gravel (representing creviced bedrock), silt loam (representing the original topsoil), a sand or sandy loam till (fill material), gravel (the seepage bed), and another layer of silt loam (the mound cover). The columns were dosed with septic tank effluent at the rate of 2 cm every 6 hours. Until crusting caused permanent ponding at the fill-gravel interface, the fill was aerobic and the silt loam at the bottom of the column was anaerobic. Redox potentials were higher in the fill (350 to 600 mV) than in the silt loam (200 to 410 mV). Moisture tension fluctuations after a 2 cm addition were greatest near the fill-gravel interface and decreased with depth in the column. After continuous ponding, tension fluctuations almost ceased, the subcrustal soil became anaerobic, and the redox potentials decreased to around -250 mV. In a separate experiment, simulating field conditions, aerobic conditions were maintained in the subcrustal fill, by perforating column walls. The moisture tension regime and the rate of crust formation were similar to nonperforated columns.

Additional Index Words: soil crusting, unsaturated water flow, redox potentials, crusting, methane.

The current Health Code in Wisconsin (Wisconsin State Board of Health, 1969) does not allow the construction of a septic tank and subsurface seepage bed when certain soil conditions prevail. An expensive holding tank operation is, at present, the only legal alternative to the septic tank at individual homes on such soils. The magnitude of the problem is apparent from the estimate that about 50% of Wisconsin soils are considered by the Health Code to be unsuited to the traditional septic tank-subsurface seepage bed system (Bouma et al., 1972). Similar limitations are being experienced in other parts of the country (USDHEW, Public Health Service, 1967).

One possible alternative is to construct a disposal field in fill on top of the unsuitable soil (the mound system). The mound system has been proposed mainly for problem situations involving sites with: (i) a high water table, (ii) a slowly permeable subsoil, and (iii) a shallow permeable soil above highly creviced bedrock (Bouma et al., 1972). With the traditional system, nutrient and pathogen contamination of ground water would occur in cases (i) and (iii) and seepage of unpurified effluent to the surface in case (ii).

When building a mound, 60 cm (2 ft) of sand is placed above the original topsoil, followed by 30 cm (1 ft) of gravel on which polyvinyl chloride (PVC) distribution pipes are laid. The pipes are covered first with a shallow layer of straw, and then with 15 to 30 cm of topsoil. The top and sides of the mound should be seeded and a vegeta-

[1] Contribution from the Soil Science Dep., Univ. of Wis., and the Geological and Natural History Survey, University-Extension Madison, Wis. 53706. This research was part of the Small Scale Waste Management Project, funded by the State of Wisconsin and the Upper Great Lakes Reg. Comm. Received 22 June 1973.

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