Relation between Ce^{144} and Sr^{90} Rainout in convective rainstorms

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ABSTRACT

Analyses were made of the time and space distribution of the concentrations of Ce^{144} and Sr^{90} in 1963 convective storms in Illinois through use of a network of 16 automatic rainwater samplers operated in conjunction with recording raingages and weather radar sets. Great space and time variability were found in the Ce^{144}/Sr^{90} ratio in these storms. A relatively strong trend was found for the above ratio to decrease during the life of storms. A relatively weak trend was noted for the average ratio of Ce^{144}/Sr^{90} to decrease with increasing amounts of total storm rainfall. Implications of these findings are discussed.

Introduction

For a research program on radioactive rainout in convective rainfall (Huff & Stout, 1964), a network of 16 automatic rainwater samplers was operated in central Illinois during the spring and summer of 1963. As part of this program, extensive sampling was made of the concentration of Ce^{144} and Sr^{90} in the rainwater from eight convective rainstorms. The ratio, Ce^{144}/Sr^{90}, provides a measure of the average age of the radioactive debris in water samples, and has been used by several investigators to calculate the source of the nuclear debris brought to the surface in specific rainstorms.

One drawback to the use of the ratio is the effect of fractionation on the theoretical ratio that must be used in debris age calculations (Walton, 1961). High altitude nuclear detonations have less likelihood of nuclide fractionation. In this paper, however, fractionation effects are not of paramount importance, since emphasis is placed upon the space and time variability of the ratio and not upon calculations of the actual age of the debris. The primary purpose of the investigation was to determine whether the radioactive rainout in convective storms tends to contain radioactive debris from single or multiple atmospheric source regions. In the case of a single source, the ratio should remain relatively constant in a storm with small changes in time and space. Thus, the degree of space and time variability of Ce^{144}/Sr^{90} provides a measure of the extent to which air associated with the development and sustenance of a thunderstorm, a line of storms, or a group of storms is consistent with respect to atmospheric environment.

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Data used

Analyses were restricted to 1963 storms in which at least four rainwater samples were obtained with the automatic samplers to define the distribution pattern of radioactive rainout through a storm. Further, analyses were limited to storms in which samples were obtained from the start to finish of rainfall. With these restrictions, 34 sets of time samples from eight storms were available for study of Ce^{144}/Sr^{90} from spring and summer field operations. Radar observations were available also to aid in the analyses of these storms.

The 1963 sampling network is shown in Fig. 1. The automatic samplers are labeled "time sampler," and a recording raingage was located at each of these. The total storm samplers are plastic tubs used to catch samples of the total storm rainfall for use in other phases of the research program. The outer border in Fig. 1 encompasses an area of approximately 6000 square miles used in April and May in support of a nationwide sampling program in cooperation with other agencies involved in research on radioactive rainout. During June and July, the outer ring of time samplers was removed and the network reduced to approximately 3000 square miles to facilitate servicing and to

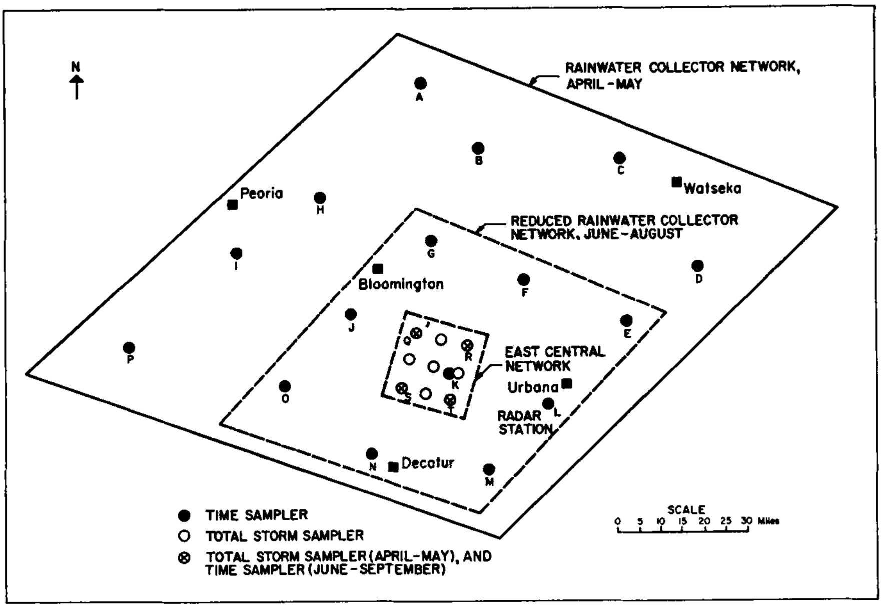

Fig. 1. 1963 rainwater sampling network.

increase the sampling density on the East Central Illinois Rainage Network of 50 raingages in 400 square miles (inner area, Fig. 1). The outer ring of samplers was used to activate stations Q, R, S, and T in Fig. 1.

Characteristics of Ce^{144}/Sr^{90}

A normalizing procedure was employed in the construction of individual Ce^{144}/Sr^{90} distribution curves in the 1963 storms to facilitate comparison of analytical results between stations within a particular storm and from one storm to another (Huff, 1964). In this procedure, the value of Ce^{144}/Sr^{90} in each sample collected at a sampling station within a particular storm was expressed as a ratio to the average value of Ce^{144}/Sr^{90} for the entire storm at the station. These calculated ratios were then plotted against cumulative percent of storm rainfall to provide a measure of the variation in Ce^{144}/Sr^{90} as a storm passed over a sampling point.

The individual distribution curves from all 34 cases for the eight 1963 storms were combined to obtain the seasonal average curve shown in Fig. 2. Because the variation between sampling stations within storms and between storms was relatively large in some cases, the 90-per cent envelope curves have also been indicated in Fig. 2 to illustrate the degree of variation about the average curve.

The average curve for all storms indicates a trend for Ce^{144}/Sr^{90} to remain relatively constant until approximately 40 per cent of the rain has fallen, after which it displays a pronounced decrease throughout the rest of the storm. In all 34 cases, Ce^{144}/Sr^{90} reached a maximum during the first half of the rainfall.

The average curve indicates a trend for the

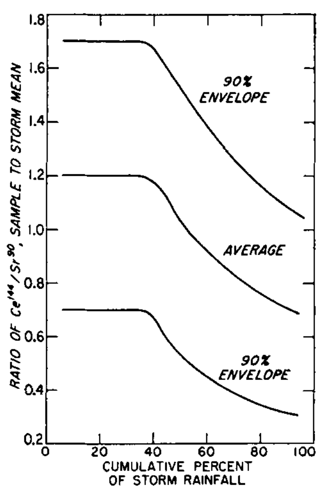

Fig. 2. Average Ce^{144}/Sr^{90} in 1963 storms.

rainfall in the latter portions of storms to contain radioactive debris of significantly different proportions and, apparently, of an older average age than is found in the early portion of storms. A satisfactory explanation of the observed trend has not been found. Initial efforts to relate the ratio distribution characteristics to several meteorological factors, such as rate and duration of rainfall, and storm type, have not revealed significant relationships.

One possibility is that the higher ratios at the start of storms are associated with low-level entrainment of surface-contaminated air into the leading edge of the storm. This low-level air, polluted with surface dirt, could be relatively rich in Ce^{144} in comparison with Sr^{90} as a result of greater leaching of the Sr^{90} fallout into the soil. This condition would then result in a relatively high ratio at the start of storms due to the recycling of previous surface fallout. Evidence presented by Huff (1964) indicates that the trend for relatively high concentrations of radioactive rainout at the start of storms is related also to the entrainment of surface-contaminated air into the leading edge of storms. However, analysis by Walton (1961) provides contrary evidence. He found resuspension only a minor factor influencing the variability of Ce^{144}/Sr^{90}, based upon rainwater and soil analyses in New Jersey.

Extensive measurements of raindrop size distributions in Illinois rainstorms in recent years by the State Water Survey have shown a trend for the raindrop distribution at the start of convective showers to have a disproportionate number of relatively large drops. Differences in the affinity of particulates rich in Ce^{144} and those rich in Sr^{90} for different sizes of raindrops and cloud droplets could contribute to the relatively high values of Ce^{144}/Sr^{90} at the start of rainstorms and to the space and time variability in this parameter.

Table 1. *First and last ratios in storm of June 10, 1963*

Station	First	Last
E	33	12
F	20	9
G	15	19
K	20	7
L	23	4
M	28	14
N	19	18
R	27	9
S	29	5
T	23	—
Median	23	9

Table 2. *First and last ratios in storm of June 13, 1963*

Station	First	Last
C	12	1+
D	29	12
E	29	—
F	—	27
L	22	5
M	—	10
N	49	17
Median	29	11

Tables 1 and 2 further illustrate the trend for smaller values of Ce^{144}/Sr^{90} near the end of storms in comparison with those at the beginning of storms. In these tables, first and last ratios are shown for sampling stations in the storms of June 10 and June 13, 1963. The storm of June 10 consisted of squall-line rainfall in advance of a cold front. The rainfall on June 13 at stations C and D (Table 2) was associated with a warm front passage, whereas the rain at the other stations occurred later in air mass thunderstorms in the warm sector of the storm system.

In four of seven storms during 1963 in which samples were obtained at four or more sampling stations, a trend was found for Ce^{144}/Sr^{90} to decrease with increasing storm rainfall amounts. In these storms, therefore, a tendency was shown for smaller ratios (older debris) to occur in the more intense rainshowers or thunderstorms in the convective storm system. Greater vertical development of the more intense cells into a layer of older debris may have taken place in these storms or, again, the variation may have been related to differences in droplet collection efficiencies within the convective cloud system. In the other three storms, no significant relationship was indicated between volume of storm rainfall and Ce^{144}/Sr^{90}.

Fig. 3 shows an example of the relationship between Ce^{144}/Sr^{90} and total storm rainfall in the storm of June 13, 1963. In this figure, a regression line has been fitted to the data from

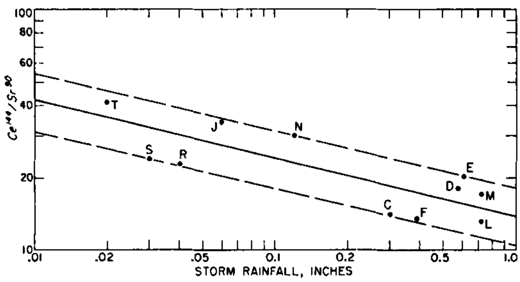

Fig. 3. Relation between Ce^{144}/Sr^{90} and storm rainfall on June 13, 1963.

the 11 sampling stations and dashed lines used to indicate the envelope of values about the mean. A distinct decrease in the ratio with increasing storm rainfall is indicated. Rain at five stations (*D*, *E*, *R*, *T*, *M*) was associated with mature convective storms. At two stations (*N*, *S*) the storms were dissipating, and at three stations (C, F, J) the storms appeared to be in a developing or intensifying stage. Characteristics of the storm over station *L* could not be determined from radar data.

Four of the five stations exposed to mature storms fall above the regression line in Fig. 3 otherwise, no distinct grouping by meteorological factors was found. The differences between stations at high rainfall values in Fig. 3 range between 13 and 20 and could have resulted primarily from radiochemical processing errors. Overall, however, the range of values among the 11 stations appears to be too great to contribute solely to processing errors, and indicate to the authors a strong likelihood that the rainstorms contained radioactive debris from multiple atmospheric source regions.

Comparison of 32 time samples at Station *F* (Fig. 1) on July 13, 1963, provided an excellent example of the radioactivity-rainfall relationships associated with the approach and passage of warm and cold fronts within the same storm system. These relationships are illustrated in Fig. 4. At station *F*, Ce^{144}/Sr^{90} ranged from 6 to 34 in 9 samples from a steady rain of 0.34 inch associated with the warm front approach, from 4 to 18 in 7 samples from a later rainshower of 0.25 inch associated with the warm front passage, and from 6 to 43 in 13 samples from a thunderstorm of 0.44 inch associated with the cold front passage. Average rainfall rate was 0.20 in/hr in the steady rain, 0.19 in/hr in the rainshower, and 2.50 in/hr in the thunderstorm. For total storm rainfall on July 13, average Ce^{144}/Sr^{90} varied from 9 to 42 among 15 network stations.

Areal and time variability of Ce^{144}/Sr^{90}

A measure of the areal variability of Ce^{144}/Sr^{90} in convective storms is provided in Table 3. Data from the 1963 sampling network (Fig. 1) were used in compiling this table which shows the average ratio for each storm, the number of stations upon which the average is based, and the coefficient of variation. The coefficient of variation provides a measure of the relative variability within the network which encompassed 6000 square miles in the April–May storms and 3000 square miles in the June–July storms. The coefficient of variation, *C.V.*, expressed as a percentage, is calculated as follows:

$$C.V. = \frac{\text{Standard deviation}}{\text{Network average}} \times 100$$

The best available information indicates a standard deviation of approximately ± 12 per

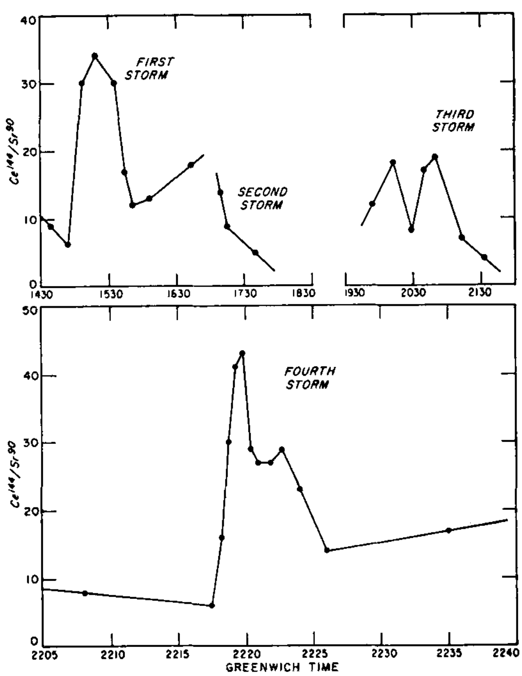

Fig. 4. Time distribution of Ce^{144}/Sr^{90} on July 13, 1963.

cent in the ratios of Table 3 due to uncertainties in the radiochemical processing of the data (Walton *et al.*, 1963). Therefore, when the coefficient of variability exceeds 12 per cent by substantial amounts, it is indicative of variability resulting from processes outside radiochemical measurements. As shown in Table 3, the coefficient of variation has a median of 36 per cent, approximately three times the radiochemical measurement error, and ranges from 18 to 49 per cent. Thus, it appears that there is substantial natural variability in the Ce^{144}/Sr^{90} ratio resulting from atmospheric processes.

Table 3 shows network ratio averages ranging from 12 to 24 with the relative variability of the ratio ranging from 18 to 49 per cent within the 10-storm sample. These data indicate that the radioactive rainout in convective storm systems usually incorporates radioactive particulates from several atmospheric source regions within distances of a few miles, if the Ce^{144}/Sr^{90} ratio truly reflects the atmospheric distribution of the two radionuclides.

Examination of the concentrations of Sr^{90} and Ce^{144} used in the calculations of Ce^{144}/Sr^{90} showed wide variation from station to station in individual storms and in the network average between storms. For example, in the storm of June 10, the Sr^{90} concentration in the total storm rainfall samples ranged from less than 50 pc/l to over 500 pc/l among stations, and the Ce^{144} concentration ranged from less than 1000 pc/l to over 6000 pc/l on the network of 6000 square miles (Fig. 1). Network averages in the 1963 storms shown in Table 3 ranged

Table 3. *Areal variability of Ce^{144}/Sr^{90} in 1963 storms*

Date	Number of stations	Average network ratio	Coefficient of variation (%)
4/17	8	18	19
4/19	8	19	42
4/22	12	21	25
5/4	6	22	33
6/10	12	18	18
6/13	9	24	40
6/19	13	15	39
7/1	13	14	19
7/13	15	19	49
7/20	12	12	44
Median		19	36

Table 4. *Time variability of Ce^{144}/Sr^{90} in storm of June 10, 1963*

Station	Number of samples	Mean ratio	Coefficient of variation (%)
E	6	20	45
G	5	19	18
K	5	15	61
M	4	19	43
N	6	19	7
R	6	19	73
T	4	22	11
Median		19	45

from 12 pc/l to 177 pc/l for Sr^{90}, and from 227 pc/l to 2922 pc/l for Ce^{144}. The highest values occurred on June 10 and the lowest on July 13.

In Table 4, the time variability in Ce^{144}/Sr^{90} is shown for several stations in the squall-line storm of June 10, 1963 to illustrate the typical variability within a thunderstorm as it passes over a point. Only stations with four or more time samples were used in this tabulation. Among the stations, the relative variability varied from a relatively low value of 7 per cent at Station *N* to a high value of 73 per cent at Station *R*. The median was 45 per cent for the seven stations. Combining 34 cases from eight storms during 1963 in which stations had four or more time samples, a median of 45 per cent was obtained for the time relative variability. The median value of areal variability for Ce^{144}/Sr^{90} was 36 per cent (Table 3). Thus, the time variability within a storm passing a point appears to be of the same general magnitude as the areal variability in storms affecting areas of 3000 to 6000 square miles. The general conclusion is that both the time and space variability in the ratio of Ce^{144}/Sr^{90} is frequently high.

Summary and conclusions

A trend was found for the ratio of Ce^{144}/Sr^{90} to be appreciably lower at the end of storms than at or near the beginning of rainfall. The cause of this trend has not been conclusively determined at this time. Two possible influences are entrainment of surface-polluted air that is relatively high in Ce^{144} into the forward edge of storm systems, and differences in the affinity of Ce^{144} and Sr^{90} particulates for various sizes of cloud droplets and raindrops.

A relatively weak trend was found for the average Ce^{144}/Sr^{90} to decrease with increasing amounts of total storm rainfall. This trend may be related to greater vertical development of the more intense storms into the stratospheric levels and stronger entrainment of air into these storms.

The variability of Ce^{144}/Sr^{90} was found to be large in both time and space. Until further research clarifies the cause of the large time and space variability observed in Ce^{144}/Sr^{90} in the 1963 storms, the use of this ratio as a reliable indicator of the debris age, and consequently of the atmospheric source of radioactive rainout in rainwater, is questionable.

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СООТНОШЕНИЕ МЕЖДУ Ce^{144} И Sr^{90}, ВЫПАДАЮЩИМИ В КОНВЕКТИВНЫХ ОСАДКАХ

Проведен анализ пространственно-временного распределения концентрации Ce^{144} и Sr^{90} в конвективных бурях 1963 года в Иллинойсе на основе использования сети 16 автоматических установок для сбора дождевой воды, работающих вместе с сетью записывающих дождемеров и метеорологических радиолокаторов. В этих бурях была обнаружена большая изменчивость в пространстве и времени для отношения Ce^{144}/Sr^{90}. Для указанного отношения была обнаружена относительно сильная тенденция к уменьшению в течении времени жизни бури. Отмечена относительно слабая для среднего отношения Ce^{144}/Sr^{90} тенденция уменьшаться с увеличением полного количества осадков в буре. Обсуждается значение обнаруженных фактов.