SULFUR CONTENT OF OKLAHOMA RAINFALL

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It has been known for a long period of time that small quantities of sulfur are present in rain (Alway 1940). One of the earlier investigations on this subject was conducted by Miller (1906) at Rothamsted, England, who collected rainfall during the six-year period from 1881 to 1887. The average rainfall during this period was 29.95 inches and the quantity of sulfur in the rain was 7.0 pounds an acre. Miller reports that the sulfur content of rain collected at Lincoln, Canterbury, New Zealand, from 1884 to 1888 was six pounds per acre. The average rainfall was 29.7 inches. Crowther and Ruston (1911) collected rainfall from November 1907 to December 1909 on the experimental farm operated by the University of Leeds near Garforth, England. The average rainfall was 25.59 inches and the total quantity of sulfur was 38.27 pounds an acre. Rainfall collected by these investigators at ten different stations located at different distances from Leeds during one year from November 1907 to October 1908 indicated that the sulfur content decreased gradually from approximately 134 pounds to 29.2 pounds an acre as the distance from the city increased.

Data collected by Kossowitsch (1913) from various locations in Russia show very clearly the effect of industrial areas on the quantity of sulfur appearing in rainfall. Two samples collected in or near St. Petersburg and one sample collected from southeast Russia contained much higher quantities of sulfur than four samples obtained from areas which were distinctly rural in character. These results are given in Table I.

TABLE I

Sulfur content of rainfall occurring in different parts of Russia

Location	No. of years	Period	Annual rainfall inches [a]	Pounds of sulfur per acre	Pounds of sulfur per acre-inch of rain
St. Petersburg	2	1909-11	24.62	26.8	1.09
Ochta	1	1909-10	24.12	28.6	1.19
Pawlowsk	1	1909-10	21.30	5.8	.27
Sapolje	1	1909-10	18.99	3.8	.20
Tula	1	1909-10	19.76	3.2	.16
Iekaterinoslav	1	1909-10	14.03	19.8	1.41
Ssamara	2	1909-11	15.75	2.6	.16

[a] Calculated

Investigations on the sulfur content of rainfall in the United States have been conducted at several agricultural experiment stations. Some of the earlier research on this subject occurred at the University of Illinois and at the University of Tennessee. Several investigators have collected rainfall for a few months or have analyzed individual rains for sulfur content and have attempted to estimate the quantity of sulfur brought to earth by applying the data obtained to annual rainfall records. Sucn information was not included in the compilation of data which are presented in Table II. Only those locations were listed where continuous records for one or more years were obtained. It is quite evident from a study of these data that the quantity of sulfur occurring in the rainfall depends to a very great extent upon the distance from densely populated areas where coal is the most important source of fuel.

The effect of industrial areas on the sulfur content of rainfall is quite evident in the comparison which was made by Eaton and Eaton (1926) who collected rainfall on the campus of the University of Chicago and at Liberty, Indiana. Nearly five times as much sulfur was found in the rainfall in Chicago as at Liberty, Indiana. In a similar study by McIntire and Young (1923) at Copperhill, Tennessee, during a three-year period, rainfall collected from an area near a smelter was compared with rainfall collected six miles away. These data show that the sulfur content of the rainfall at a distance of six miles from the point of origin had decreased to less than ⅛ of that occurring in the rain collected near the smelter. A second comparison on the effect of an industrial area on the sulfur content of rainfall was also made by these investigators at Knoxville, Tennessee, and at a location seven miles from the city. Although this comparison did not cover the same period of time, the sulfur content of rain collected for a two-year period seven miles from the city was only 20% of the sulfur content of rain falling at the weather bureau station in Knoxville. At Crossville, Tennessee, the sulfur content of rain was very similar to that obtained by Kossowitsch (1913) in the rural areas of Russia. The effect of densely populated areas on the sulfur content of rainfall was also studied by Alway et al. (1937) who found that 196.7 pounds of sulfur were obtainea in rainfall at Minneapolis, Minnesota, during the 1936 season, and only 3.1 pounds were secured during the same period at Bemidji, Minnesota, which is located approximately 180 miles north of Minneapolis. Riffenburg (1925) analyzed 23 samples of rainwater collected at Washington, D. C., in 1923 and 1924. He found that these samples varied in SO_4 content from 1 to 17 parts per million and the pH value varied from a minimum of 4.4 to a maximum of 7.7. This author also stated that the average SO_4 content of rain, as compiled from a large number of references which were not given in his manuscript, was 5 parts per million. Collins and Williams (1933) are of the opinion that much of the rain falling in thinly populated areas does not contain more than 2 parts per million of sulfate which is equivalent to .15 pounds of sulfur per acre-inch of rain.

TABLE II

Sulfur content of rainfall at various locations in the United States where continuous records for one or more years have been obtained

PLACE	No. of years	Period	Average annual rainfall in inches	Average quantity of sulfur in pounds per acre	Pounds of sulfur per acre-inch of rain
ILLINOIS					
Chicago (Eaton 1926)	1	1921-22	36.8	227.4	6.18
Urbana (Stewart 1920)	7	1912-19	33.0	45.1	1.36
Urbana (Stewart 1920)	3	1917-19	38.1	40.8	1.07
INDIANA					
Liberty (Eaton, 1926)	1	1921-22	49.3	46.0	.93
IOWA					
Ames (Erdman 1923)	1	1921	30.4	14.9	.49
KENTUCKY					
Greenville (Johnson 1924)	2	1921-22	45.5	36.0	.79
Lexington (Johnson 1924) Weather Bureau	2	1921-22	44.9	41.2	.92
Lexington (Johnson 1924) Van Meter farm	2	1921-22	45.6	23.8	.52
Mayfield (Johnson 1924)	2	1921-22	49.6	25.9	.52
Paducah (Johnson 1924) Lone Oak	2	1921-22	42.8	33.8	.79
Russellville (Johnson 1924)	2	1921-22	43.5	29.8	.69
Shelbyville (Johnson 1924) Lincoln Institute	2	1921-22	41.4	17.1	.41
MINNESOTA					
Bemidji (Alway 1937)	1	1936	....	3.13	.....
Minneapolis (Alway 1937)	1	1936	....	196.70[a]	.....
Page (Alway 1937)	1	1936	....	5.51	.....
University Farm (Alway 1937)	1	1936	....	25.96[a]	.....
NEW YORK					
Alfred (Wilson 1926)	2	1923-35	32.3	49.7	1.54
Brockport (Wilson 1926)	3	1923-26	31.7	77.5	2.44
Ithaca (Wilson 1926)	8	1918-26	29.0	38.2	1.32
Geneva (Collison 1932)	10	1919-28	35.6	41.2	1.16
TENNESSEE					
Columbia (McIntire 1923)	3	1919-21	58.29	26.2	.45
Cooperhill (McIntire 1923) Weather Bureau	3	1919-21	59.68	232.4	3.89
Cooperhill (McIntire 1923) 6 mi. away	3	1919-21	59.68	72.2	1.21
Crossville (McIntire 1923)	3	1919-21	59.58	12.7	.21
Jackson (McIntire 1923)	3	1919-21	50.73	59.2	1.17
Knoxville (McIntire 1923) Weather Bureau	7	1913-21	48.44	94.5	1.95
Knoxville (McIntire 1923) University farm	8	1915-21	50.42	51.5	1.02
Knoxville (McIntire 1923) 7 mi. from city	2	1919-20	58.14	18.6	.32
Louden (McIntire 1923)	3	1919-20	55.83	19.3	.35
McGhee (McIntire 1923)	3	1919-20	52.65	24.2	.46

[a] Personal note from F. J. Alway indicated that these values were "much too high" because of sulfur dioxide in air reacting with metallic surface of rain gauge.

TABLE II—Continued.

Sulfur content of rainfall at various locations in the United States where continuous records for one or more years have been obtained

PLACE	No. of years	Period	Average Annual rainfall in inches	Average quantity of sulfur in pounds per acre	Pounds of Sulfur per acre-inch of rain
TEXAS					
Angleton (Fraps 1930	4	1924-27	39.37	9.3	.24
Balmorhea (Fraps 1930)	4	1924-27	12.57	3.9	.31
Beaumont (Fraps 1930)	4	1924-27	44.16	14.8	.34
Beeville (Fraps 1930)	4	1924-27	26.24	6.4	.24
College Station (Fraps 1930)	4	1924-27	37.50	11.8	.31
Denton (Fraps 1930)	4	1924-27	28.84	9.7	.34
Lubbock (Fraps 1930)	4	1924-27	17.70	5.9	.33
Nacogdoches (Fraps 1930)	4	1924-27	41.82	11.2	.27
Spur (Fraps 1930)	4	1924-27	22.87	6.3	.27
Temple (Fraps 1930)	4	1924-27	33.49	8.0	.24
Troup (Fraps 1930)	4	1924-27	38.29	8.1	.21
VIRGINIA					
Blackburg (Ellett 1929)	6	1923-28	38.63	16.9	.34

Since the quantity of sulfur in the atmosphere is increased as a result of the liberation of sulfur dioxide from the combustion of coal, more sulfur should be found in the rain occurring in the vicinity of densely populated areas during the winter months. Quarterly data obtained for a two-year period in Kentucky by Johnson (1934) show that approximately sixty percent of the sulfur occurred in the fall and winter precipitation. Similar data were obtained by McIntire (1923) in Tennessee from nine different locations which did not include the results from Copper Hill. Wilson (1926) analyzed the rainfall collected at Ithaca, New York, during a three-year period and found that the average sulfur content of the rain from November to May was 15 parts per million, whereas the sulfur content of precipitation from May to November was 3.4 parts per million. Fraps (1930) analyzed rainwater collected at 11 different stations in Texas at monthly intervals during the four-year period from 1924 to 1927. The average annual rainfall for these different locations and the average quantity of sulfur calculated in pounds an acre for each location indicate that the sulfur content of the rain is approximately .3 of a pound for each inch of rain. The variation for the different locations was .21 to .37 inches of sulfur per inch of rain. Some of the high-rainfall areas in Texas contained less sulfur in the rainfall than other areas which received a much lower quantity of total precipitation. In areas quite remote from industrial development or where natural gas and oil are the important sources of fuel, the quantity of sulfur appearing in the rain should be somewhat constant and approach a value which is dependent, to a very great extent, upon the quantity of water which falls. None of the values obtained in the United States are quite as low as some of the values obtained from rural areas in Russia. In the eastern part of the United States a high percentage of the rainfall in rural areas contains an average of .5 to 1 pound of sulfur for each acre-inch of rain.

It has been pointed out by Alway et al. (1937) that the type of container in which the rain is collected may affect the quantity of sulfur which is obtained. Sulfur dioxide occurring in the atmosphere will react with metal surfaces. Rain collected in metal containers will dissolve the soluble sulfates and the quantity of sulfur measured as barium sulfate will be higher than the quantity of sulfur which is actually present in the rainfall. Galvanized iron gave results which were higher than those obtained in brass cylinders. These investigators also found that moist soils will absorb sulfur dioxide from the atmosphere and that the sulfur dioxide content of the atmosphere can be measured by exposing a cylinder coated with lead peroxide to the air under a suitable shelter to protect it from wind and rain. It is quite probable that a positive error for the sulfur content of rainfall has been obtained by some investigators where rain gauges constructed all or in part of galvanized iron have been used. Data collected at Tennessee and reported by McIntire and Young (1923) were obtained from rain gauges coated with asphalt. The rain gauges used by Stewart (1920) at the University of Illinois were lined with tin.[1]

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The first analyses of Oklahoma rainfall for sulfur content were made in 1927. Rainfall was collected for a five-month period before any analyses were made. The rain gauge was located about 1¼ miles west of Stillwater, Oklahoma, on the Oklahoma Agricultural Experiment Station Agronomy Farm. Since 1931 a woven wire fence located about six feet from the north and west side of the rain gauge greatly reduced any possible contamination of the rain water from bird droppings (Johnson 1925). A standard rain gauge was used to collect the water in this experiment. Except for rains which were larger than 2.2 inches the water was collected in a brass cylinder and did not come in contact with a galvanized iron surface. It is quite probable that some of the large rains which caused the brass cylinder to overflow would collect some sulfur from the inside of the rain gauge; however, the free movement of air under such conditions is greatly restricted and the error from this effect is very likely to be rather small. Since it is quite probable that some sulfur dioxide may be absorbed by the soil from the air, any slight error due to sulfur dioxide reacting with the zinc surface on the interior of the rain gauge may not be a serious matter in this region.

Data were obtained at quarterly intervals after the first season during the 15-year period from 1928 to 1942 except in 1935 when semi-annual records were secured. No results were obtained in 1940. Information concerning the quantity of rain for each three month's period, the sulfur content of the rainfall in parts per million and pounds per acre, the annual rainfall and pounds of sulfur per acre-inch of rain are given in Table III.

The method of analyses for sulfur in the rain water was as follows: One liter of filtered water was treated with 5 ml of 1% KCl and 1 ml of concentrated nitric acid, placed on an electric hot plate or steam chest and evaporated to dryness. Hot distilled water was added to dissolve the sulfate, the solution was filtered to remove any insoluble residue, the volume of the filtrate was increased to 150 ml and sufficient hydrochloric acid was added to make the pH of the solution about 1.0. The solution was heated to boiling, 5 ml of 10% barium chloride were added, and the beaker was covered with a watch glass. After precipitation, the sulfate precipitate was placed on a steam chest for two or three hours and then allowed to stand overnight before filtering through a No. 42 Whatman filter paper. The weight of the barium sulfate was determined by ignition in a porcelain crucible.

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[1] Information obtained by letter from Dr. E. E. DeTurk, Urbana, Illinois.

It will be observed that the quantity of sulfur in Oklahoma rainfall was much lower than the sulfur content of rainfall occurring in the eastern part of the United States. The data compare favorably with results published by the Texas Agricultural Experiment Station. The lowest quantity of sulfur in the rainfall during this 15-year study occurred in 1934 when the rainfall was about 5 inches below the average annual precipitation for this locality, and the highest value was obtained in 1933 when rainfall was about 1½ inches below the average for the 49-year period during which rainfall records have been obtained at this station.

Considerable variation occurred in the quantity of sulfur occurring in rainfall for different seasons of the year and for the different years in this study. High rainfall did not correlate with an increase in the quantity of total sulfur obtained. Under central Oklahoma conditions the quantity of winter rainfall is much lower than summer rainfall. More total sulfur, as well as a higher concentration of sulfur in the rainfall, was secured during the fall months of October, November, and December than during January, February, and March. Fifty-seven percent of the total sulfur occurred in rains falling during the second and third quarters. This is almost the reverse of conditions occurring in Kentucky and Tennessee. The average quantity of sulfur in the rain was slightly lower in the spring as compared with the fall and winter months. Rains observed during July, August, and September contained less sulfur than at any other season of the year. There is no accurate explanation for the variation in the sulfur content of rainfall for the different years. The sources of the air masses in which the rainstorms developed might give a plausible explanation of the differences which were observed.

Some coal is burned in Oklahoma but natural gas and oil are the most important types of fuel consumed in most of the larger cities in the state. Three smelters producing zinc and lead from sulfide ores also liberate considerable quantities of sulfur dioxide to the atmosphere. Since these smelters are located north and east of Stillwater, Oklahoma, and the prevailing wind in this area is from the south or southeast in the summer and northwest in winter, it is quite improbable that sulfur dioxide liberated from these smelters would cause any appreciable increase in the sulfur content of the rainfall in this area under average conditions.

TABLE III

Studies on the sulfur content of rainfall at Stillwater, Oklahoma, from 1927 to 1942

Year	Jan., Feb., Mar.			Apr., May, June			July, Aug., Sept.			Oct., Nov., Dec.			Total rainfall inches	Total sulfur lb/A	Lbs. of sulfur per acre-inch of rain
	Rain in.	Sulfur		Rain in.	Sulfur		Rain in.	Sulfur		Rain in.	Sulfur				
		p. p. m. [a]	lb/A		p. p. m. [a]	lb/A		p. p. m. [a]	lb/A		p. p. m. [a]	lb/A			
1927 [b]													38.10	6.85	.18
1928	4.75	1.80	1.94	13.82	.91	2.95	4.29	1.49	1.45	9.66	.98	2.15	32.52	8.49	.26
1929	6.04	1.91	2.62	19.07	1.42	6.14	5.75	1.03	1.34	6.22	1.15	1.62	37.08	11.72	.32
1930	4.63	1.15	1.21	11.06	1.20	3.03	4.79	1.01	1.10	5.57	1.57	1.98	26.05	7.32	.28
1931	4.05	2.13	1.96	7.14	1.45	2.35	8.75	1.01	1.96	10.02	.94	2.14	29.78	8.41	.28
1932	6.03	2.11	2.89	9.10	.58	1.20	12.66	.51	1.46	5.60	1.22	1.55	33.39	7.10	.21
1933	7.77	1.79	3.15	4.42	3.10	3.11	10.74	1.82	4.43	8.50	1.03	1.99	31.43	12.68	.40
1934	3.89	1.72	1.52	6.64	1.72	2.59	10.57	.28	.67	6.01	1.02	1.39	27.11	6.17	.23
1935 [b]													31.89	7.85	.25
1936	.28	1.54	.10	7.78	1.54	2.72	6.39	1.09	1.58	4.24	3.48	3.35	18.69	7.75	.41
1937	1.82	.61	.25	11.10	1.92	4.83	7.84	1.61	2.86	6.59	3.19	4.62	27.35	12.56	.46
1938	9.70	1.11	2.44	15.29	1.30	4.51	9.93	1.22	2.75	3.47	2.09	1.64	38.39	11.34	.30
1939	4.30	1.48	1.44	9.18	.82	1.71	6.47	1.01	1.48	3.89	2.09	1.84	23.84	6.47	.27
1940 [c]													30.63		
1941	2.91	1.58	1.04	14.18	.56	1.81	7.91	.65	1.17	12.66	.76	2.18	37.66	6.20	.17
1942	2.00	.99	.45	20.50	1.03	4.79	11.97	.69	1.87	4.21	3.14	3.00	38.68	10.11	.26
Average		1.52	1.61		1.35	3.21		1.03	1.86		1.74	2.26		8.73	

[a] p. p. m.=parts per million.
[b] Rainfall not collected quarterly.
[c] No data obtained in 1940.

The sulfur content of rainfall collected at Goodwell, Guthrie, Heavener, and Lone Grove, Oklahoma, for different seasons from 1931 to 1939 is given in Table IV. In the High Plains area which is represented by Goodwell, the rainfall is low but the average sulfur content of the rain collected was higher than the average for the central part of the state. The data obtained at Guthrie were lower than results secured at Stillwater in 1931, 1933, and 1938. Slightly higher results were secured in 1934 and 1939. The sulfur content of the rainfall at Lone Grove which is 12 miles west of Ardmore, Oklahoma, is similar to other data where seasonal variations of 50 to 100 percent have been obtained. The sulfur content of the rainfall at Heavener in eastern Oklahoma is characteristic of that occurring in the more humid portion of the United States.

All of the sulfur which occurs in rain may not be absorbed by the soil. During seasons of torrential rainfall, a considerable quantity of the water falling on sloping land is lost by runoff; consequently, the efficacy of the sulfur in rain, as an aid to crop production, will vary considerably on different types of soil and under different climatic conditions.

Since the rain falling in the Great Plains region originates from masses of air moving inland from the Gulf of Mexico, and since the opportunity for contamination of this air with sulfur dioxide from industrial areas is limited, it is quite probable that the sulfur content of the atmosphere in this region will never be very high. Analyses made by F. J. Alway at the University of Minnesota to determine the quantity of sulfur dioxide absorbed by lead peroxide candles exposed on the Oklahoma Agricultural Experiment Station Agronomy Farm 1½ miles west of Stillwater, Oklahoma, during 1941 and 1942, indicate that the sulfur content of the atmosphere in this area is very low. Data obtained with the lead peroxide candles are similar to results secured from a measurement of the sulfur in rain although some rains contain much larger quantities of sulfur

TABLE IV

Studies on the sulfur content of rainfall at different locations in Oklahoma

Place and year	Annual rainfall inches	Pounds of sulfur per acre	Pounds of sulfur per acre-inch of rain
Goodwell			
1933	12.62	5.9	.50
1934	14.27	5.8	.40
Guthrie [a]			
1931	29.20	7.3	.25
1933	31.40	5.6	.18
1934	35.30	7.5	.21
1937	24.13	12.6	.52
1938	31.36	5.9	.19
1939	23.56	8.9	.38
Heavener			
1934	32.03	17.0	.53
Lone Grove			
1934	35.51	7.6	.21
1936	28.58	11.7	.41

[a] Rainfall collected by Soil Conservation Service Staff 5½ miles south of Guthrie, Oklahoma.

than others depending upon the frequency of the rain and the opportunity of recently washed air masses to mix with air from which the sulfur dioxide has not been removed by recent precipitation.

Experiments have shown that the sulfur content of Oklahoma soil is being decreased as a result of cultivation. Since sulfur is an essential plant nutrient and occurs in the surface soil of humid regions chiefly in organic compounds which are insoluble in water, the quantity of sulfur occurring in the rainfall may determine whether sulfur should be added to the soil to maintain crop production. Ellett and Hill (1929) show that more sulfur will occur in the drainage from a shallow layer of soil than will be obtained from a deeper profile. Where runoff occurs, much of the sulfur in the rainwater is not absorbed by the soil. In high rainfall areas some of the sulfur is leached through the soil profile; consequently, there are many areas where the total quantity of sulfur in the rain cannot be used as an index of the sulfur available for plant growth. Alfalfa hay will contain from 4 to 6 pounds of sulfur per ton. Three tons of alfalfa hay will remove more sulfur than is added to many Oklahoma soils by rainfall. As soils decline in fertility, fertilizers such as ordinary superphosphate, which contains large quantities of calcium sulfate, must be applied to maintain the available phosphorus supply of the soil. Ammonium sulfate is a nitrogen fertilizer which is used extensively in fertilizer mixtures. In regions where fertilizers containing sulfur are applied to increase crop production, loss of sulfur as a result of tillage is not an important problem. Thomas and Hill (1937) found that alfalfa plants will absorb sulfur dioxide from the atmosphere but computations by Alway et al. (1937) indicate that under average conditions crops cannot obtain sufficient quantities of sulfur through the stomata of the leaves to supply the requirements for optimum growth, assuming that some relation occurs between the rate of absorption of sulfur and carbon dioxide. In areas where the crop requirements for sulfur are high and the quantity of sulfur occurring in rainfall is low, a sulfur deficiency for crop production may appear before a phosphorus deficiency exists where the available phosphorus content of the soil is high and the organic-matter content is low.

SUMMARY

The sulfur content of rainfall at Stillwater, Okahoma, was studied for 15 years from 1927 to 1942. The largest quantity of sulfur in the rain occurred during October, November, and December. The largest quantity of total sulfur was obtained during April, May, and June which is the period of highest precipitation in this region. The average quantity of sulfur in the annual rainfall was 8.73 pounds per acre with a maximum of 12.68 and a minimum of 6.17 in 1933 and 1934, respectively. The average sulfur content per acre-inch of rain was similar to that obtained in other areas of limited industrial development. The sulfur content of rainfall collected for six years, 5 miles south of Guthrie, Oklahoma, and for two years near Lone Grove, Oklahoma, was similar to the results obtained at Stillwater. Sulfur data on rainfall at Goodwell, Oklahoma, were lower than at Stillwater, although the concentration of sulfur in the rain falling in this area was nearly twice as high. Seventeen pounds of sulfur per acre was present in 32 inches of rainfall collected at Heavener, Oklahoma, in 1934.

Since the sulfur content of Oklahoma soils is gradually decreasing as a result of cultivation, a sulfur deficiency may develop on some areas when crops like alfalfa, which has a sulfur requirement that exceeds the sulfur added to the soil by rainfall, are grown.

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