

$^{87}\text{Sr}/^{86}\text{Sr}$ RATIOS AND TOTAL STRONTIUM CONCENTRATIONS
IN SURFACE WATERS OF THE SCIOTO RIVER
DRAINAGE BASIN, OHIO¹

JOHN B. CURTIS, JR., AND ALAN M. STUEBER

Department of Geology, Miami University, Oxford, Ohio 45056

ABSTRACT

A total of 23 water samples from the Scioto River and its tributaries has been analyzed for total strontium concentrations and strontium-isotopic compositions in order to investigate the nature of strontium in surface waters of the Scioto River drainage basin, Ohio. Total Sr concentrations were quite variable and, in the northern portion of the basin, they were unusually high compared to most North American rivers, probably due to weathering of celestite in the Silurian carbonate bedrock and overlying glacial till.

The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the Scioto River throughout its length are constant at about 0.7084, whereas tributaries have a range of values from about 0.708 to about 0.715. The bedrock of the Scioto River basin consists of Paleozoic carbonates, with $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of about 0.708, and Paleozoic clastic sediments with a significantly higher range of values. Tributaries north of the Pleistocene glacial boundary, underlain either by carbonate or clastic bedrock as well as glacial till, have uniformly low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of about 0.708, indicating the dominant presence of carbonate-type strontium. The effect of bedrock on the strontium-isotopic compositions of these waters is apparently superseded by the effect of readily soluble carbonate and celestite in the glacial till. Tributaries in the southern portion of the basin, underlain by clastic bedrock alone, have relatively high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, indicating the presence of strontium leached from the bedrock. Based on this survey, application of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio as a tracer of surface-water provenance in Ohio should be restricted to unglaciated terrain.

INTRODUCTION

The isotopic ratio $^{87}\text{Sr}/^{86}\text{Sr}$ has been used as a tracer in the study of geologic materials for over ten years. Geochemists have focused their efforts primarily upon rocks and minerals, but applications have included studies of fresh-water systems (e.g., Faure *et al.*, 1963; Hart and Tilton, 1966; Faure *et al.*, 1967). Strontium isotopes in Ohio surface and ground waters have been investigated by Eastin and Faure (1970) and by Stueber *et al.* (1972); a discussion of the concepts involved in the application of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio as a tracer of water movement can be found in these references and in Curtis (1972).

To summarize briefly, with the passage of time ^{87}Rb produces ^{87}Sr through radioactive decay, so that the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in a rock increases at a rate proportional to its Rb/Sr ratio and the decay constant of ^{87}Rb . Chemical weathering of rocks releases strontium into solution in ground water, whose isotopic composition depends upon the geologic ages and Rb/Sr ratios of the rocks contributing the strontium. After the ground water reaches the surface and becomes a part of rivers and streams, its original $^{87}\text{Sr}/^{86}\text{Sr}$ ratio should persist, unless the water is mixed with waters of different isotopic composition. Thus the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of water in a stream reflects the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and the relative proportions of the various ground-water components which contribute to the stream.

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Because the bedrock of Ohio is predominantly composed of rubidium-poor carbonate and rubidium-rich detrital sediments of Paleozoic age, and because these materials should have significantly different present-day $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, the potential exists for finding variable $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in Ohio surface and ground waters. Stueber *et al.* (1972) made a survey of such waters over the entire state in order to investigate this potential and as the first step in evaluating the usefulness of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio as a tracer of water provenance in Ohio. Their results showed that relatively high ratios of 0.711 to 0.713 occur in waters of eastern and southern Ohio, whereas lower ratios of 0.708 to 0.709 are found in waters to the north and west. This trend is apparently due in part to the change in the dominant bedrock lithology across Ohio, clastic sediments predominating in the east and south and carbonates predominating in the north and west.

The location of the Pleistocene glacial boundary evidently also has an effect on the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of Ohio waters. Relatively high ratios occur only in one area north of the glacial boundary (Ashtabula County, which lies far east in the clastic-sediment terrain), whereas low carbonate-type ratios occur in areas which are underlain by clastic bedrock. Apparently the presence of readily soluble carbonate material in the glacial till overlying clastic bedrock has had a strong effect on the strontium-isotopic composition of waters in such areas.

The survey study of Stueber *et al.* (1972) demonstrated that a significant variability in $^{87}\text{Sr}/^{86}\text{Sr}$ ratios does exist in Ohio waters and that this variability shows a correlation with type of bedrock and overburden. The next step in evaluating the usefulness of the $^{87}\text{Sr}/^{86}\text{Sr}$ parameter as a tracer of water provenance in Ohio was a study of a single drainage basin underlain by different rock types.

The Scioto River drainage basin, located in central and south-central Ohio (fig. 1), was chosen for the study. Its watershed, the second largest within the state, drains an area of 6517 square miles, almost 16 percent of the state's area. The boundary marking the southern limit of Pleistocene glaciation crosses through the lower portion of the basin (fig. 1). North of this boundary, the bedrock consists primarily of Silurian- and Silurian-Devonian-age carbonate rocks (Niagaran and Monroe) west of the Scioto River, and Devonian and Mississippian limestones, shales, and sandstones (Columbus and Delaware Limestones, Olentangy and Ohio Shales, and Waverly Group) east of the Scioto. South of the glacial boundary, the basin is underlain almost entirely by clastic sediments of Mississippian age (Waverly Group). A relatively flat, glaciated till plain occupies the northern two-thirds of the basin where glacial drift, predominantly of Wisconsin age, ranges in thickness from zero to several hundred feet.

The present study is an attempt to determine whether variations exist in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the surface waters of the Scioto River basin and an attempt to evaluate the effects of rock types within the basin upon these ratios. Another objective of this study was an attempt to evaluate the effect of the input of strontium from tributaries on the isotopic composition of the strontium already present in the Scioto River.

ANALYTICAL PROCEDURES AND RESULTS

Water samples were obtained from the Scioto River at 16 locations distributed over the entire length of the river, from its source near Foraker to its confluence with the Ohio River at Portsmouth. Single samples taken from each of 15 tributaries of the Scioto River were also collected. The locations of all sampling points are shown in Figure 1. The Scioto River was sampled both above and below the point at which a tributary enters it, whereas the tributaries were sampled just above their confluence with the Scioto. This procedure was employed in order to monitor the effect of the input of strontium from a tributary upon the strontium of the Scioto River.

Two separate samples of water were obtained at each collection point. A

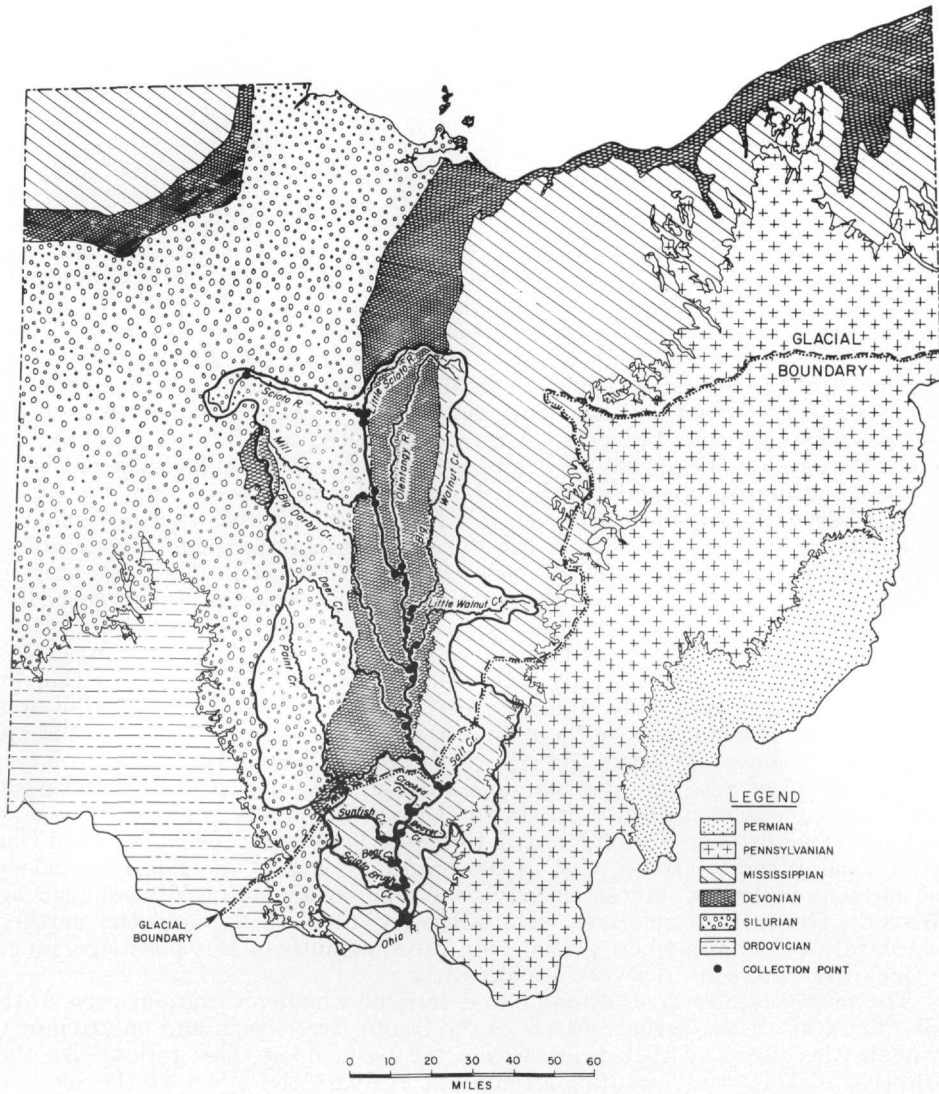


FIGURE 1. Geologic map of Ohio, showing the location of the Scioto River drainage basin.

500-ml portion was collected for the purpose of determining the total strontium concentration, and a two-liter portion was collected for the strontium-isotopic analysis. All samples were stored in polyethylene containers which had been rinsed three times with the water to be collected before the sample was taken.

The samples were collected on two days in early August, 1971, during a period of relatively light rainfall, in order to minimize the contribution of direct surface runoff to the water in a stream. Three additional tributaries south of the glacial boundary were sampled in mid-December, after it became apparent that this boundary had a controlling effect on the isotopic composition and concentration of strontium in the surface waters. Scioto Brush Creek, which had been sampled in August, was sampled again in December in order to evaluate the variability of strontium in a single tributary during this interval.

The concentrations of strontium were determined by atomic-absorption spectrophotometry, according to the method of Fishman and Downs (1966). The analyses were carried out on a Perkin-Elmer Model 303 unit. The U.S.G.S. standard water-reference sample number 26 was run with each set of water samples analyzed as a test of the accuracy and reproducibility of the total-Sr determinations. The values obtained compare favorably with the accepted value, and are essentially the same as those reported by Stueber *et al.* (1972).

The separation of strontium from the water samples was carried out by cation-exchange chromatography, and the isotopic compositions were determined on a solid-source mass spectrometer with a 12-inch radius of curvature, using double rhenium filaments. A detailed description of these analytical methods was presented by Stueber *et al.* (1972).

Twenty-three water samples were analyzed for Sr concentrations and for $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios. The results for the Scioto River samples, listed in order of sampling from source to mouth, are presented in Table 1. The results of the analyses of the tributaries of the Scioto River are presented in Table 2. These

TABLE 1
Analyses of the Scioto River

Sample Location	$^{87}\text{Sr}/^{86}\text{Sr}^*$	Sr (mg/l)
Near Foraker, Hardin Co.	0.7084	5.70
Above Little Scioto R.	0.7082	4.38
Below Olentangy R.	0.7086	1.98
Below Big Walnut Cr.	0.7086	1.05
Below Paint Cr.	0.7085	1.06
Above Scioto Brush Cr.	0.7084	0.42
Above Ohio R.	0.7090	0.33

*Normalized to $^{88}\text{Sr}/^{86}\text{Sr}=8.375$.

TABLE 2
Analyses of Tributaries of the Scioto River

Tributary	$^{87}\text{Sr}/^{86}\text{Sr}^*$	Sr (mg/l)
Tributaries North of Glacial Boundary		
Mill Cr.	0.7081	4.65
	0.7079	
Big Darby Cr.	0.7078	4.37
Deer Cr.	0.7080	1.96
Paint Cr.	0.7079	0.56
Little Scioto R.	0.7080	8.00
Olentangy R.	0.7084	1.16
Big Walnut Cr.	0.7087	0.37
Little Walnut Cr.	0.7084	0.90
Tributaries South of Glacial Boundary		
Crooked Cr.	0.7139	0.13
Sunfish Cr.	0.7110	0.19
Bear Cr.	0.7105	0.04
Scioto Brush Cr. (collected 8/71)	0.7147	0.05
(collected 12/71)	0.7149	0.04
Salt Cr.	0.7127	0.04
Beaver Cr.	0.7136	0.06
Ohio R. (N. of confluence)	0.7116	0.22

*Normalized to $^{88}\text{Sr}/^{86}\text{Sr}=8.375$.

have been grouped according to position with respect to the glacial boundary for convenience in later discussion.

The accuracy of the isotopic data was evaluated by analyzing the inter-laboratory Eimer and Amend SrCO_3 standard. Six analyses performed during the course of this study yielded a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.7079 ± 0.0001 , which is in excellent agreement with analyses reported by other laboratories. The reproducibility of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios is indicated by the agreement of the duplicate analyses, including chemical processing, of the Mill Creek water sample (Table 2). Measured $^{87}\text{Sr}/^{86}\text{Sr}$ ratios which differ by one part in 700 or more are clearly significant beyond the limits of experimental uncertainty.

Because three water samples from tributaries were obtained in December, 1971, several months after the other water samples had been collected, it was necessary to demonstrate that seasonal effects on the isotopic composition of strontium in surface waters in this area were negligible. Eastin and Faure (1970) analyzed Sr from the Olentangy River at Columbus four times in a single year, and found that the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio varied from 0.7086 to 0.7116. Scioto Brush Creek, which is located in the area of our later sampling, had been sampled initially in August, 1971, and was sampled again in December. The total strontium content of Scioto Brush Creek was essentially the same at both times (Table 2). The isotopic composition of the two strontium samples was identical, within experimental error; in fact, the reproducibility of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio was as good as that of a single water sample (Table 2, Mill Creek). These results support the validity of direct comparison of analyses of water samples obtained south of the glacial boundary in August and in December, 1971.

DISCUSSION

The Scioto River

The total strontium concentration of the Scioto River decreases rapidly downstream, from 5.70 mg/l near its source to less than 0.5 mg/l at its mouth (Table 1). This rapid decrease can be correlated with the types of rocks contacted by waters of the Scioto River system. Carbonate bedrock relatively high in strontium predominates in the upper reaches of the drainage basin, whereas low-strontium clastic sediments become increasingly important downstream. However, the concentration of strontium in the Scioto River near its source is unusually high, compared to that of most North American rivers. This fact was pointed out by Eastin and Faure (1970), who showed that the ratio of concentration of strontium to calcium in the Scioto River at Columbus was also unusually high. They suggested that most of the strontium was derived from weathering of the glacial till, which may contain significant amounts of the mineral celestite (SrSO_4), transported southeastward by the Wisconsin glaciers from the Bass Islands-Sandusky area of Ohio.

Feulner and Hubble (1960) found unusually high strontium concentrations in surface waters of Champaign County, located partially within the Scioto basin, which they attributed to the weathering of celestite in the Silurian bedrock and in local glacial materials. It is probable that the weathering of celestite is primarily responsible for the unusually high Sr concentrations found in the upper portions of the Scioto River.

With the exception of one sample, the isotopic composition of strontium in the Scioto River is constant, within experimental error, over its entire length (Table 1). The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of about 0.7084 is typical of the isotopic composition of strontium in Paleozoic marine carbonate rocks (Peterman *et al.*, 1970). It is probably also representative of the celestite, which is associated with marine carbonates and evaporites, and should have inherited its $^{87}\text{Sr}/^{86}\text{Sr}$ ratio from Paleozoic oceans. The strontium present in carbonate material and in celestite is leached more rapidly by ground waters than is the more radiogenic strontium present

in detrital-silicate components of clastic sediments. This fact, coupled with the predominance of carbonate- and celestite-rich rocks in the Scioto basin, is probably what produces the observed constancy of low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the Scioto River.

The $^{87}\text{Sr}/^{86}\text{Sr}$ value for the Scioto River just above its confluence with the Ohio River is slightly higher than the values obtained elsewhere (Table 1). The difference is significant beyond experimental error and is thought to be due to a mixing of Scioto and Ohio River waters, since the sample was obtained quite near their confluence. Analysis of a sample of the Ohio River obtained just above its confluence with the Scioto yielded a relatively high $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.7116, so that if Scioto and Ohio River waters were to be mixed, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the Scioto near Portsmouth could quite conceivably be raised to a value of 0.7090.

The data for the Scioto River are presented graphically (fig. 2) by plotting $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and total Sr concentrations as a function of the location of the collection sites downstream from the source of the river. The $^{87}\text{Sr}/^{86}\text{Sr}$ data are represented by a horizontal line in Figure 2, indicating the constancy of the

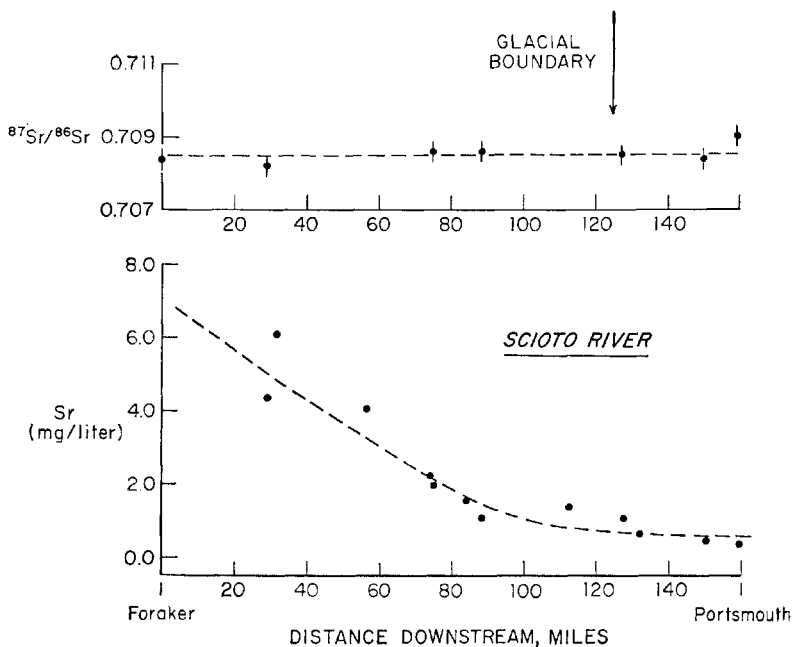


FIGURE 2. Strontium in the Scioto River.

strontium-isotopic composition. No observable change in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio appears at the position of the Pleistocene glacial boundary, indicated by a vertical arrow. The exponential decline in the Sr concentration down the Scioto River suggests that the Sr content, and therefore presumably the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the Scioto, has been established near its source. The contribution of relatively radiogenic strontium by tributaries which drain from areas of clastic sediments farther downstream has apparently not affected the observed isotopic composition of the Scioto River.

Scioto River Tributaries

In the northern portion of the Scioto basin, the strontium concentrations in the tributaries are unusually high (Table 2). Such anomalous values are probably

due to the weathering of celestite in the Silurian carbonate bedrock and glacial till. The highest Sr concentrations occur in the northernmost tributaries, the concentrations generally decreasing in a southerly direction. The amount of celestite in the glacial till should also decrease in a southerly direction within the Scioto basin, because the principal source was probably in the Bass Islands-Sandusky area of northern Ohio (Eastin and Faure, 1970). The strontium concentrations of tributaries south of the glacial boundary are significantly lower than are those of other tributaries (Table 2), and are more comparable to those found in most North American rivers (Skougstad and Horr, 1963).

In contrast to the constant isotopic composition of strontium in the Scioto River, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of its tributaries (Table 2) show considerable variability, ranging from 0.7078 to 0.7147. This range reflects the input of strontium leached from detrital components of clastic bedrock present in the Scioto basin, and is comparable to the variability of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios found in surface waters across the entire state (Stueber *et al.*, 1972).

A comparison of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the tributaries with the nature of the underlying bedrock reveals some anomalous relationships. The first five tributaries listed in Table 2 enter the Scioto from the west and north and are underlain by carbonate bedrock and glacial till. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of these five streams are quite consistent and indicate the predominance of strontium derived from carbonate-type sources. The next three tributaries listed in Table 2 enter the Scioto from the east and are underlain by predominantly clastic bedrock, overlain by glacial till. The isotopic compositions of their waters, however, do not indicate the presence of strontium leached from clastic sediments. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are essentially the same, within experimental uncertainty, as those found in the tributaries to the west. Thus strontium from local bedrock does not determine the strontium-isotopic compositions of these streams; another strontium source must exist. It would seem that this source must be the glacial till, and there is indeed a relationship between tributary strontium and the position of the glacial boundary.

The tributaries south of the glacial boundary (Table 2) are not underlain by glacial till, and drain predominantly clastic Devonian and Mississippian bedrock. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of all these streams are significantly higher than are those of tributaries north of the glacial boundary. The higher ratios must be due, at least in part, to strontium which has been leached from the underlying clastic bedrock. The total strontium concentrations of these tributaries are quite low, especially when compared with the Sr concentrations of tributaries which are underlain by the same clastic sediments only a few miles north of the glacial boundary.

According to Flint (1971, p. 174), it has been established that much of the coarse fraction of glacial till consists of material of fairly local lithology, and thus it is believed that this till is predominantly of local origin. Therefore it is probable that the glacial till of the Scioto basin contains large amounts of carbonate material, due to the abundance of carbonate bedrock in the basin. Eastin and Faure (1970) cited a study by Schmidt (1958), which showed that more than 80 percent of the pebbles in the till in the vicinity of Columbus are dolomite. The carbonate in the till, as well as the celestite discussed previously, provide sources of readily soluble strontium which establish the isotopic composition of the surface waters north of the glacial boundary, regardless of the nature of the underlying bedrock. Any contribution of radiogenic ^{87}Sr leached from the minor Precambrian components of the till is masked by strontium from the more abundant carbonate and celestite.

The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and total Sr concentrations obtained for the tributary streams have been plotted in a manner similar to that employed for the Scioto River data (fig. 3). The influence of the position of the glacial boundary on these parameters is strikingly evident. The tributaries north of the glacial boundary

all have relatively constant, carbonate-type $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, regardless of whether the bedrock is predominantly carbonate or predominantly clastic in nature. South of the glacial boundary, however, the tributaries have significantly higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and thus do reflect the presence of strontium leached from the underlying clastic sediments. Thus the position of the glacial boundary appears to be the controlling factor in determining the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of tributary waters in the Scioto basin.

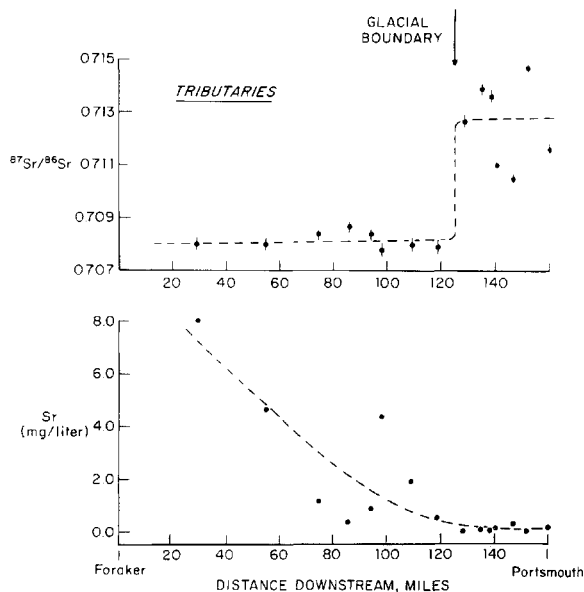


FIGURE 3. Strontium in tributaries of the Scioto River.

The total Sr concentrations of the tributaries south of the glacial boundary are extremely low when compared with those of tributaries north of the boundary (fig. 3), so that, even though these tributaries have relatively high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, their input has no effect upon the observed isotopic composition of the Scioto River, which retains a low carbonate-type $^{87}\text{Sr}/^{86}\text{Sr}$ ratio throughout its length (fig. 2). The only detectable effect of the input of these tributaries is a dilution of the total strontium concentration of the Scioto River.

The $^{87}\text{Sr}/^{86}\text{Sr}$ Ratio as a Tracer of Water Provenance

The use of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio as a tracer of water provenance in the Scioto basin depends upon a demonstrated variability of this parameter in surface waters and a relationship between such variability and the nature of the bedrock within the basin. The isotopic analyses of strontium in surface waters of the Scioto basin reported here do show a significant variability, but the relationships of these data with bedrock lithologies are complicated by the presence of glacial till throughout much of the basin. The dominance of glacial till over bedrock lithology in determining $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of surface waters north of the glacial boundary is shown in Figure 4, which is a bedrock lithology map of Ohio on which the Scioto River basin has been outlined. The basin has been subdivided into the drainage areas of the tributaries whose waters have been analyzed for their strontium-isotopic compositions.

South of the glacial boundary, where till is absent, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in surface waters indicate the presence, to some degree, of strontium which has been leached from the detrital Rb-bearing components of the underlying clastic bedrock. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in all surface waters north of the glacial boundary, however, indicate the presence of strontium dissolved only from carbonate material, although carbonate bedrock is present to the west of the Scioto River, whereas clastic bedrock predominates to the east. Where glacial drift is present, readily soluble carbonate material and celestite in the till appear to control the strontium supplied to surface waters. Any strontium leached from underlying clastic bedrock will be overwhelmed by strontium from the till and will have no detectable effect on

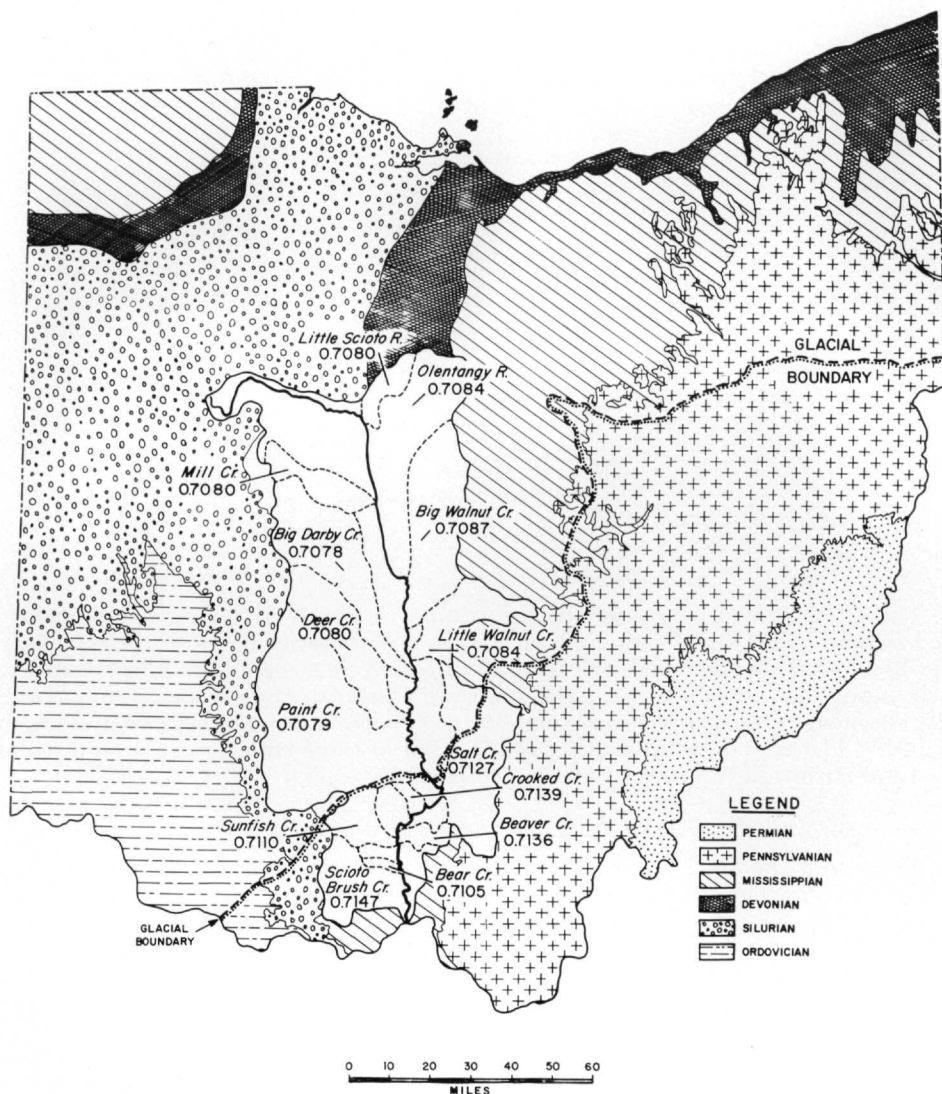


FIGURE 4. Geologic map of Ohio, with $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of surface waters in the Scioto River basin. The basin has been subdivided into the drainage areas of the tributaries whose waters have been analyzed.

the observed isotopic compositions. Where carbonate bedrock is overlain by glacial till, both lithologies will presumably contribute significant amounts of strontium to surface waters.

These relationships indicate that the application of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio as a tracer of water provenance in Ohio should be limited to unglaciated terrain where both clastic and carbonate bedrock are present. Unfortunately, such a geologic setting is not present within the Scioto River basin (fig. 1).

CONCLUSIONS

Investigation of strontium in surface waters of the Scioto River drainage basin, Ohio, has revealed unusually high total-strontium concentrations for the Scioto River and its tributaries in the northern portion of the basin, probably due to weathering of celestite in the Silurian carbonate bedrock and overlying glacial till. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the Scioto River water is essentially constant throughout its length at a value of about 0.7084, due to the dominating presence of strontium weathered from carbonate and celestite sources. Scioto River tributaries, however, show a relatively wide range of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, but the distribution of this variability is more dependent upon the position of the sampling relative to the Pleistocene glacial boundary than upon the bedrock lithology within the basin. The usefulness of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio as a tracer of surface water provenance in the Scioto River basin is therefore limited by the presence of readily soluble Sr-bearing materials in the glacial till which covers much of the basin. Future work of this type in Ohio should be confined to unglaciated terrain where both carbonate and clastic bedrock are present.

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