RADIOACTIVITY IN COAL MINE DRAINAGE

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Natural Radioactivity in the Kiskiminetas River

Although only required by regulation to monitor their effluents, most health groups at nuclear facilities also monitor the off site environment. They do this to (1) assure themselves that their effluent monitoring is effective, (2) demonstrate permissible off site exposure and (3) provide a background base in case of an accidental release. So, along with air, fallout and vegetation monitoring, we sample our receiving stream, the Kiskiminetas River, at weekly intervals.

NUMEC's nuclear fuel plants are located about 30 miles northeast of Pittsburgh. Figure 1, which gives the major physiographic divisions of Pennyslvania[1], shows the Kiskiminetas River flowing into the Allegheny River just north of Pittsburgh. Our plutonium plant is about 10 miles up the Kiski, near Leechburg, the uranium plant is in Apollo, five miles further up stream. Western Pennsylvania is part of the Appalachian Plateau and is characterized by high, flat-topped divides separated by steep-sided valleys in which flow deeply entrenched streams. The rocks beneath the plateau are a uniform succession of Carboniferous and Devonian sediments and have under gone little folding or faulting. Coal mining was once the principal and is still an important industry.

We sample the Kiskiminetas at three bridges, one above our uranium plant, one between the plants, and another below the plutonium plant. The 1966 average concentrations are given in Table I.

Table I

Alpha Activity Levels

Kiskiminetas River - 1966

Location	Average Concentration pCi/liter	Concentration Range pCi/liter
Apollo	13.14	.3 - 102.7
Vandergrift	13.36	.45 - 101.0
Leechburg	10.77	.9 - 46.0

This table, and all other data in this report, gives only alpha radioactivity levels. Our interest is primarily in alpha radiation, since nuclear fuel nuclides are alpha emitters.

The averaged gross alpha concentrations are higher than for most eastern United States Rivers[2]. Since the average flow in the Kiski River is more than 3,000 cubic feet per second, the Apollo concentration represents about 100 millicuries per day. This is 20 times the radioactivity that we are permitted to release per day from our combined plants.

The range in concentrations from tenths of a picocurie per liter to more than 100 picrocuries per liter intrigued us. Apparently the upstream source was fluctuating. Figure 2 shows the monthly averaged concentration for all three locations. We obtained the monthly average flow rate from the Hydrology Section of the Army Corps of Engineers, Federal building, Pittsburgh, Pennsylvania. The calculated millicuries per day shows that the total activity per day is a stronger function of flow rate than concentration.

The mystery of the upstream radioactivity proved irresistible, so in May, 1966, we took a two day survey by canoe of the Kiski Watershed. We repeated the survey in

June, 1967 and also conducted surveys of the west branch of the Sesquehanna River in August and November of 1967.

Survey and Analytical Methods

The Kiski area contains a number of coal mines, whose drainage creates a high acid content (pH = 2 - 5) in the Kiskiminetas River. Several geological publications[3,4,5] have described the association of uranium and coal in Western Pennsylvania. Estimates of uranium in coal ranged from 10 to 140 parts per million. Since coal mine drainage might be the source of the stream's radioactivity, we planned to survey water from several mines.

Figure 3 shows the detail of our 1966 survey. We took river samples from the Allegheny to the Loyalhanna reservoir. All large side streams were sampled and samples were obtained directly from two mines and a boney pile. The term, boney pile, refers to the shale overcover removed from coal.

Most of the samples were obtained by canoe or flat-bottomed fishing boat. The Kiski has a shallow draft from late spring to fall.

We collected one liter samples at each location. The acidity of each sample was tested with wide range pH test strips. Most mines and side streams showed high acidity. The Kiskiminetas River is one of the most acid rivers in Pennsylvania[6].

The samples were analyzed for gross alpha radioactivity following the approximate procedures used by the Public Health Services[7]. The suspended matter was removed by centrifuging and filtration. The filtered water samples were evaporated and transferred to stainless planchets for counting. Prior to evaporation the samples were treated with 1 normal HNO_3 to digest organic matter. Beaker rinses were combined with the sample concentrate and evaporated to dryness. The planchets were flamed to dull red, cooled and weighed. Alpha counting was done in an alpha scintillation counter (Eberline SAC-3). Correction was made for self absorption for samples exceeding 10 mg/cm^2.

Suspended solids in the samples were also analyzed for alpha activity. While self absorption uncertainies were present, most of the activity appeared in the dissolved solids.

Results of River Surveys

The results of the May, 1966 survey of the Kiski watershed are given in Table 2.

There are striking radioactivity concentrations in coal mine drainage. The levels are appreciable considering that the MPC_w for unidentified radionuclides is 10 pCi/liter. We subjected several samples to radiochemical analysis and found the radioactivity predominantly from $^{234, 238}U$; less than 10% was from ^{226}Ra.

The variation in concentration among the various samples is interesting. River samples 1 and 20, stream samples G, H, I, K, N and O are all upstream from mine drainage and all low in gross alpha activity.

We repeated the Kiski watershed survey in June, 1967. The results are given in Table 3. We took only side stream samples on this survey, attempting to gain additional detail over our 1966 survey. While it is not as dramatic as the 1966 data, the 1967 data confirmed the high concentrations in coal mine drainage.

Our review of the Pennsylvania geological literature indicated that other watersheds might produce radioactivity in mine drainage. Consequently, we surveyed the west branch of the Sesquehanna River in August and November 9, 1967. Results of this survey are in Table 4. It is immediately apparent that the Sesquehanna watershed is much less radioactive. However, this may be misleading since many of the samples were collected following heavy rains. In any case, the increased activity in some of the coal mine drainage is evident.

Origin or Radioactivity in Coal Mine Drainage

Several authors[3,4,5,7,8,10,11] have referred to the association of uranium and coal. Large reserves of uranium-bearing coal and lignite are known in western North and South Dakota and southeastern Montana. In 1954, considerable tonnages of ore-grade uraniferous lignite were discovered in the Cave Hills area of western South Dakota.[10] These western United States discoveries led the United States Geological Survey to explore eastern coal reserves for uranium occurrences.[4,5]

Uranium was found in uneconomic concentrations in Pennsylvania coal. Estimates ranged from 10 to 140 parts per million, although some eastern Pennsylvania sedimentary deposits contain up to 0.3 percent U_3O_8.[3] McCauley[3] concluded that only the absence of an eastern ore concentrating mill and the dispersal of the mineral leases among a large number of individuals prevents the economic mining of uranium in Pennsylvania.

The coexistence of uranium and coal is explained by the descending meteoric hypothesis for uranium occurrences.[3] Downward moving solutions leached small amounts of uranium and other metals from syngenetic accumulations in the over-lying sediments and these metals ultimately were precipitated in carbonaceous zones by organically produced hydrogen sulfide.

Figure 4 shows a simple scheme to explain radioactivity in coal mine drainage. Uranium concentrated in the coal and shale overcover is exposed to weathering and ground water during mining. The acid drainage leaches the uranium out of the coal bed. The uranium leaves the coal bed, enters the watershed and redeposits at a favorable location downstream.

The Significance of Radioactivity in Coal Mine Drainage

Coal mine drainage has severely affected the Pennsylvanian ecology. Acid mine water has destroyed the life of most our state's streams. Whether the presence of radioactivity in coal mine drainage adds an increment of disadvantage is problematic. The Kiski and other mine polluted streams are not directly used for public water supplies. The first user of water from the Kiski watershed is the city of Pittsburgh which obtains much of its water from the Allegheny River. Hursh[2] demonstrated that water treatment effectively removed radium from the Allegheny River.

The presence of uranium in coal itself may be important to public health. Certainly, coal mines should be surveyed for radon daughter air activity. Thousands of tons of coal are burned in plants each year. While it has been demonstrated that most of the uranium remains in the ash,[4] ^{226}Ra or other uranium daughters may be dispersed from coal-burning stacks. We have detected higher gross alpha fallout near coal burning power stations or closer to highways where coal ashes have accumulated.[12]

From a regulatory viewpoint, the coal mine drainage is interesting. Several mines in our study exceeded the permissible concentration for ^{226}Ra. The whole Kiskiminetas River usually exceeds the Public Health Service Guide of 3 alpha disintegrations per liter. It is certainly useful to NUMEC to demonstrate its innocence for the high Kiski concentrations. Nuclear facilities located in coal mining areas would be well advised to sample routinely above and below their discharge points in receiving streams.

Probably the most significant feature of the radioactive coal mine drainage is geologic. More than 30 tons of uranium per year has been leached from Kiski coal mines during the past 50-60 years. This uranium may be depositing in favorable locations in the Kiski or Allegheny River beds. Man has accelerated the natural leaching and reconcentrating cycle. Some distant generation of uranium users may benefit.

Table 2

Kiskiminetas River Alpha Radioactivity Survey

May, 1966

Sampling Point*	Concentration pCi/liter	Remarks
River 1	2.7	Allegheny River
" 2	18.9	
" 3	26.9	
" 4	45.2	
" 5	25.5	
" 6	28.2	
" 7	142.7	Old mine drainage
" 8	16.8	
" 9	33.6	
" 10	17.3	
" 11	46.4	Downstream of principal mine drainage
" 12	119.1	" " " " "
" 13	81.8	" " " " "
" 14	11.8	
" 15	3.6	
" 16	14.5	
" 17	16.4	
" 18	17.3	
" 19	12.7	
" 20	1.8	Upstream of all mines
Stream A	32.7	Mine drainage
" B	30.5	" "
" C	21.6	
" D	29.3	
" E	20.5	
" F	12.7	
" G	1.0	
" H	2.9	
" I	1.0	
" J	163.6	Downstream of Boney Pile
" K	1.0	
" L	10.0	
" M	15.5	
" N	4.5	Upstream of all mines
" O	3.4	" " " "
" P	17.3	Reservoir, some mine drainage
Mine # 1	174.1	
Mine # 2	120.0	
Boney Pile	180.0	Shale overcover of coal

* See figure 3 for sampling point location

Table 3 (Watershed)

Kiskiminetas, Alpha Radioactivity Survey

June, 1967

Sample Location	Concentration, pCi/liter
Conemaugh Reservoir	9.1
Mine stream, 1 mile below dam	24.5
Side stream, 2 miles below dam	15.4
Side stream, 3 miles below dam	10.0
Side stream at Deep Rapids	20.0
River, Saltsburg	16.4
Mine stream, Saltsburg	31.8
Fresh stream, 1 mile below Saltsburg	0.9
Side stream, 1.5 miles below Saltsburg	8.2
Mine stream, 2 miles below Saltsburg	24.5
Mine drainage, 2 miles below Saltsburg	14.5
Mine stream, Salina	32.7
Mine drainage, Brownstown	7.3
Mine drainage, 1 mile below Brownstown	19.1
Mine drainage, 1.5 miles below Brownstown	27.3
Beaver Run, W. Apollo	9.1
Mine Stream, N. Apollo	41.8
Pine Run Creek	6.4
Side stream, River Road	8.2
Mine Drainage, N. Vandergrift	12.7
Stitt's Run	11.8
Storm Culvert #1	3.2
Storm Culvert #2	3.6
Hungry Hollow Creek	18.2
Fresh Water Spring, Leechburg	Bkg.
Mine stream, Leechburg	60.0
Mine stream, Baghdad #1	21.8
Mine stream, Baghdad #2	53.6
Strip Mine, 2 miles above Allegheny River	32.7
Strip Mine stream	30.9
Side stream Kiski Junction	11.8

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Table 4

West Branch, Sesquehanna River Watershed

Alpha Radioactivity Survey

August, November 1967

Sample Location	Concentration, pCi/liter
Mine drainage, W. Decatur	Bkg.
Moshannon Creek, Phillipsburg	1.66
Moshannon Creek, Route 153	0.77
Lick Run, Shawsville	0.11
Deep Mine drainage, Osceola Mills	14.55
Sesquehanna River, Clearfield (above Mine pollution)	Bkg.
Sesquehanna River, above Shawsville power station	Bkg.
Sesquehanna River, below Shawsville	0.31
Trout Run, Mine stream	0.31
Mine Settling Ponds, Karthaus	2.47
Mosquito Creek, unpolluted	Bkg.
Deer Creek, unpolluted	Bkg.
Shintown Run, Mine Stream	Bkg.
Kettle Creek, Mine Stream	0.33
Cooks Run, Mine Stream	2.66
Strip Mine, Chilcote farm	8.06
Indian Caverns	Bkg.

Figure 1 - Physiographic divisions of Pennsylvania. The Appalachian Plateau is heavily mined for coal.

Figure 2 - Natural radioactivity levels in the Kiskiminetas River. Individual data points are averages for a given month.

Figure 3 - Sample locations for May, 1966 survey of Kiskiminetas watershed. Refer to Table 2 for results.

Figure 4 - Origin of radioactivity in coal mine drainage. Trace uranium, concentrated in coal zones, is leached by acid drainage into watershed.

PHYSIOGRAPHIC DIVISIONS OF PENNSYLVANIA

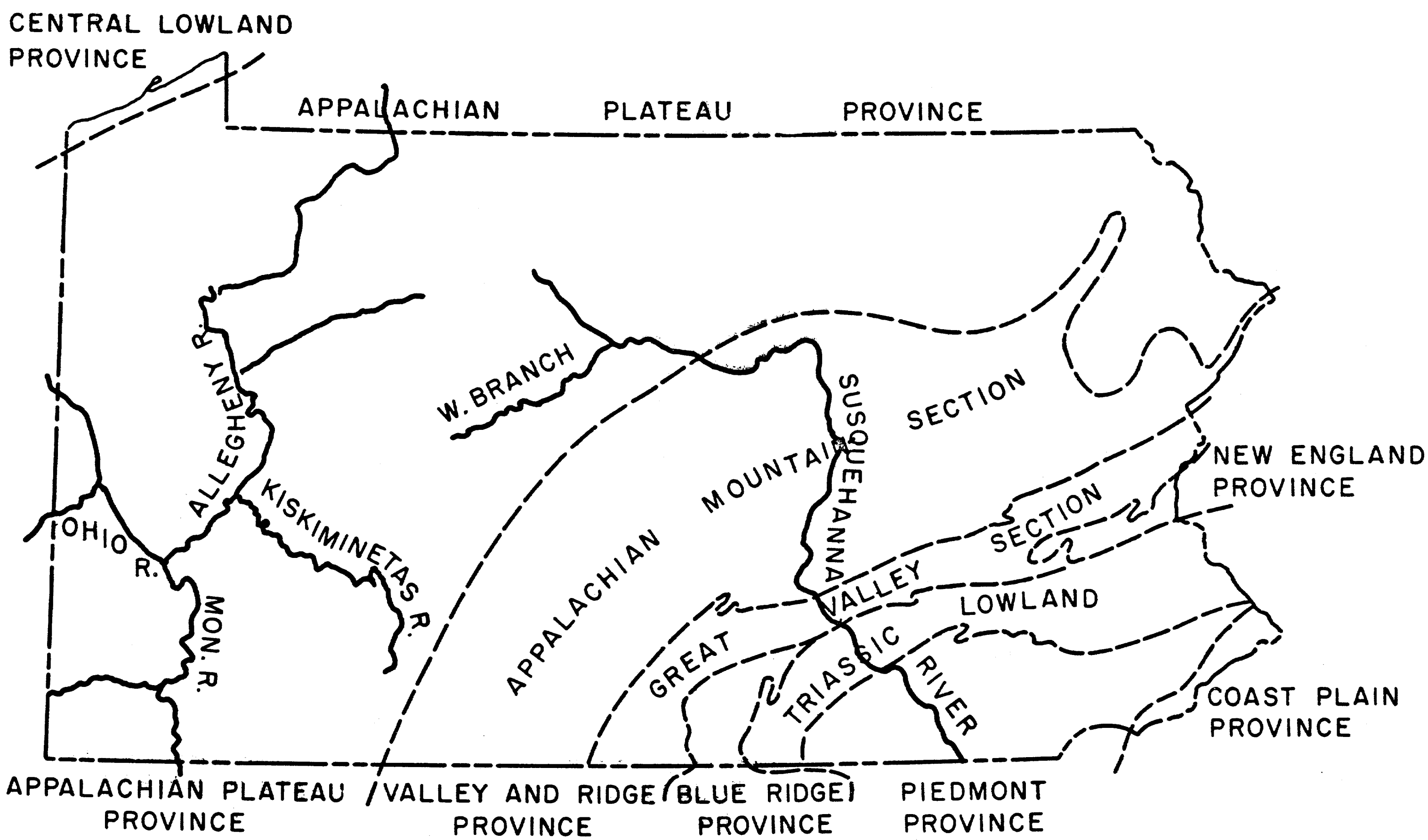

Figure 1

ALPHA RADIOACTIVITY IN THE KISKIMINETAS RIVER

July, 1966 - July, 1967

APOLLO-VANDERGRIFT-LEECHBURG

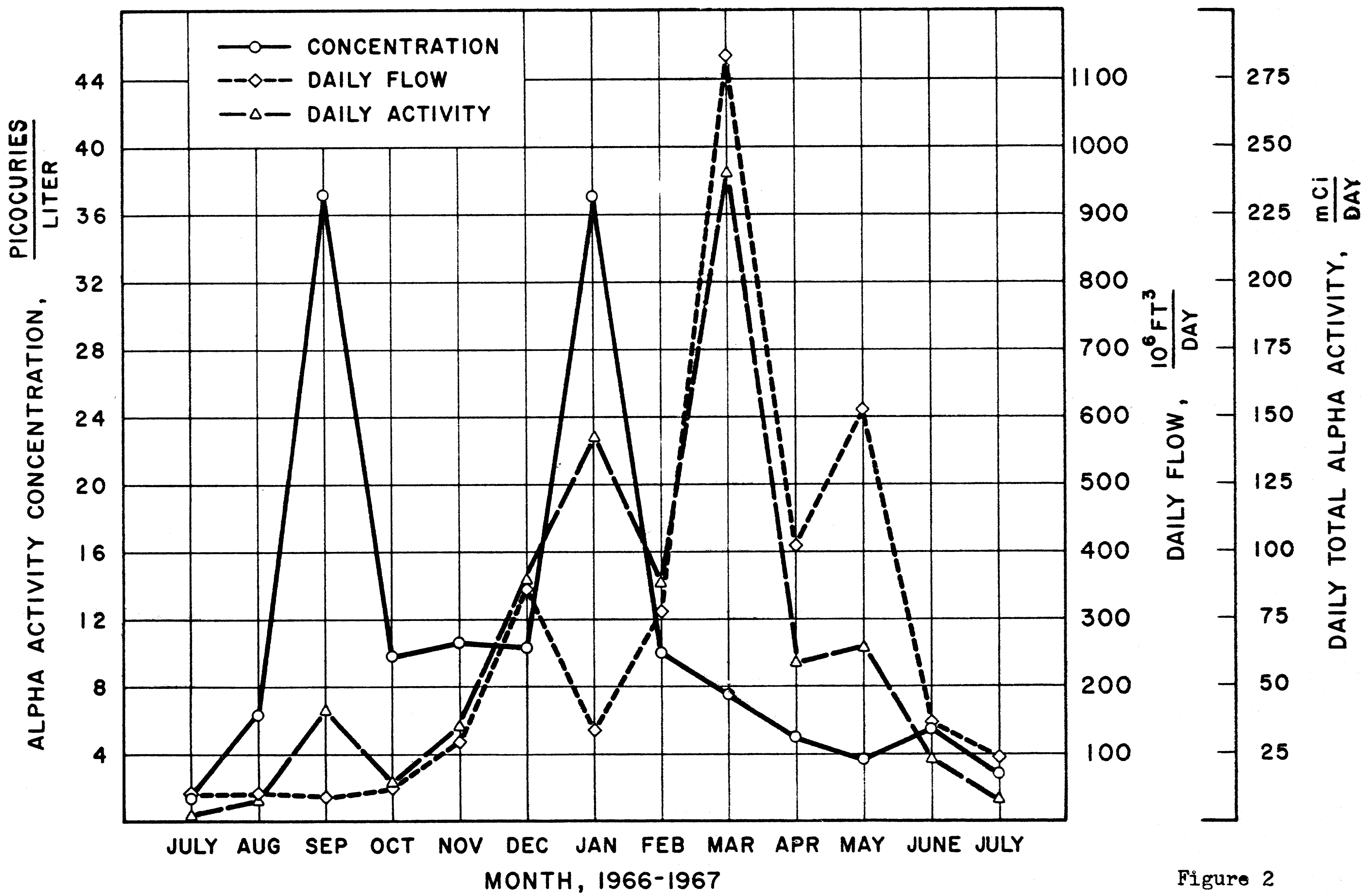

Figure 2

RADIOACTIVITY SURVEY - KISKIMINETAS RIVER

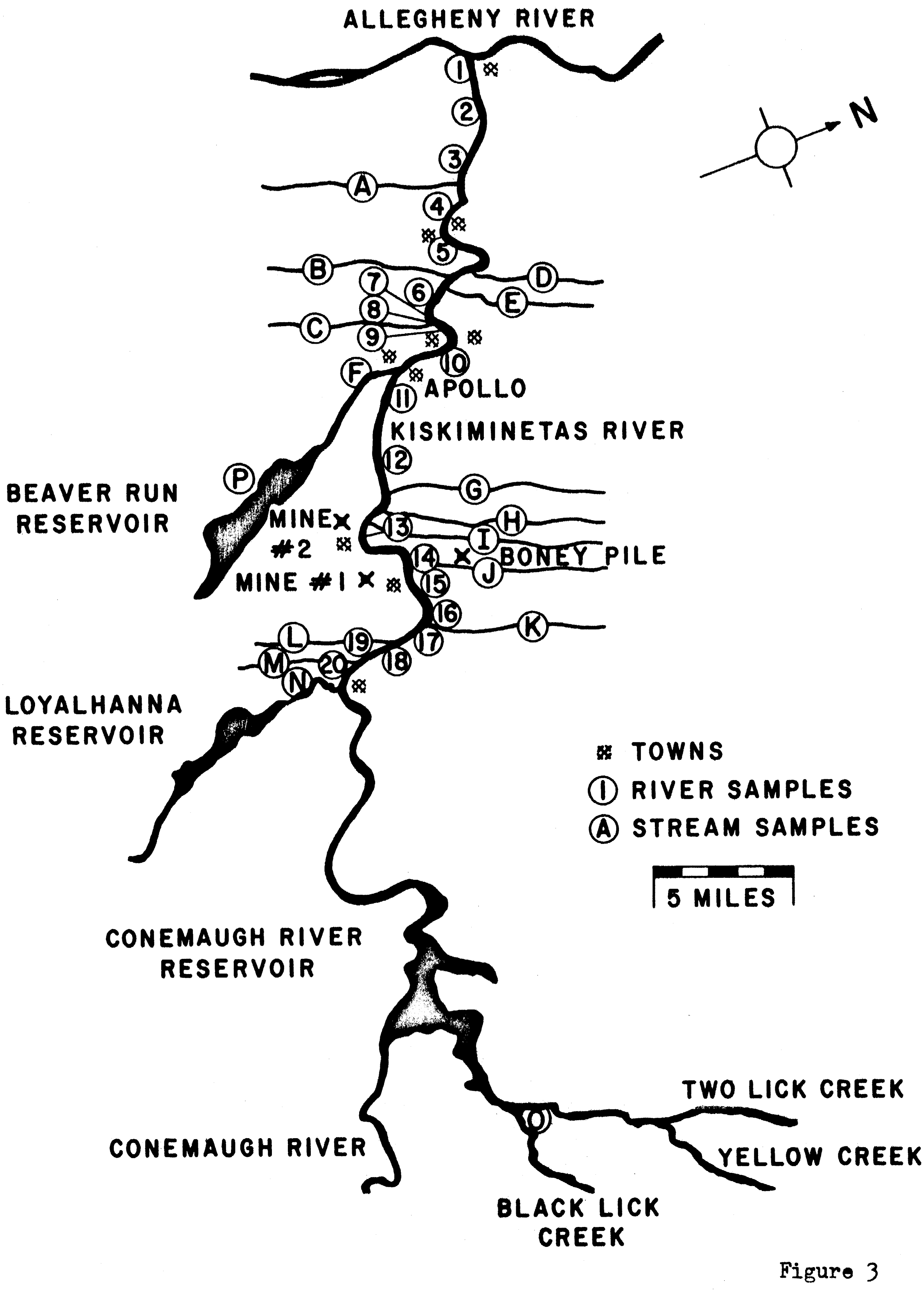

Figure 3

RADIOACTIVITY IN STRIP MINE DRAINAGE

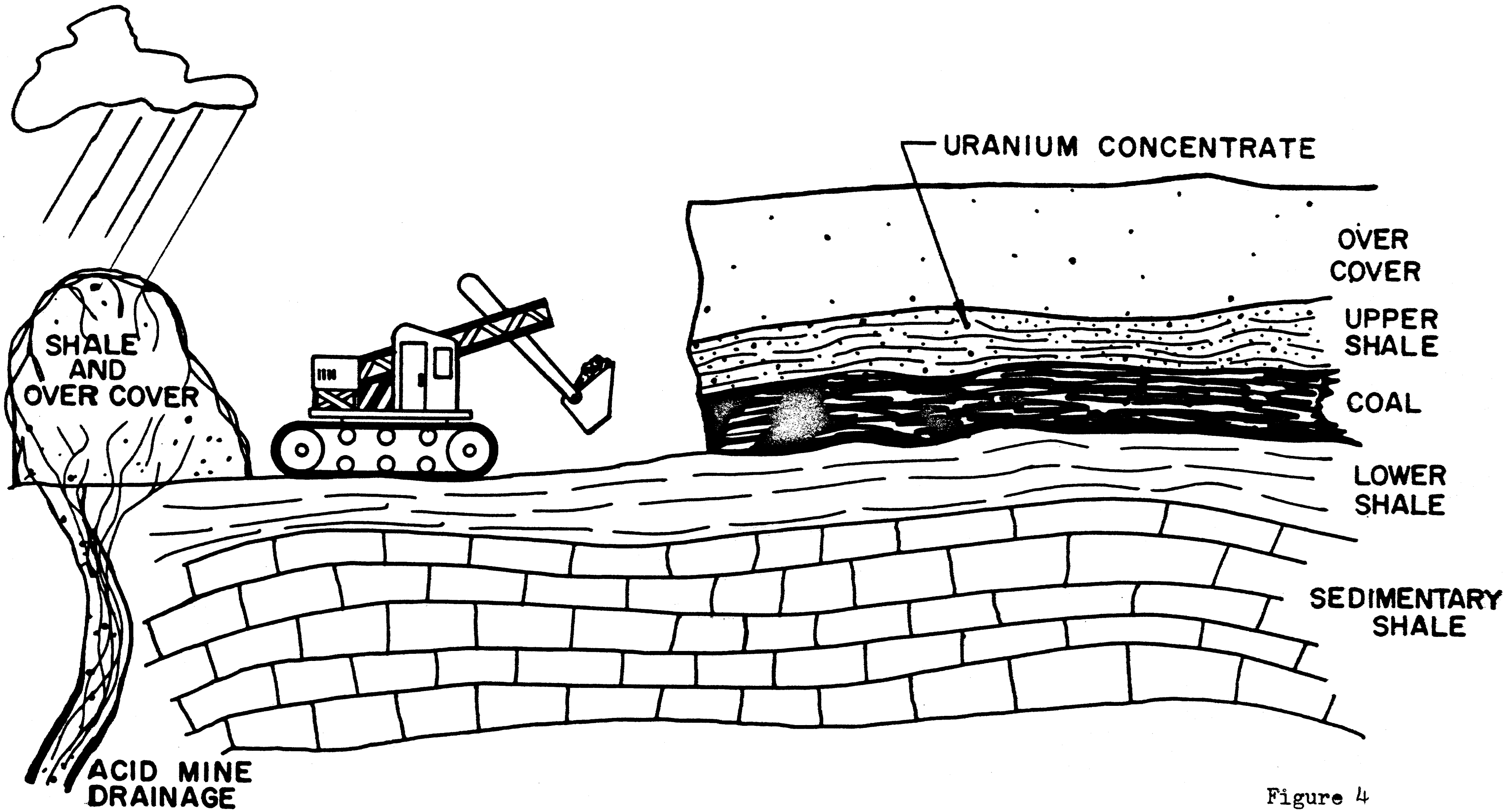

Figure 4