Research and Development

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## **Project Summary**

# Coal Mine Related Stream Sediment Geochemistry in Southern Illinois

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The stream sediment geochemistry of several elements, specifically cadmium. cobalt, copper, iron, manganese, nickel and zinc, has been examined in a sixcounty area of Southern Illinois. This area has been greatly impacted by coalmining activities. Arsenic and chromium were also investigated, but only in isolated areas. The survey of these elements in stream sediments indicates that in most areas having four times the median value for cadmium, cobalt, iron, nickel and zinc, are generally located downstream from areas of coalrelated uses. In detailed study of these streams, there occur marked relationships among the pH of mine drainage water, the concentrations of metals in the water, and the concentrations of metals in corresponding stream sediment. Water with low pH reaches seasonal summer highs in metal content and can enter streams already carrying large concentrations of metals. In those parts of a mine-impacted stream where low pH is maintained, metals can be leached from the stream sediment to further increase the concentrations in water. On the other hand, if the pH of a stream is increased, metals will tend to precipitate out of the water and become part of the stream sediment.

This Project Summary was developed by EPA's Industrial Environmental Research Laboratory, Cincinnati, OH, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

### **Background**

Southern Illinois is an area that for many years has been extensively mined for coal. Coal-bearing Pennsylvanianaged rocks crop out in a broad band that curves gradually across the southern part of the state. Where the coal beds in these rocks are shallow, surface mining has been widespread. North of the shallow outcrop area, where the coal beds are deeper, underground mines are abundant. As of mid-1971, 21,987 hectares of the six-county area of this study had been affected by surface mining activities and surface disturbances related to underground mining. This amounts to three percent of the land area in these counties.

Prior to passage of various State and Federal reclamation laws, some of the mining practices used in Illinois resulted in the creation of problem sites which have generated large amounts of acid mine drainage. In the southern Illinois area, acid mine drainage typically arises in one of two general settings. First, old, coarse refuse (gob) piles and fine refuse (slurry) impoundments which have been exposed to the weathering processes have created areas of intense production of acid drainage and related pollutants. Second, there are some areas in which the overburden has a high potential for the production of acid drainage and little potential to neutralize it. Strip mines exposing such material to air and water can create large non-point source areas of acid mine drainage.

The polluted waters generated from these sources are known to carry appreciable concentrations of several potentially



toxic trace elements. Monitoring programs by the Illinois Environmental Protection Agency, the Greater Egypt Regional Planning and Development Commission, and numerous studies of individual sites, have shown that appreciable quantities of zinc, cadmium, nickel, cobalt, iron, manganese and minor amounts of copper in the streams are draining such areas. However, not all of the contaminants moved by a stream are dissolved in the water. Many are carried as part of the particulate suspended and bed load of a stream, to be picked up, dropped, or rolled along as circumstances dictate. The literature suggests that such elements may be either removed from solution to become part of the particulate load or may be dissolved from solids and reintroduced into the water, depending upon the chemical environment. Indeed, some studies suggest that much of the trace element load of a stream is carried in such a non-dissolved state. Thus, a monitoring program that examines only the dissolved portions of a polluted stream may miss large quantities of metals being transported in another manner. This study attempts to ascertain the extent of distribution of metals in the stream sediment of the mine-impacted portion of southern Illinois, and to determine the conditions under which metals move from the water to become a part of the sediment, and vice versa.

#### **Approach**

This investigation of the geochemistry of cadmium and other metals related to coal mining is restricted to a six-county area of southern Illinois. The counties involved are Gallatin, Jackson, Perry, Randolph, Saline, and Williamson (Figure 1). The amount of land that has been disturbed by surface mining and the surface effects of underground mining of coal in these counties range from over seven percent in Perry County to less than one percent in Gallatin County. Table 1 indicates the amount of surface disturbance related to coal-mining activities.

### **General Geology**

Coal-bearing formations of the Pennsylvanian age crop out across southern Illinois in a west-to-northwest-trending arc (Figure 2). Their dip is toward the north and northeast, resulting in an older-to-younger progression of formations from south to north. This simple pattern is complicated somewhat by several structural features, including several fault systems and two major

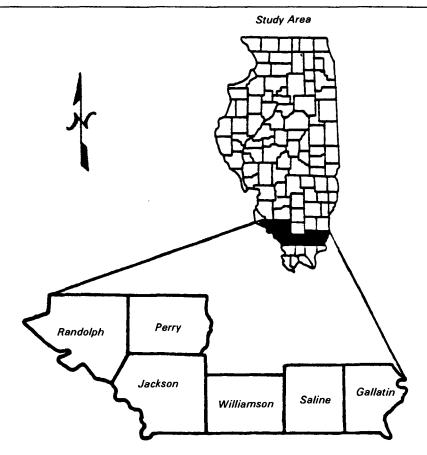


Figure 1. Location of study area.

Table 1. Areas Disturbed by Coal Mining Activities

County	Hectares in County	Area Disturbed	Percent
Gallatin	84,955	600	0.71
Jackson	156,181	1,983	1.27
Perry	114,740	8,222	7.17
Randolph	153,851	2,195	1.43
Saline	99,459	3,846	3.87
Williamson	110,596	5,140	4.65

folds. Of the fault systems within the study area, the Cottage Grove Fault System, the Wabash Valley Fault System, and the Fluorspar Area Fault System displace the coal beds on the order of a few meters to a few tens of meters. The Shawneetown Fault, on the other hand, has displacements estimated to be on the order of hundreds to thousands of meters. South of the Shawneetown Fault, the coal-bearing rocks are folded into a U-shaped outcrop pattern which has brought several of the coals to the surface in a relatively small area. When encountered,

all of these features affect the mining of the coals to some extent and at times establish the physical limits of a mining operation.

Another structural feature of major importance is the DuQuoin Monocline. Unlike the previously mentioned features, this structure was active during the time of deposition of the Pennsylvanian rocks and separated a relatively stable area which was dominated by shallow marine influences on the west (western shelf area) from one on the east which was gradually subsiding. The subsiding eastern area was the site of active delta building during the time of coal deposition. The different types of environments resulted in somewhat different overburden rocks associated with the coal beds, and this in turn has influenced the water quality in the different parts of the area. West of the monocline, there are numerous limestone beds; east of the structure limestones are sparse.

The important coal-bearing formations are the Spoon and the Carbondale Formations. The Spoon Formation consists largely of sandstone, siltstone, and gray

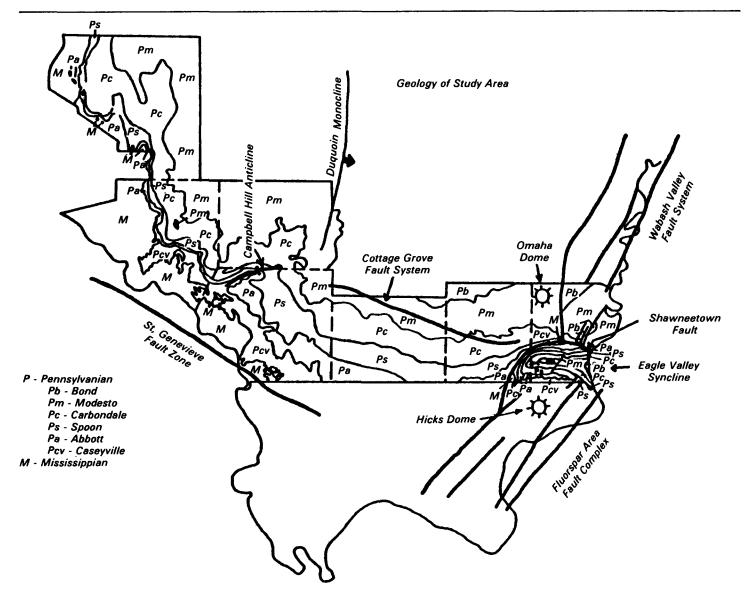


Figure 2. Geology of study area.

shale, with very little limestone. Two important coal beds, the Davis and De-Koven coals, are commercially important in the southern parts of Williamson and Saline Counties. The fact that there is only about six meters of material between these two coals allows them to be mined simultaneously. At some locations, the stratigraphic sequence between the coals is partially made up of a pyrite-rich black shale that contains up to twelve percent sulfur. When exposed at the surface, this pyritic shale can produce considerable amounts of acid. Extensive mining of the Davis and DeKoven coals has produced areas of intense pollution because of this unfortunate stratigraphy.

The Carbondale Formation overlies the Spoon Formation and differs from it by having larger proportions of limestone and shale. The limestones tend to become more abundantly higher in the stratigraphic section and west of the DuQuoin Monocline. Two important coals are also present in this formation. These are the Harrisburg (Number 5) and the Herrin (Number 6) coals. The Number 6 coal is stratigraphically above the Number 5. East of the DuQuoin Monocline, in the area of Williamson and Saline counties, the stratigraphic interval between the two coal beds is on the order of 43 meters. and the two coals are mined separately. The generally low dip of the strata in this area results in the outcrop lines of the two coals being separated by as much as 1.5 kilometers, giving rise to two series of parallel strip mines. Further to the west, particularly in the vicinity of the DuQuoin Monocline, the stratigraphic interval between these coals becomes much thinner. In Perry and Randolph counties, only about 10 meters of rock separate the beds and the two coals are commonly removed in a single mining operation.

## **Elemental Distribution Results**

The regional distribution of metals within the study area are discussed on an element-by-element basis.

#### Cadmium

In the Saline River area, there are numerous high cadmium values in the stream sediments. Most of these are concentrated along the South Fork, with another area of concentration in Eagle Creek. Two high values were noted in the westernmost part of the area; these occur downstream from a coal-fired powerplant. Another cluster of high values occurs in the northeast part of the basin, associated with underground coal mines in the area. In the Crab Orchard Creek area, numerous high values of cadmium occur. As before, clusters of high points are located downstream from coal mining activities. A pair of high values occurs in the southern part of the area. Although no mining is presently taking place in that area, several small and very old mines are scattered around the hills, and it is possible that these two points are downstream from one or more of these unmapped mines. Not shown on the map are several underground mines in the central part of the area. Several of the high values are below the old underground mines. In the Beaucoup Creek area, only four scattered, high cadmium values occur, and three of these are clearly associated with strip mines. The fourth occurs in the northernmost part of the area and does not appear to be related to mining activities. In the Marys River area, several high values are present. Virtually all of these are adjacent to or downstream from mines.

#### Copper

In the Saline River area, only a few anomalously high copper values occur. Of these, two are downstream from a known gob pile, one is adjacent to a strip mine, and the fourth is not known to be related to mining. Three high values occur in the Crab Orchard Creek area. The first is several kilometers downstream from mines; however, there are many low values between it and the mines. The other two are scattered, apparently unrelated to mines. The two high concentrations which appear in the Beaucoup Creek area are scattered and are spatially unrelated to mining. Finally, in the Marys River area, no high values occur, although a few scattered intermediate values show no relationship to mining.

#### Cobalt

In the Saline River area, six high cobalt values are present. One of these, located in the north center portion of the area, is in the vicinity of an old gob pile and is likely related to it. A second is located

near the mouth of Eagle Creek, a stream contaminated by mining, while a third, in a tributary of the Saline River, receives drainage from a mining area. Two anomalously high values do not appear to be spatially related to coal mining. In the Crab Orchard Creek area, there are four more high values. Three of these four locations are downstream from mining activities; however, the fourth is not. In the Beaucoup Creek area, three high values occur in the northern part of the area but appear to be unrefated to mining. Lastly, in the Marys River area, the one high value is in close proximity to a strip mine, but is upstream from the mining activity.

#### Iron

In the Saline River area, only three exceptionally high iron values occur. Two are downstream from mines while one is not. Several intermediate concentrations are located downstream from mining activities. In the Crab Orchard Creek area, a few high values are present, all of which occur downstream from coal mines. No exceptionally high values occur in the Beaucoup Creek area or in the Marys River area.

#### Manganese

In the Saline River area, four high values of manganese occur in scattered locations, three of which occur downstream from coal mines. If intermediate values are considered, only a few are downstream from coal mines. Seven high values occur in the Crab Orchard Creek area, mainly downstream from mines; intermediate values occur frequently. Five high values occur in the Beaucoup Creek area, although only two of these are downstream from mines. Intermediate values appear to be unrelated to mining. Finally, two high manganese values occur in the Marys River area. Both of these are adjacent to strip mines, but do not appear to be downstream from them. Intermediate values are randomly scattered across the entire area.

#### Nickel

It can be seen that most of the twenty anomalous concentrations of nickel in the Saline River area and the Crab Orchard Creek area are downstream from coal mining activities. In the Beaucoup Creek area, there are only two high values, one of which is downstream from a coal mine. In the Marys River area, the only high value is adjacent to a coal strip mining area.

#### Zinc

Zinc is similar in geochemistry to cadmium, and not surprisingly the distribution of these two elements is similar. High zinc values are present in the saline river area, mostly downstream from strip mining or downstream from the coalfired power plant located in the area. Other high concentrations occur in Eagle Creek, an area heavily impacted by mining. Another neighborhood with high zinc values is located near an old gob pile in the northeast part of the area. Many high values also occur in the Crab Orchard Creek area. With few exceptions, these are closely associated with mining. High values in the Beaucoup Creek area, with two exceptions, are also closely associated with mining. Lastly, the one high value of zinc found in the Marys River area is downstream from a strip mine.

#### **Conclusions**

The stream sediment geochemistry of several elements; specifically, cadmium, cobalt, copper, iron, manganese, nickel, and zinc, has been examined in a six-county area of Southern Illinois that has been greatly impacted by coal-mining activities. Arsenic and chromium were also investigated, but only in isolated areas.

A regional survey of these elements in stream sediments indicates that most areas of anomalously high concentrations (four times the median value) of cadmium, cobalt, iron, nickel and zinc generally occur downstream from coal mines or in other areas of coal-related land uses. Most such anomalously high stream sediments are within eight kilometers of the coal-related area. Only manganese, and to a lesser extent copper, show distribution patterns unrelated to mining.

In three streams singled out for more detailed study, marked relationships exist among the pH of mine drainage water, the concentrations of metals in the water, and the concentrations of metals in corresponding stream sediment. Water with low pH shows seasonal summer highs in metal content and can enter streams already carrying large concentrations of metals that have either been released during the oxidation of pyrite or leached from the rocks with which the acid drainage has come into contact. In those parts of a mine-impacted stream where low pH is maintained, metals can he leached from the stream sediment to further increase the concentrations in water. On the other hand, if the pH of a stream is increased, metals will tend to precipitate out of the water and become part of the stream sediment. The precipitation reactions can occur at considerable distances downstream from the source of pollution, and account for the observed regional distribution of anomalously high concentrations of metals in sediment.

Based on the data obtained in this study, the following conclusions can be made:

- Various metals such as arsenic, cadmium, cobalt, copper, iron, manganese, nickel, and zinc are present in the rocks of the coal mining region of southern Illinois, in amounts generally equal to or slightly greater than the content in similar rocks around the world.
- Sulfuric acid created by the oxidation of pyrite can mobilize these elements, allowing them to move into nearby surface drainages.
- Once in the stream, the fate of a particular metal depends on geochemical factors, such as pH, which determine whether it will stay in solution or become a part of the stream sediment.
- Those parts of streams less than 8 kilometers downstream from coal mining activities frequently have trace element contents in bottom sediment significantly higher than those parts of the stream not influenced by coalrelated activities.

Parts of streams having low pH values consistently have higher concentrations of the trace elements in water than do areas with high pH values.

Sampling sites with low pH values commonly show a strong seasonal variation of concentration of elements in water, marked by an increase in concentration of all trace elements in the summer and fall months. Sites with high pH values do not develop a seasonal trend; instead they have various peaks and lows in concentration occurring throughout the year.

Sites with low pH, and a seasonal trend in water quality, show strong interelemental behavioral similarity. Conversely, sites with high pH values that lack the seasonal trend do not have strong interelemental behavioral similarity.

Concentrations of all trace elements in water decrease with greater distance from the source, due to dilution from additional surface water or groundwater and/or to the precipitation of elements out of solution.

Rainfall may dilute the concentrations of the trace elements in water. The

influence of this factor increases in streams of greater drainage area.

Sampling sites with low pH values consistently have lower concentrations of the trace elements in stream sediment than sites with high pH values.

None of the streams studied showed significant seasonal variation for trace elements in sediments. Most values fluctuate throughout the year, with erratic peaks and lows. However, high or peak values occur more often in summer months.

Concentrations of most of the trace elements in sediment decrease with increasing distance downstream from a polluting source. When high pH-high sediment concentration streams mix with low pH-low sediment concentration streams, the resulting values for metals in sediment are not necessarily intermediate. Some elements are of lower values than either contributing stream, suggesting a dissolution of these elements from the sediment. Iron and chromium are frequently in higher concentration in the sediment below the confluence than in either contributing stream, indicating precipitation of these elements. The specific behavior of the metals mainly depends on the pH of the water below the mixing zones.

The higher trace element content of stream sediment below coal mines is a result of leaching of the elements in or near the mines by acidic water coupled with reprecipitation when the acidic water is neutralized. Consequently, it is recommended that monitoring stream sediment for trace element pollutants not be required, because pH and trace element content in the mine effluent will control mine-related downstream changes in the trace element content of the sediments.

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Thomas J. Powers is the EPA Project Officer (see below).

The complete report, entitled "Coal Mine Related Stream Sediment Geochemistry in Southern Illinois," (Order No. PB 84-153 444; Cost: \$13.00, subject to change) will be available only from:

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