Lead Isotope Guides
For Mississippi Valley
Lead-Zinc Exploration

By RALPH S. CANNON, Jr., and ARTHUR P. PIERCE

CONTRIBUTIONS TO ECONOMIC GEOLOGY

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A review of correlations of isotopic compositions of ore-lead and tonnages of known ore in Mississippi Valley lead-zinc deposits

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CONTRIBUTIONS TO ECONOMIC GEOLOGY

LEAD ISOTOPE GUIDES FOR MISSISSIPPI VALLEY LEAD-ZINC EXPLORATION

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ABSTRACT

Lead in statiform lead-zinc deposits in many regions of the world is found to vary notably in isotopic composition. These lead isotope variations are potential guideposts for the exploration geologist searching for major lead deposits of this class. In the Eastern Hemisphere most such deposits in marine sedimentary rocks of Phanerozoic age contain ordinary lead of reasonable model age. In the Mississippi Valley, however, stratiform lead-zinc deposits contain so-called J-lead, relatively enriched in radiogenic Pb^{206}, Pb^{207}, and Pb^{208}. For each of the four most important lead-producing districts of this region we can define a favorable range of lead isotope composition within which one may concentrate the search for major lead deposits. In two of these districts we can delineate geographic areas within which we are most likely to find lead of favorable compositions. Within a third district more intensive investigations have detected three-dimensional gradients of lead isotope variation in individual mineralized areas. Such gradients should prove helpful in finding and exploring tracts of mineralized ground. Lead isotope data from these four districts define the compositions of lead most likely to guide the prospector to new major lead discoveries in the Mississippi Valley region. In current literature we note other localities from which favorable lead isotope analyses have been published.

INTRODUCTION

On September 19, 1968, the senior author presented a paper on lead isotope variations (Cannon and Pierce, 1968) at a session on geochemistry applied to Mississippi Valley lead-zinc exploration, sponsored by the Society of Mining Engineers, at the Fall Meeting of the American Institute of Mining, Metallurgical and Petroleum Engineers at Minneapolis, Minn. The substance of that talk is reproduced herein to make the information available to a wider audience.

OUTLOOK

The use of lead isotope data may prove to be helpful in future exploration for mineral deposits. The isotopic composition of lead in lead minerals of ore deposits varies widely, and studies of these variations have for 30 years been making important contributions to better
understanding of the various geologic processes that form ore deposits (Cannon and others, 1961; Brown, 1965). Such new scientific knowledge of the genesis of ores will surely help to make mineral exploration more effective in the future. Meanwhile, there are tangible opportunities to put lead isotope variations to work in exploration now, by using lead isotope data empirically, even though we may not understand the geologic causes of the variations. If we are able to identify lead isotope variations that show positive correlations with quantitative information on known distribution of minable ore, then we evidently have a useful guide to help us explore for new ore.

Until now, such possibilities have been generally ignored, for a variety of reasons. One reason is that lead isotope analyses are expensive—one commercial laboratory quotes $175 per analysis. Second, people in exploration have seldom had occasion to review the lead isotope story in enough detail to appreciate its potential. And third, people engaged in lead isotope measurement have seldom given attention to practical applications.

We believe that as soon as a breakthrough is made in cost of lead isotope analyses, this kind of information will be put to use on a broad front. In this report we wish to suggest several kinds of applications that we anticipate in future exploration for lead and zinc in the Mississippi Valley region.

**ISOTOPIC EVOLUTION OF LEAD**

The isotopic composition of lead (fig. 1) varies in lead ores in significant ways. The least abundant isotope, Pb$^{204}$, is inherited from the original or primeval lead of the solar system; whereas the three most abundant isotopes, Pb$^{206}$, Pb$^{207}$, and Pb$^{208}$, are in part original, in part radiogenic. These three continue to increase in abundance because they are stable end-products of the radioactive decay of uranium and thorium. Figure 1 shows how the average composition of lead in earth materials may have changed during earth history by decay of U$^{238}$ to Pb$^{206}$, U$^{235}$ to Pb$^{207}$, and Th$^{232}$ to Pb$^{208}$. This idealized model implies that about two-thirds of earth-lead is original, and about one-third is radiogenic. Some such isotopic evolution of lead is the primary cause of the variation of isotopic composition that we find in lead of ore deposits.

**INTRODUCTION TO ORE-LEAD DATA**

For simplicity we present data in this report in only two ways. A useful index of the radiogenic component in any one sample is the

1 "To extract the utmost information from lead data, simultaneous variations of all four isotopes must be compared,” (Cannon and others, 1961, p. 6).
Figure 1.—Isotopic relationships between uranium, thorium, and lead in the earth's crust. Black represents primeval lead; gray represents U and Th decayed to radiogenic Pb during 4.55 billion years; white represents radioactive parent isotopes that still remain. Reprinted with permission from Economic Geology (Cannon and others, 1961, fig. 1).

ratio $\text{Pb}^{206}/\text{Pb}^{207}$, which compares the two isotopes that are formed at different rates by decay of $\text{U}^{238}$ and $\text{U}^{235}$. Our second way of representing data involves computing $\text{Pb}^{206} + \text{Pb}^{207} + \text{Pb}^{208} = 100$. Differences or trends within a group of samples are easily visualized if the three major isotopes are thus calculated to 100 percent and plotted on triangular coordinates. Figure 2 shows such a plot of 1,280 analyses, mostly of samples of galena. These points represent all analyses of lead from lead minerals that were available to us as of 1960 (Cannon and others, 1961, p. 11-12). About 95 percent of the analyses plot within the smaller triangle that represents less than 1 percent of the entire triangular field.
FIGURE 2.—Isotopic composition of lead from 1,280 ore minerals—plotted in terms of Pb\(^{206}\)+Pb\(^{207}\)+Pb\(^{208}\)=100. Reprinted with permission from Economic Geology (Cannon and others, 1961, fig. 5). Small triangle added to represent the area enlarged in figure 3.

BEST-FIT CURVE

The heavily populated smaller triangular area is here magnified 10 times (fig. 3) so that we can contour the distribution of analyses. Nearly 85 percent of all analyses plot inside the contour lines shown here. A curve has been calculated to best fit the contoured data (Cannon and Pierce, 1967, p. 428). The calculation represents the isotopic evolution of lead from the primeval kind of lead that has been found in iron meteorites to the most common kind found in geologically young ore deposits. It represents evolution during a span of 4,550 million years in a closed system containing Pb, U, and Th in proportions reasonable for either the earth’s crust or mantle. This best-fit curve represents the same model as the bar diagram in figure 1.

ORDINARY LEAD

About 75 percent of all analyses fall on this curve, or within limits of experimental error. These samples plainly fit a simple history of isotopic evolution in the earth, and are called “ordinary lead” (Russell and others, 1954, p. 301). Most samples from major ore deposits are exactly such ordinary leads. For most ordinary leads, model ages calculated from this model are in accord with geologic evidence of
their age. For the exploration-minded geologist, figuratively anywhere inside the contours of this diagram is good hunting ground for big ore deposits (Cannon and others, 1962, p. 120–121).

**J-LEAD**

About 10 percent of all analyses fall beyond the zero end of the best-fit curve, within the contours at the upper apex of figure 3. Most such samples represent galenas from the major lead and lead-zinc districts of the Mississippi Valley. Such leads have been nicknamed “J-leads” because they were first detected in galena samples from Joplin, Mo. They evidently represent lead that has somehow become exceptionally enriched in radiogenic Pb$^{206}$, Pb$^{207}$, and Pb$^{208}$. 
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U-LEADS

Another type of exceptional lead with some similarity to J-lead is represented by 5 percent of the analyses. We call them "U-leads" because they are abnormally enriched in the decay products of uranium, Pb\textsuperscript{206} and Pb\textsuperscript{207}. Figure 2 shows the gross relationships between the field of U-leads, the field of J-leads, and, within the smaller triangle, the field of ordinary leads. The analyses plotted within the U-lead field of figure 2 have unique significance for uranium exploration, because they represent analyses of galenas from uranium ore deposits. Consequently, any prospector who finds galena containing lead of such a composition will know that the odds are very high that he is on the trail of a uranium deposit (Cannon and others, 1958).

ISOTOPIC VARIETIES OF LEAD IN STRATIFORM LEAD-ZINC DEPOSITS

Several years ago for a symposium sponsored by the Society of Economic Geologists on the worldwide class of stratiform lead-zinc-barite-fluorite deposits of the "Mississippi Valley type," we plotted on triangular coordinates all lead isotope data for such deposits in North America, and for such deposits in the rest of the world, principally Europe and North Africa (Cannon and Pierce, 1967, p. 428). We found that some deposits so classified contain ordinary lead, almost invariable in isotopic composition, whereas other deposits of this class contain J-type lead with notable variations in composition.

ISOTOPIC COMPOSITION VERSUS GEOGRAPHIC DISTRIBUTION

Figure 4 shows the result for North America. We see a considerable range of values. In general, the values of J-leads from the four principal mining districts of the Mississippi Valley depart farthest from ordinary lead of zero model age. At the top of the diagram is the Upper Mississippi Valley (Wisconsin) lead-zinc district, then in order of decreasing "J-ness," Tri-State lead-zinc, Southeast Missouri lead, and the Illinois-Kentucky fluor spar district with subordinate lead and zinc. On downward, we find data representing Central Kentucky, East Tennessee zinc, a few data from other Eastern States, and finally, almost on the ordinary lead curve, the Pine Point lead-zinc district of Canada. Figure 12 presents a less cluttered and more up-to-date picture for the four major Mississippi Valley districts alone.

In other parts of the world, the range of values is even greater. Uppermost in figure 5 are plotted some relatively minor occurrences of extreme J-leads from Sweden and the Ukraine, then the important Laisvall lead-zinc deposits of Sweden and Norway, and at the bottom
a tight cluster of more than a hundred analyses that fall within a very small area around the zero end of the ordinary lead curve.

These two pictures are similar in one sense and opposite in another. In North America all three major lead districts of the Mississippi Valley produce lead rather similar to Joplin J-lead, whereas Pine Point produces ordinary lead. In the Eastern Hemisphere only Laisvall produces lead rather similar to Joplin J-lead, whereas lead mined from dozens of stratiform deposits in the rest of Europe and North Africa is ordinary lead with model ages close to zero.

**ISOTOPE COMPOSITION VERSUS TONNAGE OF LEAD**

Thus, we have a continuous spectrum of isotopic composition, but with some suggestion of a bimodal distribution of major lead production. To evaluate this distribution, we have attempted to estimate total tons of lead—that is, production plus reserves—in terms of isotopic composition, for worldwide Mississippi Valley type deposits. Figure 6 is the result, plotted in terms of $\text{Pb}^{206}/\text{Pb}^{207}$ ratios. The principal tonnages of lead have a bimodal distribution that is quite impressive. So here is our first tangible lead-isotope guide for exploration for deposits of the Mississippi Valley type. If you are looking for large tonnages of lead in a Mississippi Valley type of geologic environment anywhere in the world, your chances of success are best if you are on
the track of ordinary lead with $\text{Pb}^{206}/\text{Pb}^{207}$ in the range between 1.15 and 1.20, or J-lead in the range between 1.29 and 1.42.²

This diagram further illustrates that $\text{Pb}^{206}/\text{Pb}^{207}$ ratios in ordinary lead range from about 0.89 in the most primitive earth-leads so far known from ancient Precambrian ores, to about 1.20 in the evolute leads of numerous younger ore deposits of the Phanerozoic. In J-leads we note a range from 1.20 for those close to ordinary lead to about 1.95 for extreme J-leads from Sweden. This ratio will be used in some of the following illustrations as an empirical index of "J-ness."

²Scientists in the U.S.S.R. evidently have been aware for some years that the presence of Joplin-type J-lead may be a guide to important lead deposits (Tugarnlov and others, 1960, p. 355–356).
ISOTOPE GUIDES FOR MISS. VALLEY LEAD-ZINC EXPLORATION

J-LEADS

ORDINARY LEADS

PINE POINT VARIETY

MISSISSIPPI VALLEY VARIETY

\[ \text{FIGURE 6.—Isotopic composition versus tons of lead in worldwide Phanerozoic stratiform lead-zinc deposits, SE Mo, Southeast Missouri district. Tri-State, the Tri-State mining region (Mo.-Kans.-Okla.). UMV, Upper Mississippi Valley district (Wis.-Ill.-Iowa).} \]

LEAD ISOTOPES IN MISSISSIPPI VALLEY DISTRICTS

Now, what has been learned so far about lead isotopes in the four major districts of the Mississippi Valley? About 250 lead isotope analyses are available, of which 50-100 are reconnaissance analyses made in early years. More recently—in the past 2 years—several large batches of data totaling about 200 analyses have been published.

TRI-STATE DISTRICT

First, the Tri-State district. The first suite of galenas ever analyzed resulted in the discovery of lead of unusual composition in samples of galena labeled "Joplin." This pioneering study was made by Alfred Nier, eminent professor of physics at the University of Minnesota. Nier's analyses (Nier, 1938; Nier and others, 1941) showed a significant range of composition among his three Joplin samples. For this reason we selected a large galena crystal from this district when we wanted to make a first test of lead isotope homogeneity within a single specimen (Cannon, Buck, and Pierce, 1963). We were amazed when we found, within a 3-inch cube of galena, a range of values half as great as the known spread of compositions for the entire district (Cannon, Pierce, and Delevaux, 1963). Figure 7 shows that we found samples from the growth center of the crystal to be least radiogenic,
and samples from outer growth layers successively more radiogenic. We inferred that the samples may represent a family of mixtures: ordinary lead mixed with increments of radiogenic lead. In this interpretation, the relative abundance of radiogenic component increased from about 9 percent of the lead present in the ore-forming solution when the crystal first started to grow, to about 11 percent when the outer growth layers were deposited. Another early study of samples from the Tri-State district, made by Farquhar and Cumming (1954)

Figure 7.—Sampling and isotopic variation of growth zones of a cross-sectioned galena crystal from the Tri-State district. A, Sample sites. B, J-leads and ordinary lead evolution on three-component diagram. C, Detail of B (×35), showing trend (dashed line) of crystal data (dots) in relation to Tri-State (white area) and Mississippi Valley (stippled area) J-leads, and evolution of ordinary lead (curve). Reprinted with permission from Science (Cannon, Pierce, and Delevaux, 1963, fig. 1).
of the University of Toronto, showed what appeared to be stratigraphic variations—an orderly progression from lower mineralized beds to higher mineralized beds. But our single crystal from M bed, near the center of the stratigraphic sequence they tested, showed just about as much variation as they reported in a suite of 11 samples from different beds. On the basis of these meager data we are not yet prepared to say how lead isotopes should be used in exploration within the Tri-State district.

**SOUTHEAST MISSOURI**

The old Lead Belt of the Southeast Missouri district, on the other hand, has had the most thorough reconnaissance of all Mississippi Valley districts. Some 10 years ago J. S. Brown and his staff of St. Joseph Lead Co. collaborated with J. L. Kulp and associates of Lamont Geological Observatory in a lead isotope study of a suite of nearly 175 galenas from the Southeast Missouri district. These results were made available to the public recently in Monograph 3 of Economic Geology, printed in 1967 but distributed in 1968. Brown reported (1967, p. 414-416) from the Southeast Missouri district systematic variations with depth which he illustrated for the Bonne Terre mine area by projecting his data onto a vertical longitudinal section. We have plotted our Pb\(^{206}/\text{Pb}\(^{207}\) ratios on Brown's section (1967, fig. 3, p. 415) and contoured them (our fig. 8). The contours fit his generalization that lead is progressively less radiogenic upward away from the Precambrian basement, and outward toward the fringes of mineralization.

If Brown's generalization is valid, then lead isotopes should give us a useful guide for exploration in Southeast Missouri. For the Bonne Terre mine we see a convex pattern of variation with progressively smaller Pb\(^{206}/\text{Pb}\(^{207}\) ratios upward and outward. A difference in isotopic composition for two galenas from the same horizon in a pair of adjacent drill holes, as shown, for example, diagrammatically in figure 8, might thus be expected to point toward the center of the area of mineralization.

Brown also reported (1967, p. 416-417) evidence that early galena in Southeast Missouri is less radiogenic, and late galena more radiogenic, as we had found in the Tri-State crystal. He believes, then, that the fringe ore of the Bonne Terre mine area was deposited early, and ore near the basement in the center of the mine area was deposited later.

We have also plotted (fig. 8) and contoured Pb\(^{206}/\text{Pb}\(^{207}\) values on the map of the Bonne Terre mine (his fig. 2, p. 414) without paying any attention to stratigraphic position of the samples. On this plan, likewise, the contours suggest a concentric pattern of isotopic
Figure 8.—Isotopic variations in plan and section. Bonne Terre mine, Southeast Missouri district, after Brown (1967, figs. 2, 3), with Pb$^{206}$/Pb$^{207}$ data and contours (heavy lines) added. Gray pattern represents mine area; dots, sampling sites; DH, hypothetical drill holes.
variation. Here again, the same pair of samples points toward the center of the mineralized area. From this example alone, lead isotopes would appear to provide an excellent tool for prospecting in Southeast Missouri. Brown's data (1967, p. 423-424) from other mines in the old Lead Belt—the Leadwood-Desloge and Federal-Flat River mine areas—appear to show variations somewhat similar, but clearly less systematic, than his picture for the Bonne Terre mine.

A few preliminary analyses from other ore deposits in Bonneterre Formation (Brown, 1967, p. 425) in extensions of the Southeast Missouri district suggest that such deposits can be expected to have much the same kinds of lead isotope variation as have been observed in the old Lead Belt. However, the traces of galena in the barite deposits in higher Cambrian and Ordovician formations are somewhat different; they are measurably richer in radiogenic lead (Brown, 1967, p. 420).

ILLINOIS-KENTUCKY

For the Illinois-Kentucky fluor spar district we have 10 analyses, eight of them published by Heyl and his colleagues in 1966. Heyl, Delevaux, Zartman, and Brock (1966, p. 947-951) noted districtwide variations in isotope ratios and contoured their data on a map of the district (their fig. 5). We have modified their illustrations slightly to show our contours of \( \text{Pb}^{206}/\text{Pb}^{207} \) values for their eight analyses. The \( \text{Pb}^{206}/\text{Pb}^{207} \) contours (our fig. 9) imply a simple gradient across the district, with increasing abundance of radiogenic isotopes toward the southeast. We note, however, that samples from major mines represent only a limited portion of this gradient. On the basis of this scant evidence we suggest that a major deposit in this district is likely to contain lead with \( \text{Pb}^{206}/\text{Pb}^{207} \) ratios in the range between, say, 1.265 and 1.285. If a pair of contours were drawn across the map for these values, they would define a belt that includes most of the larger mines of the district.

UPPER MISSISSIPPI VALLEY DISTRICT

Heyl, Delevaux, Zartman, and Brock (1966, p. 938) also published lead isotope analyses from 16 mines and prospects in the Upper Mississippi Valley district, together with similar lead isotope contour maps of the district (their fig. 2, p. 940-941). In our figure 10 we have likewise drawn \( \text{Pb}^{206}/\text{Pb}^{207} \) contours on Heyl’s district map. Here again, a gradient of lead isotope values appears to sweep across the district rather systematically from west to east, with the least radiogenic samples near the northwest corner and the most radiogenic toward the northeast. Two lines of independent evidence tend to suggest that some such gradient is real. First, most lead isotope analyses
from other investigators (Bate and Kulp, 1955; Russell and Farquhar, 1960) fit quite well the contours based on Heyl's data. Second, we have made two different attempts to look for lead isotope variations at a single locality in this district, and the maximum variations found are equal to approximately one contour interval on this map—only one-tenth of the total range that Heyl found across the district. Many years ago we obtained analyses of seven samples from a vertical range of stratigraphic positions in the large Bautsch ore body near the town of Galena, Ill. As compared with Heyl's Pb$^{206}$/Pb$^{207}$ value for a sample from the Bautsch mine (near the bottom of the map) of 1.381, our seven Bautsch analyses ranged from 1.369 to 1.385. Then, in 1968 we sampled a large galena cube from the Bautsch mine and obtained a value of 1.363 from the exterior of the crystal and a value of 1.383 from the interior. This recent pair of analyses would seem to imply that early lead is more radiogenic, and later lead is less radiogenic—just the opposite of evidence from Tri-State and Southeast Missouri districts.

The 13 small mines or prospects that Heyl and his colleagues sampled yielded a broad range of values, ranging from 1.30 to 1.50. The three large mines that they tested yielded values ranging only from 1.38 to 1.41. As before, these data suggest that lead in most large lead-
zinc ore bodies of this district will fall within a limited range of values, such as the 1.38 and 1.42 contours. One simple test of this suggestion is to see from what parts of the district most of the production has come. In figure 11 we have plotted the positions of the 30 largest mines (Heyl and others, 1959; Mullens, 1964) in the district. Of these 30 mines, 26 fall between the same two contours. These 26 mines represent about 50 percent of the district production of lead-zinc ore. When adjacent smaller mines are taken into account, something like 80 percent of the lead-zinc production of the district has come from the area between these contours. Lead in the composition range 1.38 to 1.42, therefore, seems to have the best chance of guiding the prospector to the discovery of a major ore body in the Upper Mississippi Valley district. Furthermore, the likeliest place to look for lead of this composition, so far as we can judge from this limited sampling, should be the area between the 1.38 and 1.42 contours. The exploration geologist
will be quick to note that the area so defined appears to point eastward beyond the southeast edge of the district toward uplands where the favorable ore horizons are mostly concealed by younger strata.

**LEAD ISOTOPE GUIDES FOR EXPLORATION**

Finally, we have generalized the data for each of the four districts on triangular coordinates (fig. 12) to present the overall picture for the Mississippi Valley. The upper dashed line encloses data for the

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**Figure 11.**—Map of Upper Mississippi Valley district, showing locations of major lead-zinc mines in relation to a selected pair of Pb$^{206}$/Pb$^{207}$ contours.
Upper Mississippi Valley district, and the gray patch near the center of this polygonal area represents our guess about "big ore" in the Upper Mississippi Valley district. This gray patch encloses all samples between the 1.38 and 1.42 contours. The long dashes at lower left enclose all Southeast Missouri samples, and the gray patch below the center of this area represents the major tonnage of lead that has been produced from the old Lead Belt. The solid line at upper center en-
closes all Tri-State samples, and the gray patch from top to center represents samples from M bed, the most important ore horizon. The lower solid line encloses samples from the Illinois-Kentucky fluorspar district, and the small gray patch represents samples from larger mines in that district. For added perspective, a fifth polygon has been plotted in order to represent the ordinary kind of lead, at the zero end of the best-fit lead isotope evolution curve, such as has been found in stratiform lead-zinc deposits in Europe and North Africa and at Pine Point, Canada.

We should like to show you more specifically how the major tonnages of Mississippi Valley J-lead are related to the trilinear coordinates and to Pb$^{206}$/Pb$^{207}$ ratios as well. Figure 13 is a chart to guide the exploration geologist who wishes to use lead isotope analyses in prospecting for lead in the Mississippi Valley region. If you hope to make a new discovery of a major tonnage of lead in the Mississippi Valley region, you might consult Figure 13 and other pertinent data for guidance.
Valley—a new mine, or a new district—it seems wise to search for J-lead of isotopic composition much like that outlined here for the Tri-State district, or as shown by the gray patches representing major tonnages of lead so far produced from the principal Mississippi Valley districts.

To illustrate this approach to prospecting, we have also plotted on figure 13 several potential lead isotope guides for the prospector, based on published lead isotope analyses. The barite deposits of Southeast Missouri, in Ordovician and underlying Cambrian formations stratigraphically above the lead-bearing Bonneterre Dolomite, are so far known to contain relatively small amounts of galena. But this lead (Brown, 1967, p. 418) is of favorable isotopic composition, as is shown by the line pattern on this diagram, and thus offers to guide the prospector to "big ore." The other examples concern northern Arkansas. Only two galenas have been analyzed from the northern Arkansas lead and zinc region (Bate and Kulp, 1955, p. 44). These analyses, represented by the two circles (fig. 13), suggest that the northern counties of Arkansas (McKnight, 1935), like the Tri-State district, may be a favorable place to look for "big ore." We suggest that these several J-lead clues deserve thoughtful consideration and further investigation.

Finally, it is conceivable that some future prospecting in the Mississippi Valley region may discover, instead of Mississippi Valley-type J-lead, ordinary lead like the Pine Point variety illustrated in figure 13. Any such discovery of ordinary lead likewise would deserve careful evaluation, inasmuch as some millions of tons of such ordinary-type lead have been found in recent years, in a distant part of the Interior Lowlands province, at Pine Point in Canada.

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