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TEPCO, Denver	2
TEPCO, RPS, Washington, (including master)	2
	70

CONTENTS

ILLUSTRATIONS

TABLES

THE ISOTOPIC VARIATION OF COMMON LEAD IN GALENA FROM THE FRONT RANGE AND ITS GEOLOGICAL SIGNIFICANCE

By George Phair and Henry Mela, Jr.

ABSTRACT

Lead extracted from thirteen samples of galena from the Colorado Front Range was analyzed mass spectrometrically. These samples included nine from the major districts in the Laramide mineral belt, three from "higher temperature" deposits of controversial ages from outlying parts of the Range, and one from the Stove Mountain pegmatite of definite late pre-Cambrian (Pikes Peak) age. The results show that: (1) galena samples from the southwestern half of the Laramide mineral belt are enriched in Pb^{206}, Pb^{207}, and Pb^{208} relative to Pb^{204} over galena from the northeastern half, (2) the three galena samples from the deposits in the outlying parts of the Front Range have isotopic compositions typical of older pre-Cambrian common lead and widely different from all Late Cretaceous or younger lead so far reported in the literature. These facts in large measure substantiate the pre-Cambrian age of the deposits inferred by Lovering and Goddard. (3) As might be expected the composition of the lead in the Stove Mountain pegmatite galena is intermediate between the Laramide and the earlier pre-Cambrian lead.

As in the Colorado Plateau lead samples studied by Stieff and Stern, the regional averages for Pb^{206} and Pb^{207} increase from east to west in the Front Range along the Laramide mineral belt. Unlike the lead from the Colorado Plateau, however, thorium-lead Pb^{208} increases along

with the uranium leads Pb^{206} and Pb^{207}. No Pb^{206} enrichment was noted in galena from nonuraniferous veins in and around centers of intrusion of uranium-rich late-stage Tertiary intrusives or in actual uraniferous mining districts.

INTRODUCTION

During the 1952 field season fresh galena was collected from 26 mines in the Colorado Front Range as part of a comprehensive study of the geological cycle of uranium and of related elements in this region carried on since 1949 by the U. S. Geological Survey on behalf of the Atomic Energy Commission. A special effort was made to obtain samples from mines then in operation. Most of the galena was coarsely crystallized, and hand-picked separates weighing 200-500 mg free from other sulfides and alteration products were readily obtained. All original exposed surfaces were chipped off and only the insides of the fragments were used. As a check on possible contamination 12 of the 13 samples which had been selected for isotopic study were analyzed chemically for uranium (table 1). Uranium was found to be negligible for the present purposes in all 12 samples. Lead separated from each of these samples as the iodide was analyzed isotopically by the Mass Assay Laboratory of the Carbide and Carbon Chemical Company's Y-12 Plant, Oak Ridge, Tenn., using modified Nier-type spectrometers. The samples were analyzed in the sequence indicated by the numbers in tables 1 and 2.

These isotope abundance studies were integrated with the broader investigations of common lead variation in the Colorado Plateau deposits carried on by L. R. Stieff and T. W. Stern of the Geological

Table 1.--Uranium content of galena samples.

Sample no.	Percent U (chemical)
GS/177	0.00080
GS/266	0.00071
GS/267	0.00104
GS/268	0.00069
GS/269	0.00092
GS/270	0.00074
GS/271	0.00035
GS/272	0.00065
GS/274	0.00053
GS/275	0.00027
GS/276	0.00046
GS/277	0.00031

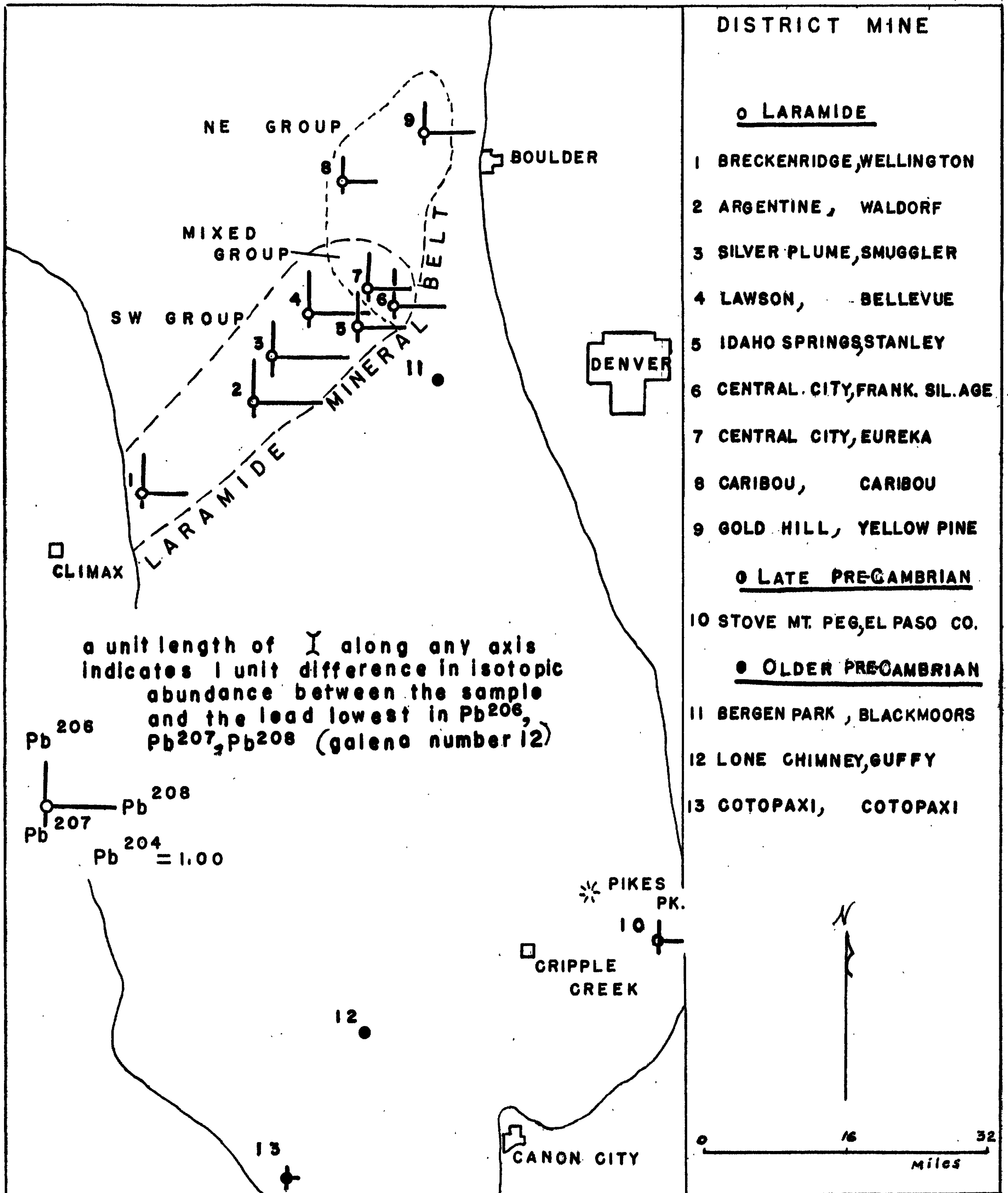
Table 2.--Results of isotopic analyses of common lead expressed in percentages.

Sample no.	District, Mine	Pb^{204}	Pb^{206}	Pb^{207}	Pb^{208}
GS/3	Caribou, Caribou	1.43	24.16	21.54	52.86
GS/177	Central City, Eureka	1.39	24.99	21.31	52.31
GS/266	Silver Plume, Smuggler	1.35	24.00	21.21	53.43
GS/267	Idaho Springs, Stanley	1.36	24.86	21.34	52.44
GS/268	Cotopaxi, Cotopaxi	1.49	23.49	22.73	52.28
GS/269	Guffy, Lone Chimney	1.52	23.35	22.68	52.44
GS/270	Breckenridge, Wellington	1.38	24.98	21.34	52.30
GS/271	Central City, Franklin-Silver Age	1.39	24.60	21.43	52.58
GS/272	Bergen Park, Blackmoor's	1.51	23.34	22.80	52.34
GS/274	Argentine, Waldorf	1.35	24.94	20.97	52.73
GS/275	Lawson, Bellevue	1.38	24.72	21.23	52.66
GS/276	Gold Hill, Yellow Pine	1.40	24.22	21.38	52.99
GS/277	El Paso County, Stove Mt. Peg.	1.45	24.01	22.13	52.40

Survey. We are indebted to these workers and to R. S. Cannon, Jr., of the Geological Survey, for numerous helpful suggestions in the course of the study. Most of the samples were collected by Phair assisted by Norman Herz. One sample (GS 3) was collected by R. U. King and Phair during the summer of 1949. Mela made the chemical analyses and separated the lead iodides for mass spectrometric study.

GALENA FROM THE LARAMIDE MINERAL BELT

Nine samples representing most of the larger districts in the Laramide mineral belt were among those analyzed. These came from mines scattered over the length of the belt (fig. 1). The sample spacing was closest in the middle part of the Front Range where uranium in the Laramide porphyries and in the associated veins has been found to reach a regional high. The distribution of uranium and of radioactivity in the early Tertiary dike rocks throughout a large part of the mineral belt is known from radiometric and uranium analyses of more than 400 samples of porphyry collected in the field and studied in the laboratory (Phair, 1952; also summary report in preparation). The trend of differentiation in the same rocks has been worked out on the basis of results of 50 standard rock analyses by rapid methods coupled with petrographic study. Besides filling gaps in the regional isotopic pattern as described by Stieff and Stern (personal communication) these studies had an additional objective. We hoped to find out whether lead from areas in and around centers of intrusion of lime-poor rhyolite known to be enriched in uranium (up to 20 x normal calc-alkaline granite) showed an unusually



THE ISOTOPIC VARIATION OF COMMON LEAD IN GALENA FROM THE COLORADO FRONT RANGE

Figure 1

high content of the uranium daughter Pb^{206}. Throughout the mineral belt we have noted evidence of close space, time, and geochemical connections linking these uranium-rich igneous rocks with actual deposits and showings of pitchblende (Phair, 1952).

Both uranium and lead tend to be concentrated in late stage differentiates and in hydrothermal solutions. Assuming that the common ancestor for the porphyries and the associated vein deposits was a magma initially rich in uranium formed by remelting or assimilation of older rocks which were themselves uranium-rich, it seems reasonable to suppose that the concentration of uranium and of the uranium daughter Pb^{206} should go hand in hand. One might expect, therefore, to find late stage differentiates enriched in uranium, vein galena enriched in the isotope Pb^{206}, and actual deposits of pitchblende closely associated with one another in space and in time. If so, the Pb^{206} abundances might serve not only as permissive indication of the uranium potential in a given area, but might also throw new light on the age-old question of the origin of magmas.

It should be remarked at the outset that this brief investigation was not designed to test whether the common lead from galena closely associated with uranium minerals in the same vein contained unusually large amounts of Pb^{206}. That such local enrichment may occur is clear from the studies of galena from (1) the Ace Creek mine (Collins, Farquahar, and Russell, 1954, table 1, p. 7); (2) the Witwatersrand by the same workers, (op. cit., table 1, p. 11); (3) the Happy Jack mine, Utah, by L. R. Stieff and T. W. Stern (personal communication). With one exception we intentionally avoided collecting galena from veins known to be uraniferous because some such lead samples may reflect post-depositional introduction of Pb^{206}. We were interested in the composition of the common lead at the time of original deposition and in the relationship of that composition to the last stages of magmatic differentiation.

The data presented in this paper (tables 2 and 3) show only that the Pb^{206} content of the galena and the uranium contents of the associated igneous rocks are not related in any simple way. Our sampling has been too incomplete and the analytical errors are too great to permit the drawing of fine distinctions.

Table 3 and the sketch map (fig. 1) show that the isotopic results do fall into a geographic pattern which, however, bears little relationship to the spatial distribution of radioactive igneous rocks and of uraniferous veins. Two galena samples from the northern part of the mineral belt, an area containing numerous radioactive centers of intrusion and abundant small showings of uranium, are poorest in Pb^{206}. Two from the central part of the belt, the area richest in uraniferous late-stage differentiates and in workable uranium deposits, have mixed characteristics--both are enriched in Pb^{206}, one is enriched in Pb^{207} and neither is enriched in Pb^{208}. Five galena samples from the southern part of the belt, the area poorest in radioactive intrusions and in actual showings of pitchblende, are enriched in all 3 radiogenic isotopes with respect to those in the north. This geographic pattern is evident in spite of any experimental error which can reasonably be expected to be present. The pattern for Pb^{207} is less well defined than for Pb^{206} and Pb^{208} because of

Table 3.--Isotopic variation in common lead from the Front Range, where Pb^{204} = 1.00. (Samples listed from northeast to southwest on map.)

Sample no.	Map no.	Mine	Pb^{206}	Pb^{207}	Pb^{208}	Excess over GS/269 Pb^{206}	Excess over GS/269 Pb^{207}	Excess over GS/269 Pb^{208}	
			Laramide galena						
GS/276	9	Yellow Pine	17.30	15.27	37.85	1.94	0.35	3.35	Less enriched
GS/3	8	Caribou	16.89*	15.06	36.96	1.53	0.14	2.46	Less enriched
GS/177	7	Eureka	17.98	15.33	37.63	2.62	0.41	3.13	Mixed
GS/271	6	Franklin-Silver Age	17.70	15.42	37.83	2.34	0.50	3.33	Mixed
GS/267	5	Stanley	18.28	15.69	38.56	2.92	0.77	4.06	More enriched
GS/275	4	Bellevue	17.91	15.38	38.16	2.55	0.46	3.66	More enriched
GS/266	3	Smuggler	17.78	15.71	39.58	2.42	0.79	5.08	More enriched
GS/274	2	Waldorf	18.47	15.53	39.06	3.11	0.61	4.56	More enriched
GS/270	1	Wellington	18.10	15.46	37.90?	2.74	0.54	3.40?	More enriched
			Late pre-Cambrian galena						
GS/277	10	Stove Mt. pegmatite	16.56	15.26	36.14	1.20	0.34	1.64	
			Older pre-Cambrian galena						
GS/272	11	Blackmoor's Ranch	15.46	15.10	34.66	0.10	0.18	0.16	
GS/269	12	Lone Chimney	15.36	14.92	34.50	0.00	0.00	0.00	
GS/268	13	Cotopaxi	15.76	15.25	35.09	0.46	0.33	0.59	

*Solid lines separate lead into two groups without overlap--one less enriched and one more enriched.

its limited variability. Pb^{207} cannot vary independently from Pb^{206}, a small change in the one being accompanied by a much larger change in the other. The precise position of the dividing lines (solid lines in table 3) which separate the Pb^{207} into two groups without overlap, one group enriched in that isotope, the other not enriched, may be open to some question. Slight shifts in position of these dividing lines from one isotope to another give rise to the intermediate group with mixed characteristics. The distinction between enriched and unenriched galena with respect to Pb^{206} and Pb^{208} is much more pronounced and less subject to error. The maverick in the geographical pattern is the Pb^{208} in sample GS/270; it is definitely low. The associated Pb^{206} and Pb^{207} from the same galena sample conform to the regional trend. Possibly a more detailed sampling would have revealed other erratics, but in terms of regional averages the trend seems clear. No internal trends are evident within any one of the three groups.

The observed pattern represents the eastward beginnings of a geographic pattern first noted by Stieff and Stern (personal communication) in the region to the west of the Front Range. On the basis of isotopic analyses of some 45 samples of lead minerals, chiefly from the Colorado Plateau and from the San Juan Mountains, they concluded that samples from any one area may show considerable variation but the regional averages for the contents of Pb^{206} increase in a westward direction. All of their samples contain more Pb^{206} than the most highly enriched galena from the Front Range. In contrast to the lead minerals studied by Stieff and Stern, however, Pb^{208} in the Front Range lead minerals increases relative to Pb^{204} along with the Pb^{206}.

The differences in isotopic composition between lead minerals from the northeastern and southwestern Front Range are much too large to be attributable to small differences in time of deposition. Lovering and Goddard (1950) have postulated that the "deep" parent magma, which in Laramide times underlay the northeastern half of the belt, differed in composition from that beneath the southwestern half. To the extent that the isotopic differences may reflect differences in lead and uranium contents of the source rocks from which these magmas were derived, these results are consistent with that interpretation.

In its low ratio of Pb^{206}, Pb^{207}, and Pb^{208} to Pb^{204}, the lead from the Caribou mine approaches a late pre-Cambrian lead in composition (cf. lead from the Stove Mountain pegmatite, table 3). Kerr and Kulp (1952) in their determination of the Pb^{206}/U^{238} age of uraninite from the Caribou mine made a generalized common lead correction and calculated an apparent age of 23 $\pm$ 10 million years. Using the actual lead from this mine in making the common lead correction, we calculated an apparent Pb^{206}/U^{238} age of 37 million years for the same sample. Because the sample analyzed by Kulp contained about 97 percent of the total lead as common lead, the mass spectrometer error becomes multiplied many fold in making any common lead deduction and a maximum uncertainty of about 14 million years is introduced into any age calculations that can be made. Considering only these sources of error the possible Pb^{206}/U^{238} age of this sample ranges from 23 to 51 million years. The upper limiting age is close to the average Pb^{206}/U^{238} age obtained on uraninites from the Central City district by several workers. Little confidence can be attached to such results. More and better samples are needed.

PRE-CAMBRIAN GALENA

Like common lead forming today the original lead present in the earth's crust is believed to have consisted of the isotopes Pb^{204}, Pb^{206}, Pb^{207}, and Pb^{208}. The last three of these isotopes are also produced by processes of radioactive decay; the Pb^{204} is nonradiogenic so far as is known. Throughout the earth's history the original lead has become increasingly enriched relative to Pb^{204} in all three radiogenic isotopes derived from the breakdown of U^{238}, U^{235} and Th^{232}. Addition of the radiogenic components has been facilitated by basic geological processes--magmatic, hydrothermal, and supergene. As a result the isotopic composition of the lead tends to reflect the geological age of the mineral assemblage in which it is now found and to which it is genetically related. The percent Pb^{204} in any lead sample (table 2) thus provides an index of its "primitiveness". Just what this index will mean in terms of absolute age depends upon geographical and geological factors, but it is generally true that the oldest leads have the highest Pb^{204} contents.

No claim is made that the isotopic composition of common lead will permit a quantitative estimate of age in terms of millions of years. Moreover, an inspection of the table, Relative isotopic abundances of common leads (Faul, 1954), reveals several exceptions to the rule that pre-Cambrian lead is characterized by primitive compositions. The same data show, however, that these exceptions are limited to certain lead of probable late pre-Cambrian age (e.g., some at Sudbury) or to lead from deposits known to be uraniferous (e.g., Witwatersrand). These exceptional lead samples seem to be local developments typical of certain mines. In two of the four Sudbury mines in which these enriched leads are found, lead having the more usual late pre-Cambrian characteristics is also present. The lead from Witwatersrand is characterized by Pb^{206}/Pb^{204} ratios far in excess of the Pb^{208}/Pb^{204} ratio, the reverse of what is found in leads from nonuraniferous deposits.

In the anomalous pre-Cambrian lead so far reported the isotopic enrichment involves only the uranium lead; the associated thorium lead is present in the abundance expectable in old age galena. The Front Range galena samples from ore deposits outside the Laramide belt in contrast to those from within the belt are remarkably uniform in composition and definitely primitive with respect to content of all three radiogenic isotopes. The published data reveal no instance of isotopic overlap of any kind between lead of definite older pre-Cambrian 1/ age from nonuraniferous veins and lead of definite Laramide age--the only age distinction we are attempting to draw in this paper. It might be argued that one should be cautious about using the relative primitiveness even on a broad scale as an age

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1/ By "older" pre-Cambrian lead we mean lead of a definitely more primitive composition than lead from a late pre-Cambrian pegmatite of Pikes Peak type the "absolute" age of which is reasonably well known. So used, the term "older" pre-Cambrian is probably synonymous with Early to Middle pre-Cambrian.

indicator because geographical trends not yet suspected may eventually turn up. In the present studies, however, we are concerned not with global or continent-wide generalizations but with relationships in a single small region the geology of which has been under study for 50 years. Our data have been obtained not on single samples but on sets of samples, and within each set the samples have been so spaced as to provide an estimate of the effect of geographical isotopic variation. The geographical variation evident among the three samples of unknown age is extremely small. The differences in isotopic composition between these and the samples from the Laramide mineral belt are much too large and much too systematic to be attributed to analytical or sampling errors. The results are in harmony both with the predominant world-wide isotopic pattern and with what is, in our opinion at least, the bulk of the geological evidence.

Scattered throughout parts of the Front Range outside the Laramide mineral belt are a number of ore deposits of unknown age thought to be pre-Cambrian by Lovering and Goddard (1950). Recently several geologists have voiced the opinion that these deposits represent only a deeper and higher-temperature phase of the Laramide metallization.

Many of these deposits have been mined in a small way for base metals, chiefly copper and zinc. Unlike most of the ores known to be Laramide these ores consist of massive sulfides in a gangue of rock-forming silicates, particularly pyroxenes and hornblendes. The "high-temperature" metallics--magnetite, pyrrhotite and iron-rich sphalerite--are scattered in these deposits but are locally abundant. The ore bodies themselves are lenticular, foliated to banded, and more or less conformable with the enclosing gneisses. During the summer of 1952 Phair visited 14 of the 18 deposits described by Lovering and Goddard (1950). Galena was found to be very scarce in most of these with the exception of those near the southern limit of the range (Cotopaxi, Lone Chimney, and Isabel). Fortunately we were able to collect fresh galena from 3 of the 4 subtypes recognized by Lovering and Goddard. Of the 4 deposits from which isotopically analyzed galena was obtained, one, Cotopaxi, has been interpreted as a magmatic segregation in mafic rocks; two, Lone Chimney and Bergen Park, as high temperature replacements; and one, Stove Mountain, as an occurrence of primary galena in granite pegmatite (J. S. Adams, personal communication). Two of these deposits--Cotopaxi and Lone Chimney, now the Bessie--were being actively worked in 1952.

The age of the Stove Mountain pegmatite is definitely late pre-Cambrian. It is a differentiate of the closely associated, largely undeformed Pikes Peak granite which cuts foliated crystalline rocks and which has been dated within the limits of 700 to 900 million years by several laboratory methods. The isotopic composition of the galena from this pegmatite is consistent with a late pre-Cambrian age but of itself does not rule out entirely an early Paleozoic age. This galena supplied by J. S. Adams, U. S. Geological Survey, was doubly valuable. Not only is syngenetic galena extremely rare in pegmatites but in addition the sample provided a relatively fixed point on the geological time scale for the late pre-Cambrian in this region with which to compare our other results. The galena was present as isolated cubes as large as half an inch on an edge in an aggregate of coarse microcline and quartz with smaller euhedra of zircon and sphene. No other recognizable sulfides nor any hydrothermal gangue minerals were present. The large cubes were covered with a whitish film which extended into the adjacent silicates, but when the cubes were broken open the insides were found to be completely fresh.

Figure 1 shows that the three other lead samples (11,12,13) from outside the belt differ markedly in isotopic composition from all those collected from mines located within the belt. In addition these lead samples from outside the belt have (1) distinctly lower contents of Pb^{206} and Pb^{208} but higher Pb^{204}, and (2) nearly equal parts of Pb^{206} and Pb^{207}. These attributes are consistent with, and in a large measure confirm, the older pre-Cambrian ages of all three samples. For the purposes of comparison the isotopic composition of galena believed to be Middle pre-Cambrian (1300 million years) by Nier (1938) is as follows:

	Pb^{204}	Pb^{206}	Pb^{207}	Pb^{208}
Galena, Great Bear Lake	1.00	15.93	15.30	35.30

Age determinations by lead-uranium, lead-thorium and lead-lead methods upon primary uraninite from a pegmatitic syenite intrusive into the basement complex in the Central City district and alpha-lead determinations upon zircon from the same intrusive indicate that the middle pre-Cambrian was a time of igneous activity in that part of the Front Range at least. These three galena samples may be older but probably are not much younger than the age found 1300 ± 100 million years (Phair and Gottfried, manuscript in preparation). In contrast to the Laramide lead samples the older pre-Cambrian lead samples show little geographic variation; two samples of galena from similar deposits 65 miles apart are so nearly identical that the differences cannot be plotted on the scale of the sketch map (fig. 1, nos. 11 and 12). The third of the older pre-Cambrian galena samples shows a very slight enrichment in radiogenic components as compared to the other two, but the difference is of the same order as the expected experimental error.

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