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Lead Isotopes of Palaeozoic, Strata-Bound to Stratiform Galena Bearing Sulfide Deposits of the Eastern Alps (Austria); Implications for their Geotectonic Setting

by V. Köppel¹ and E. Schroll²

Abstract

The lead isotope compositions of galena lead from stratiform to strata-bound ore deposits in Palaeozoic host rocks in the Eastern Alps indicates that the source of the lead must be sought in crustal rocks. In one stage of their history these rocks were upper crustal rocks with high U/Pb and Th/Pb ratios. In a later second stage, prior to the extraction of the lead, the U/Pb ratios was lowered probably due to a uranium loss either during a high grade metamorphism or during wheathering and sedimentation of the source material. From a comparison of feldspar-lead isotope data, which at present do not cover the entire area sufficiently, with those of ore-lead one may tentatively conclude that the most likely source rocks are the pre-Ordovician metasediments.

Keywords: lead, galena, Pb-isotopes, stratiform deposits, Eastern Alps.

1. INTRODUCTION

This investigation forms part of a study of the lead isotope systematics of ore deposits in the Eastern and Southern Alps that was initiated with the aim of finding possible source beds for the lead of the economically important Pb-Zn deposits in Triassic carbonates. Other types of deposits were included in this study in order to obtain a survey of the lead isotope patterns of the galena bearing deposits.

The results show that lead isotope ratios of galena bearing ores vary roughly according to the, admittedly often assumed, age of the deposits. This correlation has already been noticed by KOVAC (1968) who carefully evaluated the data produced during the first period of lead isotope measurements of ores. The re-

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Table 1 Summary of the characteristic features of stratiform to strata-bound Palaeozoic ore deposits of the Eastern Alps.

No.	Locality	Tectonic position	Stratigraphic position	Main mineral constituents	Grade of metamorphism	Literature
1–8	Palaeozoic of Graz Styria	UEA	ID, calcareous or graphitic schists, argillaceous or cherty limestone	gn (Ag), sph, pyr,(po), bar, sid	epizonal	WEBER (1977,19
9	Ramingstein Styria	MEA crystalline complex	OP (?) garnet-mica- schists	gn (Ag), sph, mesozonal par, chpyr, (Mo)		WEISS (1951) TUFAR (1971)
10	Meiselding Carinthia	UEA Gurktal nap	Or, (Cm) sericite schists, meta- volcanites	gn (Ag), sph chpyr, po	epizonal	TUFAR (1974)
11	Moosburg Kamuda Carinthia	MEA crystalline complex	OP (?) banded marble	gn, sph, aspyr, sid,	diaphtorized	HOMANN (1962) TUFAR (1974)
12	Rettenbach, Mittersill Salzburg	MEA Grauwacken- zone	UOrd, Sil (?) quartz phyl- lites, green- schists	pyr, po, chpyr, aspyr, gn, sph, sulphosalts	epizonal	UNGER (1969)
13	Walchen, Oeblarn, Styria	MEA Grauwacken- zone	WOrd, Sil (?) sericite - quartzite greenschists	pyr, po, chpyr, aspyr, gn, sph, sulphosalts	epizonal	UNGER (1968)
14	Striedalmer Plaike, Carinthia	MEA Kreuzeck crystalline complex	Ord (?) volcano- sedimentary sequences	pyr, po, chpyr, gn, sph	mesozonal	FRIEDRICH (1963
15	Kaser Wieserl, Gnoppnitz Carinthia	MEA Kreuzeck crystalline complex	Ord (?) volcano- sedimentary sequences	pyr, po, chpyr, gn, sph	mesozonal	FRIEDRICH (1963
16	Knappen- stube Carinthia	MEA Kreuzeck crystalline complex	Ord (?) volcano- sedimentary sequences	pyr, po, aspyr, gn, sph	mesozonal	FRIEDRICH (1963)
17	Panzendorf Eastern Tyrol	MEA Defereggen crystalline complex	Ord (?) volcano- sedimentary sequences	pyr, po, chpyr, gn, sph	mesozonal	TORNQUIST (1935 PRASAD (1969)
18	Stockenboi Buchholz- graben, Carinthia	MEA	OP (?), seri- cit quartzite, meta-volca- nites .	cin, pyr, chpyr, po, po (gn)	epizonal	SCHULZ (1969) POLEGEGG (1971)

Table 1 Continued

No.	Locality	Tectonic position	Stratigraphic position	Main mineral constituents	Grade of metamorphism	Literature
19	Koprein, Leppengraben, Eisenkappel Carinthia	MEA	Ord (?) greenschists of Eisen- kappel, limestone	gn (Ag), sph, chpyr, pyr	epizonal	×
20	Schneeberg, (Monte Neve) South Tyrol	MEA Oetztal-Stubai crystalline complex	Sil or pre-Sil	gn (Ag), sph, po, chpyr, sulphosalts mangano-phyllit, garnet, tremolit biotit, Zn-anda- lusit	mesozonal	FOERSTER (1963
21	Arzberg Fröschnitz Styria	LEA Stuhleck Kirchberg nap	pre-Alpidic connected with Mürztaler Grobgneis	sid, bar, gn, sph, pyr, tetr	epizonal	TUFAR (1963)
22	Kreiteralm Obertal Schladming Styria	MEA Schladming- crystalline complex	OP (?)	gn (Ag), sph, tetr, chpyr	epizonal	FRIEDRICH (193

Abreviations:

UEA = Upper East Alpine unit, MEA = Middle East Alpine unit, LEA = Lower East Alpine unit, OP = Old Palaeozoic, Cm = Cambrian, Ord = Ordovician, Sil = Silurian, L = lower, U = upper.

Gn galena, sph sphalerite, pyr pyrite, po pyrrhotite, chpyr chalcopyrite, aspyr arsenopyrite, mo molybdenite, cin cinnabarite, tetr tetraedrite, sid siderite, bar barite, flu fluorite.

sults furthermore show that the μ (= ²³⁸U/²⁰⁴Pb) and the W (= ²³²Th/²⁰⁴Pb) values of the reservoirs in which the ore leads developed are different for the Southern Alps and East Alpine nappe units on the one hand and the Penninic area on the other hand (Köppel, 1983). The trend to lower μ and W values observed in the Penninic realm continues northwards into Germany (LIPPOLT et al., 1983, WEDEPOHL et al., 1978).

Most, if not all, stratiform to strata-bound deposits in the Alps have formerly been considered to be of epigenetic origin and related to the Alpidic magmatism and/or metamorphism. Their stratiform to strata-bound nature was explained as either due to selective metasomatism or possibly due to late Alpidic deformation and metamorphism. During the past decades, however, a reexamination of the concept of the Alpidic metallogenesis in the light of syngenetic theories brought a change in outlook. To-day, these deposits are considered to be of syngenetic to syndiagenetic origin (compare e.g. SCHULZ, 1974, 1983, HÖLL and MAUCHER, 1976, TUFAR, 1974 and 1980). In this paper we present the results of polymetallic, stratiform to stratabound sulfide deposits occurring in metasedimentary to metavolcanic sequences in East Alpine nappe units. In order to substantiate the interpretation of these data it will be necessary to mention the results obtained from other ore types from Alps and from lead of feldspars from the Southern Alps.

As can be seen from table 1, in which the main features of the deposits are summarized, the host rocks of these deposits are either late Precambrian to Cambrian metasediments or Palaeozoic metasediments the oldest of which has been biostratigraphically dated as upper Ordovician (see SCHARBERT and SCHÖNLAUB, 1980 and SCHÖNLAUB, 1980).

Between the time of deposition of the late Precambrian to Cambrian sequence and the Palaeozoic sediments there occurred during the Ordovician a period of magmatism and metamorphism, the so-called Caledonian event. The *pre-Caledonian sediments* contain mainly metapelites and metapsammites with 1500 to 2700 my old detritus as is evident from the zircon age patterns (e.g. KÖPPEL and GRÜNENFELDER, 1971). In the Gleinalpe amphibolites interfingered with plagioclasegneisses occur. The latter yielded a Rb-Sr whole rock isochron age of 500 ± 45 my and an initial 87Sr / 86Sr ratio of $0.7044 \pm .0012$ which precludes an origin of these rocks from continental crustal material (FRANK et al., 1976). They may have originated in an island arc situation. Lead isotope analyses of amphibolites from the Ivrea zone (CUMMING and KÖPPEL, in prep.) indicate the presence of MORB type basalts. BULETTI (1983) reported the presence of amphibolites in the Ceneri zone (Southern Alps) which according to their chemistry originate from MORB type rocks.

The Palaeozoic sequence also contains basic and acid volcanics, especially at its upper Ordovician base and in Devonian rocks of the Palaeozoic of Graz. The presence of an oceanic magmatism has until now not been established. The sediments consist of psammitic and pelitic rocks as well as of carbonates. The Palaeozoic sequence is unmetamorphosed in the Carnian Alps, elsewhere the Hercynian and/or Alpidic metamorphism reached greenschist to amphibolite grade. To distinguish between the Palaeozoic rocks and the pre-Caledonian sequence then becomes often difficult.

In view of the polymetamorphic nature of the pre-Mesozoic rocks in many areas and the difficulty in determining the time of deposition of sedimentary

Fig. 1 Sketch map of the Eastern and Southern Alps (after FREY et al., 1974) showing the investigated galenabearing ore mineralizations of Pre-Alpine ages. (See localities of the numbers 1-21 in table 1.)

¹ Penninic domaine

² Austroalpine domaine and Southern Alps, folded, but not affected by Alpine metamorphism

³ Permo-Mesozoic cover metamorphized (including anchimetamorphic rocks)

⁴ Tertiary intrusives

⁵ Pre-Triassic basement of the Pennic domaine

⁶ metamorphosed palaeozoic cover and Pre-Alpine crystalline basement



and volcanic series the assignment of ages to these stratiform and strata-bound ore deposits often rests solely on lithostratigraphic considerations and the presence or absence of high grade metamorphism.

2. RESULTS

The results are listed in table 2 and plotted in fig. 2. The sample localities are indicated on the tectonic sketch map (fig. 1) and the deposits are characterized in table 1. The lead isotope ratios exhibit the following features: (i) The high μ and W values indicate that the lead evolved for a significant period in rock reservoirs with high U/Pb and Th/Pb ratios when compared to the values pertaining to average crustal leads. (ii) The 207Pb/206Pb model ages are higher than the 208Pb/204Pb model ages. Both these feature are typical for all galena bearing ore deposits of the Southern Alps and East Alpine nappe units (KöPPEL, 1983). (iii) In the two diagrams of fig. 2 the data points form two clusters. One is defined by the samples 1–14 and the second by 15–19. The first group has 207Pb/206Pb model ages ranging from about 550–660 my and 208Pb/204Pb model ages of 170–360 my, whereas in the second group they range from 330–440 my and from 20–150 my respectively.

The two clusters do not correspond to the two periods of mineralizations, i.e. the pre-Caledonian and the Palaeozoic ones. The deposits in the Palaeozoic of Graz, for which biostratigraphic evidence proves a lower Devonian age (SCHÖNLAUB, 1980), all contain isotopically identical leads with a 207Pb/206Pb model age of 550 my. A comparison of the lead model ages (table 2) with the assumed ages (table 1) shows that in some cases the model ages roughly agree with the assumed ages whereas is others the lead model ages are older. Again this feature has been observed in all types of galena bearing ore deposits of the Southern Alps and in the East Alpine Nappe units.

The lead isotope ratios indicate that they were not severly disturbed during the subsequent periods of metamorphism by the addition of radiogenic lead from the surrounding host rocks. Young or even negative model ages were only observed in deposits that contain very little lead or where the emplacement of the ore is demonstrably related to the Alpidic orogeny (Köppel and Schroll, 1983), as for example in the gold bearing vein deposits of the Hohe Tauern.

3. DISCUSSION

There are basically two ways of interpreting the data: (i) either by creating a new lead isotope evolution model such that the lead model ages agree with the age of the deposits determined by other means, or else (ii) by accepting the pres-

Table 2 Lead isotope composition of galenas from stratiform to strata-bound Palaeozoic ore deposits of the Eastern Alps.

				· · · · · · · · · · · · · · · · · · ·			
Sa	ample Occurrence No	206 _{Pb} / ²⁰⁴ Pb	207 _{Pb} / ²⁰⁴ Pb 2	⁰⁸ Pb/ ²⁰⁴ Pb	t ₂ * (my)	μ ₂ *	^w 2*
Pa	laeozoic of Graz:			u.			
1 2 3 4 5 6 7 8	Rabenstein Guggenbach Haufenreith Schrens Burgstall Rechberg Raebstollen Arzwaldgraben	18.055 18.040 18.046 18.060 18.046 18.074 18.051 18.058	15.679 15.674 15.679 15.685 15.675 15.685 15.677 15.680	38.255 38.240 38.253 38.291 38.244 38.294 38.252 38.266	-		
Me	ean	18.054 ±.011	15.679 ± .004	38.263 ± .020	580	10.15	40.9
Ot	her Palaeozoic occur	rences:					
9 10 11 12 13 14 15 16 17 18 19 20	Ramingstein Meiselding Moosburg Rettenbach Walchen Striedalmer Plaike Kaser Wieserl Knappenstube Panzendorf Stockenbai Koprein Schneeberg/ Monte Neve	17.817 17.968 17.959 17.890 17.886 17.933 17.920 17.924 17.908 18.279 18.226 18.276	15.621 15.669 15.666 15.637 15.625 15.635 15.622 15.627 15.637 15.691 15.669	37.970 38.185 38.123 38.061 38.072 38.053 38.038 38.107 38.481 38.479 38.469 38.469 38.469	645 625 620 600 585 570 580 610 440 440	10.0 10.1 10.1 10.0 10.0 9.9 9.9 10.0 10.1 10.0	40.0 41.0 40.7 40.7 40.0 39.9 39.6 39.6 40.4 40.5 40.5
	" "	18.264 18.238 18.237	15.675 15.664 15.660	38.477 38.425 38.397	410	10.0	40.0
21 22	Arzberg Kreiteralm	18.388 18.314	15.692/ 15.648	38.591 38.432	440 330	10.0 9.9	40.5 39.1

The isotope ratios are corrected for fractionation effects. The SRM 981 of the National Bureau of Standards was used to determine the fraction factors:

²⁰⁶ Pb/ ²⁰⁴ Pb	:	1.0030
207 _{Pb/} 204 _{Pb}		1.0043
²⁰⁸ Pb/ ²⁰⁴ Pb	:	1.0060

The reproducibility of the isotopic ratios are $\pm .5$ % for the 206 Pb/ 204 Pb and 207 Pb/ 204 Pb ratios and ± 1 % for the 208 Pb/ 204 Pb ratio.

*) according to the Stacey A. Kramers (1975) Pb isotope evolution model.

ently employed evolution models and interpreting the data on the basis of the geochemical characteristics of the U/Pb and Th/Pb ratios which govern the long term growth of the lead isotope ratios. A lead isotope evolution model can be fitted to the data of the Palaeozoic of Graz. According to WEBER (1983) these are synsedimentary, volcanogenic deposits of lower Devonian age and are the





Solid curves: Evolution curves according to the model of STACEY and KRAMERS (1975); broken curves according to the model of CUMMING and RICHARDS (1975).

MS: Data field of feldspar lead from metasediments of the Southern Alps (5 points) and the Eastern Alps (3 points). OG: Data field of orthogneisses and pegmatites of the Southern Alps (3 data points). The feldspar data point of the amphibolite of Nibbio is not shown $(^{206}Pb/^{204}Pb = 18.728, ^{207}Pb/^{204}Pb = 15.533, ^{208}Pb/^{204}Pb = 37.971$; MORB-type Pb).

biostratigraphically best dated pre-Mesozoic ores of the Eastern Alps. Furthermore they most likely are the biggest of the pre-Mesozoic deposits. They contain an extremely homogeneous lead. A Stacey-Kramers type evolution model with a μ_1 value of 8 from 4.57 to 3.05 by and a μ_2 of 9.8 for the second stage starting at 3.05 by would yield a ²⁰⁷Pb/²⁰⁶Pb model age of 390 my. However, by this model the lead-lead ages of the second cluster would be lowered to about 200 my and those of the economically important syndiagenetic Pb–Zn deposits in Triassic host rocks to about 100 my. Obviously there exists no single evolution model that applies to all stratiform to strata-bound deposits of the Eastern Alps. This indicates that probably none of these deposits contains a lead that is isotopically representative for the Alpine crust. The question whether the data can legitimately be discussed in terms of the accepted evolution models must therefore be solved by examining the isotope pattern of rock leads. So far not enough rock data exist from the Eastern Alps to test the validity of the accepted evolution models. A reasonable number of data is, however, at hand from the western part of the Southern Alps (CUMMING and KÖPPEL in prep.) where the same type of ore lead is again observed in vein-deposits. The data shows that only feldspars of the pre-Ordovician metasediments have a lead isotope pattern similar to the galena ores of the Eastern Alps. Lead of other rock types does not exhibit these high μ and W values. On the basis of these data one therefore has to conclude tentatively that a new lead isotope evolution model for the lead in the stratiform deposits of the East Alpine units as well in other types of deposits in the Eastern and Southern Alps is not justified.

Several models which have been proposed during the past years differ considerably in their approach to define an average lead isotope growth curve. Nevertheless, the evolution curves are all very similar because of the general agreement that the massive stratiform volcanogenic sulfide deposits contain isotopically an average crustal lead. In addition deep sea sediments and manganese nodules also contain well mixed leads of average crustal compositions (STACEY and KRAMERS, 1975, CUMMING and RICHARDS, 1975, DOE and ZART-MAN, 1979, ZARTMAN and DOE, 1981).

The investigations by ZARTMAN (1974), in the Western United States and by SATO and SASAKI (1973, 1980) on the Kuroko and Besshi type deposits in Japan revealed that the lead isotope pattern reflects the broad tectonic setting in which ore and rock forming processes take place. The plumbotectonics model by DOE and ZARTMANN (1979) accounts for these observations by distinguishing 3 types of U-Pb and Th-Pb reservoirs with distinct μ and W values. In the upper crust U and Th are normally enriched and the μ and W values are high leading thus to an accelerated growth of the isotope ratios. The lower crust (granulite facies rocks) has low μ values, the growth of the uranogenic lead isotopes is therefore retarded, and W values similar to upper crustal rocks. In the oceanic mantle the μ and W values are low compared to upper crustal rocks.

The data of stratiform to strata-bound deposits of the Eastern Alps, as well as from other types of deposits from this unit and the Southern Alps, show that the main source of the lead must be sought in rocks with a long crustal history. The high μ and W values demonstrate a prolonged residence time of the lead in an upper crustal environment. The data point from the Palaeozoic of Graz, for example, indicates that the upper crustal development of the lead must have started before 1300 my (Stacey-Kramers model) or 1200 my (Cumming-Richards model). Such old source material could conceivably be contained in the pre-Ordovician metasediments which all contain detrital zircons with approximate primary ages of 2000 to 2700 my (Köppel and GRÜNENFELDER, 1971). As already mentioned above the feldspar-lead of these metasediments is isotopically similar to the ore-lead (fig. 2) and must therefore have evolved in a similar way. It should be noted here that the 87Sr/86Sr initial ratio reported by HUNZI-

KER and ZINGG (1980) of $0.7086 \pm .0008$ for metasediments of the Ivrea zone and $0.7107 \pm .0018$ for metasediments of the Ceneri zone do not comport with a long crustal history of the sedimentary material as deduced from the age of detrital zircons and from the lead isotopic composition of the feldspars. This discrepancy is not exceptional. Similar cases are reported by GRAUERT (1969) for paragneisses of the East Alpine Silvretta nappe and JÄGER (1979) commented upon the observation that the low initial 87Sr / 86Sr ratios of all analyzed paragneisses from central Europe seem to indicate a sedimentation age of 500 to 900 my, provided that the sediments exchange Sr with the sea water. Such a mechanism implies that the strontium balance of the sediments was largely governed by newly formed or transformed clay minerals (CLAUER, 1979). The sediments possibly also contained significant amounts of material derived from basic volcanics with high strontium concentrations, but low 87Sr / 86Sr ratios and low lead concentrations.

The lead of the stratiform to strata-bound ore deposits did, however, not evolve entirely in an upper crustal environment. The generally too high 207Pb/ ²⁰⁶Pb model ages require that the source material at one stage in its history lost uranium; thereby the U/Pb ratio was lowered and the growth of the uranogenic isotopes retarded. Such an event may have been coupled with a high grade, i.e. granulite facies metamorphism during which uranium that is not tightly held in crystall-lattices tends to escape with the fluid phase whereas the lead is retained in the feldspars. Thorium is more readily accepted in crystalline phases than uranium, and therefore the generally high W values of the upper crustal stage are not severely changed during high grade metamorphism. The ²⁰⁸Pb / ²⁰⁴Pb ratio continues to develop at an accelerated rate leading to low ²⁰⁸Pb/²⁰⁴Pb model ages. Another possibility is that the source material of the pre-Ordovician sediments lost uranium during erosion and deposition as sandstones or greywackes in the foreland throughs. During such processes intergranular uranium and uranium in easily soluble phases may be removed and consequently the u values will be lowered. According to the data of WEDEPOHL (1969) the average U/Pb ratios of sandstones, including beach sands, correspond to a present-day μ value of about 7 with a large spread from 2 to 12 (average crustal value appr. 10). Also the Th/Pb values of sandstones, greywackes and recent beach sands are lower, 20 to 30, than the average crustal value of appr. 37. On the basis of the lead isotope pattern a uranium loss under high grade metamorphic condition is favored to explain the high 207Pb/206Pb and low 208Pb/204Pb model ages. On geological grounds the second explanation appears attractive, but it needs testing by comparing rock leads from metapsammites with lead from metapelites.

Implications regarding the tectonic setting

From the foregoing discussion it follows that the pattern of the lead isotope ratios of galena-bearing ore deposits in the East Alpine nappe units, and also in the Southern Alps, require the dominant presence of early Proterozoic source material that acted as a source bed for lead. In the adjacent region situated today to the North, the Penninic area, and further northwards the lead isotope pattern follows a normal trend in accordance with the accepted growth curves (KÖPPEL, 1983) One may therefore speculate that in pre-Ordovician to late Proterozoic time there existed in the South an old continent bordered towards the North by geosynclinal troughs which acted as repositories for detritus from the old continental mass. These sediments are now the metasediments of the Southern Alps and the East Alpine nappe units. They also contain amphibolites. So far only the lead from feldspars of the amphibolite from Nibbio (Ivrea zone) has been analyzed (CUMMING and KÖPPEL, in prep.) and the results strongly suggest that it represents a MORB type basalt. The formation of the geosynclinal foreland trough was therefore possibly accompanied by ocean floor rifting. (In contrast to this amphibolite the ultramafic rocks of the Ivrea zone, the peridotites, the layered series and the associated Ni-Cu-Fe ores as well as the Gabbros exhibit a lead isotope pattern distinct from oceanic mantle lead, CUMMING and KÖPPEL in prep.) According to HUNZIKER and ZINGG the sedimentary pile was metamorphosed 478 ± 20 my ago (see also ZINGG, this volume). U-Pb zircon ages (Köppel and Grünenfelder, 1971) and Rb-Sr whole rock ages (Bo-RIANI et al., 1981) of the orthogneisses indicate a period of magmatism 440 to 470 my ago. The feldspar lead from the orthogneisses distinguishes itself from the lead observed in feldspars from the metasediments by its normal μ and W values which preclude an origin of these magmas by melting of metasedimentary material. In the context of the plumbotectonic model these leads would argue for a mature island arc or an oceanic-continental plate boundary situation. Similarly FRANK et al. (1976) argued on the basis of 87Sr/86Sr initial values of leucocratic gneisses interfingered with amphibolitic layers in Gleinalpe region for island arc situations. There is at present no evidence available for the existence of post-Ordovician mafic oceanic crust within the East Alpine nappe units or the Southern Alps.

In the Penninic realm pre-Ordovician metasediments are present but they not appear to have acted as important source beds for lead probably because they play quantitatively not such an important role as they do in the Southern Alps and the East Alpine nappes. Island arc situations probably prevailed during the Silurian and lower Devonian (Höll and MAUCHER, 1976) and the lead isotope data especially from the Tauern area favor this idea (Köppel, 1983, Köppel and Schroll, 1983).

Although these conclusions are speculative, it appears that the lead isotopes

in conjecture with other methods will play in the future an important role in deciphering the pre-Mesozoic history of the Alps.

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