Chapter 5

Lead Isotopes as Tracers of Metal Sources and Timing of the Carbonate-Hosted Pb-Zn Deposits in the Nappes Zone, Northern Tunisia

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Additional information is available at the end of the chapter

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Abstract

The polymetallic deposits in the Nappes zone, Northern Tunisia occur in the contact between Triassic-Miocene/Eocene carbonate rocks and in the Upper Cretaceous limestones. They can be divided into two groups: one is Pb-Zn mineralization with Hg and As in fractures with local intrusions of Neogene volcanics (e.g., Fej Hassene, Oued Maden), and the second is stratabound and karst Pb-Zn mineralization with arsenic and antimony hosted in the continental Neogene strata or situated immediately below them (Aïn Allega, Sidi Embarek, Jebel Hallouf-Sidi Bou Aouane, Bazina, Jalta and Jebel Ghozlane). Pb-isotopic compositions of galenas display a homogeneous Pb isotope signature. Generally, Pb isotope ratios on ores from the Jalta, Jebel Ghozlane, Jebel Hallouf, Oued Maden and Fedj Hassene plot between samples of the Late Miocene igneous rocks and the sedimentary country rocks of the Nefza area and between the upper crust and orogen curves. This intermediate position may imply potential mixing between end-member sources. Because the Pb-Zn mineralization is fault-controlled and spatially associated with the post-nappe Miocene series and the calculated model age is about 10.86 Ma, one is led to argue that the mineralization in the Nappes zone deposits occurred during the last paroxysmal phase of the Alpine folding (i.e., Miocene age).

Keywords: Pb isotopes, carbonate-hosted Pb-Zn deposits, mixing sources, Upper Miocene age, Nappes zone

1. Introduction

The Nappes zone (**Figure 1**), which constitutes the eastern prolongation of the Atlas orogenic belt of North Africa, is composed of thrust sheets that resulted from a major Neogene tectonic

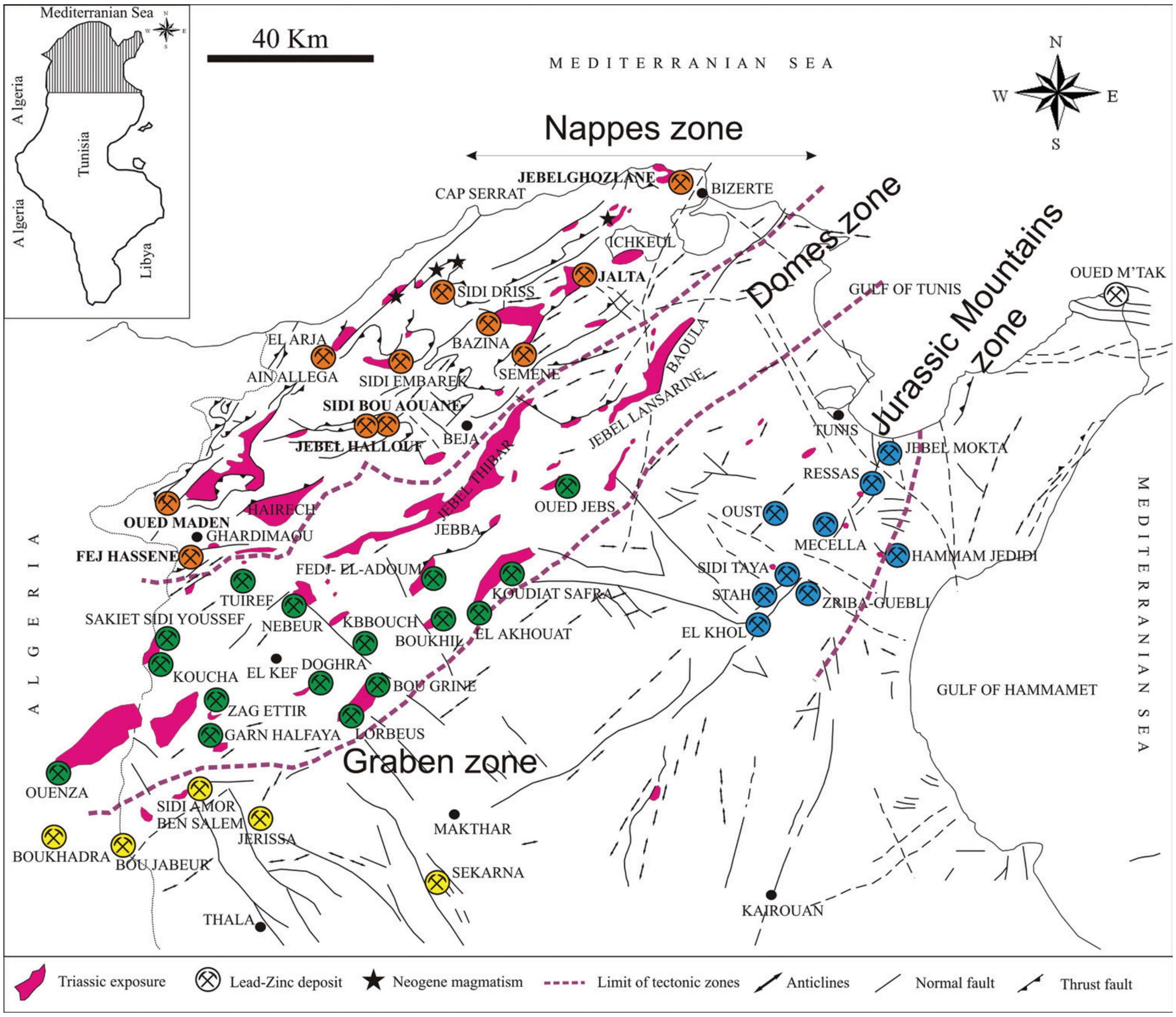

Figure 1. Geological zones, Triassic exposures and Pb-Zn deposits in Northern Tunisia (adapted from Jemmali et al. [1].) Brown circle: Nappes zone Pb-Zn-Hg (As) deposits. Green circle: Domes zone Pb-Zn (Ba-Sr) deposits. Yellow circle: Reef Aptian zone Pb-Zn deposits. Blue circle: F-Ba (Pb-Zn) deposits.

event [2]. The thrust sheets moved during the Early to Middle Miocene from NW to SE along regional westerly dipping fault planes in shear zones that vary in strike from E-W to SW-NE [2, 3]. The Nappes zone comprises Mesozoic and Tertiary sediments and is characterized by a major thrust sheet of Oligo-Miocene "Numidian" flysch that overrides younger flysch strata mainly of Cretaceous and Eocene age [4]. This zone includes the following units: (1) the Numidian nappe consists of a thick series of siliciclastic flysches made up of sandstones and argillites, which is of Oligocene to Burdigalian age; (2) the Tellian units dated Late Cretaceous to Early Eocene. It is composed of three formations: the Abiod (marl and limestones), El Haria (marl and shale) and Metlaoui (limestones); and (3) the Miocene-Pliocene Molasse includes coarse conglomerates and red sandy shales [4]. The Late Miocene major orogeny resulted in the Neogene basins along NNE-SSW directions and the emplacement of the nappes. This compressive event was followed by the development of longitudinal faults and bimodal volcanism [5, 6]: (1) granodiorite and rhyodacites were likely emplaced in a Serravallian-Tortonian

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compressive context, and (2) basalts may relate to the following Messinian rifting event. In the Nappes zone, the Triassic outcrops (variegated clays, sandstones, dolostones, gypsum breccias) often occur at the base of the overlapping units (Aïn Jantoura, Hédil), as well as diapiric units cross-cutting the allochthonous formations (structural trend of Ghardimaou-Ain Draham-Cap Serrat, Jebel Zouza) [7].

The Nappes zone hosts numerous polymetallic deposits (**Figure 1**). The mineralization is associated either with the Late Miocene orogeny (emplacement of the nappes) or with the Neogene volcanism. Two groups of deposits are present there: (1) stratabound and karst Pb-Zn mineralization with arsenic and antimony hosted in the continental Neogene strata or situated immediately below them (Aïn Allega, Sidi Embarek, Jebel Hallouf-Sidi Bou Aouane, Bazina, Semene, Jalta, Bechateur, Ain el Bay, Chouichia) and (2) Pb-Zn mineralization with arsenic and mercury in fractures with local intrusions of Neogene volcanics (Fej Hassene, Oued Maden, Jebel Arja).

The current study represents a synthesis of Pb isotope data collected from papers published by Jemmali et al. [1, 8–10] of Jalta, Jebel Ghozlane, Jebel Hallouf, Oued Maden and Fedj Hassene deposits in the Nappes zone (**Figure 1**) and comparison to previously published Pb isotope ratios of Neogene igneous rocks and sedimentary cover rocks from the same zone. This will help constrain the source(s) of metals and the possible age of mineralization.

2. Setting and characteristics of the Nappes zone Pb-Zn deposits: case study

2.1. Jalta

The lithostratigraphic units consist mainly of (**Figure 2**): (1) the Triassic series consist of chaotic shales and dolomitic limestones containing gypsum and alunite; (2) the Upper Cretaceous and Eocene series are composed of marls and limestone, respectively; (3) Middle to Upper Miocene strata are represented by continental, detrital facies composed of gray conglomerates and lenticular limestones, lacustrine limestones, and blue marls alternating with conglomerates, overlying the Triassic, Cretaceous and Eocene deposits; (4) the continental Pliocene series lies on the Upper Miocene conglomerates; and (5) the Quaternary continental deposits are represented by alluvium.

At the regional scale, the location of the basin of Jalta is mainly controlled by two main structural trends [12]: N30 Ras El Korane-Thibar major fault on the northwest side and Messeftine N-S fault on the eastern side accompanied on the southern side by the secondary N140E Mateur fault. At the deposit scale, the main structural features in and around the Jalta district consist mostly of strike-slip faults and few normal faults [4, 13], having different orientations (**Figure 2**). NE-SW trending sinistral strike-slip faults with N40-N60 exist far from the Jalta mine. In the northwestern part of the district, NE-SW trending sinistral strike-slip faults are considered as a deep fault where Triassic rocks are in contact with Miocene series. NW-SE trending dextral strike-slip faults with N120-N150, with distensive components, cut the Jalta

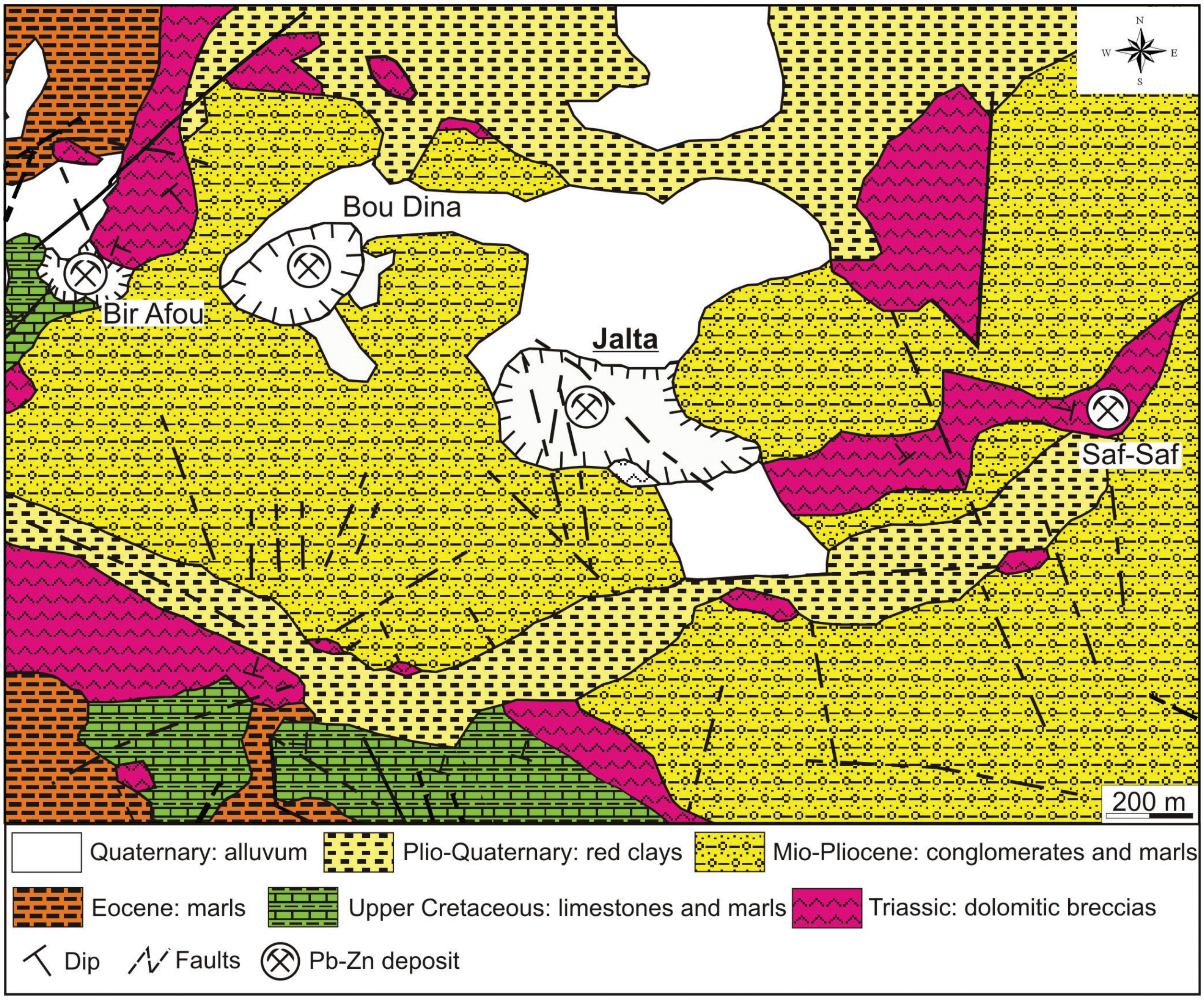

Figure 2. Geological map of Jalta district (adapted after Crampon [11].).

mine and other areas around it. These sinistral and dextral strike-slip faults host mineralization. In contrast, E-W trending normal faults, with a tectonic style of horst-and-graben structures, are more expressed outside west of the district and do not host Pb-Zn mineralization. All these sets of faults, especially the sinistral strike-slip faults, are due to the N-S directed deformation that affected the region during Miocene [4].

The Jalta mine has produced 75,000 tons of ore grading 59% Pb. The Pb-Zn mineralization in the Jalta deposit ([1, 10]; **Figure 2**) is located mainly near and along the contact of the Triassic rocks with the Miocene series. The mineralization mainly hosted by brecciated Triassic dolostones is composed of galena, barite and minor pyrite, jordanite and sphalerite occurring as veins, stockworks, disseminated, karstic and breccias cement [10].

2.2. Jebel Ghozlane

Based on several studies [14, 15], the stratigraphy of Jebel Ghozlane consists mainly of (**Figure 3**): (1) the Triassic breccias composed of variegated clays, sandstones and dolostones; (2) the Upper

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Cretaceous rocks consist of massive limestones with marl intercalations; (3) the Maastrichtian-Paleocene is represented by a marl-bearing series; (4) the Lower Eocene rocks consist of marine limestones with *Globigerina* sp.; (5) the Upper Eocene consists of marls and (6) the Quaternary is represented by marine facies. NNE-SSW trending dextral strike-slip fault characterizes the contact between the Triassic and Eocene at Jebel Ghozlane, but in other parts of the area the main contact between them is an unconformity. At Jebel Daouda, thrust faults trending NE-SW separate the Upper Eocene rocks from the Cretaceous and Triassic rocks, and the Upper Eocene rocks from the Lower Eocene rocks at Jebel Touila.

The Jebel Ghozlane deposit, which is among the largest of the Pb-Zn deposits in and near the Bechateur district, is situated along faults and a thrust-sheet boundary (**Figure 3**). It produces ca. 6680 tons of Pb and 53,128 tons of Zn pure metals. The deposit is hosted by Triassic dolostones and Lower Eocene dolomitic limestones. The orebodies, which occur as vein, dissemination and breccia styles, are localized along N150-160 dextral strike slip-fault contact between the Triassic and Eocene rocks (**Figure 2**). The ore of the deposit consists of galena, sphalerite and minor pyrite, with barite and celestite as gangue minerals.

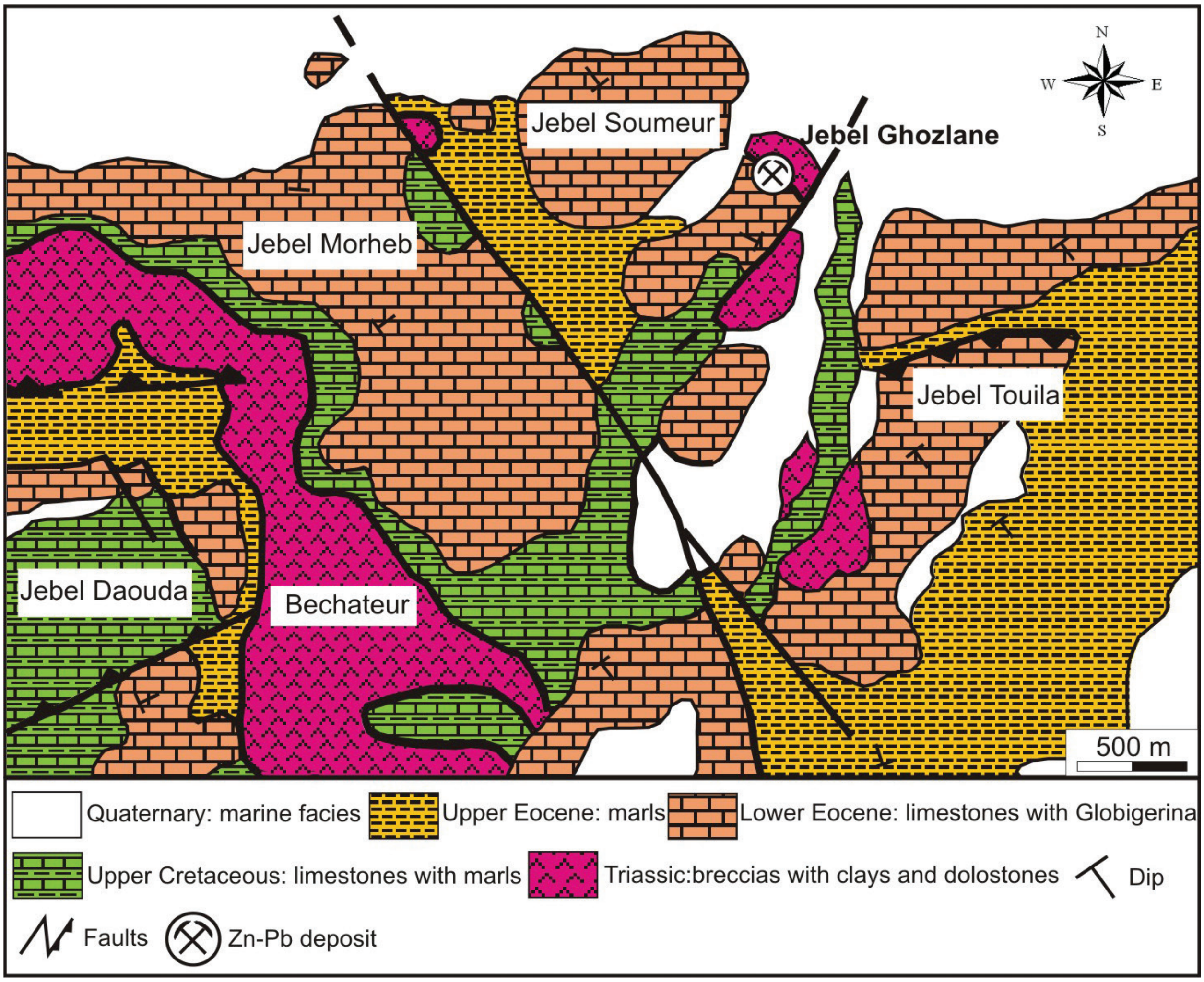

Figure 3. Geological map of Jebel Ghozlane. (Adapted from Melki et al. [15].)

2.3. Jebel Hallouf

The stratigraphy of Jebel Hallouf consists of a Triassic-Miocene series overlain in fault contact by the Kasseb Paleocene-Oligocene allochthonous unit, which in turn is overlain by the Neogene post-nappe continental series [4, 16] (**Figure 4**). The Triassic series consists of dolomitic breccia and evaporates, unconformably overlain by Lower-Upper Cretaceous clay-rich limestones. Upper Cretaceous-Paleocene series consists mainly of marls, which in turn are overlain by Eocene limestones. The overlying Oligocene-Miocene strata are made of sandstones. Five successive extensional and compressional episodes have characterized the tectonic evolution of Jebel Hallouf [17]: NE-SW-trending upright isoclinal open to closed folds, and a succession of closed-to-tight N30°–40°E oriented saline rock-cored anticlines and broad E-W to N-S-trending open-to-gentle synclines, truncated and/or accompanied by a series of ENE-WSW, NW-SE, E-W and N-S trending faults.

The Jebel Hallouf deposit was mined by the SOTEMI mining company between 1965 and 1986 and has produced since its exploitation in 1910 the equivalent of 326,541 tons of Pb and 14,207 tons of Zn metals. Mineralization is hosted by Campanian-Maastrichtian limestones. The style of mineralization consists of cavity-filling of karstic features that cut the mentioned host rocks on the north side of the Jebel Hallouf anticline. Vertical cavities developed from WSW-ENE and NNW-SSE joints were partially filled up by stratified mineralized calcite. The mineral association consists of calcite, sphalerite, galena, jordanite and pyrite.

2.4. Oued Maden

The lithostratigraphic units consist of a series of stacked nappe structures produced by regional westward intra-Miocene tangential over-thrusting and Upper Miocene to Pliocene-Quaternary post-nappe tectonic phases [18]. Two main nappes are recognized [4]: (1) the

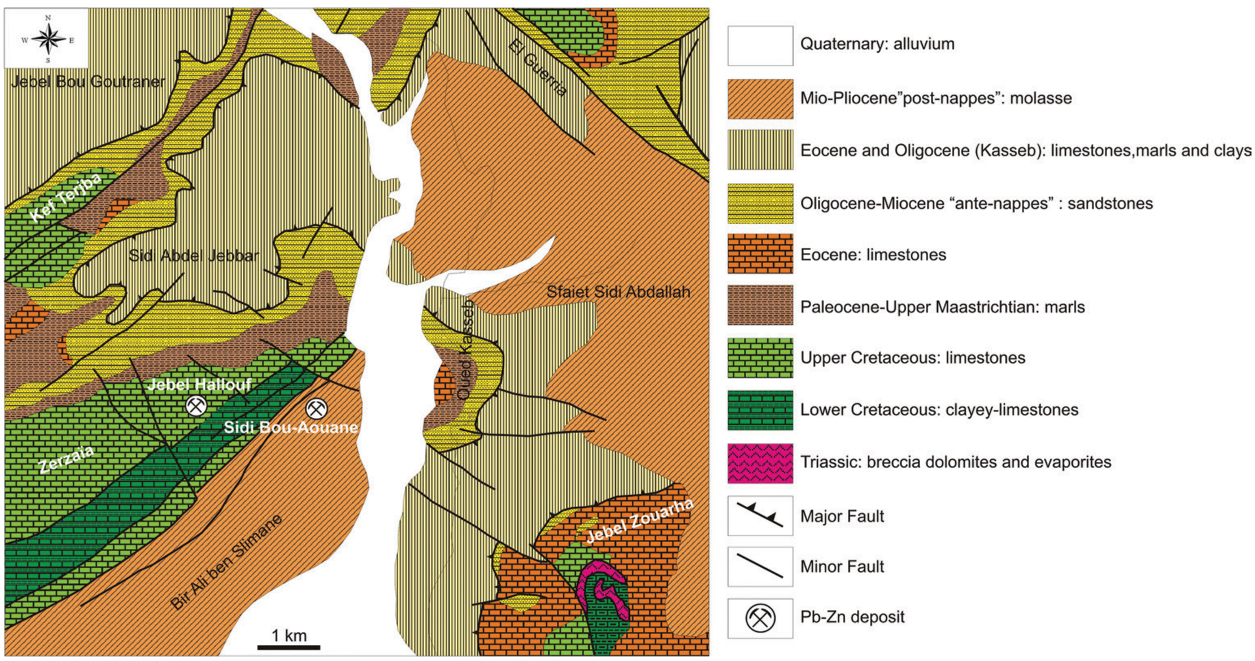

Figure 4. Geological map of Jebel Hallouf (adapted from [16].).

Numidian nappe consisting of Oligocene clay-rich sandstones and (2) the Ed-Diss nappe comprising Senonian (Upper Campanian-Lower Maastrichtian) and Eocene sediments. The autochthon beneath these nappes displays a normal stratigraphic sequence consisting of Santonian-Maastrichtian (Late Cretaceous) rocks (**Figure 5**). The Upper Santonian-Lower Campanian consists of thinly bedded gray marls with rare intercalations of marly limestones. These series are in fault contact with the underlying Triassic rocks and are overlain by thick succession of marls and limestones of Upper Campanian-Lower Maastrichtian age, followed by thick gray sulfide-rich limestones of Middle Maastrichtian age.

From 1900 to 1955, about 11,500 tons of Pb and 89 tons of Cu pure metals (with ca. 350 g/t Ag) were produced from open pit and underground workings [18]. Mineralization occurs as open-space fillings of veinlets and stockwork structures superimposed on the NE-SW-trending Groura and Ferza faults cutting across the Triassic dolostone and the Campanian-Maastrichtian limestone country rocks. The Groura and Ferza faults (**Figure 5**) are part of a regional major structure referred to as the Ghardimaou-Cap Serrat fault (**Figure 1**). The ore mineralogy is dominated by galena, sphalerite, pyrite and sulfosalts (mainly tetrahedrite).

2.5. Fedj Hassene

The lithostratigraphic units of Fedj Hassene (**Figure 6**) consist of: (1) Triassic series consist of limestone, dolostone and gypsum-bearing red argillite breccia, locally truncated by regional E-W striking faults; (2) Aptian gypsum-bearing dolomitic breccias; (3) Albian marls and limestones; (4) Cenomanian limestones and marls capped by a Turonian succession of marls and limestones; (5) Eocene nummulitic limestones; (6) Oligocene marls and sandstones; and (7) continental Neogene marls, sandstones and conglomerates. The Cretaceous ends with thick Coniacian-Santonian marls followed by a succession of Campanian-Maastrichtian marls and limestones. The major structure in the Fedj Hassene deposit is the Ain el Kohla ESE-WNW-trending fault.

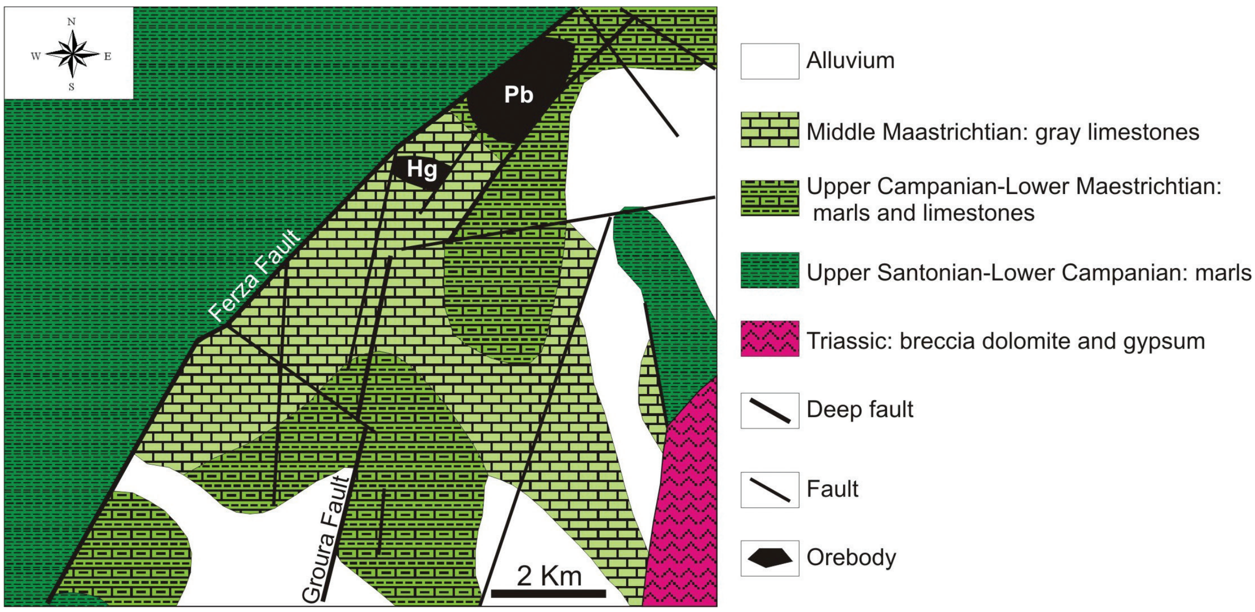

Figure 5. Geological map of Oued Maden. (Adapted from Slim-Shimi [19].)

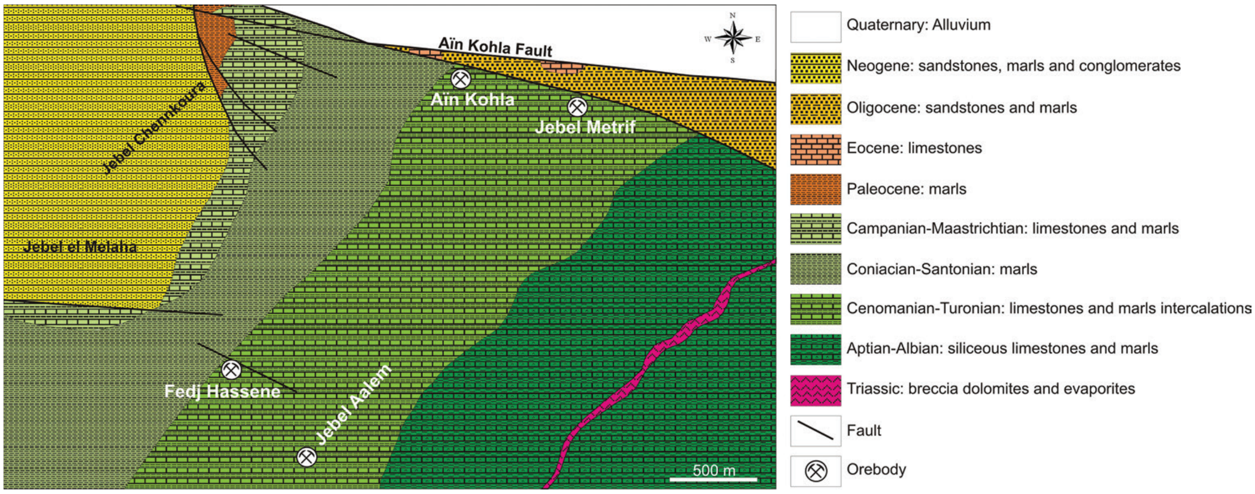

Figure 6. Geological map of Fedj Hassene. (Adapted from Sainfeld [20].)

During the lifetime of the mine that exploited the Fedj Hassene deposit until 1992, about 55,600 tons of Zn and about 300 tons of Pb pure metals have been produced. The Zn-Pb mineralization mainly occurs as ESE-WNW-trending veins and stockworks enclosed in Upper Cretaceous limestones. The mineralization consists mainly of sphalerite and galena with minor amounts of pyrite and chalcopyrite.

3. Data source

The lead isotopic compositions obtained on galenas from the abovementioned deposits are listed in **Table 1** and plotted on conventional covariation diagrams in **Figures 7** and **8**. The Pb isotope ratios range between 18.695 and 18.894 for $^{206}Pb/^{204}Pb$, 15.661 and 15.684 for $^{207}Pb/^{204}Pb$, and 38.718 and 38.917 for $^{208}Pb/^{204}Pb$. When it is plotted on conventional isotopic diagrams, Pb in the ore samples from the two deposits (Jalta and Jebel Ghozlane) defines two distinct fields (**Figures 7** and **8**). However, the radiogenic nature of the Pb characterizes the Oued Maden. The ore samples are quite homogeneous for each deposit and have higher $^{206}Pb/^{204}Pb$ values than samples from the Late Miocene igneous rocks of the Nefza area. The $^{207}Pb/^{204}Pb$ versus $^{206}Pb/^{204}Pb$ ratios plot between the orogene and upper crustal curves but close to the upper crustal reservoir in the plumbotectonics model of Zartman and Doe [22] (**Figure 7**), whereas the $^{208}Pb/^{204}Pb$ ratios versus $^{206}Pb/^{204}Pb$ plot slightly above the orogen curve. The narrow range of data may suggest a well-mixed source (see discussion). The compositional variation of Jalta and Jebel Ghozlane (two distinct fields), Jebel Hallouf, Oued Maden and Fedj Hassene may be due to an input of Pb from different sources and mixing of multiple metal-bearing brines. The samples from Jebel Hallouf, Jalta, and Jebel Ghozlane suggest ore deposition from a similar hydrothermal fluid. A comparison of the Pb isotope ratios of ore samples in the studied deposits has been made with previously published Pb isotope values of Late Miocene igneous rocks and sedimentary country rocks from the Nefza area (**Figure 7**). Some of the ore samples have Pb isotope ratios that plot very close to previous results by Decrée et al. [21].

Deposit name	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$
Jalta	18.8210	15.6770	38.8740
	18.8230	15.6760	38.8750
	18.7200	15.6650	38.7300
	18.8200	15.6740	38.8708
	18.8510	15.7290	39.0610
Oued Maden	18.8900	15.6800	38.9070
	18.8940	15.6840	38.9170
	18.8840	15.6750	38.8880
Fedj Hassene	18.7920	15.6660	38.8490
	18.7880	15.6630	38.8620
	18.7880	15.6630	38.8380
Jebel Hallouf	18.8200	15.6780	38.9140
	18.8200	15.6800	38.9130
	18.8200	15.6760	38.8690
Jebel Ghozlane	18.7140	15.6680	38.7250
	18.7030	15.6680	38.7430
	18.7020	15.6670	38.7390
	18.7020	15.6670	38.7320

Table 1. Lead isotope composition of galena from Jebel Ghozlane [1, 8], Jalta [1, 10], Oued Maden, Fedj Hassene and Jebel Hallouf [8].

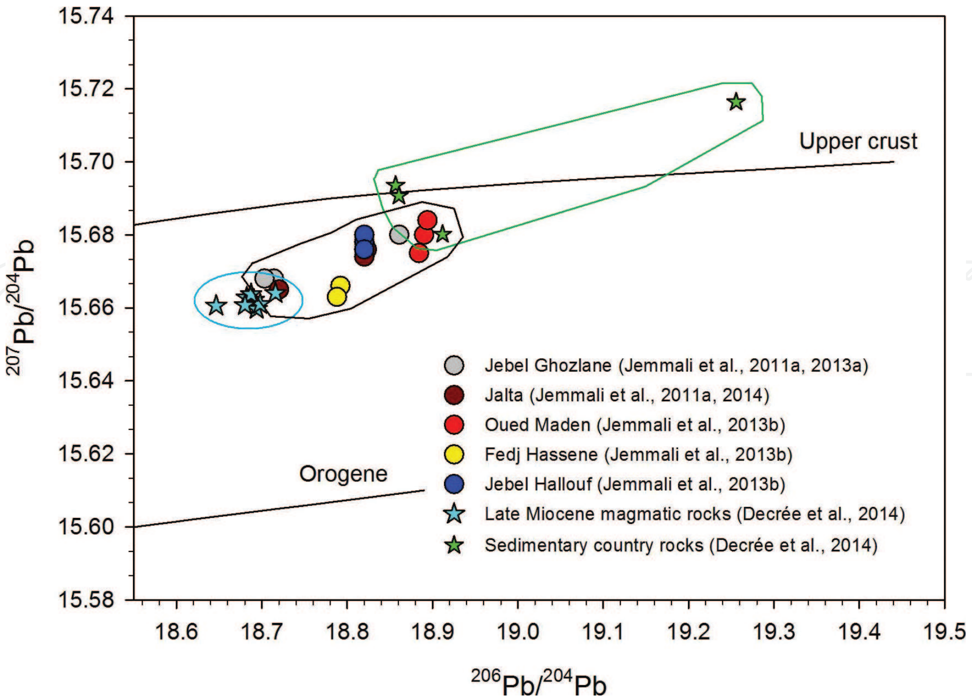

Figure 7. Plots of $^{207}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ for the Pb-Zn deposits in Jalta, Jebel Ghozlane, Jebel Hallouf, Oued Maden and Fedj Hassene compared to the Late Miocene igneous rocks and sedimentary country rocks from the Nefza area [21]. Curves of growth trends for Pb isotope ratios are from the plumbotectonic model of Zartman and Doe [22].

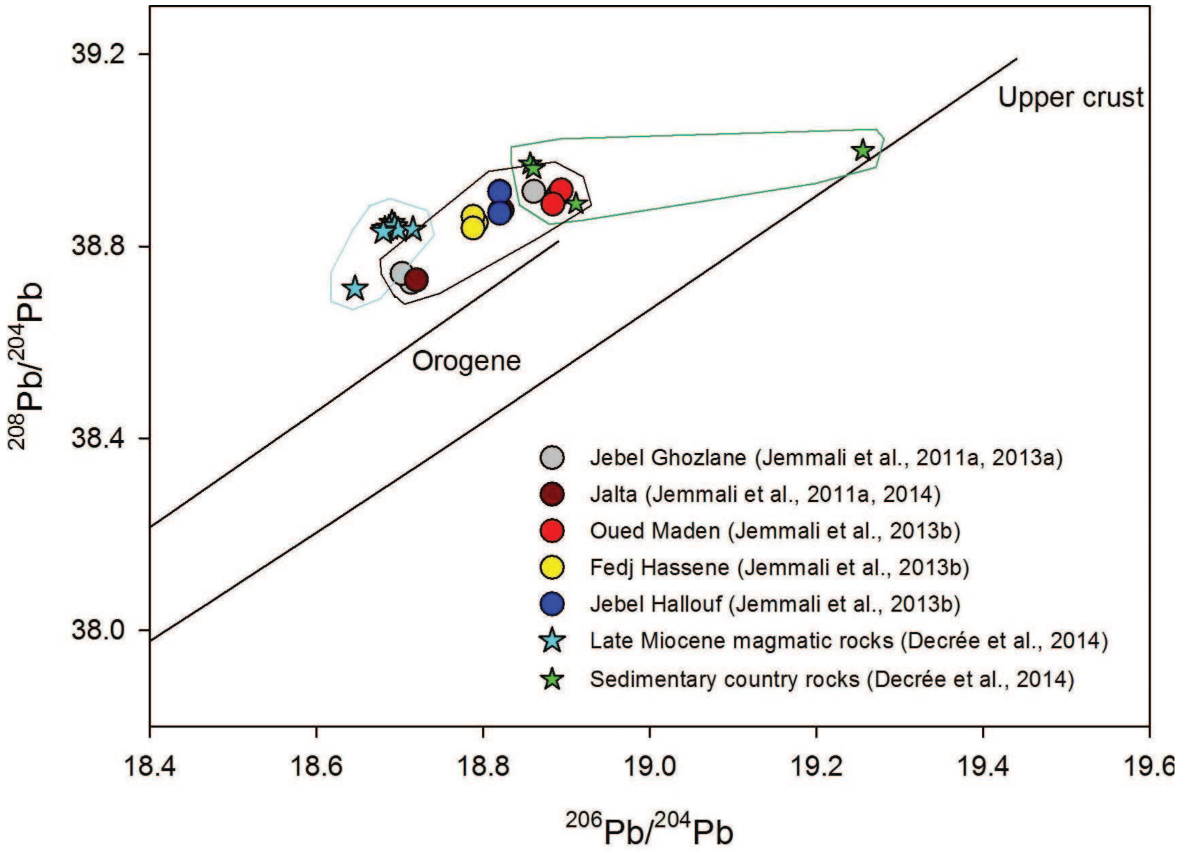

Figure 8. Plots of $^{208}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ for the Pb-Zn deposits in Jalta, Jebel Ghozlane, Jebel Hallouf, Oued Maden and Fedj Hassene compared to the Late Miocene igneous rocks and sedimentary country rocks from the Nefza area [21]. Curves of growth trends for Pb isotope ratios are from the plumbotectonic model of Zartman and Doe [22].

4. Discussion

4.1. Source(s) of metals

The restricted range of isotopic ratios in the studied ore deposit cluster may suggest that Pb originated from a well-mixed source. Overall, the Pb isotope ratios of ore samples plot within the field defined by Late Miocene igneous rocks and sedimentary country rocks implying potential mixing between end-member sources originating from the abovementioned similar metal source(s) (**Figure 7**). The Late Miocene igneous rocks correspond to the basement of the Atlasic foreland from which they are originated [23]. The plausibility of basement rocks as the deep-seated source of Pb in the studied ore deposits is supported by the similarity of Pb isotopic ratios between the Late Miocene igneous rocks and the abovementioned deposits, as well as the presence of jordanite, orpiment and realgar minerals. Jordanite is also present in the Lengenbach Pb-Zn-As-Tl-Ba deposits, which are hosted in Triassic dolostones in the Swiss Alps, for which it was proven that Pb and other metals were leached from basement rocks [24]. Another support for the plausibility of basement rocks as the deep-seated source of radiogenic Pb in the studied deposits is the presence of inherited faults connected to deep-seated faults cutting the Mesozoic-Cenozoic cover and the Triassic salts (**Figure 3**) and

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possibly extending into the basement, like the NE-SW Ghardimaou-Cap Serrat, the NE-SW Ras El Korane-Thibar and the N-S basement faults. However, the occurrence of the studied deposits mainly in Upper Cretaceous limestones and Miocene conglomerates also suggests plausible late remobilization of metals.

Nevertheless, the positions of the $^{208}Pb/^{204}Pb$–$^{206}Pb/^{204}Pb$ data slightly above the orogen curve (**Figure 8**) imply some contribution of Th-derived Pb (cf. [25]). The likely source of Th-derived Pb is the rocks in the Hercynian fold belt of North Africa, which include monazite-bearing Paleozoic metasediments and Hercynian granitoids [26]. These Paleozoic rocks are host to some of the world's economically significant base metal deposits [27], have been deformed and/or emplaced during the Hercynian orogeny, and form the basement exposed in scattered inliers of the Atlas systems in Morocco and Algeria [28, 29]. Thus, it is plausible that basement rocks in Northern Tunisia are similar to those in Algeria and Morocco. If that is the case, and following the plumbotectonic models [22], the source of Pb in galena of the studied deposits can be inferred as follows. Lead in the northern African Paleozoic basement rocks, which were deformed and/or emplaced during the Hercynian orogeny, became part of a well-mixed multi-source upper crust reservoir. Evidence to this is the rather homogenous Pb isotope data from Zn-Pb ores hosted in pre-Hercynian metasediments and from Hercynian granitoids [27]. Then, Pb in the well-mixed multi-source upper crust reservoir was partly recycled back into the mantle and/or passed on to favorable host rocks in an orogeny [22, 30]. The orogeny in this case is the Maghrebides fold-thrust belt, which was formed during the Alpine orogeny and where the studied Pb-Zn deposits are hosted primarily in Triassic carbonate rocks and partly also in the cover rocks. A second source may be the sedimentary cover rocks, and the Pb isotope ratios of the studied ore deposits are plotting near to the Pb isotope values defined by the second source (**Figure 7**). The current Pb isotope data support Decrée et al. [23] conclusion, suggesting that mixing between end-member sources originating from the basement igneous rocks and the sedimentary cover rocks was responsible for providing metals for the Nappes zone deposits.

4.2. Age of mineralization

The governing mechanisms during the first phase of halokinesis which has taken place likely during Jurassic to Middle Cretaceous [31] were presumably controlled by Jurassic-Lower Cretaceous normal faults inherited from Tethyan rifting [32]. The salt diapirs have been active again during the Alpine orogeny (Early-Middle Miocene compressional events produced folds, nappe emplacement and bimodal volcanism and restarted the halokinetic phenomena [31]). Because the Pb-Zn mineralization in the Nappes zone is associated with the post-nappe Miocene series, one is led to conclude, therefore, that the mineralization is related to the last paroxysmal phase of the Alpine folding (i.e., Miocene age). Because the Pb-Zn mineralization in the studied deposits is fault-controlled and spatially associated with the post-nappe Miocene series, one is led to hypothesize that the mineralization occurred during the last paroxysmal phase of the Alpine folding (i.e., Miocene age). The calculated model ages using the Pb isotope model of Stacey and Kramers [33] range from 2.7 to 21.6 Ma with a median of 10.86 Ma (excluding negative values of Oued Maden), indicating an Upper Tertiary-Quaternary age. This age, which was attributed to the F-(Ba-Pb-Zn) ores of the Zaghouan district [14, 34, 35] and to the polymetallic mineralization of the Nappes zone [23, 36] and which is similar to that recently proposed for the world-class Touissit-Bou Beker district of northeastern Morocco [37], coincides with the Serravallian-Tortonian magmatism event [38] and with the mid-Miocene Alpine compressional tectonics in Northern Tunisia [39]. Thus, the Tertiary orogeny in this case, as mentioned before, is represented by the Maghrebides fold-thrust belt, which was formed during the Alpine orogeny. Similarly, a genetic link between the Messinian mafic magmatism and Sidi Driss Pb-Zn deposit has been proposed by Decrée et al. [40]. In the Oued Maden deposit, the Pb-isotopic data give negative model ages. This implies that an anomalous Pb was introduced to characterize ore Pb in the Oued Maden deposit, which gave negative or excess model age suggesting significant radiogenic contamination. This kind of highly radiogenic Pb known as J-type Pb [41] could be derived from high U and Th crustal source reservoirs. Accordingly, resulting radiogenic Pb was likely stored in basement rocks before being remobilized and redeposited in the cover rocks in the studied area.

5. Conclusions

This study presents a lead isotope database of 18 galena samples from the selected deposits belongs to the Nappes zone, Northern Tunisia. The isotopic signatures of these carbonate-hosted Pb-Zn deposits reflect the source(s) of metals and the probable age of mineralization. The current Pb isotope data support suggests that mixing between end-member sources originating from the basement igneous rocks and the sedimentary cover rocks was responsible for providing metals for the Nappes zone deposits. The calculated model indicates an Upper Tertiary-Quaternary age for the emplacement of the mineralization. This study may be useful for mineral exploration and archaeological correlation of metal artifacts.

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