which should be cited to refer to this work.

Lead isotope analyses of gold−silver ores from Roşia Montană (Romania): a first step of a metal provenance study of Roman mining activity in *Alburnus Maior* (Roman Dacia)

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The Roşia Montană ore deposit (Apuseni Mountains, Romania) is Europe's largest Au−Ag deposit. It also corresponds to the Roman *Alburnus Maior* mining site, known by historians and archaeologists due to the discovery of dozens of Roman wooden wax tablets during the underground works carried out during the 18th and 19th centuries.

The present geochemical research is based on a detailed archaeological and geological study of the Roman mines at Roşia Montană, making use of archaeologically and geologically documented ore samples. The geochemical analyses allowed us to establish an accurate database for the ores exploited during Roman times at Roşia Montană (and probably before). This approach represents a contribution towards improving the accuracy of metal provenance studies of gold−silver ores during antiquity in Romania, and also at an European level, because the studied ore samples represent remnants of the original ores used by the Romans for the production of precious metals.

Twenty-nine ore samples and one litharge roll have been selected, prepared and analysed by MC-ICP-MS (high-resolution measurements). A specific Roşia Montană Pb isotope signature of gold−silver ores extracted by the Roman miners was obtained. This signature is distinct when compared with other ore deposits from the Apuseni Mountains, as well as within a broader region (Maramureş ore district).

A litharge roll discovered in a Roman inclined adit situated close to the surface, which attests the presence of metallurgical workshops, has also been analysed. The different lead isotope values of the litharge roll and the Roşia Montană gold−silver ores suggest that other ore sources from the South Apuseni Mountains or from elsewhere were also employed by the gold metallurgy developed at Roşia Montană during Roman times.

1. Introduction

At the beginning of the 2nd century AD, a new economic space was established in the north-eastern part of the Roman Empire, more specifically in the new Roman province of Dacia. This province was conquered by Rome in 106 AD after the victory of Trajanus against Decebalus, the last Dacian king. The assets of this new territory were mainly based on the important gold and silver resources located in the so-called Apuseni Mountains, part of the Carpathian chain (Fig. 1a). Due to the abundance of Au−Ag epithermal ore deposits, the southern part of the Apuseni Mountains was labelled "Metalliferous Mountains", being also known as the "Golden Quadrilateral" (Ghiţulescu and Socolescu, 1941). This area covers about 900 km^2 and represents the richest province of Europe in terms of Au−Ag ores. Cook and Ciobanu (2004) estimated a total gold production of the area from ancient times to the present of about 1500−2000 tons, very similar to other major gold provinces around the world (Goldfarb et al., 2001). The Roşia Montană (RM) ore deposit is situated in the northern part of the Metalliferous Mountains (Fig. 1a and b); this area was a very important

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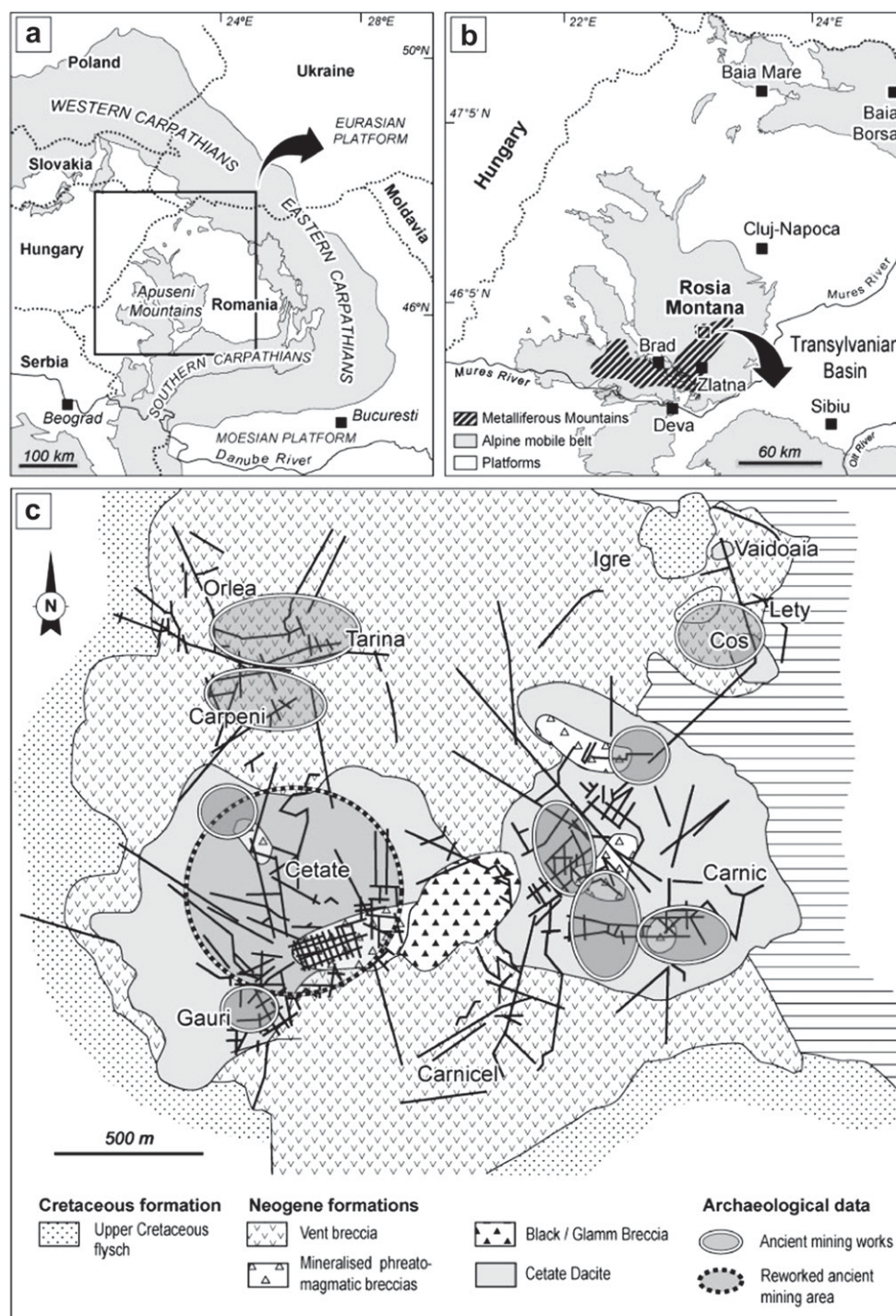 *E-mail address:* sbaron@univ-tlse2.fr (S. Baron).

Fig. 1. Map location of a) Apuseni Mountains in Romania; b) Roşia Montană in the Metalliferous Mountains; c) the Roman works in the different massifs/mining fields from Roşia Montană.

mining centre since antiquity. According to Manske et al. (2006), and taking into account the present reserve calculation (www. gabrielresources.com), RM is Europe's largest Au−Ag ore deposit.

The detailed archaeological study of the RM site began ten years ago and, consequently, new questions emerged, such as how to estimate the gold and silver production of the mine during Roman times, how to follow the absorption of these metals in the trade taking place at the time, or whether these mines were active before the Roman conquest. To answer these questions, we began our research with a study of the Roman mines, and not of the objects, as

the common practice has been up to now. Indeed, several attempts at authentication and metal provenance determination of Dacian gold artefacts have been performed based on comparisons of elementary contents (Hauptmann et al., 1995; Bugoi et al., 2008; Constantinescu et al., 2008, 2009). However, both the limited database of ore sample compositions and the lack of archaeological and geological contexts for the samples render the conclusions of these studies questionable. Lead isotopic signatures are commonly used for metal provenance studies within the field of archaeometallurgy. This approach is based on the comparison of the lead

isotope signatures of ores from known deposits with those of metal objects (e.g. Baron et al., 2009; Klein et al., 2009; Renzi et al., 2009). A prerequisite for this approach is the existence of a database of lead isotope ratios for the ore deposits. Presently, only one example of lead isotopic data is available for the RM deposit (Marcoux et al., 2002), and this is examined from a strictly geological perspective, with no archaeological purpose.

The present research is based on a geological study that consists of detailed mapping and sampling of the ore bodies still present on the face lines, the walls and the roofs of the Roman mines attested by archaeology. The sampling of the ancient mining surfaces was carried out during the study of different ore bodies employed by the Roman exploitation and verified by the archaeological excavations (Fig. 2a). By this interdisciplinary protocol of combined archaeological and geological studies of the ancient mining works, it was possible to recognise different phases of ore deposition, their spatial development, as well as chronological relationships between different ore bodies exploited by the Roman miners. At the same time, a relevant sampling database for the Roman ores was established. Each ore sample from the database is rigorously referenced according to archaeological and geological standards. Lead isotope analyses were performed on each sample using high-
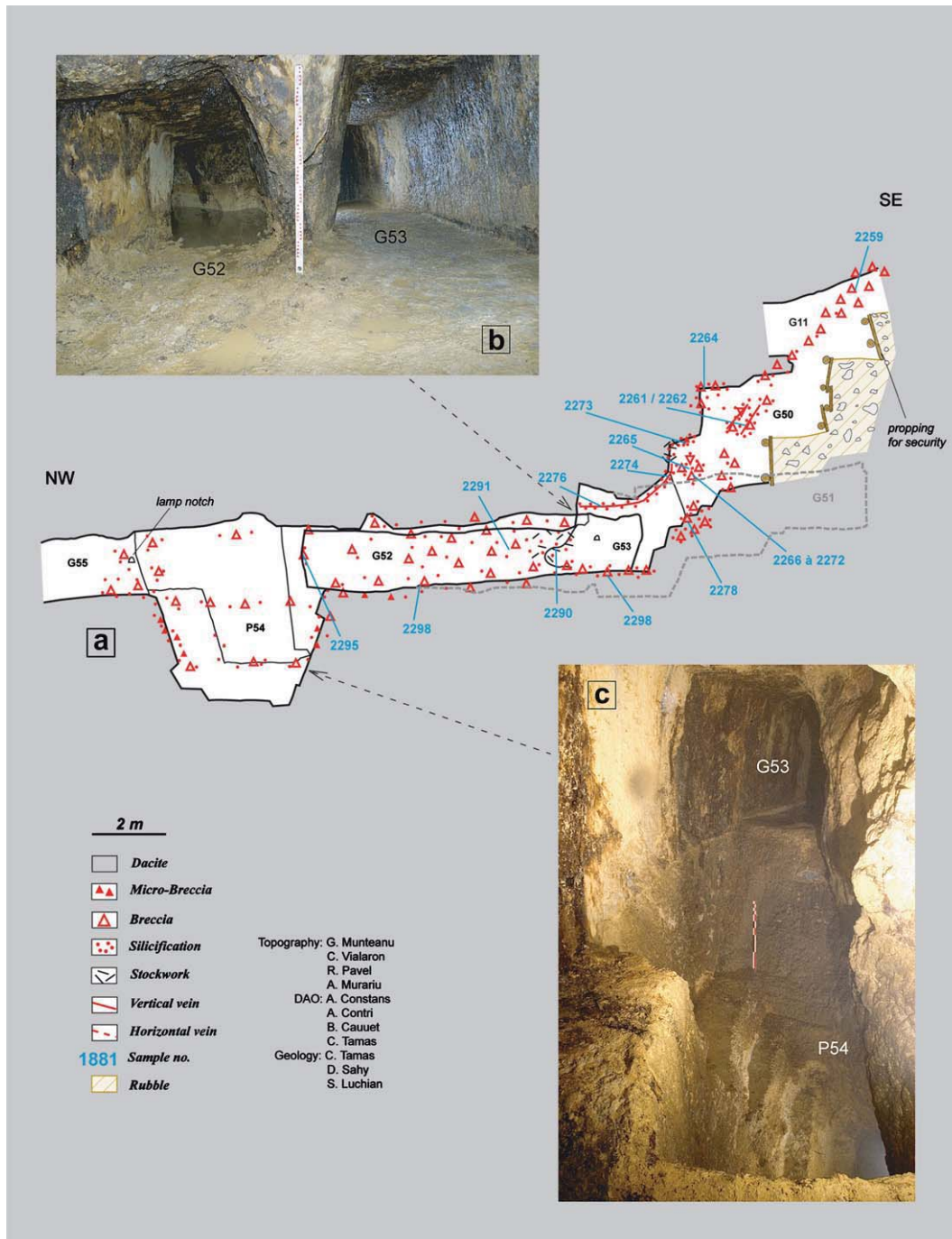
Fig. 2. Cârnic 3 — Lower G11. a) Longitudinal geological cross section of the Roman mining works; b) The pillar between the adits G52 and G53 (© B. Cauuet); c) View of the P54 shaft (© G. Munteanu).

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resolution measurements. The present provenance study uses this database of relevant samples by means of mineralogical and lead isotope analyses.

The aims of this paper are:

1) To provide a relevant signature of the ores mined at RM by the Roman and perhaps Dacian miners on the basis of both geological and archaeological studies.
2) To provide the first data on the origin of a litharge roll discovered in the Roman underground works from RM.

At the same time, this approach provides us with the opportunity to initiate a provenance study addressing the ores, as well as the metallurgical by-products, and to estimate the uses of raw material within this important mining site.

2. Historical, archaeological and geological context of RM mines

2.1. The historical context of RM mines

After the Roman conquest, a group of mining villages flourished in the RM area, identified as *Alburnus Maior* by numerous epigraphic stone inscriptions discovered at the site during modern building or agricultural works. With regards to the Roman underground mining works, archives from the 16th century until today (Slotta et al., 2002) documented that the ancient mines were reworked and partially re-exploited during all these historical periods. During these re-exploitations, numerous accidental discoveries of archaeological artefacts (mining tools and wooden equipment) have been made.

The most remarkable discoveries from RM (among votive altars, funeral monuments/steles, etc.) are the wooden wax tablets with engraved texts, found during the 18th and the 19th centuries in many galleries. These tablets represent different types of contracts (house sales, marriage, employment, etc.) agreed upon by the inhabitants, the miners and the mine contractors (Slotta et al., 2002). Of about 40 tablets discovered, 25 were preserved and studied. The oldest one dates back to the year 131 AD and the most recent one to 167 AD (Mrozek, 1989).

As a result of epigraphic studies, numerous Latin and Greek inscriptions (tablets and stones) revealed the presence of an Illyrian population, who, after the Roman conquest, came to work in the mines as free workers in great numbers. These first migrants had to live among the native Dacian population, probably quickly assimilated and unmentioned in the epigraphic data available, with the exception of the very same name of *Alburnus Maior*, *Alburnus* being considered to be of Dacian origin (Wollmann, 1989).

Named Getes by the Greeks and Dacians by the Romans, this population was first mentioned by Herodotus (Larcher, 2005, *Herodotus, IV, 93*) in the context of Darius' expedition against the Scythians at the end of the 6th century BC. The Decebalus' Dacian kingdom ended with the Roman conquest by Trajanus in 106 AD. The Roman administration was withdrawn from the province of Dacia by Aurelian in 271 AD. From political, economic and technological—historical points of view, it would be very important to establish also if the primary deposits (veins, breccias) from the Apuseni Mountains (RM, Bucium, Brad, etc.) were already known and exploited by the Dacians before the Roman conquest. It is possible that the Romans promoted the wars against Dacia in order to take control of the gold and silver mines already in activity.

Several hoards with silver spiral bracelets from the Dacian period (2nd and 1st centuries BC) were discovered, such as those from Coada Malului (Prahova county) or Senereuş (Hunedoara county) (Sîrbu and Florea, 2000). Recently, Dacian gold spiral bracelets have been found by clandestine treasure hunters using metal detectors near Sarmizegetusa Regia, the capital of the Dacian kingdom (Constantinescu et al., 2008, 2009). Furthermore, the Dacians minted a great number of copies of Roman silver *denarius* coins (Preda, 1973). The question thus arises of the contribution of the precious metals recovered from primary ore deposits within Apuseni Mountains in the production of Dacian gold and silver objects. Moreover, the abundance of Dacian silver objects is also worth mentioning and one should keep in mind that the primary silver metallurgy was much more elaborated than that of gold. Were the Dacians able to produce silver from primary ores or did they simply re-cast silver objects traded from abroad (ingots, coins, jewellery, etc.)?

A certain contemptuous assertion is linked to the idea that the Barbarians could only recover gold from placers, because their mining technology was too primitive to allow them to produce gold from primary ore deposits (veins). It is already proved that at least at Limousin, Central France, the Celts obtained most of their gold from primary deposits starting from the 6th/5th century BC, as well as during the Second Iron Age (Cauuet, 2004b). Furthermore, the Celts were contemporaneous with the Geto-Dacians from Transylvania since the 4th century BC (Sîrbu, 2006). One issue of particular importance presented by the study of the ancient mines from RM is the possibility of identifying pre-Roman mining activity at the site. In addition to archaeological data from attested Dacian mines, another way to elucidate this question is to characterise, by isotopic analyses, the precious metals from the mines with assumed Dacian activity, and to compare them with Dacian objects.

2.2. The archaeological study of RM gold—silver mines

The significant mining potential of the RM area is attested by the density of the ancient to recent mining works revealed during the last decade by a large scale archaeological research program. Nine archaeological campaigns were conducted at the site in order to study the mining networks located in seven massifs which surround the actual RM village: Cârnic, Cârnicel, Cetate, Coş, Orlea, Ţarina, Carpeni (Fig. 1c) (Cauuet et al., 2003; Cauuet, 2005, 2008).

The study of the different underground networks revealed an extensive Roman mining activity during the 2nd and 3rd centuries AD. In the south-western part of the Cârnic massif, more than 4 km of linear Roman mining works were discovered. Considered in their entirety, these ancient works have a 98 m vertical development and cover a surface of 13,600 m^2. According to the present state of the research, the different ancient mining fields studied at RM include galleries, downward sloping adits, narrow vertical works, inclined or staged stopes (Fig. 2c), chambers with pillars, and some helicoidal shafts. The variety of underground works is in agreement with the context of the ore deposit (Cauuet, 2004a). The Roman works are usually of very good quality and they systematically present trapezoidal sections, which allow their rapid identification (Fig. 2b). Tool marks on the walls of the mining works are also well preserved. The presence of lamp notches in the walls also provides evidence of the use of lighting equipments during the Roman period, because oil lamps arrived at *Alburnus Maior* only at the beginning of the 2nd century AD, with the arrival of the Romans (Cauuet, 2005).

The current research carried out in the northern part of the site (Carpeni, Orlea and Ţarina massifs) revealed, in the Păru-Carpeni area, a group of four interconnected rooms dug one on top of the other. These underground rooms, situated at about 30 m under the surface, are equipped with drainage machineries composed of a range of lifting wooden wheels, dated to the 2nd century AD by the ^{14}C method (Cauuet, 2008). Almost all these vestiges were dated to Roman times. Several unexpected radiocarbon ages around 2100 ± 50 BP (Cauuet et al., 2003, and unpublished data)

obtained on wooden fragments and charcoals discovered in some mining networks from the Cârnic massif suggest that the mine was in activity during the Dacian period as well. To date, no ceramic or other type of artefacts belonging to the Dacian Culture were discovered, either in underground mining work, or in the surface excavations of dwellings or necropolises. For the moment, the Dacian mining activity remains more of an assumption than a demonstrated fact. However, the scale of the mines at *Alburnus Maior* and the incredible speed of their progression at the beginning of the 2nd century AD both suggest that the deposit was well known and partly in exploitation before the Roman invasion. If archaeology must bring more accurate evidence of a pre-Roman mining activity, another way to indirectly demonstrate this hypothesis is to verify it by geochemical studies.

2.2.1. The treatment of the gold−silver ores at RM

Only crushing and grinding workshops have been discovered so far. The metallurgical workshops where the separation of gold and silver from the gangue and other metals took place are still not located. In any case, during the underground archaeological excavation, a litharge roll has been discovered in a Roman underground network from the Cârnic massif (Fig. 1c). It was found within a secondary backfilling deposit of an inclined adit situated very close to the surface. The filling material of the ancient work represents flooding events with deposition of interlayer sand and clay sequences originating from the surface. The litharge roll (Fig. 5) was located very close to the floor of the inclined plane and it might have been carried away by a mud flow arriving in the underground from the surface. Litharge is initially composed of lead oxide (PbO), which can be easily transformed by weathering, depending on burial conditions. This artefact presents all the characteristics of the litharge rolls found in great quantities within the sites of lead argentiferous metallurgy of the Roman period located in the south of the Iberian Peninsula, at the La Loba mine (Domergue, 1990; Blazquez Martinez et al., 2002), and also in Greece, for example in the Laurion mining district (Conophagos, 1980).

2.2.2. The treatment of gold−silver ores, i.e. the "chaîne opératoire"

The *chaîne opératoire* of gold production in antiquity is only partially known, and the available information is essentially based on the ancient texts of Pliny the Elder (Zehnacker, 1999, *Gaius Plinius Secundus, 33, 77*) and Diodorus (Bommelaer, 1989, *Diodorus Siculus, 3, 14*). In addition, only few examples of archaeological evidence concerning the various steps of the gold−silver metallurgy are reported in the literature for prehistoric and ancient periods (Piccottini, 1994; Bachmann, 1999; Cauuet and Tollon, 1999; Ramage and Craddock, 2000; Domergue, 2008). In a brief summary of this process, after mining, the gold−silver ore is crushed and then roasted in order to oxidise the sulphides and better disintegrate the material. The roasted gold−silver ore is ground and then concentrated (probably by panning). The Au−Ag concentrate is charged in crucibles with specific additives, like lead metal or lead oxide, to facilitate the smelting process and to extract from the ore the two noble metals, i.e. Au and Ag; these will pass into the lead metal. Then, the lead−gold−silver metal is poured into moulds and subjected to cupellation to separate the noble metals from the alloy by oxidising the lead. The last step involved the separation or parting of gold and silver by the cementation process, using reagents such as different salts including sodium chloride, antimony sulphides, nitrates, etc.

From the above-mentioned gold *chaîne opératoire*, the litharge roll found inside a Roman gallery from RM suggests that it might have been used as an additive for gold−silver extraction. This archaeological evidence may suggest that some metallurgical processes could have been developed at RM in the Roman period:

(i) cupellation process for Ag separation (process from a possible lead *chaîne opératoire*), or (ii) the use of lead as additive in the charge at the beginning of the *chaîne opératoire* of gold, i.e. as additive to the gold−silver ores for the reduction smelting step, to collect the noble metal.

Only further excavations will allow confirming one or both hypotheses. Nevertheless, the present isotopic study will contribute to the understanding of the origin of this lead: local production at the site or import.

2.3. The geological context of the RM ore deposit

2.3.1. Geological setting

RM is situated in the Apuseni Mountains, an Au−Ag province located in the heart of the Romanian Carpathian Mountains (Fig. 1a). The second Romanian gold province is situated north of this one, in the Maramureş region, close to the border with the Ukraine. Within this northern Au−Ag province, there are two main mining districts, Baia Mare and Baia Borşa (Fig. 1b).

Three main ore deposit districts are known in the Metalliferous Mountains, Brad−Săcărâmb, Zlatna-Stănija, and Roşia Montană−Bucium. The ore deposits from these districts consist of porphyry copper and Au−Ag epithermal deposits, all related to Neogene volcanism/magmatism (Ghiţulescu and Socolescu, 1941; Ianovici et al., 1976; Boştinescu, 1984; Ciobanu et al., 2004a).

The RM ore deposit (Fig. 1c) is hosted by a Neogene maar−diatreme complex (Leary et al., 2004; Tămaş, 2007) that pierced a Cretaceous flysch. The unexposed crystalline basement occurs as fragments in breccia pipe structures, which are well developed at the ore deposit scale. The Neogene volcanic activity in the RM area consists of two major events, confirmed by means of K/Ar datings (Pécskay et al., 1995; Roşu et al., 1997, 2004): emplacement of Cetate dacite at 13.5 ± 1.1 Ma (two mushroom-like domes − Cetate and Cârnic, and related volcanoclastics), and Rotunda andesites at 9.3 ± 0.47 Ma (rooted body, lava flows, and related volcanoclastics). $^{40}Ar/^{39}Ar$ datings of ore-related adularia from several veins in the Cetate massif (Manske et al., 2004) indicated an age of about 12.7 Ma (12.78 ± 0.09 Ma and 12.71 ± 0.13 Ma) for the mineralisation.

2.3.2. The Roşia Montană mineralisation

RM is an epithermal deposit that shows a transition from a low to an intermediate sulphidation state (Mârza et al., 1997; Leary et al., 2004; Manske et al., 2006; Tămaş et al., 2006; Wallier et al., 2006; Tămaş, 2007). Various types of ore bodies are known: veins, breccia structures (breccia pipes and breccia dykes), stockworks, and impregnations. From the first microscopic study carried out by Petrulian (1934) until recent contributions (Tămaş, 2002; Ciobanu et al., 2004b; Tămaş et al., 2004, 2006; Bailly et al., 2005), more and more Au−Ag mineral assemblages and minerals have been reported. In addition to electrum and free gold, common sulphides (pyrite, chalcopyrite, sphalerite, galena, marcasite, arsenopyrite, alabandite, tetrahedrite, etc.) and Ag−minerals (argentite, proustite, pearceite, polybasite, etc.) also occur. Tellurides (hessite, sylvanite, petzite, altaite), as well as an endemic Te-bearing argyrodite, have recently been identified at RM (Ciobanu et al., 2004b; Tămaş et al., 2004, 2006). With the exception of pyrite, the other sulphides occur in minor amounts within the RM deposit. The tellurides have been observed only in two veins with rhodochrosite−rhodonite gangue. These vein structures are dominated by tetrahedrite, while sphalerite, galena, chalcopyrite and tellurides are subordinate.

3. Materials and methods

The geological investigation of the entire ore deposit indicated several mineralising events which have spanned a period of about

500,000 years (Manske et al., 2006). At the geological scale, this represents a short period of time, which cannot generate significant isotopic variation related to radioactive decay (Faure, 1986). However, the events may have some different lead isotopic composition, due either to various ore sources, or to fluid contamination by interaction with different wallrocks during circulation. In order to carry out a representative lead isotopic investigation and obtain a significant isotopic signature, 29 ore samples were collected from different mining fields of the RM deposit − Cârnic, Cetate, Țarina and Carpeni − related to the Roman mining works under consideration (Fig. 1c).

3.1. Sampling of chrono- and geo-referenced materials

Archaeological excavations were performed intensively in the Cârnic, Cetate, Țarina and Carpeni massifs. To provide an accurate signature for future metal provenance studies, a chrono- and geo-referenced ore sampling pertaining to the Roman exploitation was performed in the Cârnic massif. Along 4 km of Roman works, as well as in the context of modern and recent works, the geological study allowed the identification of four mineralisation events according to the crosscutting relationships among the ore bodies and their mineralogical peculiarities (Tămaş et al., 2006):

#1 − early phreatic brecciation with high grade gold, low grade silver (electrum, pyrite, polybasite, tetrahedrite, galena, chalcopyrite, sphalerite) breccia dykes with quartz−adularia gangue (average grades of 30−140 g/t Au and 20−70 g/t Ag);
#2 − gold−silver (electrum, polybasite, pyrite, chalcopyrite, sphalerite, tetrahedrite, marcasite, covellite) rich veins (up to 120 g/t Au and 150 g/t Ag) with quartz−adularia gangue;
#3 − rebrecciated breccia structures with high grade silver (acanthite, stephanite, polybasite−pearceite, native silver, tetrahedrite, galena, pyrite, chalcopyrite, sphalerite, bornite), and medium grade gold (electrum), with up to 220 g/t Ag and 9 g/t Au. This type of ore body also contains Ge-bearing minerals (argyrodite) and galena with Te traces. The gangue of these breccia bodies is made of quartz−adularia, together with a black hydrothermal cement (referred to as chinga by the local miners) rich in carbon.
#4 − extremely rich silver with some gold grade veins (tetrahedrite, sphalerite, galena, pyrite, hessite, altaite, sylvanite, Te-argyrodite, electrum, marcasite), with up to 1150 g/t Ag and 5 g/t Au. The gangue is dominated by rhodonite−rhodochrosite, but minor quartz also occurs.

From 29 samples used for isotopic analyses, only 21 are from the Cârnic massif. They reflect the ore deposition events as follows: ten samples were collected from the mineralisation event #1, only one sample from the mineralisation event #2, seven samples from the mineralisation event #3, and three samples from the mineralisation event #4.

According to the archaeological studies, it is clear that the Romans mined the mineralisation events #1, #2 and #3, but not #4. For this reason, the ores are clearly chrono- and geo-referenced. Phase #4 samples, as well as eight samples from other ore fields at RM (Cetate, Țarina, Carpeni) have also been analysed in order to obtain a general coverage of the Pb isotope signatures of the entire RM deposit.

3.2. Sub-sampling for lead isotopic studies

The ore samples do not contain lead minerals. If available, galena was separated by hand-picking. If not, small pieces of gold−silver ore were crushed, hand-picked, and powdered. The litharge roll was sampled using a diamond mini-drill to collect few milligrams of powder.

3.3. Samples dissolution

The litharge roll and the galena samples were prepared according to the following protocol: dissolution in a Teflon® vessel using 0.5 mL of concentrated HNO_3 (bi-distilled) and set at 100 °C overnight.

For the rest of the ore samples, few milligrams of gold−silver ore powder (according to the lead concentration in each sample) were dissolved in a Teflon® vessel using 0.3 mL of concentrated HNO_3 (bi-distilled) and 0.3 mL of concentrated HF (Merck Suprapur quality) and were set at 100 °C overnight. The samples were evaporated at 60 °C. For ore samples containing some chinga cement (C-rich), the residues were taken back with 0.05 mL of 30% H_2O_2 (Merck Suprapur quality) and 0.5 mL of concentrated HNO_3 in order to volatilise the carbon and were set at room temperature overnight. This procedure was repeated several times.

For all samples, after the last evaporation (60 °C), the residues were taken up in a 1 mL of 0.9 M HBr (bi-distilled) and left at room temperature overnight in order to homogenise the solution with the residue. Pb was separated from the other elements by ion exchange, using the AG1X8 resin (Strelow and Walt, 1981). After separation, the solution was evaporated at 60 °C, and the residues were taken back in 1−5 mL of 0.3 N HNO_3 (Merck Suprapur quality).

In order to determine the total external uncertainties, three samples have been triplicated (analytical error margins located in isotopic diagrams).

3.4. Lead isotopes measurements

The lead isotopic composition was measured with a MC-ICP-MS (Neptune, VG Instruments, at the LMTG laboratory, Toulouse, France), following the procedure reported by Baron et al. (2009). Repeated measurements of the NIST NBS 981 Pb reference material yielded a reproducibility (2× standard deviations) better than 125 ppm for all the reported Pb isotope ratios (Table 1). The repeated measurements on the three triplicates of three samples allowed estimating a total external uncertainty better than 150 ppm (2× standard deviations) for each reported Pb isotope ratio (Table 2).

4. Results and discussion

All lead isotopic data concerning gold−silver ores, galena, and the litharge roll from RM are reported in Table 2.

4.1. Lead isotopic signatures of the entire RM ore deposit

The lead isotopic compositions of all gold−silver ores and the few galena-rich samples from the different mining fields studied at RM are presented in Table 2. The ranges of lead isotopic values of the entire RM deposit are 2.07569−2.08365 for $^{208}Pb/^{206}Pb$ ratio, 1.18769−1.19197 for $^{206}Pb/^{207}Pb$ ratio, 38.709−38.821 for $^{208}Pb/^{204}Pb$ ratio, 15.6558−15.6695 for $^{207}Pb/^{204}Pb$ ratio, and 18.6002−18.6770 for $^{206}Pb/^{204}Pb$ ratio. The data obtained by Marcoux et al. (2002) on one ore sample from RM fall within these ranges. The different ore events identified in the Cârnic massif and in the other mining fields of RM are distinguishable by their different lead isotopic compositions, due to the fact that high-resolution measurements were employed (Fig. 3). Thus, the high precision of the data will enable a relevant provenance study at the scale of the entire RM area.

However, at a regional scale, by comparison with other data available for Romanian ore districts (Fig. 4), the lead isotope

Table 1
Lead isotopic compositions of the NIST SRM 981 Pb reference material during different analytical sessions.

NIST SRM 981 Pb	^{208}Pb/^{206}Pb	^{207}Pb/^{206}Pb	^{208}Pb/^{204}Pb	^{207}Pb/^{204}Pb	^{206}Pb/^{204}Pb
SRM 981 Pb[a]	2.16768 ± 23	1.09310 ± 07	36.722 ± 08	15.4980 ± 25	16.9408 ± 21
July 2008	2.16750	1.09310	36.720	15.4985	16.9414
2× sd ($n = 15$)[b]	0.00007	0.00002	0.002	0.0007	0.0010
RSD ppm	33	24	50	46	62
November 2008	2.16749	1.09310	36.720	15.4985	16.9413
2× sd ($n = 87$)[b]	0.00020	0.00007	0.002	0.0008	0.0018
RSD ppm	93	68	61	50	103
February 2009	2.16755	1.09303	36.716	15.4971	16.9388
2× sd ($n = 36$)[b]	0.00016	0.00004	0.005	0.0015	0.0009
RSD ppm	72	45	123	96	56
November 2009	2.16746	1.09309	36.720	15.4985	16.9413
2× sd ($n = 23$)[b]	0.00003	0.00001	0.001	0.0005	0.0005
RSD ppm	15	11	36	30	30

[a] Referenced value by Double-spike TIMS (Thirlwall, 2002).
[b] 2× sd from the mean.

Table 2
Lead isotopic compositions of the ores (gold–silver ores and galena) from different mining fields at Roşia Montană.

Ores sample id.	Host	^{208}/^{206}Pb	^{206}/^{207}Pb	^{208}/^{204}Pb	^{207}/^{204}Pb	^{206}/^{204}Pb
Ores from Carnic massif/mining field						
Ore deposition phase #1						
2018 A − 2	Dark *chinga*	2.07865	1.19098	38.780	15.6646	18.6562
1908 − 2	Dark *chinga*	2.07847	1.19099	38.778	15.6649	18.6567
1908 − 4	Dark *chinga*	2.07864	1.19104	38.781	15.6643	18.6569
2207	Dark *chinga*	2.07672	1.19057	38.709	15.6558	18.6396
2014	Dark grey *chinga*	2.07656	1.19165	38.755	15.6619	18.6633
2050	Pyrite & chalcopyrite	2.07624	1.19187	38.753	15.6601	18.6650
2018 A − 1	Dark *chinga*	2.07827	1.19114	38.776	15.6640	18.6580
2018 B	Dark *chinga*	2.07770	1.19146	38.774	15.6631	18.6620
2054	Dark *chinga*	2.07644	1.19131	38.728	15.6560	18.6511
2050	Pyrite & chalcopyrite	2.07638	1.19183	38.758	15.6619	18.6661
Ore deposition phase #2						
2210	Quartz	2.08007	1.19047	38.795	15.6666	18.6506
Ore deposition phase #3						
1486	Dark *chinga*	2.07569	1.19197	38.741	15.6583	18.6643
2124 A	Dark *chinga*	2.07819	1.19107	38.776	15.6654	18.6585
2124 B	Quartz & Pyrite	2.07855	1.19070	38.765	15.6630	18.6499
1572	Quartz	2.08160	1.18949	38.797	15.6688	18.6380
2170 Fe	Iron-rich phase	2.07686	1.19135	38.746	15.6597	18.6562
2170	Quartz	2.07854	1.19099	38.775	15.6636	18.6550
2170	Quartz & Iron/Quartz	2.07811	1.19112	38.769	15.6628	18.6561
Ore deposition phase #4						
3198 − 1	Sequence 1 (gangue)	2.08157	1.18959	38.792	15.6659	18.6360
3198 − 2	Pyrite sequence 2	2.08201	1.18924	38.795	15.6684	18.6335
3198 − 3	Au−Ag sequence	2.08174	1.18933	38.791	15.6675	18.6338
Ores from other massifs/mining fields						
Cetate Massif						
129	Galena	2.08365	1.18769	38.756	15.6608	18.6002
1328	Galena	2.08350	1.18776	38.756	15.6611	18.6015
106	Galena	2.08351	1.18779	38.760	15.6620	18.6032
154 − 5	Sulphide rich quartz amethyst	2.08187	1.18893	38.764	15.6609	18.6198
Tarina Massif						
2456	Pyrite	2.07994	1.19075	38.806	15.6685	18.6573
2581 Bis	Quartz	2.07962	1.19091	38.805	15.6681	18.6594
2500	Galena	2.07873	1.19174	38.815	15.6683	18.6725
Carpeni Massif						
2953	Pyrite + Quartz	2.07856	1.19193	38.821	15.6695	18.6770
Litharge roll						
Lith RM	−	2.08587	1.19206	38.598	15.6434	18.6478
Total external uncertainties[a]		0.00020	0.00013	0.006	0.0016	0.0034

[a] Twice the standard deviation from the mean of tree triplicates. These error bars are reported on Pb/Pb diagrams.
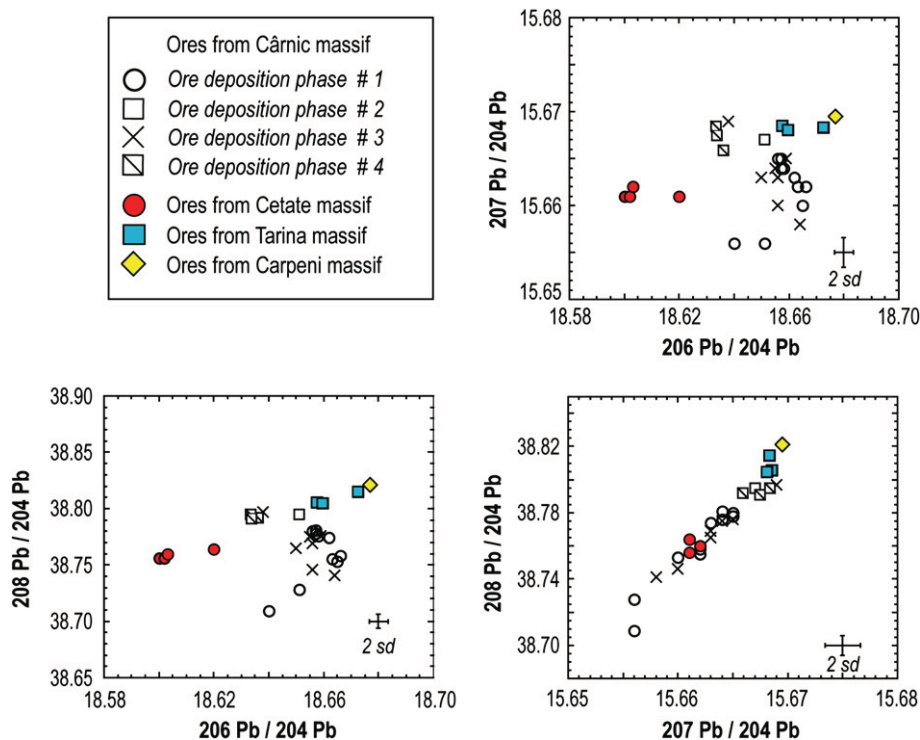
Fig. 3. Pb/Pb diagrams of the ores from different mining fields/massifs from Roşia Montană.

compositions of gold−silver ores and galena from RM are homogeneous, indicating the same source of fluids for the epithermal mineralisations throughout the RM deposit. These results are consistent with previous studies carried out on fluid inclusions presenting a constant $\delta^{18}O$ value ($\delta^{18}O$ of 4.5−5.0 per mil) and indicating a common magmatic component (Wallier et al., 2006). The $^{207}Pb/^{204}Pb$ vs $^{206}Pb/^{204}Pb$ diagram (Fig. 4) shows that the lead isotope compositions measured for RM in this study are slightly more radiogenic than the values reported for the Apuseni Mountains as a whole (Marcoux et al., 2002), but overlapped with a small part of them (Fig. 4).

In comparison with the Baia Borşa and Baia Mare epithermal mineralisations of north-western Romania (Maramureş county), the Apuseni Mountains group shows less radiogenic values (Cook and Chiaradia, 1997) (Fig. 4). Concerning the isotopic composition of the Baia Borşa mining district, two very distinct groups are discernible. These two signatures are clearly related to both different sources and ages of the mineralisation (Cook and

Chiaradia, 1997). This example reemphasises the importance of a geological study, with the identification of the mineralisation events allowing a relevant sampling, which in turn enables an exhaustive isotopic characterisation of a given mining district. This approach, coupled with archaeological evidence for ancient mining, is a prerequisite for any metal provenance study.

4.2. Lead isotopic signature of the gold−silver ores mined by the Romans

The archaeological and geological studies certified that the Romans mined the ore deposition events #1, #2 and #3, but they did not exploit the #4 one. The lead isotopic compositions of the ore deposition events #1, #2 and #3 respectively display ranges of values of 18.6396−18.6580, 18.6506 and 18.6380−18.6643 for the $^{206}Pb/^{204}Pb$ ratio. The ranges of values for the three events are not significantly different from one another, as indicated by their overlapping. Hence, as these values are homogeneous, we can
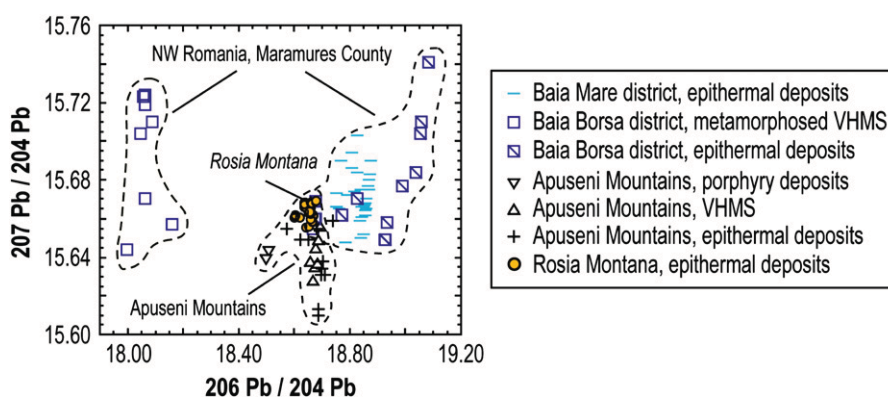

Fig. 4. $^{207}Pb/^{204}Pb$ vs $^{206}Pb/^{204}Pb$ diagram of Roşia Montană compared with available lead isotope data from Romania (Cook and Chiaradia, 1997; Marcoux et al., 2002).

provide a mean signature for the three events of the gold–silver ores mined by the Roman miners: 2.07787 ± 0.00148 (1σ) for the ^{208}Pb/^{206}Pb ratio, 1.19111 ± 0.00059 (1σ) for the ^{206}Pb/^{207}Pb ratio, 38.764 ± 0.023 (1σ) for the ^{208}Pb/^{204}Pb ratio, 15.6625 ± 0.0035 (1σ) for ^{207}Pb/^{204}Pb ratio, and 18.6558 ± 0.0078 (1σ) for the ^{206}Pb/^{204}Pb ratio.

The Baia Mare and Baia Borşa mining districts were also assumed to have been exploited during ancient times, but there is no archaeological excavation to confirm this hypothesis yet. In any case, the results presented in this study will allow improving the discrimination of the provenance of ancient metal in Romania.

4.3. Origin of the litharge roll found in the Roman mining network

The lead isotopic compositions of the litharge are 18.6478 ± 0.0034 (2σ) for the ^{206}Pb/^{204}Pb ratio, 15.6434 ± 0.0016 (2σ) for the ^{207}Pb/^{204}Pb ratio, and 38.589 ± 0.006 (2σ) for the ^{208}Pb/^{204}Pb ratio. In order to identify the source of this litharge, we need to compare its isotopic signature to Pb-rich ores from different mining fields at RM, and especially with the phase #3 ores rich in Ag-bearing galena. Unexpectedly, the litharge displays significantly different lead isotopic ratios as compared to the data available for the entire RM ore deposit (Fig. 5). This result suggests that the litharge lead source is allochthonous with respect to the RM deposit. Furthermore, the scarcity of galena in the RM ore deposit and particularly in the ores mined by the Romans (0.3wt%, $n = 30$, unpublished data) confirms the Pb isotope analyses of the litharge. The lead content present in the gold–silver ores is not sufficient to allow the extraction of the noble metals (Th. Rehren, personal communication).

Consequently, cupellation processes seem to have not been carried out at RM, and it is now clear that the litharge roll found in the Cârnic massif was brought from another area. These facts point towards the hypothesis that the litharge, with an unknown provenance for the moment, but certainly not from RM, served during the smelting of gold–silver ores at the site. Several deposits with significant galena occurrences as compared with RM are located in the Apuseni Mountains, south of RM (Fig. 1b). Some lead isotopic data reported by Marcoux et al. (2002) on these deposits are very close to the signature of the litharge, suggesting that the lead composing the litharge roll might originate from these Apuseni Mountains deposits. However, studying these mining sites appears necessary; even if numerous ancient mining works are known in the Apuseni Mountains, they have been insufficiently studied to date (Cauuet, unpublished data). Taking into account that the litharge originated from a mine located in the southern part of the Metalliferous Mountains, this aspect suggests that a wider mining management was carried out on a regional scale. This is in agreement with the historical documented setup of *Auraria Dacicae*, a territory comprising the mines from the entire range of the Apuseni Mountains. This territory was the Emperor's property and was ruled by a *procurator aurariarum*, seated at *Ampelum* (today Zlatna, Fig. 1b), approximately 35 km south-east of RM (Sîntimbrean, 1989).

The further challenge consists of identifying metallurgical wastes (slags) at RM in order to i) understand the metallurgical process that took place at this important mining site; ii) estimate the isotopic contamination due to the addition of lead for the smelting process of gold–silver ores, which needs to be taken into account in future metal provenance studies. The development of our research at the scale of the Apuseni Mountains will allow us to confirm if the source of the lead from the litharge is located in this area or abroad.

5. Conclusions

The gold–silver mining vestiges of Roşia Montană, dating back to Roman Dacia (106–271 AD), have been studied by means of archaeology, geology and geochemistry. The archaeological research allowed the accurate dating of the mining works and facilitated the identification of the ores exploited by the Roman miners by joint geological research. Four ore deposition events were identified in the Cârnic massif by geological studies that also
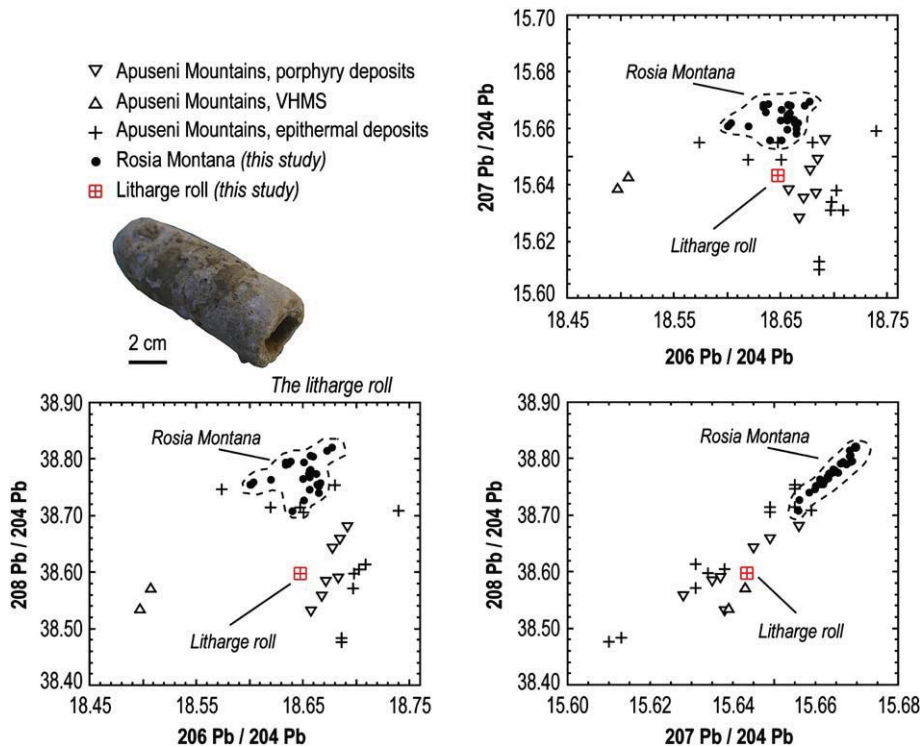

Fig. 5. Pb/Pb diagrams of Roşia Montană, the litharge and data from other ore deposits in the Apuseni Mountains (Marcoux et al., 2002).

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allowed a detailed mineralogical characterisation of the exploited ores. The Romans mined the ores corresponding to the depositional events #1, #2 and #3, but they did not exploit the ores from the #4 one. The geochemical analyses carried out in the context of the present study revealed the lead isotopic compositions of the gold–silver ores mined by the Romans in the Cârnic massif, as well as in other mining fields (Cetate, Țarina, Carpeni). The signature of the entire RM mining area is distinct from the other deposits in the Apuseni Mountains, allowing the establishment of a more accurate database for further metal provenance studies on a regional scale. As there are reasons to suspect that the RM deposit has been exploited by the Dacian population, these lead isotopic data could also be compared with Dacian gold–silver products and/or objects.

The lead isotopic composition of the litharge roll discovered at RM is significantly different as compared to the compositions of Au–Ag ores from the entire RM ore deposit. It suggests that i) some lead was used in the gold *chaîne opératoire* in order to extract Au–Ag metals from the ores; ii) other lead ore sources, possibly situated in the Apuseni Mountains, contributed to the metallurgical processes developed at the Roşia Montană site during Roman times.

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