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The origin of lead artifacts from Novae: applications of Pb isotopes in identifying the provenance of Roman artifacts from N. Bulgaria

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Abstract

This study identifies the lead ores used to produce lead artifacts used by the Romans between the first and eighth centuries AD during the construction of the fort and then the town of Novae (N. Bulgaria). For this purpose, lead samples were taken from pipes, joints of columns and pedestals, and from a lead ingot. The samples were analyzed for lead isotopes and the results were compared to literature data for Roman mines from what is now Bulgaria, Greece, North Macedonia, Montenegro, Bosnia and Herzegovina, Italy, Germany, and Romania. Pb isotope results indicate that during the earlier stages of Novae's establishment, lead was most likely supplied from several different mines located in the Balkan area. Several samples also show Pb isotopes indicating mixing of lead from mines in the Balkan area. Then in the fourth—fifth century AD lead began to be supplied mainly from mines located in NW Bulgaria, with one sample possibly from deposits in German. This is evidenced by the matching of the results obtained for the ores to the data for deposits from these regions. Two possibly recycled samples were also identified. Deposits from other European regions did not match samples from Novae, indicating that majority of the lead was sourced from mines in the Balkan region.

Keywords Lead isotopes, Novae, Lead origin, Lead provenance, Roman artifact, Artifacts origin, N. Bulgaria

Introduction

One of the strategic factors affecting the economic development of a state is the availability of raw materials. The Roman Empire's economic growth was fueled by wide access to various raw materials located within its vast territory. Overall the origin of raw materials for the production of objects such as sewage pipes is less investigated than materials needed to make ceramics [1, 2], mirrors [3], or coins [4–6]. This study investigates the origin of the ores used in the production of lead in the structural and architectural elements at the Novae site (N. Bulgaria).

Novae was one of the most important military and civil centers of the Roman Empire on the Danube (Moesia Inferior province) between the first and sixth centuries AD. [7–12]. It was an important point for controlling

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traffic on the Danube and patrolling the Danube Plain. Despite the archaeological research conducted at this site since the 1960s, many questions related to the life and functioning of the fortress and then the city remain to be investigated. These include the origin of lead which was widely used by the Romans for the production of pipes (water supply, sewage), as well as a binder in columns. Lead isotope analysis is a standard, long-established method for determining the provenance of archaeological metals, including lead [13–21].

Determining the source of the ore in the Balkans can be problematic. This is due in part to the large number of polymetallic deposits present in the region from N Romania to S Greece. Some of them have been exploited since ancient times, including the Roman Period, to modern day [22–30]. However, Pb isotope analyses have been conducted on only a small number of these deposits. The largest Pb isotopic databases are published by the OXALID laboratory and in the GlobaLID [16, 20, 21, 31–47].

Another factor is recycling of lead used by the Romans [14, 48–51]. This fact makes it extremely difficult or even impossible to determine the provenance of the studied artifacts. To check whether recycled lead was used to make given artifacts, a chemical analysis should be performed to identify variation in the content of other metals, especially tin [48–51]. The increased Sn content may indicate the use of recycled lead [48–51].

In this work we attempted to determine the region from which the ore used to produce the studied Novae archaeological lead artifacts was most likely derived. For this purpose, lead isotope analysis of lead samples from the Novae site (Fig. 1) and lead ore samples from polymetallic mines in Madan (Bulgaria), Trepča (Kosovo), and Rudnik Mine (Serbia) have been conducted, and compared to published data obtained for ores from Bulgaria, Greece, North Macedonia, Montenegro, Bosnia and Herzegovina, Serbia (including, Kosovo) Italy, Germany, and Romania.

The Novae archaeological site

The first camp installations of wood and earth were erected by the August VIII Legion in 45 AD, while part of the province of Moesia. After the death of Nero (54–98)

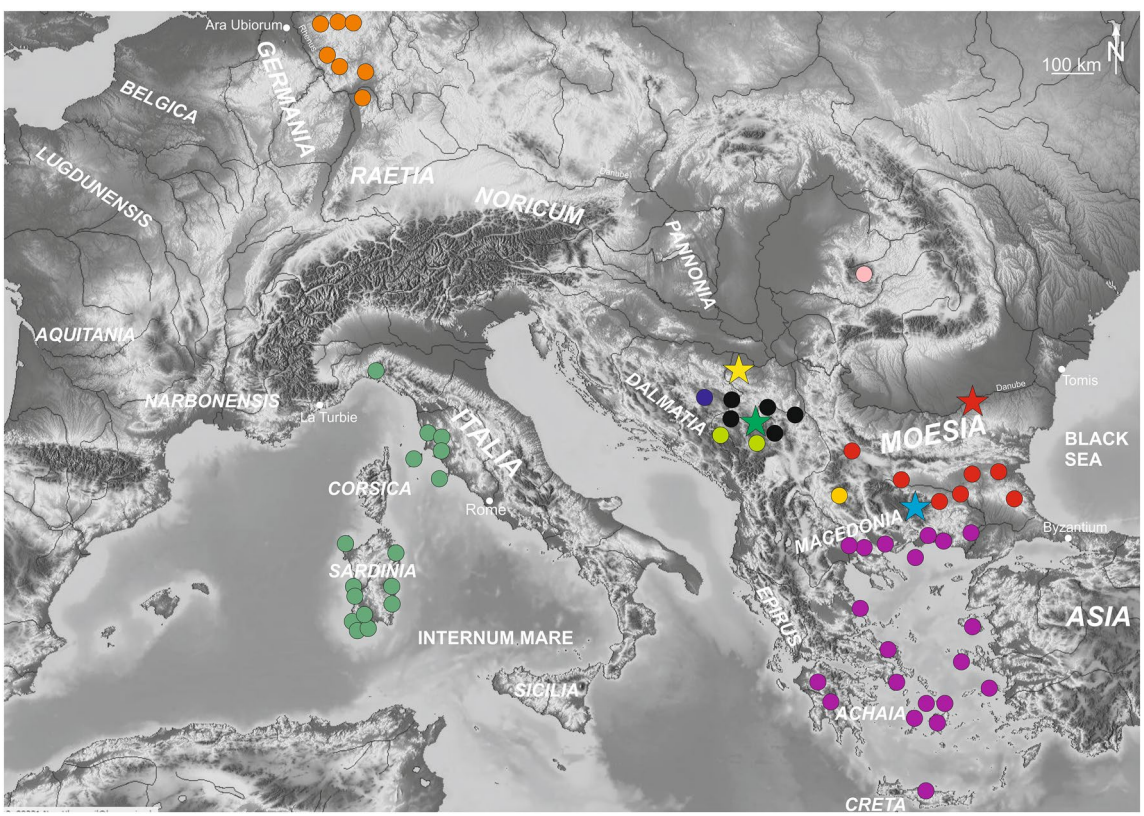

Fig. 1 Location of studied sites. Research sites of Roman colonies are marked by red star—Novae in Svistov (N. Bulgaria). New data for deposits: blue star—Madan (S Bulgaria); green star—Trepča (Kosovo); yellow star—Rudnik (Serbia). Ore deposits used for comparison are marked by circles: red circle—Bulgaria; purple circle—Greece; black circle—Serbia (including Kosovo); light green circle—Montenegro, blue circle—Bosnia and Herzegovina; yellow circle—North Macedonia; pink circle—Romania; orange circle—Germany; green circle—Italy

and the reform of the army, Augustus VIII Legion was sent to Gaul, and in 69 was replaced by the Legion I of Italy. After the division of the province in 86, Novae was one of the most important fortifications of the Roman limes in the province of Lower Moesia. Before the Dacian wars of Trajan (98–117), the fortress was fortified with stone walls, although most of the buildings inside had already been built of stone. In this way, a protected area of over 18 hectares was established. Trajan himself apparently visited this place because lead seals from his luggage were found in the area. The peaceful times lasted almost 250 years until Kniva, the chief of the Goths, conducted raids in the province. Although Novae was not conquered, the area was heavily damaged. By the beginning of the fourth century, the fortress's importance began to decline and the military crew was reduced. In the same period numerous civilian buildings, both residential and industrial, were constructed. In 376 and 441 the region was subjected to new barbarian raids, from Goths and Huns, respectively, after which the Novae site lost its importance. Later on, during the time of Justinian (527–565) the port on the Danube River at Novae was thriving and became the seat of the bishopric with a significant presence of military from the Byzantine army. In the post-Justinian period, the site became inhabited by Slavs.

Regular archaeological excavations began in 1960. Over a period of 60 excavation seasons, the remains of many buildings have been uncovered, the most important of which is the legionary hospital (valetudinarium), commandant's office (principia), legionary baths (thermae), and coarse barracks. The work was carried out by scientists from Warsaw, Poznań, Wrocław and Sofia. Hundreds or even thousands of different items made of lead were found during the work. Most of the lead artifacts found at the site were lead pipes. Lead was also used extensively in architectural features.

Sampling and methodology

Ten lead samples were taken directly from columns (Fig. 2A–H, J) from Novae. Additionally, one lead sample was collected from a lead ingot (Fig. 2I) found during excavations at the Novae site and which was stored in the warehouse of the Center for Archaeological Research In Novae at Warsaw University, Poland. Two additional samples come from water pipes from the archaeological site of Novae. Lead ore samples from polymetallic mines in Madan (Bulgaria), Trepča (Kosovo), and Rudnik Mine (Serbia) were also examined. The samples of ores were taken from specimens in the collections of the University of Warsaw in Warsaw (Poland) and the Institute of Archeology in Belgrade (Serbia).

To determine the mineral composition of the complex multi-mineral ore samples from Rudnik Mine (Serbia), powder X-ray diffraction (XRD) analysis as performed using an X'Pert PRO MPD. The High Score Plus program and the COD database were used to interpret the XRD results.

To perform chemical analysis of lead samples, preparation was made in the form of a 1-inch disk with embedded fragments of individual samples. The chemical composition of the metal fragments was obtained using Electron Probe Microanalysis (EPMA) in the Laboratory of Electron Microscopy, Microanalysis, and X-Ray Diffraction, Faculty of Geology, University of Warsaw. The chemical analyses were carried out by Cameca SXFiveFE instrument equipped with five wavelength dispersive spectrometers (WDS). The samples were embedded in epoxy resin, polished, and carbon-coated. The operating conditions of the electron microprobe were: 15 kV accelerating voltage, 15 nA probe current, and a focused electron beam. The following analytical crystals and X-ray lines (in brackets) were used: TAP (Al Kα, As Lα, Se Lα), LPET (S Kα, Pb Mα, Si Kα, Ca Kα, P Kα, Sb Lα, Sn Lα, Ag Lα, Au Mα, Te Lα, Cd Lα, Hg Lα, Pb Lα, Bi Mα,) and LLIF (Fe Kα, Mn Kα, Cu Kα, Ni Kα, Co Kα, Cr Kα, Zn Kα). The natural and synthetic standards used in the analysis were as follows: orthoclase (Si, Al), wollastonite (Ca), ZnS (S, Zn), PbTe (Pb), Fe_2O_3 (Fe), rhodonite (Mn), Cu_2O (Cu), NiO (Ni), CoO (Co), Cr_2O_3 (Cr), $ZnAs_2$ (As), Bi_2Te_3 (Bi), CdSe (Cd, Se), Ag (Ag), Au (Au), HgTe (Hg), SnO_2 (Sn), Pd (Pd), InSb (Sb) and $EuPO_4$ (P). The Φ (ρZ) correction model (X-PHI in the electron microprobe software) developed by [52] was used to quantify the chemical composition of the investigated metal fragments. The results of the analysis are presented in Table 1.

The lead samples were cleaned of any external dirt and patina associated with metal corrosion. This was done to reduce errors during isotope analyses. The ore samples (S1, S2, PbS—galena; Or—galena + sphalerite + pyrrhotite) were crushed and then ground. All samples were dissolved in ultrapure nitric acid. Due to the fact that lead was the predominant element in both metal and ore samples, no chromatographic resins were used to purify the samples.

Lead isotope ratios were measured at the University of Warsaw Biological and Chemical Research Centre using a multicollector inductively coupled plasma mass spectrometer (MC-ICP-MS, Plasma II, Nu Instruments, Wrexham, UK). Isotopic analyses of lead were performed using Tl NIST 997 as an internal standard. Pb isotope analyses were performed in dry plasma mode with Aridus 3 desolvation nebulizer (Cetac, Omaha) as a way of introducing the sample. As lead

Fig. 2 Photos of lead sampling sites from columns and pedestals (**A**–**H**, **J**) at the Novae site (N. Bulgaria). **I**—**a** lead ingot found at the Novae site

content in the original samples was very high, no lead matrix separation was performed. As long as lead content is high enough a very good accuracy of Pb isotopic results without matrix separation can be achieved [53]. Sensitivity for ^{208}Pb was 0.8–1.0 V per µg/L, so measurements were taken from solutions at approximately 50 µg/L of total Pb. The results of the measurement of the NIST 981 standard are presented in Table 2.

Results

The results of chemical analyses (Table 1) of most samples indicate pure lead without significant admixtures (above 0.1 wt%) of other metals. Elevated Sn content was noted only in samples no. 2 and 15, and elevated content of Sb in sample no. 2. All samples showed a significant silicon content. This is probably due to contamination of the sample with abrasive material. All samples show trace amounts of Te (< 0.1 wt%). Most samples show an

Table 1 Results of chemical analyses of lead samples from Novae (N. Bulgaria). The results were averaged over 5 measurements for each sample. BDL—below detection limit

Weight%	Fe	Cu	Co	Ni	Cr	Zn	Mn	S	Pb	Si	Te	Al	Se	Ag
S3_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.039	94.566	0.679	0.063	0.058	BDL	BDL
S4_av	BDL	0.084	BDL	BDL	BDL	BDL	BDL	0.043	92.772	0.980	0.046	0.097	BDL	BDL
S5_av	BDL	0.096	BDL	BDL	BDL	BDL	BDL	0.048	91.609	0.990	0.057	0.073	BDL	BDL
1_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.057	93.698	0.016	0.057	BDL	BDL	BDL
2_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.047	91.646	0.185	0.050	BDL	BDL	BDL
3_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.051	90.842	1.616	0.061	0.071	BDL	BDL
5_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.045	91.133	1.878	0.049	0.054	BDL	BDL
7_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.043	91.731	1.320	0.069	0.105	BDL	BDL
8_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.051	93.396	0.779	0.052	0.060	BDL	BDL
9_av	0.044	BDL	BDL	BDL	BDL	BDL	BDL	0.046	93.605	0.964	0.044	0.122	BDL	BDL
10_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.043	87.504	2.692	0.045	0.065	BDL	BDL
15_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.042	93.066	1.431	0.052	0.050	BDL	BDL
32_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.044	90.589	1.139	0.057	0.140	BDL	BDL
BDL_av	0.041	0.077	0.054	0.063	0.035	0.097	0.049	0.014	0.144	0.014	0.021	0.018	0.013	0.027

Weight%	Cd	As	Au	Hg	Sn	Pd	Bi	Sb	P	Ca	Total
S3_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.033	95.595
S4_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.052	94.198
S5_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.058	93.117
1_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.016	94.046
2_av	BDL	0.040	BDL	BDL	0.115	BDL	BDL	0.172	BDL	0.020	92.424
3_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.059	92.886
5_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.066	93.442
7_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.062	93.516
8_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.048	94.658
9_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.064	95.074
10_av	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	0.069	90.591
15_av	BDL	BDL	BDL	BDL	0.139	BDL	BDL	BDL	BDL	0.053	95.059
32_av	BDL	BDL	0.051	BDL	BDL	BDL	BDL	BDL	BDL	0.094	92.311
BDL_av	0.034	0.031	0.049	0.070	0.021	0.025	0.068	0.027	0.016	0.011	

elevated content of Al (< 0.25 wt%), Ca and S (< 0.1 wt%). Cu, Ag, and Au can be indicated as trace admixtures in individual samples. The rest of the analyzed elements in average values are below the detection limit.

Due to the very high plasticity of lead, the abrasive material used to polish the sample adhered to the metal. This resulted in an uneven surface of the sample and the addition of SiC. Additionally, the uneven surface and surface oxidation of the sample resulted in low totals (around 90–97 wt%).

The results of lead isotope analyses for lead samples are in the following ranges: $^{208}Pb/^{206}Pb$ 2.0719 to 2.0824; $^{207}Pb/^{206}Pb$ 0.8355 to 0.8456; $^{206}Pb/^{204}Pb$ 18.469 to 18.741; $^{208}Pb/^{204}Pb$ 38.402 to 38.867; $^{207}Pb/^{204}Pb$ 15.617 to 15.670. The results of lead isotope analyses for ores fall within the following ranges: $^{208}Pb/^{206}Pb$ 2.0784 to 2.0887; $^{207}Pb/^{206}Pb$ 0.8386 to 0.8500; $^{206}Pb/^{204}Pb$ 18.380 to 18.686; $^{208}Pb/^{204}Pb$ 38.334 to 38.901; $^{207}Pb/^{204}Pb$ 15.618 to 15.666. Detailed results of sample measurement are presented in Table 2 and Fig. 3.

The results obtained for lead samples can be divided into three main groups. Sample no. 8 has the highest values of lead isotope ratios of $^{208}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$ and elevated $^{207}Pb/^{204}Pb$, and so could be assigned to a different group. But overall the Pb isotope values in no. 8 are relatively close to the majority of the Novae samples, particularly when one considers the overall variations observed in ores in the region (Figs. 3, 4, 5, 6, 7). Sample no. 32 (ingot) shows overall lower Pb isotope values compared to the rest of the samples. This indicates that this sample most likely comes from a different source than the rest of the samples analyzed in this work.

X-ray diffraction analysis showed that the ore sample from Rudnik Mine (Serbia) consists of galena, sphalerite,

Table 2 Table presenting the description, archaeological dating, and results of lead isotopic analyses of artifacts made of lead from Novae (S3, S4, S5, 1, 2, 3, 5, 7, 8, 9, 10, 15, 32) and lead ores from Madan (S1), Trepča (S2) and Rudnik Mine (PbS, Or)

Sample name	Country	Region	Type	Principal Metal	Description	Chronology	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$
S3	Bulgaria	Novae	Lead	Pb	Water pipe	No dating	2.0771	0.8387	18.683	15.666	38.806
S4	Bulgaria	Novae	Lead	Pb	South pipe	No dating	2.0799	0.8406	18.628	15.654	38.743
S5	Bulgaria	Novae	Lead	Pb	32/93 w	No dating	2.0824	0.8415	18.610	15.656	38.754
1	Bulgaria	Novae	Lead	Pb	VICTORIA pedestal—lead; InGL No. 46; section X; Episcopal Basilica	184 CE	2.0771	0.8379	18.679	15.657	38.797
2	Bulgaria	Novae	Lead	Pb	Signum orignis—InGL lead No. 47 found in 1976, section X; Episcopal Basilica	208 CE	2.0719	0.8379	18.701	15.670	38.748
3	Bulgaria	Novae	Lead	Pb	Signum orignis—InGL lead No. 47 found in 1976, section X; Episcopal Basilica	208 CE	2.0773	0.8393	18.653	15.655	38.748
5	Bulgaria	Novae	Lead	Pb	Column base; section XI; PRINCIPIA, in situ	1st half of the second century	2.0822	0.8410	18.604	15.645	38.736
7	Bulgaria	Novae	Lead	Pb	Altar; section XI; PRINCIPIA	No dating	2.0775	0.8384	18.679	15.624	38.805
8	Bulgaria	Novae	Lead	Pb	Altar; section XI; PRINCIPIA	No dating	2.0739	0.8355	18.741	15.658	38.867
9	Bulgaria	Novae	Lead	Pb	Section X; Episcopal Basilica but it may come from an earlier facility, e.g. a bathhouse	1st half of the second century	2.0766	0.8381	18.677	15.652	38.785
10	Bulgaria	Novae	Lead	Pb	Section VIIIA—villa extra muros	1st half of the second century	2.0768	0.8382	18.676	15.655	38.786
15	Bulgaria	Novae	Lead	Pb	Column bases from the house, inscription INEGRO, INERONTE; section XI; PRINCIPIA	1st half of the 3nd century	2.0774	0.8385	18.666	15.650	38.776
32	Bulgaria	Novae	Lead	Pb	Lead bar; 056\83 w; section IV, Horreum	395–402	2.0793	0.8456	18.469	15.617	38.402
S1	Bulgaria	Madan	Ore	Pb	Galena		2.0819	0.8386	18.686	15.666	38.901
S2	Serbia	Trepca	Ore	Pb	Galena		2.0857	0.8500	18.380	15.618	38.334
PbS	Serbia	Rudnik Mine	Ore	Pb	Galena		2.0784	0.8387	18.657	15.648	38.777
Or	Serbia	Rudnik Mine	Ore	Pb	Galena + sphalerite + pyrrhotite		2.0789	0.8387	18.667	15.656	38.805
NIST 981							2.1661	0.9145	16.932	15.484	36.677

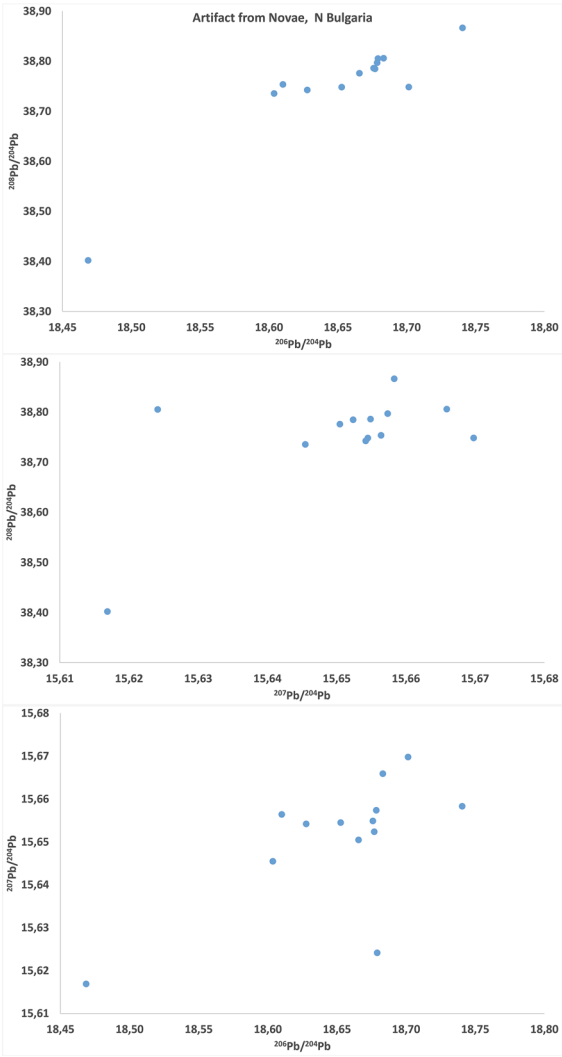

Fig. 3 Diagrams showing Pb isotope ratio results for archaeological samples from Novae

and pyrrhotite, and so lead is dominantly hosted by galena (PbS).

The full dataset is included in the Additional files 1 and 2.

Discussion

The elevated (> 0.1 wt%) Sn content of samples no. 2 and no. 15 suggests that these objects were formed from recycled metal. Similar values were reported for objects made of lead in other parts of the Roman Empire [e.g. 48–50]. They were described as recycled lead pipes soldered with lead–tin solders [e.g. 48–50]. Accordingly, samples no. 2 and no. 15 were omitted from the ore source assessment.

Lead isotope analysis was employed to determine the source of the ore used by people inhabiting Novae between the first and eighth centuries AD. The results were compared to analogous results published by the OXALID laboratory obtained for lead ores from the territory of today's Bulgaria, Greece and Italy [20, 21, 32–47, 54, 55] and additional data for ores from present-day Germany [56, 57], Romania [25], Serbia [26, 54, 55, 58–60], North Macedonia [54, 55], Montenegro [54, 55] and Bosnia and Herzegovina [54, 55] used by the Romans. In addition, new analyses (Table 2) from several well-known polymetallic ore mines (Madan (Bulgaria), Trepča (Kosovo), and Rudnik Mine (Serbia)) were included in the database. The interpretation of the results was based on the analysis of binary isotope ratio plots ($^{208}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$; $^{207}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$; $^{208}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$) and on numerical data of calculated Euclidean distances (Additional file 1). The following criteria were adopted:

i. A good match was considered if over 70% of the Euclidean distance results are below the value of 0.1, including over 40% of the results are below 0.05;
ii. A slight match was considered if at least 50% of the Euclidean distance results were below the value of 0.1;
iii. A very slight match was considered if less than 50% of the Euclidean distance results were below the value of 0.1.

Novae lead versus Bulgarian ores

The lead isotope results obtained for the lead samples from Novae overall plot within the Pb isotope range published for Pb deposits in the area of present-day Bulgaria [34, 35, 54, 55] (Fig. 4), specifically:

- There is a good match between sample no. S3 and the Zvezdel and Zvezdel-Pcheloyad (East Rhodope) deposits and a slight match with the Madan, Spahievo (East Rhodope) and Gradishteto, Malko Turnovo (South East);
- There is a good match between sample no. 8 and the Zvezdel and Spahievo (East Rhodope), Madjarovo [South (Kardzhali)] deposits and a slight match with the Madan, Madzharovo and Zvezdel-Pcheloyad (East Rhodope), Laki and Madan-Thermes (Central Rhodopes);
- There is a good match between samples no. 1 and no. 7 and the Zvezdel-Pcheloyad (East Rhodope) deposits and a slight match with the Zvezdel and Spahievo (East Rhodope) and Gradishteto, Malko Turnovo (South East);

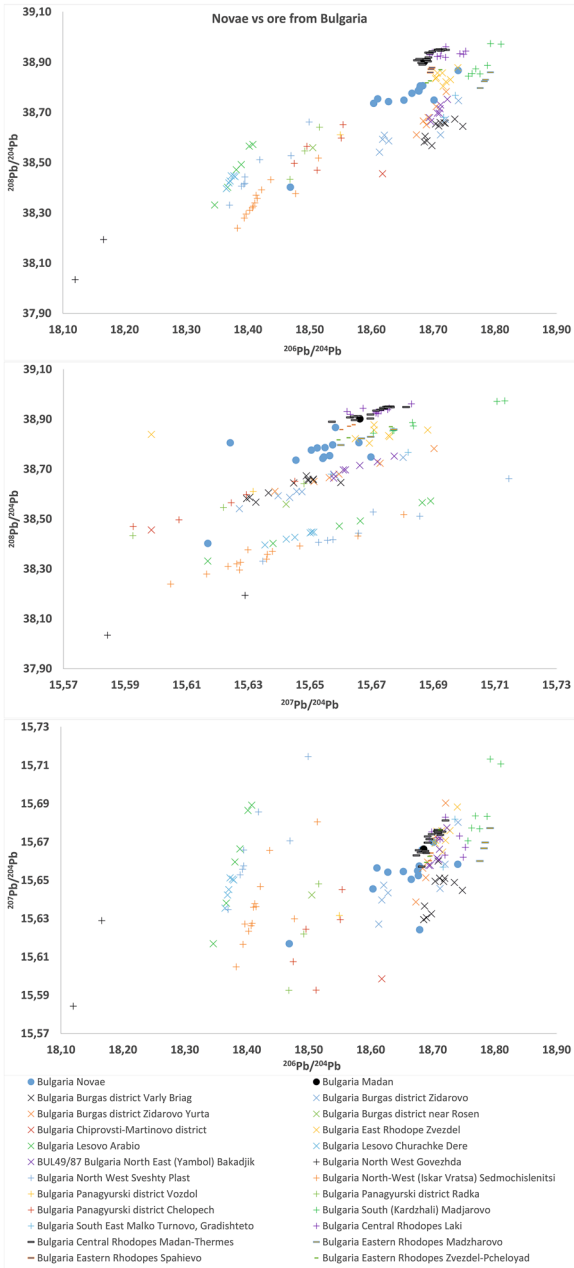

Fig. 4 Comparative charts showing the results from Novae compared to the results from the deposits in Bulgaria [33, 34, 54, 55]

- There is a good match between samples no. 9 and 10 and the Zvezdel-Pcheloyad (East Rhodope) deposits and a slight match with the Zvezdel and Spahievo (East Rhodope), Bakadjik [North East (Yambol)] and Gradishteto, Malko Turnovo (South East);
- There is a slight match between sample no. S4 and the Zidarovo Yurta (Burgas district) and Bakadjik [North East (Yambol)];
- There is a slight match between sample no. 3 and the deposits of Zidarovo Yurta (Burgas district), Zvezdel-Pcheloyad (East Rhodope), Bakadjik [North East (Yambol)] and Gradishteto, Malko Turnovo (South East);
- There is a slight match between sample no. 32 and the Sveshty Plast and Sedmochislenitsi (North West).
- The remaining samples (no. S5, no. 5) yielded no matches with Bulgarian lead ores.

Novae lead versus Serbia ores

A number of lead samples from Novae show a very good match with the ore results presented in this work and published data for ores from the Rudnik deposit (Serbia) [26, 54, 55, 58–60] (Fig. 5). Samples no. S3, no. 1, no. 9, and no. 10 show a good match with part of the data from the Rudnik mine. Moreover, samples no. 3, no. 7 and no. 8 have a slight match to the Rudnik deposits.

Additional lead mining areas in Serbia are Kopaonik (Novo Brdo, Raska, Stari Trg, Belo Brdo) and Lece (Lece). There is a good match of sample no. 8 to the deposits from Raška, Stari Trg, and Kopaonik Mountain, and samples no. S4, no. S5 to the Novo Brdo deposit. Samples no. S3, no. 1, no. 7, no. 9, and no. 10 show a slight match to the Novo Brdo, Raska, Stari Trg, Belo Brdo, and Lece deposits. However, samples no. 3 and no. 5 show a slight match to the Novo Brdo deposit. Additionally, sample no. 8 shows a slight match to the Novo Brdo and Lece deposits. Sample no. 32 has no match.

Novae lead versus North Macedonia ores

Comparing the results of samples from Novae to samples from North Macedonia [54, 55] (Fig. 5), only matches to ores from Zletovo (Zletovo) are visible (good match—no. S3, no. 1, no. 3, no. 7, no. 9, and no. 10; slight match—no. S4, no. S5, and no. 8).

Novae lead versus Montenegro ores

The comparison charts (Fig. 5) of samples from Novae to data from Montenegro [54, 55] demonstrate the lack of matching of the results.

Novae lead versus Bosnia and Herzegovina ores

Results for samples from Novae compared to samples from Bosnia and Herzegovina [54, 55] (Fig. 5) match only with the Srebrenica deposit (Drina/Podrinje) (good match—no. S3, no. 1, no. 7, no. 9 and no. 10; slight match—no. S5, no. 3, and no. 8).

Novae lead versus Romanian ores

Samples no. S4, no. S5 and no. 5 shows a good match with the data for Cetate Massif (Rosia Montana,

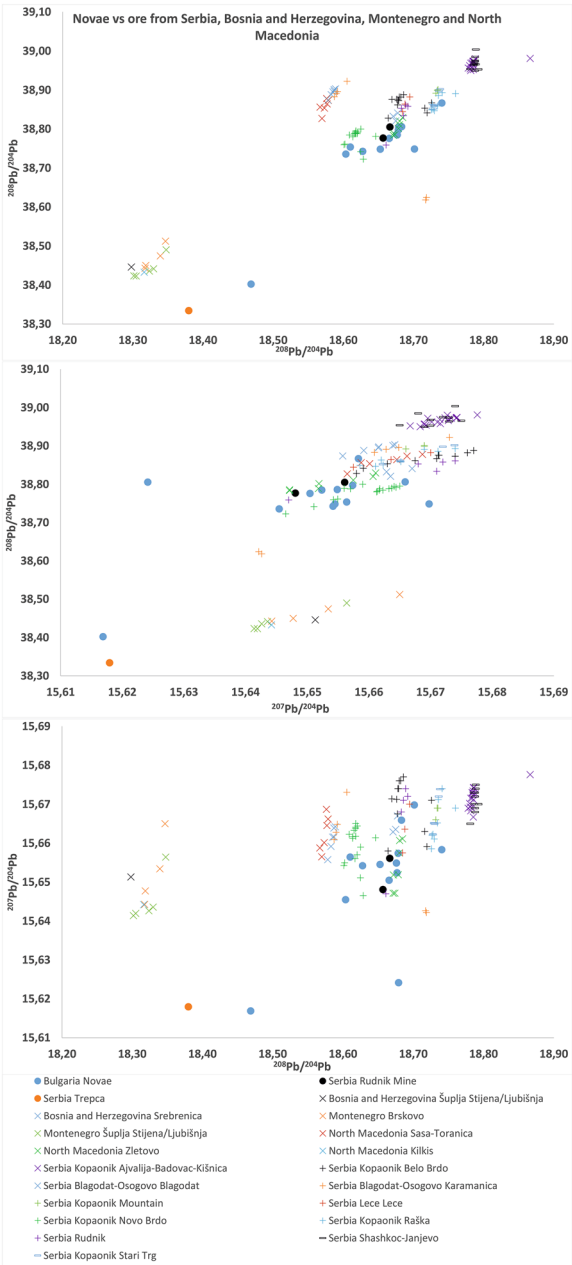

Fig. 5 Comparative charts showing the results from Novae compared to results from deposits in Serbia [26, 54, 55, 58–60], North Macedonia [54, 55], Montenegro [54, 55] and Bosnia and Herzegovina [54, 55]

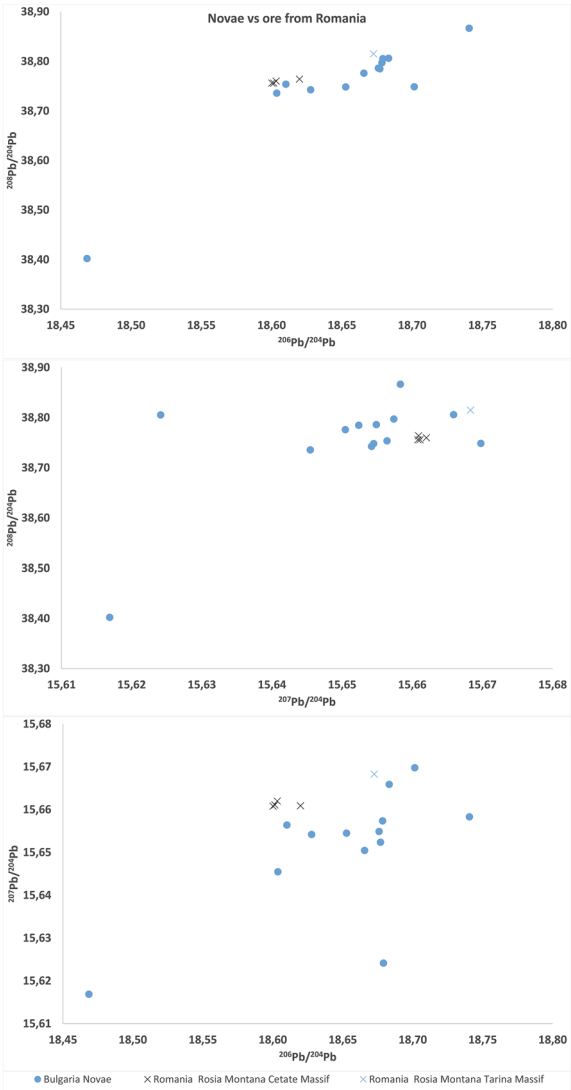

Fig. 6 Comparative charts showing the results from Novae compared to results from deposits in Romania [25]

Romania) ores and a slight match to Tarina Massif (Rosia Montana, Romania) ores. Contrarily, samples no. S3, no. 1, no. 7, no. 9, and no. 10 show a good match with the data from Tarina Massif (Rosia Montana, Romania) and a slight match to Cetate Massif (Rosia Montana, Romania) ores (Fig. 6). Sample no. 3 show a slight match to both deposits, while sample no. 8 has a slight match to the Tarina Massif deposit (Rosia Montana, Romania).

Sample no. 32 from Novae shows no match with ores from the Rosia Montana deposit. However, it is important to note that the mineralization is mainly Au–Ag ore in quartz veins with a subordinate amount of sulfide minerals such as pyrite, sphalerite, and galena. Therefore, if any Pb–Zn ore was mined in Rosia Montana it would likely be relatively minor quantities. Additionally, it is quite likely that Pb mined in the deposit was used locally during the cupellation of the Au–Ag ores. Overall, it is rather unlikely that a large amount of lead was mined and exported from Rosia Montana during the Roman period.

Novae lead versus Greek ores

Comparing the results obtained for samples from Novae to the results for deposits from Greece [38–47, 54] (Fig. 7), one can identify possible matches with four main deposits with good and/or poor fit:

- For deposits from the Rhodope region, a good match of samples no. S3, no. 1, no. 7, no. 9, and no. 10, and a slight match of samples no. S4 and no. 3 are visible;
- For deposits from the Thrace region, there is a good match to samples no. S3, no. 1, no. 7, no. 9, and no. 10 and a slight match to samples no. S4, no. 3, and no. 8;
- For deposits from the Pangeon Mt region. There is a good match to samples no. S3 and no. 1 and a slight match to samples no. 3, no. 7, no. 8, no. 9, and no. 10;
- For deposits from the Euboea region, a good match to samples no. S3, no. 1, no. 7, no. 9, and no. 10 and a slight match to samples no. 3 and no. 8 are visible.

Moreover, there is a good match of sample no. 8 to deposits from the N. Aegean, Thasos, and Xanthi regions and a slight match of samples no. S3 and no. 1 to deposits from the Xanthi region. The rest of the samples show no or very slight match.

Novae lead versus Italian ores

The results obtained for the samples from Novae are compared to published data for Italian deposits [32, 33, 37] (Fig. 8). Overall, the samples from Novae show more radiogenic Pb isotopes compared to the majority of Italian deposits. Few Italian deposits show Pb isotopes close to the artifacts (Fig. 8), suggesting that no Pb from ores from present-day Italy were utilized in Novae. Only samples no. S4, no. S5 and no. 5 show a similarity to ore from the Capo Marargiu (SS) deposit in Sardinia (Italy). However, it is unlikely that lead will be transported from Sardinia to Bulgaria at the time.

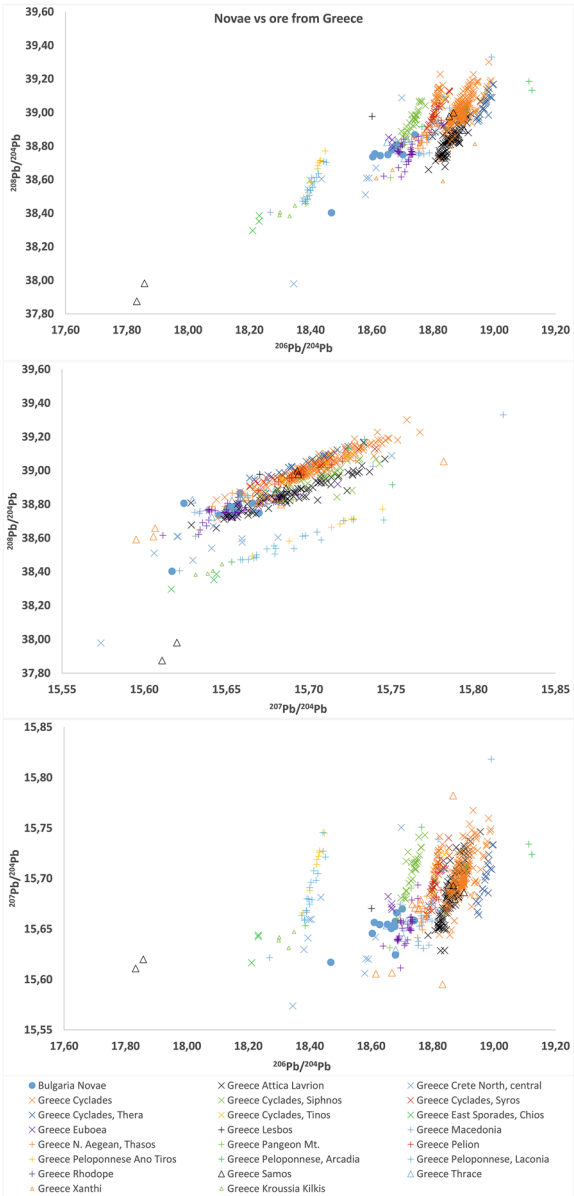

Fig. 7 Comparative charts showing the results from Novae compared to results from deposits in Greece [38–47, 54]

Novae lead versus German ores

Novae samples show distinctly more radiogenic Pb isotopes when compared to German ores (Fig. 9). Only one sample (no. 32, the ingot from Novae) plots within the range of the German ores. The ingot sample matches the radiogenic Pb isotope values reported for German ores from the vicinity of Siegerland and Hunsrueck [56, 57] (Fig. 9). However, the ingot sample also plots within the range for the Bulgarian deposits so it is not possible to distinguish solely on Pb isotopes alone if the metal used in the ingot was from German or Bulgarian ores. However, it is unlikely that lead will be transported from Germany at the time, given that the rest of the samples show origin from regional (Bulgaria-Serbia-Romania) deposits.

Interpretation of results

The data presented in Figs. 3, 4, 5, 6, 7, 8, 9 and Table 3 indicate that the majority, if not all, of the Pb artifacts in Novae were made of from ores of the Balkan region. Lead isotope patterns between lead samples from Novae and lead ores allow for division into 3 main groups:

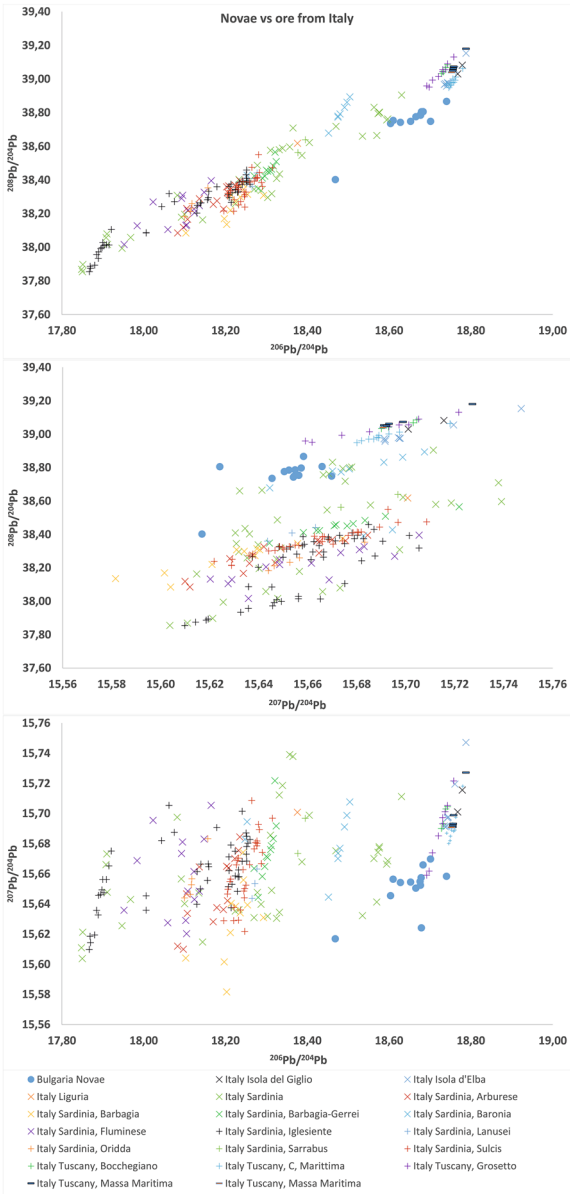

Fig. 8 Comparative charts showing the results from Novae compared to results from deposits in Italy [32, 33, 37]

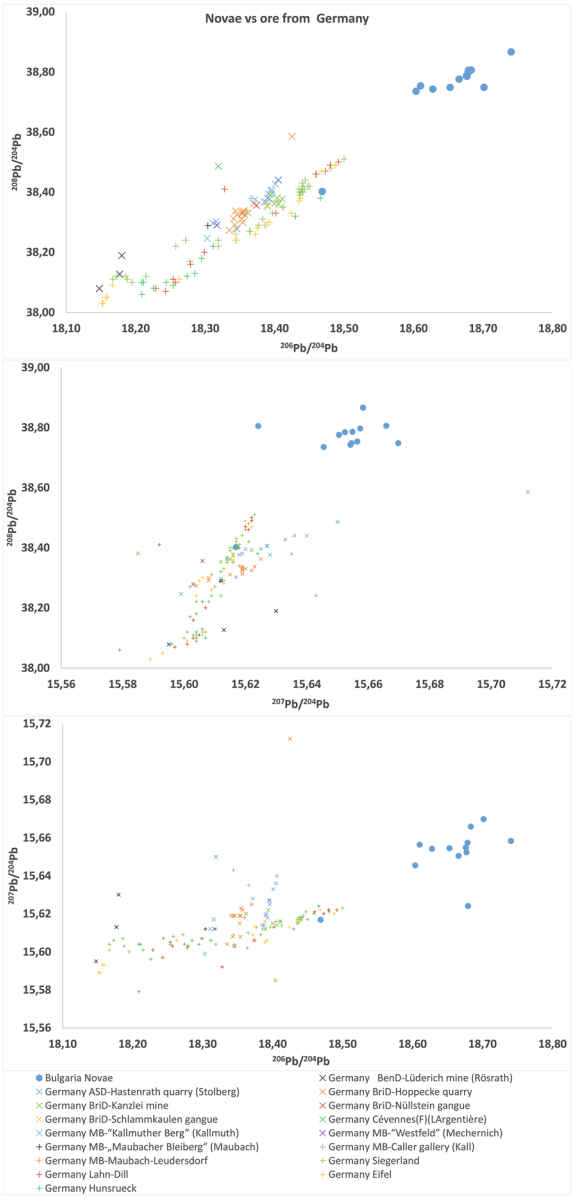

Fig. 9 Comparative charts showing the results from Novae compared to results from deposits in Germany [56, 57]

- Group 1 consists of samples (no. S4, no. S5) that show possible match to two deposits.
- Group 2 consists of samples (no. S3, no. 1, no. 7, no. 9, no. 10), that overall match a number of deposits in Bulgaria, Serbia, Greece, North Macedonia, Bosnia and Herzegovina, and Romania.
- Group 3 consists of samples no. 3, no. 5, no. 8, no. 32, which show overall distinct matching patterns compared to group 1 and 2samples.

Group 1 is consistent with only two locations—Novo Brdo in Serbia or Cetate Massif, Rosia Montana in Romania. However, considering the nature of the deposits (Novo Brdo—lead deposit; Cetate Massif—gold deposit) and trace amounts of only Cu in the chemical analysis, we can confidently identify Novo Brdo as the source of the ore for these samples. If the ore came from a polymetallic deposit, the metal would be expected to also contain trace amounts of other metals (Ag, Au, Bi, Sn, Sb, etc.).

Table 3 Table presenting the match of lead isotope results obtained for particular artifacts from Novae to literature data obtained for lead ore deposits from Bulgaria, Serbia, North Macedonia, Montenegro, Bosnia and Herzegovina, Greece, Italy, Germany, and Romania

No.		Bulgaria												
		Burgas district				Burgas district near Rosen	Chiprovsti-Martinovo district	East Rhodope				Lesovo		North East (Yambol)
		Madan	Varly Briag	Zidarovo	Zidarovo Yurta	Meden Rid	Veneca	Zvezdel	Madzharovo	Spahievo	Zvezdel-Pcheloyad	Arabio	Churachke Dere	Bakadjik
Novae	S3	x		o	o			+	o	x	+			o
	S4				x/o						o			x
	S5													
	1			o	o			x	o	x	+			o
	2		o	o	+			x	o		x			+
	3			o	x			o			x			x
	5													
	7				o			x		x	+			o
	8	x			o			+	x	+	x			
	9			o	o			x		x	+			x
	10			o	o			x		x	+			x
	15			o	o			x		o	x			x
	32													

No.		Bulgaria										Comments
		North West			Panagyurski district			South (Kardzhali)	South East	Central Rhodopes		
		Govezhda	Sveshty Plast	Sedmochislenitsi	Vozdol	Radka	Chelopech	Madjarovo	Malko Turnovo, Gradishteto	Laki	Madan-Thermes	
Novae	S3							o	x		o	
	S4											
	S5											
	1							o	x		o	
	2								+			Probable recycling
	3								x			
	5						o					
	7							o	x		o	
	8							+		x	x	
	9								x			
	10								x			
	15								x			Probable recycling
	32		x	x		o	o					

Table 3 (continued)

No.		Serbia											
		Kopaonik					Kopaonik Moutain	Blagodat-Osogovo		Lece	Rudnik	Shashkoc-Janjevo	Trepca
		Ajvalija-Badovac-Kišnica	Belo Brdo	Novo Brdo	Raška	Stari Trg		Blagodat	Karamanica	Lece			
Novae	S3		x	x	x	x	o			x	+		
	S4		o	+							o		
	S5		o	+							o		
	1		x	x	x	x				x	+		
	2		o	x						o	x		
	3		o	x							x		
	5			x							o		
	7		x	x	x	x				x	x		
	8	o	x		+	+	+			x	x	o	
	9		x	x	x	x				x	+		
	10		x	x	x	x				x	+		
	15		o	x	o	o				x	+		
	32												

No.		North Macedonia			Montenegro		Bosnia and Herzegovina		Romania		Italy	Germany	
		Blagodat-Osogovo	Zletovo	Kroussia	Northeast Montenegro		Northeast Montenegro	Drina/Podrinje	Rosia Montana				
		Sasa-Toranica	Zletovo	Kilkis	Brskovo	Šuplja Stijena/ Ljubišnja	Šuplja Stijena/ Ljubišnja	Srebrenica	Cetate Massif	Tarina Massif			
Novae	S3		+					+	x	+	o		
	S4		x					o	+	x	o		
	S5	o	x					x	+	x	o		
	1		+					+	x	+	o		
	2		x					x	x	x			Probable recycling
	3		+					x	x	x	o		
	5	o	o						+		o		
	7		+					+	x	+			
	8		x					x		x			
	9		+					+	x	+	o		
	10		+					+	x	+	o		
	15		+					x	x	+	o		Probable recycling
	32											o/x	

Table 3 (continued)

No.		Greece												
		Attica Lavrion	Crete North, central	Cyclades	Cyclades, Siphnos	Cyclades, Syros	Cyclades, Thera	Cyclades, Tinos	East Sporades, Chios	Euboea	Lesbos	Macedonia	N. Aegean, Thasos	Pangeon Mt
Novae	S3				o					+		o	o	+
	S4		o		o					o		o		
	S5		o		o					o		o		
	1				o					+		o	o	+
	2				o					x		o	o	x
	3		o		o					x		o		x
	5		o		o					o		o		
	7				o					+		o	o	x
	8	o		o	o	o				x		o	+	x
	9				o					+		o	o	x
	10				o					+		o	o	x
	15				o					+		o	o	x
	32													

No.		Greece									
		Pelion	Peloponnese	Peloponnese, Arcadia	Peloponnese, Laconia	Rhodope	Samos	Thrace	Xanthi	Kroussia	
Novae	S3					+		+	x		
	S4					x		x			
	S5					o		o			
	1					+		+	x		
	2					+		x	o		Probable recycling
	3					x		x			
	5					o		o			
	7					+		+	o		
	8	o				o		x	+		
	9					+		+	o		
	10					+		+	o		
	15					+		x	o		Probable recycling
	32										

+: good match, x: slight match, o: very slight match, no match

Group 2 has good matches with deposits such as Zvezdel-Pcheloyad in Bulgaria, Rudnik in Serbia, Zletovo in North Macedonia, Tarina Massif, Rosia Montana in Romania and the Euboea, Rhodope and Thrace regions in Greece. This makes it difficult to identify solely based on Pb isotopes which deposit was the ore source. Considering the nature of the deposit and the mined ore can provide more constraints on the possible sources. Several of the deposits are focused on the extraction of gold (including Thrace, Rosia Montana, Rhodope). Other are silver (including Zvezdel-Pcheloyad, Rudnik, Euboea, Thrace), or copper (including Rudnik, Euboea, Thrace, Rhodope) and/or lead and zinc (including Rudnik, Zletovo, Euboea, Thrace, Rhodope). However, data published on Mindat.org show that galena is present in each of these deposits. Moreover, all of them were exploited during the Roman rule in this area. Literature data also indicate that, for example, Rudnik is considered one of the sources of lead in the Roman period. Chemical analysis data show trace amounts of Au and Te in the samples and virtually no Ag, Cu, and Zn. This suggests that either the lead came from a lead deposit with elevated Au content lacking Ag, Cu, and Zn, or that the lead came from Au deposits, where Pb was also mined as a secondary commodity, or alternatively, lead came from polymetallic deposits, but all metals except Au admixtures were effectively removed from it. Therefore, additional data is required to determine which Balkan deposit was the source for Group 2.

Group 3 consists of samples with different matches. Sample no. 3 has a good match only to the Zletovo deposit in North Macedonia. The results of chemical analyses of the samples (Additional file 2) show trace amounts of Ag, Al, Au, and Fe, which may reflect the chemistry of the deposit. Sample no. 5 also has only one good match to Cetate Massif, Rosia Montana in Romania. This is interesting because there are no traces of Au in the chemical composition. In individual analytical points, trace values of Sn ($\ll 0.1$ wt%), Se, Cu, and Bi can be observed (the average value of these elements is below the detection limit). This would indicate more of a polymetallic deposit consisting of Pb, Bi, and Cu sulphosalts than a gold mine. Sample no. 8 has the best match in group 3 to Zvezdel, Spahievo, and Madjarovo in Bulgaria, also to Raška, Stari Trg, and Kopaonik Mountain in Serbia, and also N. Aegean, Thasos and Xanthi in Greece. However, it should be noted that the Greek deposits have the weakest match of these three regions. Analyzing the chemical composition (trace amounts of Ag, Al, Au, and Cu—Additional file 2) may indicate the polymetallic origin of the ore. Both deposits in Bulgaria and Serbia have similar chemistry. This makes it impossible to directly indicate the source of the ore. Sample no. 32 does not have any good match. It has slight matches to the Sveshty Plast and Sedmochislenitsi deposits in Bulgaria and Siegerland in Germany. Judging by the content of trace elements in the sample such as Au, it can be assumed that the most likely source will be Sveshty Plast, described as a Pb-Au mine [33, 34].

Only 4 samples have good matches to individual deposits. However, sample no. 5, despite a good match, has a chemical composition that does not fully match the chemistry of the deposit in Romania (no Au in the lead composition). Six samples have good matches to several deposits. They may be the result of mixing ores from different sources (mixing lead bars from different deposits), although there are no mixing patterns evident in the trace elements. Alternatively, they may come from one of the indicated deposits, but their isotopic and chemical composition are similar, making it difficult to determine a single source. Sample no. 32 is the most problematic. It does not yield a good match any of the deposits considered in this study. Slight matches indicate NW Bulgarian or German deposits. This may be due to the use of TIMS for lead analyses of ores versus MC-ICP-MS for the lead metal samples in this study. Isotope fractionation may occur during TIMS analysis [61–64], thereby distorting the results.

Patterns emerge when one considers the age and use of the lead objects analyzed. Within one object (Signum orignis) we have lead derived possibly from recycling (no. 2) and along with lead from another deposit (no. 3). Lead samples from the columns, despite dating to the same period, were derived from different sources (no. 5, no. 9, no. 10) or possibly recycling (no. 15). In later periods, lead was probably imported from other regions of the Balkans or slight possibility for outside of the Balkans, as evidenced by the data for sample no. 32. Similar patterns were observed by researchers studying lead pipes at Roman sites in Portugal and Spain [48–51] with pipes from a single building having been produced from several deposits.

When comparing the Pb isotope results of samples from Novae to data from Sardinia and Montenegro, no significant matches were observed.

Conclusions

Comparing the obtained isotope results for the lead samples from Novae with the lead isotope results for various deposits from Bulgaria, Greece, North Macedonia, Montenegro, Bosnia, and Herzegovina, Italy, Germany, and Romania, several matches of the metal results with the ore results can be observed. This outlines at least 4 possible directions for lead transport to Novae. Additionally, the isotope data also shows some changes through time in the ore source for metal production. Lead samples dated to the second—third

century AD came from several sources at the same time. The first source was the Novo Brdo mine in Serbia. The results obtained for Novo Brdo show a very good match to the results obtained for two lead samples (no. S4, no. S5). Another very likely source of lead is the Zletovo deposit in North Macedonia. Sample no. 3 has a good match for this deposit. The rest of the samples (no. S3, no. 1, no. 7, no. 8, no. 9, no. 10) from this period most likely come from deposits in the Balkan region that share similar isotopic compositions One lead sample from this period (no. 5) remains enigmatic with Pb isotope values that match very well Rosia Montana in Romania, but the Au-free composition is inconsistent with this source. Later, in the fourth century CE lead began to be supplied mainly from NW Bulgaria deposits, with one sample (no. 32) that possibly can also be from deposits in Germany. No clear match between samples from Novae and samples from Sardinia or Montenegro was observed.

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s40494-024-01151-2.

Additional file 1. Table with the results of Pb isotope analyzes for lead samples from Novae and literature data for comparison and determination of provenance. The tables also present results for the calculation of Euclidean distances between samples from Novae and lead ore data from individual deposits. Distances indicating a good match are marked in red, and distances indicating a slight match are marked in yellow. No color means no match.

Additional file 2. Table with raw data for chemical analyzes of lead samples from Novae.

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Author contributions
JR—chief archaeologist, initiator of research, responsible for organizing excavations, collecting samples at the archaeological site, dating samples, archaeological analysis of the obtained results. PS—sampling at the archaeological site, archiving samples, preparing the manuscript, preparing figures in the manuscript. JK—preparation of samples and analysis of Pb isotopes, description of measurement methods. GK—assistance in interpretation of results, preparation of the manuscript. WP—assistance in the interpretation of results, preparation of the manuscript, transfer of literature data for better interpretation. BMM—performing chemical analyzes using FE-EMPA, describing the measurement method. MK—sampling at an archaeological site, preparation of samples for Pb isotope analyses, interpretation of results, preparation of the manuscript, organization of work while writing the manuscript, corresponding author.

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Availability of data and materials
The dataset(s) supporting the conclusions of this article is(are) included within the article [and its additional file(s)].

Declarations

Competing interests
The authors declare no competing interests.

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