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# Central-Eastern Europe as a centre of Middle Ages extractive metallurgy

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## ABSTRACT

Central-eastern to southeastern Europe, from Bohemia to Greece is home to some of the richest ore deposits on earth, with archaeological evidence suggesting a long history of metal use. However, the exact timing and extent of past metal processing activities remains unclear. The Middle Ages and Early Modern period (c. 500–1800 common era (CE)) in Europe, saw the expansion of metal use at an unprecedented scale, continent-wide. Here we analysed rates of past atmospheric lead (Pb) deposition in six peat bogs from Romania, Serbia and Greece. We show that after 1000 CE, the redevelopment of central European mining industry was synchronous with Pb pollution in southeastern Europe, with the onset of metal pollution occurring in the area prior to central Europe. Therefore, southeastern Europe may have led regional mining developments, with technological advances rapidly shifting from east to west through the Middle Ages. This indicates how south-eastern Europe should be included in future discussions of Middle Age metallurgy not simply as a contributor, but at times as a leader in metal production.

## 1. Introduction

The measurement of lead (Pb) concentration in sedimentary archives such as peat and lake sediments has helped to shed light on the extent of past environmental pollution (Dudka and Adriano, 1997; Hansson et al., 2015; Longman et al., 2020; Shotyk et al., 1998; Thevenon et al., 2011). Pb is released into the atmosphere as a by-product of mining, smelting, and metallurgy (Dudka and Adriano, 1997). By analysing the Pb content in peats, the earliest environmental pollution in the Bronze age (Longman et al., 2018; Martínez Cortizas et al., 2013), the rise of atmospheric Pb associated with the Industrial Revolution (Le Roux et al., 2004, 2005), and the Pb burden of leaded gasoline in the 20th Century have been fingerprinted (Shotyk et al., 2016; Weiss et al., 1999a). Recent work using ice core records has taken this approach a step further, introducing a level of analytical and temporal resolution previously unobtainable, which is helping to shed light even on the short-term impacts of societal development, and the role pandemics and conflicts played in economic activities and consequently environmental pollution (McConnell et al., 2018, 2019; More et al., 2017). The primary advantage of these records is that they are usually continuous, and can provide information from periods when historical records and traditional finds-based archaeology are lacking (Longman et al., 2020).

One period for which reconstructions of environmental pollution are of particular interest is the Middle Ages, between 500 and 1700 CE. In Europe, this was a time of considerable societal development, covering the transition from a largely agricultural society following the Roman Empire collapse, to the industrial world in which we live today (McConnell et al., 2019). Our understanding of environmental pollution through this time is based largely on ice core records, which have been taken from either Greenland, Artic or from Alpine glaciers (More et al., 2017; McConnell et al., 2018, 2019). These records provide a high-resolution, integrated signal of European pollution, and so are ideal for investigating short term large-scale drivers of change in the period such as the fall of the Roman Empire, the Black Death and the rise of metal producing empires (More et al., 2017; McConnell et al., 2018, 2019). However, their distance from main pollution centres means such records do not consider the spatial variability of Pb pollution, but instead represent an average signal. Peat and lake sediment records show divergence from these integrated ice core records, indicating considerable inter-site Pb variability, and potentially divergent local

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palaeopollution trends that must be further assessed (Kylander et al., 2005; Longman et al., 2018; De Vleeschouwer et al., 2009; Le Roux et al., 2004, 2005; Shotyk et al., 1998).

For example, the first appearance of a Middle Age Pb pollution peak occurs at different times in records across Europe, suggesting strong spatial variability in metal extraction and processing, most likely in line with past economic development at regional scales. In southern Germany, the first signs of a Medieval increase in atmospheric pollution occurs c.1050 CE (Le Roux et al., 2005), whilst in northern Spain there is evidence of pollution from c.1500 CE on (Martínez Cortizas et al., 2002), and in Scotland there is little signature of pollution until the Industrial Revolution (Cloy et al., 2005, 2008). This regional variability in Pb fallout means that when interpreted together, multiple peat or lake records of pollution can be used to identify regional changes (Renberg et al., 2001). For example, a synthesis of 89 European palaeopollution records demonstrates how 81 of the records contain a Roman lead peak, but 44 of these may be representative of local sources and so represent regional Pb depositional variability (Silva-Sánchez and Armada, 2023).

Clearly, to fully understand the history of metallurgical development across Europe, sites in all regions are required. The record of metal pollution from the period 500–2000 CE is well reconstructed in western Europe from a range of archives (Hansson et al., 2015; Longman et al., 2020; Renberg et al., 2001 and references therein), but studies are lacking from central and eastern Europe, with only a single study of this region presented to date (Longman et al., 2018). This first study of central-eastern palaeopollution through the Middle Ages clearly identifies a different trend compared to western European records (Longman et al., 2018). High overall levels of Middle Age Pb pollution suggest local exploitation of ores, and a lack of slowdown in metal pollution following the collapse of the Western Roman Empire. From this record alone, it appears that south-eastern Europe's palaeopollution history is divergent from western Europe, with the data suggesting an advanced metallurgical industry in southeastern Europe during the period 500–1800 CE (Longman et al., 2018). This is supported by the limited traditional archaeological evidence available, which suggests that mining in the Metalifieri Mountains of Romania and at Rosia Montana began from the 11th Century onwards (Cauuet et al., 2003; Maghiar and Olteanu, 1970). By the 12th Century, the arrival of Saxon workers and the

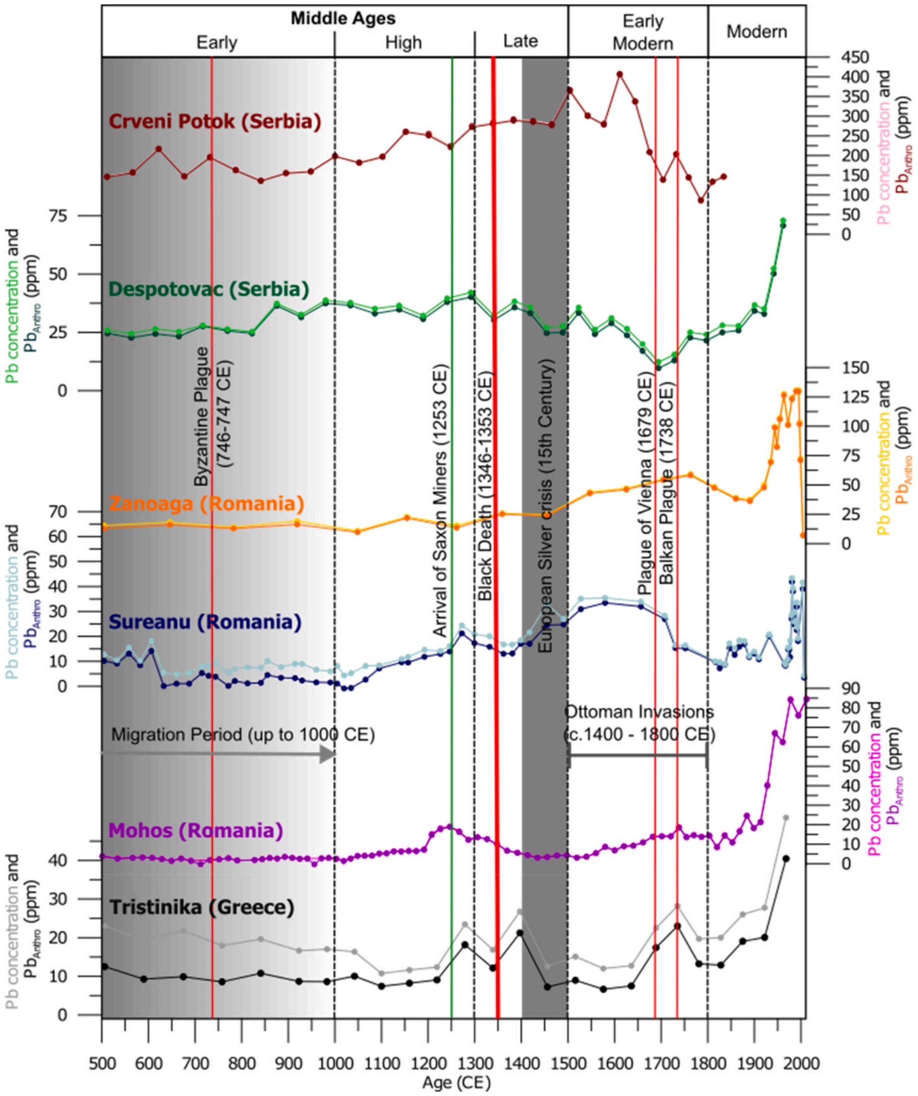

**Fig. 1. Reconstructed anthropogenic Pb pollution for each of the bogs analysed in this study.** Bog names and locations are labelled in the colour of the graph. For exact locations of each bog, see Fig. 3. For each record, we present here the raw Pb concentration, and the calculated anthropogenic Pb level through time (Pb$_{Anthro}$), see methods for information of derivation. Also highlighted are aspects of European history which may be relevant to the Pb chronologies presented here such as plagues (red lines) the arrival of Saxon miners in the Balkans (green line) and other aspects of Balkan history. The sites in Romania (Zanoaga, Sureanu and Mohos) are all north of the Danube, with the remainder south of the river. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

demand for silver in the Hungarian Kingdom meant mines in Transylvania and the Balkan peninsula were extremely important sources of metal to the continent (Borcoş and Udubaşa, 2012; Fine, 1994; Stojković, 2013). However, without further regional studies to fill the gaps in this patchy archaeological record, it has been challenging to ascertain whether the previous local record from Serbia (Longman et al., 2018) is a record of purely local changes or representative of the region as a whole.

To address this challenge, and to provide a more robust investigation of the spatio-temporal variability of European pollution and metallurgical development in the Middle Ages, we present new past pollution data from six well-dated peat and wetland records from southeastern Europe (Fig. 1).

## 2. Material and methods

### 2.1. Site descriptions

Sureanu (SUR) peat bog (45.581°N 23.507°E) is located in the Southern Carpathians (Romania) (Fig. 4), at an altitude of 1840m above sea level (a.s.l.). It is a small bog (200m by 100m) located adjacent to a small lake at the base of a glacial cirque. The climate of the area is temperate continental (Trufas, 1986), with between 900 and 1800 mm rainfall per year. The site is covered by snow for much of the year (roughly 100 days at lower altitudes and 200 days above 2000m a.s.l.). The surrounding geology consists of Late Proterozoic- Early Paleozoic gneiss from the Getic-Supragetic nappe. A 579 cm long core was taken, alongside samples of the neighbouring lake sediment, and background rock. The record grades from lacustrine gyttja in the lowermost 200 cm, before the gradual transformation into a raised bog with the accumulation of *Sphagnum* peat (Longman et al., 2017a). For the upper 400 cm, the core is *Sphagnum*-dominated.

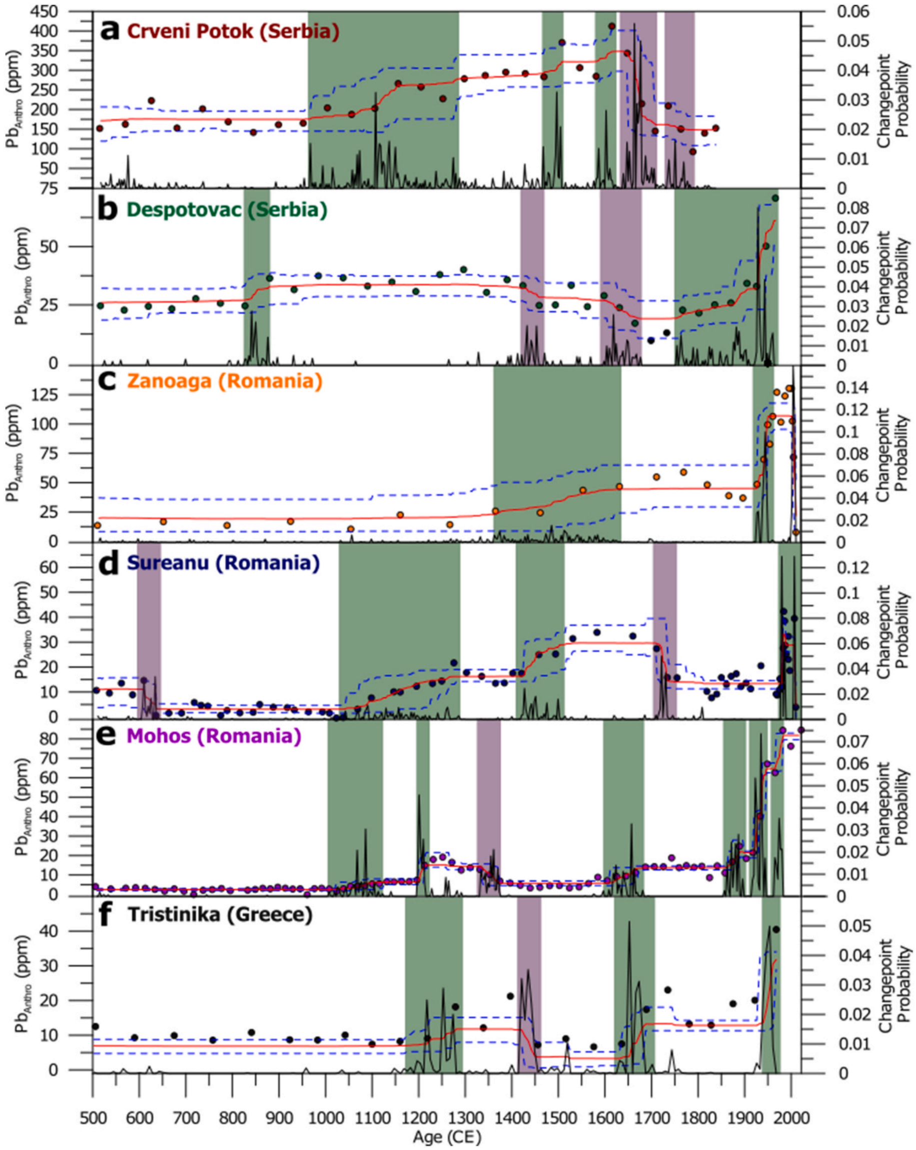

**Fig. 2. Downcore Pb pollution levels in the bogs studied here, along with the output of changepoint modelling.** Panel **a** is Crveni Potok (Serbia), **b** Despotovac (Serbia), **c** Zanoaga Rosie (Romania), **d** Sureanu (Romania), **e** Mohos (Romania) and **f** Tristinika (Greece). For each panel black circles indicate calculated anthropogenic lead (Pb$_{Anthro}$), with the model output shown by a red line indicating the mean and dashed blue lines the 5th and 95th percentiles. Changepoints are indicated by the solid black lines, with positive changes highlighted with green boxes and negative with purple. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

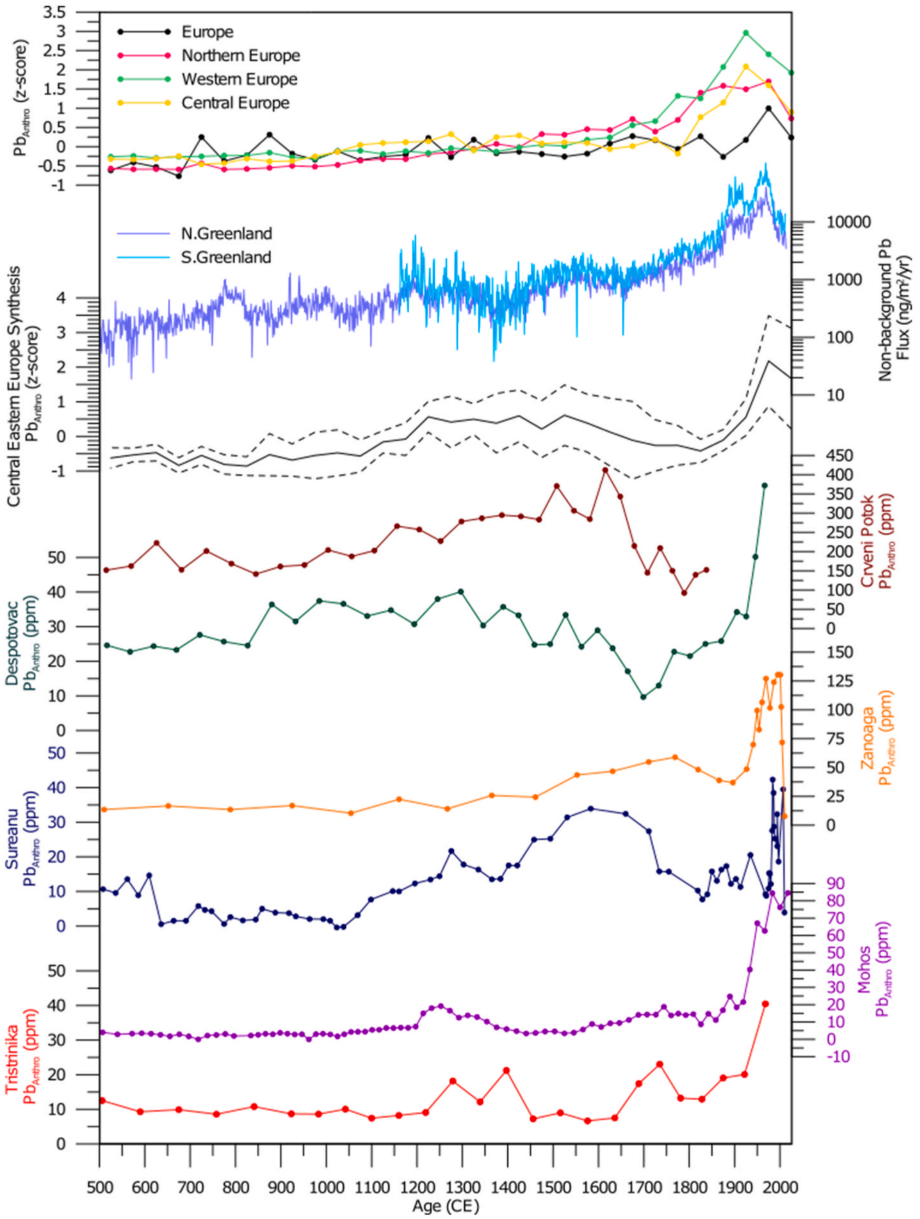

**Fig. 3. Comparison of Central-Eastern European records with syntheses of atmospheric pollution in Europe.** In the upper panel are compiled synthesis data of regional (grey lines) and Europe-wide (black line) Pb pollution (from Longman et al., 2020). In the second panel is data from Greenland, assumed to represent an integrated European Pb pollution signal (from McConnell et al., 2019), with potential contribution from North America (Pérez-Rodríguez et al., 2018). The third panel is the output of the synthesis of all anthropogenic Pb concentrations in the central-eastern European peat bogs studied here (see Methods and Materials for further information). The solid black line is the average for each 50-year bin, and the dashed lines represent the average ±1SD. Below are anthropogenic Pb values for all sites studied here.

Mohos (MOH) peat bog (46.050°N, 25.550°E, 1050 m a.s.l.) is located in the Eastern Carpathian Mountains (Fig. 1). It fills a large volcanic crater, related to volcanic activity from the Ciomadul volcano, with Mohos crater last eruption c. 60,000 years ago (Karátson et al., 2016). The surrounding geology comprises dacites, with occasional pyroclastic slope deposits, and thick soil coverage. The bog covers roughly 80 ha and the ca. 10 m thick peat is almost entirely *Sphagnum*-dominated (Longman et al., 2017b). Data used here have previously been published elsewhere (Longman et al., 2022).

Crveni Potok (CP) bog (43.914°N, 19.420°E, 1090m a.s.l.) is a small peatbog (c. 3 ha), located in the Tara National Park of western Serbia, close to the border with Bosnia-Herzegovina (Fig. 4). Approximately 270 cm of peat was recovered in overlapping cores, with the uppermost 100 cm comprising fibrous sedge/*Sphagnum* dominated peat. There is no permanent water inlet and so all nutrients are supplied by rainfall and runoff. The vegetation surrounding the mire is dominated by *Abies*, *Picea* and *Fagus* (Finsinger et al., 2017). The bedrock is massive and banked Triassic calcareous limestone, and with local Cretaceous marble and ultrabasic intrusions. We use previously published data from this site (Longman et al., 2018).

Zanoaga (ZNG) bog (45.176°N, 22.056°E, 1400 m a.s.l.) is a blanket bog c. 1 km$^2$ in area, located on the Semenic Mountains in the south-western part of the Romanian Carpathians (Fig. 4), with the *Sphagnum* peat deposit ca 200 cm thick. Yearly average precipitation averages 1400 mm, and the surrounding vegetation is dominated by *Fagus* and *Carpinus* forests. Bedrock consists of metamorphic (crystalline schist) and occasional plutonic (granite and granodiorite) intrusions (Cocoş, 1997).

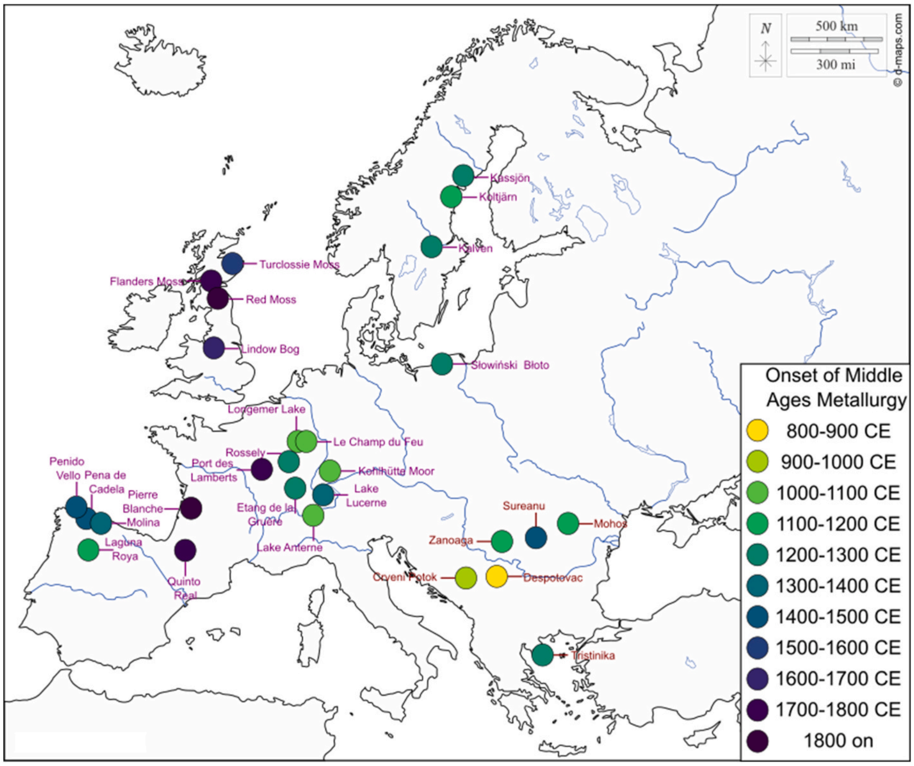

**Fig. 4. Timing of the onset of Middle Age metallurgy across Europe.** Coloured circles represent each of the sediment and peat records used in this study, with the fill relating to the timing of the first positive changepoint in the record for the period 800–1800 CE (see Methods). Studies analysed here are labelled with red writing, whilst those collated from literature are highlighted in purple. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Despotovac (DES) mire is located to the east of the town of Despotovac (44.094°N, 21.458°E, c.100m a.s.l.). It is a small wetland area (c. 5 ha) formed in a limestone depression, and local vegetation is dominated by grasses and sedges. Average yearly precipitation is roughly 600 mm, typically falling as snow between December and February. This wetland was cored in August 2015, with 4 m of sandy and silty sediment collected.

Tristinika marsh (39°55′55″N, 23° 52′ 22.8″E) is located on the peninsula of Sithonia, in northern Greece, roughly 120 km southwest of Thessaloniki (Fig. 1). The marsh is situated at just above sea level, and is periodically flooded (Zalidis and Mantzavelas, 1996). In June 2014, the marsh was cored close to its centre using a hand driven chamber corer, and 7 m of sediments were retrieved (Panajiotidis and Papadopoulou, 2016).

### 2.2. Trace metal analysis

For each record, the existing age models were used to select samples covering the past 1500 years. Samples from Despotovac, Sureanu, Crveni Potok, Zanoaga and Mohos were digested and analysed according to the methodology detailed in (Longman et al., 2018). Briefly, this involved a multi acid ($HNO_3$-HCl-HF) microwave-assisted digestion prior to analysis on a PerkinElmer Optima 8000 Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) at Northumbria University. Montana soil 2711 and peat standard NIMT/UOE/FM/001 were digested and analysed alongside samples, in addition to blanks. Recoveries of each standard and each element are detailed in Table S1. Samples from Tristinika were digested via a similar multi-acid process (HF-$HNO_3$-HCl) on a benchtop hotplate prior to analysis on a PerkinElmer 2000B Inductively Coupled Plasma Mass Spectrometer (ICP-MS) at the University of Oxford. Again, peat standard NIMT/UOE/FM/001 was run alongside samples, with recoveries in Table S2.

### 2.3. Chronology

For all cores, peat chronology was developed using $^{14}$C dating, typically performed on bulk peat samples, via accelerator mass spectrometry (see Table S3 for all dates used here and Figs. S1–S6 for all age models). Analyses were performed at either the HEKAL AMS Laboratory, MTA ATOMKI Institute for Nuclear Research of the Hungarian Academy of Sciences in Debrecen (Molnár et al., 2013), or at the $^{14}$C CHRONO centre at Queen’s University Belfast, with further methodology in Longman et al. (2021). Age-depth modelling was completed using the R package Bacon (Blaauw and Christen, 2011), using the IntCal20 calibration curve (Reimer et al., 2020). Outliers were removed when it is clear they represent reversals (see Table S3, Fig. S2).

The accuracy and precision of the age models varies from site to site, and the period of interest is typically the uppermost section of a deeper core (Figs. S1–S6). The best-dated site (Sureanu), has 8 dates covering the last 2000 years, whilst the least well-dated (Crveni Potok and Despotovac) have only 2 dates in this period. However, all have earlier dates which can be used to help guide the age depth modelling. Clearly, the sites with fewer dates have less precise age models, and so age uncertainty is a potential issue. For this reason, when discussing changepoint timing we round our estimates to the nearest 50 years, and group the timing of Middle Age metallurgy into hundred-year intervals.

### 2.4. Statistical methods

To remove the signal of natural input of base metals into the bog through detrital input, two calculations have been made; the Lithogenic/Anthropogenic contribution (Shotyk et al., 2000).

Such calculations are made using background (crustal) values (Rudnick and Gao, 2013), of Pb and the conservative, lithogenic element Zr via the following equation (Eq.1):

$$Pb_{Litho} = Zr_{sample} \times \left(\frac{Pb_{Crust}}{Zr_{Crust}}\right)$$

Equation (1)produces a Pb value which may be related to lithogenic processes ($Pb_{Litho}$). Therefore, the anthropogenic contribution ($Pb_{Anthro}$) may be determined as:

$$Pb_{Anthro} = Pb_{Total} - Pb_{Litho}$$

### 2.5. Changepoint analysis

To infer statistically where changes occur in the Pb datasets, we use a changepoint modelling approach (Gallagher et al., 2011; Longman et al., 2020), completed on the $Pb_{Anthro}$ data from our sites, and a compilation of other locations across Europe (see Table S4 for further details). We use these data to infer when the onset of Middle Age metallurgy occurred across the European continent. This was completed via analysis of the raw posterior probability of each changepoint model output for each site. The mean and standard deviation of this dataset (the changepoint probability) was calculated, and a cutoff of mean plus 2 standard deviations was used to indicate when statistically significant positive changepoints occurred (Table S4). This allows us to determine when the first significant rise in Pb occurred in the period 500–1800 CE, and determine variability seen in Middle Age metal pollution.

### 2.6. Central-Eastern European synthesis

To enable the interpretation of broad trends seen in all sites in this work, and to enable comparison to datasets which discuss extra-regional Pb pollution, we synthesise the $Pb_{Anthro}$ data (Fig. 3). For this, and following the method of Longman et al. (2020), we convert all data to z-scores to reduce the impact of absolute concentration variability between sites. These z-score data are then binned at 50-year resolution and averaged, and the standard deviations of each bin calculated.

## 3. Results and discussion

### 3.1. Contrasting developments in metal processing north and south of the danube in the middle ages

As shown in the Pb record of Crveni Potok bog in Serbia (Longman et al., 2018), and supported here by new Serbian data from Despotovac mire (Fig. 1), there is no evidence for cessation of smelting activities south of the Danube during the so-called ‘Dark Ages’ post 500 CE (Edmondson, 1989). This is the time corresponding to the shift of the Roman political and economic power from the Italian Peninsula to southeastern Europe controlled by the Byzantine Empire (Grig and Kelly, 2012). The Byzantine Empire at the time largely used the Danube as their northern frontier (De Frankopan, 1997; Stephenson, 1999), and our data suggest this north-south divide extended to regional shifts in metal processing as well, with increasing (or stable) Pb concentrations south of the Danube, and decreases in the north (Fig. 1).

Indeed, our reconstructions from peat records north of Danube in Romania (Mohos, Sureanu, Zanoaga; Fig. 1) show statistically significant negative changepoints (reflecting decreases in anthropogenic Pb content; see Methods) in anthropogenic Pb ($Pb_{Anthro}$) from ca. 400 CE to around 1000 CE (Fig. 2). This trend is more similar to the trend of Pb pollution reconstructed from records across Western Europe from Spain to Sweden (Brännvall et al., 1999a, 2001; Kylander et al., 2005; Martínez Cortizas et al., 2002). Locally it represents the reduction in mining activities associated with the end of the Roman Empire, and most likely the slow decline of regional ore extraction north of Danube during the Migration Period (Benea, 2008), a period of large-scale migration across the European continent driven by repeated invasions by various tribes (Benea, 2008).

### 3.2. The re-emergence of region-wide metal processing in the late middle ages

In all our records, there are several positive changepoints indicating statistically significant increases in anthropogenic Pb pollution from Romania to Greece spanning the period 800–1300 CE (Figs. 1 and 2). These represent evidence for the re-exploitation of metal ores in central and southeastern Europe (Fig. 1). The earliest of the positive changepoints are documented in Despotovac and Crveni Potok records, at 840 CE and 970 CE, respectively (Fig. 2), suggesting an increase in the intensity of local metallurgy (Longman et al., 2018) in the Balkan Peninsula. Across our Romanian sites, positive changepoints are reconstructed at c.1050 CE at Mohos and 1100 CE at Sureanu, clearly linked to re-activation of mining in Transylvania under the Kingdom of Hungary with evidence for mining in the Metaliferi Mountains (Maghiar and Olteanu, 1970), supported by $^{14}$C dating of mine timberworks at Rosia Montana to 775–1030 CE (1090 ± 60 BP) (Cauuet et al., 2003) (Fig. 2). In Greece, the southernmost of our sites, this rise in Pb does not occur until 1250 CE, suggesting asynchronous development in metal extraction development throughout the region.

It has long been considered that re-emergence of mining economic activities in southeastern Europe has been linked with knowledge brought by Saxon mining communities that had been settled in the mining areas of Hungary, Transylvania and Serbia in the 13th Century (Borcoş and Udubaşa, 2012; Fine, 1994; Stojković, 2013). Our data suggest however that many southeastern European mines were reopened prior to the arrival of Saxon miners, and that early metal processing activities in southeastern Europe developed alongside those in the Erzgebirge and Harz. Our data supplement the limited archaeological surveys in Romania which suggest a thriving metallurgical society at this time (Maghiar and Olteanu, 1970). This lack of clear archaeological evidence may be due to the signature of metal processing having been overprinted in archaeological records by later extractive activity, such as has occurred in the Erzgebirge region after Soviet exploitation of Late Middle Age mining locations (Meinrath et al., 2003; Wolkersdorfer, 1996), or at Rosia Montana in Romania during the communist time that destroyed extensive Roman and Medieval mining vestiges (Baron et al., 2011). Alternatively, the archaeological evidence has not yet been properly assessed.

Atmospheric Pb pollution across central-eastern Europe continued to rise until 1200 CE, when pollution levels began to level out in general (Fig. 1), but with a nuanced site-specific variability. This further indicates that arrival of Saxon miners supplemented rather than instigated early mining economic activities during the ‘Dark Ages’ in this region. Some archaeological evidence may support this conclusion, with evidence of the development of metal extraction across Transylvanian ore fields by the Kingdom of Hungary from the 12th Century CE onwards (Borcoş and Udubaşa, 2012; Maghiar and Olteanu, 1970). The first mentions of many northern Transylvanian Middle Age mining centres are made during middle 13th Century CE, but our data suggest active mining activities have been conducted prior (from 1100 CE onwards) to their first recording of mining towns in official documents (Zsamboki, 1985). Part of the reason for the development of Transylvanian metal processing is the opening of new silver and copper mines in the Baia Mare area in NW Romania (Borcoş and Udubaşa, 2012), with archaeological evidence of large-scale mining complexes in Bistrita (Rădulescu, 2004) and around the mining town of Rodna (Niedermaier, 2004).These mining regions were largely unaffected by the economic collapse induced by repeated Ottoman invasions to the southern Balkans and became some of the most important centres of extraction and processing of non-ferrous minerals in 14th Century Europe (Kacsó and Pop, 2010). In the 15th Century, driven by the demand for silver in western Europe, there was a crisis in the metal’s availability (Bailly-Maître, 2002; Pamuk, 2000). We suggest the high levels of Pb across our sites at this time reflect the exploitation of central-eastern silver mines to alleviate the crisis. This assertion is supported by archaeological evidence which suggests a number of Serbian mines opened at this time supplying the Venetians (Pamuk, 2000; Stojković, 2013).

### 3.3. Central-Eastern European metallurgy after 1600 CE

Following the persistent Late Middle Age Pb enrichment, a series of negative changepoints across all our records may be observed in the 17th Century at Sureanu, Despotovac and Crveni Potok (Fig. 1). These negative changepoints attest to the impact of Ottoman wars of occupation, which led to political unrest and economic collapse across the Balkans and central-eastern Europe (Longman et al., 2018; Stojković, 2013). For example, the conquest of the Balkans led to a complete cessation of metallurgical activities across the region (Hategan, 2005). Across Transylvania in the period 1600–1800 CE, and especially after c.1650 CE, there was a significant decrease in mining activities (Wollmann, 1999). This decrease in metalworking, and associated $Pb_{Anthro}$ decrease across most our records, continues until the first positive changepoints associated with the Industrial Revolution are observed, from 1800 CE onwards (Figs. 1 and 2). This rise is most dramatic in the later 19th Century (Fig. 1), linked to the interest in mining of the Habsburg administration (Wollmann, 1999). Under the Habsburgs, a coherent policy of mining and quarrying was established, allowing metal production to resume (Rădulescu, 2004; Wollmann, 1999), particularly in Transylvania as reflected in the slow rise in $Pb_{Anthro}$ across all sites after 1800 CE (Figs. 1 and 2). A final peak in Pb pollution is observed in the second half of the 20th Century CE (Fig. 1), with some of the best expressed positive changepoints. This is linked to the appearance of leaded gasoline (petrol containing added tetraethyl Pb), which became the largest source of lead pollution in developed countries after its adoption in the 1930s (Kovarik, 2005; Landrigan, 2002).

### 3.4. Comparison of records at European scale

Our results provide a large amount of data on palaeopollution for regions of Europe previously unstudied, hinting at the development of a strong local culture of metal extraction before the onset of the Middle Ages in southeastern Europe. For this we synthesise the PbAnthro data collected from all sites (Fig. 3), and compare to two syntheses of European Pb pollution, first an integrated ice core record (McConnell et al., 2019), and secondly from a set of Europe-wide bog records (Longman et al., 2020) (Fig. 3) We also compare our results with a number of regionally important records of palaeopollution to construct a spatial analysis of Middle Age pollution (Fig. 4). Comparison to Europe-wide syntheses clearly demonstrates how Central-Eastern Europe diverges in the Middle Ages (Fig. 3), with Pb pollution appearing earlier, and the peak lasting longer than in western Europe. We discuss the emergence of Middle Age pollution in further detail below, but the scale and extent of the Pb peak in the period 1200–1700 CE is of particular interest.

As discussed above, this peak is likely linked with the development of Hungarian and Transylvanian mining activities in the Baia Mare region, in combination with the re-emergence of large-scale mining in the Apuseni Mountains. Similar features may be observed in records from central Europe, previously considered as centres of Middle Age metallurgy and suggest enhanced metallurgical activities in south-eastern Europe also occurred in the period 1200–1700 CE (Fig. 3). A measure of the level of regional pollution at this time may be gleaned via comparison of Middle Age pollution levels to those in the era of leaded gasoline, thought to be the period of greatest Pb pollution (Shotyk et al., 2016; Weiss et al., 1999b). At Sureanu, a high-altitude bog located c. 220 km from the Baia Mare region and 100 km from the Apuseni Mountains, the levels of Pb in the 17th century CE are similar in scale to levels observed during the latter half of the 20th Century, when the use of leaded gasoline resulted in intense atmospheric Pb pollution (Figs. 1 and 3). As a result, the scale of metalworking in the Baia Mare region and the Apuseni Mountains must have been considerable. Economic geology studies do suggest southeastern Europe as one of the most significant metalworking regions in Europe also during the Middle Ages, with periodic increased metal output to offset the reductions or stagnations occurring in many central European locations during this time (Longman et al., 2018; Stojković, 2013).

A second major feature of the southeastern European records is that there is little evidence for metal pollution linked to the early Industrial Revolution (Figs. 1 and 3). In the Pb syntheses (Hansson et al., 2015; Longman et al., 2020; Renberg et al., 2001), there is a gradual rise in Pb pollution after the Middle Ages, with data from Greenland showing little evidence for a decrease in Pb deposition after 1600 CE (Fig. 3). In contrast, at Serbian and Romanian (although not Zanoaga) sites there is a marked decline from 1650 CE, with especially Sureanu and Crveni Potok detailing very low levels of Pb pollution until roughly 1900 CE. This is despite the development of widespread metallurgical technology such as bloomeries in 1619 CE and the blast furnace from 1670 onwards (Tylecote, 2002). Our data details the negative influence of laws and taxes in eastern Europe enacted by the Ottoman Empire (Stojković, 2013), as well as collapse of the mining sector in these regions following the discovery of Americas and input of cheaper overseas metals. Clearly the industrial revolution lagged significantly in central-eastern Europe compared to western Europe, with the majority of developments, such as the widespread adoption of coal use (McConnell and Edwards, 2008), led by the British and other western European powers.

### 3.5. Where did middle age metal pollution begin in Europe?

The clearest difference between existing Pb syntheses (1, 13) and our data is the emergence of a large Early Middle Age peak at c.1000 CE in several of our central and south-eastern European records. It is presumed that Middle Age metallurgy re-emerged in central European sites such as the Harz mountains in Germany (Asmus, 2012; Kempter and Frenzel, 2000). By compiling Pb records from across Europe and determining the first statistically significant (see Methods) positive changepoints in the period 500–1800 CE in anthropogenic Pb content, we can reconstruct the spatial development of Middle Age metallurgy's emergence.

This exercise indicates that the only locations with statistically significant positive changepoints prior to 1000 CE are located in the Serbian records in Crveni Potok and Despotovac (Fig. 4), at c.800 and c.1000 CE, respectively. This is despite previous work having suggested Middle Age pollution began with the exploitation of the Melle mines of France during the reign of Charlemagne from 620 CE (Téreygeol, 2013), and of mining activity in Derbyshire (Blanchard, 2005), UK from 780 CE onwards. We see no evidence for the influence of any of this activity, even in locations close to the purported metalworking (Fig. 4), suggesting mining activities in southeastern Europe were much more pervasive at this time than in the west, supporting the assertion that central-eastern European mines would have been available to alleviate the silver crisis in coming centuries (Pamuk, 2000). Our data also suggest that large-scale metallurgy never ceased in southeastern Europe during the 'Dark Ages' as compared with the first positive changepoints in German locations in the Black Forest at c.1050 CE (Fig. 4). These occur at a similar time to the onset of metal pollution in the Vosges mountains in central-eastern France, suggesting a locus of metallurgy developed in this region in the 11th Century (Mariet et al., 2018a, 2018b).

Interestingly, for the period 1100–1300 CE, positive changepoints are seen in Romanian locations and those in central Europe such as Switzerland (Fig. 4), along with a single location in north-western Spain and in Sweden (Bindler et al., 2009; Brännvall et al., 1999b). The spatial variability of these changepoints suggests that Middle Age pollution did not radiate, and that there were multiple locations of development at the onset of the period. It may be that some Romanian sites were opened and developed by Saxon miners arriving after 1250 CE (Longman et al., 2018; Stojkovic, 2010), but this does not explain how Serbian locations show pre-Saxon mining development, and how Spanish records occur prior to any influence being possible from central Europe. The remainder of central European and Iberian sites indicate Middle Age pollution began before 1400 CE, in line with archival data which suggests the widespread adoption of silver smelting (McConnell et al., 2019), a boom in population and widespread iron and lead smelting (Brännvall et al., 1999a; McConnell et al., 2019).

The currently accepted view of metallurgical development in the Middle Ages as being led only by mining activities in central or western Europe is not supported by our work. In contrast, we suggest metallurgical technological development was a much more complex process, with multiple locations of extractive metallurgy emerging continent wide from 800 to 1200 CE (Fig. 4). To reach this conclusion, our work provides data for a region which was previously uninvestigated for palaeopollution in environmental archives. By analysing 6 bogs from Serbia, Romania and Greece, we are able to demonstrate that metallurgical technology in central-Eastern Europe did not develop at the same time as in western Europe. By investigating new bogs and via comparison to numerous extant records, it is possible to demonstrate the regional nature of environmental Pb records. Our findings highlight how more research is required to fill gaps in the palaeopollution record, both spatially and temporally.

## CRediT authorship contribution statement

**Jack Longman:** Writing – review & editing, Writing – original draft, Visualization, Resources, Project administration, Methodology, Investigation, Formal analysis, Conceptualization. **Daniel Veres:** Writing – review & editing, Writing – original draft, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Vasile Ersek:** Writing – review & editing, Writing – original draft, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Calin G. Tamas:** Writing – review & editing, Writing – original draft, Methodology, Formal analysis. **Aritina Haliuc:** Writing – review & editing, Writing – original draft, Methodology, Investigation. **Eniko Magyari:** Writing – review & editing, Writing – original draft, Investigation. **Florin Gogaltan:** Writing – review & editing, Writing – original draft, Resources, Methodology, Formal analysis. **Sampson Panajiotidis:** Writing – review & editing, Writing – original draft, Resources, Methodology, Investigation. **Maria Papadopoulou:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis.

## 4. Open research

All data are available in the main text or the supplementary materials.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jas.2024.106093.

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