

# Iron Oxides Prehistoric Mines A European Overview

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## Résumé

Cette recherche offre une synthèse sur les anciennes exploitations d'oxydes et d'hydroxydes de fer connus sur le continent européen. L'objectif est de synthétiser les informations publiées pour chaque site. Les sites préhistoriques d'extraction de ces minéraux sont rares. En Europe, on compte cinq mines préhistoriques d'oxydes de fer connues : Tzines (Grèce), Rydno (Pologne), Lovas (Hongrie), Bad Sulzburg (Allemagne) et Grotta della Monaca (Italie). Ces cinq mines couvrent une période entre la phase finale du Paléolithique supérieur et le Néolithique final. Ces mines, qui ont survécu aux destructions liées aux exploitations ultérieures, donnent un aperçu de la distribution géographique et du cadre chronologique de cette activité, de même qu'elles livrent des informations sur les techniques d'extraction et les outils utilisés. Par contre, en l'absence de liens avec des habitats bien identifiés, l'état actuel des recherches ne permet pas de comprendre l'organisation sociale et économique des groupes qui ont développé ce type d'exploitation, exception faite du site de la mine de Rydno. Malheureusement, peu de choses sont connues à ce jour sur le traitement de la matière première après son extraction. Seule une phase de broyage direct in situ ou à proximité du site d'extraction a pu être observée. Un nombre important de sites en grotte et de plein air livrent des évidences de l'utilisation d'oxydes de fer comme pigments, abrasifs et agents siccatifs. D'autres hypothèses sont possibles mais, en l'absence de preuves directes, elles sont tirées de sources historiques et de comparaisons ethnographiques : peinture du corps, travail de la peau, conservation de substances organiques, remèdes médicaux, entre autres. Quoiqu'il en soit, l'importance évidente de ces matériaux durant la Préhistoire laisse à penser que les sources d'approvisionnement étaient très activement recherchées.

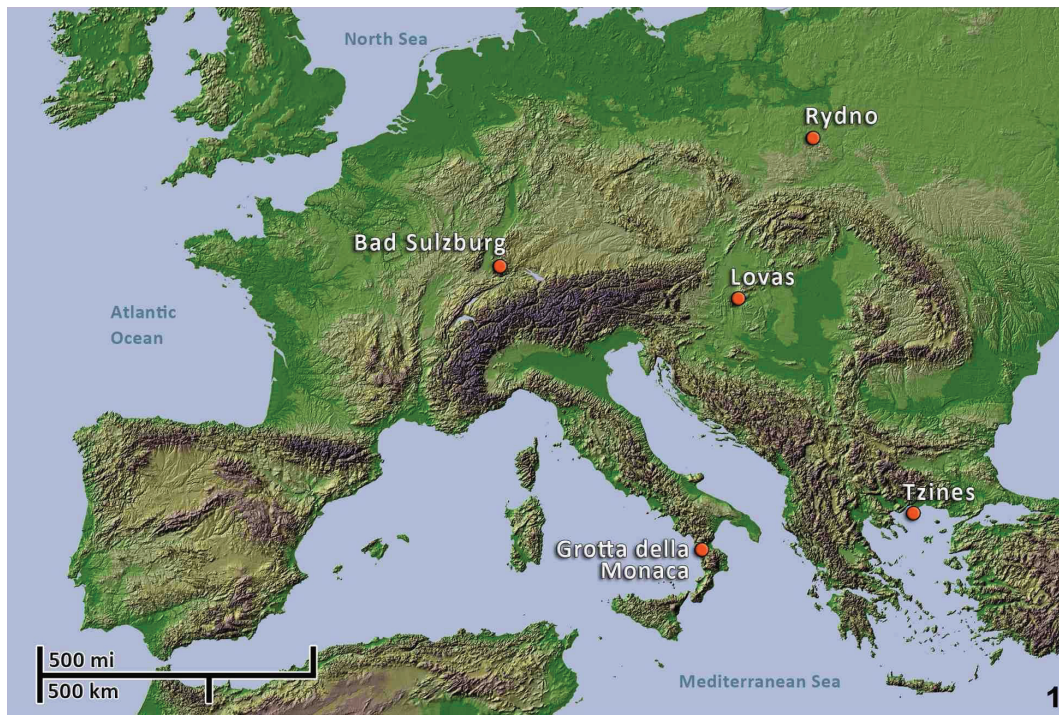
**Mots-clés :** Minières, mines, oxydes de fer, Préhistoire, Tzines (Grèce), Rydno (Pologne), Lovas (Hongrie), Bad Sulzburg (Allemagne), Grotta della Monaca (Italie), Europe.

**Keywords:** Mines, Iron oxides, Prehistory, Tzines (Greece), Rydno (Poland), Lovas (Hungary), Bad Sulzburg (Germany), Grotta della Monaca (Italy), Europe.

## 1. INTRODUCTION

Iron oxides and hydroxides are among the most widespread chemical compounds in nature. They are composed of Fe combined with O, in the case of iron oxides, or OH, in the case of iron hydroxides. Nowadays, they are used mainly as ores in iron and steel industry and as pigments (Cornell & Schwertmann, 2003: 1-2, 511-524). Some can be obtained by thermal transformation: for example maghemite ( $\gamma\text{Fe}_2\text{O}_3$ ), which can derive from the heating of some iron oxides associated with organic matters, or hematite ( $\alpha\text{Fe}_2\text{O}_3$ ), that can be produced by heating goethite [ $\alpha\text{FeO}(\text{OH})$ ] at a temperature between 250°C and 300°C. Typical acicular nano-porosities caused by a dehydration process allow to distinguish natural hematite from the heated one (Cornell & Schwertmann, 2003: 6-7, 369-373; Pomiès *et al.*, 1998).

These minerals are often connected with the term *ochre*. Ochre is a generic word that has not an univocal and received definition. In literature it is often described as any colouring matter varying in colour from yellow to brown, without any reference to its mineralogical composition. One useful definition is suggested by Elias, Charrière, Prévot, Garay and Vignaud (2006: 70-71) for commercial natural ochres. The authors state: "Ochres contain varying amounts of octahedral iron oxides, namely hematite ( $\alpha\text{Fe}_2\text{O}_3$ ) or/and goethite [ $\alpha\text{FeO}(\text{OH})$ ], and of white pigments (alumino-silicate as kaolinite or illite, quartz and calcium compounds as calcite, anhydrite, gypse or dolomite). When the hematite is the main iron oxide, a red colour is observed, whereas the ochre is yellow when the goethite dominates". This specific description is useful because it underlines the heterogeneous mineral composition



**Fig 1** – 1. Geographical setting of the European prehistoric iron oxides mines (drawing by F. Breglia © 2013); 2. Entrance of Tzines “T1” mine (Thasos Island, Greece) (Image courtesy of the Deutsches Bergbau Museum Bochum); 3. Mining stone tools from the Tzines “T1” mine (Thasos Island, Greece) (Images courtesy of the Deutsches Bergbau Museum Bochum, modified).

**Fig 1** – 1. Localisation géographique des mines préhistoriques d’oxydes de fer en Europe (dessin par F. Breglia © 2013); 2. L’entrée de la mine “T1” de Tzines, dans l’île de Thasos (Grèce) (Droit de reproduction du Deutsches Bergbau Museum de Bochum); 3. Outils de mineur en pierre provenant de la mine “T1” de Tzines (île de Thasos, Grèce) (Droit de reproduction du Deutsches Bergbau Museum de Bochum, modifié).



of ochres and links its colour with the presence of iron oxides, clay minerals and carbonates.

The use of iron oxides, especially hematite, and ochres in general has been largely attested worldwide within numerous prehistoric sites. However, taking into account all the activities connected with the production cycle of iron minerals – used for different purposes from smelting ones – it is possible to observe the rarity of the hitherto known sites in which extraction activities, essential for the subsequent phases of mineral processing, were performed.

This paper offers a concise summary of a wider work about European and non-European iron oxides prehistoric mines known in literature, raised after some archaeological investigation carried out on the iron hydroxide Neolithic exploitation of Grotta della Monaca, a cave located in southern Italy (Levato, 2012). The aim is to summarize in a single contribution the information published for each site known in Europe, focusing the attention on geographic distribution, geological setting, chronology, mining techniques, and – where it has been possible – on mineralogical analysis and socio-economic organization.

## 2. IRON OXIDES PREHISTORIC MINES

In Europe are known at least five iron oxides prehistoric mines, concentrated in the southern and north-eastern European belts: Tzines (Greece), Rydno (Poland), Lovas (Hungary), Bad Sulzburg (Germany), and Grotta della Monaca (Italy) (Fig. 1:1). These five mines cover a time span ranging from the final phase of the Upper Palaeolithic to the end of Neolithic Age.

The site of Tzines is an extraordinary example of underground mine, the most ancient one today known in Europe. It is located in Thasos, an island of the northern Aegean Sea, close to the coast of East Macedonia and Thrace. The site sets in an area with rich iron-manganese deposits under the Ais Matis-Kastrou marble horizon. Iron ore is deposited erratically in the marble rock. Hematite contains small hollows in which iron oxidised and on whose surface the iron oxide weathered in powdery form is deposited (Koukouli-Chrysanthaki & Weisgerber, 1999: 130-131). Archaeological investigations were carried out between 1982

and 1993 by the 18th Ephorate of Prehistoric and Classical Antiquities of Kavala, represented by Chaido Koukouli-Chrysanthaki, in collaboration with Gerd Weisgerber of the Deutsches Bergbau Museum of Bochum and Georgios Gialoglou of the Greek Institute of Geology and Mineral Exploration. Several mining sites were discovered, four of which were excavated. The best investigated ones are the underground mines called “T1” and “T2”. Prehistoric miners exploited powder hematite starting from the surface and then deepened the extraction through underground mining (Koukouli-Chrysanthaki & Weisgerber, 1999: 129-134). The T1 mine is composed of two galleries, 11 metres in length on the whole, with variable height and width (Fig. 1:2). In this site twenty-two layers, distinct by very hard upper surfaces, reflect different phases of frequentation with time gaps between one mining period and the next one. The surfaces compactness is a consequence of the deposition of waters rich in carbonates percolating from the roof and the weight of the bodies of prehistoric miners that crawled along the central axis of the cavity. Miners left *in situ* no longer useful tools placing them alongside the cave walls. The deposit provided over 500 finds made both of hard animal material, as antlers, horns, ribs, and of stone, such as flint and especially pebbles. According to Koukouli-Chrysanthaki, Weisgerber, and Cierny (Koukouli-Chrysanthaki & Weisgerber, 1999; Weisgerber *et al.*, 2008), antlers – mainly of *Cervus elaphus* – were employed as wedges or chisels, leaving digging traces on the mine walls, whilst pebbles – mainly made of marble and gneiss without any manufacturing traces – were used as hammers without handle and probably were collected in the neighbouring streams (Fig. 1:3). Other tools such as bone spatulas and flint blades might have been used respectively to collect and scrape hematite. The T2 mine is composed by a chamber 3 x 4 metres and by three small branches. A different extractive technique has been recognized within it. It was based on the direct percussion of the rock walls with the exclusive use of handheld pebbles equally lacking any manufacturing traces (Koukouli-Chrysanthaki & Weisgerber, 1999: 131-134; Weisgerber *et al.*, 2008). The lack of pottery, charcoal, and diagnostic flint tools made the dating on this context very difficult. However, two radiocarbon dates have been carried out on finds coming from T1 mine: the first, executed measuring C14 levels of deer antlers, has yielded a date

of around 6400 years BC (HD-8528-8509); the second, on an animal bone, has been realized by accelerator mass spectrometer (AMS), giving a dating of  $20350 \pm 160$  years BP (ETH-11573) (Tab.1). This last one is confirmed by the presence of a *Saiga tatarica* horn which dates back to

the Late Glacial when Thasos was still attached to the mainland (Koukouli-Chrysanthaki & Weisgerber, 1999: 135-136; Weisgerber *et al.*, 2008). Unfortunately, the interruption of the research does not allow adding other information, even if a new series of studies is to be hoped.

<i>Lab. no</i>	<i>Material</i>	<i>Context</i>	<i>Radiocarbon dating</i>
ETH-11573	Bone	Tzines T1 mine	$20350 \pm 160$ BP
Gd-2002	Charcoal	Rydno Quarry I/1980	$19280 \pm 280$ BP
Gd-4306	Charcoal	Rydno Quarry I/1980 Magdalenian (?)	$12960 \pm 210$ BP
Gd-725	Charcoal	Rydno Quarry III/1979 Hamburghian/Magdalenian (?)	$12290 \pm 210$ BP
Bln-2037	Charcoal	Rydno Quarry I/1977 Hamburghian (?)	$11970 \pm 125$ BP
Gd-724	Charcoal	Rydno Quarry III/1979 Hamburghian (?)	$11940 \pm 300$ BP
Gd-713	Charcoal	Rydno Quarry III/1979 Arch Backed Piece	$10910 \pm 220$ BP
Gd-714	Charcoal	Rydno Quarry III/1979 Arch Backed Piece/Masovian (?)	$10710 \pm 250$ BP
Gd-710	Charcoal	Rydno Quarry I/1977 Masovian	$10360 \pm 320$ BP
Gd-719	Charcoal	Rydno Quarry III/1979 Masovian (?)	$9840 \pm 370$ BP
MAMS-21718	Bone	Lovas Mine	$11941 \pm 44$ BP
MAMS-21719	Bone	Lovas Mine	$11918 \pm 41$ BP
MAMS-21720	Bone	Lovas Mine	$11825 \pm 41$ BP
ETH-15199	Bone	Lovas Mine	$11740 \pm 100$ BP
MAMS-21722	Bone	Lovas Mine	$11728 \pm 46$ BP
MAMS-21721	Bone	Lovas Mine	$11469 \pm 40$ BP
Beta-105503	Charcoal	Bad Sulzburg Mine	$6250 \pm 60$ BP
Beta-105505	Charcoal	Bad Sulzburg Mine	$6180 \pm 60$ BP
Beta-105504	Charcoal	Bad Sulzburg Mine	$5810 \pm 60$ BP
LTL-3583A	Charcoal	Grotta della Monaca/Ramo delle vaschette	$5183 \pm 50$ BP
LTL-3584A	Charcoal	Grotta della Monaca/Ramo delle vaschette	$5010 \pm 50$ BP
LTL-3582A	Charcoal	Grotta della Monaca/Buca delle impronte	$4935 \pm 45$ BP
LTL-3581A	Charcoal	Grotta della Monaca/Buca delle impronte	$4880 \pm 45$ BP

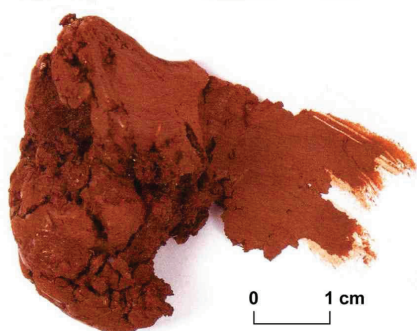
**Tab. 1** – Uncalibrated radiocarbon dating acquired on charcoal and bones from the mines of Tzines, Rydno, Lovas, Bad Sulzburg and Grotta della Monaca (from Dobosi, 2006; Goldenberg *et al.*, 2003; Larocca, 2010; Sajó *et al.*, 2015; Schild *et al.*, 2011; Weisgerber *et al.*, 2008).

**Tab. 1** – Datations radiocarbones non calibrées sur charbon de bois ou os issus des mines de Tzines, Rydno, Lovas, Bad Sulzburg et Grotta della Monaca (d'après Dobosi, 2006 ; Goldenberg *et al.*, 2003 ; Larocca, 2010 ; Sajó *et al.*, 2015 ; Schild *et al.*, 2011 ; Weisgerber *et al.*, 2008).



Rydno is the only mine for which it was possible to reconstruct exhaustively the socio-economic context, thanks to the data coming from flint assemblage and the presence of campsites surrounding the mining area. This site is located along the northern foothills of the Holy Cross Mountains, in the Kamienna River Valley (central and southern Poland). The river cuts into Triassic and Liassic sandstone as well as fluvioglacial sand and gravel deposited during the maximal phase of the Odra Glaciation (Early Saale). On the top of the so-called "Łyżwy-Nowy Młyn Hill" the Quaternary deposits were removed by slope processes linked to the Last Glacial Maximum. Here sandstone and red Triassic clay outcrop under a thin soil, while on the northern and southern hill-sides slope deposits thicken downwards interbedding with redeposited red clay. The quarry area is located just on the north-eastern hillside. Here a thin slope deposit covers beds of polygenetic red conglomerate – up to 1 metre – interbedded with fine grained Variegated Sandstone. Conglomerate is composed by quartz gravel sand and pebbles mixed with hematite, chert, sandstone, igneous gravel in a red clayey matrix. The Variagated Sandstone is considered a local variant of the Upper Variegated Sandstone of the Upper Roethian (Lower Triassic). According to their genesis, this sandstone formed in shallow sea, conglomerate in beach zone, while hematite gravel and pebbles enclosed in the conglomerate derive from an older eroding seashore (Schild *et al.*, 2011: 49-50, 55). The mining area was discovered by Stefan Krukowski in 1945 but it was excavated, after some time, between the 1970s and 1980s, by the Department of Stone Age of the Institute of Archaeology and Ethnology, Polish Academy of Science, under the direction of Romuald Schild (Schild *et al.*, 2011: 46-48). In an area of 110 x 30-40 metres a series of wide open pits, different in size, were dug by prehistoric miners into the beds of conglomerate (Schild *et al.*, 2011: 19, 55; Fig. 2:1-2). Quantitative spectrographic analysis carried out on hematite gravel show that it contains between 45 % and 70.12 % by weight of Fe, while structural X-ray analysis show that it is composed by hematite and lepidocrocite [indicated respectively as  $\text{Fe}_2\text{O}_3$  and  $\text{FeO}(\text{OH})$ ] (Hensel, 2011; Tab. 2, Tab. 3). Nine radiocarbon dating set mining activities between  $19280 \pm 280$  (Gd-2002) and  $9840 \pm 370$  (Gd-719) years BP (around 23000-11500 years calBP), that

is between shortly after the Last Maximum Glacial and the Early Holocene (Tab.1) (Schild *et al.*, 2011: 57-58). None of the evidence can be associated to the oldest date. Flint tools from the quarry are related with certainty at least to two major taxonomic units, the Arch Backed Piece and the Younger Tangent Point (Masovian) technocomplexes, that are also the most numerous and best represented in the surrounding campsites. They include in many cases blanks, cores and rare retouched tools, similar in shape to the campsites ones. This attests that camping and living activities were carried out alongside hematite and red clay mining. Furthermore, gravitational concentrations of charcoal at the pits bases suggest that miners did not refill them immediately after their exploitation (Schild & Królik, 1981: 63-69; Schild *et al.*, 2011: 52, 57-58, 101, 400). Arch Backed Piece technocomplex is set in the Allerød, a temperate phase dominated by taiga forests in Central Poland (Greenland Interstadial 1c-1a, 13950-12700 years calBP) (Schild *et al.*, 2011: 394, 399). The Arch Backed Piece groups of Rydno located their camps above all south-east of the mine, forming multiunit aggregations with several basin huts. Raw material assemblage shows the main use of Upper Oxfordian so-called "chocolate" flint, coming from the western section of the chocolate belt (located about 20 kilometres from the site), and of Jurassic flint from the Cracow-Częstochowa Upland. Other small, single camps around the mine show a nearly exclusive use of erratic chocolate flint collected from the surface, maybe due to the poor territorial knowledge or the lacking access to the nearest flint sources. This dichotomy may reflect a distinction within the two groups, the "controllers" and the "users" of the mine. The large communities of "controllers" needed supplementary resources of goods, including foodstuff, and this was possible through a monopolistic control over the mine, allowing its access or bartering hematite and red clay (Schild & Królik, 1981: 77-81; Schild *et al.*, 2011: 72, 223, 394-395, 400-401; Sulgostowska, 2006: 36). The Masovian phase occupies in Poland a time span between the beginning of the Younger Dryas and the end of the Preboreal Oscillation (Greenland Stadial 1 - Greenland Holocene 11.2 event, 12700-11200 years calBP) (Schild *et al.*, 2011: 395, 399). Park tundra and birch-pine woods covered the plain of central Poland. Seasonal nomadic movement connected with reindeer and



**Fig. 2** – 1. Archaeological excavation within the mining area of Rydno, Poland (Cut 1/1977, Pit 1 and Pit 2) (from Schild et al., 2011: fig. 4.5); 2. Red clay and hematite gravel from the mining area of Rydno, Poland (from Schild et al., 2011: fig. 4.7, modified); 3. *Alces alces* ulna, on the left, and bone point, on the right, from the so-called “Pit 2” of Lovas, Hungary. The ulna is 209 mm long; the bone point is 132 mm long, with a maximum width of 9.5 mm (from Mészáros & Vértes, 1959: 9, 18, fig. 11, modified).

**Fig. 2** – 1. Fouilles archéologiques à l’intérieur de l’aire minière de Rydno, Pologne (Cut 1/1977, Puits 1 et Puits 2) (d’après Schild et al., 2011 : fig. 4.5) ; 2. Argile rouge et gravier d’hématite de l’aire minière de Rydno, Pologne (modifiée d’après Schild et al., 2011 : fig. 4.7) ; 3. Cubitus de *Alces alces*, à gauche, et pointe en os, à droite, du « Puits 2 » de Lovas, Hongrie. Le cubitus mesure 209 mm de long ; la pointe mesure 132 mm de long, avec une plus grande largeur de 9,5 mm (modifiée d’après Mészáros & Vértes, 1959 : 9, 18, fig. 11).

Rydno - Spectrographic Analysis								
No	Sample no	Percentages by weight						
		Fe	Si	Mg	Ca	Al	Ti	Mn
1	6570 A	70.12 50.76	12	2.0	0.38	1.8	0.44	0.02
2	6570 B	64	12	2.0	0.30	2.2	0.60	0.003
3	6571 A	66	14	2.0	0.27	2.2	0.72	0.09
4	6571 B	68	8	1.4	0.28	1.7	0.20	0.05
5	6572 A	52	11	1.5	0.25	2.0	0.51	0.03
6	6572 B	45	17	1.3	0.25	2.5	0.52	0.02

**Tab. 2** – Hematite gravel samples from Rydno: results of spectrographic analysis as determined by Classical Chemical Methods (from Hensel, 2011: 406, tab. 2).

**Tab. 2** – Échantillons de gravier d'hématite de Rydno : les résultats de l'analyse spectrographique par méthodes chimiques classiques (d'après Hensel, 2011 : 406, tab. 2).

horse hunting was the main mode of life. The emergence of this group at Rydno is characterized by large Upper Oxfordian chocolate flint processing workshops. Small units formed camps, located near the southern and northern bank of the Kamienna River, and show the use of chocolate flint coming from distinct sections of the outcrop. The sites placed near the Kamienna fords certainly lied in a strategic position and controlled the mine to a certain extent. The different access to flint sources is thought a result of social, ethnic, or territorially distinct bands occupying each bank of the Kamienna River (Schild & Królik, 1981: 81; Schild *et al.*, 2011: 76, 395, 399, 401). The “last revival” of Rydno occurred during the Late Mesolithic Vistulian (Janislavician) phase (Early Atlantic, 8200-7500 years calBP) (Schild *et al.*, 2011: 402). Subsequent Neolithic and Bronze Age occupations indicate short-lived campsites not necessary associated with hematite exploitation (Schild *et al.*, 2011: 350, 372-388). Chocolate flint from the Holy Cross Mountain was exported hundreds kilometres in the territory of Arch Backed Piece and Masovian groups. Hematite from Rydno was most likely exported in the Late Glacial campsites of Całowanie, 100 kilometres to north-north-east (Hensel, 1981: 100; Hensel, 2011: 407; Schild *et al.*, 2011: 403; Sulgostowska, 2006). According to Schild *et al.* (2011: 403), since “ochre” sources

are less widespread than those of flint, hematite gravel and red clay are the goods for which enormous aggregations of settlements were formed at Rydno. This can explain the presence of raw material coming from faraway lands, such as radiolarite from Pieniny Mountains (southern Poland) or from Slovakia, Cretaceous flint from Volhynia, Ukraine, and Podlaskia (eastern Poland), obsidian from southern Slovakia or Hungary (Schild & Królik, 1981: 67; Schild *et al.*, 2011: 78; Sulgostowska, 2006: 38, 40). As stated by Schild *et al.* (2011: 58), the date obtained for the Arched Backed Piece mining activities of Rydno is similar to that of Lovas mine.

The Lovas mine is located in the homonym municipal district, south of the Bakony Mountains and north of the Balaton Lake, in the Veszprém County (central and western Hungary). The site sets in a Triassic dolomitic plateau 280-290 metres above sea level that borders the southern limb of the Bakony Mountains. The first archaeological excavation was carried out between 1951 and 1952 by Gyula Mészáros of the Veszprém County Museum and László Vértes of the Hungarian National Museum. Their research revealed the existence of prehistoric mining activities dated to the Middle Palaeolithic, making Lovas at that time the most ancient “ochre” mine of Europe. In 1977



<i>Rydno - Structural X-Ray Analysis. Identification characteristic of hematite and lepidocrocite as compared with radiographic data</i>						
No	$d_{hkl}$ Actual	<i>I</i>	$d_{hkl}$ Fe <sub>2</sub> O <sub>3</sub>	<i>I</i>	$d_{hkl}$ FeOOH	<i>I</i>
1	6.1	100			6.25	100
2	3.64	20	3.66	10		
3	3.30	20			3.29	80
4	2.70	80	2.69	70		
5	2.51	60	2.51	40	2.46	70
6	2.20	60	2.18	20		

**Tab. 3** – Hematite gravel samples from Rydno: identification of phase composition by structural X-ray analysis as compared with radiographic data (from Hensel, 2011: 406, tab. 3).

**Tab. 3** – Échantillons de gravier d'hématite de Rydno : l'identification de la composition de phases par des analyses structurelles aux rayons X (d'après Hensel, 2011 : 406, tab. 3).

Viola T. Dobosi and István Vörös of the Hungarian National Museum carried out further surveys to find new prehistoric mines, but without success because modern mining, active up to 1960, probably destroyed other further evidence. Therefore, they concentrated their attention on the revision of the chronology and of the faunal remains discovered during the Fifties. Recent study, published in 2015, has shedded new light on radiocarbon dating, ochre mineral composition and ochre processing. Three open pits – “Pit 1”, “Pit 2” and “Pit 3” – differing in size were dug by prehistoric miners within the Triassic dolomitic rock containing red beddings (Mészáros & Vértes, 1959: 1-7; Dobosi, 2006: 29-30; Sajó *et al.*, 2015). According to mineralogical analysis carried out in the Fifties (analytical method unspecified) the red beddings

were composed by clay with fragments of dolomite. In particular, it is composed by the 74.3 % of CaCO<sub>3</sub>, identified as “finely grained calcite”, and the 16.8 % of Fe(OH)<sub>3</sub>, identified as “limonite with colloidal graining” (Mészáros & Vértes, 1959: 24-25; Tab. 4). According to mineralogical analysis carried out at the end of the Nineties (analytical method unspecified) it is composed by a percentage of Fe comprised between 11.49 % and 4.61 % of Fe (Dobosi, 2006: 35; Tab. 5). Quantitative evaluation of XRD spectra, published in 2015, shows that ochre is composed by dolomite (about 80 w/w %) with smaller quantities of quartz, clay and mica, and 5 % of hematite; dolomite dominate also the red dolomite veins. XRD spectra of a sample of red ochre particoles (<10µm) near a

<i>Lovas - Micromineralogical analysis of 1954</i>				
CaCO <sub>3</sub>	Fe(OH) <sub>3</sub>	SiO <sub>2</sub>	heavy fraction	coal, bone
74.3 %	16.8 %	6.7 %	1.0 %	1.2 %

**Tab. 4** – Red sediment from Lovas: micromineralogical results (analytical method unspecified) (from Mészáros & Vértes, 1959: 24).

**Tab. 4** – Sédiment rouge de Lovas : résultats microminéralogiques (méthode analytique non spécifiée) (d'après Mészáros & Vértes, 1959 : 24).

<i>Lovas - Mineralogical analysis of 1999</i>					
Mg	Al	Si	P	Ca	Fe
11.50	0.88	3.71	/	72.43	11.49
1.51	/	/	/	93.87	4.61

**Tab. 5** – Ochre from Lovas: mineralogical results (analytical method unspecified) (from Dobosi, 2006: 35).

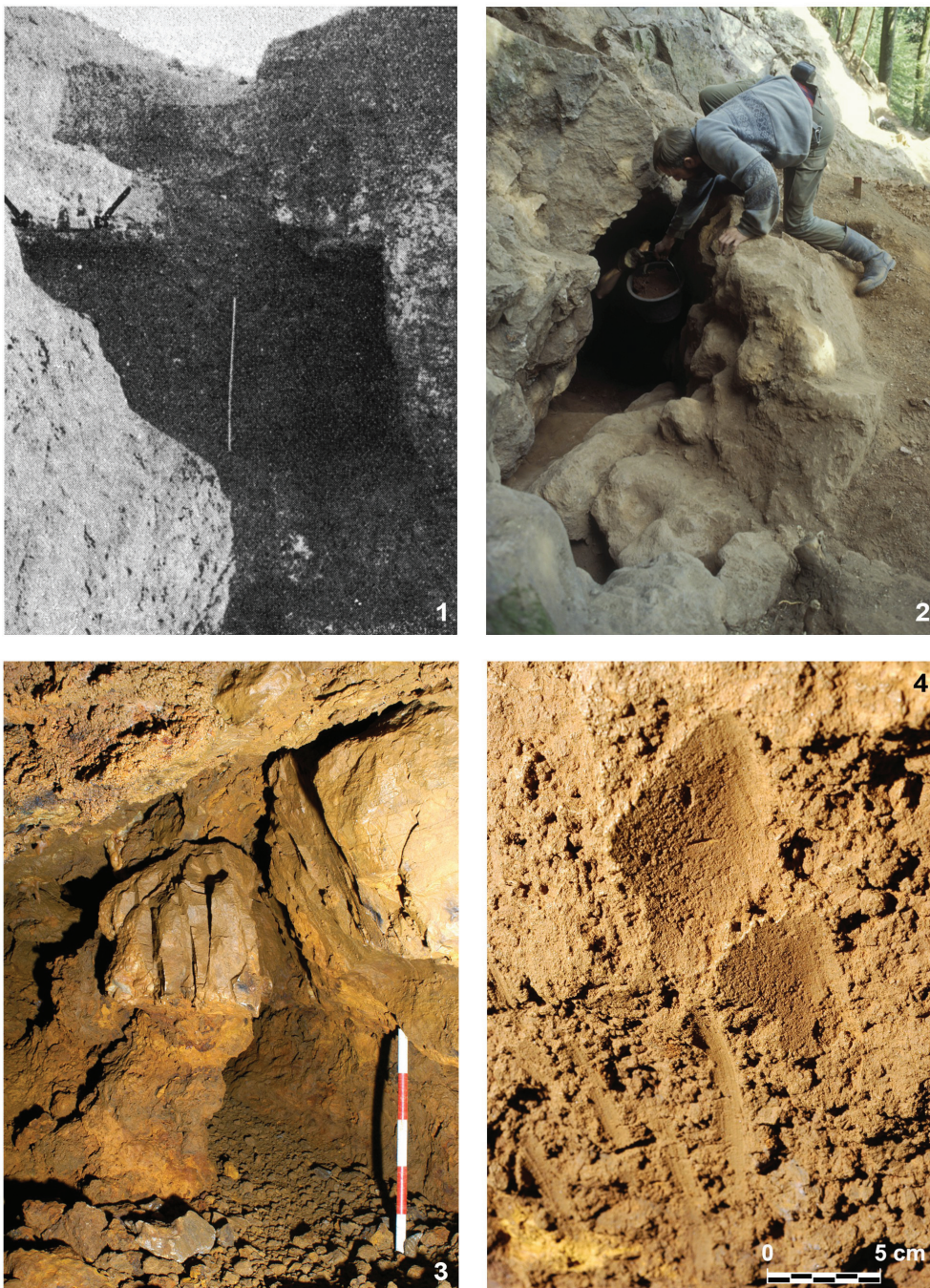
**Tab. 5** – Ocre de Lovas : résultats minéralogiques (méthode analytique non spécifiée) (par Dobosi, 2006 : 35).

bone tools shows that ochre is composed by dolomite (c. 70 w/w %), smaller quantities of quartz, clay, calcite and 12 % of hematite. SEM analysis equipped with energy-dispersive spectrometer on red ochre show a 52.1 wt % of Ca and 4.2 wt % of Mg as dolomite, partly coated by the 22.7 wt % of Fe, as hematite. The ochre show a particular structure, formed by a “core” of dolomite or quartz grains and an external “shell” of hematite. Since this structure does not occur in the *in situ* pigment, researchers suggest that prehistoric miners ground the red material in order to obtain finer particles and exploited the natural oxidation process through which hematite nano-particles coated the core of sand or silt-sized grains, obtaining in this way a major volume of pigmented material (Sajó *et al.*, 2015). The archaeological assemblage includes lithic and bone implements. Lithics are about twenty and are not diagnostic, except for a bifacial leaf scraper classified by Vértes and Mészáros (1959) in the Transdanubian Szeletian group, known as Jankovichian Culture, attributed by them in the Würm I-II Interstadial. This setting on the base of a single tool in the Middle Palaeolithic is now widely rejected. Raw material shows use of radiolarite coming from sources 20 kilometres far from the site (Dobosi, 2006: 34-35). Lovas distinguishes for its bone assemblage, characterized by tools specifically made for mining. On 130 definable animal bones, the 80 % are of *Alces alces*, above the 16 % of *Cervus elaphus*, and the remaining above 4 % equally divided between *Rangifer tarandus*, *Capra capratorum*, *Sus scrofa*, *Equus sp.*, and *Grus grus*. Among ninety-two individual bone objects there are awls, antler tools, scapulas probably used as shovels, polished ribs used as levers, spoon chisels from tubular bones. However, peculiar elements of the mining toolkit are 23 *Alces alces* ulnae; their morphology reflects a planned digging function and it is influenced by their anatomic features, since the natural arch of the bone allows to resist to great strain (Fig. 2:3). Bone working techniques show cutting, carving, pointing and polishing traces (Dobosi, 2006: 31-36; Dobosi & Vörös, 1979; Mészáros & Vértes, 1959: 7-25). Mining activities were carried out extracting first ochre veins outcropping on the surface and then the one placed within the soil. As attested by the best preserved and excavated Pit 2 (above 6 metres long, with an height and a width of 2.5 metres) the last phase of exploitation corresponds to the lowest layer, upon which other distinct layers indicate its use as camp after

the end of mining activities (Dobosi, 2006: 35; Dobosi & Vörös, 1979: 21; Mészáros & Vértes, 1959: 1-7, 24-25; Sajó *et al.*, 2015) (Fig. 3:1). The chronological setting of the site was difficult and controversial. However, five new AMS radiocarbon dating on *Alces alces* bone confirm the previous single radiocarbon date known for this site ( $11740 \pm 100$  years BP, ETH-15199). In fact they yielded a date between  $11941 \pm 44$  (MAMS-21718) and  $11469 \pm 40$  (MAMS-21721) years BP (13800-13200 years calBP) (Tab. 1), setting mining activities during the Allerød Interstadial GI-1-b-d (Dobosi, 2006: 32; Sajó *et al.*, 2015).

The Holocene iron oxides mining includes the sites of Bad Sulzburg and Grotta della Monaca. Bad Sulzburg is located in the southern slope of the Black Forest, in the federal state of Baden-Württemberg (south-western Germany). This mine sets in an area in which hydrothermal veins containing hematite with low content of non ferrous or precious ore outcrop. The iron ore deposits are associated with a strong silicification of the rock wall that leads to the formation of “reef-like” rocks emerging from the neighbouring gneiss. Hematite is finely distributed within “hornstone-like” quartz but it occurs also as joint fillings or compact masses. Under these rock formations detrital slopes are characterized by ground substrates and siliceous blocks pigmented by hematite. Bad Sulzburg mine was discovered in 1980 by Norbert Kindler and excavated by the Institute of Pre- and Proto-History of the Freiburg University in 1997 (Goldenberg *et al.*, 2003: 179-180). Apart from shallow cavities in the rocky outcrop, it is characterized by a small pit – 4 metres long, 1 metre wide and 3 metres deep – dug for hematite exploitation (Fig. 3:2). This pit (called “Pit 1”) was refilled with mining debris, containing stone implements and fragments, a flint blade and charcoals. Between them, 33 more or less completely preserved stone hammers were recovered. They are mainly made of quartzite or quartzitic sandstone rounded pebbles that could be acquired in the gravel banks of the Rhine, 15 kilometres far from the mine. Some samples were handheld, other show notches and trace of picking. Within 16 hafted hammers 13 shows trace of use after their breakage, i.e. they were employed in mining activities after their damage. The only flint blade might have been used to repair hammer hafting; while two grindstones, respectively made of amphibolite and gneiss, could be connected with hematite





**Fig. 3** – 1. Southern profile of Lovas mine “Pit 2”, Hungary (from Mészáros & Vértes, 1959: fig. 2.1); 2. Entrance of the so-called “Pit 1” of Bad Sulzburg mine, Germany (Image courtesy of Gert Goldenberg - Institut für Archäologien, Universität Innsbruck); 3. Goethite supporting pillar occurring within a Final Neolithic mining sector of Grotta della Monaca (Italy) (photo by F. Larocca); 4. Imprints of scapula shovels on a goethite vein outcropping within a Final Neolithic mining sector of Grotta della Monaca (Italy) (photo by F. Larocca).

**Fig. 3** – 1. Profil méridional du « Puits 2 » de la mine de Lovas, Hongrie (d’après Mészáros & Vértes, 1959 : fig. 2.1) ; 2. Entrée du « Puits 1 » de la mine de Bad Sulzburg, Allemagne (Droit de reproduction de Gert Goldenberg - Institut für Archäologien, Universität Innsbruck) ; 3. Pilier de soutien en goéthite au sein d’un secteur minier datant de la fin du Néolithique de la Grotta della Monaca (Italie) (photo de F. Larocca) ; 4. Empreintes de petites pelles en cubitus sur un filon de goéthite affleurant à l’intérieur d’une zone d’extraction, vers la fin du Néolithique, Grotta della Monaca (Italie) (photo de F. Larocca).



processing, such as the *in situ* mineral grinding for the testing of iron oxide quality. Mining techniques were based on the crushing of rock rich in hematite using stone hammers (Goldenberg *et al.*, 2003: 181-185). Three radiocarbon AMS dating on oak wood charcoals place mining activities between 6250±60 (Beta-105503) and 5810±60 (Beta-105504) years BP (end of VI-beginning of V millennium calBC) (Tab. 1). They set mining activities in the Linear Ceramic Culture of the Upper Rhine Valley (5500-4800 years BC) (Goldenberg *et al.*, 2003: 182). In this period organized mining activities were at their first development, reaching the most advance expression during the Late Neolithic, within the extensive mining complexes of Michelsberg Culture. However, evidence from Bad Sulzburg mine allows to recognize an already specialized extractive method. Probably hematite was reserved for the consumption of the local group that maybe could have come from the Markgräflerland region, located in the western border between the Black Forest and the Rhine River (Goldenberg *et al.*, 2003: 182, 183, 185).

Grotta della Monaca is a karstic cave located in the municipality district of Sant'Agata di Esaro (CS), Calabria, southern Italy. Its impressive entrance is located on the hydrographic left of the Esaro River. This cavity sets in the north-western sector of the Calabrian-Peloritan Arc, along a fault called "Sanginetto line" marking the boundary between the metamorphic crystalline Calabrian units and the Apennines carbonate domains. It develops within Mesozoic carbonate units (Triassic) for about 500 metres in length and it is composed by galleries, chambers and narrow passages (Dimuccio, 2005: 25-26). Being highly visible and easily reachable, it is cited in some literary sources and explorative reports dating back to the 19th and 20th centuries. Inspired by these writings, surveys were carried out in the 1990s by the "Enzo dei Medici" Regional Centre of Speleology in collaboration with the "Aldo Moro" University of Bari and led to the discovery of ancient mining activities, giving birth to speleo-archaeological researches ongoing since 2000, under the direction of Felice Larocca (Larocca & Dimuccio, 1997; Larocca & Lorusso, 1998). The cavity was frequented during the late Neolithic for the exploitation of iron hydroxides, such as goethite, that outcrops isolated or associated to lepidocrocite, its polymorph (Larocca, 2010: 267-268). The carbonate substrate in which Grotta della Monaca

develops (Unità di San Donato) contains, among others, scattered primary iron sulfide mineralizations (e.g. pyrite - FeS<sub>2</sub>) (Amodio Morelli *et al.*, 1976; Bonardi *et al.*, 1982; Boni *et al.*, 1990; Dimuccio, 2005; Letto *et al.*, 1992; Lorenzoni *et al.*, 1983), which form in oxygen-poor environments. It seems plausible to assume that the iron oxide/hydroxide minerals (essentially massive dyke/stratiform facies), observed inside the cave (Larocca & Dimuccio, 1997; Dimuccio *et al.*, 2005), are the result of pyrite oxidation in oxygen-rich environment. Grotta della Monaca seems to be a typical example of polygenetic cave (i.e. has experienced more than one type of origin) where the mixing of hypogenic ascending flows (rising along fractures and inclined bedding planes) with probable infiltrating oxygen-rich epigenetic meteoric waters (descending vadose flows) caused the cave development (by corrosion) and a concurrent precipitation of massive ferruginous mineralizations. An important role of microbial activity, especially in catalysing the precipitation of iron during the active speleogenetic phases, cannot be excluded (Dimuccio Luca Antonio, personal communication). The archaeological excavation of two underground mining sectors, called "Buca delle impronte" and "Ramo delle vaschette", revealed evidence of iron hydroxides exploitation, AMS dated between 5183±50 (LTL-3583A) and 4880±45 (LTL-3581A) years BP (end of V-first half of IV millennium calBC) (Tab. 1). The main feature of this mining activity is the use of natural cavities as means of access to iron hydroxide sources. In this sense, caves are though "containers" of precious mineral sources, as already suggested by Gerd Weisgerber in the Nineties (Larocca, 2010: 269; Weisgerber, 1997). Underground mining required goethite pillars to support the unstable roof preventing this way its potential collapse (Fig. 3:3). This technique reflect a mining tradition similar to Neolithic flint exploitation one (Di Lernia & Galiberti, 1993: 33-36). The tools used for mining have not been found but they left several digging traces both on veins and on small goethite boulders, scattered on the ground and within the archaeological deposit. It is possible to identify with certainty blows left by picks made of deer antler and by shovels of mammal scapulas (Larocca & Levato, 2013: 23-24; Fig. 3:4). For the underground lighting miners used torches made of small *Pinus sylvestris* branches (Larocca, 2012: 254). Recent techno-functional analyses on lower and upper grindstones – probably of the Final Ne-

olithic and coming from the entrance – have revealed grinding and crushing activities of goethite blocks (Caricola, 2013: 157-159). Unfortunately, it is still unknown where these minerals were exported to and what was their use.

### 3. CONCLUSION

The actual state of the researches on the currently known iron oxides prehistoric mines does not allow understanding the socio-economic context that developed such exploitations; especially because of the lack of settlements linked to the mining sites, except for Rydno. Unfortunately not much is known about mineral processing phases after its extraction; only a subsequent grinding phase carried out *in situ* or in the immediate surroundings is attested, as at Bad Sulzburg, Grotta della Monaca, probably Lovas, and Rydno. In the latter, thin sandstone palettes used for grinding and mixing hematite have been found, such as quartzite and granite upper grinding stones that bear traces of ochre (Schild *et al.*, 2011: 96). Still more difficult is to determine in what shape, in which way and where the finished product was carried. Ethnographic record suggests that aborigines miners of Wilgie Mia (Western Australia) separated “ochre” from the spoil or directly *in situ* or in places surrounding the mines. The finely ground mineral was mixed with water to form cakes, ready for barter only if they showed a bright red colour when dry (Davidson Sutherland, 1952: 84; Sagona, 1994: 137). While mining at Wilgie Mia was carried out by men, hematite from Toolumbunner mine (Tasmania) was excavated by women, that ground hematite using pestles and discoid faceted lithic bases (the so-called “ballywinne”). Powdered mineral was transported within hides bags downhill, where men made a paste mixing “ochre” and fat used for body painting (Sagona, 1994: 114-117, 143-144). According to Schild *et al.* (2011: 404), hematite mining organization at Rydno shows close analogies with Parachilna (Bookartoo) mine, in South Australia. This mine was controlled by Blinman aborigines that allowed the access to the source in return to gifts. Organized expeditions followed established trade routes, carrying hematite in form of cakes weighing above 30-40 kilograms. The central role of iron oxide from Parachilna within aborigines communities was related not only to its quality but also to its traditional and spiritual value, since it

was originated by the blood of the big dog *Marindi* according to aboriginal mythology (Schild *et al.*, 2011: 404; Smith & Fankhauser, 2009: 4-5). From the archaeological point of view, the use iron oxides is widely attested in shelters, caves, and open-air settlements. Evidence shows their use as pigments, abrasives, and drying agents (Salomon *et al.*, 2008; Wadley, 2005; Philibert, 1994). Nevertheless, in absence of direct evidence, their finding within the archaeological sites allows further hypothesis based on historical and ethnographic sources: body painting, hide processing, organic matter preservation, medical remedy. The wide use of “ochre” – above all of its red variety – during Prehistory is often explained with different utilitarian and/or symbolic interpretations (Wadley, 2005). However, it can be assumed that the two functions do not exclude each other and that the utilitarian use of ochre might have been associated with a parallel development of its colour symbolism (Martini, 2010: 158-163). The relevance of iron oxides and “ochres” in general among prehistoric communities suggests a more in-depth research on raw material sources in close connection, where possible, with mining and settlement evidence, in order to reconstruct the social and economic context of the human groups that sought a mineral, evidently precious, even in the underground.

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