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## MEDIEVAL LEAD SMELTING ON THE MONT LOZÈRE, SOUTHERN FRANCE.

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*Key words* : lead metallurgy, medieval, palynology, slag, isotope, petrology, pollution.

**ABSTRACT** : During a survey on the western part of the Mont Lozère (Cévennes National Park), about 50 lead smelting sites were mapped. Pluridisciplinary research activities were engaged : archaeology with small excavations to check extension of sites, presence of archaeological levels, ... ; survey around the granitic massif on mineralisations and mining traces ; characterization, typology of metallurgical wastes (slags, lining, ...) ; lead isotopes on mineralisation, slag, soil, plant, ... ; palynology and geochemistry on core collected in peatland.

Five radiocarbon dates on charcoal linked to slags gave a medieval age (between 985 and 1277 AD). All investigated sites show the same slag typology. The main facies are a black one, often vitreous, and a grey one, white coated, with bigger Pb prills. Chemical data have to be interpreted as complex genesis for slag, *i.e.* for smelting process : participation of arena, addition of iron (oxydes ?), lime, ... but the signification of the different slag facies is not yet elucidated.

Lead isotope data are in agreement with exploitation of mineralisations located in the S-SW border of Mont Lozère Granite but not with exploitation of mineralisations located in northern one.

A new unexpected conclusion resulted from results on a peat core : another period of lead smelting, with the same chemical and Pb isotopic characters occurred during late La Tène.

## INTRODUCTION

This progress report concern a multidisciplinary work about the lead metallurgy whose traces are located on the western part of the granitic Mont Lozère (Cévennes National Park).

The Mont Lozère Granite is a late-hercynian one at the S-E of the Massif Central. As his southern brother, Mont Aigoual Granite, it intruded the metamorphic basement (Cévennes Schistes). After a long period of weathering and erosion, a new cover was deposited (liasic and jurassic formations, mainly lime and dolomite, which form the Causses). Mineralisations occur all around these granites, both in the basement and in the cover but only in the first decametres of the cover (Fig.1). Most of mineralisation (vein or stratiform) bear Pb as main metal (galena) but also may contain As, Cu, Zn, Sb, and Ag as minor constituent. Associated minerals are quartz and/or barytine (4).

During preliminary survey on the western part of the Mont Lozère about 50 lead smelting sites (Fig.2) were mapped inside a seven square kilometres area (J. Peytavin, S. Lhuillier). Few sites were added during subsequent surveys, but a new area was defined three kilometres eastward. Altitudes of all slag sites are between 1360 and 1430 m. Most sites show a very poor vegetation cover compared to neighbouring areas with no slags.

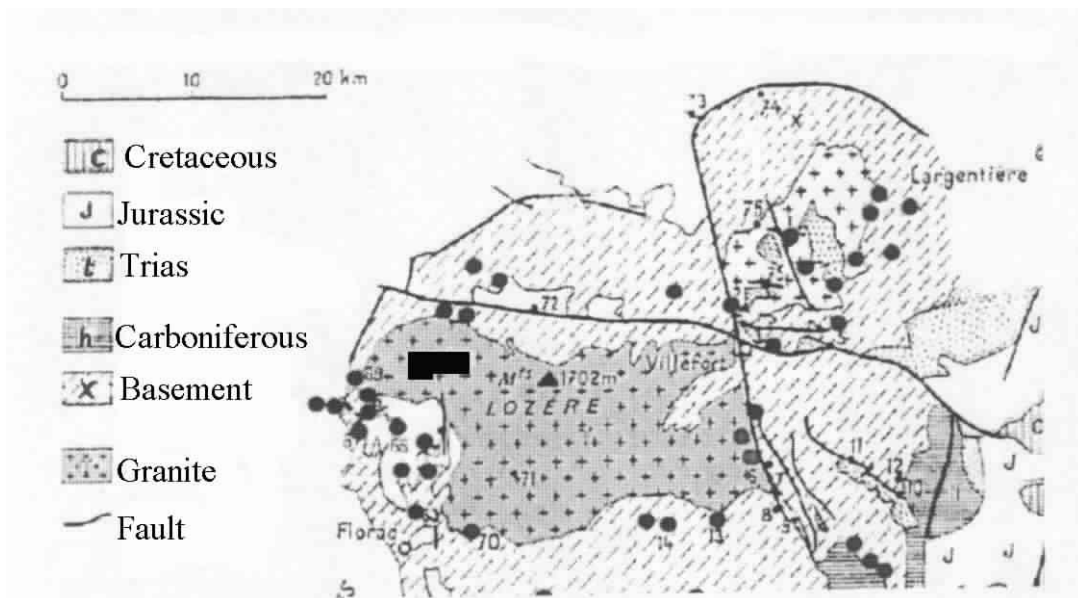


Fig. 1 : Geological sketch. Black dots are main Pb (Ag) mineralisations. Black rectangle is the slag area.

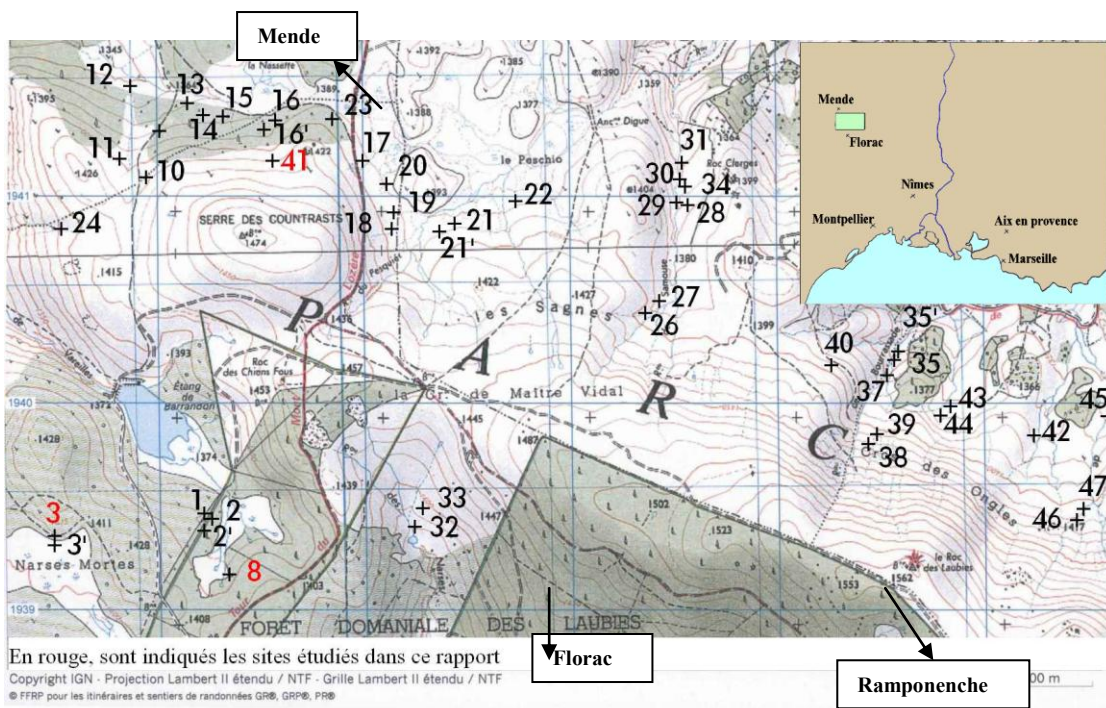


Fig. 2 : slag sites in the Barrandon Pond area.

Five  $^{14}\text{C}$  dates conducted on charcoal fragments linked to slags gave a medieval age : between 985 and 1277 AD, overlapping the five dates with 95% confidence level, but four of them are focused on the first half XIIIth. C. Because all sites seem show the same characters and typology, we are enclined to believe that these ages are relevant for the whole set of slag sites. But don't forget that a Galloroman mine (dated by oil-lamp, Le Bleyard-Neyrac area) and Renaissance mining (radiocarbon age near Cocurès) exist (Fig.4). Complementary ways of research were engaged : archaeology with small excavations to check extension of sites, presence of archaeological levels, diagnostic for excavation, ...; survey around the granitic massif on mineralisations and mining traces ; characterization, typology of metallurgical wastes (slags, lining, ...) ; lead isotopes on

mineralisation, slag, soil, plant, ... ; palynology and geochemistry on core collected in peatland (Narses Mortes) ; heavy metals pollution in and around smelting workshops.

## Historical context

Medieval Gevaudan had about the same extension as the present *Département de la Lozère* (48). After migrations times during early Middle Ages, Charlemagne (*Carolus Magnus*) created Aquitaine kingdom for his son Louis (*Ludovicus*) the VIIth ; this kingdom included Gevaudan. At about 1100, Gevaudan was binded to the County of Toulouse. During Medieval period, from the VIIth C. until Philippe the IVth (socalled Le Bel, end XIIIth C. / beginning XIVth), silver was the main metal for coinage ; consequently a renewal of interest for mining developed notably in the Cévennes as all along the southern border of the Massif Central (1). The Coinage Hall in Mende might be created about 1170 (attribution of *regalia* to the Bishop of Mende by Louis the VIIth.). At about the same time Aldebert, the bishop of Mende, tried to lay hands on mines and mining right in his diocese, against local powers (baronnies, priory, ...). Last coinage from Mende dated about from 1316. Relationship of mining in Gevaudan and vicissitudes during the Albigeois Crusade are not yet known but we have to emphasize coincidence with the last part of our present datations span (Meaux-Paris treaty : 1229). During the XIIIth C., the Capetian power has tried progressively to recover mining right. In 1306, an *acte de paréage* between the Bishop and the king Philippe the IVth fixed a joint-authority on Gevaudan.

Very few archives concern directly the Mont Lozère area and its mining/smelting activity, and they are later than the expected time for it. More precision on the datation of this medieval mining/smelting activity is required but archaeological investigations (excavation, ...) have not yet given enough information. Also, very few information were obtained about workshop operation.

## FROM ORE TO WORKSHOP

One of the aims of this study is to track the source of Pb ores which supplied the smelting sites using the Pb isotopic composition. The lead isotopic composition of galena collected in most surrounding ores is very similar to that of different slag samples (lead prills) sampled on workshop sites (Fig.3). On the other hand, the high precision of the results allowed us to select the mineralised areas which were probably the ore sources. The Pb isotopic composition of slags is even more homogeneous ( $^{208/206}\text{Pb} = 2.092 \pm 0.002$  ;  $^{206/207}\text{Pb} = 1.179 \pm 0.001$  ;  $^{208/204}\text{Pb} = 38.663 \pm 0.025$  ;  $^{207/204}\text{Pb} = 15.665 \pm 0.006$  ;  $^{206/204}\text{Pb} = 18.476 \pm 0.023$ ), and will allow source tracing in the environment. If we assume that no fractionnement occur during the metallurgical *chaîne opératoire*, these values, may be used to characterize the lead which was produced on the Mont Lozère.

The slag samples show an homogeneous group unlike the whole set of ores samples. The ores that have supply the metallurgical activity are issue of North-Eastern Montmirat and the Western and Northern Bondons areas (Fig.4). The others ones might be to exclude because of the different lead isotopic composition. If further investigations will confirm that, in spite of topological proximity, northern and north-eastern mineralisations did not supply ore to eastern medieval smelting workshops on the Mont Lozère, the place of the Barony of Tournel in the mining game will be difficult to situate.

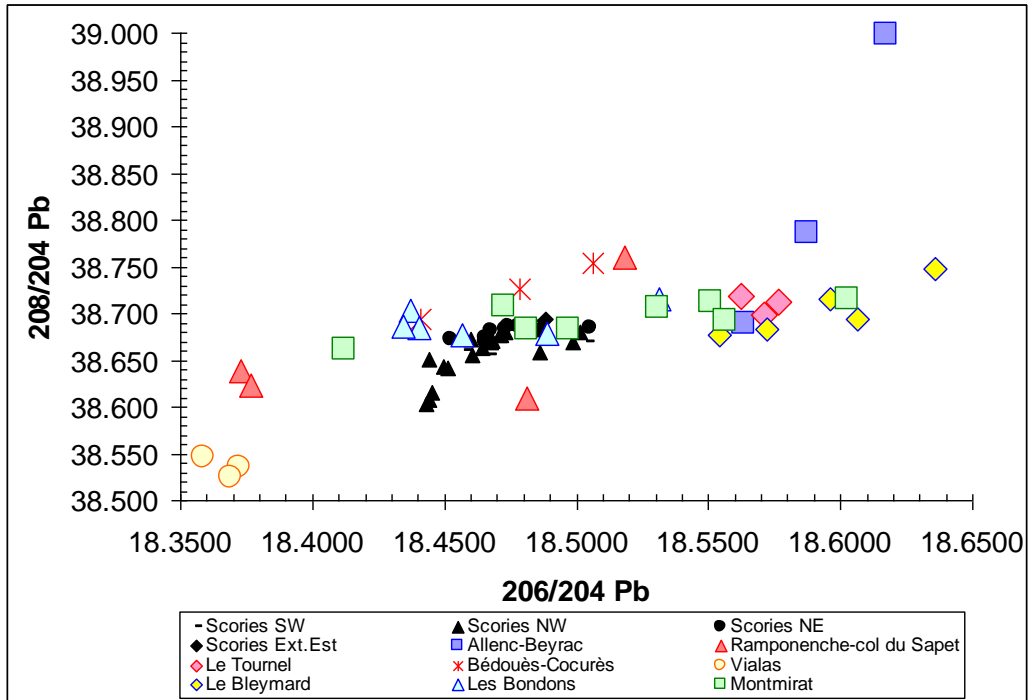


Fig. 3: Diagram  $^{208}\text{Pb}/^{204}\text{Pb}$  versus  $^{206}\text{Pb}/^{204}\text{Pb}$ . We used the NIST 981 Pb reference material and normalised on NIST 997 Tl (7). The RSD (ppm) is 149 ppm on the  $^{208}\text{Pb}/^{206}\text{Pb}$  ratio (<0.015%).

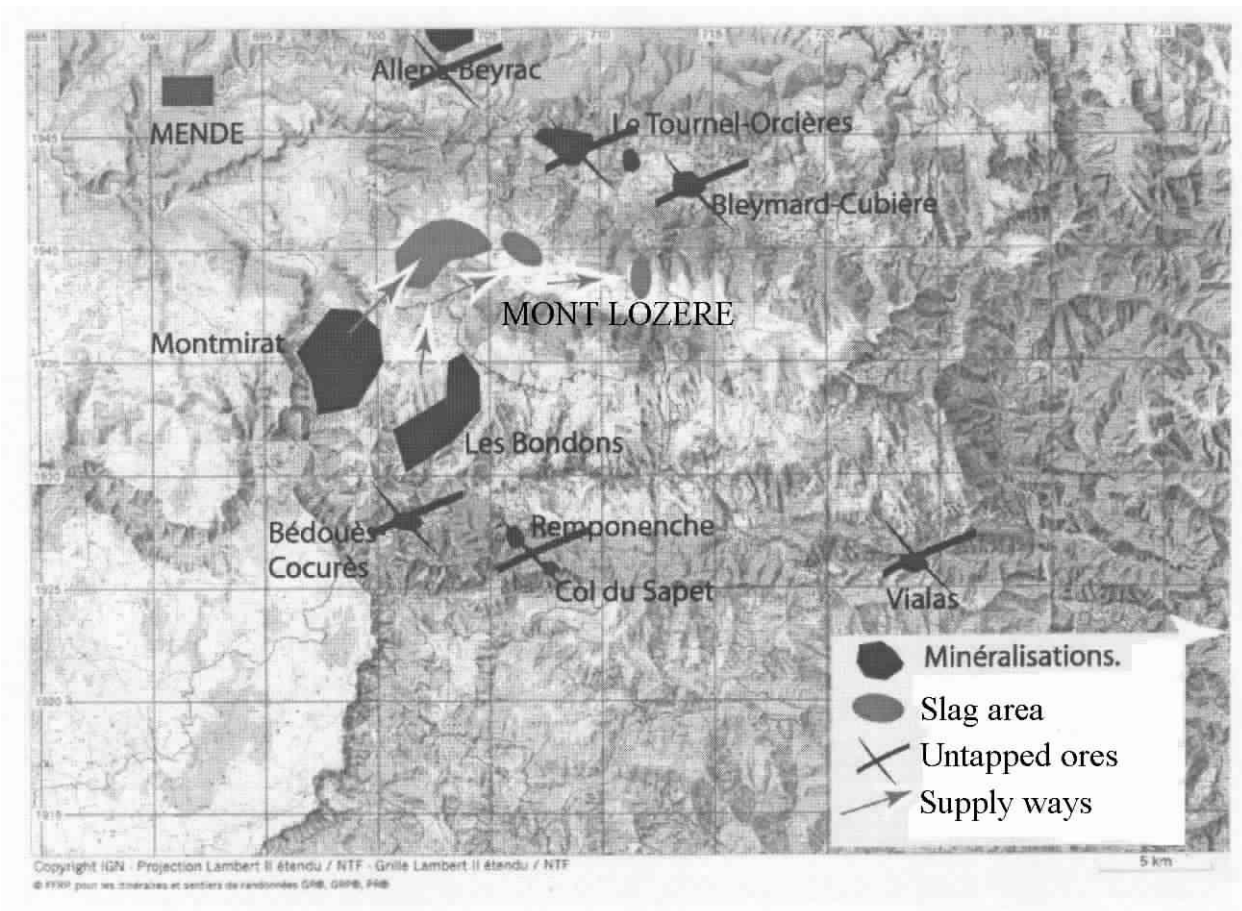


Fig. 4 : Relationship between mineralisation and smelting areas after Pb-isotopic signals.

## INFORMATION FROM SLAG

On smelting sites of Mont Lozère, slags are studied by macroscopical and microscopical observations and by chemical analysis :

- pieces of lining and tuyère. They are composed by a clayey matrix with a granitic arena (quartz, potassic feldspar, ...)
- two main types of slags, black vitreous slags (with inclusions of quartz) and grey slags which present a white patina (Fig. 5 and 6).
- locally, other types of slag may be found. *e.g.* on site 12, there are also many black and crystallised slags, some of them are flat and only one centimetre thick. Iron bloomery slags have been found on sites 8 and 61 but no evident traces of bloomery workshop were not yet discovered on this part of Mont Lozère.



Fig. 5 (left) : black glassy slag, with quartz and feldspar inclusions.

Fig. 6 (right) : white coated slag, with Pb prill in the bottom of a channel.

Because of the very high fragmentation of the samples, it is difficult to give details concerning the shape of the furnaces. However some specimens are enough big and can be interpreted as moulding a basin (diameter about 30 cm). Following Biringuccio such basin could be placed at before or on the side of the furnace. Tuyère might indicate that there was using forced ventilation, but a structure which look like a furnace have two stony walls as a funnel in front of one of dominant wind, and sites are in cleared and windy area. It is also the case of the sites located in the area of Swaledale in Yorkshire (5).

For petrological and chemical study of the various types of slag and lining, all the major and traces elements were taken into account in order to highlight various contributions within slags by means of diagrams of correlation. *E.g.* :

- Si, Al and K are correlated, which may imply that they have the same origin. However lining are made up almost exclusively by these three elements. One can thus consider that the contribution of the molten wall can be represented by the parameter " $P = SiO_2 + Al_2O_3 + K_2O$ ".
- CaO,  $P_2O_5$  and MgO present a very good correlation. This suggests a contribution of a material containing these elements. It could be dolomite  $(Ca, Mg)CO_3$ , because this mineral constitutes part of the gangue of mineralisation. BaO does not show a clear correlation with other elements, although there is a small tendency to a positive correlation with  $CaO + MgO + P_2O_5$ . We can think that BaO comes from another material, and it could act of the barytine  $BaSO_4$  which is also a mineral composing the gangue of the ore. One represents the contribution of the gangue of the ore by the parameter " $G = CaO + MgO + P_2O_5 + BaO$ ".
- The contribution of the ore itself is given by the content of PbO which comes directly from PbS

crystal. The content of PbO in slags is very variable and disturbs strongly the reading of the diagrams. Also, in order to be able to highlight the various types of additions, we chose to use the chemical analyses recomputed at 100% without lead (Fig. 7) :

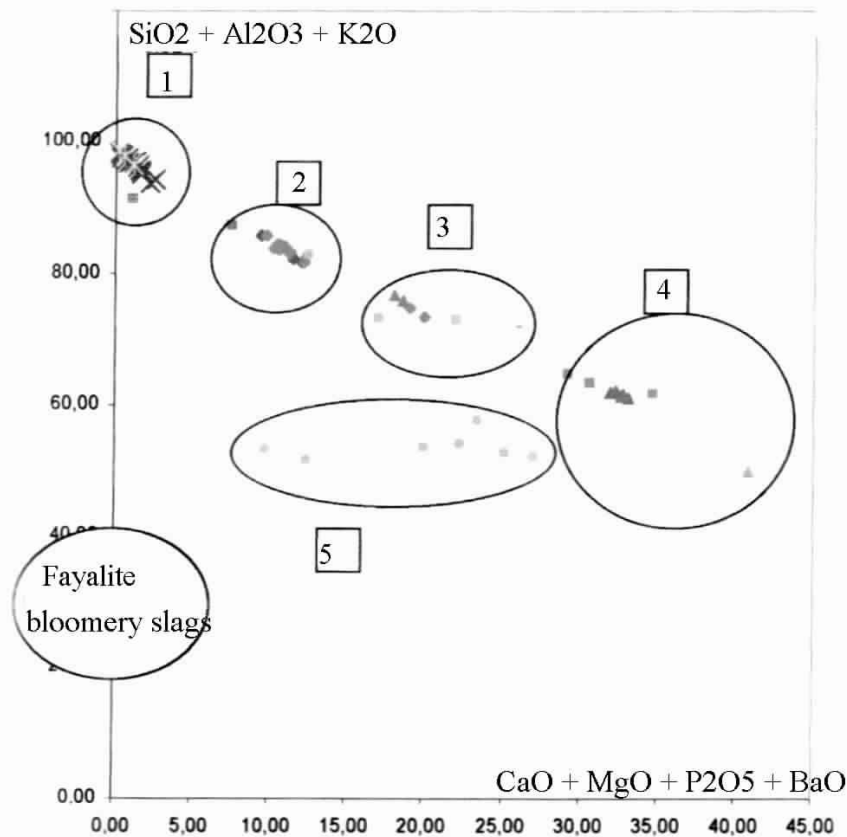


Fig. 7 : Chemical typology of slags (analysis are recalculated without Pb).

In this way, almost all samples are distributed on a line (virtual correlation since the totality of the elements taken into account add up 100% of the analysis). Group 1 represents the composition of burnt or molten lining (without contamination by slag). Group 4 represents the load of the furnace *i.e.* the elements brought by the ore and the associated gangue. Groups 2 and 3 are in intermediate position between these the first two groups. They could be amalgams between the load of the furnace and the melted lining with a lining component more significant (group 2) or a composition charges dominant (group 3).

The presence of remainders of gangue in the charge seems important. According to Birringuccio, it would be preferable to use a cleanest possible ore. According to the experimental work of F Tereygeol (in progress), one could realize that obtaining a pure ore could be done easily during crushing on skins of animals. However the variable report/ratio of (CaO+MgO)/BaO seems to indicate one "bad" washing of the ore that a voluntary addition of products. In some ore galena may be too scatered to be washed perfectly or a part of gangue was left voluntarily ?

Another group 5 is not on this line. They are black and crystallized slags of site 12. Some of these slags are very iron rich. The high content in iron is all the more significant as one moves away from the right-hand side. The diagrams carried out did not show any correlation with the other chemical elements. Iron thus comes here from another contribution that of the walls or from that of, the gangue of the ore. The alignment of slags is done towards another contribution : may be slags characteristic of the reduction of the iron ores in bloomery. As said beforef, bloomery slags have been found on sites 8 and 61. One can thus put forth the assumption that bloomery slags were added to ore. In its treaty, Birringuccio mentioned this feature (XVIth C.).

## INFORMATION FROM PEATLAND

(the word "peatland" is used instead of "peat-bog", seeing that they are minerotrophic)

### Palynology :

Several palynological studies have been conducted in the French Massif central over the last decades. The postglacial vegetation history is now well known, and the major human disturbances on the vegetation dynamics are documented at a regional scale (2, 3, 6). However, less attention was given to the role of specific human activities on plant communities during the last thousand years. Several lead smelting sites found on the western part of the Mont Lozère are located near peatlands, allowing the opportunity to examine at a fine temporal scale the role of past metallurgical activities on the forest dynamics using paleobotanical indicators (pollen, plant-macrofossils). These activities used large quantity of wood for combustion, especially beech, a tree species currently absent from the altitudinal range where lead smelting sites were found. When did metallurgical activities begin on Mont Lozère ? What was the impact on the vegetation ? To answer these questions, we have analysed the pollen content of sediments collected from numerous peatlands. For some of these sites, geochemical analyses were also conducted.

Narses Mortes peatland is located at an altitude of 1400 m on the south-western edge of Mont Lozère (44°26'N; 3°36'E). Two sediment cores were collected : one at the centre of the site where the thickness of organic matter was maximum (140 cm), and another one near the edge of the peatland (lateral core; 138 cm). Pollen analyses were conducted on the lateral core at 2 cm intervals for the upper part of the core (0-74 cm), and at 4 cm intervals for the basal part (74-138 cm). Geochemical analyses were measured on the same levels (*infra*). Seven samples were radiocarbon dated by the AMS method. Dates are presented in calendar years.

The lateral core provides a record of more than 2000 years. Four major periods in the vegetation history were defined based on pollen records. During the first period (Zone 1; >275 BC), a forested environment is deduced from the abundance of birch (*Betula*), deciduous oak (*Quercus*) and beech (*Fagus*). The pollen representation (pollen percentages and concentrations) of all tree species declined rapidly after *circa* 275 BC (Zone 2). The opening of the vegetation cover is associated to a major increase in Pb anomalies reaching a maximum at depth 112 cm. Subsequently, lead anomalies began to drop progressively at the same time of an increase in the pollen representation of *Fagus*, *Quercus* and fir (*Abies*) indicating an expansion of the forest cover. Zone 3 (after *circa* 710 AD) is characterized by a second opening of the forest cover characterized by a significant decrease of all tree species and a maximum in the abundance of Poaceae and anthropogenous herbaceous species. Finally, the upper part of the diagram (Zone 4) show high values of *Pinus* corresponding to its introduction during the last decades on abandoned cultivated fields. High lead anomalies reflect modern atmospheric metal inputs.

Pollen analyses show that past metallurgical activities played a strong and significant role on the vegetation dynamics of Mont Lozère. Two major periods of deforestation occurred during the last 2000 years. The first one was characterized by a decrease of all tree species associated to a peak in lead anomalies. This deforestation is closely associated to metallurgical activities, and occurred during the Antic period, as indicated by the radiocarbon dates and the presence of pollen of planted tree species like chestnut (*Castanea*) and walnut (*Juglans*). The second episode happened more recently during the Medieval period leading to the definitive disappearance of beech and oak in the area where lead smelting sites are located. However, this deforestation is firstly attributed to agropastoral activities because of the high representation of Poaceae and ruderal taxa, but it is likely probable that metallurgical activities also played a role.



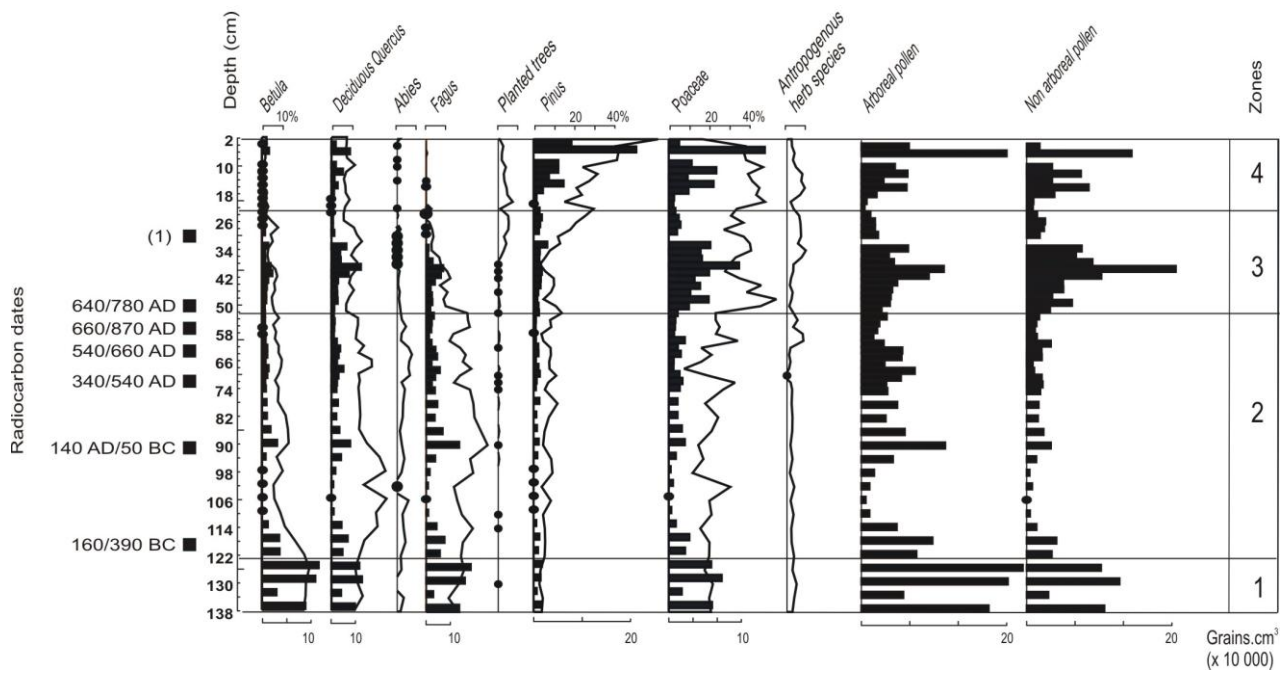


Fig. 8: Simplified pollen diagram of Narses Mortes peatland (lateral core). Results are expressed as pollen percentages (curves) and pollen concentrations ( $\text{grains.cm}^{-3}$ ; bars). Dots represent less than 1%. Pollen zones correspond to the regional vegetation history. (1) :  $^{14}\text{C}$  determination for this sample (level 31 cm) cannot be interpreted as an age because it contains high recent plants content. The level line, in between zones 3 and 4, is also the summit of the chemical profiles (*infra*). After these profiles, the antic metallurgy began at the 119 cm level until -95 cm (or 91) level at least. The medieval one might begin at 41 cm but it is overshadowed by industrial pollution from level 35 cm.

#### Chemical and Pb isotopic investigation :

The "lateral core" was split into two longitudinal sections, one for palynological investigation (*supra*) whereas the second half core was retrieved and divided into 58 individual samples, at 2 cm intervals, from level 23 cm to 139 cm (too much recent plant in upper part). The "Narses Mortes" peatland is strongly minerotrophic and contains 8 to 60 % ash. Minerotrophic peatland records both atmospheric deposition, soils leaching and the groundwater influence. The measured metal concentrations are normalised to Al contents of peat samples and the metal/Al ratios are compared to that of the Mont-Lozère granite : relative excess in metal concentrations are found in peat samples. The Narses Mortes peatland presents an enrichment for most of the heavy metals for the upper samples, which might be attributed to atmospheric inputs during the last centuries. Only Pb and As are enriched in some deeper samples suggesting a contribution from the surrounding activities. Hence two phases of metallurgical activities can be distinguished : one beginning at 119 cm depth and another one at 41 cm depth (Fig. 9-11). The  $^{206}\text{Pb}/^{207}\text{Pb}$  vs depth diagram shows two anthropogenic periods illustrated by less radiogenic isotopic composition than the background composition. The oldest period seems to have long activity and may correspond to a single event because of the same isotopic composition during 30 cm. The second smelting activity began around 41 cm depth but the isotopic composition is not the same along the profile. Other inputs are also recorded (Fig. 12 and 13). These two smelting activities, at different periods, have used the same ores. According to radiocarbon dates and to palynological data, the medieval smelting activity record began at about 41 cm depth. However, a very important activity, recorded in deeper samples (Latenian level, may be continued in Galloroman time) remains being confirmed by archaeological arguments.

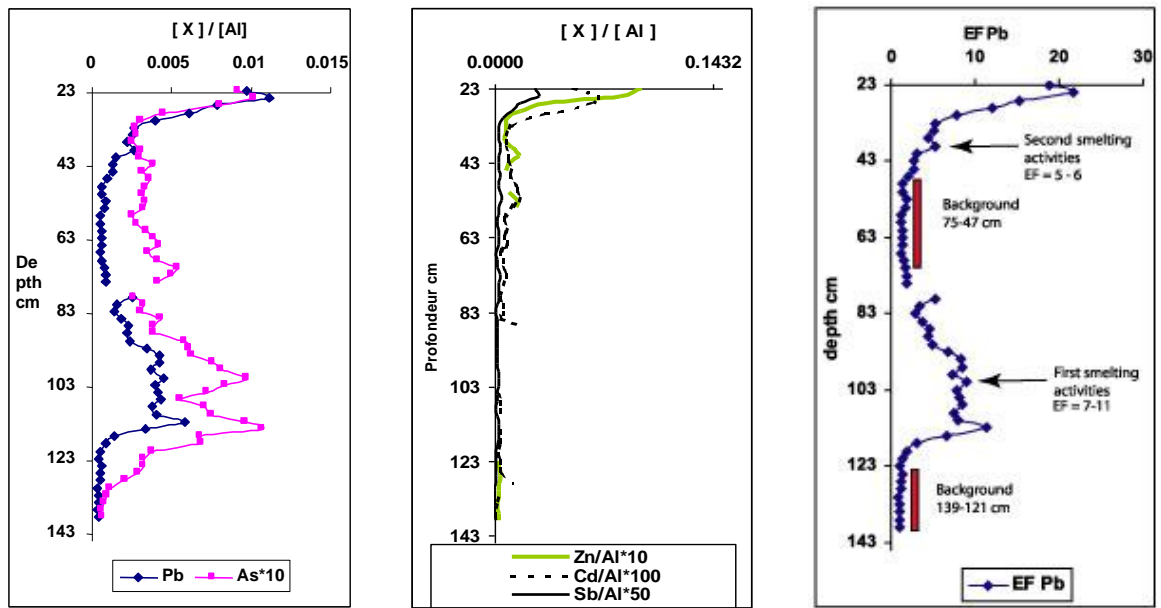


Fig. 9, 10, 11: Elemental chemical analysis. Fig.9 shows the Pb and As concentrations in the peat core and then their correlation. Fig. 10 shows the Zn, Sb and Cd concentrations. Fig. 11 shows the Enrichment Factor (EF) for Pb. EF correspond to :  $( [Pb] / [Al] )_{sample} / ( [Pb] / [Al] )_{granite}$ . The normalisation allows to estimate the excess Pb, then the anthropogenic resulting from metallurgical activities.

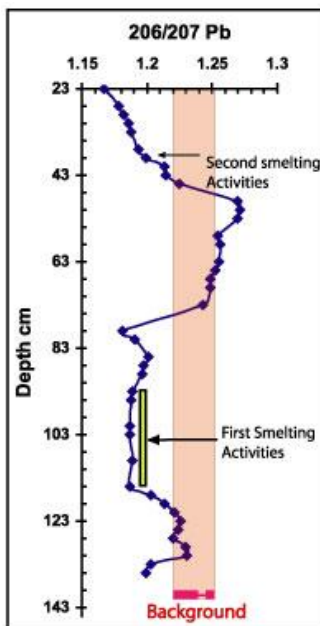


Fig. 12 : Diagram  $^{206}Pb/^{207}Pb$  versus depth. We can see the phases of anthropogenic activities illustrated by the less radiogenic signature than the background value. At 79 and 81 cm depth, a modern contamination due to circulation of waters occurs.

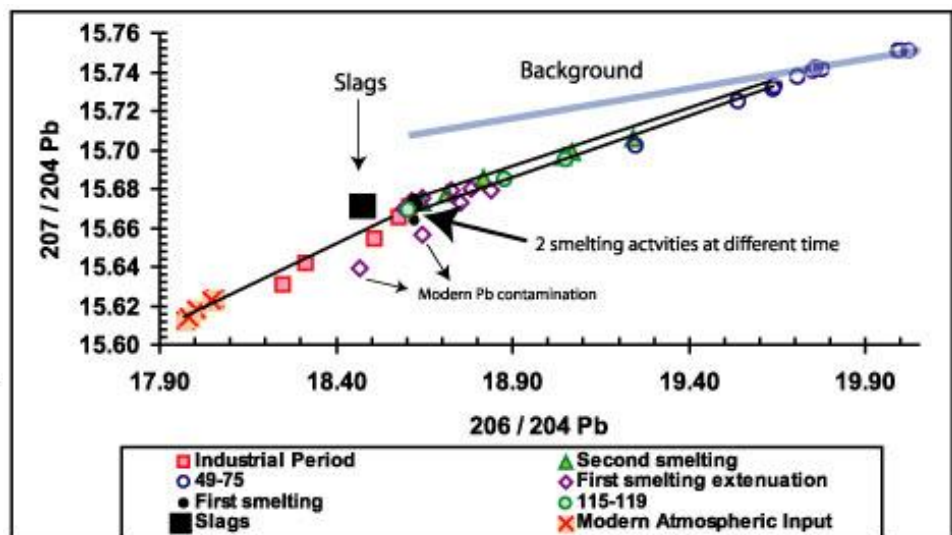


Fig. 13 : Diagram  $^{207}Pb/^{204}Pb$  versus  $^{206}Pb/^{204}Pb$ . The same ore have been used but at different time. Even a wide background, it is possible to see the history of this core link with the anthropogenic activity on the Mont-Lozère massif.

Minerotrophic peatlands can be used for proxies of atmospheric inputs. The atmospheric metal inputs are not disturbed by the leaching of surrounding soils or groundwaters because of the high isotopic gradient measured throughout the entire profil.

## CONCLUSIONS / PROSPECT

Pollen analyses show that past metallurgical activities played a strong and significant role on the vegetation dynamics of Mont Lozère. Two periods with lead metallurgy are recorded by peatland ; however only the medieval one have yet been determined by the radiocarbon dates ; at this time the Antic one is presently "invisible" on the Mont Lozère. Both were using same ores (or ores with the same Pb-isotopic signal). Where are antic workshops? Are their characters similar to medieval ones ? Now, we have to multiply the radiocarbon dates on the whole area.

Two main types of slag are always associated (dark ones and white coated ones) : did they produced after two different operations , *e.g.* as in Yorkshire (5), or during only one operation ? Participation of arena, addition of iron (oxydes ?), lime, ...are proved in some cases, but which ones may be generalised ? What role do such additions ? At this time, no well preserved workshops have been excavated.

We have also to confirm that northern and north-eastern ore deposits did not supplied the Mont Lozère workshops ... and to understand this feature.

We have also to determine data for estimating Pb (Ag) production.

And, last but not least, such ancient pollution in and from smelting workshop may be used as a model for the evolution of present industrial pollution.

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