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Proceedings of the 43rd Annual Conference on Computer Applications and Quantitative Methods In Archaeology

edited by

Stefano Campana, Roberto Scopigno,
Gabriella Carpentiero and Marianna Cirillo

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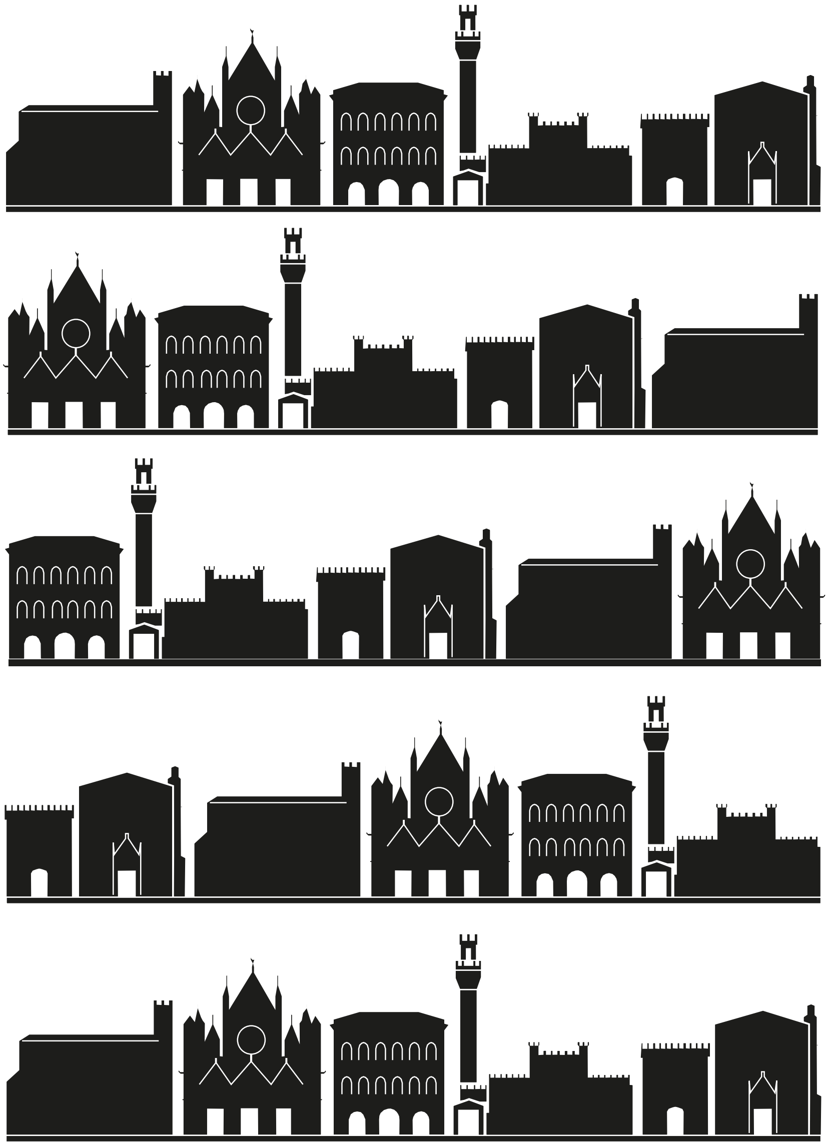

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Computer Applications & Quantitative Methods in Archaeology

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Archaeological and Physicochemical Approaches to the Territory: On-site Analysis and Multidisciplinary Databases for the Reconstruction of Historical Landscapes

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Abstract: The 'Colline Metallifere', a wide territory located in the south-west of Tuscany, has been characterized by a great development of mining activity over the centuries; this is due to the presence of a large mineral deposit, mainly constituted by mixed sulfide ores, that were exploited since Eneolithic age for the extraction of copper, silver, lead and iron.

The territory has extreme relevance for the study of pre-industrial mining and smelting processed. Here, archeological research has been developed since '80 through archaeological digs and surveys. The project and the results we are going to briefly present are the result of an established collaboration between archaeologists and chemists of the University of Siena. It implies the comprehensive study of ancient mining sites, combining classic historical and archaeological observations with physical-chemical measurements and statistical methods, as well as the development of a multidisciplinary-relational database.

Keywords: Interdisciplinary methodology, Archaeological surveys, Smart Database, Physical-chemical analysis, Mining territory

1 Aims of the project (L.D., A.D., V.V., A.B.)

The research project we are going to present has been developed and is still going on over a wide territory located in southern Toscana, the 'Colline Metallifere' district (Fig. 1). The area is very well known for the presence of a large mineral deposit, mainly constituted by mixed sulfide ores, that were exploited for the production of copper, silver, lead and iron. As a consequence, the area has known a great development of mining activity, that has left outstanding remains (mining shafts, galleries, slag heaps) dating back to a long range of time. Beside this, considerable alunite deposits underwent to a systematic extraction activity historically recorded from Late Middle Age to early 19th century, even though archaeological research is discovering a much longer exploitation tradition. In the Colline Metallifere district, the University of Siena has undertaken archeological researches since 1980 through excavations and surveys, covering a territory of more than 145km^2, recording and describing over 2500 sites (about 50% of these were ancient mining and/or smelting sites). The data collected both from digs and fieldworks have contributed to produce a general understanding of the mining exploitation techniques and ore transformation and commercialization over time (Dallai 2013; Bianchi *et al*. 2013). From an environmental point of view, the area offers many research suggestions; among these, the presence of arsenic and heavy metals contamination associated to the relevant remains of the long-

Fig.1. Location of sites referred to in the text.

standing tradition of ore mining and processing has addressed the development of a multidisciplinary project, in which a combination of archaeological and physicochemical analyses and the statistical treatment of the data are used to describe and interpret this peculiar historical landscape (Dallai *et al*. 2013). A number of scientific techniques applied on environmental matrices (i.e. soil, stream sediments, water, plants) have been used with both predictive and descriptive goals. In particular, high throughput techniques have been used in multi-scale investigations (*intra-situ* and medium-large territorial scale) in order to obtain detailed chemical and/or geochemical maps. The merging of these data with archaeological maps will give a more accurate interpretation of the historical context and can be very advantageous in the excavation planning of a site. This model need a specific care in the classification, storage and recovering of the data. Consequently the production of a 'smart database' is mandatory in order to have a correct inter-exchangeability of the data and flexibility in the data-mining.

In the present work the methodological approach of our research is reported together with key studies demonstrating its validity and robustness.

2 The territory of the southern Colline Metallifere: historical mining, metallurgical tradition and archaeological investigations (L.D.)

Investigations in the Colline Metallifere Grossetane, specifically the southern part of the hill-chain that stretches from the province of Livorno (north of Populonia), to Grosseto, began in the mid-1990s, with the planning of a series of topographic campaigns in sample areas that have a strong connection with mining. During the years, several archaeological digs in key sites (i.e. fortified settlements and productive structures) have been undertaken as well. Since the very beginning, the aim of the project has been to propose a reconstructions of changes through time in settlement patterns and exploitation techniques (both mining and metallurgical ones), stressing the existing relations between natural resources and settlements, with a special focus on the Medieval time. The reach mineralization was crucial in addressing the gradual increase in the importance of inland areas, such as the hilly ones, compared with coastal plains. This phenomenon is documented in the archeological record ever since the later Roman time, reaching a peak in the medieval period (Marasco 2013). Polymetallic deposits in the Massa Marittima and Montieri area became of primary importance in the medieval period, for the productions of so-called 'coinable metals'; in this same period, together with intensive mining, specific regulations, the so called *Ordinamenta super artem fossarum rameriae et argenteriae civitatis Massae,* drafted at the end of the 13th century, were produced and included within the statute of the Comune of Massa Marittima (1311-1325). The *Ordinamenta* constitute one of the oldest examples in Europe of mining regulations, and represent a rich source of technical information relating to the management and control of mining activities (Dallai 2014). In the same area, out of the many different research in-depths proposed by the project, some of the mines of Massa Marittima have become the terrain for a pilot study that has produced a better understanding of both mining techniques, productive steps, and the environmental impact of ancient tips on the surrounding area. Underground surveys and explorations have revealed a number of technical characteristics used at these mine-workings as well (Aranguren *et al*. 2007). Chemical analyses carried out on waste material have revealed that, in the presence of marked differences in the nature of the mining deposit in question, initial selection operations at the mine head were capable of eliminating minerals regarded as useless for the purposes of production (e.g. calamine) which is clearly a sign of skill, specifically in the systematic setting aside of zinc minerals from the extraction and metallurgical process (Dallai *et al*. 2013).

The project is still going on, with a special focus on multidisciplinary approaches to the territory. In some cases (a good example is the territory of Montieri), new inspections are currently being planned, to increase the amount of information available. In particular field-surveys on the hill of Montieri have revealed a very large number of mine openings that are now filled in. Although it is not possible to establish a definite date for these, it is known that there were many profitable mines active here in the medieval period (there are also references to these in the well-known *Brevi* of Montieri, written documents dated to the first quarter of the 13th century), which were mined for silver.

Access to the underground areas in the Montieri district has been possible thanks to the presence of mine-workings datable to the early years of the 19th century (in the Poggio area), or to the second half of the 18th century (in the area known as Le Carbonaie), which used parts of older workings. The more recent mine-workings, often intended for the production of green vitriol, have revealed the existence of older tunnels and chambers, probably dug by miners seeking copper and silver minerals, and the presence of waste rock stored within the chambers themselves (Fig. 2) (Dallai *et al*. 2012).

In the majority of the sampled areas of the Colline Metallifere, where researchers have identified ancient mines features and the remains of industrial structures related to metallurgical production, these have become a very good 'data sets' on which the project team could perform combined archaeological and physicochemical analyses. A statistical interpretation of chemical analyses on residues of mineral processing has made it possible to add new pieces of information to our understanding of economic exploitation in the pre-industrial era.

2.1 Data base structure and data collection: physico-chemical, archaeological and geological data (A.B.)

Given the multidisciplinary nature of the project, the storage and management data system has to deal with data originated from different sources: archaeological, chemical and geological in particular. Moreover, it has to be taken in account that the aim of the database is not merely limited to a data consultation, but fosters an interaction between the different sections in order to obtain extra information.

The data base structure is summarized as follows:

- disciplinary sections designed to store information derived from different sources: geological, historical, chemical and archaeological in particular.
- possible interactions between the different sections.
- geographic data management.
- tools for data analysis in each disciplinary section and through the different ones.

The construction of this archive has started with the choice of the RDMS software (*https://it.wikipedia.org/wiki/Relational_database_management_system*), that allows us to build a quite complex relational structure with the possibility to organize geographical data. The next step has been the selection of the Open source MySQL software, as well as MsAccess and

Fig. 2. Ancient mines located in the study area: underground inspections (LTTM, University of Siena).

LibreBase softwares, with the aim of allowing data viewing and consulting. For the construction of the database structure the choice has privileged the open source software Workbanch, which allowed us the development of a conceptual model.

Regarding the three main sections of the database (the geological, chemical and archaeological ones), the 'state of the art' was quite different from one section to the other; as an example, considering the archaeological data, the project could profit of an already existing storage structure which required only a general revision, according to the new needs of the research and the interaction with the others sections. On the contrary, as regards to the chemical and geological sections, the structure had to be completely built up. The final conceptual model of this storage system can be define as a 'star', at whose centre we have the so called 'localization', that is a couple of geographic coordinates. In order to clarify this concept we can use the example of the on-site handheld XRF analyses; once the data have been acquired, they will be stored in a separate table containing the site coordinates; through internal relations, the table is linked to the rest of the database structure.

The nature of data acquired to the project can vary a lot, depending on the different needs of each discipline; if on the one hand for the archaeological and geological sections we deal with descriptive alphanumeric information, which must be digitized by an operator, on the other hand the chemical analyses produce only numeric data. It is clear that this different nature of data turns to be a problem both for their management and comparison. In order to obtain comparative analyses and spatial analysis developed with the GIS software, that is one of the main goals of this project, it is therefore mandatory that the nature of data, even if originally very different, became similar. For this reason we have defined a 'conversion field' voice that enable the translation of alphanumeric values into numbers by giving a value, where possible, to each descriptive definition.

This method can obviously produce errors; therefore the 'weight' of the field may vary, according to the different contexts.

For the statistical analysis, we have chosen the so called 'Cube OLAP methodology' (*https://it.wikipedia.org/wiki/Cubo_OLAP*); the OLAP cube is defined as a multidimensional spreadsheet where the values stored in each face can interact in a virtually unlimited way with the values of the other faces (Fig. 3). Through the use of Pivot Tables, this methodology is capable to analyze a large amount of data in a short time, and can also deal with values of different nature.

Finally, the database scheme is designed to follow three different steps: data input; management and consulting and data processing. This last final step runs partially outside the structure itself and interacts with other software for data exchange. The database can be query in order to obtain a selection of information; thanks to the query, we can obtain sets of data ready to be analyzed by other programs, such as the spreadsheet that supports the function OLAP (i.e. Excel or LibreCalc) and GIS software for spatial analysis. This goal is obtained with a mechanism of data import in which an external spreadsheet regularly updated can developed OLAP queries, graphs and statistical analyses. This sheet can also generate a point file based on geographic coordinates, ready to be imported into GIS software.

Mentioning geographical data storage, the database has also a part dedicated to this aim. MySql software allows us to create specific tables for the geographical data storage that can be related to other tables for data management and interrogation. DBMS bridges between the queries required from the client and the DB storage; this can be particularly useful when managing large volumes of queries. One of the possible goals in the use of this database is also the identification and selection of key areas that satisfy given characteristics (Fig. 4). This process, which is typical of predictive archaeology, follows standardized procedures. The availability of chemical and geological data expands the possible combinations of information, usually limited to the geomorphological ones.

To conclude, the structure that we have designed can also deal with a difficult aspect of data management, the so called indexes. The term index identify the 'weight' assigned to a particular feature, or to a particular combination of elements, in addressing or modifying the development of a given area. The definition of this 'weights' is complex and strictly related to the territory in which they are identified. It is hard to imagine or to employ similar indexes to read different territories; even if possible biases have to be taken in account, we think that they can be a powerful tool for reading homogeneous contexts, especially for predictive analysis.

3 A key study area: Montieri and the medieval silver production

3.1 An archaeological and historical overview (L.D.)

Archaeological investigation in the Montieri district, one of the most relevant silver, lead and copper mining area of the Colline Metallifere, began in 2007, with archaeological digs (emergency excavation works and archaeological digs undertaken both in the town center and in the parish church named 'La Canonica'); surveys on crucial territorial samples, such as the Poggio of Monteri itself and other main mining areas; physico-chemical analysis performed both on wide areas (such as on sediments sampled along the streams of the Poggio itself), as well as on archaeological digging sites (i.e. La Canonica, mentioned above) (Benvenuti *et al*. 2014). The

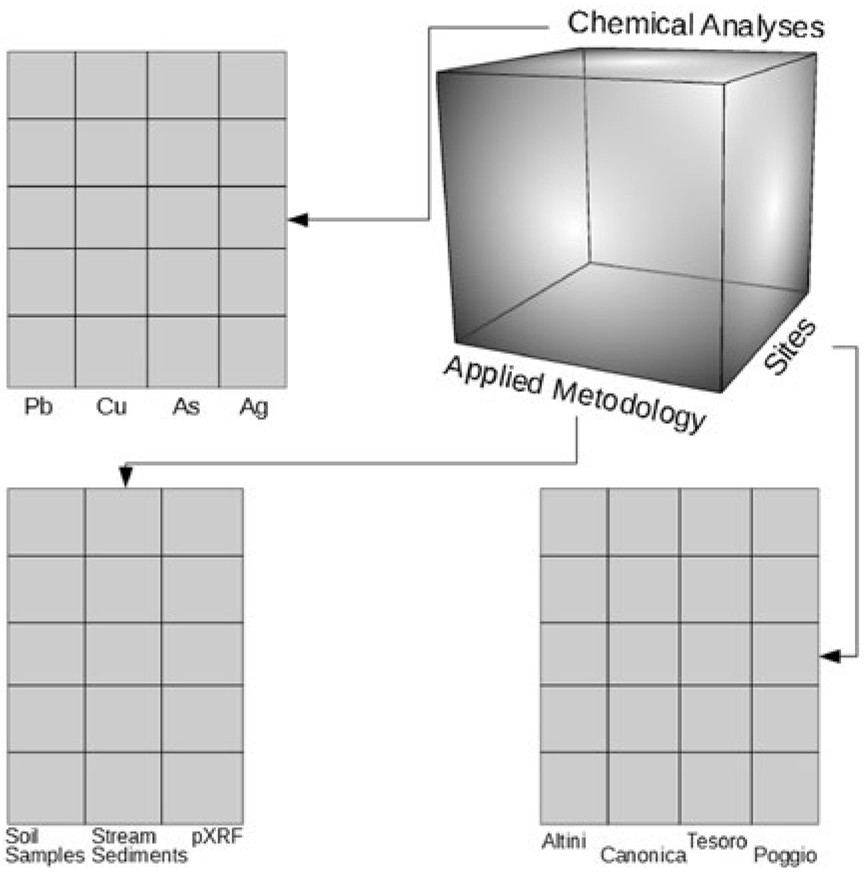

Fig. 3. OLAP cube structure: an example from the project.

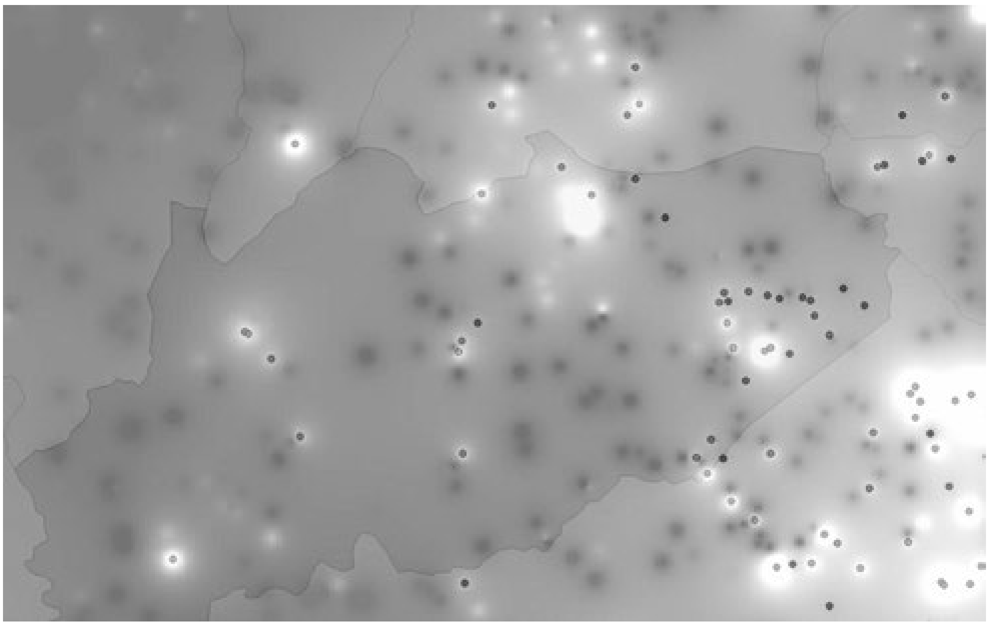

Fig. 4. In white: areas with high values of antimony, arsenic and silver.

bishop of Volterra was the main political figure in this area, and it is due to his influence if the castle of Montieri developed as one of the main mining centers in the Colline Metallifere. The castle was the core of a complex system of ore exploitation and mineral production sites and its economical importance is testified by the building quality and by the presence of a mint which struck coins on behalf of the bishop between the end of the 12th century and the first half of 13th (Bianchi *et al*. 2013).

The multidisciplinary research chose this area as a key one in order to perform multi-scale environmental analyses. In this territory, mineral processing for silver production has led to the accumulation of a significant amount of slag, partly positioned where the modern-day town center stands. A series of test analyses have been recently conducted, relating to both the settlement area and the surrounding territory, as well as on-site physico-chemical analyses realized on the site of La Canonica, a religious complex linked to the bishop of Volterra, attested from written documents since 1133. The church with its extremely peculiar central plan surrounded by six apses and the fine stone architecture, shows the economical effort sustained by the bishop in the mid-11th century. Modifications went on in the next centuries, until the final abandonment of the site, occurred just before the 15th century. At La Canonica site, a very large-scale sampling grid of 1 x 1 m was applied to two different digging areas; the analysis results revealed a marked presence of anomalies (in particular Fe, Pb and Cu) in an area where archaeological research discovered the presence of archaeo-metallurgical structures (specifically: forges, and the remains of a bell furnace). The integrated approach, involving scientific analysis and a historical-archeological input, has made it possible to get very significant results also on a larger scale; the distribution of the geochemical anomalies of trace elements has enabled the study of extensive extraction or transformation sites from the pre-industrial era.

3.2 Environmental data collection: on-site and territorial strategies (A.B., V.V.)

From an environmental point of view, the Montieri territory turned out to be ideal to define a standard methodology of sampling that could allow a coherent and statistically reliable data collection.

After some tests, we could define a standard method in order to identify the areas which were matching given criteria, and therefore were suitable to collect significant measurements directly on the field, as well as ideal to sample soils and stream sediments. The identification of these areas is finalized to perform predictive analyses as well. As said, the predictive methodology used in archaeology is based mainly on the definition of morphological parameters: i.e. slope, exposure, asperity index.

Regarding the Montieri area, the agricultural resource availability introduces a single powerful variable capable to call in question all the others based on geomorphological parameters. For a proper map construction, it is therefore necessary to consistently reduce the weight of the most common parameters and to identify new ones, in order to design predictive maps more coherent and effective.

Once that the areas of interest were selected we were able to establish a field analysis and sampling strategy; obviously, the methodology is different dealing with a single archaeological site or with a large territory (Fig. 5). In the first case, as mentioned before, the data (XRF measurements) have been collected following a grid of 1 x 1 square m; inside each square we have collected from a minimum of 3 to a maximum of 4 XRF measures or more, if necessary. When possible, 1 or 2 samples of soils have been collected as well. In the case of territorial analyses, we have proceeded with a systematic soil and stream sediments sampling; in fact, in a wooden territory, as the Montieri one, streams are particularly useful both as natural paths to cross the vegetation, and, moreover, as heavy metal collectors. From the samples, which are normally taken every 50m, the laboratory analyses and the different techniques applied (i.e. X-ray fluorescence, Atomic Absorption Spectroscopy (AAS) and ICP-MS spectrometry) could extract interesting physicochemical details to produce territorial maps highlighting the main concentration of given elements (Fig. 6).

Fig. 5. Montieri: on-site XRF analyses (LTTM, University of Siena).

3.3 Slags and heavy metal contamination: using environmental data for a historical reconstruction (V.V., A.D.)

Regarding the large scale investigations, arsenic (As) has been fruitfully used as tracer elements to study not only the environmental contamination but, as we will see, even the historical landscape: indeed, this can be considered a very good example of the effectiveness of a multidisciplinary approach to the study of the territory.

Arsenic is ubiquitous in mixed sulfide mineralization and it was proved that its soil concentration is closely related to the extraction and processing of these minerals (Donati *et al*. 2005).

The concentration of arsenic and other heavy metals in soil and sediments is usually measured with purpose of environment monitoring. In fact, part of our data concerning the distributions of trace elements was originally used for their ‘natural mean ground level’ determination in the Colline Metallifere territory. However, this kind of analysis has proved to be also very interesting from the historical and archaeological perspective, particularly to determine the production ‘vocation’ of the

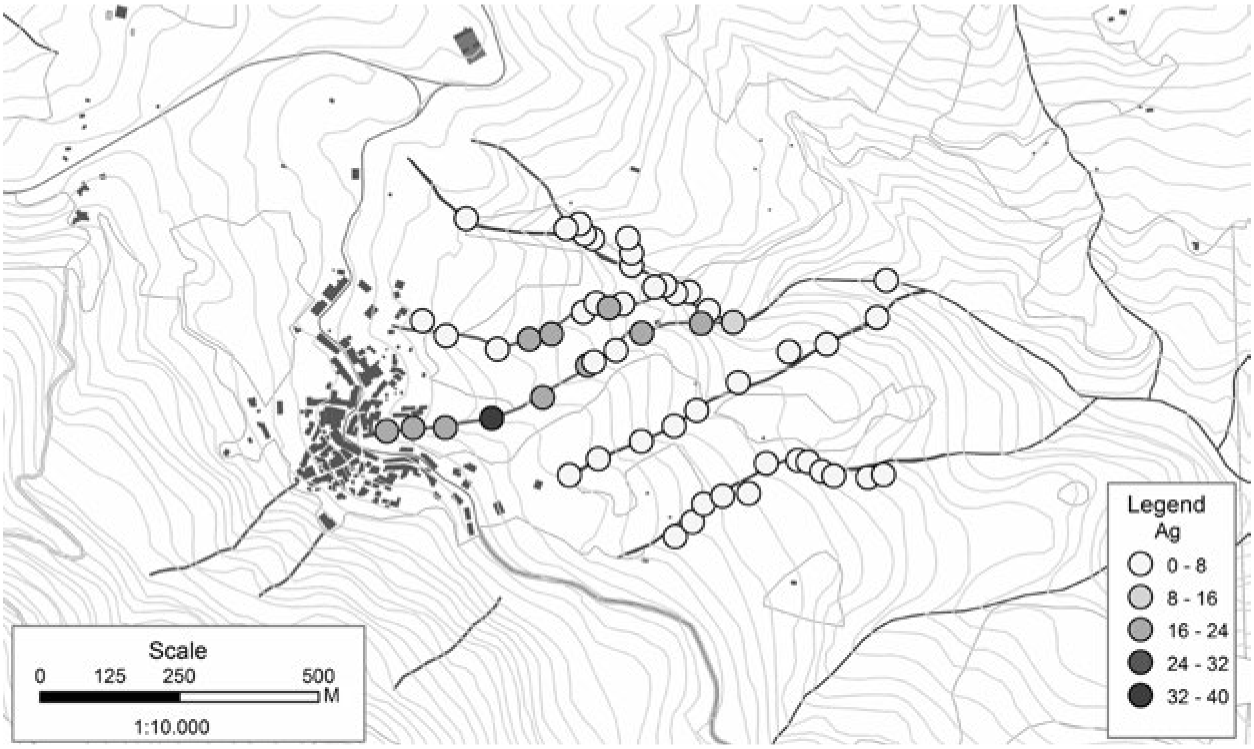

Fig. 6. Montieri's vector topographic map. It shows the results of Atomic Absorption Spectroscopy (AAS) analysis of silver distribution in the river sediments. In the legend, the concentration ranges considered.

individual mining sites, setting out from an analysis of the minerals present in the waste dumps.

Therefore this protocol could also become a valuable instrument for monitoring the local area, with a view to safeguarding the cultural heritage; to this end, the case study of the Tesoro site, in the municipality of Massa Marittima is significant: here, the major anomaly of arsenic levels indicates the probable location of an extraction area and/or mineral processing area that has never been described before (Donati *et al*. 2004, 2007).

Regarding the intra-site scale approach, an early analysis of archaeological database was done together with an assessment of other environmental parameters. After this preliminary work, a number of model sites in the 'Colline Metallifere' territory were chosen.

Among the others, one of the most interesting cases was the Altini site. This mine was located very close to Massa Marittima and it was cyclically exploited in the past. Now only few traces of the ancient activity are visible. The analysis of arsenic distribution revealed a non-homogeneous anomaly indicating the different location of the productive elements (extractive sink, mine tails deposit, processing area) and their functional connection (Fig. 7).

The environmental data collection described in the previous paragraph helped to define, on a smaller scale, the topographic limits of a large slag area, very well known from historical descriptions, nowadays partly covered by the standing town of Montieri. In fact the 18th century descriptions indicate that the mass of waste visible at that time in the city center reached exceptionally large proportions. The slags are the result of the ore extraction and metallurgical processes that went on around Montieri's village during the Medieval period. The place, as said above, was in that historical phase in fact an important mining and metallurgical center, and numerous silver mines are still partly visible in the surrounding hills and have been identified by the archaeological surveys.

To estimate the extent of this pile of slag with an initial level of topographical approximation, a large-scale screening was conducted on the river sediments, using the natural leaching process of contaminated soil as a guide. In the case in hand, lead was used as the 'tracer' element; indeed, lead is very closely connected with silver extraction (production from silver-bearing galena). Five gullies (seasonal streams), that begin lower down the town of Montieri, have been chosen for the analysis. The first four are located just below the slag heap while the fifth, named 'Madonna's river' is positioned to the east. Along the streams it was made a systematic stream sediment sampling for the laboratory analysis through the Atomic Absorption Spectroscopy (AAS) technique.

Results from the analyses showed high concentrations of lead (Pb) on the central rivers compared with the peripheral ones. This indicates that the contamination originated from an area approximately definable. The subsequent level of refinement has included chemical measurements of the soil; this has led us to define with greater precision the area where the slag probably accumulated, as detailed in Benvenuti *et al*. (2014)**.** The area that can be calculated on the basis of these figures is around 7,500 m^2. with a depth of around 4m in the southern extremity to a few dozen cm in the northern extremity (Fig. 8) (as in Fig. 3, Dallai *et al*. in press). This research has been important to determine some important chemical feature for

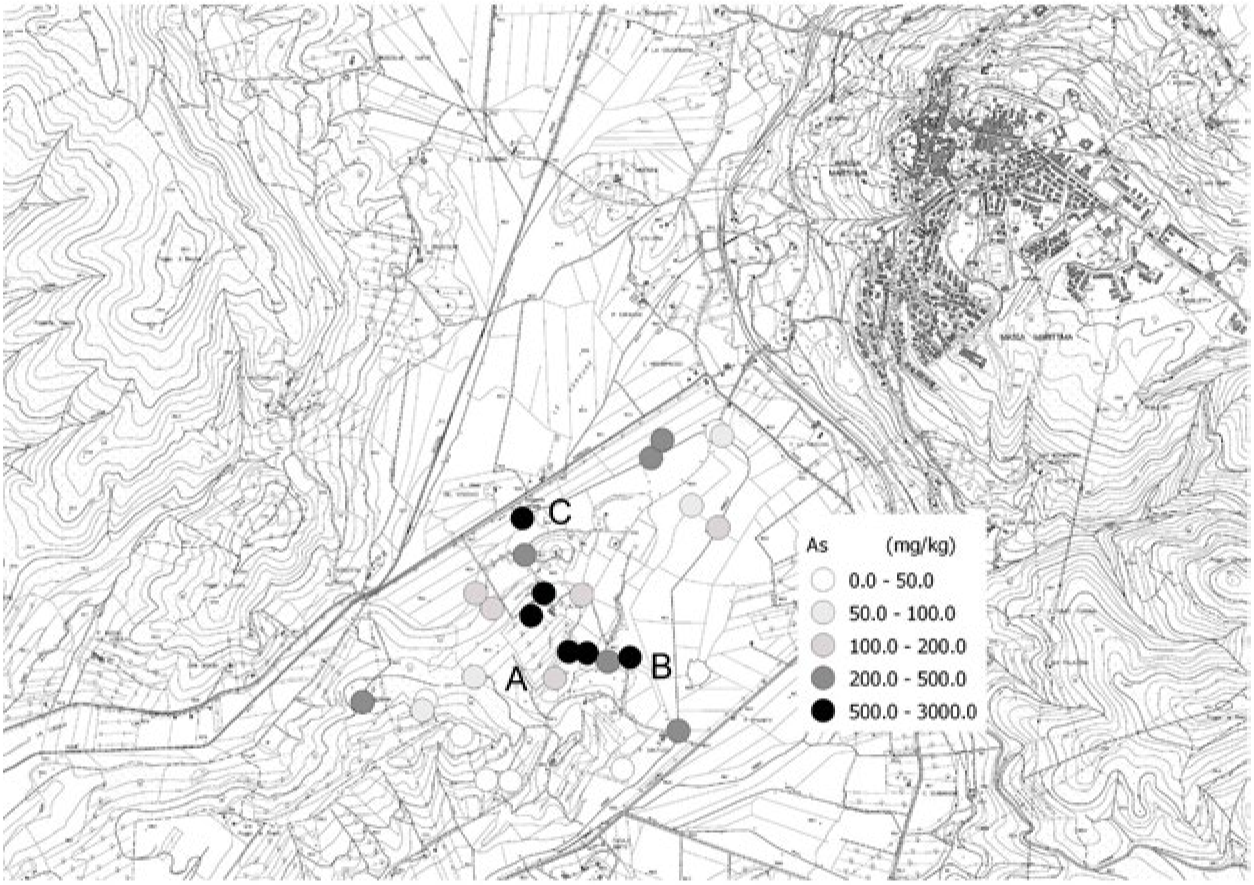

Fig. 7. Arsenic concentration in soil samples collected in the Altini site (Massa Marittima – GR). The distribution of As was non-homogeneous and with a clear north-south direction reflecting the location of the main elements of the mining activity: extraction sink (A), mine tails deposit (B) and processing area (C).

a better understanding of the local historical mining context. In particular it is crucial to find out on the one hand some productive characteristics that can help in defining the amount of mineral worked and, consequently, of metal produced. On the other hand, behind the archaeological aspect, it is useful to verify the spread of contamination and to understand to what extent the presence of metallurgical slags has affected the local environment, providing general indications on the toxic elements speciation in the area.

4 Conclusions (L.D., A.D., A.B., V.V.)

As described in the paragraphs above, the proposed research approach was applied to a context, the Colline Metallifere territory with strong peculiarities; an important mining field in the south-west of Tuscany where silver, copper, lead, iron and alum have been cyclically exploited for centuries contributing to the economic development that characterized the area through the centuries. This mining field was also recently exploited (the last pyrite mine was dismissed in 1995 year) and it is predictable that in the future mixed metal sulfides will be extracted again.

As we have seen, the territory can be properly studied by the proposed multidisciplinary approach, gathering together documental information, archaeological surveys and physico-chemical data in a smart database. Using this database, merged maps were produced and cross-correlations between ancient mining and smelting sites and the distribution of metal contamination were obtained.

This approach can be used at different scales: a context (large/medium) or local (intra-site) scale, as seen in par. 3.3. Both of them give accurate information about the localization of sites and their main use and about the mineral exploited. The difference between them resides in the respective level of detail that can be reached.

Moreover, the proposed methodology also has a great potential as predictive analysis. Indeed, it is possible to use chemical and environmental data in synergy with the databases already available for the local area, to highlight the possible consistency between the presence of geochemical anomalies, and the location of archaeo-mining and archaeo-metallurgical sites that have not yet been documented.

Once again, the Montieri area and the so called ‘Poggio’ (see par. 2), can offer good examples of cross-data management,

After a careful analysis that has evaluated historical and cartographic data, and a preliminary on-site fieldwork described above, we could define significant differences between three

Fig. 8. The concentration of lead in soil slag heap. The polygon represent the waste area.

main areas. The first one is located on the East slope of the hill; here, the combination of archaeological evidences related to mining exploitation and metallurgical activities, as well as the historical record, testify the existence of a long archaeo-mining tradition. The chemical analyses in particular have stressed the presence of high concentrations of lead (Pb), copper (Cu), silver (Ag) and arsenic (As) all over the territorial sample, with specific peaks recorded closed to possible ancient mines and metallurgical structures (i.e.: Pb 2271 ppm; Ag 38 ppm; Cu 477 pm; As 435 ppm).

The same analyses could discriminate a much lower concentration of heavy metals in a second sampled area, south of Montieri village, where the historical record again describe the existence of ancient mines. Nevertheless, their impact on the environment appears to be of less importance (i.e.: Pb 103 ppm; Ag4 ppm; Cu 67 ppm; As 100 ppm). The third and final key area of the 'Poggio' is located on the western slope, where 'La Canonica' stands. Here, the chemical values evidence, once more, only very low concentrations of heavy metals (i.e.: Pb113 ppm; Ag 1,5 ppm, Cu 30 ppm; As 118 ppm). From all the data collected in the area we could finally establish in which area of the Poggio the mining activities went on and where, on the contrary, this cannot be assess on a scientific base, although the written sources suggest the contrary.

High throughput physico-chemical data were collected (from 2002 to 2015) by using classic geo-referenced sampling of soil and/or sediments (analyzed by laboratory techniques: GF AAS; ICP-MS; XRD; FESEM – EDX) or by using on site pXRF technique. Data arising from previous geochemical prospecting and exploring activity and from environmental monitoring were also used. This gave the possibility to carry out an extremely detailed screening of the areas under investigation, and to get a very precise picture of the distribution of tracer (indicator) elements, both within an individual site and across larger geographical areas. Further comparison with data gathered during archeological monitoring, still under way, will certainly be useful to specify with even greater precision the conclusions that the analytical data are already suggesting.

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