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# MONITORING MINERAL EXTRACTION AND PROCESSING SITES IN WEST AND SOUTH WEST ROMANIA BY REMOTE SENSING-DERIVED INFORMATION AND LABORATORY ANALYSES

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**Abstract:** Samples collected from eight mineral extracting and processing sites, representing commodities of different origin and in different environments (lignite, bituminous coal, porphyry copper and gold extraction mines, copper flotation, metallurgic waste dump), were analyzed in the laboratory for: mineralogy on thin sections, X ray diffraction (XRD), gamma spectrometry, density and spectral reflectance measurements. In sample locations, estimated ground reflectance spectra were extracted from Landsat-TM images, in order to verify the OH-FeOx anomalies, obtained by processing the satellite images with a methodology previously developed for mapping mining wastes at regional scale. The processed satellite images highlighted, by means of the extent and type of OH-FeOx anomalies, the area coverage of the deposited mined material and pointed out the modifications in time. Diagnostic spectral features given by iron ferric/ferrous ions, OH-metal and/or molecular water stay at the basis of the remote sensing OH-FeOx anomalies and the minerals which they indicated, were confirmed either by the microscopic observations on thin sections, or XRD, or both. A differentiation of the sites was performed by statistically analyzing the remote sensing anomalies and comparing with the results of the microscopic analyses and XRD.

Keywords: Romania, satellite image, spectrum, Principal Components Analysis, mineralogy, XRD

## **1. Introduction**

Extraction and processing of solid raw minerals in the Romanian territory is known before Roman times, and the importance and variety of mineral resources determined mining to be a traditional activity of the population. At the end of 1989 in Romania there were 278 mines and open pits in operation and 70 processing plants: 30 for metal ores, 34 for non-metal ores and 6 for coal preparation, spread over 41 mining basins. In that period the mining sector produced 150 million tons of coal, metal ores, non-metal ores and salt per year and the mining activity ensured the daily subsistence of 10% of country population (Fodor, 2005). The extensive and intensive exploitation of ore deposits has been a characteristic of the mining industry in Romania which resulted in the occupation of large land surfaces by mining industry with mine facilities (in operation or abandoned) as waste dumps, tailing ponds, access roads, specific equipments etc. Many of these adversely affected the environment and constituted intense sources for geohazards (subsidence, terrain slides, dust and gas emissions, acid mining waters, erosion, heavy metals into the soil etc.). Passing towards the market economy implied the necessity of assessing the profitability of the mines and the problems they pose to the environment. Therefore, the mining industry restructuring strategy, approved by the Romanian Government, stipulated that only 112 mines were to be maintained into operation: 68 for coal, 34 for metal ores, 3 for uranium and 7 for salt (Fodor, 2005). The rest of the mines were closed and the reserves were put into conservation. According to the European Union legislation, remediation measures were mandatory to be applied for the closed mining facilities.

This paper presents part of the work carried out in line with this policy for the Ministry of Research and Education within the framework of the National Programme for Research and Development part II (2008-2010). The project had the purpose of making up a database of mineral extraction and processing sites (both active and closed), characterized by physical, chemical, mineralogical parameters (point information), linked with remote sensing-derived information, which gives the spatial extent of mined material, together with a qualitative characterization of the sites (fig. 1). The outputs of the remote sensing based methodology consist in GIS-integrated maps of waste material distribution for the moment of time  $t_i$  (i=1,2...n). These are linked with the database of sample laboratory measurements, which on their turn are referred to a moment of time  $t_i$  (where  $j \ge 1$ ). An ideal situation would be when the samples are collected in the same day with the remote sensing image. In reality this can be very rarely achieved, as clouds and adverse meteorological conditions might hamper the acquisition of good quality images from optical sensors over the area of interest. A good approach consists in using available old satellite images from archives, which have also the advantage of a low cost, and satellite images of the same year (preferable, if possible, the same season) as the ground collected samples. This would allow a validation of the remote-sensing OH-FeOx anomalies and the maps of changes in waste material distribution.



Fig. 1. Flow chart of the processing chain and outputs.

### 2. Study area

The field campaign took place in 2008, when eight sites (fig. 2) were visited for sample collection and ground observations. The first site, Rovinari, represents a lignite quarry, still in operation, which is by far the biggest as surface. It is located in the Subcarpathian Hills, in the Dacic Basin, Getic Zone. In an alternation of sands, sandy clays, clays and carbonaceous clays belonging to the clay horizon of Dacian-Romanian age (Borcos et al., 1984), the lignite occur as beds. A sample (PR1) was collected from the active ash storage site located between the power plant and Rovinari East quarry. Another sample (PR2) was collected from a waste dump located approximately 300 m close to an area in operation, belonging to the Gârla quarry, nearby the European road E79. The dump is made up of unstable material, which was deposited in open-pit benches, affected by weathering and gullies caused by runoff. The tailings were not fixed and there were not noted yet signs of ecological measures. However, between the dump and the road there is an agricultural cultivated field, as this part of Rovinari mining district has been already restored (Rotunjanu et Olariu, 2009).

In the second site, Vulcan, located in the posttectonic cover, in the Petrosani Depression of South Carpathians, the basin of Jiu Valley, the exploitation is done underground, the mine being active. The bituminous coal is found in beds of up to 30 m thick in the middle horizon (Upper Oligocene) of the continental-lacustrine-deltaic molasse deposits (Borcos et al., 1984). The host rocks consist of an alternation of clays, sandy marls, marls, bituminous marls, clay shales, carbonaceous shales. Two samples (PR3 and PR4) were collected from Vulcan dump and decanting pond, at a very small distance between one another. At the time of field visit, the water has been evaporated in great part, the pond appearing as a gray-coloured field made up of waste, lacking vegetation and showing lots of drying cracks. The samples are made up of rock fragments representing mine wastes in different stages of alteration, as well as black bituminous coal fragments.

The third site, Deva, represents the dump site of the porphyry copper mine, presently closed. It belongs also to the South Carpathians (Poiana Rusca Mts.), the volcanic zone of Mures Valley. The mineralization occurs as impregnations and stockwork hosted in the Miocene biotite amphibole andesites in subvolcanic facies, feldspathized, biotitized, argillized, belonging to the Pades Series. A zonation of the mineralization was observed (Borcos et al., 1984): an internal zone with alkali feldspar, biotite, bornite, chalcopyrite, magnetite (at depth with pyrite, anhydrite) and an external zone with clay minerals, quartz, locally pyrite. A sample (PR5) was collected from the dump site located north of Deva, bordered in the West by the European road E68 and the railway, and in the East by Mures river. In order to stabilize the slopes of

the deposited wastes, shrubs were planted, but without important beneficial results due to the lack of cohesion of the mineral grains, which have the aspect of light-gray, yellowish fine-grained sand. The dump is now covered with patches of grass on its upper part, and the fine material can be seen only on slopes. collecting the water and the poorly consolidated material, carried downwards by runoff. The material is visually very similar to that of Deva dump, but on the top of Uibanesti dump the vegetation is missing, only water with suspended material can be seen. The great instability of the deposited material and its lack of cohesion (sample PR6)



Fig. 2. Localization of investigated sites.

The next two sites represent dumps of deposited material originating from the mines located nearby Brad, in the Brad-Sacâramb Neogene volcanic zone of Western Carpathians. The first dump lies north-west of Brad, downstream of Uibanesti, while the other is situated in the south-east, upstream of Criscior. The dumps are made up of mined waste coming most probably from the mines of Musariu, Musariu Nou, Bradisor, Valea Morii, Ciresata, Valea Morii Veche, Caraci-Magura Tebei and Magura Tebei. All these mines are presently closed, but till twenty-thirty years ago they were operated for extracting gold and silver, accompanied in most of the cases by lead, zinc and copper. The genesis of the ores is hydrothermal, the veins, impregnations and stockworks of Miocene age being hosted in Badenian-Sarmatian-Pannonian andesites and quartz-andesites propylitized, chloritized, adularized, sericized, argillized, silicified (Borcos et al., 1984).

The dump of Uibanesti has its slopes strongly affected by gullies, although acacia trees had been planted on those. The gullies created by rain runoff are so deep that presently, at the leg of the slope, a cement channel is being built, for the purpose of represent a real danger for the urban settlements located downstream.

The dump upstream of Criscior has two water ponds on the top, the biggest one having on its shores, on about a strip of 1 m wide, some swamp vegetation. The deposited material is consolidated, but presents strong weathering. The sample (PR7) was collected from the upper part of the dump, in the first 20 cm depth from the surface. On the surface of the dump, a thin layer of soil was formed, on which scarce grass is growing.

The sixth site represents the quarry for gold at Rosia Montana, where the exploitation is temporarily stopped, waiting for a decision of the government, as its continuation, by licensing the operation to Rosia Montana Gold Corporation, implies many environmental and social problems. The mineralization of gold and silver, occurring as hydrothermal veins, stockworks and impregnations of Miocene age, is hosted in the Badenian dacites intensely adularized, sericitized, argillized, silicified, pyritized, as well as in the alternations of Badenian marls, sandstones, pyroclastics, rhyodacites and Sarmatian marls and sands, mostly argillized (Borcos et al., 1984). The big open-pit benches show the exposed rocks, which suffer from strong weathering, their colours varying from yellow to reddish-brown. The sample (PR8) was collected from the north part of the quarry, nearby a former gallery.

The next site is the porphyry copper quarry of Rosia Poieni, located, as the precedent site, in the Neogene volcanic zone of Rosia Montana – Bucium – Baia de Aries. The ore occurs as stockwork and impregnations of Miocene age in the Sarmatian amphibolic andesites in subvolcanic facies, feldspatized, biotitized, argillized, propylitized (Borcos et al., 1984). The exploitation is active and two samples were collected from the site: one (PR9) from the slope of a median open-pit bench, which shows the signs of hydrothermal alteration and weathering, visible in the whole quarry as colour variations from yellow to reddish-brown, sometimes grayish, and the other (PR10) from the flotation residues, which have a very dark colour.

The last site is the waste dump from the mineral processing platform in Zlatna, located also in the Western Carpathians, Metaliferous Mts., southern of Rosia Poieni and Rosia Montana and north of Mures valley. The city had a long tradition both in mineral extracting and mineral processing, lying in the Zlatna - Stanija metallogenetic district. Gold was initially extracted, followed in time by other metals: lead, zinc, copper. The old mineral processing plant (presently closed) produced concentrates of gold, lead, zinc, copper and pyrite. Another mineral processing plant, the biggest on the industrial platform, was active in the field of copper and chemistry, possessing a smelter (active till 1991) and units for producing copper for converters, electrolytic copper, sulfates of copper, iron and magnesium, aluminum dust and sulphuric acid. Some reduced sections are still active nowadays. Emissions of gas and heavy particles caused smoke and vapors in the surrounding area, the almost complete destruction of the vegetation and the pollution of Ampoi river with residues. The metallurgic waste dump of Zlatna (fig. 3) shows a strongly altered material with intense reddish-dark brown colours given by iron oxides (sample PR11), both at the surface, as well as in the depth. The complete lack of vegetation on the dump slopes is an indicator of its high toxicity.

## 3. Materials and methods

## 3.1. Laboratory analyses

The samples were specially prepared in order to be analyzed on thin sections in polarized light using Jenapol microscope. The mineralogical analysis were completed by XRD performed with the Bruker D8 Advance instrument. Density determinations were done using the volume displacement method and the samples were also analysed by gamma spectrometry in order to detect the content in  $U^{238}$ , Th<sup>232</sup> and K<sup>40</sup> using a digital multi-channel analyzer DSPEC jr.2.0 - Ortec with 16684 channels. Spectral reflectance measurements were done using the GER 2600 spectroradiometer, operating in the 0.35-2.5 µm range. The methodology consisted in measuring the light reflected by the surface of the sample and dividing the values to those reflected by a standard white material, which has a uniform diffuse reflection and no absorption features in the operating range of the spectroradiometer. The relative reflectance is obtained this way and, because the instrument is taking the measurement in 640 channels of very narrow bandwidth (1.5 nm for the spectral range 350-1050 nm and 11.5 nm for the spectral range 1050-2500 nm, showing practically the variation of this parameter with the wavelength, it is referred to as the relative spectral reflectance.



Fig. 3. Zlatna dump site.

While the spectral reflectance measurements were performed in order to correlate the remote sensing anomalies with the samples mineralogy determined by microscopy on thin sections and XRD, the measurements of density and gamma spectrometry had the purpose of completing the database of mineral extraction and processing sites (fig. 1) with additional parameters.

## 3.2. Remote sensing methodology

For this stage of the project we adopted a methodology developed at the Joint Research Centre (JRC) of the European Commission, Institute of Environment and Sustainability (IES), Ispra, Italy within the project PECOMINES "Inventory, regulations and environmental impact of toxic mining wastes in pre-accession countries" (Vîidea et al., 2004). The method was developed for the purpose of using multi-temporal Landsat Thematic Mapper (TM) images, which represent by their medium spatial resolution (30m) a detailed and costefficient tool for mapping mining wastes at regional level, extending up to national and continental level. Due to the good time coverage of Landsat scenes (since 1983), TM and Enhanced Thematic Mapper (ETM) data have the advantage of detecting the changes occurred in the mining zones and monitoring the material deposited in waste dumps, decanting ponds etc.

Identifying mining wastes, exclusively using remote sensing data, is based on the anomalously high concentration of the deposited material both in iron oxides/hydroxides (ferryhydrite, goethite, hematite, limonite) and in secondary alteration minerals containing hydroxyl group (OH). The remote sensing data processing consists of applying the Principal Components Analysis (PCA) in a selective way, on TM bands atmospherically and geometrically corrected. The variant of the PCA used was called Feature-Oriented Principal Components Selection (FPCS), elaborated by Crosta and McMoore (1989), subsequently modified by Loughlin (1990, 1991) and adapted by Vîjdea et al. (2004), for processing whole satellite scenes in vegetation conditions specific for the climate of Central and Eastern Europe.

The idea of using this method came from the results obtained by applying it in exploration geology. The anomalies obtained in areas with hydrothermally altered rocks and acidifications, basically reflect the same mineralogical components as in mining wastes, the most relevant groups belonging to the iron oxides/hydroxides and to those containing hydroxyl, both frequently associated to the process of pyrite weathering. This fact led to the idea of the potentiality of the method in identifying areas with waste dumps, tailing ponds, quarries, where the exposed material is much oxidized and frequently contains secondary minerals indicators of the alteration and/or acidification processes. The surface accumulation of these spectrally distinct materials is generally much higher in the case of waste dumps than in natural outcrops. Therefore, Landsat TM/ETM images have a high potential in detecting even small mining waste, if the exposed area is at least one pixel size (30 X 30m). The chain of processing Landsat scenes with this methodology is presented in figure 4.

The atmospherical corrections were performed with an open source programme developed at JRC and the University of Trier, which takes into account: 1) the different response of the satellite sensor (at-sensor radiance) during its lifetime, due to the variability in the detectors sensibility; 2) the characteristics of the atmosphere which influence the propagation of incident electromagnetic radiation, and after its interaction with the objects on Earth surface, the reflected radiation reaching the sensor. The programme is based on a modified 5S model (Simulation of Satellite Signal in Solar Spectrum), created by Tanré et al. (1990) with further developments (Hill, 1993; Lacaze et al., 1996; Hill and Mehl, 2003).

The geometrical corrections were done also by an open source program (Hill and Mehl, 2003), which had the advantage of speeding very much the tedious work of Ground Point Collection (GCPs), but any commercial software can be used, as is was the case for the PCA processing and all the rest of remote sensing applied methodology.

## 4. Results and Discussion

The objective of the remote sensing methodology was to map all oxidized areas, where there are different levels in OH-bearing secondary alteration minerals. The output anomaly OH-FeOx image has 20 anomaly classes expressed in the image legend and represent combinations of high levels of iron oxides and various levels (from low to high) of OH-bearing minerals against a gray background. The content in FeOx increases as we go from FeOx class 1 to FeOx class 4 (figure 5b) due to the fact that two Principal Components were combined for giving the FeOx image, in order to keep all types



Fig. 4. Remote sensing processing chain.

of oxidized surfaces, even those with very low reflectance. All eight investigated sites were identified on the processed images as spots with OH-FeOx anomalies, some having a higher proportion of the classes low in OH-bearing minerals and with high content in iron oxides (simple oxidation classes), while at others the classes showing high OH-bearing minerals dominate. For example, in figure 5 it is shown the tailing pond of Uibanesti (situation is referring to 1992, the satellite acquisition year), where 95.62% of anomalous pixels belong to classes 17-20. A comparison with Google image showed that the pond walls were much thicker in 2003, and the top with suspended sediments in less water. The field visit of 2009 confirmed the lack of the vegetation on the top and the big gullies on the pond slopes, despite the acacia trees planted as restoration measure. The arrow indicates the sample collection point, while white vectors represent localities of GIS integrated topographic database.

A site selected for its prevalent oxidation anomalies (classes 1-8) is the metallurgical dump site of Zlatna (figure 6), located South-East of the locality. On the dump (ground photo in fig. 3) these oxidation classes represent 79.69% of the total anomalies, although there are present also some pixels high in OH-bearing minerals (class 20), which account for 1.07%.

It must be noted that the pixels with higher content in OH-bearing minerals (classes 10-12 and 17) on figure 6b represent the pollution, expressed as smoke and gaseous emissions plus particles in the air and on the roofs, trees etc. of Zlatna, coming from the units of the processing plant which were still in operation at image date (25.09.1992).

In order to further check the remote sensing anomalies, spectra were extracted (using commercial software tools – in this case ERDAS IMAGINE) from the multispectral Landsat-TM images in the location of the samples. A comparison was made



Fig. 5. Uibanesti decanding pond. a) Landsat-TM image (4,5,3-RGB); b) OH-FeOx anomaly image.

with the laboratory measured spectra, resampled to the center bandwidth of Landsat 5 Thematic Mapper sensor (0.485, 0.56, 0.66, 0.83, 1.65 and 2.215  $\mu$ m).

Figure 7 shows the image and laboratory spectra for the samples collected at Uibanesti and Zlatna dump sites. The high content in iron oxides is proved in all 4 spectra by the sharp fall of the reflectance from TM band 3 to TM band 1. The image spectrum of Uibanesti shows an extra spectral feature: an increase of the reflectance in TM2, which is caused by the mixture water - suspended minerals, as it can be seen on the top of the dump in fig. 5a. The combination of  $Fe^{2+}$  and  $Fe^{3+}$  absorption features in these minerals cause probably also the profound minimum centered between TM3 and TM4.

The deep decrease of reflectance between TM5 and TM7 caused by the absorptions in TM band 7 are very evident on the image and laboratory measured spectrum of Uibanesti and explain the intense OH anomalies. The combined interpretation of mineralogical analyses on thin sections and XRD indicated the presence of sericite, illite, as well as carbonates, all these minerals possessing absorption features in TM7. For Zlatna, the microscopic analyses and XRD indicated epidote and undifferentiated clay minerals, as well as oxides which are responsible for the almost complete opacity of the sample. These oxides are also the cause of the low reflectance of the sample, the lowest of all measured samples.

In order to compare the spatial distribution of OH-FeOx anomalies for each studied site, the percentage of the anomaly class of the total anomalous pixels of the site was computed. This distribution is shown in figure 8, where the analysis done for the site of Vulcan at two time intervals was included. This was the only site falling in two adjoining available scenes: 184/29 (15.10.1990 – containing the sites of Rovinari and Vulcan) and 185/29 (25.09.1992 – containing Vulcan, Deva, Uibanesti, Criscior, Rosia Montana, Rosia Poieni and Zlatna).

In figure 8 it can be seen that the investigated sites are grouped in two main categories: one with dominant oxidation classes (Rovinari, Vulcan at both dates, Deva, Criscior and Zlatna) and another where the anomalies, high in both iron oxides and OH-bearing secondary minerals (classes 17-20), are the majority (Uibanesti, Rosia Montana, Rosia Poieni). The result was confirmed by a correlation



Fig. 6. Zlatna metallurgical dump site a) Landsat-TM image (4,5,3-RGB); b) OH-FeOx anomaly image.



Fig. 7. Comparison of samples reflectance for Uibanesti and Zlatna sites.

analysis, computed using these percentage of anomalies. The correlation coefficient values are positive between members of the first group, ranging from 0.18 to 0.90, with a media of 0.37. Against members of the second group, the values are slightly negative, ranging from -0.09 to -0.01, with a media of -0.08. The second waste dump group (Uibanesti, Rosia Montana, Rosia Poieni) displays positive values of the correlation coefficients between themselves (ranging from 0.18 to 0.79, with a media of 0.51) and negative values against members of the first group (ranging from -0.21 to -0.01, with a media of -0.08).



Fig. 8. Percentage distribution of OH-FeOx anomaly classes in the investigated sites.

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## **5.** Conclusions

The OH-FeOx anomalies that mapped the mineral extraction and processing sites were confirmed by the analyses done in the laboratory (even though the time difference was rather great and some modifications at the surface of the sites caused by remediation measures are to be expected). The samples represented one-point observation and the surface mapped by anomalous pixels reached in some cases tens of hectares. The remote sensing anomalies represent a useful tool for: 1) planning the field campaign by selecting in advance the sample collection points in representative anomalous areas; 2) checking by using time series (multitemporal) satellite data the efficiency of the restoration measures that were taken, by detecting the changes occurred in time.

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