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EPR, SEM and XRD investigation of ornamental limestone and marbles from some renowned Romanian quarries.

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Ornamental limestone and marble samples were collected and analysed by means of Electron Paramagnetic Resonance (EPR), Scanning Electron Microscopy (SEM) and X-Ray Diffraction (XRD), in order to evidence any systematic peculiarities able to be used in further provenance studies as well as to get more detailed information regarding geochemistry and mineralogy of three of the most important deposits from Romania. In this respect, 20 samples of limestone (Arnota quarry, Capatani Mountains and Mateias South quarry, Iezer Mountains) and 9 of calci-dolomitic marble (Porumbacu de Sus quarry, Fagaras Mountains) were collected over a significant sampling area.

EPR spectroscopy, primarily used to asset the degree of homogeneity of considered samples, evidenced, for both Arnota and Mateias South limestone, the presence of a typical six hyperfine lines spectrum of Mn^{2+} ions in calcite but no traces of Fe ferromagnetic clusters. A more careful investigation has showed that although within the same quarry, there were no significant differences regarding EPR spectra, the resonance lines were systematic narrower in the case of Mateias South samples which suggested a lower content of divalent manganese ions. The Porumbacu calci-dolomitic marble, presented a more intricate Mn^{2+} spectrum, consisting of a superposition of typical dolomitic and calcitic spectra. Again, the EPR spectra were almost identical, attesting, as in the previous cases, a relative uniform distribution of paramagnetic Mn^{2+} ions within quarry.

In the case of SEM, scattered, back scattered and absorbed electron modes were used to visualise the mineral formations on the sample surfaces while an EDAX quantitative analysis was used to determine the content of the most abundant elements. Although, at a first inspection, both groups of limestone looked almost similar, displaying a great variety of randomly orientated micro-crystalline agglomeration, only in the case of Arnota samples, we have noticed the presence of some micron size graphite inclusions, potential proxies for further provenance studies. The Porumbacu South marble showed a different pattern, characterized by a more uniform crystallite distribution, all of them presenting almost perfect cleaving surfaces. EDAX results evidenced, excepting the dominant Ca and Mg (the last one in the case of Porumbacu de Sus marble), the presence, in small quantities, of some other element such as Fe, Ni, Cu and Zn whose content represent also a good provenance proxy.

XRD investigation evidenced not only of the dominant calcite and dolomite mineral phases, but also other minor mineral fraction, whose presence could be well related to the content of mentioned trace elements.

Principal Component and Cluster Analysis, finally used to classify all investigated samples, allowed us to group them in three cluster in accordance with their provenance.