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Historic Gold Mining in the Apuseni Mountains Recorded in Stalagmite Geochemistry

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In antiquity "*Patrulaterul de Aur*" (The Golden Quadrilateral), ca. 550 km^2 of the Apuseni Mountains was one of Eurasia's richest gold and silver mining region. It is estimated that 60 – 64 M ounces of gold have been extracted from Romania during the past 2 ka, about 75% of which was sourced from this particular area (Gratian et al, 2002). Activities such as mining and smelting of ores and the associated biomass burning produced ample amounts of aerosols that were carried downwind and accumulated as wet or dry deposition. Meteoric water remobilized these trace elements by leaching from the soil as solute or particle/colloidal phase in aqueous solution through the bedrock and into the cave. Here we present the trace elemental compositions (22 elements, obtained by means of LA-ICP-MS) in a ca. 2 ka old stalagmite from the Frumoasă Cave (Trascău Mountains, SE Apuseni Mountains; Fig. 1) as a proxy for historical Au, Ag, and Cu mining and smelting. The most prominent sites (Roșia Montană, Zlatna, and Abrud) where ores were extracted and/or processed over the past 2 ka are < 30 km upwind of the Frumoasă Cave.

Fig. 1 1. Location of the study site (star).

Geologically, the area of interest pertains to the Southern Apuseni Mountains (SAM). Frumoasă Cave is hosted by Tithonic Stramberk-type limestone of the Bedeleu Nappe (Ianovici et al, 1976). Neogene magmatic rocks range from basaltic-andesites to dacites, with subordinate trachyandesites and associated intrusives (Rosu et al, 2004). Neogene gold mineralization types in the SAM are: 1. Au and Au-Ag, 2. Au-Cu-As, 3. Au-Ag-Te (Bi), 4. Au-Ag-Pb-Zn-Cu, and 5. Cu-Au (Mo), present in vein systems, breccia pipes, and stockworks (Gratian et al, 2002).

Four U/Th dates were obtained on a 30.4-cm tall stalagmite (PF109), with the lowermost sample (at 3.5 cm above its bottom) indicating an age of 2,034 ± 48 years BP. Using the age-depth model of (Scholz and Hoffmann), we show that PF109 grew between 2136 and 332 years before present, with a maximum growth rate of 132 µm/year and includes a ca. 300-year hiatus (ca. AD 1650-1350).

The stalagmite section corresponding to the Early Dacian Kingdom (ca. 125 BC to AD 87) all measured trace elements show a visible increase in concentration. During Decebalus's reign (AD 87 – 106) a new, relatively short-lived (element-dependent) spike in concentration is detected in both siderophile and chalcophile elements, while absent from the lithophile group. During the interval corresponding to the Roman rule of Dacia (AD 106 – 272) the first significant increase in elemental concentrations is present. Between AD 272 and up to the Medieval Period concentrations are maintained at relatively high levels. During the Medieval Period all concentrations slowly decreased back to base level, but rise sharply again in the Early Modern Period (a significant quasi-simultaneous spike in concentration), comparable to the behavior of the earth metals for the same interval.

Trace elemental concentrations in stalagmite PF109 are far too high to be explained by bedrock dissolution alone. Unquestionably, the elements we measured interacted with both organic and inorganic matter in the soil and bedrock, but elemental concentrations observed throughout the speleothem are well above normal values for limestone bedrock, suggesting that atmospheric input was the primary source. A covariation between the most significant concentration spikes of trace elements measured as proxies for anthropic activity (lithophiles, siderophiles, and chalcophiles) and those measured as proxies for climate variability (Mg, Sr, Ba, and P) is observed over several time intervals. This relationship is best explained by the existence of a common forcing (*i.e.*, local and regional climate dynamics): aerosols generated above the mining region accumulate downwind on the surface during drier intervals and are flushed into the caves during the wet periods.

The interpretation of the trace elemental geochemical dataset could be enhanced by an improved age model (additional U/Th dates), as well as by comparing the dataset to similar data from the bedrock, overlying soil, and clay accumulated within the cave. It would be ideal to monitor the drip water geochemistry at the tip of the "*soda straw*" that fed PF109. Stable isotope data (*e.g.*, Pb) would be an equally valuable tool to fingerprint the mineral ores from which the trace elements identified in PF109 originated.

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