

## Geochemistry of fluids from the Eastern Carpathians and Transylvanian Basin boundary (Romania)-constraints on the origin of mineral waters and dissolved gases

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The Eastern Carpathians, along the Rodna-Bârgăului subvolcanic area and Călimani-Gurghiu-Harghita volcanic chain, with the Transylvanian Basin boundary, host important resources of Romanian CO<sub>2</sub>-rich mineral waters. Results of a comprehensive study on the volatiles dissolved in artesian thermal waters discharged over a 200 km-long transect show large contents of CO<sub>2</sub>-dominated gases.

The circulation of fluids on the study area is enhanced by tectonic fragmentations which are considered to be the main upraising path for CO<sub>2</sub> and mineral waters. The geochemical features of the gas phase extracted by water samples reveal amounts of CO<sub>2</sub> up to about 2000 cm<sup>3</sup> STP/L<sub>H2O</sub>, and helium up to 0.02 cm<sup>3</sup> STP/L<sub>H2O</sub>. Our investigations show that the wide range of chemical and isotopic composition can be explained in terms of contemporary occurrence of Gas-Water Interactions (GWI) affecting the circulating waters after their infiltration. Carbon ( $\delta^{13}\text{C}_{\text{TDC}}$  Total Dissolved Carbon, ranging from -17 to +10‰ vs PDB) and He systematics (He isotopes in the range of 0.38–0.9Ra, Ra = air-normalized <sup>3</sup>He/<sup>4</sup>He ratio), coherently indicate the presence of fluids from sources located at different depths in the crust (e.g. sediments, oil reservoirs) besides minor but detectable contributions of mantle/magmatic-derived fluids.

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## Chalcophile element partitioning between silicate and sulphide liquids

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We report the partitioning of the elements Cu, In, Tl, Pb, Ag, Zn, Cr, Co, Ni, Sb, Mn and Cd between FeS-rich sulphide liquids and anhydrous basaltic melts at high P and T. There are simple relationships between the FeO contents of the silicate melts and the sulphide-silicate partition coefficients for the individual trace elements. These relationships can be generally represented as follows:

$$\log D_{M}^{\text{sulph} / \text{sil}} \approx A + \frac{n}{2} \log [\text{FeO}]$$

where A is a constant related to the free energy of Fe-M exchange, n is a constant related to the valence of the element and [FeO] is the FeO content of the silicate melt in mole fraction or weight %. At 1.5 GPa and 1400°C, with [FeO] in weight %, we report the following values of n and A:

At 1.5 GPa and 1400°C we found, with [FeO] in weight %, the following values of A and n/2: Cu (3.33; -0.82); In (2.24; -1.12); Tl (1.86; -0.76); Pb (2.64; -1.09); Ag (3.47; -0.82); Zn (1.15; -0.79); Cr (1.23; -0.87); Co (2.76; -1.09); Ni (3.65; -0.84); Sb (2.56; -1.23); Cd (2.69; -0.93); Mn (0.46; -0.59).

We calculated the composition of the putative Hadean sulphide matte extracted from primitive mantle during the final stages of accretion and possibly responsible for the current "spiky" abundance pattern of chalcophile elements in silicate Earth. Starting with the current primitive mantle abundances of these elements and calculating matte composition, however, we find that it is not possible to generate an initial abundance pattern which approximates chondritic. The simple "Hadean matte" model is inadequate.

We calculated Ce/Pb and Nd/Pb ratios of basalts generated by mantle melting. Calculated Nd/Pb is essentially constant over wide ranges of partial melting and fractional crystallization with a value of ~18.6 if we assume that depleted mantle contains 65 ppb Pb. Calculated Ce/Pb varies slightly during batch partial melting from 21–29 with the canonical value of 25 being achieved at ~10% partial melting. These trends are in excellent agreement with measurements of oceanic basalt glasses. Our partitioning relationships enable calculation of the concentrations of a number of incompatible chalcophile trace elements in depleted mantle. These are as follows: 30 ppm Cu, 65 ppb Pb, 7.6 ppb Ag, 12 ppb In, 23 ppb Cd, 1.6 ppb Sb and 1.3 ppb Tl.