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A Clusius Column in the Crust? Anomalous Noble Gas Isotope Ratios in Exhalations from the East Carpathians, Romania

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We have investigated the noble gas inventory of five gas samples and five mineral waters from the South Harghita Mountains in Transylvania, Romania. The Harghita Mountains, part of the East Carpathians, are the youngest volcanic region of eastern Europe and are regarded to have been active even subrecently. The volcanic activity of the Harghita chain was induced by subduction processes due to the closure of a Tethys remnant by collision of the Tisia-Dacia microplate and the European platform (Csontos, 1995). Close to the youngest volcano, the Ciomadul, abundant mineral water wells and dry gas exhalations (mofettes) occur.

The gas phases of all samples are dominated by CO_2 (94 - 99% vol.), accompanied by nitrogen, hydrogen and methane. The concentrations of He and Ar are small, ranging from 10 to 13 ppmv and from 14 to 71 ppmv, respectively. All water samples are oversaturated with respect to CO_2.

Elevated ^3He / ^4He ratios ranging from 1.2 to 3.2 R_A (Atmospheric ratio $R_A = 1.39 * 10^{-6}$) are evidence for the presence of a mantle component in all gas samples and in at least one water sample. Likewise, the ^{40}Ar / ^{36}Ar ratios are higher than atmospheric in three gas and three water samples. Quantitative estimates show that there is more ^{40}Ar present than can be expected based on a typical crustal ^4He / ^{40}Ar production ratio. This excess ^{40}Ar is caused by admixture of Ar from both crustal and mantle sources. Ne isotopic compositions are close to atmospheric except in two gas samples originating from dry mofettes (see below).

In most samples the elemental abundance patterns of the noble gases follow those expected for a dissolution equilibrium in water at approximately 50 ºC, except for He which is enriched by up to three orders of magnitude. In contrast, noble gas concentrations in the two mofettes mentioned above strongly deviate from this pattern showing distinct enrichments of the lighter over the heavier noble gases. Likewise, the light isotopes of Ne and Ar are up to 6.5% overabundant in these two samples. These results cannot be reconciled with mixtures of noble gases from atmospheric, crustal and mantle sources, but can only be explained by isotope fractionation. Three-isotope diagrams of Ar and Ne support mass-dependent fractionation as a possible reason for the deviations from atmospheric ratios.

Similar anomalous isotope ratios have been observed before (e.g. Nagao et al., 1981; Piperov et al., 1994), but no conclusive explanation has been given. Isotopic fractionation of that extent cannot be the result of a single step Rayleigh-type process. We propose fractionation by thermodiffusion in a setting analogous to a Clusius column. In the Clusius column, developed in 1938, combined thermodiffusion and convection effects were efficiently used for gas isotope fractionation (Clusius, 1950). In a closed system they lead to the quantitative separation of noble gas isotopes within a few weeks. The column consists of a vertical 30 - 100 m long glass tube with a diameter of 1 cm, having cooled walls and a central heating wire. The light isotopes concentrate close to the heated wire due to thermodiffusion, whereas the heavy isotopes tend to accumulate near the walls. Convection causes the gases to circulate, further amplifying the separation process and concentrating the lighter isotopes towards the top of the column.

In our model the Clusius column is replaced by gas conduits in the crust below the mofette exhalations. The elongated heat source is represented by the hot, continuous, upward moving gas stream, whereas the column wall is formed by the cooler crustal rocks. This setting will cause a thermodiffusion effect between the hot center of the gas flow and the cool wall rocks. The light isotopes are concentrated in the center of the conduit and become enriched near its upper end. Of course the efficiency of this system would be much smaller than for a laboratory device, but due to the long pathway through the crust, a significant enrichment of the light over the heavier isotopes is likely to occur.

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