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Physicochemical characteristics of drip waters: Influence on mineralogy of recent cave carbonate precipitates

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Speleothems are one of the most intensively explored archives of palaeoclimate variability in continental settings. Considerable advances with respect to climatic and cave forcing of drip characteristics and related speleothem proxy data have been made during the last decades. The parameters, however, that control speleothem mineralogy and its changes with time and space are still poorly understood. In order to shed light on processes influencing speleothem mineralogy, precipitation experiments of recent carbonate crystals on watch glasses and glass plates were performed in seven selected caves. These include three caves in Germany as well as Morocco and one cave in Romania, which are situated in both limestone and dolostone.

Drip water sites of these caves were analysed for their fluid Mg/Ca molar ratio, pH, degree of saturation for calcite and aragonite and drip rates. Corresponding precipitates were analysed with respect to their mineralogy using a high resolution scanning electron microscope (SEM). The following results are found: High fluid Mg/Ca ratios are observed only for caves situated in dolostone, hence the hostrock lithology indirectly controls the carbonate mineralogy of speleothems. The precipitation of aragonite in place of calcite occurred only in dolostone caves and is bound to very specific conditions, which are: high fluid Mg/Ca ratios (> 0.5), high fluid pH (> 8.2) and low fluid saturation indices for calcite (< 0.8). These specific conditions are induced by slow drip rates of < 0.2ml/min (often under more arid conditions), causing the precipitation of calcite / aragonite prior to reaching the stalagmite top. Due to this, fluid chemistry is altered, which in turn leads to changes in carbonate mineralogy and geochemistry on the stalagmite top. Interestingly, all of the above mentioned factors must act in a concerted manner. If this is not the case, calcite is the dominant phase. The threshold, where only aragonite precipitates is at fluid Mg/Ca ratios exceeding 2.4. Generally, calcite growth is inhibited at high fluid Mg/Ca ratios and hence, aragonite precipitation is kinetically stabilized. On the other hand, aragonite precipitation is possibly inhibited by some types of organic compounds leading to calcite precipitation, whilst it is induced in other cases. Based on the data shown here, the parameters inducing aragonite precipitation are now clearly better understood. Thus, conclusions of drip water palaeo-conditions from aragonite speleothems can be drawn, which leads to an improved understanding of aragonite speleothems as climate archives.