

## Paleohydrogeochemical study of the deep granitic rocks by using carbon isotope and morphology of carbonate minerals

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This study aimed to develop a methodology for assessing the hydrochemical evolution of a groundwater system, using fracture-filling and fracture-lining calcite. Fracture calcite in deep (to c. 1,000 m) granitic rocks of the Tono area, central Japan, was investigated by optical and electron microscopy, chemical and isotopic analysis. Coupled with geological evidence, isotopic and morphological data imply three main origins for the waters that precipitated the calcite, 1) relatively high-temperature hydrothermal solutions, 2) seawater, 3) fresh water. The calcite morphological data, coupled with isotopic data, could provide a powerful palaeohydrogeological tool in such circumstances.

This study aimed to develop a methodology for assessing the hydrochemical evolution of a groundwater system, using fracture-filling and fracture-lining calcite. Fracture calcite in deep (to c. 1,000 m) granitic rocks of the Tono area, central Japan, was investigated by optical and electron microscopy, chemical and isotopic analysis. Coupled with geological evidence, these new data imply three main origins for the waters that precipitated the calcite:

- relatively high-temperature hydrothermal solutions, precipitating calcite distinguished by  $\delta^{18}\text{OSMOW}$  from -3 to c. 10 permil and with  $\delta^{13}\text{CPDB}$  from c. -18 to -7 permil.

- seawater, probably partly of Miocene age, which precipitated calcite distinguished by  $\delta^{13}\text{CPDB}$  of c. 0 permil and  $\delta^{18}\text{OSMOW} > \text{c. } 20$  permil.

- fresh water, with a variable  $\delta^{13}\text{CPDB}$  composition but which precipitated calcite distinguished by  $\delta^{13}\text{CPDB}$  that was significantly  $< 0$  permil and as low as c. -29 permil and  $\delta^{18}\text{OSMOW} > \text{c. } 17$  permil.

The isotopic data also imply that fresh water flushed marine water from the deep sub-surface. However, the marine calcite of probable Miocene age shows no evidence for dissolution. Thus, the deep groundwaters probably remained calcite-saturated during the flushing, owing to buffering by water/rock interactions up-stream along the flow paths followed by the fresh groundwater. Some of the calcite also contains  $^{14}\text{C}$ , indicating that it formed less than 50,000 years ago and suggesting that this buffering has occurred continuously since the Miocene. Consequently, many different calcite crystal forms have been preserved. Studies of other groundwater systems have correlated similar crystallographic variations with variations in the salinity of coexisting groundwaters. When this correlation is applied to the Tono observations, the calcite crystal forms imply a similar range of groundwater salinity to that inferred from the isotopic data. Thus, the present study suggests that even in presently low-salinity groundwater systems, calcite morphological variations may record variations in the salinity of coexisting groundwaters. It is suggested that calcite morphological data, coupled with isotopic data, could provide a powerful palaeohydrogeological tool in such circumstances.