

# U Isotope Systematics in Four European Stalagmites: New Insights and Implications for Palaeoclimatic Reconstruction

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U-series dates have been used to provide the chronology for many recent studies of continental palaeoclimate as recorded by speleothems. However, the accompanying precisely measured  $^{234}\text{U}/^{238}\text{U}$  ratios, as records of the U isotopic composition in cave drip-water, contain additional insights that have yet to be explored fully. A recent study by Kaufman et al. (1998) for example, revealed that time-series variations in  $(^{234}\text{U}/^{238}\text{U})_i$  in speleothems from the Soreq cave in Israel exhibit a broad positive correlation with  $\delta^{18}\text{O}$ , such that drier conditions produced higher  $(^{234}\text{U}/^{238}\text{U})_i$  and higher  $\delta^{18}\text{O}$ . Here we present new data for four European Holocene speleothems to explore further the possibility that U isotope ratios can provide useful palaeoclimatic information.

U contents and  $(^{234}\text{U}/^{238}\text{U})_i$  ratios vary widely between the four stalagmites, but intra-stalagmite ranges are quite restricted. Typical ranges are listed in Table 1.

$(^{234}\text{U}/^{238}\text{U})_i$  ratios in the Pere Noel stalagmite (Belgium) decrease systematically with time from 12,877 to 3672 years ago. Interestingly, Sr isotope ratios in this stalagmite also exhibit smooth decreases with time, from 0.7090 to 0.70881; interpreted by Verheyden et al. (in press) as mixing between limestone- and weathered soil-derived Sr. In this interpretation, decreases in  $^{87}\text{Sr}/^{86}\text{Sr}$  with time require a higher contribution of Sr from the limestone relative to the more radiogenic soil silicate phases. The uranium and strontium isotope data can be reconciled if the  $(^{234}\text{U}/^{238}\text{U})$  ratio of uranium contributed by the weathered material decreased with time due to preferential leaching of  $^{234}\text{U}$ . If this decrease was sufficient to counter-balance any increase in the amount of limestone-derived U it could result in an overall decrease with time in the mixed  $(^{234}\text{U}/^{238}\text{U})$  ratio. An alternative explanation would be that if the climate had been getting wetter through the time (as proposed by Verheyden et al. in press), drip-waters would have progressively shorter water-rock interaction times, resulting in lower  $(^{234}\text{U}/^{238}\text{U})$  due to decreased recoil-emitted  $^{234}\text{U}$ .

The stalagmites from Crag (S.W. Ireland) and Grotte de Clamouse (S.E. France) have higher  $(^{234}\text{U}/^{238}\text{U})_i$  ratios when

stalagmite growth-rates are high. McDermott et al. (1999) demonstrated that higher growth rates coincide with higher  $\delta^{18}\text{O}$  values. In principle high  $\delta^{18}\text{O}$  values might reflect warmer conditions, or in semiarid regions, high  $\delta^{18}\text{O}$  could predominantly reflect drier conditions (e.g. Soreq Cave, Israel, Bar-Matthews et al. 1999). Because high  $\delta^{18}\text{O}$  is accompanied by higher speleothem growth-rates at Crag and Grotte de Clamouse, McDermott et al. (1999) argued that temperature was the dominant control on  $\delta^{18}\text{O}$  at these sites. We propose that during the warmer periods, weathering rates were higher, and so the relative contribution of U from fresh unweathered minerals increased to give higher  $(^{234}\text{U}/^{238}\text{U})$  in the drip-waters. Therefore, we infer that increased chemical weathering rates in response to warmer conditions were responsible for shifts to higher  $(^{234}\text{U}/^{238}\text{U})$  in the Grotte de Clamouse and Crag stalagmites.

In contrast with all of the other stalagmites studied here, the  $(^{234}\text{U}/^{238}\text{U})_i$  ratios in the Grotta di Ernesto (N.E. Italy) stalagmite show a clear negative correlation with Th/U ratio and Th contents. New dissolved organic carbon (DOC) data indicate that organic material incorporated into this low-U stalagmite may be responsible for the observed variations in  $(^{234}\text{U}/^{238}\text{U})$  and Th concentrations. Overall, this study offers new insights into the factors that control U isotope ratios in stalagmites and shows that site-specific effects must be well understood before this proxy can be reliably applied.

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