

## Precise measurement of $^{226}\text{Ra}/^{230}\text{Th}$ disequilibria in deep-sea sediments by high-sensitivity ICP-MS

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We describe a new method suitable for the precise and accurate determination of  $^{226}\text{Ra}$  in porewater and sediment samples using a single-collector sector field ICP-MS (ThermoFisher Element XR) equipped with an Apex-Q desolvation device and a high-sensitivity Jet-X interface. In combination with  $^{230}\text{Th}$  measurements in parallel sediment samples, this method allows precise and accurate quantification of the  $^{226}\text{Ra}/^{230}\text{Th}$  disequilibria in surface sediment cores, thereby enabling the use of this isotope pair as a tracer of solute transfer across the sediment-water interface in the deep ocean. The method integrates a step of isotope dilution with  $^{228}\text{Ra}$  as an internal spike, a pre-concentration of Ra and Ba by  $\text{MnO}_2$  precipitation, and an efficient separation of Ra from other undesirable elements using a cation exchange resin and a Triskem Sr-spec resin. With the inclusion of one or two additional cation resin columns and the use of up to 16 bed-volumes of a lower molarity (1.7 M) HCl eluent, our procedure eliminates the complicated matrix effects persistently encountered in previous studies, and provides a highly purified solution suitable for  $^{226}\text{Ra}$  measurement using an Element XR ICP-MS apparatus. Consequently, we are able to determine the activity of  $^{226}\text{Ra}$  in ~20-50 ml of porewater or 100 mg of sediment with an internal precision of ~1.0% and an accuracy of ~99.2%. The precise measurements of porewater and solid phase  $^{226}\text{Ra}$  in a sediment core from the North Pacific Ocean allowed the distribution coefficient ( $K_d$ ) of  $^{226}\text{Ra}$  to be constrained tightly within a range of 4700-11600 ml g<sup>-1</sup>. Moreover, with the aid of a one-dimensional exchange model, the combination of the  $^{226}\text{Ra}$  and  $^{230}\text{Th}$  measurements allowed us to estimate a  $^{226}\text{Ra}$  flux of  $1140 \pm 20$  dpm m<sup>-2</sup> y<sup>-1</sup> from the sediment core.