Routine high-precision U-Th isotope analyses for paleoclimate chronology

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A number of proxy archives, such as speleothems have annual banding. Often their utility for annual scale reconstruction is limited by lack of precision in the absolute ages in part due to poor analytical uncertainties. These analytical uncertainties may result from a number of factors, including: 1) low ionization efficiency, 2) Faraday-multiplier gain stability during multiple jumps of small signals (^{234}U, ^{233}U and ^{236}U, for example), 3) mass fractionation, especially for ICP-MS measurements where fractionation can be severe. Although ICP ionization results in much higher ion yield than thermal ionization for thorium, up to a factor of 50 for dry aerosol, a number of the isotopes of interest are still too small to be measured on Faraday cups with 10^{11} Ω feedback resistors. Here we describe an analytical approach that uses a new generation of 10^{12} Ω feedback resistors that bridge the gap between 10^{11} Ω feedback resistors and electron multipliers. These 10^{12} Ω resistors provide a factor of 10 improvement in gain with only a factor of three increase (square root of resistance) in the Johnson noise (thermal voltage noise), resulting in about a factor of 3 improvement in the signal/noise ratio. The experiments were carried out using the Finnigan Neptune MC ICP-MS.

Given the significant signal/noise ratio advantage of the 10^{12} Ω resistors two strategies have been considered. In the first case the minor spike peaks, ^{229}Th, ^{233}U and ^{236}U were measured on Faraday cups, with the following arrangement: ^{238}U (10^{10} Ω), ^{236}U (10^{12}), ^{235}U (10^{11}), ^{234}U (SEM), ^{233}U (10^{12}), ^{230}Th (channeltron), ^{229}Th (10^{12}). The ^{236}U/^{233}U ratio is used to correct U and Th mass fractionation. The added advantage in analyzing U and Th isotopes simultaneously is that any possible effects of instrumental fluctuations on U-Th isotope ratios are minimized. Using this scheme we obtained the following preliminary data for a mixed U [CRM-145 (NBL-112a)]-Th standard: δ^{234}U -36. 6 ± 0.3 (2-σ) (accepted value -37.1 ±1), ^{230}Th error of 0.3% (2-σ) on about 180 ng of U corresponding to 10 pg of ^{234}U and ~ 0.34 pg of ^{230}Th using an Aridus desolvating nebulizer (DSN), and δ^{234}U -37. 1 ± 0.5 (2-σ), ^{230}Th error of 0.5% (2-σ) for the same sample sizes without using a DSN. These values were obtained after establishing the Faraday-SEM and Faraday-channeltron gain. This approach should result in significant improvements in paleoclimate chronology at all time scales.

Data for the second approach, in which all U isotopes are measured on Faradays, will be presented at the meeting.