Precise determination of U-Th-Ra disequilibria in natural samples using TIMS with sequential chromatographic separation of U, Th and Ra

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U-series disequilibria in natural samples is an important tracer to understand various geological processes occurring on a timescale from 10^6 years to <1 year. Recent technical improvements of multi-collector ICP-MS demonstrated a potential of precise determination for U-Th-Ra disequilibria in natural samples better than TIMS. However, TIMS still has some advantages compared to ICP-MS: 1) smaller instrumental mass fractionation, 2) negligible memory effects, 3) smaller isobaric interferences due to hydrocarbons and/or hydrides, 4) enough very high abundance sensitivity to eliminate ^{232}Th tailing, and 5) larger ionization efficiency for Ra. Here we present precise determination of U-Th-Ra disequilibria in natural samples by isotope dilution TIMS with an improved method of sequential chromatographic separation of U, Th and Ra.

Good isolation of U, Th, and Ra is indispensable for both ICP-MS and TIMS analyses to avoid various analytical problems on mass spectrometry. A powdered silicate rock sample (~0.5 g) was weighed and mixed with ^{229}Th, ^{233}U, and ^{228}Ra enriched spikes simultaneously. Spike addition prior to decomposition is very important to prevent potential sample loss during decomposition. The sample was decomposed with $HF\text{-}HClO_4$ and finally conditioned by 5M HNO_3. The solution was loaded on 0.5 mL of U/TEVA·spec resin, and rinsed in the order of 4M HNO_3, 5M HCl and 0.1M HNO_3 to collect major elements (containing Ra), Th, and U, respectively. Th and U fractions were further purified by anion exchange resin (AG1X8). Major elements in the Ra fraction were removed by passing through cation exchange resin (AG50WX12). Ra was then separated from Ba, LREE, and organic matters by using Sr·spec, TRU resin and CG71. General recovery yields were ~90% for U, Th, and Ra.

TIMS analyses were performed on a Finnigan MAT262 equipped with RPQ_{plus} ("*INU*"). ^{230}Th/^{232}Th and Th content were simultaneously determined in a single run, as were ^{234}U/^{238}U and U abundance. Analytical reproducibility was tested by using a GSJ rock standard (JB-3), and was 0.7%, 0.2%, 0.4%, 0.1%, and 0.6% for ^{230}Th/^{232}Th, ^{234}U/^{238}U, Th content, U content, and ^{226}Ra content, respectively. Ionization efficiencies were comparable or higher (especially for Ra) than previous TIMS analyses, which were achieved by the excellent purification of U, Th and Ra by our technique. Therefore, our separation method must be also effective for the determination of U-Th-Ra disequilibria using MC-ICP-MS.

A demonstration of our technique for U-Th-Ra systematics of Miyakejima volcano, Izu arc, Japan, will also be presented.

Large sea-level excursions during the Marine Isotope Stages 4 and 3 obtained from Huon Peninsula uplifted coral terraces

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Huon Peninsula, Papua New Guinea, is situated at a tectonically active location. The uplifted terraces enable collection of sea-level records from the Last Ice age when sea-levels were significantly lower than present. We have been investigating the sea-level history of this period by dating fossil corals. Here we report further evidence of sea-level changes during the MIS3 as well as during MIS4. We dated over 20 samples using the Neptune Multi-Collector Sector ICP-MS at the RSES, ANU. The results show the following: (i) sea-levels during the MIS3 varied between about –50 to –90m, (ii) the transition between MIS5 and MIS4 was characterized by a very large and rapid sea-level fall (iii) initial uranium 234/238 values are lower than those for modern corals but consistent with results from our earlier study using TIMS. The large sea-level fall (about 50-60m in 6 kyr) at the end of the MIS5, which consistent with the observations from Barbados terraces (Cutler et al., 2003; Potter et al., 2001), indicates that continental ice sheets were able to grow very rapidly. The magnitude of this sea-level fall is equivalent to the previously reported sea-level excursion at the MIS6 to 5 boundary from Aladdin's Cave (Esat et al., 1999) and also similar to the MIS3 to MIS2 transition determined from Huon Peninsula, PNG and Bonaparte Gulf, Australia (Yokoyama et al., 2000; Lambeck et al., 2002). This suggests that continental ice sheets may be very sensitive to the changes in global climate.

References

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