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High-precision stable isotope measurements of tungsten and molybdenum in single sample aliquots combined with optimized separation for mixed double spikes

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Mass-dependent fractionation of W isotopes has been an emerging tool in recent years that can potentially constrain a wide range of geochemical processes in the crust/mantle systems and low temperature environments. In particular, this isotope system is expected to improve our understanding of the Earth system in combination with the Mo isotopes, which has successfully been established in the past two decades as a proxy, particularly for the oxygenation of Earth's ocean and atmosphere, but also still been expanding its applications to the solid earth and other areas including ore deposit, oil, and anthropogenic tracing. Here, we establish a combined double-spike (DS) methods for W (^{180}W-^{184}W spike) and Mo (^{97}Mo-^{100}Mo spike) to perform simple, efficient, and robust isotope measurements of these two chemically analogous elements in single sample aliquots, which could be a robust basis for the development of the stable isotope geochemistry of W and Mo.

Based on the previous column chemistry, we optimized two-stage anion-exchange procedures to remove matrix elements, particularly the critical interferences of Ta and Hf on ^{180}W, and to collect sharply separated W and Mo fractions. The obtained recoveries are quantitative for both elements, and their purities are sufficiently high to achieve high-precision measurements comparable to previous DS measurements of individual elements. The reproducibility of our isotope measurements for in-house standard solutions (2SD) were ±0.02‰ for δ^{186}W and ±0.03‰ for δ^{98}Mo. We applied our method to 27 geochemical reference materials including 10 igneous rocks (AGV-2, JA-3, JR-1, JB-1, JB-1a, JB-2, JB-3, W-2a, TDB-1, WGB-1), 9 sediments (Nod-A-1, Nod-P-1, JMn-1; JMS-1, JMS-2, CRM7302-a, HISS-1, MESS-4, PAC-3), and 8 sedimentary and metasedimentary rocks (SDC-1, SDO-1, SBC-1, SCO-1, SCO-2; JSL-1, JSL-2, IOC-1) to produce a comprehensive data set. The data set confirmed the accuracy of our measurements and expands the reference materials available for interlaboratory comparisons of δ^{186}W and δ^{98}Mo. The data set also indicates potential pitfalls in sample preparations for particular sample types, and shows several variations of W and Mo isotopes possibly related to low-/high-temperature geochemical processes.