Pushing the analytical limits of Si isotope analysis using a novel double spike approach

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The advent of multi-collector ICPMS in the early 2000's marked a major step forward in analytical precision of silicon isotope measurements compared to traditional gas source mass spectrometry. Relatively small Si isotope fractionation during high-temperature processes could now be resolved and used to study, for instance, planetary accretion and differentiation. Despite these marked improvements, the reproducibility and accuracy of Si isotope measurements are still hampered by matrix effects, fractionation during sample decomposition or Si separation and interlaboratory bias. Double spiking has been the solution to most of these problems, but can conventionally only be applied to elements with at least four isotopes. It is, however, possible to extend the double spike technique to 3-isotope elements if double spike and sample are added in a "critical mixture" [1] and this has recently been demonstrated for Mg isotope analyses [2].

We report on the development of the critical mixture double spike technique for Si isotope analyses with the aim to achieve 20 ppm (2 SD) reproducibility. Such improved precision opens a range of possibilities for geochemical research. A main objective is testing the giant impact hypothesis for the origin of the Moon, as previous work could not resolve a significant difference between the Moon and the bulk silicate Earth [3]. In addition, the Si isotope composition of seawater is notoriously difficult to measure due to strong matrix effects, which would be obviated when employing the double spike approach.

Measuring Si isotopes at 20 ppm reproducibility requires maximising the internal precision of individual measurements. In addition to well-resolved polyatomic interfering species such as NO^+ on ^{30}Si, we found that presence of ^{28}SiH and tailing from the NO^+ beam are significant and have to be properly resolved or corrected for. We will illustrate the importance of these effects and present new double spike measurements now these impediments have been addressed.

[1] Coath et al. (2017) *Chem. Geol.* **451** p. 78-89. [2] Hin et al. (2015) *AGU abstract V23D-06.* [3] Armytage et al. (2012) *GCA* **77** 504-514.