Hydrogen and Triple Oxygen Stable Isotope Composition of Water in Hydrated Carbonaceous Chondrites

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Much of what is known about the origin and evolution of water in the Solar System comes from measurements of H stable isotope compositions in meteorites and other astromaterials. Yet, considerable uncertainty remains about the origin of the Solar System's water (and thus Earth's) and whether it came from water-rich chondritic meteorites that coalesced in the inner Solar System, or those that formed further out in the region of the gas giant planets. The H and O stable-isotope ratios ($^{2}H/^{1}H$, $^{17}O/^{16}O$, $^{18}O/^{16}O$; expressed here as δ values) of water and hydroxyl (OH-) in hydrated minerals of meteorites provide important clues about the water's origin. Limitations in previous techniques to analyse H isotopes in these materials lead to comingling of H from different minerals and organics into a bulk pool, severely limiting interpretations. Furthermore, it has not been previously possible to measure the isotopic compositions of O and H in the same samples despite the scientific advantages such a technique would provide. Here, we detail the application of a TGA-IRIS (thermogravimetry + isotope spectroscopy) analytical system to measure mineral-specific water and hydroxyl-bound H and O isotope compositions and abundances in hydrous minerals from terrestrial samples and carbonaceous chondrites.

TGA-IRIS results from a suite of terrestrial minerals (gypsum, lizardite, kaolinite, goethite, serpentine, cronstedtite, greenalite, chamosite, nontronite, saponite) give reproducibility in the range of $\delta^{2}H \pm 0.5$ to 2.3‰, $\delta^{18}O \pm 0.08$ to 0.48‰, $\delta^{17}O \pm 0.07$ to 0.27‰, and $\Delta^{17}O \pm 0.043$ to 0.100‰. The minimum sample size ranges from 3 mg to 30 mg for materials with 20 to 2 wt.% water. Experiments using rehydration of anhydrite for conversion to gypsum with water of known $\delta^{2}H$, $\delta^{17}O$ and $\delta^{18}O$ isotopic composition indicate that TGA-IRIS is able to recover gypsum hydration water with fractionation factors that match literature values. In Murchison, preliminary isotope values of water in Fe (oxy)hydroxides are $\delta^{2}H$=-55 ‰, $\delta^{18}O$=7.0 ‰, and $\Delta^{17}O$=0.17‰, suggesting that this water is of terrestrial origin. While in phyllosilicates (cronstedtite) $\delta^{2}H$=43 ‰, $\delta^{18}O$=10.8 ‰, and $\Delta^{17}O$=-1.09‰, suggesting an extraterrestrial origin. These values are similar to the one available literature measurement of volatile oxygen in Murchison (Baker et al., 2002, Anal. Chem.). TGA-IRIS presents the potential to generate thermal water release profiles, and coupled H and O isotopic data from water in meteorites from across the known spectrum of types, as well as materials from the upcoming sample return missions of Hyabusa-2 and OSIRIS-Rex.