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Determination of Water D/H in Hydrated Chondrites using NanoSIMS Imaging

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Introduction: Hydrogen isotopic compositions (D/H or □□D) in chondrites are a powerful tool for deciphering the source of water delivered to terrestrial planets (1). CM-type carbonaceous chondrites contain up to ~10wt.% H_2O, retained as OH in phyllosilicates. The D/H ratio of phyllosilicates (a direct proxy for water) in chondrites cannot be determined directly using whole rock measurements, because their matrices also accreted D-rich organics which are mixed with D-poor phyllosilicates at the sub-micrometer scale. To address this issue, water D/H has been estimated by in-situ measurements of both D/H and C/H in hydrated chondrites, which define a mixing line in a D/H vs. C/H plot. The intercept gives the isotopic composition of the phyllosilicate alone (1). However, SIMS measurements of water D/H using this method can be compromised by *(i)* contamination and *(ii)* limited dispersion of the phyllosilicates/organics ratio measured with a large primary beam.

Methods: We addressed both issues using the Wash U NanoSIMS50 which allows us to obtain coordinated isotopic and elemental data with high-spatial resolution. H^-,D^- with $^{12}C^-$,$^{12}C^{14}N^-$,$^{12}C^{15}N^-$,$^{28}Si^-$ are collected using magnetic-field peak-jumping in "Combined Analysis" mode. Centering of the secondary ions beam in Cy and P2/P3 planes of the secondary column changes between the low and high masses, resulting in misaligned ion images. So, we used AutoHotkey scripts to send a different Cy voltage for every B-field set up through the virtual keyboard of the NanoSIMS. To separate phyllosilicate-rich from organic-rich pixels, we assume that D/H is not simply a linear function of C/H, but in general D/H is approximated by a function using all measured species: $D/H = C^a N^b Si^c H^d$. The true phyllosilicate composition [C,N,Si,H] is estimated from the data and is then used to estimate the water D/H composition from the linear regression model. NanoSIMS isotopic analyses were carried out in a matrix area of the CM Maribo and our analytical conditions were the same as outlined in (2).

Results: First, we calculated a □□D value of −178±46‰ (2σ) for the phyllosilicates in Maribo using the D/H vs. C/H correlation from the resized pixels. This value is higher than previous measurements using SIMS [□□D ≈ −420 to −270‰, (2, 3)], demonstrating that D/H ratio of phyllosilicate cannot be simply determined using the D/H *vs.* C/H line in this matrix area. Second, we calculated the □□D value of the phyllosilicates in Maribo using all the measured species and the

linear regression model described above. We found that the phyllosilicate D/H is best correlated for dominant contributions of N, Si and H (b=0.14, c=0.58 and d=−0.86) and minor contributions of C (a=0.06). We calculated a □□D value of −286+/-60‰. This value is consistent with those previously determined by SIMS, demonstrating that our method can be used to precisely determine the water D/H on very small areas.

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