Supporting Figure S2 <u>Methods:</u> The phantom experiment was performed on a 7T whole-body MRI scanner (GE Healthcare, Waukesha, WI). An RF surface coil was used for both transmission and reception. The thermal ^{13}C phantom was made using a 21.7 mm diameter cylinder syringe filled with ethylene glycol (HOCH$_2$CH$_2$OH, anhydrous, 99.8%, Sigma-Aldrich, St. Louis, Missouri). A gradient-echo ^{1}H image was acquired with TE/TR = 2.5/100 ms, nominal flip angle = 30°, FOV = 80 × 80 mm^2, matrix size = 256 × 128 (zero filled to 256 × 256), and slice thickness = 3 mm. 2D dynamic

MRSI scans with circular full k-space coverage and prospective two-fold undersampling were both performed with TE/TR = 2.9/80 ms, flip angle = 20º, FOV = 3 × 3 mm^2, matrix size = 8 × 8, slice thickness = 20 mm, no averages, readout bandwidth = 10000 Hz, and 5 frames. <u>Results:</u> The top panel shows a prospectively two-fold undersampled k-space (white) overlaid on another fully acquired k-space (purple). The bottom panel shows the full k-space and CS reconstructed spectra. The voxel-wise signal intensities of undersampled k-space data were linearly correlated with those of the full k-space data (slope = 1.02 and R^2 = 0.94 across 31 voxels in abovementioned two datasets), confirming the feasibility of the proposed method. The triplet spectra of ethylene glycol (J_{C-H} = 150 Hz) were well recovered by our method. The two bright bold lines indicate the location of the surface ^{13}C RF transceiver coil used.