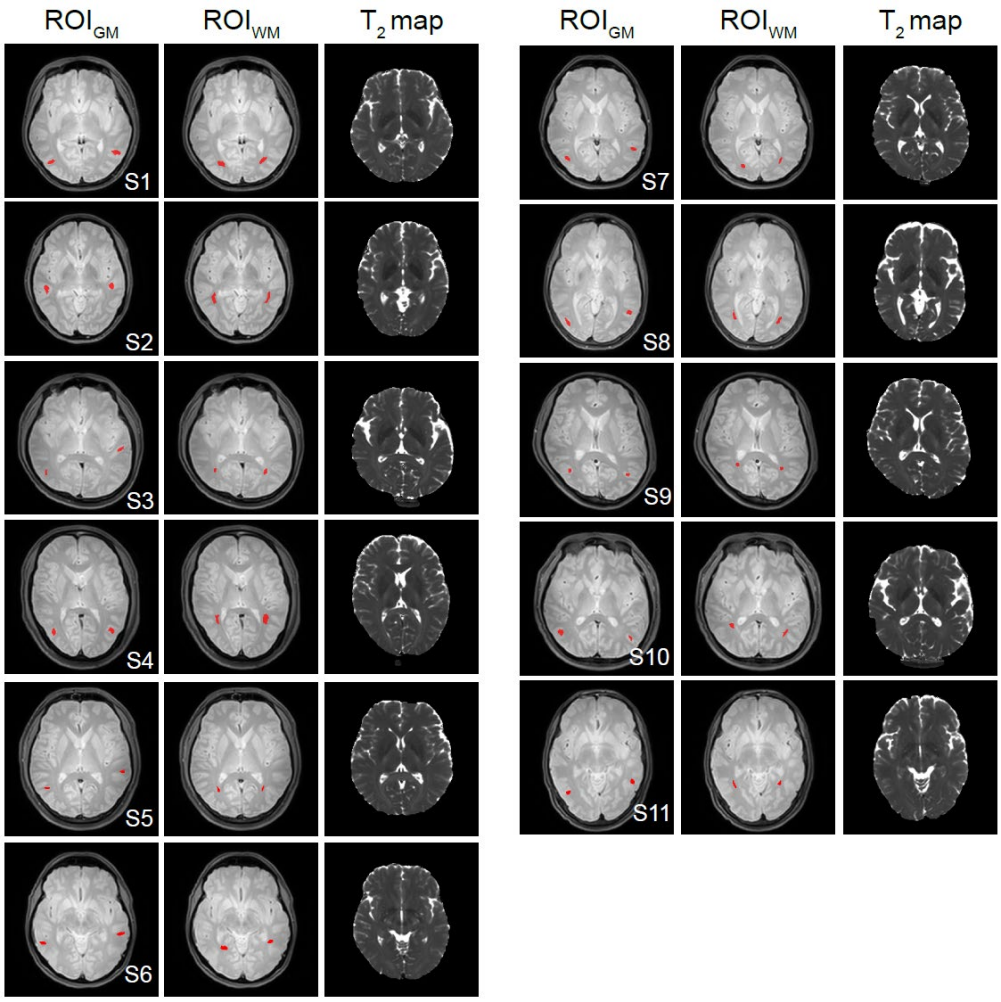
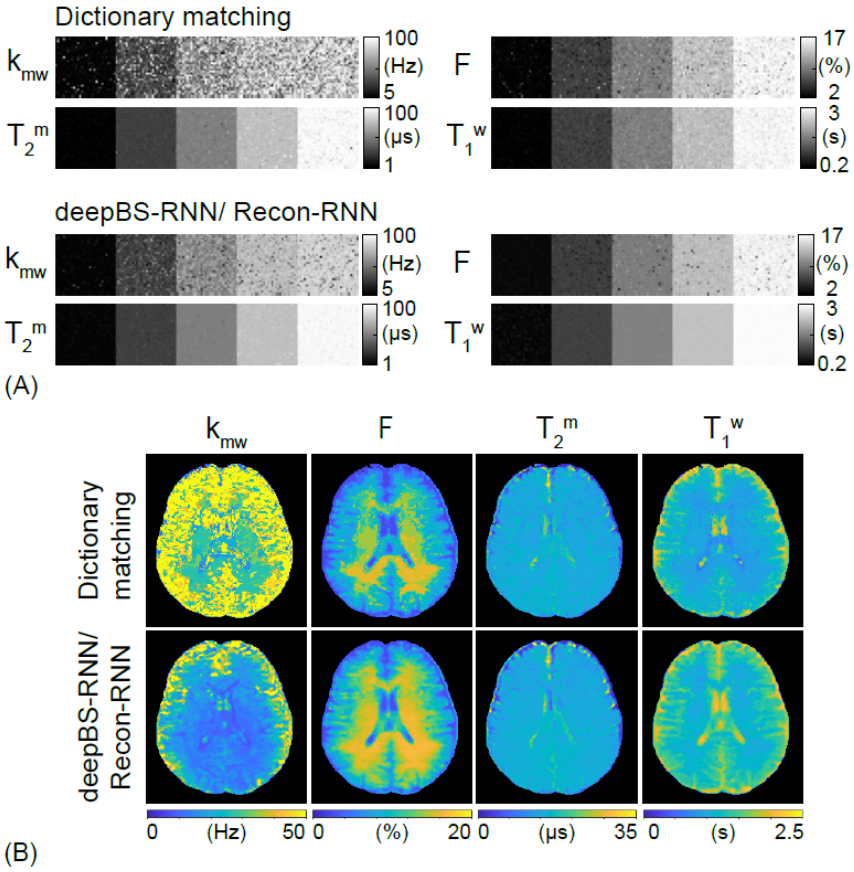
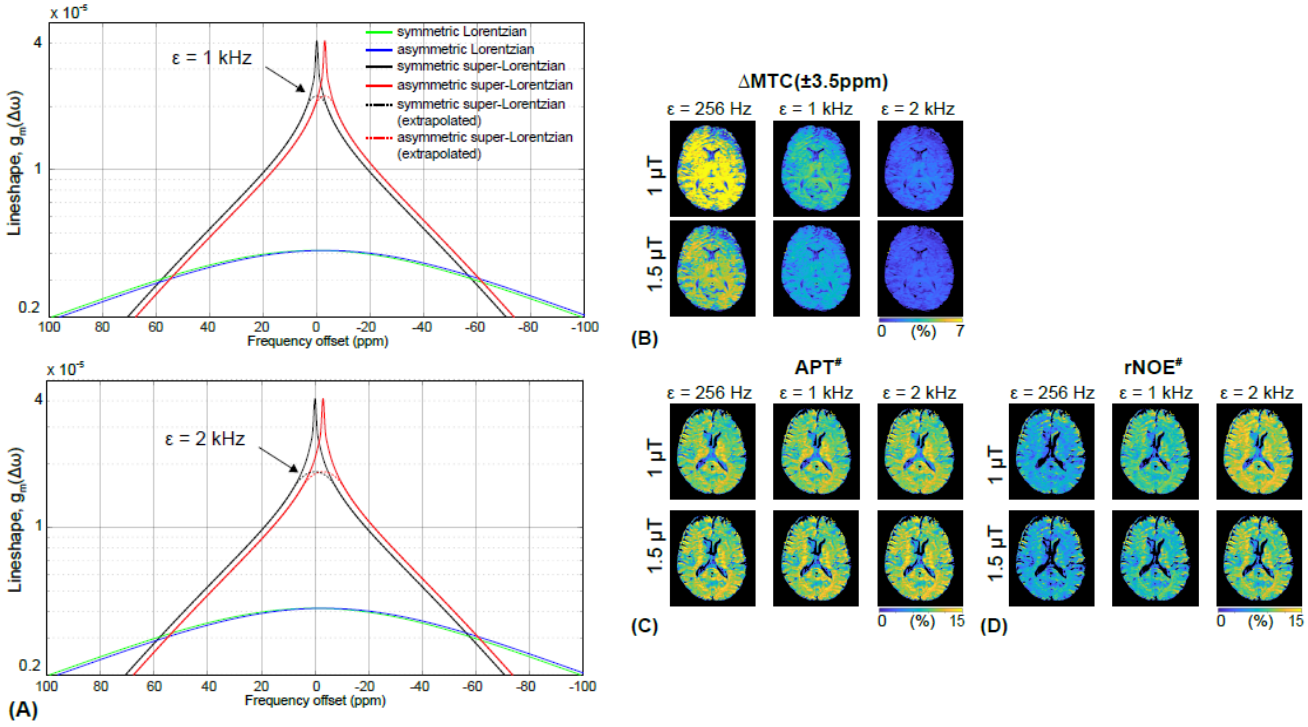
Supporting Information Figure S1. ROIs were drawn on T_2w images. Eight subjects (S1 - S8) were participated in the test-retest study, while three subjects (S9 - S11) were participated in the MTC asymmetry study. T_2 maps are shown as the reference.



Supporting Information Figure S2. Comparison of the deepBS-RNN/Recon-RNN with the dictionary matching method**.** (A) Bloch equation-based numerical phantom results. The nRMSE values of the dictionary matching method were 18.9% for k_{mw}, 6.2% for F, 2.5% for T_2^m, and 3.4% for T_1^w, while nRMSE values of the Recon-RNN were 9.1% for k_{mw}, 2.4% for F, 1.1% for T_2^m, and 0.8% for T_1^w. (B) *In vivo* tissue parameter maps of a healthy volunteer brain. Note that the reconstruction accuracy of the dictionary matching approach depends directly on the resolution of the dictionary (size = 200k).



Supporting Information Figure S3. Semisolid absorption lineshapes (Lorentzian and Super-Lorentzian) with/without MTC asymmetry (e.g., Δ_{mw} = 3 ppm). The super-Lorentzian functions are extrapolated from two cut-off frequencies (ε = 1 kHz vs. 2 kHz) to model the semisolid macromolecular pool lineshape during on-resonance RF irradiation because the super-Lorentzian absorption lineshape has an on-resonance singularity (at Δw =0). With an asymmetric MTC, the lineshape center is shifted upfield from the water resonance, thus, the amplitude of the lineshape is increased at upfield frequency offsets but decreased at downfield frequency offsets (black vs. red for super-Lorentzian or green vs. blue for Lorentzian). Consequently, the shift can affect the overall MTC (or Z_{ref}), APT#, and NOE# signal intensities.



Supporting Information Table S1. Estimated semisolid MTC parameters, free bulk water T_1 relaxation time, MTC at ±3.5 ppm, APT#, and rNOE# signal intensities (mean ± standard deviation and 95% confidence interval) obtained from Bloch fitting, BS/Recon-FCNN, BS/Recon-RNN and deepBS-RNN/Recon-RNN methods for white matter and gray matter of the human brain. Mean and standard deviations values were obtained across eight subjects participated in test-retest study.

Methods	ROIs	k_{mw} (Hz)	F (%)	T_2^m (μs)	T_1^w (s)	B_1 = 1 μT			B_1 = 1.5 μT		
						MTC (%)	APT# (%)	rNOE# (%)	MTC (%)	APT# (%)	rNOE# (%)
Bloch Fitting	WM	23.9 ± 1.5	16.0 ± 1.6	15.6 ± 0.2	1.2 ± 0.1	37.9 ± 0.6	7.3 ± 0.6	9.4 ± 0.5	55.3 ± 0.6	5.4 ± 0.7	5.7 ± 0.6
	GM	38.3 ± 4.6	9.5 ± 1.0	15.2 ± 0.3	1.5 ± 0.1	33.1 ± 1.0	7.2 ± 0.7	9.1 ± 0.6	51.1 ± 1.1	4.6 ± 0.7	4.8 ± 0.7
BS/Recon-FCNN	WM	11.4 ± 0.5	18.0 ± 0.5	14.1 ± 0.4	1.1 ± 0.1	34.4 ± 0.6	10.8 ± 0.8	12.9 ± 0.9	49.7 ± 0.8	11.1 ± 1.0	11.3 ± 1.0
	GM	15.7 ± 2.0	12.1 ± 1.6	14.3 ± 0.4	1.3 ± 0.1	32.0 ± 0.9	8.3 ± 0.7	10.2 ± 0.7	48.1 ± 1.1	7.6 ± 0.8	7.8 ± 0.8
BS/Recon-RNN	WM	14.2 ± 0.6	17.6 ± 0.6	15.8 ± 0.4	1.1 ± 0.1	36.7 ± 0.5	8.5 ± 0.6	10.6 ± 0.6	52.6 ± 0.6	8.1 ± 0.8	8.4 ± 0.7
	GM	19.0 ± 1.9	11.8 ± 1.5	15.2 ± 0.4	1.3 ± 0.1	33.1 ± 1.0	7.2 ± 0.8	9.1 ± 0.8	49.7 ± 1.1	6.0 ± 0.9	6.2 ± 1.0
deepBS-RNN/Recon-RNN	WM	14.6 ± 0.9	17.0 ± 0.6	14.6 ± 0.5	1.1 ± 0.1	35.6 ± 0.4	9.5 ± 0.5	11.7 ± 0.5	51.8 ± 0.5	9.0 ± 0.7	9.3 ± 0.6
	GM	19.8 ± 3.0	12.1 ± 1.3	14.9 ± 0.6	1.3 ± 0.1	32.9 ± 0.9	7.3 ± 0.9	9.3 ± 0.9	49.5 ± 1.2	6.1 ± 1.0	6.3 ± 1.1

Supporting Information Table S2. Effect of symmetric vs. asymmetric MTC on semisolid MTC parameters, free bulk water T_1 relaxation time, MTC at +3.5 ppm, APT#, and rNOE# signal intensities (mean ± standard deviation) in white matter and gray matter of the healthy volunteer human brain. All quantitative tissue parameters and signal intensities were estimated using Recon-RNN. Mean and standard deviations values were obtained across additional three subjects participated in the study of asymmetric analysis.

ROIs	MTC model	k_{mw} (Hz)	F (%)	T_2^m (μs)	T_1^w (s)	B_1 = 1 μT			B_1 = 1.5 μT		
						MTC (%)	APT# (%)	rNOE# (%)	MTC (%)	APT# (%)	rNOE# (%)
WM	Symmetric	13.4 ± 0.0	17.5 ± 0.3	15.7 ± 0.3	1.1 ± 0.0	36.4 ± 0.5	7.7 ± 0.5	10.3 ± 0.2	52.2 ± 0.6	7.5 ± 0.5	8.1 ± 0.2
	Asymmetric	8.2 ± 0.2	20.4 ± 0.4	16.2 ± 0.3	1.1 ± 0.0	33.3 ± 0.4	10.7 ± 0.5	9.7 ± 0.2	48.2 ± 0.4	11.5 ± 0.5	9.4 ± 0.3
GM	Symmetric	19.2 ± 0.9	11.4 ± 0.3	14.2 ± 0.4	1.3 ± 0.0	32.4 ± 0.7	7.0 ± 0.5	9.6 ± 0.4	49.2 ± 0.8	5.9 ± 0.4	6.8 ± 0.2
	Asymmetric	10.5 ± 0.5	14.2 ± 1.0	15.5 ± 0.5	1.2 ± 0.1	30.5 ± 0.7	8.9 ± 0.5	8.2 ± 0.3	45.9 ± 0.6	9.2 ± 0.4	7.5 ± 0.3

Supporting Information Table S3. Between-subject coefficient of variance (CoV) of the estimated semisolid MTC parameters, free bulk water T_1 relaxation time, MTC at ±3.5 ppm, APT#, and rNOE# signal intensities obtained from deepBS-RNN/Recon-RNN.

ROIs	Coefficient of variance									
	k_{mw}	F	T_2^m	T_1^w	B_1 = 1 μT			B_1 = 1.5 μT		
					MTC	APT#	rNOE#	MTC	APT#	rNOE#
WM	4.1 %	3.1 %	3.2 %	4.8 %	1.1 %	5.8 %	4.7 %	0.9 %	7.7 %	6.6 %
GM	14.6 %	11.0 %	3.6 %	6.8 %	2.6 %	11.5 %	8.6 %	2.3 %	15.2 %	16.3 %

Supporting Information Table S4. Intraclass correlation coefficient (ICC) of the estimated semisolid MTC parameters, free bulk water T_1 relaxation time, MTC at ±3.5 ppm, APT#, and rNOE# signal intensities obtained from Bloch fitting, BS/Recon-FCNN, BS/Recon-RNN and deepBS-RNN/Recon-RNN methods. Mean ICC values were 0.6 ± 0.3, 0.6 ± 0.3, 0.6 ± 0.2 and 0.6 ± 0.1 corresponding to Bloch Fitting, BS/Recon-FCNN, BS/Recon-RNN and deepBS-RNN/Recon-RNN respectively.

		Intraclass correlation coefficient									
						B_1 = 1 μT			B_1 = 1.5 μT		
		k_{mw}	F	T_2^m	T_1^w	MTC	APT#	rNOE#	MTC	APT#	rNOE#
Bloch Fitting	WM	0.07	0.60	0.61	0.65	0.92	0.71	0.65	0.94	0.76	0.73
	GM	0.13	0.83	-0.12	0.70	0.78	0.71	0.79	0.71	0.80	0.75
BS/Recon-FCNN	WM	0.15	0.64	0.64	0.82	0.20	0.91	0.85	-0.13	0.83	0.80
	GM	0.48	0.90	0.53	0.62	0.68	0.85	0.80	0.42	0.87	0.80
BS/Recon-RNN	WM	0.31	0.55	0.81	0.76	0.77	0.62	0.48	0.73	0.52	0.30
	GM	0.22	0.86	0.23	0.63	0.79	0.79	0.79	0.70	0.84	0.79
DeepBS-RNN/Recon-RNN	WM	0.29	0.61	0.60	0.83	0.76	0.68	0.73	0.62	0.67	0.78
	GM	0.70	0.89	0.55	0.52	0.58	0.74	0.62	0.49	0.63	0.55