LC-MS traces of ligands, complexes	.S2
Titration of MnCl <sub>2</sub> to isolated ligand	.S4
Gd(III)-based contrast discussed in main text	.S4
MRI SNR vs time curves for various organs	.S5



**Figure S1**. LC trace of monomer ligand at 280 nm detection (black) and MS chromatogram (blue) of extracted  $m/z^+$  = 399.2.



**Figure S2**. LC trace of hexamer ligand at 280 nm detection (black) and MS chromatogram (blue) of extracted  $m/3z^+$  = 840.8.



**Figure S3**. LC trace of monomeric complex at 280 nm detection (black) and MS chromatogram (blue) of extracted  $m/z^+$  = 452.0.



**Figure S4**. LC trace of hexameric complex at 280 nm detection (black) and MS chromatogram (blue) of extracted  $m/3z^+$  = 946.2.



**Figure S5**. MnCl<sub>2</sub> titrated to a solution of the monomeric (left) and dendrimeric (right) chelates. The sharp increase in  $1/T_2$  occurs when [Mn] > [L]. The molecular weight of the isolated ligand was determined by dividing mol Mn present at the point of inflection by weight of dissolved ligand material.



[Gd(DTPA-BMEA)(H<sub>2</sub>O)]

[Gd(BOPTA)(H<sub>2</sub>O)]<sup>2-</sup>

[Gd(EOB-DTPA)(H<sub>2</sub>O)]<sup>2-</sup>

Figure S6. Clinically utilized Gd(III) complexes discussed for comparison in this study.



**Figure S7**. Signal-to-noise ratio (SNR) vs time curves for blood, kidney, liver, and gallbladder. After injection of both Mn(II) dendrimer and  $[Gd(DTPA)(H_2O)]^{2-}$  the blood pool signal exhibited an immediate increase, followed by rapid decrease to baseline. The increase in vascular SNR one minute post injection compared to baseline resulted in a 255±79% SNR increase for  $[Gd(DTPA)(H_2O)]^{2-}$  compared with a 507±73% SNR increase for the Mn dendrimer, a 2-fold improvement. The time course of signal

enhancement in the kidneys, liver, and gallbladder demonstrated both renal and hepatobiliary clearance for Mn(II) Hexamer, while  $[Gd(DTPA)(H_2O)]^{2-}$  is exclusively cleared via the kidneys.