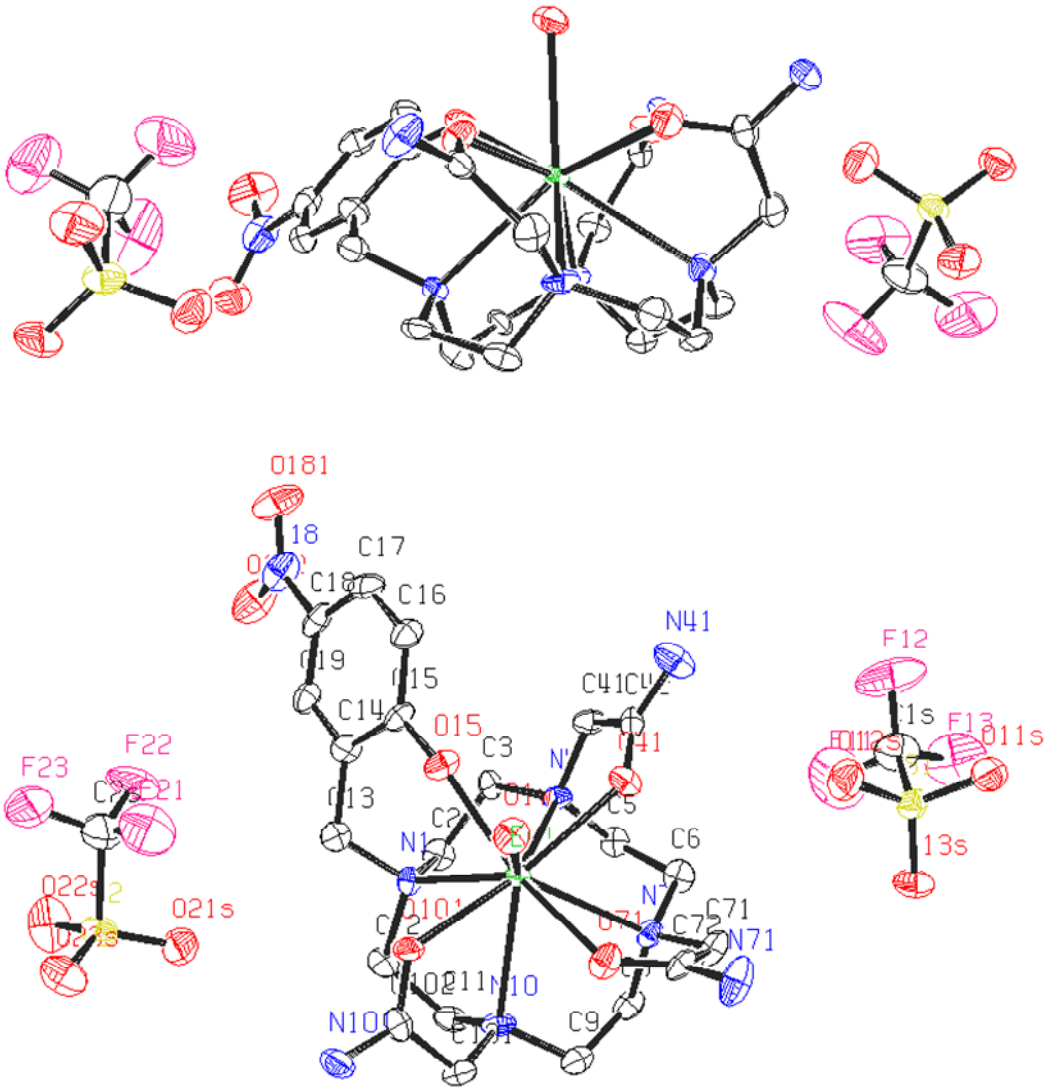
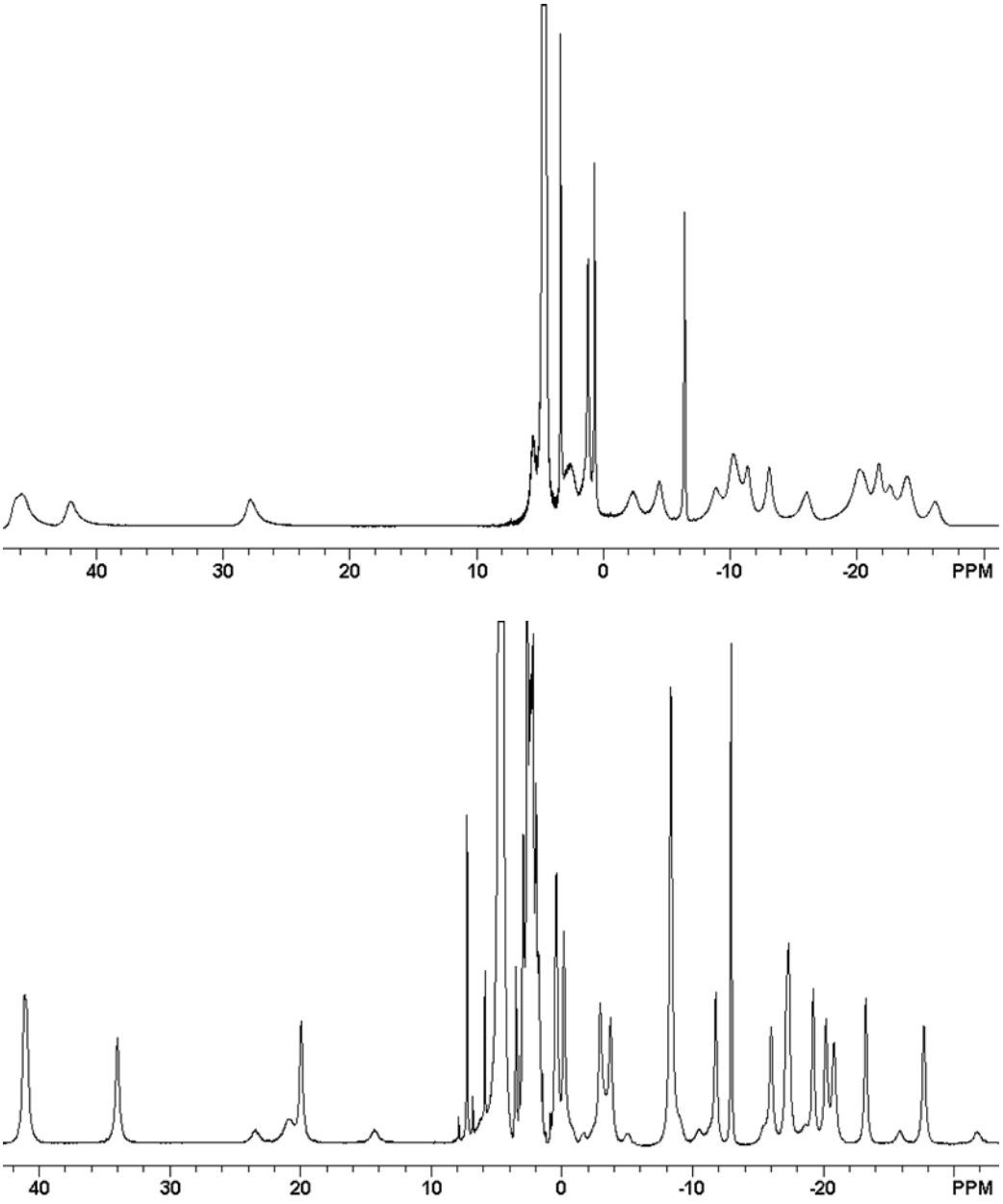
Supplementary Information - S 1.



ORTEP renderings of the crystal structure of Eu(NP-DO3AM).H_2O.$(CF_3SO_3^-)_2$ showing 50% ellipsoids.

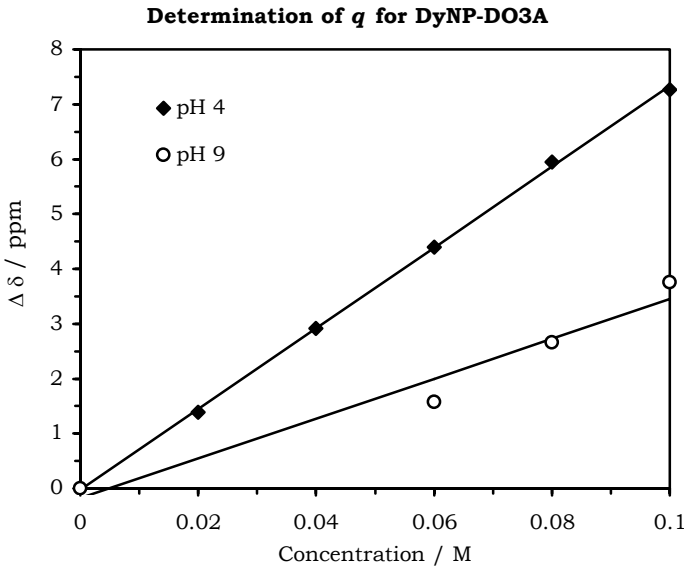
Supplementary Information - S 2.

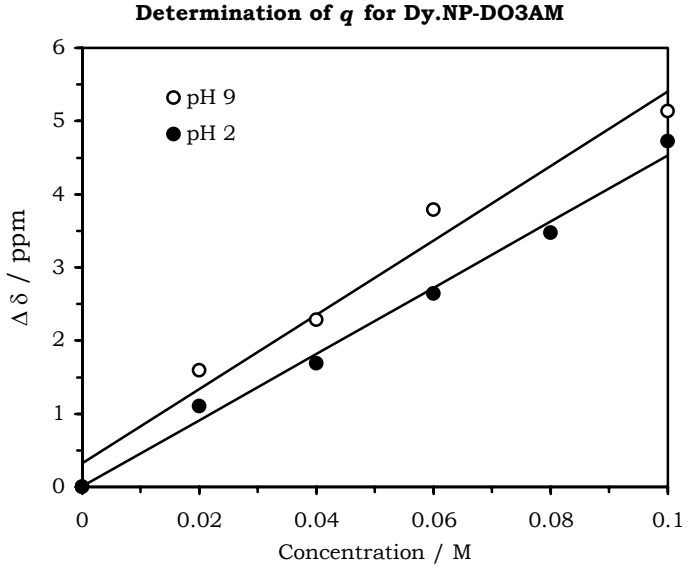


^{1}H NMR spectra of Eu(NP-DO3A) (bottom) and Eu(NP-DO3AM) (top) recorded on at 0°C and 500 MHz in D_2O at pD = 8.4 and pD = 6.5, respectively. ^{1}H NMR spectra of the corresponding Yb complexes recorded over the pH range 4 – 9 exhibited similar spectra but did not change in appearance as a function of pH.

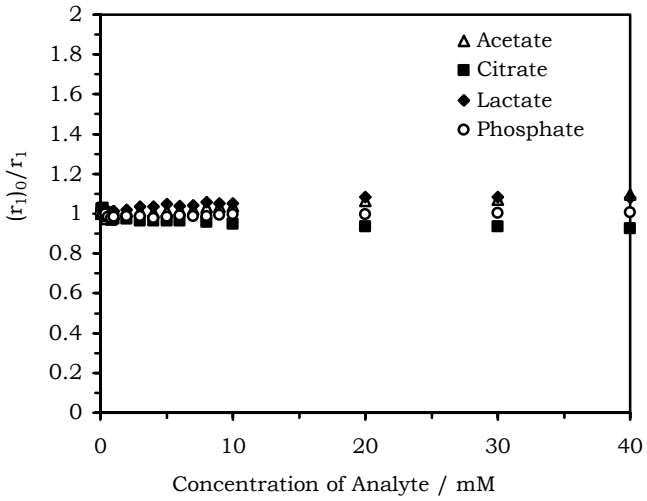
Supplementary Information – S 3

Determination of the hydration states of the Dy complexes by ^{17}O NMR

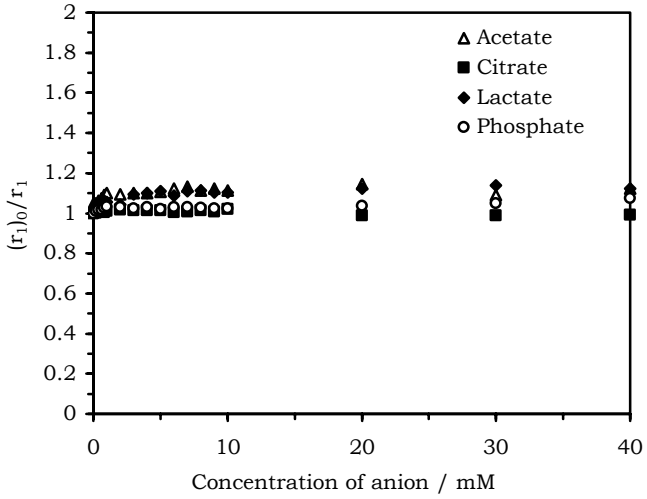




Supplementary Information - S 4.



The effect of anion concentration on relaxivity at pH 4. For a 1 mM solution of GdNP-DO3A at pH 4 the value of the initial relaxivity $(r_1)_0$ over the relaxivity (r_1) is shown as a function of the concentration of anion.



The effect of anion concentration on relaxivity at pH 9. For a 1 mM solution of GdNP-DO3A at pH 9 the value of the initial relaxivity $(r_1)_0$ over the relaxivity (r_1) is shown as a function of the concentration of anion.