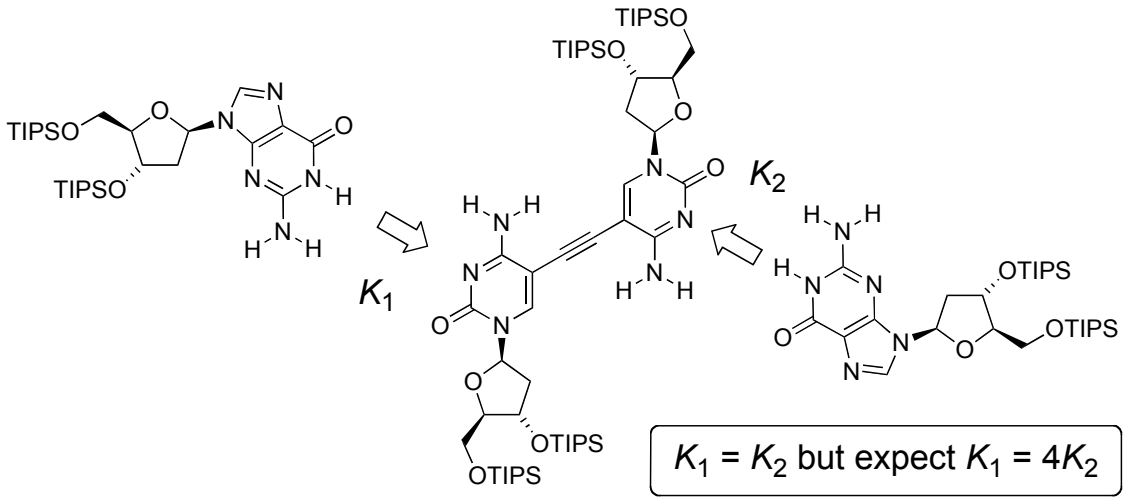
Graphical Abstract



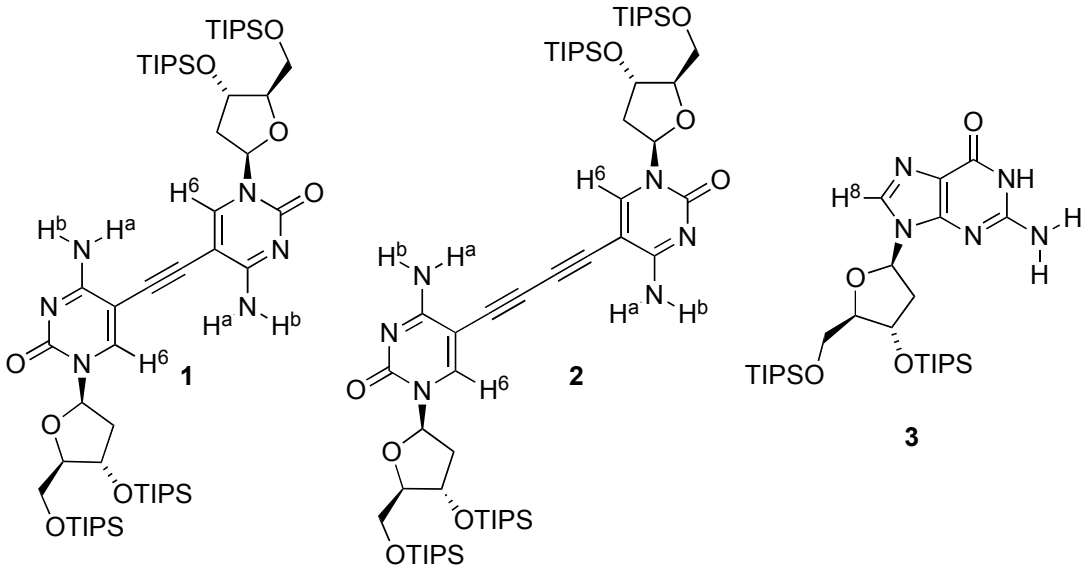


Figure 1. Receptors **1**, **2** and guanosine derivative, **3** used for binding studies; TIPS = triisopropylsilyl.

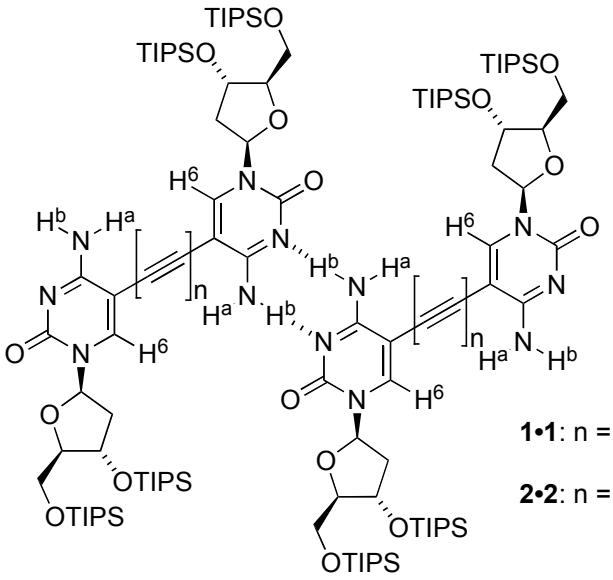


Figure 2. Dimers of **1** and **2, 1•1** and **2•2**; respectively.

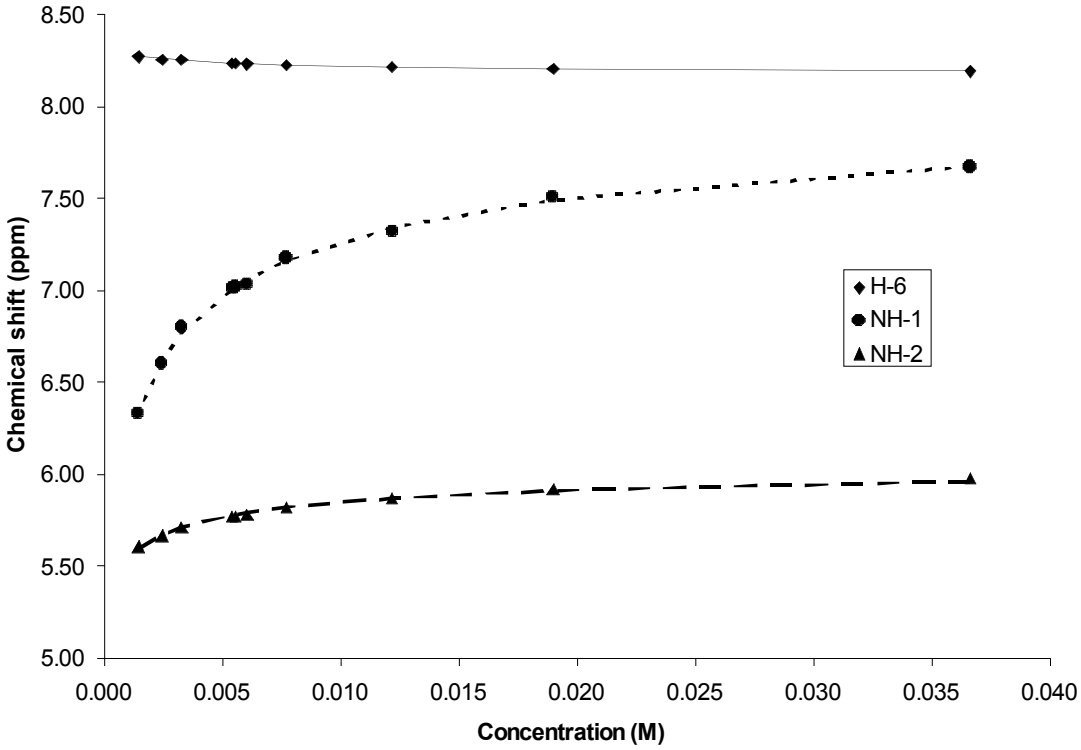


Figure 3. Curve fitting from NMR dilution of **2**; $K_{\mathbf{2}\cdot\mathbf{2}} = 340\pm7\ M^{-1}$.

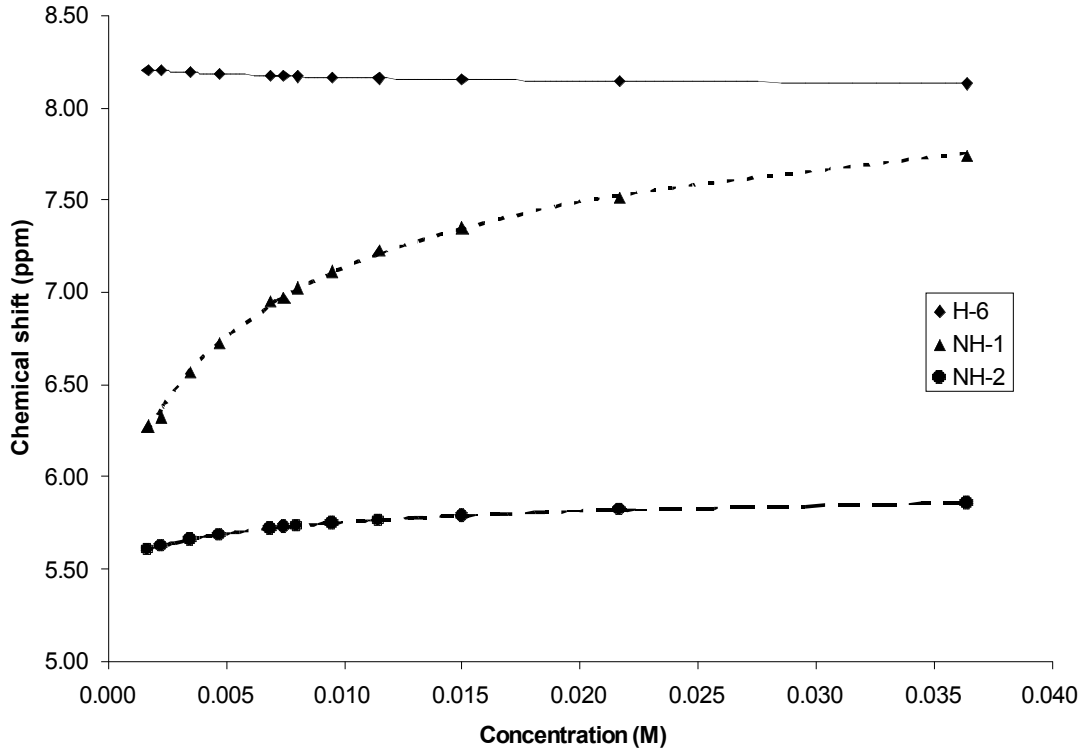


Figure 4. Curve fitting from NMR dilution of **1** in $CDCl_3$; $K_{\mathbf{1}\cdot\mathbf{1}} = 83\pm3\ M^{-1}$.

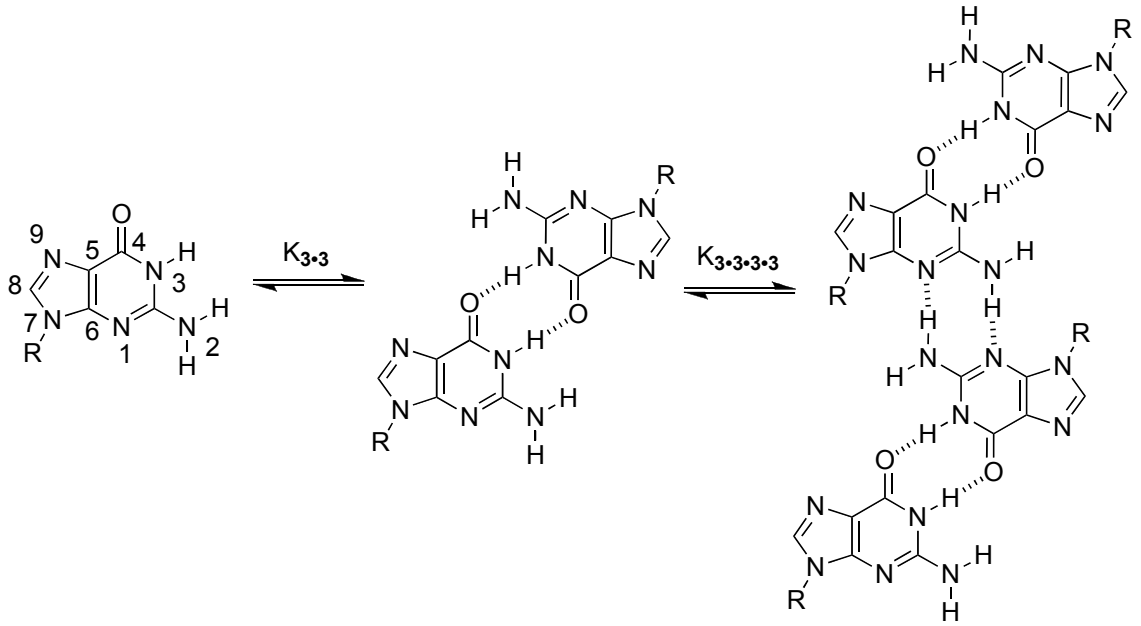
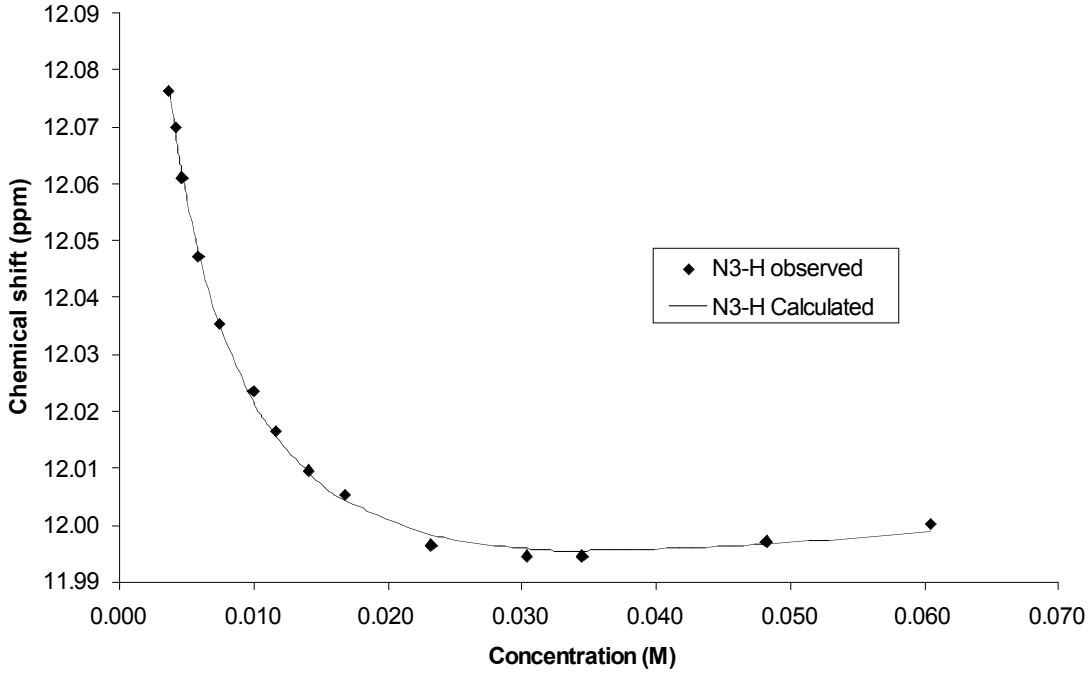
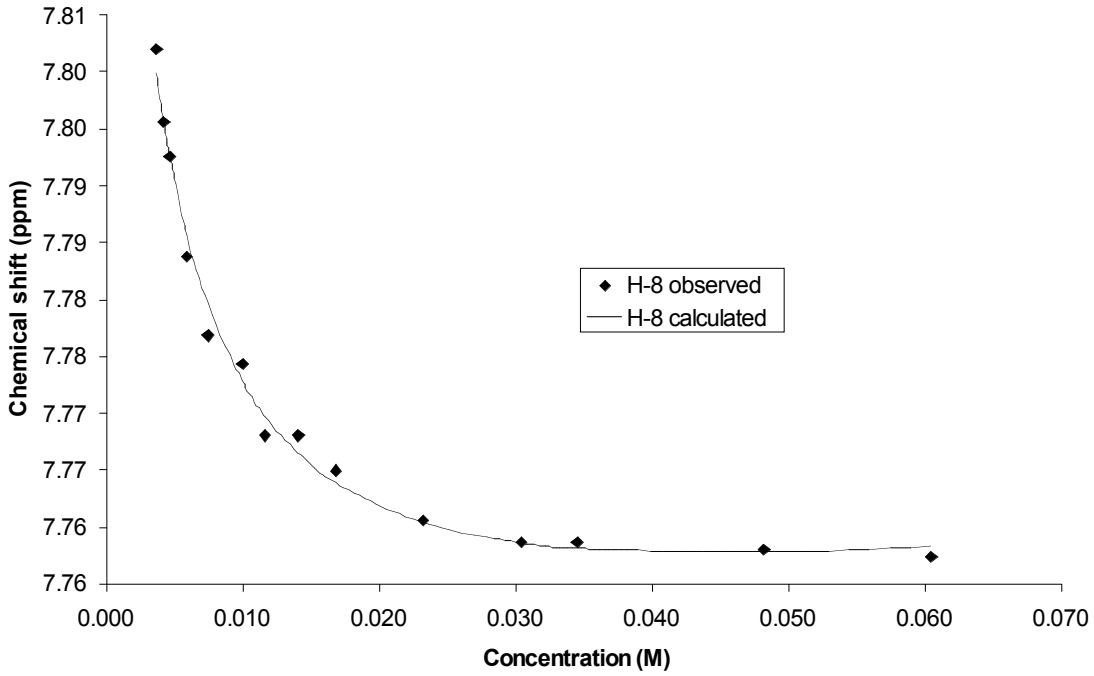


Figure 5. Self-association model of lipophilic guanosine **3** in chloroform.


12.09
12.08
12.07
12.06
12.05
12.04
12.03
12.02
12.01
12.00
11.99
Chemical shift (ppm)
0.000
0.010
0.020
0.030
0.040
0.050
0.060
0.070
Concentration (M)
N3-H observed
N3-H Calculated


7.81
7.80
7.80
7.79
7.79
7.78
7.78
7.77
7.77
7.76
7.76
Chemical shift (ppm)
0.000
0.010
0.020
0.030
0.040
0.050
0.060
0.070
Concentration (M)
H-8 observed
H-8 calculated

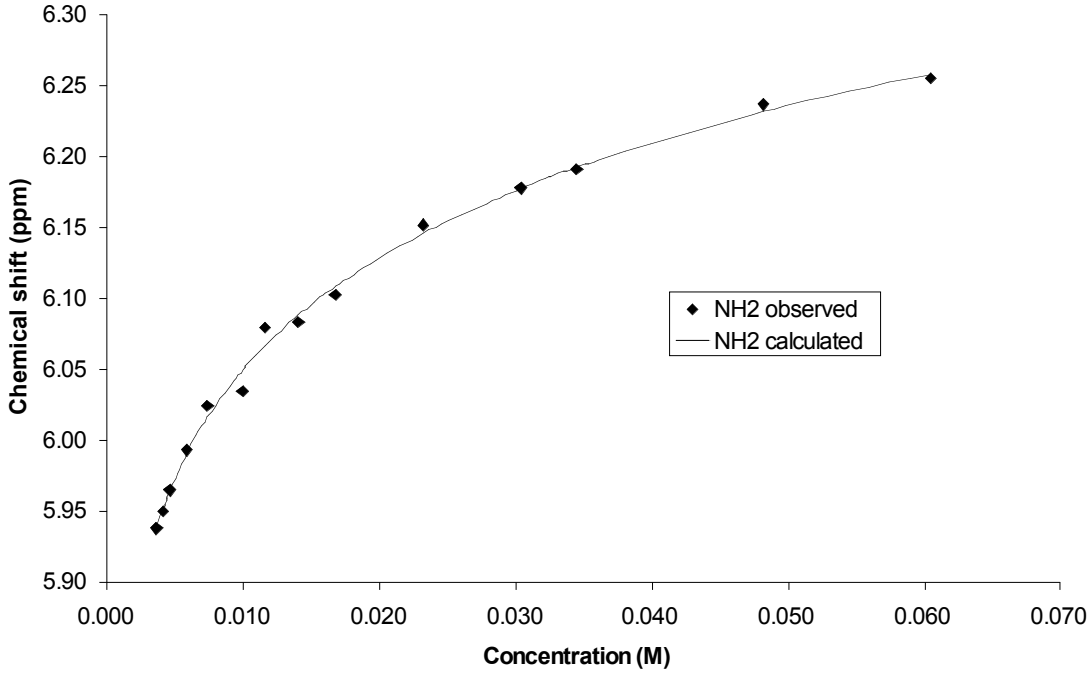


Figure 6. Curve fitting for NMR dilution of **3** in $CDCl_3$. The three titration curves (**A**) N3-H (**B**) H-8 and (**C**) NH_2 are fitted simultaneously to give $K_{\mathbf{3}\cdot\mathbf{3}}$ of 370±72 and $K_{\mathbf{3}\cdot\mathbf{3}\cdot\mathbf{3}\cdot\mathbf{3}}$ of 15±1 M^{-1}.

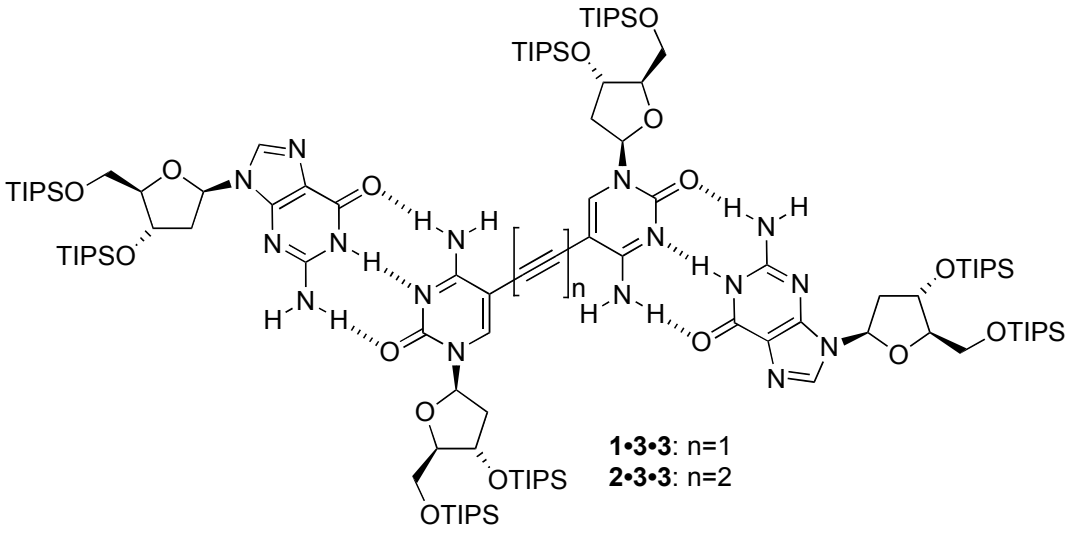
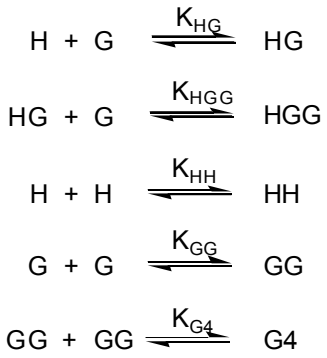


Figure 7. Complexation of receptors **1** or **2** with **3**; TIPS = triisopropylsilyl.



Scheme 1. Equilibria in the binding studies between **2** and **3**.

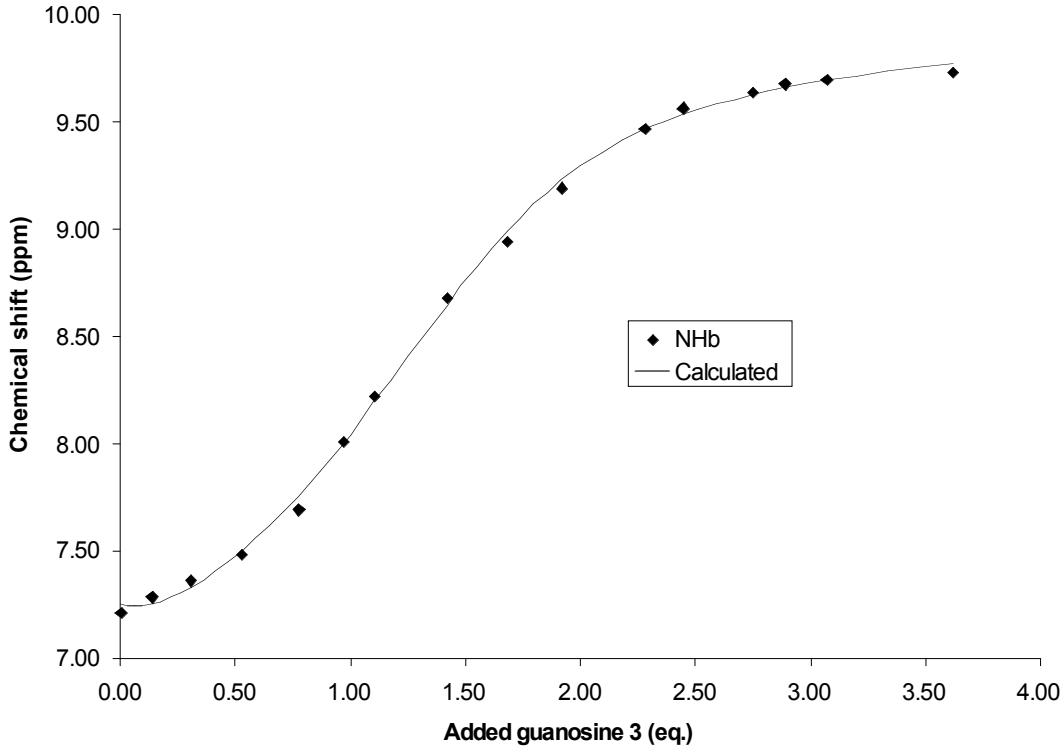


Figure 8. Curve fitting of NMR titration in $CDCl_3$ between **2** and **3**. $K_{\mathbf{2}\bullet\mathbf{3}}$ 8100 ± 380; $K_{\mathbf{2}\bullet\mathbf{3}\bullet\mathbf{3}}$ 1170 ± 80 M^{-1}.

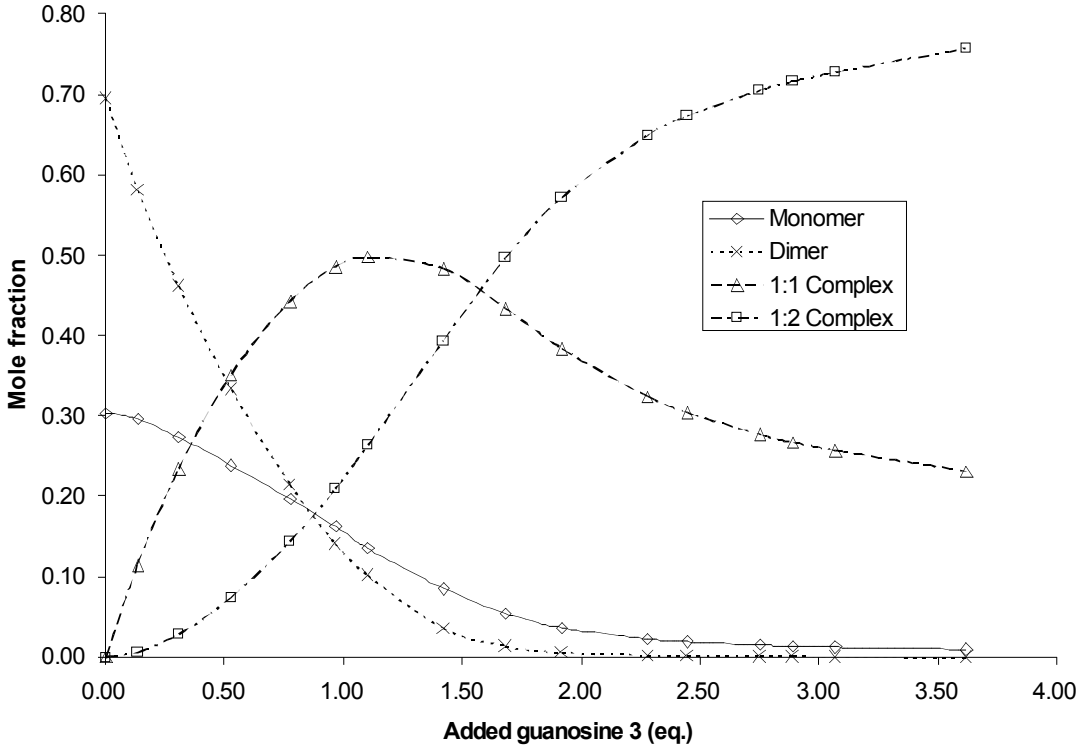


Figure 9. Speciation curve for receptor **2** upon addition of **3** in $CDCl_3$, showing monomeric **2**, dimeric **2,** 1:1 complex (**2•3**) and 1:2 complex (**2•3•3**).

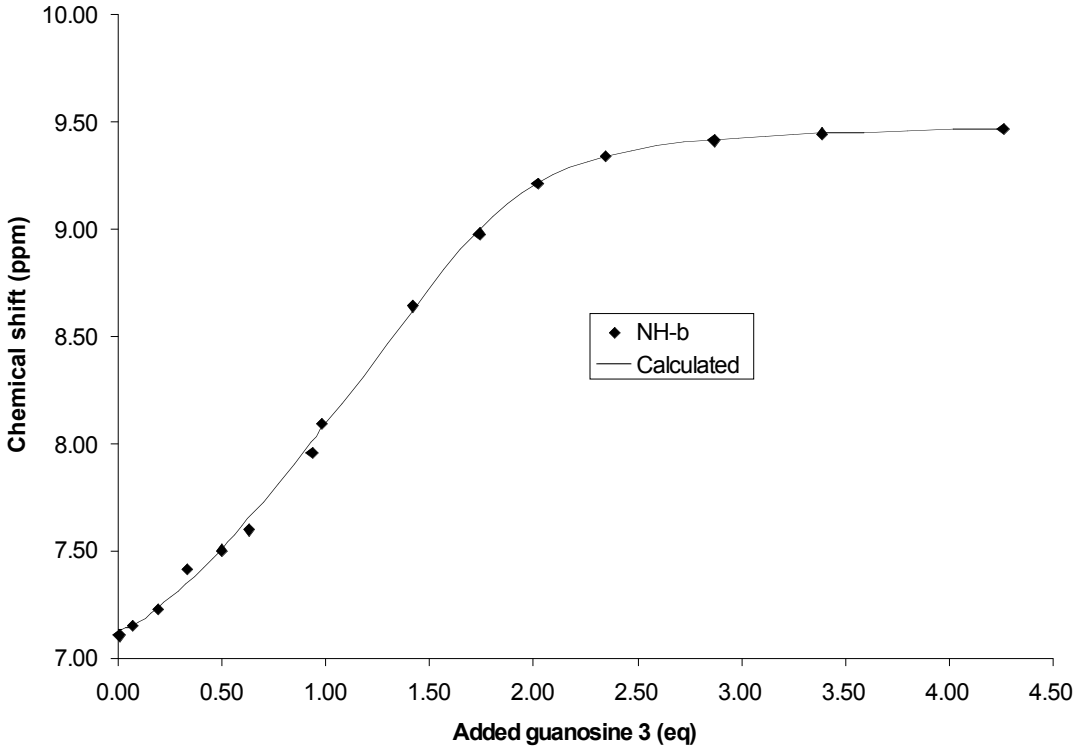


Figure 10. Curve fitting of NMR titration in $CDCl_3$ between **1** and **3**. $K_{\mathbf{1}\cdot\mathbf{3}} = 5180 \pm 210$, $K_{\mathbf{1}\cdot\mathbf{3}\cdot\mathbf{3}} = 4800 \pm 170\ M^{-1}$.

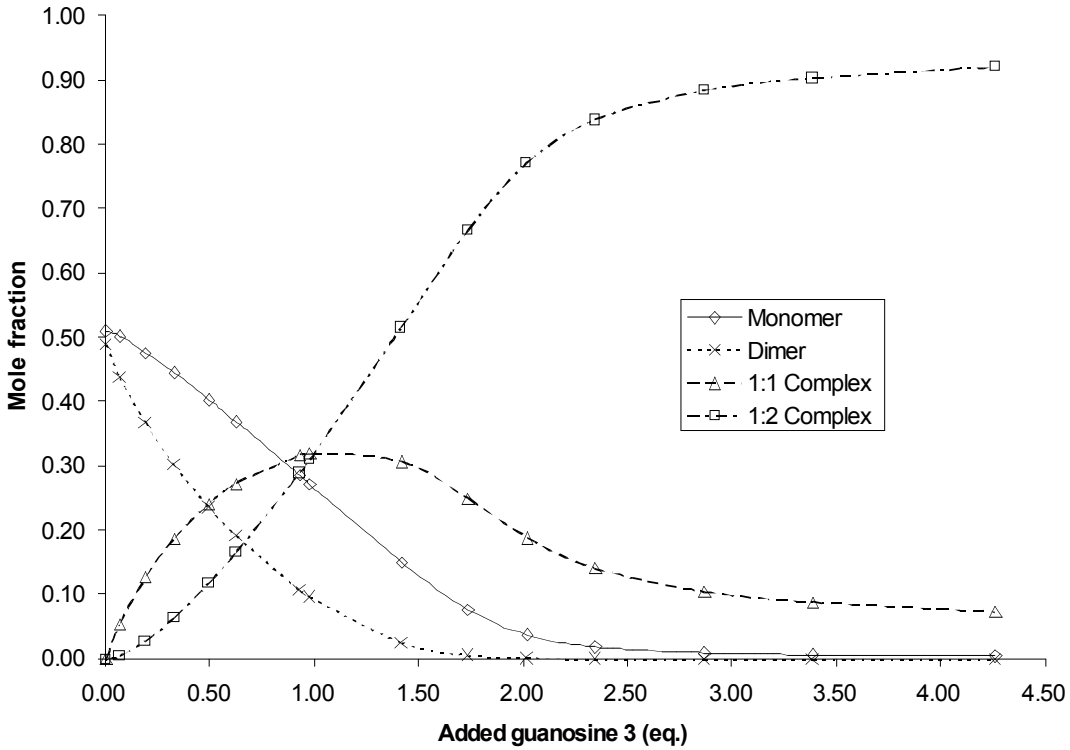


Figure 11. Speciation curve for **1** upon addition of **3** in $CDCl_3$.

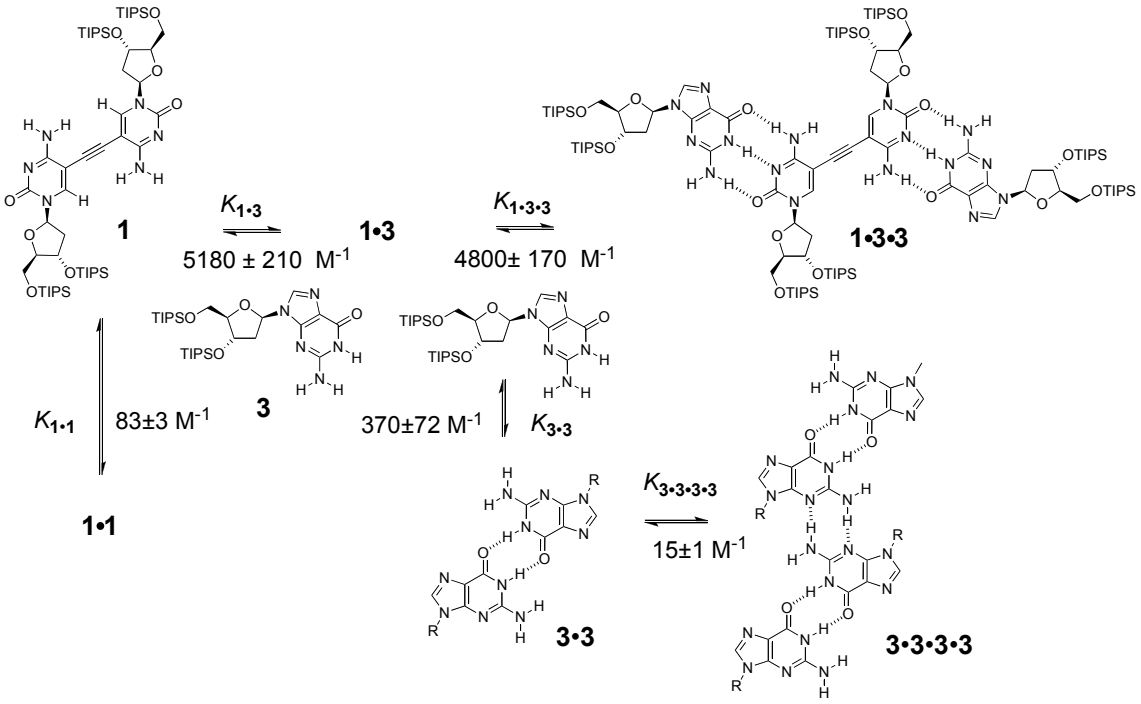


Figure 12. Summary of equilibria in deuterochloroform for monoalkyne receptor **1.**

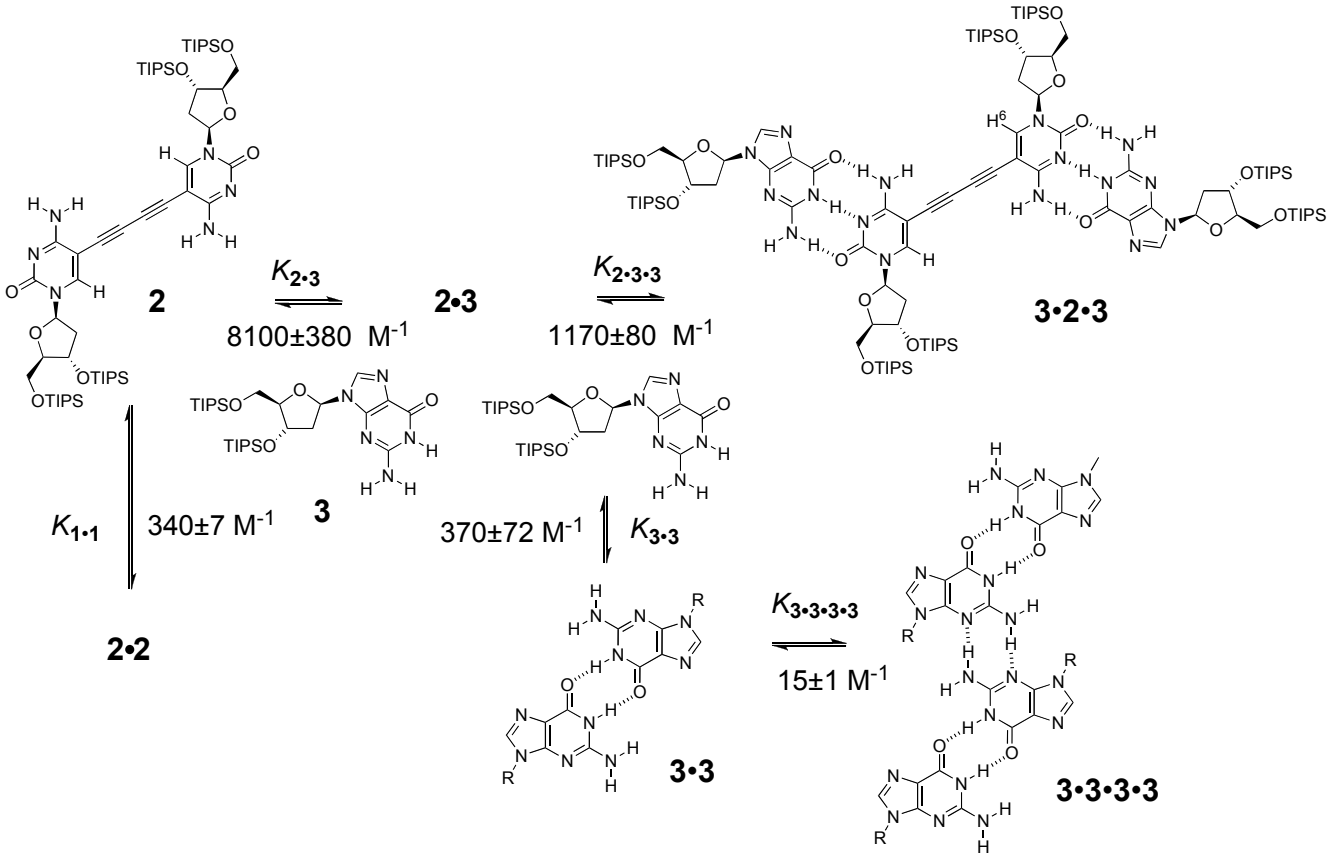


Figure 13. Summary of equilibria in deuterochloroform for dialkyne receptor **2**.

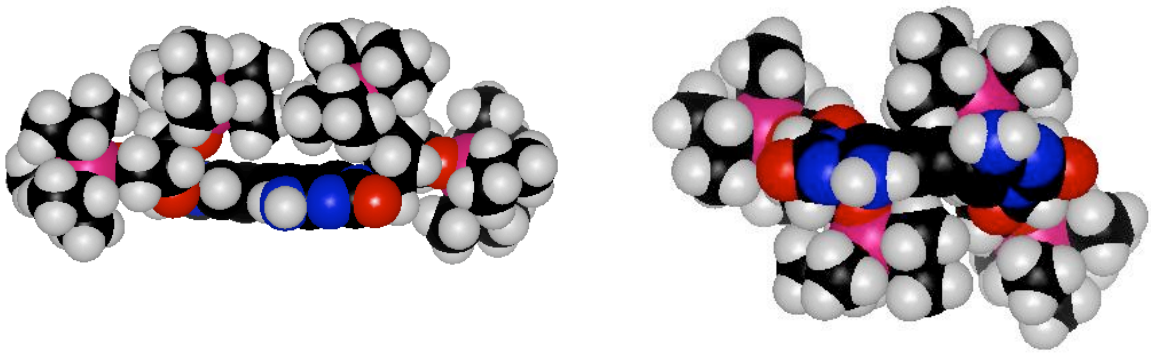


Figure 14. C-P-K models of **1** at E_{min}(left) dihedral ≈ 30° showing interactions between TIPS groups and E_{max} (right) dihedral ≈ 165°.

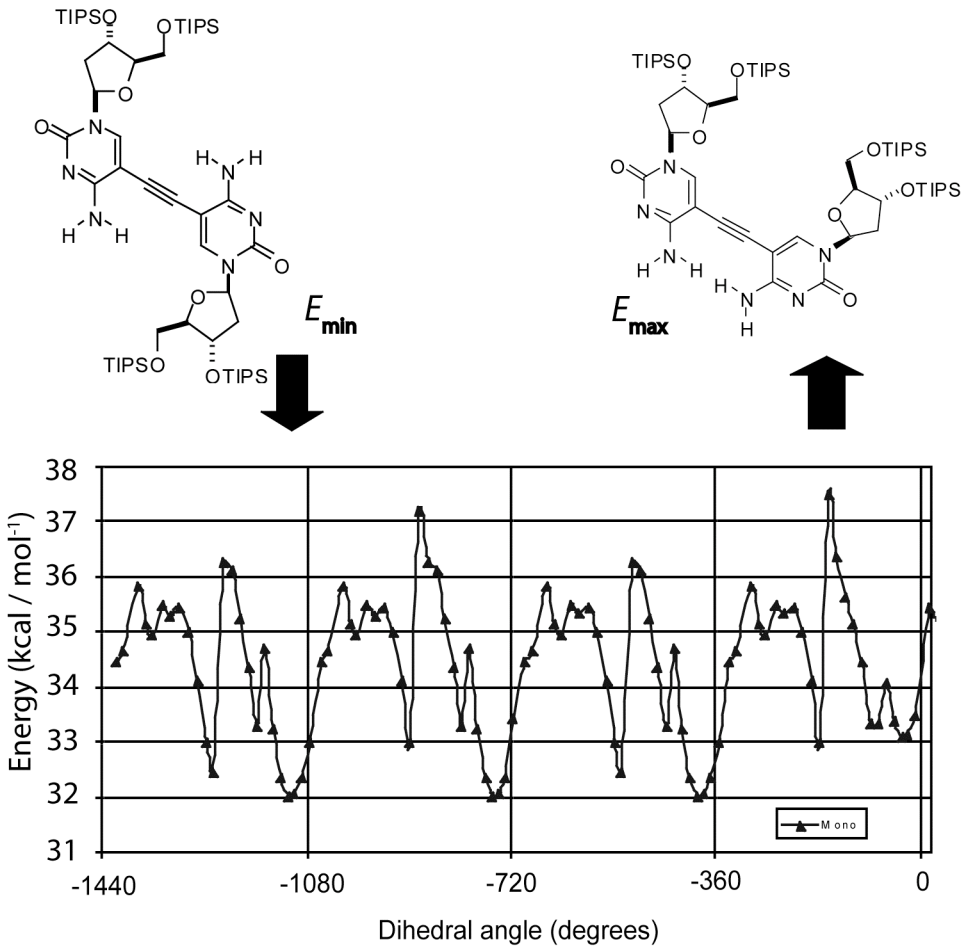


Figure 15. Torsional energy *vs*. dihedral angle for monoalkyne **1**

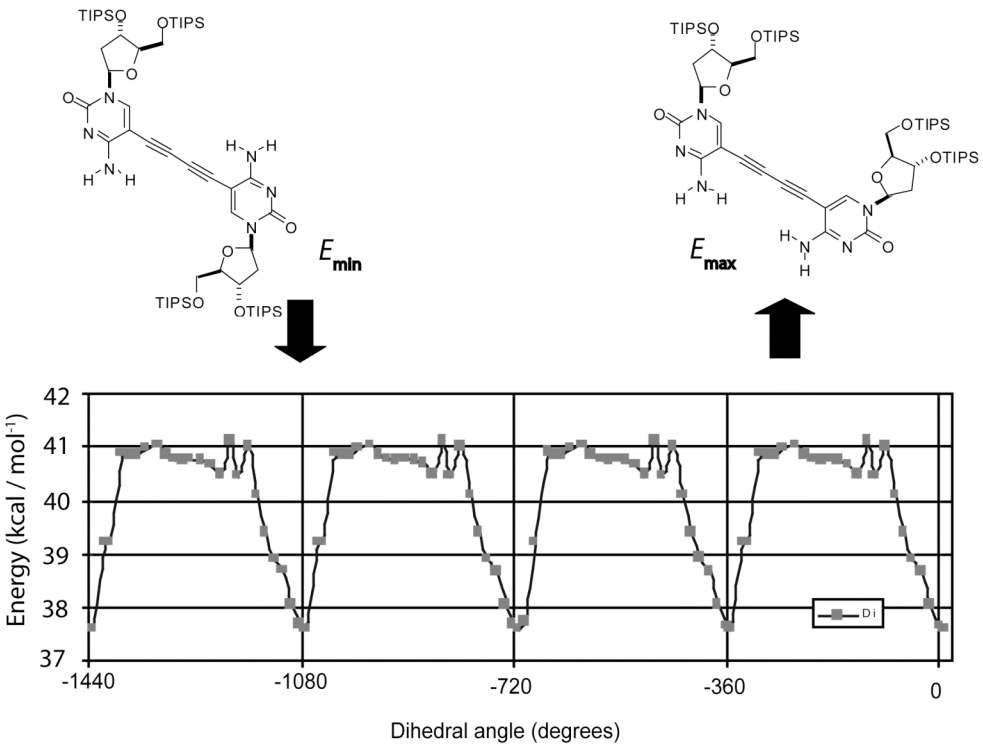


Figure 16. Torsional energy *vs*. dihedral angle for dialkyne **2**