**Table S3.** $^{1}$H NMR Chemical Shift Assignments for the 1:1 DNA–Drug Complex [d(GGTACC)$_2$ACRAMTU][a]

| residue | H6/H8 | H2/H5/ $CH_3$ | H1' | H2' | H2" | H3' | H4' | H5' H5" |
|---|---|---|---|---|---|---|---|---|
| G1 | 7.74 | | 5.64 | 2.38 | 2.38 | 4.76 | 4.13 | 3.65<br>3.65 |
| G2 | 7.80 | | 5.89 | 2.66 | 2.62 | 4.95 | 4.36 | 4.10<br>4.07 |
| T3 | 7.21 | 1.49 | 5.77 | 2.09 | 2.35 | 4.84 | 4.26 | n. a.[b]<br>4.14 |
| A4 | 8.22 | 7.51 | 6.16 | 2.73 | 2.81 | 4.98 | 4.41 | n. a.<br>4.19 |
| C5 | 7.37 | 5.40 | 5.97 | 2.08 | 2.40 | 4.75 | 4.27 | 4.19<br>4.17 |
| C6 | 7.62 | 5.73 | 6.19 | 2.25 | 2.56 | 4.50 | 4.11 | 4.13<br>4.03 |

| ACR | H1/H8 | H2/H7 | H3/H6 | H4/H5 | H9 | H10 | H11 | H12 |
|---|---|---|---|---|---|---|---|---|
| | 7.86<br>(-0.54)[c] | 6.61<br>(-0.94) | 7.09<br>(-0.87) | 7.00<br>(-0.75) | 4.10<br>(-0.34) | 4.03<br>(-0.35) | 3.09<br>(0.21) | 3.09<br>(0.24) |

[a] 500 MHz, $D_2O$, 303 K, pH* 7.0; shifts are relative to DSS. [b] Not assigned due to signal overlap.
[c] Numbers in parentheses are $\Delta\delta = \delta_{\text{adduct}} - \delta_{\text{free ACRAMTU}}$ (ppm). At 303 K the free duplex was partially melted, and $\Delta\delta$ values are meaningless and are not given.