Table S2. ^{13}C-NMR spectral data (δ in ppm) of the cyclic =CYY′ systems in $CDCl_3$ at room temperature

Cmpd	Y	Y′	Y	Y′	C_β [a]	C_α [a]	R
5a	CN	CO_2Me	116.30 (s)	51.57 (q, J = 147.0 Hz, OMe), 168.50 (s, C=O)	56.14	172.16	43.41 (d of t, J_t = 147.6 Hz, J_d = 5.3 Hz, NCH_2), 69.38 (t, J = 155.7 Hz, OCH_2)
5b	$CO_2CH_2F_3$	$CO_2CH_2CF_3$	59.95 (q of t, J_{C-H} = 150.0 Hz, H_{C-F} = 36.7 Hz, CH_2), 123.23 (q, J = 277.4 Hz, CF_3), C=O signal not observed		74.80	172.24	42.49 (d of t, J_t = 146.7 Hz, J_d = 5.5 Hz, NCH_2), 69.56 (t, J = 155.3 Hz, OCH_2)
5c[b]	Barbituric acid moiety		150.81 (s, C=O), 164.80 (s, 2C=O)		78.57	170.11	43.06 (t, J = 146.8 Hz, NCH_2), 70.11 (t, J = 155.9 Hz, OCH_2)
5d	Meldrum's acid moiety		26.44 (q, J = 128.6 Hz, Me), 103.35 (m, J = 4.6 Hz, C), 160.83 (s, C=O), 166.36 (s, C=O)		72.88	171.59	42.40 (t, J = 147.8 Hz, NCH_2), 70.35 (t, J = 156.3 Hz, OCH_2)
5e	CN	CN	113.54 (s)	115.70 (s)	34.79	173.96	43.42 (t, J = 147.9 Hz, NCH_2), 71.00 (t, J = 156.4 Hz, OCH_2)
5f	CO_2Et	COMe	14.34 (q, J = 126.7 Hz, Me), 59.64 (t, J = 142.5 Hz, CH_2), 167.09 (s, C=O)	30.04 (q, J = 124.9 Hz, Me), 196.28 (s, C=O)	88.94	171.49	42.50 (t, J = 146.9 Hz, NCH_2), 68.29 (t, J = 156.0 Hz, OCH_2)
5g	CONHPh	COMe	120.66 (d, J = 162.1 Hz), 123.09 (d, J = 159.4 Hz), 128.63 (d, J = 166.3 Hz), 139.02 (t, J = 9.2 Hz), 167.69 (s, C=O)	32.09 (q, J = 128.3 Hz, Me), 195.33 (q, J = 5.7 Hz, C=O)	89.27	173.03	42.43 (t, J = 146.6 Hz, NCH_2), 68.52 (t, J = 154.8 Hz, OCH_2)
5h	$SO_2C_4F_9$	COMe	110.49-118.65 (m, C_4F_9)	29.58 (q, J = 129.5 Hz, OMe), 194.78 (t, J = 6.6 Hz, C=O)	90.15	171.44	42.92 (t, J = 147.1 Hz, NCH_2), 69.62 (t, J = 155.4 Hz, OCH_2)
5i	COMe	COMe	31.50(Me), 194.05(C=O)		99.46	171.81	42.45(NCH_2), 68.09(OCH_2)
5j[b]	Dimedone moiety		28.38(q, J= 120.6Hz, Me), 30.39(s, C), 52.10(t, J= 127.2Hz, CH_2), 193.88(s, C=O)		96.45	170.01	42.63 (t, J = 147.1 Hz, NCH_2), 69.36 (t, J = 155.9 Hz, OCH_2)

Table S2 (continued)

5k	CN	$CONMe_2$	118.62 (s)	37.88 (q, J = 137.4 Hz, NMe), 168.07 (s, C=O)	55.93	173.46	43.33 (t, J = 141.9 Hz, NCH_2), 68.66 (t, J = 152.4 Hz, OCH_2)
5l	CN	CONHMe	118.83	26.45(Me), 166.74(C=O)	55.13	170.66	44.31(NCH_2), 69.07(OCH_2)
5m[b]	CN	$CONH_2$	119.02	166.37	55.08	170.30	42.80(NCH_2), 69.25(OCH_2)
5n	$CO_2CH_2F_3$	CO_2Me	59.70 (q of t, J_{C-H} = 149.9 Hz, J_{C-F} = 36.2 Hz, CH_2), 123.35 (q, J = 277.5 Hz, CF_3), 163.20 (s, C=O)	51.35 (q, J = 146.5 Hz, OMe), 162.45 (s, C=O)	75.34	171.74	42.42 (d of t, J_t = 146.7 Hz, J_d = 5.3 Hz, NCH_2), 69.25 (t, J = 155.3 Hz, OCH_2)
5o	CO_2Me	CO_2Me	51.26 (q, J = 146.8 Hz, Me)		76.28	171.31	42.38 (t, J = 147.2 Hz, NCH_2), 69.01 (t, J = 156.2 Hz, OCH_2)
6a[c]	CN	CO_2Me	119.18 (s)	51.62 (q, J = 146.9 Hz, OMe), 166.25 (s, C=O)	65.24	175.13	29.94 (t, J = 147.3 Hz, NCH_2), 52.40 (t, J = 142.6 Hz, SCH_2)
6b	$CO_2CH_2F_3$	$CO_2CH_2CF_3$	59.90 (t of 1, J_{C-H} = 151.8 Hz, J_{C-F} = 35.8 Hz, CH_2), 124.13 (q, J_{C-F} = 277.5 Hz, CF_3), 165.67 (s, C=O)		83.39	177.85	29.08 (t, J = 145.74 Hz, NCH_2), 50.61 (t, J = 148.0 Hz, SCH_2)
6c[b]	Barbituric acid moiety		150.65, 163.90 , 165.13 (s, C=O)		87.66	175.58	28.97 (t, J = 146.0 Hz, NCH_2), 49.92 (t, J = 147.2 Hz, SCH_2)
6d	Meldrum's acid moiety		24.42 (q, J = 128.6 Hz, Me), 103.72 (m, J = 4.6 Hz, C), 163.12 (s, C=O), 164.83 (s, C=O)		81.92	177.73	29.36 (t, J = 145.6 Hz, NCH_2), 49.19 (t, J = 145.1 Hz, SCH_2)
6e	CN	CN	115.59 (s)	117.46 (s)	42.37	177.34	31.82 (t, J = 148.2 Hz, NCH_2), 52.08 (t, J = 145.7 Hz, SCH_2)
6f Z **E**	CO_2Et	COMe	14.34 (Me), 60.07 (CH_2),168.18 (C=O) 14.32 (Me), 59.96 (CH_2), 169.25 (C=O)	31.27 (Me), 196.02 (C=O) 31.05 (Me), 194.13 (C=O)	98.62 ov with Z-isomer	177.57 174.50	28.80 $(NCH_2)_3$, 49.49 (SCH_2) 29.19 (NCH_2), 47.99 (SCH_2)
6g[b]	CONHPh	COMe	119.63 (d, J = 162.7 Hz), 123.39 (d, J = 152.8 Hz), 129.06 (d, J	28.58 (q, J = 127.2 Hz, Me), 190.64 (q, J = 5.7 Hz,	104.18	171.07	29.16 (t, J = 144.1 H, NCH_2), 49.81 (t, J = 145.1 Hz, SCH_2)

Table S2 (continued)

			= 160.3 Hz), 140.06 (t, J = 8.2 Hz), 167.66 (s)	C=O)			
7a	CN	CO_2Me	117.49 (s)	51.15 (q, J = 146.9 Hz, OMe), 168.29 (s, C=O)	57.94	169.75	20.41 (t, J = 130.9 Hz, CH_2), 37.52 (t, J = 144.1 Hz, NCH_2), 66.51 (t, J = 152.2 Hz, OCH_2)
7b	$CO_2CH_2F_3$	$CO_2CH_2CF_3$	59.70 (t of q, J_{C-H} = 149.9 Hz, J_{C-F} = 36.6 Hz, CH_2), 123.2 (q, J_{C-F} = 277.4 Hz, CF_3), 166.08 (s, C=O)		76.04	167.55	20.27 (t, J = 132.2 Hz, CH_2), 37.57 (t, J = 144.3 Hz, NCH_2), 66.18 (t, J = 151.8 Hz, OCH_2)
7c[b]	Barbituric acid moiety		150.42 (s, C=O), 164.84 (s, 2C=O)		80.25	167.85	19.96 (t, J = 132.6 Hz, CH_2), 37.67 (t, J = 146.8 Hz, NCH_2), 67.06 (t, J = 153.3 Hz, OCH_2)
9a	CN	CO_2Me	115.79 (s)	51.85 (q, J = 147.4 Hz, OCH_3), 166.32 (s, C=O)	61.35	169.16	67.87 (t, J = 153.0 Hz, OCH_2), 115.28 (d, J = 161.2 Hz), 118.21 (s), 124.76 (d, J = 160.7 Hz), 125.20 (d, J = 164.5 Hz), 129.80 (d, J = 163.3 Hz), 130.98 (s)
9b	$CO_2CH_2CF_3$	$CO_2CH_2CF_3$	60.19 (t of q, J_{C-H} = 148.9 Hz, J_{C-F} = 37.1 Hz, CH_2), 122.98 (q, J_{C-F} = 277.8 Hz, CF_3), 163.55, 167.46 (2s, C=O)		78.97	165.00	67.56 (t, J = 152.6 Hz, OCH_2), 115.55 (d, J = 161.4 Hz), 118.81 (s), 124.63 (d, J = 160.3 Hz), 125.07 (d, J = 164.5 Hz), 129.78 (d, J = 162.8 Hz), 131.71 (s)
11a E	CN	CO_2Me	116.16	52.48 167.81	77.18	177.97	114.60, 119.52, 125.93, 126.75, 137.03, 140.04, 163.54 (C=O)
Z			113.3	52.58 163.79	73.36	168.90	110.64, 117.74, 126.16, 130.54, 137.71, 137.98, 153.51 (C=O)
11c[b,d]	Barbituric acid moiety						

[a] All signals are singlets. [b] In DMSO-d_6. [c] The E-Isomer signals not observed. [d] No signals were observed because of low solubility in all solvents.