Electronic Supplementary Information

Towards understanding intermolecular interactions in hydantoin derivatives: case of cycloalkane-5-spirohydantoins tethered with a halogenated benzyl moiety

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Table S1 Puckering parameters[1] of the investigated compounds

Compound		q_2, Å	q_3, Å	φ_2, °	φ_3, °
1		0.342(3)		184.8(6)	
2		0.343(4)		4.1(8)	
3		0.004(2)	–0.562(2)	296(30)	
4		0.004(5)	–0.555(5)	106(63)	
5	**A**	0.290(4)	0.599(3)	131.4(8)	346.6(4)
	B	0.504(3)	0.623(3)	272.0(3)	272.8(3)
6	**A**	0.498(4)	0.624(5)	271.9(5)	272.5(4)
	B	0.274(6)	0.597(5)	234.1(12)	14.5(6)

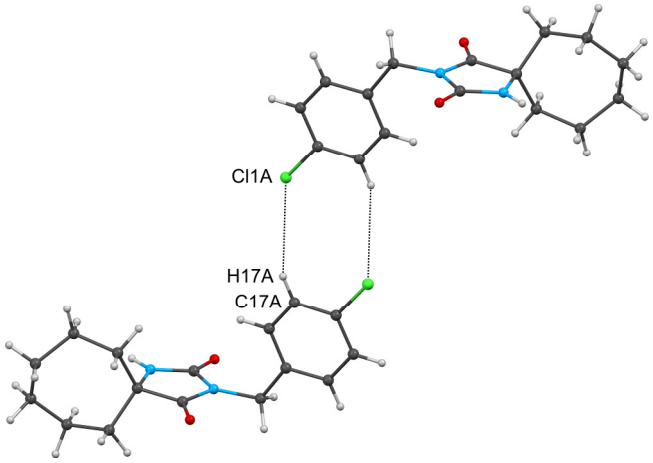

Fig. S1 Dimeric moiety in the crystal structure of **5**.

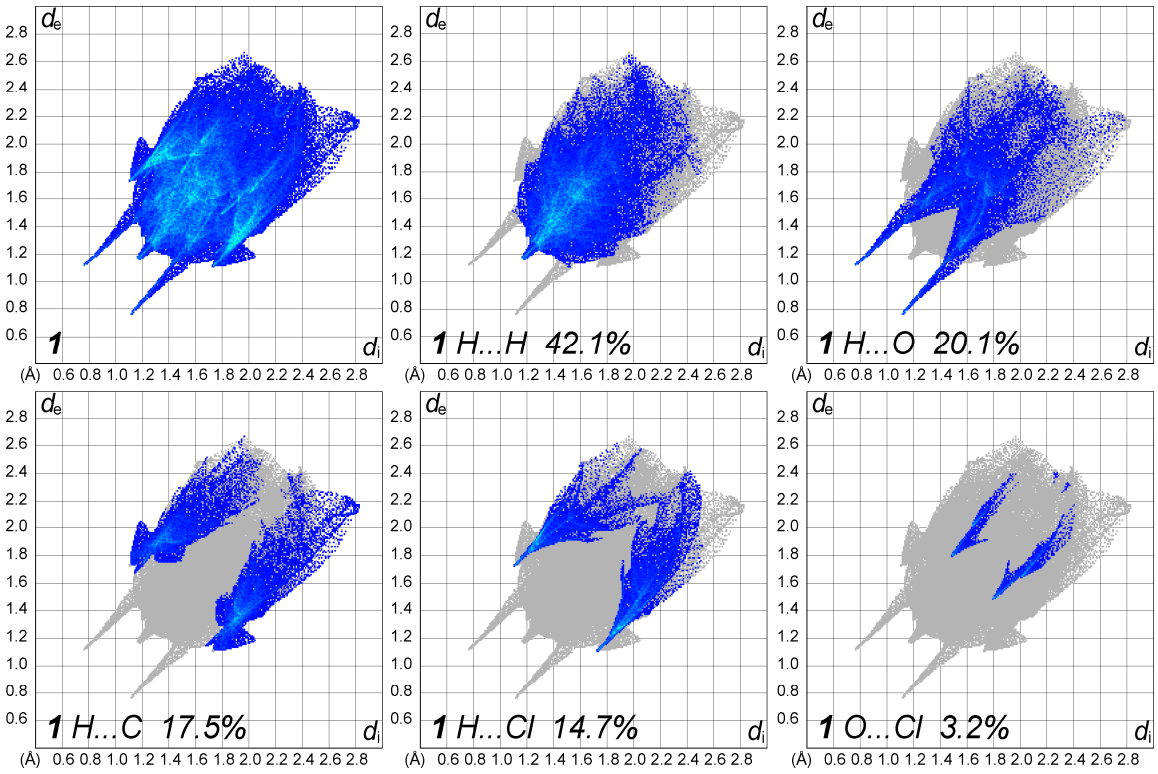

Fig. S2 2D fingerprint plots and decomposed 2D fingerprint plots of the observed atom–atom contacts for **1**.

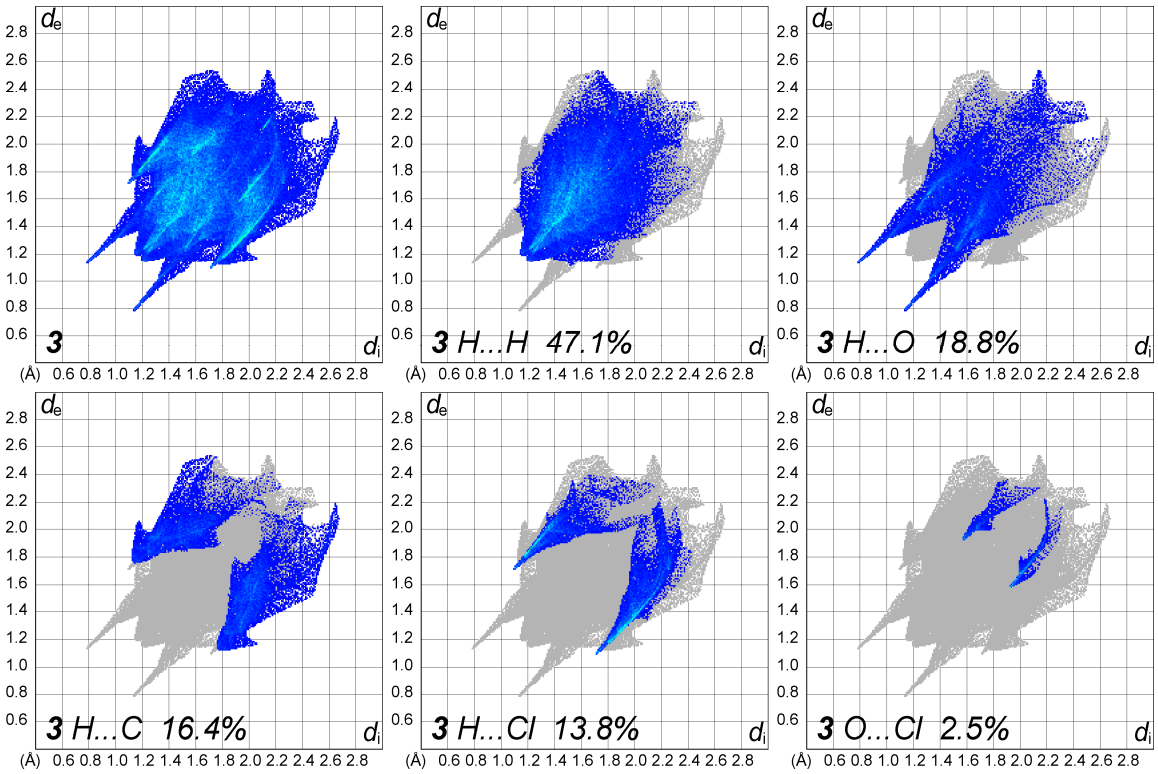

Fig. S3 2D fingerprint plots and decomposed 2D fingerprint plots of the observed atom–atom contacts for **3**.

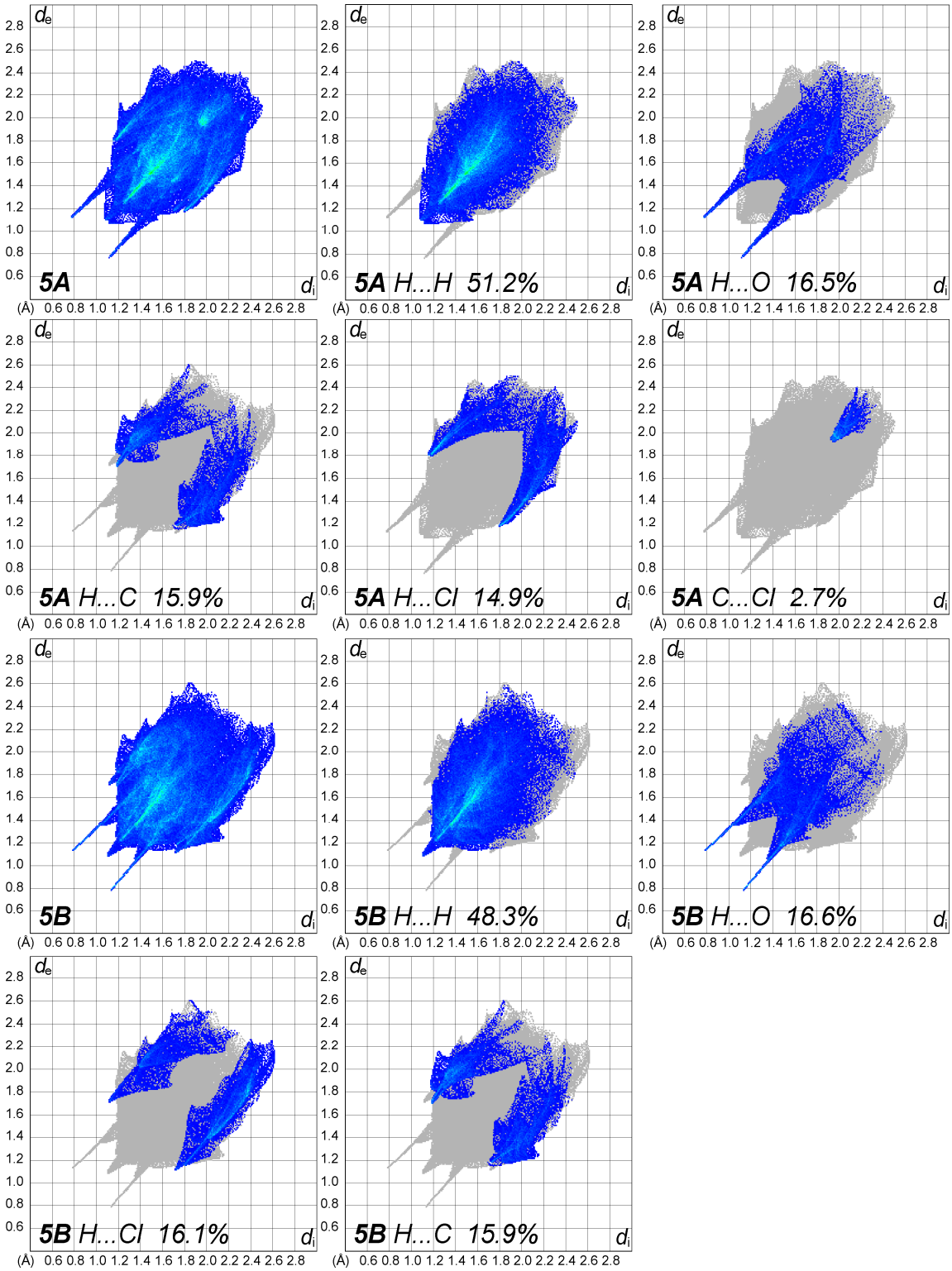

Fig. S4 2D fingerprint plots and decomposed 2D fingerprint plots of the observed atom–atom contacts for two molecules of **5**.

Physico-chemical characterization of the investigated compounds

3-[(4-Chlorophenyl)methyl]-1,3-diazaspiro[4.4]nonane-2,4-dione (**1**). yield: 62%. m.p. 128–131 °C. FTIR (KBr) ν (cm^{-1}): 3211 (NH), 1771 (C=O), 1711 (C=O). ^{1}H NMR (200 MHz, DMSO-d_6): δ = 8.61 (s, 1H, NH), 7.40 (d, 2H, *J* = 10 Hz, C_6H_4), 7.26 (d, 2H, *J* = 10 Hz, C_6H_4), 4.53 (s, 2H, CH_2), 1.94–1.74 (m, 8H, C_5H_8). ^{13}C NMR (50 MHz, DMSO-d_6): δ = 177.7, 155.6, 136.1, 132.3, 129.4, 128.8, 67.6, 40.6, 37.4, 24.8. Elemental analysis, found: C, 60.40; H, 5.45; N, 10.00. Calc. for $C_{14}H_{15}N_2O_2Cl$: C, 60.33; H, 5.42; N, 10.05%.

3-[(4-Bromophenyl)methyl]-1,3-diazaspiro[4.4]nonane-2,4-dione (**2**). yield: 68%. m.p. 162–165 °C. FTIR (KBr) ν (cm^{-1}): 3216 (NH), 1768 (C=O), 1709 (C=O). ^{1}H NMR (200 MHz, DMSO-d_6): δ = 8.61 (s, 1H, NH), 7.54 (d, 2H, *J* = 8 Hz, C_6H_4), 7.20 (d, 2H, *J* = 8 Hz, C_6H_4), 4.51 (s, 2H, CH_2), 1.95–1.75 (m, 8H, C_5H_8). ^{13}C NMR (50 MHz, DMSO-d_6): δ = 177.6, 155.6, 136.6, 131.7, 129.8, 120.7, 67.6, 40.7, 37.4, 24.8. Elemental analysis, found: C, 52.10; H, 4.70; N, 8.70. Calc. for $C_{14}H_{15}N_2O_2Br$: C, 52.03; H, 4.68; N, 8.67%.

3-[(4-Chlorophenyl)methyl]-1,3-diazaspiro[4.5]decane-2,4-dione (**3**). yield: 66%. m.p. 185–186 °C. FTIR (KBr) ν (cm^{-1}): 3232 (NH), 1773 (C=O), 1710 (C=O). ^{1}H NMR (200 MHz, DMSO-d_6): δ = 8.83 (s, 1H, NH), 7.40 (d, 2H, *J* = 8 Hz, C_6H_4), 7.24 (d, 2H, *J* = 8 Hz, C_6H_4), 4.52 (s, 2H, CH_2), 1.68–1.03 (m, 10H, C_6H_{10}). ^{13}C NMR (50 MHz, DMSO-d_6): δ = 176.8, 155.7, 136.1, 132.2, 129.3, 128.8, 61.3, 40.4, 33.5, 24.3, 21.0. Elemental analysis, found: C, 61.57; H, 5.89; N, 9.52. Calc. for $C_{15}H_{17}N_2O_2Cl$: C, 61.54; H, 5.85; N, 9.57%.

3-[(4-Bromophenyl)methyl]-1,3-diazaspiro[4.5]decane-2,4-dione (**4**). yield: 70%. m.p. 193–194 °C. FTIR (KBr) ν (cm^{-1}): 3216 (NH), 1768 (C=O), 1709 (C=O). ^{1}H NMR (200 MHz, DMSO-d_6): δ = 8.83 (s, 1H, NH), 7.53 (d, 2H, *J* = 8 Hz, C_6H_4), 7.18 (d, 2H, *J* = 8 Hz, C_6H_4), 4.50 (s, 2H, CH_2), 1.67–1.03 (m, 8H, C_6H_{10}). ^{13}C NMR (50 MHz, DMSO-d_6): δ = 176.8, 155.7, 136.6, 131.7, 129.6, 120.7, 61.3, 40.5, 33.5, 24.5, 21.0. Elemental analysis, found: C, 53.48; H, 5.02; N, 8.35. Calc. for $C_{15}H_{17}N_2O_2Br$: C, 53.43; H, 5.08; N, 8.31%.

3-[(4-Chlorophenyl)methyl]-1,3-diazaspiro[4.6]undecane-2,4-dione (**5**). yield: 65%. m.p. 170–173 °C. FTIR (KBr) ν (cm^{-1}): 3231 (NH), 1769 (C=O), 1721 (C=O). ^{1}H NMR (200 MHz, DMSO-d_6): δ = 8.68 (s, 1H, NH), 7.39 (d, 2H, *J* = 8.4 Hz, C_6H_4), 7.23 (d, 2H, *J* = 8.6 Hz, C_6H_4), 4.49 (s, 2H, CH_2), 1.85–1.55 (m, 12H, C_7H_{12}). ^{13}C NMR (50 MHz, DMSO-d_6): δ = 178.0, 155.6, 136.2, 132.2, 129.3, 128.8, 63.9, 40.4, 37.1, 28.9, 22.3. Elemental analysis, found: C, 62.59; H, 6.27; N, 9.10. Calc. for $C_{16}H_{19}N_2O_2Cl$: C, 62.64; H, 6.24; N, 9.13%.

3-[(4-Bromophenyl)methyl]-1,3-diazaspiro[4.6]undecane-2,4-dione (**6**). yield: 67%. m.p. 178–180 °C. FTIR (KBr) ν (cm^{-1}): 3232 (NH), 1770 (C=O), 1720 (C=O). ^{1}H NMR (200 MHz, DMSO-*d*6): δ = 8.68 (s, 1H, NH), 7.52 (d, 2H, *J* = 8.4 Hz, C_6H_4), 7.17 (d, 2H, *J* = 8.4 Hz, C_6H_4), 4.47 (s, 2H, CH_2), 1.85–1.55 (m, 12H, C_7H_{12}). ^{13}C NMR (50 MHz, DMSO-d_6): δ = 178.0, 155.6, 136.6, 131.7, 129.7, 120.7, 63.9, 40.5, 37.1, 28.9, 22.3. Elemental analysis, found: C, 54.68; H, 5.48; N, 8.03. Calc. for $C_{16}H_{19}N_2O_2Br$: C, 54.71; H, 5.45; N, 7.98 %.

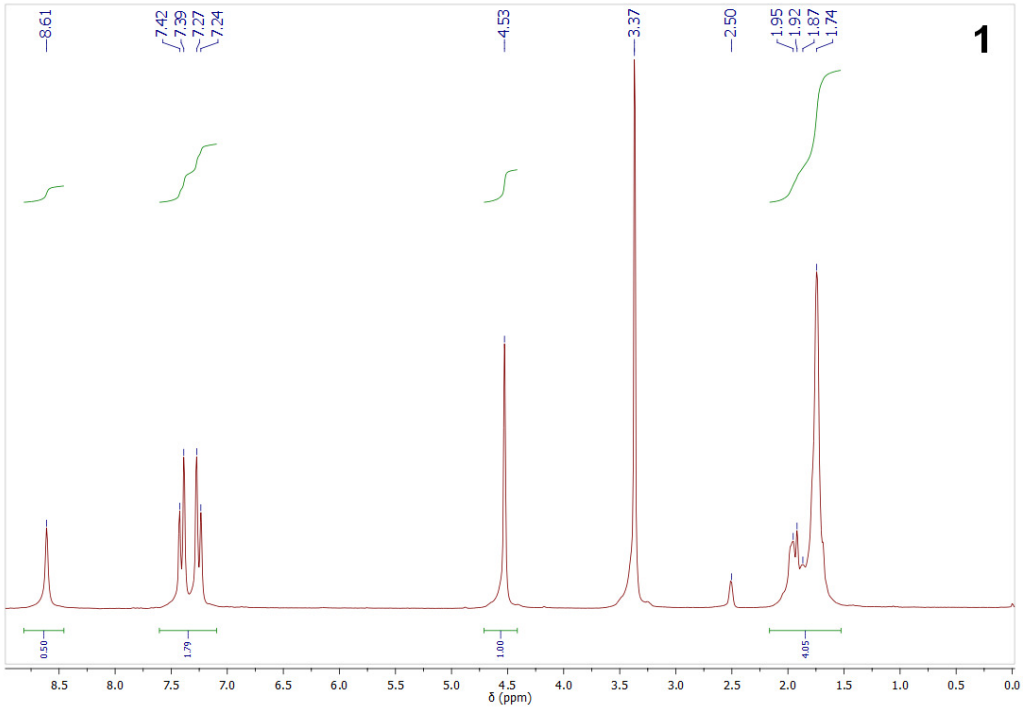

Fig. S5 ^{1}H NMR spectrum of 3-[(4-chlorophenyl)methyl]-1,3-diazaspiro[4.4]nonane-2,4-dione.

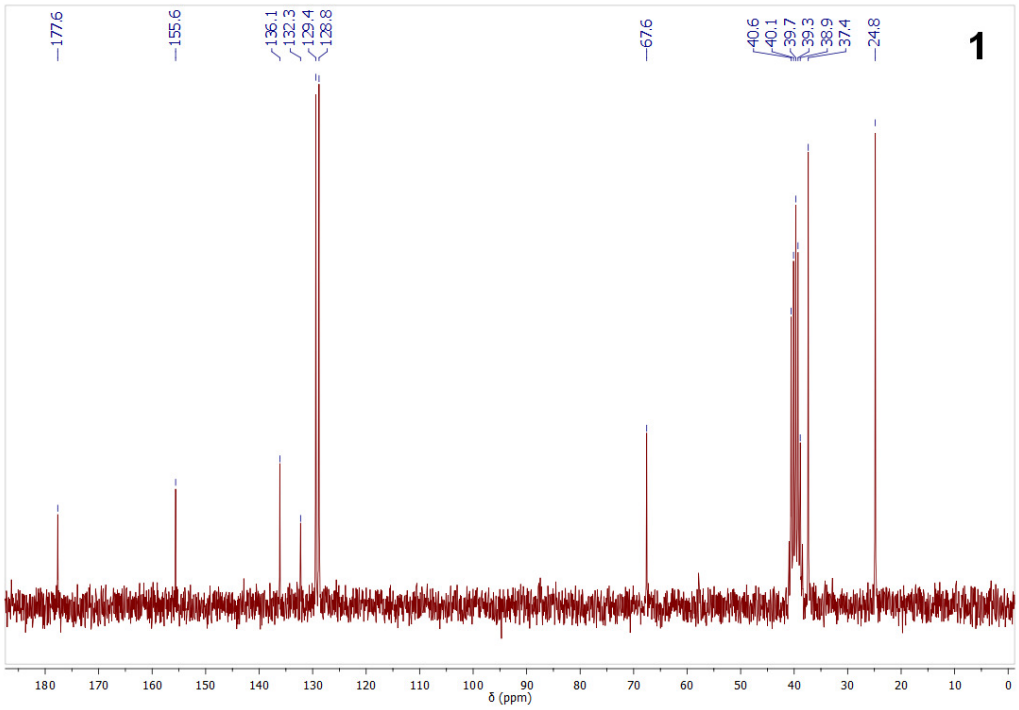

Fig. S6 ^{13}C NMR spectrum of 3-[(4-chlorophenyl)methyl]-1,3-diazaspiro[4.4]nonane-2,4-dione.

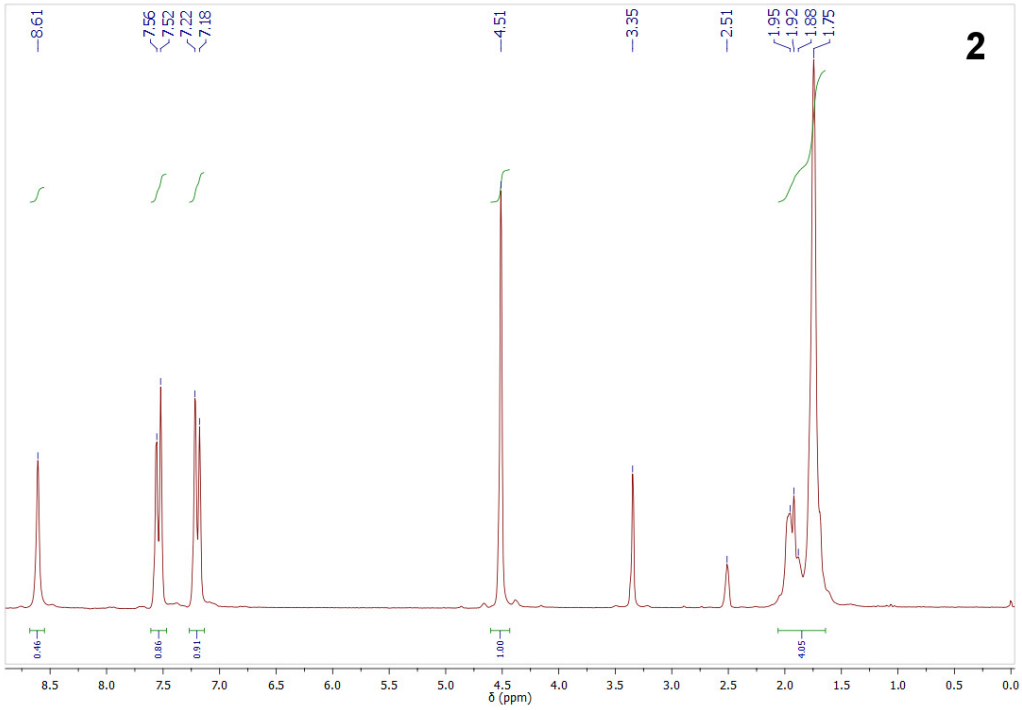

Fig. S7 ^{1}H NMR spectrum of 3-[(4-bromophenyl)methyl]-1,3-diazaspiro[4.4]nonane-2,4-dione.

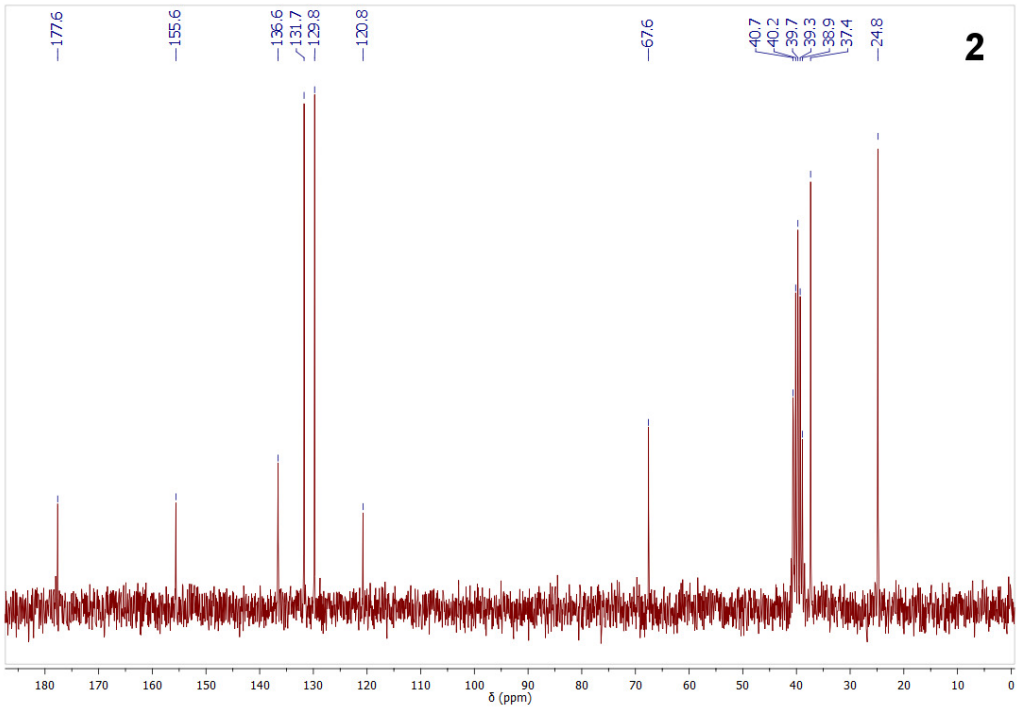

Fig. S8 ^{13}C NMR spectrum of 3-[(4-bromophenyl)methyl]-1,3-diazaspiro[4.4]nonane-2,4-dione.

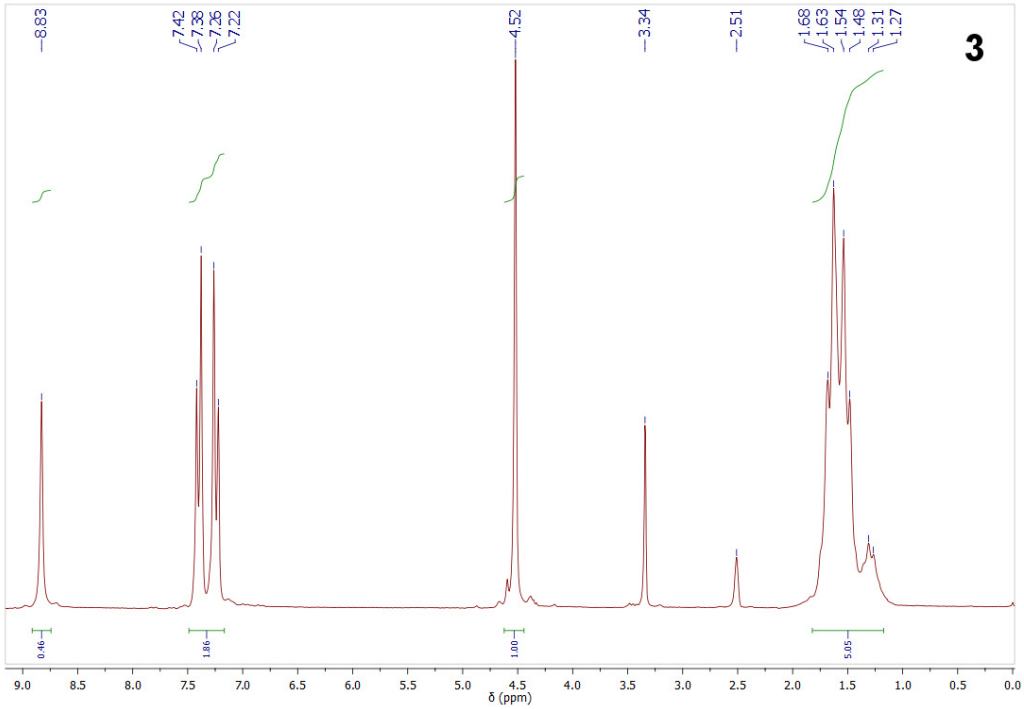

Fig. S9 ^{1}H NMR spectrum of 3-[(4-chlorophenyl)methyl]-1,3-diazaspiro[4.5]decane-2,4-dione.

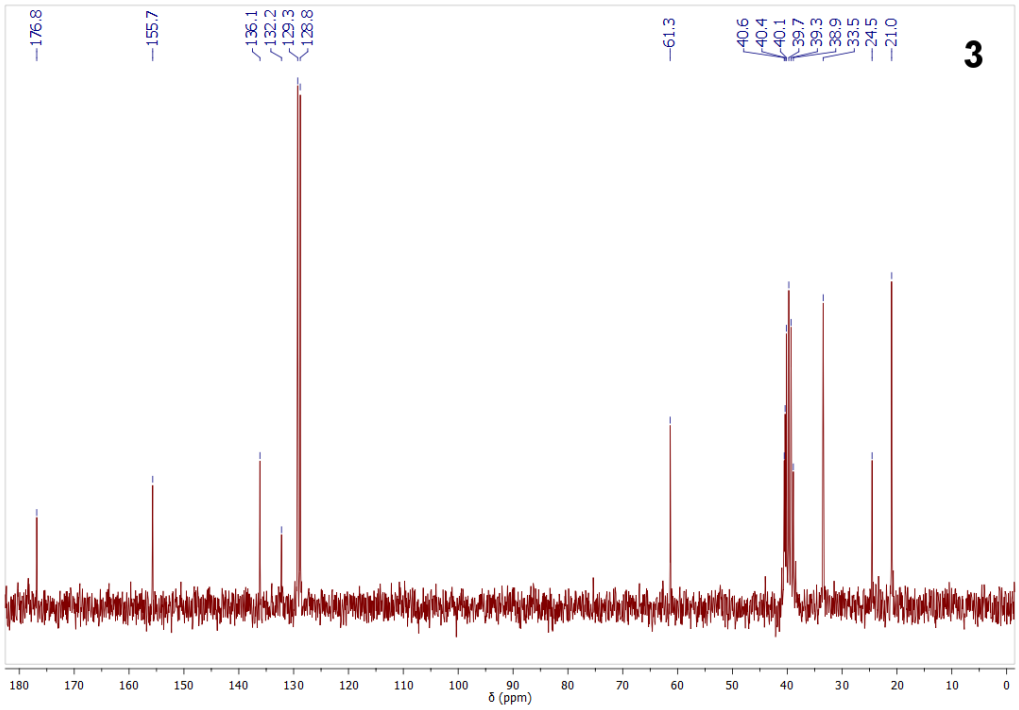

Fig. S10 ^{13}C NMR spectrum of 3-[(4-chlorophenyl)methyl]-1,3-diazaspiro[4.5]decane-2,4-dione.

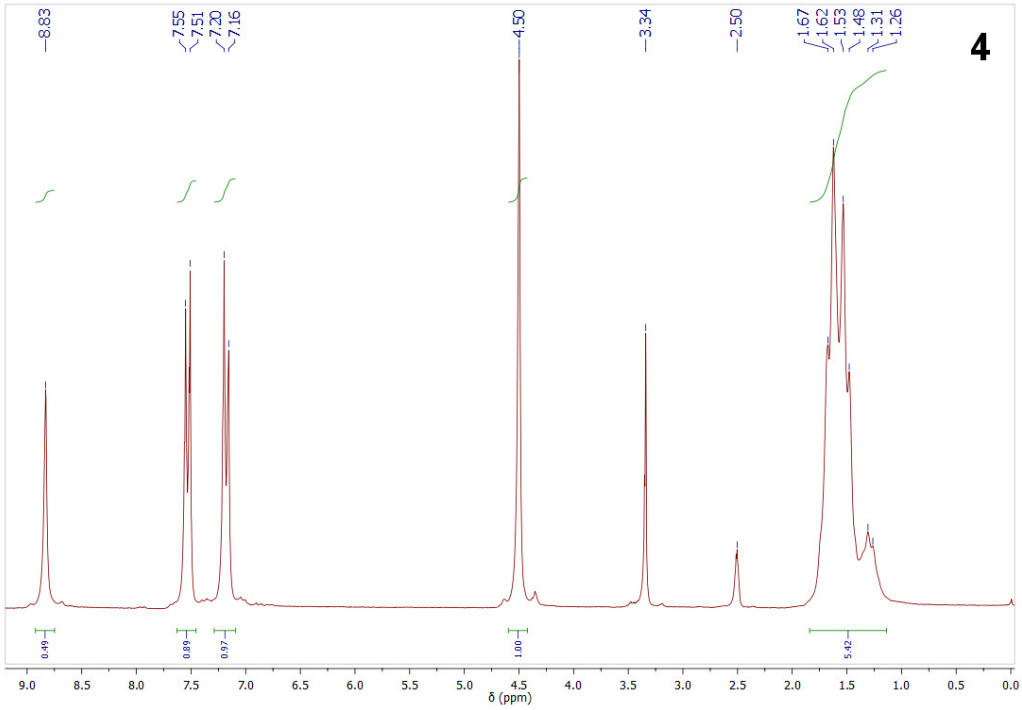

Fig. S11 ^{1}H NMR spectrum of 3-[(4-bromophenyl)methyl]-1,3-diazaspiro[4.5]decane-2,4-dione.

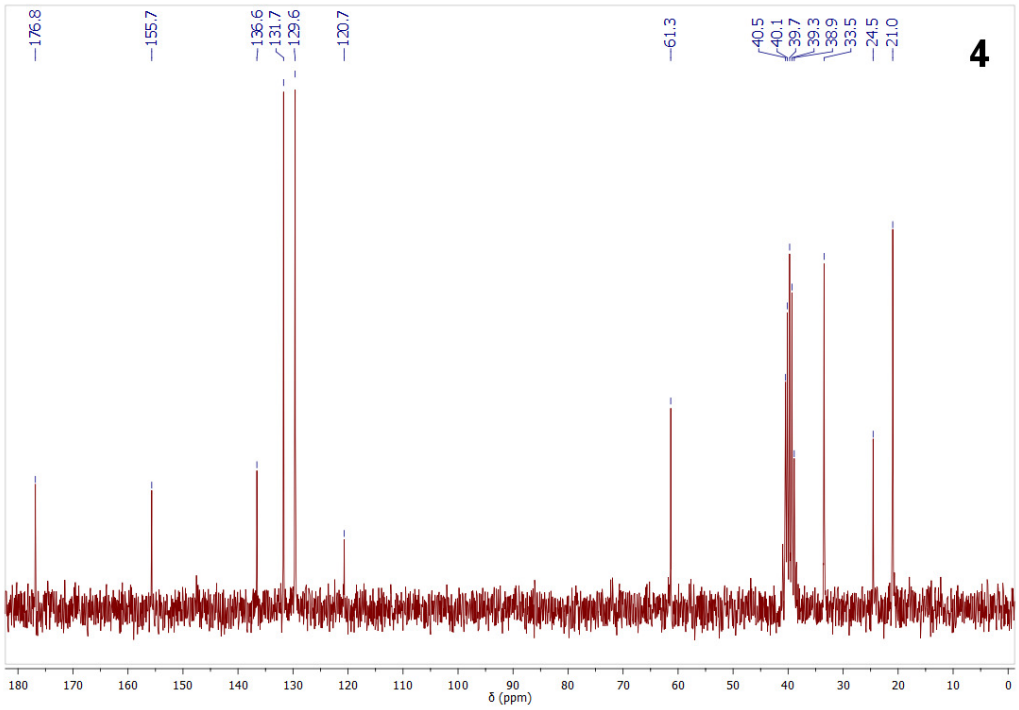

Fig. S12 ^{13}C NMR spectrum of 3-[(4-bromophenyl)methyl]-1,3-diazaspiro[4.5]decane-2,4-dione.

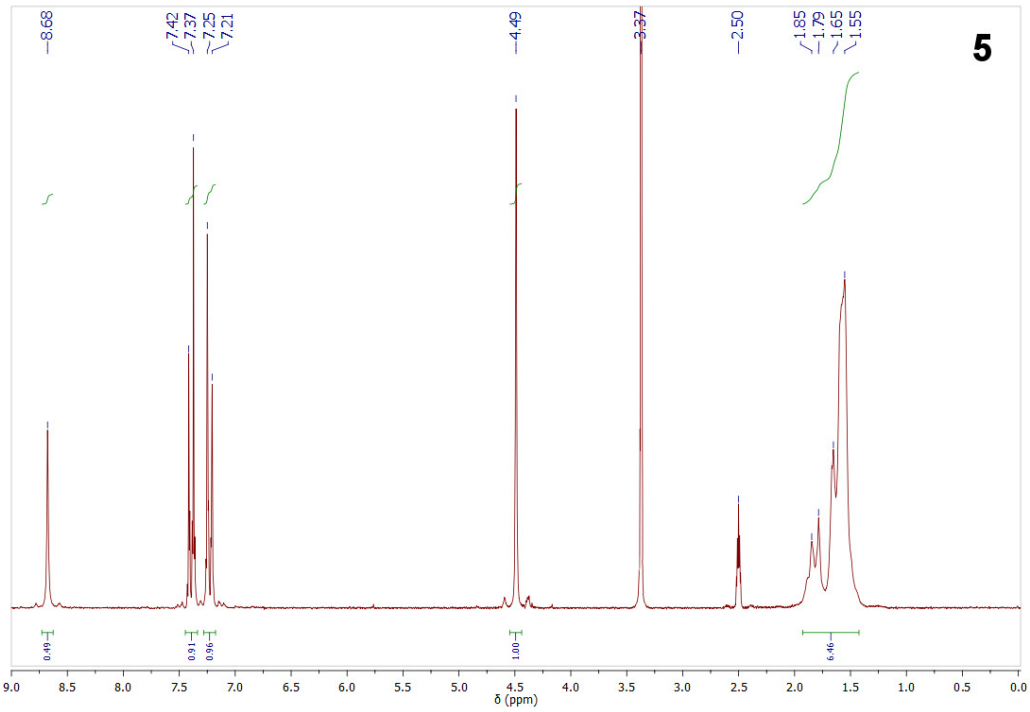

Fig. S13 ^{1}H NMR spectrum of 3-[(4-chlorophenyl)methyl]-1,3-diazaspiro[4.6]undecane-2,4-dione.

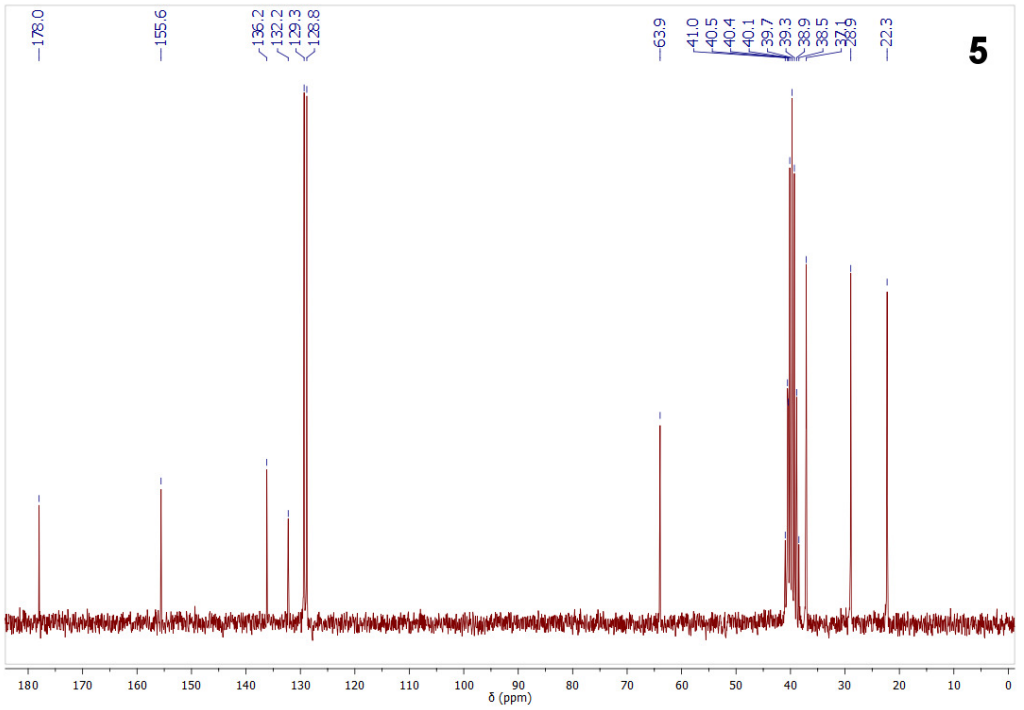

Fig. S14 ^{13}C NMR spectrum of 3-[(4-chlorophenyl)methyl]-1,3-diazaspiro[4.6]undecane-2,4-dione.

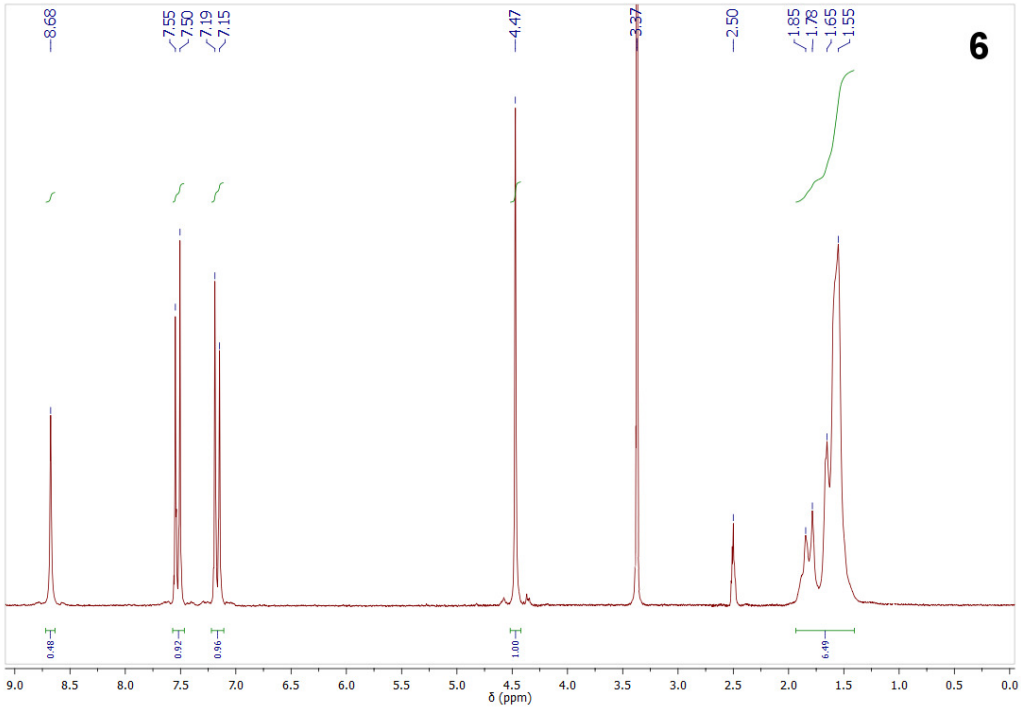

Fig. S15 ^{1}H NMR spectrum of 3-[(4-bromophenyl)methyl]-1,3-diazaspiro[4.6]undecane-2,4-dione.

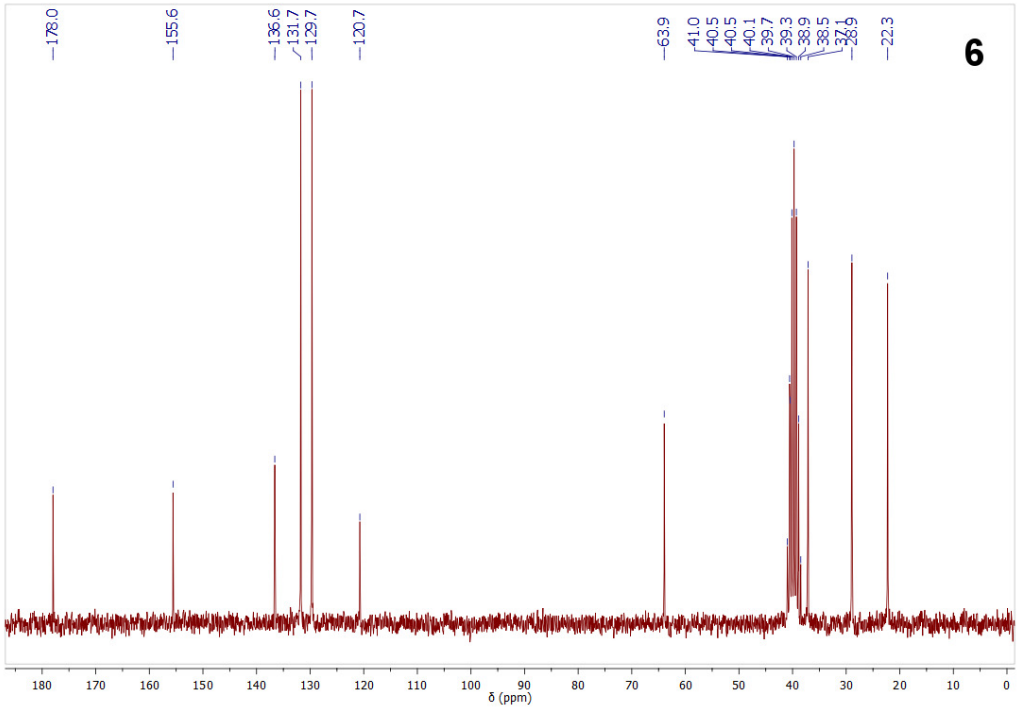

Fig. S16 ^{13}C NMR spectrum of 3-[(4-bromophenyl)methyl]-1,3-diazaspiro[4.6]undecane-2,4-dione.

References

1 J. B. Hendrickson, *J. Am. Chem. Soc.*, 1967, **89**, 7036.