Supporting Information

Solvent-free, mechanochemically scalable synthesis of 2,3-dihydroquinazolin-4(1H)-one using Brønsted acid catalyst

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Contents

Number of pages: **24**

Number of Figures: **16**

Number of schemes: 4

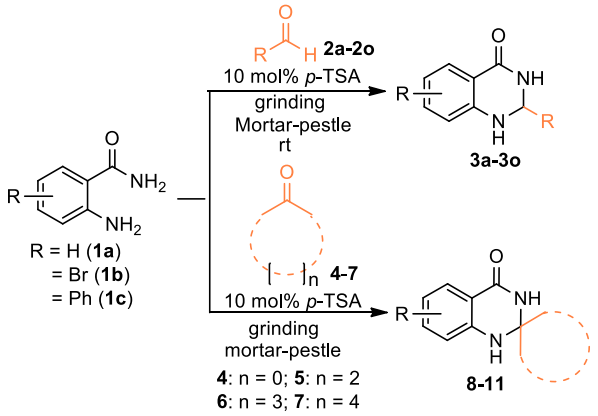
Number of Tables: **5**

General Aspects

All chemicals were received from Sigma Aldrich, Spectrochem, S.D Fine chemicals, Avra chemicals and used as such without further purification. The reactions were monitored by TLC carried out on Merck silica gel (60F254) by using UV light and phosphomolybdic acid as visualizing and developing agents. Proton nuclear magnetic resonance spectra (^{1}H NMR) were obtained at 400 MHz on Agilent spectrometer. ^{13}C NMR spectra were obtained at 100 MHz on Agilent spectrometer. Spectra were recorded in $CDCl_3$ and DMSO-d_6 solutions. Chemical shifts are reported in ppm, referenced to tetramethylsilane (TMS) as the external reference. Hydrogen coupling patterns are described as singlet (s), doublet (d), triplet (t) and multiplet (m). Coupling constants (J) are reported in Hertz. Melting points of all the compounds were recorded by Analab ThermoCal melting point apparatus in the open capillary tube. All grinding reactions are carried out in mortar and pestle at room temperature. Ball milling was performed in tumbler ball milling instrument purchased from S.F. Engineering works having frequency 40 rpm, voltage 230 V, motor power 18.5 kW, and current 24 A. Internal diameter of milling vessel was 5 cm with 200 ml capacity.

General Method For The Preparation Of The 2,3-dihydroquinazolin-4(1H)-one (3a-3o, and 8-11)

Scheme S1. Reaction of anthranilamides (**1a-c**) with aldehydes (**2a-o**) or ketones (**4-7**) under grinding in mortar pestle.

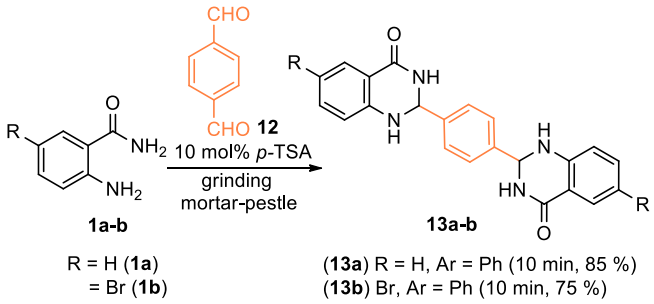


Procedure: To a mixture of anthranilamide **1** (0.100g, 0.73mmol, 1 equiv.), and *p*-nitrobenzaldehyde **2** (0.122g, 0.81mmol, 1.1 equiv.) was added *p*-TSA catalyst (10 mol %) and ground in a mortar-pestle at room temperature. Upon completion of the reaction, as monitored by TLC, water was added to the reaction mixture to remove the *p*-TSA, and the product was filtered off. The residual solid was washed with water and then with 2% EtOAc-hexane mixture to remove excess aldehyde and to isolate 0.183 g (93%) of the desired product.

Similar procedure was followed for the synthesis of the compounds **3a-3o** and **8-11**.

General procedure for the preparation of bisquinazolinone (13a-b)

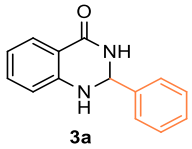
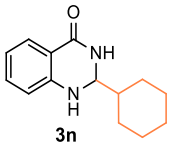
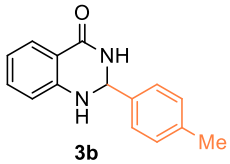
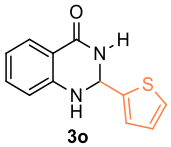
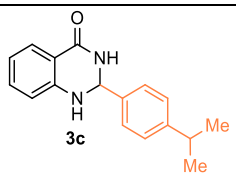
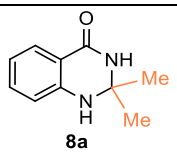
Scheme S2. Reaction of anthranilamides (**1a-b**) with terephthaldehyde (**12**) under grinding in mortar pestle.

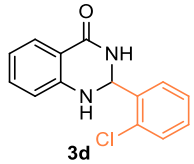
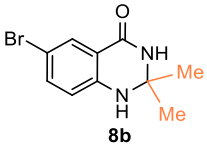
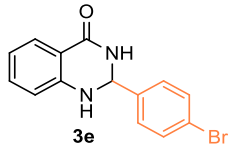
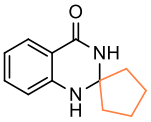
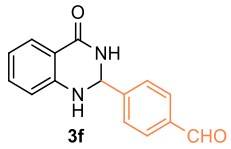
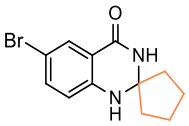
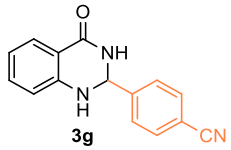
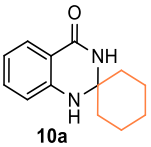
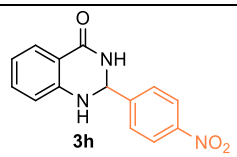
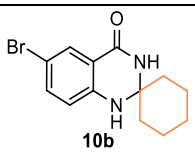
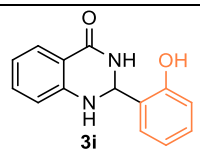
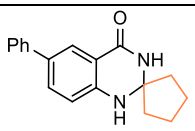
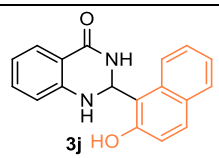
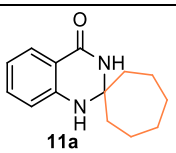
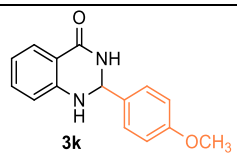
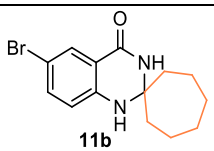
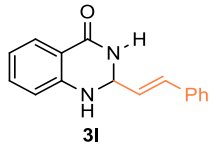
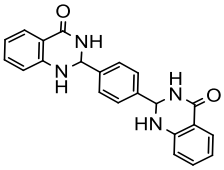
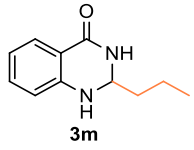
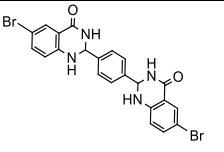


Procedure: To a mixture of terephthaldehyde **12** (0.100g ,0.74mmol, 1 equiv.) and anthranilamide **1** (0.223g, 1.64mmol, 2.2 equiv.) was 10 mol% of *p*-TSA catalyst and ground in a mortar-pestle. The progress of the reaction was monitored by TLC. Upon completion of the reaction, water was added to remove the *p*-TSA, and the product was filtered off. The residual white solid was washed with water and then purified by silica gel column chromatography using DCM:MeOH as eluent to get hold of 0.22 g (80%) of the desired product.

Melting points of the synthesized 2,3-dihydroquinazolin-4(1H)-one 3a-3o, 8-11 and 13 and comparison with the literature reported data

Table S1. Melting point of the synthesized 2,3-dihydroquinazolin-4(1H)-one **3a-3o**, **8-11** and **13**

Sl. No.	Structure	Melting point		Sl. No.	Structure	Melting point	
		Observed	Reported			Observed	Reported
1	 **3a**	220-222 °C	221-223 °C[9,13]	14	 **3n**	178-180 °C	178-180 °C[9,17]
2	 **3b**	220-222 °C	224-225°C[2,14]	15	 **3o**	210-212 °C	213-215 °C[2,14]
3	 **3c**	162-164 °C	162-164 °C[10,15]	16	 **8a**	182-184 °C	182-183 °C[4,21]

4	 3d	200-202 °C	202-204 °C[2,16]	17	 8b	226-228 °C	
5	 3e	198-196 °C	198-199 °C[5,17]	18	 9a	250-252 °C	251-253 °C[3,4]
6	 3f	185-187 °C	185-187 °C[10,18]	19	 9b	210-212 °C	-
7	 3g	250-252 °C	250-252 °C[8,19]	20	 10a	215-217 °C	217-219 °C[3,4]
8	 3h	198-200 °C	198-200 °C[7,17]	21	 10b	218-220 °C	-
9	 3i	216-218 °C	218-219 °C[1,19]	22	 10c	250-252 °C	-
10	 3j	170-172 °C	172-174 °C[10,19]	23	 11a	202-204 °C	204-205 °C[3,17]
11	 3k	180-182°C	181-182 °C[2,17]	24	 11b	206-208 °C	-
12	 3l	224-226 °C	224-226 °C[11,20]	25		245-247°C	245-246 °C[5,8]
13	 3m	173-175 °C	175-178 °C[5,8]	26		>260°C	-

Characterization data of the 2,3-dihydroquinazolin-4(1H)-one derivatives

2-Phenyl-2,3-dihydroquinazolin-4(1H)-one (3a):[9,13] Color = White solid (0.156 g, 95% yield); MP = 220-222 ºC (purified by washing with PE/EA = 98:2); IR (KBr): 3305, 3180, 3059, 1665, 1615, 1535, 1450 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) δ: (in ppm) 5.72 (s, 1H), 6.76 – 6.57 (m, 2H), 7.64 – 7.00 (m, 8H), 8.24 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 66.9, 114.8, 115.4, 117.5, 127.3, 127.8, 128.8, 128.9, 133.7, 142.1, 148.3, 164.0.

2-(*p*-Tolyl)-2,3-dihydroquinazolin-4(1H)-one (3b):[2,14] Color = White solid (0.140 g, 80% yield); MP = 224-225°C (purified by washing with PE/EA = 98:2); IR (KBr): 3311, 1657, 1607, 1508, 1484, 1383 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) δ: (in ppm) 2.30 (s, 3H), 5.73 (s, 1H), 6.77-6.66 (m, 2H), 7.06 (s, 1H), 7.27-7.19 (m, 3H), 7.39 (d, J = 7.6 Hz,2H), 7.63 (d, J = 8.0 Hz, 1H), 8.24 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 21.2, 66.9, 114.9, 115.5, 117.5, 127.3, 127.8, 129.3, 133.7, 138.2, 139.1, 148.4, 164.1.

2-(4-Isopropylphenyl)-2,3-dihydroquinazolin-4(1H)-one (3c):[10,15] Color = White solid (0.141 g, 85% yield); MP = 162-164 °C (purified by washing with PE/EA = 98:2); ^{1}H NMR (400 MHz, DMSO-d_6) δ: (in ppm) 1.16 (d, J = 6.9 Hz, 6H), 2.85 (dt, J = 13.7, 6.8 Hz, 1H), 5.69 (s, 1H), 5.69 (s, 1H), 6.64 (t, J = 7.4 Hz, 1H), 6.70 (d, J = 8.0 Hz, 1H), 7.02 (s, 1H), 7.27 – 7.17 (m, 3H), 7.39 (d, J = 8.1 Hz, 2H), 7.58 (d, J = 6.8 Hz, 1H), 8.19 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 24.3, 33.7, 67.0, 114.8, 115.4, 117.5, 126.7, 127.4, 127.8, 133.7, 139.4, 148.4, 149.2, 164.1.

2-(2-Chlorophenyl)-2,3-dihydroquinazolin-4(1H)-one (3d):[2,9] Color = Off white solid; (0.171 g, 90%); MP = 200-202 °C (purified by washing with PE/EA = 98:2); ^{1}H NMR (400 MHz, DMSO-d_6) δ: (in ppm) 6.11 (s, 1H), 6.72 (dd, J = 16.3, 7.5 Hz, 2H), 6.98 (s, 1H), 7.22 (d, J = 6.9 Hz, 1H), 7.37 (s, 2H), 7.46 (s, 1H), 7.63 (d, J = 5.6 Hz, 2H), 8.18 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 64.1, 115.0, 115.1, 117.9, 127.8, 127.9, 129.2, 130.0, 130.7, 132.3, 133.9, 138.3, 148.1, 164.0.

2-(4-Bromophenyl)-2,3-dihydroquinazolin-4(1H)-one (3e):[5,16] Color = White solid (0.207 g, 93% yield); MP = 196-198 °C (purified by washing with PE/EA = 98:2); ^{1}H NMR (400 MHz, DMSO-d_6) δ: (in ppm) 5.76 (s, 1H), 6.68 (t, J = 7.5 Hz, 1H), 6.75 (d, J = 8.1 Hz, 1H), 7.15 (s, 1H), 7.25 (t, J = 7.7 Hz, 1H), 7.44 (d, J = 8.4 Hz, 2H), 7.60 (t, J = 7.4 Hz, 3H), 8.34 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 67.7, 116.4, 116.8, 119.2, 123.5, 129.3, 131.0, 133.1, 135.3, 143.0, 149.5, 165.4.

4-(4-Oxo-1,2,3,4-tetrahydroquinazolin-2-yl)benzaldehyde (3f):[10,17] Color = White solid (0.148 g, 80% yield); MP = 185−187 °C (purified by washing with PE/EA = 98:2); IR (KBr): 3296, 3190,3070, 1656, 1606 cm^{-1}; ^{1}H NMR ($CDCl_3$ + DMSO-d_6 300 MHz) δ : (in ppm) 5.87 (s, 1H), 6.75−6.67 (m, 2H), 7.24−7.19 (m, 2H), 7.38 (s, 1H), 7.85−7.67 (m, 5H), 9.95 (s, 1H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ: (in ppm) 66.3, 114.9, 115.3, 117.8, 127.9, 130.0, 133.9, 136.6, 147.9, 148.7, 163.8, 193.2.

4-(4-Oxo-1,2,3,4-tetrahydroquinazolin-2-yl)benzonitrile (3g):[8,18] Color = Pale-yellow crystal (0.164 g, 90% yield); MP = 250-252 °C (purified by washing with PE/EA = 98:2); ^{1}H NMR (400 MHz, DMSO-d_6) δ: (in ppm) 5.83 (s, 1H), 6.67 (t, *J* = 7.28 Hz, 1H), 6.73 (d, *J* = 8.04 Hz, 1H), 7.23 (t, *J* = 7.28 Hz, 1H), 7.26 (s, 1H), 7.58 (d, *J* = 7.52 Hz, 1H), 7.63 (d, *J* = 8.04 Hz,2H), 7.84 (d, *J* = 8.28 Hz, 2H), 8.45 (br s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 65.9, 111.5, 114.9, 115.3, 117.9, 127.8, 128.1, 132.8, 134.0, 147.7, 147.8, 163.8.

2-(4-Nitrophenyl)-2,3-dihydroquinazolin-4(1H)-one (3h):[7,16] Color = Yellow solid (0.183 g, 93% yield); MP = 198-200 °C (purified by washing with PE/EA = 98:2); ^{1}H NMR (400 MHz, DMSO-d_6) δ: (in ppm) 5.88 (s, 1H), 6.74 - 6.64 (m, 2H), 7.21 - 7.28 (m, 2H), 7.58 (d, *J* = 4 Hz, 1H), 7.71 (d, *J* = 8 Hz, 2H), 8.22 (d, *J* = 8 Hz, 2H), 8.47 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 65.7, 115.0, 115.3, 117.9, 124.0, 127.8, 128.4, 134.0, 147.7, 147.8, 149.7, 163.7.

2-(2-Hydroxyphenyl)2,3-dihydroquinazolin-4(1H)-one (3i):[1,18] Color = White solid (0.158 g, 90% yield); MP = 216-218 °C (purified by washing with PE/EA = 98:2); ^{1}H NMR (400 MHz, DMSO-d_6) δ: (in ppm) 5.98 (s, 1H), 6.64 (t, *J* = 7.52 Hz, 1H), 6.71 (s, 1H), 6.75 (t, *J* = 8.28 Hz, 1H), 6.78 (d, *J* = 7.52 Hz, 1H), 6.83(d, J = 8.04 Hz, 1H), 7.13 (t, *J* = 6.76 Hz, 1H), 7.20 (t, *J* = 7.04 Hz, 1H), 7.31 (d, *J* =7.8 Hz, 1H), 7.59 (d, *J* = 7.76 Hz, 1H), 7.90 (s, 1H), 9.83 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 61.6, 115.0, 115.2, 115.8, 117.4, 119.2, 127.6, 127.7, 127.7, 129.7, 133.6, 148.5, 155.0, 164.4.

2-(2-Hydroxynaphthalen-1-yl)-2,3-dihydroquinazolin-4(1H)-one (3j):[10,18] Color = Orange solid (0.191 g, 90% yield) ; MP =170-172 °C (purified by washing with PE/EA = 98:2); ^{1}H NMR (400 MHz, CD_3OD) δ: (in ppm) 6.82 (d, *J* = 9.28Hz, 1H), 7.22−7.30 (m, 2H), 7.45 (t, *J* = 8.28 Hz, 1H), 7.56 (t, *J* = 8.04 Hz,1H), 7.61 (d, *J* = 7.76 Hz, 2H), 7.66 (d, *J* = 8.28 Hz, 1H), 7.75 (d, *J* = 9.28 Hz, 1H), 8.18 (d, *J* = 8.56 Hz, 1H), 9.35 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 109.2, 119.5, 120.7, 123.5, 123.9, 126.0, 127.0, 128.6, 128.8, 129.4, 129.5, 131.6, 134.0, 137.8, 142.4, 154.2, 169.6, 172.6.

2-(4-Methoxyphenyl)-2,3-dihydroquinazolin-4(1H)-one (3k):[2,16] Color = White solid (0.112 g, 60% yield); MP = 180-182 °C (purified by washing with PE/EA = 98:2); ^{1}H NMR (400 MHz, DMSO-d_6) δ: (in ppm) 3.75 (s, 3H), 5.72 (s, 1H), 6.68 (s, 1H), 6.75 (d, *J* = 8.1Hz, 1H), 6.96 (s, 2H), 7.02 (s, 1H), 7.25 (s, 1H), 7.43 (d, *J* = 8.6Hz, 2H), 7.62 (d, *J* = 7.6Hz, 1H), 8.19 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 57.1, 68.2, 115.5, 116.3, 116.9, 119.0,129.2, 130.1, 135.1, 135.4, 149.9, 161.3, 165.6.

(E)-2-Styryl-2,3-dihydroquinazolin-4(1H)-one (3l):[11.19] Color = Pale yellow solid (0.091 g, 50% yield) ; MP = 224-226 °C (purified by washing with PE/EA = 98:2); ^{1}H NMR (500 MHz, DMSO-d_6) δ: (in ppm) 5.31 (d, *J* = 6.7 Hz, 1H), 6.37 (dd, *J* = 15.8 Hz and 6.8Hz, 1H), 6.67 (dd, *J* = 14.9 Hz and 6.5 Hz, 2H), 6.75 (d, *J* = 8.0 Hz, 1H), 6.89 (br s, 1H), 7.29-7.23 (m, 2H), 7.34 (t, *J* = 7.5Hz, 2H), 7.45 (d, *J* = 7.5 Hz, 2H), 7.62 (d, *J* = 7.6 Hz, 1H), 8.14 (brs, 1H); ^{13}C NMR (125 MHz, DMSO-d_6) δ: (in ppm) 65.8, 114.5, 114.8, 117.1, 126.6, 127.3, 128.1, 128.3, 128.7, 131.6, 133.2, 135.7, 147.7, 163.3.

2-Propyl-2,3-dihydroquinazolin-4(1H)-one (3m):[5,8] Color = Red solid (0.125 g, 90% yield); MP = 173-175 °C (purified by washing with PE/EA = 98:2); IR (KBr): 3214, 3021, 1670 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$) δ: (in ppm) 1.01 (t, *J* = 4.0 Hz, 3H), 1.52-1.46 (m, 2H), 1.78 (q, 2H), 4.22 (brs, 1H), 4.90 (t, *J* = 4.0 Hz, 1H), 6.24 (brs, 1H), 6.67 (d, *J* = 4.0Hz, 1H), 6.87 (t, *J* = 6.0 Hz, 1H), 7.31 (t, *J* = 4.0 Hz, 1H), 7.89 (d, *J* = 4.0 Hz, 1H); ^{13}C NMR ($CDCl_3$, 100 MHz) δ: (in ppm) 14.0, 17.6, 37.8, 65.3, 114.9, 116.1, 119.6, 128.8, 134.1, 147.6, 165.7.

2-Cyclohexyl-2,3-dihydroquinazolin-4(1H)-one (3n):[9,16] Color = White solid (0.135 g, 80% yield); MP = 178-180 °C (purified by washing with PE/EA = 98:2); ^1H NMR (400MHz, DMSO-d_6) δ: (in ppm) 1.11 (s, 5H), 1.70 (d, *J* = 10.6Hz, 6H), 4.44 (s, 1H), 6.54 (s, 1H), 6.60 (s, 1H), 6.74 (d, *J* = 8.1Hz, 1H), 7.19 (s, 1H), 7.55 (d, *J* = 7.6Hz, 1H), 7.87 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 27.4, 27.5, 27.8, 28.6, 28.9, 41.2, 41.4, 41.6, 44.7, 70.5, 116.0, 116.7, 118.3, 129.1, 134.9, 150.2, 165.6.

2-(Thiophen-2-yl)-2,3-dihydroquinazolin-4(1H)-one (3o):[2,14] Color = White Solid (0.143 g, 85% yield); MP = 223-224°C (purified by washing with PE/EA = 98:2); IR (KBr): 3287, 1650, 1607, 1515, 1487, 763 cm-1; ^1H NMR (400 MHz, DMSO-d_6) δ: (in ppm) 6.03 (s, 1H), 6.79 -6.70 (m, 2H), 6.99 (t, *J* = 3.6 Hz, 1H), 7.14 (d, *J* = 2.8 Hz, 1H), 7.29-7.25 (m, 2H), 7.46 (d, *J* = 4.8 Hz,1H), 7.64 (d, *J* = 8.0 Hz, 1H), 8.46 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 63.0, 115.2, 115.6, 118.0, 126.2, 126.4, 126.9, 127.8, 133.8, 146.9, 147.7, 163.6.

2,2-Dimethyl-2,3-dihydroquinazolin-4(1H)-one (8a):[4,20] Color = white solid (0.110 g, 85% yield); MP = 182-184 °C (purified by crystallization in methanol); IR (KBr): 3255, 1632, 1485, 1270, 1175, 750 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$) δ: (in ppm) 1.58 (s, 6H), 4.25 (br s, 1H), 6.64 (d, *J* = 7.6 Hz, 1H), 6.83 (t, *J* = 7.2 Hz, 1H), 7.05 (brs, 1H), 7.31 (t, *J* = 8.0 Hz, 1H), 7.89 (d, *J* = 7.2 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ: (in ppm) 29.6, 67.6, 114.6, 114.7, 118.7, 128.3, 133.9, 146.0, 164.7.

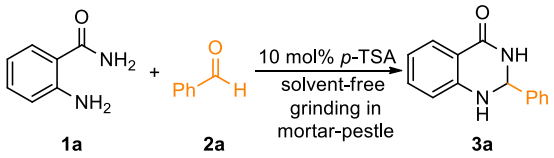
1'H-Spiro[cyclopentane-1,2'-quinazolin]-4'(3'H)-one (9a):[3,4] Color = White solid (0.133 g, 90% yield); MP = 250-252 °C (purified by crystallization in methanol); IR (KBr):3292, 3163, 1639,1616 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) δ: (in ppm) 1.64–1.68 (m, 4 H), 1.77–1.79 (m, 4 H), 6.63 (t, *J* = 7.6 Hz, 1 H), 6.70 (d, *J* = 8.0 Hz, 1 H), 6.74 (s, 1 H), 7.21 (t, *J* = 8.4 Hz, 1 H), 7.57 (d, *J* = 8.0 Hz, 1 H), 8.10 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 22.4, 77.5, 114.8, 115.0, 117.0, 127.7, 133.4, 147.9, 163.9.

1'H-Spiro[cyclohexane-1,2'-quinazolin]-4'(3'H)-one (10a):[3,4] Color = White solid (0.142 g, 90% yield); MP = 215-217 °C (purified by crystallization in methanol); IR (KBr): 1614, 1643, 3183, 3286 cm^{-1}. ^1H NMR (400 MHz, DMSO-d_6) δ: (in ppm) 1.22 (s, 1H), 1.38 (s, 1H), 1.53 (br s, 6H), 1.70 (s, 2H), 6.56 (br s, 2H), 6.77 (d, *J* = 4 Hz, 1H), 7.17 (br s, 1H), 7.52 (d, *J* = 5.7 Hz, 1H), 7.86 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 21.3, 25.1, 37.6, 68.2, 114.9, 115.0, 116.9, 127.5, 133.5, 147.2, 163.6.

1'H-spiro[cycloheptane-1,2'-quinazolin]-4'(3'H)-one (11a):[3,16] Color = Off white solid (0.143 g, 85% yield); MP = 202-204 °C (purified by crystallization in methanol); ^{1}H NMR (400 MHz, DMSO-d_6) δ: (in ppm) 1.50 (s, 8H), 1.87 (s, 4H), 6.59 (s, 1H), 6.71 (s, 2H), 7.20 (t, *J* = 7.6Hz, 1H), 7.54 (d, *J* = 7.7Hz, 1H), 8.02 (s, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) δ: (in ppm) 22.7, 31.1, 42.9, 73.8, 116.1, 116.2, 118.2, 135.0, 148.6, 164.5.

2,2'-(1,4-Phenylene)bis(2,3-dihydroquinazol-4(1H)-one) (13a):[5,8] **Color =** Light brown solid (0.234 g, 85% yield); MP = 245-247 °C (purified by silica-gel column chromatography using DCM/Methanol = 9.3:0.7); IR (KBr): 3416, 3212, 1663, 1669 cm-1; ^{1}H NMR($CDCl_3$, 400 MHz): δ 5.89 (s, 1H), 5.96 (s, 1H), 5.96 (s, 1H), 6.77 (d, *J* = 8.0 Hz, 1H), 6.85(t, *J* = 8.0 Hz, 2H), 6.91 (br s, 1H), 7.32 (t, *J* = 6.0 Hz, 2H), 7.43 (s, 2H), 7.62 (s, 1H), 7.78(d, *J* = 8.0 Hz, 2H), 7.87 (d, *J* = 8.0 Hz, 2H), 7.94 (d, *J* = 8.0 Hz, 2H), 10.04 (s, 1H); ^{13}C NMR ($CDCl_3$ + DMSO-d_6, 100 MHz): δ = 66.3, 114.2, 114.7, 117.1, 126.6, 127.2, 129.3, 133.0, 141.6, 147.4, 164.0.

Table S2. Reproducibility of the reaction between anthranilamide 1a and benzaldehyde 2a



Sl. No.	Time required for complete conversion of **1a**	% yield
1	3	95
2	3	94
3	3	95

Solubility profile of anthranilamide and benzaldehyde in molten *p*-TSA

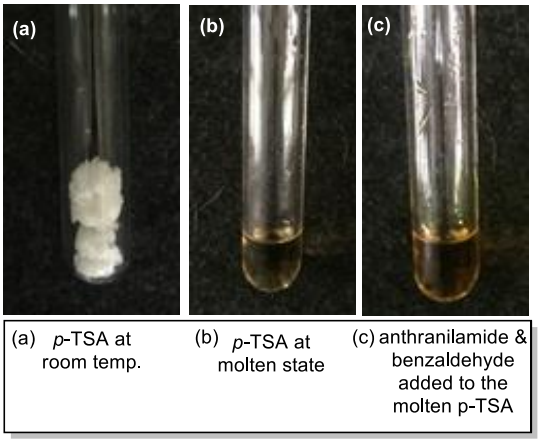


Figure S1. Solubility profile of anthranilamide and benzaldehyde in molten *p*-TSA.

Calculation Of The Green Chemistry Metrics for the reaction between anthranilamide 1a and *p*-nitrobenzaldehyde 2h in presence of 10 mol% *p*-TSA

Scheme S3. Synthesis of 2-(4-nitrophenyl)-2,3-dihydroquinazolin-4(1H)-one (**3h**) from anthranilamide and *p*-nitrobenzaldehyde by grinding in mortar pestle

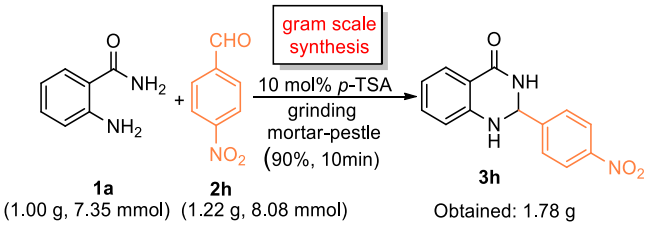


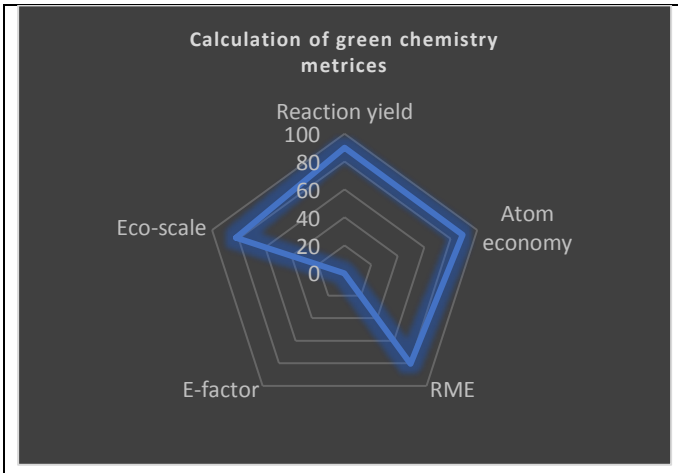
Table S3. Calculation Of The Green Chemistry Metrics.

1	**Atom economy** [12]	
	Atom Economy = (Exact molecular mass of desired product (C) / Exact Molecular mass of all reactant (A+B)) X 100%	= (269.26/136.15 + 1.1 (151.12) X 100% AE = **89.04%**
2	**Reaction mass efficiency** [12]	
	Reaction Mass Efficiency = (Mass of product (C)/ Mass of reactant (A) + Mass of reactant (B)) X 100	= (1.786/ (1+1.220)) X 100 RME = **80.45%**
3	**E-factor** [12]	
	E-Factor = Amount of waste/ Amount of product	Total amount of reactant = A+B = (1.0 + 1.220) g = 2.220 g Amount of final product = 1.786 g Amount of waste = (Total amount of reactant – amount of final product) = (2.220 – 1.786) g = 0.434 g E-factor = (0.434 /1.786) = **0.2430**
4	**Ecoscale** [12]	
	Ecoscale = (100 – Sum of individual penalties)	Eco-scale = 100–18.07 = **81.93**

Table S4. Calculation of the penalty points for the ecoscale calculation.

Calculation of penalty points		
Sr. No	Parameters	Penalty points
1	**Reaction Yield**	
	Yield = 89%	5.07
2	**Price of reaction components (to obtain 10mmol of final product)**	

	Anthranilamide	3
	4-Nitro Benzaldehyde	0
	p-TSA	0
3	**Safety**	
	Anthranilamide	5
	4-nitro Benzaldehyde	5
	p-TSA	0
4	**Technical setup**	
	Common setup	0
5	**Temperature/Time**	
	Room temperature	0
6	**Work and Purification**	
	Water wash and Simple filtration	0
7	**Total penalty points**	18.07



1	Reaction yield	90%
2	Atom economy (AE)	89.04
3	Reaction Mass Efficiency (RME)	80.45
4	E-factor	0.243
5	Eco-Scale score	81.93

Figure S2. Green chemistry metrices for the synthesis of 2-(4-nitrophenyl)-2,3-dihydroquinazolin-4(1H)-one (**3h**) from anthranilamide and *p*-nitrobenzaldehyde by grinding in mortar pestle.

Process Intensification: Mechanochemical scalable synthesis of 2-(4-nitrophenyl)-2,3-dihydroquinazolin-4(1H)-one (3h) by ball milling

Ball milling was carried out in tumbler ball milling instrument purchased from S.F. Engineering works having frequency 40 rpm, voltage 230 V and motor power 18.5 kW and current 24A. The internal diameter of milling vessel was 5 cm with 200ml capacity.

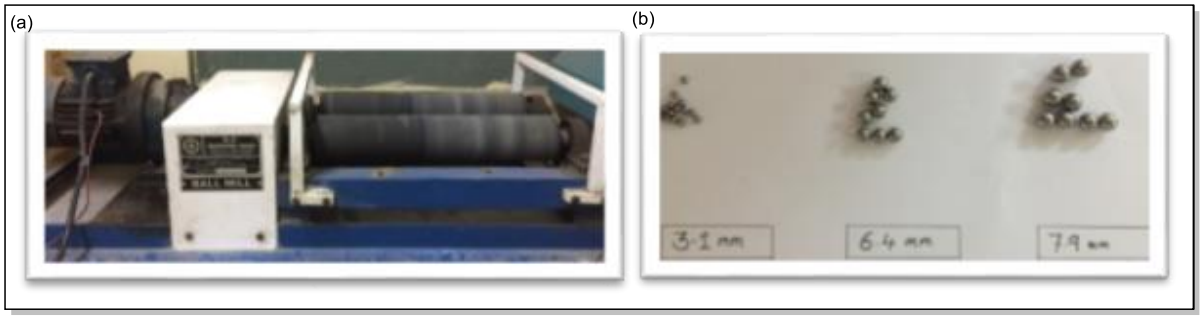


Figure S3. (a) Tumbler ball mill; (b) Milling balls of variable sizes.

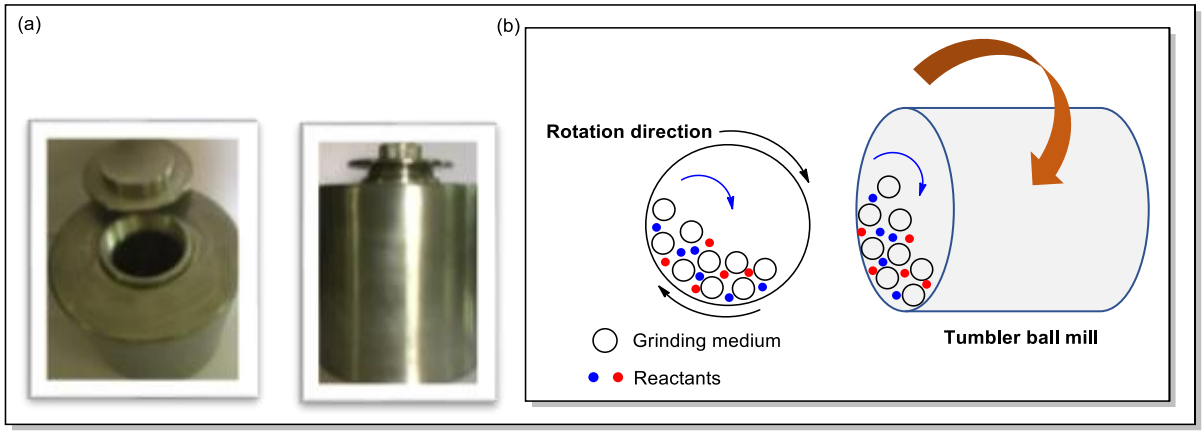
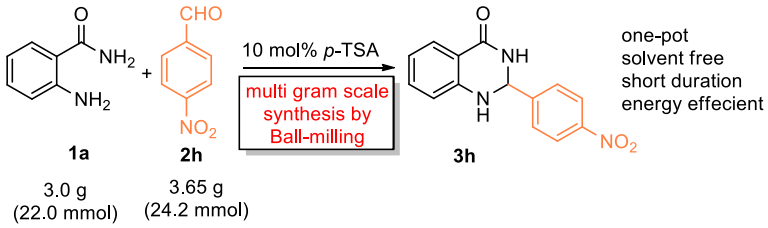


Figure S4. (a) Ball milling vessel; (b) Representation of mechanical ball milling operation in tumbler ball mill.

Scheme S4. Multigram synthesis of 2-(4-nitrophenyl)-2,3-dihydroquinazolin-4(1H)-one (**3h**) by mechanochemical ball milling.



Procedure: To a mixture of anthranilamide **1a** (0.1 g, 0.73 mmol, 1 equiv.) and *p*-nitrobenzaldehyde **2h** (0.122 g, 0.81 mmol, 1.1 equiv.) was added 10 mol% of *p*-TSA catalyst and the reaction mixture was milled in a ball mill consisting of stainless-steel balls. The progress of the reaction was montiored by TLC and HPLC analyses and on completion of the reaction, water was added, and the product was filtered off and dried to get hold of the desired product in excellent yield and purity.

Similar ball milling experiments were subsequently carried out with different ball diameters.

Table S5. Mechanochemical synthesis of 2-(4-nitrophenyl)-2,3-dihydroquinazolin-4(1H)-one (**3h**) by ball milling under several conditions.

Sl. No.	Time (mins)	Conversion of anthranilamide **1a** under different milling conditions[b]			
		Condition A[c,d]	Condition B[c,d]	Condition C[c,d]	Condition D[c,d]
1	10	60	30	70	55
2	20	65	50	75	60
3	30	70	55	80	70
4	40	75	65	85	75
5	50	78	70	87	80
6	60	83	75	90	85
7	120	86	79	93	90
8	240	90	83	98	93

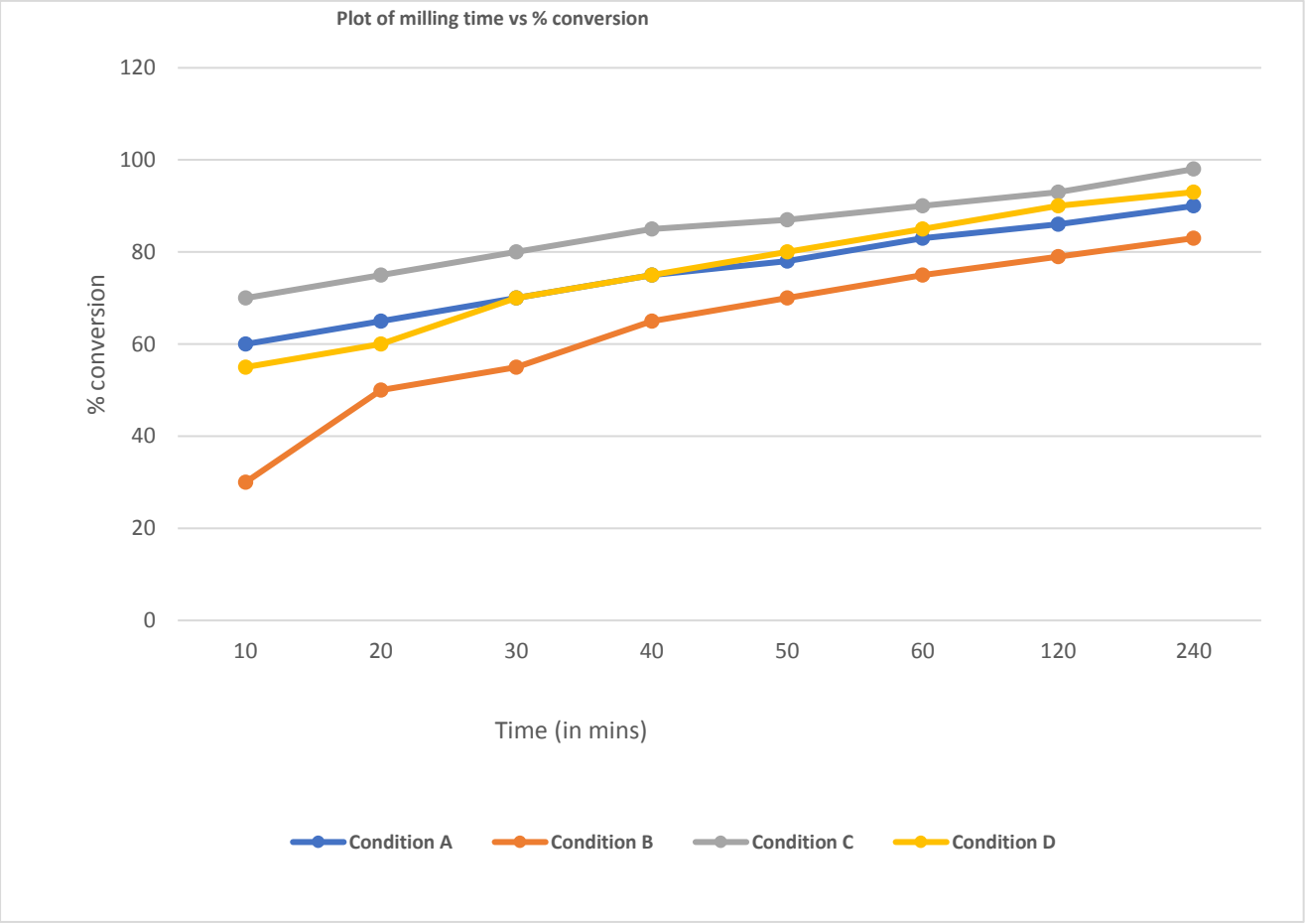


Figure S5. Graphical plot of the conversion of anthranilamide vs time interval (in mins) during the mechanochemical milling process.

^{1}H and ^{13}C NMR Spectral Reproductions Of Representative 2,3-Dihydroquinazolin-4(1H)-one (3a, 3c, 3d, 3h, 8b, 9b, 10a, 10b, 10c, 11b and 13b)

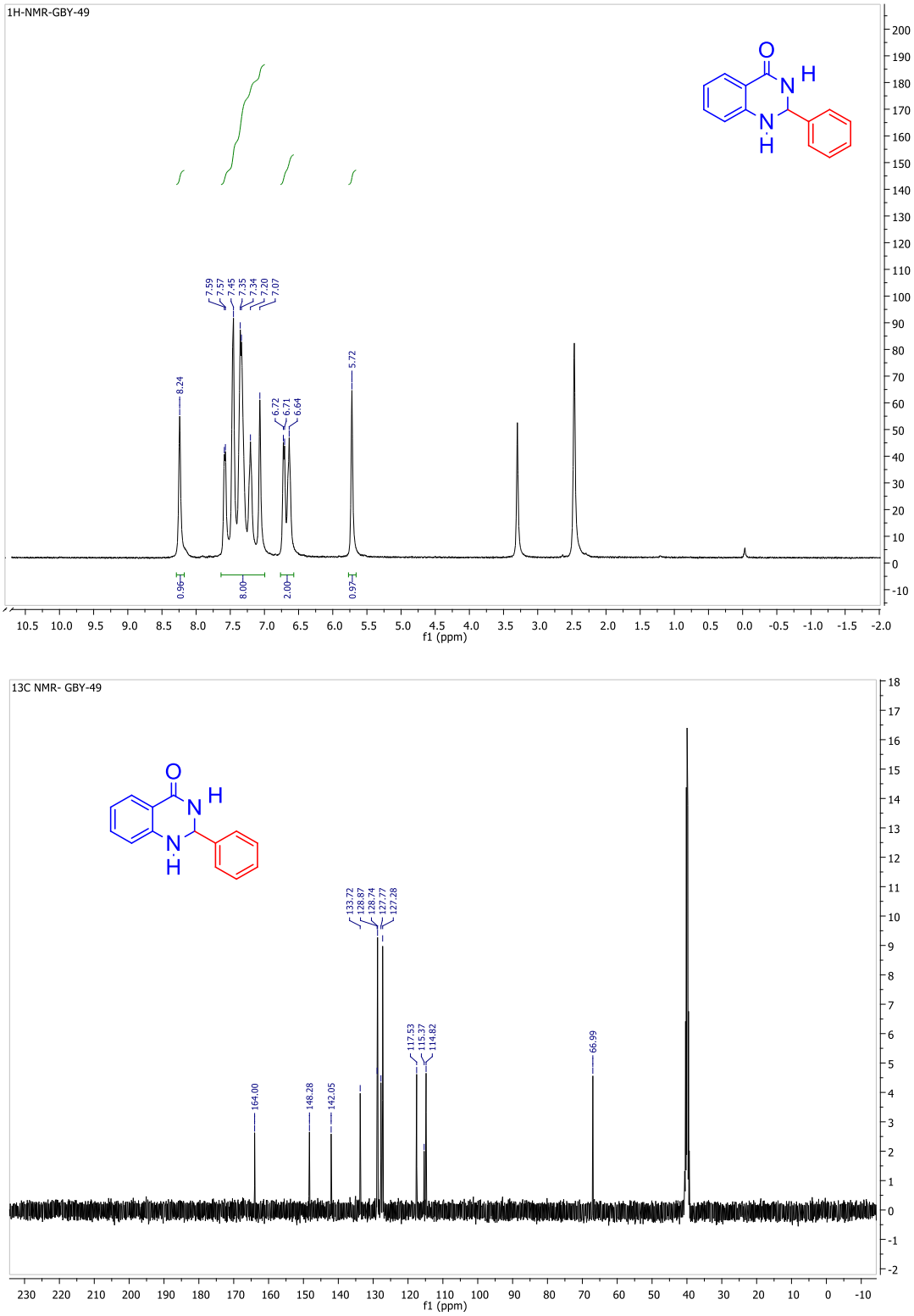
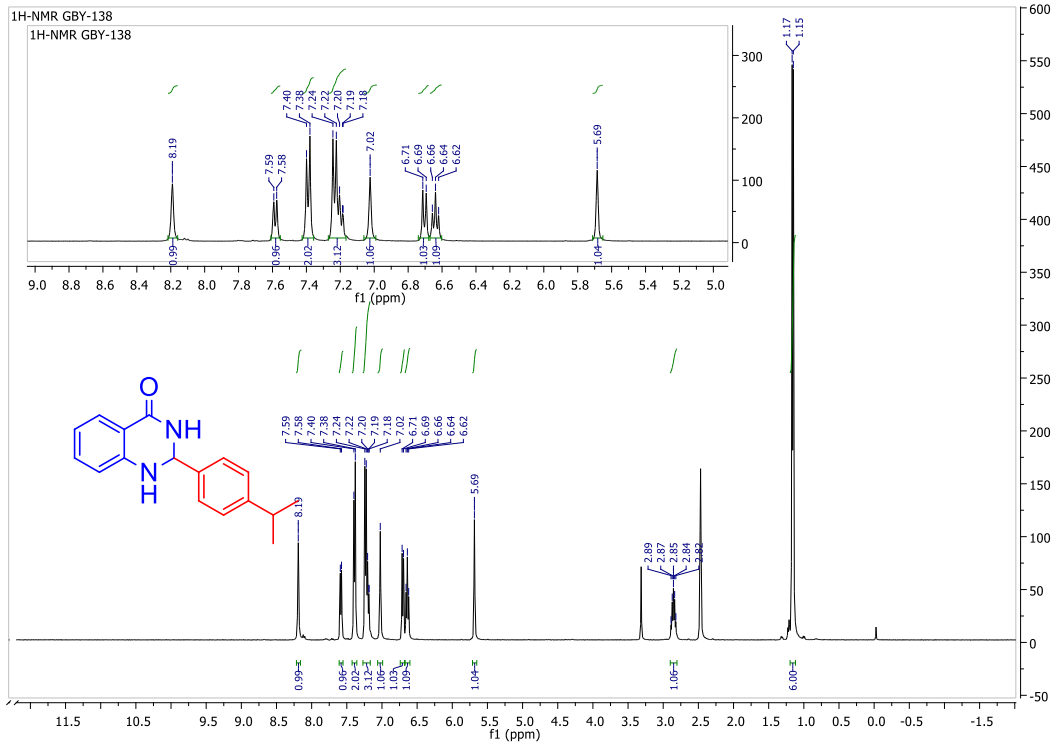


Figure S6. ^{1}H (400 MHz, DMSO-d_6) and ^{13}C NMR (100 MHz, DMSO-d_6) spectral reproduction of 2-phenyl-2,3-dihydroquinazolin-4(1H)-one (**3a**).



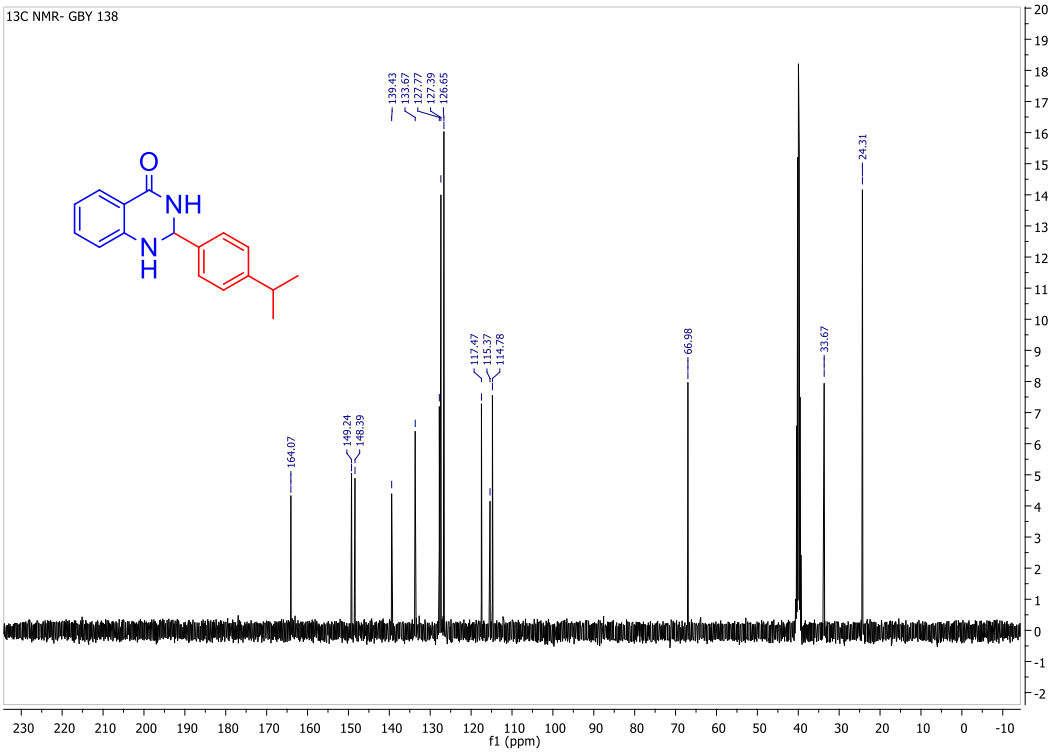


Figure S7. ^{1}H (400 MHz, DMSO-d_6) and ^{13}C NMR (100 MHz, DMSO-d_6) spectral reproduction of 2-(4-isopropylphenyl)-2,3-dihydroquinazolin-4(1H)-one (**3c**).

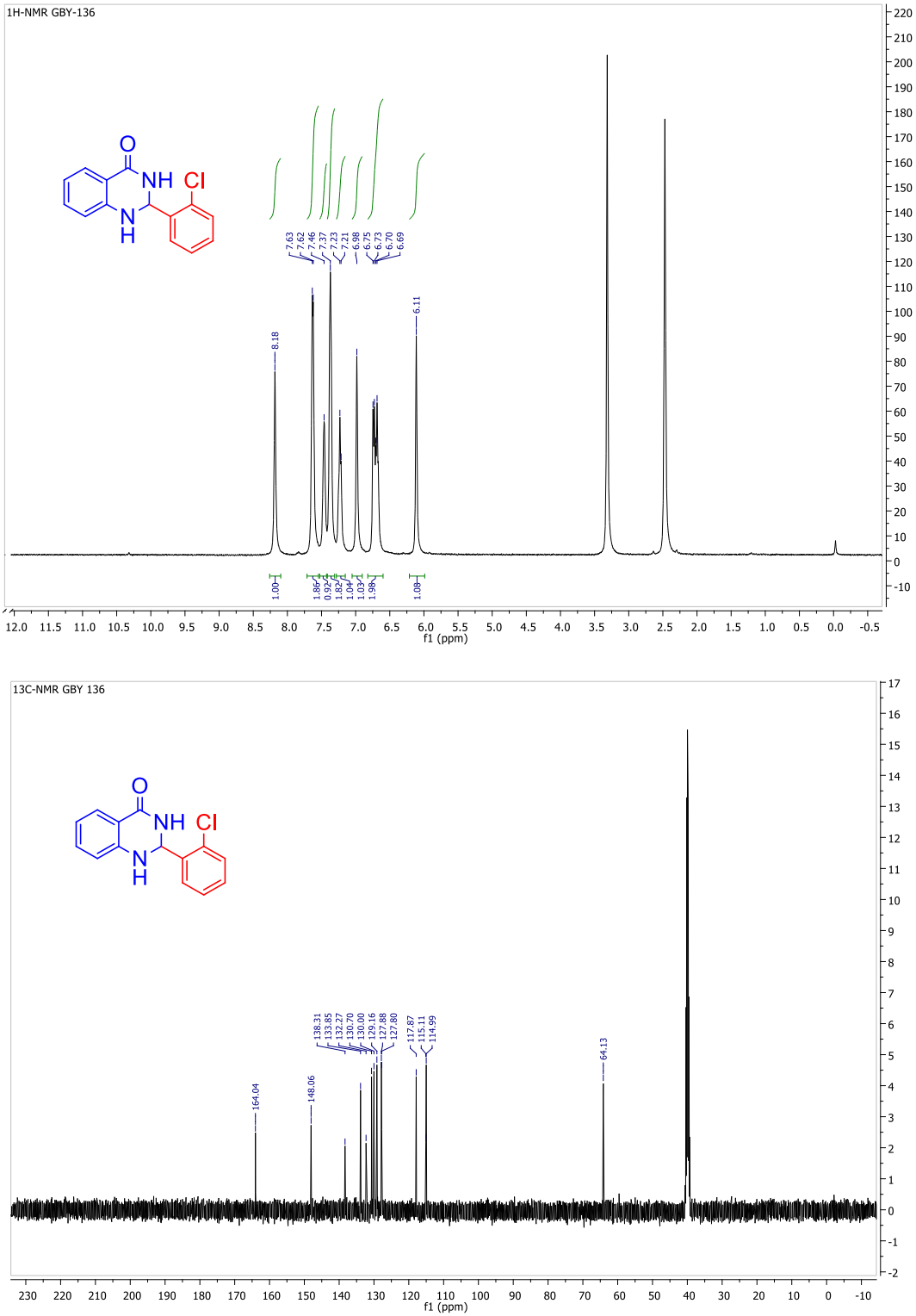


Figure S8. ^{1}H (400 MHz, DMSO-d_6) and ^{13}C NMR (100MHz, DMSO-d_6) spectral reproduction of 2-(2-chlorophenyl)-2,3-dihydroquinazolin-4(1H)-one (**3d**).

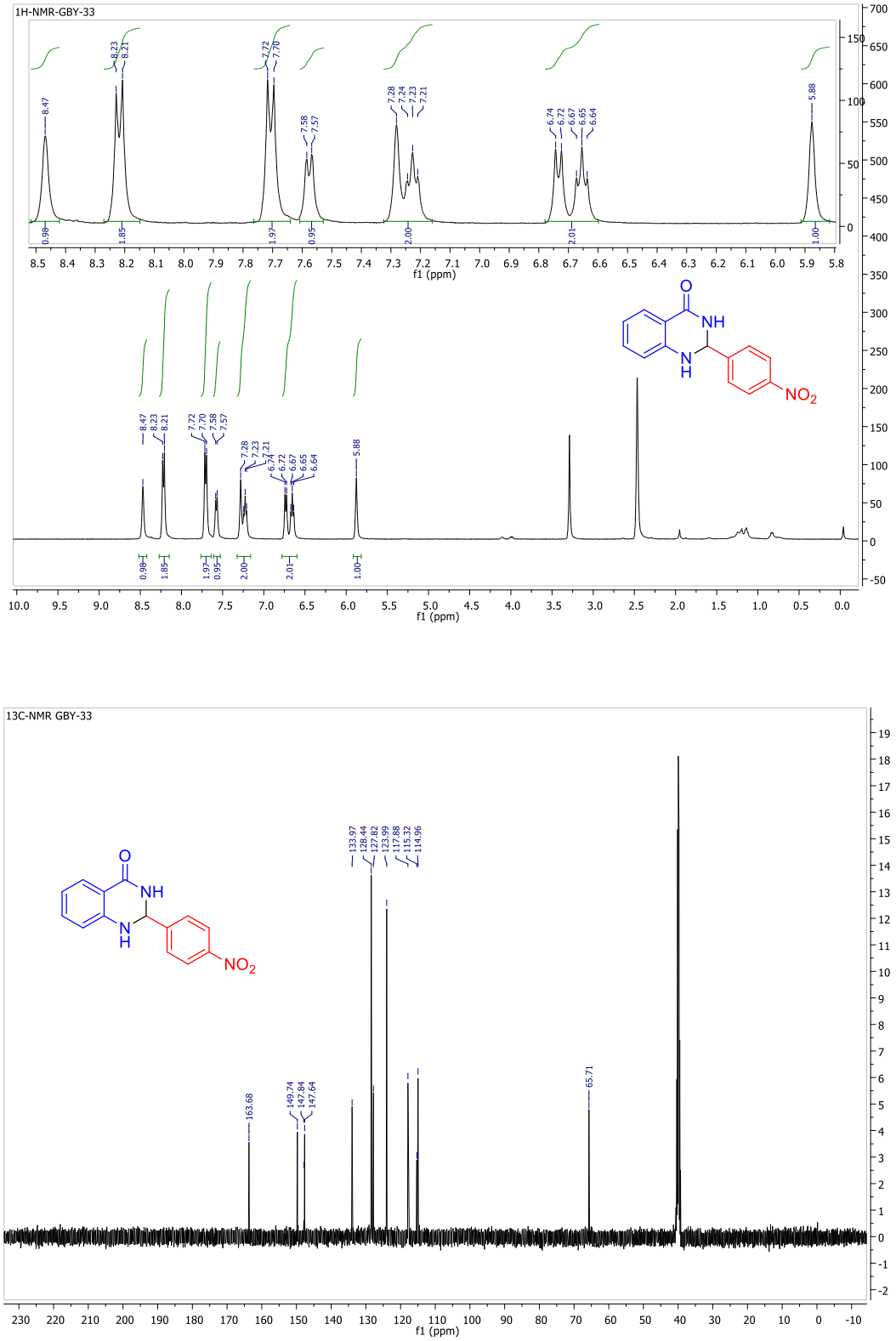


Figure S9. ^{1}H (400 MHz, DMSO-d_6) and ^{13}C NMR (100 MHz, DMSO-d_6) spectral reproduction of 2-(4-nitrophenyl)-2,3-dihydroquinazolin-4(1H)-one (**3h**).

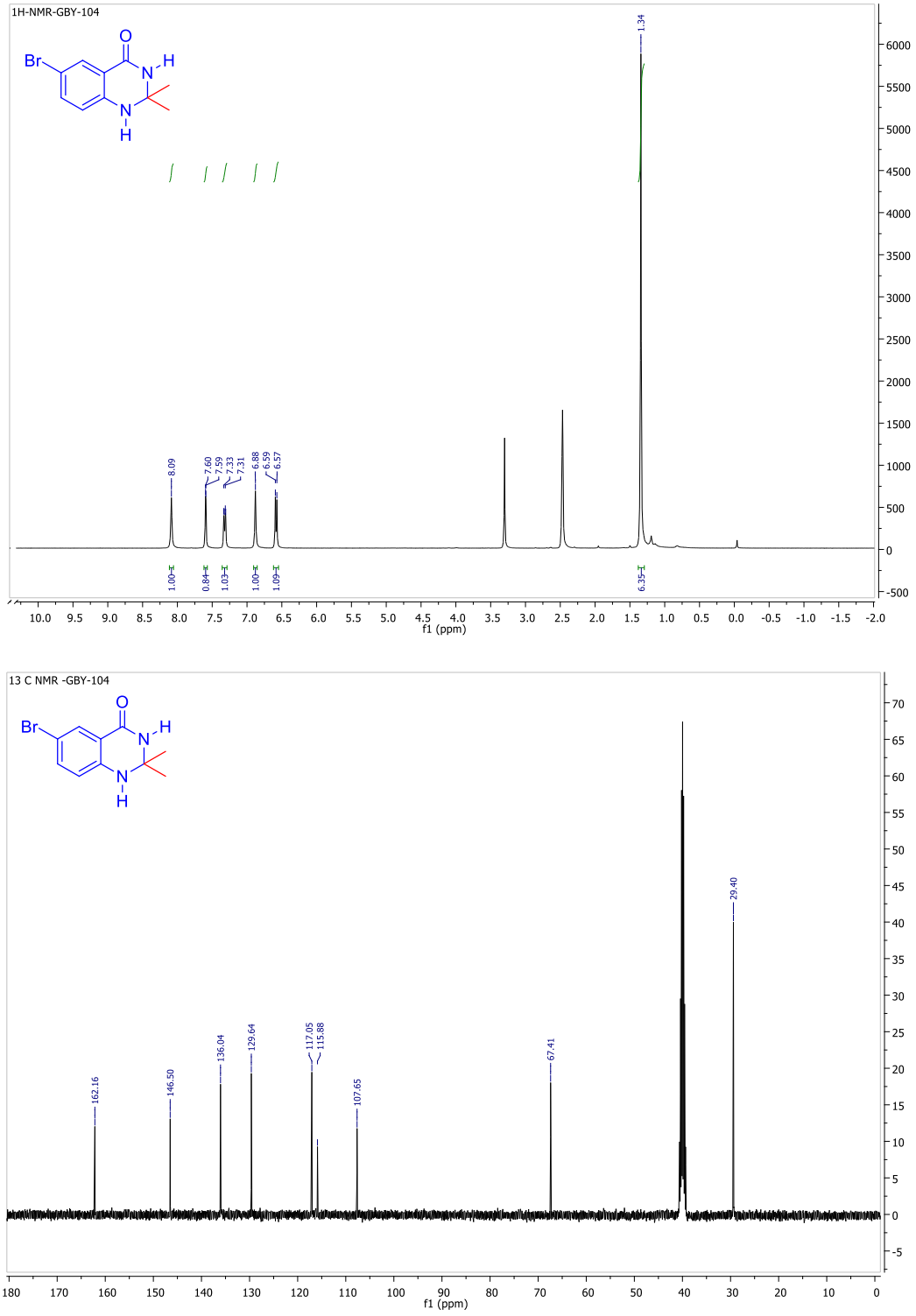


Figure S10. ^{1}H (400 MHz, DMSO-d_6) and ^{13}C NMR (100 MHz, DMSO-d_6) spectral reproduction of 6-bromo-2,2-dimethyl-2,3-dihydroquinazolin-4(1H)-one (**8b**).

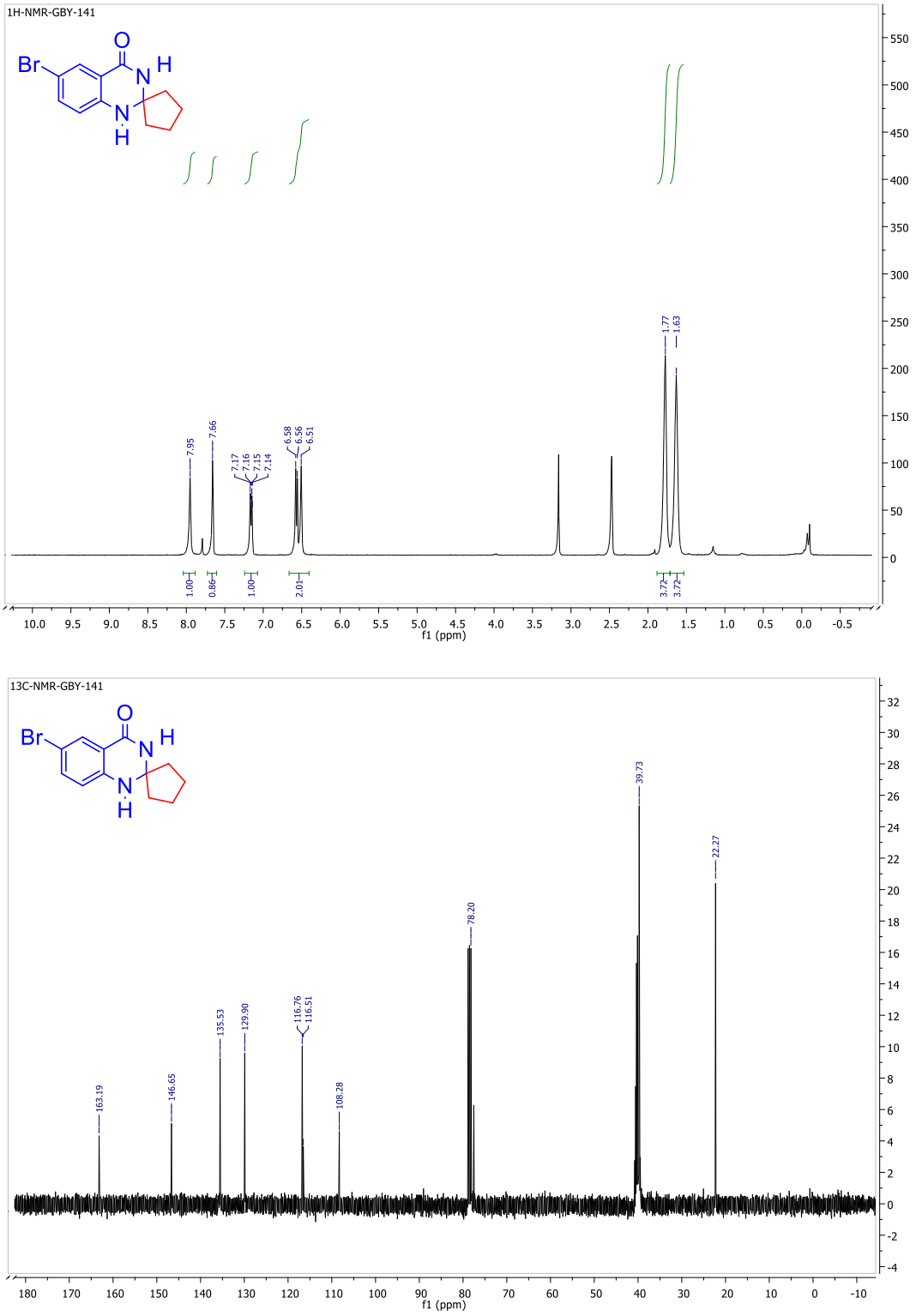


Figure S11. ^{1}H (400 MHz, DMSO-d_6) and ^{13}C NMR (100 MHz, DMSO-d_6) spectral reproduction of 6'-bromo-1'H-spiro[cyclopentane-1,2'-quinazolin]-4'(3'H)-one (**9b**).

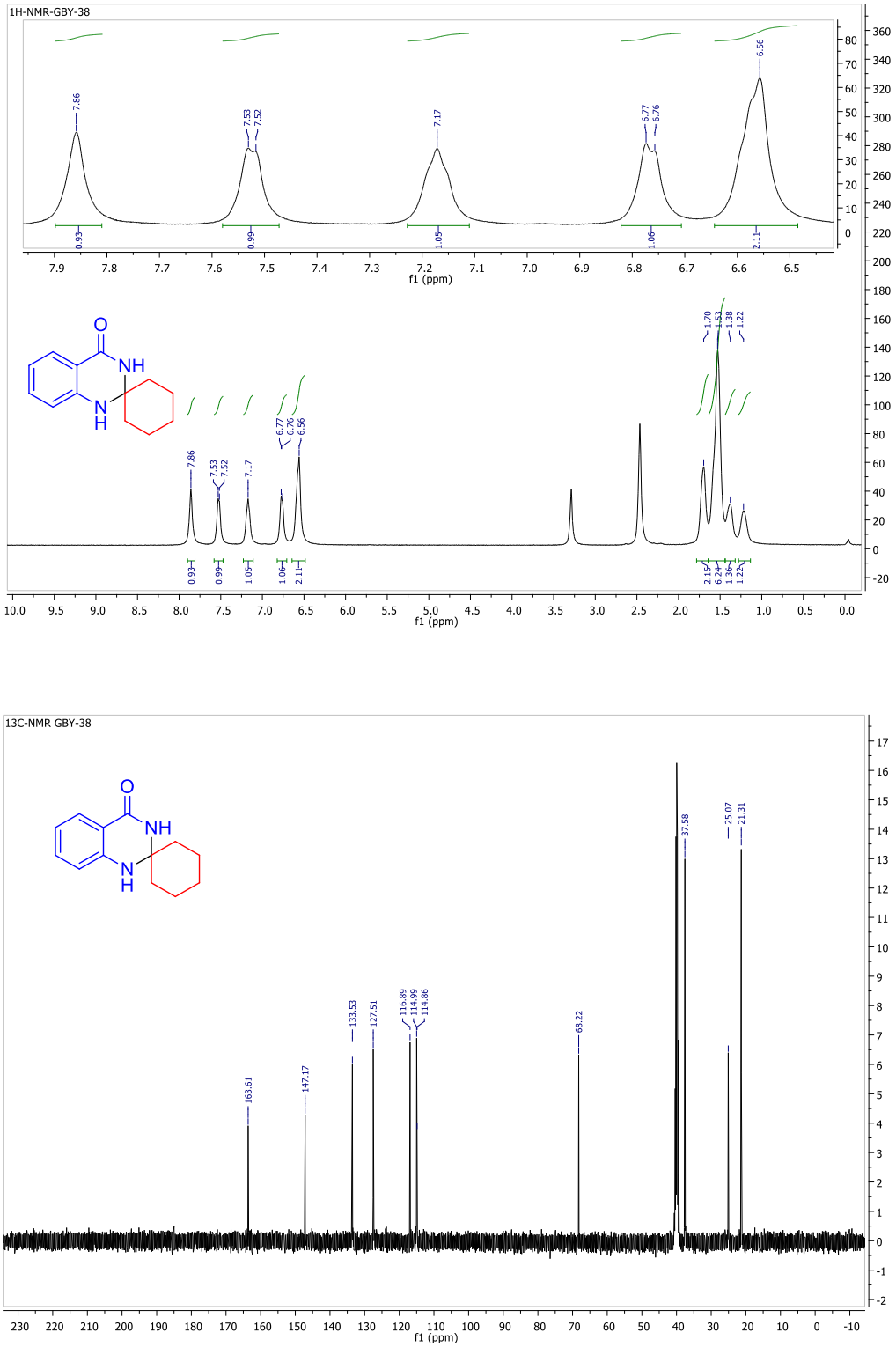


Figure S12. ^{1}H (400 MHz, DMSO-d_6) and ^{13}C NMR (100 MHz, DMSO-d_6) spectral reproduction of 1'H-spiro[cyclohexane-1,2'-quinazolin]-4'(3'H)-one (**10a**).

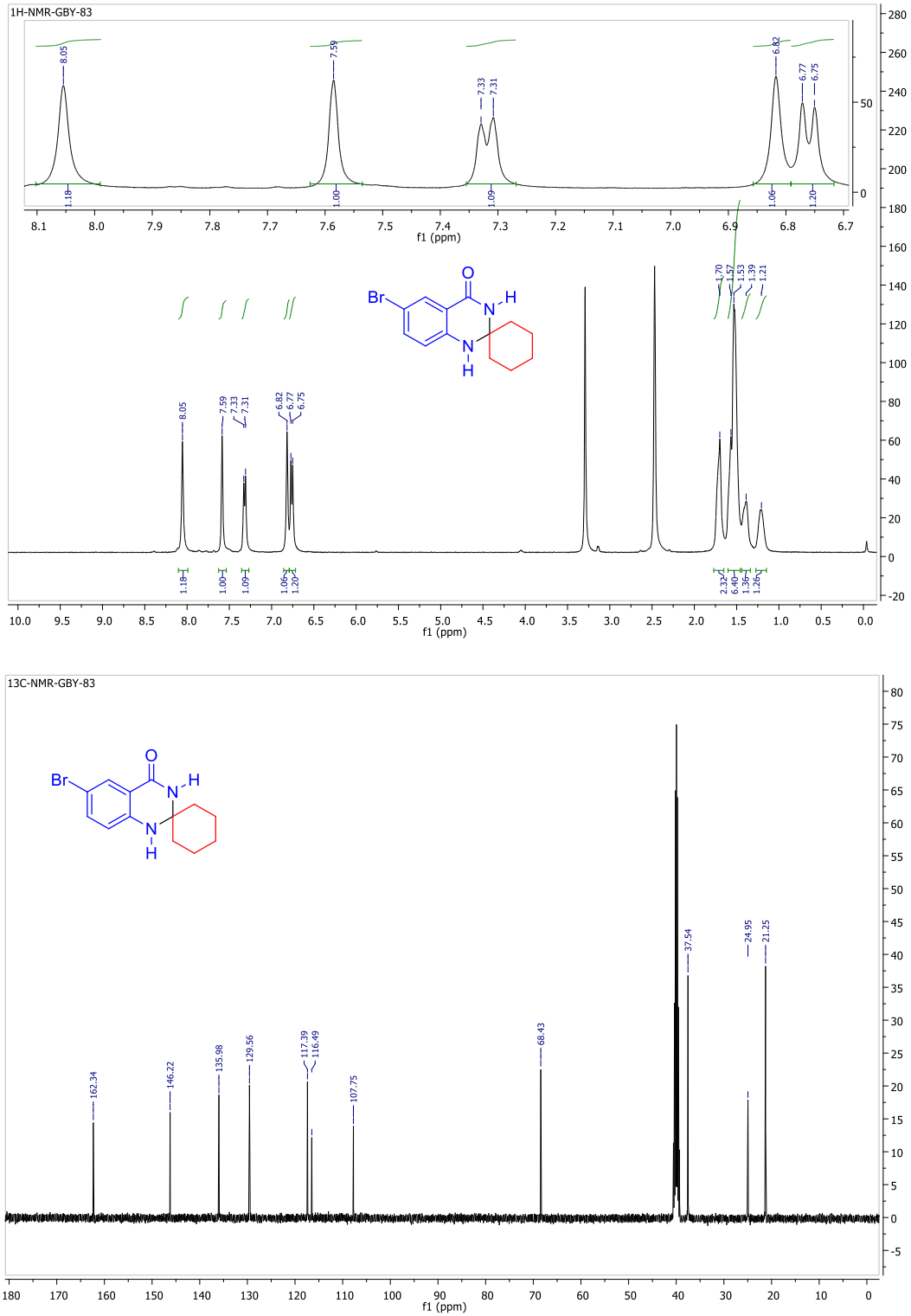


Figure S13. ^{1}H (400 MHz, DMSO-d_6) and ^{13}C NMR (100 MHz, DMSO-d_6) spectral reproduction of 6'-bromo-1'H-spiro[cyclohexane-1,2'-quinazolin]-4'(3'H)-one (**10b**).

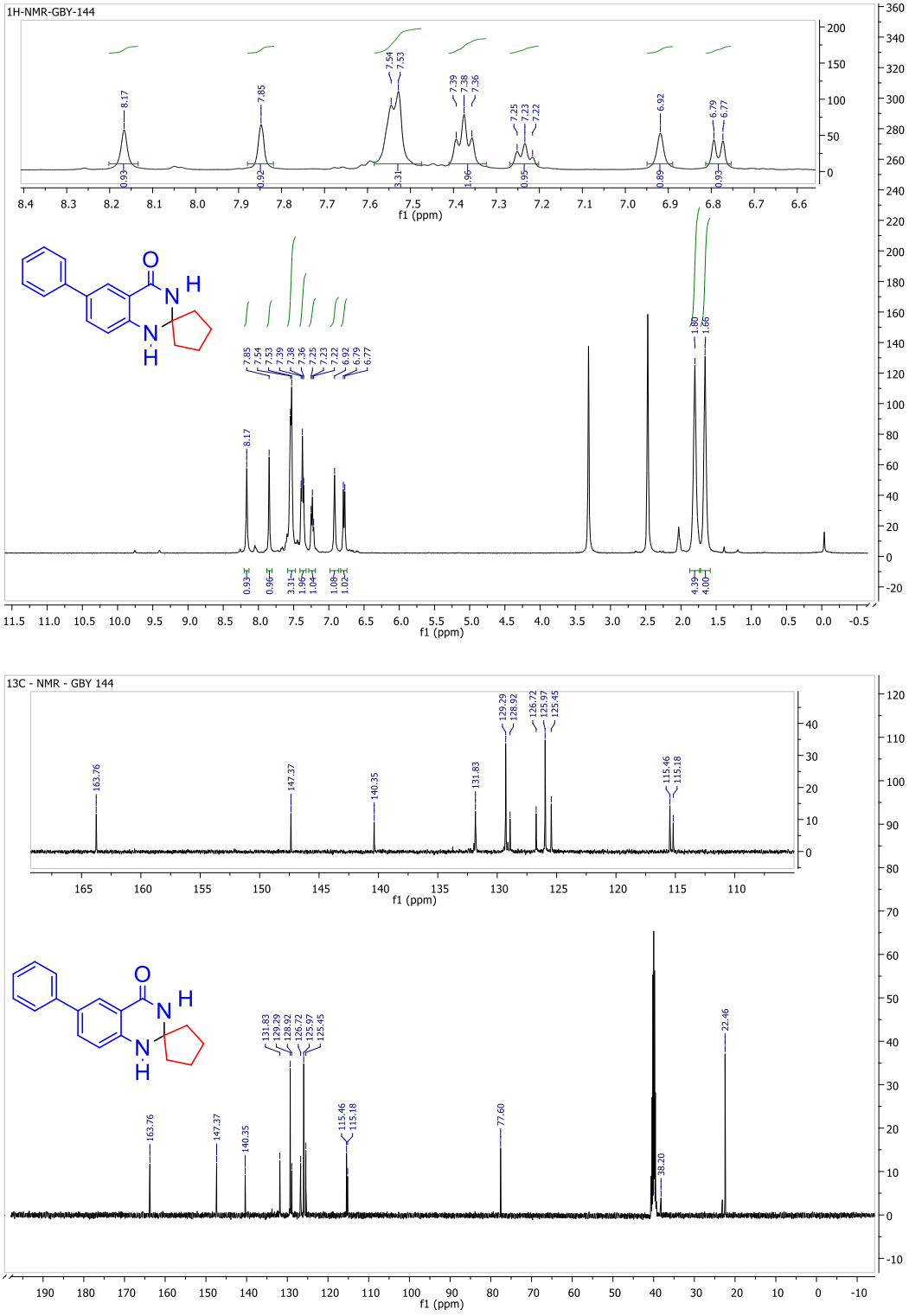
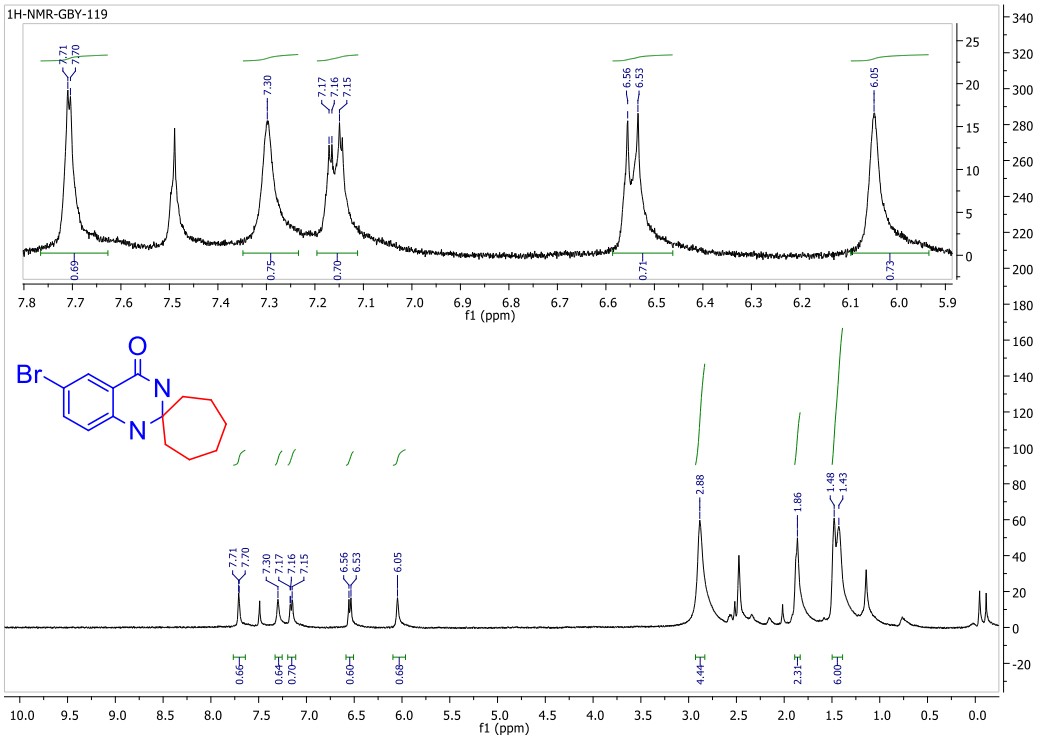


Figure S14. ^{1}H (400 MHz, DMSO-d_6) and ^{13}C NMR (100 MHz, DMSO-d_6) spectral reproduction of 6'-phenyl-1'H-spiro[cyclopentane-1,2'-quinazolin]-4'(3'H)-one (**10c**).



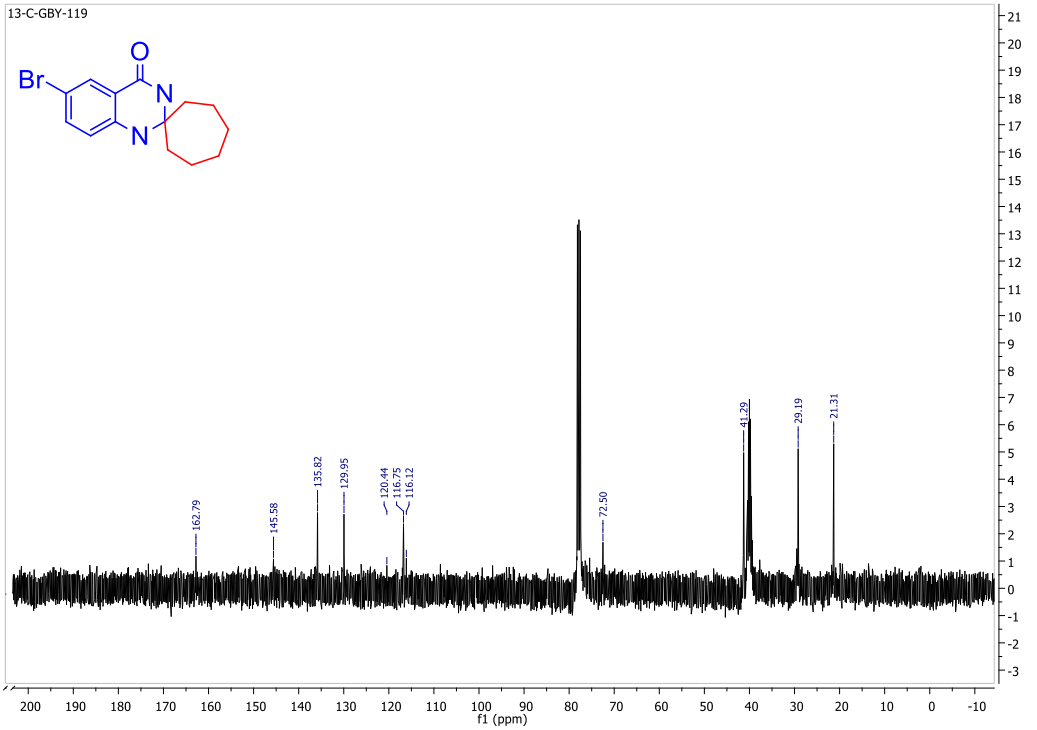


Figure S15. ^{1}H (400 MHz, DMSO-d_6) and ^{13}C NMR (100 MHz, DMSO-d_6) spectral reproduction of 6'-bromo-1'H-spiro[cycloheptane-1,2'-quinazolin]-4'(3'H)-one (**11b**).

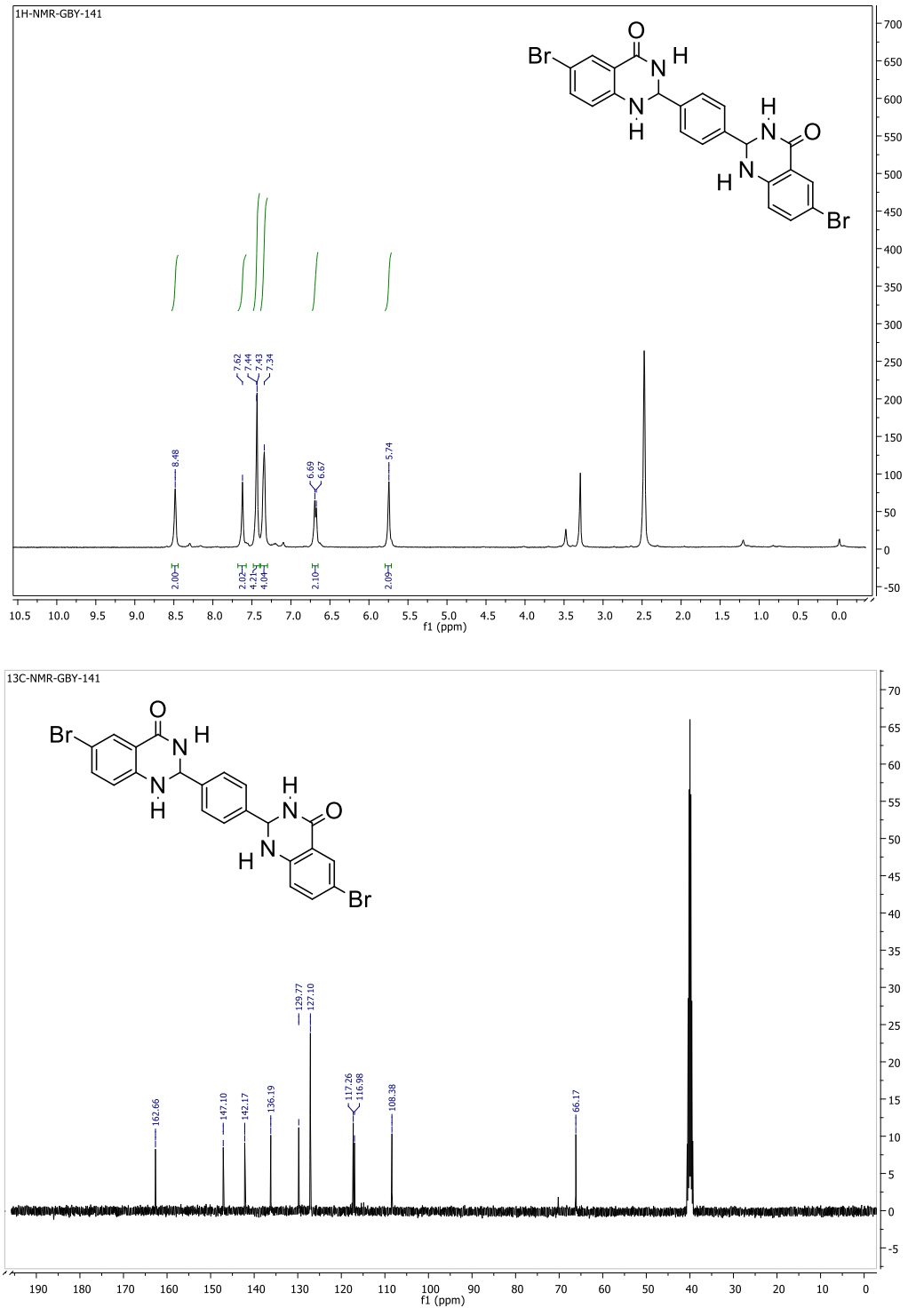


Figure S16. ^{1}H (400 MHz, DMSO-d_6) and ^{13}C NMR (100 MHz, DMSO-d_6) spectral reproduction of 2,2'-(1,4-phenylene)bis(6-bromo-2,3 dihydroquinazolin-4(1H)-one (**13b**).

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