

## Supporting Information

### Facile synthesis of triazolo/benzazolo[2,1-*b*]quinazolinones derivatives catalyzed by a new deep eutectic mixture based on glucose, pregabalin and urea

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#### **6,6-dimethyl-9-phenyl-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-*b*]quinazolin-8(4*H*)-one (1a):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3445, 2962, 1650, 1594, 1374, 1252, 721  $\text{cm}^{-1}$ .  **$^1\text{H}$  NMR** (400 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 0.95 (s, 3H,  $\text{CH}_3$ ), 1.03 (s, 3H,  $\text{CH}_3$ ), 2.05 (d,  $J=16$  Hz, 1H,  $\text{CH}_2$ ), 2.19 (d,  $J=16$  Hz, 1H,  $\text{CH}_2$ ), 2.52-2.59 (m, 2H,  $\text{CH}_2$ ), 6.19 (s, 1H, CH), 7.17-7.29 (m, 5H-Ar), 7.68 (s, 1H-Ar), 11.14 (s, 1H, NH);  **$^{13}\text{C}$  NMR** (100 MHz,  $\text{DMSO-d}_6$ ):  $\delta$  26.77, 28.45, 32.16, 49.74, 57.89, 105.55, 126.92, 127.69, 128.23, 141.55, 146.82, 150.24, 150.39, 192.96 ppm.

#### **9-(4-chlorophenyl)-6,6-dimethyl-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-*b*]quinazolin-8(4*H*)-one (2a):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3436, 3094, 2961, 1649, 1578, 1367, 1254, 760  $\text{cm}^{-1}$ ;  **$^1\text{H}$  NMR** (400 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 0.71 (s, 3H,  $\text{CH}_3$ ), 0.79 (s, 3H,  $\text{CH}_3$ ), 1.81 (d,  $J=16$  Hz, 1H,  $\text{CH}_2$ ), 1.95 (d,  $J=16$  Hz, 1H,  $\text{CH}_2$ ), 2.25-2.34 (m, 2H,  $\text{CH}_2$ ), 5.97 (s, 1H, CH), 6.95 (d,  $J=8$  Hz, 2H-Ar), 7.09 (d,  $J=8$  Hz, 2H-Ar), 7.45 (s, 1H-Ar), 10.95 (s, 1H, NH);  **$^{13}\text{C}$  NMR** (100 MHz,  $\text{DMSO-d}_6$ ):  $\delta$  26.87, 28.36, 32.17, 49.71, 57.34, 105.16, 128.25, 128.86, 132.26, 140.49, 146.76, 150.17, 150.55, 192.98 ppm.

#### **9-(2-Chlorophenyl)-6,6-dimethyl-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-*b*]quinazolin-8(4*H*)-one (3a):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3224, 3085, 2923, 1643, 1573, 1365, 1257, 748  $\text{cm}^{-1}$ ;  **$^1\text{H}$  NMR** (250 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 0.97(s, 3H,  $\text{CH}_3$ ), 1.02 (s, 3H,  $\text{CH}_3$ ), 2.04-2.05 (m, 1H,  $\text{CH}_2$ ), 2.19 (d, 1H,  $J=17.5$  Hz,  $\text{CH}_2$ ), 2.51-2.60 (m, 2H,  $\text{CH}_2$ ), 6.55 (s, 1H, CH), 7.21-7.36 (m, 4H, ArH), 7.65 (s, 1H, =CH), 11.20 (s, 1H, NH);  **$^{13}\text{C}$  NMR** (62.9 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 25.66, 27.38, 30.95, 48.62, 55.23, 103.38, 116.89, 125.99, 128.26, 128.43, 137.05, 145.72, 148.91, 149.90, 191.71 ppm.

#### **9-(2,4-Dichlorophenyl)-6,6-dimethyl-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-*b*]quinazolin-8(4*H*)-one (4a):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3090, 2941, 1643, 1610, 1438, 1261, 839  $\text{cm}^{-1}$ ;  **$^1\text{H}$  NMR** (300 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 0.99 (s, 3H,  $\text{CH}_3$ ), 1.09 (s, 3H,  $\text{CH}_3$ ), 2.08 (q,  $J=14.17, 23.03$  Hz, 2H,  $\text{CH}_2$ ), 2.49 (s, 2H,  $\text{CH}_2$ ), 6.48 (s, 1H, CH), 7.22-7.31 (m, 2H, Ar-H), 7.45 (s, 1H, Ar-H), 7.59 (s, 1H, Ar-H), 10.92 (s, 1H, NH);  **$^{13}\text{C}$  NMR** (75 MHz,  $\text{CDCl}_3$ )  $\delta$ : 13.11, 15.71, 59.48, 60.52, 110.18, 123.35, 125.12, 129.47, 141.86, 161.39 ppm.

#### **9-(4-bromophenyl)-6,6-dimethyl-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-*b*]quinazolin-8(4*H*)-one (5a):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3436, 3090, 2965, 1650, 1584, 1367, 1255, 759  $\text{cm}^{-1}$ ;  **$^1\text{H}$  NMR** (400 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 0.95 (s, 3H,  $\text{CH}_3$ ), 1.03 (s, 3H,  $\text{CH}_3$ ), 2.05 (d,  $J = 16$  Hz, 1H,  $\text{CH}_2$ ), 2.19 (d,  $J = 16$  Hz, 1H,  $\text{CH}_2$ ), 2.54-2.58 (m, 2H,  $\text{CH}_2$ ), 6.19 (s, 1H, CH), 7.13 (d,  $J = 8$  Hz, 2H-Ar), 7.47 (d,  $J = 8$  Hz, 2H-Ar), 7.70 (s, 1H-Ar), 11.19 (s, 1H, NH);  **$^{13}\text{C}$  NMR** (100 MHz,  $\text{DMSO-d}_6$ ):  $\delta$  26.87, 28.36, 32.18, 49.71, 57.43, 105.12, 120.85, 129.22, 131.18, 140.91, 146.75, 150.18, 150.56, 192.99 ppm.

**6,6-dimethyl-9-(4-nitrophenyl)-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-*b*]quinazolin-8(4*H*)-one (6a):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3412, 3090, 2964, 1646, 1579, 1351, 1253, 729  $\text{cm}^{-1}$ ;  **$^1\text{H}$  NMR** (400 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 0.95 (s, 3H,  $\text{CH}_3$ ), 1.03 (s, 3H,  $\text{CH}_3$ ), 2.05(d,  $J = 16$ Hz, 1H,  $\text{CH}_2$ ), 2.19 (d,  $J = 16$ Hz, 1H,  $\text{CH}_2$ ), 2.49 (d,  $J = 16$ Hz, 1H,  $\text{CH}_2$ ), 2.56 (d,  $J = 16$ Hz, 1H,  $\text{CH}_2$ ), 6.36 (s, 1H, CH), 7.47 (d,  $J = 8$ Hz, 2H-Ar), 7.73 (s, 1H-Ar), 8.14 (d,  $J = 8$ Hz, 2H-Ar), 11.31 (s, 1H, NH);  **$^{13}\text{C}$  NMR** (100 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 26.92, 28.26, 32.21, 49.63, 57.49, 104.72, 123.55, 128.46, 146.81, 146.89, 148.38, 150.40, 150.97, 193.01 ppm.

**6,6-dimethyl-9-(3-nitrophenyl)-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-*b*]quinazolin-8(4*H*)-one (7a):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3436, 3082, 2960, 1645, 1580, 1367, 1256, 732  $\text{cm}^{-1}$ .  **$^1\text{H}$  NMR** (400 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 0.756 (s, 3H,  $\text{CH}_3$ ), 0.829 (s, 3H,  $\text{CH}_3$ ), 1.86 (d,  $J = 16$  Hz, 1H,  $\text{CH}_2$ ), 1.99 (d,  $J = 16$  Hz, 1H,  $\text{CH}_2$ ), 2.28-2.40 (m, 2H,  $\text{CH}_2$ ), 6.20 (s, 1H, CH), 7.37-7.44 (m, 2H-Ar), 7.52 (s, 1H-Ar), 7.84- 7.90 (m, 2H-Ar), 11.09 (s,1H, NH);  **$^{13}\text{C}$  NMR** (100 MHz,  $\text{DMSO-d}_6$ ):  $\delta$  26.85, 28.31, 32.23, 49.65, 57.39, 104.59, 121.66, 122.81, 130.04, 133.71, 143.45, 146.77, 147.58, 150.42, 151.13, 193.09 ppm.

**9-(4-methoxyphenyl)-6,6-dimethyl-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-*b*]quinazolin-8(4*H*)-one (8a):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3436, 2961, 1649, 1580, 1367, 1246, 745  $\text{cm}^{-1}$ .  **$^1\text{H}$  NMR** (400 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 0.75 (s, 3H,  $\text{CH}_3$ ), 0.82 (s, 3H,  $\text{CH}_3$ ), 1.83 (d,  $J = 16$  Hz, 1H,  $\text{CH}_2$ ), 1.98 (d,  $J = 16$  Hz, 1H,  $\text{CH}_2$ ), 2.28-2.37 (m, 2H,  $\text{CH}_2$ ), 3.47 (s, 3H,  $\text{OCH}_3$ ), 5.93 (s, 1H, CH), 6.60 (d,  $J = 8$  Hz, 2H-Ar), 6.87 (d,  $J = 8$  Hz, 2H-Ar), 7.45 (s, 1H-Ar), 10.87 (s,1H, NH);  **$^{13}\text{C}$  NMR** (100 MHz,  $\text{DMSO-d}_6$ ):  $\delta$  26.79, 28.49, 32.15, 49.78, 54.99, 57.30, 105.71, 113.54, 128.09, 133.82, 146.72, 150.14, 158.66, 192.94 ppm.

**9-(4-hydroxyphenyl)-6,6-dimethyl-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-*b*]quinazolin- 8(4*H*)-one (9a):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3348, 3096, 2932, 1631, 1587, 1370, 1269, 731  $\text{cm}^{-1}$ ;  **$^1\text{H}$  NMR** (400 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 0.96 (s, 3H,  $\text{CH}_3$ ), 1.03 (s, 3H,  $\text{CH}_3$ ), 2.05 (d,  $J = 16$  Hz, 1H,  $\text{CH}_2$ ), 2.18 (d,  $J = 16$  Hz, 1H, CH,  $\text{CH}_2$ ), 2.57-2.68 (m, 2H,  $\text{CH}_2$ ), 6.08 (s, 1H, CH), 6.62 (d,  $J = 8$  Hz, 2H-Ar), 6.96 (d,  $J = 8$  Hz, 2H-Ar), 7.65 (s, 1H-Ar), 9.39 (s, 1H, OH or NH), 11.04 (s, 1H, OH or NH);  **$^{13}\text{C}$  NMR** (100 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 26.79, 28.51, 32.14, 49.80, 52.99, 57.33, 105.86, 114.84, 132.21, 128.07, 146.69, 149.83, 150.03, 156.87, 192.94 ppm.

**9-(4-hydroxy-3-methoxyphenyl)-6,6-dimethyl-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-*b*]quinazolin-8(4*H*)-one (10a):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3081, 2968, 1655, 1589, 1375, 1242, 764  $\text{cm}^{-1}$ ;  **$^1\text{H}$ -NMR** (400 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 1.01 (s, 3H, $\text{CH}_3$ ), 1.08 (s, 3H,  $\text{CH}_3$ ), 2.11 (q,  $J = 13.62, 22.79$  Hz, 2H,  $\text{CH}_2$ ), 2.43 (s, 2H,  $\text{CH}_2$ ) 3.70 (s, 3H,  $\text{OCH}_3$ ), 6.71 (s, 1H, CH), 7.11-7.20 (m, 2H, Ar-H), 7.37 (s, 1H, Ar-H), 7.49 (s, 1H, Ar-H), 11.21 (s, 1H, NH);  **$^{13}\text{C}$  NMR** (75

MHz, CDCl<sub>3</sub>)  $\delta$ : 25.37, 27.56, 30.72, 48.10, 49.33, 101.17, 122.48, 126.51, 131.77, 133.12, 146.57, 149.44, 155.83, 157.46, 189.74 ppm.

**6,6-dimethyl-9-(p-tolyl)-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-b]quinazolin-8(4H)-one (11a):**

**FT-IR** (KBr, cm<sup>-1</sup>): 3380–3022, 2957, 2922, 1711, 1610, 1461 cm<sup>-1</sup>; **<sup>1</sup>H NMR** (400 MHz, DMSO-d<sub>6</sub>)  $\delta$ : 0.98 (s, 3H, CH<sub>3</sub>), 1.11 (s, 3H, CH<sub>3</sub>), 2.17 (d,  $J$  = 5.7 Hz, 1H, CH<sub>2</sub>), 2.24 (d,  $J$  = 5.7 Hz, 1H, CH<sub>2</sub>), 2.38 (s, 3H, CH<sub>3</sub>), 2.62–2.70 (m, 2H, CH<sub>2</sub>), 6.09 (s, 1H, CH), 7.03 (s, 2H-Ar), 7.03 (s, 2H-Ar), 7.70 (s, 1H-Ar), 11.05 (s, 1H, NH); **<sup>13</sup>C NMR** (100 MHz, DMSO-d<sub>6</sub>)  $\delta$ : 22.3, 24.8, 29.2, 48.5, 51.6, 57.2, 108.1, 126.0, 128.6, 129.9, 143.6, 144.1, 149.0, 154.5, 192.9 ppm.

**6,6-Dimethyl-9-(o-tolyl)-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-b]quinazolin-8(4H)-one (12a):**

**FT-IR** (KBr, cm<sup>-1</sup>): 3085, 2925, 1643, 1581, 1365, 1257, 748 cm<sup>-1</sup>; **<sup>1</sup>H NMR** (250 MHz, DMSO-d<sub>6</sub>)  $\delta$ : 0.97(s, 3H, CH<sub>3</sub>), 1.03 (s, 3H, CH<sub>3</sub>), 2.12 (q, 2H,  $J$  = 17.5 Hz, CH<sub>2</sub>), 2.48–2.60 (m, 5H, CH<sub>3</sub> and CH<sub>2</sub>), 6.42 (s, 1H, CH), 6.95–7.09 (m, 4H-Ar), 7.63 (s, 1H-Ar), 11.09 (s, 1H, NH); **<sup>13</sup>C NMR** (62.9 MHz, DMSO-d<sub>6</sub>)  $\delta$ : 18.89, 26.83, 28.48, 32.18, 49.73, 54.12, 106.14, 126.18, 126.64, 127.43, 129.96, 135.56, 140.32, 146.55, 149.87, 150.42, 193.00 ppm.

**9-(phenyl)-5,6,7,9-tetrahydro-1,2,4-triazolo[5,1-b]quinazolin-8(4H)-one (13a):**

**FT-IR** (KBr, cm<sup>-1</sup>): 3344, 2990, 1715, 1638, 1584, 1549, 1412, 1365 cm<sup>-1</sup>; **<sup>1</sup>H NMR** (300 MHz, DMSO-d<sub>6</sub>)  $\delta$ : 1.85-2.66 (m, 6H, 3CH<sub>2</sub>), 6.22 (s, 1H, CH), 6.91-7.30 (m, 5H-Ar), 7.69 (s, 1H, CH), 11.19 (br. s, 1H, NH); **<sup>13</sup>C NMR** (75 MHz, DMSO-d<sub>6</sub>)  $\delta$ : 21.12, 26.85, 36.77, 58.18, 107.05, 127.45, 150.82, 128.15, 128.72, 128.87, 141.97, 147.22, 150.51, 153.03, 193.76 ppm.

**9-(4-chlorophenyl)-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-b]quinazolin-8(4H)-one (14a):**

**<sup>1</sup>H NMR** (400 MHz, DMSO-d<sub>6</sub>)  $\delta$ : 1.88-1.99 (m, 2H CH<sub>2</sub>), 2.20- 2.34 (m, 2H, CH<sub>2</sub>), 2.64-2.71 (m, 2H, CH<sub>2</sub>), 6.24 (s, 1H, CH), 7.24 (d,  $J$  = 8.4 Hz, 2H, Ar-H), 7.35 (d,  $J$  = 8.8 Hz, 2H, Ar-H), 7.71 (s, 1H, Ar-H), 11.20 (s, NH, 1H); **<sup>13</sup>C NMR** (100 MHz, DMSO-d<sub>6</sub>)  $\delta$ : 20.6, 26.3, 36.2, 57.17, 106.1, 2 x 128.2, 2 x 128.9, 132.2, 140.4, 146.6, 150.1, 152.7, 193.2 ppm.

**9-(3,4,5-Trimethoxyphenyl)-5,6,7,9-tetrahydro-1,2,4-triazolo[5,1-b]quinazolin-8(4H)-one (15a):**

**FT-IR** (KBr, cm<sup>-1</sup>): 3433, 2956, 2924, 2834, 1645, 1595, 1507, 1460, 1416, 1328, 1239, 1128, 1009, 888, 832, 580 cm<sup>-1</sup>; **<sup>1</sup>H NMR** (400 MHz, DMSO-d<sub>6</sub>)  $\delta$ : 1.90–2.03 (m, 2H, CH), 2.28–2.31 (m, 2H, CH), 2.60–2.57 (m, 2H, CH), 3.61 (s, 3H, OCH<sub>3</sub>), 3.71 (s, 6H, OCH<sub>3</sub>), 6.81 (s, 1H, CH), 6.48 (s, 2H, ArH), 7.70 (s, 1H, ArH), 11.13 (s, 1H, NH) ppm; **<sup>13</sup>C NMR** (100 MHz, DMSO-d<sub>6</sub>)  $\delta$ : 20.7, 26.4, 36.3, 55.8, 57.6, 59.8, 104.3, 106.1, 136.9, 146.6, 149.9, 152.7, 193.4ppm.

**9-(4-bromophenyl)-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-b]quinazolin-8(4H)-one (16a):**

**FT-IR** (KBr, cm<sup>-1</sup>): 3429, 3129, 2883, 1648, 1578, 621 cm<sup>-1</sup>; **<sup>1</sup>H NMR** (400 MHz, DMSO-d<sub>6</sub>)  $\delta$ : 1.89–2.01 (m, 2H, CH<sub>2</sub>), 2.22–2.34 (m, 2H, CH<sub>2</sub>), 2.61–2.71 (m, 2H, CH<sub>2</sub>), 6.23 (s, 1H, CH), 7.18 (d,  $J$  = 8.4 Hz, 2H-Ar), 7.49

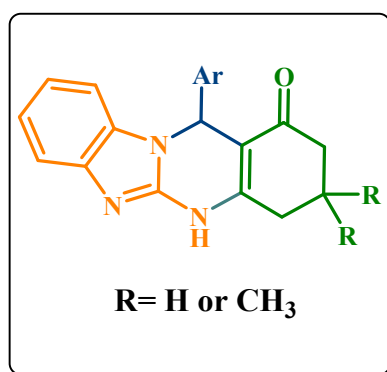
(d,  $J = 8.4$  Hz, 2H-Ar), 7.71 (s, 1H-Ar), 11.21 (s, 1H, NH);  $^{13}\text{C}$  NMR (100 MHz, DMSO- $d_6$ )  $\delta$ : 21.1, 26.8, 36.7, 44.4, 57.7, 106.6, 121.3, 129.7, 131.6, 141.3, 147.1, 150.6, 153.2, 193.8 ppm.

**9-(4-methyl-phenyl)-5,6,7,9-tetrahydro-1,2,4-triazolo[5,1-*b*]quinazolin-8(4H)-one (17a):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3425, 2922, 2990, 1643, 1575, 1550, 1412, 1361  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (300 MHz, DMSO- $d_6$ )  $\delta$ : 1.88-2.66 (m, 6H, 3CH<sub>2</sub>), 2.08 (s, 3H, CH<sub>3</sub>), 6.18 (s, 1H, CH), 7.07 (m, 4H-Ar), 7.67 (s, 1H, CH), 11.14 (br. s, 1H, NH);  $^{13}\text{C}$  NMR (75 MHz, DMSO- $d_6$ )  $\delta$ : 21.08, 21.14, 26.84, 36.78, 57.89, 107.17, 127.34, 129.24, 137.41, 139.16, 147.18, 150.42, 152.85, 193.73 ppm.

**9-(3-nitro-phenyl)-5,6,7,9-tetrahydro-1,2,4-triazolo[5,1-*b*]quinazolin-8(4H)-one (18a):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3410, 3220, 2930, 1620, 1557, 1521, 1465, 1405, 1347  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (300 MHz, DMSO- $d_6$ )  $\delta$ : 1.93- 2.02 (m, 2H, CH<sub>2</sub>), 2.20-2.33 (m, 2H, CH<sub>2</sub>), 2.66-2.75 (m, 2H, CH<sub>2</sub>), 6.42 (s, 1H, CH), 7.57-7.68 (m, 2H, CH-Ar), 7.74 (s, 1H-Ar), 8.07-8.12 (m, 2H-Ar), 11.33 (br. s, 1H, NH);  $^{13}\text{C}$  NMR (75 MHz, DMSO- $d_6$ )  $\delta$ : 21.10, 26.92, 36.68, 57.77, 106.10, 122.25, 123.27, 130.49, 134.14, 143.88, 147.18, 148.09, 150.91, 153.73, 193.92 ppm.



**3,3-Dimethyl-12-phenyl-3,4,5,12-tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2H)-one (1b):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 1569, 1647, 2956, 3367  $\text{cm}^{-1}$ .  $^1\text{H}$  NMR (300 MHz, DMSO- $d_6$ )  $\delta$ : 1.06 (s, 3H, CH<sub>3</sub>), 1.17 (s, 3H, CH<sub>3</sub>), 2.23-2.37 (dd,  $J = 16.43, 30.48$  Hz, 2H, CH<sub>2</sub>), 2.67 (s, 2H, CH<sub>2</sub>), 6.49 (s, 1H, CH), 7.09-7.65 (m, 9H, Ar-H), 10.87 (s, 1H, NH).  $^{13}\text{C}$  NMR (75 MHz, DMSO- $d_6$ )  $\delta$ : 26.73, 26.43, 31.79, 35.92, 49.99, 58.15, 112.56, 119.77, 125.57, 125.67, 126.19, 127.44, 127.66, 128.13, 132.02, 132.87, 142.05, 158.48, 158.53, 162.57, 195.73 ppm.

**3,3-Dimethyl-12-(4-chloro-phenyl)-3,4,5,12-tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2H)-one (2b):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3413, 3065, 2965, 1767, 1606, 1571, 1478, 1344, 1021  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (300 MHz, DMSO- $d_6$ )  $\delta$ : 0.96 (s, 3H, CH<sub>3</sub>), 1.15 (s, 3H, CH<sub>3</sub>), 2.24 (s, 2H, CH<sub>2</sub>), 2.67 (s, 2H, CH<sub>2</sub>), 4.84 (s, 1H, C-H), 7.19-7.54 (m, 2H), 7.68 (d,  $J = 8.4$  Hz, 3H), 8.01-8.26 (m, 3H) 11.31 (brs, N-H, 1H) ppm.  $^{13}\text{C}$  NMR (75 MHz, DMSO- $d_6$ )  $\delta$ : 18.76, 27.12, 28.13, 34.65, 51.55, 115.82, 120.60, 122.98, 125.01, 127.09, 129.87, 133.65, 145.12 (two peaks), 146.42, 147.56, 152.26, 167.01, 196.50 ppm.

**3,3-Dimethyl-12-(4-bromo-phenyl)-3,4,5,12-tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-one (3b):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3232, 3048, 2932, 2863, 1648  $\text{cm}^{-1}$ ;  **$^1\text{H NMR}$**  (300 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 0.94 (s, 3H,  $\text{CH}_3$ ), 1.05 (s, 3H,  $\text{CH}_3$ ), 2.00-2.30 (m, 2H,  $\text{CH}_2$ ), 2.60-2.70 (m, 2H,  $\text{CH}_2$ ), 6.45 (s, 1H, CH), 6.98 (t,  $J = 8.0$  Hz, 1H, H-Ar), 7.06 (t,  $J = 8.0$  Hz, 1H, H-Ar), 7.25-7.30 (m, 4H, H-Ar), 7.35-7.45 (m, 2H, H-Ar), 11.15 (s, 1H, NH);  **$^{13}\text{C NMR}$**  (75 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 27.9, 32.1, 43.1, 52.4, 58.5, 112.0, 114.4, 118.2, 123.6, 123.1, 130.3, 139.2, 143.2, 144.8, 1463.3, 148.8, 154.6, 199.8 ppm.

**3,3-dimethyl-12-(4-hydroxyphenyl)-3,4,5,12-tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-one (4b):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3448, 3255, 2890, 1641, 1612, 1589, 1566  $\text{cm}^{-1}$ ;  **$^1\text{H NMR}$**  (300 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 0.95 (s, 3H,  $\text{CH}_3$ ), 1.06 (s, 3H,  $\text{CH}_3$ ), 2.04 (d,  $J = 8.94$  Hz, 1H,  $\text{CH}_2$ ), 2.24 (d,  $J = 9.24$  Hz, 1H,  $\text{CH}_2$ ), 2.50-2.73 (m, 2H,  $\text{CH}_2$ ), 6.19 (s, 1H, CH), 6.50-7.50 (m, 8H, H-Ar), 9.32 (s, 1H, OH), 11.01 (brs, 1H, NH).  **$^{13}\text{C NMR}$**  (75 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 23 26.2, 47.4, 53.9, 110.5, 115.6, 122.9, 130.6, 137.9, 141.5, 154.3, 156.3, 156.3, 197.6 ppm.

**3,3-Dimethyl-12-(2,4-dichlorophenyl)-3,4,5,12-tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-one (5b):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3238, 3061, 2963, 2931, 1650, 1615, 1595, 1573, 1563, 1459, 1374, 1270, 737;  **$^1\text{H NMR}$**  (400 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 0.95 (s, 3H,  $\text{CH}_3$ ), 1.06 (s, 3H,  $\text{CH}_3$ ), 2.04 (d,  $J = 16.0$  Hz, 1H, H-4), 2.24 (d,  $J = 16.0$  Hz, 1H, H'-4), 2.29 (d,  $J = 12.0$  Hz, 1H, H-2), 2.33 (d,  $J = 16.0$  Hz, 1H, H'-2), 6.65 (s, 1H, H-12), 6.96 (t,  $J = 8.0$  Hz, 1H, Ar-H), 7.03 (s, 1H, Ar-H), 7.08-7.12 (m, 1H, Ar-H), 7.38 (s, 1H, Ar-H), 7.40 (s, 1H, Ar-H), 7.48 (d,  $J = 8.0$  Hz, 1H, Ar-H), 11.25 (s, 1H, NH) ppm.

**3,3-Dimethyl-12-(3,4,5-trimethoxyphenyl)-3,4,5,12-tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-one (6b):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3235, 3096, 2958, 2866, 1647  $\text{cm}^{-1}$ ;  **$^1\text{H NMR}$**  (300 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 1.00 (s, 3H,  $\text{CH}_3$ ), 1.08 (s, 3H,  $\text{CH}_3$ ), 2.11-2.28 (m, 2H,  $\text{CH}_2$ ), 2.60-2.70 (m, 2H,  $\text{CH}_2$ ), 3.66 (s, 9H,  $\text{OCH}_3$ ), 6.36 (s, 1H, CH), 6.64 (s, 2H-Ar), 7.00 (dt, 1H-Ar), 7.06 (dt, 1H-Ar), 7.37 (d, 1H-Ar), 7.42 (d, 1H-Ar), 10.90 (s, 1H, NH);  **$^{13}\text{C NMR}$**  (75 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 26.5, 28.8, , 32.1, 40.0, 50.1, 54.1, 105.1, 106.3, 110.2, 116.9, 120.4, 121.7, 132.0, 137.0137.2, 142.0, 145.4, 150.5, 152.7, 192.5 ppm.

**3,3-dimethyl-12-(4-methyl-phenyl)-3,4,5,12-tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-one (7b):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3245, 3040, 2952, 2895, 1648  $\text{cm}^{-1}$ ;  **$^1\text{H NMR}$**  (300 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 0.94 (s, 3H,  $\text{CH}_3$ ), 1.05 (s, 3H,  $\text{CH}_3$ ), 2.10 (s, 3H,  $\text{CH}_3$ ), 2.00-2.30 (m, 2H,  $\text{CH}_2$ ), 2.50-2.65 (m, 2H,  $\text{CH}_2$ ), 6.35 (s, 1H, CH), 6.95 (t,  $J = 7.6$  Hz, 1H, H-Ar), 7.02-7.06 (m, 3H, H-Ar), 7.20-7.28 (m, 3H, H-Ar), 7.38 (d,  $J = 8.6$  Hz, 1H, H-Ar), 11.06 (s, 1H, NH);  **$^{13}\text{C NMR}$**  (75 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 16.9, 20.9, 26.8, 47.3, 53.5, 110.2, 115.4, 122.9, 129.1, 134.0, 134.1, 134.7, 138.1, 141.3, 152.8, 198.4 ppm.

**3,3-Dimethyl-12-(2-methyl-phenyl)-3,4,5,12-tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-one (8b):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3540, 3095, 1642, 1610, 1563  $\text{cm}^{-1}$ ;  **$^1\text{H NMR}$**  (300 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 0.92 (s, 3H,  $\text{CH}_3$ ), 1.05 (s, 3H,  $\text{CH}_3$ ), 2.13 (q,  $J = 16.23$  Hz, 2H,  $\text{CH}_2$ ), 2.56 (q,  $J = 16.35$  Hz, 2H,  $\text{CH}_2$ ), 2.64 (s, 3H,  $\text{CH}_3$ ), 6.51 (s, 1H, CH), 6.89-7.38 (m, 8H-Ar), 11.15 (s, 1H, NH) ppm.

**3,3-Dimethyl-12-(3-nitrophenyl)-3,4,5,12-tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-one (9b):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3394, 2970, 1660, 1648, 1615, 1598  $\text{cm}^{-1}$ ;  **$^1\text{H NMR}$**  (300 MHz,  $\text{DMSO-d}_6$ ):  $\delta$ : 0.92 (s, 3H,  $\text{CH}_3$ ), 1.08 (s, 3H,  $\text{CH}_3$ ), 2.08 (d, 1H,  $J = 16.2$  Hz, one proton of diastereotopic protons in  $\text{CH}_2$ ), 2.29 (d, 1H,  $J = 16.2$  Hz, one proton of diastereotopic protons in  $\text{CH}_2$ ), 2.55–2.72 (m, 2H,  $\text{CH}_2$ ), 6.67 (s, 1H, CH), 6.98 (td, 1H,  $J = 7.8$  Hz, 1.2, HAr), 7.08 (td, 1H,  $J = 7.1, 0.9$  Hz, HAr), 7.31 (d, 1H,  $J = 7.5$  Hz, HAr), 7.41 (d, 1H,  $J = 7.5$  Hz, HAr), 7.57 (t, 1H,  $J = 7.8$  Hz, HAr), 7.74 (d, 1H,  $J = 7.8$  Hz, HAr), 8.03–8.08 (m, 1H, HAr), 8.29 (t, 1H,  $J = 1.8$  Hz, HAr), 11.30 (br., 1H, NH) ppm.

**12-phenyl-3,4,5,12-tetrahydrobenzo[4,5]imidazo[2,1-*b*]quinazolin-1(2*H*)-one (10b):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 1560, 1666, 2968, 3370  $\text{cm}^{-1}$ ;  **$^1\text{H NMR}$**  (300 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 1.86–1.93 (m, 2H,  $\text{CH}_3$ ), 2.23–2.29 (m, 2H,  $\text{CH}_3$ ), 2.59–2.61 (d,  $J = 5.05$  Hz, 2H,  $\text{CH}_2$ ), 6.49 (s, 1H, CH), 7.09–7.90 (m, 9H, Ar-H), 11.11 (s, 1H, NH);  **$^{13}\text{C NMR}$**  (75 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 18.55, 19.73, 26.39, 30.14, 35.97, 49.99, 58.15, 112.64, 119.67, 125.56, 125.67, 126.28, 127.34, 127.54, 128.22, 132.13, 132.89, 142.08, 159.05, 159.09, 162.74, 196.04 ppm.

**12-(3,4,5-trimethoxyphenyl)-3,4,5,12-tetrahydrobenzo[4,5]imidazo[2,1-*b*]quinazolin-1(2*H*)-one (13b):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3430, 2998, 2890, 1641, 1612, 1589, 1555  $\text{cm}^{-1}$ ;  **$^1\text{H NMR}$**  (400 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 1.89–2.01 (m, 2H), 2.23–2.34 (m, 2H), 2.63–2.77 (m, 2H), 3.55 (s, 3H), 3.68 (s, 6H), 6.39 (s, 1H), 6.63 (s, 2H), 6.97–7.01 (m, 2H), 7.37 (d,  $J = 7.6$  Hz, 1H), 7.45 (d,  $J = 7.6$  Hz, 1H), 11.08 (s, 1H);  **$^{13}\text{C NMR}$**  (100 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 20.7, 26.5, 36.4, 53.9, 55.8, 59.8, 104.5, 107.2, 110.2, 116.8, 120.4, 121.7, 132.0, 136.8, 137.1, 141.8, 145.1, 152.6, 192.9 ppm.

**12-(4-nitrophenyl)-3,4,5,12-tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-one (15b):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3223, 3040, 2949, 1646, 1515, 898, 822  $\text{cm}^{-1}$ ;  **$^1\text{H NMR}$**  (400 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 1.80–1.97 (m, 2H,  $\text{CH}_2$ ), 2.15–2.31 (m, 2H,  $\text{CH}_2$ ), 2.65–2.68 (m, 2H,  $\text{CH}_2$ ), 6.59 (s, 1H, CH), 6.92 (t,  $J = 8.0$  Hz, 1H, ArH), 7.02 (t,  $J = 7.2$  Hz, 1H, ArH), 7.17 (d,  $J = 7.6$  Hz, 1H, ArH), 7.35 (d,  $J = 8.0$  Hz, 1H, ArH), 7.56 (d,  $J = 8.8$  Hz, 2H, ArH), 8.06 (d,  $J = 8.8$  Hz, 2H, ArH), 11.25 (s, 1H, NH) ppm.

**12-(4-fluorophenyl)-3,4,5,12-tetrahydrobenzimidazo[2,1-*b*]quinazolin-1(2*H*)-one (16b):**

**FT-IR** (KBr,  $\text{cm}^{-1}$ ): 3227, 3043, 2956, 1644, 1510, 890, 759  $\text{cm}^{-1}$ ;  **$^1\text{H NMR}$**  (400 MHz,  $\text{DMSO-d}_6$ )  $\delta$ : 1.83–1.97 (m, 2H,  $\text{CH}_2$ ), 2.15–2.31 (m, 2H,  $\text{CH}_2$ ), 2.63–2.65 (m, 2H,  $\text{CH}_2$ ), 6.41 (s, 1H, CH), 6.92 (t, 1H,  $J = 7.2$  Hz, ArH), 6.99–7.04 (m, 3H, ArH), 7.20 (d, 1H,  $J = 7.6$  Hz, ArH), 7.31–7.34 (m, 3H, ArH), 11.10 (s, 1H, NH) ppm.

