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SUPPLEMENTARY MATERIAL TO Synthesis of novel N-substituted benzyl N-(1,3-benzothiazol-2-yl) acetamides and their *in vitro* antibacterial activities

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N-[2-Methylphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3a). Yield: 70%. M.p. 132-133°. FT-IR: 3042, 1610, 1597, 1530, 1505 (Fig.S1). 1H NMR(500 MHz, DMSO-d6, δ) : 2.45 (s, 3H, CH3-Benzothiazole), 2.65 (s, 3H, Ar-CH3), 7.34 (dd, 1H, 4J12,14=1.28 Hz, J11,13=8.23 Hz, H12), 7.38 (d, 1H, J14,15=7.81 Hz, H15), 7.39 (t, 1H, J12,14=8.93 Hz, H13), 7.53 (dt, 1H, 4JH14-H12=1.31 Hz, J13,15=7.42 Hz, H14), 7.83 (s, 1H, H7), 7.84 (d, 1H, J5,4=8.44 Hz, H5), 8.12 (d, 1H, J4,5=7.81 Hz, H4), 9.34 (s, 1H, -CH=N-) (Fig.S2) (The numbering of protons of the compound is given in Fig. 1). 13C NMR(125 MHz, DMSO-d6, δ): 19.07, 21.24, 117.88, 118.67, 121.28, 125.83, 126.45, 126.88, 127.04, 128.68, 130.90, 131.15, 136.48, 138.58, 150.54, 164.16. Anal. Calcd for C16H14N2S: C 72.15, H 5.30, N 10.52, S 12.04 %; found: C 72.00, H 5.35, N 10.54, S 11,96 %.

N-[2-Methoxyphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3b). Yield: 76%. M.p. 134-136°. FT-IR: 3045, 1615, 1595, 1551 (Fig.S3). 1H NMR(500 MHz, DMSO-d6, δ) : 2.46 (s, 3H, CH3-Benzothiazole), 3.96(s, 3H, Ar-OCH3), 7.14 (t, 1H, J15,14=7.51 Hz, H14), 7.24 (d, 1H, J13,12=8.41 Hz, H12), 7.35 (dd, 1H, 4J5,7=1.34 Hz, J4,5=8.31 Hz, H5), 7.66 (dt, 1H, 4J13,15=1.75 Hz, J12,14=7.24 Hz, H13), 7.83 (d, 1H, J4,5=8.30 Hz, H4), 7.85 (s, 1H, H7), 8.11 (dd, 1H, 4J15,13=1.65 Hz, J15,14=7.76 Hz, H15), 9.30 (s, 1H, -CH=N-)(Fig.S4 and fig. S5). 13C NMR (125 MHz, DMSO-d6, δ): 21.21, 56.53, 112.93, 121.50, 122.35, 122.71, 122.77, 128.03, 128.56, 134.62, 135.43, 136.11, 149.86, 161.08, 161.73, 171.33. ESI-MS m/z= Calculated for (C16H14N2SO + H+) 283.1, observed 283.18 (Fig.S8). Anal. Calcd for C16H14N2SO: C 68.06, H 5.00, N 9.92, S 11.36 %. Found: C 68.20, H 5.20, N 9.98, S 11.35 %.

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N-[2-hyroxyphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3c). Yield: 90%. M.p. 137-138°. FT-IR: 3360, 3040, 1620, 1583, 1570, 1545 (Fig.S9). 1H NMR(500 MHz, DMSO-d6, δ): 2.44 (s, 3H, CH3-Benzothiazole); 7.01(d, 1H, J15,13=7.48 Hz H14), 7.04 (d, 1H, J13,12=8.14 Hz, H12), 7.34 (d, 1H, J5,4=8.43 Hz, H4), 7.52 (dt, 1H, 4J13,15=1.27 Hz, J12,14=8.40 Hz, H13), 7.83 (d, 1H, J4,5=8.20 Hz, H5), 7.86 (s, 1H, H7), 7.93 (dd, 1H, 4J15,13=1.21 Hz, J15,14=7.80 Hz, H15), 9.45 (s, 1H, -CH=N-), 11.5 (s, 1H, PhOH)(Fig.S10 and S11). 13C NMR(125 MHz, DMSO-d6, δ): 21.58, 117.41, 120.06, 120.31, 122.40, 122.70, 128.61, 131.66, 134.47, 135.62, 135.98, 149.73, 160.98, 166.04, 169.73 (Fig.S12). Anal. Calcd. for C15H12N2OS: C 67.14, H 4.51, N 10.44, S 11.95 %. Found: C 67.10, H 4.56, N 10.49, S 11,40 %.

N-[4-hydroxyphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3d). Yield: 72%. M.p. 136-138°. FT-IR: 3350, 3044, 1618, 1585, 1572, 1543. 1H NMR(500 MHz, DMSO-d6, δ): 2.40 (s, 3H, CH3-Benzothiazole), 6.96 (d, 2H, J11,13=8.55 Hz, H12 and H14), 7.30 (d, 1H, J5,4=8.11 Hz, H4), 7.77 (d, 1H, J4,5=8.45 Hz, H5), 7.79 (s, 1H, H7), 7.94 (d, 2H, J12,14=8.56 Hz, H11 and H15), 9.02 (s, 1H, -CH=N-), 10.5 (s, 1H, Ar-OH)(Fig.S13). 13C NMR(125 MHz, DMSO-d6, δ): 21.54, 116.63, 122.27, 122.36, 126.37, 128.34, 133.08, 134.36, 135.07, 149.92, 163.24, 166.49, 171.41(Fig.S14). Anal. Calcd. for C15H12N2OS: C 67.14, H 4.51, N 10.44, S 11.95 %. Found: C 67.02, H 4.54, N 10.40, S 11,45 %.

N-(2-methylbenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4a). Yield: 87%. M.p. 102-103°. FT-IR: 3412, 1605, 1594, 1571. 1H NMR(500 MHz, DMSO-d6, δ): 2.32 (s, 3H, CH3-Benzothiazole), 2.34 (s, 3H, Ar-CH3), 4.55 (d, 2H, JCH2-NH=5.44 Hz, Ar-CH2NH-), 7.03 (dd, 1H, 4J5,7=1.31 Hz, J4,5=8.15 Hz, H5), 7.18 (m, 3H, H12, H-14 and H15), 7.27 (d, 1H, J5,4=8.11 Hz, H4), 7.32 (d, 1H, J12,14=7.29 Hz, H13), 7.47 (s, 1H, H7), 8.25 (t, 1H, JNH-CH2=5.43 Hz, NHCH2-Ar)(Fig.S15). 13C NMR(125 MHz, DMSO-d6, δ): 19.12, 21.22, 45.92, 118.20, 121.35, 126.27, 126.99, 127.61, 128.28, 130.51, 130.98, 136.43, 137.05, 150.81, 165.78 (Fig.S16). ESI-MS m/z= Calculated for (C16H16N2S + H+) 269.1, observed 269.1 (Fig.S19). Anal. Calcd for C16H16N2S: C 71.60, H 6.01, N 10.44, S 11.95 %. Found: C 71.52, H 6.03, N 10.50, S 11.90 %.

N-(2-methoxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4b). Yield: 90%. M.p. 125-127°. FT-IR: 3430, 1608, 1567(Fig.S20). 1H NMR(500 MHz, DMSOd6, δ): 2.31 (s, 3H, CH3-Benzothiazole), 3.83 (s, 3H, Ar-OCH3), 4.53 (d, 2H, JCH2-NH=5.67 Hz, Ar-CH2NH-), 6.92 (t, 1H, J15,13=7.28 Hz, H14), 7.02 (d, 2H, J12,13=J4,5=8.09 Hz, H12 and H4), 7.25 (d, 2H, J4,5 J12,14=7.03 Hz, H5 and H15), 7.29 (t, 1H, J12,14=7.13 Hz H13), 7.46 (s, 1H, H7); 8.22 (t, 1H, JNH-CH2=5.67 Hz, NHCH2-Ar) (Fig.S21 and S22). 13C NMR(125 MHz, DMSO-d6, δ): 21.22, 42.89, 55.82, 111.06, 118.18, 120.62, 121.32, 126.79, 126.96, 128.46, 128.81, 130.44, 130.97, 150.83, 157.32, 166.00 (Fig.S23). ESI-MS m/z=

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Calculated for (C16H16N2SO + H+) 285.1, observed 285.1. Anal. Calcd for C16H16N2SO: C 67.58, H 5.67, N 9.85, S 11.28 %. Found: C 67.28, H 4.58, N 9.86, S 10.40 %.

N-(2-hyroxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4c). Yield: 90%. M.p. 170-172°. FT-IR: 3525, 3409, 1595, 1570(Fig.S28). 1H NMR(500 MHz, DMSO-d6, δ): 2.32 (s, 3H, CH3-Benzothiazole), 4.47 (d, 2H, JCH2-NH=5.796 Hz, Ar-CH2NH-), 6.74 (dt, 1H, 4J14,12=0.79 Hz J15,13=7.41 Hz, H14), 6.86 (dd, 1H, 4J12,14=0.65 Hz, J13,12=7.64 Hz, H12), 7.06 (dd, 1H, 4J15,13=1.36 Hz, J14,15=8.24 Hz, H15), 7.10 (dt, 1H, 4J13,15=1.61 Hz, J12,14=7.69 Hz, H13), 7.24 (dd, 1H, 4J5,7=1.16 Hz, J4,5=7.53 Hz, H5), 7.26 (d, 1H, J5,4=8.11 Hz, H4), 7.47 (s, 1H, 4J7,5=0.97 Hz, H7), 8.30 (t, 1H, J=5.70 Hz, NHCH2-Ar), 9.75 (br. s, 1H, Ar-OH) (Fig.S29). 13C NMR(125 MHz, DMSO-d6, δ): 21.22, 43.14, 115.85, 118.00, 119.37, 121.41, 125.34, 127.04, 128.69, 129.36, 130.55, 130.81, 150.45, 155.61, 166.29. Anal. Calcd. for C15H14N2OS: C 66.64, H 5.22, N 10.36, S 11.86 %. Found: C 66.60, H 5.21, N 10.33, S 11.84 %.

N-(4-hyroxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4d). Yield: 93%. M.p. 168-170°. FT-IR: 3542, 3430, 1591, 1556. 1H NMR(500 MHz, DMSO-d6, δ): 2.46 (s, 3H CH3-Benzothiazole), 4.49 (d, 2H, JCH2-NH=5.20 Hz, Ar-CH2NH-), 6.74 (d, 2H, J11,13=8.43 Hz, H12 and H14), 7.02 (d, 1H, J4,5=7.16 Hz, H5), 7.2 (d, 2H, J12,14=8.41 Hz, H11 and H15), 7.24 (d, 1H, J5,4=6.84 Hz, H4), 8.28 (t, 1H, JNH-CH2=5.44 Hz, NHCH2-Ar), 9.30 (br. s, 1H, Ar-OH) (Fig.S30 and S31). 13C NMR(125 MHz, DMSO-d6, δ): 21.22, 47.39, 115.54, 118.15, 121.32, 16.98, 129.34, 129.49, 130.42, 130.92, 150.86, 156.95, 165.91 (Fig.S32). Anal. Calcd. for C15H14N2OS: C 66.64, H 5.22, N 10.36, S 11.86 %. Found: C 66.61, H 5.18, N 10.31, S 11.82 %.

2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2-methylbenzyl] acetamide (5a). Yield: 76%. M.p. 188-190°. FT-IR: 1698, 1600, 1509(Fig.S36). 1H NMR(500 MHz, DMSO-d6, δ): 2.49 (s, 6H, CH3-Benzothiazole and Ar-CH3), 4.55 (s, 2H, Ar-CH2N-), 5.50 (s, 2H, N(CO)CH2Cl), 6.76 (d, 1H, J14,15=7.59 Hz, H15), 7.11 (t, 1H, J12,14=7.54 Hz, H13), 7.18 (t, 1H, J15,13=7.36 Hz, H14), 7.24 (d, 1H, J13,12=8.20 Hz, H12), 7.28 (d, 1H, J4,5=8.63 Hz, H5), 7.62 (d, 1H, J5,4=8.27 Hz, H4), 7.84(s, 1H, H7) (Fig.S37). 13C NMR(125 MHz, DMSO-d6, δ): 19.99, 22.30, 44.62, 49.49, 122.16, 122.9, 124.72, 127.64, 128.37, 128.95, 131.69, 133.31, 134.12, 135.15, 135.54, 136.40, 152.03, 168.63 (Fig.S38). ESI-MS m/z= Calculated for (C18H17CIN2SO + H+) 345.1, observed 345.1(Fig.S40). Anal. Calcd for C18H17CIN2SO: C 62.69, H 4.97, N 8.12, S 9.30 %. Found: C 62.48, H 5.006, N 8.339, S 9.265 %.

2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2methoxybenzyl]acetamide (5b). Yield: 82%. M.p. 146-149°. FT-IR: 1683, 1605, 1520 (Fig.S41). 1H NMR(500 MHz, DMSO-d6, δ): 2.40 (s, 3H, CH3-Benzothiazole), 3.53 (s, 3H, Ar-OCH3), 4.75 (s, 2H, CH2N), 5.43 (s, 2H, N(CO)CH2Cl), 6.87 (t, 1H, J12,14=7.35 Hz, H13), 6.92 (d, 1H, J13,12=6.80 Hz, H12), 7.08 (d, 1H, J14,15=8.21 Hz, H15), 7.25 (d, 1H, J4,5=8.94 Hz, H5); 7.29 (t, 1H, J15,13=7.30, H14), 7.63 (d, 1H, J5,4=8.26 Hz, H4), 7.81 (s, 1H, H7) (Fig.S42, fig.S43 and S44). 13C NMR(125 MHz, DMSO-d6, δ): 21.48, 43.77, 46.79, 55.98, 61.29, 111.43, 121.02, 121.34, 121.61, 123.94, 126.50, 128.09, 129.16, 133.22, 134.24, 145.99, 157.00, 167.86 (Fig.S45). ESI-MS m/z= Calculated for (C18H17CIN2SO2 + H+) 361.1, observed 361.1(Fig.S48). Anal. Calcd for C18H17CIN2SO2: C 59.91, H 4.75, N 7.76, S 8.89 %. Found: C 59.72, H 4.64, N 7.63, S 8.99 %.

2-[(Chloroacetyl)(6-methyl-1,3-benzothiazol-2-yl)aminomethyl]phenyl chloroacetate (5c). Yield: 78%. M.p. 159-161°. FT-IR: 1775, 1668, 1600, 1505. 1H NMR(500 MHz, DMSO-d6, δ): 2.40 (s, 3H, CH3-Benzothiazole), 4.70 (s, 2H, Ar-CH2N-), 4.80 (s, 2H, O(CO)CH2Cl), 5.50 (s, 2H, N(co)ch2Cl), 7.07 (d, 1H, J14,15=7.69 Hz, H15), 7.10 (d, 1H, J4,5=7.49 Hz, H5), 7.20 (t, 1H, J15,13=7.49 Hz, H14), 7.30 (d, 1H, J13,12=8.03 Hz, H12), 7.40 (t, 1H J12,14=7.89 Hz, H13), 7.65 (d, 1H, J5,4=8.28 Hz, H4), 7.85 (s, 1H, H7). Anal. Calcd. for C19H16Cl2N2O3S: C 53.90, H 3.81, N 6.62, S 7.58 %. Found: C 53.92, H 3.84, N 6.59, S 7.55 %.

4-[(Chloroacetyl)(6-methyl-1,3-benzothiazol-2-yl)aminomethyl]phenyl chloroacetate (5d). Yield: 82%. M.p. 156-158°. FT-IR: 1762, 1663, 1593, 1520. 1H NMR(500 MHz, DMSO-d6, δ): 2.43 (s, 3H, CH3-Benzothiazole), 4.69 (s, 2H, Ar-CH2N), 4.76 (s, 2H, O(CO)CH2Cl), 5.57 (s, 2H,N(co)ch2Cl), 6.74 (d, 2H, J11,15=7.69 Hz, H12 and H14), 7.10 (d, 2H, J12,14=8.5 Hz, H11 and H15), 7.28 (d, 1H, J4,5=7.35 Hz, H5), 7.69 (d, 1H, J5,4=8.24 Hz, H4), 7.83 (s, 1H, H7), 8.59 (s, 1H, Ar-OH). Anal. Calcd. for C19H16Cl2N2O3S : C 53.90, H 3.81, N 6.62, S 7.58 %. Found: C 53.88, H 3.83, N 6.61, S 7.55 %.



Figure S-1.FT-IR spectrum of N-[2-Methylphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3a).

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Figure S-2. 1H NMR spectrum of N-[2-Methylphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3a)



Figure S-3. FT-IR spectrum of N-[2-Methoxyphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3b)



Figure S-4. 1H NMR spectrum of N-[2-Methoxyphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3b)



Figure S-5. 1H NMR spectrum of N-[2-Methoxyphenyl)methylidene]-6methyl-1,3-benzothiazol-2-amine (3b)



Figure S-6. 1H-1H COSY spectrum of N-[2-Methoxyphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3b).



Figure S-7. HETCOR spectrum of N-[2-Methoxyphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3b)

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Figure S-8. ESI-MS spectrum of N-[2-Methoxyphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3b)



Figure S-9. FT-IR Spectrum of N-[2-hyroxyphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3c)



Figure S-10. 1H NMR spectrum of N-[2-hyroxyphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3c)



Figure S-11. 1H NMR spectrum of N-[2-hyroxyphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3c)



Figure S-12. 13C NMR spectum of N-[2-hyroxyphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3c).



Figure S-13. 1H NMR spectrum of N-[4-hydroxyphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3d)



Figure S-14. 13C NMR spectrum of N-[4-hydroxyphenyl)methylidene]-6-methyl-1,3-benzothiazol-2-amine (3d).



Figure S-15. 1H NMR spectrum of N-(2-methylbenzyl)-6-Methyl-1,3benzothiazol-2-amine (4a)



Figure S-16. 13C NMR spectrum of N-(2-methylbenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4a)



Figure S-17. COSY spectrum of N-(2-methylbenzyl)-6-Methyl-1,3benzothiazol-2-amine (4a)





Figure S-18. HETCOR spectrum of N-(2-methylbenzyl)-6-Methyl-1,3benzothiazol-2-amine (4a)



Figure S-19. ESI-MS spectrum of N-(2-methylbenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4a)

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Figure S-20. FT-IR spectrum of N-(2-methoxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4b).



Figure S-21. 1H NMR spectrum of N-(2-methoxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4b).



Figure S-22. 1H NMR spectrum of N-(2-methoxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4b).



benzothiazol-2-amine (4b).

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Figure S-24. COSY spectrum of N-(2-methoxybenzyl)-6-Methyl-1,3benzothiazol-2-amine (4b)



Figure S-25. COSY spectrum of N-(2-methoxybenzyl)-6-Methyl-1,3benzothiazol-2-amine (4b)





Figure S-26. HETCOR spectrum of N-(2-methoxybenzyl)-6-Methyl-1,3benzothiazol-2-amine (4b)



Figure S-27. HETCOR spectrum of N-(2-methoxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4b)

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Figure S-28. FT-IR spectrum of N-(2-hyroxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4c).



Figure S-29. 1H NMR spectrum of N-(2-hyroxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4c).



Figure S-30. 1H NMR spectrum of N-(4-hyroxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4d).



Figure S-31. 1H NMR spectrum of N-(4-hyroxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4d).



Figure S-32. 13C NMR spectrum of N-(4-hyroxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4d).



Figure S-33. COSY spectrum of N-(4-hyroxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4d).



Figure S-34. COSY spectrum of N-(4-hyroxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4d).

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Figure S-35. HETCOR spectrum of N-(4-hyroxybenzyl)-6-Methyl-1,3-benzothiazol-2-amine (4d).



Figure S-36. FT-IR spectrum of 2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2-methylbenzyl] acetamide (5a).



Figure S-37. 1H NMR spectrum of 2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2-methylbenzyl] acetamide (5a).



Figure S-38. 13C NMR spectrum of 2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2-methylbenzyl] acetamide (5a).

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Figure S-39. HETCOR spectrum of 2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2-methylbenzyl] acetamide (5a).



Figure S-40. ESI-MS spectrum of 2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2-methylbenzyl] acetamide (5a)

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Figure S-41. FT-IR spectrum of 2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2-methoxybenzyl]acetamide (5b).



Figure S-42. 1H NMR spectrum of 2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2-methoxybenzyl]acetamide (5b).



Figure S-43. 1H NMR spectrum of 2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2-methoxybenzyl]acetamide(5b).



Figure S-44. 1H NMR spectrum of 2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2-methoxybenzyl]acetamide(5b).



Figure S-45. 13C NMR spectrum of 2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2-methoxybenzyl]acetamide (5b).



Figure S-46. COSY spectrum of 2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2-methoxybenzyl]acetamide (5b).

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Figure S-47. COSY spectrum of 2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2-methoxybenzyl]acetamide (5b).



Figure S-48. ESI-MS spectrum of 2-Chloro-N-[6-methyl-1,3-benzothiazol-2-yl]-N-[2-methoxybenzyl]acetamide (5b).