Electronic Supplementary Material (ESI) for RSC Advances.

A Facile and Green Approach for the Synthesis of Spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile *via* One-Pot Three-Component Condensation Reaction using DBU as Catalyst

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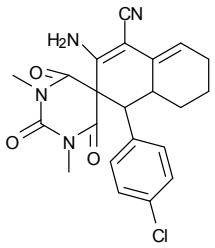
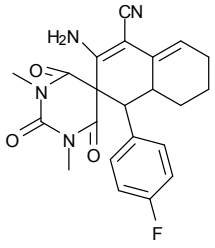
Supplementary data

3-amino-1-(4-chlorophenyl)-1',3'-dimethyl-2',4',6'-trioxo-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{22}H_{21}ClN_4O_3$, 4a)



White solid; Yield: 89%; M.p. (°C): 286-288; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2195, 1676, 1426, 1376 ; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 7.39 (d, 1H, *J*= 7.63 Hz, Ar-H), 7.32 (d, 1H, *J*= 7.63 Hz, Ar-H), 6.93 (d, 1H, *J*= 8.39 Hz, Ar-H), 6.88 (d, 1H, *J*= 7.63 Hz, Ar-H), 6.53 (s, 2H, -NH_2), 5.59 (s, 1H, vinylic), 3.35 (m, 1H, -CH), 3.07 (d, 1H, *J*= 12.21 Hz, -CH), 2.91 (s, 3H, -CH_3), 2.89 (s, 3H, -CH_3), 2.16-2.12 (m, 1H, -CH_2), 2.04-2.03 (m, 1H, -CH_2), 1.63-1.62 (m, 1H, -CH_2), 1.37-1.28 (m, 2H, -CH_2), 0.79-0.70 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.5, 166.93, 150.8, 149.6, 133.95, 132.9, 132.1, 130.6, 128.4, 127.8, 117.8, 117.4, 82.11, 60.6, 55.1, 32.2, 28.2, 28.1, 27.3, 24.9, 21.5;LCMS (ESI) *m/z* calcd. for $C_{22}H_{21}ClN_4O_3$: 424.129 [M^+]; found: 425.1362 [M+H]$^+$.

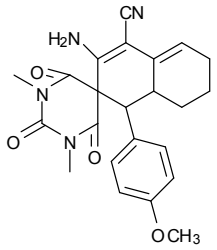
3-amino-1-(4-fluorophenyl)-1',3'-dimethyl-2',4',6'-trioxo-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{22}H_{21}FN_4O_3$, 4b)



White solid; Yield: 85%; M.p. (°C): 258-260; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2206, 1674, 1595, 1385, 846; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 7.16-7.13 (dt, 1H, Ar-H), 7.08-7.04 (dt, 1H, Ar-H), 6.94-6.91 (t, 1H, Ar-H), 6.89-6.86 (t, 1H, Ar-H), 6.50 (s, 2H, -NH_2), 5.57 (s, 1H, vinylic), 3.33 (m, 1H, -CH), 3.30 (d, 1H, *J*= 12.9 Hz, -CH), 2.88 (s, 3H, -CH_3), 2.86 (s, 3H, -CH_2), 2.15-2.10 (m, 1H, -CH_2), 2.03-1.97 (m, 1H, -CH_2), 1.63-1.61 (m, 1H, -CH_2), 1.37-1.28 (m, 2H, -CH_2), 0.77-0.68 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.5, 167.0, 161.7, 150.9,

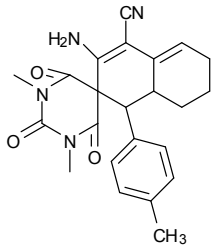
149.7, 132.3, 131.1, 130.7, 128.0, 127.9, 117.7, 117.3, 115.2, 82.1, 60.8, 55,1, 32.2, 28.1, 28.1, 27.3, 24.9, 21.6; LCMS (ESI) *m/z* calcd. for $C_{22}H_{21}FN_4O_3$: 408.1621 [M^+]; found: 409.1695 [M+H]$^+$.

3-amino-1-(4-methoxyphenyl)-1',3'-dimethyl-2',4',6'-trioxo-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{23}H_{24}N_4O_4$, 4c)



White solid; Yield: 92%; M.p. (°C): 253-255; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2206, 1688, 1436, 1264; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 6.87-6.77 (m, 4H, Ar-H), 6.49 (s, 2H, -NH_2), 5.57 (s, 1H, vinylic), 3.68 (s, 3H, -OCH_3), 3.35 (m, 1H, -CH), 2.99-2.96 (m, 1H, -CH), 2.90 (s, 3H, -CH_3), 2.88 (s, 3H, -CH_3), 2.16-2.12 (m,1H, -CH_2), 2.04-1.90 (m, 1H, -CH_2), 1.63 (bs, 1H, -CH_2), 1.35 (bs, 2H, -CH_2), 0.73-0.69 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.8, 167.1, 159.0, 151.1, 149.7, 131.4, 131.0, 126.9, 126.52, 117.8, 117.1, 114.1, 113.2, 82.1, 61.0, 55.4, 55.1, 32.3, 28.1, 28.1, 27.5, 24.9, 21.6; LCMS (ESI) *m/z* calcd. for $C_{23}H_{24}N_4O_4$: 420.1795 [M^+]; found: 421.1868 [M+H]$^+$.

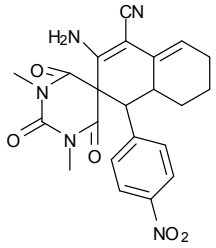
3-amino-1',3'-dimethyl-2',4',6'-trioxo-1-p-tolyl-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{23}H_{24}N_4O_3$, 4d)



White solid; Yield: 90%; M.p. (°C): 241-243; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2206, 1659, 1443, 1372; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 7.10 (d, 1H, *J*= 7.63 Hz, Ar-H), 7.02 (d, 1H, *J*=7.63 Hz, Ar-H), 6.76 (d, 1H, *J*= 8.39 Hz, Ar-H), 6.68 (d, 1H, *J*=7.63 Hz, Ar-H), 6.47 (s, 2H, -NH_2), 5.56 (s, 1H, vinylic), 3.33 (m, 1H, -CH), 2.96 (d, 1H, *J*= 12.21 Hz, -CH), 2.87 (s, 3H, -CH_2), 2.84 (s, 3H, -CH_2), 2.20 (s, 3H, -CH_3), 2.15-2.09 (m, 1H, -CH_2), 2.03-1.96 (m, 1H, -CH_2), 1.62-1.59

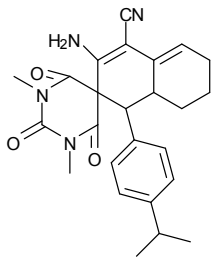
(m, 1H, -CH_2), 1.39-1.29 (m, 2H, -CH_2), 0.73-0.63 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.7, 167.1, 151.1, 149.7, 137.5, 131.8, 131.0, 130.2, 128.8, 125.6, 117.8, 117.1, 82.1, 60.8, 55.7, 32.3, 28.1, 28.1, 27.5, 24.9, 21.6, 20.7; LCMS (ESI) *m/z* calcd. for $C_{23}H_{24}N_4O_3$: 404.1853 [M^+]; found: 405.1926 [M+H]$^+$.

3-amino-1',3'-dimethyl-1-(4-nitrophenyl)-2',4',6'-trioxo-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{23}H_{24}N_4O_3$, 4e)



Pale yellow solid; Yield: 91%; M.p. (°C): 251-253; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2206, 1681, 1429, 1349; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 8.18 (dd, 1H, *J*= 8.77 and 3.05 Hz, Ar-H), 8.11 (dd, 1H, *J*= 8.39 and 2.29 Hz, Ar-H), 7.20 (d, 1H, *J*= 8.39 Hz, Ar-H), 7.16 (d, 1H, *J*=9.92 Hz, Ar-H), 6.54 (s, 2H, -NH_2), 5.60 (s, 1H, vinylic), 3.24 (d, 1H, *J*= 12.3 Hz, -CH), 2.97-2.95 (m, 1H, -CH), 2.90 (s, 3H, -CH_3), 2.87 (s, 3H, -CH_3), 2.16-2.11 (m, 1H, -CH_2), 2.02-1.97 (m, 1H, -CH_2), 1.63-1.61 (m, 1H, -CH_2), 1.41-1.35 (m, 1H, -CH_2), 1.25-122 (m, 1H, -CH_2), 0.81-0.72 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.2, 166.7, 150.6, 149.5, 147.2, 142.9, 131.9, 130.2, 127.7, 123.5, 123.3, 117.7, 117.6, 82.1, 60.3, 55.2, 32.2, 28.2, 28.2, 27.2, 24.8, 21.5; LCMS (ESI) *m/z* calcd. for $C_{22}H_{21}N_5O_5$: 435.1561 [M^+]; found: 436.1632 [M+H]$^+$.

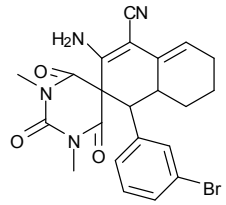
3-amino-1-(4-isopropylphenyl)-1',3'-dimethyl-2',4',6'-trioxo-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{25}H_{28}N_4O_3$, 4f)



White solid; Yield: 88%; M.p. (°C): 207-209; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2206, 1674, 1436, 1372; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 7.16 (d, 1H, *J*=8.39 Hz, Ar-H), 7.06 (d, 1H, *J*=9.16 Hz, Ar-H), 6.80 (d, 1H, *J*= 8.39 Hz, Ar-H), 6.70 (d, 1H, *J*= 7.63 Hz, Ar-H), 6.46 (s, 2H, -NH_2), 5.56 (s,

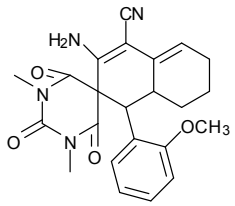
1H, vinylic), 3.52-3.43 (m, 1H, -CH(CH_3)$_2$), 3.30 (s, 1H, -CH), 2.99-2.95 (m, 1H, -CH), 2.83 (s, 3H, -CH_3), 2.95 (s, 3H, -CH_3), 2.15-2.10 (m, 1H, -CH_2), 2.05-1.91 (m, 1H, -CH_2), 1.62-1.57 (m, 2H, -CH_2), 1.40-136 (m, 1H, -CH_2), 1.10 (s, 3H, -CH_3), 1.08 (s, 3H, -CH_3), 0.79-0.71 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.7, 167.1, 151.1, 149.5, 148.7, 132.0, 130.9, 130.1, 126.0, 125.9, 125.7, 117.8, 117.1, 82.1, 61.0, 55.8, 33.1, 32.0, 28.2, 28.1, 27.5, 24.9, 23.8, 23.7, 21.6; LCMS (ESI) *m/z* calcd. for $C_{25}H_{28}N_4O_3$: 432.2167 [M^+]; found: 433.2238 $[M+H]^+$.

3-amino-1-(3-bromophenyl)-1',3'-dimethyl-2',4',6'-trioxo-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{22}H_{21}BrN_4O_3$, 4g)



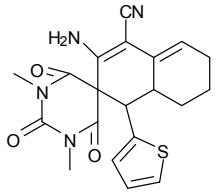
White solid; Yield: 82%; M.p. (^{o}C): 250-253; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2202, 1676, 1434, 1382, 746; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 7.47 (d, 1H, *J*= 7.63 Hz, Ar-H), 7.23 (td, 1H, *J*= 7.63 Hz, Ar-H), 7.05 (s, 1H, Ar-H), 6.89 (dd, 1H, *J*= 7.63 Hz, Ar-H), 6.50 (s, 2H, -NH_2), 5.58 (s, 1H, vinylic), 3.29-3.26 (m, 1H, -CH), 3.06 (d, 1H, *J*= 12.21 Hz, -CH), 2.94-2.85 (m, 6H, 2(CH_3)), 2.15-2.11 (m, 1H, -CH_2), 2.04-1.97 (m, 1H, -CH_2), 1.64-1.61 (m, 1H, -CH_2), 1.43-1.30 (m, 2H, -CH_2), 0.83-0.71 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.4, 168.3, 167.0, 166.7, 150.7, 149.6, 137.6, 132.8, 131.4, 130.6, 129.4, 128.6, 117.7, 117.6, 82.1, 60.7, 55.5, 32.0, 28.2, 27.2, 24.9, 21.5; LCMS (ESI) *m/z*calcd.for $C_{22}H_{21}BrN_4O_3$: 468.0789 [M^+]; found: 469.0855 $[M+H]^+$,471.0856 $[M+(H+2)]^+$.

3-amino-1-(2-methoxyphenyl)-1',3'-dimethyl-2',4',6'-trioxo-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{23}H_{24}N_4O_4$, 4h)



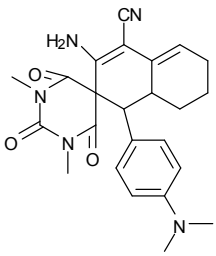
White solid; Yield: 91%; M.p. (°C): 243-246; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2202, 1682, 1426, 1382; ^1H NMR (400 MHz, DMSO-d_6) δ_H: 6.87 (d, 1H, *J*= 8.39 Hz, Ar-H), 6.79-6.70 (m, 3H, Ar-H), 6.46 (s, 2H, -NH_2), 5.55 (s, 1H, vinylic), 3.66 (s, 3H, -OCH_3), 3.30-3.27 (m, 1H, -CH), 2.95 (d, 1H, *J*= 12.97 Hz, -CH), 2.88 (s, 3H, -CH_3), 2.86 (s, 3H, -CH_3), 2.14-2.09 (m, 1H, -CH_2), 2.04-1.96 (m, 1H, -CH_2), 1.65-1.60 (m, 1H, -CH_2), 1.33-1.30 (m, 2H, -CH_2), 0.73-0.64 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.84, 167.2, 159.0, 151.1, 149.7, 131.4, 131.0, 126.9, 126.5, 117.8, 117.1, 114.1, 113.3, 82.1, 61.0, 55.4, 55.1, 32.4, 28.2, 28.1, 27.5, 24.9, 21.6; LCMS (ESI) *m/z* calcd. for $C_{23}H_{24}N_4O_4$: 420.18 [M^+]; found: 421.1874 [M+H]$^+$.

3-amino-1',3'-dimethyl-2',4',6'-trioxo-1-(thiophen-2-yl)-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{20}H_{20}N_4O_3S$, 4i)



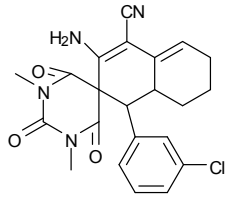
Pale pink color; Yield: 87%; M.p. (°C): 236-239; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2210, 1667, 1449, 1382; ^1H NMR (400 MHz, DMSO-d_6) δ_H: 7.42 (s, 1H, Ar-H), 6.90-6.58 (m, 1H, Ar-H), 6.68-6.67 (m, 1H, Ar-H), 6.50 (s, 2H, -NH_2), 5.55 (s, 1H, vinylic), 2.94 (s, 3H, -CH_3), 2.91 (s, 3H, -CH_3), 2.86-2.75 (m,1H, -CH), 2.48 (d, 1H, *J*= 3.81 Hz, -CH), 2.15-2.09 (m, 1H, -CH_2), 2.03-2.01 (m, 1H, -CH_2), 1.65-1.63 (m, 1H, -CH_2), 1.44-1.42 (m, 1H, -CH_2), 1.35-1.31 (m, 1H, -CH_2), 0.83-0.74 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.7, 167.1, 166.1, 150.6, 150.1, 149.8, 130.4, 130.0, 125.9, 124.7, 117.6, 82.0, 60.6, 59.3, 36.9, 34.3, 28.4, 27.3, 24.9, 21.5; LCMS (ESI) *m/z* calcd. for $C_{20}H_{20}N_4O_3S$: 396.1248 [M^+]; found: 397.1325 [M+H]$^+$.

3-amino-1-(4-(dimethylamino)phenyl)-1',3'-dimethyl-2',4',6'-trioxo-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{24}H_{27}N_5O_3$, 4j)



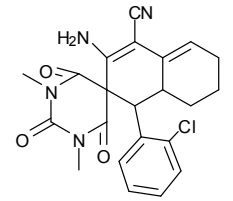
Crime color; Yield: 89%; M.p. (°C): 272-274; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2210, 1667, 1419, 1361; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 6.67-6.57 (m, 3H, Ar-H), 6.50-7.47 (d, 1H, *J*= 8.39 Hz, Ar-H), 6.44 (s, 2H, -NH_2), 5.54 (s, 1H, vinylic), 3.34 (m, 1H, -CH), 3.29-3.25 (m, 1H, -CH), 2.88 (s, 3H, -CH_3), 2.86 (s, 3H, -CH_3), 2.80 (s, 6H, -$N(CH_3)_2$), 2.13-2.09 (m, 1H, -CH_2), 2.02-2.00 (m, 1H, -CH_2), 1.60 (bs, 1H, -CH_2), 1.35-1.33 (m, 2H, -CH_2), 0.70-0.62 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 169.4, 167.7, 151.7, 150.4, 150.2, 131.7, 131.2, 126.6, 121.7, 118.3, 117.3, 112.4, 111.7, 82.5, 61.6, 56.0, 32.9, 28.6, 28.0, 25.4, 22.1; LCMS (ESI) *m/z* calcd. for $C_{24}H_{27}N_5O_3$: 433.2132 [M^+]; found: 434.2206 [M+H]$^+$.

3-amino-1-(3-chlorophenyl)-1',3'-dimethyl-2',4',6'-trioxo-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{22}H_{21}ClN_4O_3$, 4k)



White solid; Yield: 82%; M.p. (°C): 275-277; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2195, 1676, 1441, 1382; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 7.38-7.27 (m, 2H, Ar-H), 6.94 (s, 1H, Ar-H), 6.90-6.84 (m, 1H, Ar-H), 6.53 (s, 2H, -NH_2), 5.60 (s, 1H, vinylic), 3.36 (m, 1H, -CH), 3.09 (d, 1H, *J*= 12.21 Hz, -CH), 2.17-2.13 (m, 1H, -CH_2), 2.05-2.02 (m, 1H, -CH_2), 1.67-1.65 (m, 1H, -CH_2), 1.42-1.32 (m, 2H, -CH_2), 0.85-0.73 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.4, 167.0, 150.7, 149.6, 137.4, 133.3, 132.9, 130.0, 128.9, 128.6, 128.3, 117.7, 117.4, 82.1, 60.6, 55.3, 32.0, 28.1, 28.0, 27.4, 24.9, 21.5; LCMS (ESI) *m/z* calcd. for $C_{22}H_{21}ClN_4O_3$: 424.129 [M^+]; found: 425.1363 [M+H]$^+$.

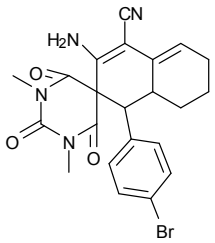
3-amino-1-(2-chlorophenyl)-1',3'-dimethyl-2',4',6'-trioxo-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{22}H_{21}ClN_4O_3$, 4l)



White solid; Yield: 93%; M.p. (°C): 242-244; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2202, 1667, 1426, 1376, 746; ^{1}H NMR (400MHz, DMSO-d_6) δ_H: 7.40 (d, 1H, *J*= 7.63 Hz, Ar-H), 7.33-7.24 (m, 2H, Ar-

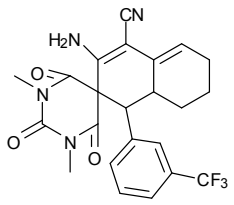
H), 6.97 (d, 1H, *J*= 7.63 Hz, Ar-H), 6.53 (s, 2H, -NH_2), 5.58 (s, 1H, vinylic), 3.66 (d, 1H, *J*= 12.21 Hz, -CH), 3.302-3.00 (m, 1H, -CH), 2.90 (s, 3H, -CH_3), 2.88 (s, 3H, -CH_3), 2.13-2.08 (m, 1H, -CH_2), 2.01-1.97 (m, 1H, -CH_2), 1.61-1.58 (m, 1H, -CH_2), 1.35-1.28 (m, 1H, -CH_2), 1.19-1.16 (m, 1H, -CH_2), 0.68-0.60 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 167.7, 167.1, 151.2, 149.9, 134.5, 132.5, 130.0, 129.7, 128.8, 127.1, 117.8, 117.5, 82.0, 60.0, 50.1, 33.2, 30.8, 28.8, 28.2, 27.4, 24.8, 21.5; LCMS (ESI) *m/z* calcd. for $C_{22}H_{21}ClN_4O_3$: 424.1298 [M^+]; found: 425.1369 [M+H]$^+$.

3-amino-1-(4-bromophenyl)-1',3'-dimethyl-2',4',6'-trioxo-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{22}H_{21}BrN_4O_3$, 4m)



White solid; Yield: 90%; M.p. (^{o}C): 261-263; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2195, 1676, 1382; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 7.50 (d, 1H, *J*= 8.39 Hz, Ar-H), 7.43 (d, 1H, *J*= 7.63 Hz, Ar-H), 6.84 (d, 1H, *J*= 8.39 Hz, Ar-H), 6.79 (d, 1H, *J*= 9.92 Hz, Ar-H), 6.49 (s, 2H, -NH_2), 5.57 (s, 1H, vinylic), 3.37 (m, 1H, -CH), 3.03 (d, 1H, *J*= 12.21 Hz, -CH), 2.14-2.10 (m, 1H, -CH_2), 2.02-1.96 (m, 1H, -CH_2), 1.63-1.60 (m, 1H, -CH_2), 1.39-1.25 (m, 2H, -CH_2), 0.76-0.67 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.5, 166.9, 150.9, 149.7, 134.3, 132.5, 131.4, 131.3, 130.6, 128.1, 121.5, 117.8, 117.5, 82.1, 60.6, 55.3, 32.2, 28.2, 24.9, 21.6; LCMS (ESI) *m/z* calcd. for $C_{22}H_{21}BrN_4O_3$: 468.0792 [M^+]; found: 469.0863 [M+H]$^+$, 471.0853 [M+(H+2)]$^+$.

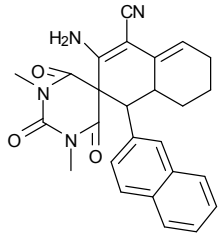
3-amino-1',3'-dimethyl-2',4',6'-trioxo-1-(3-(trifluoromethyl)phenyl)-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{23}H_{21}F_3N_4O_3$, 4n)



White solid; Yield: 85%; M.p. (^{o}C): 244-246; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2202, 1676, 1434, 1376, 753; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 7.68-7.66 (m, 1H, Ar-H), 7.55 (td, 1H, *J*= 7.63 Hz,

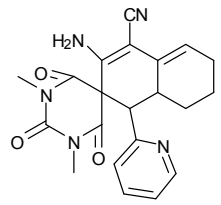
Ar-H), 7.26 (d, 1H, *J*= 7.63 Hz, Ar-H), 7.22-7.17 (t, 1H, *J*= 9.92 Hz, Ar-H), 6.55 (s, 2H, -NH_2), 5.61 (s, 1H, vinylic), 3.25-3.20 (m, 1H, -CH), 3.00 (bs, 1H, -CH), 2.88-2.83 (m, 6H, 2(CH_3)), 2.17-2.13 (m, 1H, -CH_2), 2.06-2.00 (m, 1H, -CH_2), 1.66-1.63 (m, 1H, -CH_2), 1.44-1.40 (m, 1H, -CH_2), 1.33-1.24 (m, 1H, -CH_2), 0.88-0.73 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.3, 166.7, 150.6, 149.5, 136.4, 134.6, 130.4, 129.4, 125.2, 117.7, 117.5, 82.2, 60.7, 55.3, 31.9, 28.2, 28.1, 27.2, 24.8, 21.5; LCMS (ESI) *m/z* calcd. for $C_{23}H_{21}F_3N_4O_3$: 458.1565 [M^+]; found: 459.1640 [M+H]$^+$.

3-amino-1',3'-dimethyl-1-(naphthalen-2-yl)-2',4',6'-trioxo-6,7,8,8a-tetrahydro-1*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{26}H_{24}N_4O_3$, 4o)



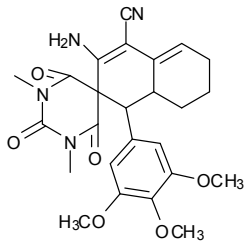
White solid; Yield: 88%; M.p. (^{o}C): 267-269; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2210, 1676, 1449, 1367; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 7.93-7.90 (m,1H, Ar-H), 7.85 (d, 1H, *J*= 8.39 Hz, Ar-H), 7.78 (d, 1H, *J*= 7.63 Hz, Ar-H), 7.52-7.45 (m, 3H, Ar-H), 7.14 (d, 1H, *J*= 6.87 Hz, Ar-H), 6.53 (s. 2H, -NH_2), 5.65 (s, 1H, vinylic), 4.04 (d, 1H, *J*= 12.21 Hz, -CH), 3.10-3.04 (m, 1H, -CH), 2.88 (s, 3H, -CH_3), 2.18-2.15 (m, 1H, -CH_2), 2.14 (s, 3H, -CH_3), 2.07-199 (m, 1H, -CH_2), 1.59-1.56 (m, 1H, -CH_2), 1.37-1.35 (m, 2H, -CH_2), 0.77-0.68 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.3, 167.3, 151.3, 149.3, 133.1, 131.0, 128.9, 128.5, 126.4, 125.7, 124.8, 124.5, 121.9, 117.8, 117.4, 82.2, 60.7, 48.2, 33.4, 28.0, 27.6, 27.0, 24.9, 21.5; LCMS (ESI) *m/z* calcd. for $C_{26}H_{24}N_4O_3$: 440.1851 [M^+]; found: 441.1923 [M+H]$^+$.

3-amino-1',3'-dimethyl-2',4',6'-trioxo-1-(pyridin-2-yl)-2',3',4',6,6',7,8,8a-octahydro-1*H*,1'*H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{21}H_{21}N_5O_3$, 4p)



White solid; Yield: 87%; M.p. (°C): 253-255; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2187, 1682, 1441, 1367; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 8.48 (s, 1H, Ar-H), 7.70 (s, 1H, Ar-H), 7.27 (s, 1H, Ar-H), 7.04 (s, 1H, Ar-H), 6.50 (s, 2H, -NH_2), 5.56 (s, 1H, vinylic), 3.35 (m, 2H, -CH), 2.96 (s, 3H, -CH_3), 2.78 (s, 3H, -CH_3), 2.10 (s, 2H, -CH_2), 1.62 (s, 1H, -CH_2), 1.32 (s, 1H, -CH_2), 1.04 (s, 1H, -CH_2), 0.84 (s, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.6, 166.4, 155.3, 151.6, 149.9, 149.0, 136.8, 131.2, 124.4, 123.3, 117.8, 116.7, 81.9, 59.9, 57.5, 32.62, 28.3, 28.0, 26.9, 24.8, 21.6; LCMS (ESI) *m/z* calcd. for $C_{21}H_{21}N_5O_3$: 391.1635 [M^+]; found: 392.1713 [M+H]$^+$.

3-amino-1',3'-dimethyl-2',4',6'-trioxo-1-(3,4,5-trimethoxyphenyl)-2',3',4',6,6',7,8,8a-octahydro-1*H*,*1'H*-spiro[naphthalene-2,5'-pyrimidine]-4-carbonitrile ($C_{25}H_{28}N_4O_6$, 4q)



White solid; Yield: 92%; M.p. (°C): 286-288; IR ($CHCl_3$) ν_{max}/cm^{-1}: 2180, 1682, 1434, 1376; ^{1}H NMR (400 MHz, DMSO-d_6) δ_H: 6.47 (s, 2H, Ar-H), 6.13 (s, 2H, -NH_2), 5.57 (s, 1H, vinylic), 3.66 (s, 3H, -OCH_3), 3.64 (s, 3H, -OCH_3), 3.56 (s, 3H, -OCH_3), 3.35 (m, 1H, -CH), 2.97-2.95 (m, 1H, -CH), 2.91 (s, 3H, -CH_3), 2.90 (s, 3H, -CH_3), 2.15-2.11 (m, 1H, -CH_2), 2.05-1.98 (m, 1H, -CH_2), 1.65-1.62 (m, 1H, -CH_2), 1.46-1.39 (m, 2H, -CH_2), 0.79-0.71 (m, 1H, -CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ_C: 168.8, 167.1, 153.0, 152.4, 151.1, 149.8, 137.2, 130.8, 130.6, 117.8, 117.3, 107.8, 102.6, 82.1, 60.84, 60.1, 56.1, 56.0, 55.8, 32.3, 30.7, 28.3, 27.3, 24.9, 21.5; LCMS (ESI) *m/z* calcd. for $C_{25}H_{28}N_4O_6$: 480.2004 [M^+]; found: 481.2077 [M+H]$^+$.

Figure 1: ^{1}H NMR spectrum of compound **4a** in DMSO.

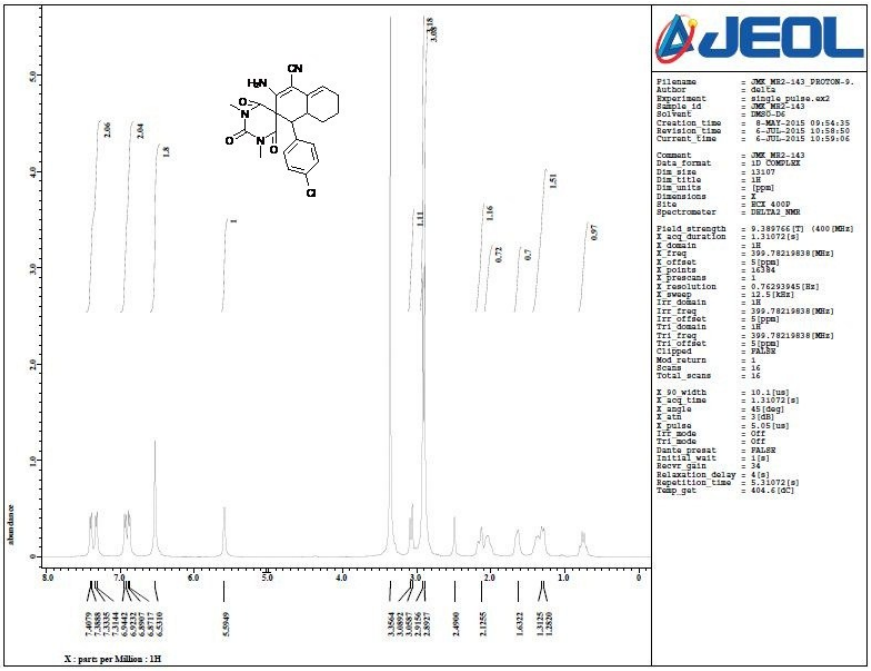


Figure 2: ^{13}C NMR spectrum of compound **4a** in DMSO.

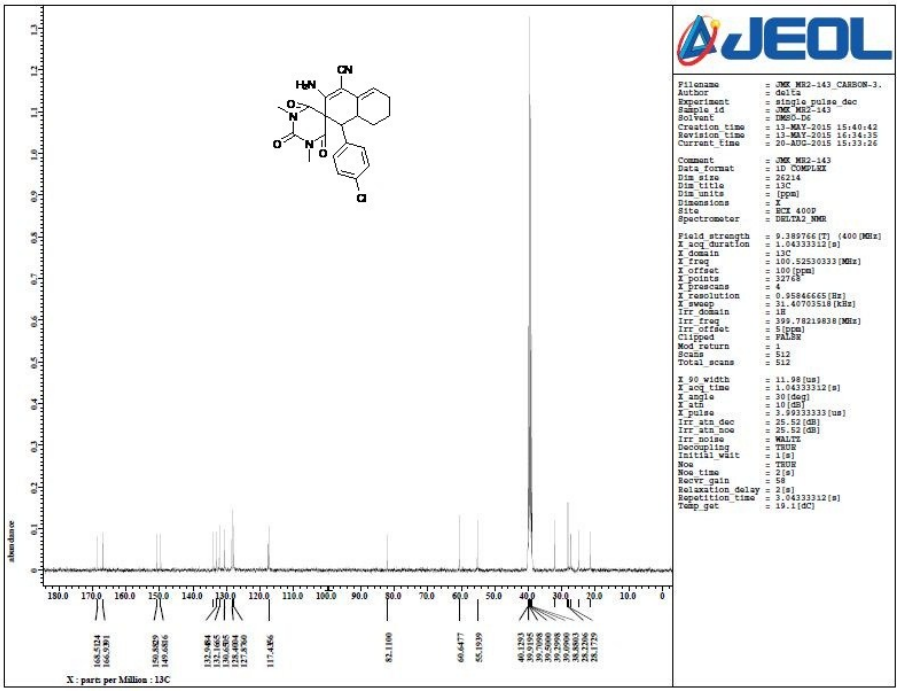


Figure 3: DEPT spectrum of compound **4a**.

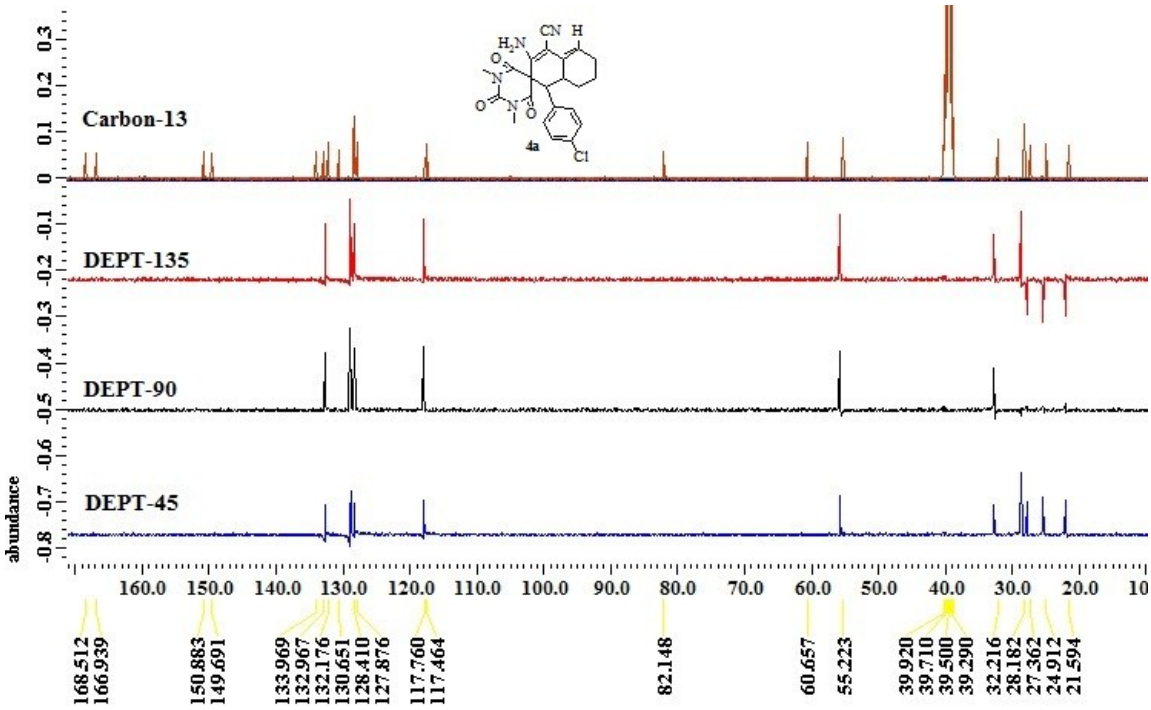


Figure 4: Mass spectrum of compound **4a**.

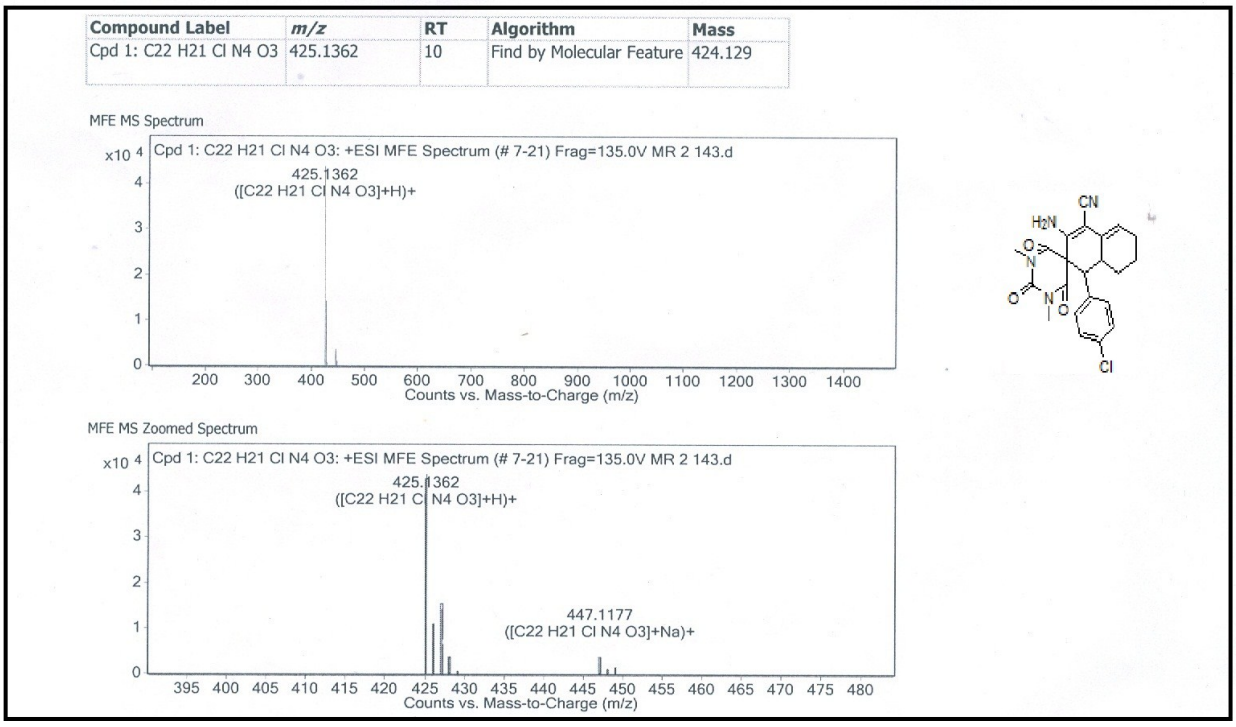


Figure 5: ^{1}H NMR spectrum of compound **4b** in DMSO.

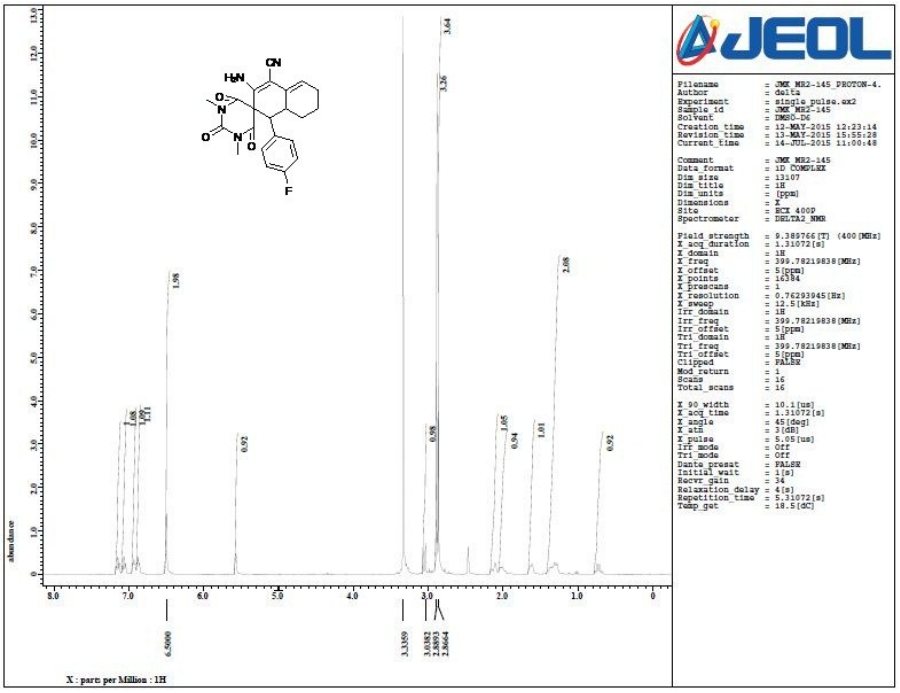


Figure 6: ^{13}C NMR spectrum of compound **4b** in DMSO.

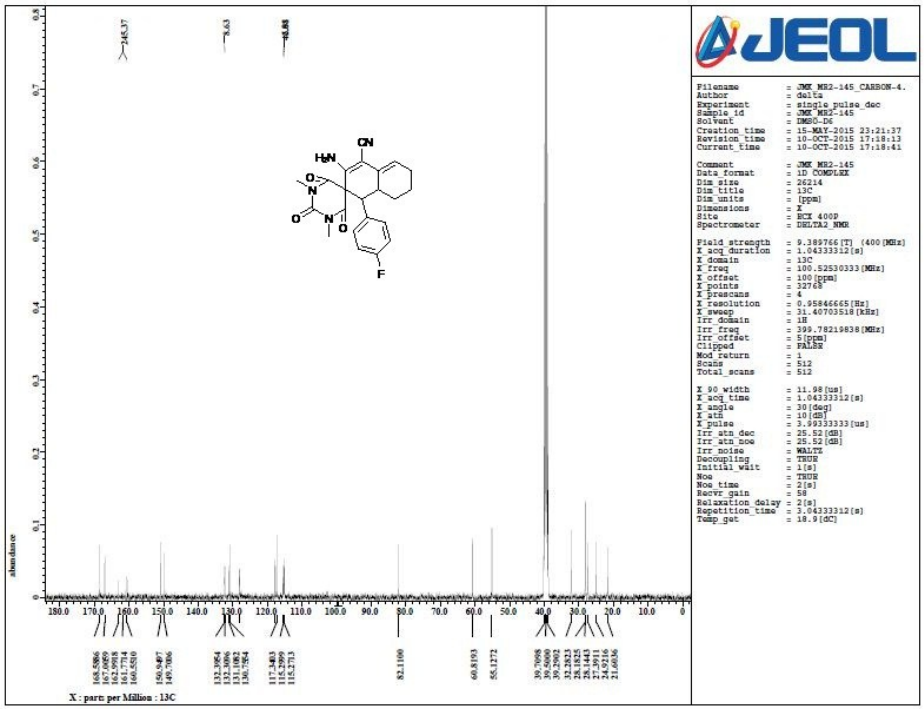
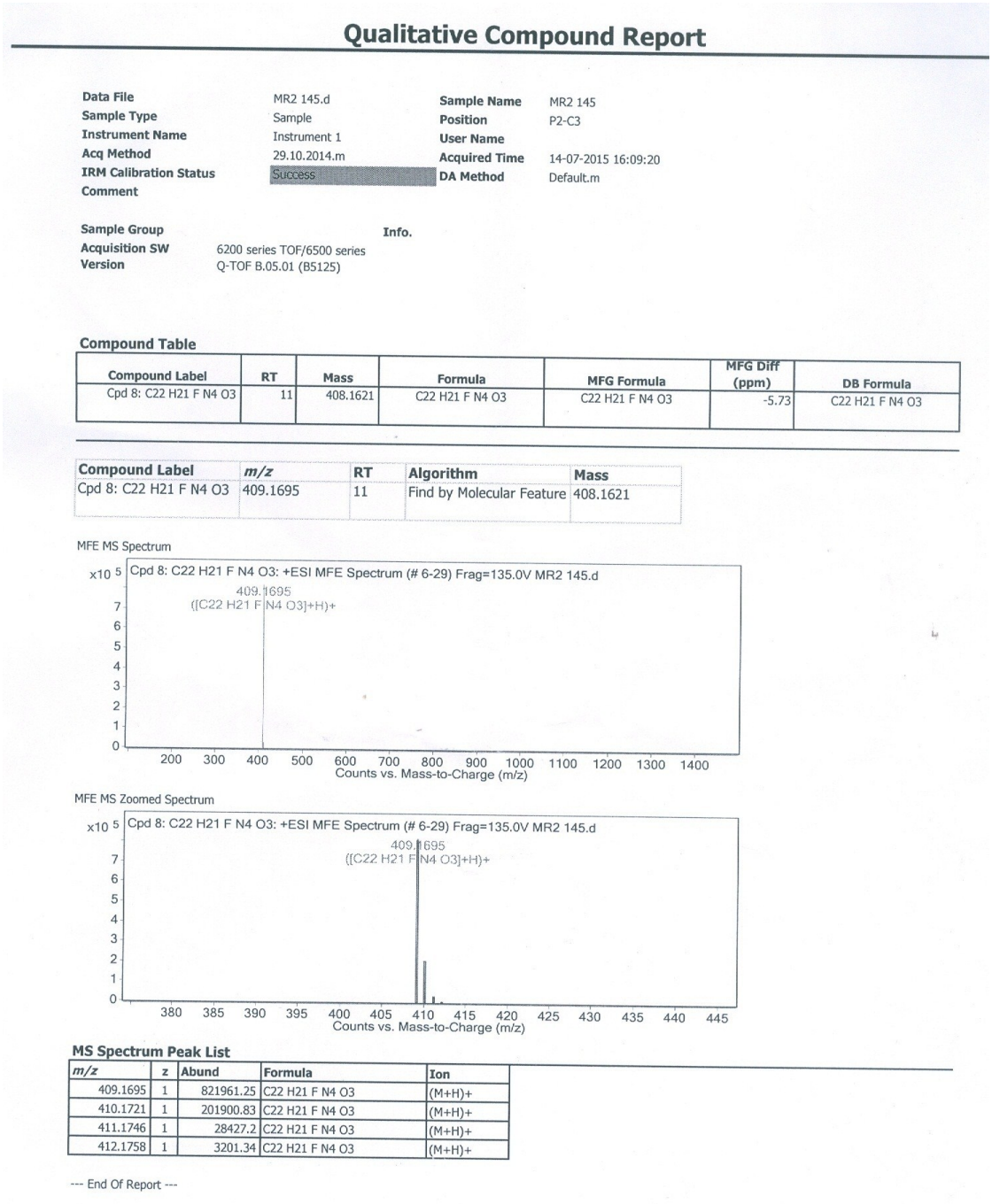


Figure 7: Mass spectrum of compound **4b**.



Qualitative Compound Report

Data File	MR2 145.d	**Sample Name**	MR2 145
Sample Type	Sample	**Position**	P2-C3
Instrument Name	Instrument 1	**User Name**	
Acq Method	29.10.2014.m	**Acquired Time**	14-07-2015 16:09:20
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

Sample Group	**Info.**
Acquisition SW Version	6200 series TOF/6500 series Q-TOF B.05.01 (B5125)

Compound Table

Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 8: C22 H21 F N4 O3	11	408.1621	C22 H21 F N4 O3	C22 H21 F N4 O3	-5.73	C22 H21 F N4 O3

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 8: C22 H21 F N4 O3	409.1695	11	Find by Molecular Feature	408.1621

MFE MS Spectrum

MFE MS Zoomed Spectrum

MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
409.1695	1	821961.25	C22 H21 F N4 O3	(M+H)+
410.1721	1	201900.83	C22 H21 F N4 O3	(M+H)+
411.1746	1	28427.2	C22 H21 F N4 O3	(M+H)+
412.1758	1	3201.34	C22 H21 F N4 O3	(M+H)+

--- End Of Report ---

Figure 8: ^{1}H NMR spectrum of compound **4c** in DMSO.

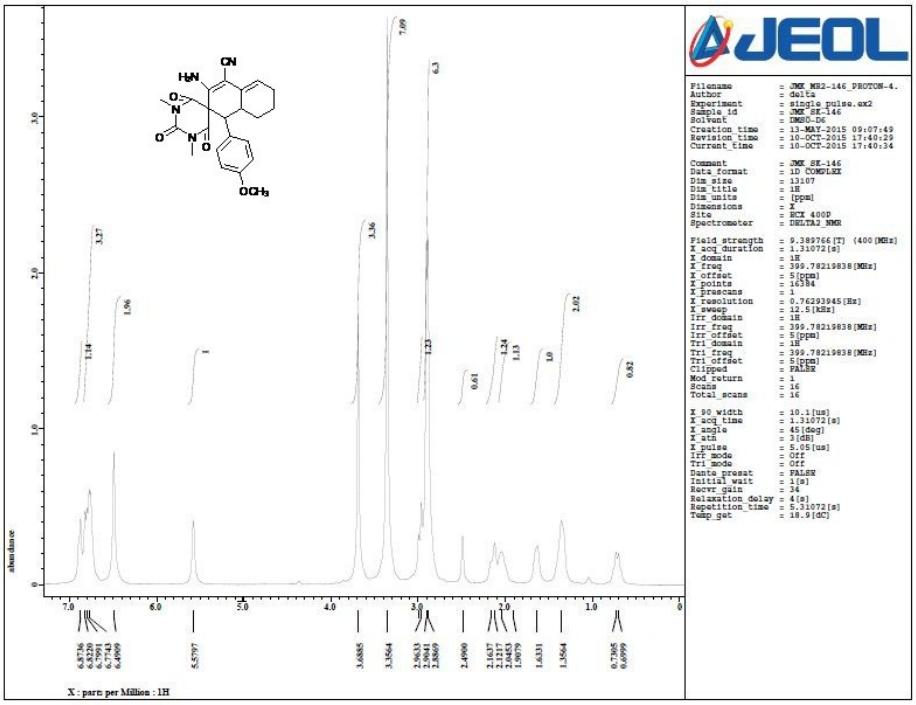


Figure 9: ^{13}C NMR spectrum of compound **4c** in DMSO.

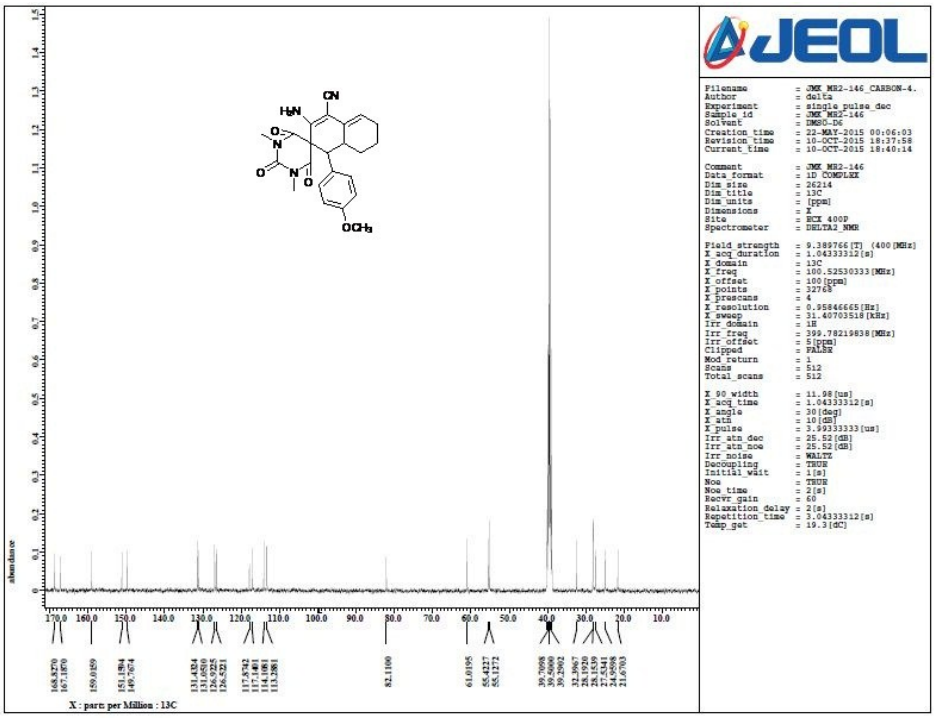
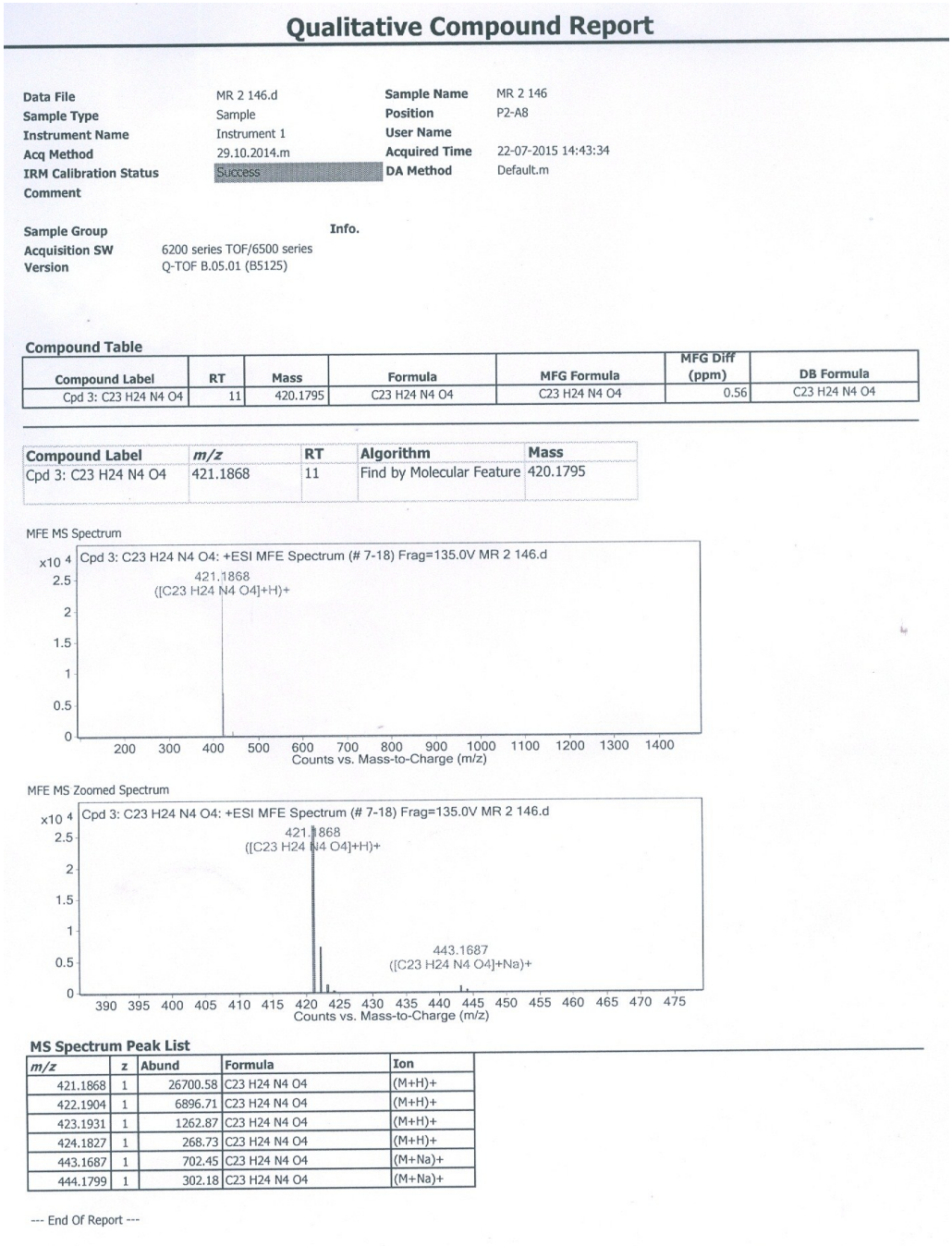


Figure 10: Mass spectrum of compound **4c**.



Qualitative Compound Report

Data File	MR 2 146.d	**Sample Name**	MR 2 146
Sample Type	Sample	**Position**	P2-A8
Instrument Name	Instrument 1	**User Name**	
Acq Method	29.10.2014.m	**Acquired Time**	22-07-2015 14:43:34
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

Sample Group **Info.**
Acquisition SW 6200 series TOF/6500 series
Version Q-TOF B.05.01 (B5125)

Compound Table

Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 3: C23 H24 N4 O4	11	420.1795	C23 H24 N4 O4	C23 H24 N4 O4	0.56	C23 H24 N4 O4

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 3: C23 H24 N4 O4	421.1868	11	Find by Molecular Feature	420.1795

MFE MS Spectrum

MFE MS Zoomed Spectrum

MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
421.1868	1	26700.58	C23 H24 N4 O4	(M+H)+
422.1904	1	6896.71	C23 H24 N4 O4	(M+H)+
423.1931	1	1262.87	C23 H24 N4 O4	(M+H)+
424.1827	1	268.73	C23 H24 N4 O4	(M+H)+
443.1687	1	702.45	C23 H24 N4 O4	(M+Na)+
444.1799	1	302.18	C23 H24 N4 O4	(M+Na)+

--- End Of Report ---

Figure 11: ^{1}H NMR spectrum of compound **4d** in DMSO.

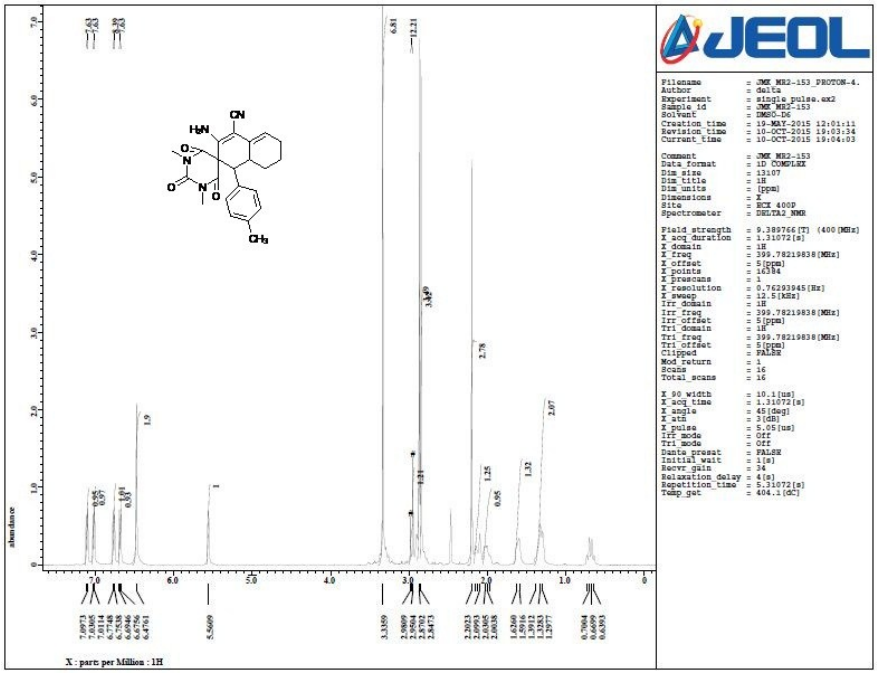


Figure 12: ^{13}C NMR spectrum of compound **4d** in DMSO.

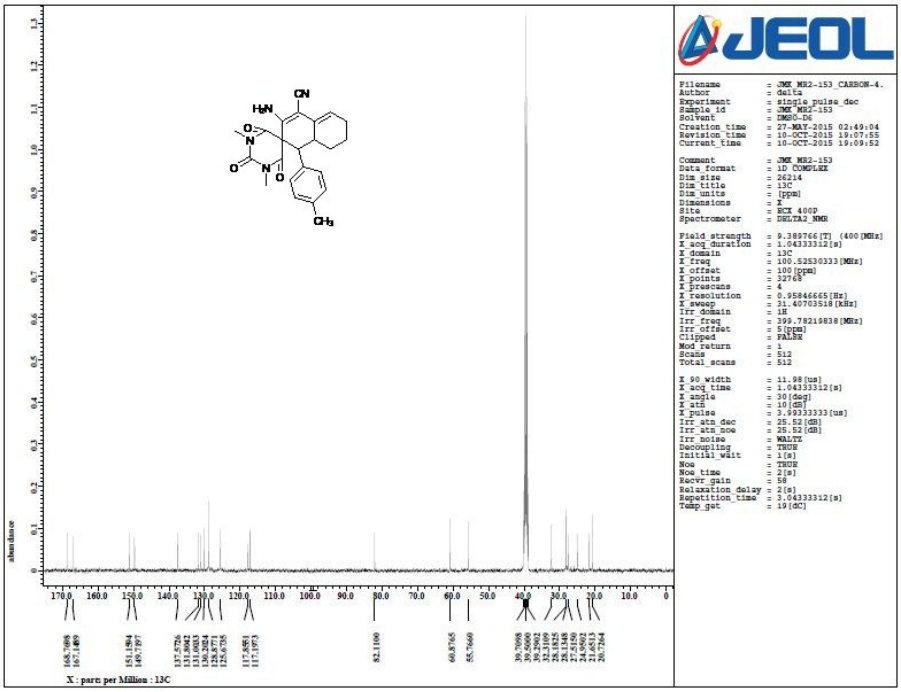
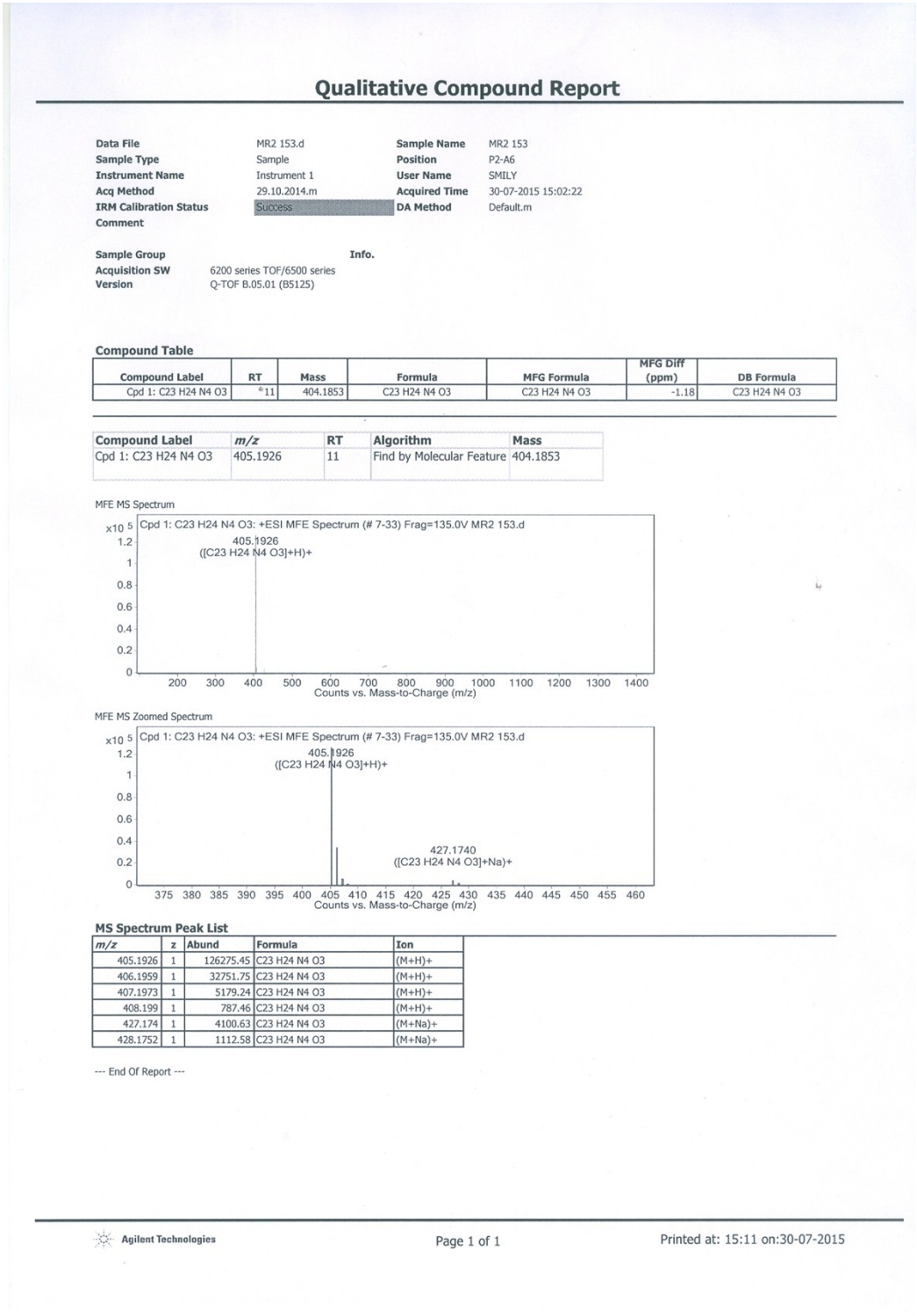


Figure 13: Mass spectrum of compound **4d**.



Qualitative Compound Report

Data File	MR2 153.d	**Sample Name**	MR2 153
Sample Type	Sample	**Position**	P2-A6
Instrument Name	Instrument 1	**User Name**	SMILY
Acq Method	29.10.2014.m	**Acquired Time**	30-07-2015 15:02:22
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

Sample Group	**Info.**
Acquisition SW Version	6200 series TOF/6500 series Q-TOF B.05.01 (B5125)

Compound Table

Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 1: C23 H24 N4 O3	*11	404.1853	C23 H24 N4 O3	C23 H24 N4 O3	-1.18	C23 H24 N4 O3

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 1: C23 H24 N4 O3	405.1926	11	Find by Molecular Feature	404.1853

MFE MS Spectrum

MFE MS Zoomed Spectrum

MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
405.1926	1	126275.45	C23 H24 N4 O3	(M+H)+
406.1959	1	32751.75	C23 H24 N4 O3	(M+H)+
407.1973	1	5179.24	C23 H24 N4 O3	(M+H)+
408.199	1	787.46	C23 H24 N4 O3	(M+H)+
427.174	1	4100.63	C23 H24 N4 O3	(M+Na)+
428.1752	1	1112.58	C23 H24 N4 O3	(M+Na)+

--- End Of Report ---

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Figure 14: [1]H NMR spectrum of compound **4e** in DMSO.

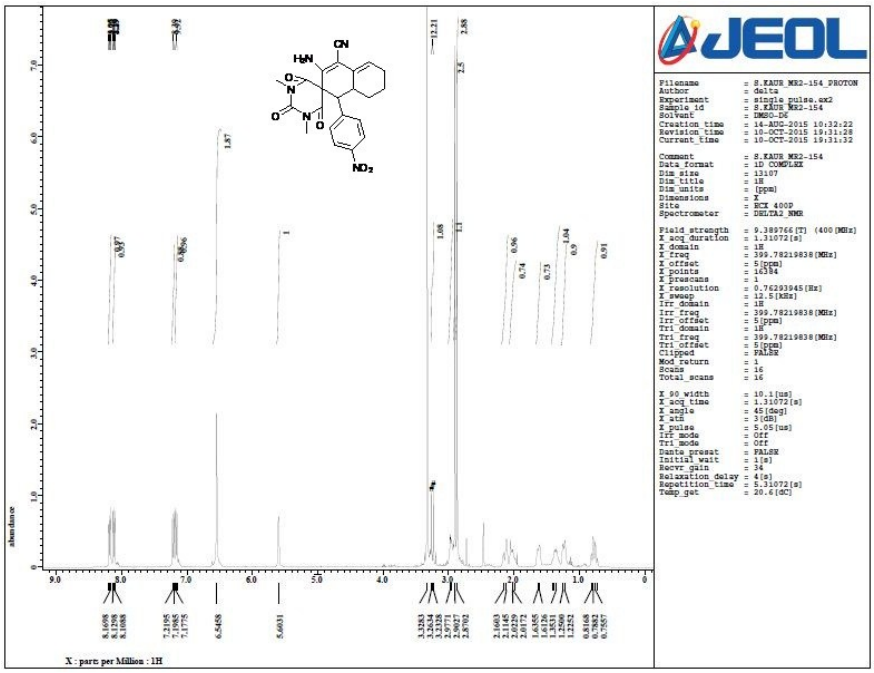


Figure 15: [13]C NMR spectrum of compound **4e** in DMSO.

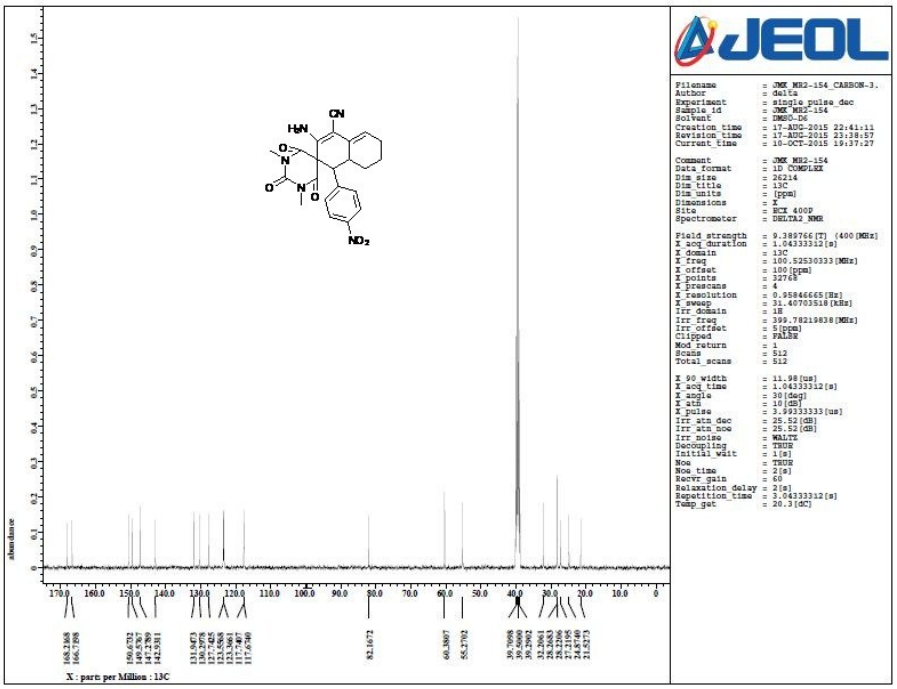


Figure 16: Mass spectrum of compound **4e**.

Qualitative Compound Report

Data File	MR2 154.d	**Sample Name**	MR2 154
Sample Type	Sample	**Position**	P1-B3
Instrument Name	Instrument 1	**User Name**	SMILY
Acq Method	29.10.2014.m	**Acquired Time**	17-08-2015 13:03:54
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

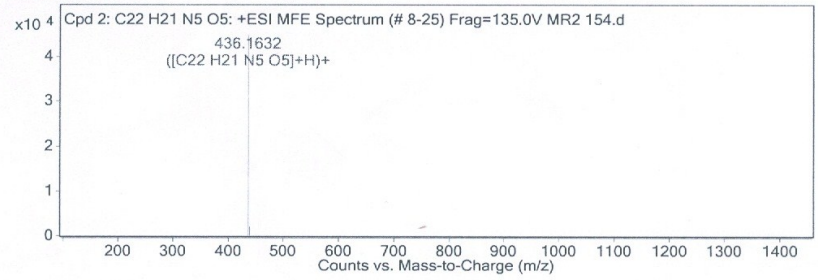
Sample Group	**Info.**
Acquisition SW Version	6200 series TOF/6500 series Q-TOF B.05.01 (B5125)

Compound Table

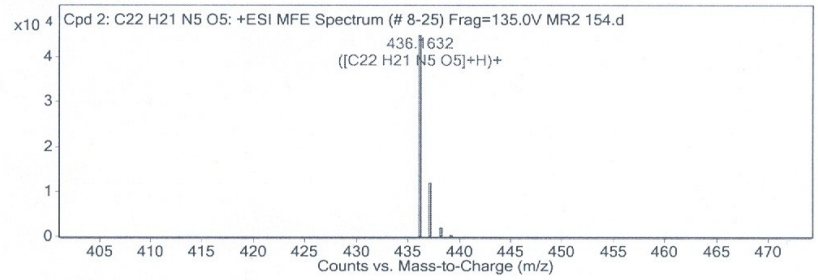
Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 2: C22 H21 N5 O5	11	435.1561	C22 H21 N5 O5	C22 H21 N5 O5	-4.28	C22 H21 N5 O5

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 2: C22 H21 N5 O5	436.1632	11	Find by Molecular Feature	435.1561

MFE MS Spectrum



MFE MS Zoomed Spectrum



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
436.1632	1	44900.22	C22 H21 N5 O5	(M+H)+
437.1671	1	11410.27	C22 H21 N5 O5	(M+H)+
438.1679	1	2152.93	C22 H21 N5 O5	(M+H)+
439.1764	1	287.69	C22 H21 N5 O5	(M+H)+

--- End Of Report ---

Figure 17: [1]H NMR spectrum of compound **4f** in DMSO.

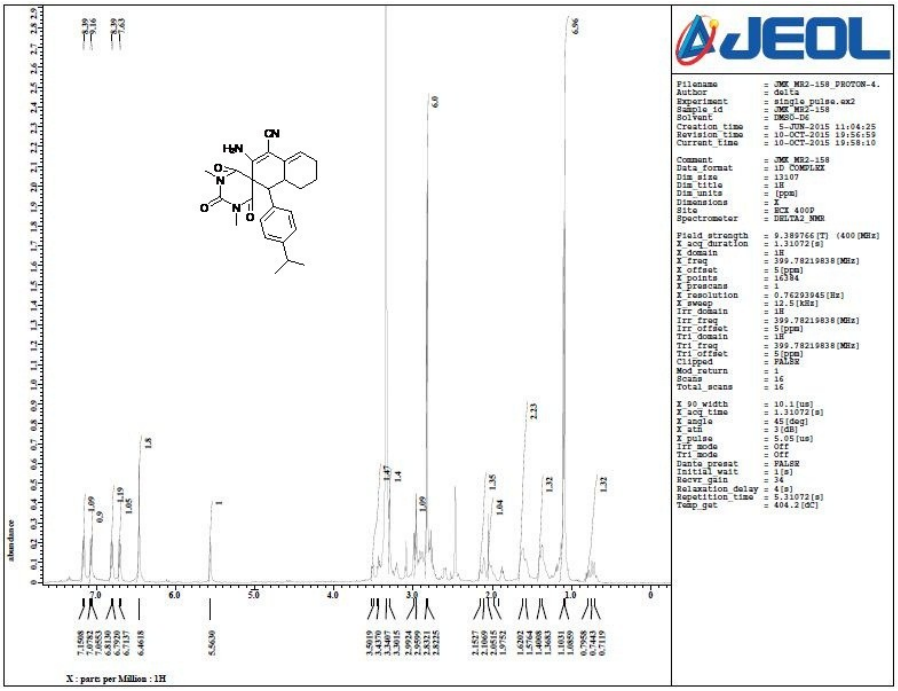


Figure 18: [13]C NMR spectrum of compound **4f** in DMSO.

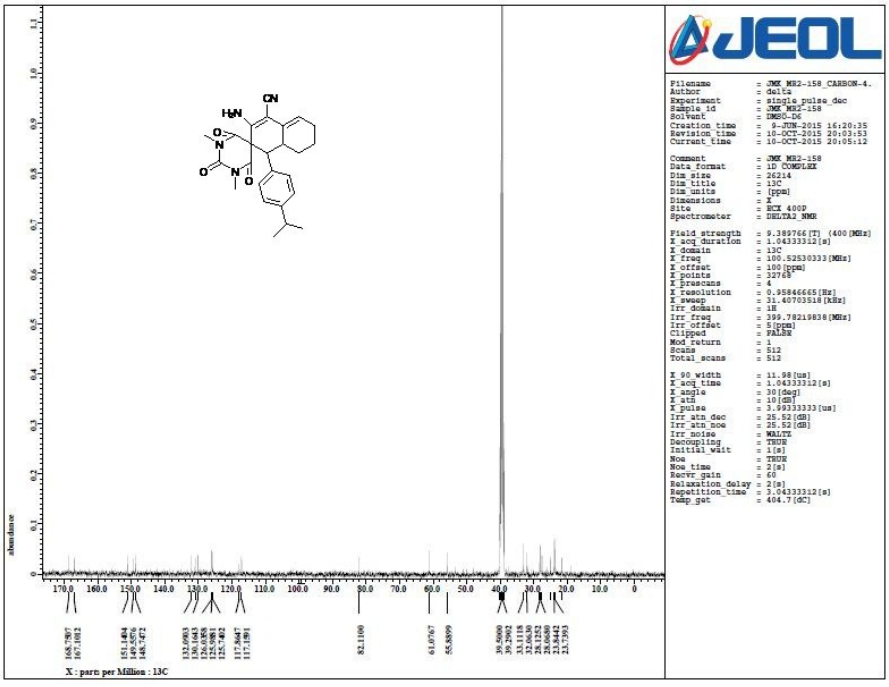


Figure 19: Mass spectrum of compound **4f**.

Qualitative Compound Report

Data File	MR2 158.d	**Sample Name**	MR2 158
Sample Type	Sample	**Position**	P1-D3
Instrument Name	Instrument 1	**User Name**	SMILY
Acq Method	29.10.2014.m	**Acquired Time**	10-08-2015 14:49:27
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

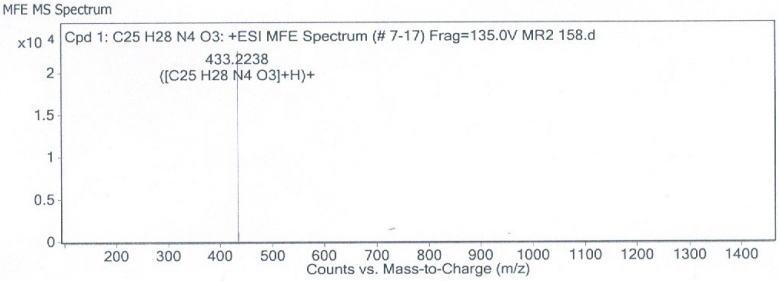
Sample Group	**Info.**
Acquisition SW Version	6200 series TOF/6500 series Q-TOF B.05.01 (B5125)

Compound Table

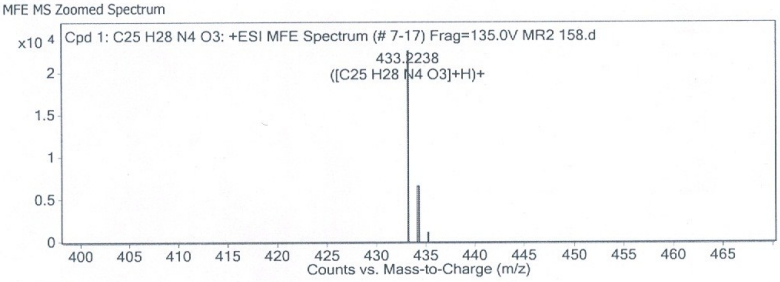
Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 1: C25 H28 N4 O3	11	432.2167	C25 H28 N4 O3	C25 H28 N4 O3	-1.3	C25 H28 N4 O3

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 1: C25 H28 N4 O3	433.2238	11	Find by Molecular Feature	432.2167

MFE MS Spectrum



MFE MS Zoomed Spectrum



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
433.2238	1	22621.44	C25 H28 N4 O3	(M+H)+
434.2277	1	6474.51	C25 H28 N4 O3	(M+H)+
435.2301	1	1109.76	C25 H28 N4 O3	(M+H)+

--- End Of Report ---

Figure 20: [1]H NMR spectrum of compound **4g** in DMSO.

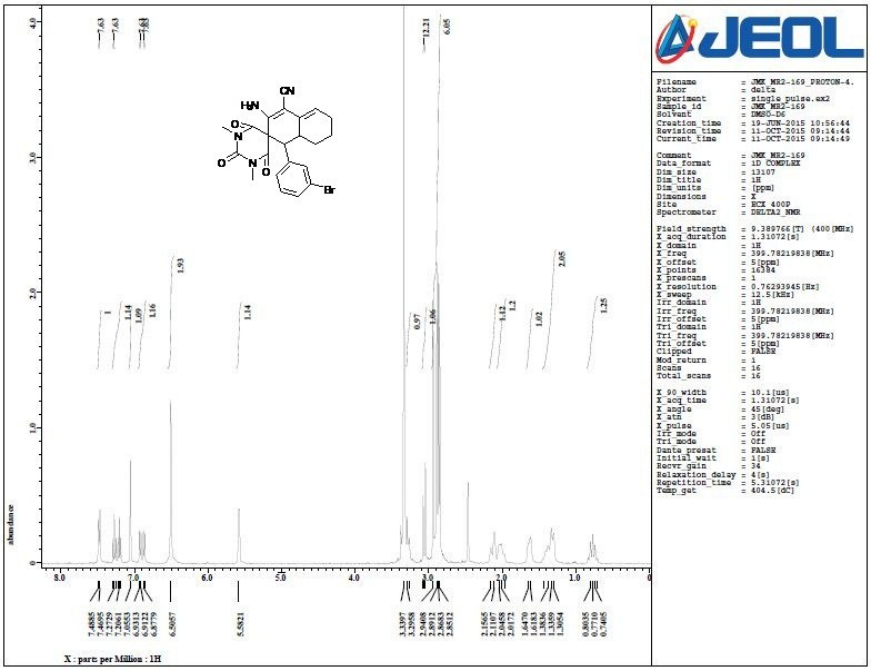


Figure 21: [13]C NMR spectrum of compound **4g** in DMSO.

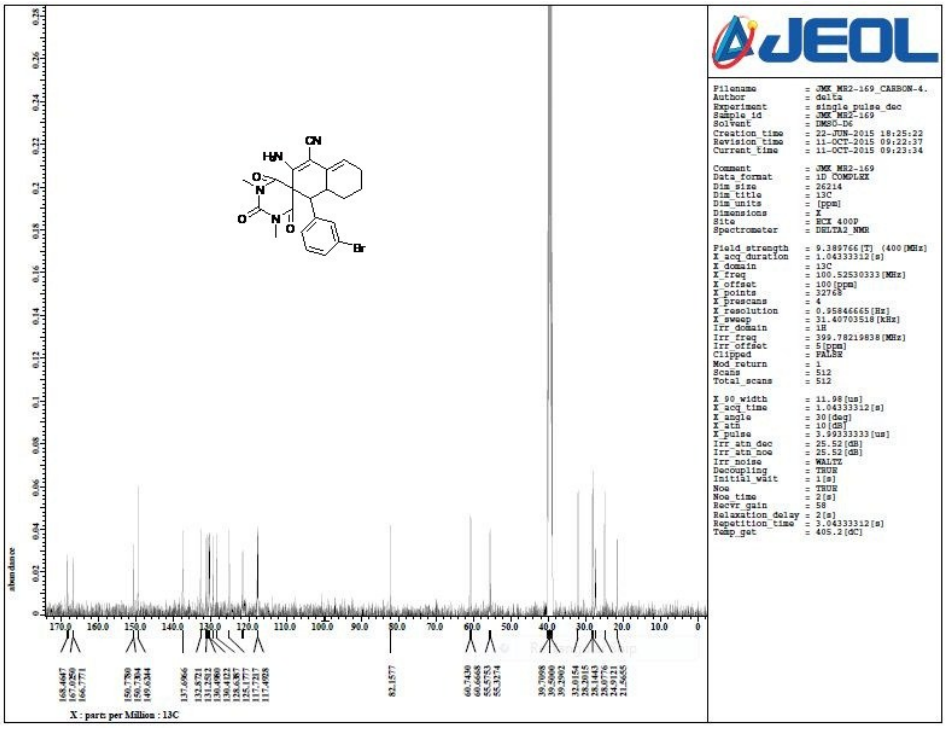
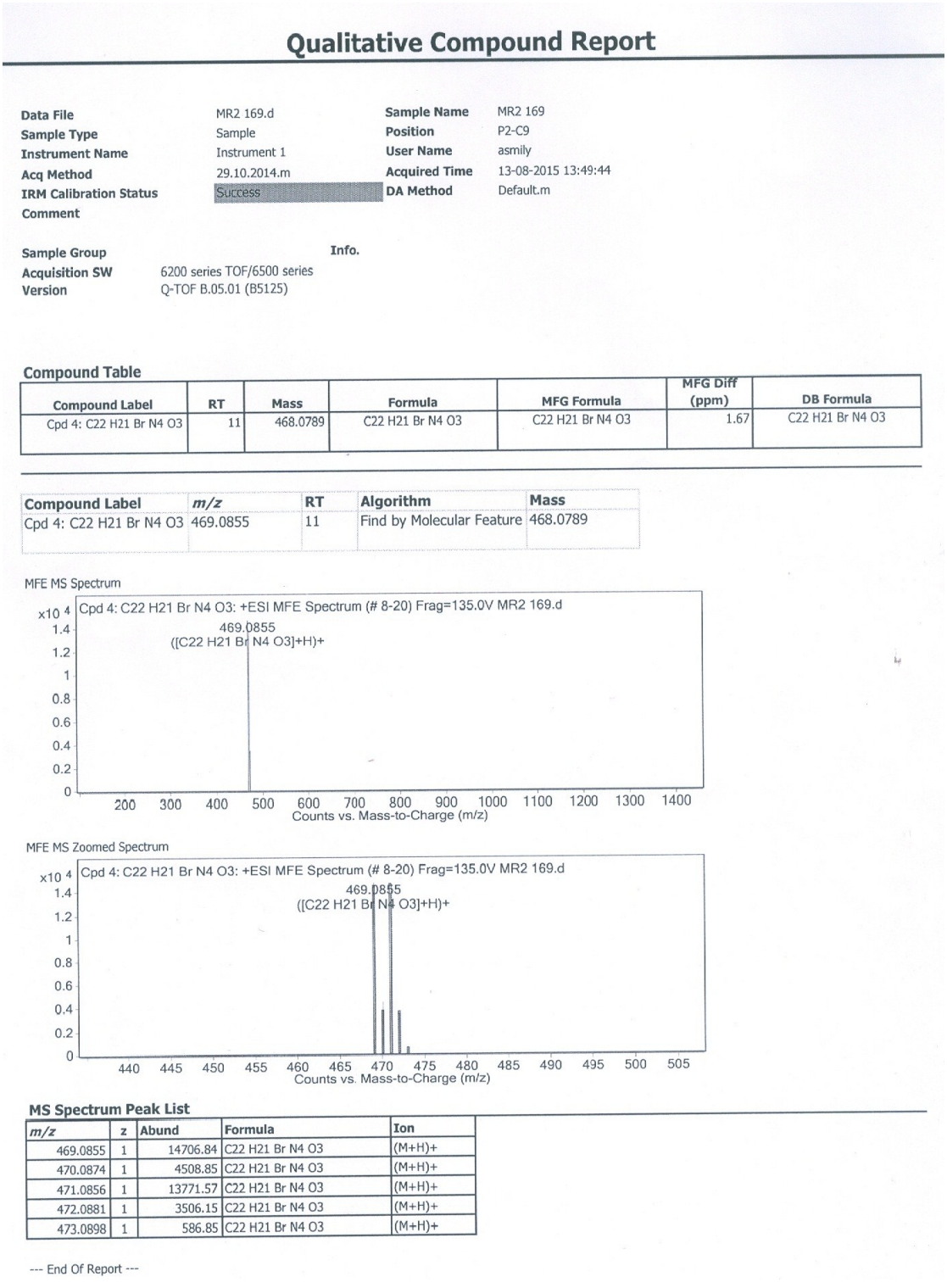


Figure 22: Mass spectrum of compound **4g**.



Qualitative Compound Report

Data File	MR2 169.d	**Sample Name**	MR2 169
Sample Type	Sample	**Position**	P2-C9
Instrument Name	Instrument 1	**User Name**	asmily
Acq Method	29.10.2014.m	**Acquired Time**	13-08-2015 13:49:44
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

Sample Group	**Info.**
Acquisition SW Version	6200 series TOF/6500 series Q-TOF B.05.01 (B5125)

Compound Table

Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 4: C22 H21 Br N4 O3	11	468.0789	C22 H21 Br N4 O3	C22 H21 Br N4 O3	1.67	C22 H21 Br N4 O3

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 4: C22 H21 Br N4 O3	469.0855	11	Find by Molecular Feature	468.0789

MFE MS Spectrum

MFE MS Zoomed Spectrum

MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
469.0855	1	14706.84	C22 H21 Br N4 O3	(M+H)+
470.0874	1	4508.85	C22 H21 Br N4 O3	(M+H)+
471.0856	1	13771.57	C22 H21 Br N4 O3	(M+H)+
472.0881	1	3506.15	C22 H21 Br N4 O3	(M+H)+
473.0898	1	586.85	C22 H21 Br N4 O3	(M+H)+

--- End Of Report ---

Figure 23: ^{1}H NMR spectrum of compound **4h** in DMSO.

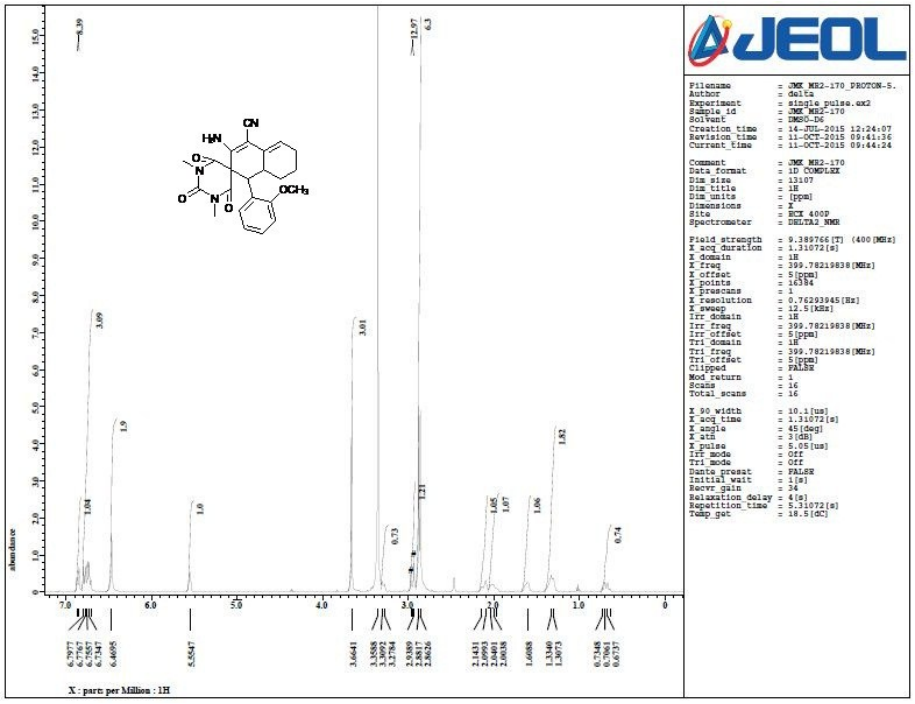


Figure 24: ^{13}C NMR spectrum of compound **4h** in DMSO.

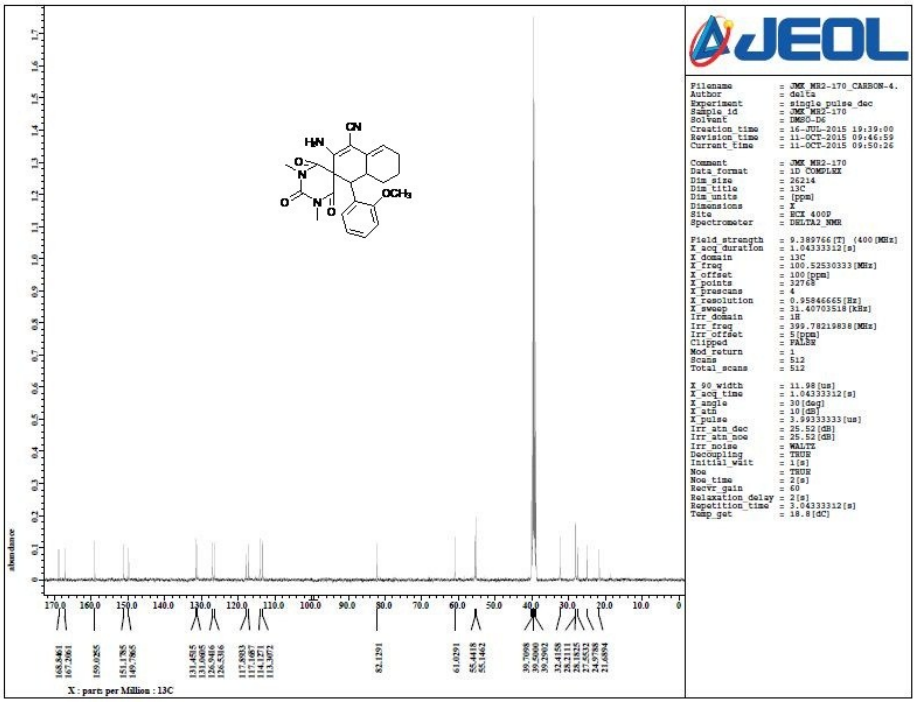


Figure 25: Mass spectrum of compound **4h**.

Qualitative Compound Report

Data File	MR2 170.d	**Sample Name**	MR2 170
Sample Type	Sample	**Position**	P1-B8
Instrument Name	Instrument 1	**User Name**	SMILY
Acq Method	29.10.2014.m	**Acquired Time**	18-08-2015 12:56:05
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

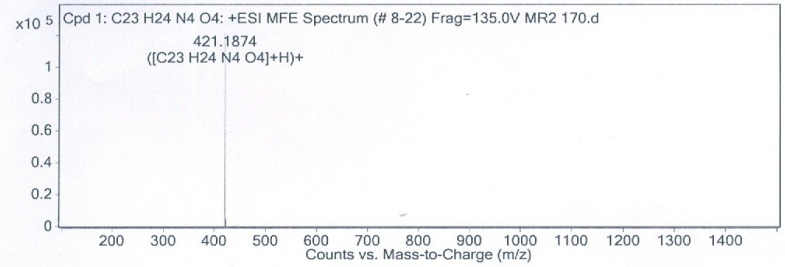
Sample Group	**Info.**
Acquisition SW Version	6200 series TOF/6500 series Q-TOF B.05.01 (B5125)

Compound Table

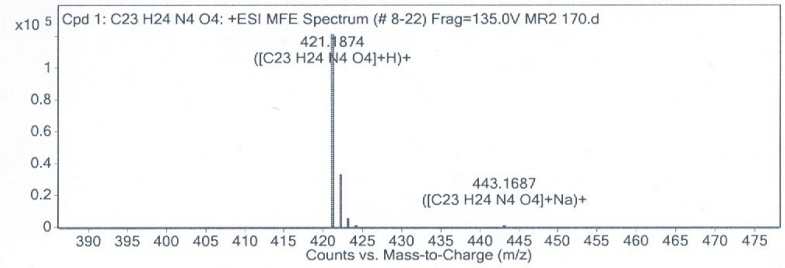
Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 1: C23 H24 N4 O4	11	420.18	C23 H24 N4 O4	C23 H24 N4 O4	-0.68	C23 H24 N4 O4

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 1: C23 H24 N4 O4	421.1874	11	Find by Molecular Feature	420.18

MFE MS Spectrum



MFE MS Zoomed Spectrum



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
421.1874	1	121135.19	C23 H24 N4 O4	(M+H)+
422.1903	1	31554.39	C23 H24 N4 O4	(M+H)+
423.1925	1	5050.15	C23 H24 N4 O4	(M+H)+
424.1935	1	652.22	C23 H24 N4 O4	(M+H)+
443.1687	1	687.06	C23 H24 N4 O4	(M+Na)+

--- End Of Report ---

Figure 26: [1]H NMR spectrum of compound **4i** in DMSO.

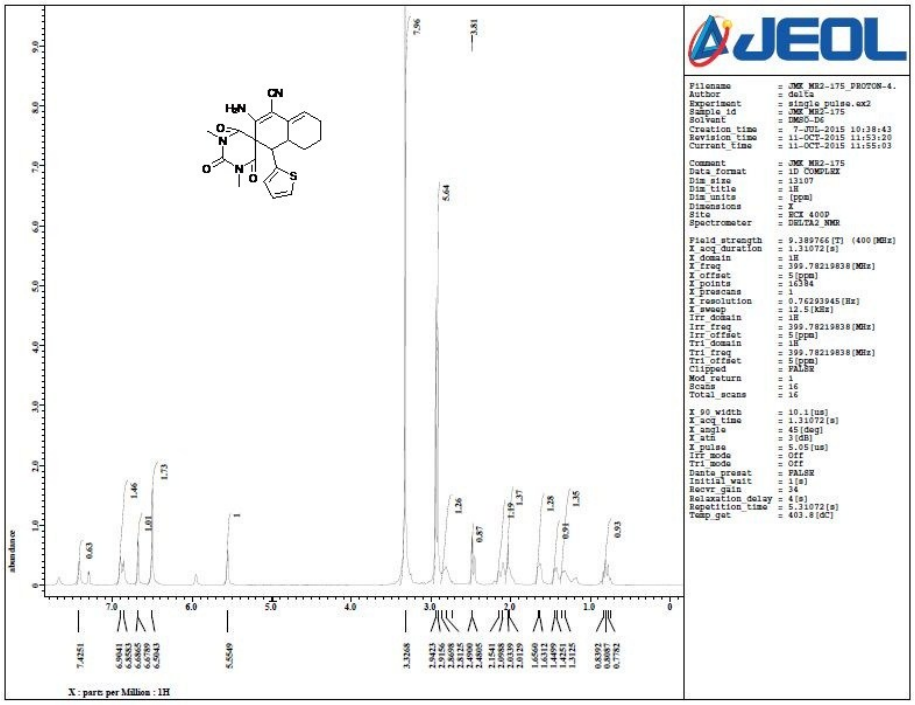


Figure 27: [13]C NMR spectrum of compound **4i** in DMSO.

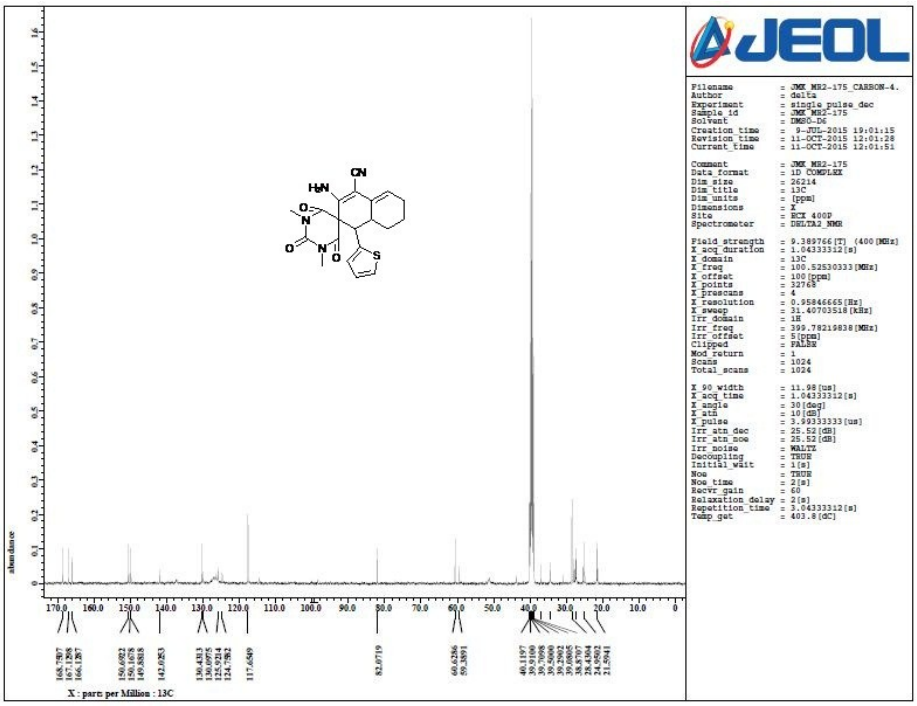


Figure 28: Mass spectrum of compound **4i**.

Qualitative Compound Report

Data File	MR2 175.d	**Sample Name**	MR2 175
Sample Type	Sample	**Position**	P1-D1
Instrument Name	Instrument 1	**User Name**	SMILY
Acq Method	29.10.2014.m	**Acquired Time**	18-08-2015 12:54:14
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

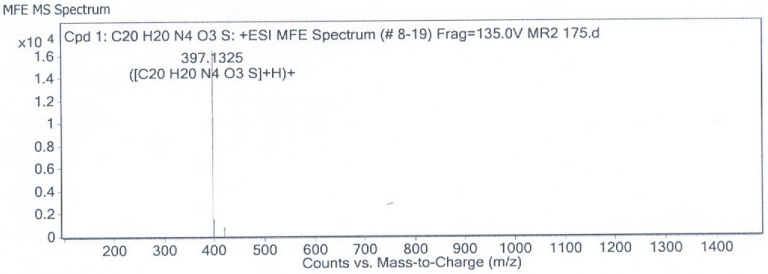
Sample Group	**Info.**
Acquisition SW Version	6200 series TOF/6500 series Q-TOF B.05.01 (B5125)

Compound Table

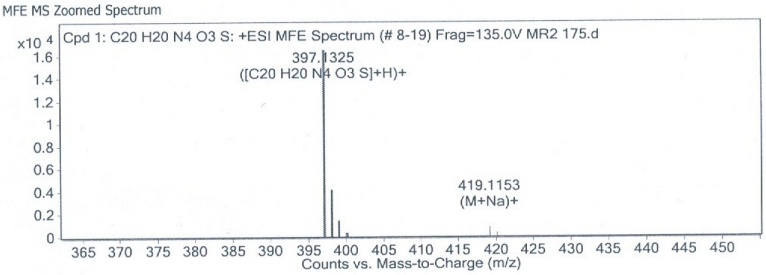
Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 1: C20 H20 N4 O3 S	11	396.1248	C20 H20 N4 O3 S	C20 H20 N4 O3 S	1.97	C20 H20 N4 O3 S

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 1: C20 H20 N4 O3 S	397.1325	11	Find by Molecular Feature	396.1248

MFE MS Spectrum



MFE MS Zoomed Spectrum



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
397.1325	1	16482.43	C20 H20 N4 O3 S	(M+H)+
398.1344	1	3926.23	C20 H20 N4 O3 S	(M+H)+
399.1305	1	1522.32	C20 H20 N4 O3 S	(M+H)+
400.1301	1	340.67	C20 H20 N4 O3 S	(M+H)+
419.1153	1	824.08		(M+Na)+
420.1152	1	375.6		(M+Na)+

--- End Of Report ---

Figure 29: ^{1}H NMR spectrum of compound **4j** in DMSO.

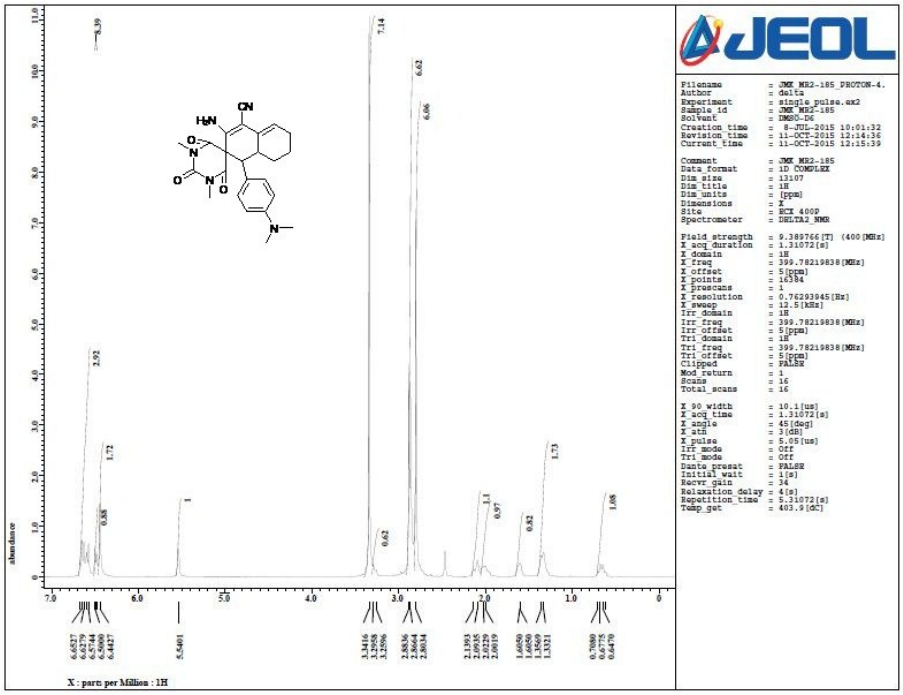


Figure 30: ^{13}C NMR spectrum of compound **4j** in DMSO.

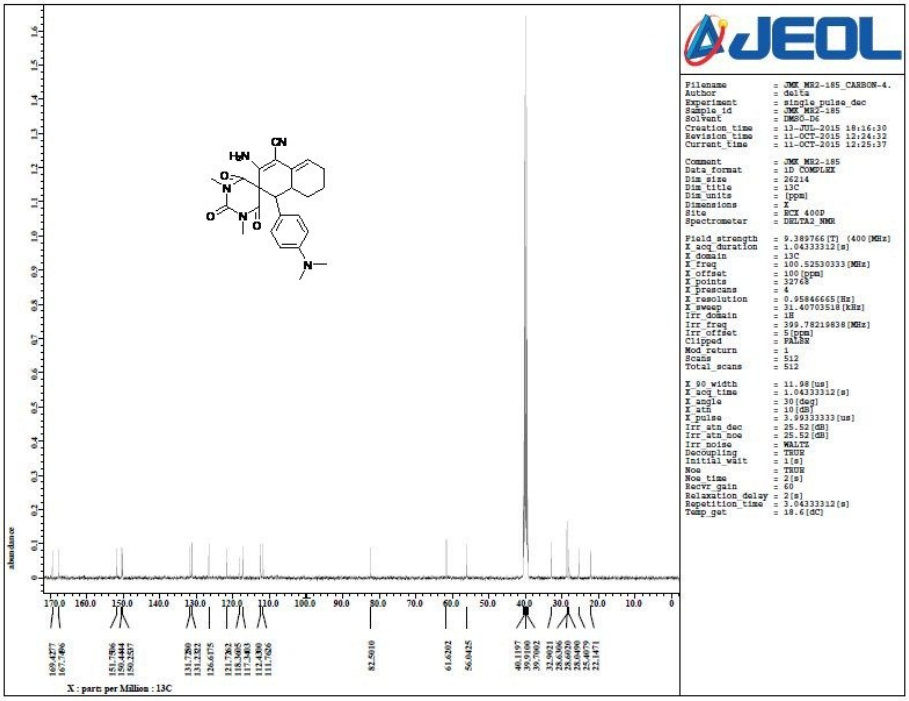


Figure 31: Mass spectrum of compound **4j**.

Qualitative Compound Report

Data File	MR 2 -1 85.d	**Sample Name**	MR 2 -1 85
Sample Type	Sample	**Position**	P1-E7
Instrument Name	Instrument 1	**User Name**	SMILY
Acq Method	29.10.2014.m	**Acquired Time**	19-08-2015 12:43:12
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

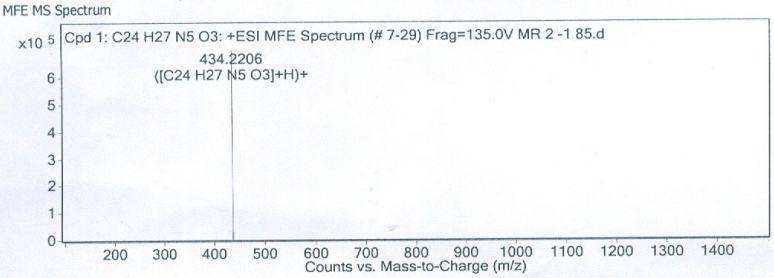
Sample Group **Info.**
Acquisition SW 6200 series TOF/6500 series
Version Q-TOF B.05.01 (B5125)

Compound Table

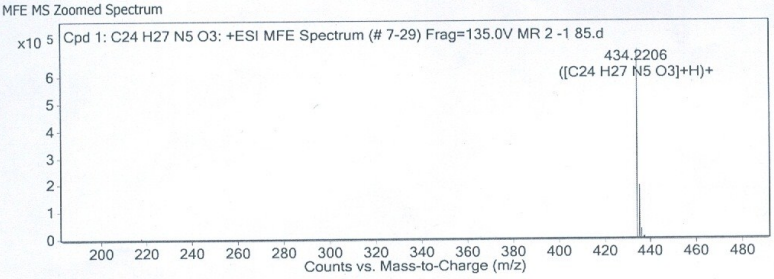
Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 1: C24 H27 N5 O3	10	433.2132	C24 H27 N5 O3	C24 H27 N5 O3	-4.13	C24 H27 N5 O3

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 1: C24 H27 N5 O3	434.2206	10	Find by Molecular Feature	433.2132

MFE MS Spectrum



MFE MS Zoomed Spectrum



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
217.6138	2	2832.23	C24 H27 N5 O3	(M+2H)+2
218.1151	2	1149.21	C24 H27 N5 O3	(M+2H)+2
434.2206	1	697641.75	C24 H27 N5 O3	(M+H)+
435.2234	1	185657.74	C24 H27 N5 O3	(M+H)+
436.2237	1	30365.02	C24 H27 N5 O3	(M+H)+
437.2241	1	4365.4	C24 H27 N5 O3	(M+H)+
456.2056	1	594.38	C21 H31 N5 O3 S	(M+Na)+

--- End Of Report ---

Figure 32: ^{1}H NMR spectrum of compound **4k** in DMSO.

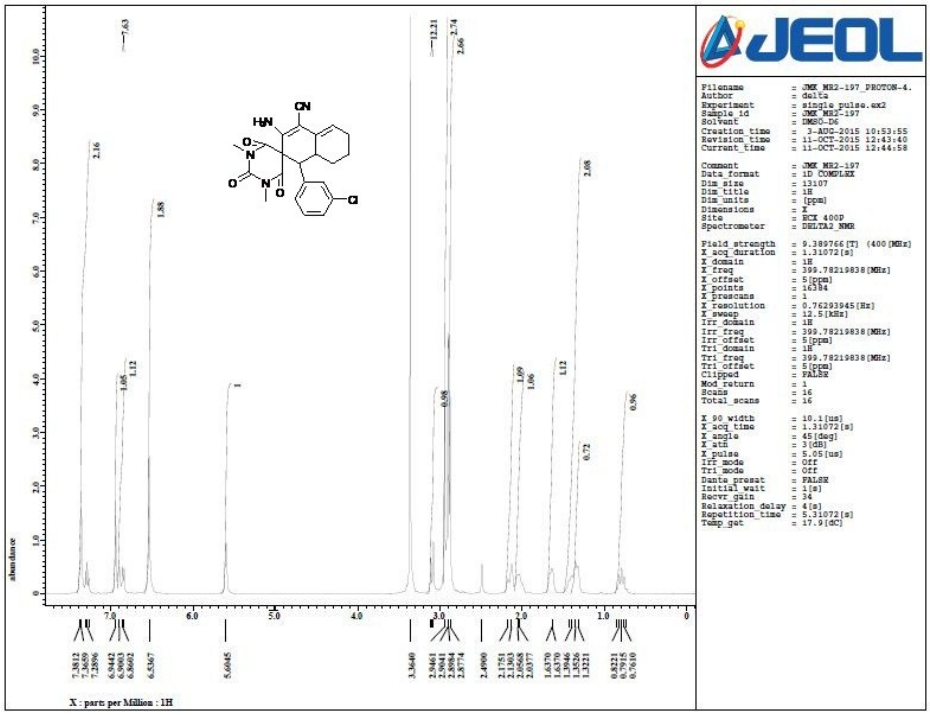


Figure 33: ^{13}C NMR spectrum of compound **4k** in DMSO.

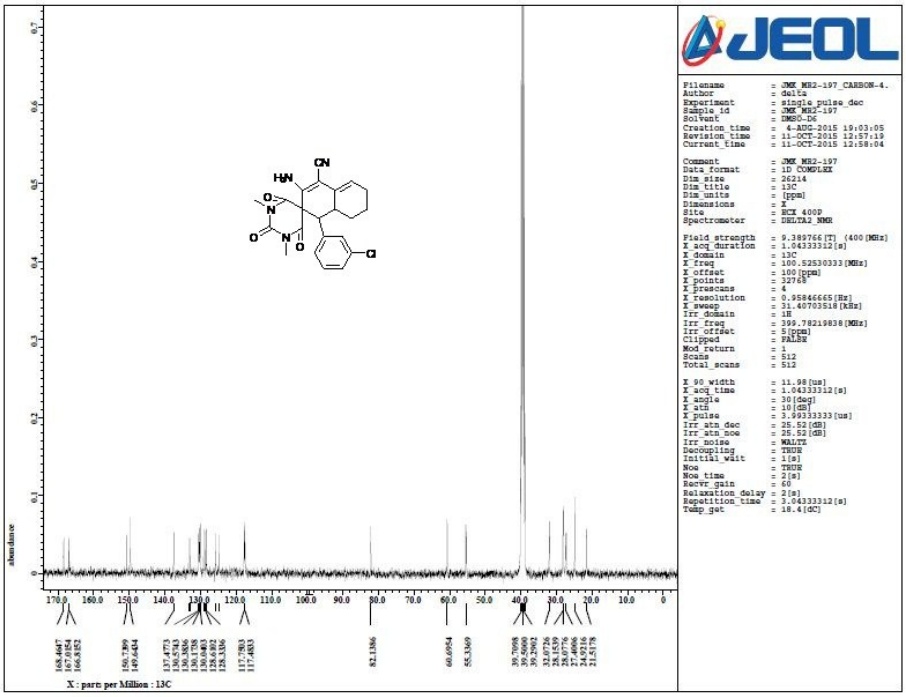
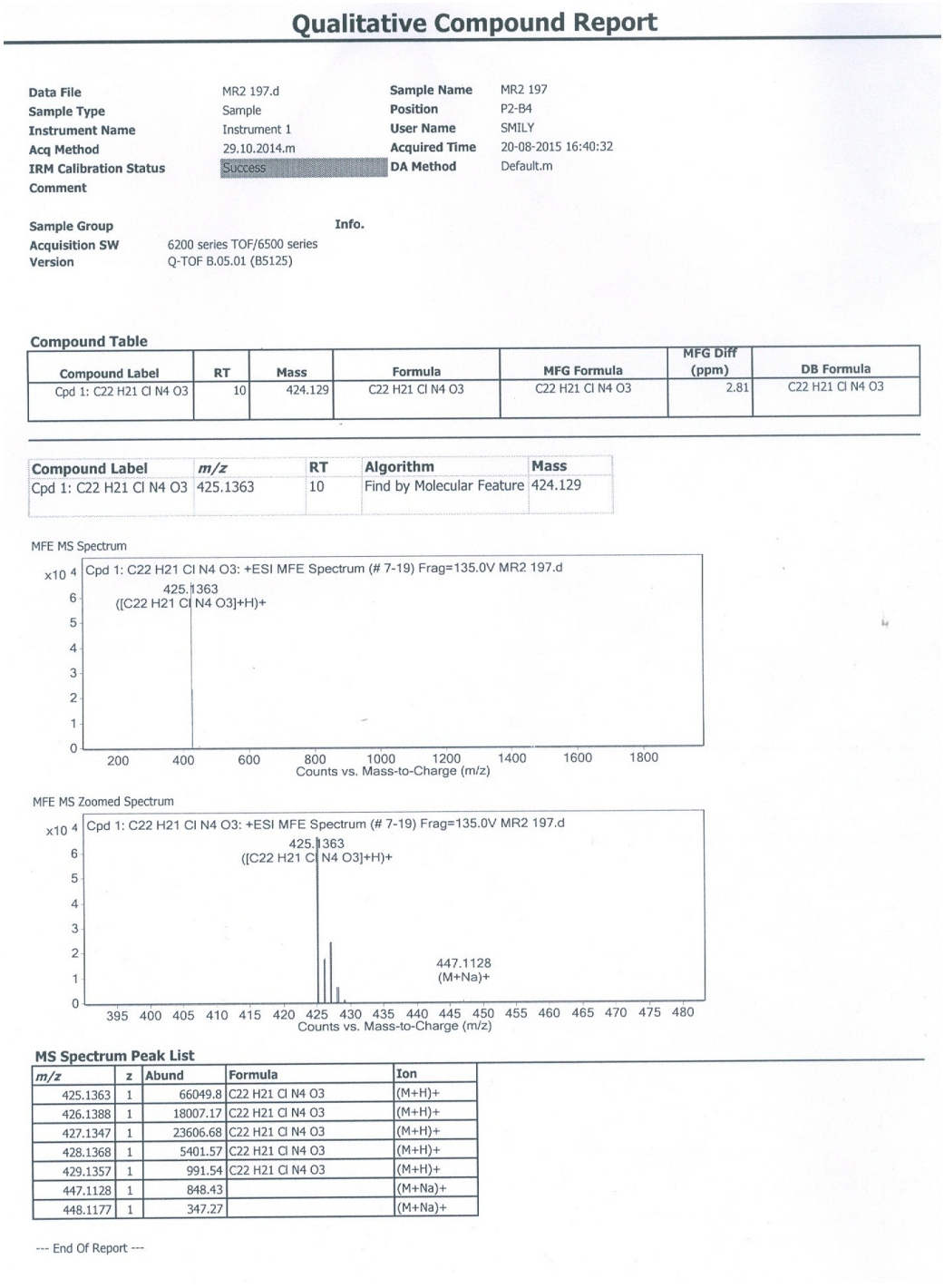


Figure 34: Mass spectrum of compound **4k**.



Qualitative Compound Report

Data File	MR2 197.d	**Sample Name**	MR2 197
Sample Type	Sample	**Position**	P2-B4
Instrument Name	Instrument 1	**User Name**	SMILY
Acq Method	29.10.2014.m	**Acquired Time**	20-08-2015 16:40:32
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

Sample Group **Info.**
Acquisition SW Version 6200 series TOF/6500 series Q-TOF B.05.01 (B5125)

Compound Table

Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 1: C22 H21 Cl N4 O3	10	424.129	C22 H21 Cl N4 O3	C22 H21 Cl N4 O3	2.81	C22 H21 Cl N4 O3

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 1: C22 H21 Cl N4 O3	425.1363	10	Find by Molecular Feature	424.129

MFE MS Spectrum

MFE MS Zoomed Spectrum

MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
425.1363	1	66049.8	C22 H21 Cl N4 O3	(M+H)+
426.1388	1	18007.17	C22 H21 Cl N4 O3	(M+H)+
427.1347	1	23606.68	C22 H21 Cl N4 O3	(M+H)+
428.1368	1	5401.57	C22 H21 Cl N4 O3	(M+H)+
429.1357	1	991.54	C22 H21 Cl N4 O3	(M+H)+
447.1128	1	848.43		(M+Na)+
448.1177	1	347.27		(M+Na)+

--- End Of Report ---

Figure 35: [1]H NMR spectrum of compound **4l** in DMSO.

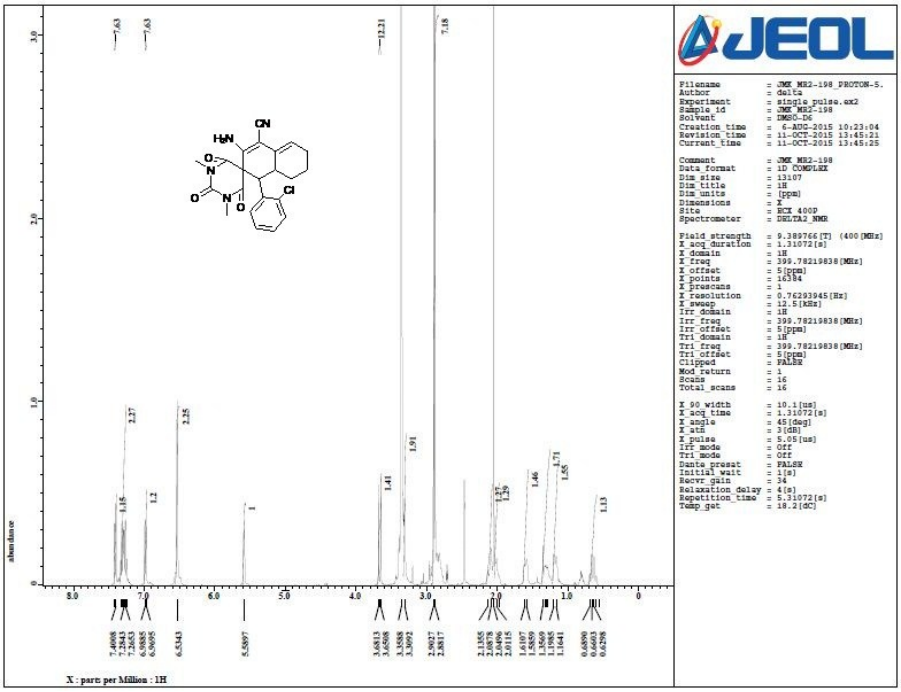


Figure 36: [13]C NMR spectrum of compound **4l** in DMSO.

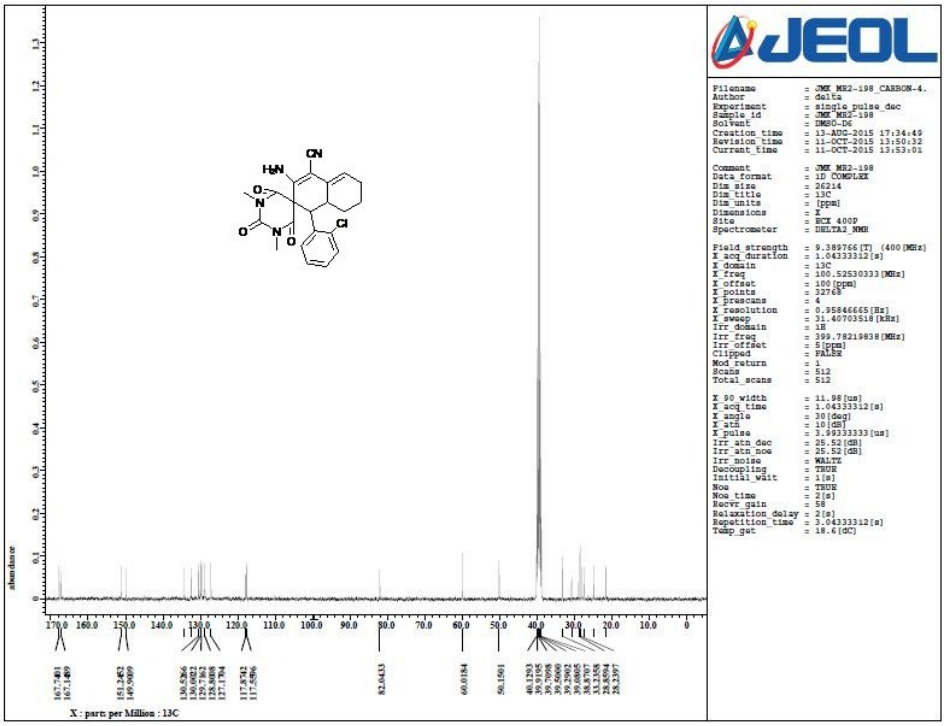


Figure 37: Mass spectrum of compound **4l**.

Qualitative Compound Report

Data File	MR2 198.d	**Sample Name**	MR2 198
Sample Type	Sample	**Position**	P2-C3
Instrument Name	Instrument 1	**User Name**	SMILY
Acq Method	29.10.2014.m	**Acquired Time**	24-08-2015 15:02:08
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

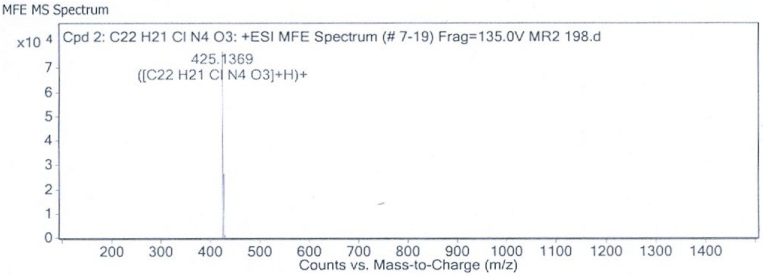
Sample Group	**Info.**
Acquisition SW Version	6200 series TOF/6500 series Q-TOF B.05.01 (B5125)

Compound Table

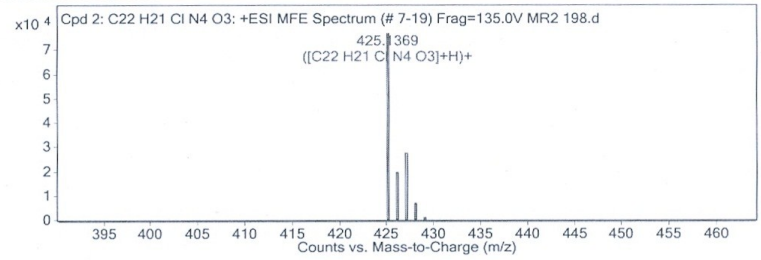
Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 2: C22 H21 Cl N4 O3	10	424.1298	C22 H21 Cl N4 O3	C22 H21 Cl N4 O3	0.94	C22 H21 Cl N4 O3

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 2: C22 H21 Cl N4 O3	425.1369	10	Find by Molecular Feature	424.1298

MFE MS Spectrum



MFE MS Zoomed Spectrum



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
425.1369	1	76850.82	C22 H21 Cl N4 O3	(M+H)+
426.1402	1	19742.42	C22 H21 Cl N4 O3	(M+H)+
427.1355	1	26531.27	C22 H21 Cl N4 O3	(M+H)+
428.1379	1	6726.85	C22 H21 Cl N4 O3	(M+H)+
429.1355	1	1026.81	C22 H21 Cl N4 O3	(M+H)+

--- End Of Report ---

Figure 38: ^{1}H NMR spectrum of compound **4m** in DMSO.

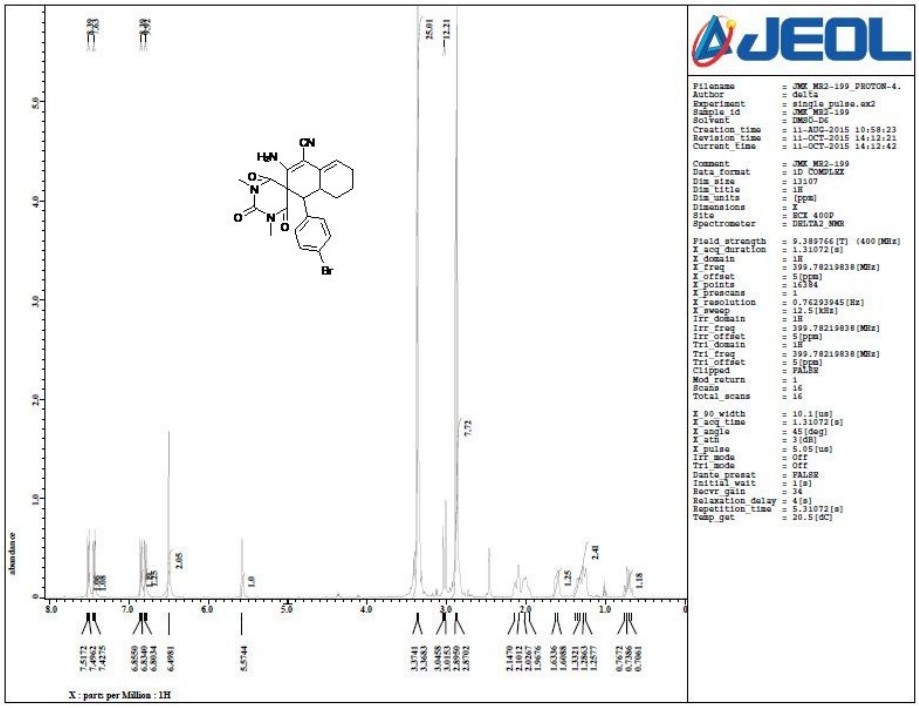


Figure 39: ^{13}C NMR spectrum of compound **4m** in DMSO.

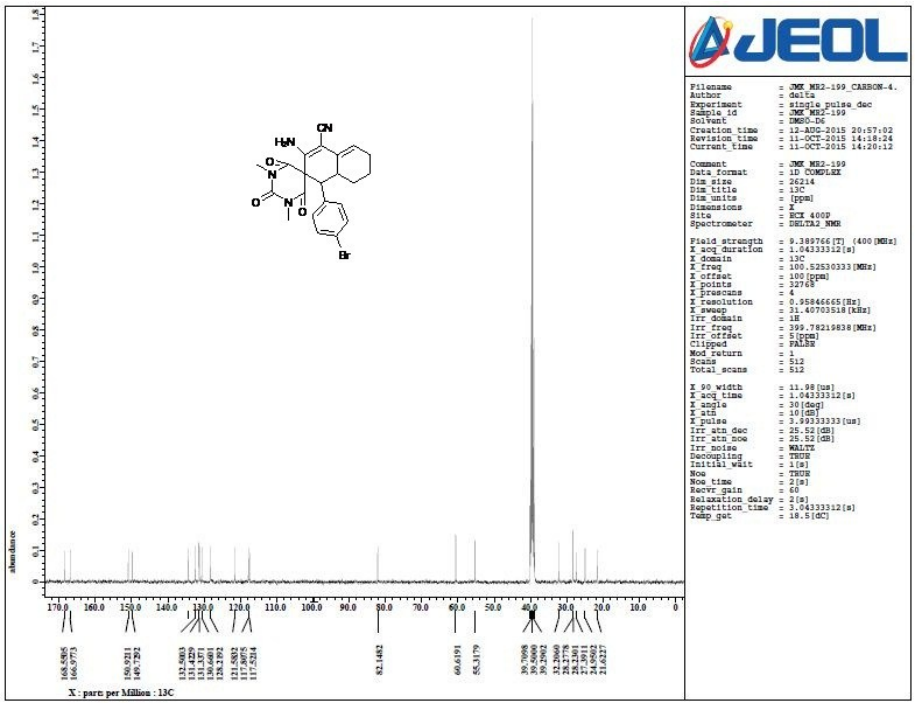


Figure 40: Mass spectrum of compound **4m**.

Qualitative Compound Report

Data File	MR2 199-.d	**Sample Name**	MR2 199-
Sample Type	Sample	**Position**	P2-B7
Instrument Name	Instrument 1	**User Name**	SMILY
Acq Method	29.10.2014.m	**Acquired Time**	26-08-2015 16:01:31
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

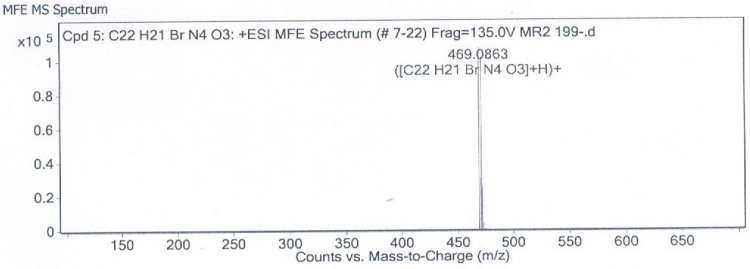
Sample Group **Info.**
Acquisition SW Version 6200 series TOF/6500 series Q-TOF B.05.01 (B5125)

Compound Table

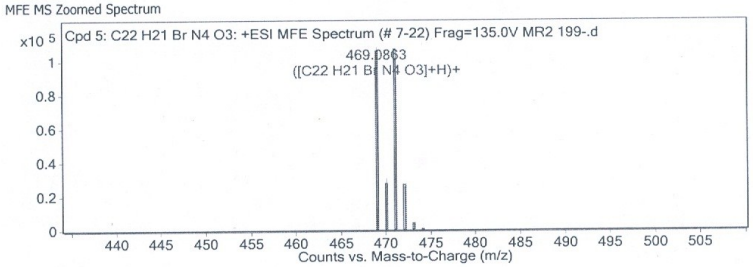
Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 5: C22 H21 Br N4 O3	11	468.0792	C22 H21 Br N4 O3	C22 H21 Br N4 O3	1.16	C22 H21 Br N4 O3

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 5: C22 H21 Br N4 O3	469.0863	11	Find by Molecular Feature	468.0792

MFE MS Spectrum



MFE MS Zoomed Spectrum



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
469.0863	1	107502.85	C22 H21 Br N4 O3	(M+H)+
470.0874	1	30697.64	C22 H21 Br N4 O3	(M+H)+
471.0853	1	101681.53	C22 H21 Br N4 O3	(M+H)+
472.0881	1	25291.63	C22 H21 Br N4 O3	(M+H)+
473.0903	1	3888.38	C22 H21 Br N4 O3	(M+H)+
474.0915	1	475.22	C22 H21 Br N4 O3	(M+H)+
475.1094	1	107.92	C22 H21 Br N4 O3	(M+H)+

--- End Of Report ---

Figure 41: [1]H NMR spectrum of compound **4n** in DMSO.

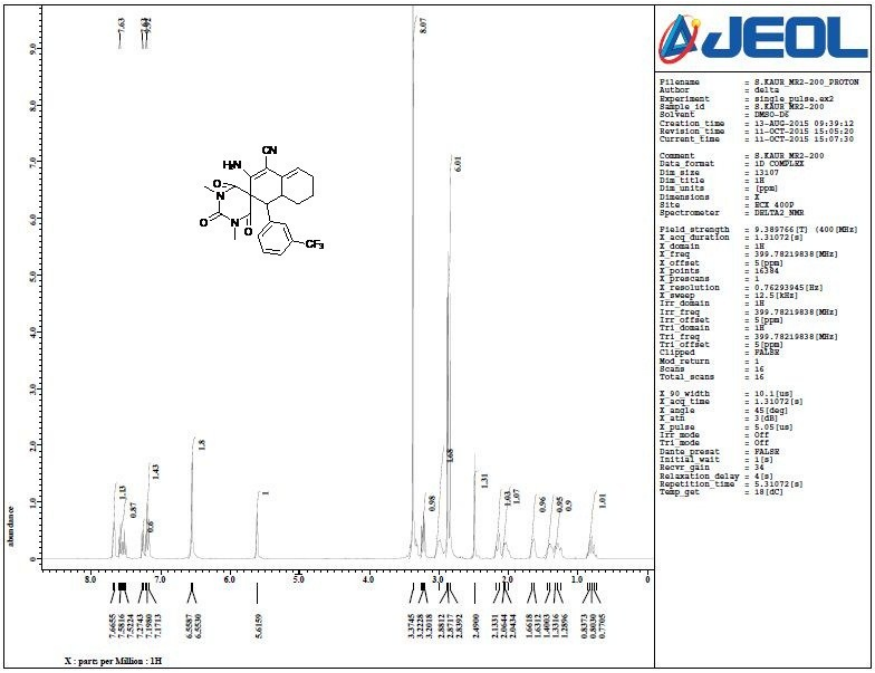


Figure 42: [13]C NMR spectrum of compound **4n** in DMSO.

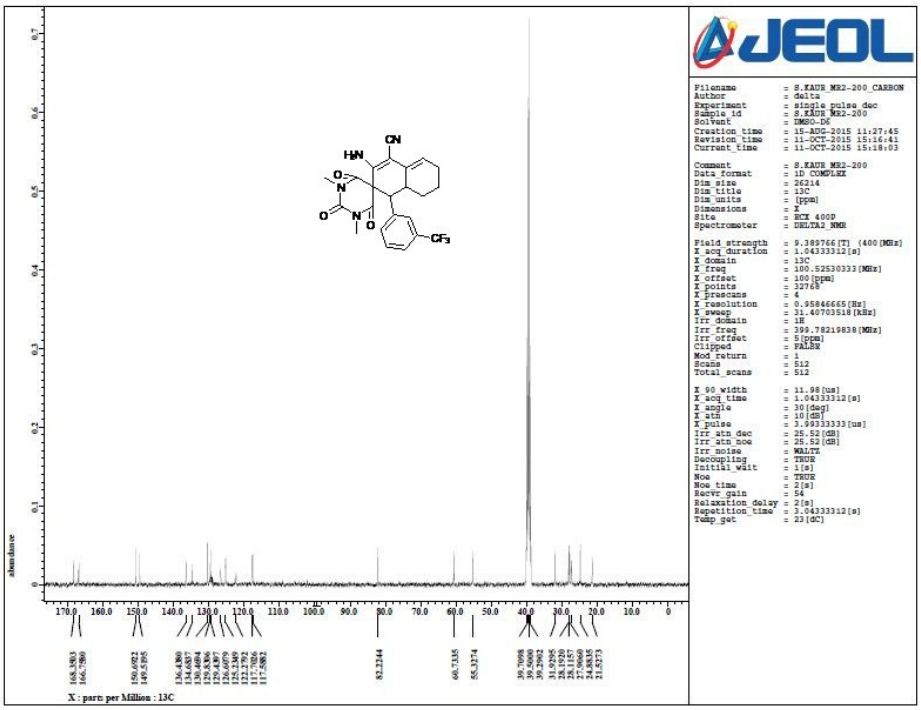


Figure 43: Mass spectrum of compound **4n**.

Qualitative Compound Report

Data File	MR2 200-.d	**Sample Name**	MR2 200-
Sample Type	Sample	**Position**	P2-B2
Instrument Name	Instrument 1	**User Name**	SMILY
Acq Method	29.10.2014.m	**Acquired Time**	26-08-2015 16:03:25
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

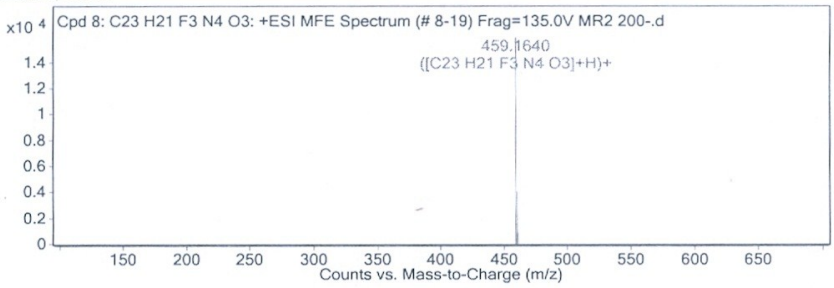
Sample Group	**Info.**
Acquisition SW Version	6200 series TOF/6500 series Q-TOF B.05.01 (B5125)

Compound Table

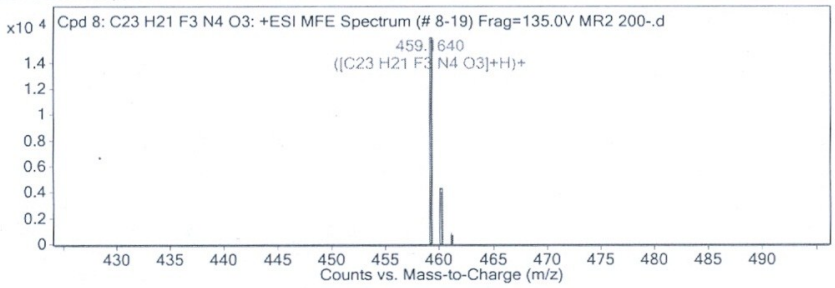
Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 8: C23 H21 F3 N4 O3	11	458.1565	C23 H21 F3 N4 O3	C23 H21 F3 N4 O3	0.24	C23 H21 F3 N4 O3

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 8: C23 H21 F3 N4 O3	459.164	11	Find by Molecular Feature	458.1565

MFE MS Spectrum



MFE MS Zoomed Spectrum



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
459.164	1	15945.24	C23 H21 F3 N4 O3	(M+H)+
460.1669	1	4006.78	C23 H21 F3 N4 O3	(M+H)+
461.1649	1	864.47	C23 H21 F3 N4 O3	(M+H)+

--- End Of Report ---

Figure 44: [1]H NMR spectrum of compound **4o** in DMSO.

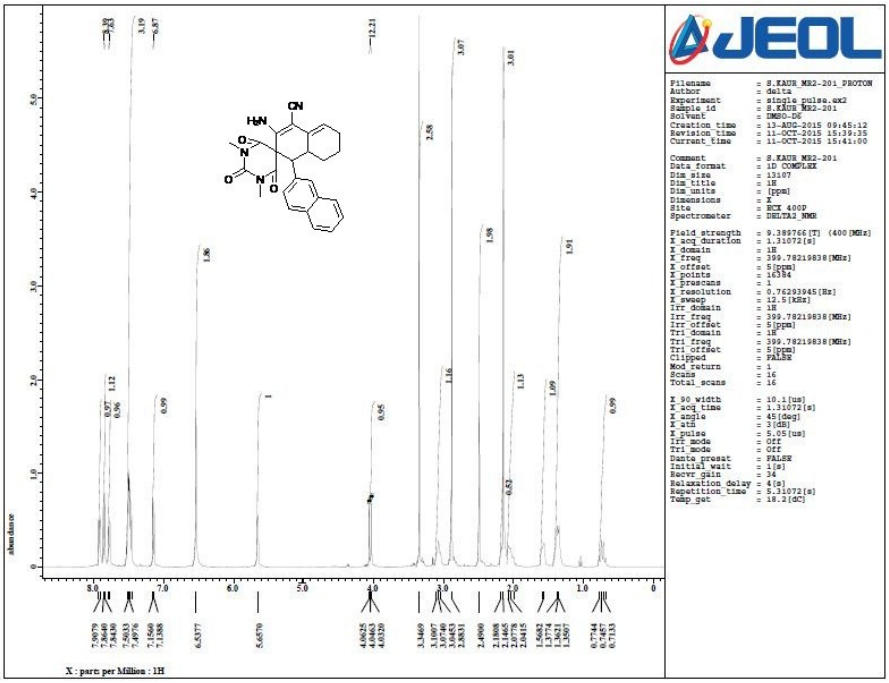


Figure 45: [13]C NMR spectrum of compound **4o** in DMSO.

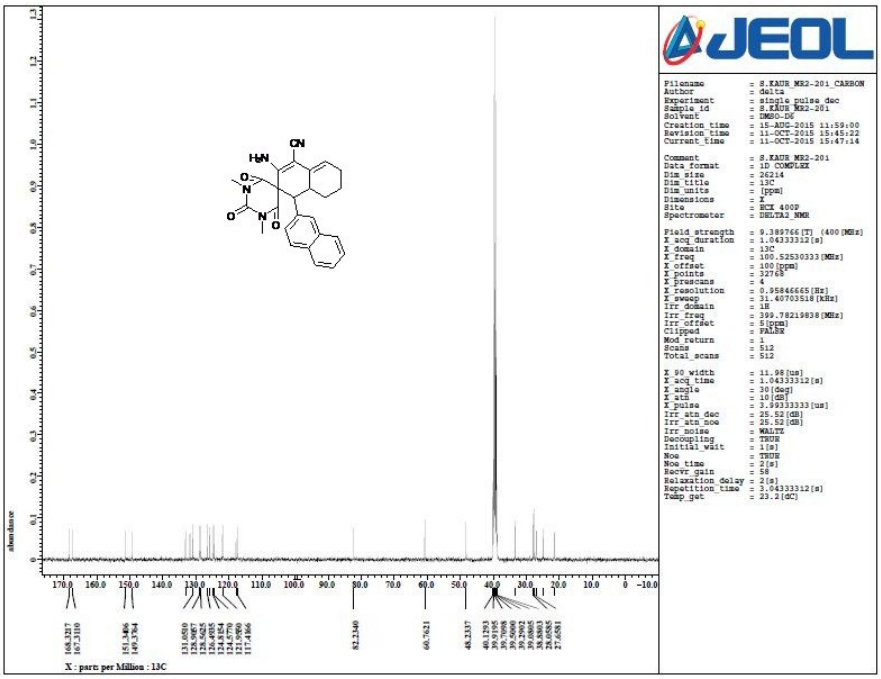
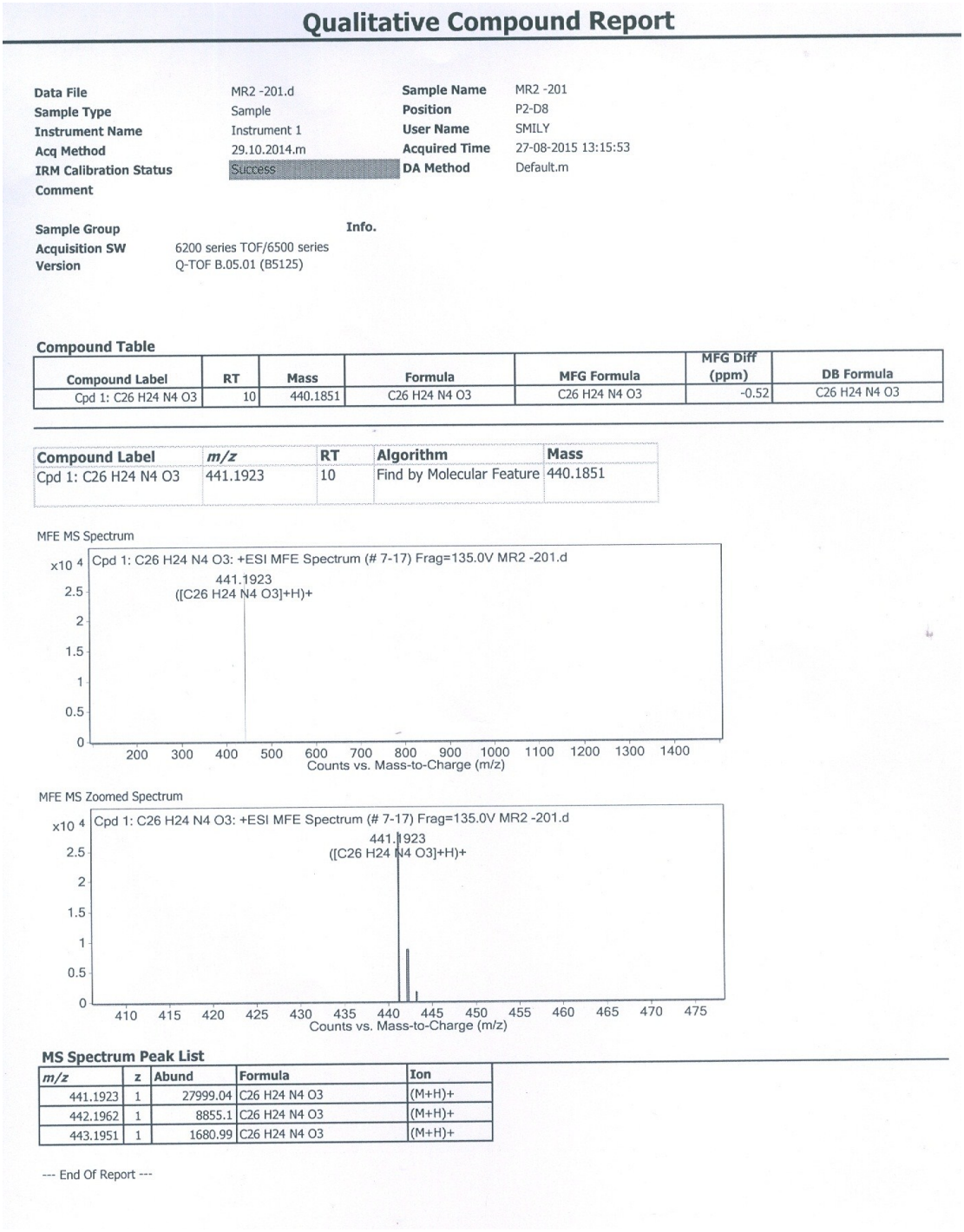


Figure 46: Mass spectrum of compound **4o**.



Qualitative Compound Report

Data File	MR2 -201.d	**Sample Name**	MR2 -201
Sample Type	Sample	**Position**	P2-D8
Instrument Name	Instrument 1	**User Name**	SMILY
Acq Method	29.10.2014.m	**Acquired Time**	27-08-2015 13:15:53
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

Sample Group **Info.**
Acquisition SW Version 6200 series TOF/6500 series Q-TOF B.05.01 (B5125)

Compound Table

Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 1: C26 H24 N4 O3	10	440.1851	C26 H24 N4 O3	C26 H24 N4 O3	-0.52	C26 H24 N4 O3

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 1: C26 H24 N4 O3	441.1923	10	Find by Molecular Feature	440.1851

MFE MS Spectrum

MFE MS Zoomed Spectrum

MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
441.1923	1	27999.04	C26 H24 N4 O3	(M+H)+
442.1962	1	8855.1	C26 H24 N4 O3	(M+H)+
443.1951	1	1680.99	C26 H24 N4 O3	(M+H)+

--- End Of Report ---

Figure 47: [1]H NMR spectrum of compound **4p** in DMSO.

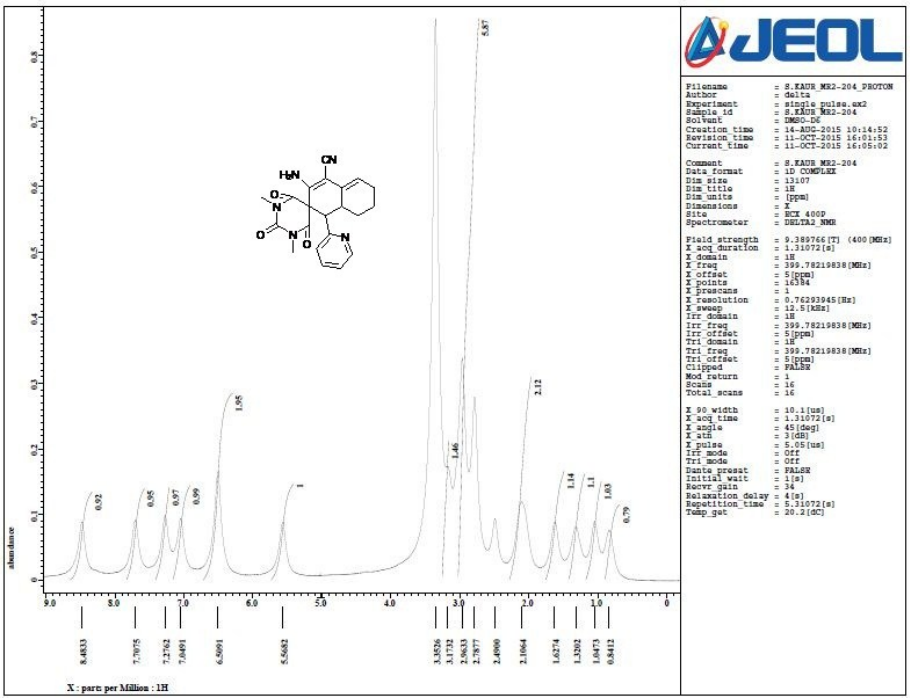


Figure 48: [13]C NMR spectrum of compound **4p** in DMSO.

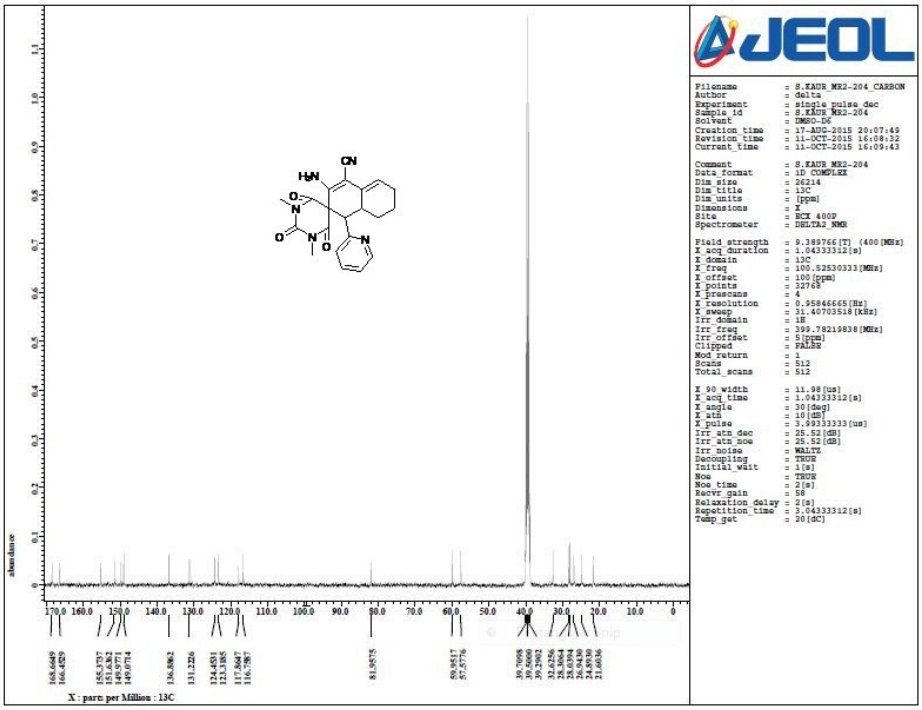


Figure 49: Mass spectrum of compound **4p**.

Qualitative Compound Report

Data File	MR2 -204.d	**Sample Name**	MR2 -204
Sample Type	Sample	**Position**	P2-A1
Instrument Name	Instrument 1	**User Name**	SMILY
Acq Method	29.10.2014.m	**Acquired Time**	31-08-2015 12:20:29
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

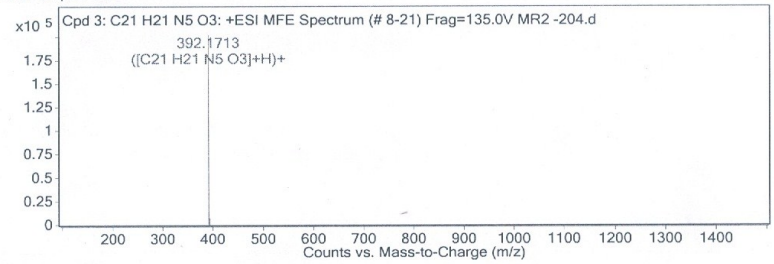
Sample Group **Info.**
Acquisition SW 6200 series TOF/6500 series
Version Q-TOF B.05.01 (B5125)

Compound Table

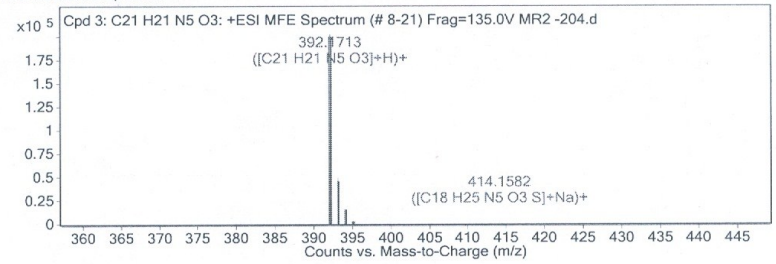
Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 3: C21 H21 N5 O3	11	391.1635	C21 H21 N5 O3	C21 H21 N5 O3	2.52	C21 H21 N5 O3

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 3: C21 H21 N5 O3	392.1713	11	Find by Molecular Feature	391.1635

MFE MS Spectrum



MFE MS Zoomed Spectrum



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
392.1713	1	201653.33	C21 H21 N5 O3	(M+H)+
393.1726	1	50209.54	C21 H21 N5 O3	(M+H)+
394.1722	1	7532.75	C21 H21 N5 O3	(M+H)+
395.1626	1	1304.68	C21 H21 N5 O3	(M+H)+
414.1582	1	506.76	C18 H25 N5 O3 S	(M+Na)+

--- End Of Report ---

Figure 50: [1]H NMR spectrum of compound **4q** in DMSO.

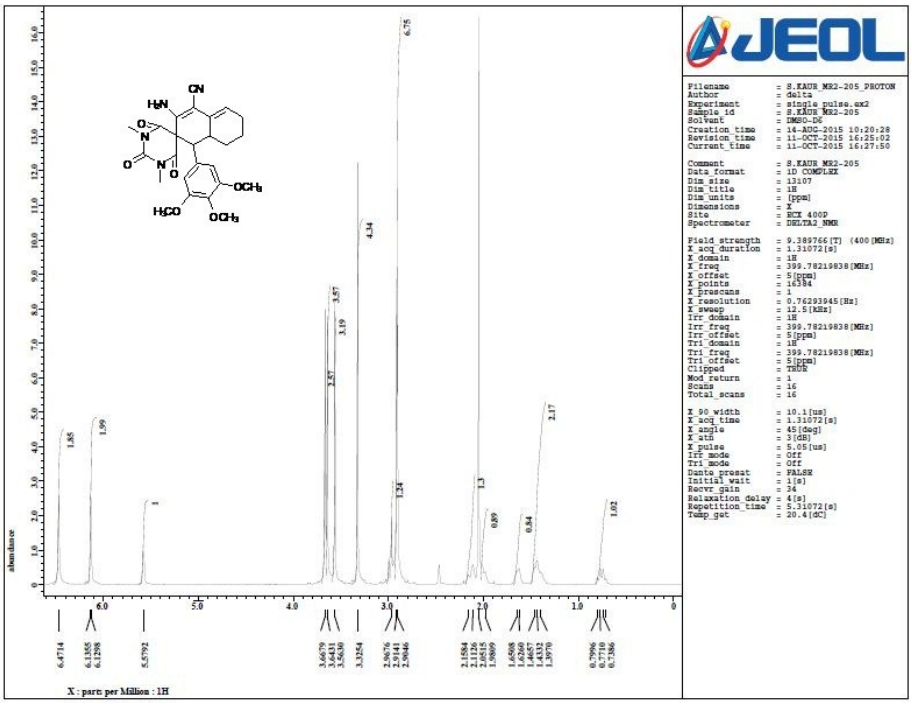


Figure 51: [13]C NMR spectrum of compound **4q** in DMSO.

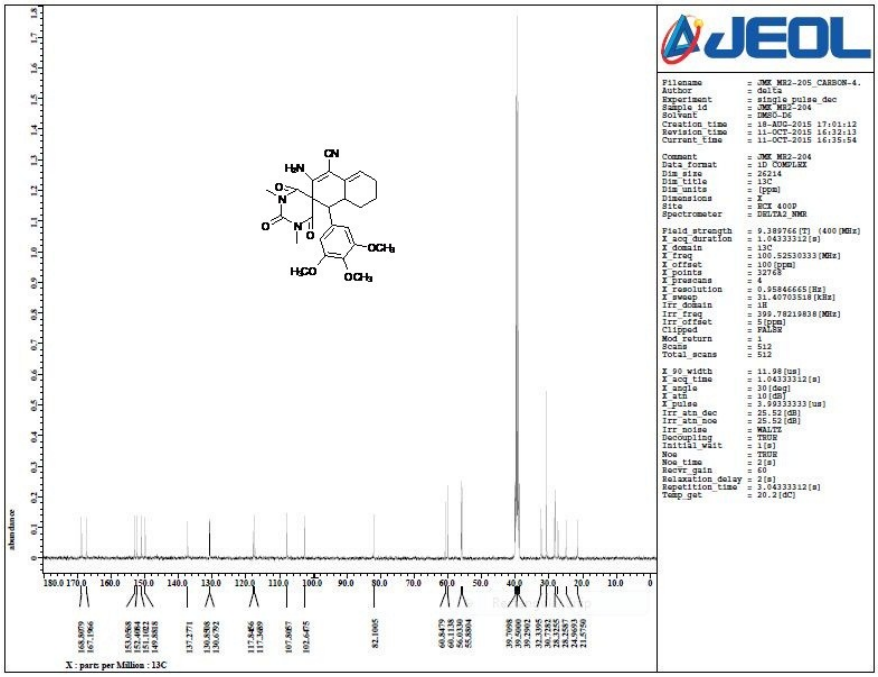
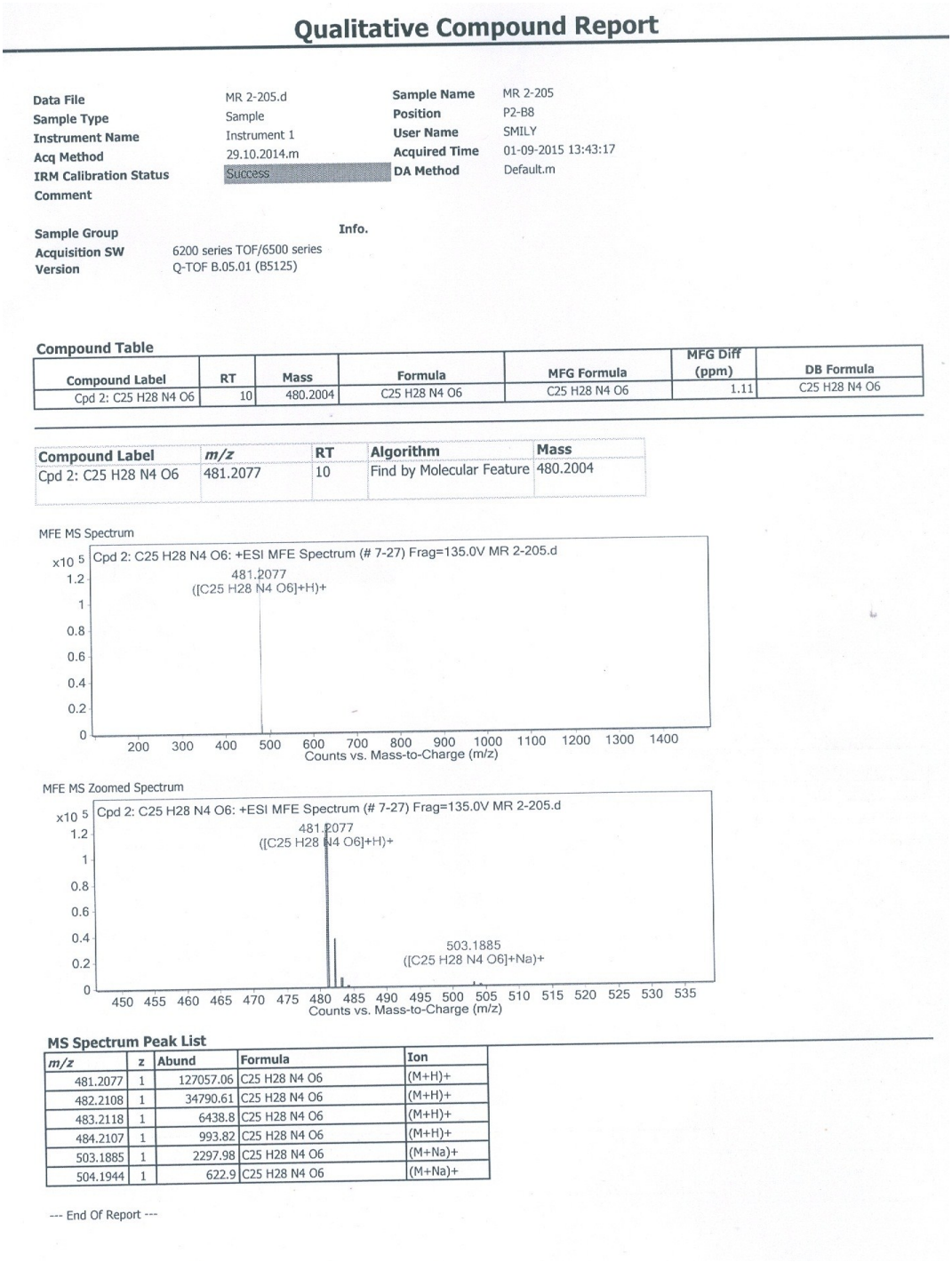


Figure 52: Mass spectrum of compound **4q**.



Qualitative Compound Report

Data File	MR 2-205.d	**Sample Name**	MR 2-205
Sample Type	Sample	**Position**	P2-B8
Instrument Name	Instrument 1	**User Name**	SMILY
Acq Method	29.10.2014.m	**Acquired Time**	01-09-2015 13:43:17
IRM Calibration Status	Success	**DA Method**	Default.m
Comment			

Sample Group **Info.**

Acquisition SW Version	6200 series TOF/6500 series Q-TOF B.05.01 (B5125)

Compound Table

Compound Label	RT	Mass	Formula	MFG Formula	MFG Diff (ppm)	DB Formula
Cpd 2: C25 H28 N4 O6	10	480.2004	C25 H28 N4 O6	C25 H28 N4 O6	1.11	C25 H28 N4 O6

Compound Label	*m/z*	RT	Algorithm	Mass
Cpd 2: C25 H28 N4 O6	481.2077	10	Find by Molecular Feature	480.2004

MFE MS Spectrum

MFE MS Zoomed Spectrum

MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
481.2077	1	127057.06	C25 H28 N4 O6	(M+H)+
482.2108	1	34790.61	C25 H28 N4 O6	(M+H)+
483.2118	1	6438.8	C25 H28 N4 O6	(M+H)+
484.2107	1	993.82	C25 H28 N4 O6	(M+H)+
503.1885	1	2297.98	C25 H28 N4 O6	(M+Na)+
504.1944	1	622.9	C25 H28 N4 O6	(M+Na)+

--- End Of Report ---

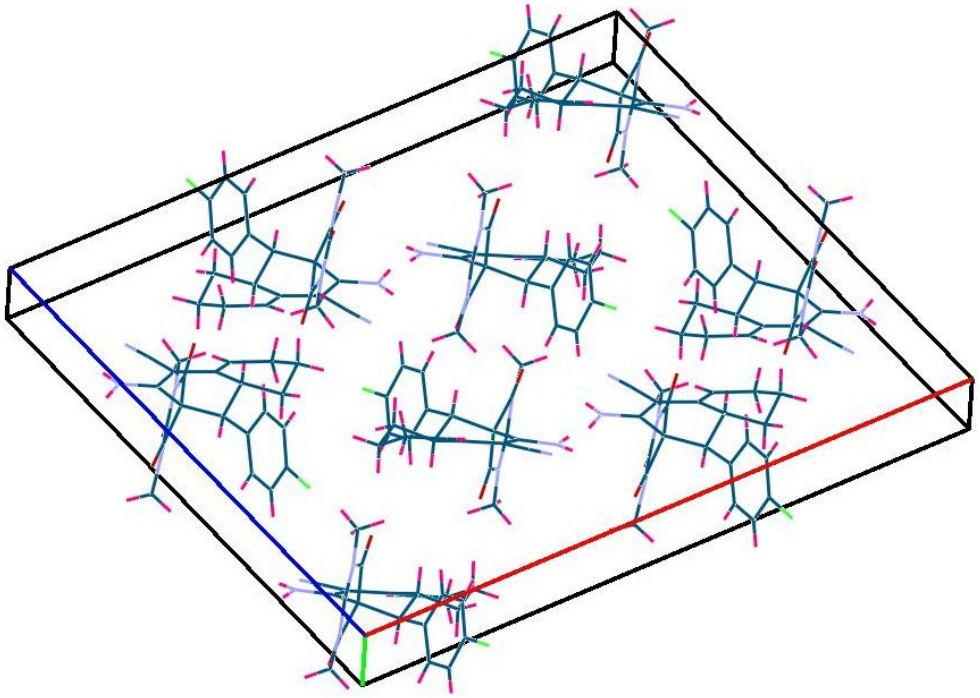
Table 1: Crystal data and structure refinement information for the compound **4a**.

Formula	C_{22} $H_{21}Cl$ N_4 O_3
Formula weight (g)	: 424.88
Temperature	: 293(2) K
Wavelength	: 0.71073 Å
Crystal system, space group	: monoclinic, I 2/a
Unit cell dimensions	: a = 22.6632(11) Å alpha = 90.00 deg. b = 10.9979(3) Å beta = 110.416(5) deg. c = 17.6446(8) Å gamma = 90.00 deg.
Volume	: 4121.6 (3)A^3
Z	: 8 g/cm^3
Absorption coefficient	: 0.217 mm^{-1}
F(000)	: 1776.0
Crystal size	: 0.04 X 0.03 X 0.01 mm
Theta range for data collection	: 2.96 to 25.00deg
Limiting indices	: Limiting indices -26<=h<=26, -13<=k<=13, -20<=l<=20
Completeness to theta	: 99.8%
Absorption correction	: Semi-empirical from equivalents
Max. and min. transmission	: 1.00000 and 0.87298
Refinement method	: Full-matrix least-squares on F2
Data / restraints / parameters	: 3623/ 0 / 279
Goodness-of-fit on F^2	: 0.983
Final R indices [I>2sigma(I)]	: R1 = 0.0761, wR2 = 0.2198
R indices (all data)	: R1 = 0.1959, wR2 = 0.3623
Largest diff. peak and hole	: 0.46 and -0.19 $e.A^{-3}$

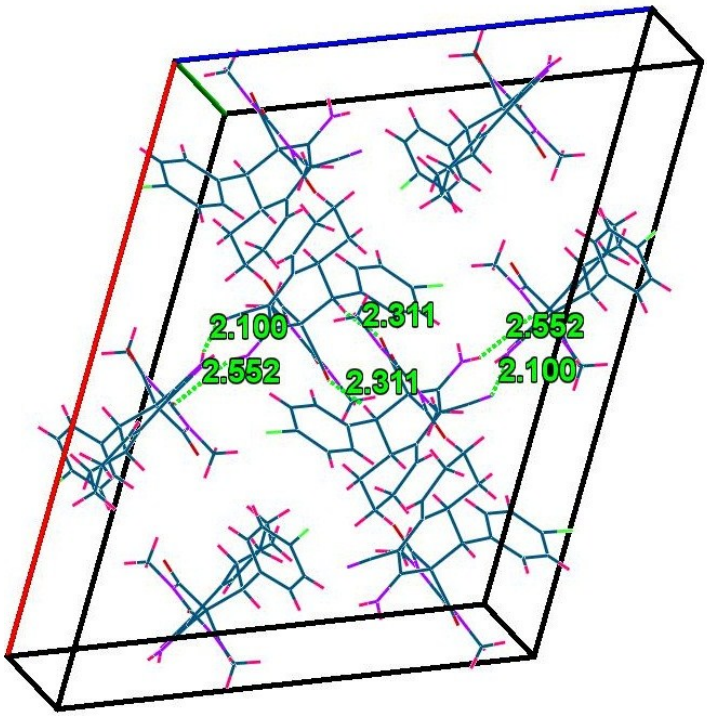
Table 2: Intermolecular and intramolecular hydrogen bonding geometries of **4a**.

D-H…A	D-H (Å)	H…A (Å)	D…A (Å)	D-H…A (°)
N(5)-H(15)…O(3)^{i}	0.81	2.56	3.203(7)	137
N(5)-H(25)…N(3)^{ii}	0.87	2.11	2.968(8)	171
C(7)-H(7)…O(2)^{iii}	0.98	2.31	3.283(6)	171
C(8)-H(8)…O(1)	0.98	2.48	3.120(6)	122
C(2)-H(2)…CL(1)^{iv}	0.93	2.80	3.622(5)	147
C(20)-H(20A)…O(3)	0.96	2.27	2.709(7)	107
C(21)-H(21A)…O(3)	0.96	2.24	2.681(7)	107

Note: D, Donor; A, Acceptor; Symmetry code: i) -x, 1/2+y, 1/2-z; ii) -x, -1/2+y, 1/2-z; iii) -x, 2-y, -z; iv) 1/2-x, y, -z



(a)



(b)

Figure 53: (a) Packing of the crystal in single unit cell (b) Intermolecular H-bonding of compound **4a.**