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Recyclization of *exo*-methylene bielectrophiles with 2-aminobenzimidazoles as an access to 3-substituted pyrimido[1,2-*a*]benzimidazol-2-ones

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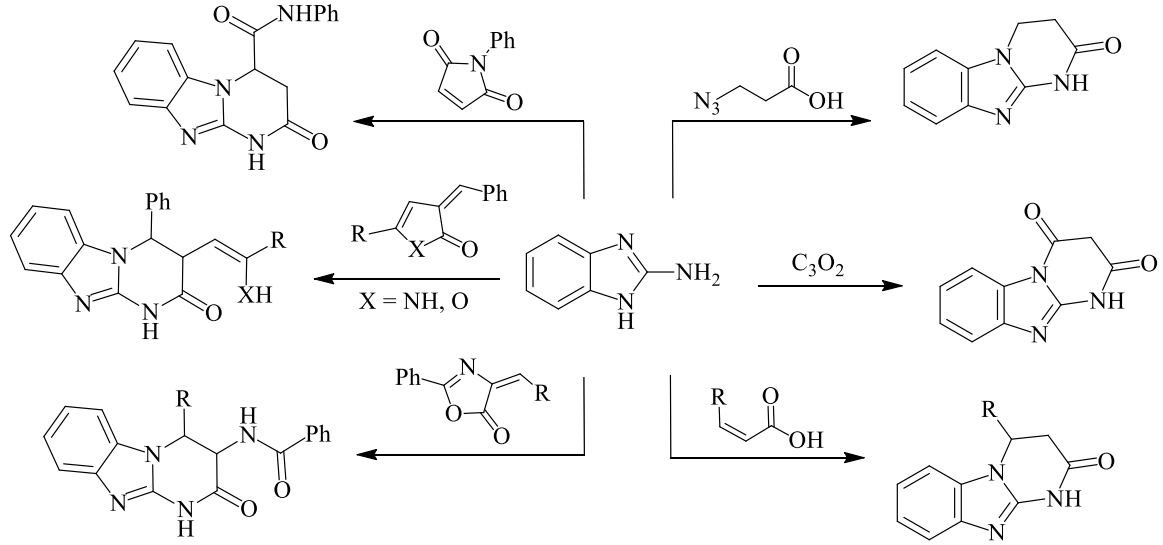
Materials and Metods

^{1}H and ^{13}C NMR spectra were registered on Bruker AVANCE II 400 (400.13 and 100.13 MHz, respectively) spectrometer in DMSO-d_6 + CCl_4, or Bruker DRX 500 (500.13 and 125.76 MHz, respectively) spectrometer in DMSO-d_6; internal standard was TMS. Melting points were determined on Stuart SMP 30. Control of reagent and products individuality, qualitative analysis of reaction mass was performed by TLC on Merck TLC Silica gel 60 F_{254} chromatographic plate; eluents: chloroform, ethylacetate, methanol and their mixtures in various ratios. The chromatograms were visualized by UV or iodine vapor.

Product purity was monitored by high performance liquid chromatography with high resolution mass spectrometric electrospray ionization detection (HPLC-HRMS-ESI) in combination with UV detection. The analyzes were performed on Agilent 1260 Infinity chromatograph (Agilent Technologies, CA, USA) and Agilent 6230 TOF LC/MS high-resolution time-of-flight mass detector. The ionization block was double electrospray; the signals were recorded in positive polarity; nebulizer N_2 20 psig; desiccant gas N_2, 6 ml min^{-1}, 325 °C; mass detection range is 50-2000 Da. Capillary voltage 4.0 kV, fragmentator +191 V, skimmer +66 V, OctRF 750 V. Poroshell 120 EC-C18 column (4.6 x 50 mm; 2.7 μm) was used. Gradient elution: acetonitrile/water (0.1% formic acid); flow rate 0.4 ml min^{-1}. Software for processing research results - MassHunter Workstation/Data Acquisition V.06.00.



Scheme S1 Literature data for the construction of pyrimido[1,2-*a*] enzimidazol-2-ones based on 2-aminobenzimidazole.

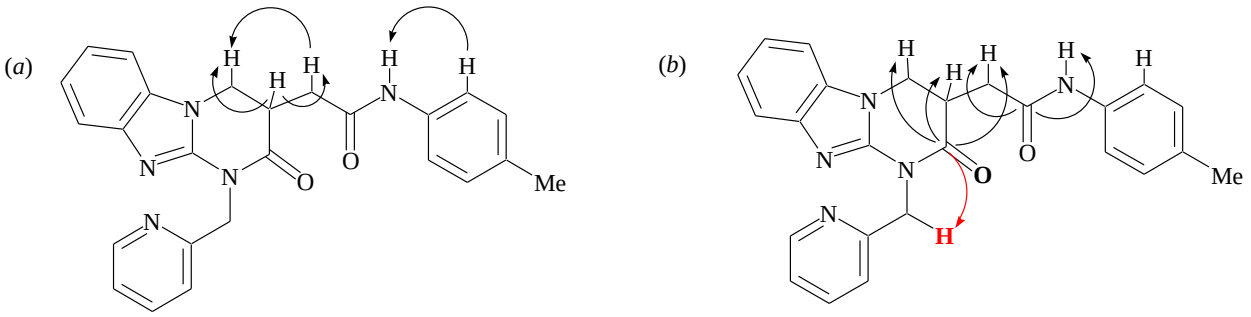


Figure S1 Principal correlations in the (*a*) ^{1}H-^{1}H NOESY and (*b*) ^{1}H-^{13}C HMBC spectra of compound **4j**

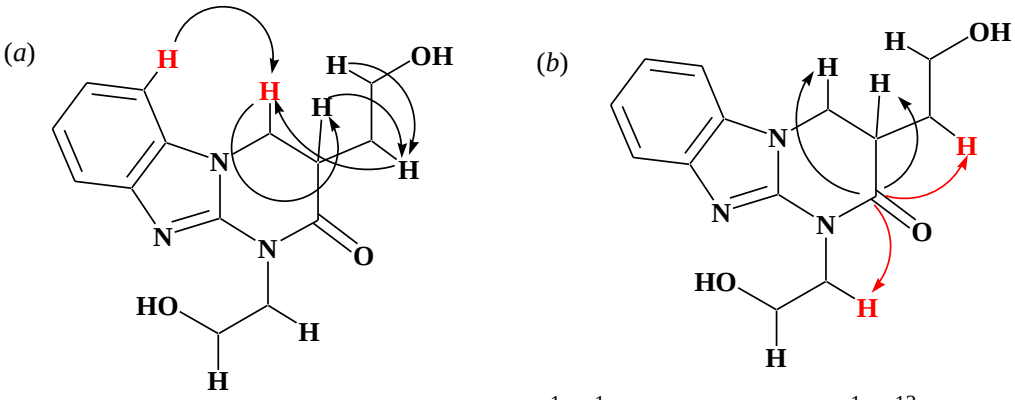
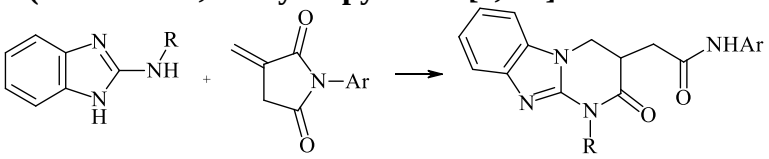


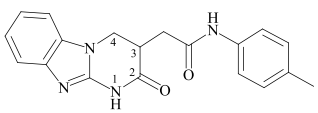
Figure S2 Principal correlations in the (*a*) ^{1}H-^{1}H NOESY and (*b*) ^{1}H-^{13}C HMBC spectra of compound **7f**.

Experimental.

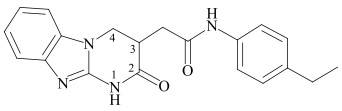
General procedure for the synthesis of 2-(2-oxo-1-*R*-3,4-dihydropyrimido[1,2-*a*]benzimidazol-3-yl)-*N*-arylacetamides.



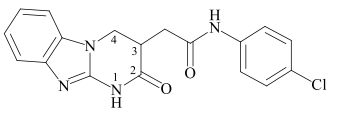
A solution of 0.005 mol of 2-*R*-aminobenzimidazole and 0.005 mol of *N*-arylitaconimide in isopropyl alcohol (30 ml) is boiled for 2-3 hours. The precipitated product is filtered, washed with isopropyl alcohol or acetone and dried.



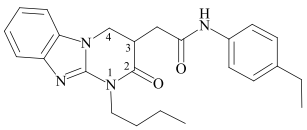
N-(4-Methylphenyl)-2-(2-oxo-3,4-dihydro-1H-pyrimido[1,2-a]-benzimidazol-3-yl)acetamide **4a.** Yield 75%, colorless powdery compound, m.p. 241-243 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 2.25 (3H, s, CH_3); 2.64 (1H, dd, *J* = 16.2, *J* = 7.3, CH_2CO); 2.94 (1H, dd, *J* = 16.2, *J* = 4.7, CH_2CO); 3.30-3.45 (1H, m, CH(3)); 4.01 (1H, t, *J* = 11.8, CH_2(4)); 4.52 (1H, dd, *J* = 11.8, *J* = 7.2, CH_2(4)); 7.02-7.13 (4H, m, 4CH-Ar); 7.30-7.35 (1H, m, CH-Ar); 7.37-7.47 (3H, m, 3CH-Ar); 9.98 (1H, s, NHCO); 11.52 (1H, s, NH(1)). ^{13}C NMR (δ, ppm): 20.51, 33.96, 36.02, 41.56, 108.71, 117.19, 119.20, 120.82, 121.42, 128.95, 132.04, 132.75, 136.52, 141.52, 147.80, 168.47, 169.61. HRMS: m/z calcd for $C_{19}H_{18}N_4O_2$, 335.1503; found, 335.1507.



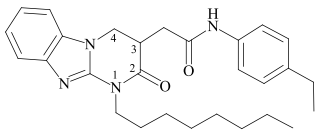
N-(4-Ethylphenyl)-2-(2-oxo-3,4-dihydro-1H-pyrimido[1,2-a]-benzimidazol-3-yl)acetamide **4b.** Yield 64%, colorless powdery compound, m.p. 238-240 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 1.17 (3H, t, *J* = 7.6, CH_3); 2.55 (2H, q, *J* = 16.1, *J* = 7.6, $ArCH_2$); 2.65 (1H, dd, *J* = 16.2, *J* = 7.6, CH_2CO); 2.96 (1H, d, *J* = 16.2, CH_2CO); 3.32-3.42 (1H, m. CH(3)); 4.00 (1H, t, *J* = 11.7, CH_2(4)); 4.52 (1H, dd, *J* = 11.7, *J* = 7.2, CH_2(4)); 7.03-7.11 (4H, m, 4CH-Ar); 7.27-7.33 (1H, m, CH-Ar); 7.37-7.42 (1H, m, CH-Ar); 7.43-7.49 (2H, m, 2CH-Ar); 9.96 (1H, s, NHCO); 11.51 (1H, s, NH(1)). ^{13}C NMR (δ, ppm): 15.70, 27.77, 34.01, 36.10, 41.58, 106.59, 117.30, 119.25, 120.75, 121,35, 127.63, 132.73, 136.74, 138.43, 141.53, 147.77, 168.40, 169.50. HRMS: m/z calcd for $C_{20}H_{20}N_4O_2$, 349.1660; found, 349.1661.



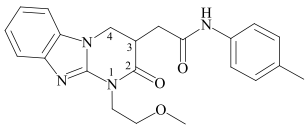
N-(4-Chlorophenyl)-2-(2-oxo-3,4-dihydro-1H-pyrimido[1,2-a]-benzimidazol-3-yl)acetamide **4c.** Yield 82%, colorless powdery compound, m.p. 242-245 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 2.68 (1H, dd, *J* = 16.3, *J* = 7.2, CH_2CO); 2.97 (1H, dd, *J* = 16.3, *J* = 4.9, CH_2CO); 3.40-3.48 (1H, m, CH(3)); 4.05 (1H, t, *J* = 11.8, CH_2(4)); 4.55 (1H, dd, *J* = 12.0, *J* = 7.3, CH_2(4)); 7.09-7.14 (2H, m, 2CH-Ar); 7.34-7.44 (4H, m, 4CH-Ar); 7.61-7.44 (2H, m, 2CH – Ar); 10.24 (1H, s, NHCO); 11.51 (1H, s, NH(1)). ^{13}C NMR (δ, ppm): 34.23, 36.03, 41.60, 108.87, 117.27, 120.72, 120.81, 121.43, 126.75, 128.64, 132.92, 138.13, 141.75, 147.99, 169.07, 169.70. HRMS: m/z calcd for $C_{18}H_{15}ClN_4O_2$, 355.0957; found, 355.0955.



2-(1-Butyl-2-oxo-3,4-dihydropyrimido[1,2-a]benzimidazol-3-yl)-N-(4-ethylphenyl)acetamide **4d.** Yield 50%, colorless powdery compound, m.p. 216-217 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 0.94 (3H, t, *J* = 7.1, CH_3); 1.17 (3H, t, *J* = 7.6, CH_3); 1.30-1-43 (2H, m, CH_2); 1.59-1.73 (2H, m, CH_2); 2.55 (2H, q, *J* = 16.5, *J* = 7.6, Ar-CH_2); 2.67 (1H, dd, *J* = 16.4, *J* = 7.1, CH_2CO); 2.95 (1H, dd, *J* = 16.1, *J* = 4.5, CH_2CO); 3.37-3.47 (1H, m. CH(3)); 3.88-3.99 (2H, m, N-CH_2); 4.03 (1H, dd, *J* = 11.8, *J* = 4.8, CH_2(4)); 4.51 (1H, dd, *J* = 11.8, *J* = 7.1, CH_2(4)); 7.03-7.14 (4H, m – 4CH-Ar); 7.29-7.35 (1H, m, CH-Ar); 7.42-7.49 (3H, m, 3CH – Ar); 9.98 (1H, s, NHCO). ^{13}C NMR (δ, ppm): 13.77, 15.72, 19.58, 27.76, 29.61, 34.35, 36.48, 41.10, 42.03, 108.71, 117.51, 119.28, 121.13, 121.42, 127.67, 133.36, 136.71, 138.50, 141.32, 148.30, 168.09, 168.28. HRMS: m/z calcd for $C_{24}H_{28}N_4O_2$, 405,2286; found, 405.2284.

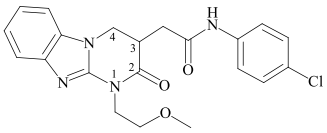


2-(1-Octyl-2-oxo-3,4-dihydropyrimido[1,2-a]benzimidazol-3-yl)-N-phenyl-acetamide **4e.** Yield 55%, colorless powdery compound, m.p. 162-163 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 0.84 (3H, t, *J* = 7.1, CH_3); 1.19-1.38 (10H, m, $5CH_2$); 1.60-1-72 (2H, m, CH_2); 1.59-1.73 (2H, m, CH_2); 2.71 (1H, dd, *J* = 16.2, *J* = 7.0, CH_2CO); 2.97 (1H, dd, *J* = 16.2, *J* = 5.1, CH_2CO); 3.49-3.57 (1H, m. CH(3)); 3.90-4.05 (2H, m, N-CH_2); 4.08 (1H, t, *J* = 11.7, CH_2(4)); 4.56 (1H, dd, *J* = 12.0, *J* = 7.2, CH_2(4)); 7.02-7.06 (1H, m, CH-Ar); 7.12-7.18 (2H, m, 2CH-Ar); 7.27-7.32 (2H, m, 2CH-Ar); 7.40-7.44 (1H, m, CH-Ar); 7.49-7.53 (1H, m, CH-Ar), 7.58-7.62 (2H, m, 2CH-Ar); 10.09 (1H, s, NHCO). ^{13}C NMR (δ, ppm): 13.98, 22.13, 26.24, 27.44, 28.67, 28.80, 31.28, 34.59, 36.43, 41.13, 42.22, 108.97, 117.62, 119.15, 121.19, 121.52, 123.17, 128.71, 133.55, 139.19, 141.46, 148.54, 168.36, 168.72. HRMS: m/z calcd for $C_{26}H_{32}N_4O_2$, 433,2599; found, 433.2599.

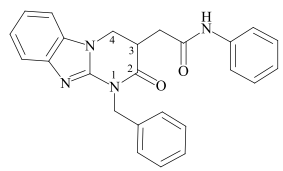


2-[1-(2-Methoxyethyl)-2-oxo-3,4-dihydropyrimido[1,2-a]benzimidazol-3-yl]-N-(p-tolyl)acetamide **4f.** Yield 62%, colorless powdery compound, m.p. 222-224 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 2.26 (3H, s, CH_3); 2.67 (2H, dd, *J* = 16.3, *J* = 7.3, CH_2CO); 2.90-3.02 (1H, m, CH_2CO); 3.21 (3H, s, OCH_3); 3.35-3.55 (3H, m, CH(3) + OCH_2)); 3.95-4.16 (3H, m, N-CH_2 + CH_2(4)); 4.52 (1H, dd, *J* = 12.1, *J* = 7.2, CH_2(4)); 7.00-7.16 (4H, m, 4CH-Ar); 7.29-7.37 (1H, m, CH-Ar); 7.40-7.52 (3H, m, 3CH-Ar); 9.95 (1H, s, NHCO). ^{13}C NMR (δ, ppm): 20.53, 27.61, 34.35, 36.48, 41.11, 57.90, 69.87, 108.66,

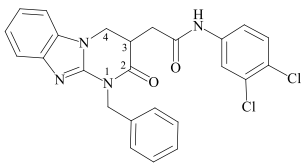
117.51, 119.19, 121.10, 121.41, 128.83, 131.84, 133.37, 136.52, 141.32, 148.29, 168.10, 168.19. HRMS: m/z calcd for $C_{22}H_{24}N_4O_3$, 393,1922; found, 393.1920.



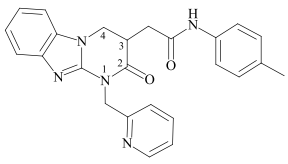
N-(4-Chlorophenyl)-2-[1-(2-methoxyethyl)-2-oxo-3,4-dihydropyrimido[1,2-a]benzimidazol-3-yl]acetamide **4g.** Yield 68%, colorless powdery compound, m.p. 227-228 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 2.69 (2H, dd, *J* = 16.4, *J* = 7.1, CH_2CO); 2.90-3.02 (1H, m, CH_2CO); 3.21 (3H, s, OCH_3); 3.40-3.45 (1H, m, CH(3)); 3.61 (2H, t, *J* = 6.2, OCH_2); 4.02 (1H, t, *J* = 12.1, CH_2(4)); 4.05-4.28 (2H, m, N-CH_2); 4.54 (1H, dd, *J* = 12.1, *J* = 7.1, CH_2(4)); 7.08-7.17 (2H, m, 2CH-Ar); 7.22-7.30 (2H, m, 2CH-Ar); 7.31-7.38 (1H, m, CH-Ar); 7.42-7.50 (1H, m, CH-Ar); 7.55-7.65 (2H, m, 2CH-Ar); 10.23 (1H, s, NHCO). ^{13}C NMR (δ, ppm): 34.41, 36.42, 41.07, 41.11, 58.03, 68.46, 108.82, 117.58, 120.67, 121.25, 121.53, 126.92, 128.38, 133.35, 137.91, 141.18, 148.25, 168.27, 168.72. HRMS: m/z calcd for $C_{21}H_{21}ClN_4O_3$, 413,1376; found, 413.1378.



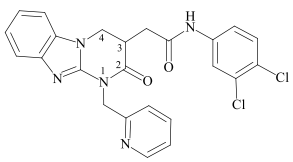
2-(1-Benzyl-2-oxo-3,4-dihydropyrimido[1,2-a]benzimidazol-3-yl)-N-phenyl-acetamide **4h.** Yield 66%, colorless powdery compound, m.p. 217-218 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 2.76 (1H, dd, *J* = 16.8, *J* = 7.6, CH_2CO); 3.00 (1H, dd, *J* = 16.2, *J* = 5.0, CH_2CO); 3.62-3.69 (1H, m, CH(3)); 4.17 (1H, t, *J* = 11.8, CH_2(4)); 4.63 (1H, dd, *J* = 12.1, *J* = 7.3, CH_2(4)); 5.21 (2H, q, *J* = 22.1, *J* = 15.3, N-CH_2); 7.02-7.06 (1H, m, CH-Ar); 7.12-7.19 (2H, m, 2CH-Ar); 7.21-7.26 (1H, m, CH-Ar); 7.28-7.32 (4H, m, 4CH-Ar); 7.37-7.40 (2H, m, 2CH-Ar); 7.43-7.46 (1H, m, CH-Ar); 7.48-7.51 (1H, m, CH-Ar); 7.58-7.62 (2H, m, 2CH-Ar); 10.11 (1H, s, NHCO). ^{13}C NMR (δ, ppm): 34.39, 36.35, 41.03, 45.20, 108.99, 117.59, 119.04, 121.24, 121.50, 123.09, 126.96, 127.17, 128.21, 128.62, 133.57, 137.04, 139.04, 141.16, 148.50, 168.50, 168,59. HRMS: m/z calcd for $C_{25}H_{22}N_4O_2$, 411,1817; found, 411.1818.



2-(1-Benzyl-2-oxo-3,4-dihydropyrimido[1,2-a]benzimidazol-3-yl)-N-(3,4-dichlorophenyl)acetamide **4i.** Yield 75%, colorless powdery compound, m.p. 191-192 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 2.78 (1H, dd, *J* = 16.3, *J* = 6.8, CH_2CO); 3.00 (1H, dd, *J* = 16.4, *J* = 5.3, CH_2CO); 3.63-3.70 (1H, m, CH(3)); 4.16 (1H, t, *J* = 11.9, CH_2(4)); 4.63 (1H, dd, *J* = 12.1, *J* = 7.4, CH_2(4)); 5.20 (2H, q, *J* = 25.1, *J* = 15.3, N-CH_2); 7.12-7.20 (2H, m, 2CH-Ar); 7.22-7.26 (1H, m, CH-Ar); 7.28-7.32 (2H, m, 2CH-Ar); 7.36-7.40 (2H, m, 2CH-Ar); 7.43-7.51 (3H, m, 3CH-Ar); 7.55-7.58 (1H, m, CH-Ar); 8.01-8.03 (1H, m, CH-Ar); 10.43 (1H, s, NHCO). ^{13}C NMR (δ, ppm): 34.45, 36.27, 40.96, 45.22, 109.00, 117.60, 119.03, 120.24, 121.25, 121.50, 124.50, 126.97, 127.17, 128.20, 130.57, 130.90, 133.55, 137.01, 139.07, 141.14, 148.45, 168.41, 169.25. HRMS: m/z calcd for $C_{25}H_{20}Cl_2N_4O_2$, 479,1037; found, 479.1039.

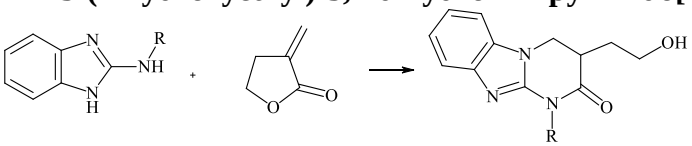


2-[2-Oxo-1-(2-pyridylmethyl)-3,4-dihydropyrimido[1,2-a]benzimidazol-3-yl]-N-(p-tolyl)acetamide **4j.** Yield 65%, colorless powdery compound, m.p. 247-249 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 2.22 (3H, s, CH_3); 2.73 (1H, dd, *J* = 15.9, *J* = 6.8, CH_2CO); 2.94 (1H, dd, *J* = 15.9, *J* = 4.8, CH_2CO); 3.56-3.66 (1H, m, CH(3)); 4.14 (1H, t, *J* = 11.8, CH_2(4)); 4.61 (1H, dd, *J* = 11.8, *J* = 7.5, CH_2(4)); 5.26 (2H, q, *J* = 37.9, *J* = 16.6); 7.02-7.15 (4H, m, 3CH-Ar + CH-Py); 7.17-7.24 (1H, m, CH-Py); 7.26-7.32 (1H, m, CH-Ar); 7.35-7.45 (4H, m, 3CH-Ar + CH-Py); 7.64-7.71 (1H, m, CH-Py); 8.39-8.44 (1H, m, CH-Py); 10.00 (1H, s, NHCO). ^{13}C NMR (δ, ppm): 20.50, 34.37, 36.47, 41.19, 46.69, 108.99, 117.58, 119.24, 120.47, 121.34, 121.58, 122.20, 128.99, 132.15, 133.58, 136.47, 136.76, 141.15, 148.54, 148.84, 155.87, 168.40, 168.62. HRMS: m/z calcd for $C_{25}H_{23}N_5O_2$, 426,1926; found, 426.1927.

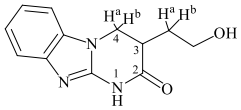


N-(3,4-Dichlorophenyl)-2-[2-oxo-1-(2-pyridylmethyl)-3,4-dihydropyrimido[1,2-a]benzimidazol-3-yl]acetamide **4k.** Yield 75%, colorless powdery compound, m.p. 226-227 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 2.78 (1H, dd, *J* = 16.3, *J* = 6.8, CH_2CO); 3.00 (1H, dd, *J* = 16.4, *J* = 5.3, CH_2CO); 3.63-3.70 (1H, m, CH(3)); 4.16 (1H, t, *J* = 11.9, CH_2(4)); 4.63 (1H, dd, *J* = 12.1, *J* = 7.4, CH_2(4)); 5.20 (2H, q, *J* = 25.1, *J* = 15.3, NCH_2); 7.12-7.20 (2H, m, 2CH-Ar); 7.22-7.26 (1H, m, CH-Ar); 7.28-7.32 (2H, m, 2CH-Ar); 7.36-7.40 (2H, m, 2CH-Ar); 7.43-7.51 (3H, m, 3CH-Ar); 7.55-7.58 (1H, m, CH-Ar); 8.01-8.03 (1H, m, CH-Ar); 10.43 (1H, s, NHCO). ^{13}C NMR (δ, ppm): 34.45, 36.27, 40.96, 45.22, 109.00, 117.60, 119.03, 120.24, 121.25, 121.50, 124.50, 126.97, 127.17, 128.20, 130.57, 130.90, 133.55, 137.01, 139.07, 141.14, 148.45, 168.41, 169.25. HRMS: m/z calcd for $C_{25}H_{20}Cl_2N_4O_2$, 479,1037; found, 479.1039.

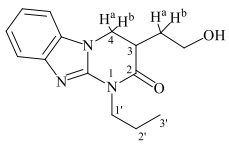
General procedure for the synthesis of 1-*R*-3-(2-hydroxyethyl)-3,4-dihydro-1*H*-pyrimido[1,2-*a*]benzimidazol-2-ones.



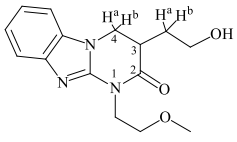
A solution of 0.005 mol 2-*R*-aminobenzimidazole and 0.8 g (0.008 mol) α-methylene-γ-butyrolactone in isopropyl alcohol (30 ml) is boiled for 2-3 hours. The solvent is removed under vacuum, the yellowish oil is dissolved in 20-30 ml of xylene. When standing or seeding, crystals are released, which are filtered and dried.



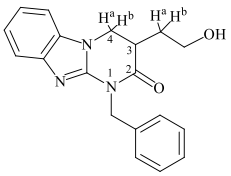
3-(2-Hydroxyethyl)-3,4-dihydro-1H-pyrimido[1,2-a]benzimidazol-2-one ***7a.*** Yield 70%, colorless powdery compound, m.p. 198-200 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 1.55-1.67 (1H, m, CH_2^a); 2.03-2.15 (1H, m, CH_2^b); 2.95-3.05 (1H, m, CH(3)); 3.60 (2H, t, *J* = 5.9, OCH_2); 3.94 (1H, dd, *J* = 12.4, *J* = 9.9, CH_2^a(4)); 4.45 (1H, dd, *J* = 12.4, *J* = 6.7, CH_2^b(4)); 4.61 (1H, s, OH); 7.05-7.15 (2H, m, 2CH-Ar); 7.31-7.44 (2H, m, 2CH-Ar); 11.57 (1H, s, CONH). ^{13}C NMR (δ, ppm): 30.90, 36.70, 41.54, 58.19, 108.69, 117.17, 120.62, 121.27, 132.83, 141.65, 147.83, 170.42. HRMS: m/z calcd for $C_{12}H_{13}N_3O_2$, 232.1081; found, 232.1080.



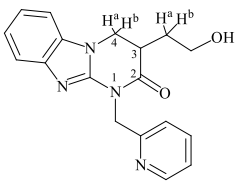
3-(2-Hydroxyethyl)-1-propyl-3,4-dihydropyrimido[1,2-a]benzimidazol-2-one ***7b.*** Yield 45%, colorless powdery compound, m.p. 142-143 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 0.87-0.99 (3H, m, CH_3(3')); 1.55-1.76 (3H, m, CH_2(2') + CH_2^a); 2.01-2.13 (1H, m, CH_2^b); 3.02-3.13 (1H, m, CH(3)); 3.56-3.62 (2H, t, *J* = 6.1, OCH_2); 3.84-4.02 (3H, m, NCH_2 + CH_2^a(4)); 4.44 (1H, dd, *J* = 12.4, *J* = 6.4, CH_2^b(4)); 4.61 (1H, s, OH); 7.05-7.18 (2H, m, 2CH-Ar); 7.33-7.51 (2H, m, 2CH-Ar). ^{13}C NMR (δ, ppm): 11.09, 20.75, 31.30, 37.06, 40.99, 43.62, 58.06, 108.84, 117.49,121.02, 121.38, 133.49, 141.43, 148.40, 169.05. HRMS: m/z calcd for $C_{15}H_{19}N_3O_2$, 274.1551; found, 274.1553.



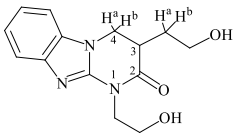
3-(2-Hydroxyethyl)-1-(2-methoxyethyl)-3,4-dihydropyrimido[1,2-a]benzimidazol-2-one ***7c.*** Yield 50%, colorless powdery compound, m.p. 113-114 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 1.56-1.68 (1H, m, CH_2^a); 2.03-2.13 (1H, m, CH_2^b); 3.04-3.14 (1H, m, CH(3)); 3.29 (3H, s, OCH_3); 3.55-3.65 (4H, m, $2OCH_2$); 3.97 (1H, dd, *J* = 12.5, *J* = 3.9, CH_2^a(4)); 3.09-4.23 (2H, m, NCH_2); 4.45 (1H, dd, *J* = 12.5, *J* = 6.5, CH_2^b(4)); 4.61 (1H, s, OH); 7.08-7.19 (2H, m, 2CH-Ar); 7.35-7.50 (2H, m, 2CH-Ar). ^{13}C NMR (δ, ppm): 31.17, 37.11, 40.93, 41.02, 58.02, 58.05, 68.52, 108.84, 117.55, 121.05, 121.40, 133.49, 141.34, 148.30, 169.12. HRMS: m/z calcd for $C_{15}H_{19}N_3O_3$, 290.1500; found, 290.1498.



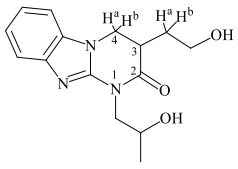
1-Benzyl-3-(2-hydroxyethyl)-3,4-dihydropyrimido[1,2-a]benzimidazol-2-one ***7d.*** Yield 45%, colorless powdery compound, m.p. 121-122 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 1.59-1.71 (1H, m, CH_2^a); 2.05-2.17 (1H, m, CH_2^b); 3.11-3.21 (1H, m, CH(3)); 3.55-3.64 (2H, m, OCH_2); 4.02 (1H, t, *J* = 10.1, CH_2^a(4)); 4.51 (1H, dd, *J* = 12.4, *J* = 6.4, CH_2^b(4)); 4.60 (1H, s, OH); 5.18 (2H, s, NCH_2); 7.05-7.17 (2H, m, 2CH-Ar); 7.18-7.24 (1H, m, CH-Ar); 7.25-7.31 (2H, m, 2CH-Ar); 7.35-7.43(3H, m, 3CH-Ar); 7.43-7.47 (1H, m, CH-Ar). ^{13}C NMR (δ, ppm): 31.12, 37.17, 41.02, 45.23, 58.05, 108.90, 117.64, 121.11, 121.42, 126.99, 127.53, 128.16, 133.66, 137.12, 141.33,148.42, 169.08. HRMS: m/z calcd for $C_{19}H_{19}N_3O_2$, 322.1551; found, 322.1551.



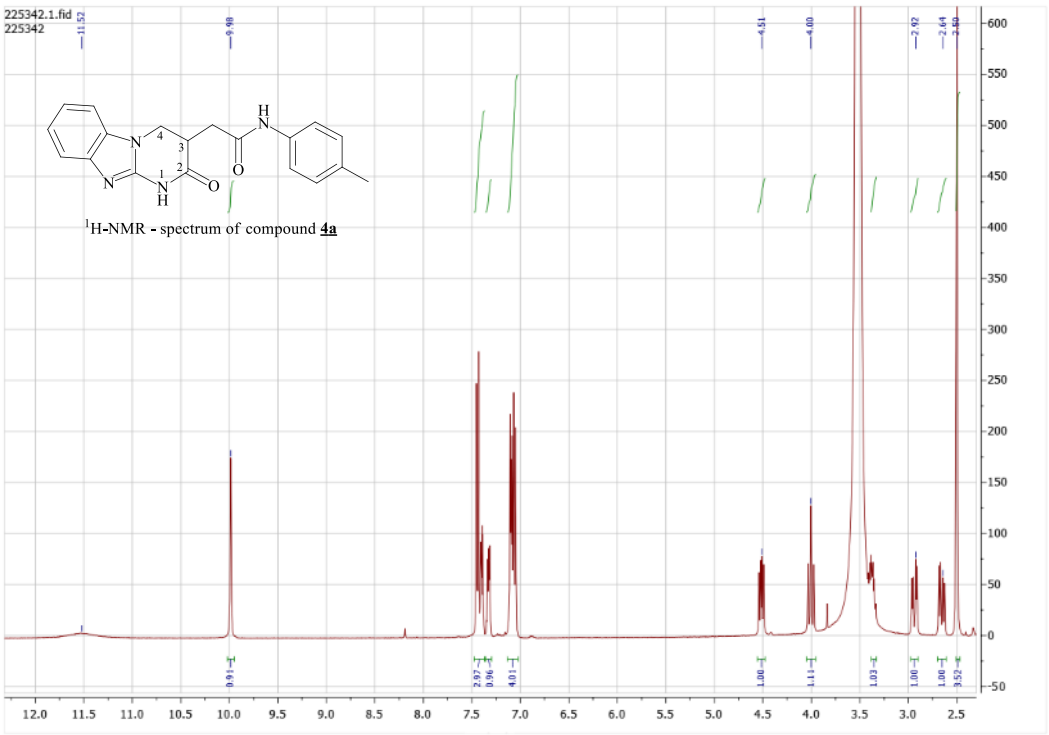
3-(2-Hydroxyethyl)-1-(2-pyridylmethyl)-3,4-dihydropyrimido[1,2-a]-benzimidazol-2-one ***7e.*** Yield 65%, colorless powdery compound, m.p. 156-157 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 1.64-1.76 (1H, m, C H_2^a); 2.10-2.21 (1H, m, CH_2^b); 3.19-3.29 (1H, m, CH(3)); 3.57-3.68 (2H, m, OCH_2); 4.09 (1H, dd, *J* = 12.3, *J* = 9.6, CH_2^a(4)); 4.56 (1H, dd, *J* = 12.4, *J* = 6.4, CH_2^b(4)); 4.66 (1H, s, OH); 5.29 (2H, q, *J* = 27.0, *J* = 16.2, NCH_2); 7.05-7.17 (2H, m, 2CH-Ar); 7.18-7.24 (1H, m, CH-Ar); 7.25-7.31 (1H, m, CH-Py); 7.35-7.46(2H, m, CH-Ar + CH-Py); 7.65-7.42 (1H, m, CH-Py); 8.41-8.46 (1H, m, CH-Py). ^{13}C NMR (δ, ppm): 31.22, 37.17, 41.11, 46.62, 58.08, 108.95, 117.56, 120.46, 121.11, 121.42, 122.05, 133.70, 136.58, 141.33, 148.62, 148.83, 155.97, 169.29. HRMS: m/z calcd for $C_{18}H_{18}N_4O_2$, 323.1503; found, 323.1503.



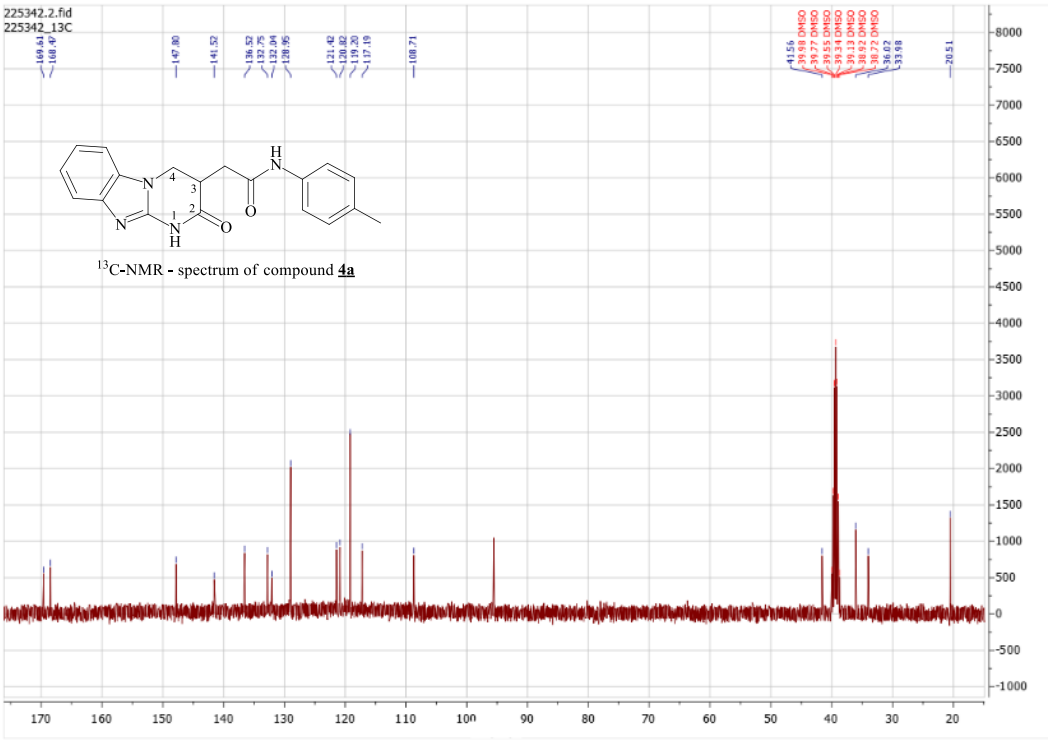
1,3-Bis(2-hydroxyethyl)-3,4-dihydropyrimido[1,2-a]benzimidazol-2-one ***7f.*** Yield 95%, colorless powdery compound, m.p. 141-142 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 1.56-1.68 (1H, m, C H_2^a); 2.04-2.16 (1H, m, CH_2^b); 3.04-3.14 (1H, m, CH(3)); 3.56-3.70 (4H, m, *J* = 5.9, OCH_2 + OCH_2); 3.96 (1H, dd, *J* = 12.4, *J* = 9.6, CH_2^a(4)); 4.01-4.16 (2H, m, NCH_2); 4.45 (1H, dd, *J* = 12.4, *J* = 6.5, CH_2^b(4)); 4.60 (1H, s, OH); 4.75 (1H,s, OH); 7.07-7.18 (2H, m, 2CH-Ar); 7.33-7.48 (2H, m, 2CH-Ar). ^{13}C NMR (δ, ppm): 31.24, 37.14, 39.14, 41.04, 44.24, 108.73, 117.46, 120.97, 121.33, 133.45, 141.33, 148.59, 169.18. HRMS: m/z calcd for $C_{14}H_{17}N_3O_3$, 276.1344; found, 276.1345.



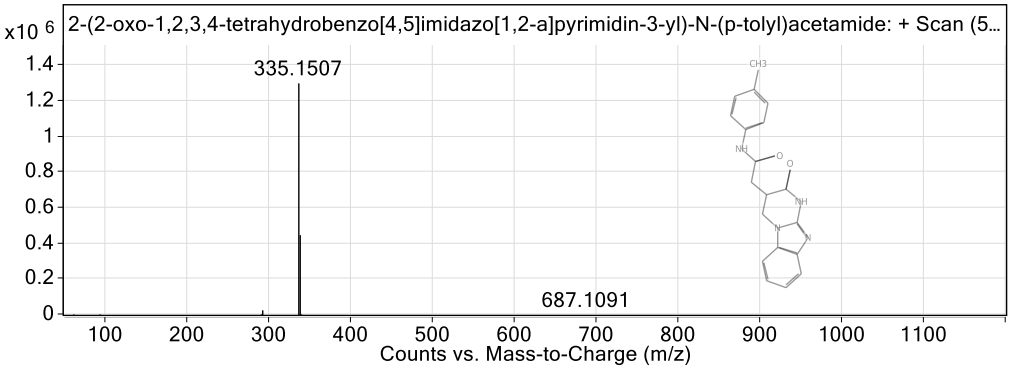
3-(2-Hydroxyethyl)-1-(2-hydroxypropyl)-3,4-dihydropyrimido[1,2-a]-benzimidazol-2-one ***7g.*** Yield 75%, colorless powdery compound, m.p. 127-128 °C. ^{1}H NMR, ((δ, ppm., *J*/Hz): 1.06-1.15 (3H, m, CH_3(3')); 1.56-1.67 (1H, m, C H_2^a); 2.03-2.15 (1H, m, CH_2^b); 3.03-3.17 (1H, m, CH(3)); 3.54-3.65 (2H, m, OCH_2); 3.76-3.89 (1H, m, OCH(2')); 3.94-4.16 (3H, m, CH_2^a(4) + NCH_2); 4.46 (1H, dd, *J* = 12.4, *J* = 6.4, CH_2^b(4)); 4.62 (1H, s, OH); 4.80 (1H,s, OH); 7.07-7.18 (2H, m, 2CH-Ar); 7.34-7.50 (2H, m, 2CH-Ar). ^{13}C NMR (δ, ppm): 20.77, 31.25, 37.21, 40.99, 49.06, 49.15, 58.11, 63.53, 63.60, 108.82, 117.44, 121.02, 121.39, 133.44, 141.22, 148.84, 169.39. HRMS: m/z calcd for $C_{15}H_{19}N_3O_3$, 290.1500; found, 290.1500.

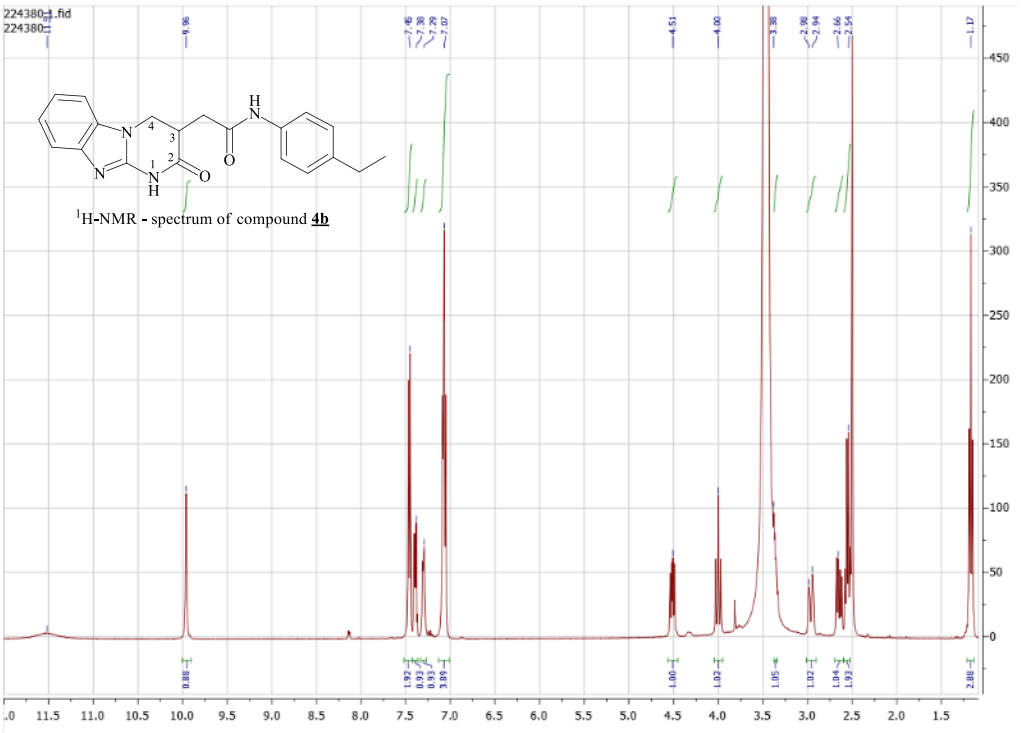


^{1}H-NMR - spectrum of compound **4a**

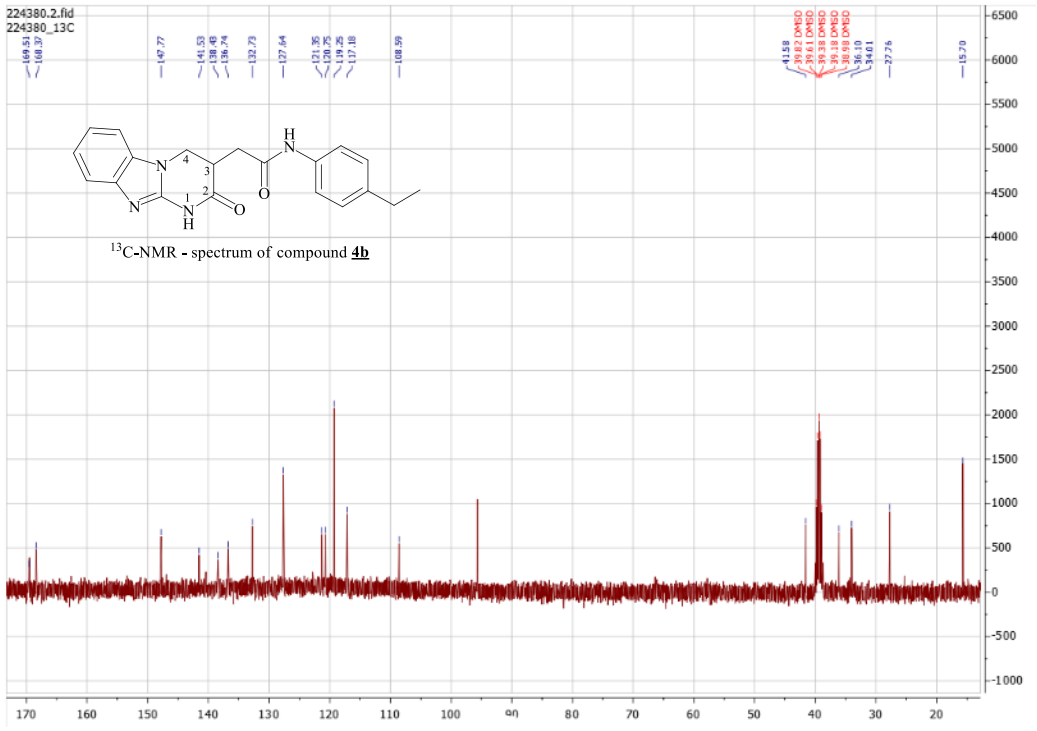


^{13}C-NMR - spectrum of compound **4a**

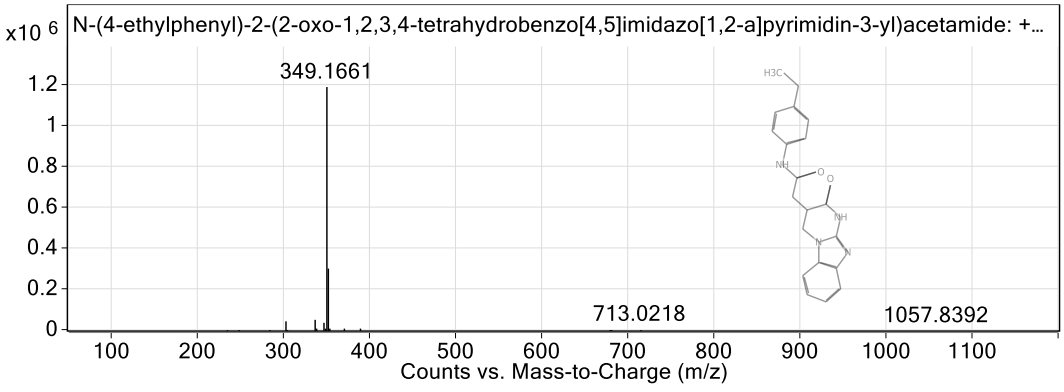


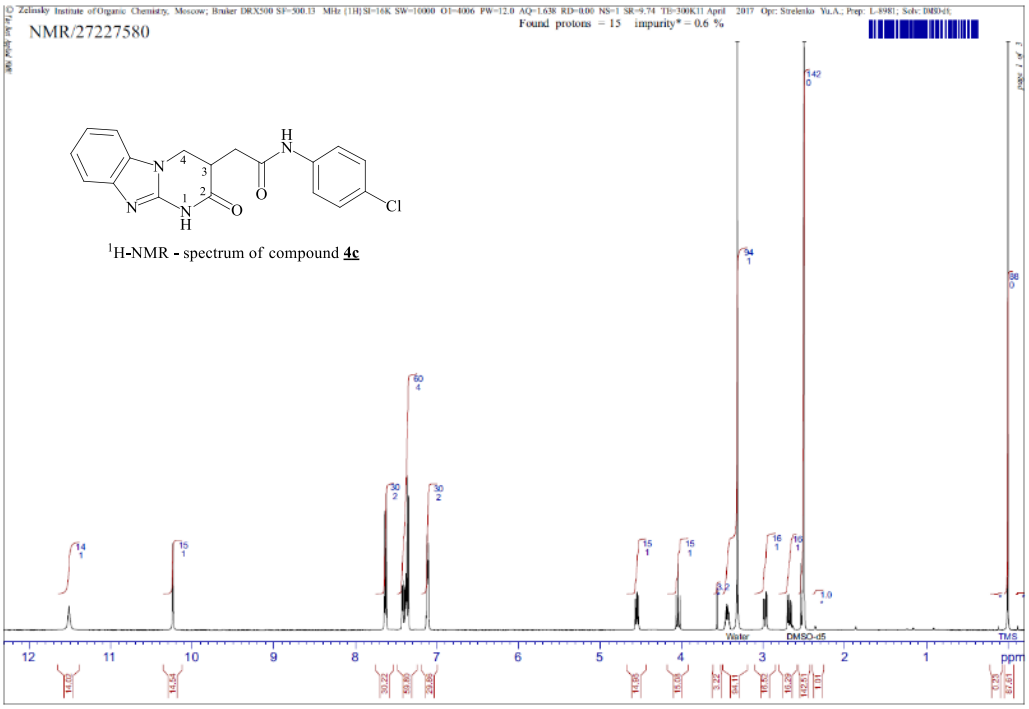


^{1}H-NMR - spectrum of compound **4b**

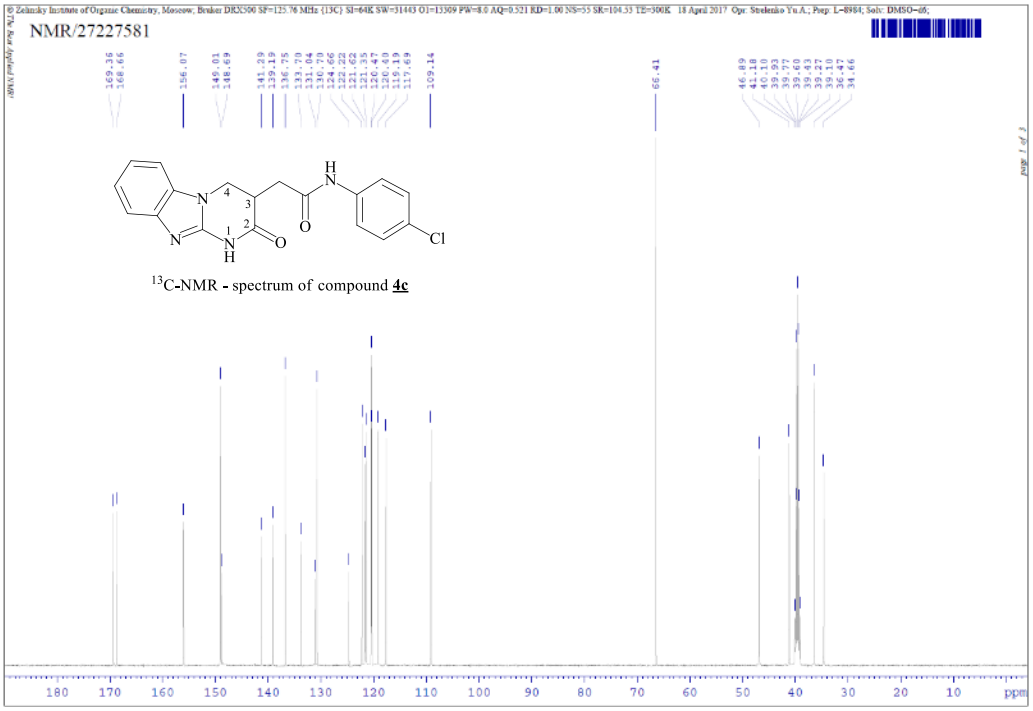


^{13}C-NMR - spectrum of compound **4b**

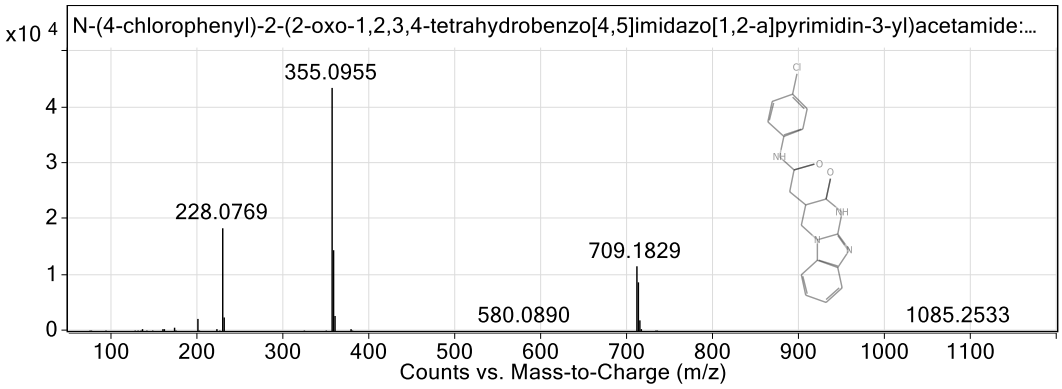


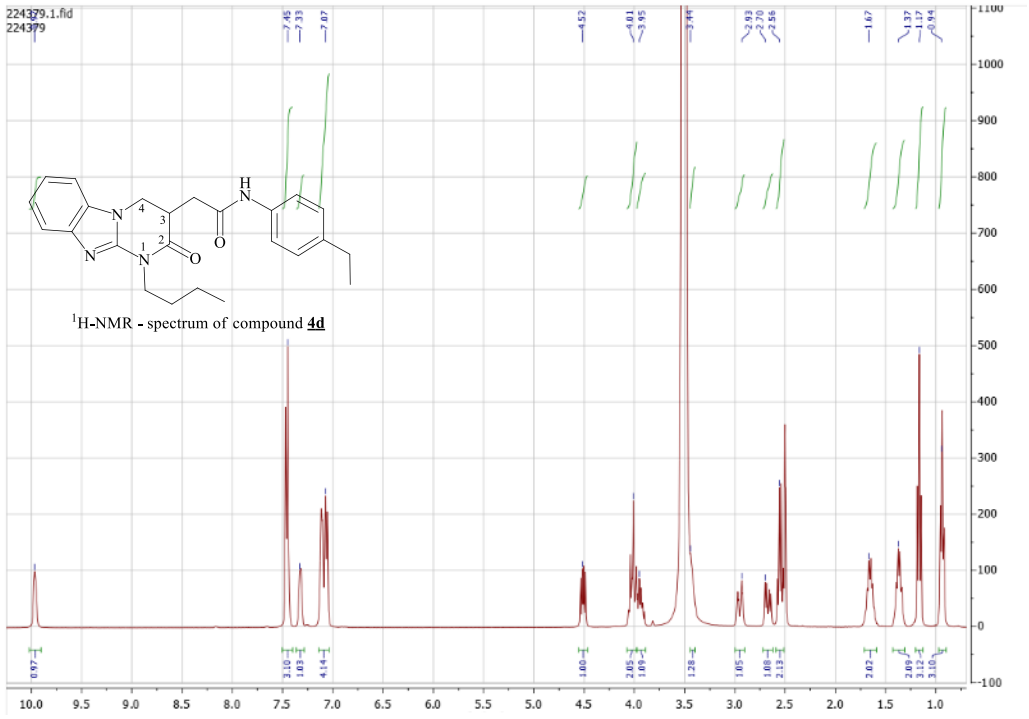


^{1}H-NMR - spectrum of compound **4c**

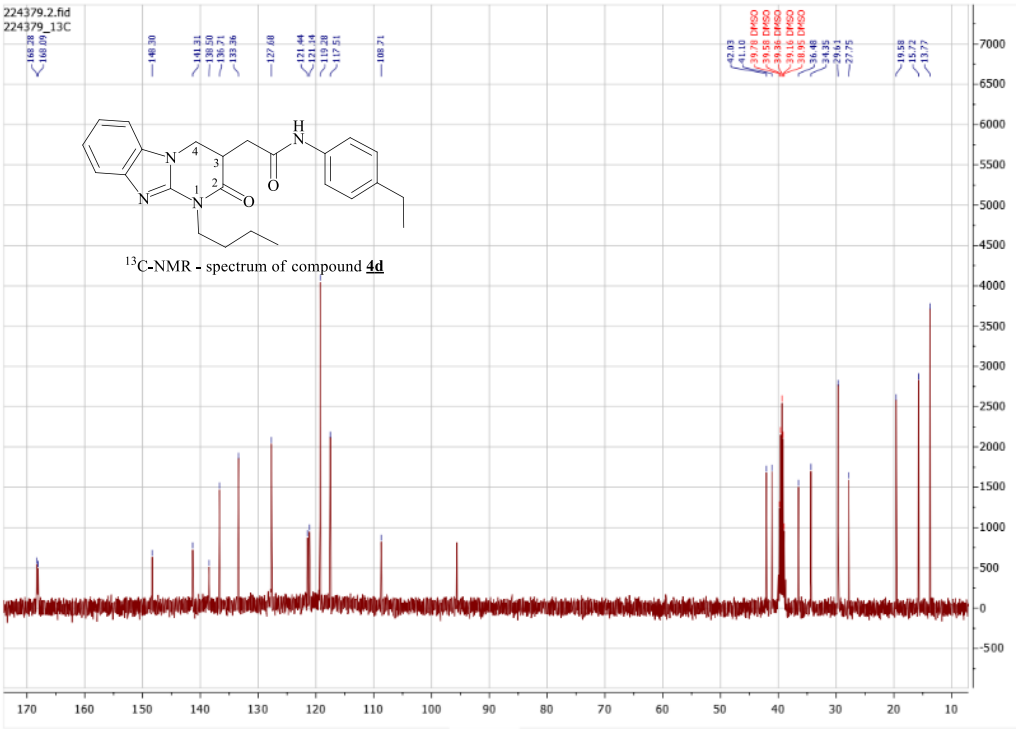


^{13}C-NMR - spectrum of compound **4c**

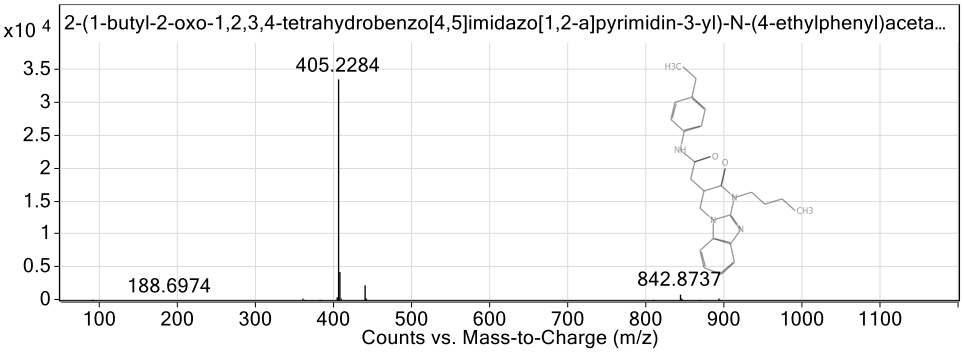


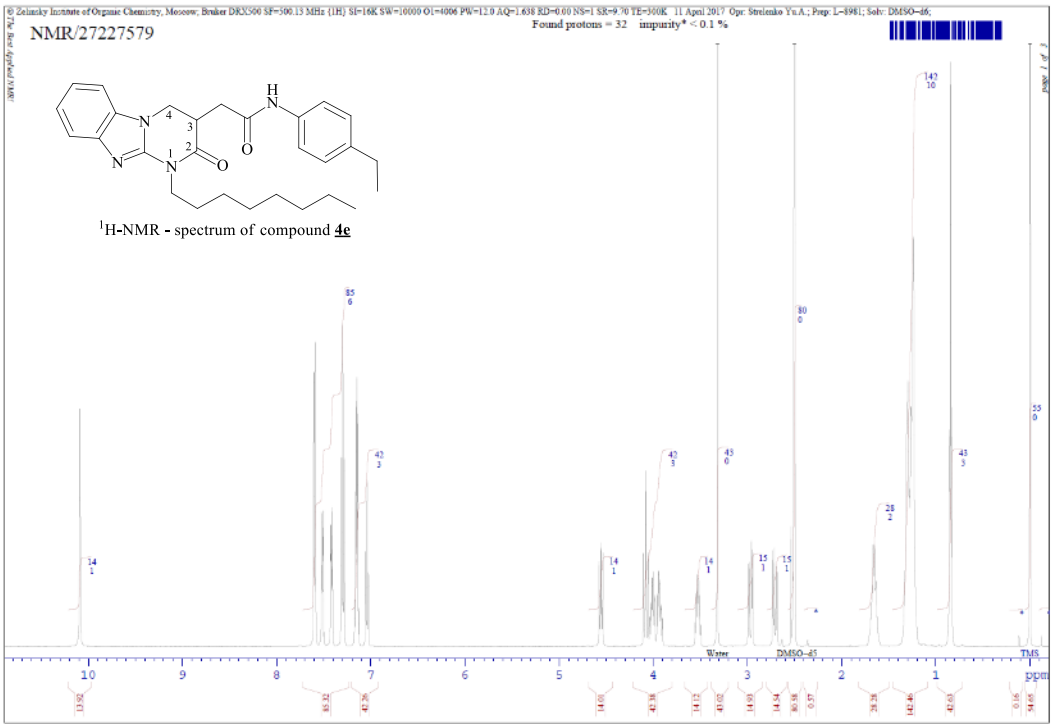


^{1}H-NMR - spectrum of compound **4d**

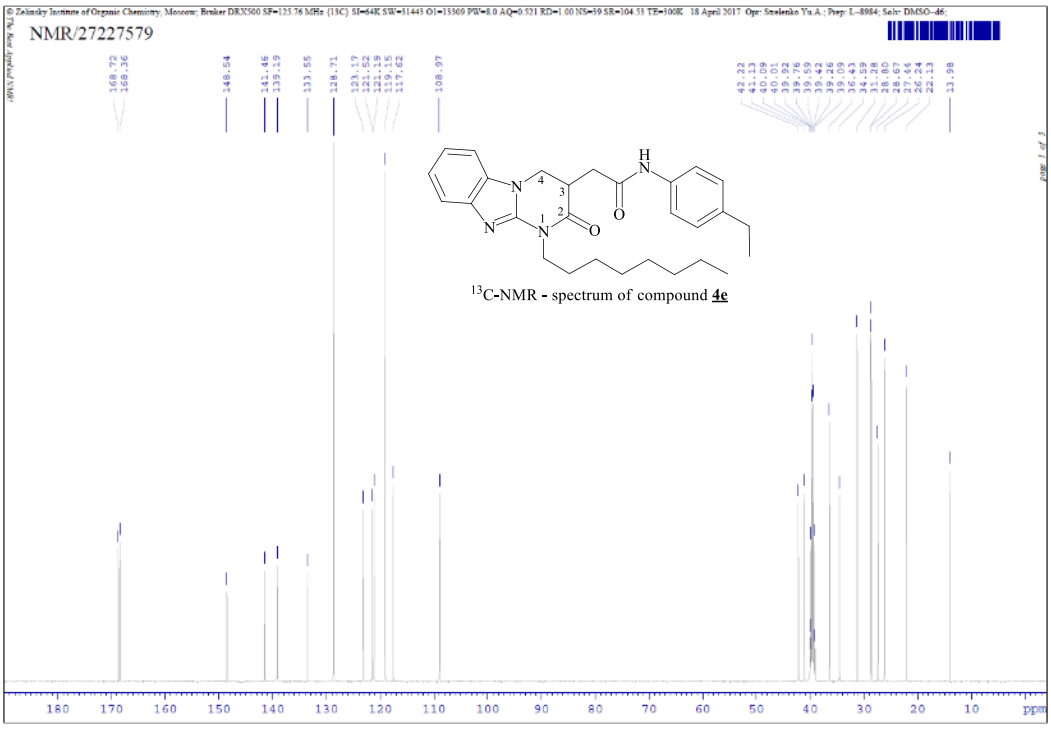


^{13}C-NMR - spectrum of compound **4d**

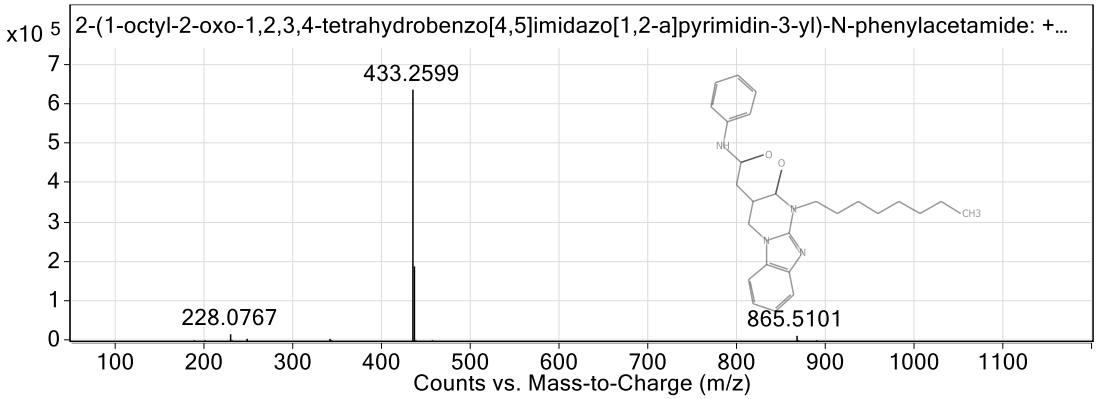


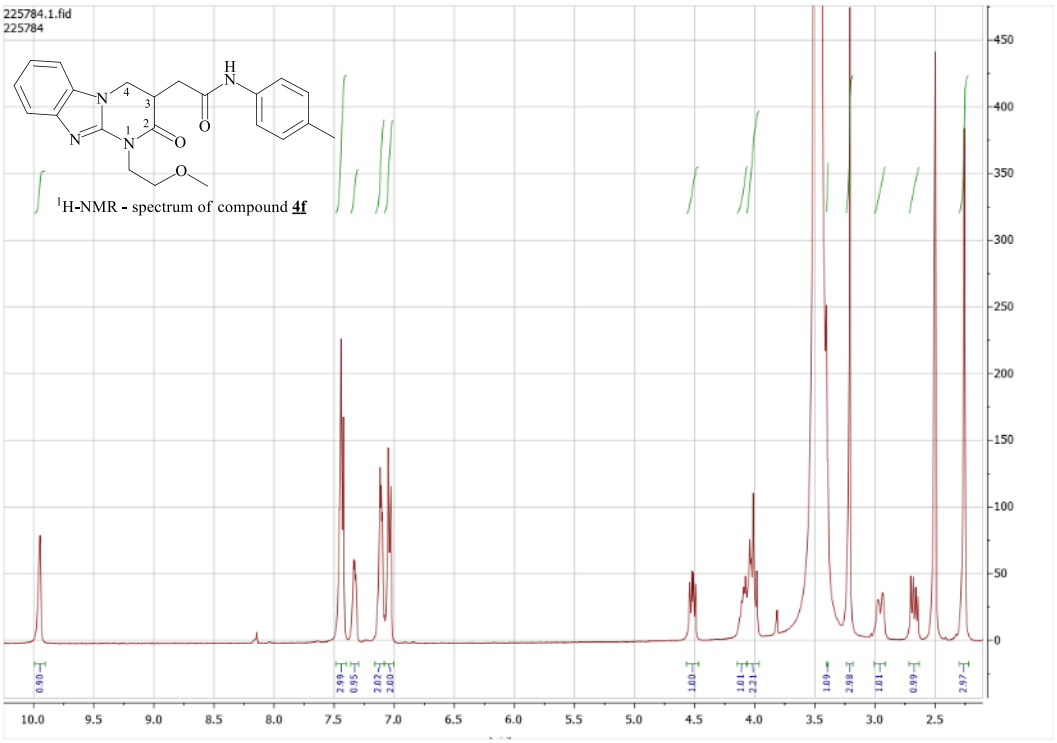


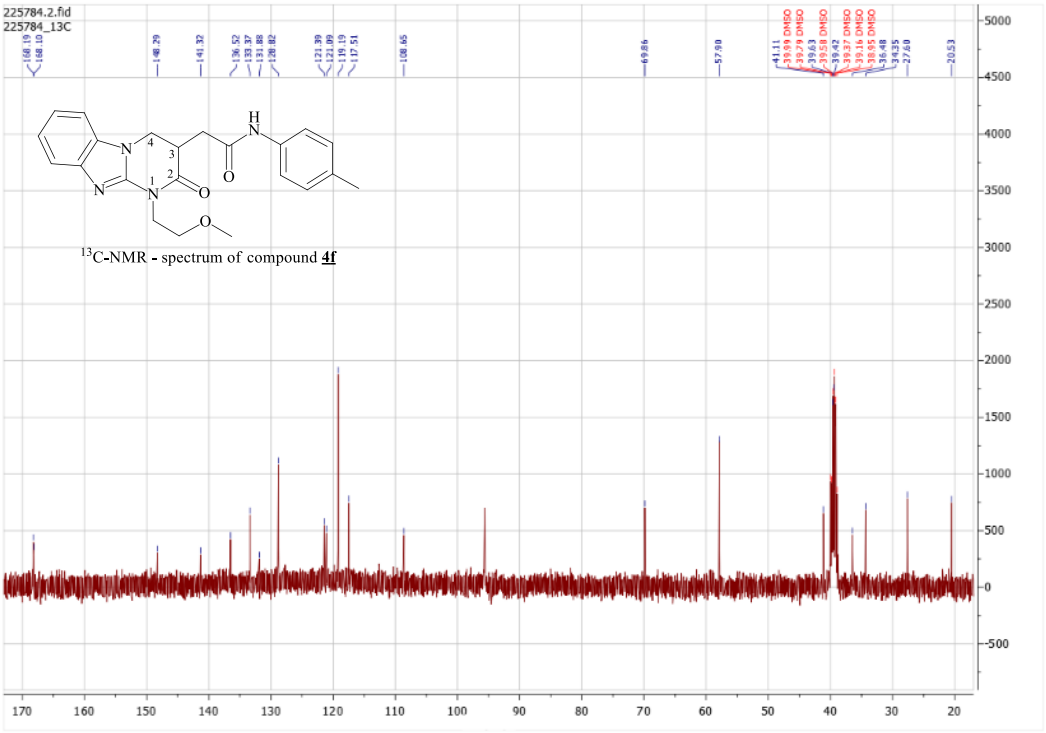
^{1}H-NMR - spectrum of compound **4e**

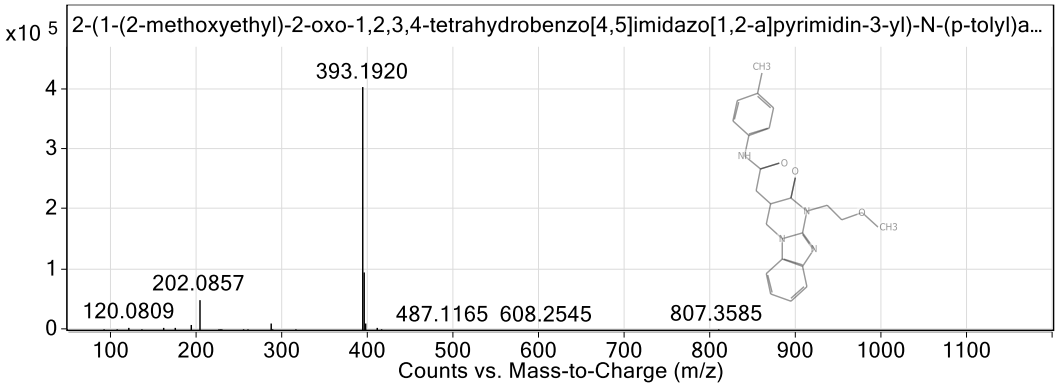


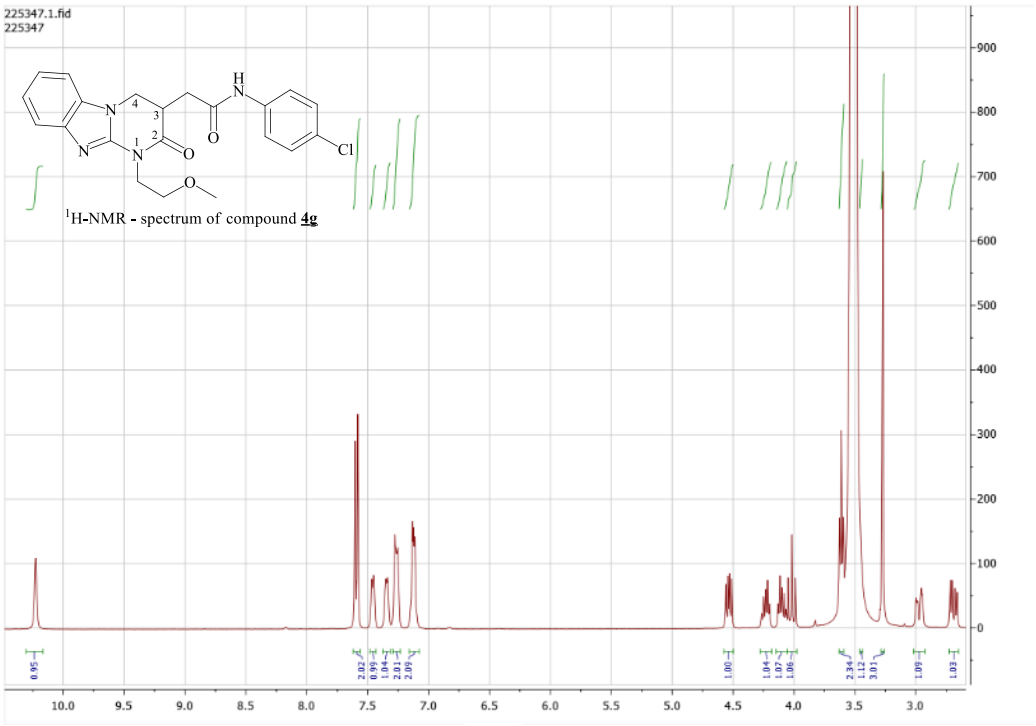
^{13}C-NMR - spectrum of compound **4e**



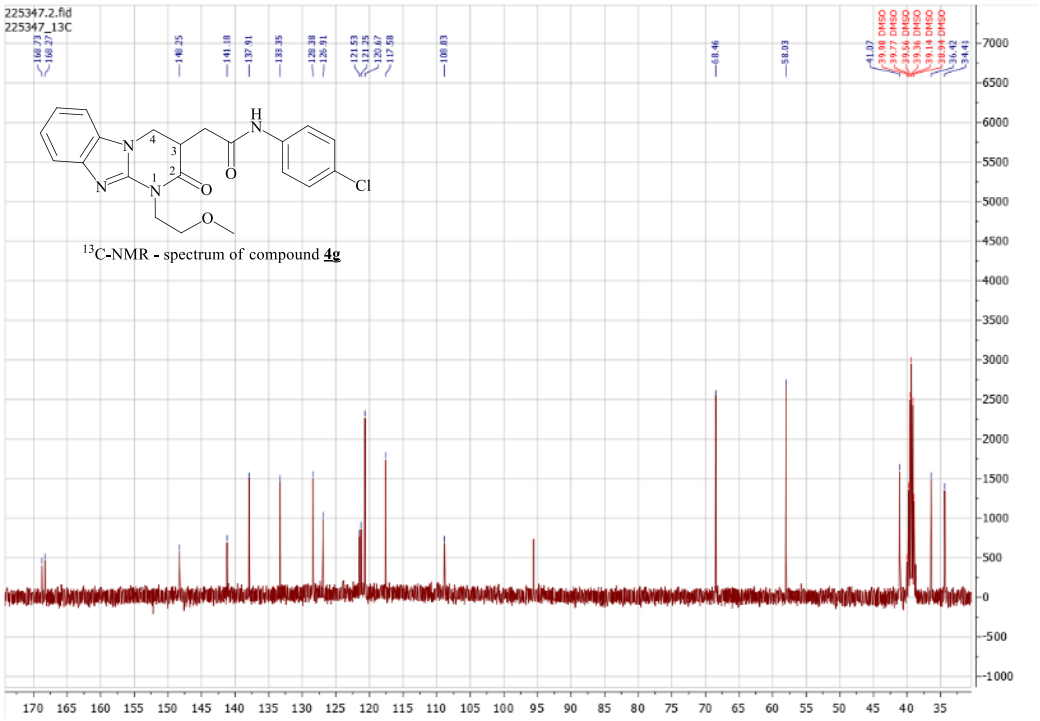

225784.1.fid
225784
^{1}H-NMR - spectrum of compound 4f


225784.2.fid
225784_13C
^{13}C-NMR - spectrum of compound 4f

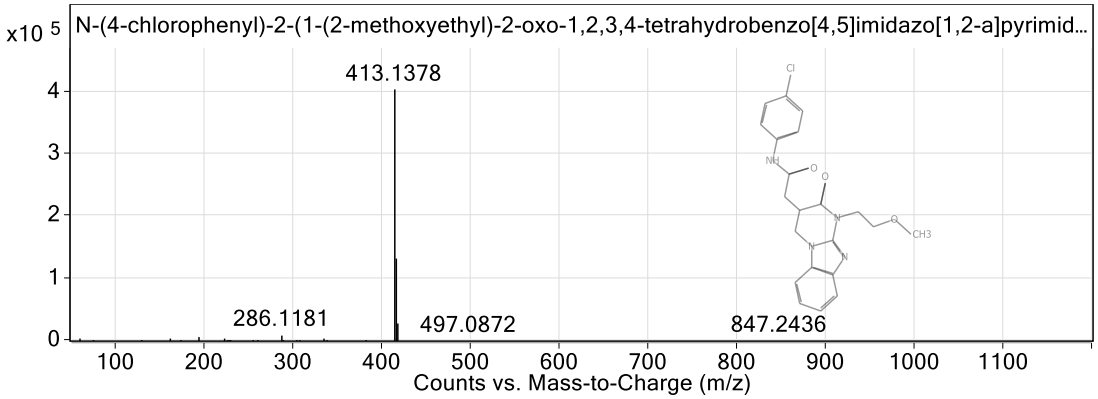

2-(1-(2-methoxyethyl)-2-oxo-1,2,3,4-tetrahydrobenzo[4,5]imidazo[1,2-a]pyrimidin-3-yl)-N-(p-tolyl)a...
x10 5
393.1920
202.0857
120.0809
487.1165
608.2545
807.3585
Counts vs. Mass-to-Charge (m/z)

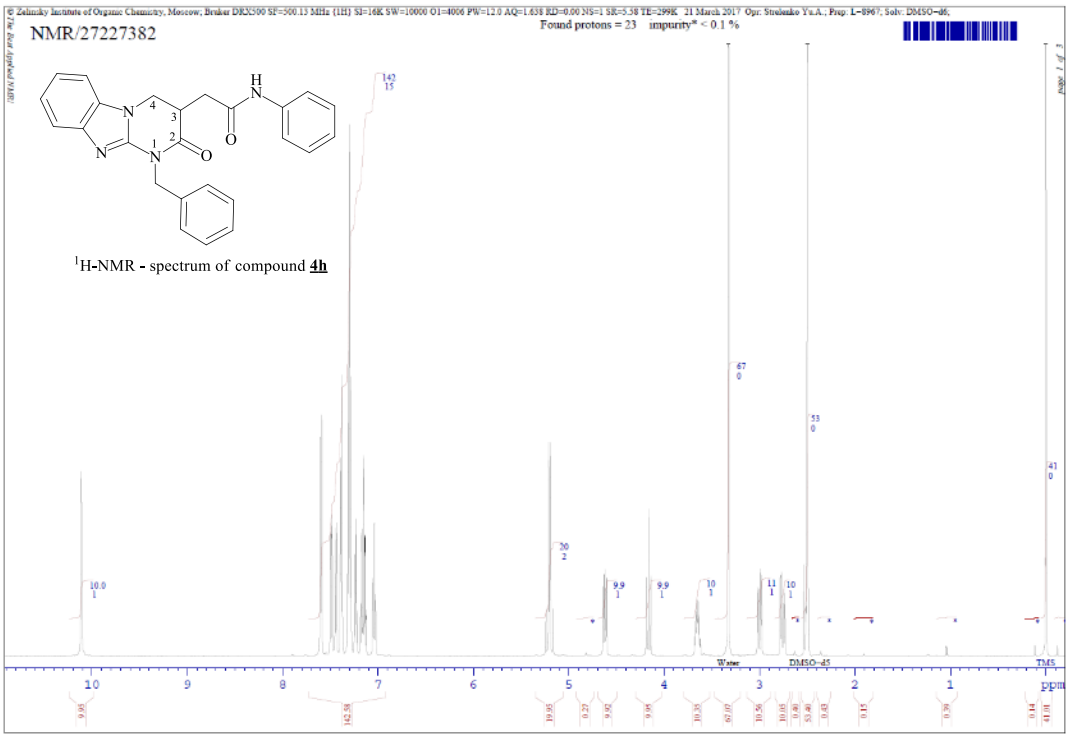


^{1}H-NMR - spectrum of compound **4g**

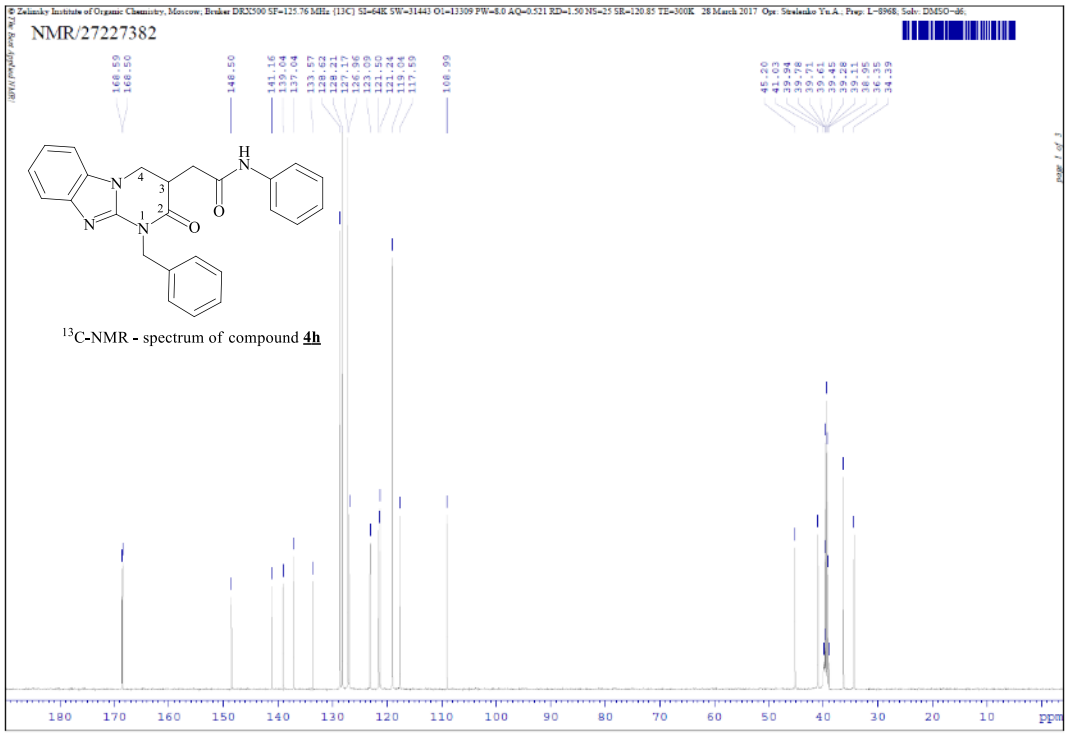


^{13}C-NMR - spectrum of compound **4g**

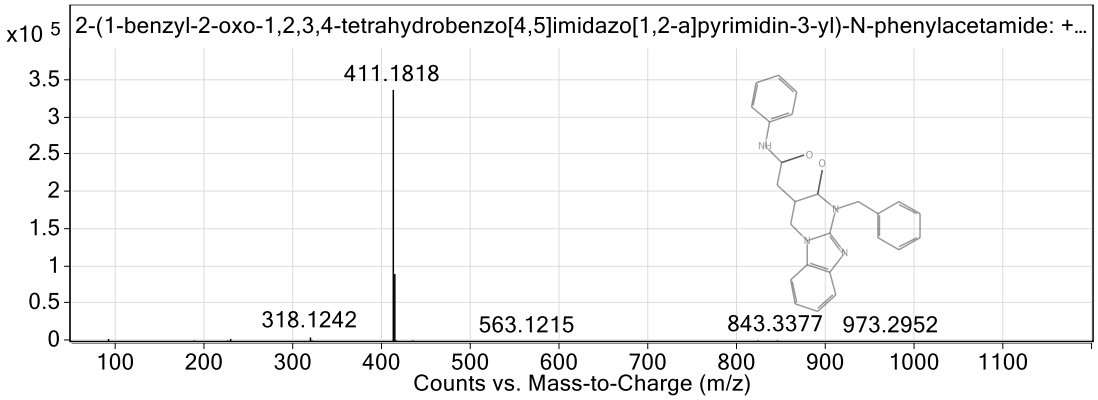


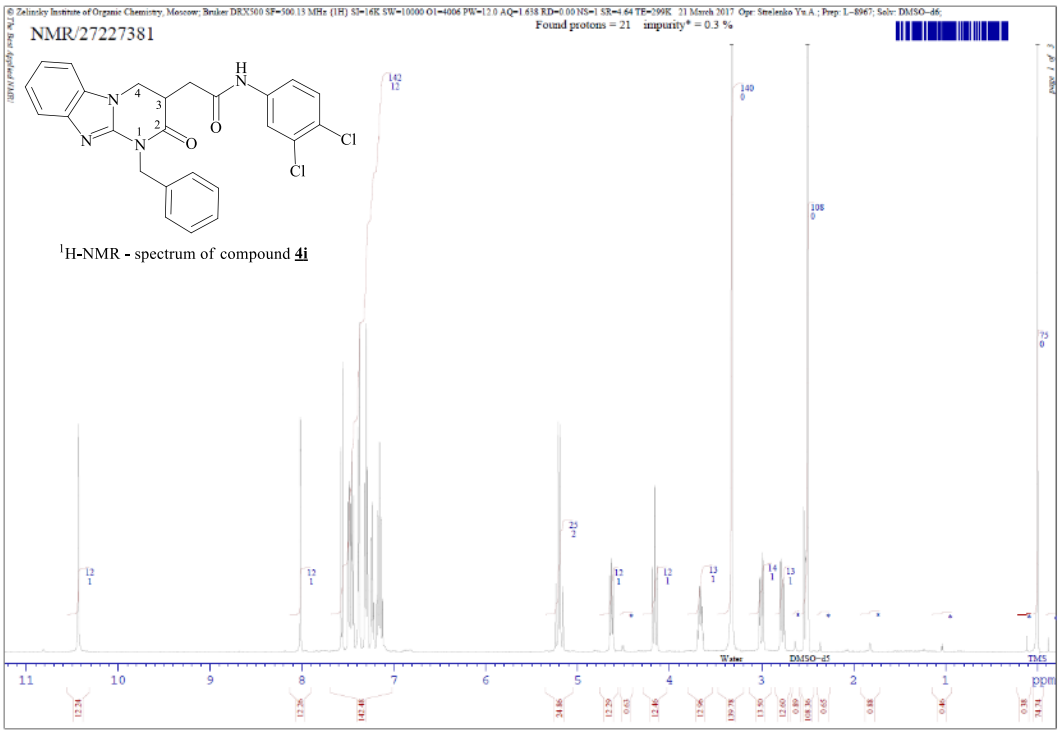


^{1}H-NMR - spectrum of compound **4h**

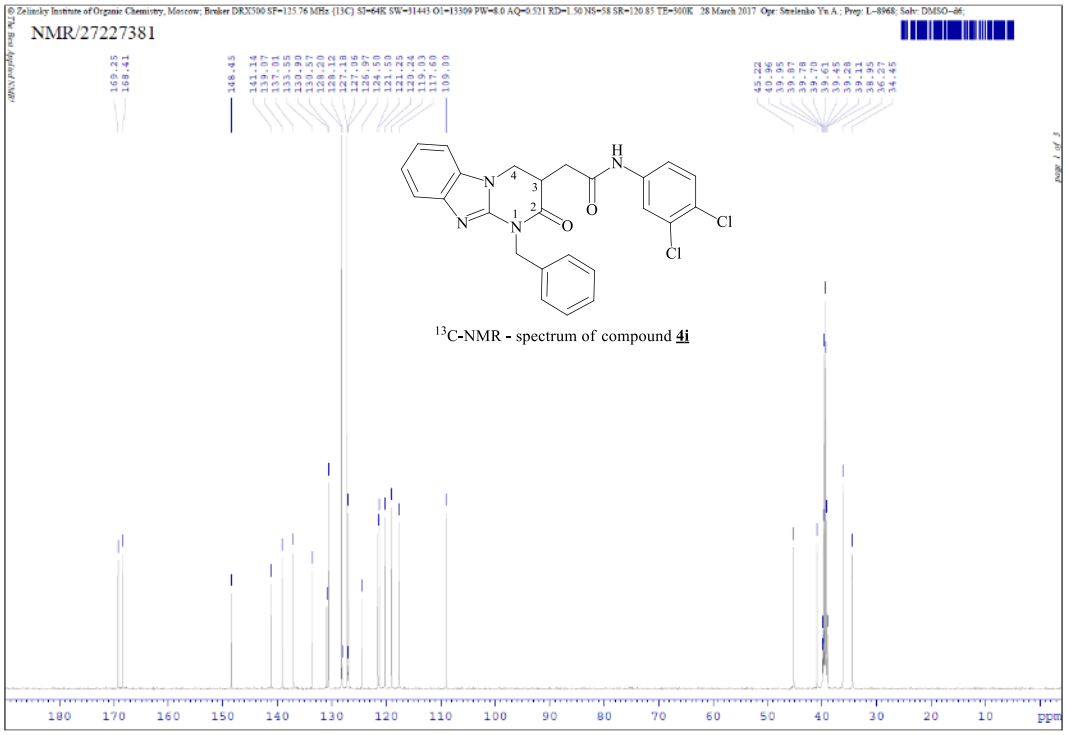


^{13}C-NMR - spectrum of compound **4h**

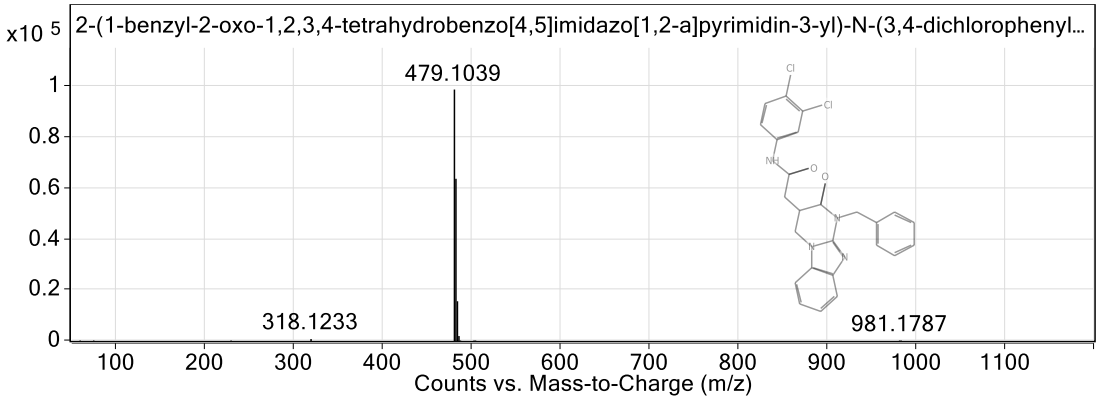


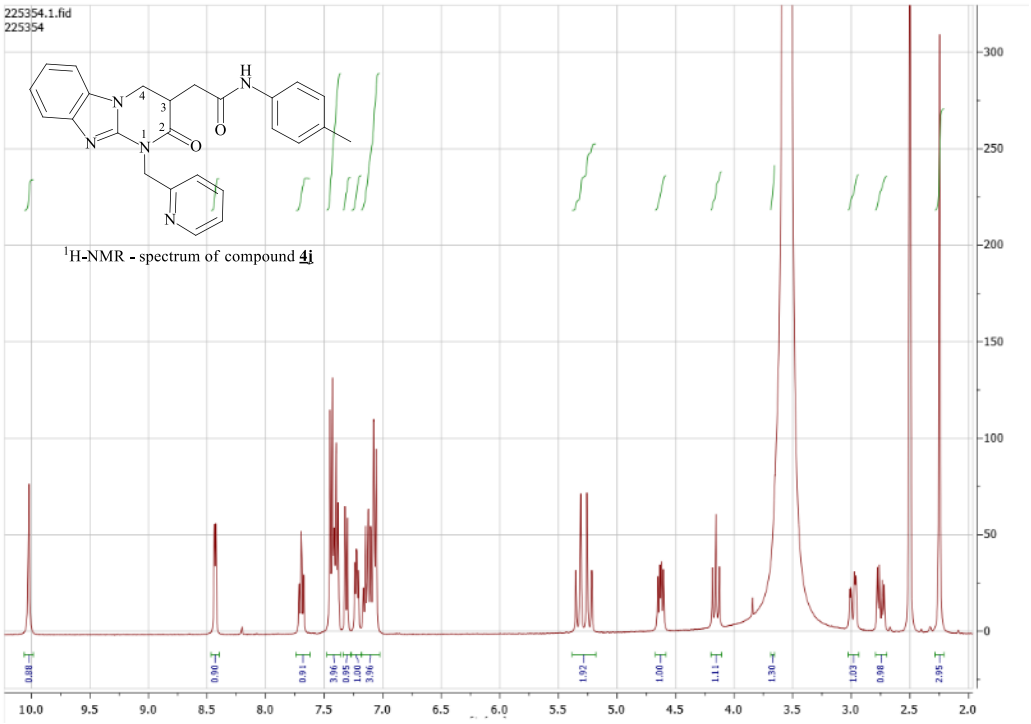


^{1}H-NMR - spectrum of compound **4i**

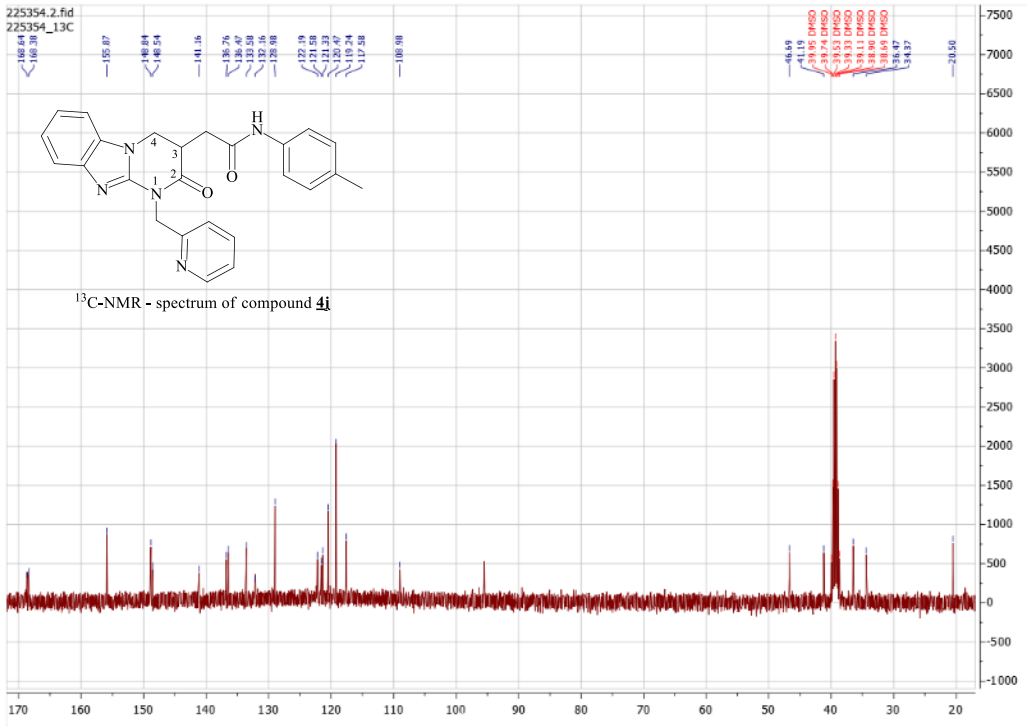


^{13}C-NMR - spectrum of compound **4i**

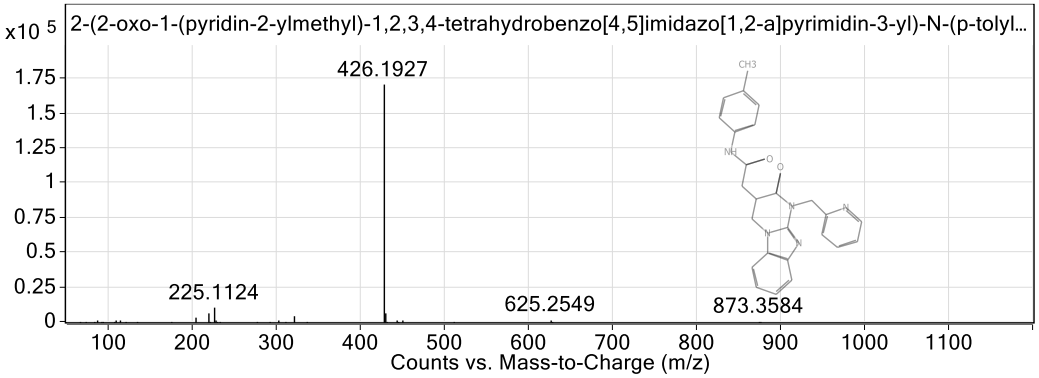


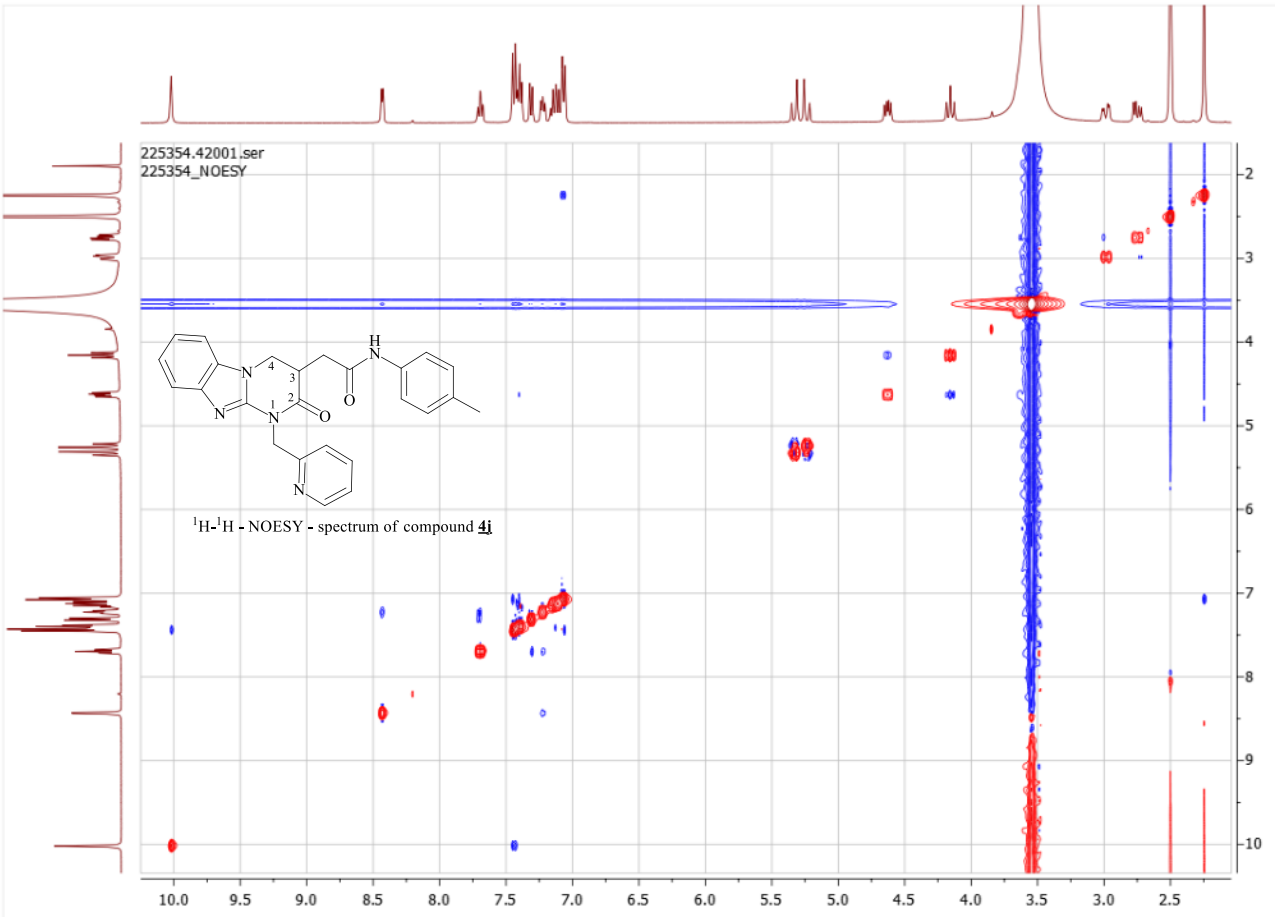


^{1}H-NMR - spectrum of compound **4j**

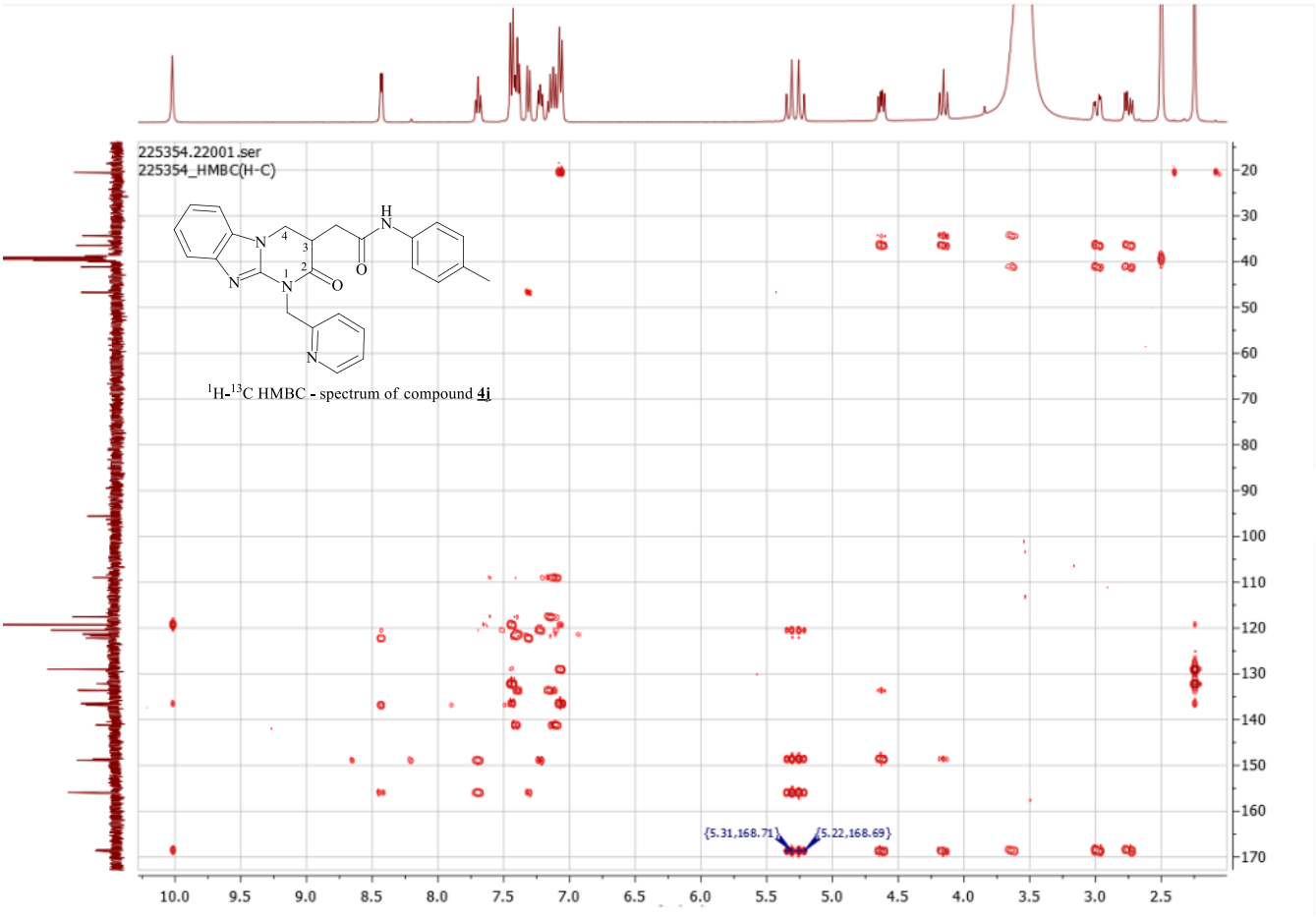


^{13}C-NMR - spectrum of compound **4j**

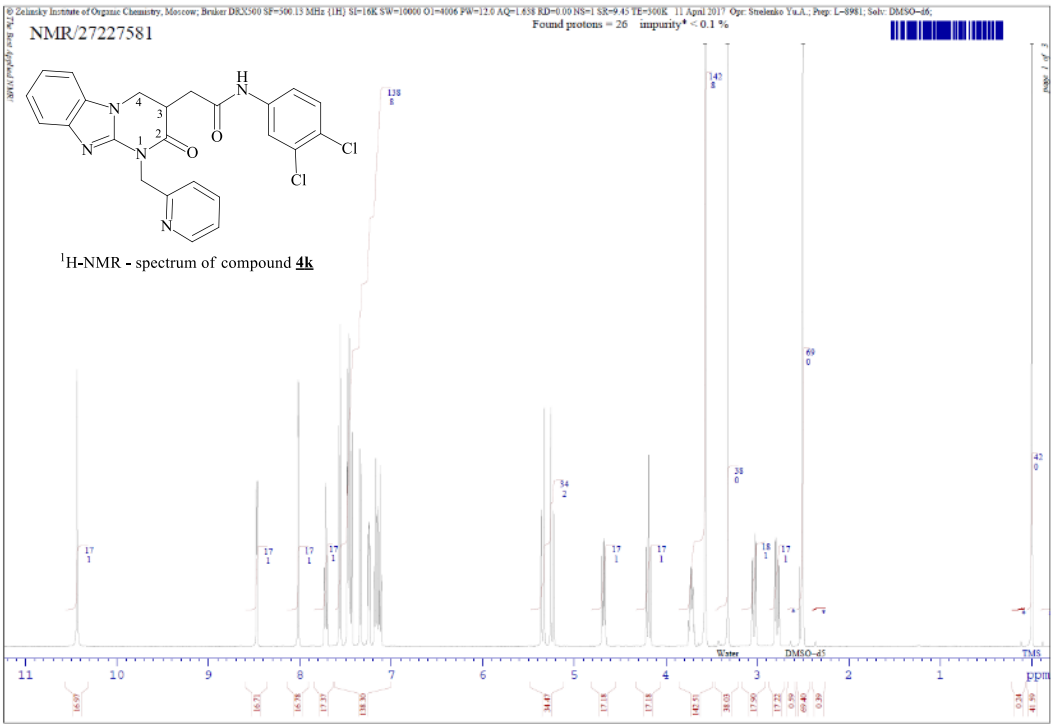




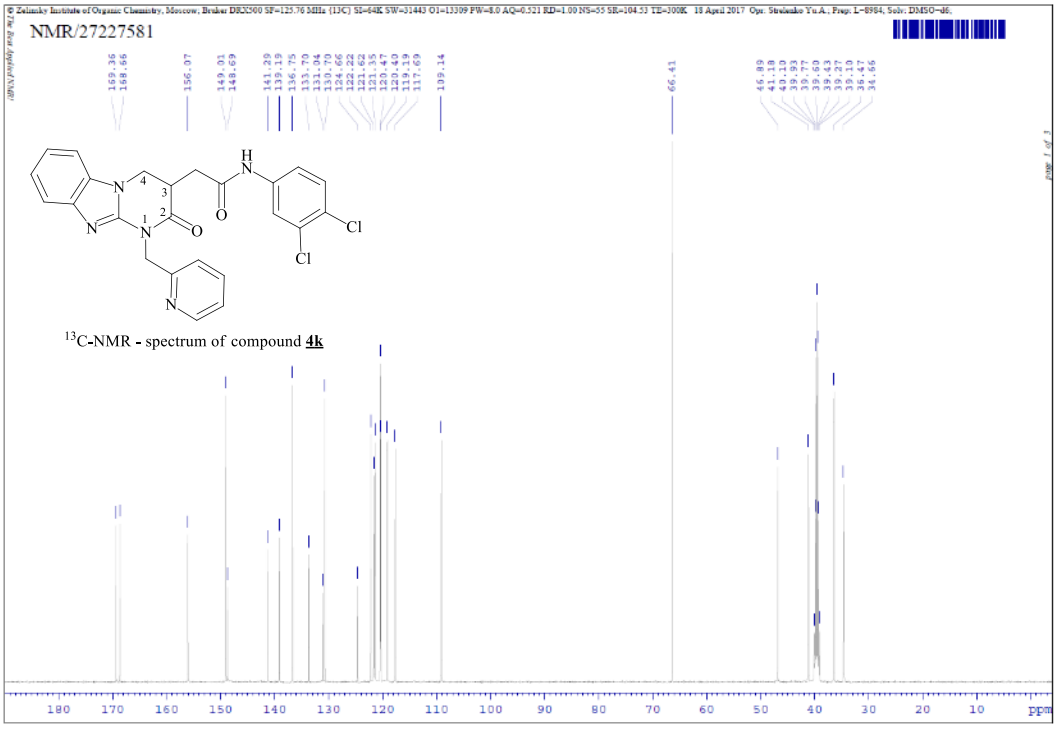
^{1}H-^{1}H - NOESY - spectrum of compound **4i**



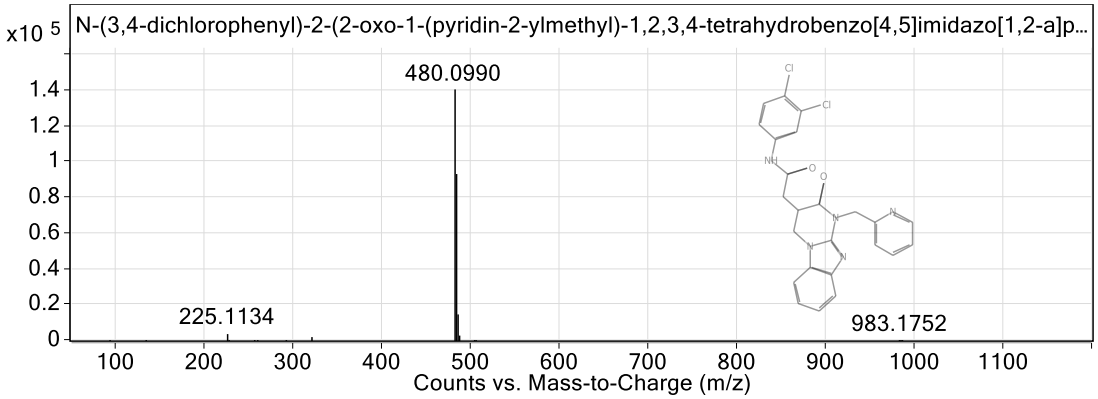
^{1}H-^{13}C HMBC - spectrum of compound **4i**

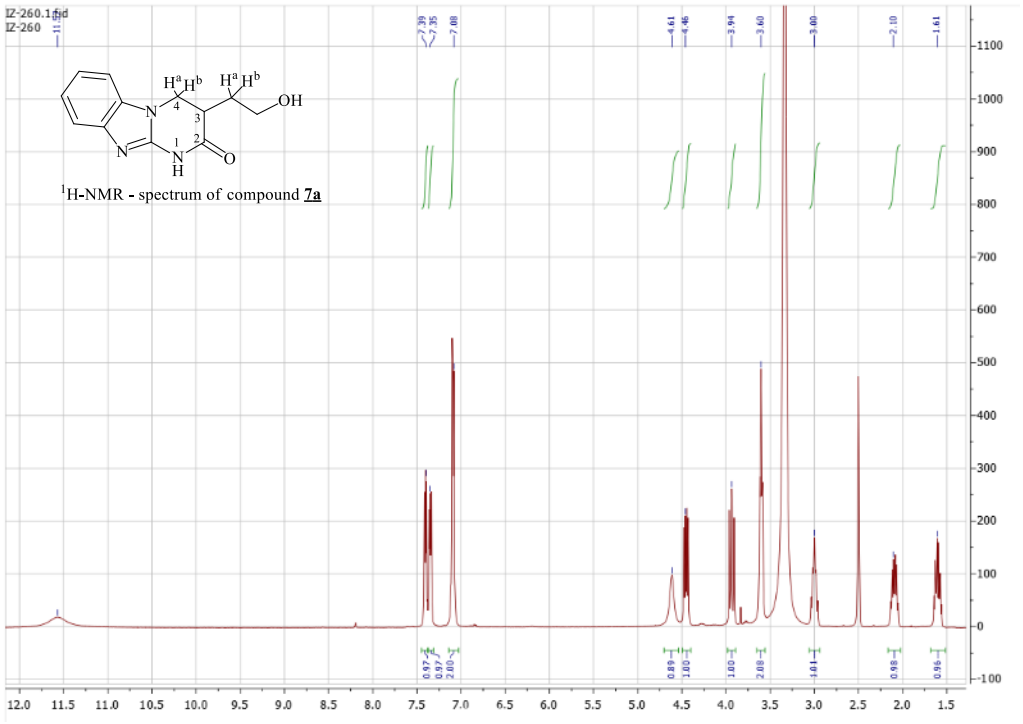


^{1}H-NMR - spectrum of compound **4k**

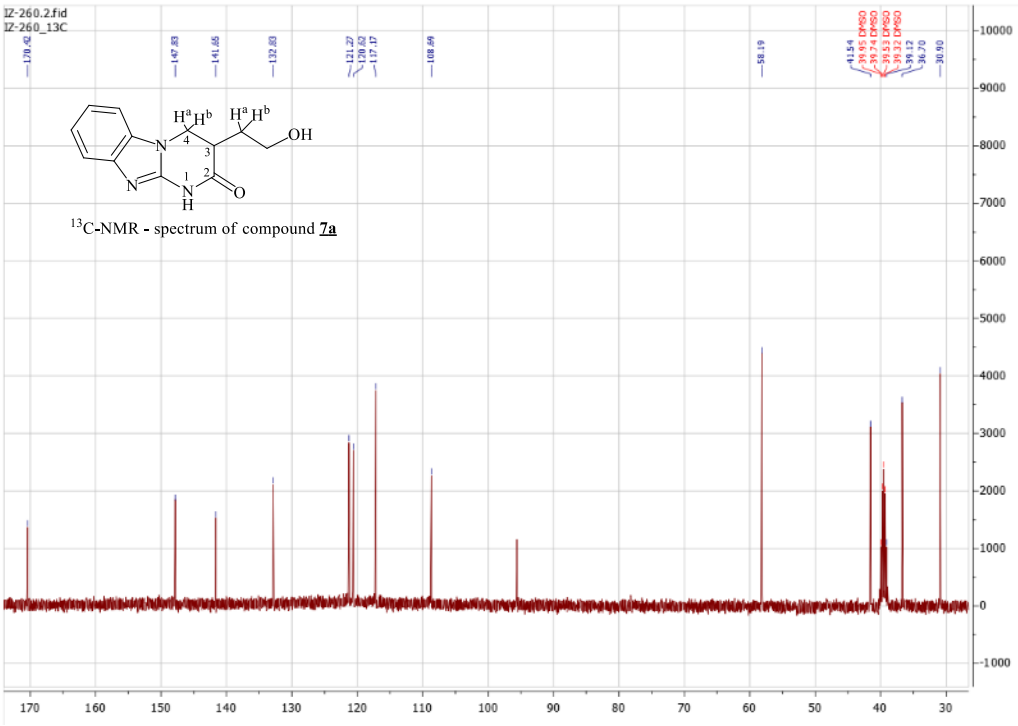


^{13}C-NMR - spectrum of compound **4k**

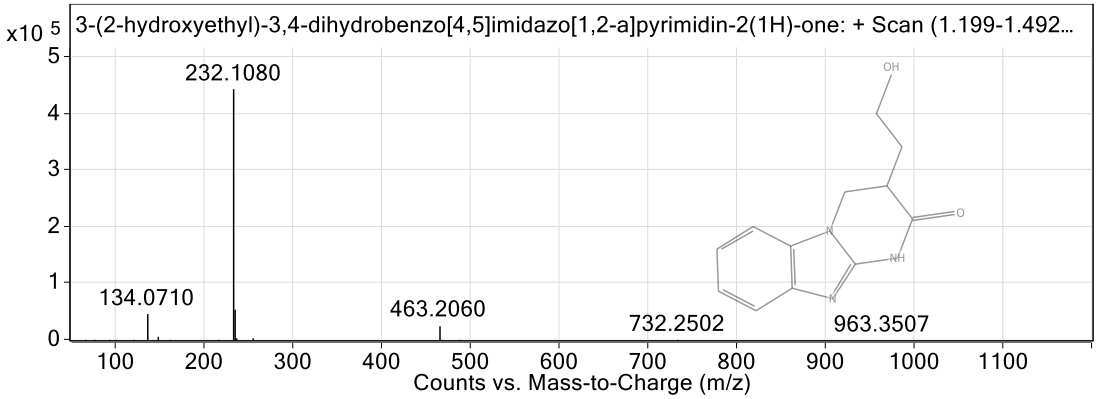


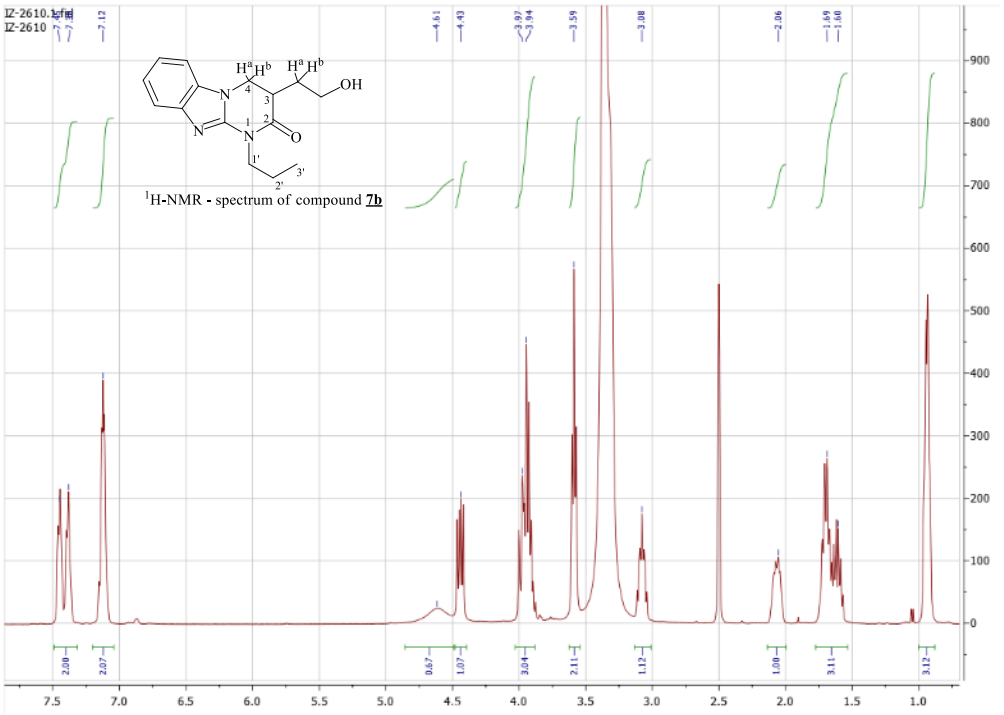


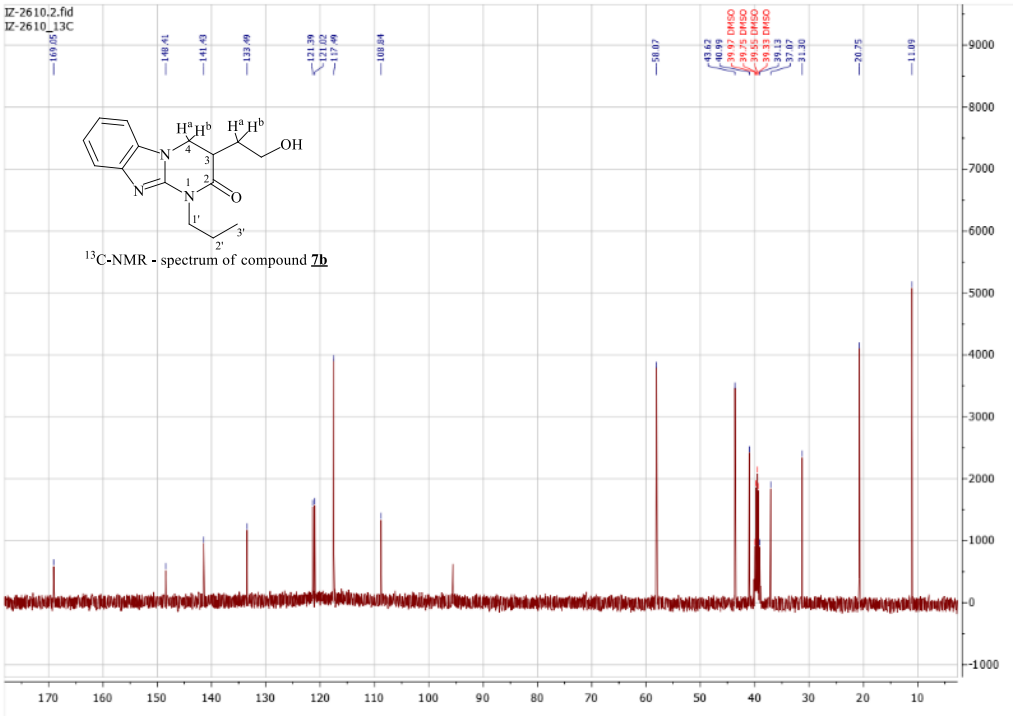
^{1}H-NMR - spectrum of compound **7a**

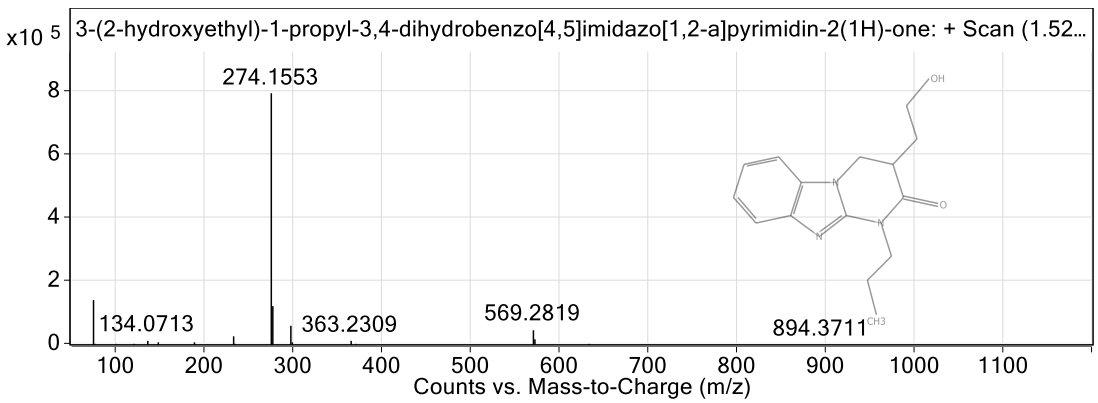


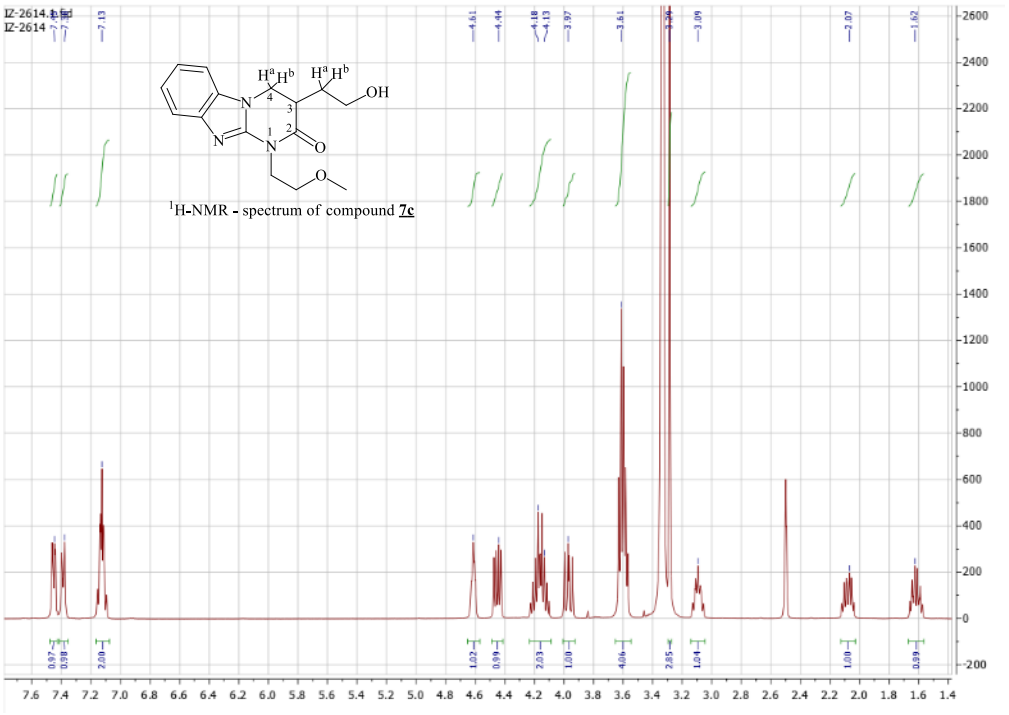
^{13}C-NMR - spectrum of compound **7a**



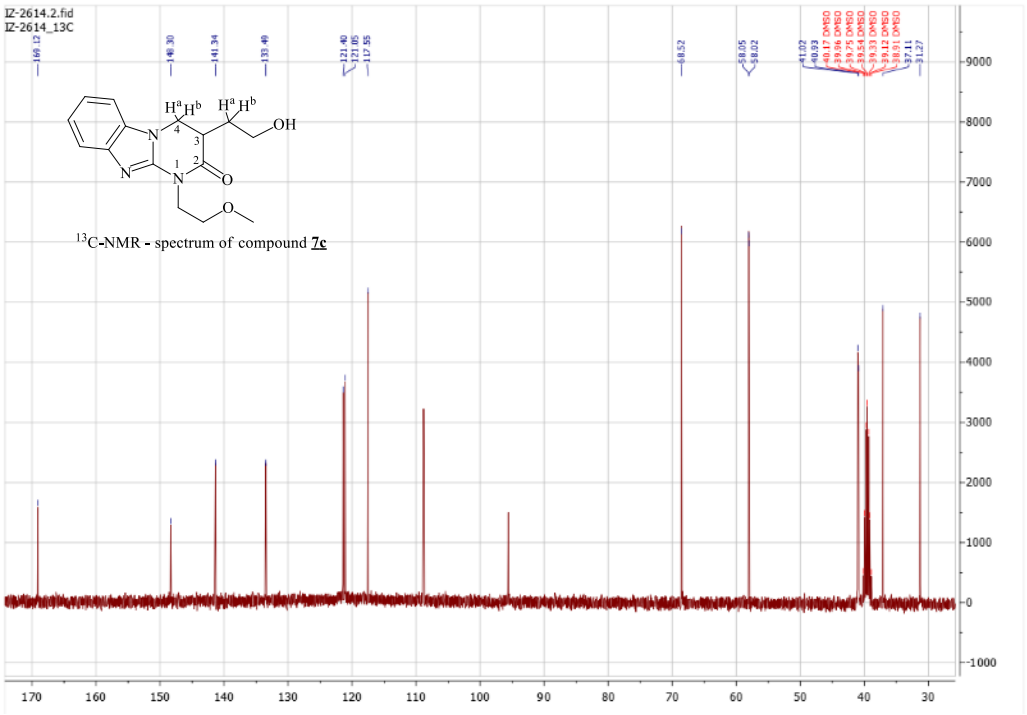

IZ-2610.1.fid
IZ-2610
OH
^{1}H-NMR - spectrum of compound 7b


IZ-2610.2.fid
IZ-2610_13C
OH
^{13}C-NMR - spectrum of compound 7b

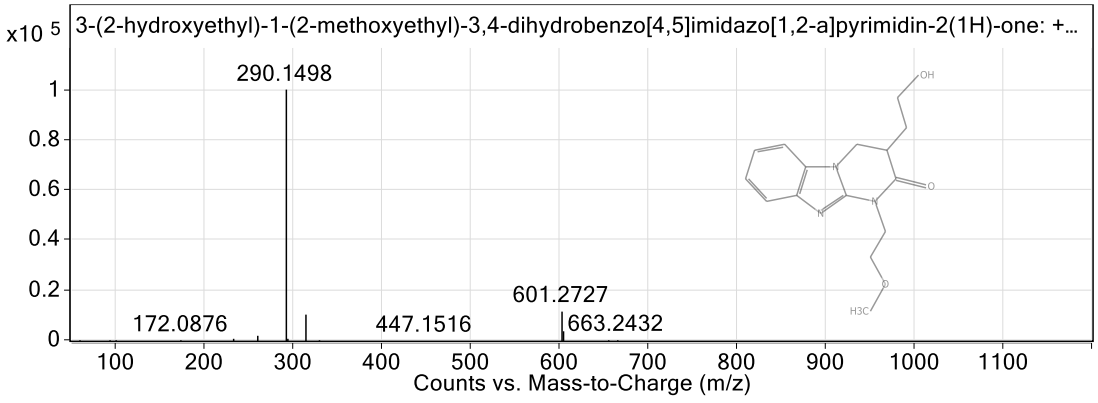

x10 5
3-(2-hydroxyethyl)-1-propyl-3,4-dihydrobenzo[4,5]imidazo[1,2-a]pyrimidin-2(1H)-one: + Scan (1.52…
274.1553
134.0713
363.2309
569.2819
894.3711
Counts vs. Mass-to-Charge (m/z)

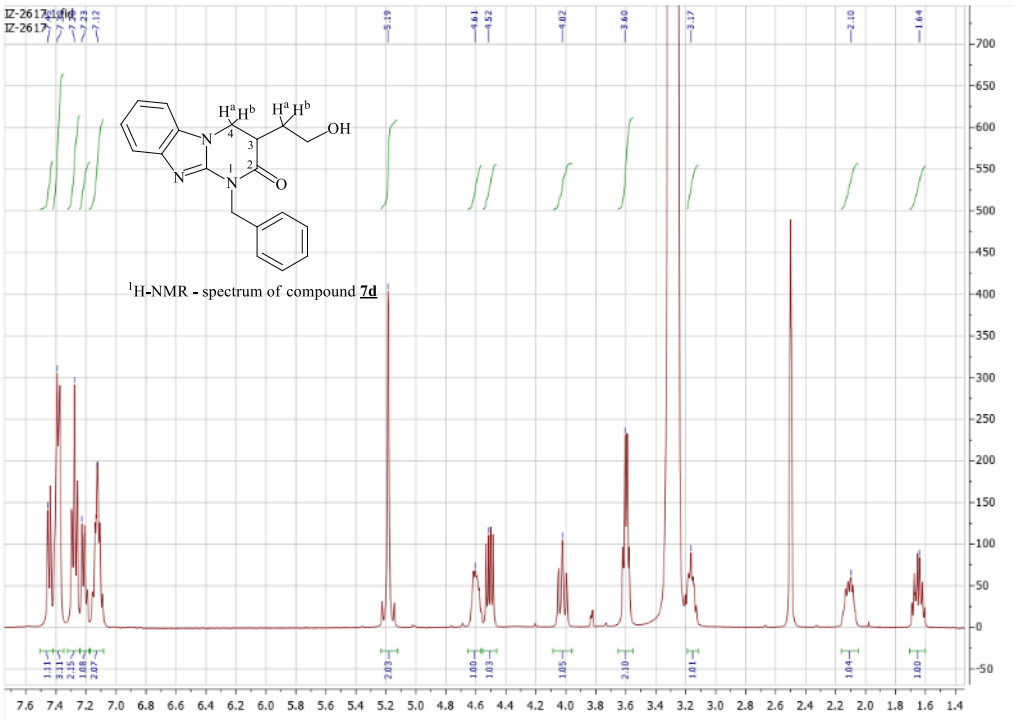


^{1}H-NMR - spectrum of compound **7c**

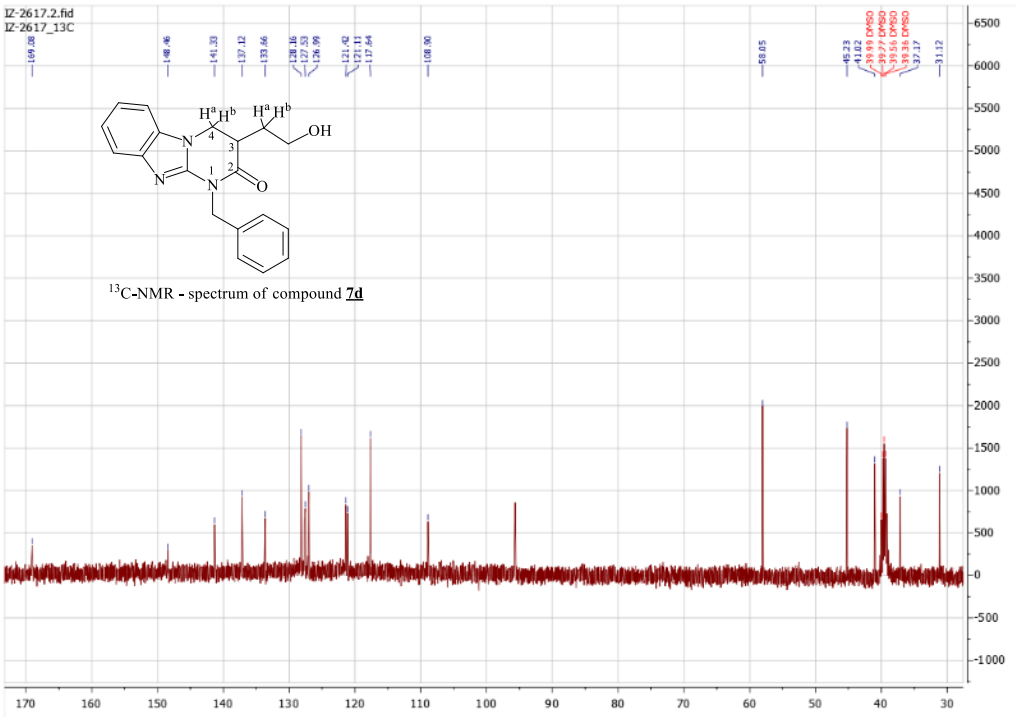


^{13}C-NMR - spectrum of compound **7c**

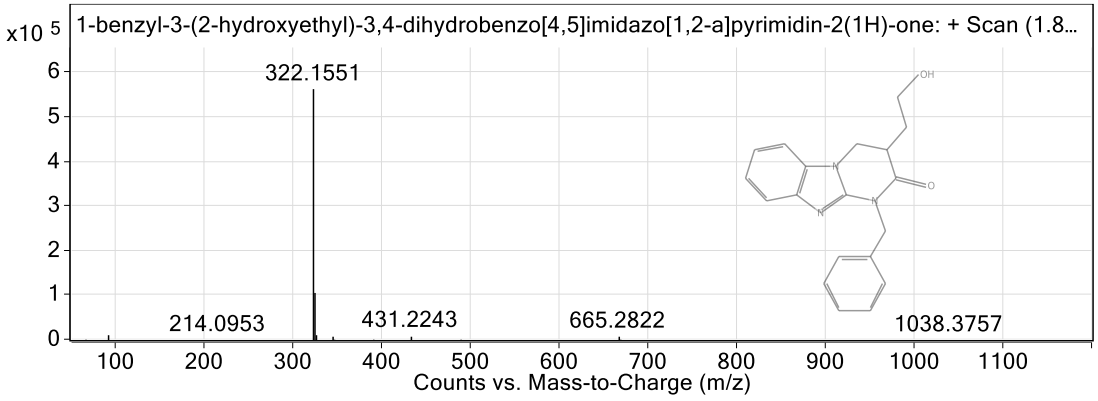


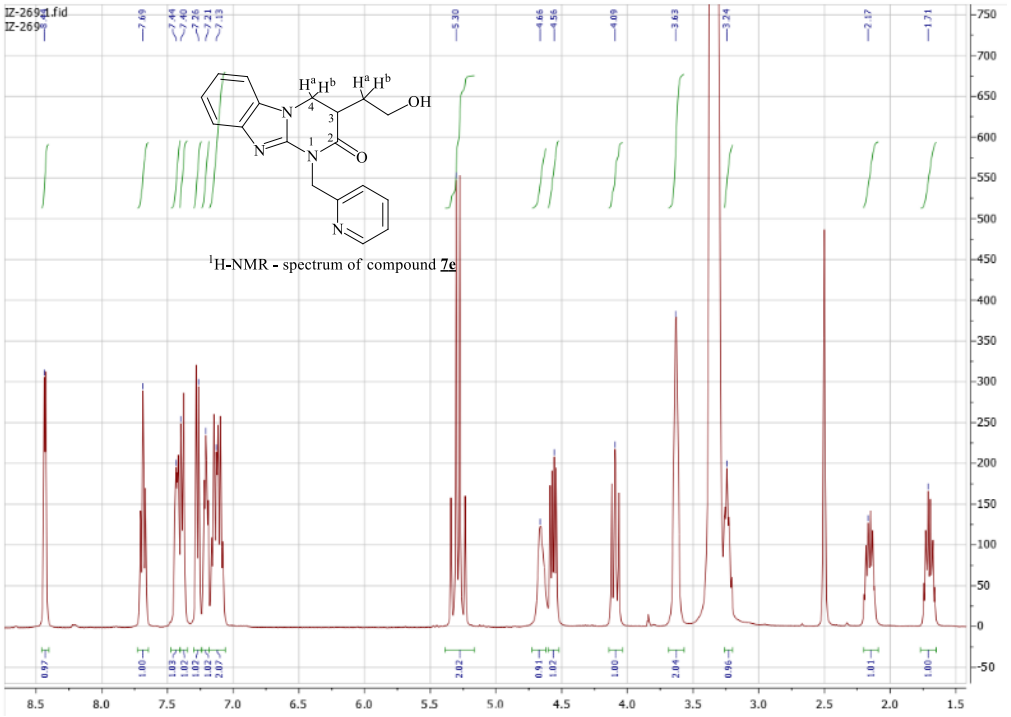


^{1}H-NMR - spectrum of compound **7d**

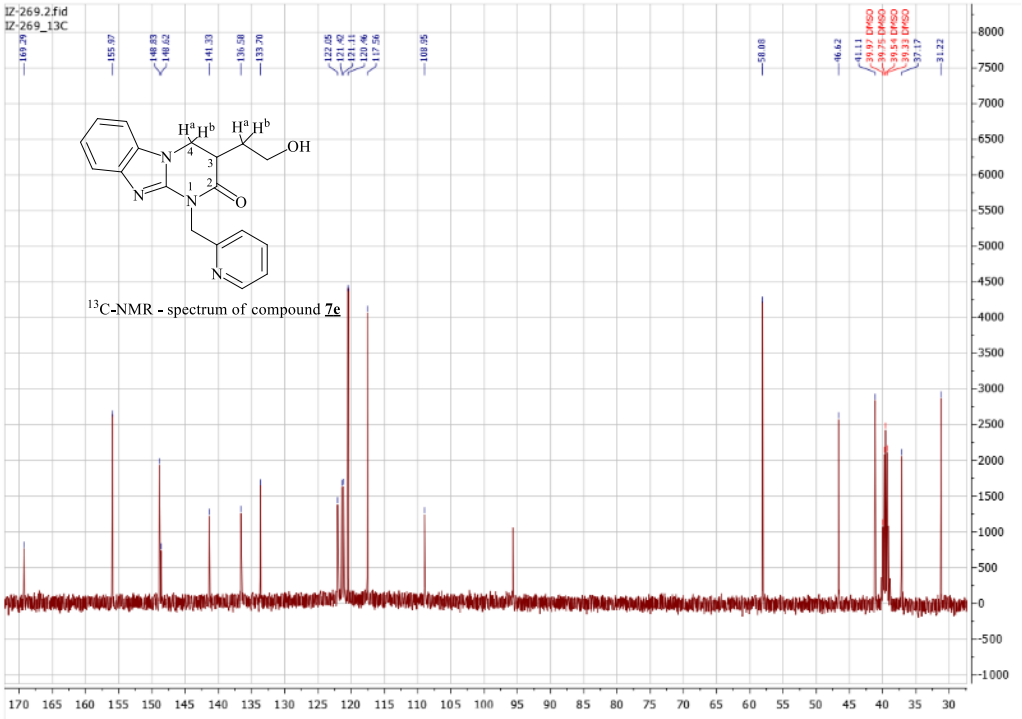


^{13}C-NMR - spectrum of compound **7d**

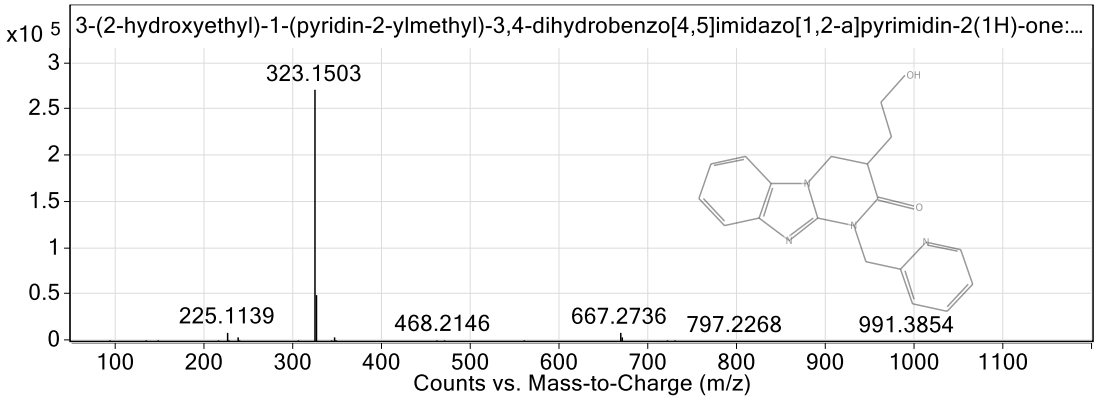


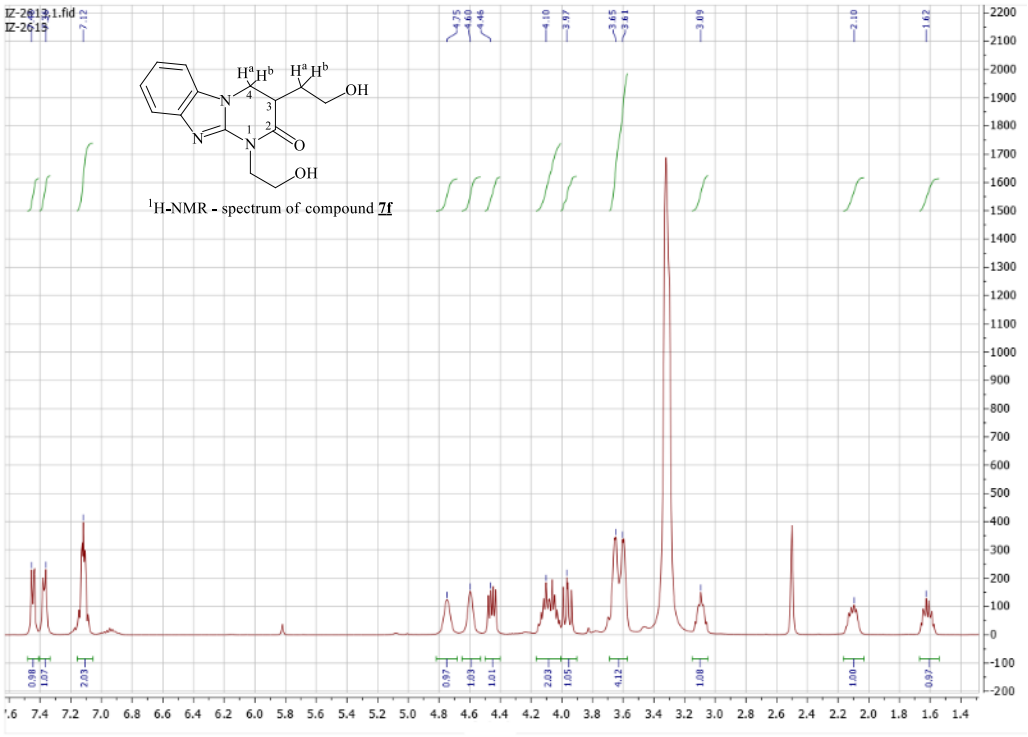


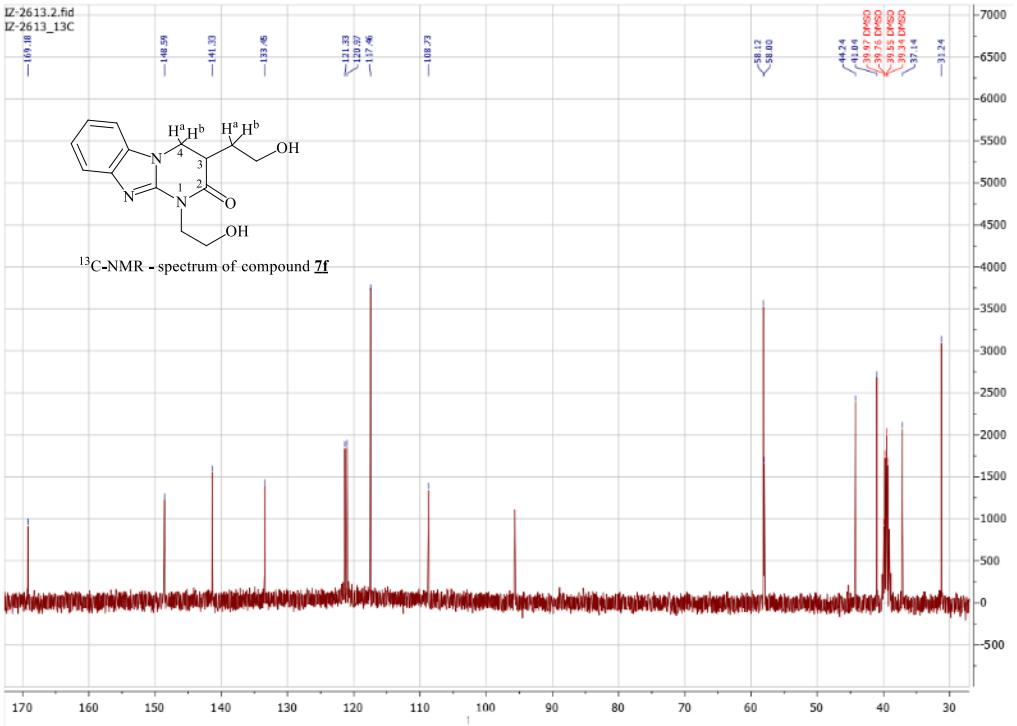
^{1}H-NMR - spectrum of compound **7e**

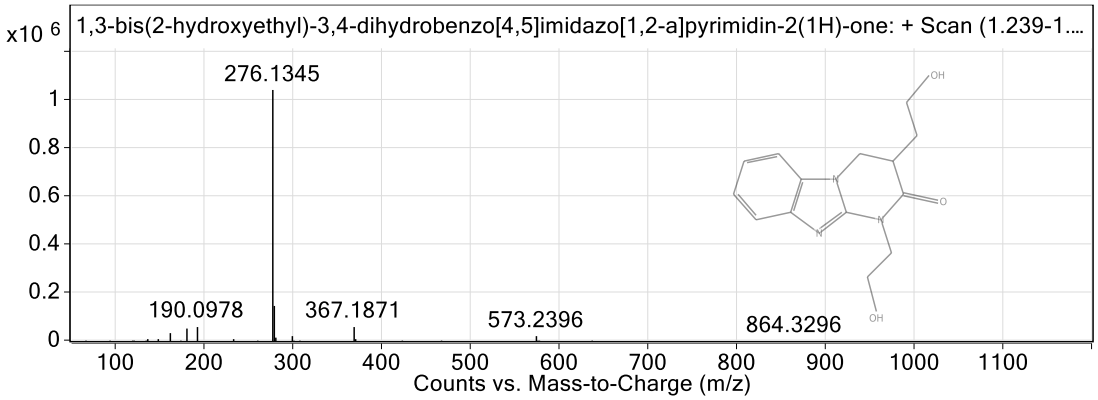


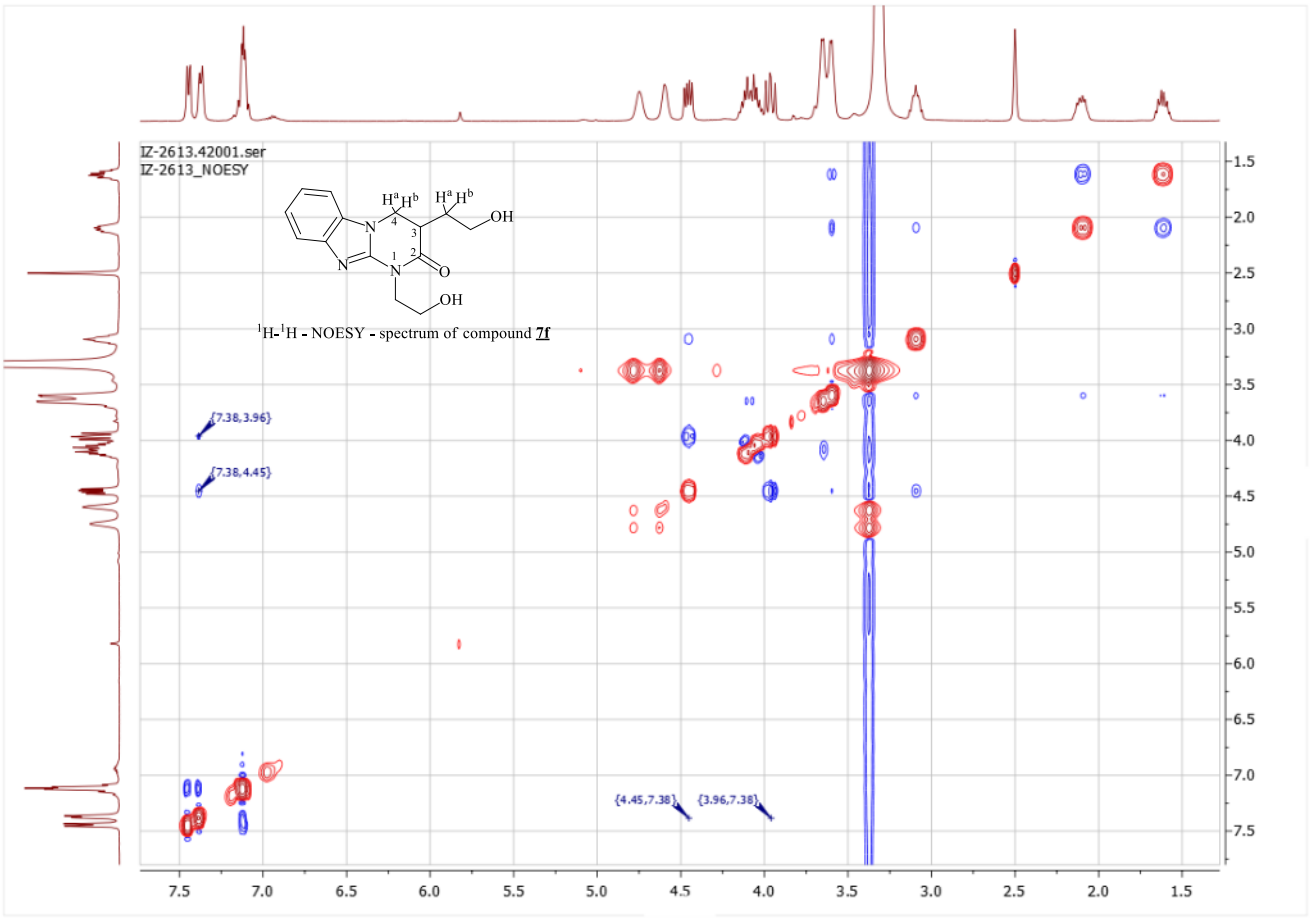
^{13}C-NMR - spectrum of compound **7e**



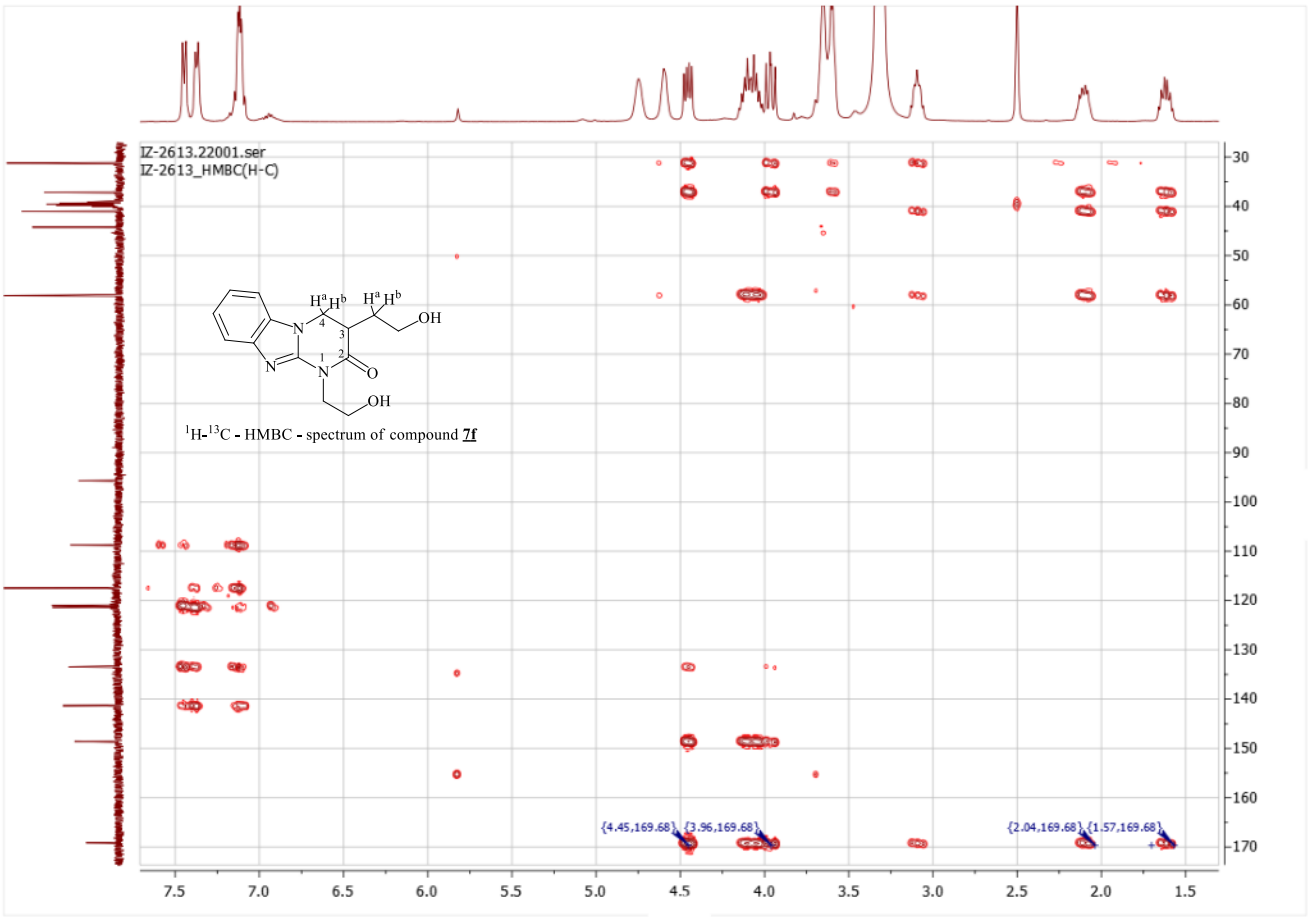

IZ-2613.1.fid
IZ-2613
OH
OH
O
^{1}H-NMR - spectrum of compound 7f


IZ-2613.2.fid
IZ-2613_13C
OH
OH
O
^{13}C-NMR - spectrum of compound 7f

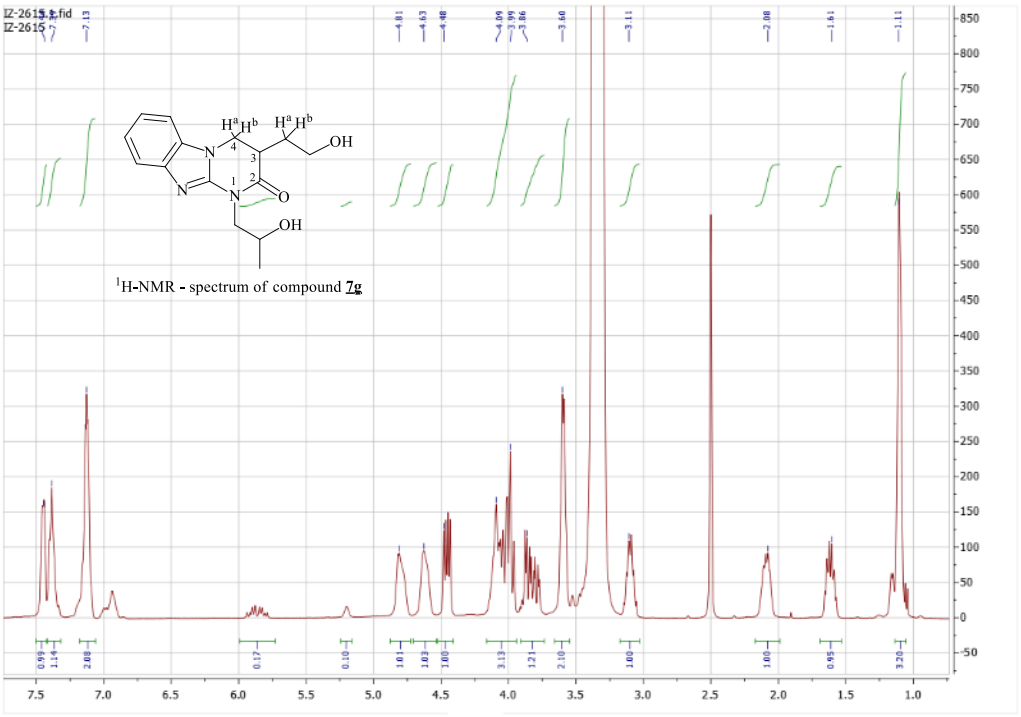

x10 6
1,3-bis(2-hydroxyethyl)-3,4-dihydrobenzo[4,5]imidazo[1,2-a]pyrimidin-2(1H)-one: + Scan (1.239-1....
276.1345
190.0978
367.1871
573.2396
864.3296
Counts vs. Mass-to-Charge (m/z)



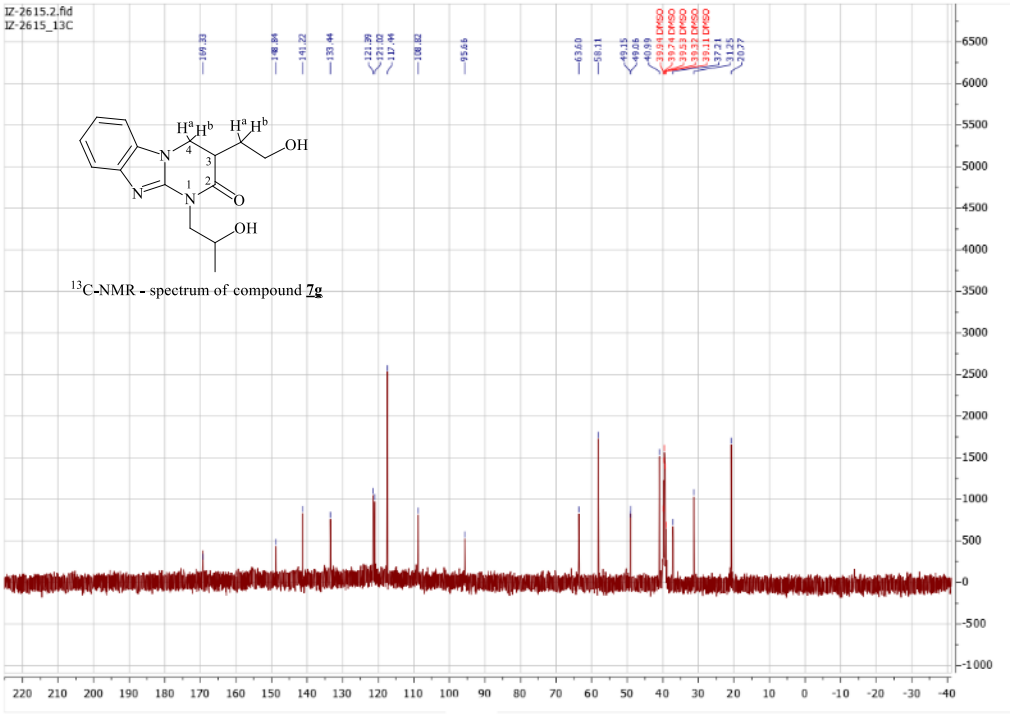
^{1}H-^{1}H - NOESY - spectrum of compound **7f**



^{1}H-^{13}C - HMBC - spectrum of compound **7f**



^{1}H-NMR - spectrum of compound **7g**



^{13}C-NMR - spectrum of compound **7g**