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Supporting Information

Carbazole-based photocatalyst (4CzIPN) as a novel donor-acceptor (D-A) fluorophore-catalyzed visible-light-induced photosynthesis of 3,4-dihydropyrimidin-2-(1*H*)-ones/thiones via a proton-coupled electron transfer (PCET) process

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Contents

1. Turnover number (TON) and turnover frequency (TOF) calculation method

There are two different kinds of yield: TON=Yield/Amount of catalyst (mol), TOF=Yield/Time/Amount of catalyst (mol). Higher TON and TOF values result in greater catalyst efficiency because less catalyst is needed to increase yields. For **4a** is a high TON: 480 and TOF: 96. Due to the study's objectives of maximizing yield, decreasing reaction time, and utilizing the least amount of catalyst possible.

2. Properties of 4CzIPN

The carbazoyl dicyanobenzene (CDCB) family, with carbazolyl (Cz) as an electron donor and dicyanobenzene as an electron acceptor, were first reported as highly efficient light collectors for organic light-emitting diodes by Adachi et al in 2012 [1]. 4CzIPN is the most promising compound for photocatalysis. As shown in Fig. 3 in the manuscript, the carbazolyl groups are significantly deformed from the dicyanobenzene ring with a large dihedral angle of about 60° due to steric hindrance. Density functional theory (DFT) calculations reveal that the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) of 4CzIPN are located on the fragment for Cz, and dicyanobenzene acceptor fragment. Separated HOMO and

LUMO create a small energy gap between the S_1 and T_1 states. Thus, the non-overlapping frontier molecular orbitals (FMOs) make 4CzIPN an efficient heat-activated hysteresis (TADF) material. More importantly, the spatially separated HOMO and LUMO allow for independent fine-tuning of the two orbitals through the modification of electron donors and electron acceptors of the cyanobenzene scaffold, whereby the photophysical properties and their electrochemistry can be tailored for specific catalytic purposes [2, 3].

In addition, the visible UV absorption and luminescence spectra of 4CzIPN, which shows the maximum emission at 507 nm. Importantly, high luminescence quantum efficiencies (up to 94.6%) and long excited state lifetimes (5.1 μs) were observed. The redox potential of 4CzIPN has also been reported [1a, 4]. Photocatalysts such as organic dyes (e.g. eosin Y, rose bengal) and polypyridyl complexes (e.g. *fac*-Ir(ppy)$_3$, [Ir[dF(CF$_3$)ppy]$_2$-(dtbbpy)](PF$_6$) and [Ru(bpy)$_3$](PF$_6$)$_2$) have found tremendous applications for photocatalytic conversions over the past decades. These compounds are excellent photocatalysts for various organic conversions. They are poor single-electron oxidizing/reducing agents in the ground state; however, the excited state under visible light irradiation is a strong single electron transfer reagent. In general, the wide redox potential window, prolonged excited state and high fluorescence quantum yield of photocatalysts can be beneficial for their catalytic activities [2]. The redox potentials and excited state lifetimes of the above-mentioned photocatalysts were compared with those of 4CzIPN. Despite being an inexpensive organic molecule, 4CzIPN shares some photophysical properties with Ru- and Ir-polypyridyl catalysts, such as long excited state time, oxygen window wide reduction and high fluorescence quantum efficiency. For example, 4CzIPN carries both the same oxidation and reduction potential as that of the Ir catalyst [Ir[dF(CF$_3$)ppy]$_2$-(dtbbpy)](PF$_6$), making it a photocatalyst very popular . In addition, the redox properties of 4CzIPN analogs can also be easily tuned by deliberate molecular design and electron donor and acceptor modifications based on property relationships [3b]. Overall, the unique properties mentioned above make 4CzIPN not

only a free alternative for transition metal photocatalysts, but also as photocatalysts irreplaceable strength in modern synthesis [2].

3. Photoredox cycle catalyzed by dye

The photoredox cycle is started when dye in the ground state is irradiated with visible light to produce the high-energy excited state of dye (Dye^*). The process of visible light photoredox catalysis is presented using two separate paths from dye in the excited state (Dye^*). In the presence of a sacrificial electron acceptor, Dye^* reductive's property can be employed. In other words, Dye^* leads the radical cation species of Dye as an electron donor. In the presence of a sacrificial electron donor, Dye^* also works as an electron acceptor (Fig. S1) [5].

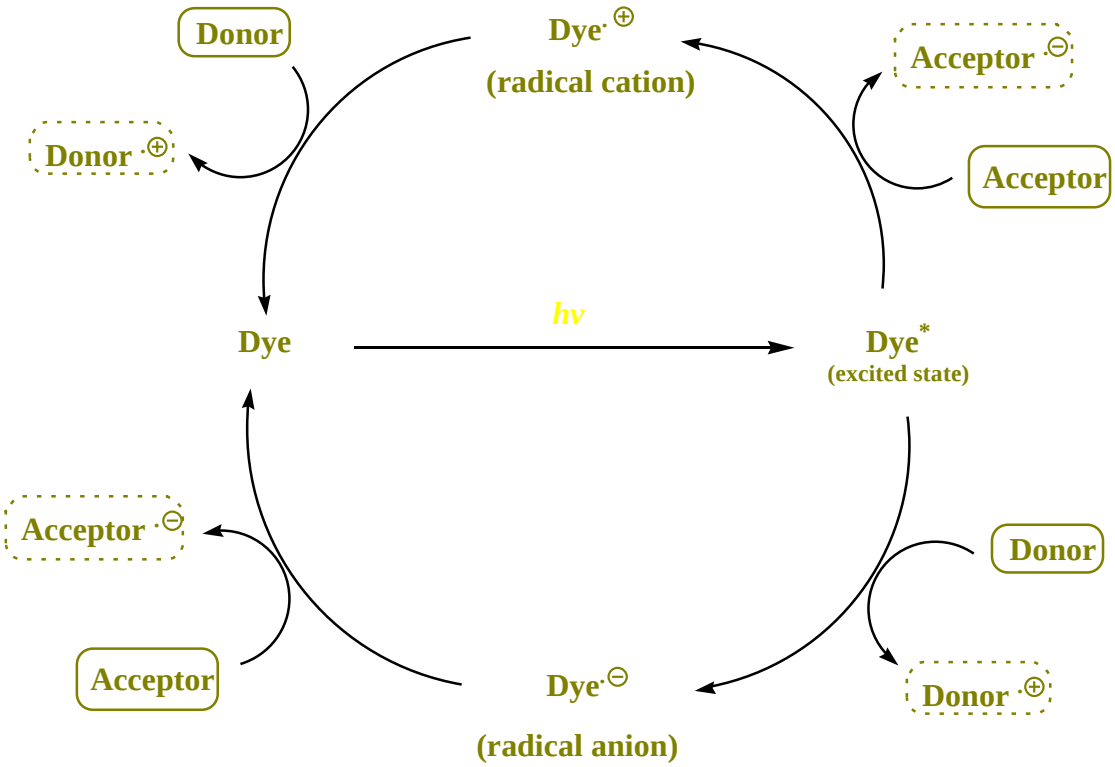


Fig. S1 Photoredox cycle catalyzed by dye [5].

4. Oxidation and reduction quenching cycle of redox catalyst

The two mechanical methods shown in Fig. S2 [6, 7] are used in most of the redox catalytic processes. In the oxidation quenching cycle, the excited state catalyst is quenched by donating an electron to the substrate or to the oxidant present in the reaction mixture; in the reduction quenching cycle, the catalyst* is extinguished by accepting an electron from the substrate or from the reducing agent. In the oxidation cycle, the reduction of [catalyst]$^{\bullet+}$ is oxidized, while in the reduction cycle, the oxidation of [catalyst]$^{\bullet\bullet}$ is reduced. Catalyst cycling can be induced by the substrate, an external redox-active reactant, or an intermediate in both cases. There are three main redox results for the substrate in both manifolds: pure oxidizing agent, pure reducing agent and pure redox neutral. An external oxidant is required for the clean oxidation reaction, which can take up electrons in the cycling steps. Likewise, in the cyclic phases of net reduction processes, an external reducing agent donates electrons. Electron transfer back to an oxidation or reduction catalyst, sometimes mediated by a redox co-catalyst, common in strong redox neutral reactions [6].

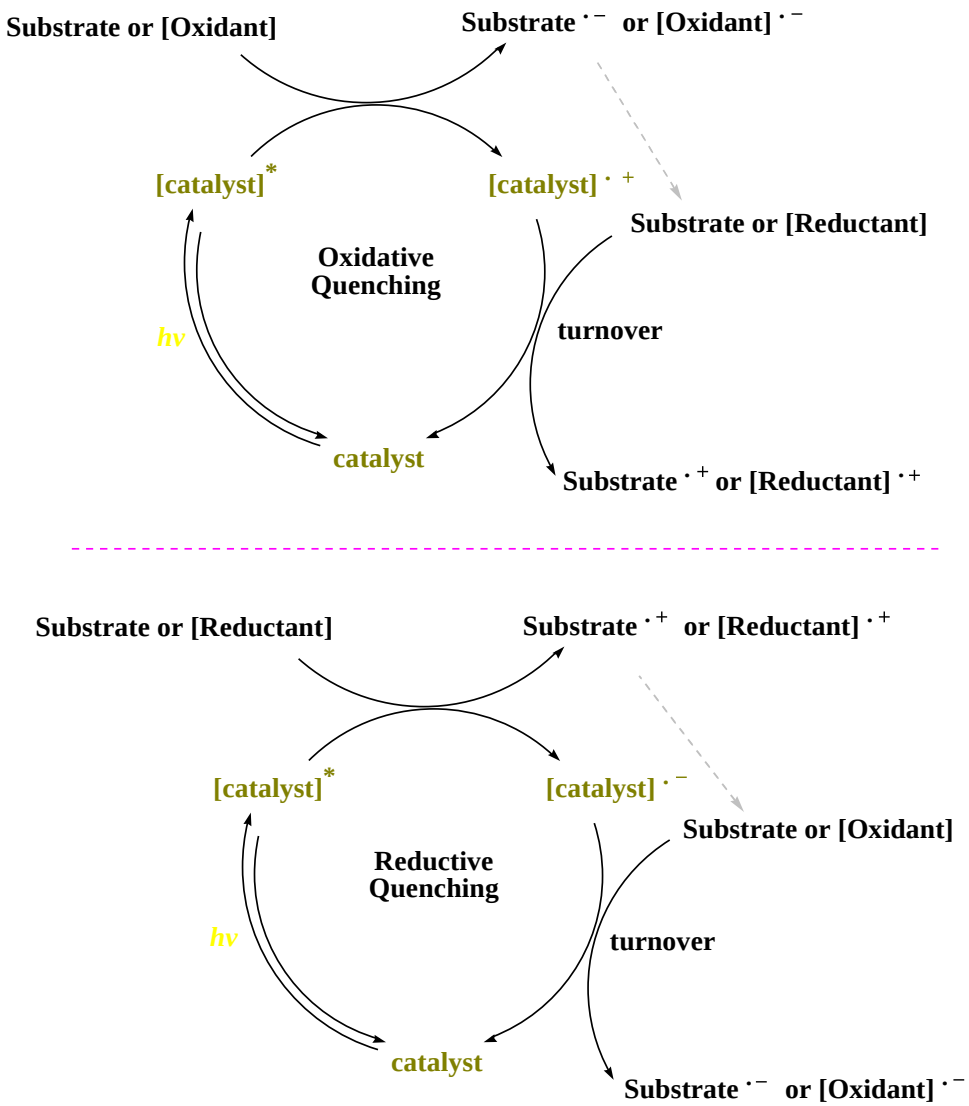
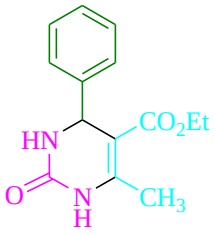


Fig. S2 Oxidation and reduction quenching cycle of redox catalyst [6].

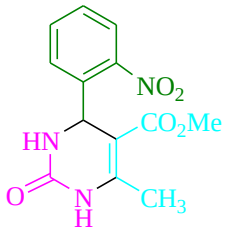
5. ^{1}HNMR, ^{13}CNMR, mass, CHN-O analyze data recorded for compounds

5-Ethoxycarbonyl-6-methyl-4-phenyl-3,4-dihydropyrimidin-2(1H)-one (4a)



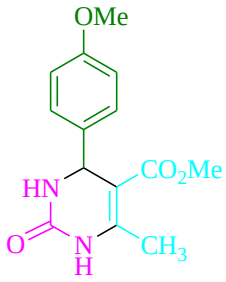
Yield: 96%; M.p. 202-204 °C; ^{1}HNMR (400 MHz, DMSO-d_6): 1.10 (3H, t, *J*= 7.2 Hz, $\underline{CH_3}CH_2$), 2.26 (3H, s, CH_3), 3.99 (2H, q, *J*=7.2 Hz, CH_2O), 5.15 (1H, s, $H_{benzylic}$), 7.26 (3H, d, *J*= 7.2 Hz, H_{Ar}), 7.33 (2H, t, *J*=7.2 Hz, H_{Ar}), 7.76 and 9.21 (2H, 2s, 2NH).

5-Methoxycarbonyl-6-methyl-4-(2-nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one (4b)



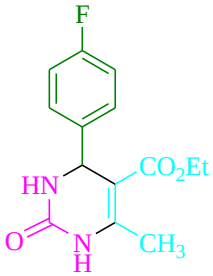
Yield: 94%; M.p. 273-275 °C; ^{1}HNMR (300 MHz, DMSO-d_6): 2.29 (3H, s, CH_3), 3.56 (3H, s, OCH_3), 5.29 (1H, s, $H_{benzylic}$), 7.51-7.53 (2H, m, H_{Ar}), 8.21-8.24 (2H, m, H_{Ar}), 7.93 and 9.40 (2H, 2s, 2NH); ^{13}CNMR (100 MHz, DMSO-d_6): 17.7 ($\underline{CH_3}$-CH=CH), 50.7 (Ar-CHN), 53.6 (CO_2-$\underline{CH_3}$), 98.9, 123.2 (CH_3-$\underline{CH}$=$\underline{CH}$), 126.7, 127.9, 128.3, 137.4, 144.5 and 148.4 (C_{Ar}), 151.9 (C=ONH), 165.8 (C=O ester).

5-Methoxycarbonyl-6-methyl-4-(4-methoxyphenyl)-3,4-dihydropyrimidin-2(1H)–one (4c)



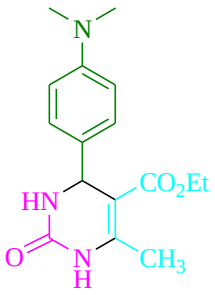
Yield: 95%; M.p. 187-189 °C; ^{1}HNMR (300 MHz, DMSO-d_6): 2.25 (3H, s, CH_3), 3.53 (3H, s, OCH_3), 3.73 (3H, s, OCH_3), 5.09 (1H, s, $H_{benzylic}$), 6.88 (2H, d, *J*= 8.8 Hz, H_{Ar}), 7.14 (2H, d, *J*= 8.8 Hz, H_{Ar}), 7.71 and 9.20 (2H, 2s, 2NH).

5-Ethoxycarbonyl-6-methyl-4-(4-fluorophenyl)-3,4-dihydropyrimidin-2(1H)–one (4d)



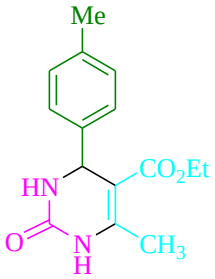
Yield: 95%; M.p. 171-173 °C; ^{1}HNMR (300 MHz, DMSO-d_6): 1.11 (3H , t, *J*= 9.6 Hz, $\underline{CH_3}CH_2$), 2.25(3H, s, CH_3), 3.99 (2H, q, *J*=9.6 Hz, CH_2O), 5.14 (1H, s, $H_{benzylic}$), 7.13-7.20 (2H, m, H_{Ar}), 7.24-7.29 (2H, m, H_{Ar}), 7.78 and 9.25 (2H, 2s, 2NH).

5-Ethoxycarbonyl-6-methyl-4-(N,N-Dimethylphenyl)-3,4-dihydropyrimidin-2(1H)-one (4e)



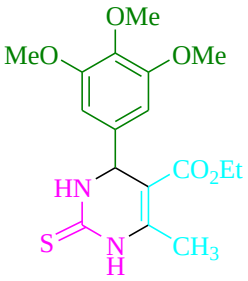
Yield: 94%; M.p. 256-258 °C; ^{1}HNMR (300 MHz, DMSO-d_6): 1.12 (3H, t, *J*= 9.2 Hz, $\underline{CH_3}CH_2$), 2.26 (3H, s, CH_3), 2.85 (6H, s, $2CH_3$), 3.99 (2H, q, *J*=9.2 Hz, CH_2O), 5.04 (1H, s, $H_{benzylic}$), 6.66 (2H, d, *J*=11.6 Hz, H_{Ar}), 7.42 (2H, d, *J*=11.6 Hz, H_{Ar}), 7.61 and 9.11 (2H, 2s, 2NH).

5-Ethoxycarbonyl-6-methyl-4-(4-methylphenyl)-3,4-dihydropyrimidin-2(1H)-one (4f)



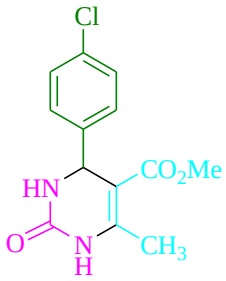
Yield: 96%; M.p. 216-218 °C; ^{1}HNMR (400 MHz, DMSO-d_6): 1.11 (3H, t, *J*= 7.2 Hz, $\underline{CH_3}CH_2$), 2.25 (3H, s, CH_3), 2.27 (3H, s, CH_3), 3.99 (2H, q, *J*=7.2 Hz, CH_2O), 5.11 (1H, s, $H_{benzylic}$), 7.13 (4H, s, H_{Ar}), 7.70 and 9.17 (2H, 2s, 2NH); ^{13}CNMR (100 MHz, DMSO-d_6): 14.0 ($\underline{CH_3}$-CH_2O), 17.6 ($\underline{CH_3}$-CH=CH), 21.1 (CH_3), 53.7 (Ar-CHN), 59.1 (CH_3-$\underline{CH_2}O$), 99.2, 123.3 (CH_3-$\underline{CH}$=$\underline{CH}$), 126.8, 127.8, 128.2, 137.3, 144.7 and 148.0 (C_{Ar}), 151.9 (C=ONH), 165.3 (C=O ester).

5-Ethoxycarbonyl-6-methyl-4-(3,4,5–trimethoxyphenyl)-3,4-dihydropyrimidin-2(1H)-thione (4g)



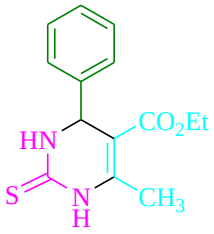
Yield: 88%; M.p. 193-195 °C; ^{1}HNMR (400 MHz, DMSO-d_6): 1.16 (3H , t, *J*= 7.2 Hz, $\underline{CH_3}CH_2$), 2.30 (3H, s, CH_3), 3.65 (3H, s, OCH_3), 3.74 (6H, s, $2OCH_3$), 4.07 (2H, q, *J*=7.2 Hz, CH_2O), 5.16 (1H, s, $H_{benzylic}$), 6.52 (2H, s, H_{Ar}), 9.65 and 10.37 (2H, 2s, 2NH); ^{13}CNMR (100 MHz, DMSO-d_6): 14.6 ($\underline{CH_3}$-CH_2O), 17.6 ($\underline{CH_3}$-CH=CH), 54.1 (Ar-CHN), 56.2, 60.1 and 60.4 ($3OCH_3$), 60.4 (CH_3-$\underline{CH_2}O$), 101 and 103.8 (CH_3-$\underline{CH}$=$\underline{CH}$), 137.4, 139.5, 139.5, 145.4, 145.6 and 153.3 (Ar), 165.6 (C=O ester), 174.9 (C=SNH); MS (EI) m/z (%): 366 (M , 118), 306 (10), 293 (77), 277 (17), 261 (12), 246 (17), 232 (12), 219 (9), 199 (100), 186 (5), 171 (47), 153 (49), 138 (4), 126 (19), 112 (18), 94 (12), 81 (11), 66 (18), 42 (32).

5-Methoxycarbonyl-6-methyl-4-(4-chlorophenyl)-3,4-dihydropyrimidin-2(1H)-one (4j)



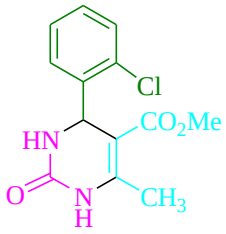
Yield: 85%; M.p. 203-205 °C; ^{1}HNMR (300 MHz, DMSO-d_6): 2.26 (3H, s, CH_3), 3.55 (3H, s, OCH_3), 5.15 (1H, s, $H_{benzylic}$), 7.25 (1H, d, *J*=11.2 Hz, H_{Ar}), 7.41 (1H, d, *J*=11.2 Hz, H_{Ar}), 7.80 and 9.28 (2H, 2s, 2NH).

5-Ethoxycarbonyl-6-methyl-4-phenyl-3,4-dihydropyrimidin-2(1H)-thione (4k)



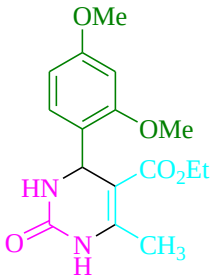
Yield: 94%; M.p. 206-208 °C; ^{1}HNMR (400 MHz, DMSO-d_6): 1.11 (3H , t, *J*= 7.2 Hz, $\underline{CH_3}CH_2$), 2.31 (3H, s, CH_3), 4.02 (2H, q, *J*=7.2 Hz, CH_2O), 5.19 (1H, s, $H_{benzylic}$), 7.23 (2H, d, *J*=7.2 Hz, H_{Ar}), 7.28 (1H, t, *J*=7.2 Hz, H_{Ar}), 7.36 (2H, t, *J*=7.2 Hz, H_{Ar}), 9.68 and 10.36 (2H, 2s, 2NH).

5-Methoxycarbonyl-6-methyl-4-(2-chlorophenyl)-3,4-dihydropyrimidin-2(1H)-one (4l)



Yield: 93%; M.p. 248-250 °C; ^{1}HNMR (400 MHz, DMSO-d_6): 2.31 (3H, s, CH_3), 3.46 (3H, s, OCH_3), 5.62 (1H, s, $H_{benzylic}$), 7.28-7.34 (3H, m, H_{Ar}), 7.42 (1H, d, *J*=7.2 Hz, H_{Ar}), 7.72 and 9.36 (2H, 2s, 2NH).

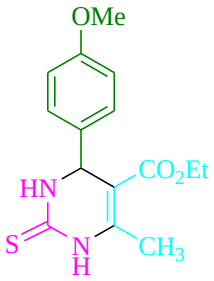
5-Ethoxycarbonyl-6-methyl-4-(2,4–dimethoxyphenyl)-3,4-dihydropyrimidin-2(1H)-one (4m)



Yield: 89%; M.p. 211-213 °C; ^{1}HNMR (400 MHz, DMSO-d_6): 1.05 (3H , t, *J*= 7.2 Hz, $\underline{CH_3}CH_2$), 2.27 (3H, s, CH_3), 3.76 (3H, s, OCH_3), 3.82 (3H, s, OCH_3), 3.93 (2H, q, *J*= 7.2 Hz, CH_2O), 5.41

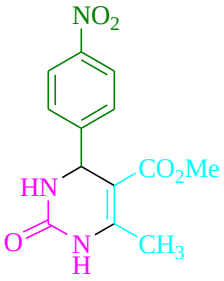
(1H, s, $H_{benzylic}$), 6.45 (1H, dd, *J*=8.4 Hz, J= 2.4 Hz, H_{Ar}), 6.54 (1H, d, *J*=2.4 Hz, H_{Ar}), 6.94 (1H, d, *J*=8.4 Hz, H_{Ar}), 7.21 and 9.08 (2H, 2s, 2NH) ppm; ^{13}CNMR (100 MHz, DMSO-d_6): 14.5 ($\underline{CH_3}$-CH_2O), 18.2 (CH_3-CH=CH), 48.9 (Ar-CHN), 55.6 and 55.7 ($2OCH_3$), 59.44 (CH_3-$\underline{CH_2}O$), 98.9, 104.8 (CH_3-CH=CH), 124.6, 124.7,128.2, 148.9, 152.6 and 158.0 (C_{Ar}), 160.3 (C=ONH), 165.9 (C=O ester); MS (EI) m/z (%): 320 (M, 27), 259 (9), 247 (100), 228 (43), 215 (28), 201 (6), 183 (47), 174 (9), 155 (43), 137 (39), 121 (11), 110 (31), 92 (16), 77 (32), 67 (10), 55 (8), 42 (30); Anal. Calcd for $C_{16}H_{20}N_2O_5$: C, 59.99; H, 6.29; N, 8.74%. Found: C, 57.36; H, 5.49; N, 8.19%.

5-Ethoxycarbonyl-6-methyl-4-(4–methoxyphenyl)-3,4-dihydropyrimidin-2(1H)-thione (4n)



Yield: 93%; M.p. 152-154 °C; ^{1}HNMR (300 MHz, DMSO-d_6): 1.13 (3H , t, *J*= 9.6 Hz, $\underline{CH_3}CH_2$), 2.29 (3H, s, CH_3), 3.74 (3H, s, OCH_3), 4.03 (2H, q, *J*=9.6 Hz, CH_2O), 5.15 (1H, s, $H_{benzylic}$), 6.77 (2H, m, H_{Ar}), 6.87 (1H, m, H_{Ar}), 7.28 (1H, t, *J*=9.6 Hz, H_{Ar}), 9.66 and 10.37 (2H, 2s, 2NH).

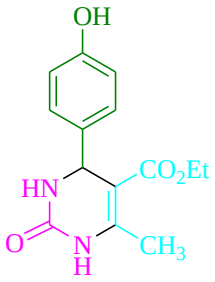
5-Methoxycarbonyl-6-methyl-4-(4-nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one (4o)



Yield: 94%; M.p. 214-216 °C; ^{1}HNMR (400 MHz, DMSO-d_6): 2.28(3H, s, CH_3), 3.55 (3H, s, OCH_3), 5.28 (1H, s, $H_{benzylic}$), 7.52 (2H, d, *J*= 8.8Hz, H_{Ar}), 8.23 (2H, d, *J*= 8.8Hz, H_{Ar}), 7.93 and

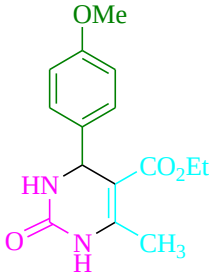
9.40 (2H, 2s, 2NH).

5-Ethoxycarbonyl-6-methyl-4-(4-hydroxyphenyl)-3,4-dihydropyrimidin-2(1H)–one (4p)



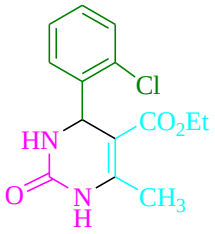
Yield: 81%; M.p. 228-230 °C; ^{1}HNMR (400 MHz, DMSO-d_6): 1.11 (3H , t, *J*= 9.6 Hz, $\underline{CH_3}CH_2$), 2.50 (3H, s, CH_3), 3.98 (2H, q, *J*=9.6 Hz, CH_2O), 5.04 (1H, s, $H_{benzylic}$), 6.68-7.04(4H, m, H_{Ar}), 7.64 and 9.13 (2H, 2s, 2NH), 9.35 (1H, s, OH).

5-Ethoxycarbonyl-6-methyl-4-(4-methoxyphenyl)-3,4-dihydropyrimidin-2(1H)–one (4q)



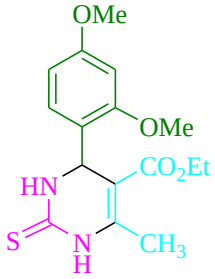
Yield: 90%; M.p.203-205°C; ^{1}HNMR (400 MHz, DMSO-d_6): 1.11 (3H , t, *J*= 9.6 Hz, $\underline{CH_3}CH_2$), 2.24(3H, s, CH_3), 3.73 (3H, s, OCH_3), 3.99 (2H, q, *J*=9.6 Hz, CH_2O), 5.09 (1H, s, $H_{benzylic}$), 6.89 (2H, d, *J*= 8.4 Hz, H_{Ar}), 7.15 (2H, d, *J*= 8.8 Hz, H_{Ar}), 7.70 and 9.18 (2H, 2s, 2NH).

5-Ethoxycarbonyl-6-methyl-4-(2-chlorophenyl)-3,4-dihydropyrimidin-2(1H)–one (4r)



Yield: 89%; M.p. 219-221 °C; ^{1}HNMR (400 MHz, DMSO-d_6): 1.00 (3H , t, *J*= 9.2 Hz, $\underline{CH_3}CH_2$), 2.31 (3H, s, CH_3), 4.02 (2H, q, *J*=9.2 Hz, CH_2O), 5.63 (1H, s, $H_{benzylic}$), 7.25-7.34 (3H, m, H_{Ar}), 7.41 (1H, d, *J*=8.8 Hz, H_{Ar}), 7.73 and 9.29 (2H, 2s, 2NH).

5-Ethoxycarbonyl-6-methyl-4-(2,4–dimethoxyphenyl)-3,4-dihydropyrimidin-2(1H)-thione (4s)



Yield: 87%; M.p. 160-162 °C; ^{1}HNMR (400 MHz, DMSO-d_6): 1.07 (3H , t, *J*= 7.2 Hz, $\underline{CH_3}CH_2$), 2.29 (3H, s, CH_3), 3.75 (3H, s, OCH_3), 3.79 (3H, s, OCH_3), 3.97 (2H, q, *J*=7.2 Hz, CH_2O), 5.42 (1H, s, $H_{benzylic}$), 6.47 (1H, dd, *J*=8.4 Hz, *J*=2.4 Hz, H_{Ar}), 6.55 (1H, d, *J*=2.4 Hz, H_{Ar}), 6.94 (1H, d, *J*=8.4 Hz, H_{Ar}), 9.20 and 10.20 (2H, 2s, 2NH) ppm; ^{13}CNMR (100 MHz, DMSO-d_6): 14.5 ($\underline{CH_3}$-CH_2O), 17.4 (CH_3-CH=CH), 49.4 (Ar-CHN), 55.7 and 56.0 ($2OCH_3$), 59.8 (CH_3-$\underline{CH_2}O$), 98.9 and 100.1 (CH_3-CH=CH), 105.0, 119.6, 128.7, 129.4, 145.4 and 160.7 (Ar), 165.7 (C=O ester), 174.3 (C=SNH); MS (EI) m/z (%): 336 (M , 58), 275 (6), 263 (100), 244 (16), 231 (17), 218 (18), 199 (29), 187 (12), 171 (18), 161 (13), 149 (10), 138 (19), 126 (22), 115 (10), 103 (11), 91 (12), 77 (19), 67 (12), 55 (8), 42 (26); Anal. Calcd for $C_{16}H_{20}N_2O_4S$: C, 57.12; H, 5.99; N, 8.33%. Found: C, 56.62; H, 4.23; N, 8.55%.

6. [1]HNMR, [13]CNMR, and mass files recorded for compounds

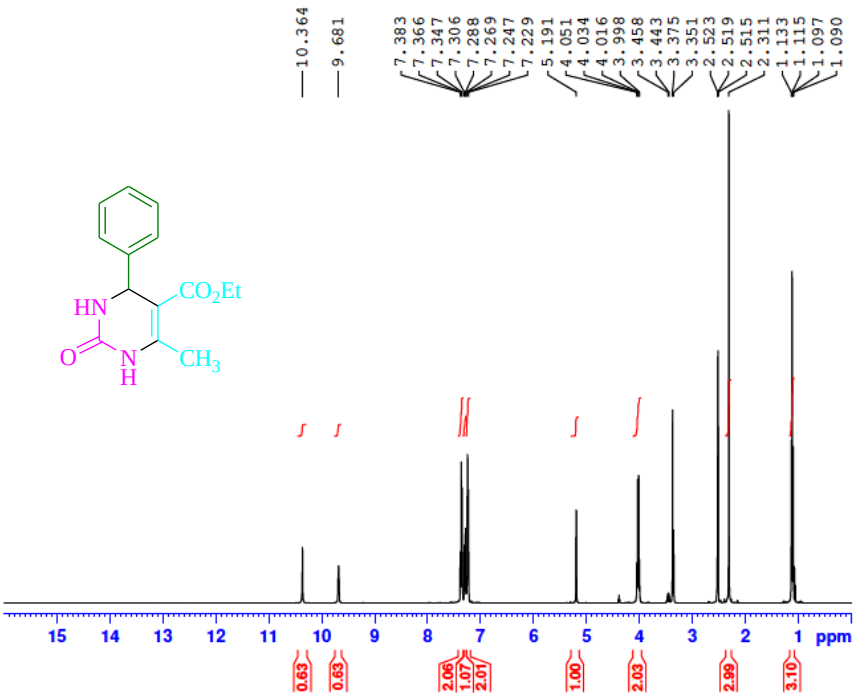


Fig. S3 [1]HNMR spectrum of compound (400 MHz, DMSO-d_6) of **4a**

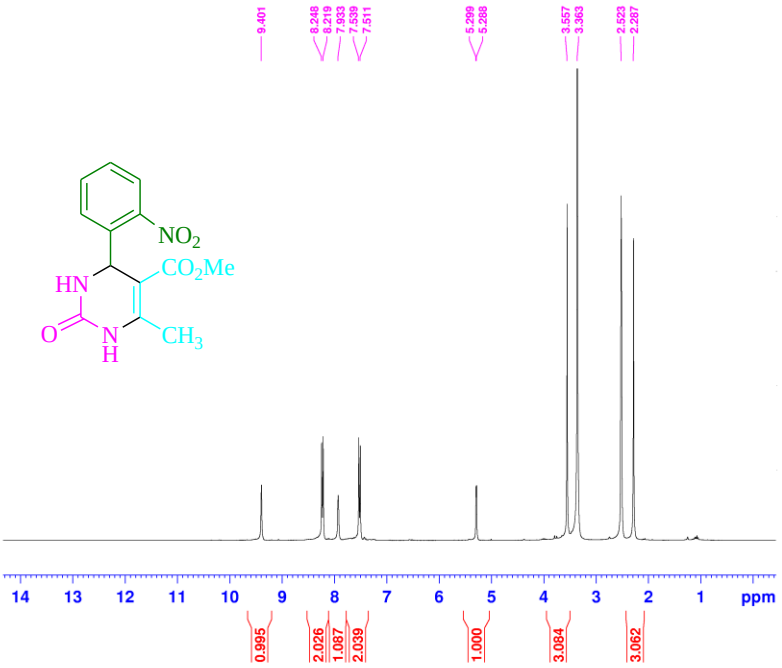


Fig. S4 ^{1}HNMR spectrum of compound (300 MHz, DMSO-d_6) of **4b**

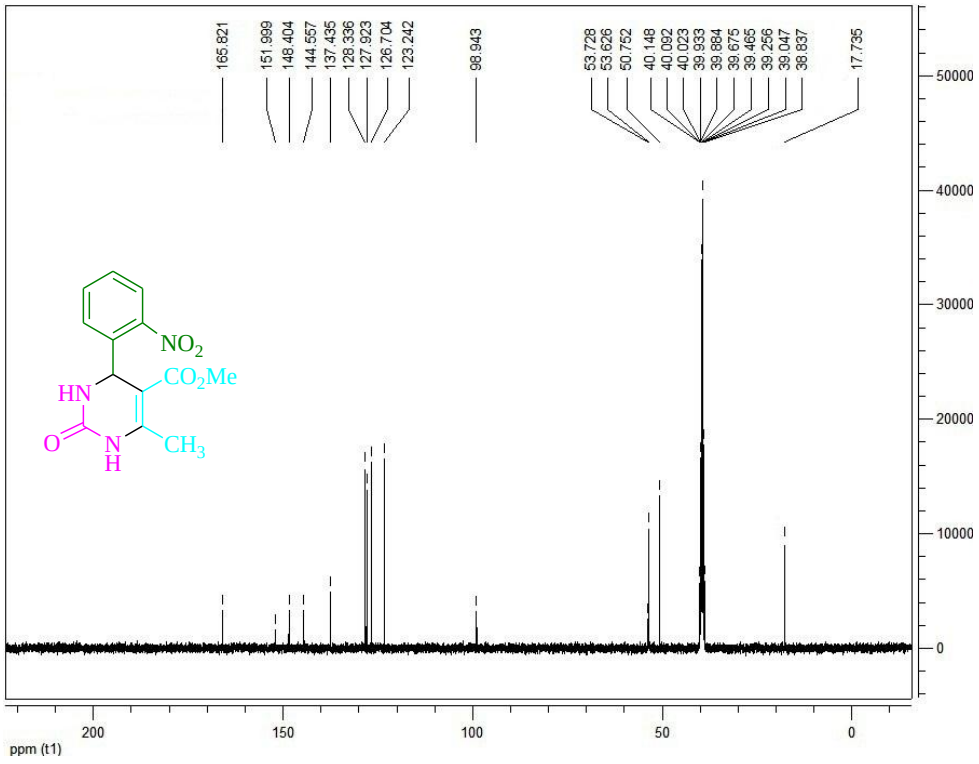


Fig. S5 ^{13}CNMR spectrum of compound (100 MHz, DMSO-d_6) of **4b**

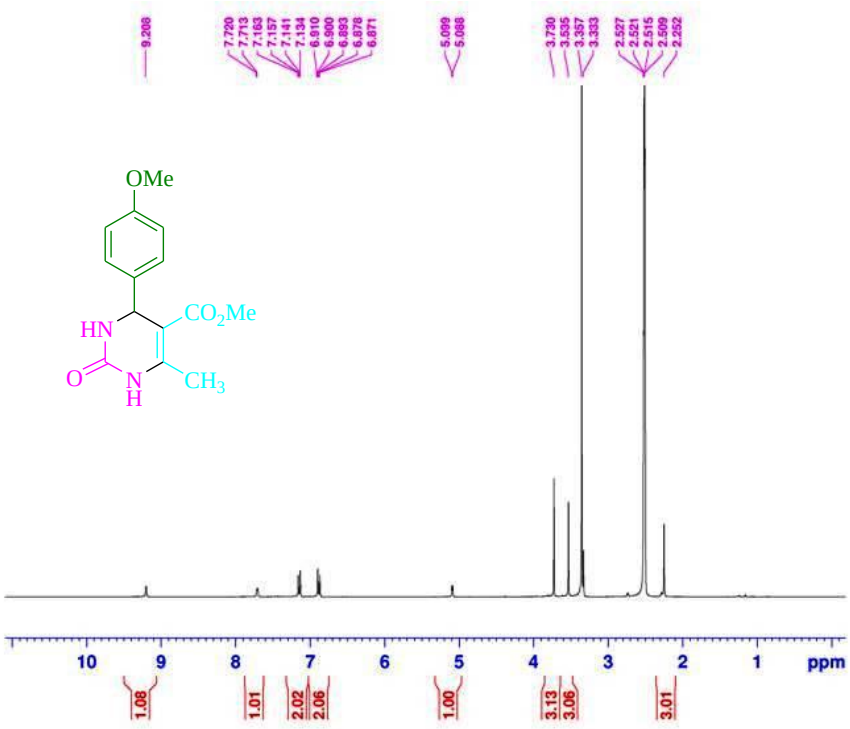


Fig. S6 [1]HNMR spectrum of compound (300 MHz, DMSO-d_6) of **4c**

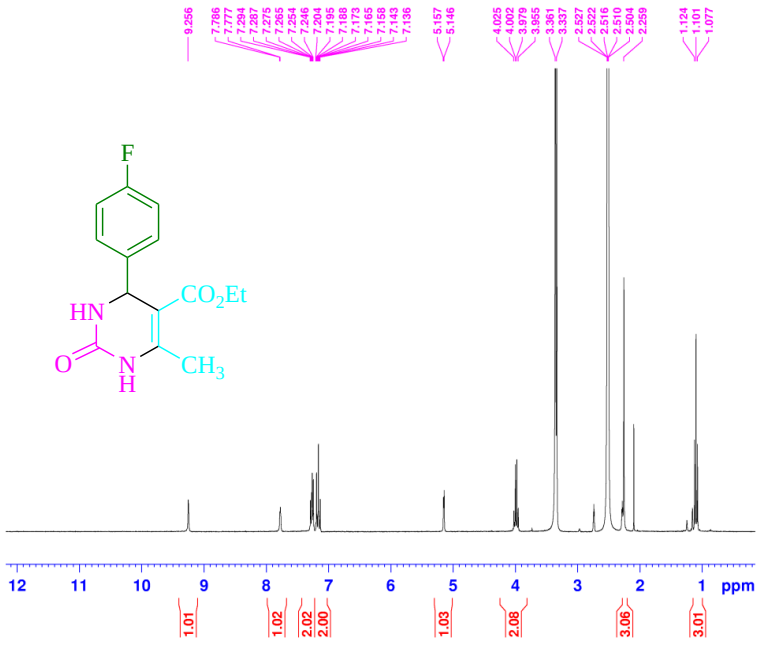


Fig. S7 [1]HNMR spectrum of compound (300 MHz, DMSO-d_6) of **4d**

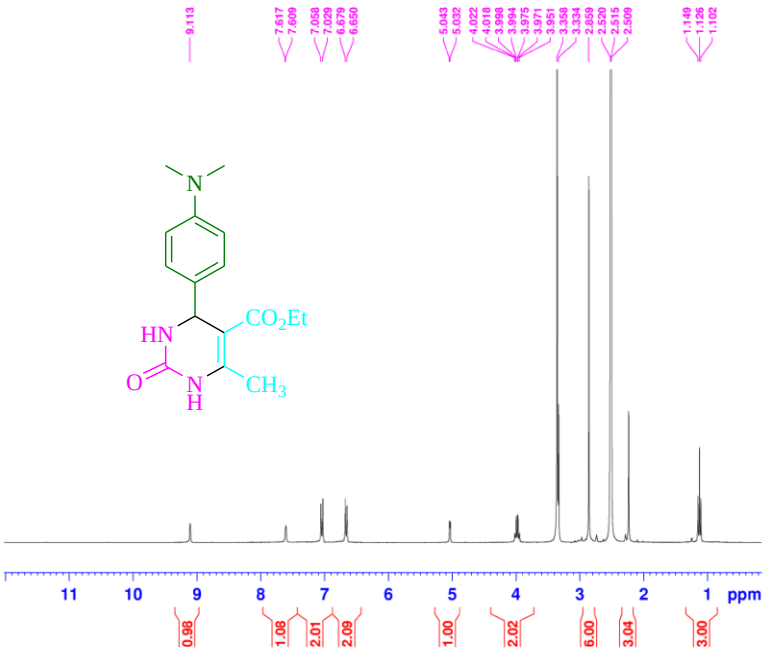


Fig. S8 [1]HNMR spectrum of compound (300 MHz, DMSO-d_6) of **4e**

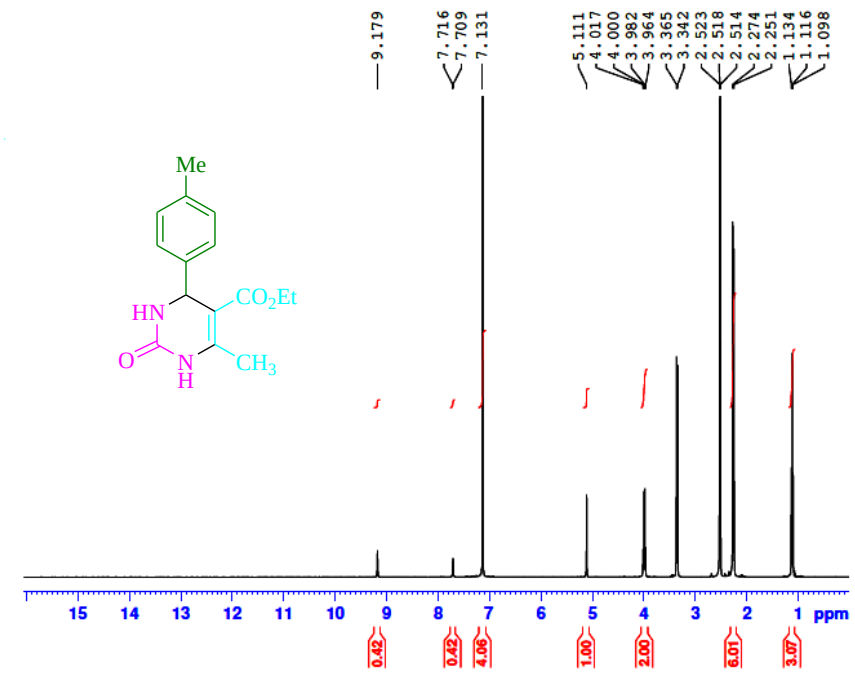


Fig. S9 [1]HNMR spectrum of compound (400 MHz, DMSO-d_6) of **4f**

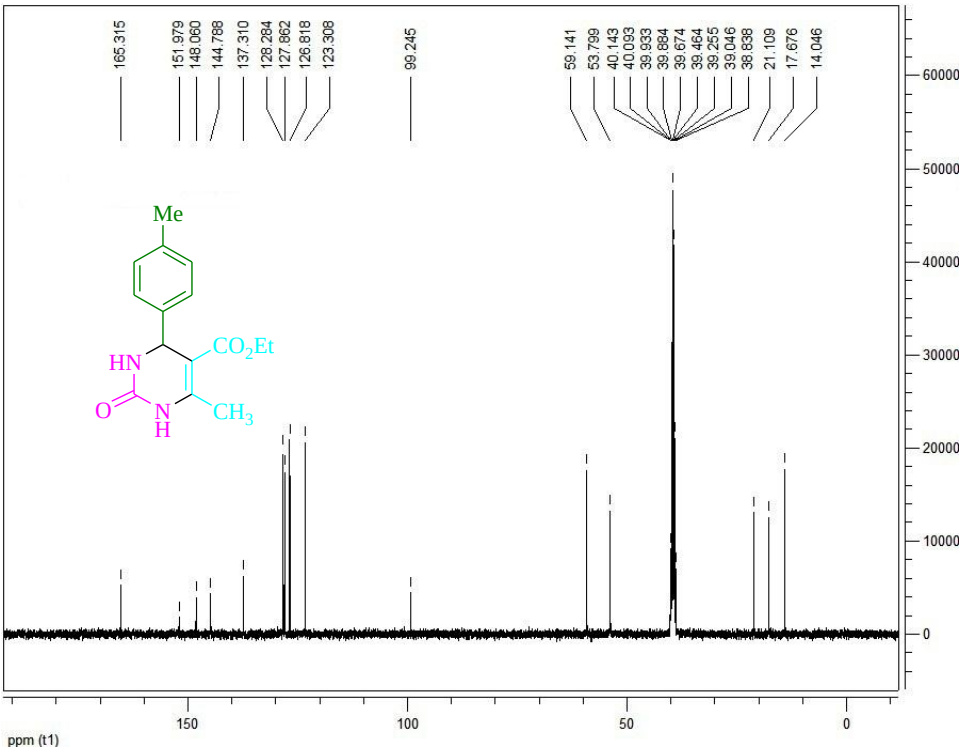


Fig S10 ^{13}CNMR spectrum of compound (100 MHz, DMSO-d_6) of **4f**

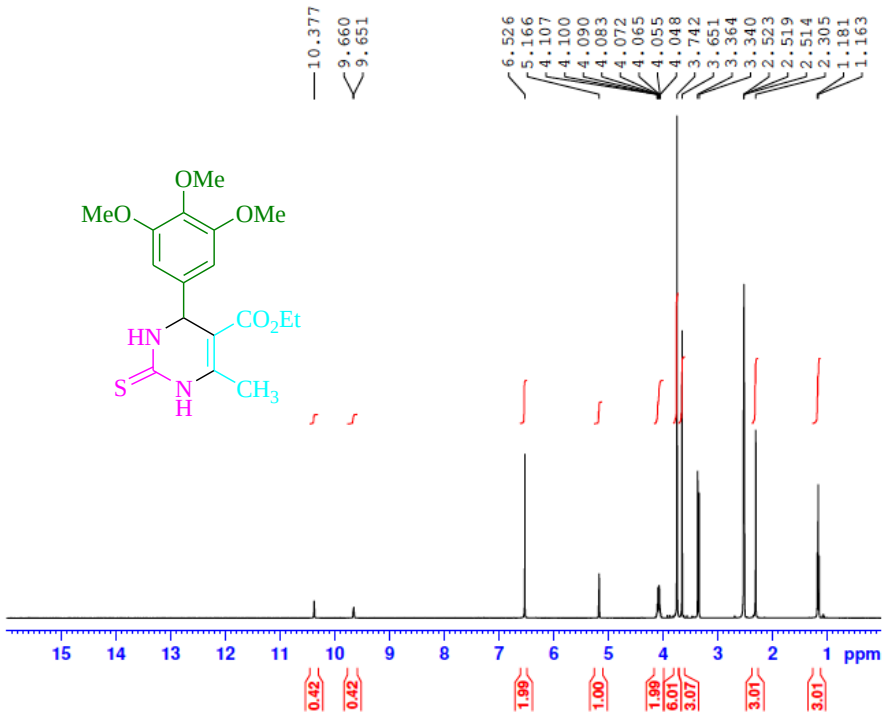


Fig. S11 ^{1}HNMR spectrum of compound (400 MHz, DMSO-d_6) of **4g**

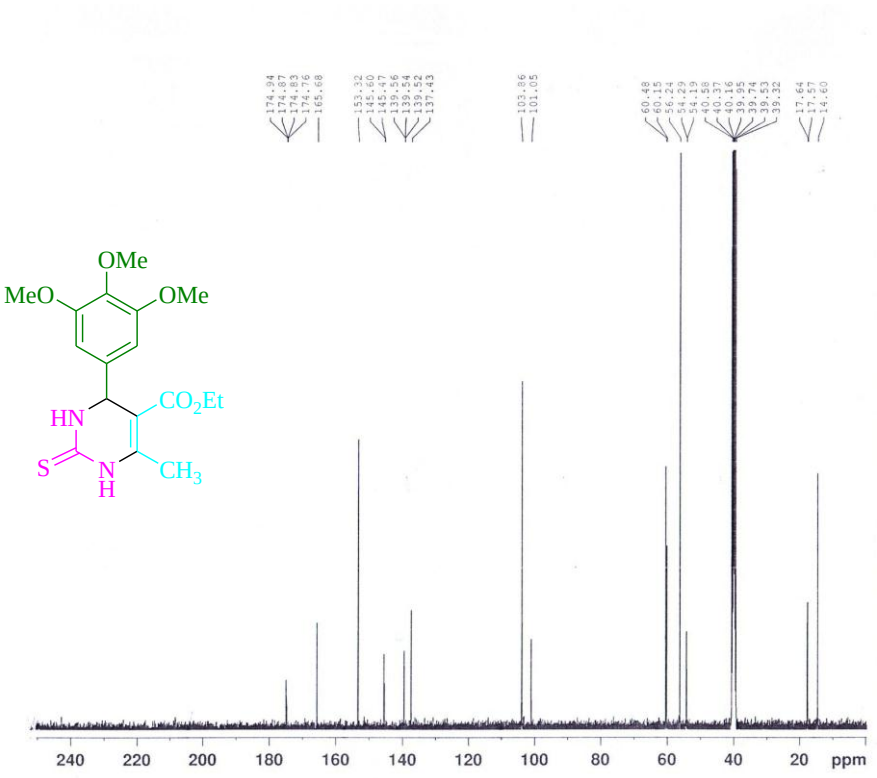


Fig. S12 ^{13}CNMR spectrum of compound (100 MHz, DMSO-d_6) of **4g**

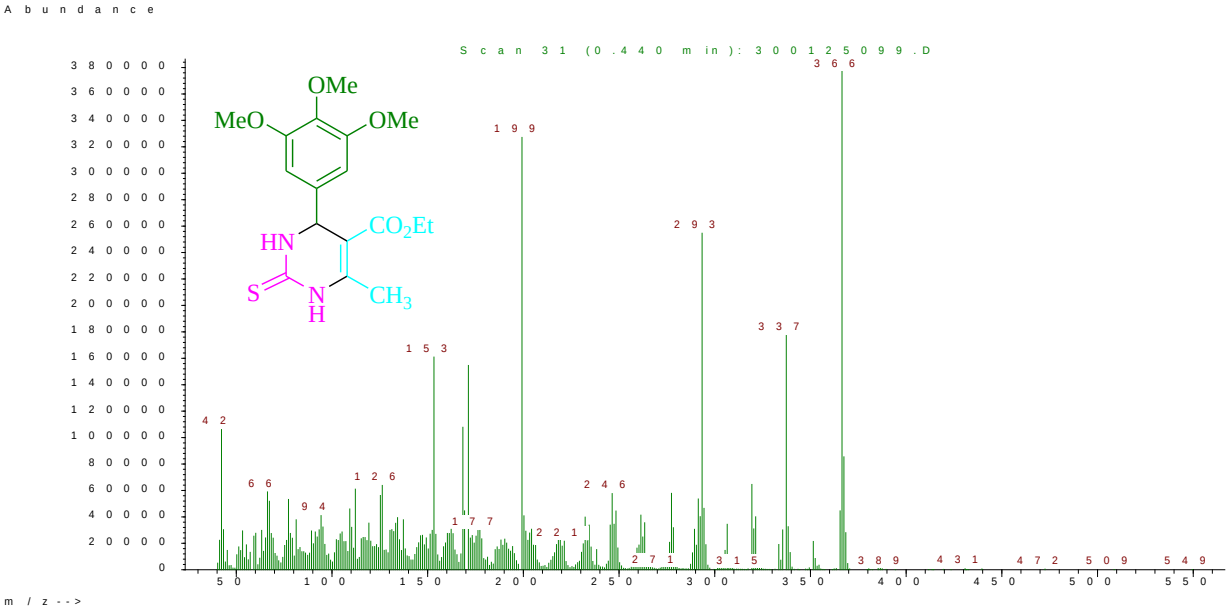


Fig. S13 Mass spectrum of compound **4g**

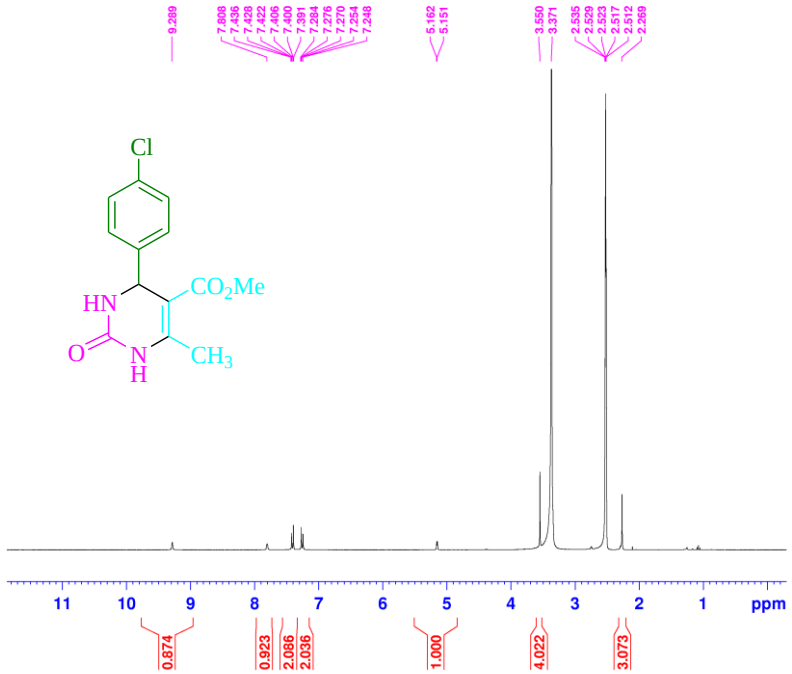


Fig. S14 [1]HNMR spectrum of compound (300 MHz, DMSO-d_6) of **4j**

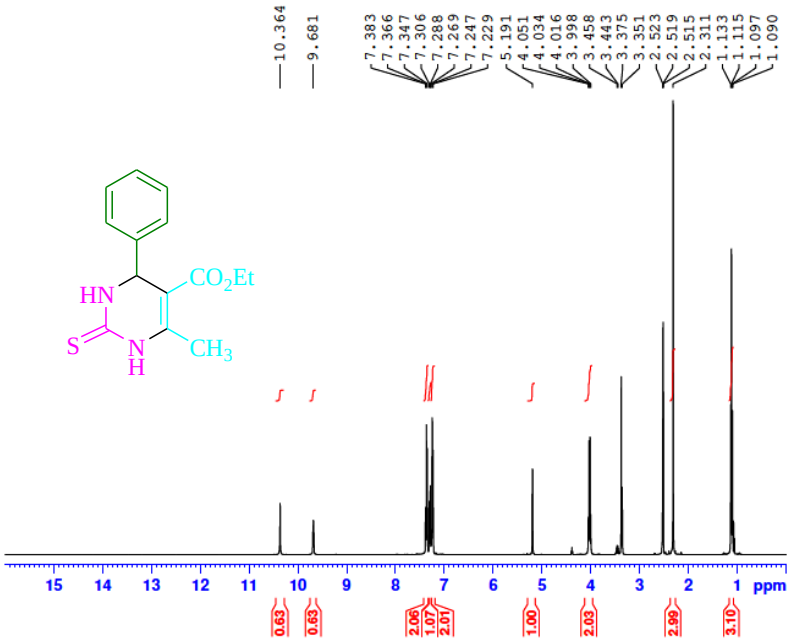


Fig. S15 [1]HNMR spectrum of compound (400 MHz, DMSO-d_6) of **4k**

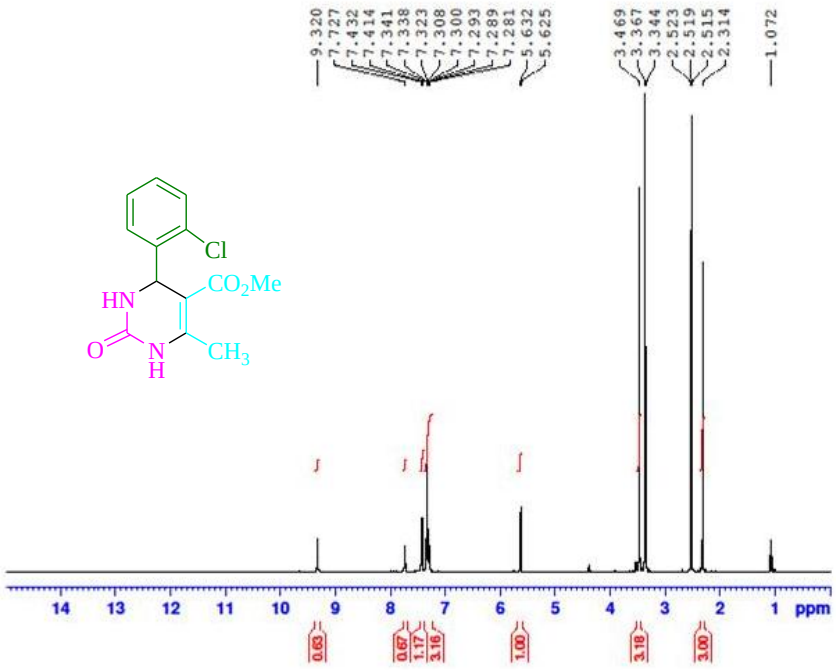


Fig. S16 [1]HNMR spectrum of compound (400 MHz, DMSO-d_6) of **4l**

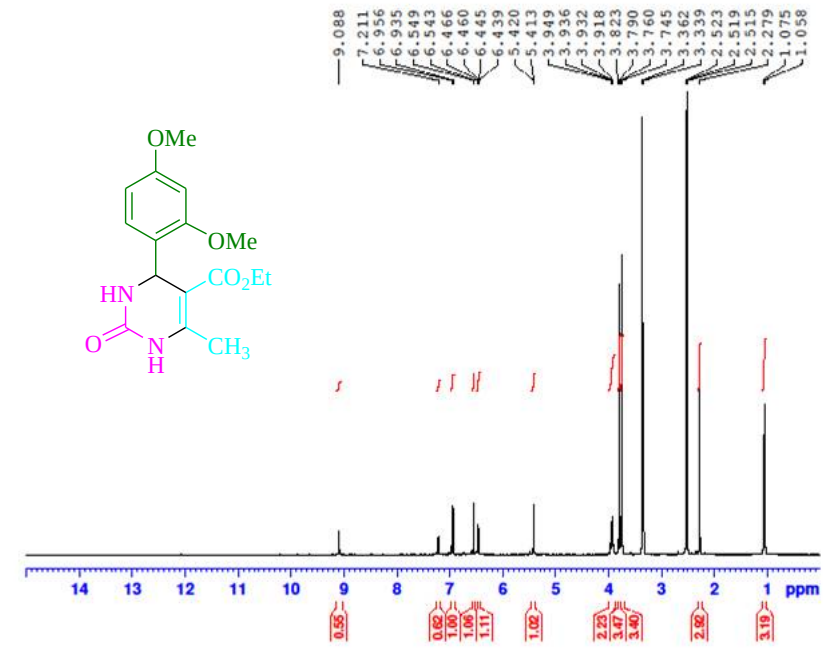


Fig. S17 [1]HNMR spectrum of compound (400 MHz, DMSO-d_6) of **4m**

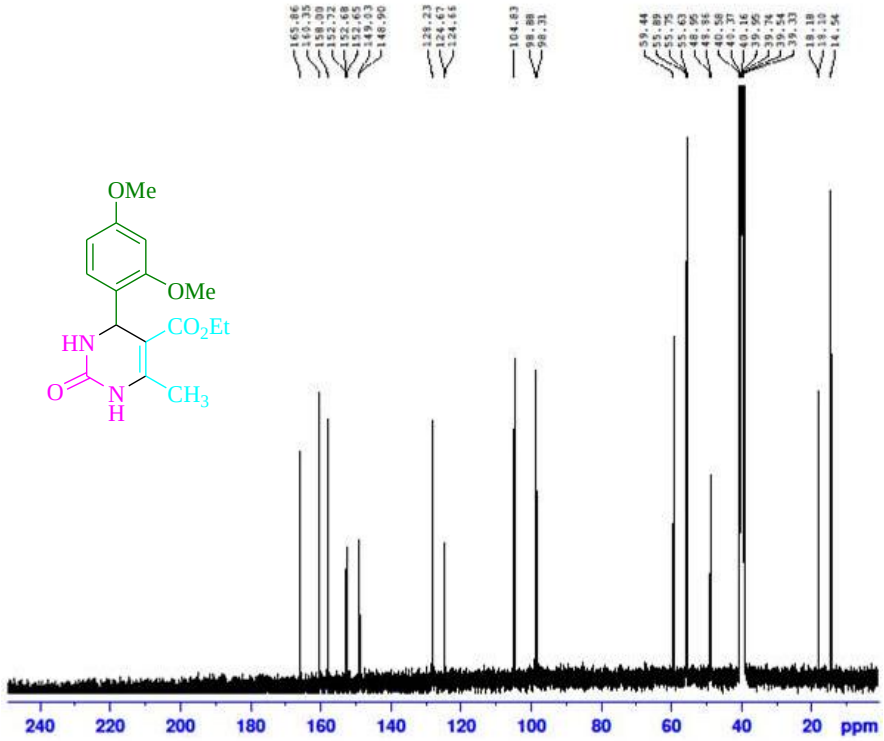


Fig. S18 ^{13}CNMR spectrum of compound (100 MHz, DMSO-d_6) of **4m**

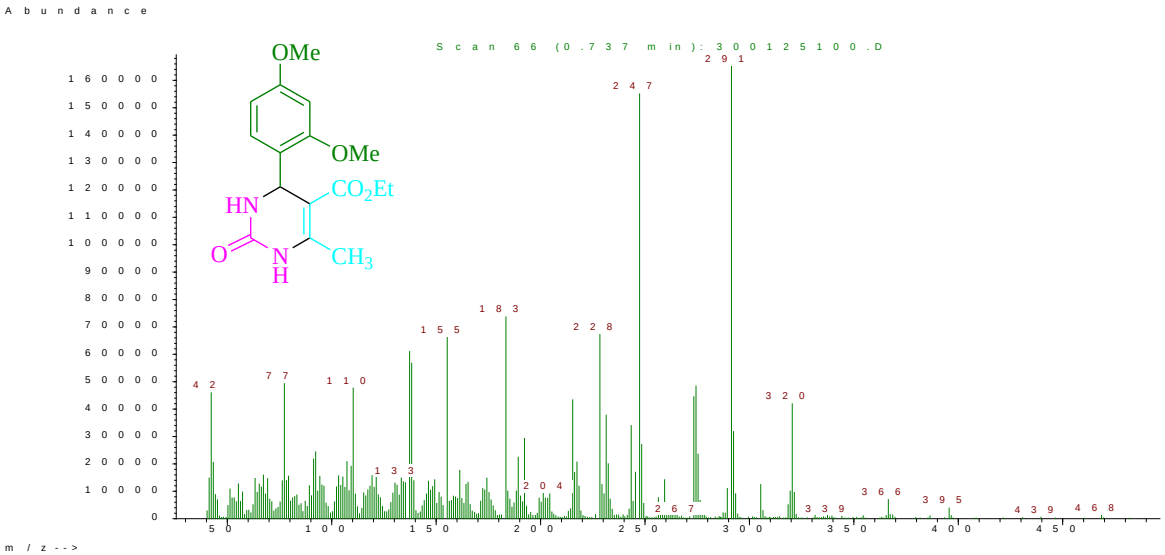


Fig. S19 Mass spectrum of compound **4m**

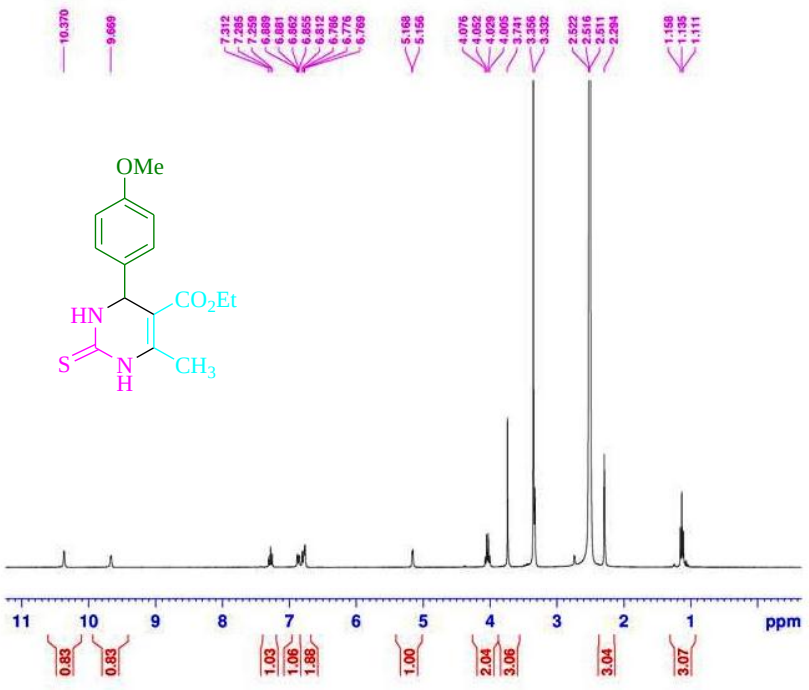


Fig. S20 [1]HNMR spectrum of compound (300 MHz, DMSO-d_6) of **4n**

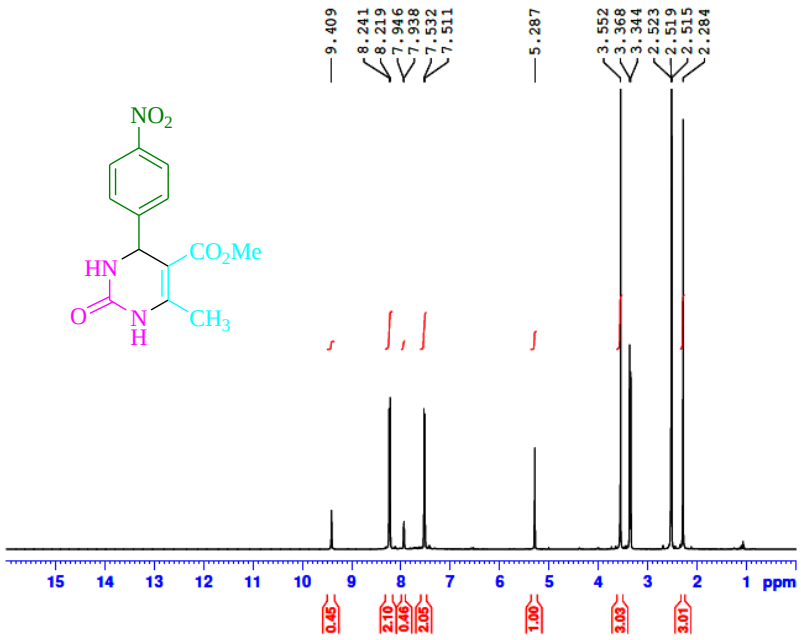


Fig. S21 [1]HNMR spectrum of compound (400 MHz, DMSO-d_6) of **4o**

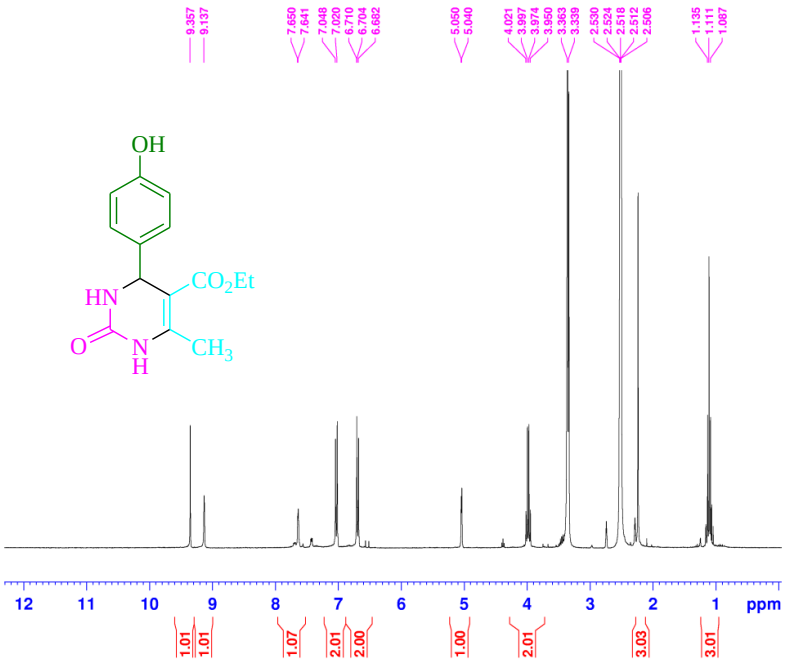


Fig. S22 [1]HNMR spectrum of compound (400 MHz, DMSO-d_6) of **4p**

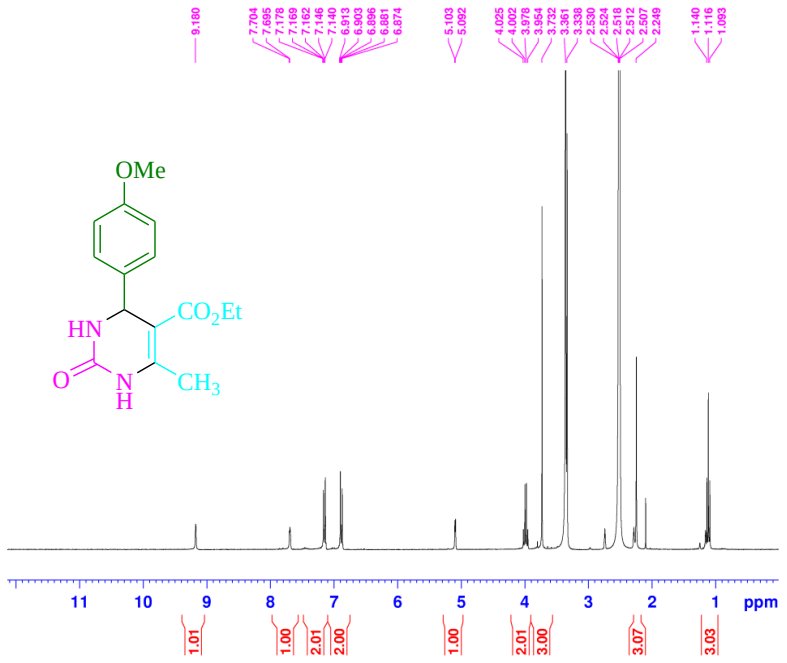


Fig. S23 [1]HNMR spectrum of compound (400 MHz, DMSO-d_6) of **4q**

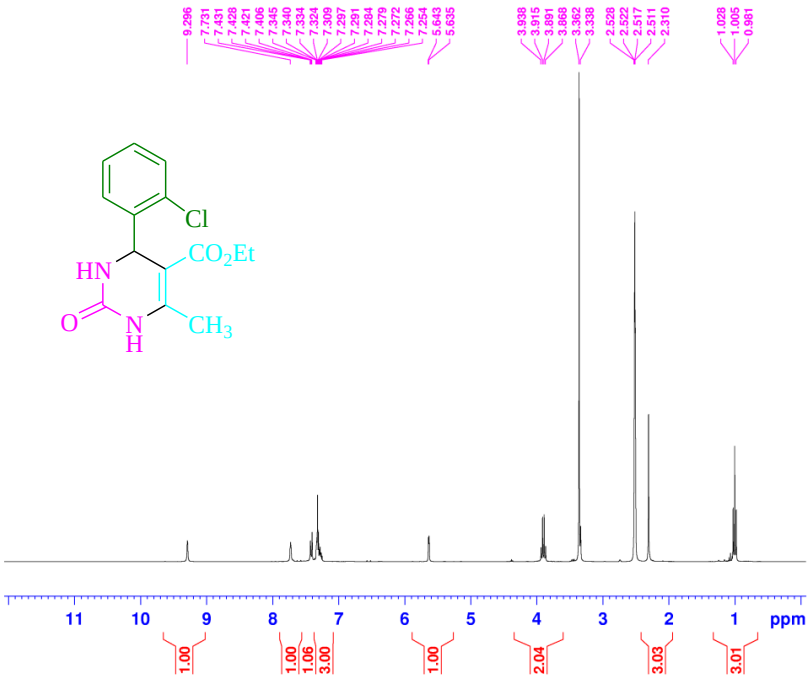


Fig. S24 [1]HNMR spectrum of compound (400 MHz, DMSO-d_6) of **4r**

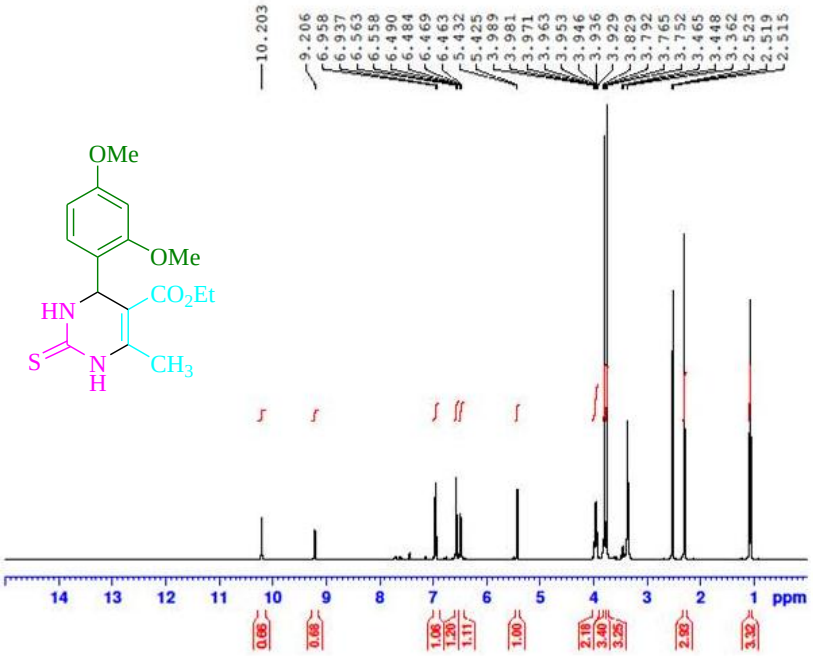


Fig. S25 [1]HNMR spectrum of compound (400 MHz, DMSO-d_6) of **4s**

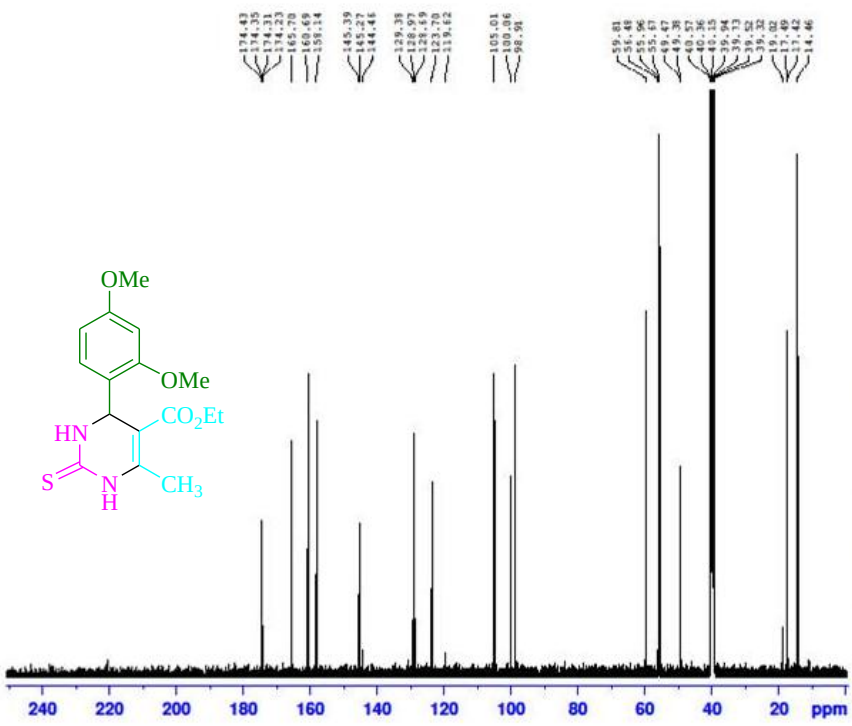


Fig. S26 ^{13}CNMR spectrum of compound (100 MHz, DMSO-d_6) of **4s**

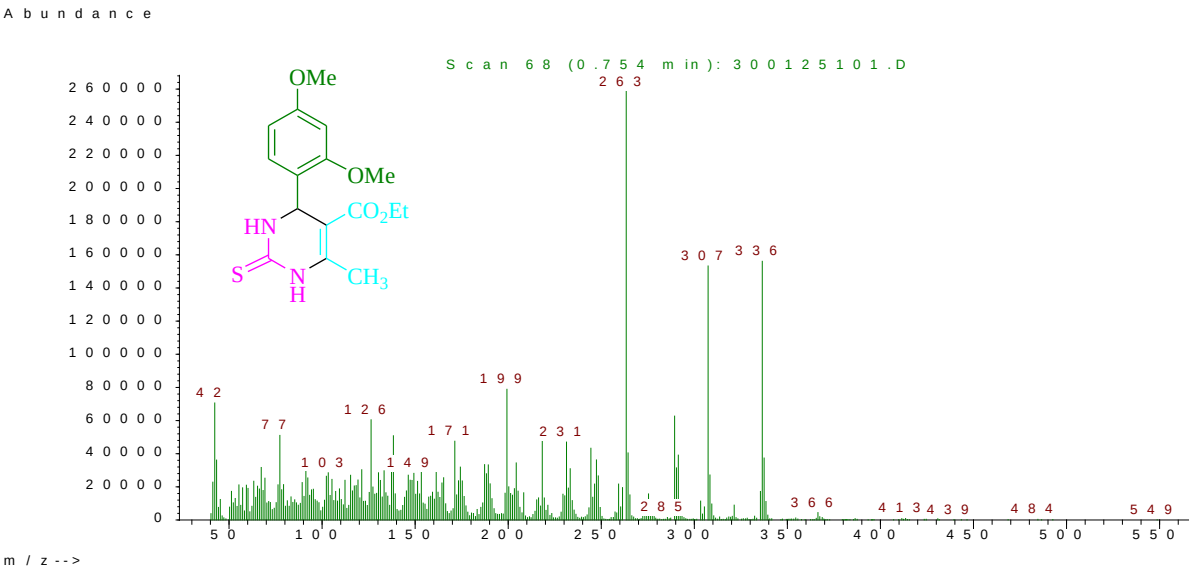


Fig. S27 Mass spectrum of compound **4s**

7. References

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