Supplementary Information (SI) for RSC Advances.
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Supporting Information

One-pot CuI/*L*-proline-catalysed multicomponent synthesis of pyrido[2',1':2,3]imidazo[4,5-*c*]quinoline derivatives from 2-(2-bromophenyl)imidazo[1,2-*a*]pyridines, NH_3 and DMSO under air

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GENERAL INFORMATION

Methods:

Unless otherwise specified, all reactions were performed under air atmosphere using oven-dried glassware. Reported reaction temperatures refer to the external bath temperature unless explicitly noted otherwise. Reaction progress was monitored by thin-layer chromatography (TLC, Merck). TLC analysis was conducted on 0.5 mm. Flash column chromatography was performed using 200–300 mesh silica gel. Analytical data of literature known compounds were in accordance with reported data.

Materials:

All commercial chemicals (purity>98%) and anhydrous solvents were obtained from commercial suppliers (Sigma Aldrich, Aladdin). Unless otherwise noted, all reagents were used without further purification.

Instrument:

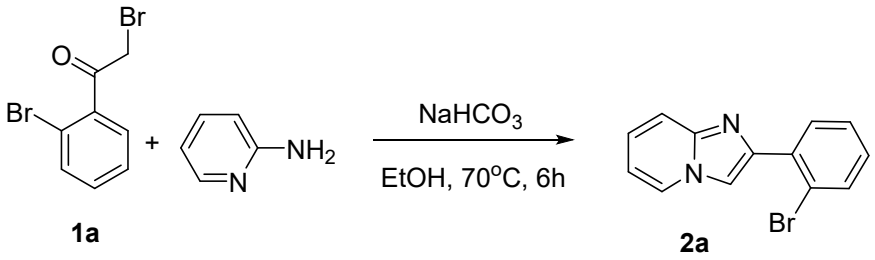
NMR spectra were measured in the Bruker Avance 600 machine (^{1}H NMR (600 MHz), ^{13}C NMR (151 MHz)). HRMS was carried out in electrospray ionization time-of-flight (ESI-TOF) mode using an Agilent 6210 mass spectrometer.

Abbreviations:

Equiv = equivalents, h = hours, min = minutes, ppm = parts per million, DMSO = dimethyl sulfoxide, NBS = N-Bromosuccinimide, EtOAc = ethylacetate, NH_4OAc = Ammonium acetate, Ph = phenyl, Me = methyl, EtOH = Ethanol, Et = ethyl, LDA = lithium diisopropylamide, OMe = tosyl.

EXPERIMENTAL SECTION

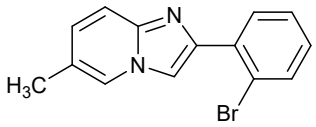
General procedure A for prepared of 2-(2-bromophenyl)imidazo[1,2-a]pyridine 3a and derivatives [1-3]



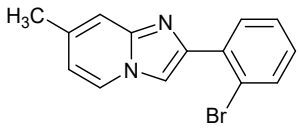
1a **2a**

To a solution of 2-bromo-1-(2-bromophenyl)ethan-1-one **1a** (500 mg, 1,8 mmol) and 2-aminopyridine (186 mg, 2 mmol) and sodium bicarbonate (151 mg, 1.8 mmol) in ethanol (4 mL). The resulting mixture was heated at 70°C and stirred for 6 hours. Upon completion, the mixture was cooled to room temperature, and the solvent was evaporated in vacuo. The residue was partitioned between ethyl acetate and water. The organic phase was separated, dried over anhydrous Na_2SO_4, filtered, and concentrated. Purification of the crude product by silica gel column chromatography

(eluent: hexanes/ethyl acetate = 4:1) afforded 2-(2-bromophenyl)imidazo[1,2-*a*]pyridine **2a** is brown syrup (403 mg, 82%). ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.26 (s, 1H), 8.14 (dd, *J* = 7.82, 1.75 Hz, 1H), 8.10 (dt, *J* = 6.79, 1.19 Hz, 1H), 7.65 (dd, *J* = 8.00, 1.26 Hz, 1H), 7.61 (dd, *J* = 9.12, 1.03 Hz, 1H), 7.40 (ddd, *J* = 7.78, 7.27, 1.26 Hz, 1H), 7.18 – 7.13 (m, 2H), 6.75 (td, *J* = 6.77, 1.18 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 144.4, 143.1, 134.3, 133.5, 131.5, 128.8, 127.4, 125.6, 124.7, 121.4, 117.5, 112.3, 111.9.

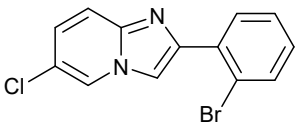


2-(2-bromophenyl)-6-methylimidazo[1,2-a]pyridine **2b** prepared following general procedure A using 2-bromo-1-(2-bromophenyl)ethanone **1a** (500 mg, 1,8 mmol) and 5-methylpyridin-2-amine (214 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 3:1) to yield **2b** (439 mg, 85 %) as a white solid. [4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.20 (s, 1H), 8.14 (dd, *J* = 7.85, 1.76 Hz, 1H), 7.93 (d, *J* = 1.84 Hz, 1H), 7.66 (dd, *J* = 8.07, 1.25 Hz, 1H), 7.53 (d, *J* = 9.20 Hz, 1H), 7.42 – 7.39 (m, 1H), 7.19 – 7.14 (m, 1H), 7.03 (dd, *J* = 9.27, 1.69 Hz, 1H), 2.33 (s, 3H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 143.6, 142.9, 134.6, 133.6, 131.6, 128.7, 128.0, 127.5, 123.4, 122.1, 121.5, 116.9, 111.8, 18.1.

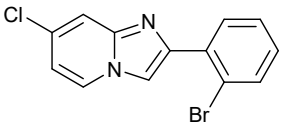


2-(2-bromophenyl)-7-methylimidazo[1,2-a]pyridine **2c** prepared following general procedure A using 2-bromo-1-(2-bromophenyl)ethanone **1a** (500 mg, 1,8 mmol) and 4-methylpyridin-2-amine (214 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 3:1) to yield **2c** (434g, 84 %) as a white solid. [4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.23 – 8.20 (m, 1H), 8.15 (dd, *J* = 7.83, 1.74 Hz, 1H), 8.03 (dd, *J* = 6.90, 1.02 Hz, 1H), 7.66 (dd, *J* = 8.00, 1.24 Hz, 1H), 7.43 – 7.37 (m, 1H), 7.17 (ddd, *J* = 8.00, 7.31, 1.76 Hz, 1H), 6.63 (dd, *J* = 6.92, 1.65 Hz,

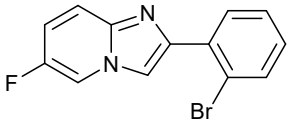
1H), 2.41 (s, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 145.0, 142.9, 135.8, 134.6, 133.6, 131.6, 128.7, 127.5, 124.9, 121.5, 115.9, 115.2, 111.5, 21.4.



2-(2-bromophenyl)-6-chloroimidazo[1,2-a]pyridine **2d** prepared following general procedure A using 2-bromo-1-(2-bromophenyl)ethanone **1a** (500 mg, 1,8 mmol) and 5-chloropyridin-2-amine (254 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 3:1) to yield **2d** (409 mg, 74 %) as a white solid. [4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.26 (s, 1H), 8.20 (dd, *J* = 1.99, 0.91 Hz, 1H), 8.11 (dd, *J* = 7.81, 1.74 Hz, 1H), 7.67 (dd, *J* = 8.00, 1.25 Hz, 1H), 7.58 (dt, *J* = 9.57, 0.82 Hz, 1H), 7.42 (ddd, *J* = 7.81, 7.32, 1.26 Hz, 1H), 7.20 (ddd, *J* = 8.02, 7.31, 1.78 Hz, 1H), 7.16 (dd, *J* = 9.55, 1.98 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 144.3, 142.9, 134.0, 133.7, 131.6, 129.2, 127.6, 126.3, 123.5, 121.5, 120.7, 118.0, 112.4.

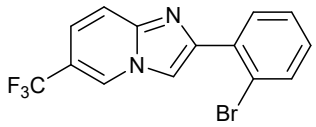


2-(2-bromophenyl)-7-chloroimidazo[1,2-a]pyridine **2e** prepared following general procedure A using 2-bromo-1-(2-bromophenyl)ethanone **1a** (500 mg, 1,8 mmol) and 4-chloropyridin-2-amine (254 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 3:1) to yield **2e** (426 mg, 77 %) as a white solid.[4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.26 (d, *J* = 5.26 Hz, 1H), 8.15 – 8.02 (m, 2H), 7.68 – 7.60 (m, 2H), 7.41 (td, *J* = 7.58, 2.23 Hz, 1H), 7.22 – 7.16 (m, 1H), 6.79 (tt, *J* = 6.94, 1.89 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 144.3, 144.2, 134.0, 133.7, 131.6, 131.2, 129.2, 127.6, 125.9, 121.5, 116.5, 114.2, 112.1.

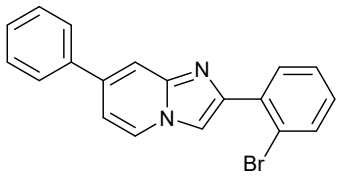


2-(2-bromophenyl)-6-fluoroimidazo[1,2-a]pyridine **2f** prepared following general procedure A using 2-bromo-1-(2-bromophenyl)ethanone **1a** (500 mg, 1,8 mmol) and 5-fluoropyridin-2-amine (222 mg, 2 mmol). The product was purified by column

chromatography (silica gel, Hexane/ethylacetate 3:1) to yield **2f** (346 mg, 66 %) as a white solid.[4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.27 (s, 1H), 8.12 – 8.06 (m, 3H), 7.66 (dd, *J* = 7.99, 1.40 Hz, 1H), 7.60 (dd, *J* = 9.85, 5.11 Hz, 1H), 7.41 (td, *J* = 7.57, 1.36 Hz, 1H), 7.20 – 7.16 (m, 1H), 7.11 (ddt, *J* = 9.97, 7.83, 1.61 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 153.25 (d, *J* = 237.03 Hz), 144.5, 142.3, 134.2, 133.7, 131.5, 129.1, 127.6, 121.5, 118.0 (d, *J* = 8.92 Hz), 116.9 (d, *J* = 25.74 Hz), 113.3, 112.3 (d, *J* = 40.54 Hz).

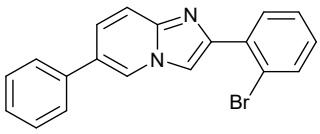


2-(2-bromophenyl)-6-(trifluoromethyl)imidazo[1,2-a]pyridine **2g** prepared following general procedure A using 2-bromo-1-(2-bromophenyl)ethanone **1a** (500 mg, 1,8 mmol) and 5-(trifluoromethyl)pyridin-2-amine (321 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 3:1) to yield **2g** (399 mg, 65 %) as a white solid.[4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.52 (s, 1H), 8.37 (s, 1H), 8.13 (dd, *J* = 7.81, 1.75 Hz, 1H), 7.73 – 7.69 (m, 1H), 7.67 (dd, *J* = 8.08, 1.26 Hz, 1H), 7.41 (td, *J* = 7.58, 1.26 Hz, 1H), 7.32 (dd, *J* = 9.45, 1.83 Hz, 1H), 7.22 – 7.17 (m, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 145.1, 144.1, 133.8, 133.6, 131.7, 129.5, 127.7, 124.9 (q, *J* = 5.70 Hz), 123.5 (q, *J* = 271.08 Hz), 121.6, 120.7 (q, *J* = 2.66 Hz), 118.3, 116.9 (q, *J* = 34.21 Hz), 113.2.

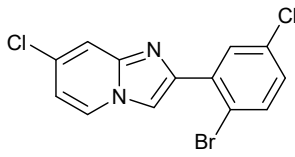


2-(2-bromophenyl)-7-phenylimidazo[1,2-a]pyridine **2h** prepared following general procedure A using 2-bromo-1-(2-bromophenyl)ethanone **1a** (500 mg, 1,8 mmol) and 4-phenylpyridin-2-amine (337 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 8:1) to yield **2h** (503 mg, 80 %) as a white solid.[4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.29 (s, 1H), 8.20 – 8.15 (m, 2H), 7.84 (s, 1H), 7.69 – 7.64 (m, 3H), 7.48 (dd, *J* = 8.44, 6.97 Hz, 2H), 7.44 – 7.38 (m, 2H), 7.20 – 7.16 (m, 1H), 7.08 (dd, *J* = 7.06, 1.83 Hz, 1H). ^{13}C NMR

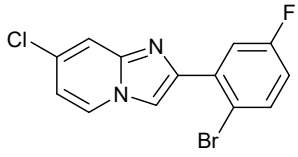
(151 MHz, $CDCl_3$) δ 149.7, 147.2, 145.8, 143.4, 137.9, 135.6, 129.8, 129.4, 129.3, 128.9, 128.7, 127.1, 126.9, 125.1, 122.8, 114.6, 112.6, 29.7.



2-(2-bromophenyl)-6-phenylimidazo[1,2-a]pyridine **2i** prepared following general procedure A using 2-bromo-1-(2-bromophenyl)ethanone **1a** (500 mg, 1,8 mmol) and 5-phenylpyridin-2-amine (337 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 8:1) to yield **2i** (509 mg, 81 %) as a white solid.[4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.34 (s, 1H), 8.32 (s, 1H), 8.17 (dd, *J* = 7.77, 1.70 Hz, 1H), 7.69 (t, *J* = 8.79 Hz, 2H), 7.59 – 7.56 (m, 2H), 7.47 (q, *J* = 8.32 Hz, 3H), 7.41 (dt, *J* = 14.65, 7.64 Hz, 2H), 7.19 (td, *J* = 7.65, 1.69 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 143.9, 143.7, 137.3, 134.4, 133.7, 131.7, 129.2, 128.9, 127.9, 127.6, 127.1, 126.9, 125.7, 123.0, 121.5, 117.5, 112.5.

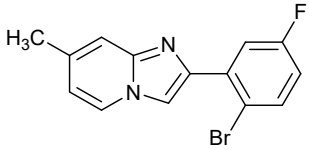


2-(2-bromo-5-chlorophenyl)-7-chloroimidazo[1,2-a]pyridine **2j** prepared following general procedure A using 2-bromo-1-(2-bromo-5-chlorophenyl)ethan-1-one **1b** (562 mg, 1.8 mmol) and 4-chloropyridin-2-amine (254 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 4:1) to yield **2j** (431 mg, 70 %) as a white solid.[4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.31 (s, 1H), 8.21 (dd, *J* = 1.99, 0.88 Hz, 1H), 8.17 (d, *J* = 2.65 Hz, 1H), 7.60 – 7.57 (m, 2H), 7.17 (ddd, *J* = 10.02, 9.03, 2.31 Hz, 2H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 143.0, 142.9, 135.5, 134.8, 133.9, 131.3, 129.1, 126.7, 123.6, 120.9, 119.1, 118.1, 112.6.

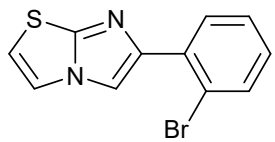


2-(2-bromo-5-fluorophenyl)-7-chloroimidazo[1,2-a]pyridine **2k** prepared following general procedure A using 2-bromo-1-(2-bromo-5-fluorophenyl)ethan-1-one **1c** (533 mg, 1.8 mmol) and 4-chloropyridin-2-amine (254 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 4:1) to

yield **2k** (369 mg, 63 %) as a white solid.[4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.28 (s, 1H), 8.14 (d, *J* = 1.44 Hz, 1H), 7.90 (dd, *J* = 9.99, 3.14 Hz, 1H), 7.57 (dd, *J* = 8.81, 5.36 Hz, 1H), 7.53 (d, *J* = 9.58 Hz, 1H), 7.13 (dd, *J* = 9.53, 1.98 Hz, 1H), 6.89 (ddd, *J* = 8.74, 7.41, 3.17 Hz, 1H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 162.0 (d, *J* = 246.63 Hz), 143.1 (d, *J* = 2.10 Hz), 142.8, 135.8 (d, *J* = 8.30 Hz), 135.0 (d, *J* = 8.07 Hz), 126.6, 123.6, 120.9, 118.2 (d, *J* = 24.58 Hz), 117.99, 116.3 (d, *J* = 22.76 Hz), 115.3 (d, *J* = 3.23 Hz), 112.5.

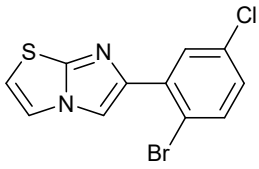


2-(2-bromo-5-fluorophenyl)-7-methylimidazo[1,2-a]pyridine **2l** prepared following general procedure A using 2-bromo-1-(2-bromo-5-fluorophenyl)ethan-1-one **1c** (533 mg, 1.8 mmol) and 4-methylpyridin-2-amine (214 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 4:1) to yield **2l** (395 mg, 72 %) as a white solid.[4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.26 (d, *J* = 0.77 Hz, 1H), 8.00 (dd, *J* = 6.94, 0.99 Hz, 1H), 7.94 (dd, *J* = 10.13, 3.14 Hz, 1H), 7.59 (dd, *J* = 8.78, 5.37 Hz, 1H), 7.36 (dh, *J* = 2.20, 1.12 Hz, 1H), 6.88 (ddd, *J* = 8.81, 7.44, 3.16 Hz, 1H), 6.62 (dd, *J* = 6.91, 1.68 Hz, 1H), 2.39 (d, *J* = 1.18 Hz, 3H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 162.0 (d, *J* = 246.41 Hz), 144.9, 141.7 (d, *J* = 2.08 Hz), 136.4 (d, *J* = 8.33 Hz), 136.0, 134.1 (d, *J* = 8.12 Hz), 124.9, 118.1 (d, *J* = 24.53 Hz), 115.8, 115.8, 115.6, 115.2 (d, *J* = 3.02 Hz), 111.6, 21.3.

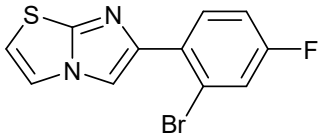


6-(2-bromophenyl)imidazo[2,1-b]thiazole **2m** were synthesized according to our previously published method.[4] Using 2-bromo-1-(2-bromophenyl)ethanone **1a** (500 mg, 1,8 mmol) and thiazol-2-amine (198 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 10:1) to yield **2m** (437 mg, 87 %) as a white solid.[4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.15 (s, 1H), 8.06 (dd, *J* = 7.86,

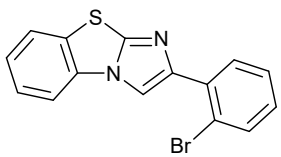
1.77 Hz, 1H), 7.62 (dd, *J* = 8.01, 1.27 Hz, 1H), 7.39 – 7.33 (m, 2H), 7.10 (ddd, *J* = 7.93, 7.27, 1.76 Hz, 1H), 6.76 (d, *J* = 4.46 Hz, 1H).[4]



6-(2-bromo-5-chlorophenyl)imidazo[2,1-b]thiazole **2n** prepared following general procedure A using 2-bromo-1-(2-bromo-5-chlorophenyl)ethan-1-one **1b** (562 mg, 1.8 mmol) and thiazol-2-amine (198 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 10:1) to yield **2n** (452 mg, 80 %) as a white solid.[4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.24 (s, 1H), 8.12 (d, *J* = 2.66 Hz, 1H), 7.54 (dd, *J* = 8.55, 0.71 Hz, 1H), 7.44 (dd, *J* = 4.47, 0.74 Hz, 1H), 7.09 (ddd, *J* = 8.43, 2.66, 0.69 Hz, 1H), 6.85 (dd, *J* = 4.47, 0.78 Hz, 1H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 149.5, 143.8, 135.9, 134.8, 133.7, 130.7, 128.2, 118.5, 118.3, 113.3, 112.5.

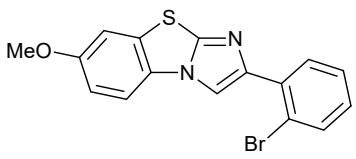


6-(2-bromo-4-fluorophenyl)imidazo[2,1-b]thiazole **2o** prepared following general procedure A using 2-bromo-1-(2-bromo-4-fluorophenyl)ethan-1-one **1d** (562 mg, 1,8 mmol) and thiazol-2-amine (198 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 10:1) to yield **2o** (401 mg, 75 %) as a white solid.[4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.12 (s, 1H), 8.04 (dd, *J* = 8.81, 6.27 Hz, 1H), 7.44 (d, *J* = 4.47 Hz, 1H), 7.38 (dd, *J* = 8.33, 2.63 Hz, 1H), 7.10 (ddd, *J* = 8.81, 7.75, 2.68 Hz, 1H), 6.84 (d, *J* = 4.49 Hz, 1H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 161.2 (d, *J* = 250.81 Hz), 149.3, 144.3, 132.1 (d, *J* = 8.22 Hz), 130.9 (d, *J* = 3.66 Hz), 120.7 (d, *J* = 3.96 Hz), 120.6 (d, *J* = 11.29 Hz), 118.5, 114.8 (d, *J* = 20.94 Hz), 112.9, 111.8.



2-(2-bromophenyl)benzo[d]imidazo[2,1-b]thiazole **2p** prepared following general procedure A using 2-bromo-1-(2-bromophenyl)ethanone **1a** (500 mg, 1,8 mmol) and benzo[d]thiazol-2-amine (297 mg, 2 mmol). The product was

purified by column chromatography (silica gel, Hexane/ethylacetate 50:1) to yield **2p** (356 mg, 60 %) as a white solid. [4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.43 (s, 1H), 8.13 (dd, *J* = 7.85, 1.76 Hz, 1H), 7.70 (ddd, *J* = 7.97, 1.15, 0.54 Hz, 1H), 7.65 (ddd, *J* = 11.45, 7.97, 1.14 Hz, 2H), 7.44 (ddd, *J* = 8.20, 7.42, 1.16 Hz, 1H), 7.40 (ddd, *J* = 7.88, 7.29, 1.29 Hz, 1H), 7.34 (ddd, *J* = 7.99, 7.38, 1.16 Hz, 1H), 7.15 (ddd, *J* = 7.98, 7.27, 1.75 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 147.3, 144.8, 134.2, 133.8, 132.2, 131.0, 130.4, 128.5, 127.5, 126.2, 125.0, 124.4, 120.7, 112.9, 111.1.

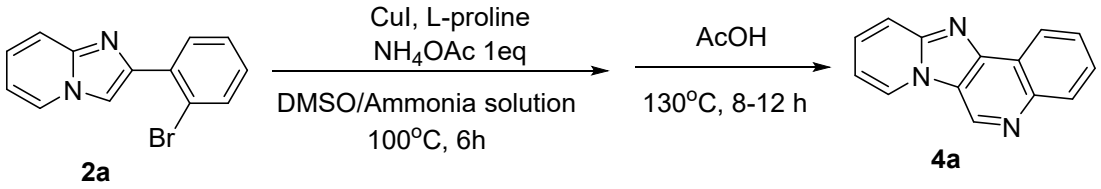


2-(2-bromophenyl)-7-methoxybenzo[d]imidazo[2,1-b]thiazole **2q** prepared following general procedure A using 2-bromo-1-(2-bromophenyl)ethanone **1a** (500 mg, 1,8 mmol) and 6-methoxybenzo[d]thiazol-2-amine (357 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 50:1) to yield **2q** (356 mg, 55 %) as a white solid.[4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.38 (s, 1H), 8.12 (dd, *J* = 7.84, 1.74 Hz, 1H), 7.66 (dd, *J* = 8.00, 1.25 Hz, 1H), 7.55 (d, *J* = 8.79 Hz, 1H), 7.39 (ddd, *J* = 7.81, 7.27, 1.27 Hz, 1H), 7.22 (d, *J* = 2.44 Hz, 1H), 7.14 (ddd, *J* = 7.99, 7.28, 1.75 Hz, 1H), 7.01 (dd, *J* = 8.81, 2.47 Hz, 1H), 3.88 (s, 3H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 157.4, 146.7, 144.4, 134.3, 133.7, 131.7, 130.9, 128.4, 127.5, 126.5, 120.6, 113.4, 113.4, 111.0, 108.8, 55.9.

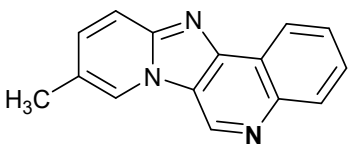
2-(2-bromophenyl)-7-fluorobenzo[d]imidazo[2,1-b]thiazole **2r** prepared following general procedure A using 2-bromo-1-(2-bromophenyl)ethan-1-one **1a** (533 mg, 1.8 mmol) and 6-fluorobenzo[d]thiazol-2-amine (297 mg, 2 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 50:1) to yield **2r** (350 mg, 56 %) as a white solid.[4] ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.41 (s, 1H), 8.11 (dd, *J* = 7.84, 1.75 Hz, 1H), 7.66 (dd, *J* = 8.02, 1.23 Hz, 1H), 7.61 (dd, *J*

= 8.81, 4.34 Hz, 1H), 7.44 (dd, *J* = 7.99, 2.51 Hz, 1H), 7.42 – 7.37 (m, 1H), 7.22 – 7.11 (m, 2H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 159.8 (d, *J* = 245.79 Hz), 146.9, 144.9, 134.0, 133.8, 131.8 (d, *J* = 10.43 Hz), 130.9, 128.8 (d, *J* = 2.25 Hz), 128.6, 127.6, 120.7, 113.9 (d, *J* = 24.66 Hz), 113.5 (d, *J* = 9.00 Hz), 111.5 (d, *J* = 27.45 Hz), 111.2.

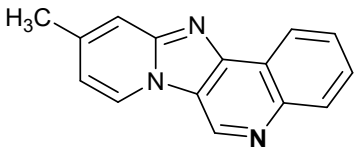
General procedure B for prepared of pyrido[2',1':2,3]imidazo[4,5-c]quinoline 4a and derivatives



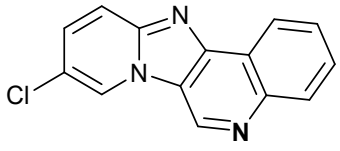
The compound 2-(2-bromophenyl)imidazo[1,2-*a*]pyridine **2a** (137 mg, 0.5 mmol), CuI (19 mg, 0.1 mmol), L-Proline (12 mg, 0.1 mmol) and NH_4OAc (39 mg, 0.5 mmol) were added into a 25 ml reaction tube. Then, DMSO solvent (1.2 ml) and ammonia solution (0.8 ml) were added. The reaction mixture was then stirred magnetically and heated at 100°C for 6 h under air atmosphere. Next, the reaction mixture was cooled to 70°C, the reaction tube was opened for 2 min, and 0.5 ml of AcOH was slowly added. The tube was sealed, and the temperature was raised to 130°C for 8-12 h. After cooling, the reaction mixture was extracted with water and ethyl acetate. The organic layer was dried with Na_2SO_4, then filtered and evaporated under reduced pressure to remove the solvent. The brown residue was purified by column chromatography (silica gel, dichloromethane/ethyl acetate 1:1) to obtain pyrido[2',1':2,3]imidazo[4,5-*c*]quinoline **4a** (77 mg, 70%) as a white solid, mp = 239°C. ^{1}H NMR (600 MHz, $CDCl_3$) δ 9.52 (s, 1H), 8.76 (ddt, *J* = 8.0, 1.7, 0.9 Hz, 2H), 8.29 (ddd, *J* = 8.2, 1.3, 0.6 Hz, 1H), 7.95 (dt, *J* = 9.2, 1.1 Hz, 1H), 7.80 (ddd, *J* = 8.4, 6.9, 1.6 Hz, 1H), 7.74 (ddd, *J* = 8.1, 6.9, 1.2 Hz, 1H), 7.62 (ddd, *J* = 9.2, 6.7, 1.3 Hz, 1H), 7.14 – 7.09 (m, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 149.2, 146.6, 145.9, 135.7, 130.4, 129.8, 128.7, 126.9, 125.3, 122.8, 122.2, 118.3, 112.7. [5, 6]



9-methylpyrido[2',1':2,3]imidazo[4,5-c]quinoline **4b** prepared following general procedure B using compound **2b** (114 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 1:1) to yield **4b** (86 mg, 74 %) as a white solid, mp = 250^{0}C. ^{1}H NMR (600 MHz, $CDCl_3$) δ 9.47 (s, 1H), 8.73 (ddd, *J* = 8.0, 1.6, 0.6 Hz, 1H), 8.52 (q, *J* = 1.4 Hz, 1H), 8.30 – 8.24 (m, 1H), 7.83 (dd, *J* = 9.2, 1.0 Hz, 1H), 7.78 (ddd, *J* = 8.4, 6.9, 1.6 Hz, 1H), 7.72 (ddd, *J* = 8.1, 6.9, 1.3 Hz, 1H), 7.46 (dd, *J* = 9.3, 1.7 Hz, 1H), 2.49 (s, 3H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 148.3, 146.5, 145.7, 135.8, 133.5, 129.7, 128.5, 126.8, 122.9, 122.7, 122.6, 122.2, 122.0, 117.5, 18.2. [5, 6]

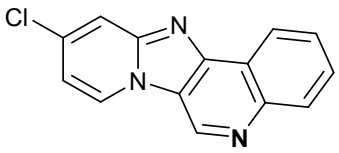


10-methylpyrido[2',1':2,3]imidazo[4,5-c]quinoline **4c** prepared following general procedure B using compound **2c** (114 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 1:1) to yield **4c** (85 mg, 73 %) as a white solid, mp = 243^{0}C. ^{1}H NMR (600 MHz, $CDCl_3$) δ 9.47 (s, 1H), 8.73 (ddd, *J* = 8.1, 1.6, 0.6 Hz, 1H), 8.61 (dd, *J* = 6.9, 1.0 Hz, 1H), 8.26 (ddd, *J* = 8.3, 1.3, 0.6 Hz, 1H), 7.78 (ddd, *J* = 8.4, 6.9, 1.6 Hz, 1H), 7.71 (ddd, *J* = 8.1, 6.9, 1.2 Hz, 1H), 7.67 (dq, *J* = 2.2, 1.1 Hz, 1H), 6.93 (dd, *J* = 6.9, 1.6 Hz, 1H), 2.55 (s, 3H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 149.8, 146.9, 145.7, 142.2, 135.4, 129.6, 128.5, 126.7, 124.4, 122.8, 122.1, 122.1, 116.5, 115.4, 22.0. [5, 6]

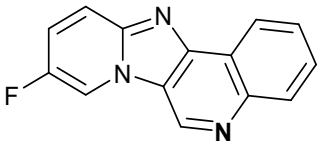


9-chloropyrido[2',1':2,3]imidazo[4,5-c]quinoline **4d** prepared following general procedure B using compound **2d** (154 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 1:1) to yield **4d** (89 mg, 70 %) as a white solid, mp = 265^{0}C. ^{1}H NMR (600 MHz, $CDCl_3$) δ 9.48 (s, 1H), 8.78 (d, *J* =

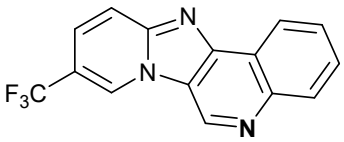
0.9 Hz, 1H), 8.73 (ddd, *J* = 8.0, 1.5, 0.6 Hz, 1H), 8.29 (ddd, *J* = 8.3, 1.2, 0.6 Hz, 1H), 7.89 (dd, *J* = 9.6, 0.9 Hz, 1H), 7.81 (ddd, *J* = 8.4, 7.0, 1.6 Hz, 1H), 7.75 (ddd, *J* = 8.1, 6.9, 1.2 Hz, 1H), 7.57 (dd, *J* = 9.6, 2.0 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 147.3, 146.9, 145.9, 135.7, 131.6, 129.9, 128.9, 127.2, 123.3, 122.7, 122.1, 122.0, 120.9, 118.6.[6]



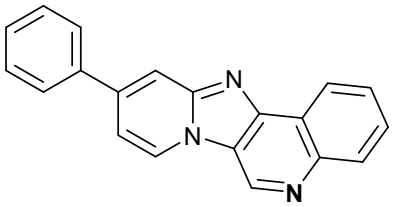
10-chloropyrido[2',1':2,3]imidazo[4,5-c]quinoline **4e** prepared following general procedure B using compound **2e** (154 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 1:1) to yield **4e** (84 mg, 66 %) as a white solid, mp = 256^{0}C. ^{1}H NMR (600 MHz, $CDCl_3$) δ 9.47 (s, 1H), 8.70 (ddd, *J* = 8.0, 1.5, 0.6 Hz, 1H), 8.64 (d, *J* = 7.2 Hz, 1H), 8.27 (d, *J* = 8.1 Hz, 1H), 7.91 (dd, *J* = 2.0, 0.8 Hz, 1H), 7.80 (ddd, *J* = 8.4, 6.9, 1.6 Hz, 1H), 7.77 – 7.72 (m, 1H), 7.08 (dd, *J* = 7.2, 2.0 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 149.0, 147.1, 145.7, 137.2, 135.4, 129.7, 129.0, 127.2, 125.5, 122.8, 121.9, 121.9, 117.1, 114.5; HRMS (ESI): *m/z* calcd for $C_{14}H_8ClN_3$ $[M+H]^+$= 254.0407; found = 254.0481.



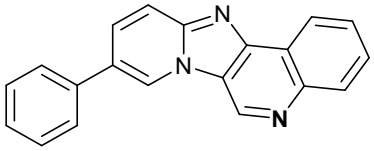
9-fluoropyrido[2',1':2,3]imidazo[4,5-c]quinoline **4f** prepared following general procedure B using compound **2f** (146 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 1:1) to yield **4f** (85 mg, 72 %) as a white solid, mp = 270^{0}C. ^{1}H NMR (600 MHz, Acetone) δ 9.74 (s, 1H), 9.41 (ddd, *J* = 4.0, 2.5, 0.8 Hz, 1H), 8.68 (ddd, *J* = 8.0, 1.6, 0.6 Hz, 1H), 8.22 (ddd, *J* = 8.3, 1.3, 0.6 Hz, 1H), 7.95 (ddd, *J* = 9.9, 5.1, 0.9 Hz, 1H), 7.81 – 7.75 (m, 2H), 7.75 – 7.72 (m, 1H). ^{13}C NMR (151 MHz, Acetone) δ 152.8 (d, *J* = 235.7 Hz), 146.8, 146.6 (d, *J* = 2.1 Hz), 145.8, 137.4, 129.9, 129.4, 128.2, 126.5, 122.4, 122.4, 122.3, 118.3 (d, *J* = 8.9 Hz), 113.9 (d, *J* = 41.5 Hz); HRMS (ESI): *m/z* calcd for $C_{14}H_8FN_3$ $[M+H]^+$= 238.0702; found = 238.0776.



9-(trifluoromethyl)pyrido[2',1':2,3]imidazo[4,5-c]quinoline **4g** prepared following general procedure B using compound **2g** (171 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 1:1) to yield **4g** (108 mg, 75 %) as a white solid, mp = 265^{0}C. ^{1}H NMR (600 MHz, Acetone) δ 9.88 (s, 1H), 9.82 (s, 1H), 8.68 (d, *J* = 8.0 Hz, 1H), 8.24 (d, *J* = 8.3 Hz, 1H), 8.05 (d, *J* = 9.5 Hz, 1H), 7.92 (dd, *J* = 9.6, 1.9 Hz, 1H), 7.86 – 7.79 (m, 1H), 7.79 – 7.72 (m, 1H). ^{13}C NMR (151 MHz, Acetone) δ 148.6, 146.9, 146.2, 137.7, 129.9, 128.6, 127.2 (q, *J* = 5.7 Hz), 126.7, 125.8 (q, *J* = 2.8 Hz), 124.0 (q, *J* = 270.2 Hz), 123.0, 122.6, 122.0, 118.4, 115.6 (q, *J* = 34.5 Hz); HRMS (ESI): *m/z* calcd for $C_{15}H_8F_3N_3$ $[M+H]^+$= 288.0670; found = 288.0744.

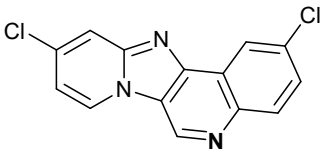


10-phenylpyrido[2',1':2,3]imidazo[4,5-c]quinoline **4h** prepared following general procedure B using compound **2h** (174 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 1:1) to yield **4h** (103 mg, 70 %) as a white solid, mp = 276^{0}C. ^{1}H NMR (600 MHz, $CDCl_3$) δ 9.50 (s, 1H), 8.76 (d, *J* = 8.0 Hz, 2H), 8.28 (d, *J* = 8.3 Hz, 1H), 8.12 (s, 1H), 7.79 (ddd, *J* = 8.4, 6.9, 1.5 Hz, 1H), 7.75 (tt, *J* = 7.0, 1.4 Hz, 3H), 7.57 – 7.53 (m, 2H), 7.51 – 7.46 (m, 1H), 7.38 (dd, *J* = 7.0, 1.8 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 145.0, 144.0, 138.7, 137.9, 134.4, 133.7, 131.7, 129.1, 128.9, 128.3, 127.6, 126.7, 125.6, 121.5, 114.3, 112.5, 111.7; HRMS (ESI): *m/z* calcd for $C_{20}H_{13}N_3$ $[M+H]^+$= 296.1109; found = 296.1183.

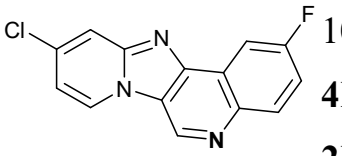


9-phenylpyrido[2',1':2,3]imidazo[4,5-c]quinoline **4i** prepared following general procedure B using compound **2i** (174 mg, 0.5 mmol). The product was purified by

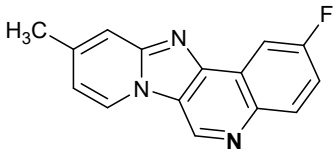
column chromatography (silica gel, Hexane/ethylacetate 1:1) to yield **4i** (96 mg, 65 %) as a white solid, mp = 268^{0}C. ^{1}H NMR (600 MHz, DMSO) δ 9.96 (s, 1H), 9.77 (s, 1H), 8.63 (d, *J* = 7.9 Hz, 1H), 8.21 (d, *J* = 8.2 Hz, 1H), 8.15 (dd, *J* = 9.4, 1.9 Hz, 1H), 8.04 (d, *J* = 9.4 Hz, 1H), 7.92 (d, *J* = 6.9 Hz, 2H), 7.81 (ddd, *J* = 8.4, 6.9, 1.6 Hz, 1H), 7.79 – 7.74 (m, 1H), 7.58 (t, *J* = 7.7 Hz, 2H), 7.51 – 7.44 (m, 1H). ^{13}C NMR (151 MHz, DMSO) δ 147.8, 145.6, 145.3, 138.2, 136.0, 130.9, 129.5, 129.1, 128.3, 128.1, 126.8, 126.4, 125.6, 124.8, 122.6, 122.3, 121.6, 117.0; HRMS (ESI): *m/z* calcd for $C_{20}H_{13}N_3$ $[M+H]^+$= 295.1109; found = 296.1193.



2,10-dichloropyrido[2',1':2,3]imidazo[4,5-c]quinoline **4j** prepared following general procedure B using compound **2j** (171 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 1:1) to yield **4j** (97 mg, 67 %) as a white solid, mp = 286^{0}C. ^{1}H NMR (600 MHz, DMSO) δ 9.86 (s, 1H), 9.75 (d, *J* = 2.0 Hz, 1H), 8.53 (d, *J* = 2.5 Hz, 1H), 8.21 (d, *J* = 8.8 Hz, 1H), 8.00 (d, *J* = 9.6 Hz, 1H), 7.83 (td, *J* = 9.6, 2.3 Hz, 2H). ^{13}C NMR (151 MHz, DMSO) δ 147.1, 144.6, 143.5, 138.7, 132.2, 131.7, 131.2, 128.8, 126.1, 122.8, 122.4, 121.1, 119.5, 118.0; HRMS (ESI): *m/z* calcd for $C_{14}H_7Cl_2N_3$ $[M+H]^+$= 288.0017; found = 288.0091.

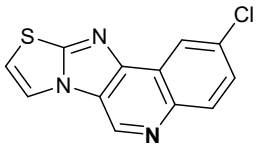


10-chloro-2-fluoropyrido[2',1':2,3]imidazo[4,5-c]quinoline **4k** prepared following general procedure B using compound **2k** (163 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 1:1) to yield **4k** (75 mg, 55 %) as a white solid, mp = 288^{0}C. ^{1}H NMR (600 MHz, $CDCl_3$) δ 9.39 (s, 1H), 8.73 (dt, *J* = 1.8, 0.8 Hz, 1H), 8.31 – 8.23 (m, 2H), 7.85 (dt, *J* = 9.6, 0.8 Hz, 1H), 7.58 – 7.50 (m, 2H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 161.2 (d, *J* = 248.9 Hz), 147.2, 146.4, 142.7, 134.9 (d, *J* = 2.8 Hz), 132.3 (d, *J* = 9.0 Hz), 131.8, 123.3, 123.1 (d, *J* = 10.5 Hz), 122.1, 121.1, 118.7, 118.4 (d, *J* = 25.2 Hz), 107.0 (d, *J* = 23.4 Hz). [6]



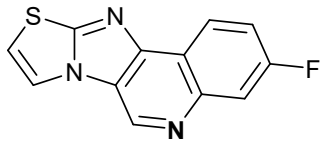
2-fluoro-10-methylpyrido[2',1':2,3]imidazo[4,5-c]quinoline **4l** prepared following general procedure B using compound **2l** (153 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 1:1) to yield **4l** (95 mg, 76 %) as a white solid, mp = 282^{0}C. ^{1}H NMR (600 MHz, DMSO) δ 9.74 (s, 1H), 9.26 (d, *J* = 6.9 Hz, 1H), 8.23 (dd, *J* = 9.1, 5.3 Hz, 1H), 8.18 (dd, *J* = 9.2, 2.9 Hz, 1H), 7.71 (s, 1H), 7.65 (td, *J* = 8.9, 3.0 Hz, 1H), 7.13 (d, *J* = 6.9 Hz, 1H), 2.51 (s, 3H). ^{13}C NMR (126 MHz, DMSO) δ 159.7 (d, *J* = 245.3 Hz), 148.8, 144.9, 142.2, 142.0, 136.5 (d, *J* = 2.5 Hz), 131.9 (d, *J* = 9.2 Hz), 122.3 (d, *J* = 10.0 Hz), 122.1, 116.9 (d, *J* = 24.9 Hz), 115.0, 114.9, 105.9, 105.7, 21.0; HRMS (ESI): *m/z* calcd for $C_{15}H_{10}FN_3$ $[M+H]^+$= 252.0859; found = 252.0933.

thiazolo[2',3':2,3]imidazo[4,5-c]quinoline **4m** prepared following general procedure B using compound **2m** (140 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 3:1) to yield **4m** (82 mg, 73 %) as a white solid, mp = 210^{0}C. ^{1}H NMR (600 MHz, $CDCl_3$) δ 9.31 (s, 1H), 8.61 (ddd, *J* = 8.0, 1.6, 0.7 Hz, 1H), 8.24 (ddd, *J* = 8.4, 1.2, 0.6 Hz, 1H), 7.95 (d, *J* = 4.5 Hz, 1H), 7.77 – 7.72 (m, 1H), 7.70 (ddd, *J* = 8.1, 6.9, 1.3 Hz, 1H), 7.07 (d, *J* = 4.5 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 158.1, 149.7, 145.0, 135.3, 129.7, 128.0, 126.9, 123.3, 122.2, 122.1, 117.9, 113.2; HRMS (ESI): *m/z* calcd for $C_{12}H_7N_3S[M+H]^+$= 226.0361; found = 226.0434.

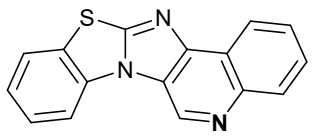


2-chlorothiazolo[2',3':2,3]imidazo[4,5-c]quinoline **4n** prepared following general procedure B using compound **2n** (157 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 3:1) to yield **4n** (78 mg, 60 %) as a white solid, mp = 230^{0}C.[5] ^{1}H NMR (600 MHz, $CDCl_3$) δ 9.28 (s, 1H), 8.58 (d, *J* = 2.4 Hz, 1H), 8.16

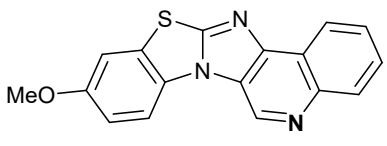
(d, *J* = 8.9 Hz, 1H), 7.96 (d, *J* = 4.5 Hz, 1H), 7.66 (dd, *J* = 8.9, 2.4 Hz, 1H), 7.10 (d, *J* = 4.5 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 158.3, 148.9, 143.3, 135.4, 132.9, 131.3, 128.9, 123.7, 122.9, 121.4, 117.9, 113.7.



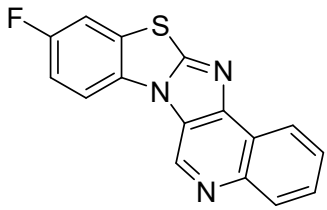
3-fluorothiazolo[2',3':2,3]imidazo[4,5-c]quinoline **4o** prepared following general procedure B using compound **2o** (149 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 3:1) to yield **4o** (79 mg, 65 %) as a white solid, mp = 225^{0}C. ^{1}H NMR (600 MHz, DMSO) δ 9.60 (s, 1H), 8.65 (d, *J* = 4.4 Hz, 1H), 8.52 (dd, *J* = 9.0, 6.3 Hz, 1H), 7.87 (dd, *J* = 10.8, 2.6 Hz, 1H), 7.62 (td, *J* = 8.8, 2.6 Hz, 1H), 7.56 (d, *J* = 4.4 Hz, 1H). ^{13}C NMR (151 MHz, DMSO) δ 161.3 (d, *J* = 244.2 Hz), 158.2, 148.4, 145.1 (d, *J* = 12.3 Hz), 138.2, 124.1 (d, *J* = 10.0 Hz), 123.1, 120.0, 118.5, 116.1 (d, *J* = 24.7 Hz), 114.3, 113.1 (d, *J* = 20.4 Hz), HRMS (ESI): *m/z* calcd for $C_{12}H_6FN_3S[M+H]^+$= 244.0266; found = 244.0341.



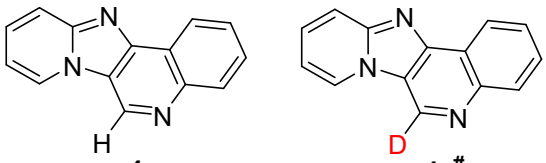
benzo[4',5']thiazolo[2',3':2,3]imidazo[4,5-c]quinoline **4p** prepared following general procedure B using compound **2p** (165 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 3:1) to yield **4p** (98 mg, 71 %) as a white solid, mp = 240^{0}C. ^{1}H NMR (600 MHz, $CDCl_3$) δ 9.61 (s, 1H), 8.63 (dd, *J* = 8.1, 1.6 Hz, 1H), 8.27 (d, *J* = 8.3 Hz, 1H), 8.12 (d, *J* = 8.0 Hz, 1H), 7.84 (d, *J* = 7.4 Hz, 1H), 7.77 (ddd, *J* = 8.4, 6.8, 1.6 Hz, 1H), 7.72 (ddd, *J* = 8.1, 6.8, 1.2 Hz, 1H), 7.66 (td, *J* = 7.8, 1.2 Hz, 1H), 7.48 (td, *J* = 7.7, 1.1 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 156.6, 149.7, 144.9, 134.8, 132.6, 129.7, 129.4, 128.2, 127.3, 127.1, 125.3, 124.7, 122.2, 122.0, 113.3; HRMS (ESI): *m/z* calcd for $C_{16}H_9N_3S[M+H]^+$= 276.0517; found = 276.0591.



10-methoxybenzo[4',5']thiazolo[2',3':2,3]imidazo[4,5-c]quinoline **4q** prepared following general procedure B using compound **2q** (180 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 3:1) to yield **4q** (101 mg, 66 %) as a white solid, mp = 243^{0}C. ^{1}H NMR (600 MHz, $CDCl_3$) δ 9.55 (s, 1H), 8.61 (ddd, *J* = 8.0, 1.6, 0.6 Hz, 1H), 8.28 – 8.23 (m, 1H), 7.98 (d, *J* = 8.8 Hz, 1H), 7.75 (ddd, *J* = 8.4, 6.9, 1.6 Hz, 1H), 7.71 (ddd, *J* = 8.1, 6.9, 1.3 Hz, 1H), 7.32 (d, *J* = 2.5 Hz, 1H), 7.18 (dd, *J* = 8.8, 2.5 Hz, 1H), 3.91 (s, 3H). ^{13}C NMR (151 MHz, $CDCl_3$) δ 157.5, 156.0, 149.3, 144.7, 134.6, 130.8, 129.7, 128.0, 127.0, 126.7, 124.3, 122.1, 114.2, 113.7, 109.2, 56.0; HRMS (ESI): *m/z* calcd for $C_{17}H_{11}N_3OS[M+H]^+$= 306.0623; found = 306.0705.



10-fluorobenzo[4',5']thiazolo[2',3':2,3]imidazo[4,5-c]quinoline **4r** prepared following general procedure B using compound **2r** (174 mg, 0.5 mmol). The product was purified by column chromatography (silica gel, Hexane/ethylacetate 3:1) to yield **4r** (106 mg, 72 %) as a white solid, mp = 260^{0}C. ^{1}H NMR (600 MHz, $CDCl_3$) δ 9.59 (s, 1H), 8.63 (ddd, *J* = 8.01, 1.64, 0.66 Hz, 1H), 8.28 (ddd, *J* = 8.27, 1.32, 0.65 Hz, 1H), 8.07 (dd, *J* = 8.82, 4.19 Hz, 1H), 7.78 (ddd, *J* = 8.39, 6.88, 1.59 Hz, 1H), 7.73 (ddd, *J* = 8.08, 6.85, 1.29 Hz, 1H), 7.58 (dd, *J* = 7.70, 2.48 Hz, 1H), 7.39 (td, *J* = 8.63, 2.53 Hz, 1H). ^{13}C NMR (151 MHz, $CDCl_3$ + CD_3OD) δ 160.2 (d, *J* = 247.61 Hz), 156.91, 149.65, 144.50, 134.55, 130.92 (d, *J* = 10.29 Hz), 129.12, 128.72, 127.6, 124.3, 122.2, 121.9, 115.3 (d, *J* = 24.75 Hz), 114.4 (d, *J* = 8.85 Hz), 112.1 (d, *J* = 27.54 Hz).[6]



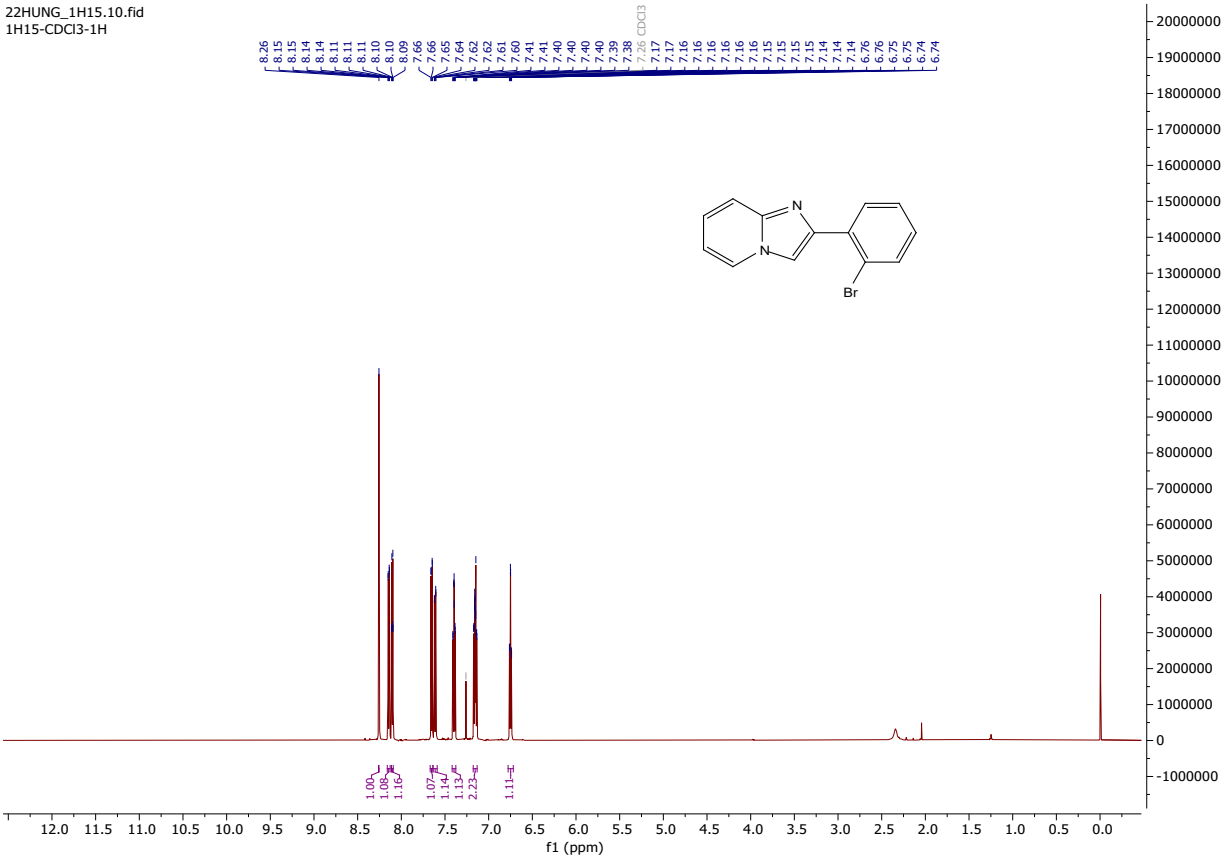
4a **4a$^{\#}$**

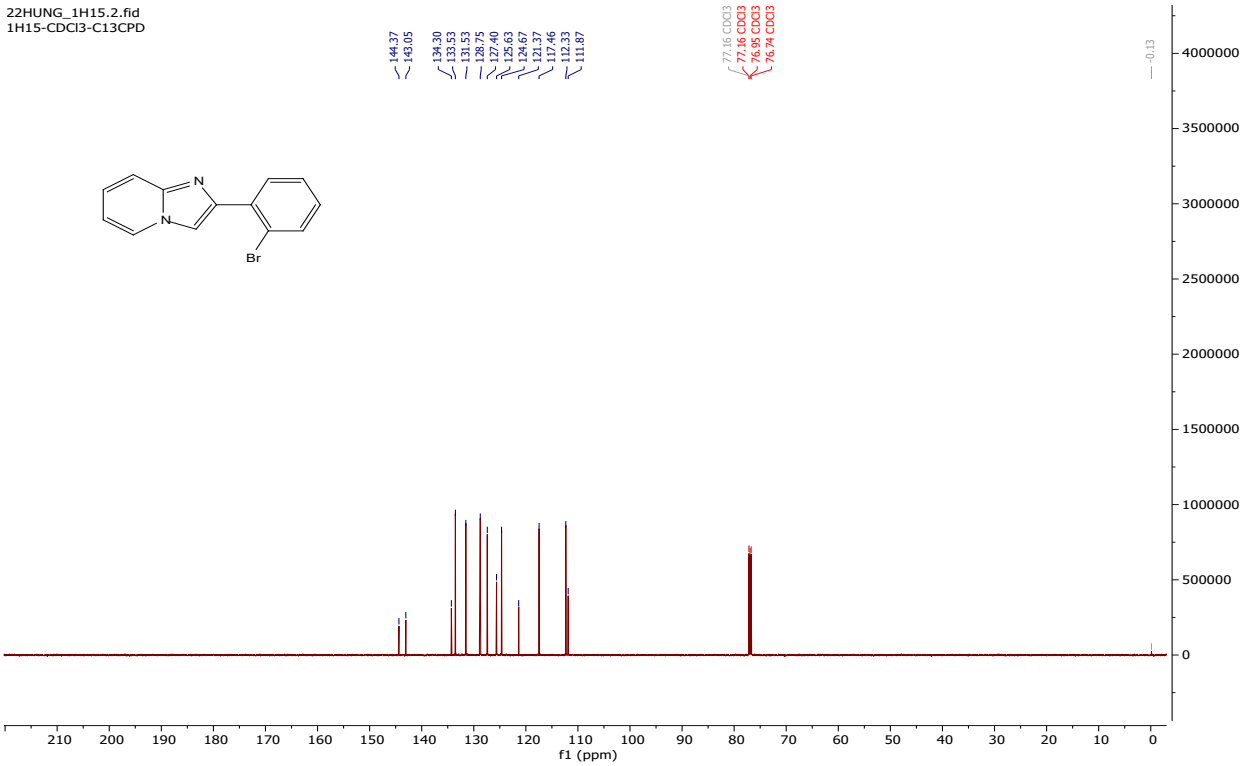
(1:0.8)

Pyrido[2',1':2,3]imidazo[4,5-c]quinoline-6-d **4a$^{\#}$** prepared similarly to compound **4a** mentioned above but using DMSO-d_6 solvent. A mixture of product **4a** and **4a$^{\#}$** was obtained in 68% isolated yield with a ratio of **4a/4a$^{\#}$** = 1:0.8.

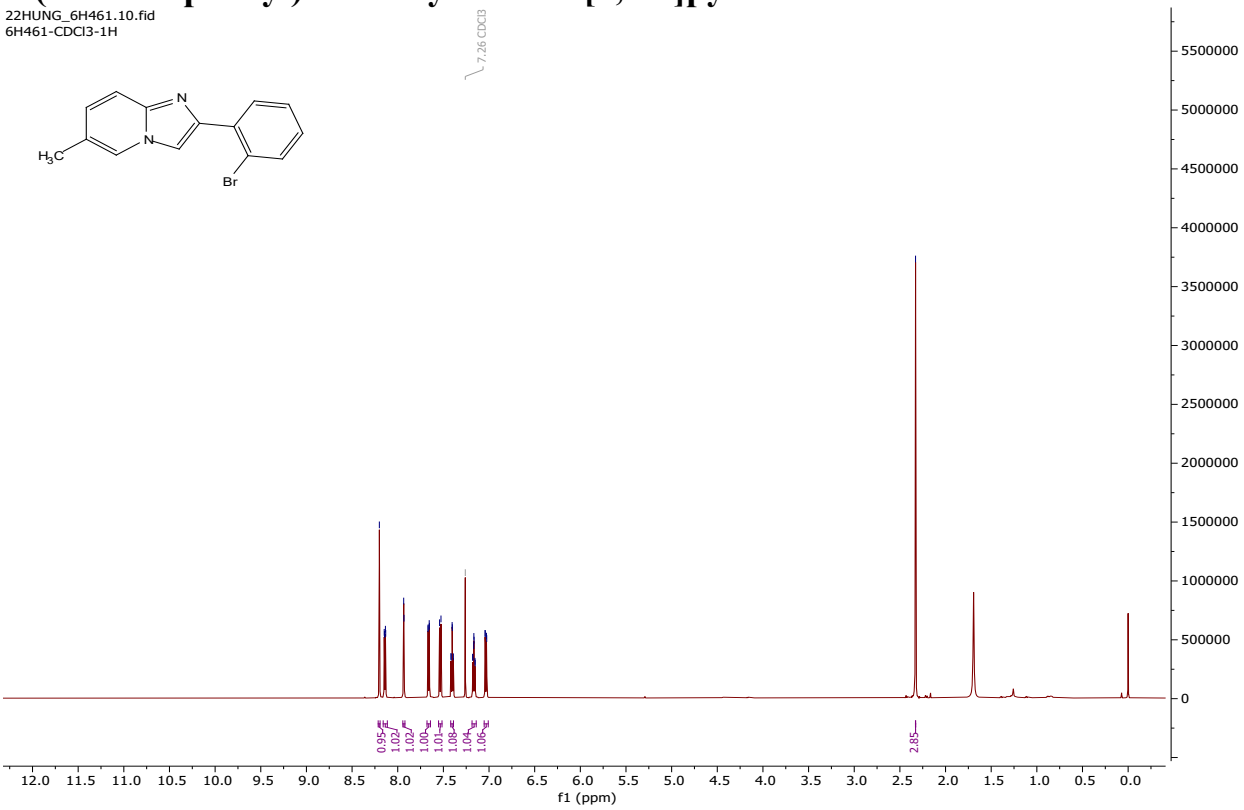
NMR SPECTRA

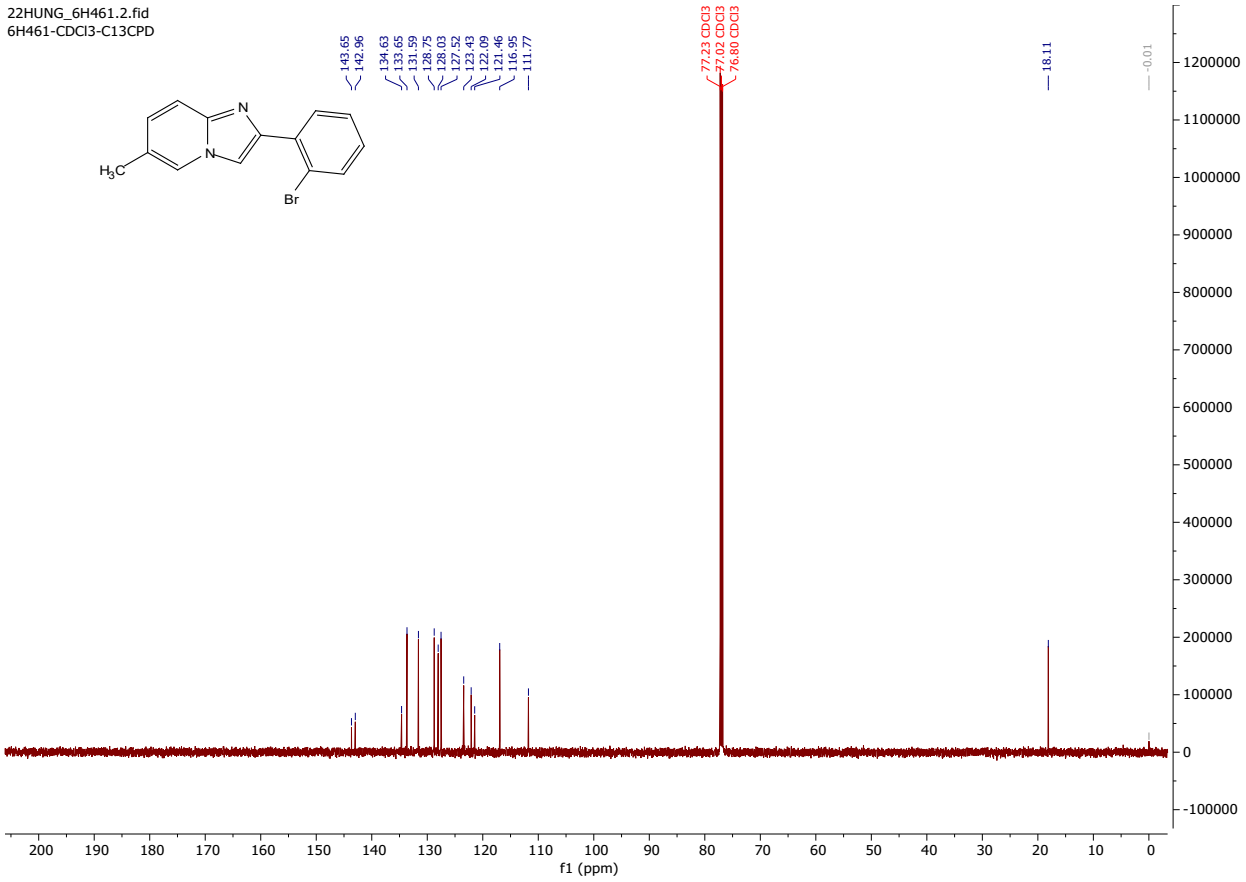
2-(2-bromophenyl)imidazo[1,2-a]pyridine 2a



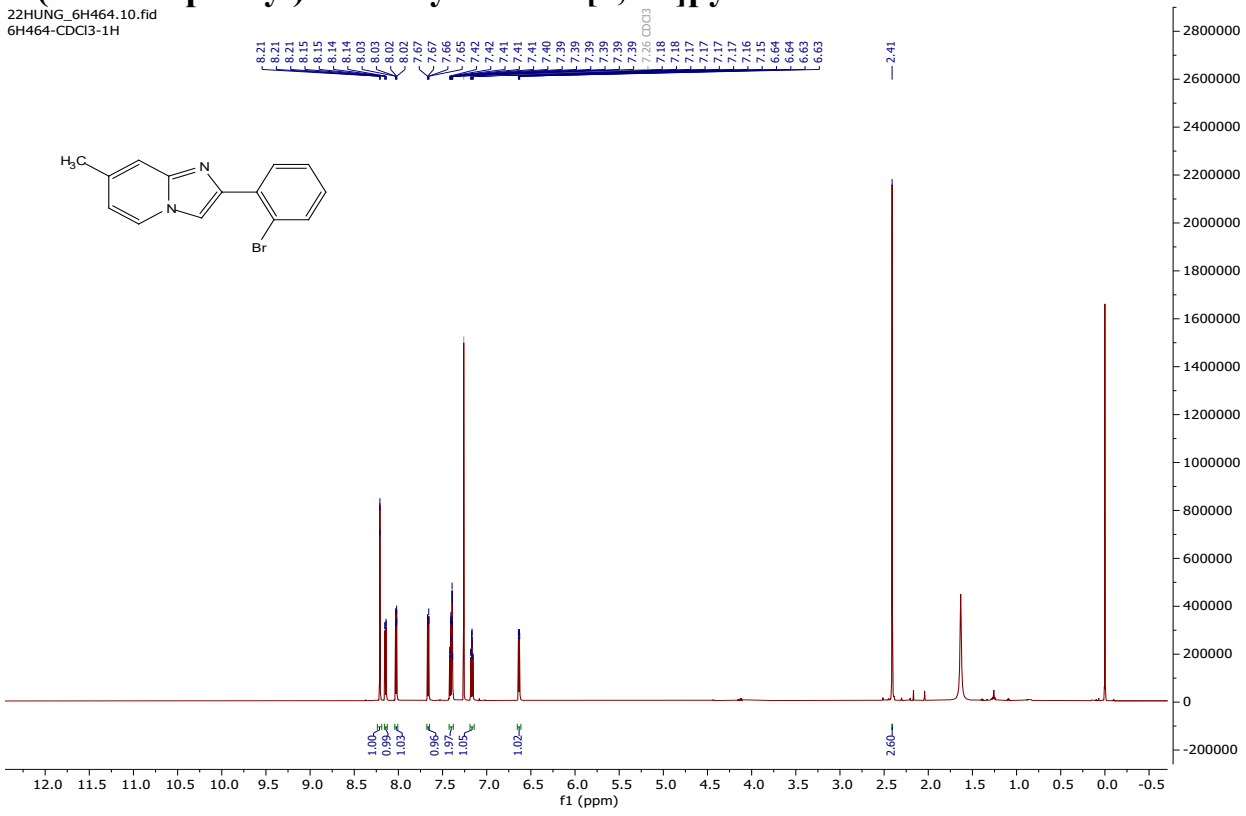


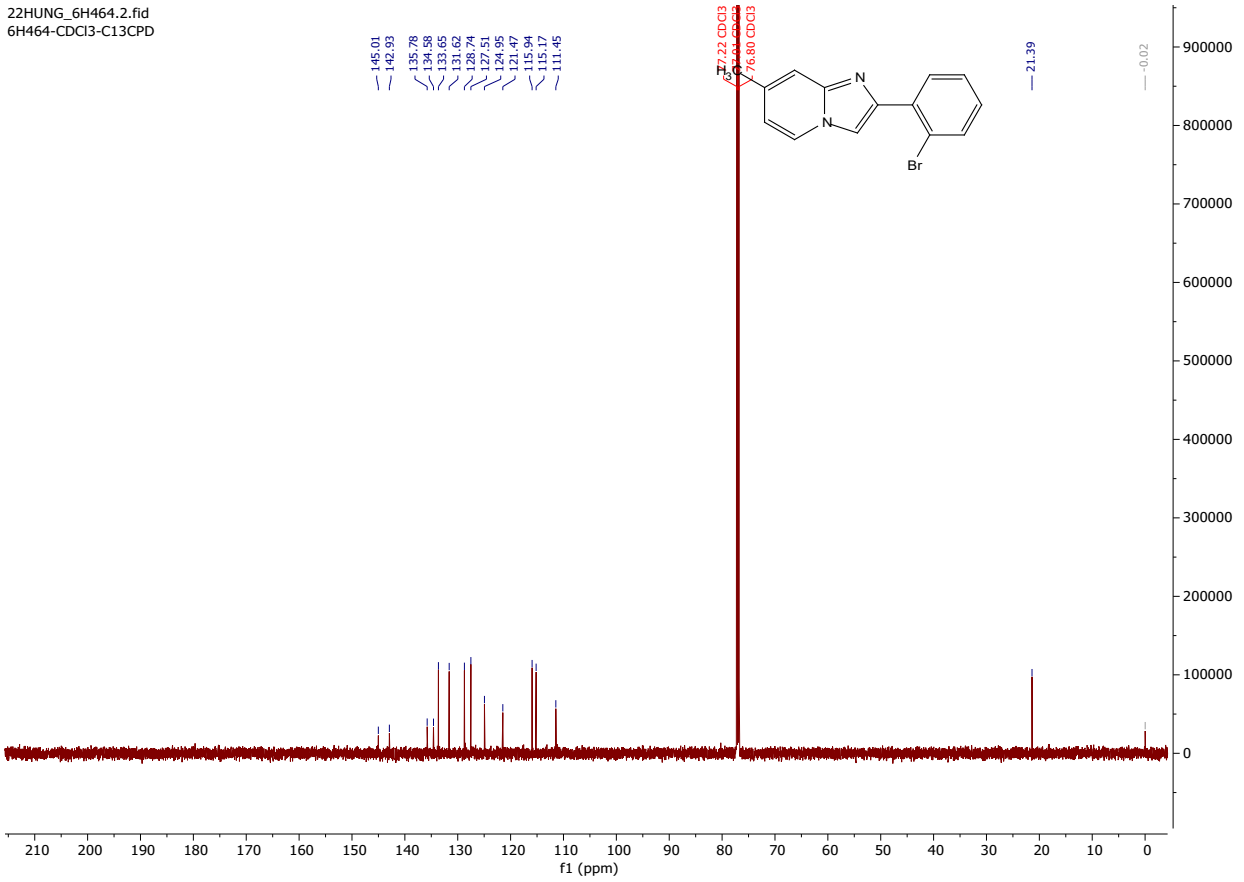
2-(2-bromophenyl)-6-methylimidazo[1,2-a]pyridine 2b



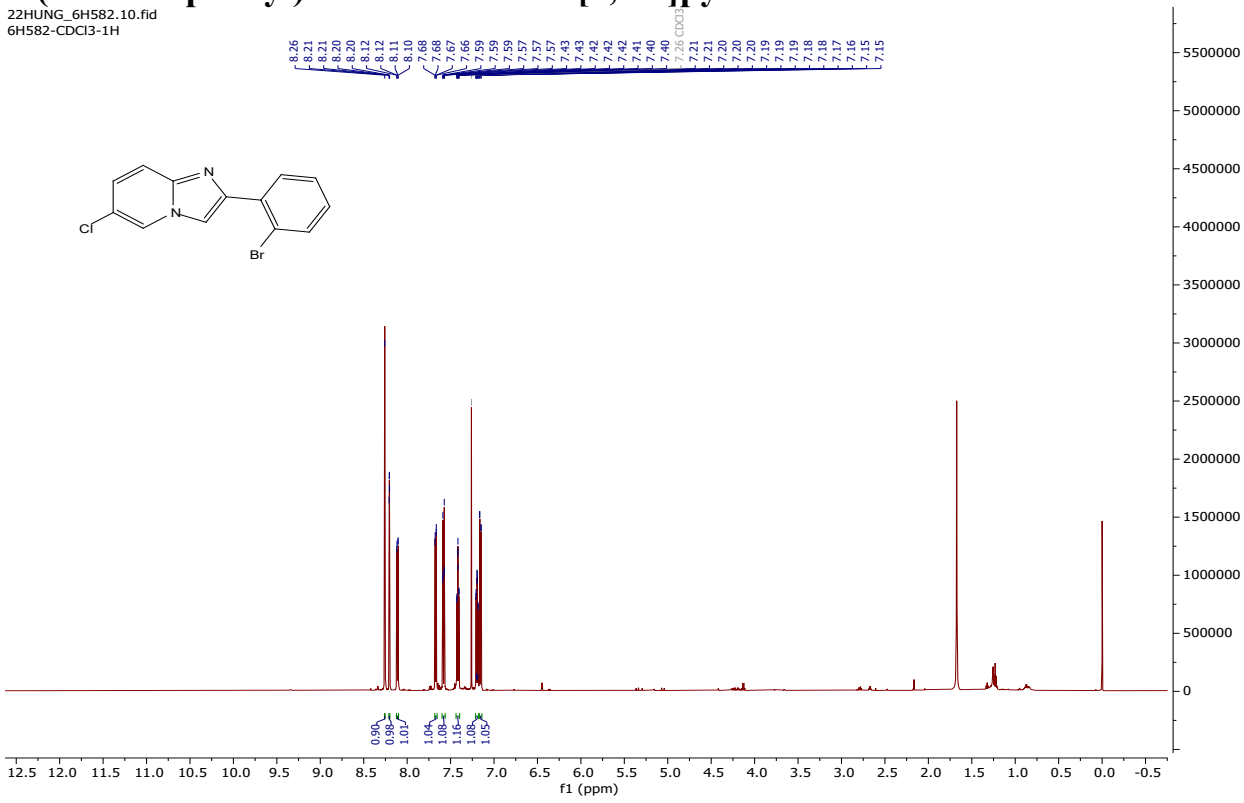


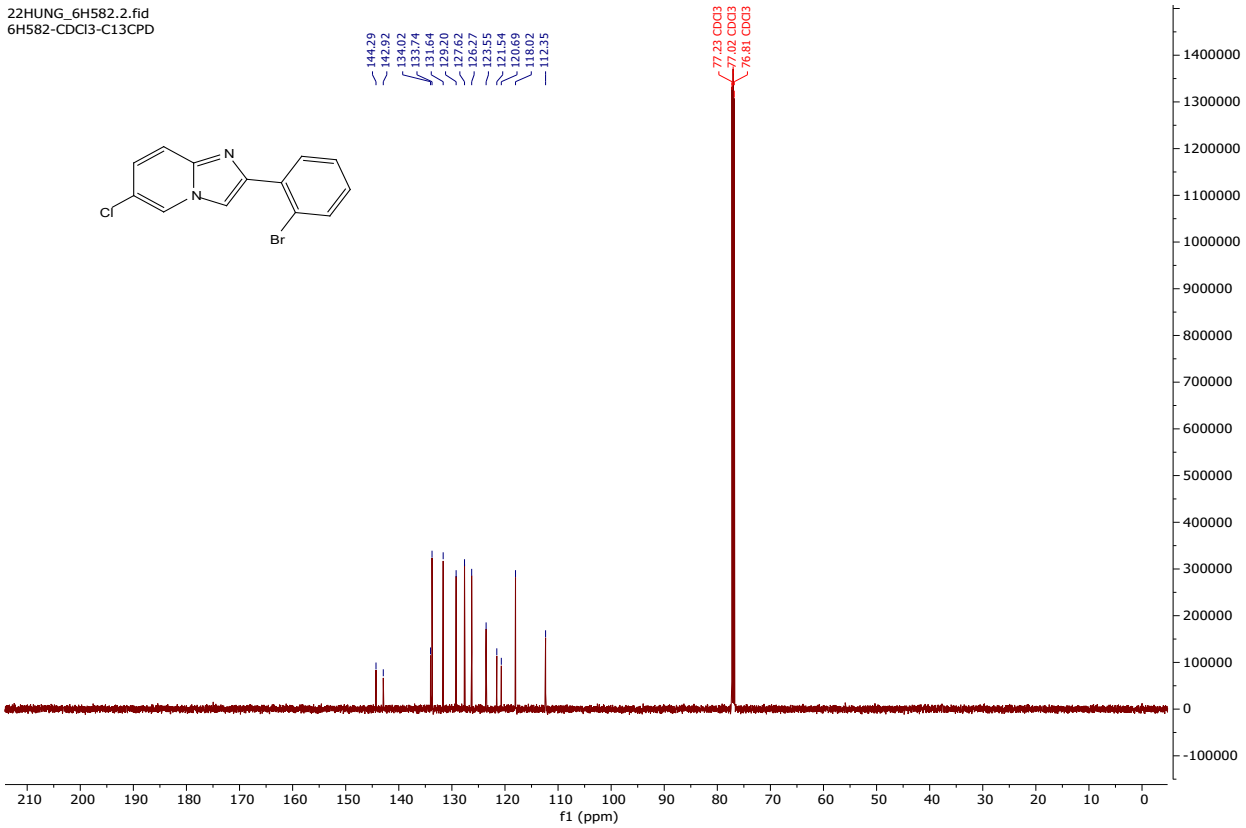
2-(2-bromophenyl)-7-methylimidazo[1,2-a]pyridine 2c



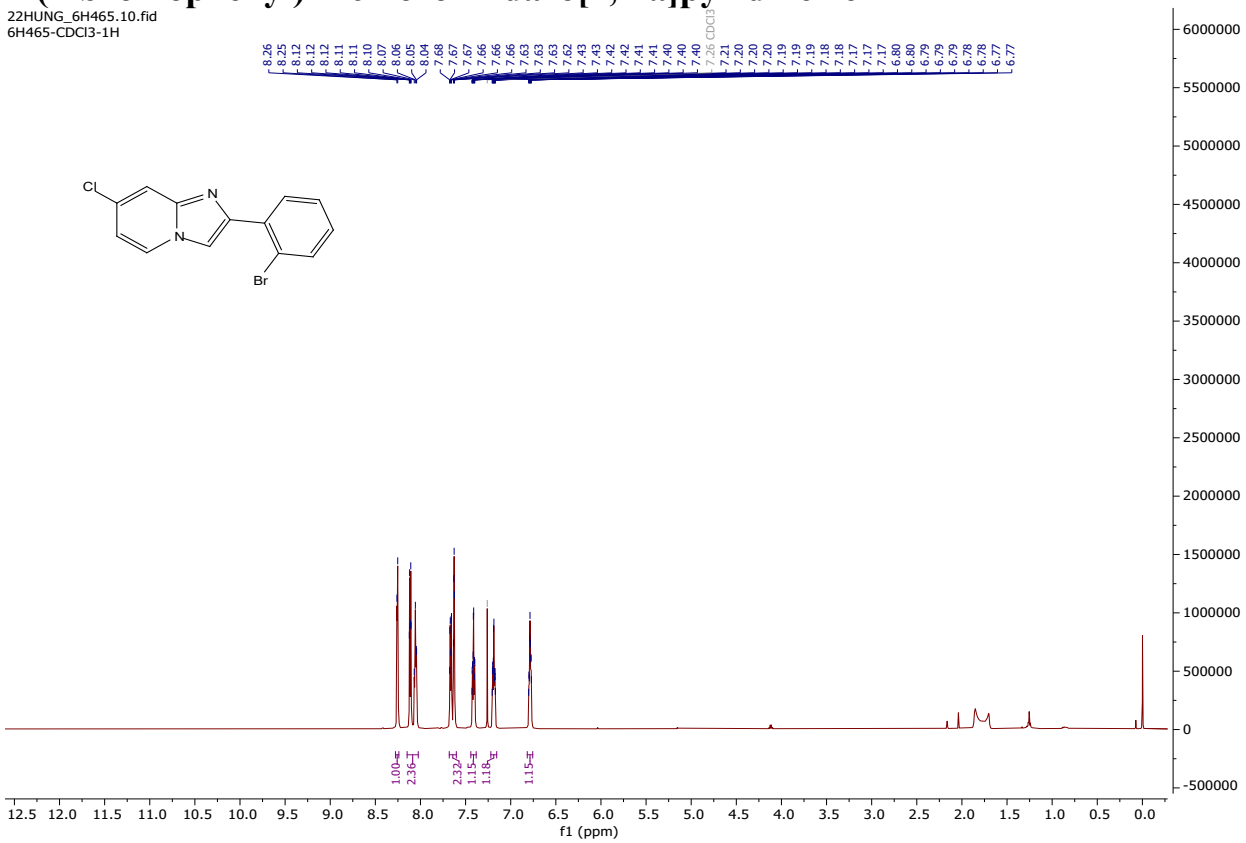


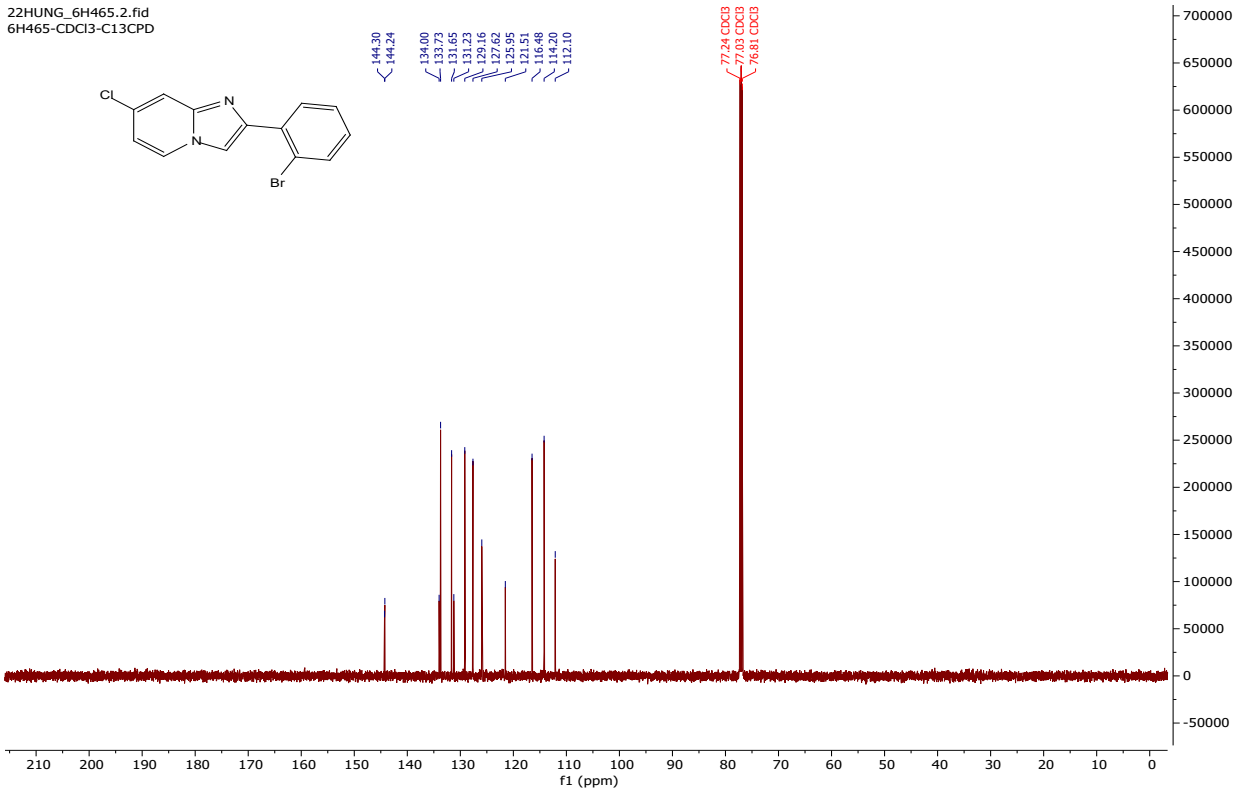
2-(2-bromophenyl)-6-chloroimidazo[1,2-a]pyridine 2d



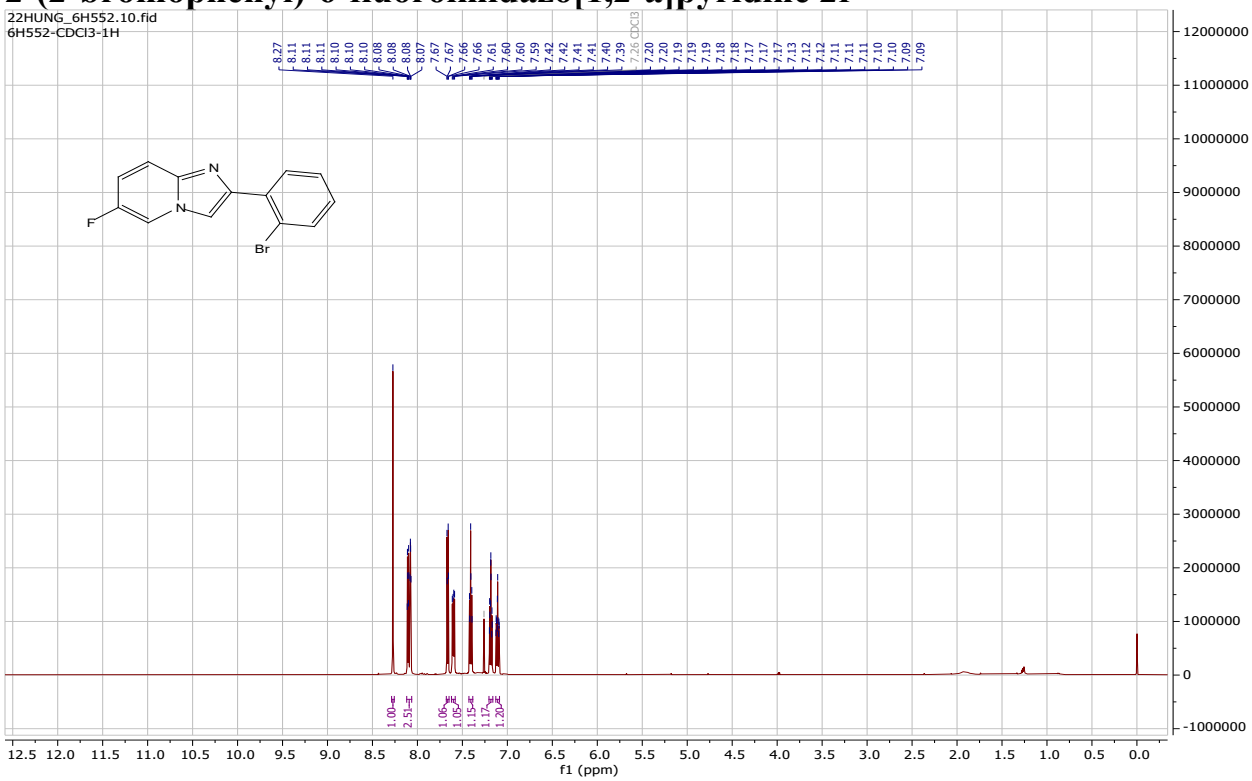


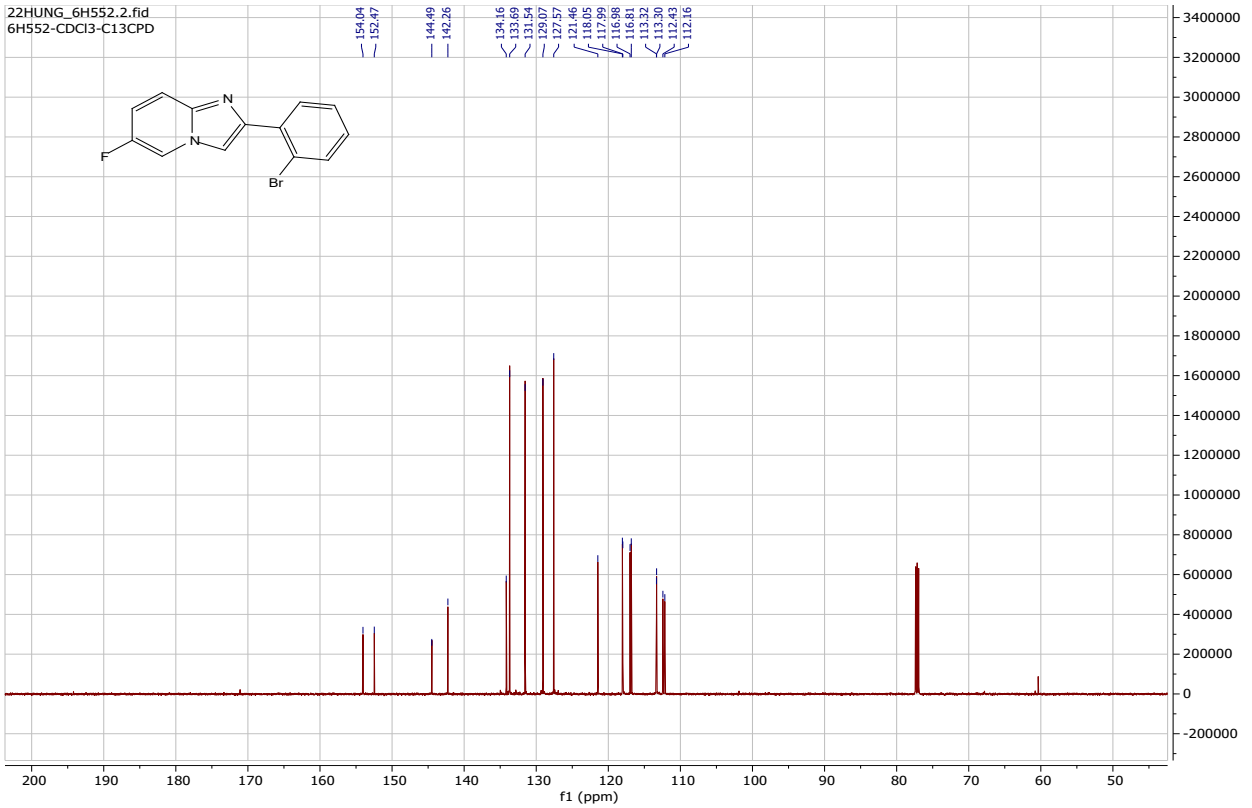
2-(2-bromophenyl)-7-chloroimidazo[1,2-a]pyridine 2e



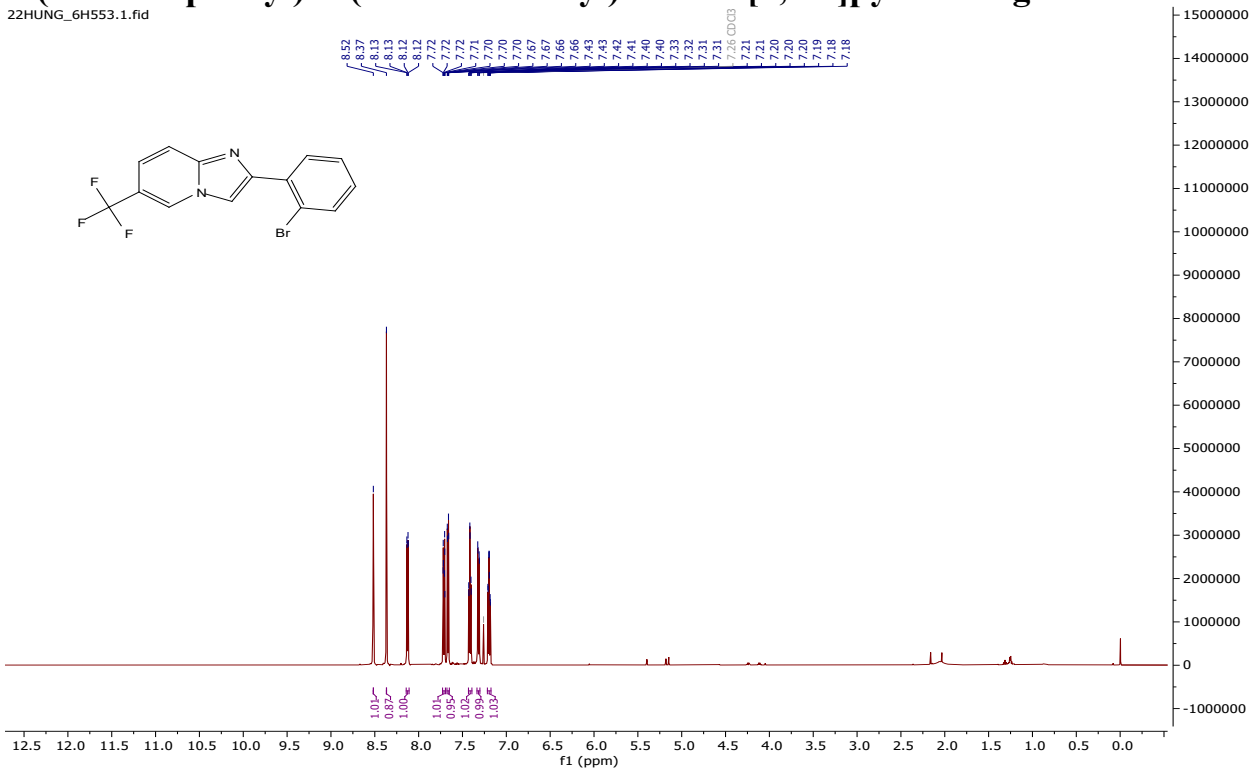


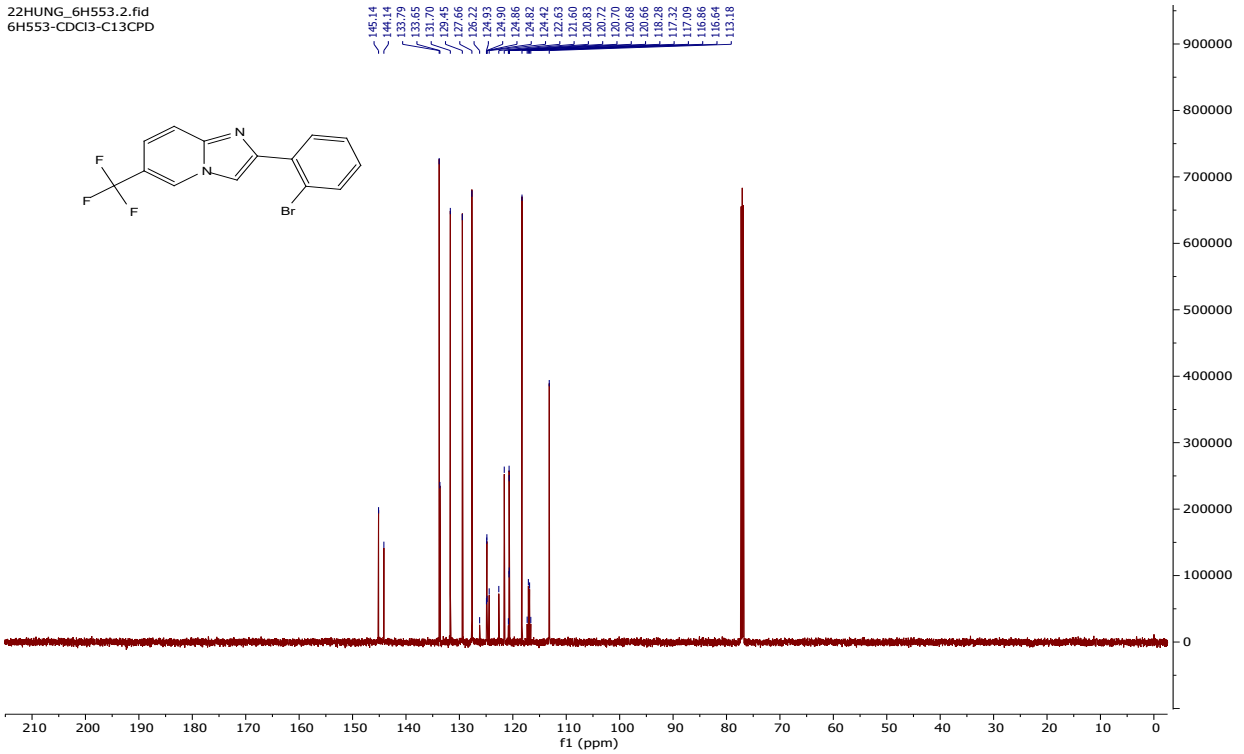
2-(2-bromophenyl)-6-fluoroimidazo[1,2-a]pyridine 2f



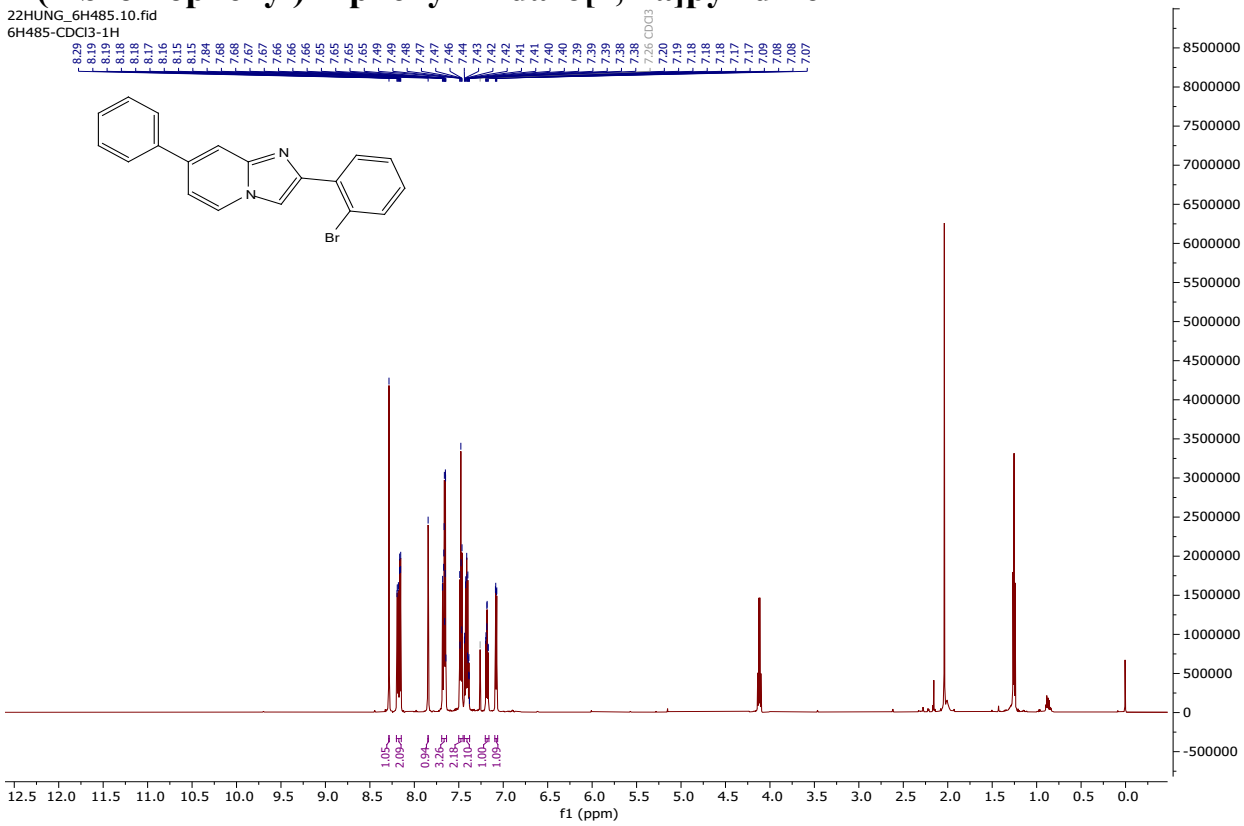


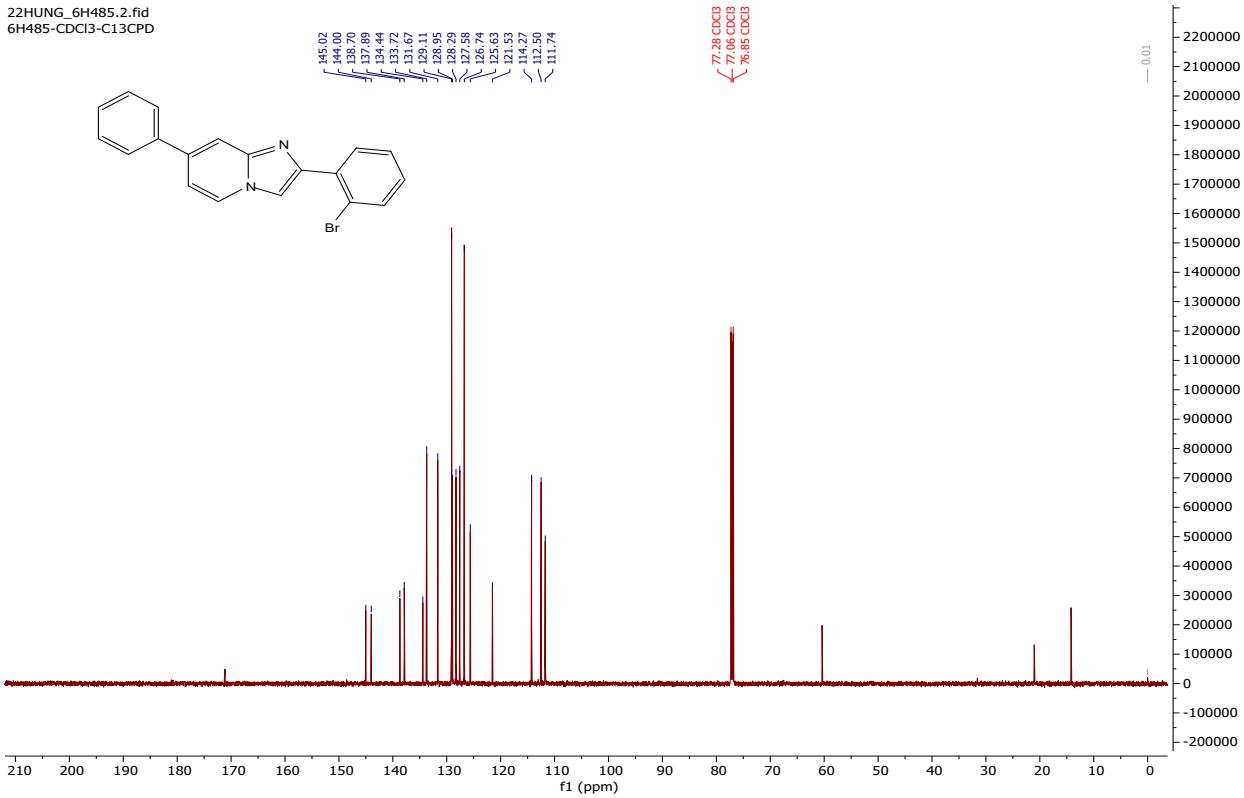
2-(2-bromophenyl)-6-(trifluoromethyl)imidazo[1,2-a]pyridine 2g



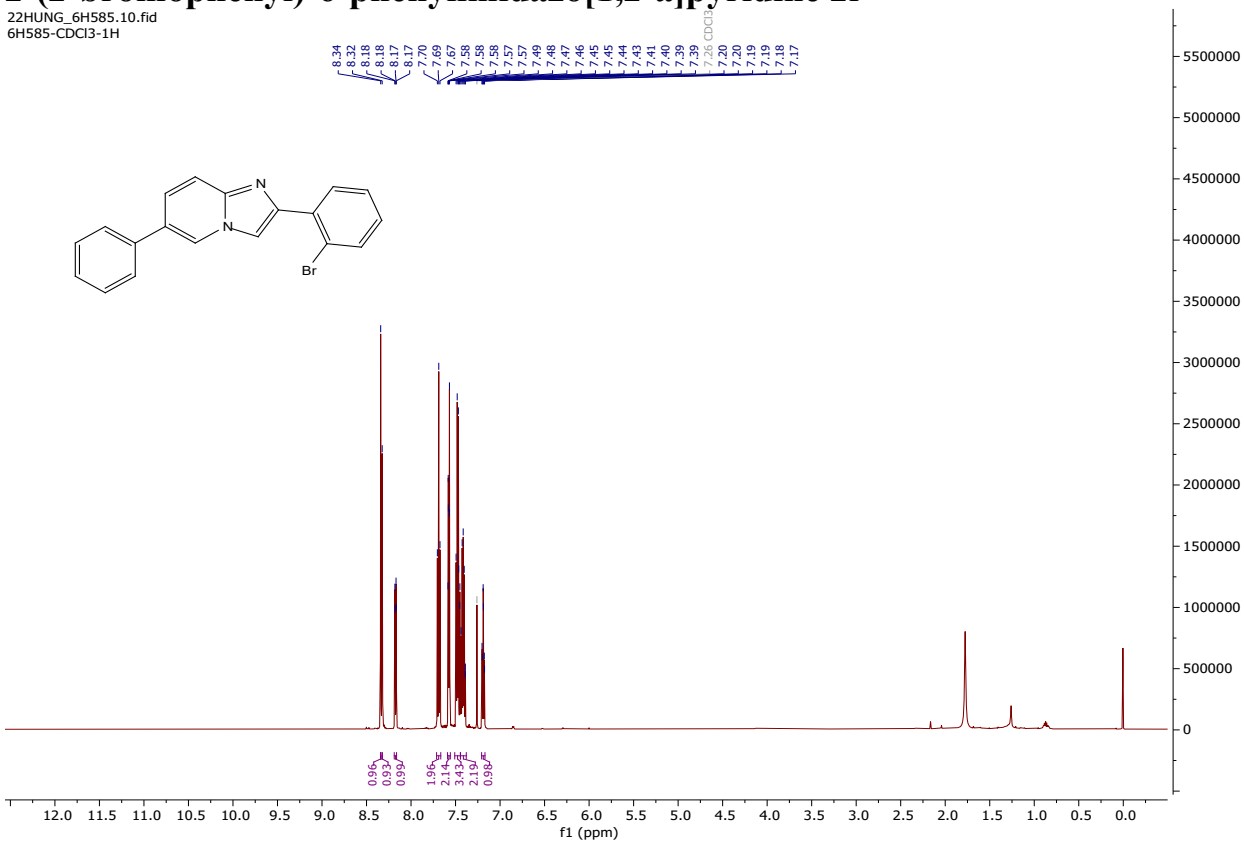


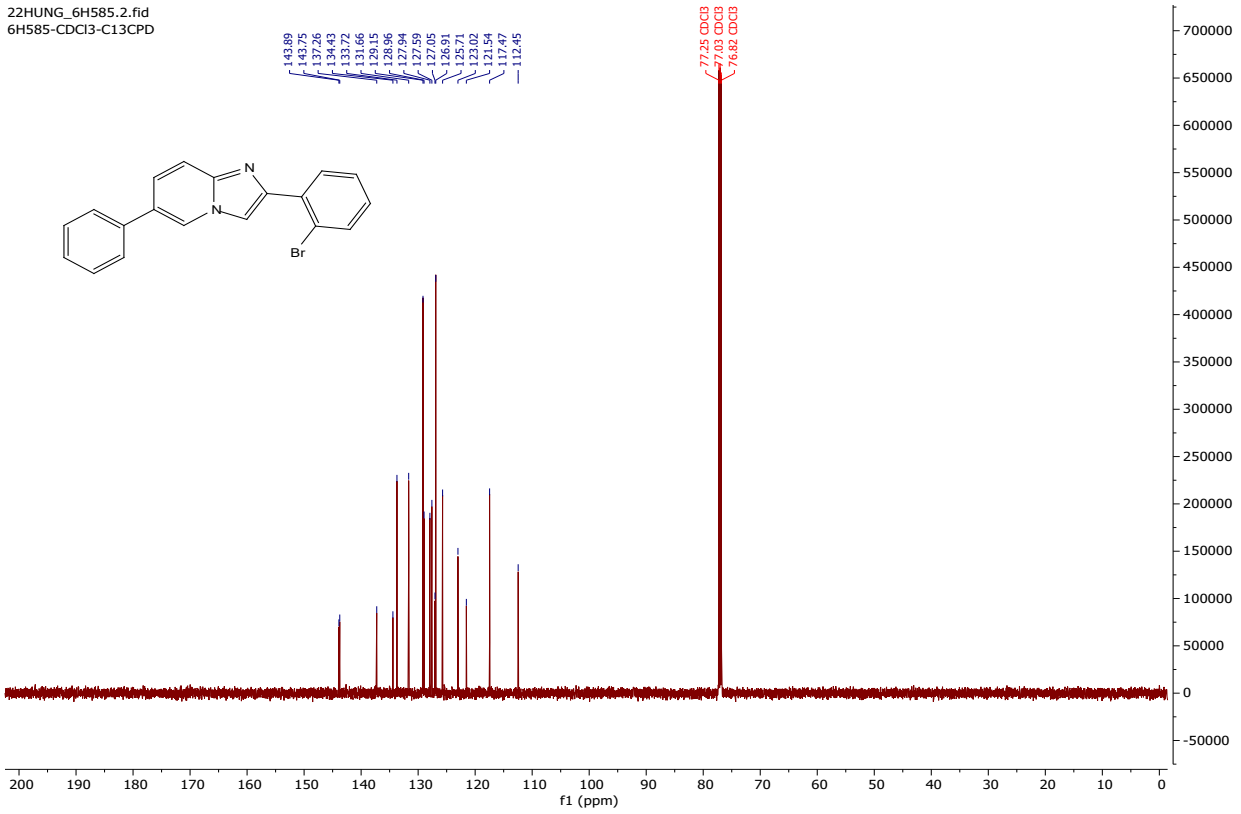
2-(2-bromophenyl)-7-phenylimidazo[1,2-a]pyridine 2h



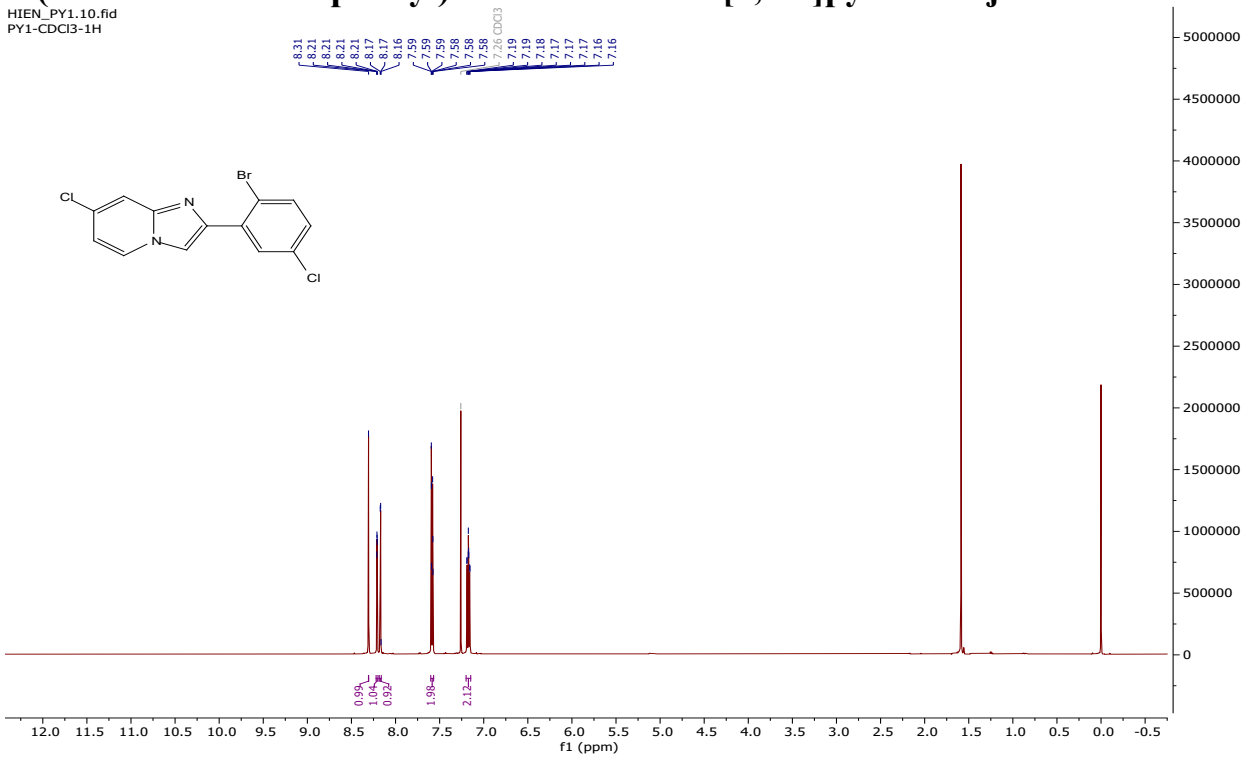


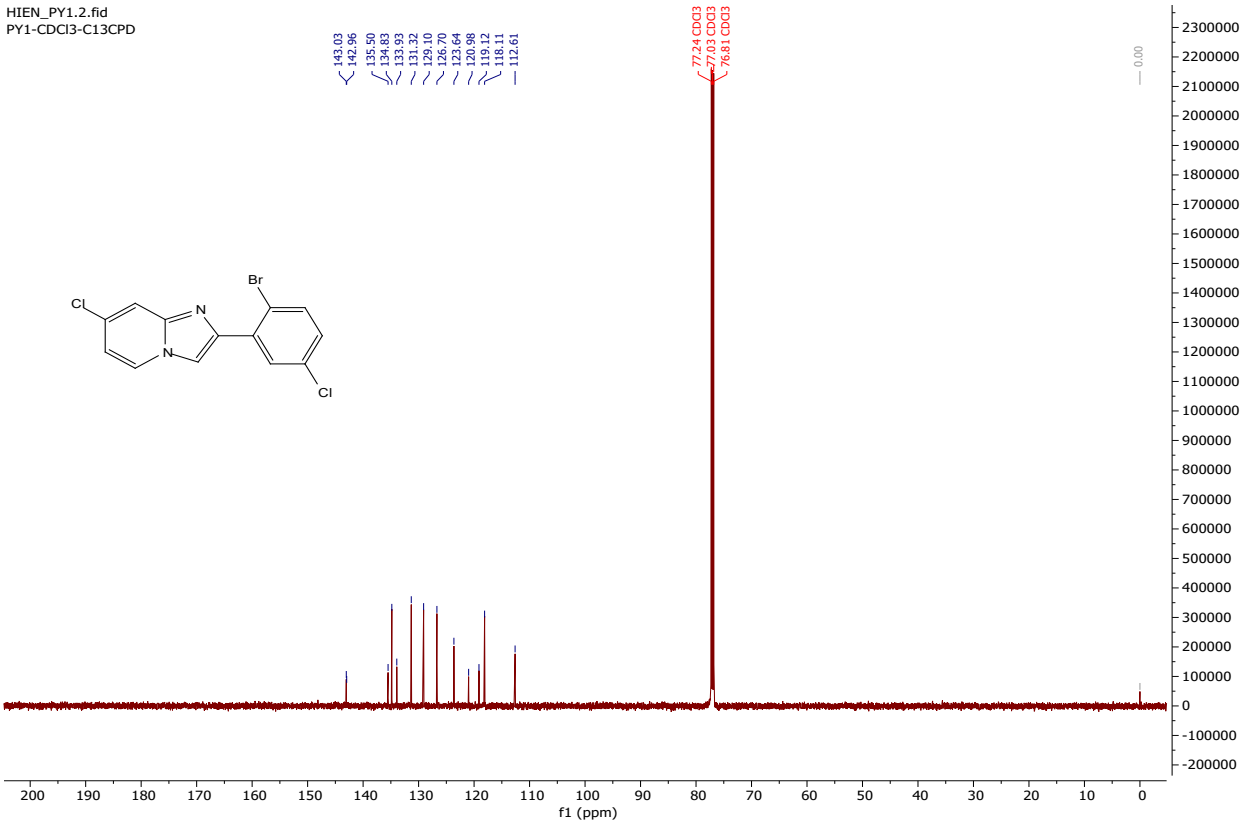
2-(2-bromophenyl)-6-phenylimidazo[1,2-a]pyridine 2i



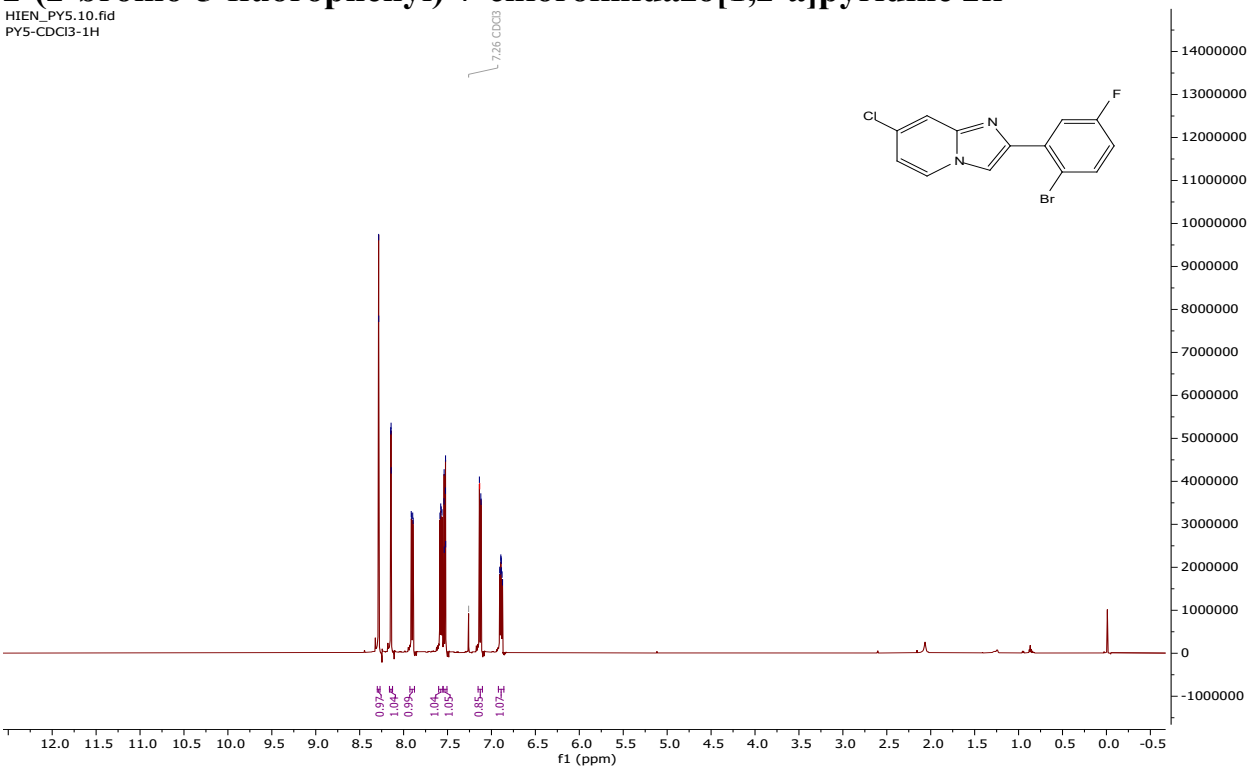


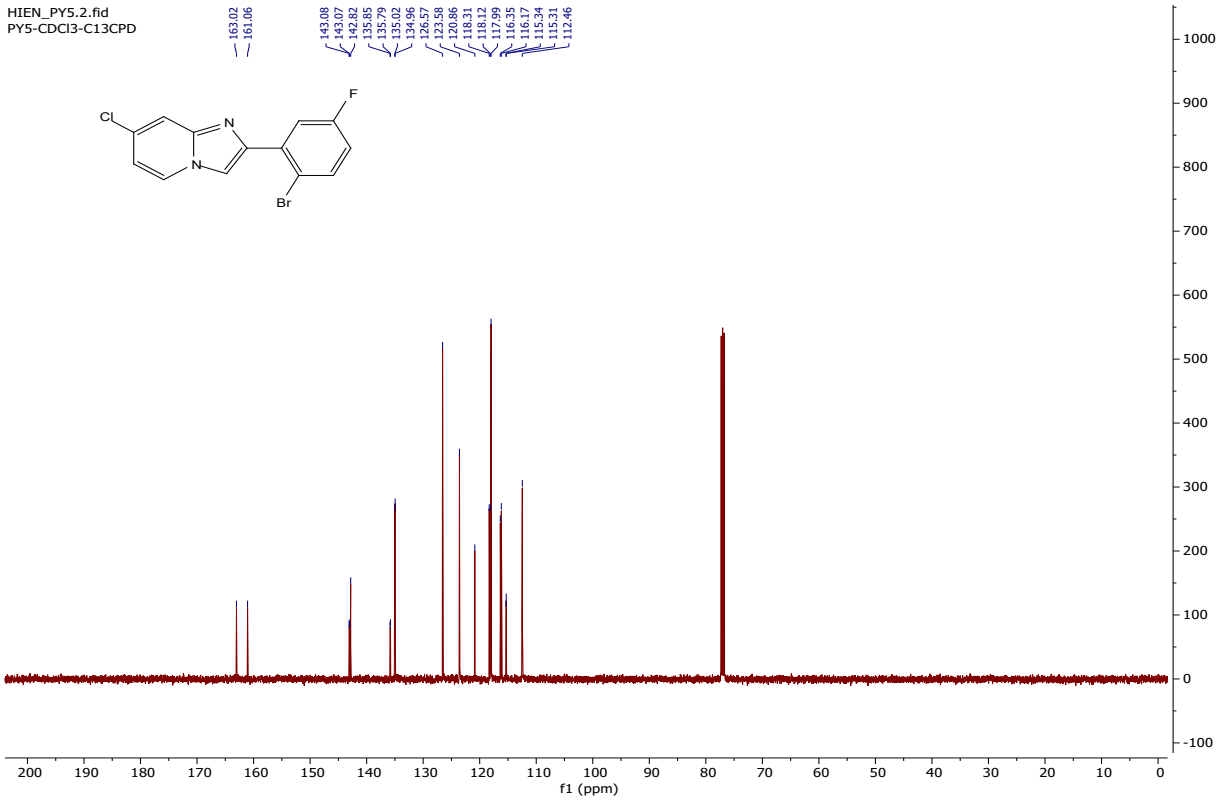
2-(2-bromo-5-chlorophenyl)-7-chloroimidazo[1,2-a]pyridine 2j



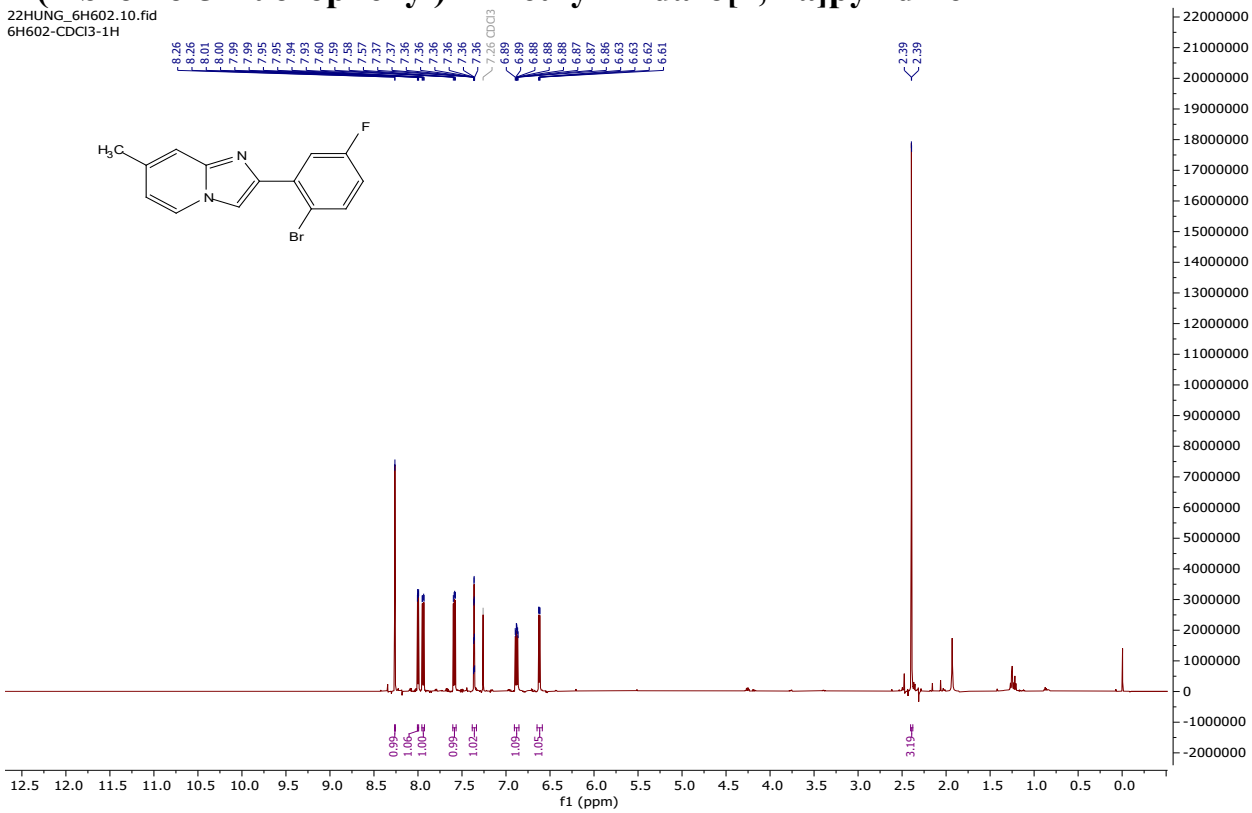


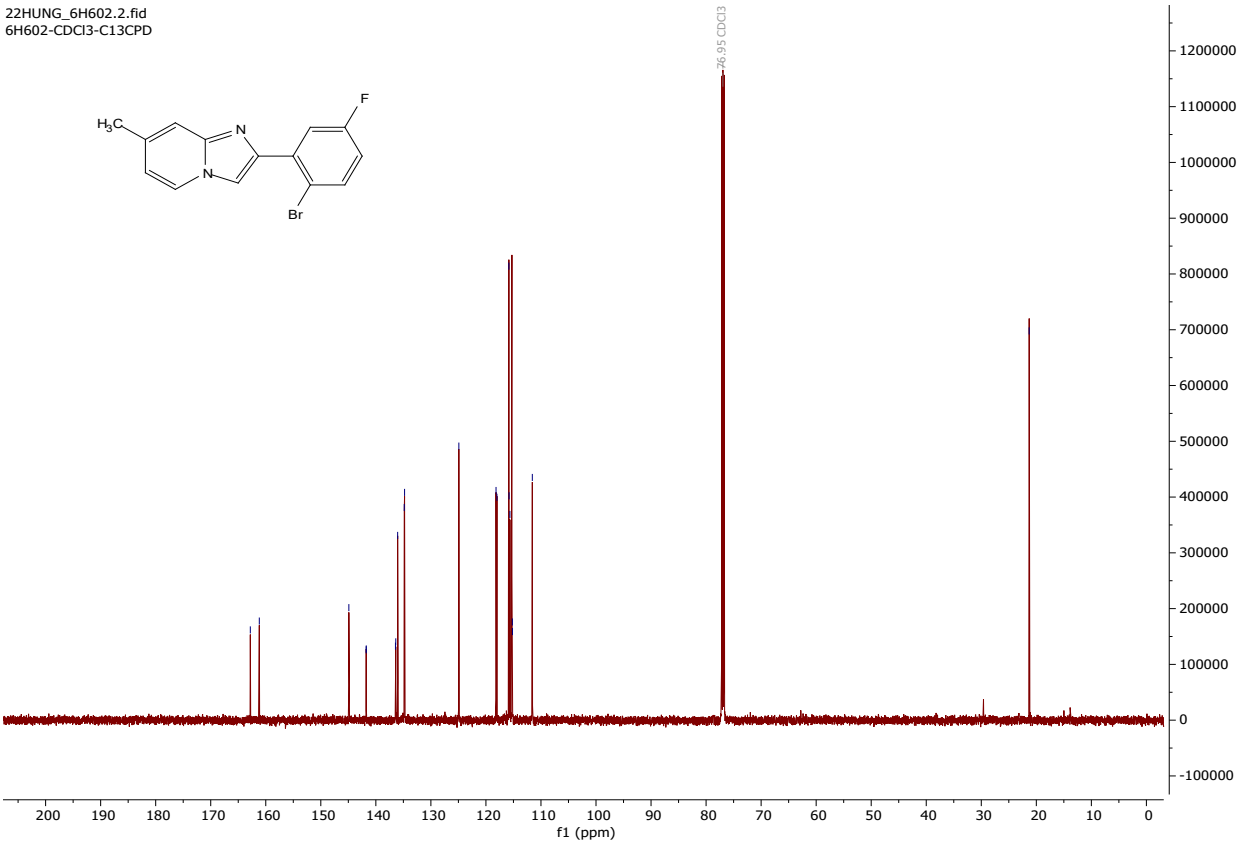
2-(2-bromo-5-fluorophenyl)-7-chloroimidazo[1,2-a]pyridine 2k



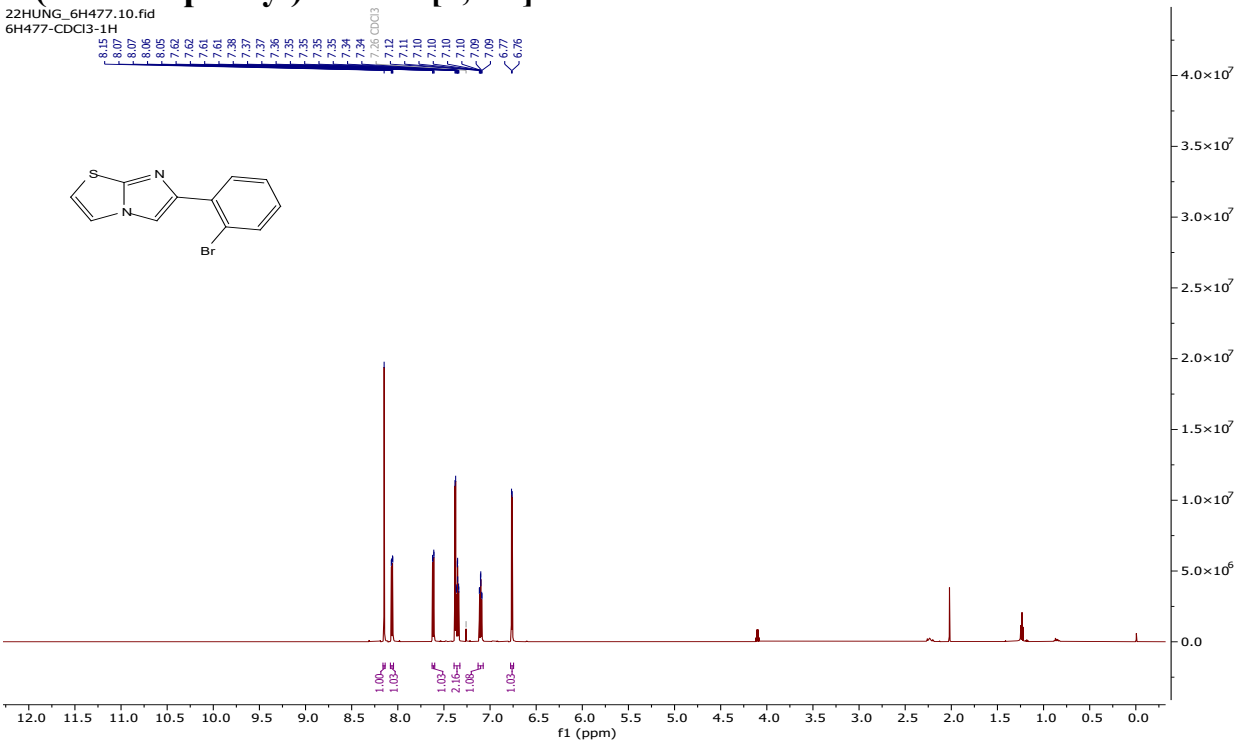


2-(2-bromo-5-fluorophenyl)-7-methylimidazo[1,2-a]pyridine 2l

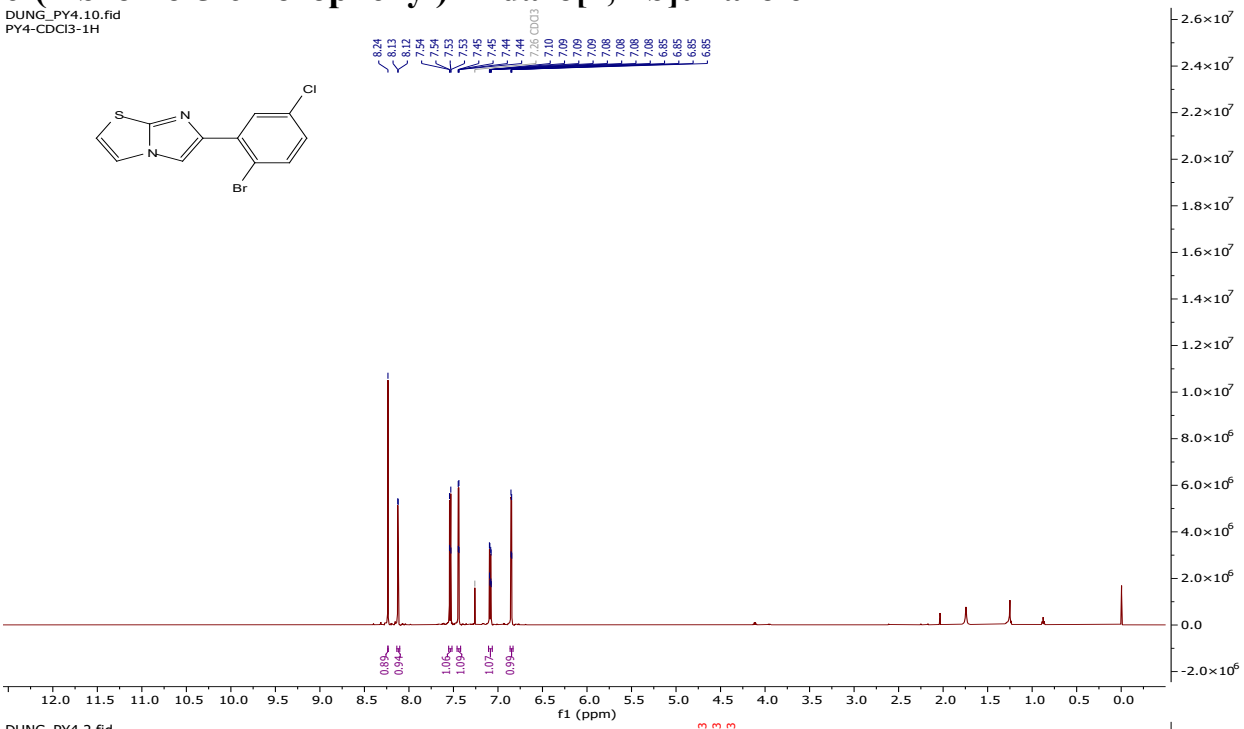


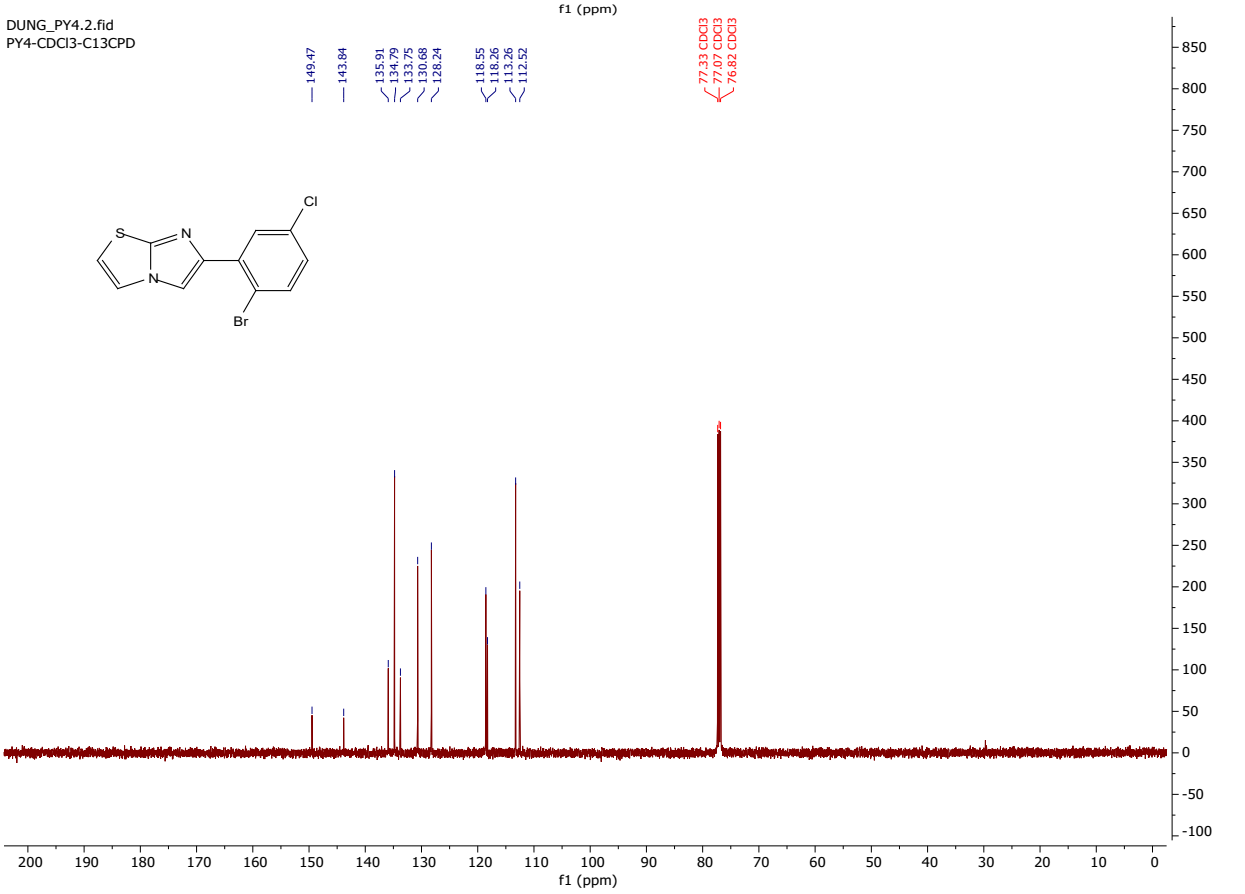


6-(2-bromophenyl)imidazo[2,1-b]thiazole 2m

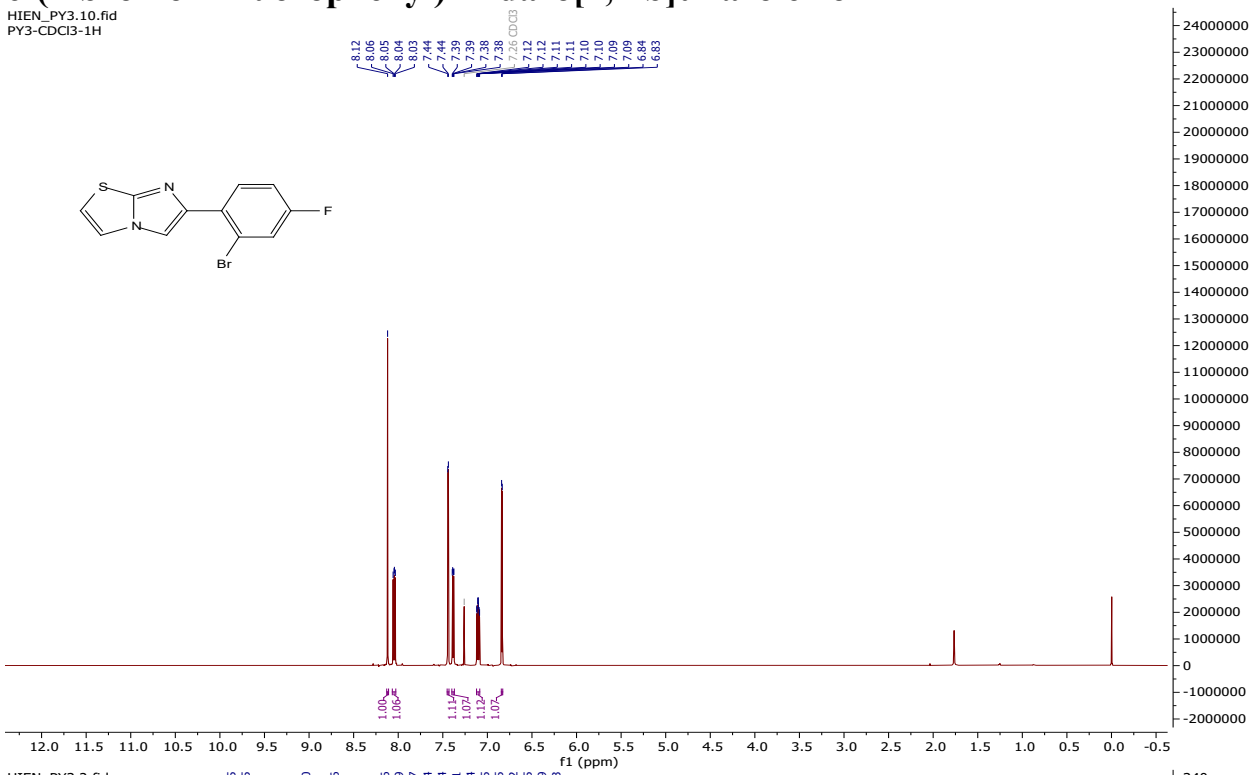


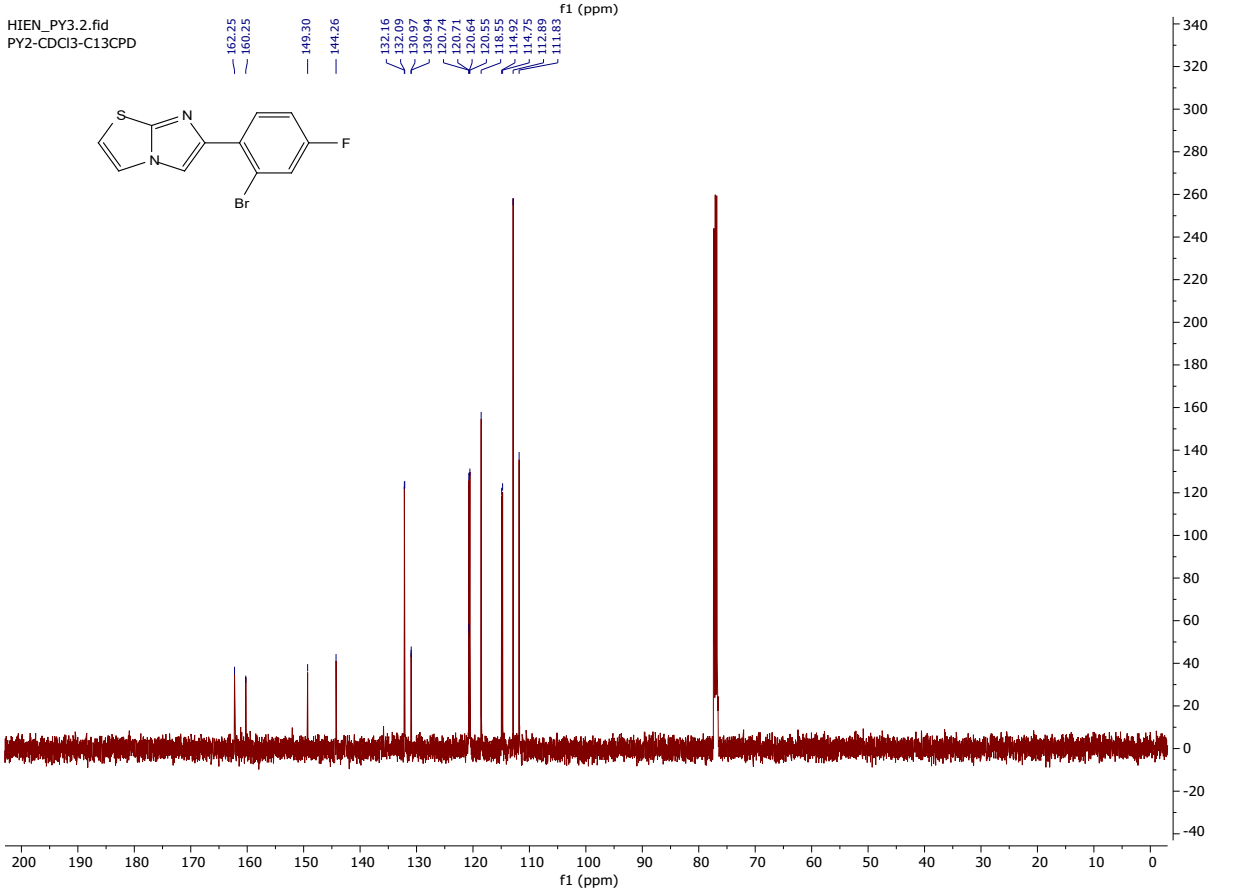
6-(2-bromo-5-chlorophenyl)imidazo[2,1-b]thiazole 2n



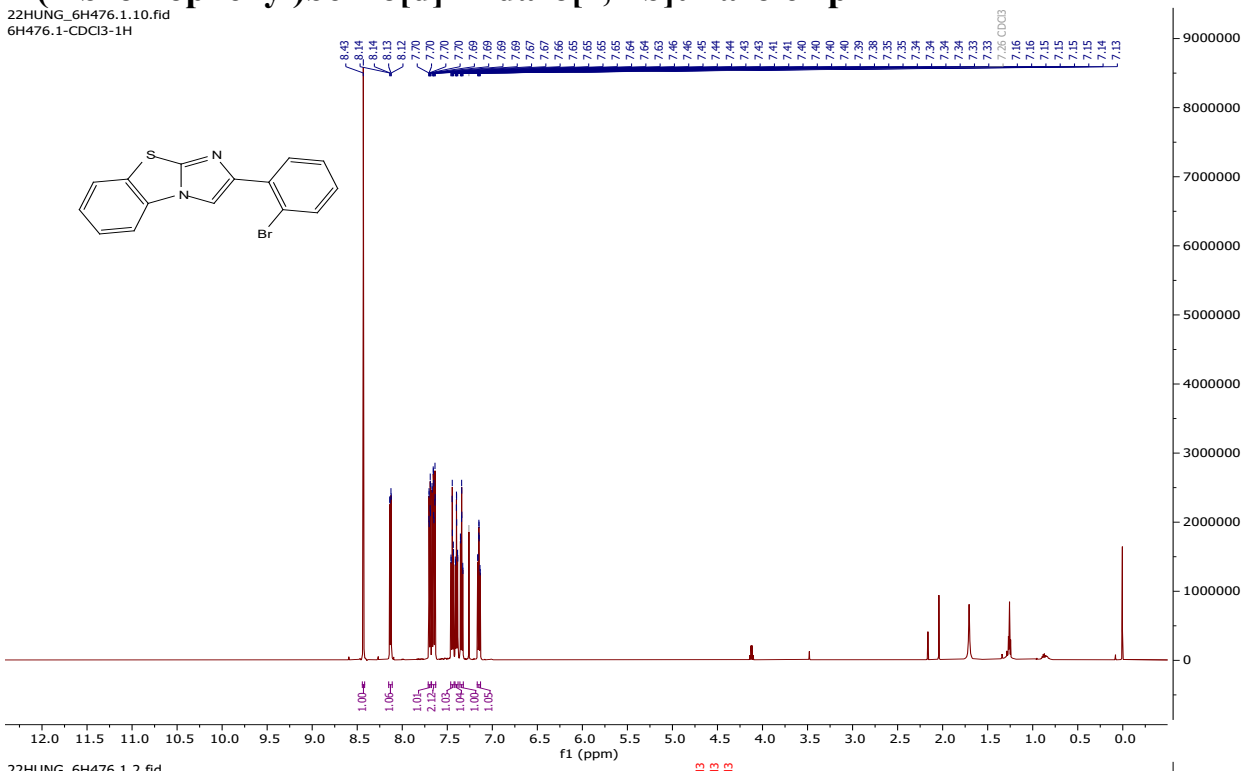


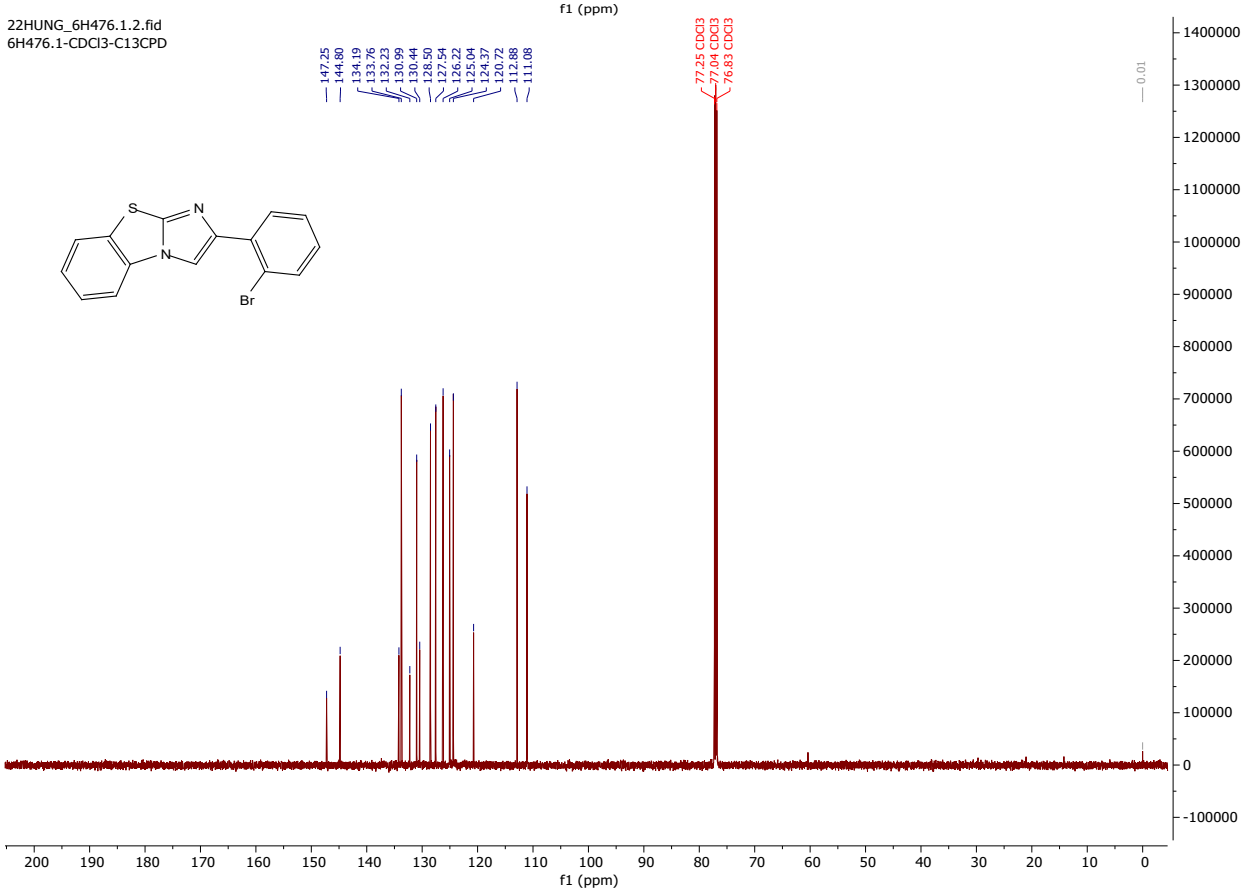
6-(2-bromo-4-fluorophenyl)imidazo[2,1-b]thiazole 2o



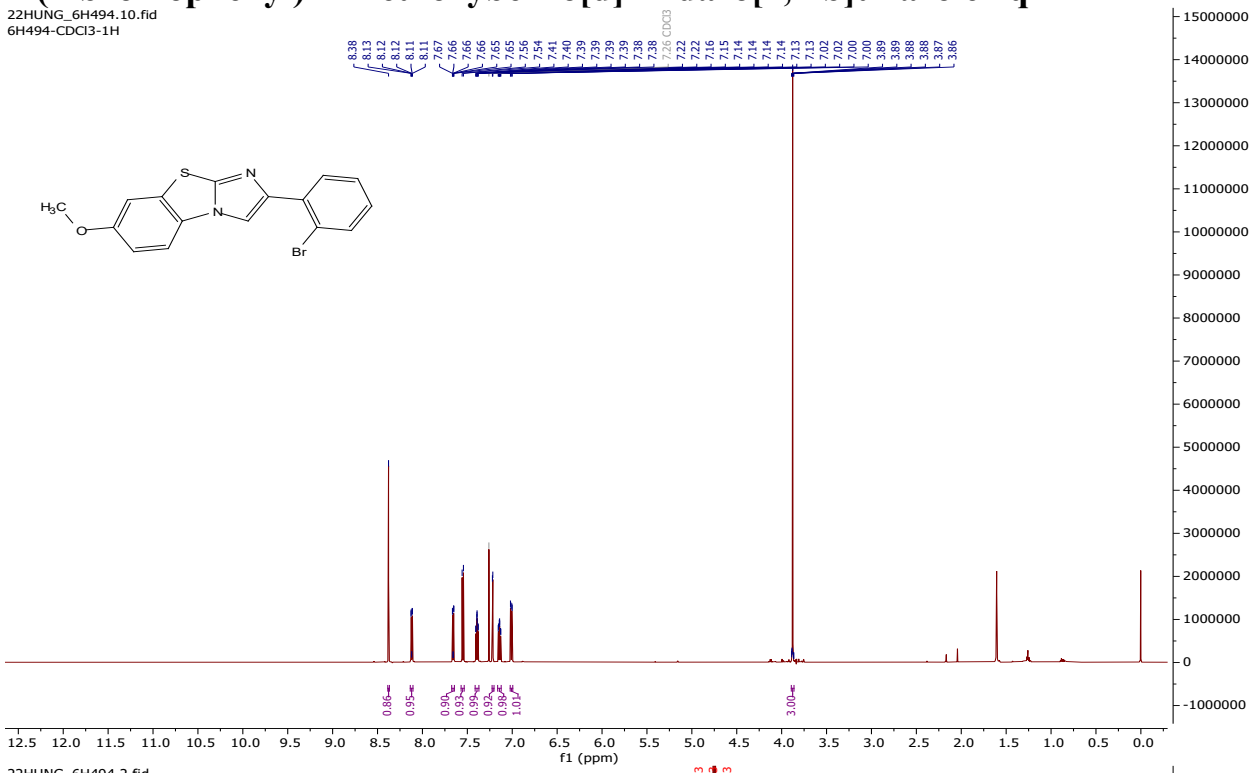


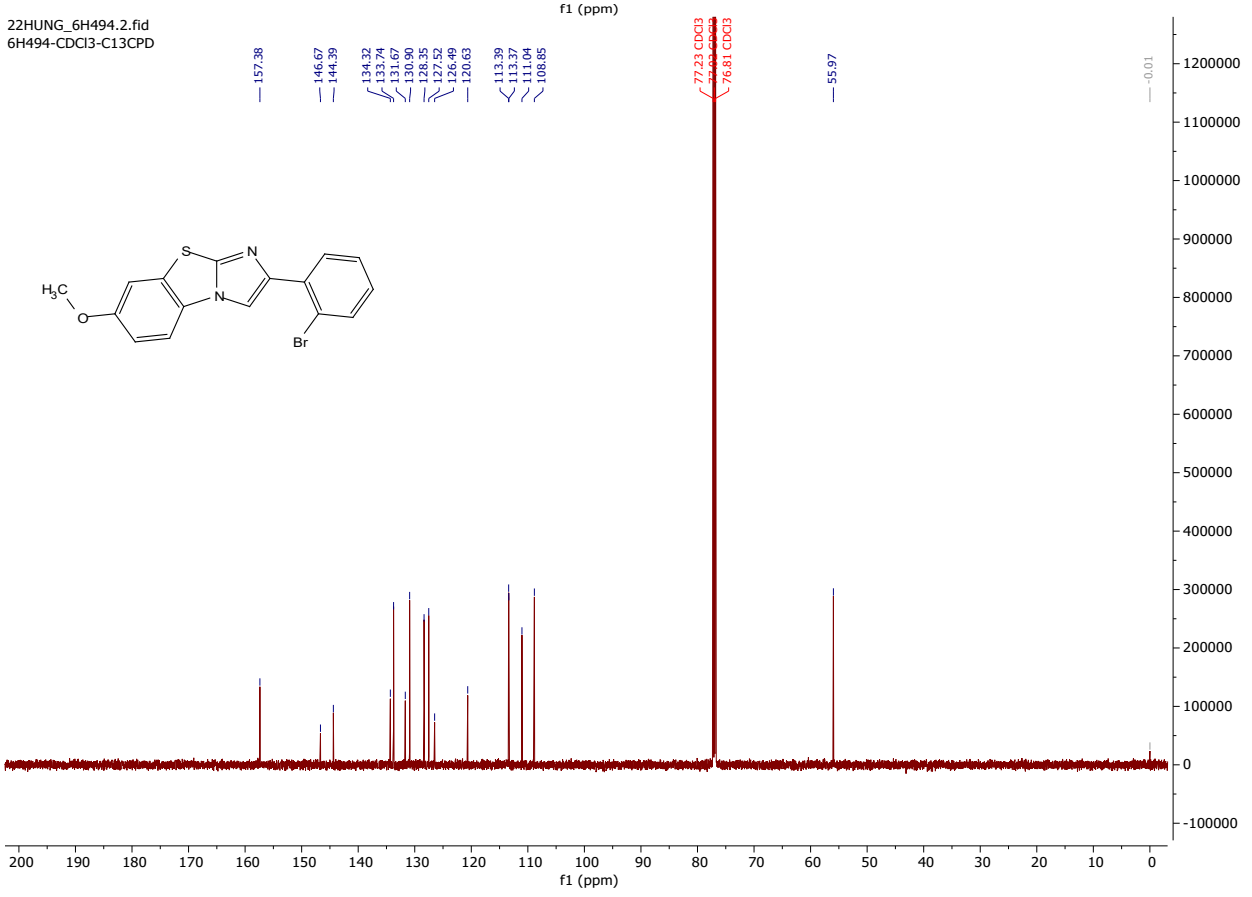
2-(2-bromophenyl)benzo[d]imidazo[2,1-b]thiazole 2p



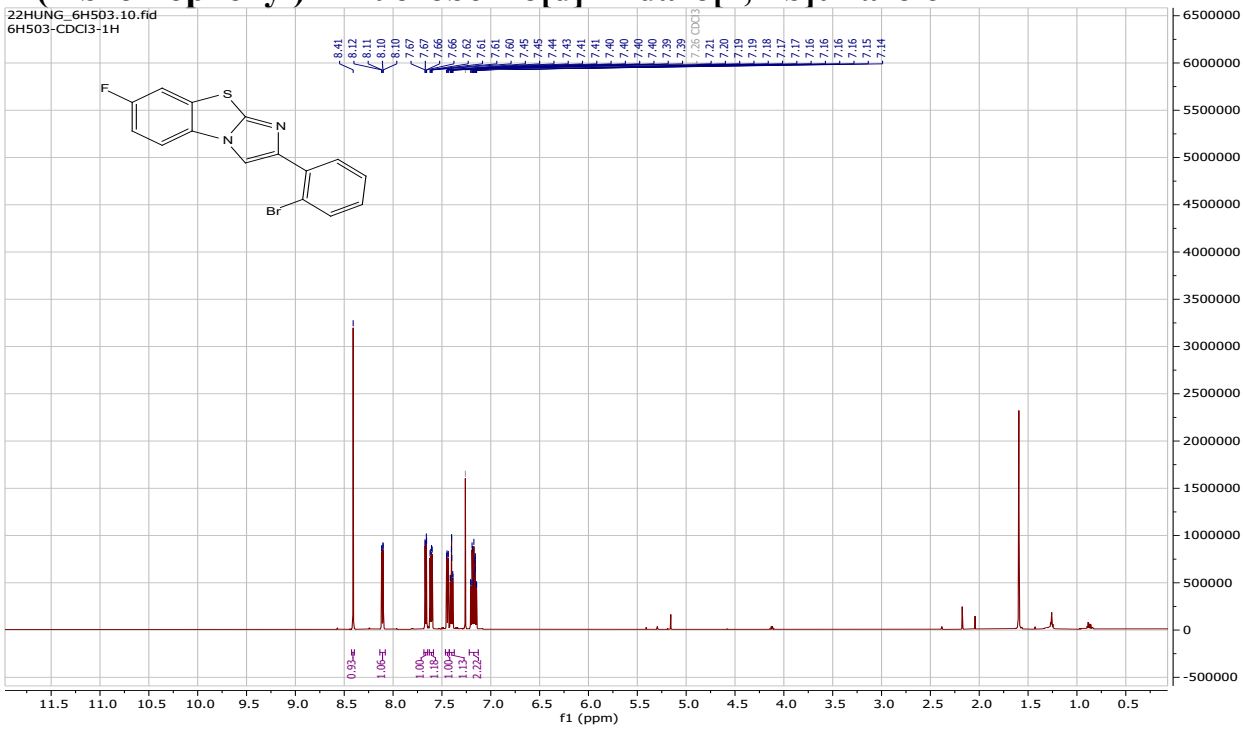


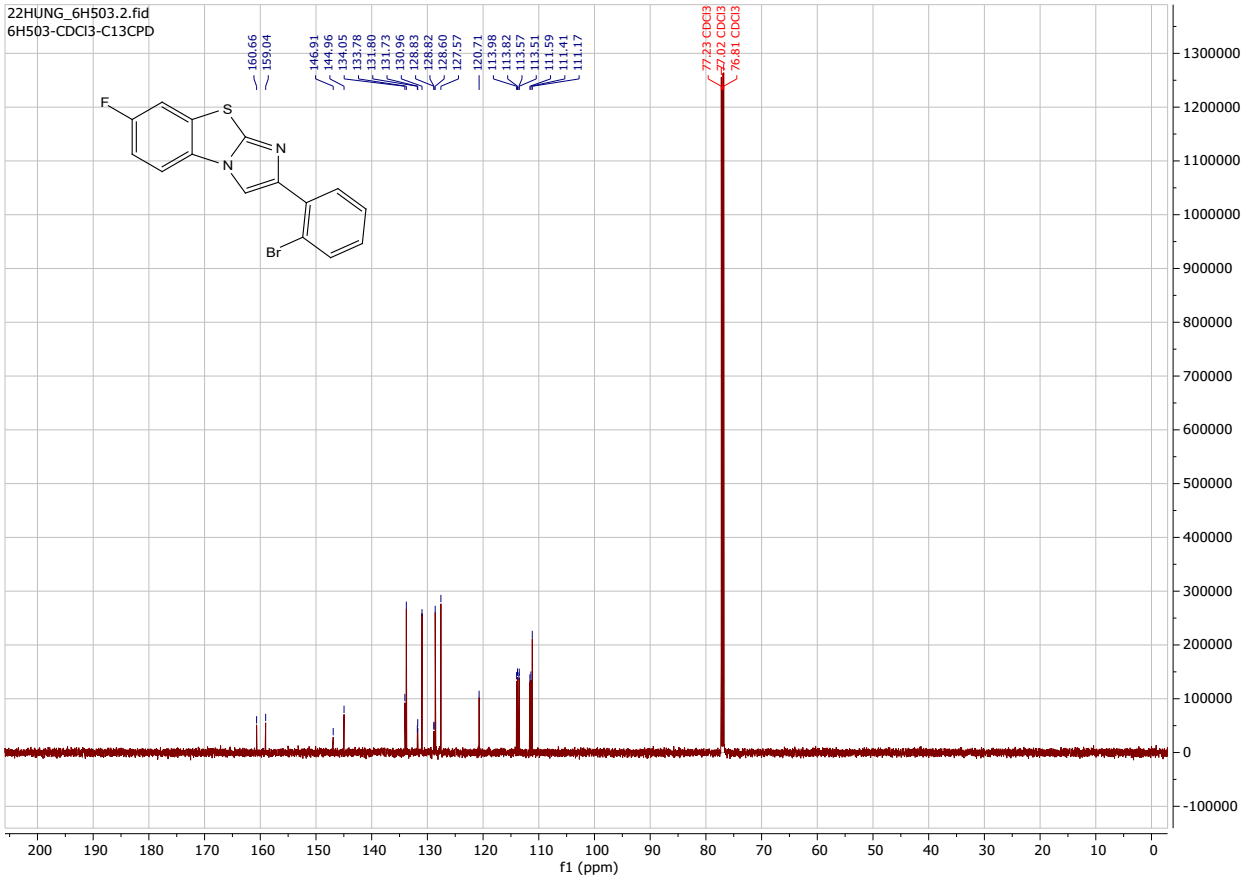
2-(2-bromophenyl)-7-methoxybenzo[d]imidazo[2,1-b]thiazole 2q



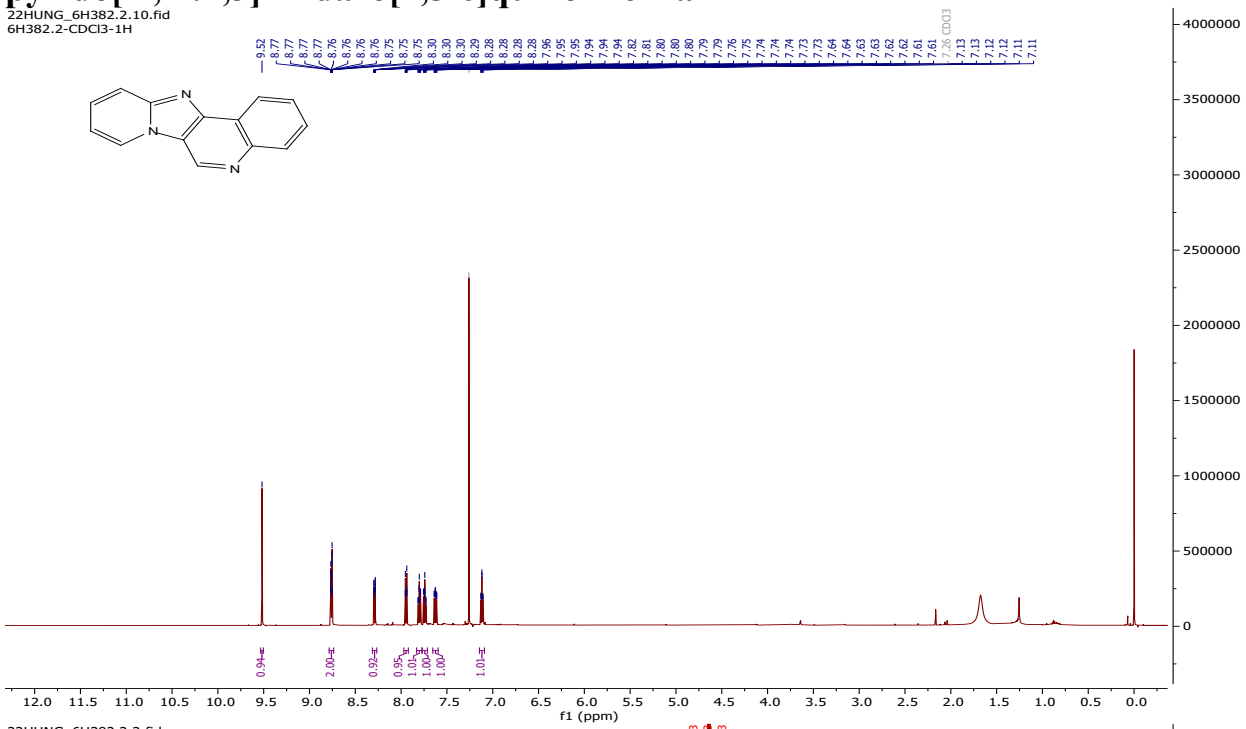


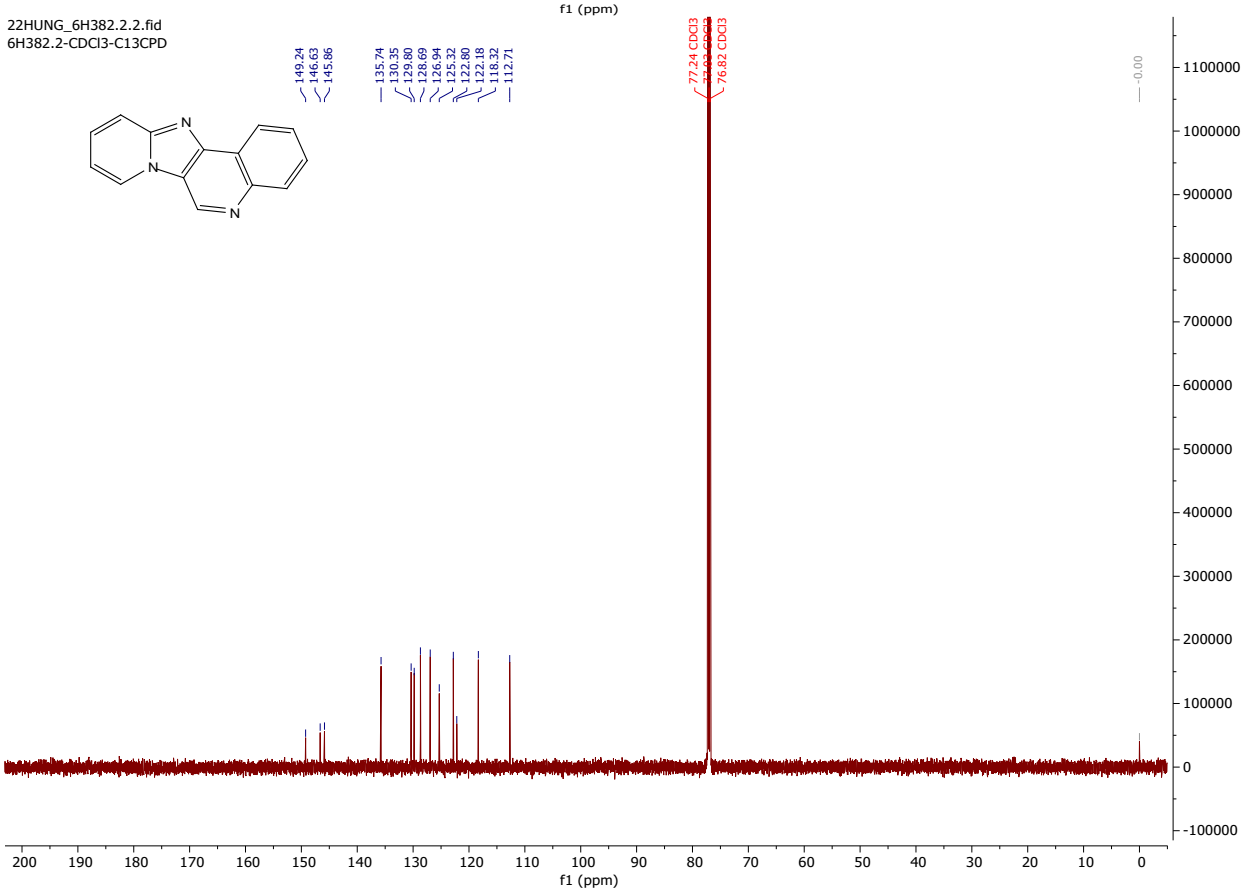
2-(2-bromophenyl)-7-fluorobenzo[d]imidazo[2,1-b]thiazole 2r



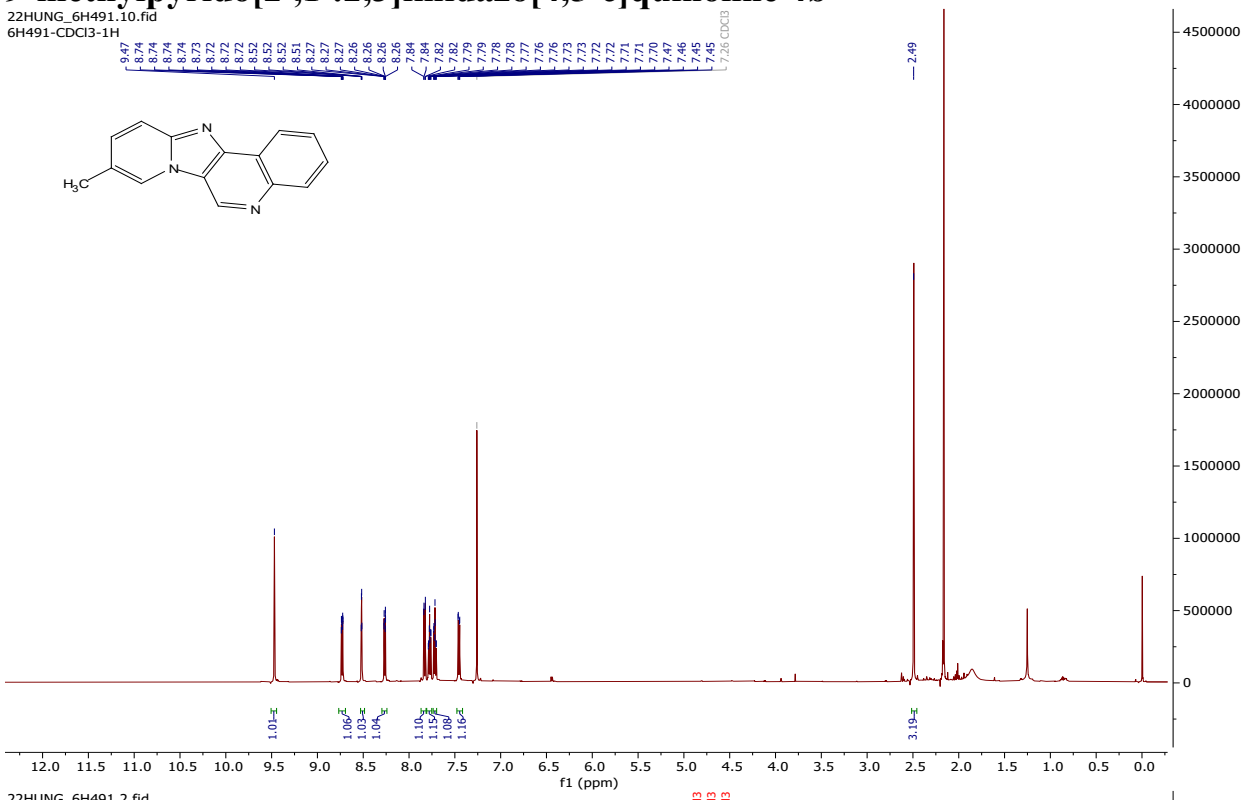


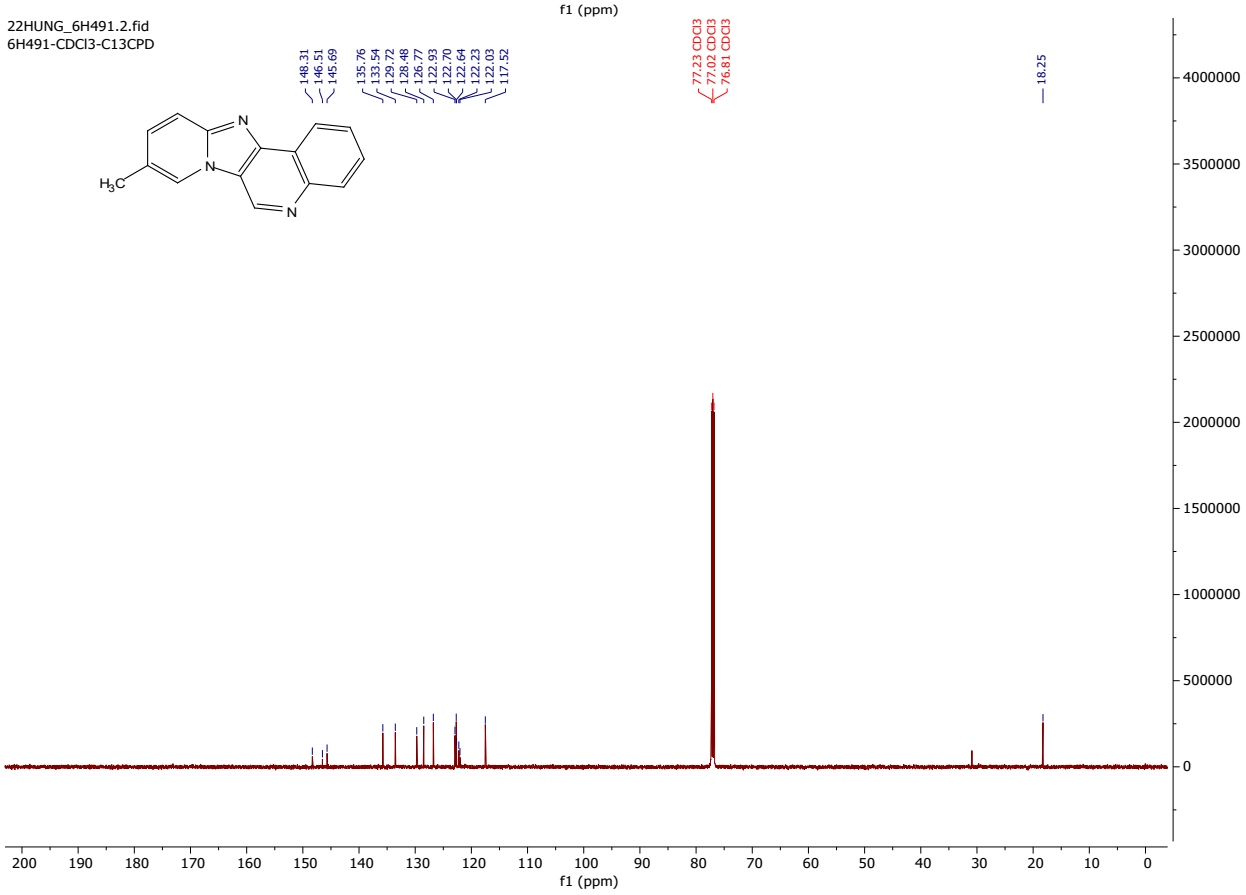
pyrido[2',1':2,3]imidazo[4,5-c]quinoline 4a





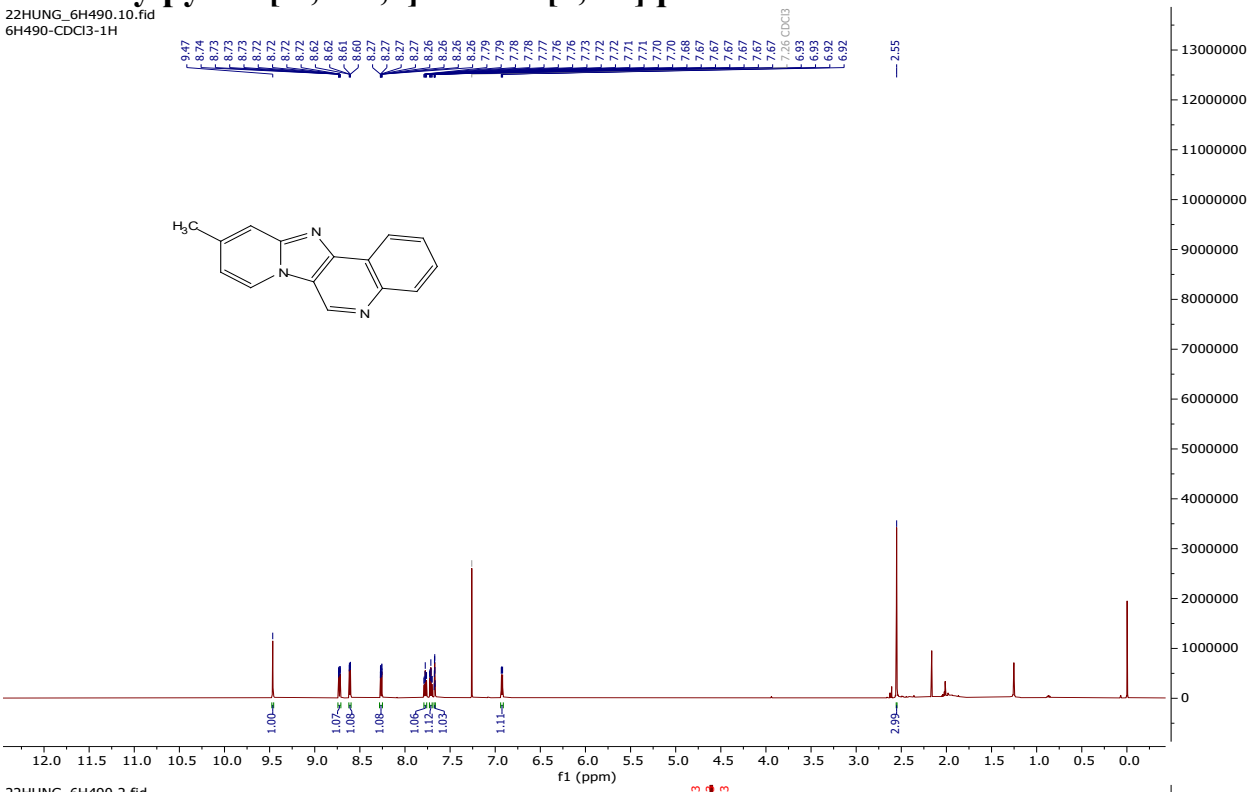
9-methylpyrido[2',1':2,3]imidazo[4,5-c]quinoline 4b





10-methylpyrido[2',1':2,3]imidazo[4,5-c]quinoline 4c

22HUNG_6H490.10.fid
6H490-CDCl3-1H



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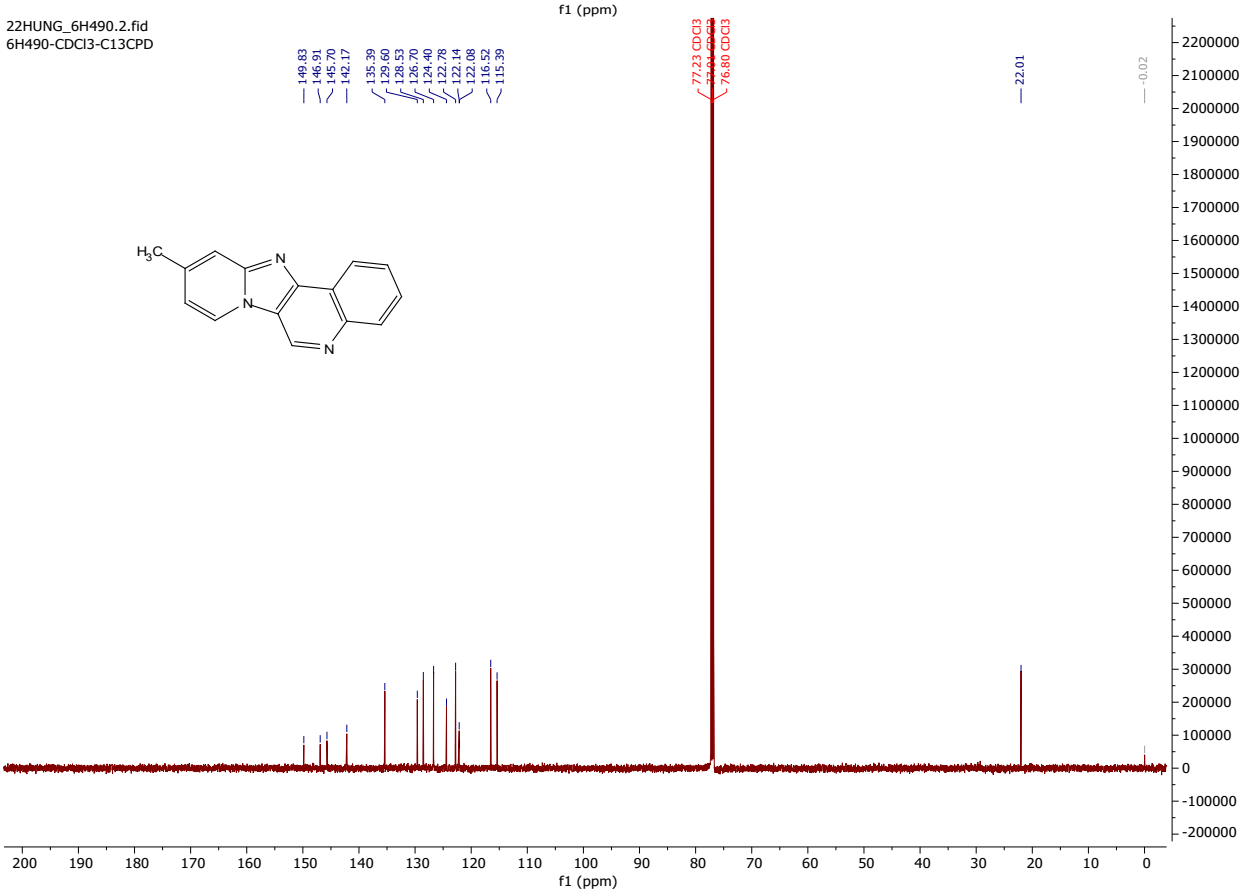
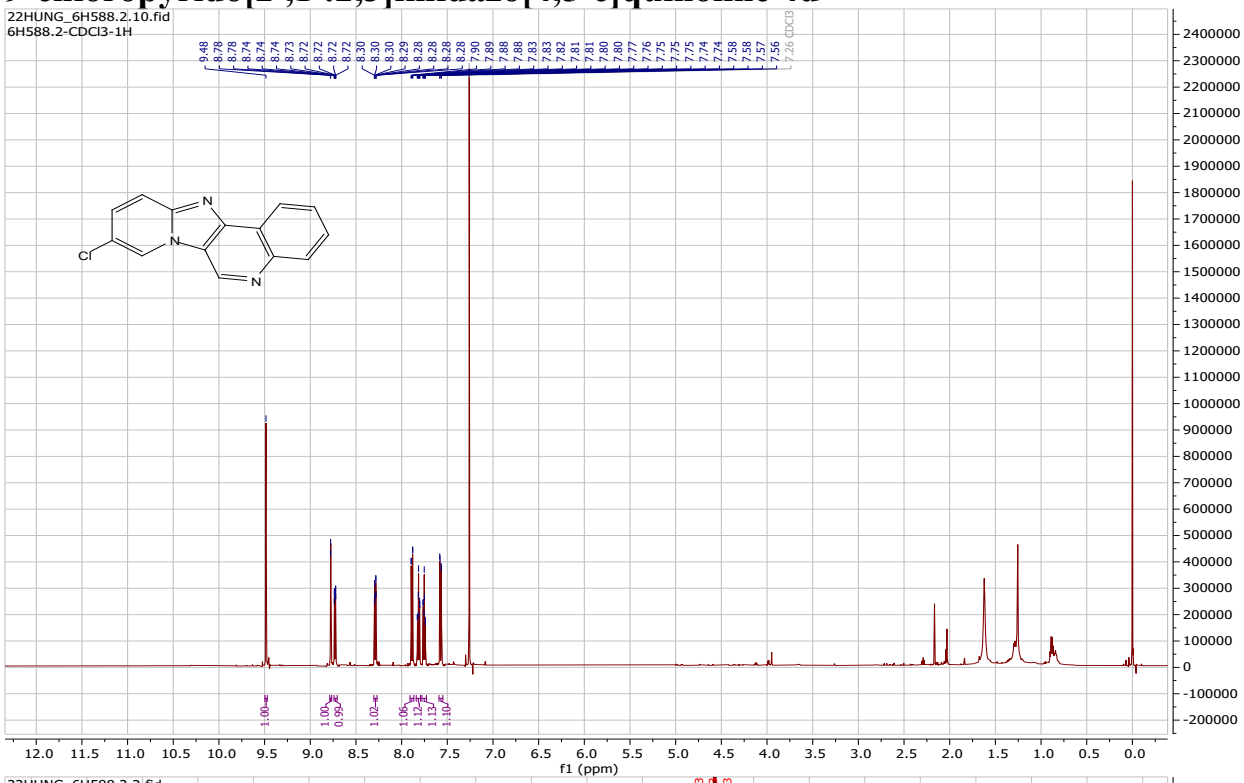
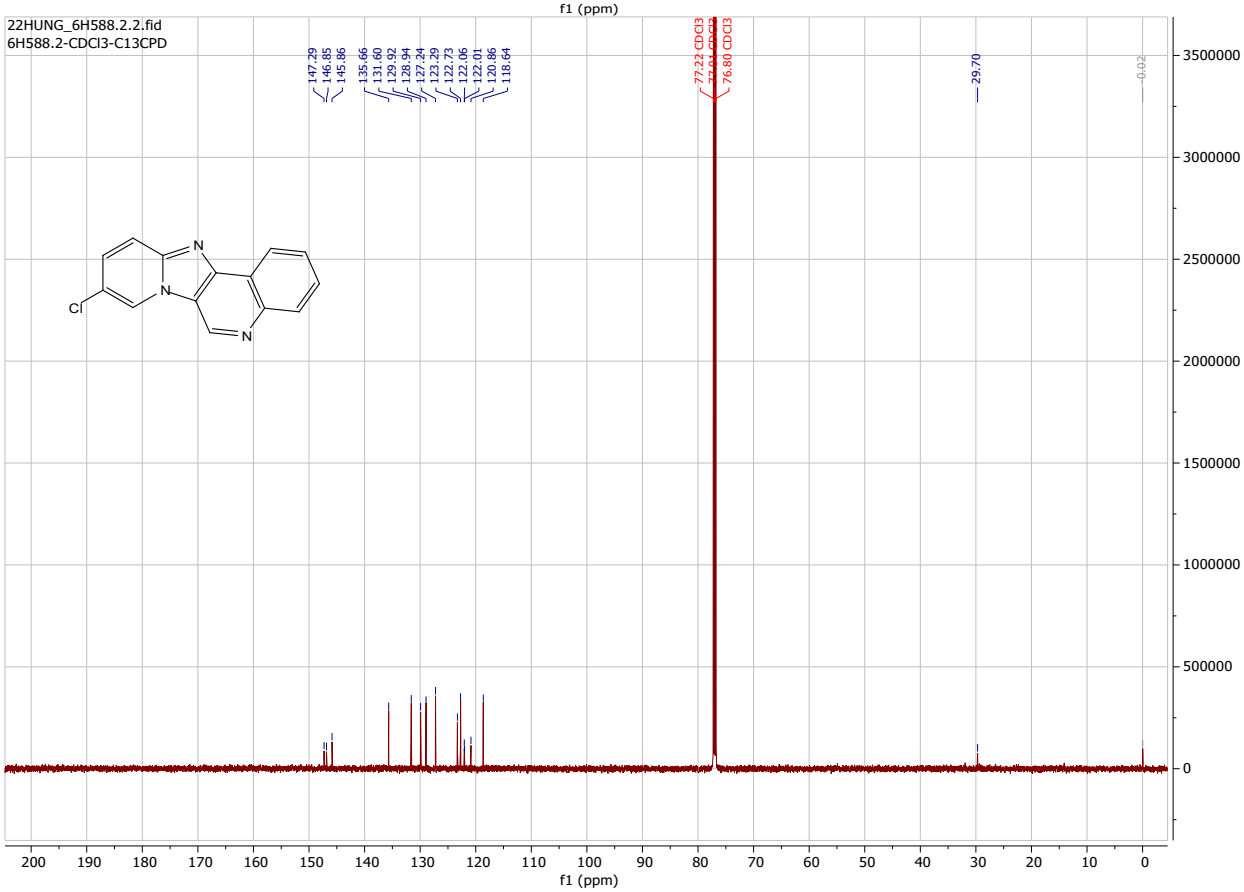
6H490-CDCl3-C13CPD

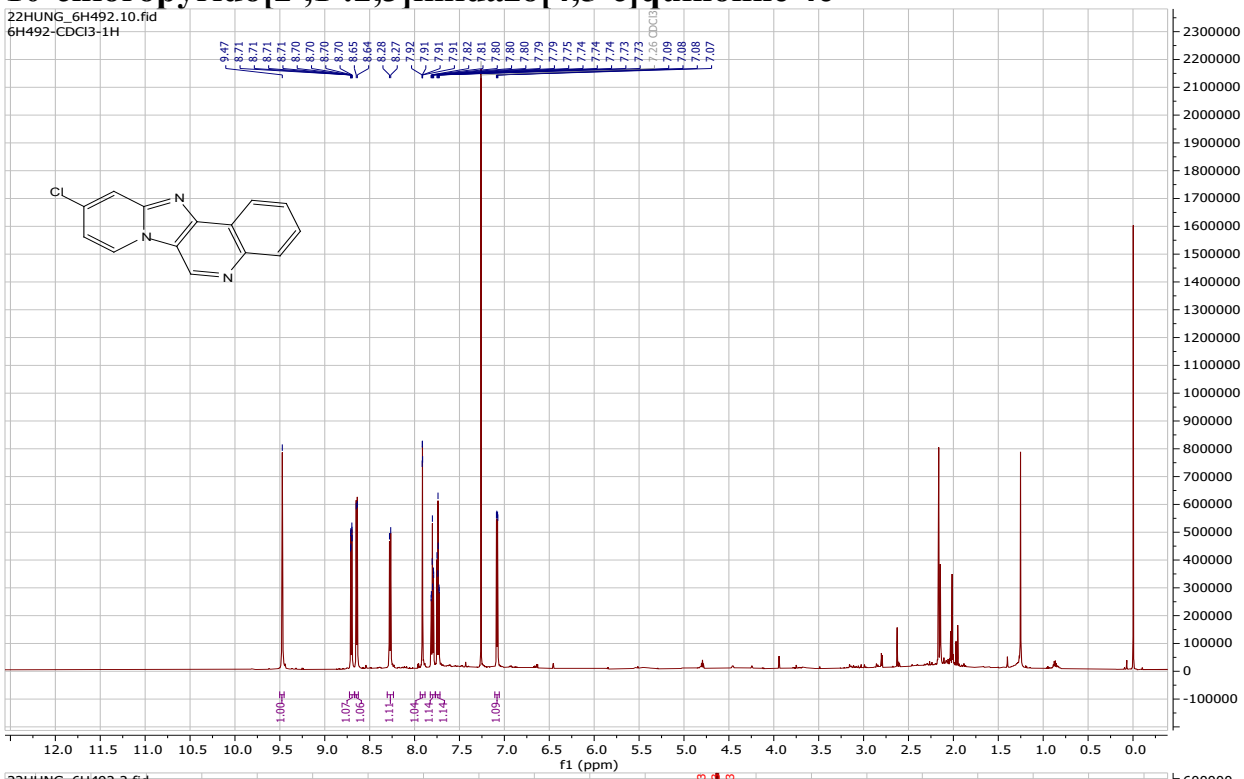


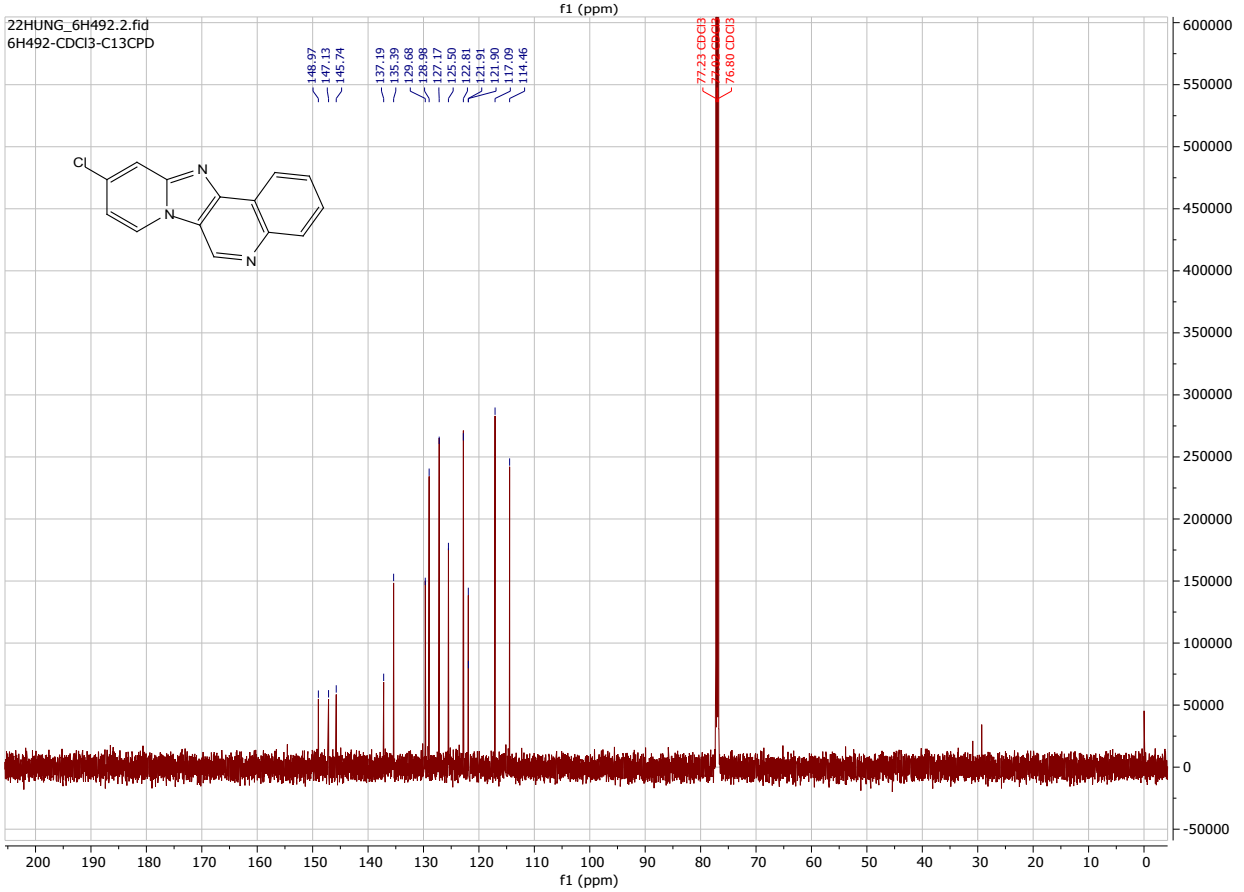
9-chloropyrido[2',1':2,3]imidazo[4,5-c]quinoline 4d

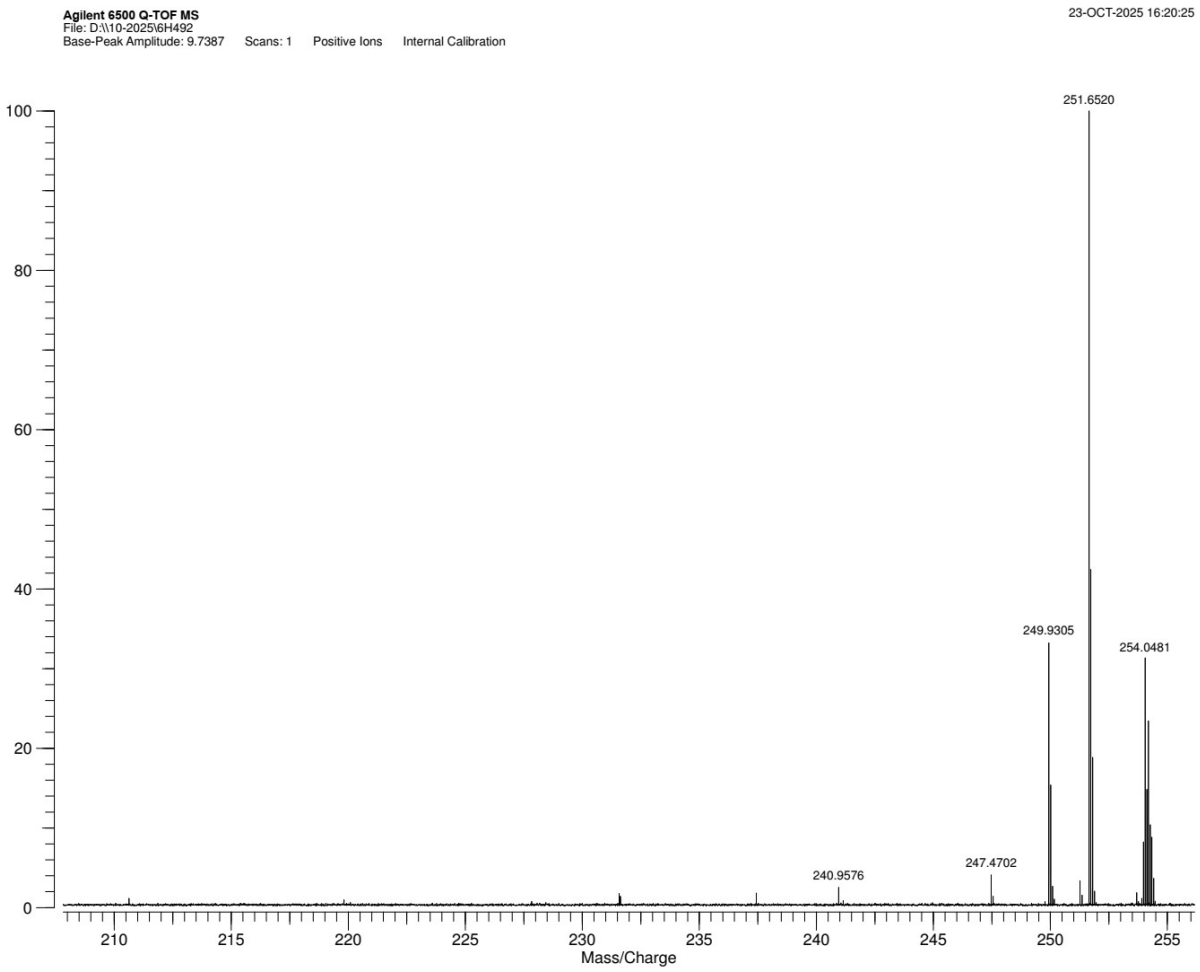




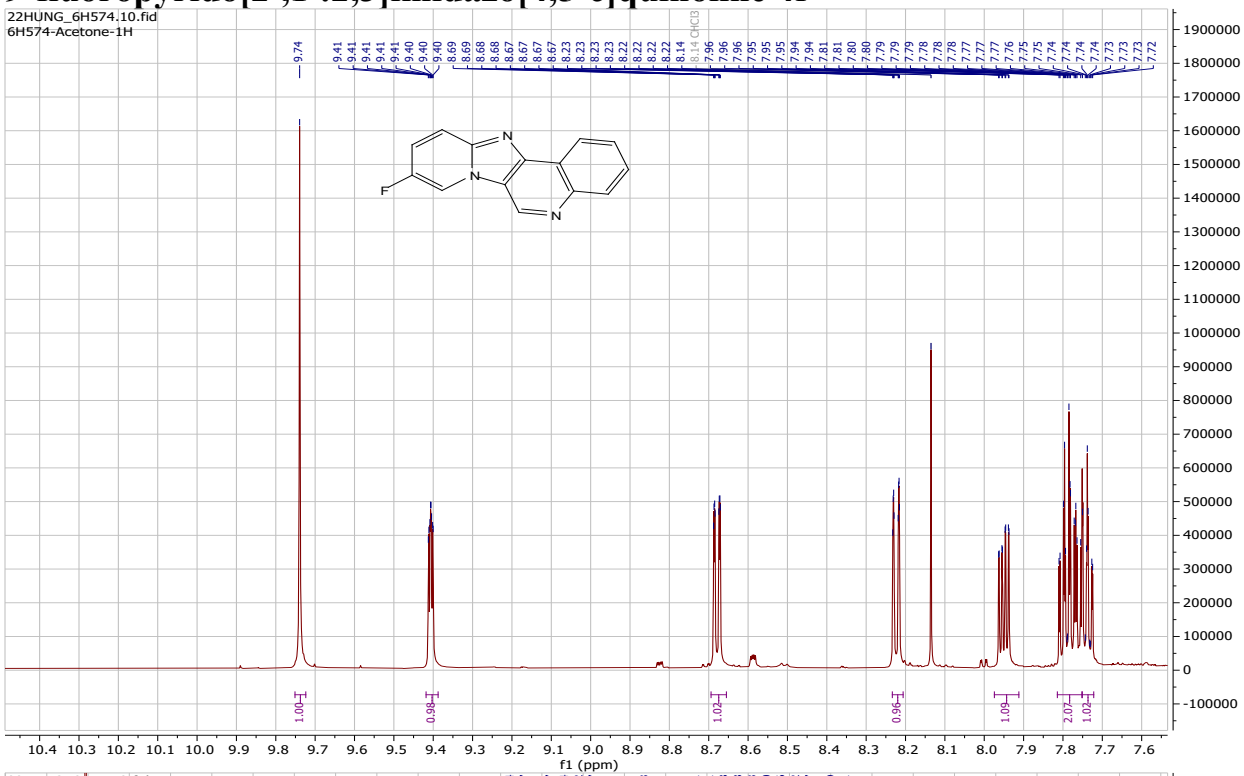
10-chloropyrido[2',1':2,3]imidazo[4,5-c]quinoline 4e

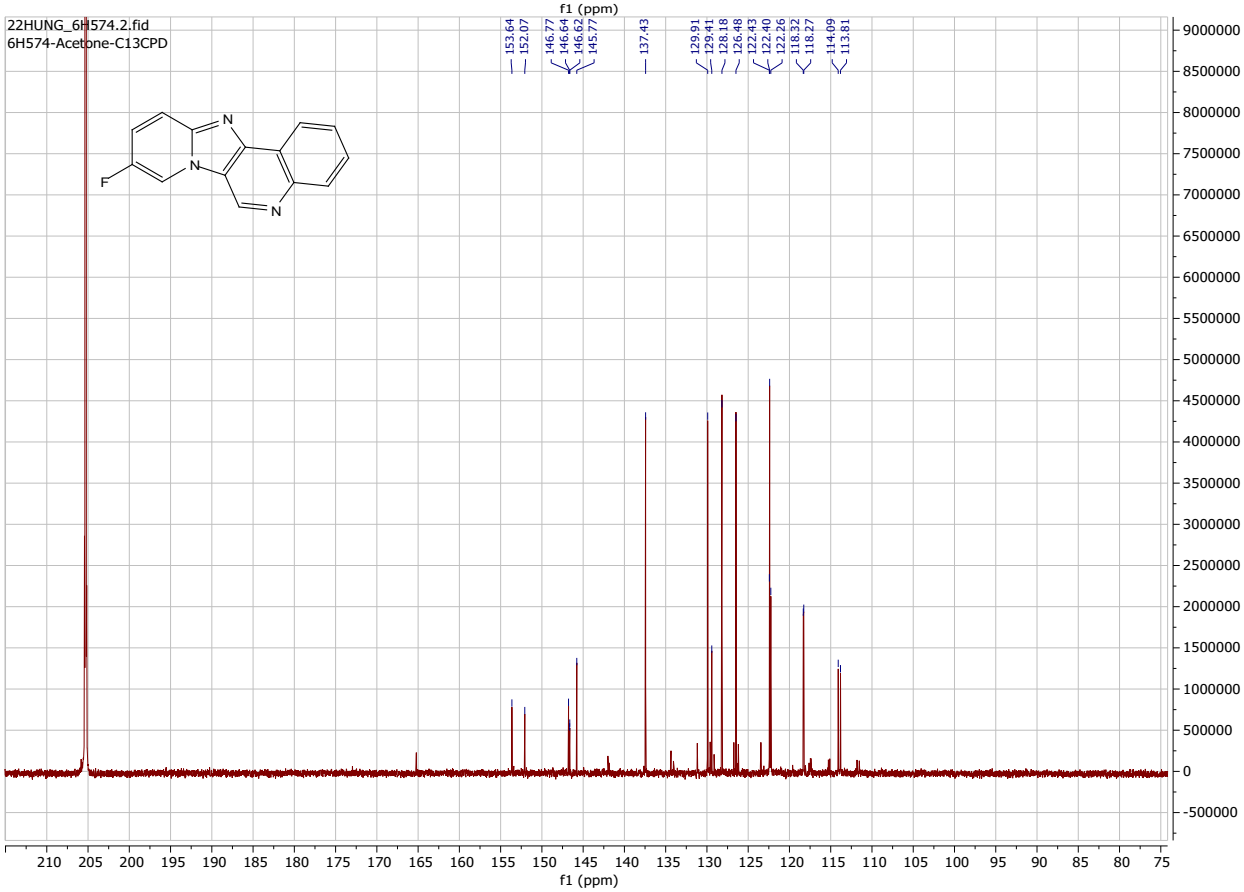


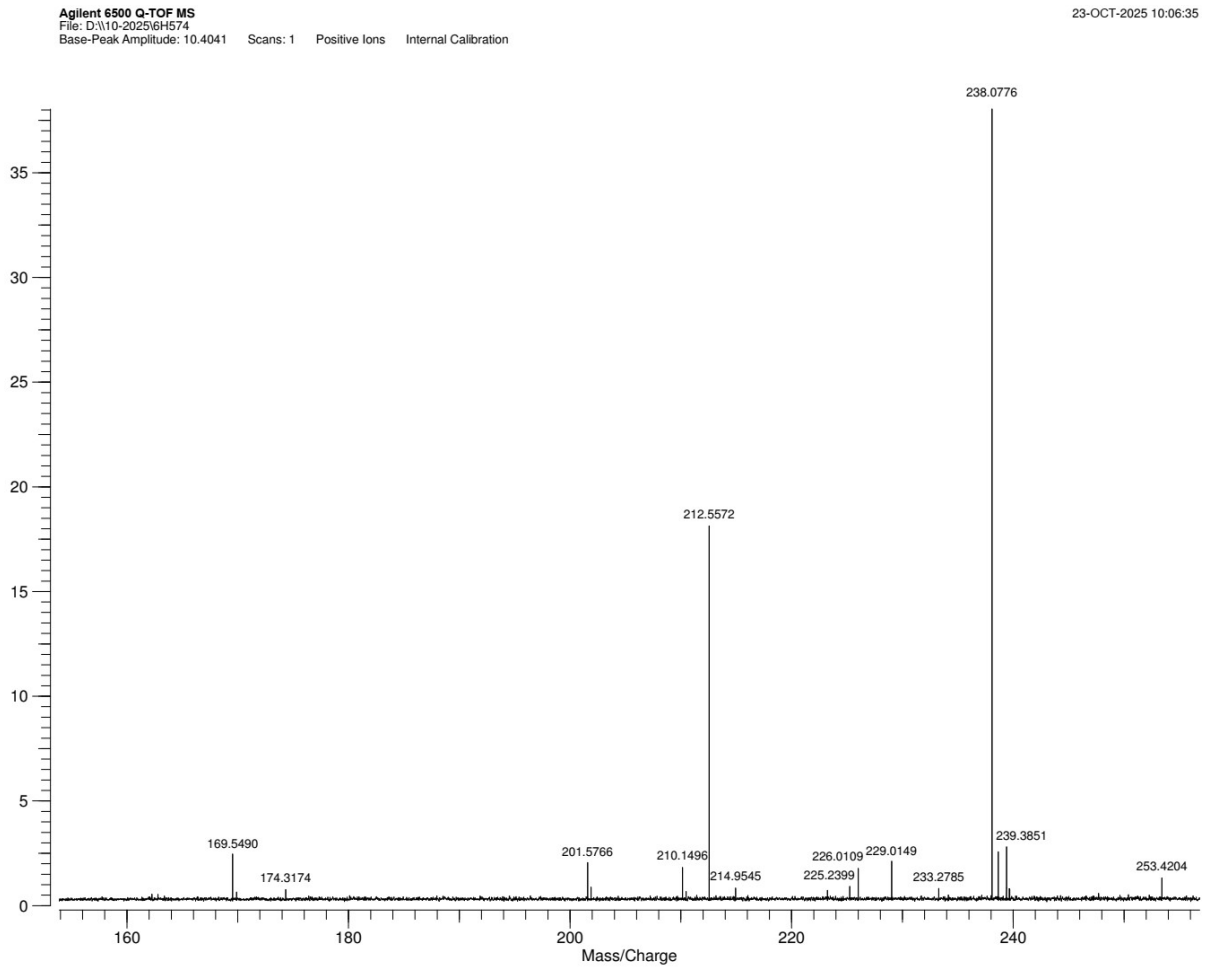



Agilent 6500 Q-TOF MS
File: D:\\10-2025\6H492
Base-Peak Amplitude: 9.7387 Scans: 1 Positive Ions Internal Calibration
23-OCT-2025 16:20:25
251.6520
249.9305
254.0481
247.4702
240.9576
100
80
60
40
20
0
210
215
220
225
230
235
240
245
250
255
Mass/Charge

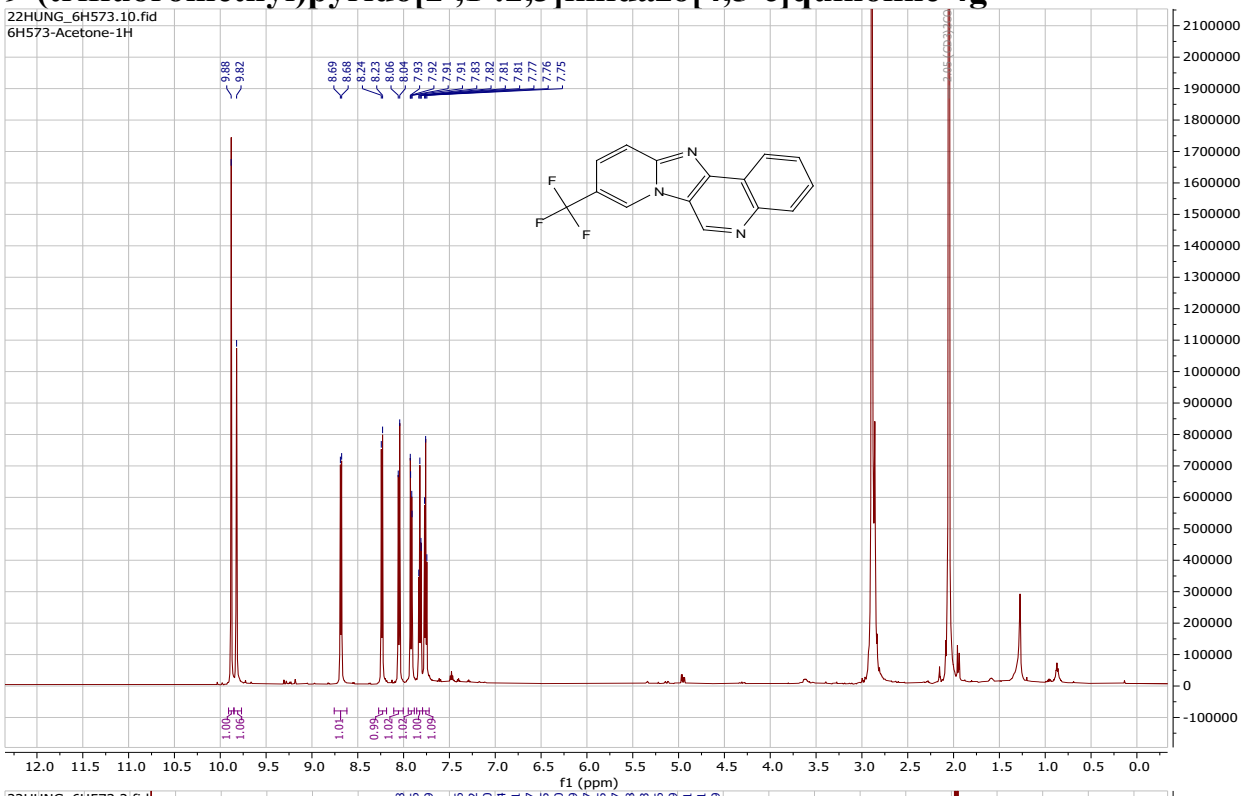
9-fluoropyrido[2',1':2,3]imidazo[4,5-c]quinoline 4f

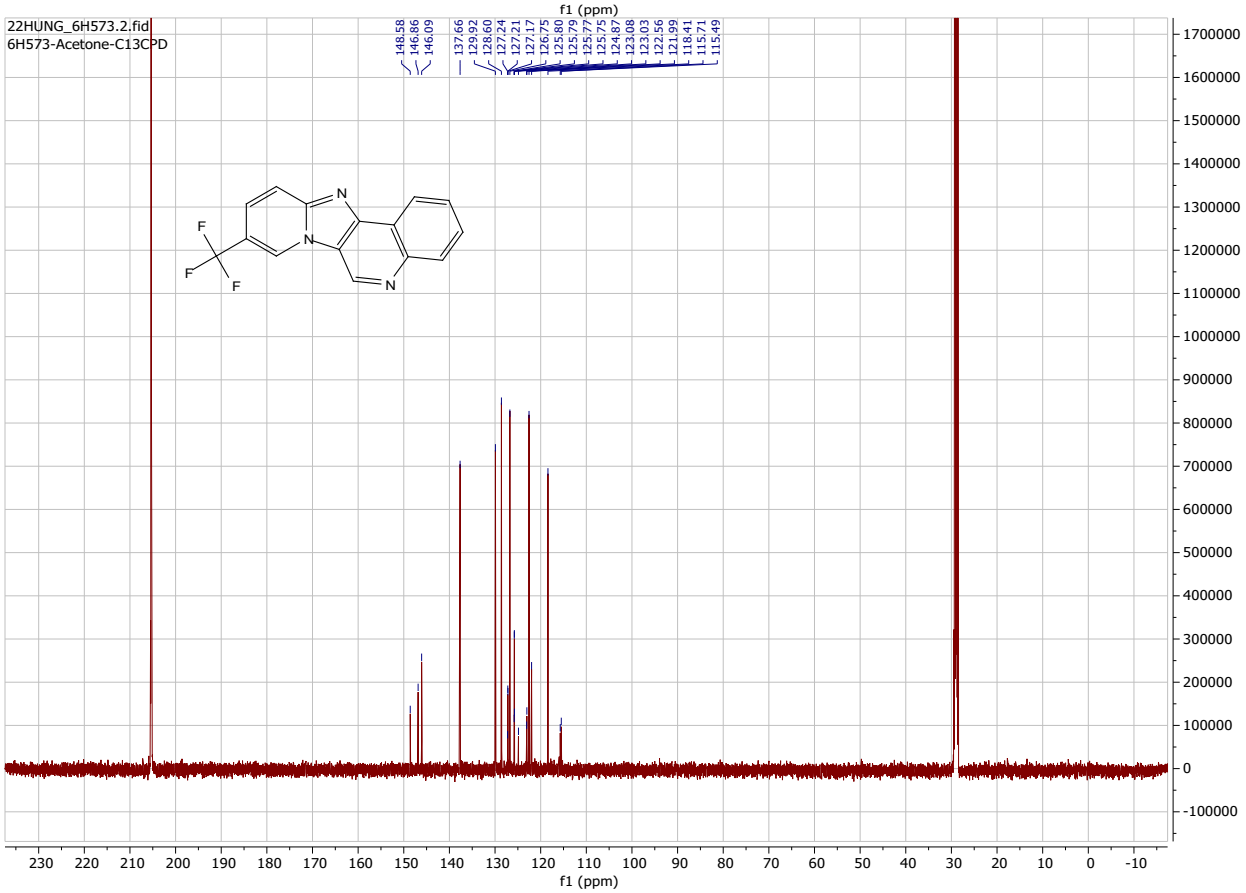


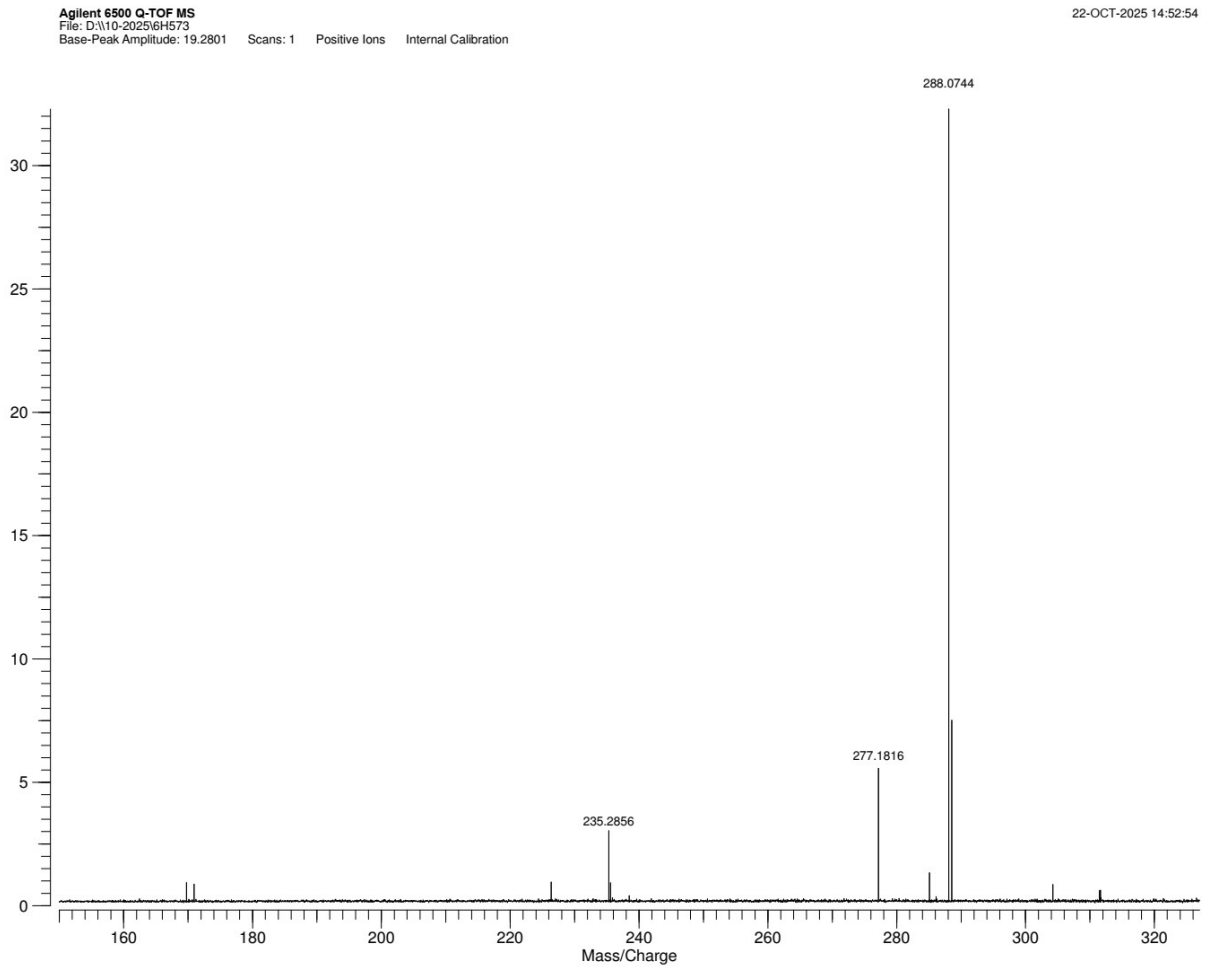



Agilent 6500 Q-TOF MS
File: D:\\10-2025\6H574
Base-Peak Amplitude: 10.4041 Scans: 1 Positive Ions Internal Calibration
23-OCT-2025 10:06:35
238.0776
212.5572
169.5490
174.3174
201.5766
210.1496
214.9545
225.2399
226.0109
229.0149
233.2785
239.3851
253.4204
Mass/Charge

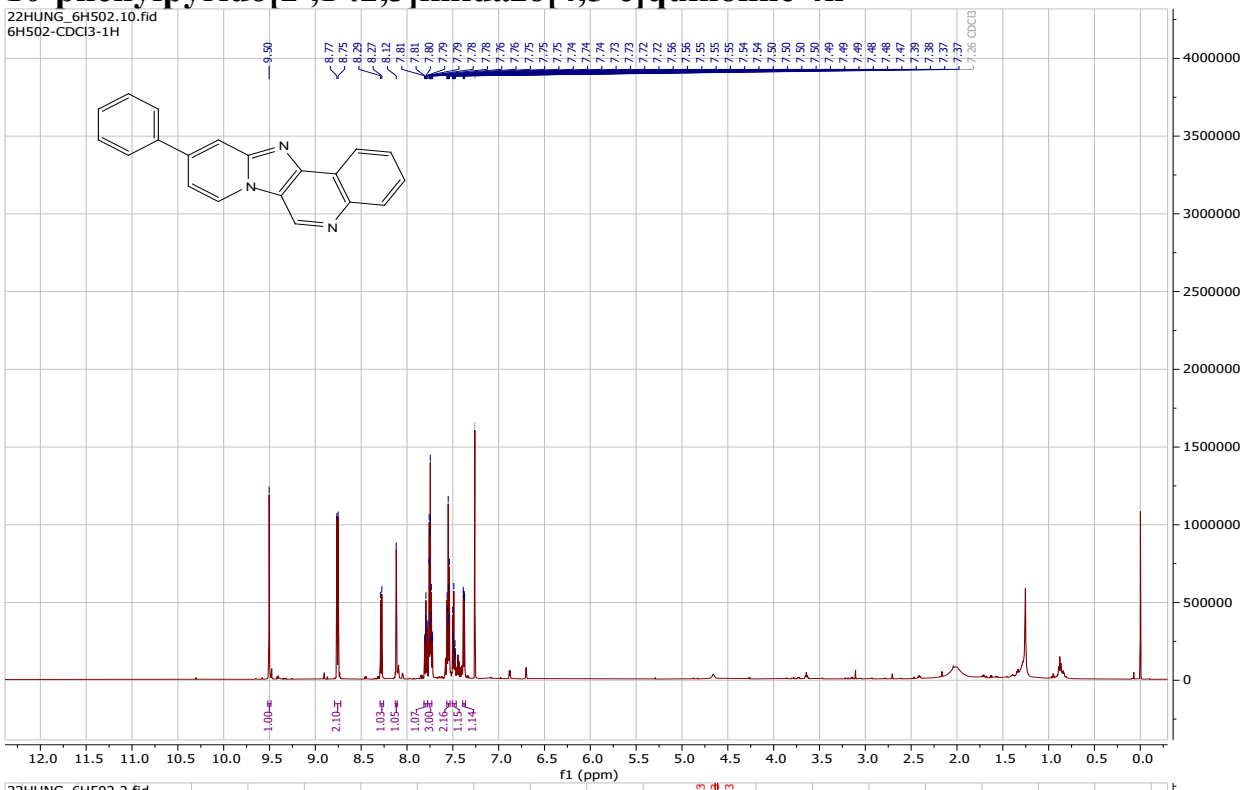
9-(trifluoromethyl)pyrido[2',1':2,3]imidazo[4,5-c]quinoline 4g

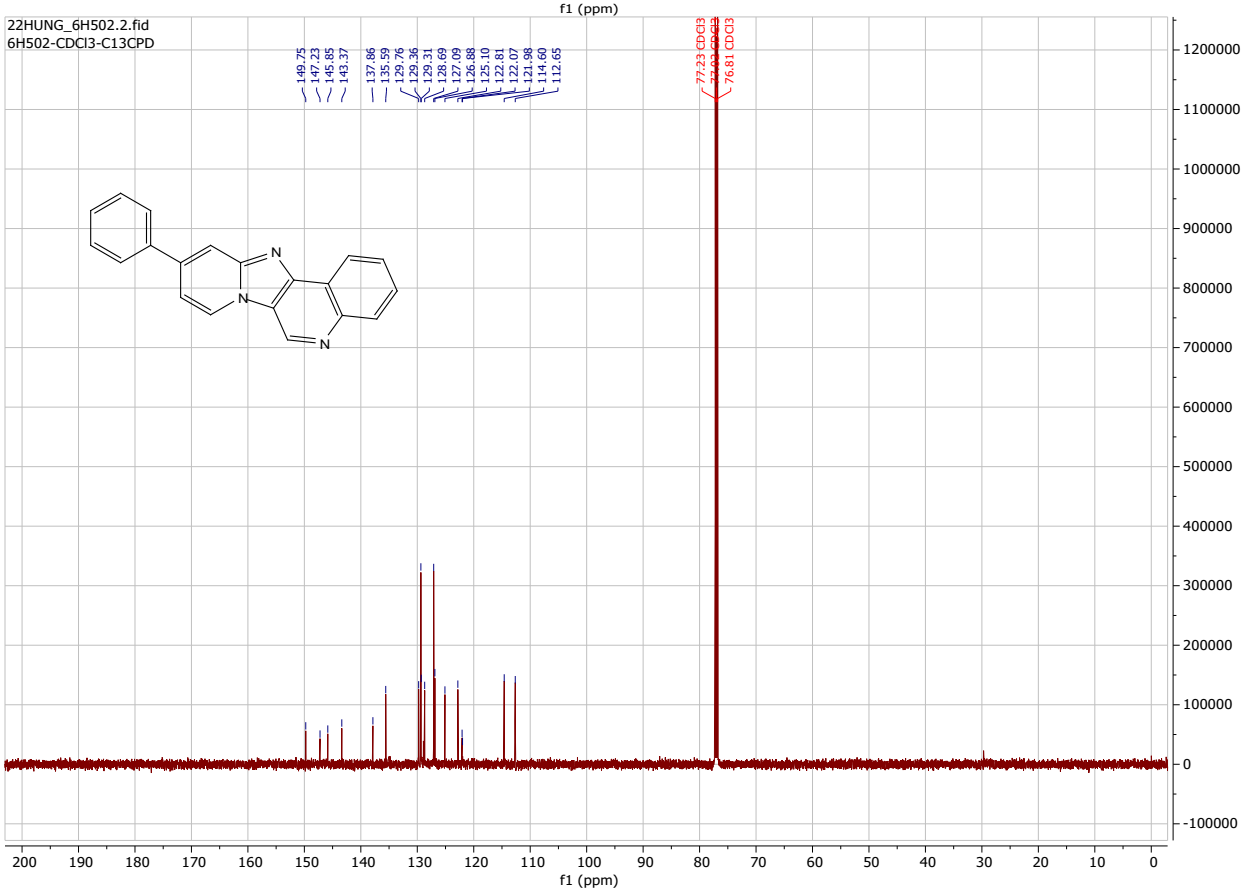


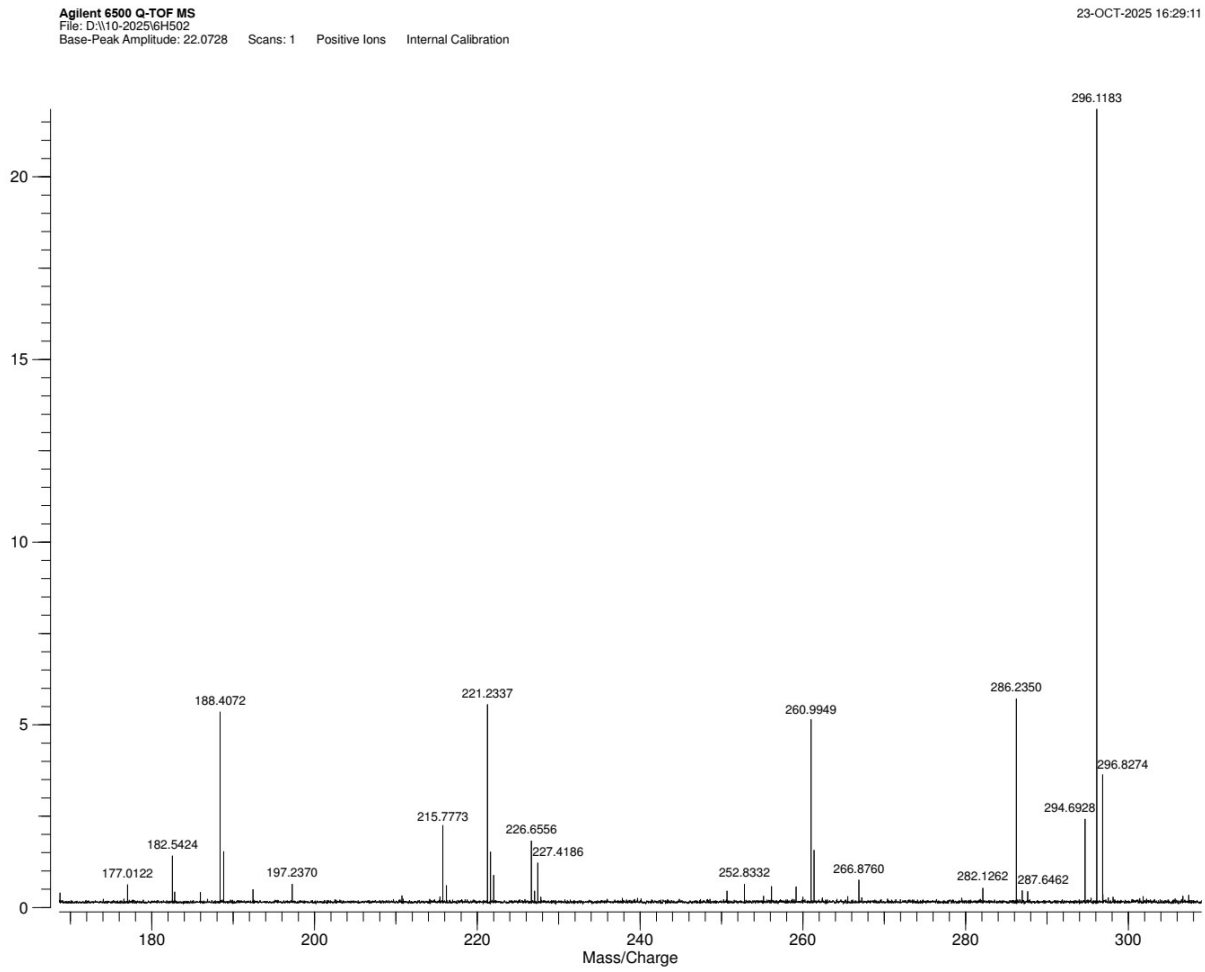



Agilent 6500 Q-TOF MS
File: D:\\10-2025\6H573
Base-Peak Amplitude: 19.2801 Scans: 1 Positive Ions Internal Calibration
22-OCT-2025 14:52:54
288.0744
277.1816
235.2856
30
25
20
15
10
5
0
160
180
200
220
240
260
280
300
320
Mass/Charge

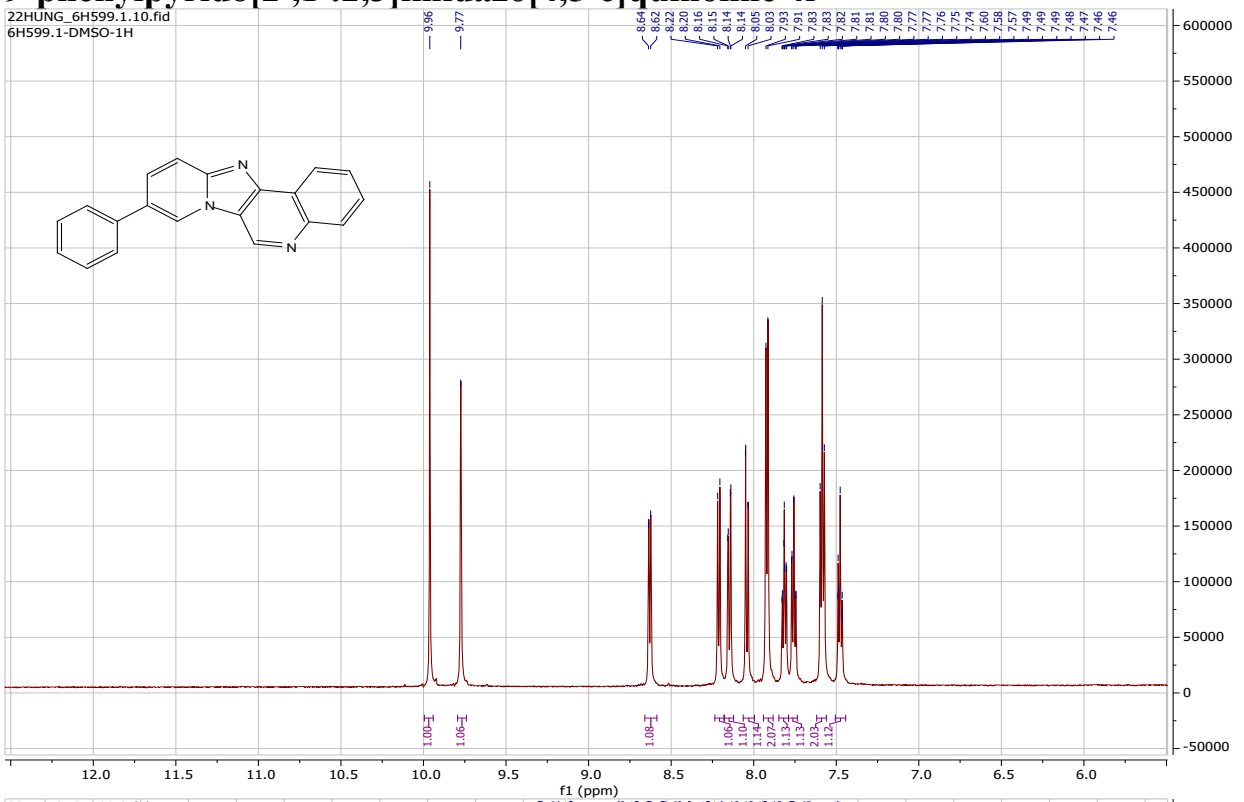
10-phenylpyrido[2',1':2,3]imidazo[4,5-c]quinoline 4h

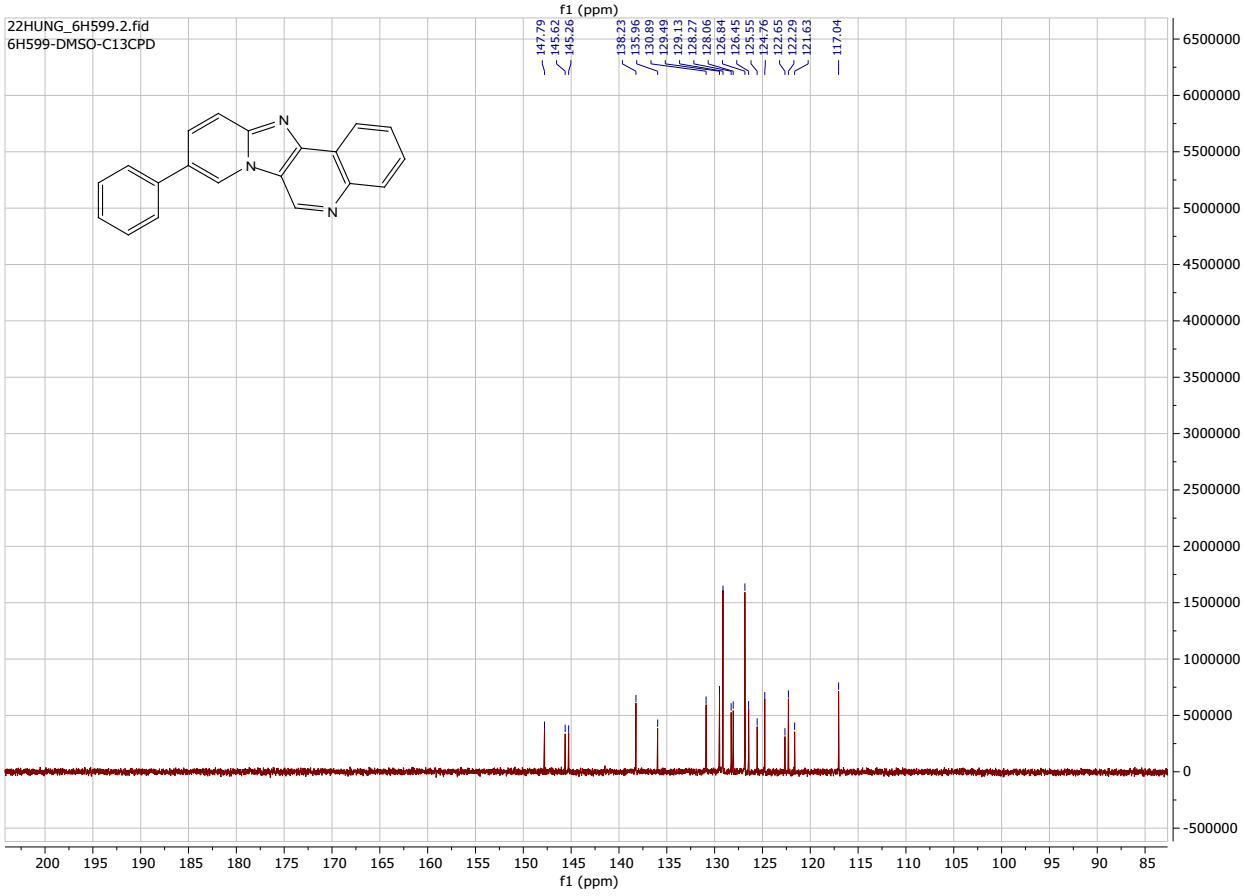


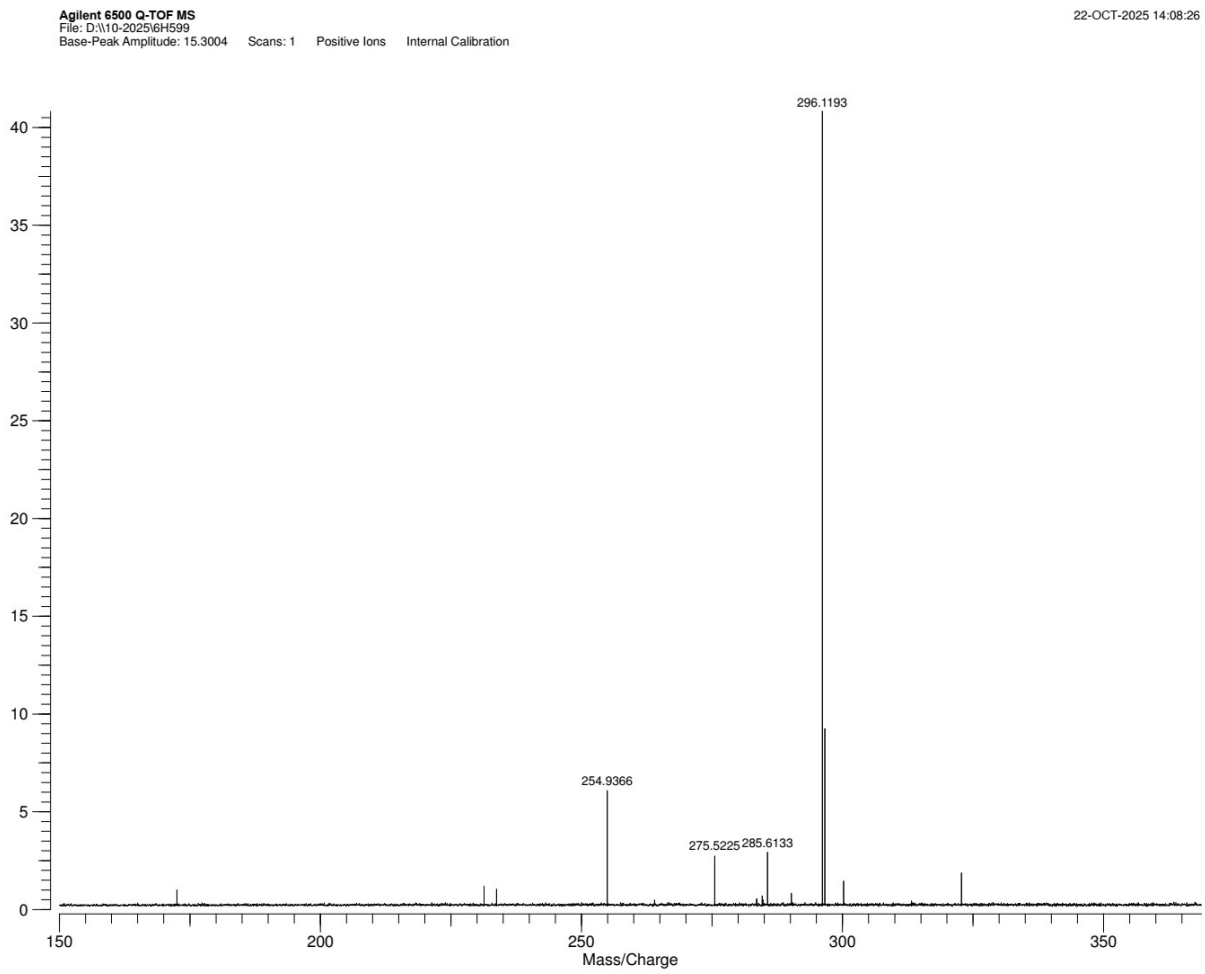



Agilent 6500 Q-TOF MS
File: D:\\10-2025\6H502
Base-Peak Amplitude: 22.0728 Scans: 1 Positive Ions Internal Calibration
23-OCT-2025 16:29:11
296.1183
221.2337
188.4072
260.9949
286.2350
296.8274
294.6928
215.7773
226.6556
227.4186
182.5424
177.0122
197.2370
252.8332
266.8760
282.1262
287.6462
Mass/Charge

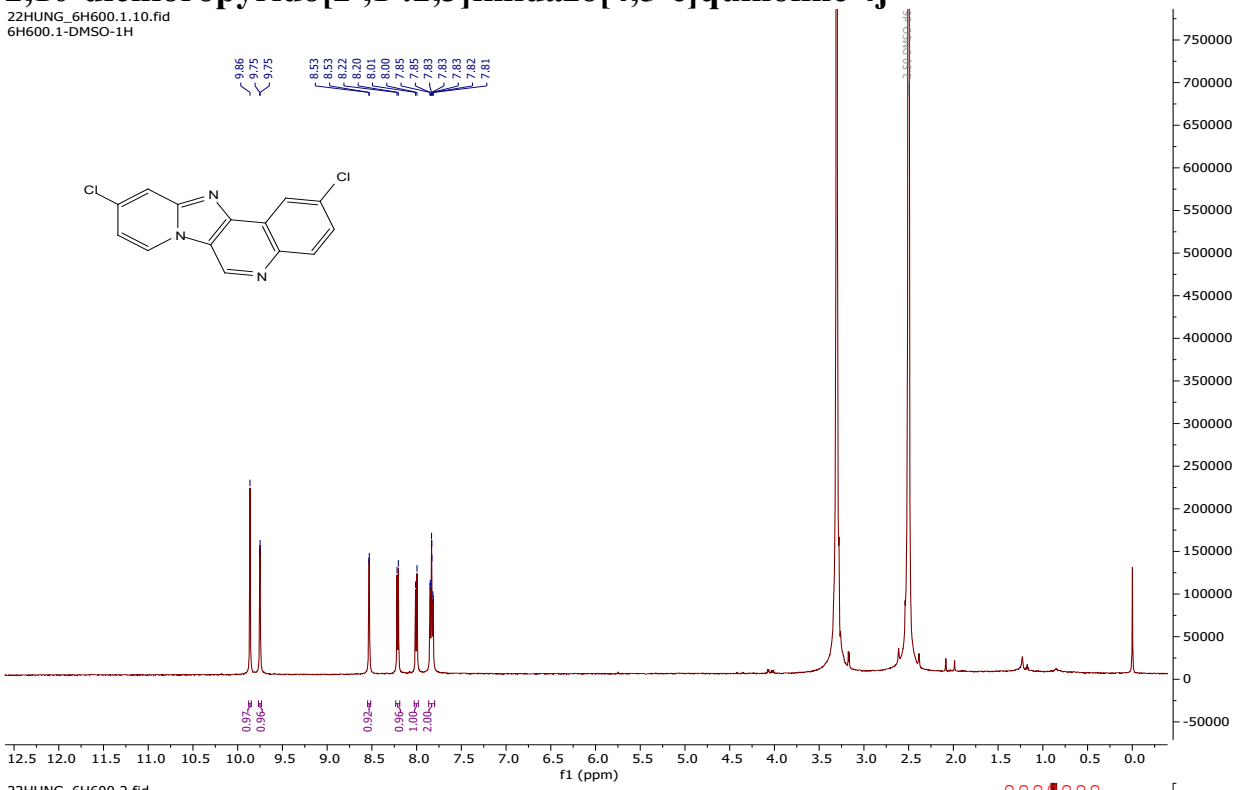
9-phenylpyrido[2',1':2,3]imidazo[4,5-c]quinoline 4i

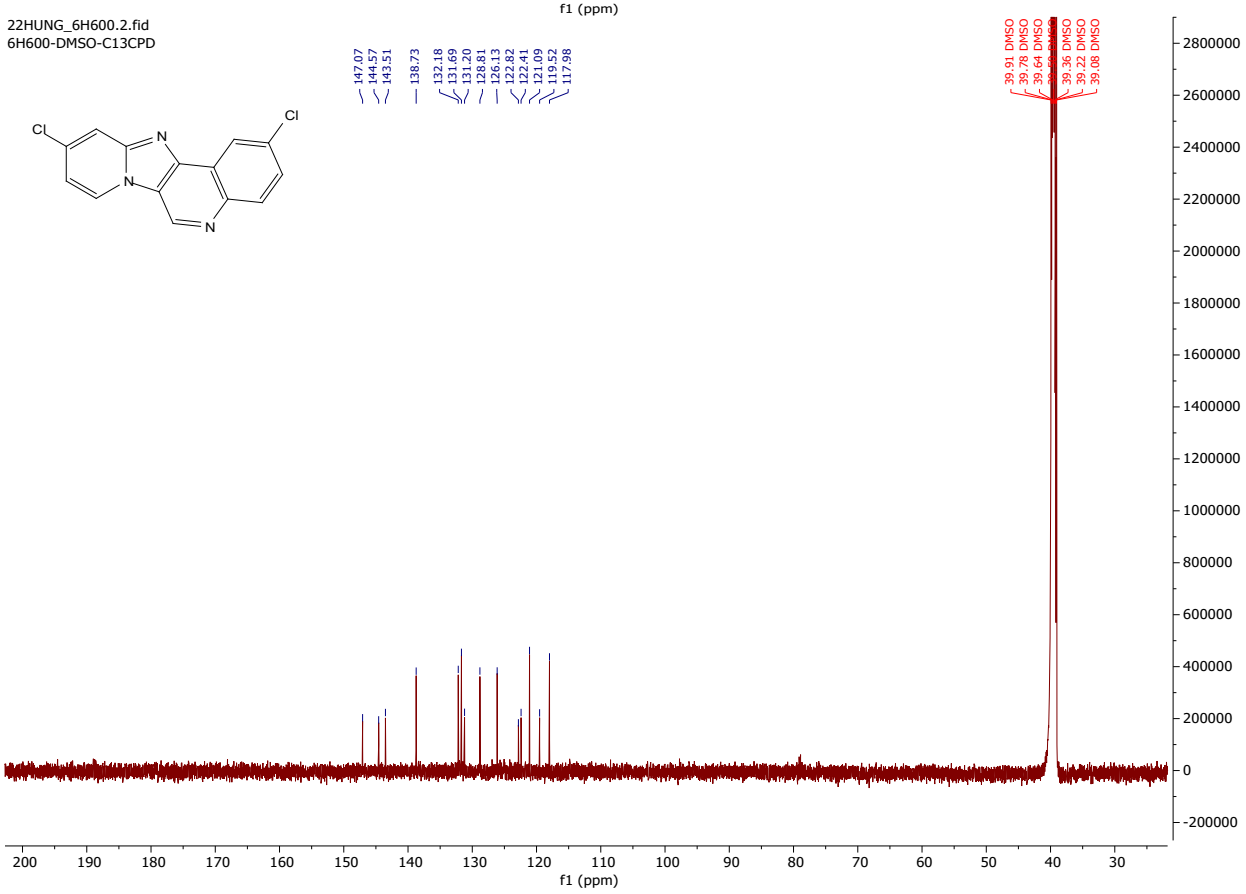


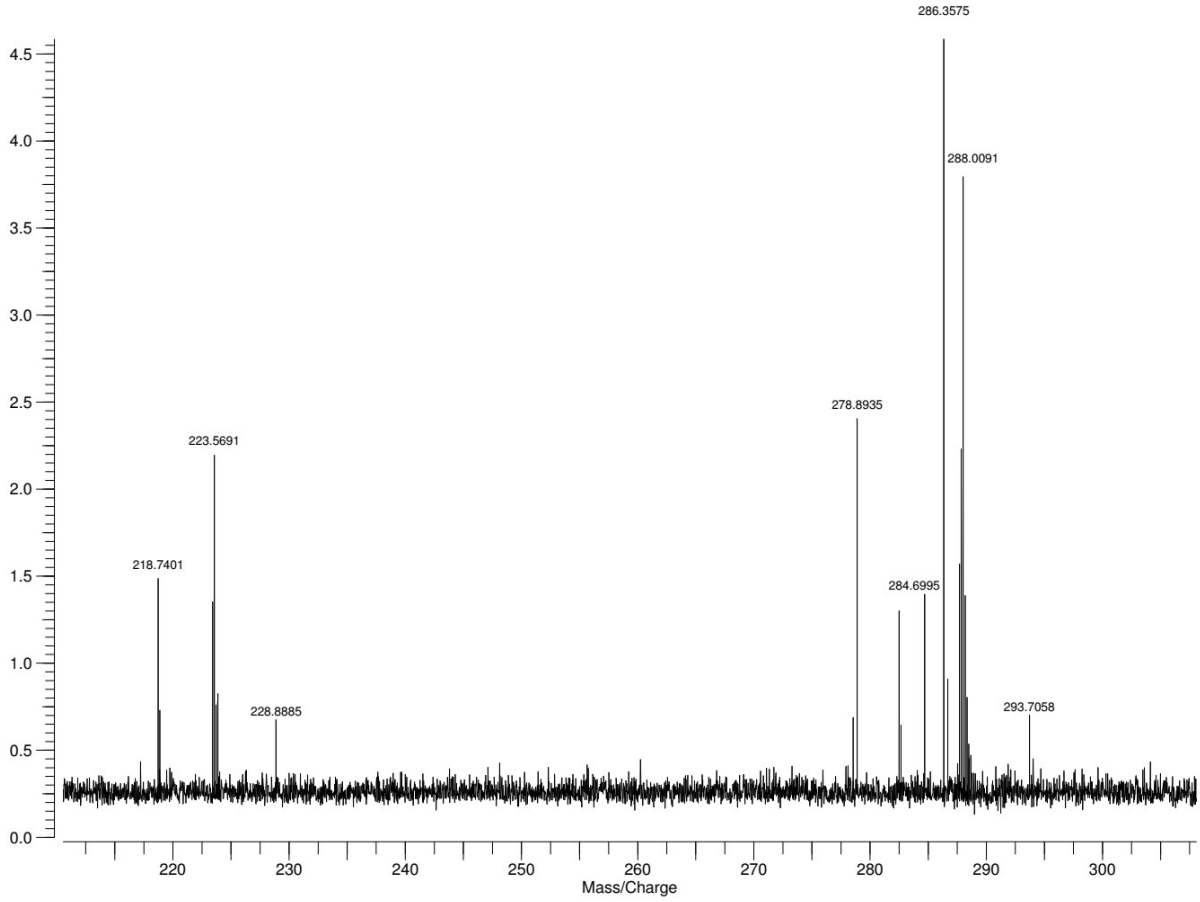



Agilent 6500 Q-TOF MS
File: D:\\10-2025\6H599
Base-Peak Amplitude: 15.3004 Scans: 1 Positive Ions Internal Calibration
22-OCT-2025 14:08:26
296.1193
254.9366
275.5225
285.6133
Mass/Charge

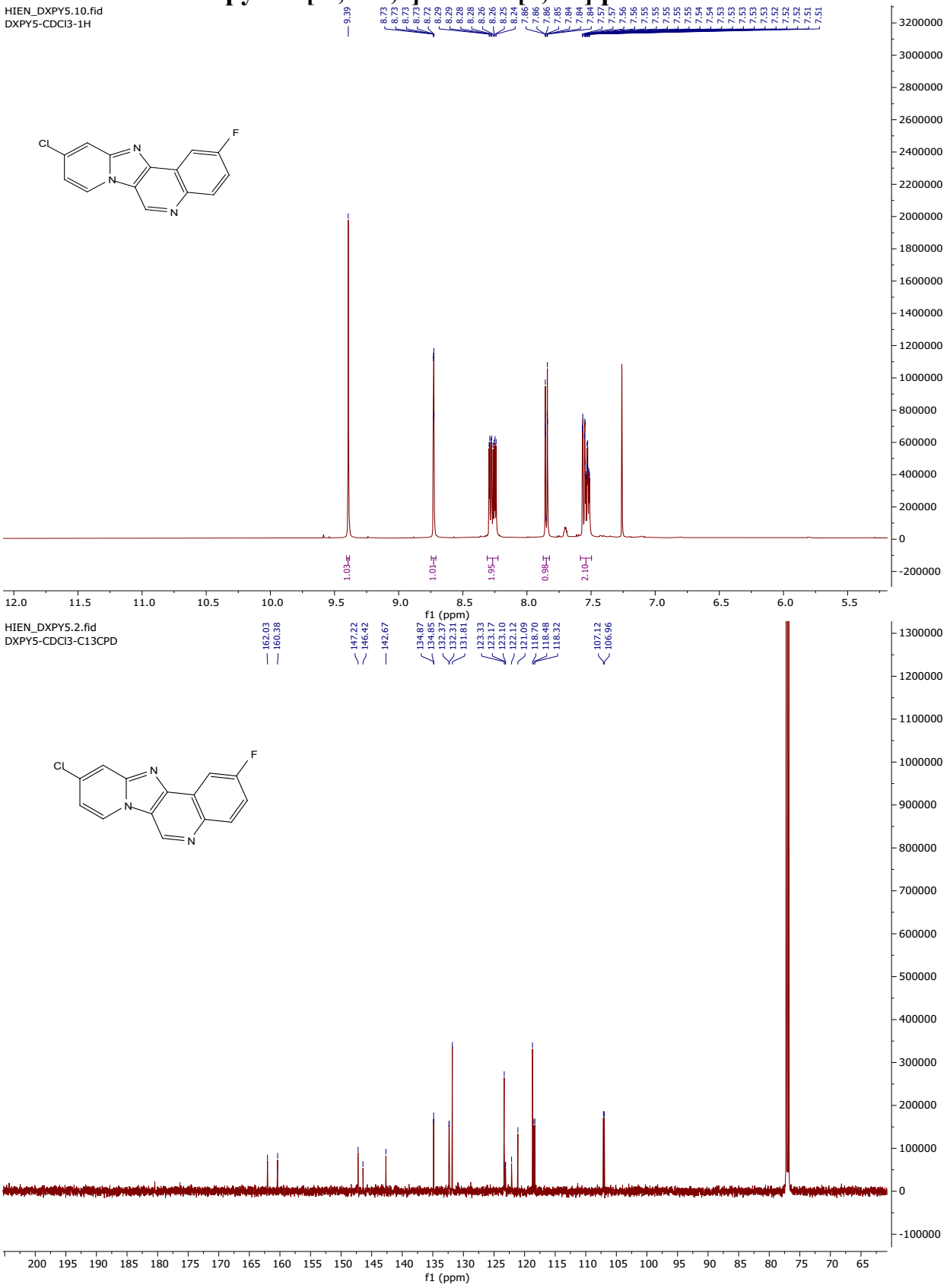
2,10-dichloropyrido[2',1':2,3]imidazo[4,5-c]quinoline 4j



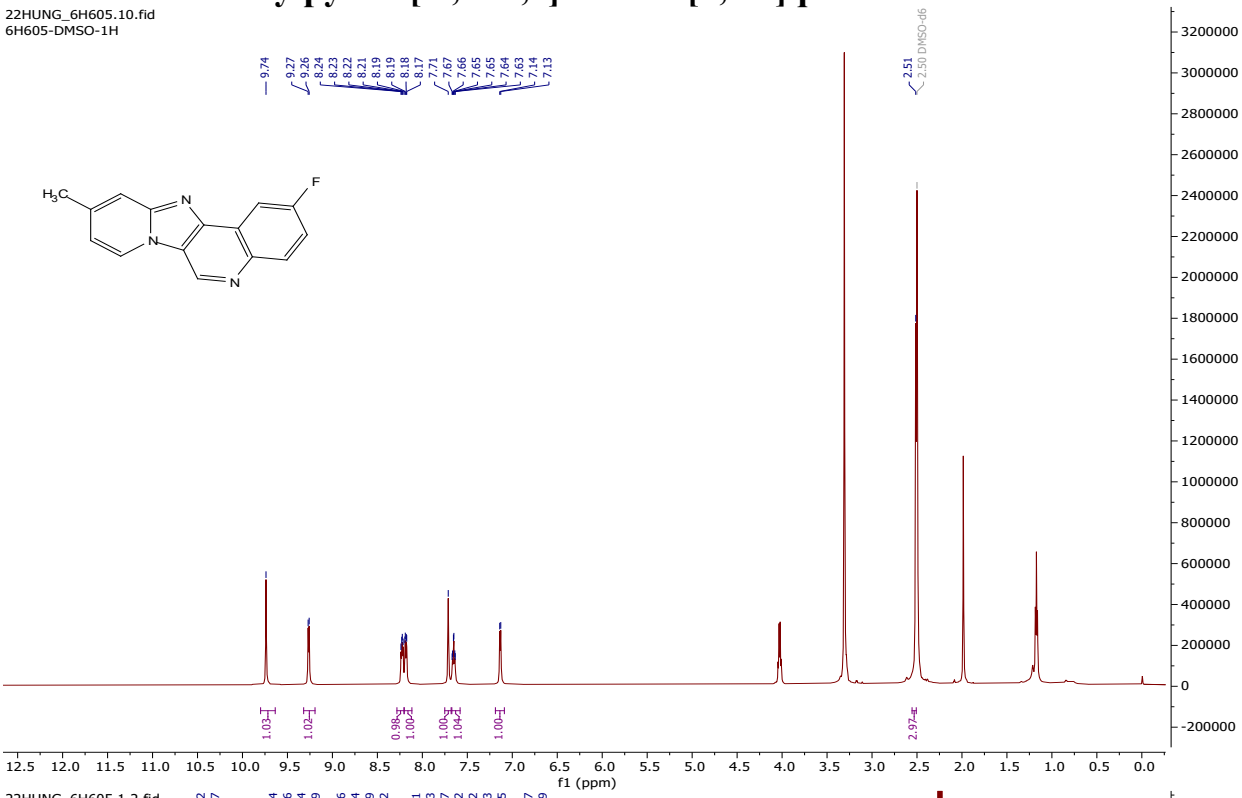


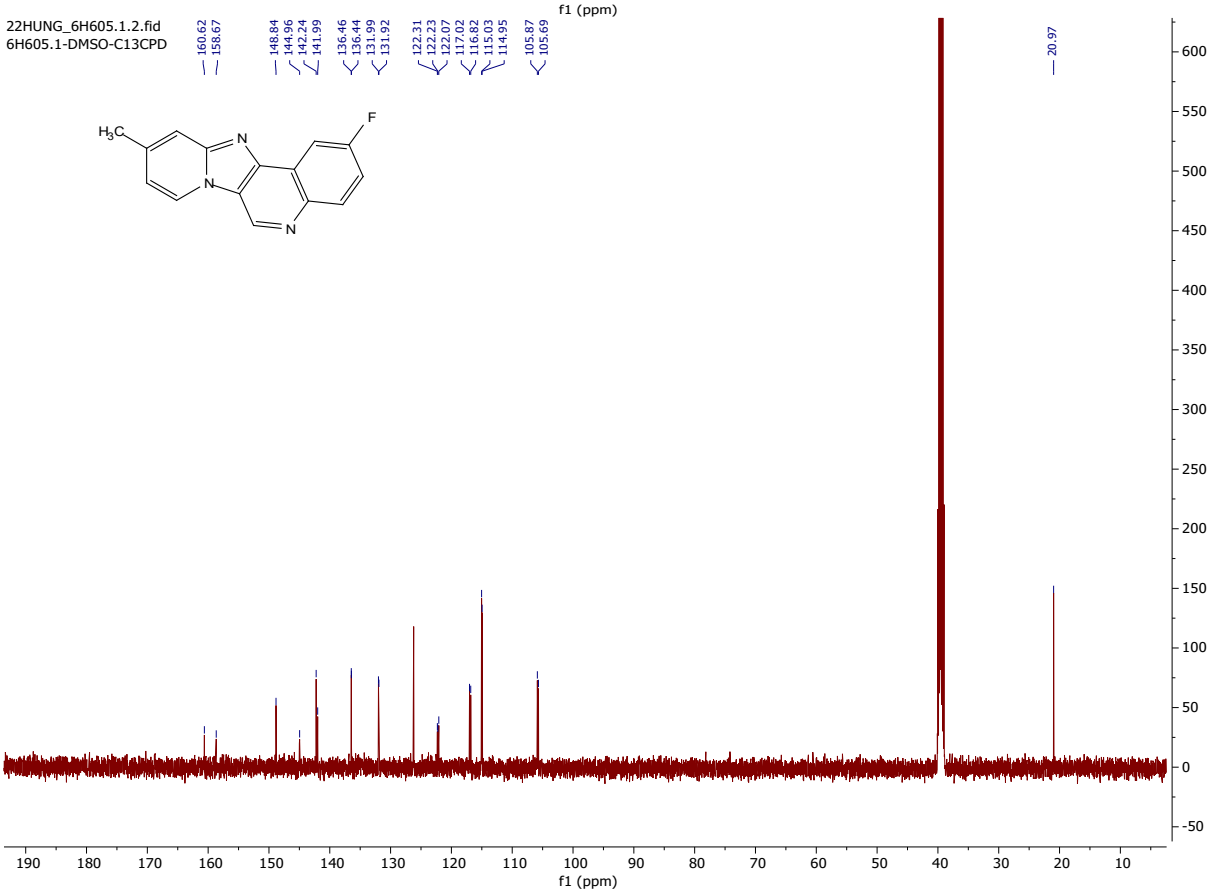

Agilent 6500 Q-TOF MS
File: D:\\10-2025\6H600
Base-Peak Amplitude: 13.0419 Scans: 1 Positive Ions Internal Calibration
23-OCT-2025 16:26:11
286.3575
288.0091
278.8935
223.5691
218.7401
284.6995
228.8885
293.7058
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
220
230
240
250
260
270
280
290
300
Mass/Charge

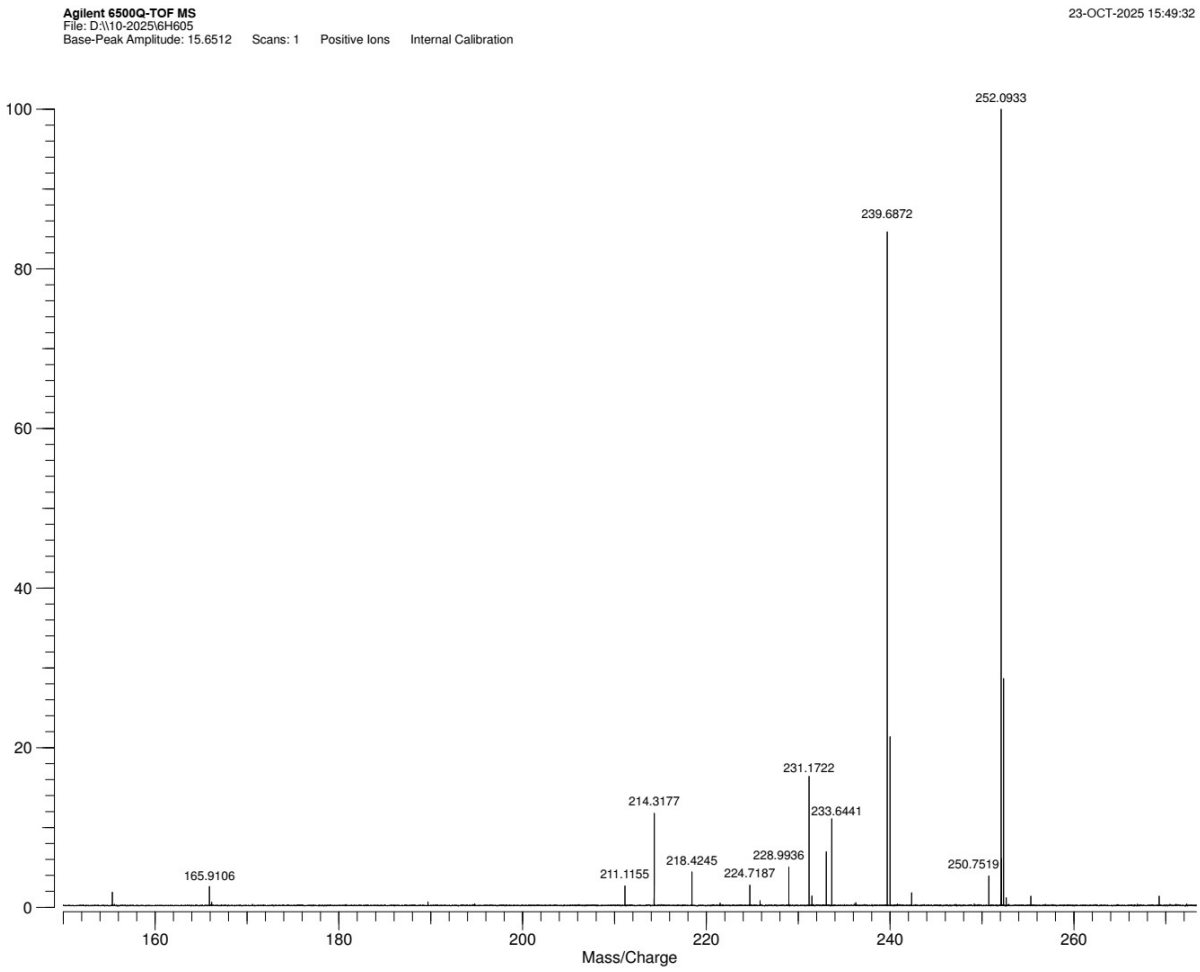
10-chloro-2-fluoropyrido[2',1':2,3]imidazo[4,5-c]quinoline 4k



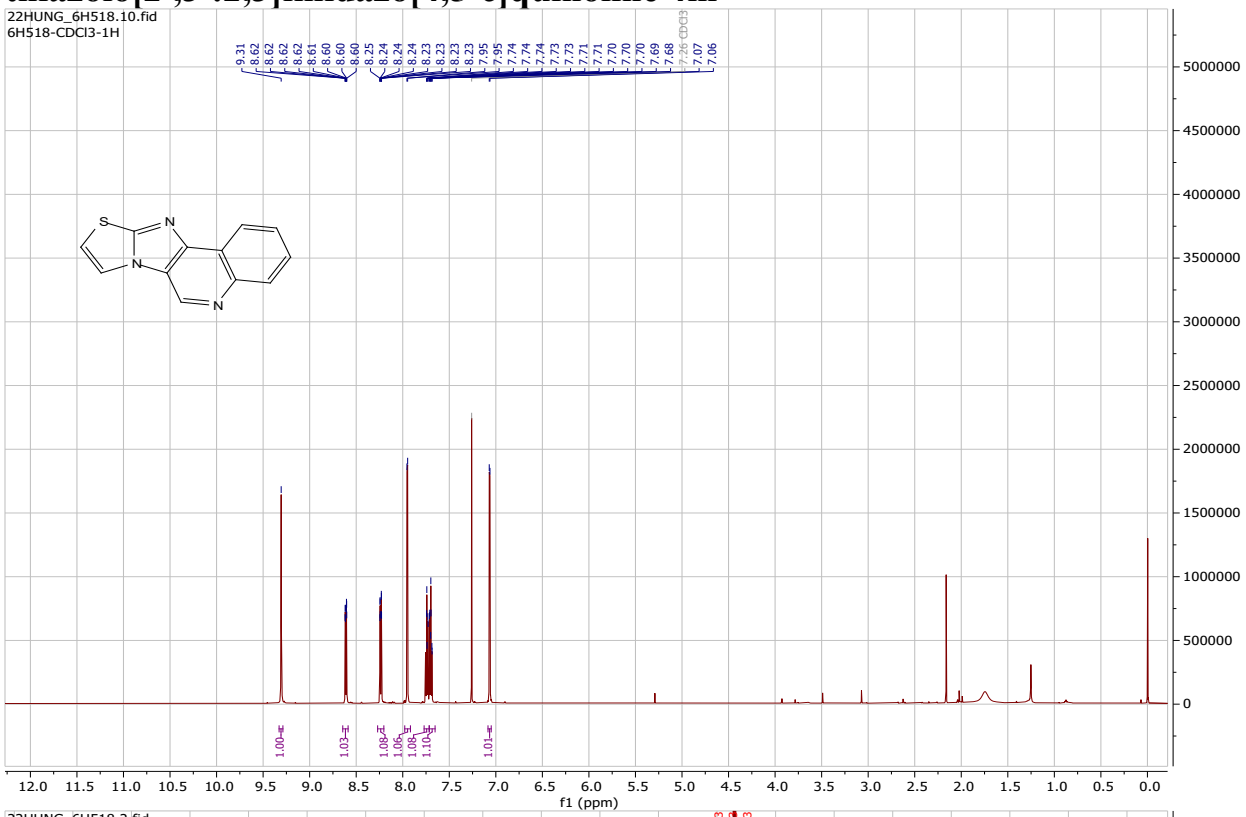
2-fluoro-10-methylpyrido[2',1':2,3]imidazo[4,5-c]quinoline 4l

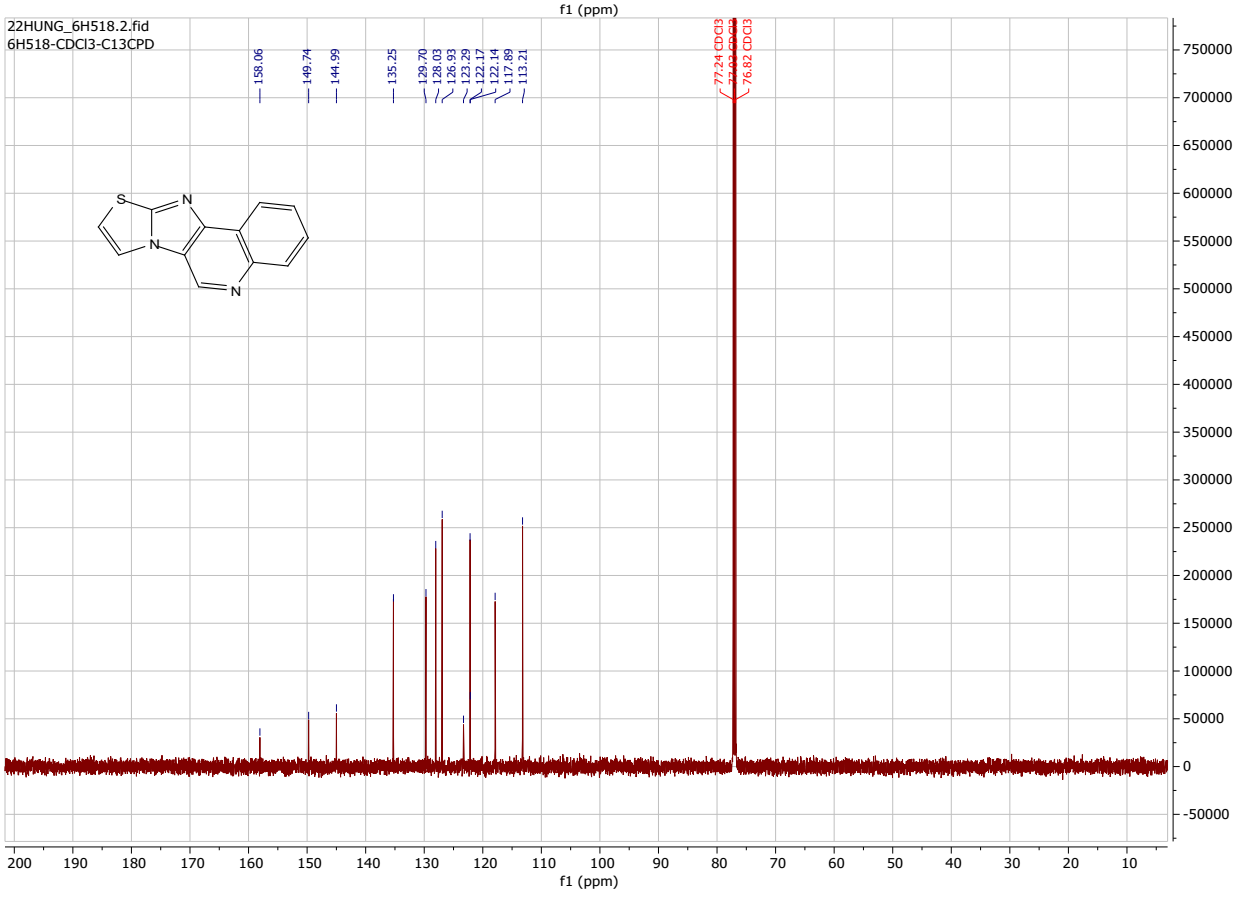


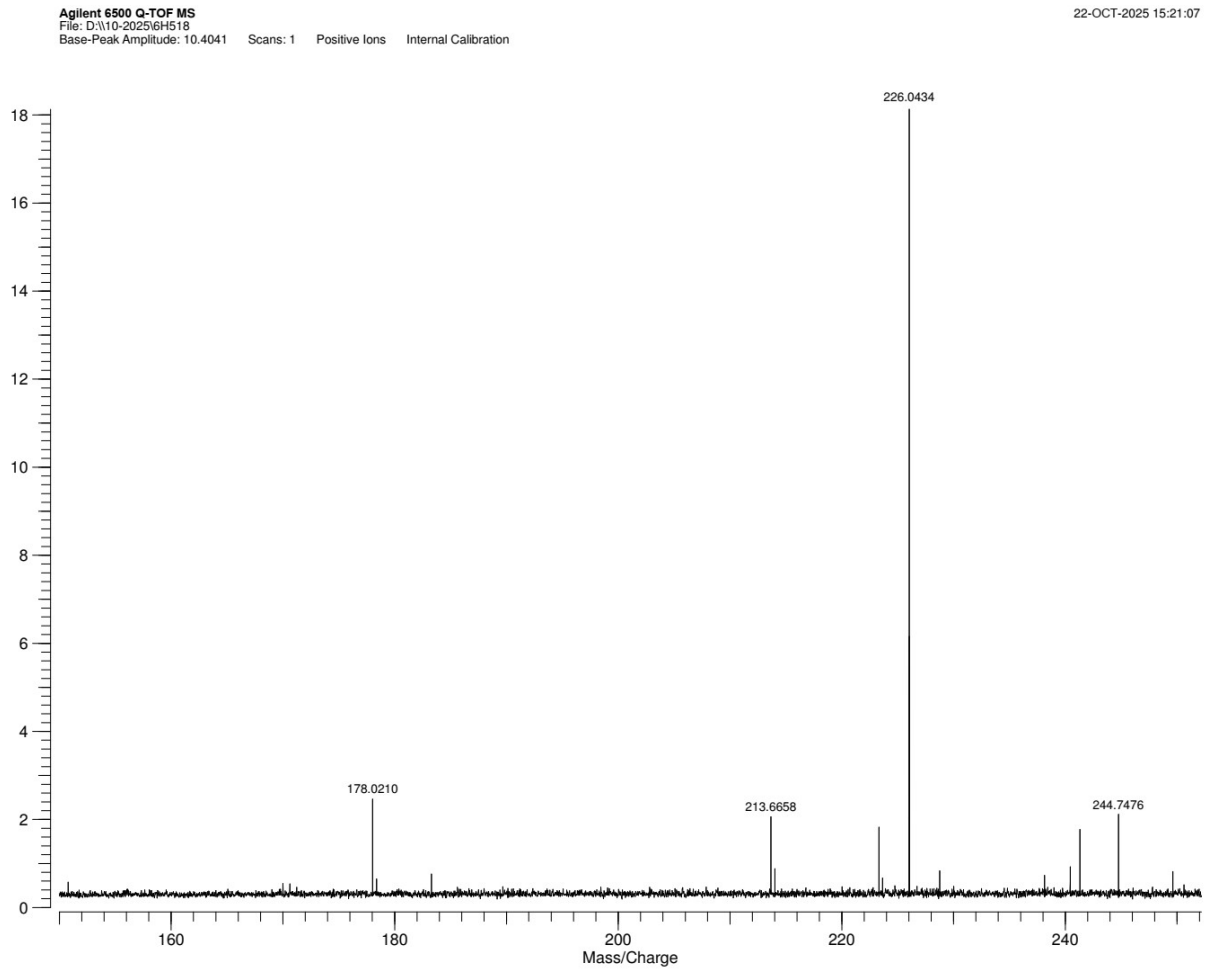



Agilent 6500Q-TOF MS
File: D:\\10-2025\6H605
Base-Peak Amplitude: 15.6512 Scans: 1 Positive Ions Internal Calibration
23-OCT-2025 15:49:32
252.0933
239.6872
231.1722
214.3177
233.6441
228.9936
218.4245
224.7187
211.1155
165.9106
250.7519
100
80
60
40
20
0
160
180
200
220
240
260
Mass/Charge

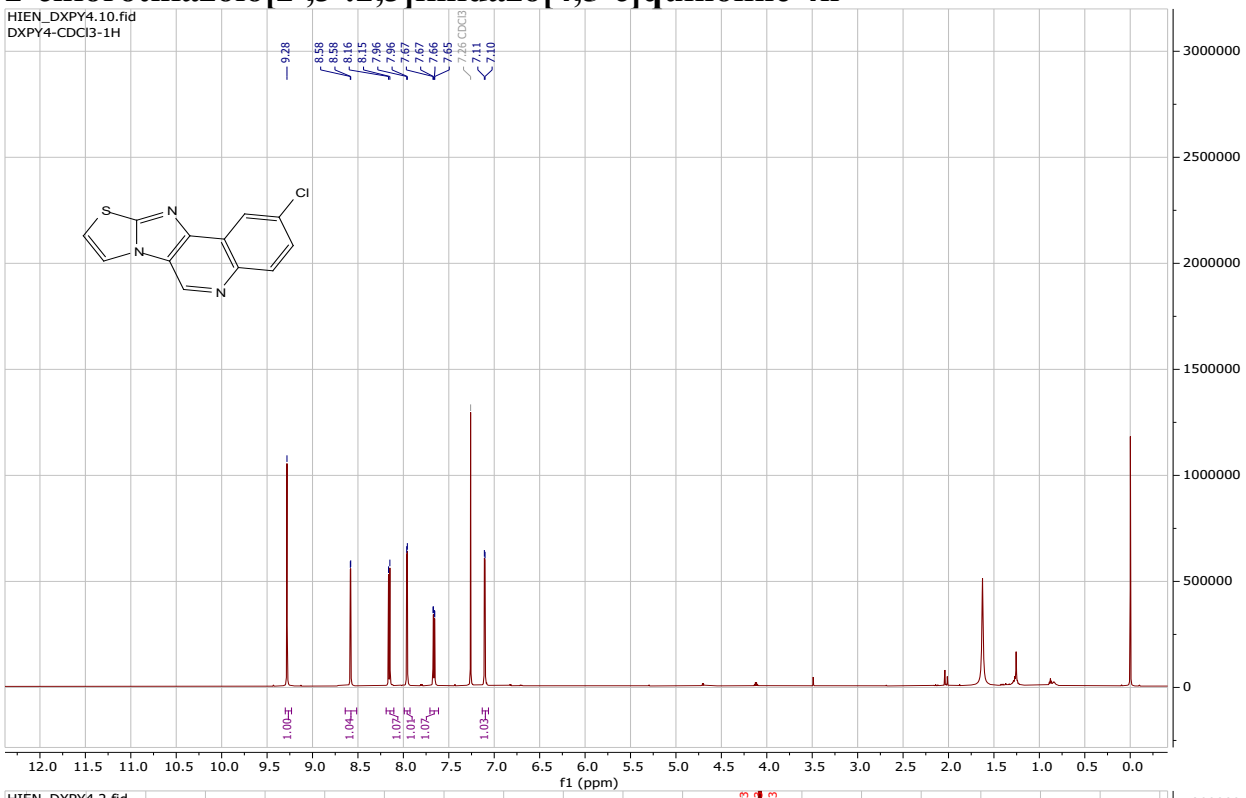
thiazolo[2',3':2,3]imidazo[4,5-c]quinoline 4m

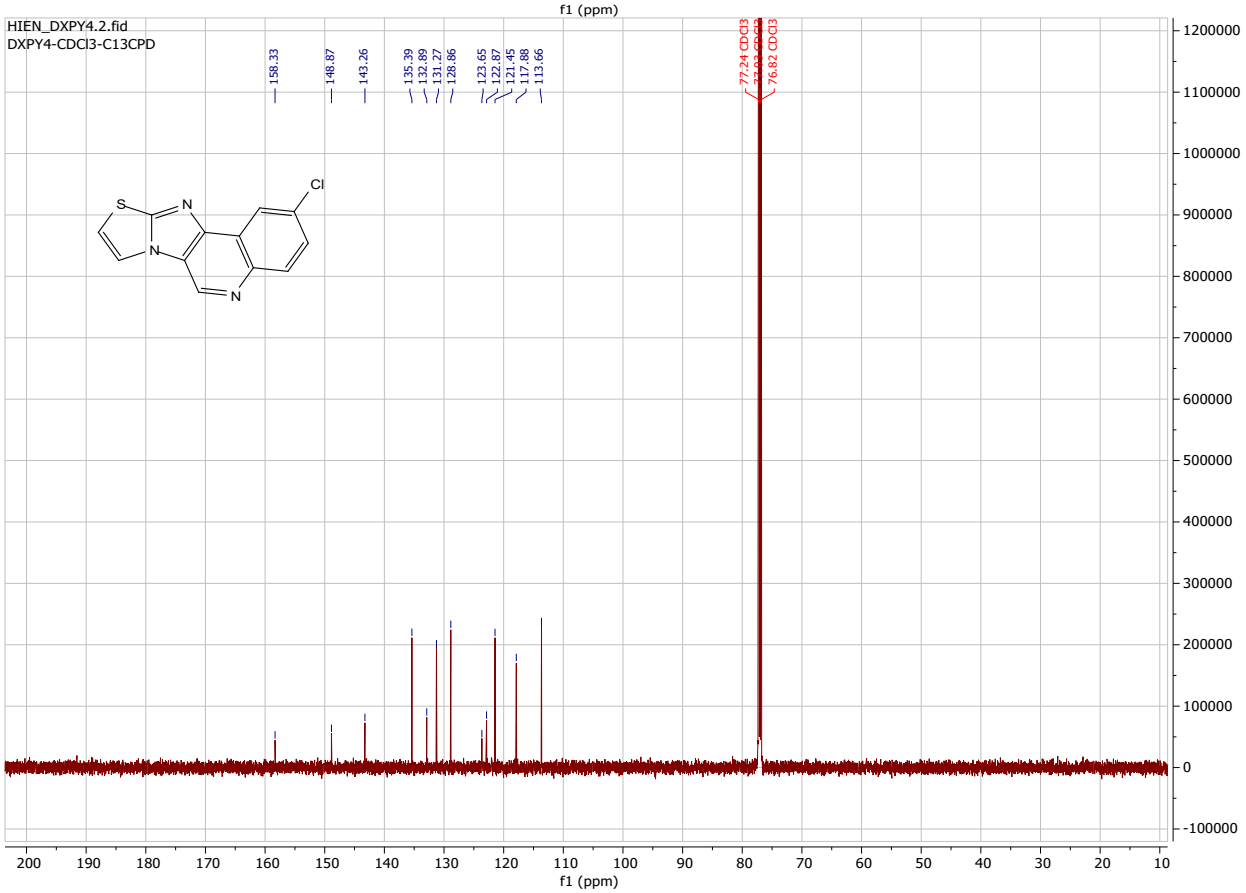




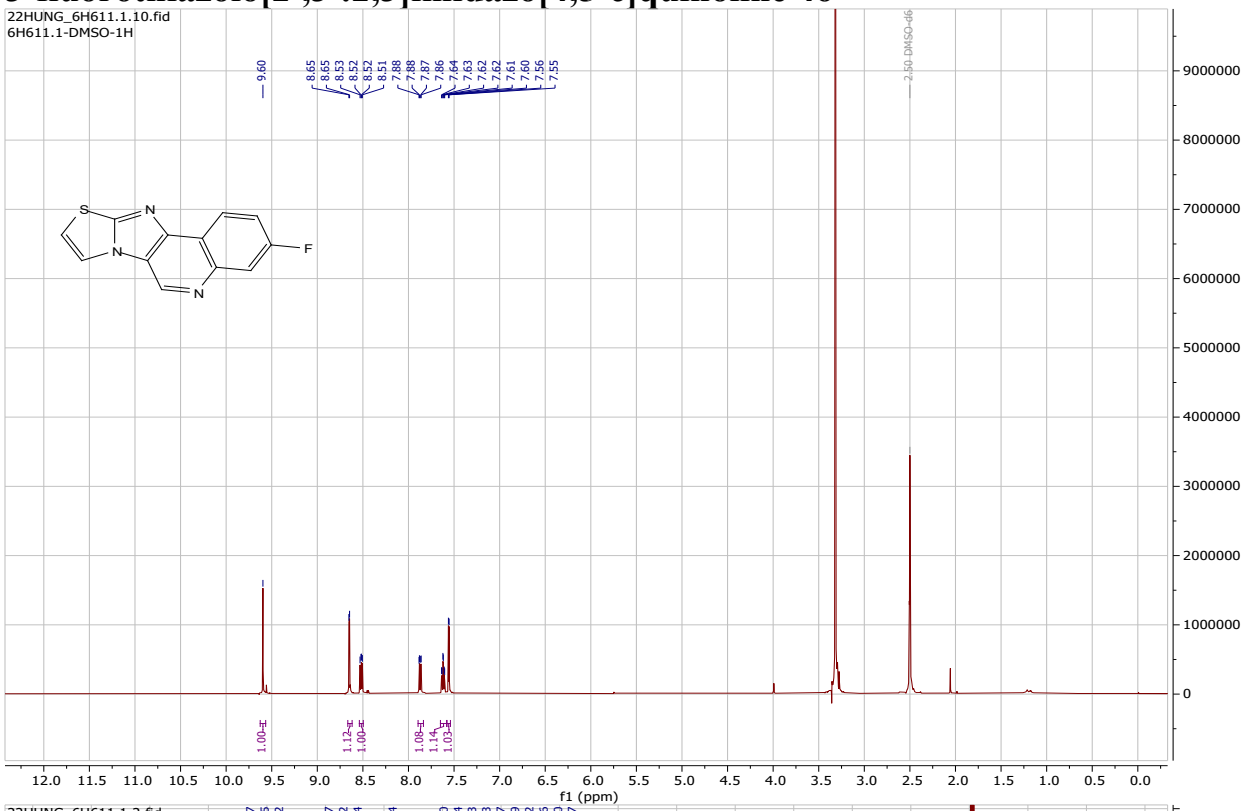

Agilent 6500 Q-TOF MS
File: D:\\10-2025\6H518
Base-Peak Amplitude: 10.4041 Scans: 1 Positive Ions Internal Calibration
22-OCT-2025 15:21:07
226.0434
178.0210
213.6658
244.7476
Mass/Charge

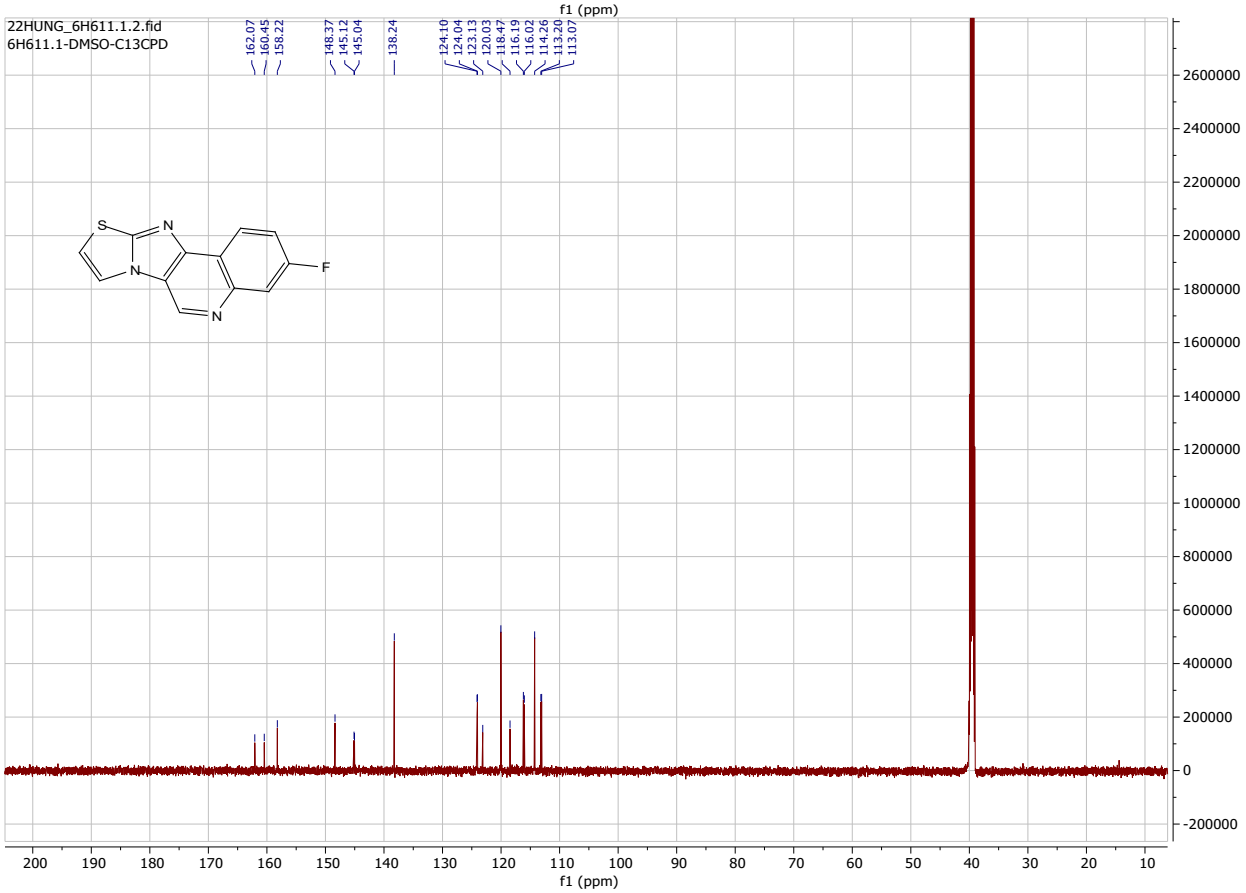
2-chlorothiazolo[2',3':2,3]imidazo[4,5-c]quinoline 4n

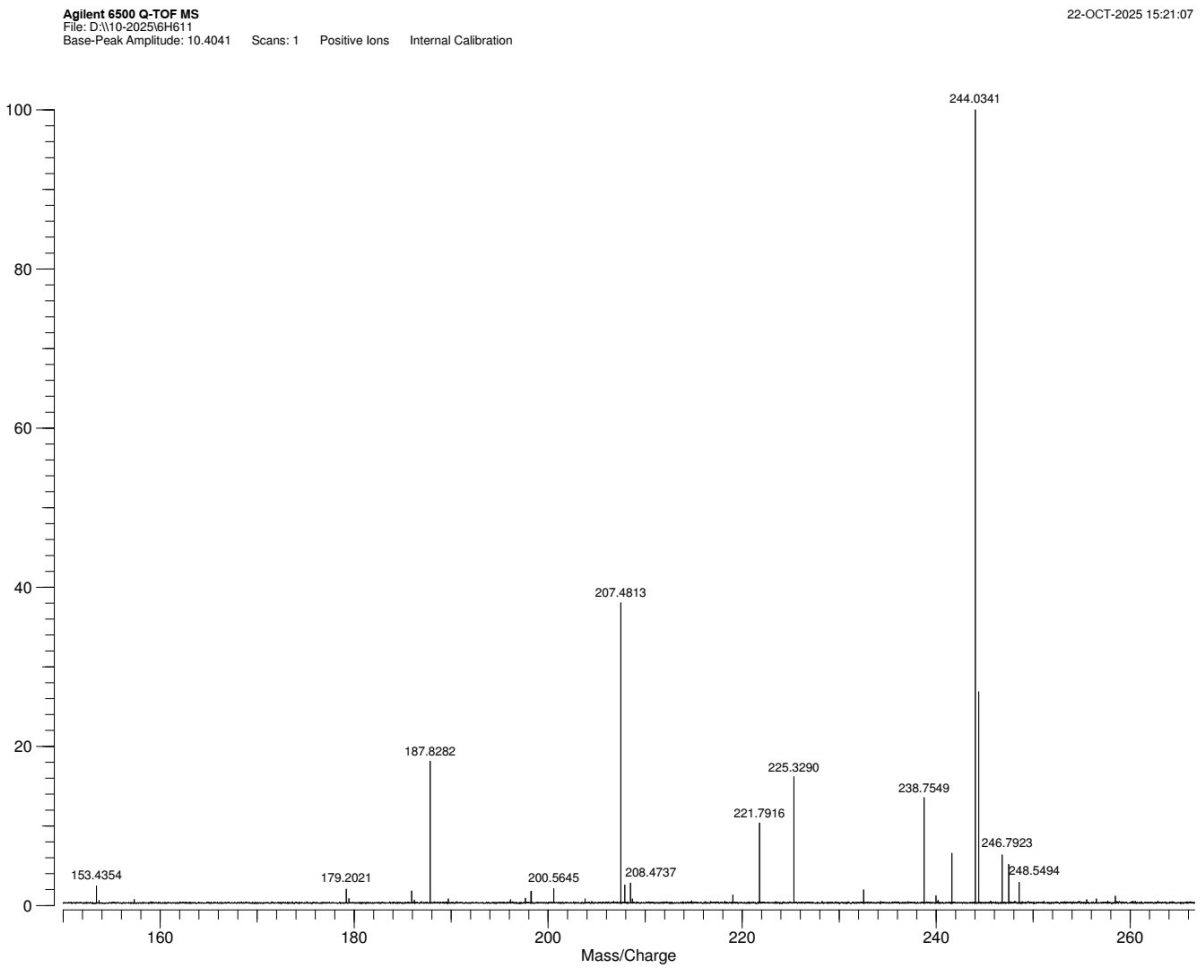




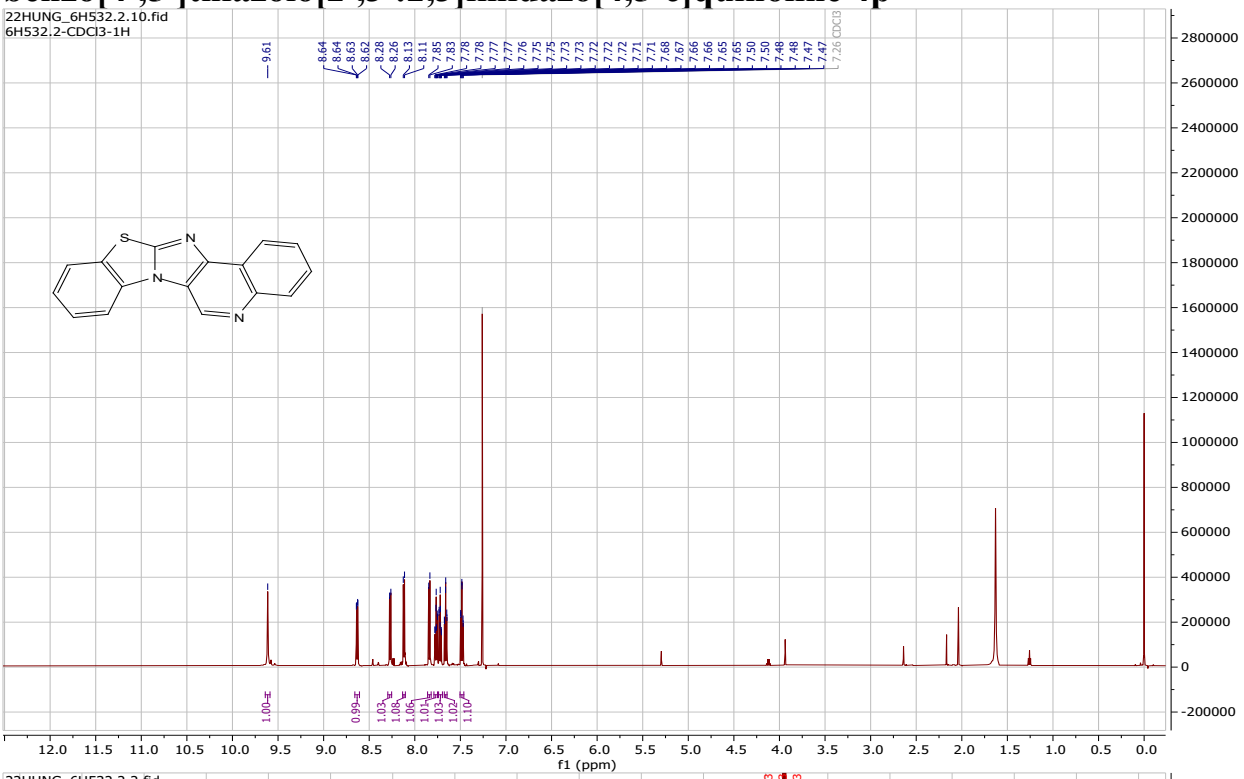
3-fluorothiazolo[2',3':2,3]imidazo[4,5-c]quinoline 4o

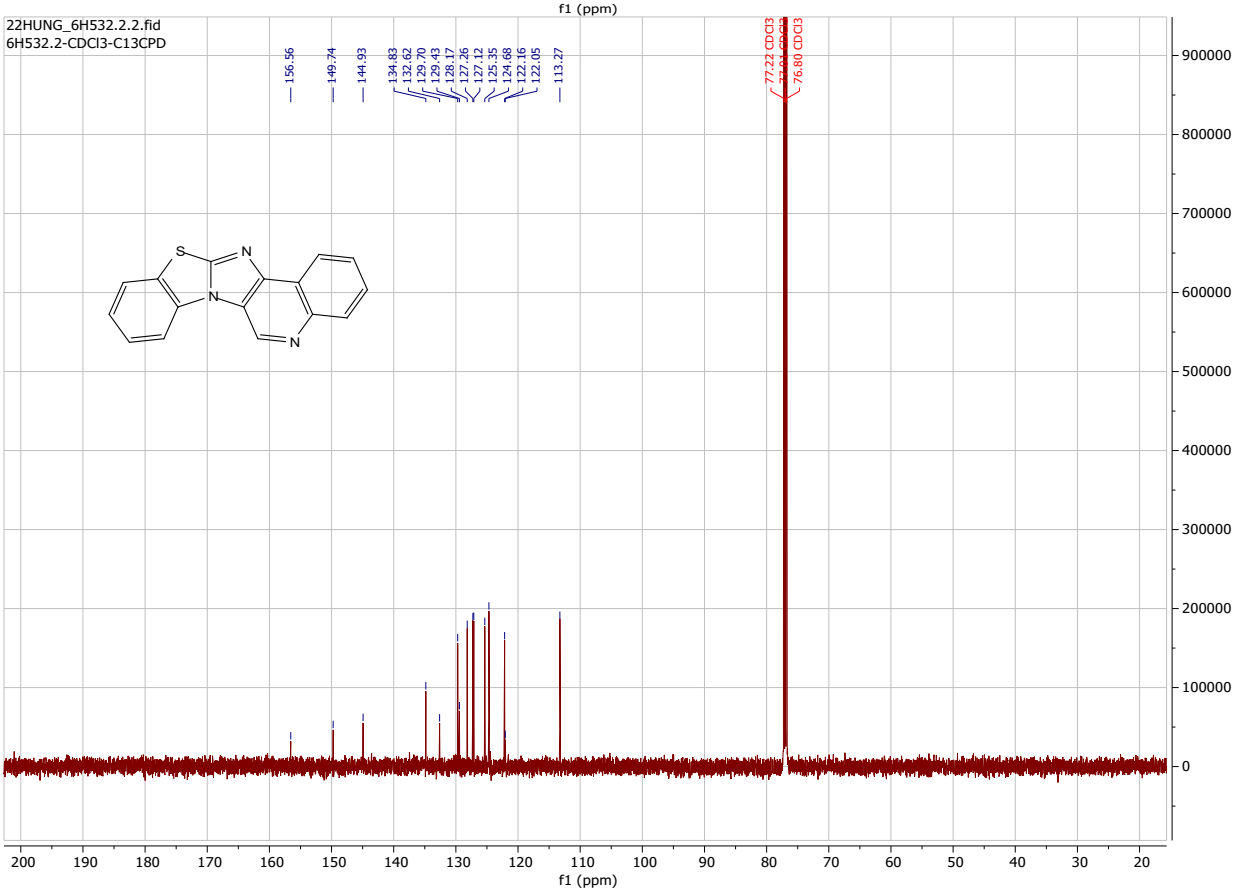


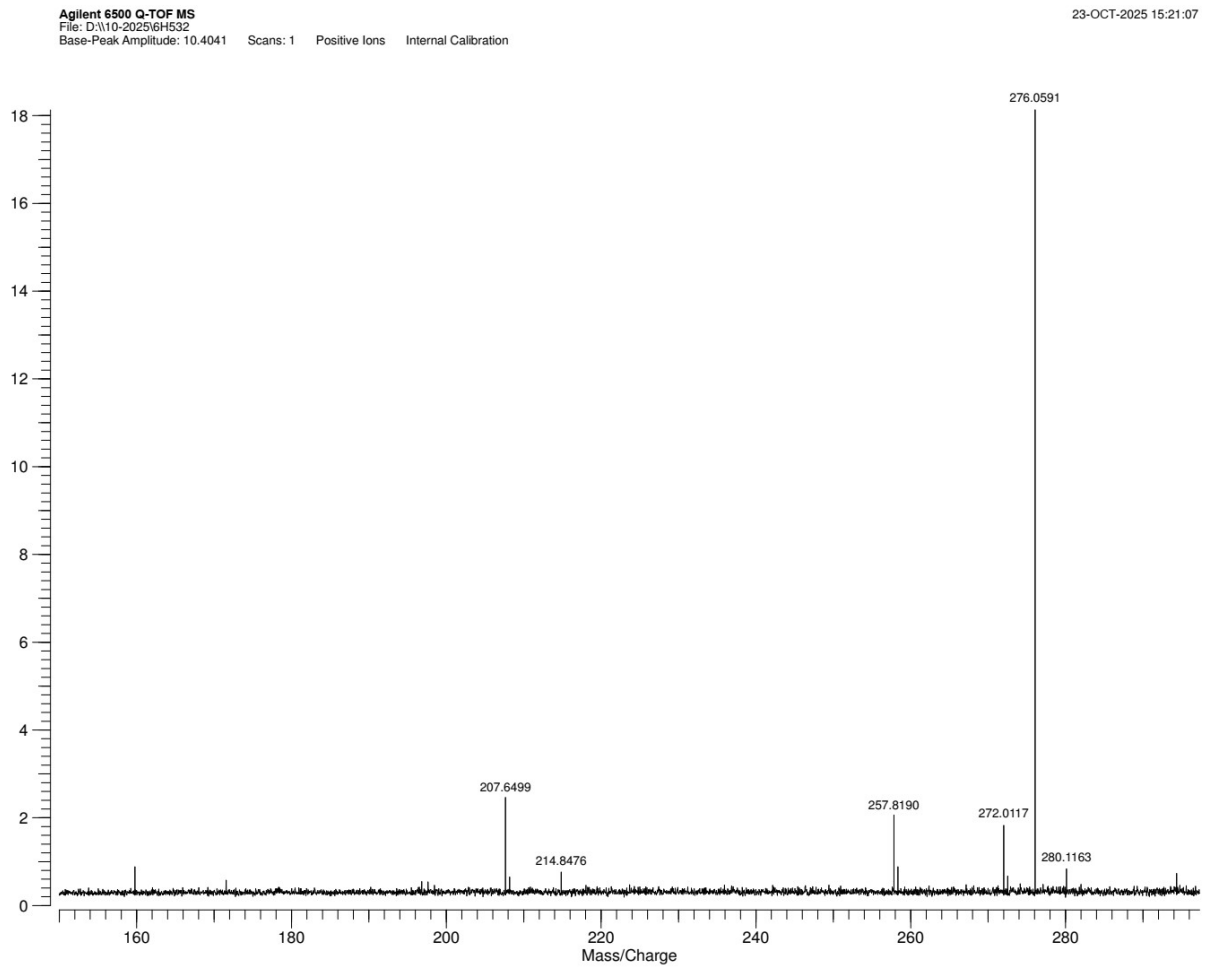



Agilent 6500 Q-TOF MS
File: D:\\10-2025\6H611
Base-Peak Amplitude: 10.4041 Scans: 1 Positive Ions Internal Calibration
22-OCT-2025 15:21:07
244.0341
207.4813
187.8282
225.3290
238.7549
221.7916
246.7923
248.5494
208.4737
153.4354
179.2021
200.5645
100
80
60
40
20
0
160
180
200
220
240
260
Mass/Charge

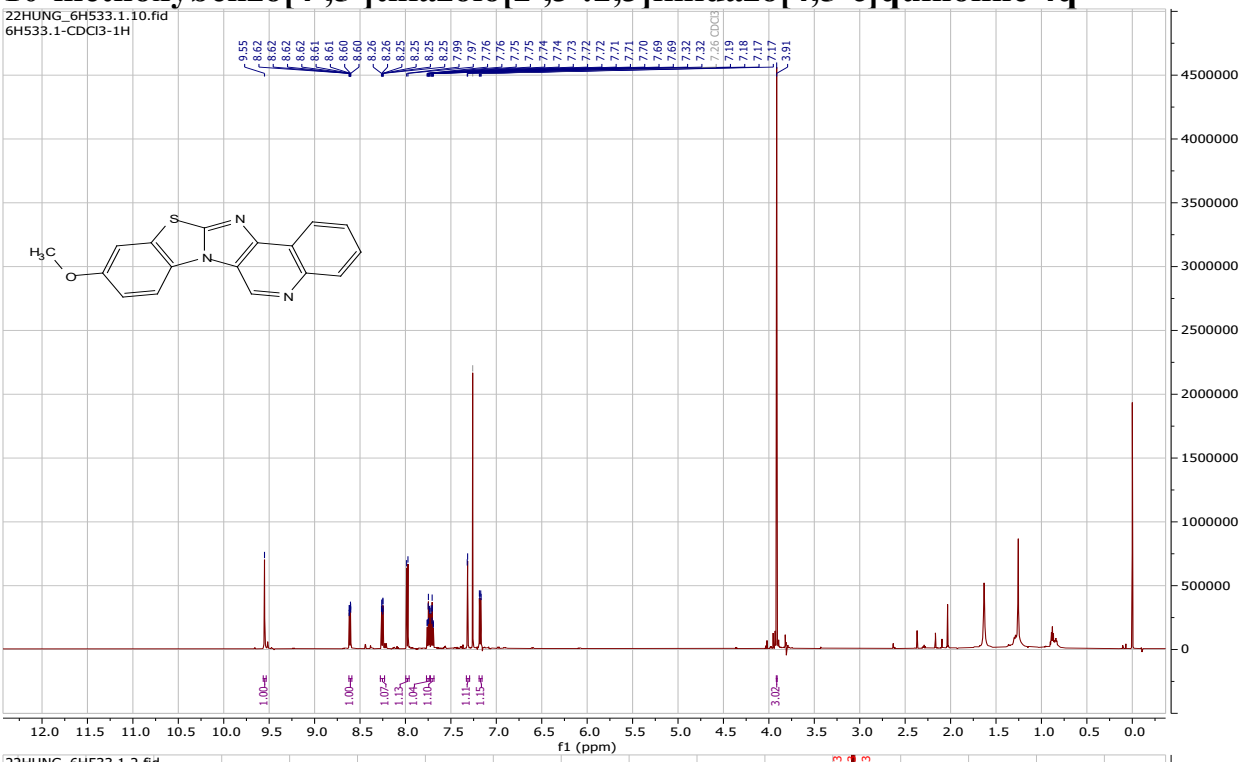
benzo[4',5']thiazolo[2',3':2,3]imidazo[4,5-c]quinoline 4p

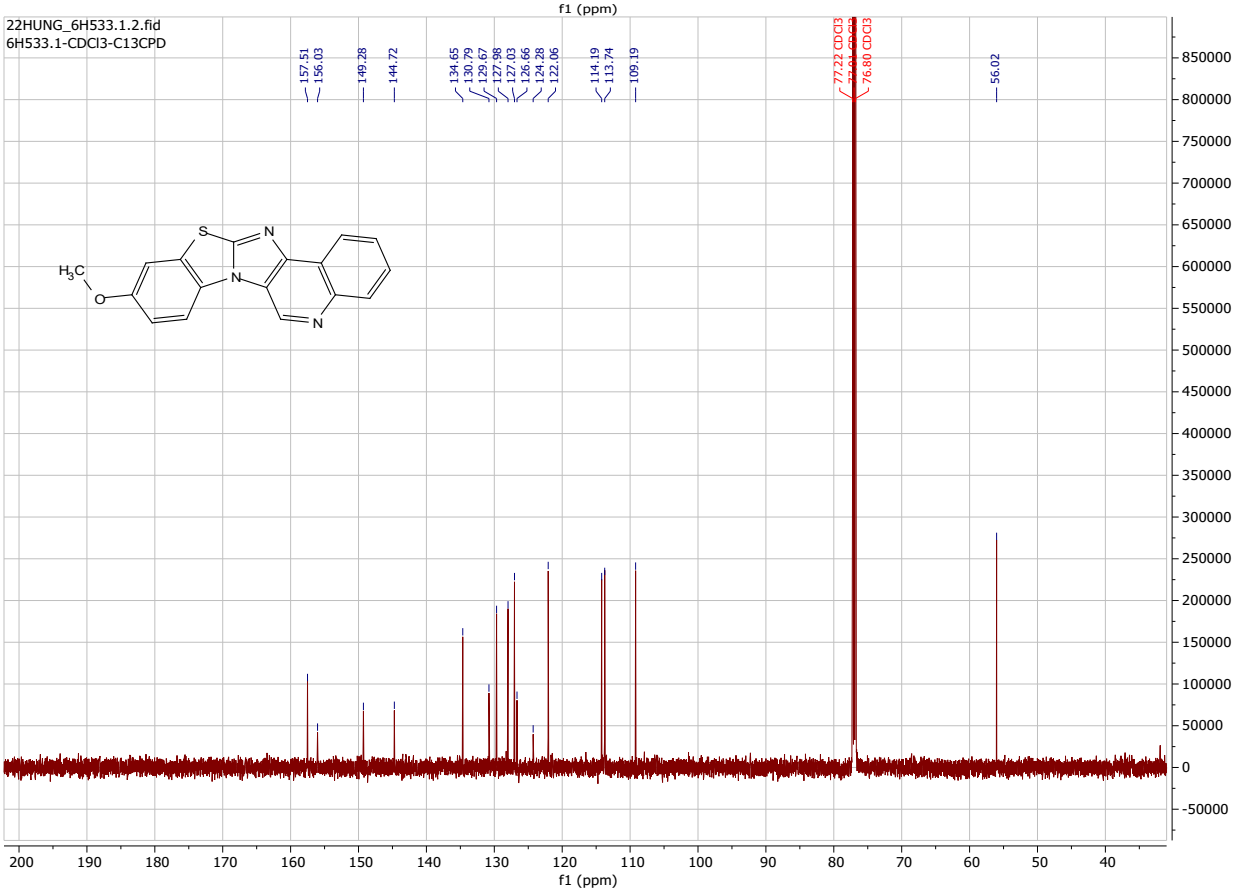


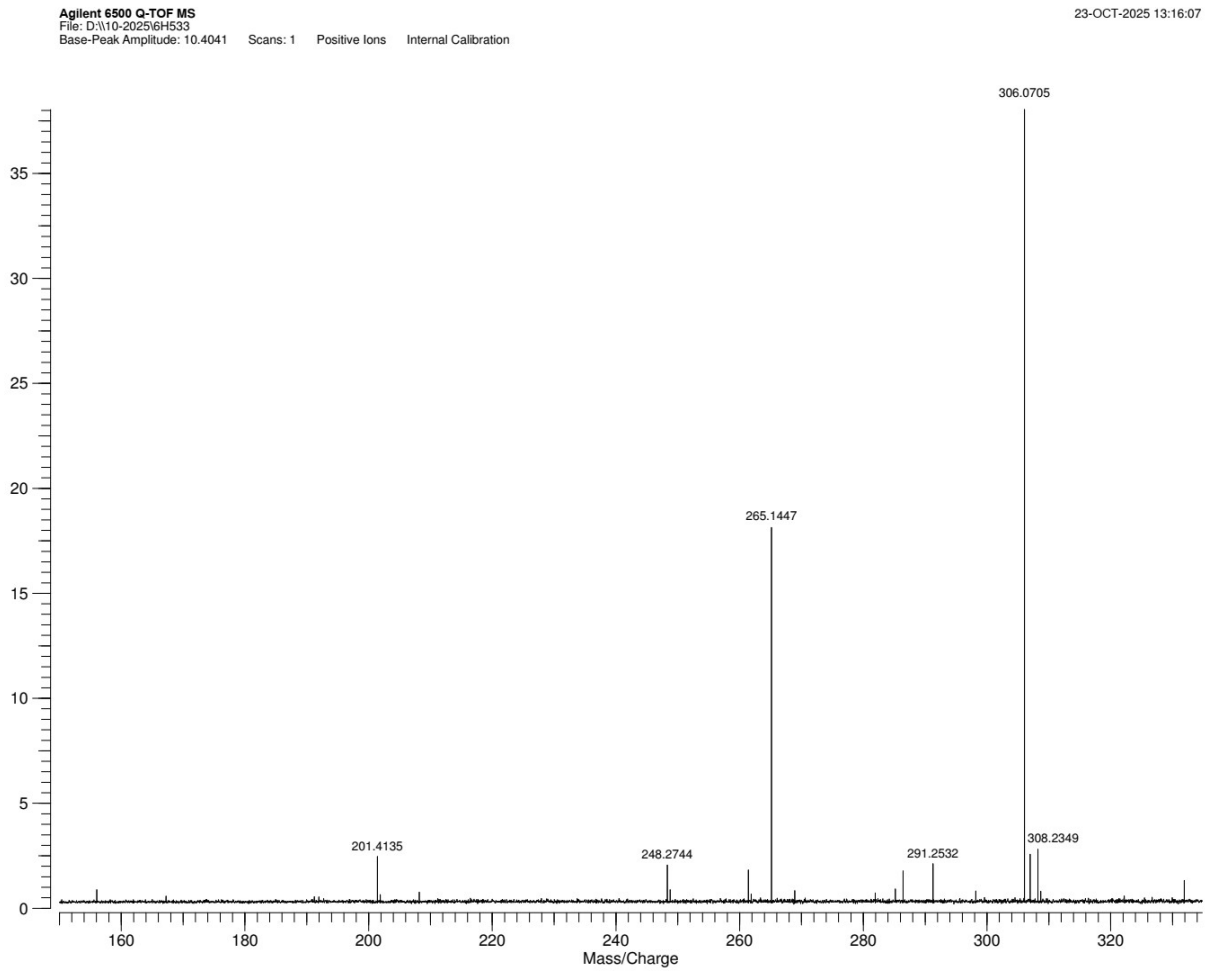



Agilent 6500 Q-TOF MS
File: D:\\10-2025\6H532
Base-Peak Amplitude: 10.4041 Scans: 1 Positive Ions Internal Calibration
23-OCT-2025 15:21:07
276.0591
207.6499
214.8476
257.8190
272.0117
280.1163
Mass/Charge

10-methoxybenzo[4',5']thiazolo[2',3':2,3]imidazo[4,5-c]quinoline 4q



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23-OCT-2025 13:16:07
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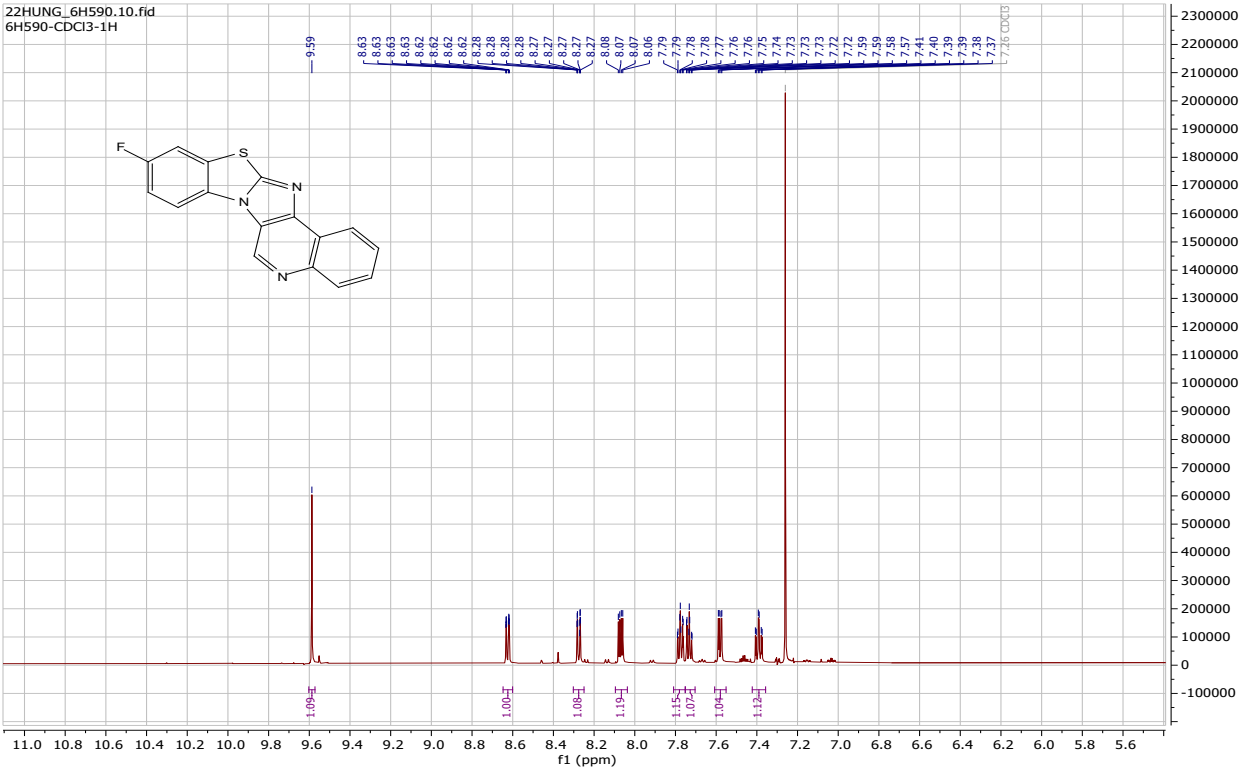
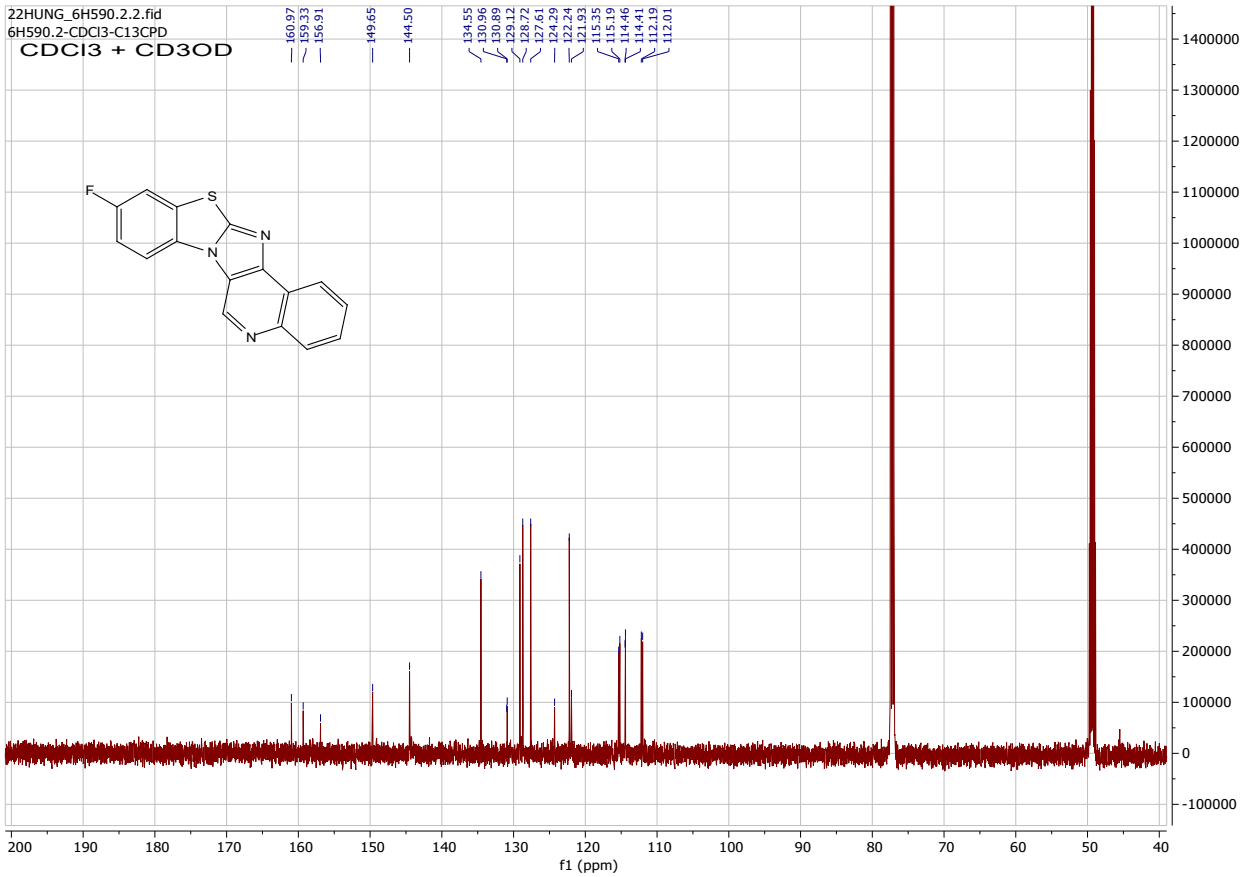
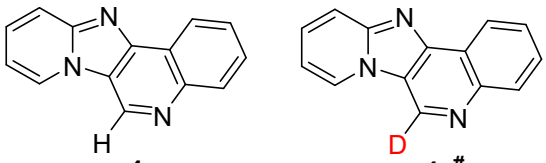
265.1447
201.4135
248.2744
291.2532
308.2349
35
30
25
20
15
10
5
0
160
180
200
220
240
260
280
300
320
Mass/Charge

10-fluorobenzo[4',5']thiazolo[2',3':2,3]imidazo[4,5-c]quinoline 4r

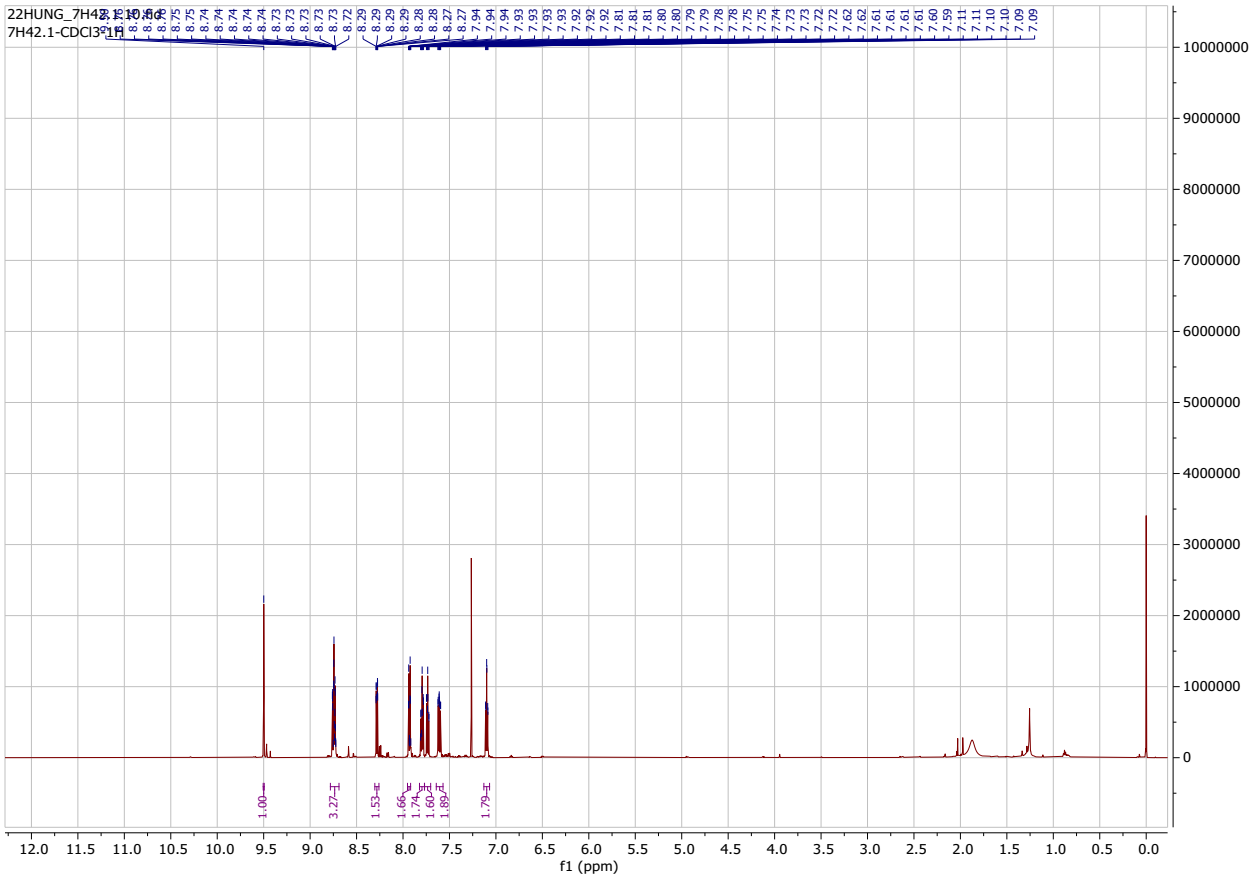






(1:0.8)

pyrido[2',1':2,3]imidazo[4,5-c]quinoline 4a & pyrido[2',1':2,3]imidazo[4,5-c]quinoline-6-d 4a#



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