1.1. Product 5a: 2-amino-7,8-dimethyl-5-oxo-1,4-diphenyl-1,4,5,6,7,8 hexahydroquinoline-3-carbonitrile

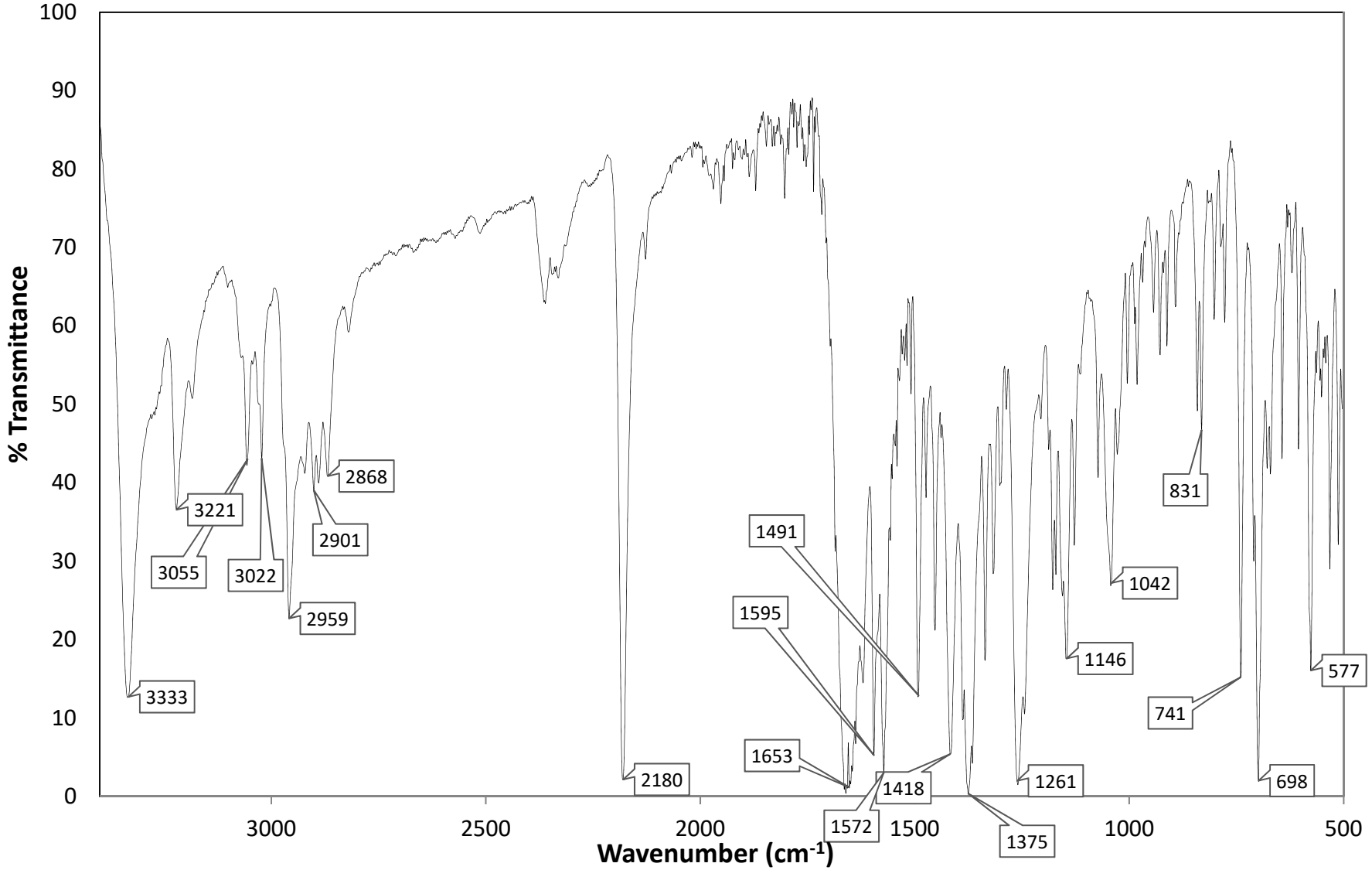


Figure S1 - IR spectrum of 5a

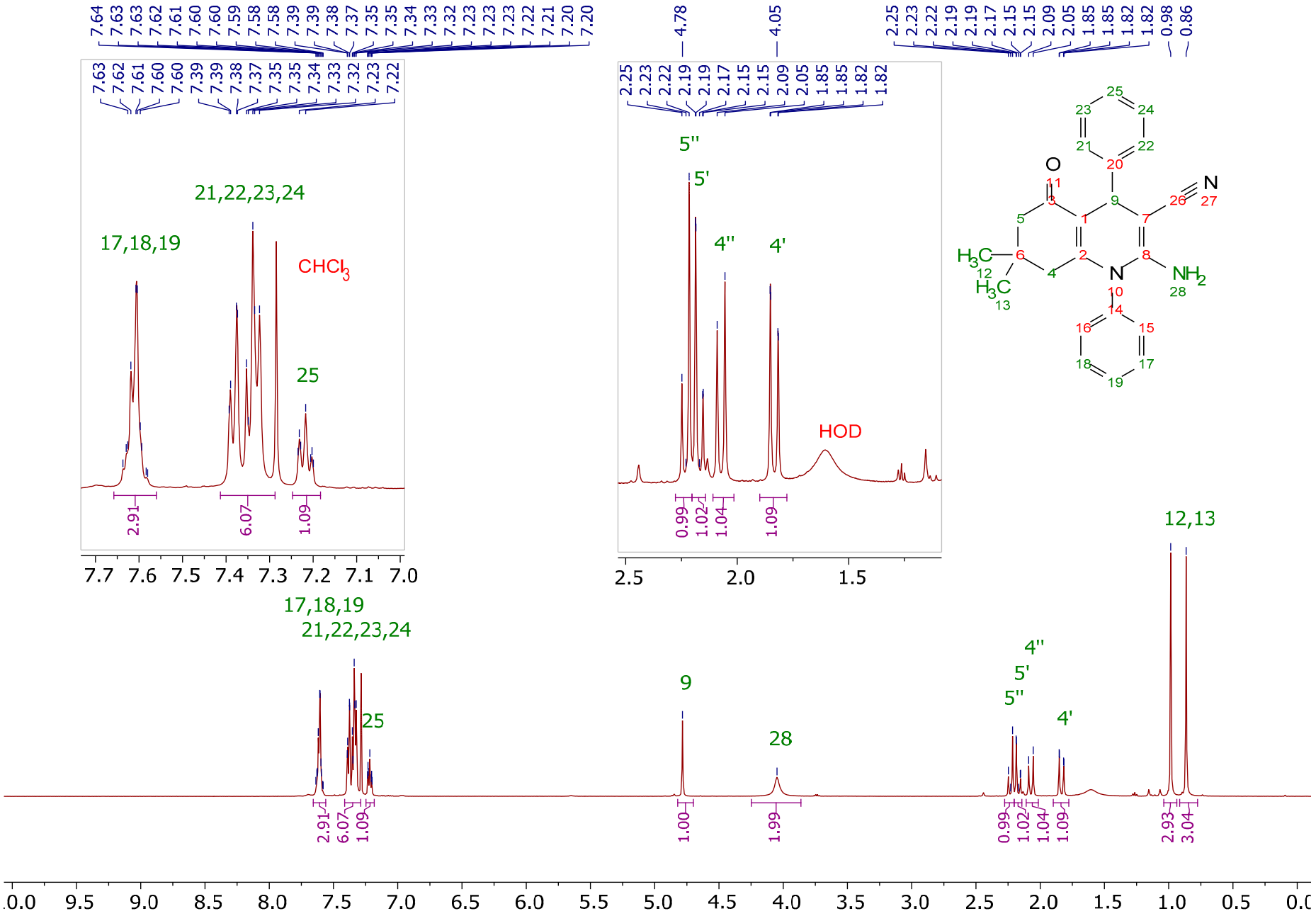


Figure S2 - ^{1}H NMR spectrum of 5a

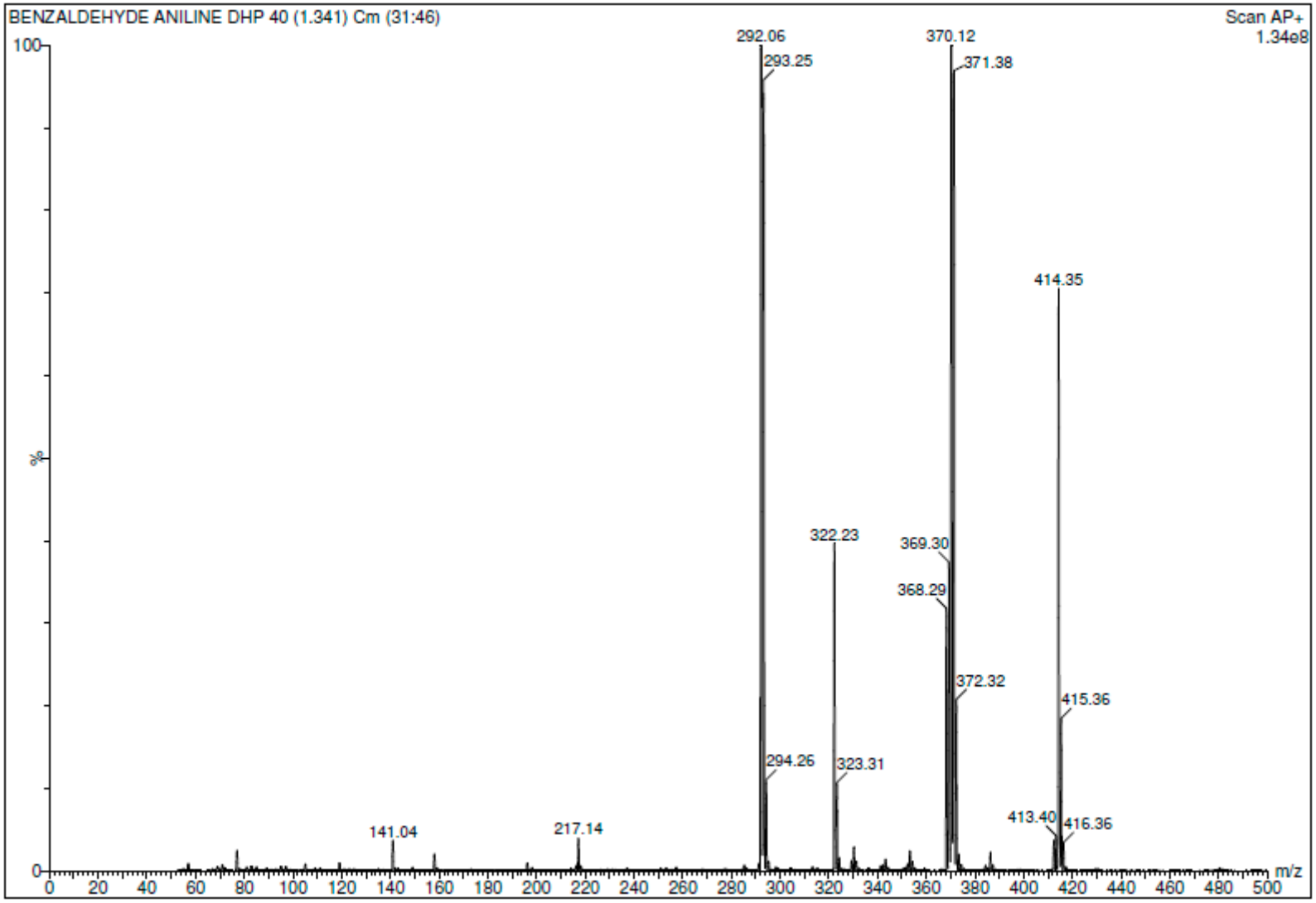
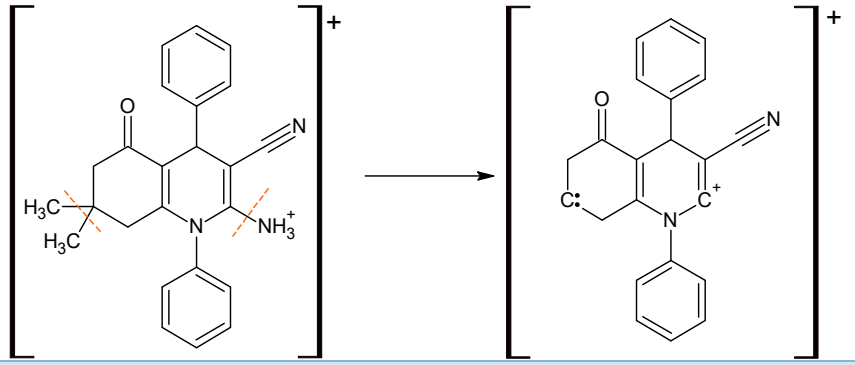
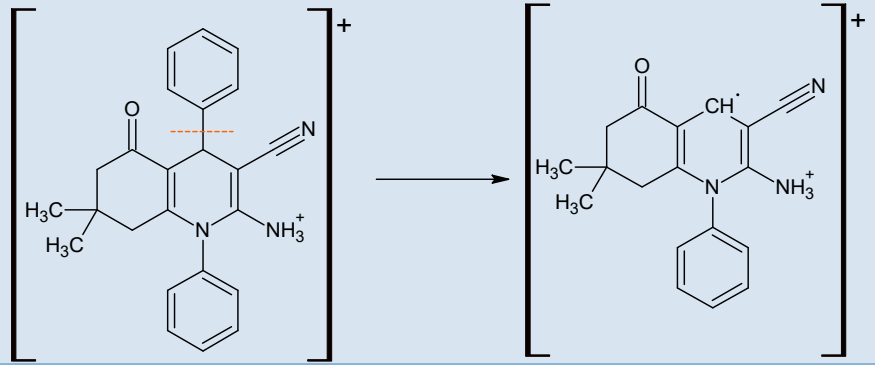
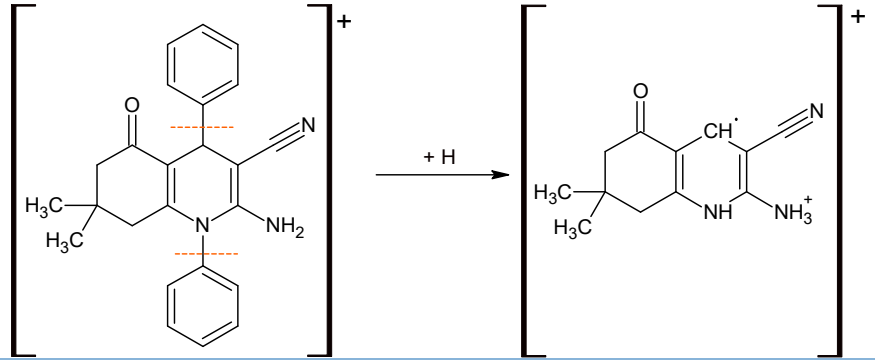


Figure S3 - MS spectrum of 5a

Table S1 - Fragmentation positions for peaks in MS spectrum of 5a

m/z	Fragmentation position and structure
370.12	$[M-H]^+$
323.31	
292.06	
217.14	

1.2. Product 5b: 2-amino-7,8-dimethyl-4-(4-methylphenyl)-5-oxo-1-phenyl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

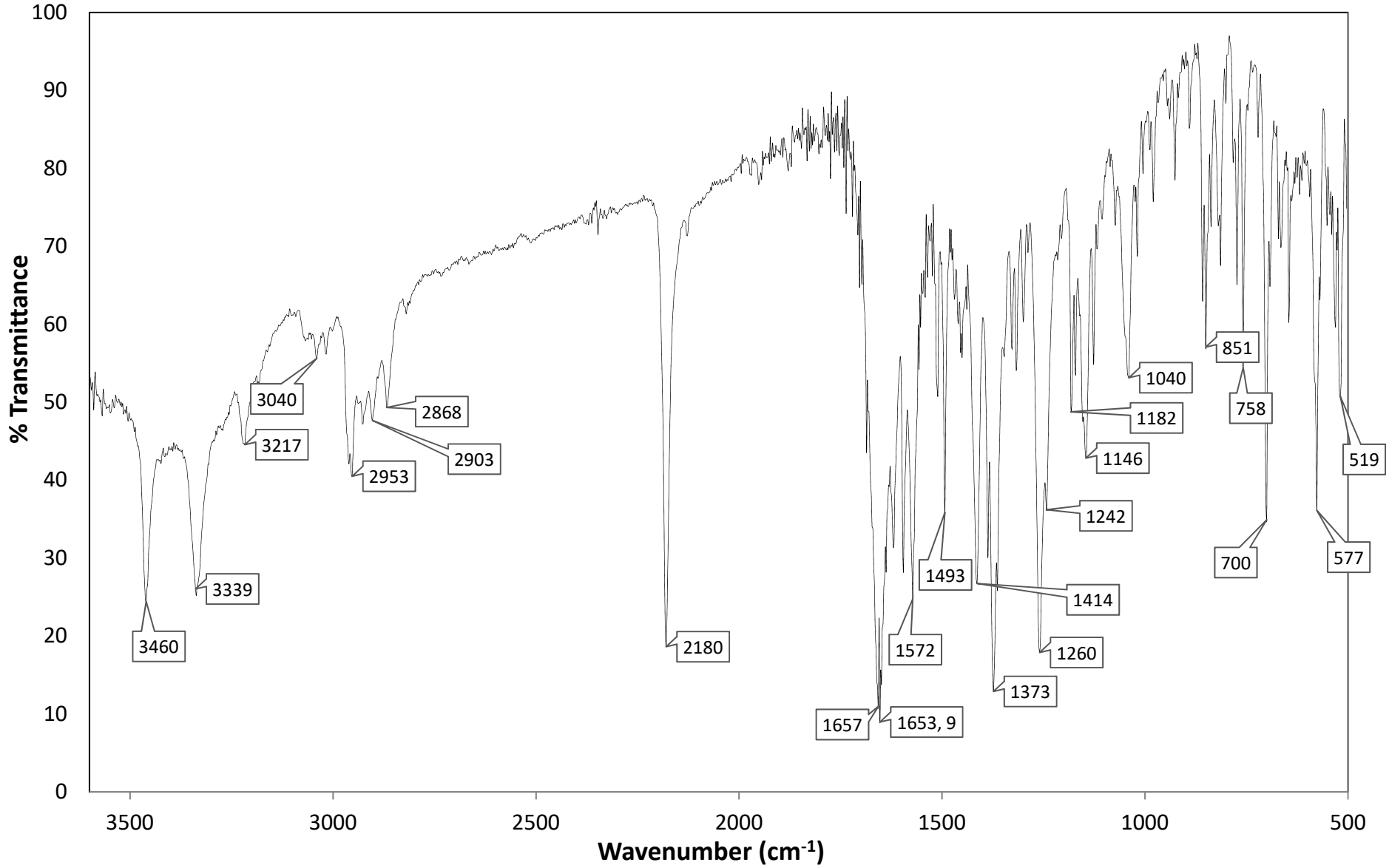


Figure S4 - IR spectrum of 5b

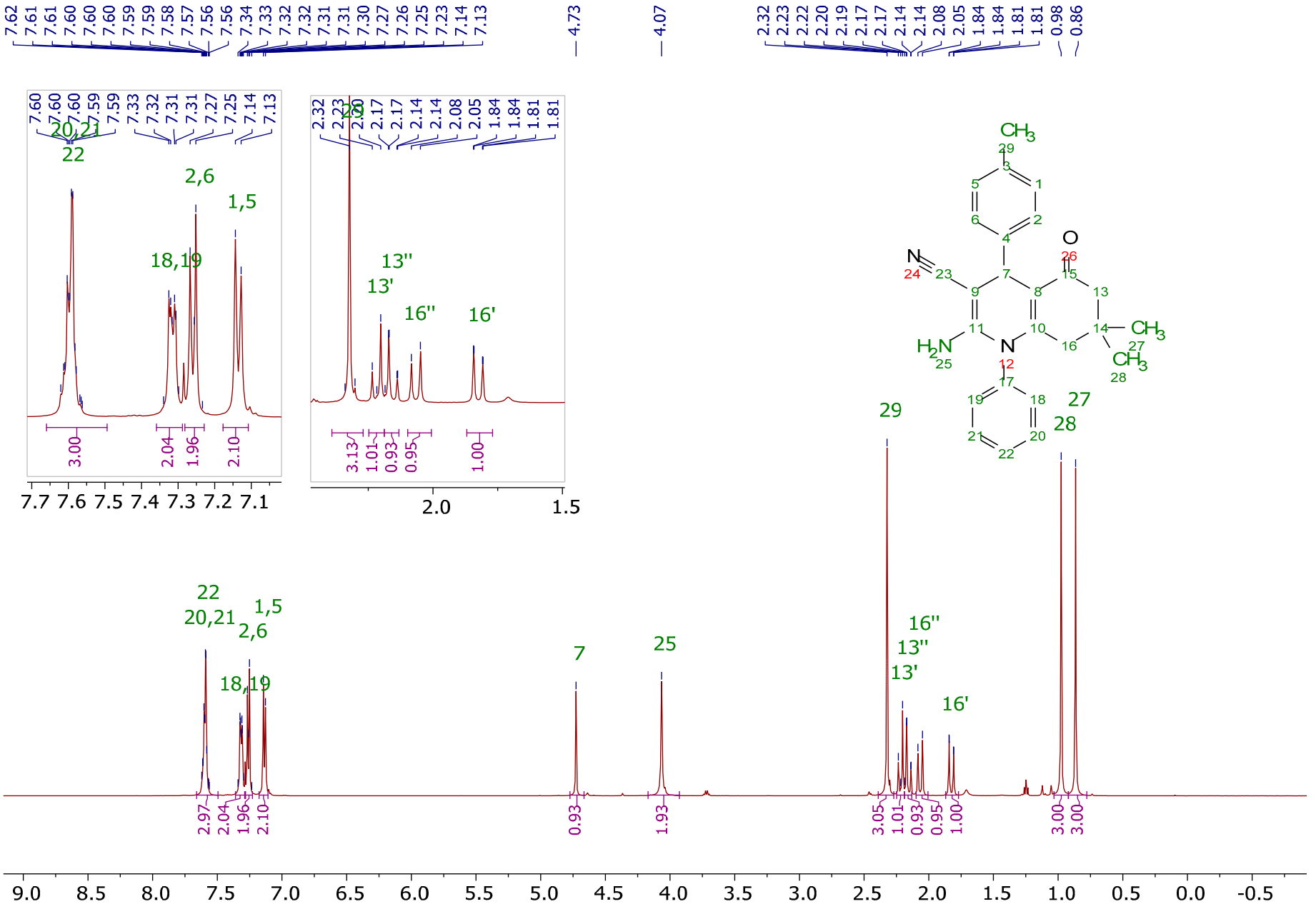


Figure S5 - ^{1}H NMR spectrum of 5b

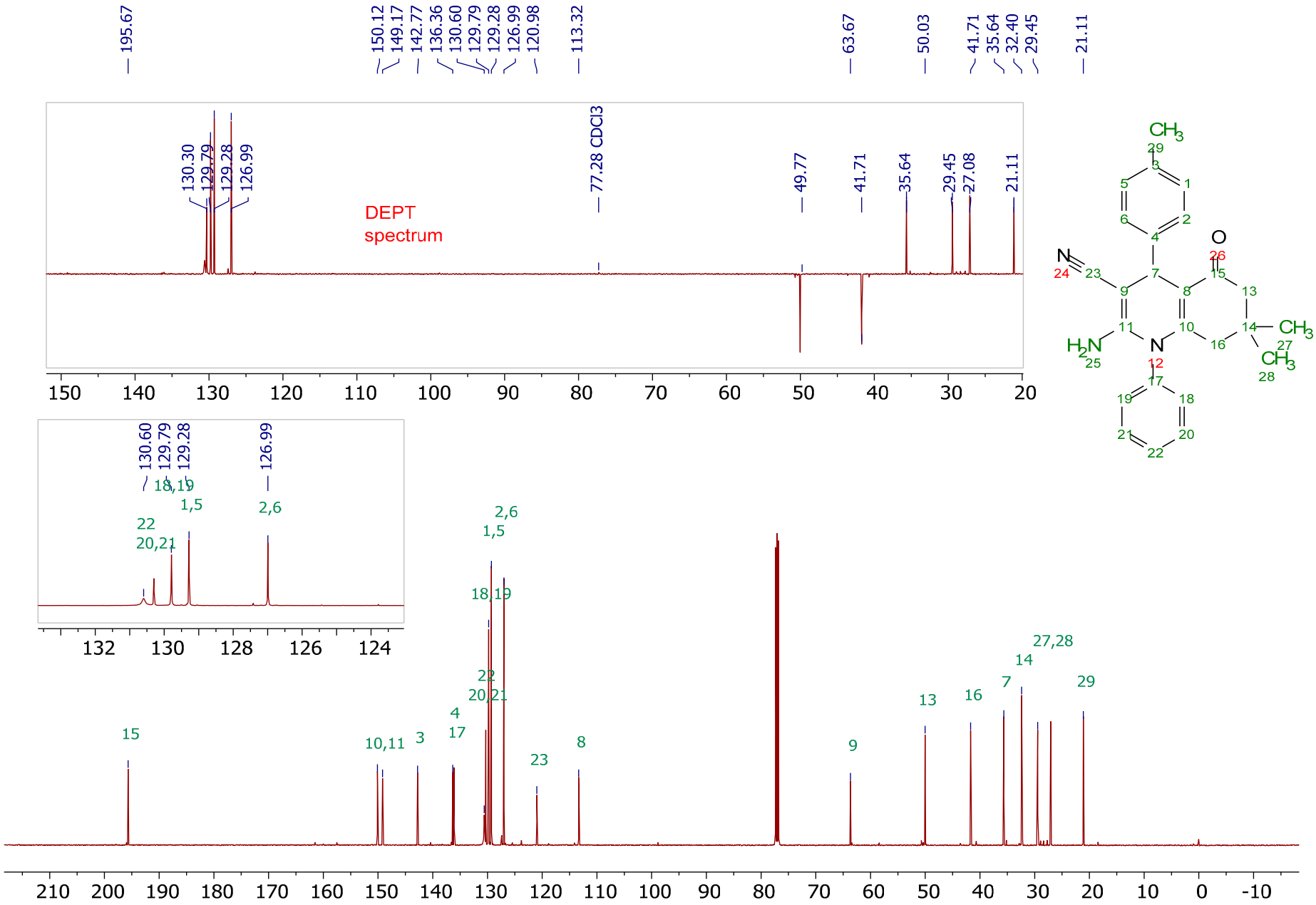


Figure S6 – ^{13}C NMR spectrum of 5b with inset expansion and DEPT spectrum

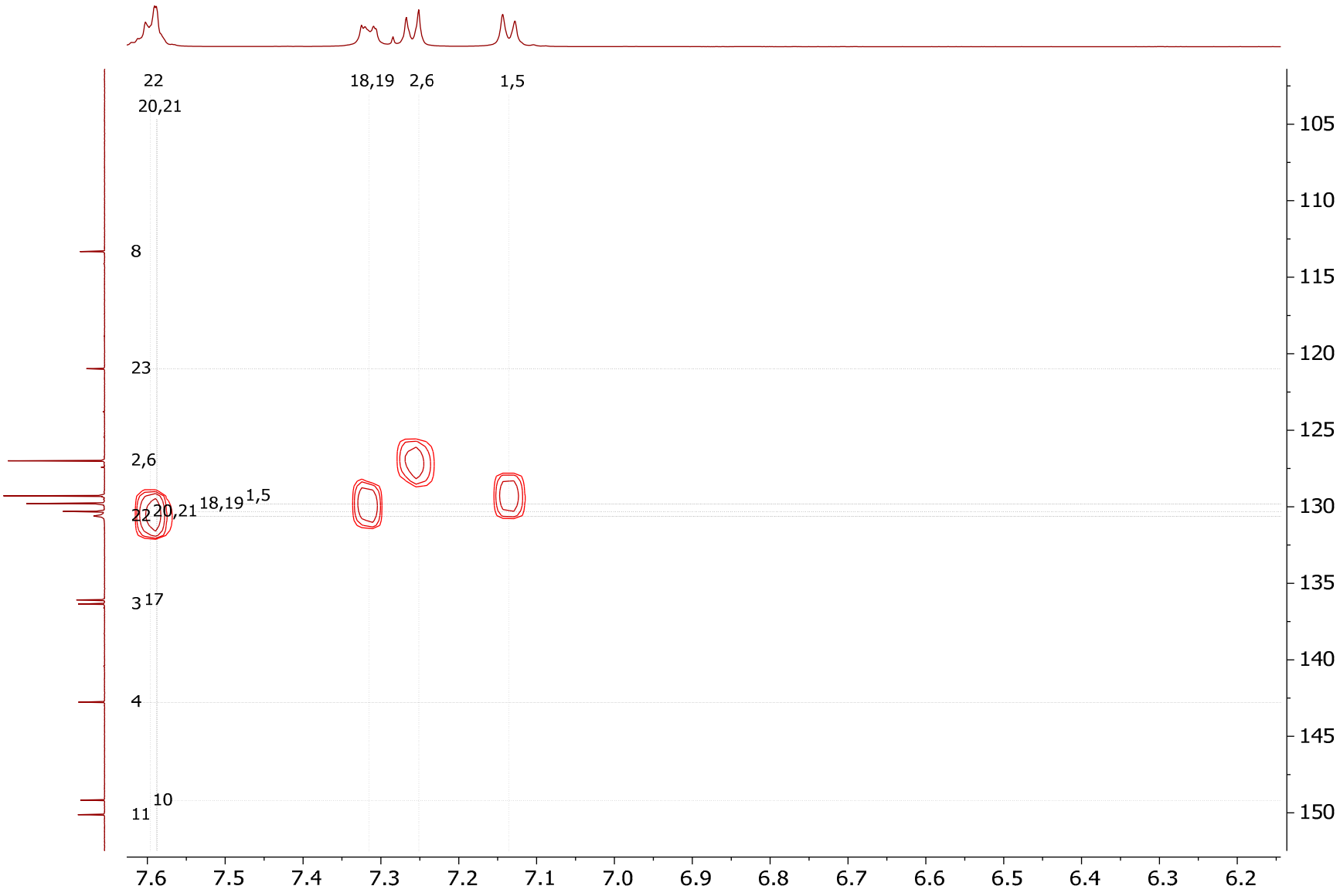


Figure S7 - HSQC spectrum of 5b

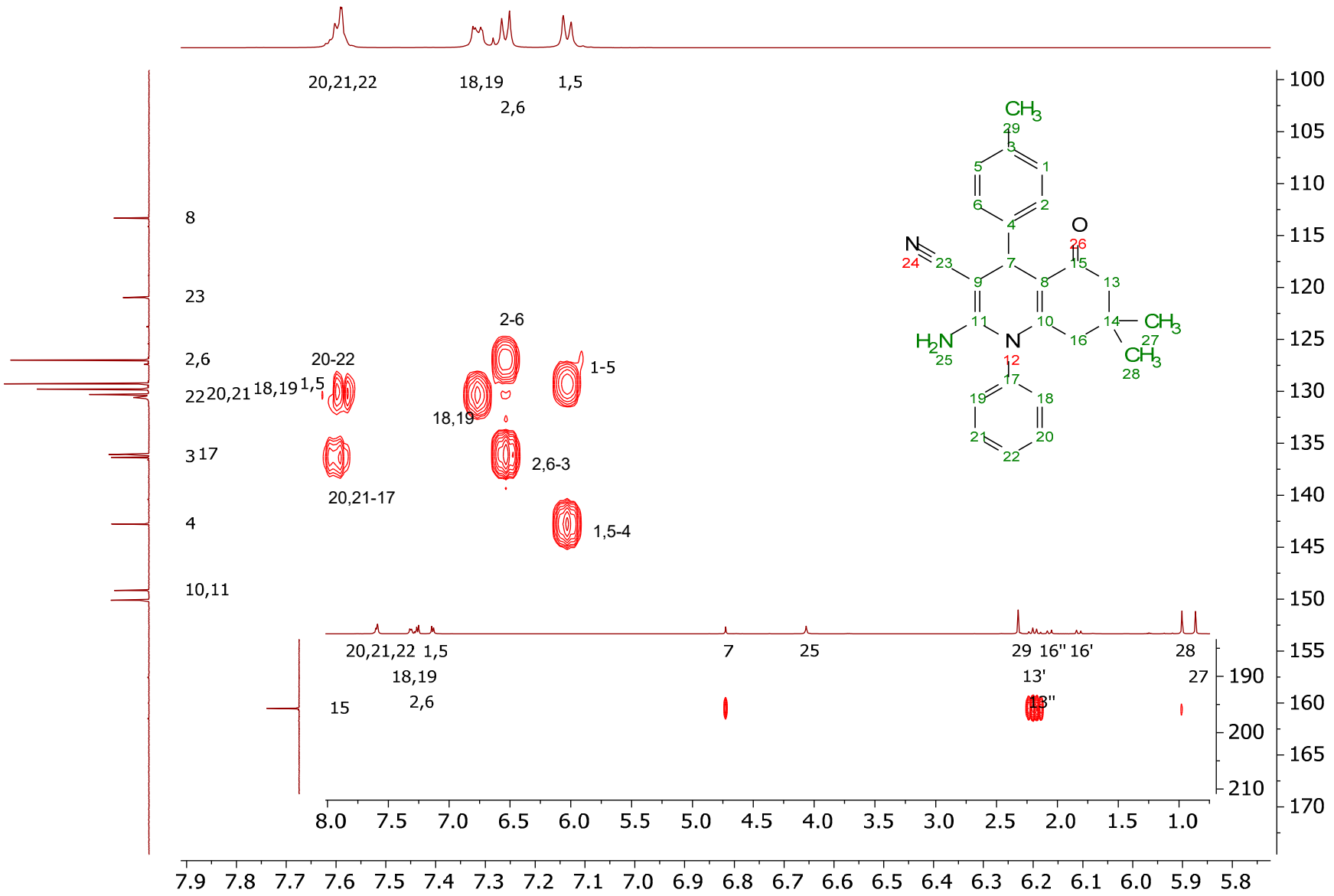


Figure S8 - Downfield region of HMBC spectrum of 5b

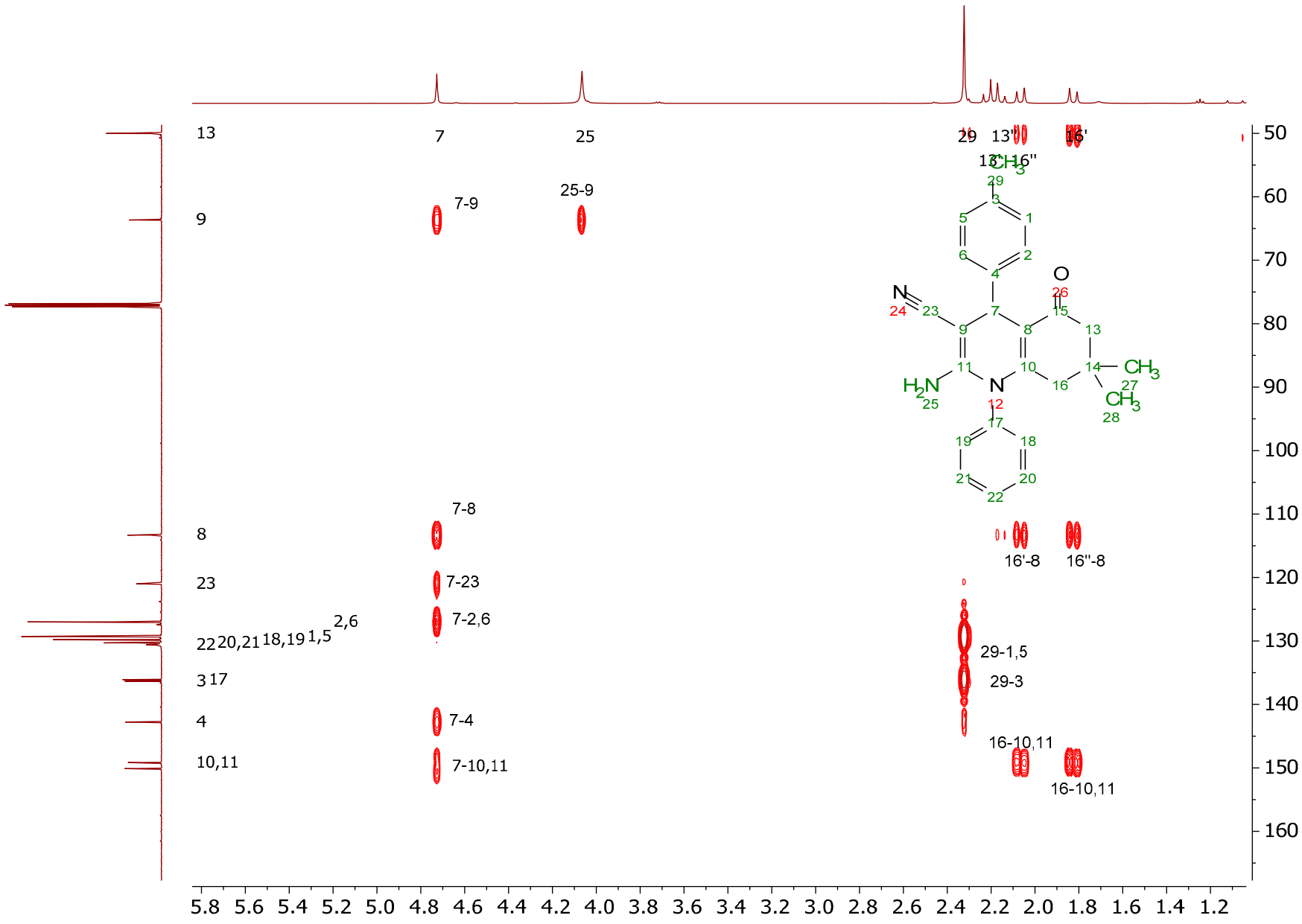


Figure S9 - Upfield region of HMBC spectrum of 5b

1.3. Product 5c: 2-amino-4-(4-chlorophenyl)-7,8-dimethyl-5-oxo-1-phenyl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

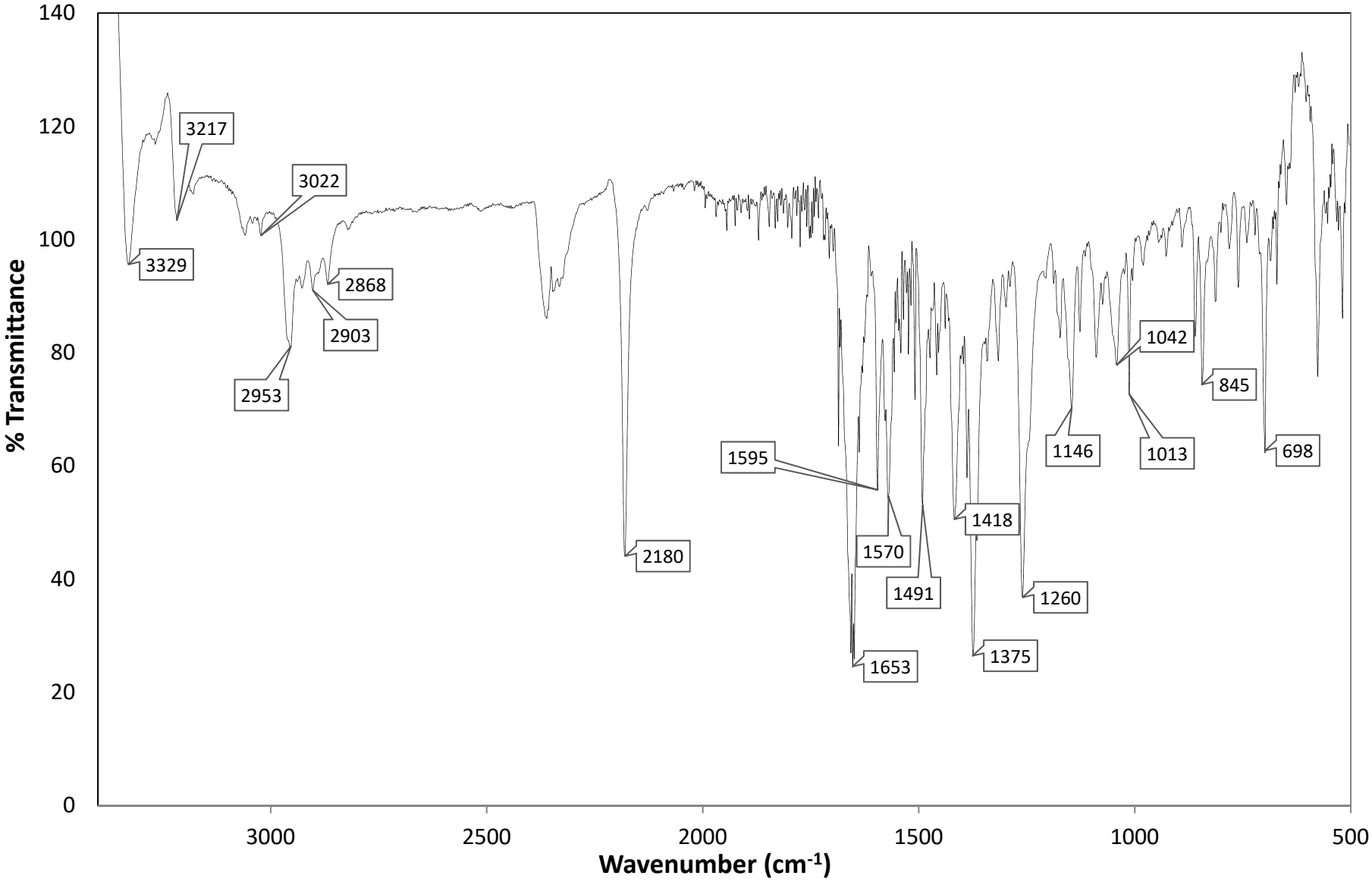


Figure S10 - IR spectrum of 5c

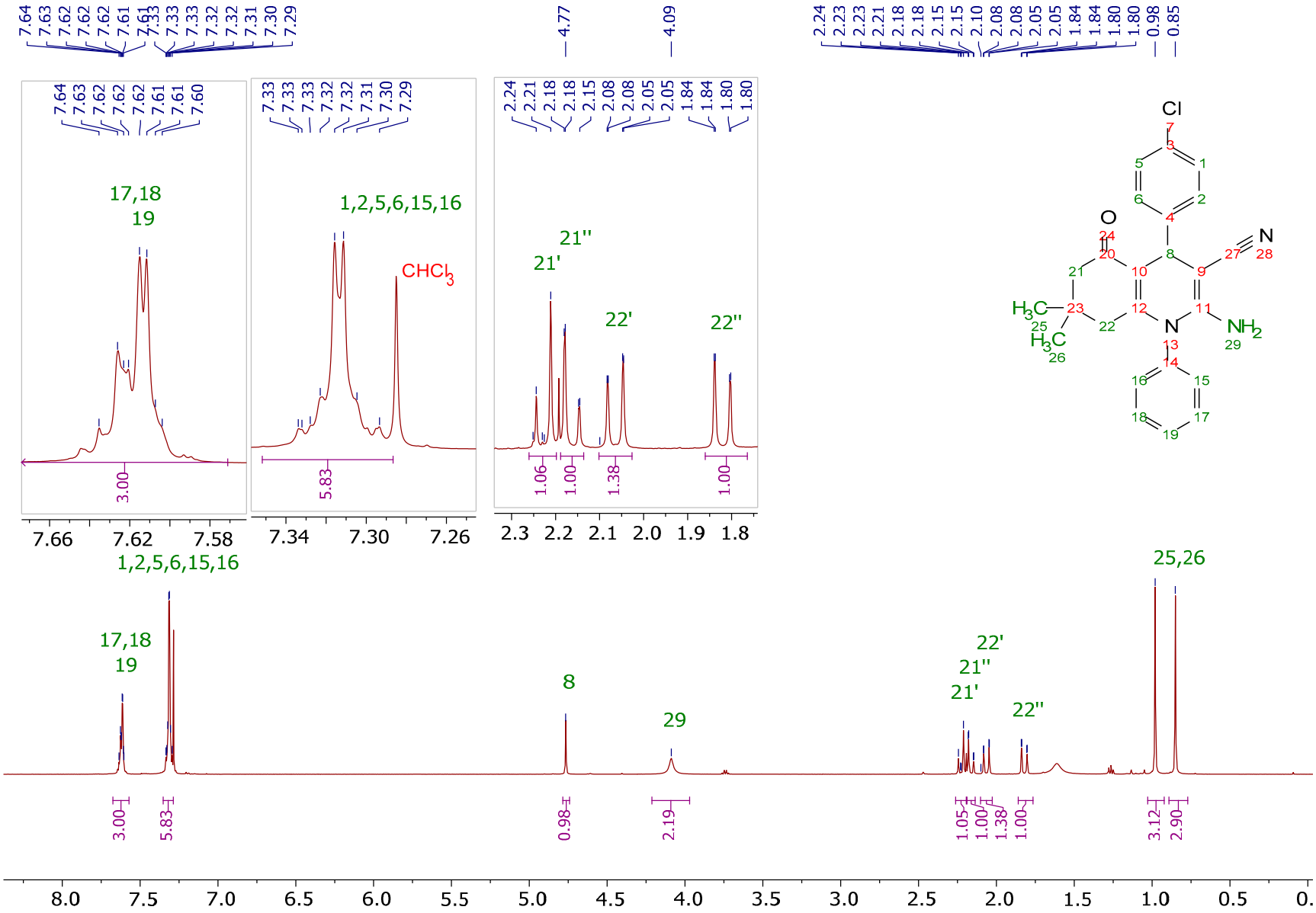


Figure S11 - ^{1}H NMR spectrum of 5c

1.4. Product 5d: 2-amino-4-(2,4-dichlorophenyl)-7,8-dimethyl-5-oxo-1-phenyl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

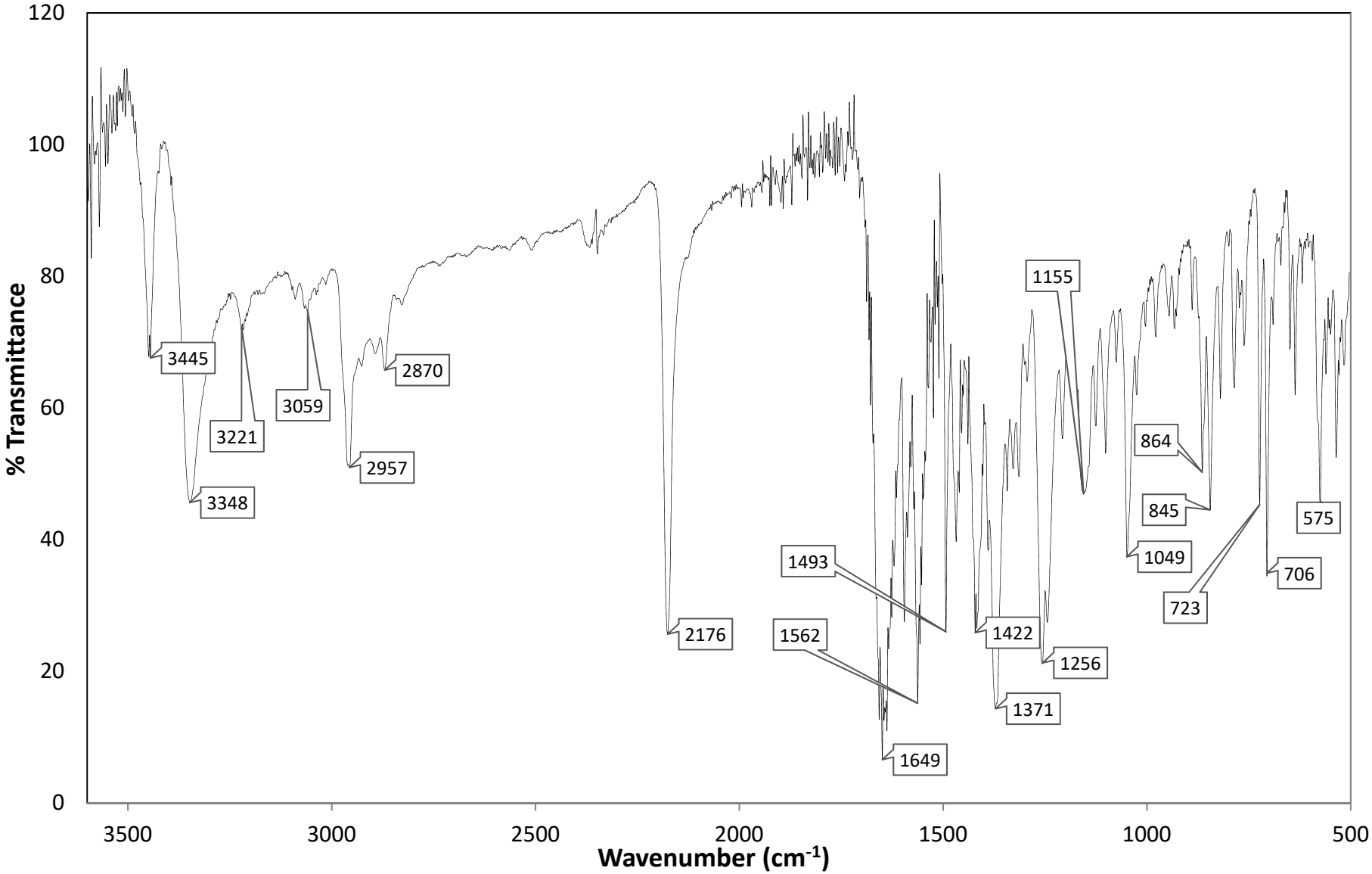


Figure S12 - IR spectrum of 5d

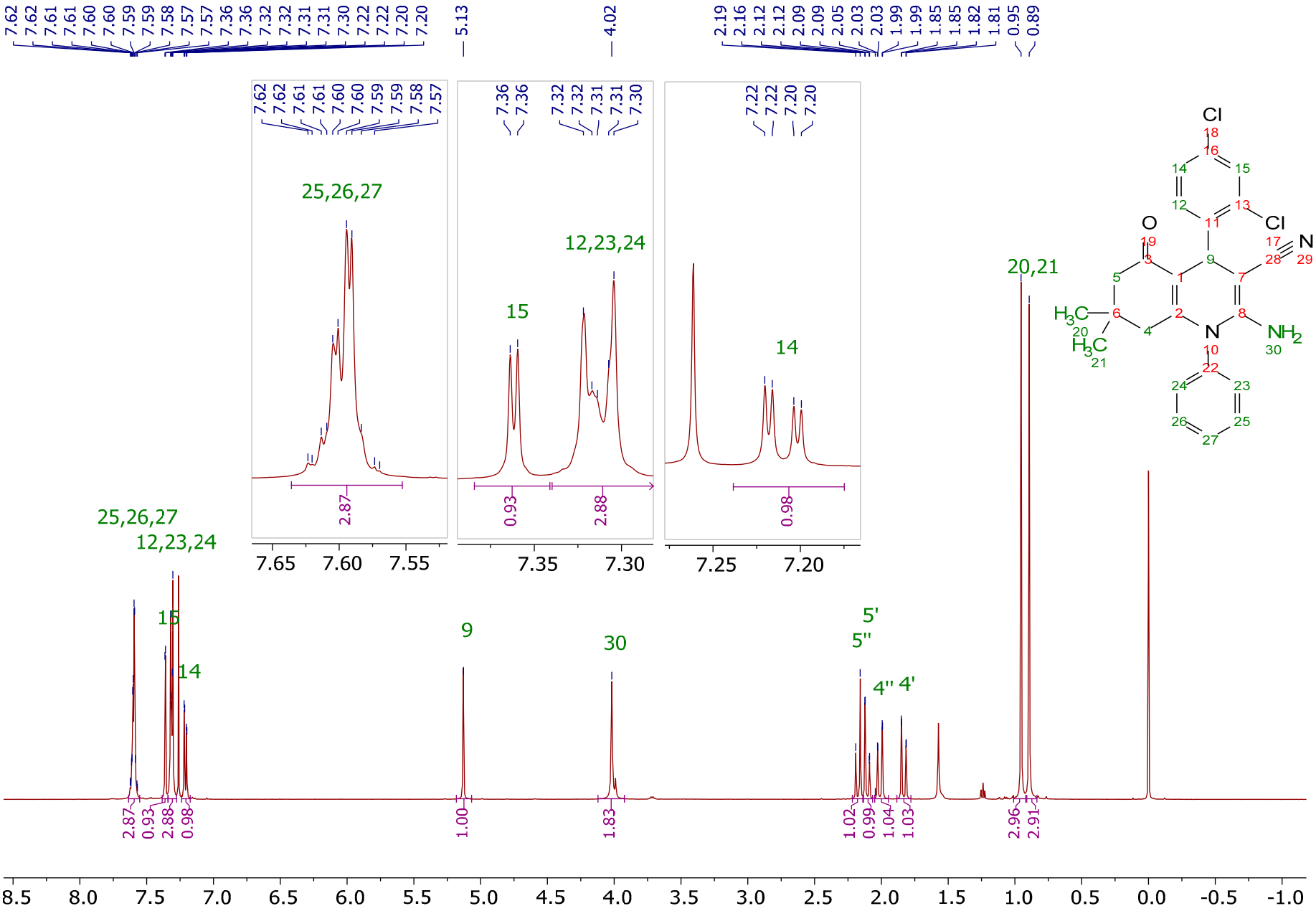


Figure S13 - ^{1}H NMR spectrum of 5d

1.5. Product 5e: 2-amino-4-(4-fluorophenyl)-7,8-dimethyl-5-oxo-1-phenyl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

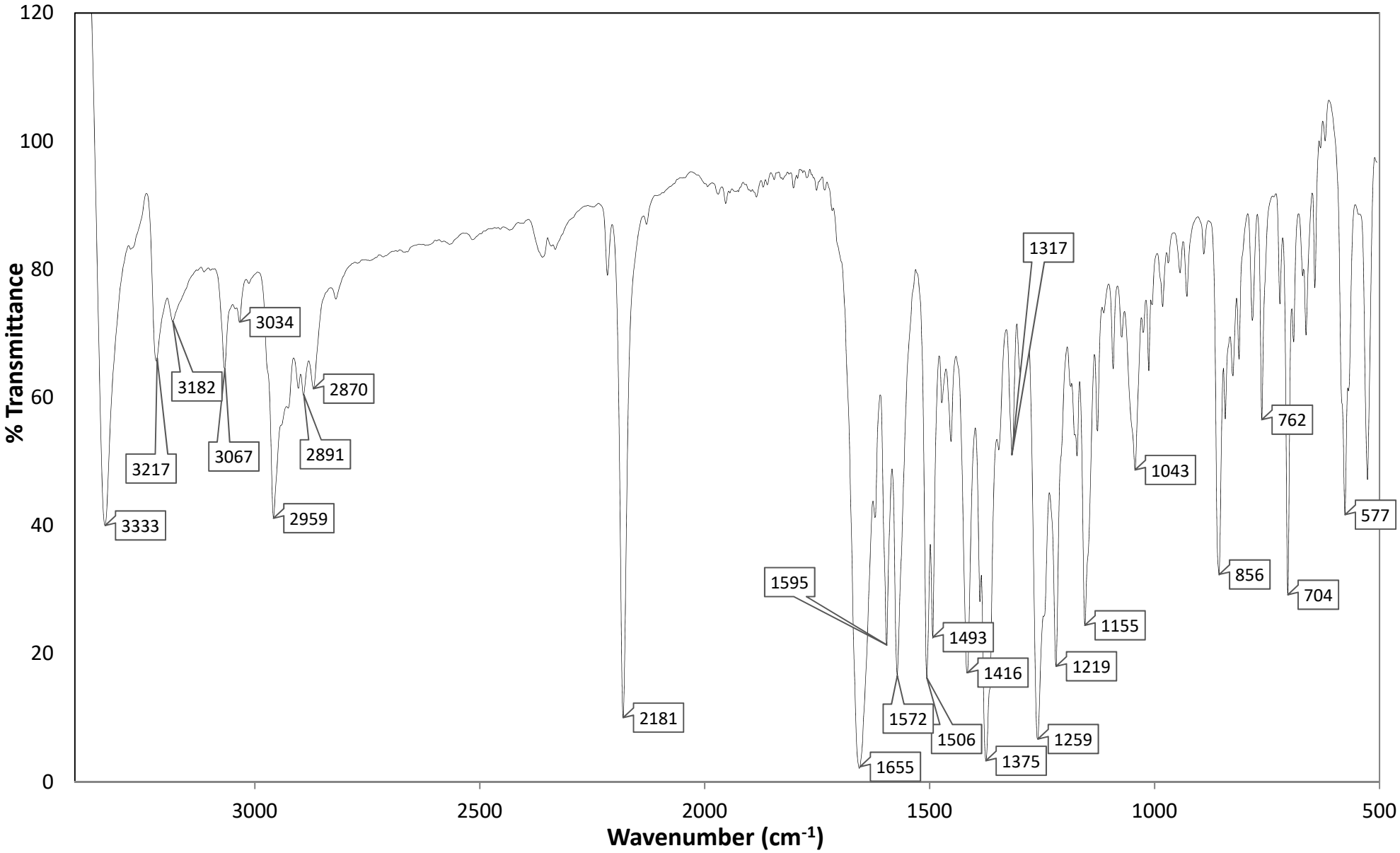


Figure S14 - IR spectrum of 5e

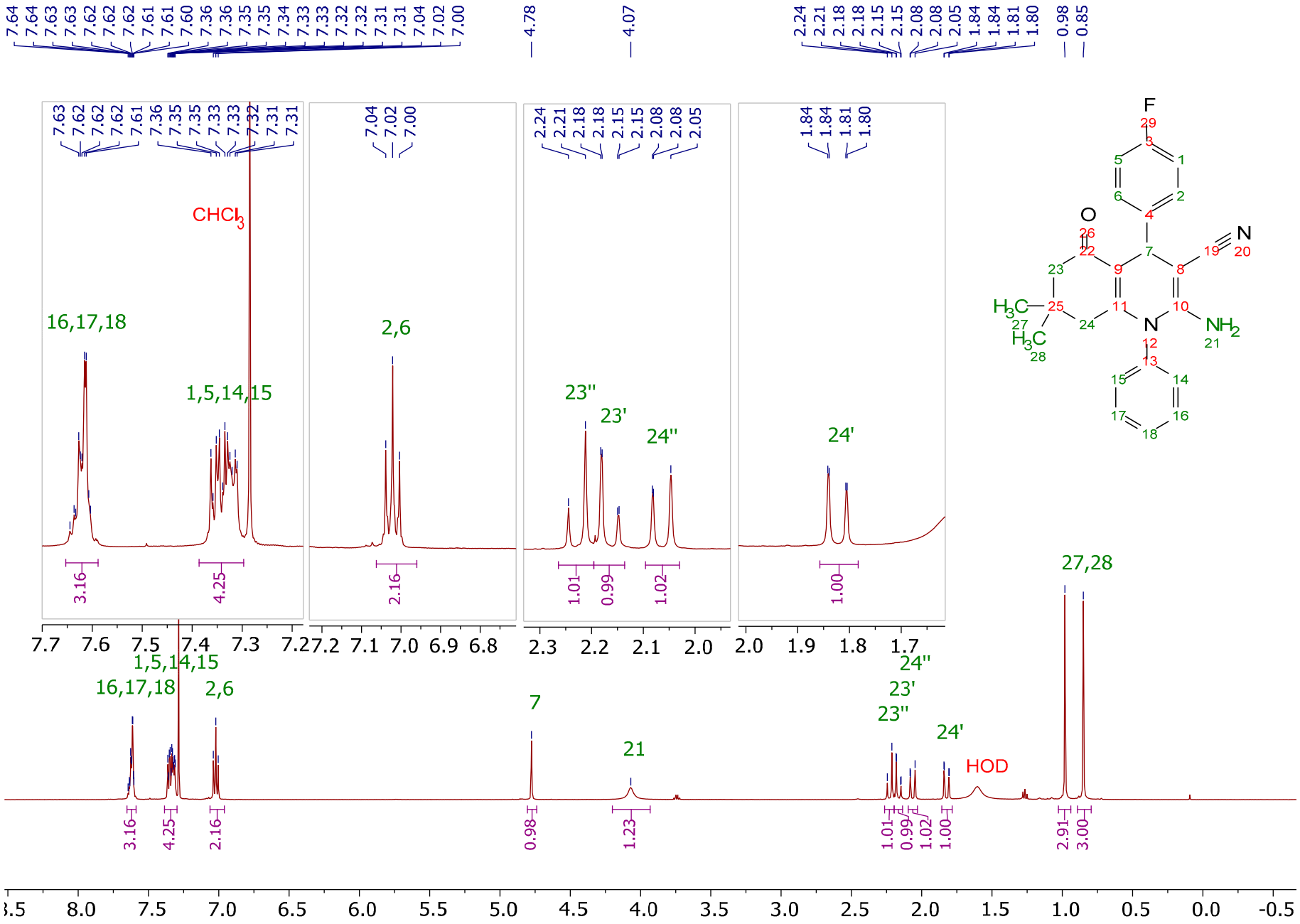


Figure S15 - ^{1}H NMR spectrum of 5e

1.6. Product 5f: 2-amino-7,8-dimethyl-5-oxo-1-phenyl-4-thiophen-2-yl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

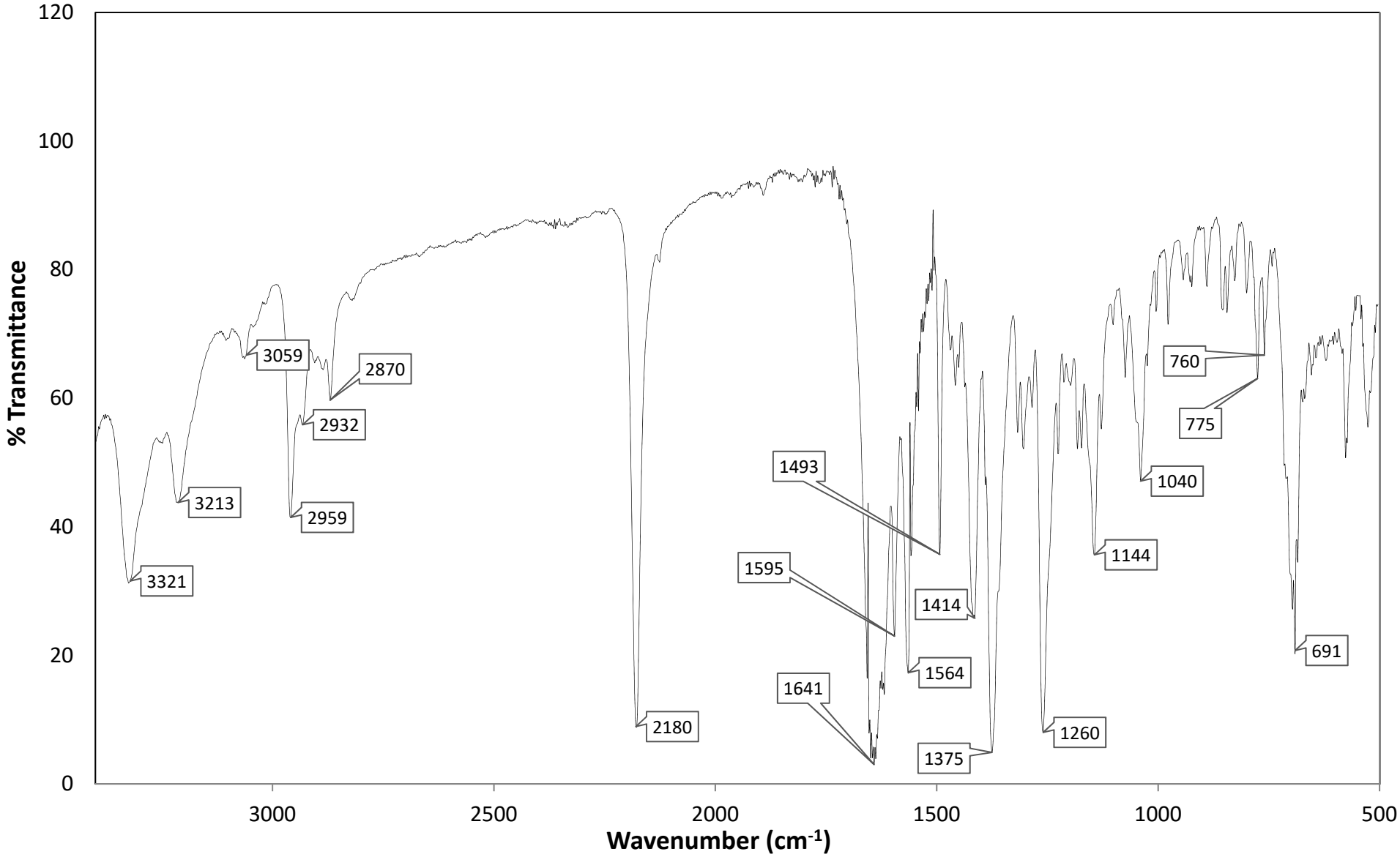


Figure S16 - IR spectrum of 5f

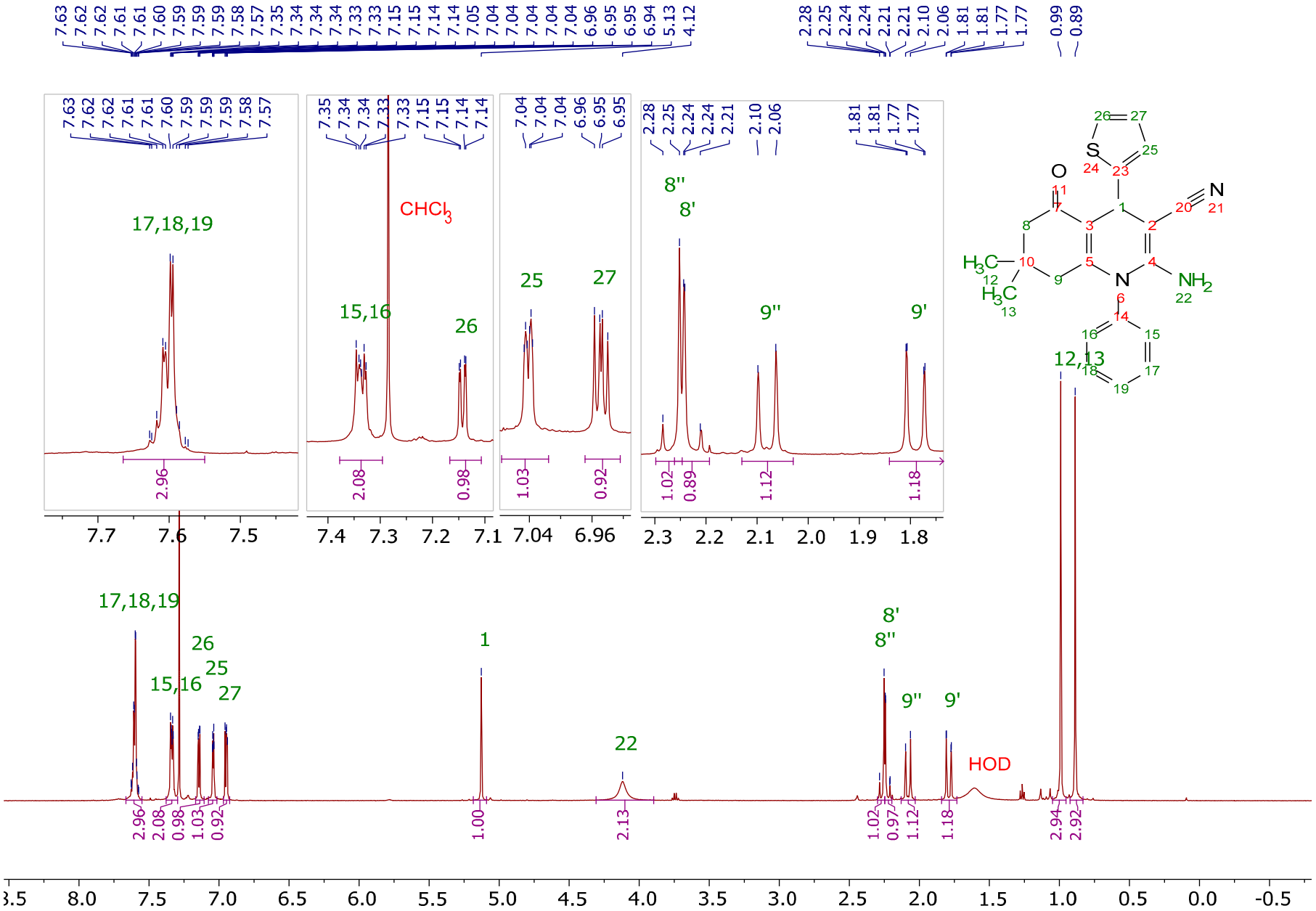


Figure S17 - ^{1}H NMR spectrum of 5f

1.7. Product 5g: 2-amino-7,8-dimethyl-5-oxo-1-phenyl-4-(3-nitrophenyl)-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

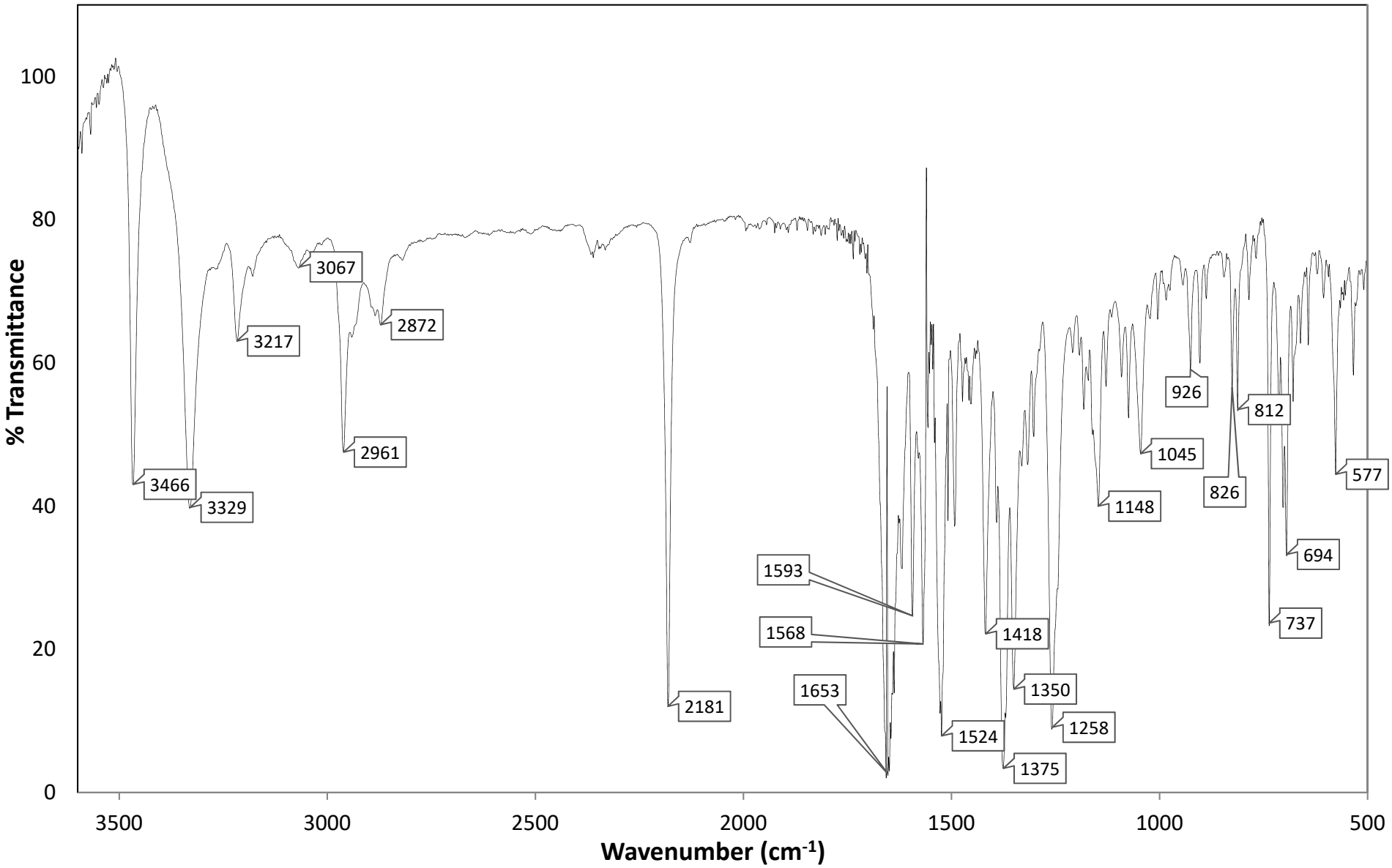


Figure S18 - IR spectrum of 5g

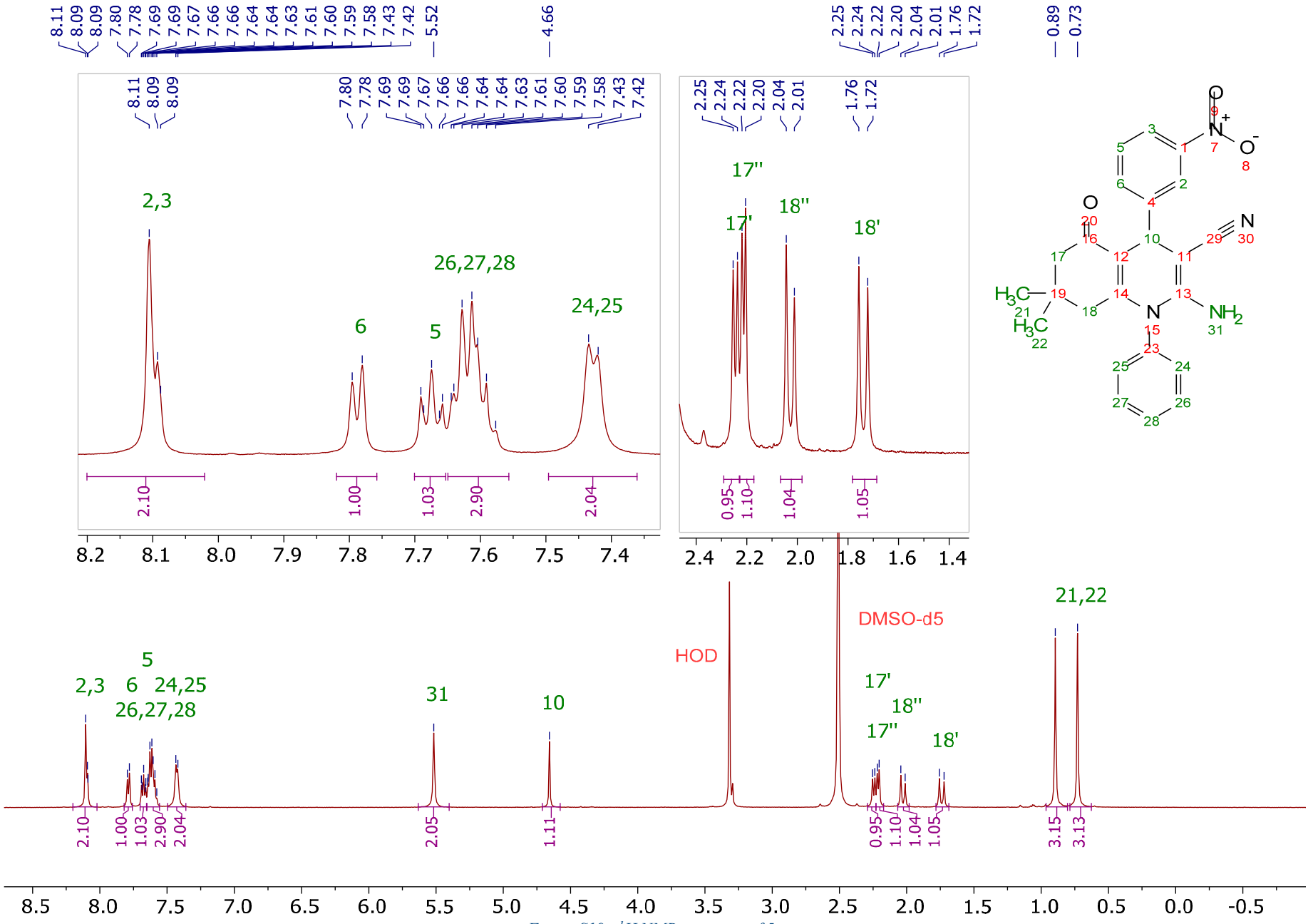


Figure S19 - ^{1}H NMR spectrum of 5g

1.8. Product 5h: 2-amino-4-(4-methoxyphenyl)-7,8-dimethyl-5-oxo-1-phenyl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

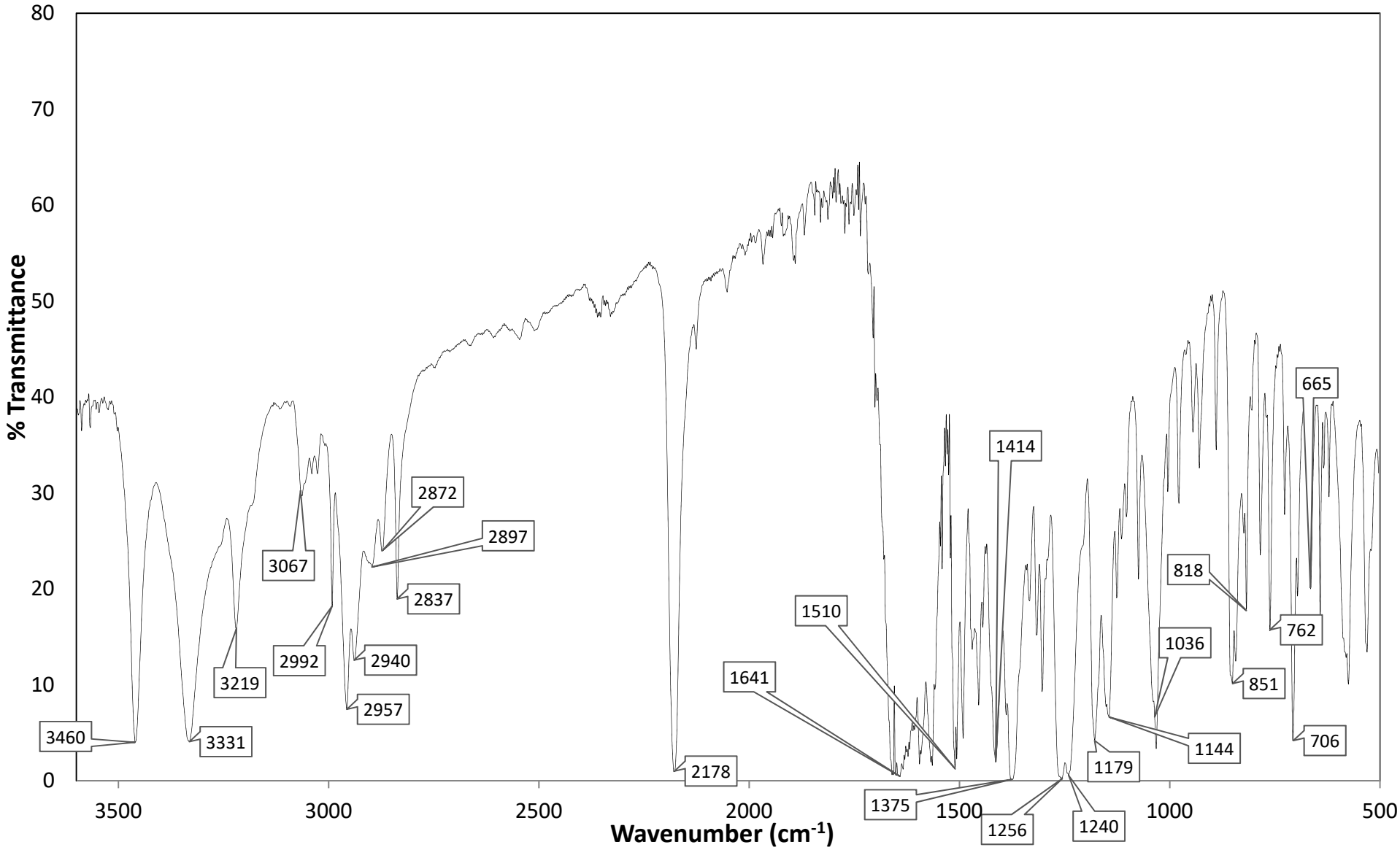


Figure S20 - IR spectrum of 5h

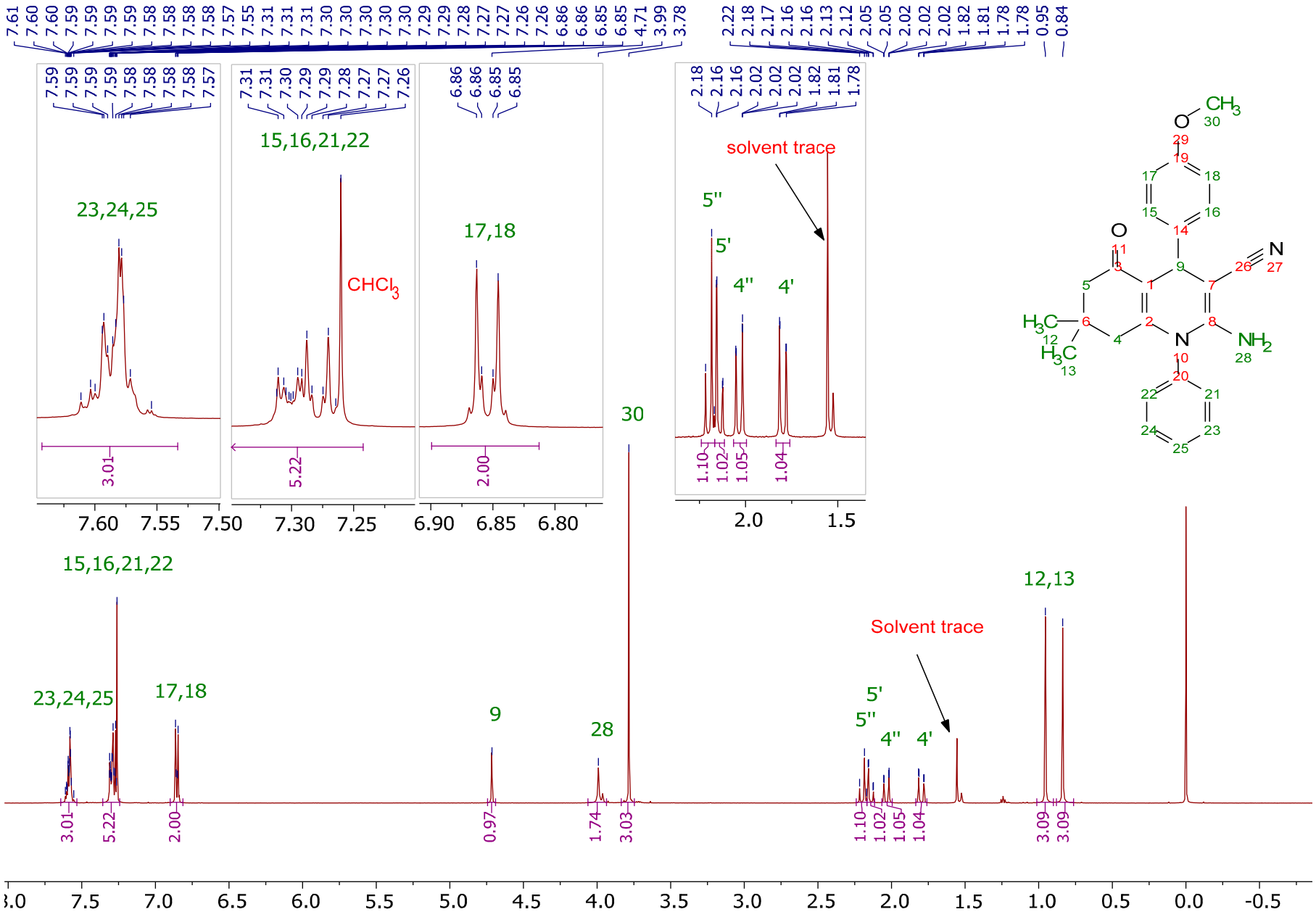


Figure S21 - ^{1}H NMR spectrum of 5h

1.9. Product 5i: Methyl 2-amino-4-(2,4-dichlorophenyl)-7,8-dimethyl-5-oxo-1-phenyl-1,4,5,6,7,8-hexahydroquinoline-3-carboxylate

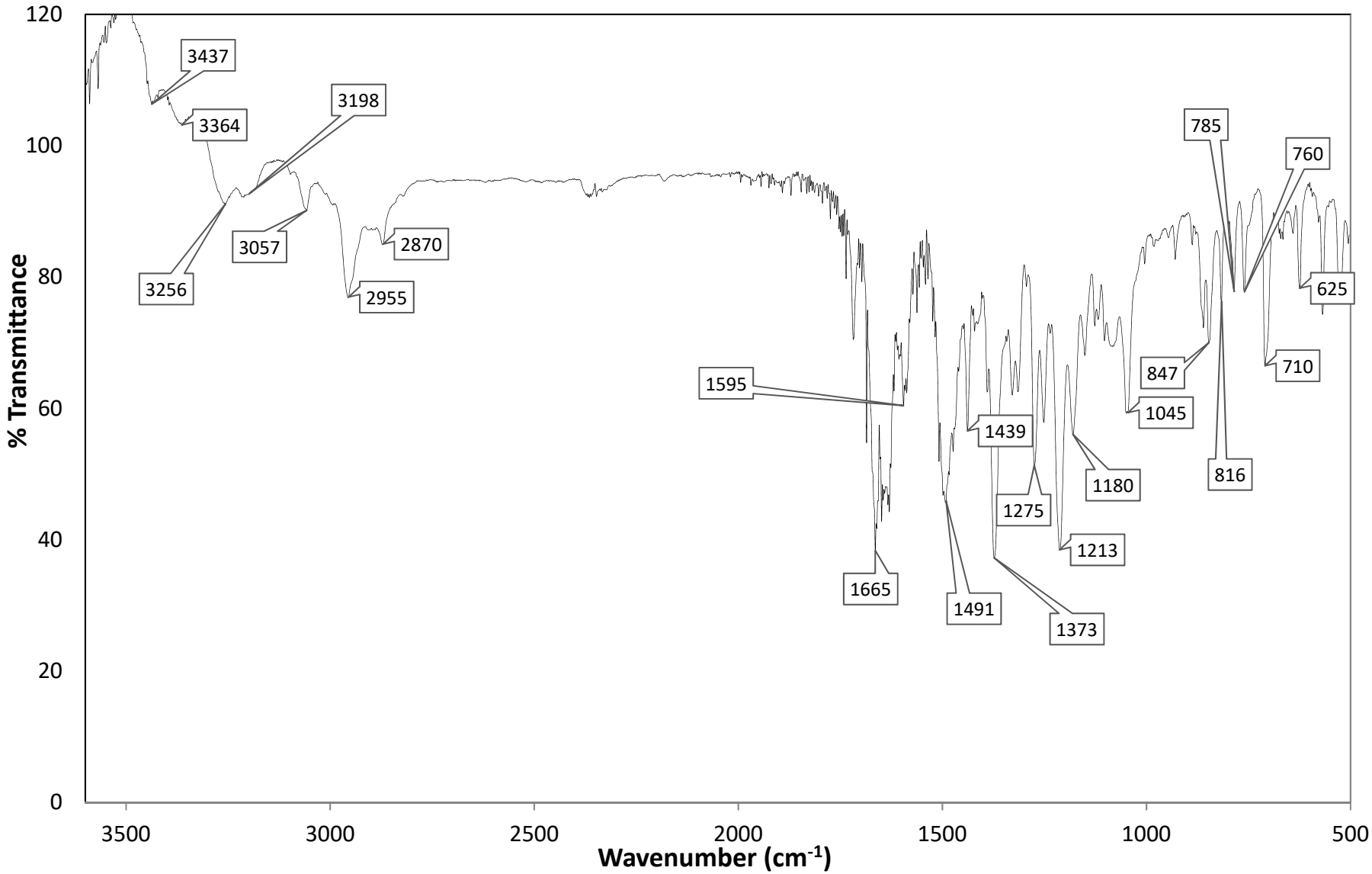


Figure S22 - IR spectrum of 5i

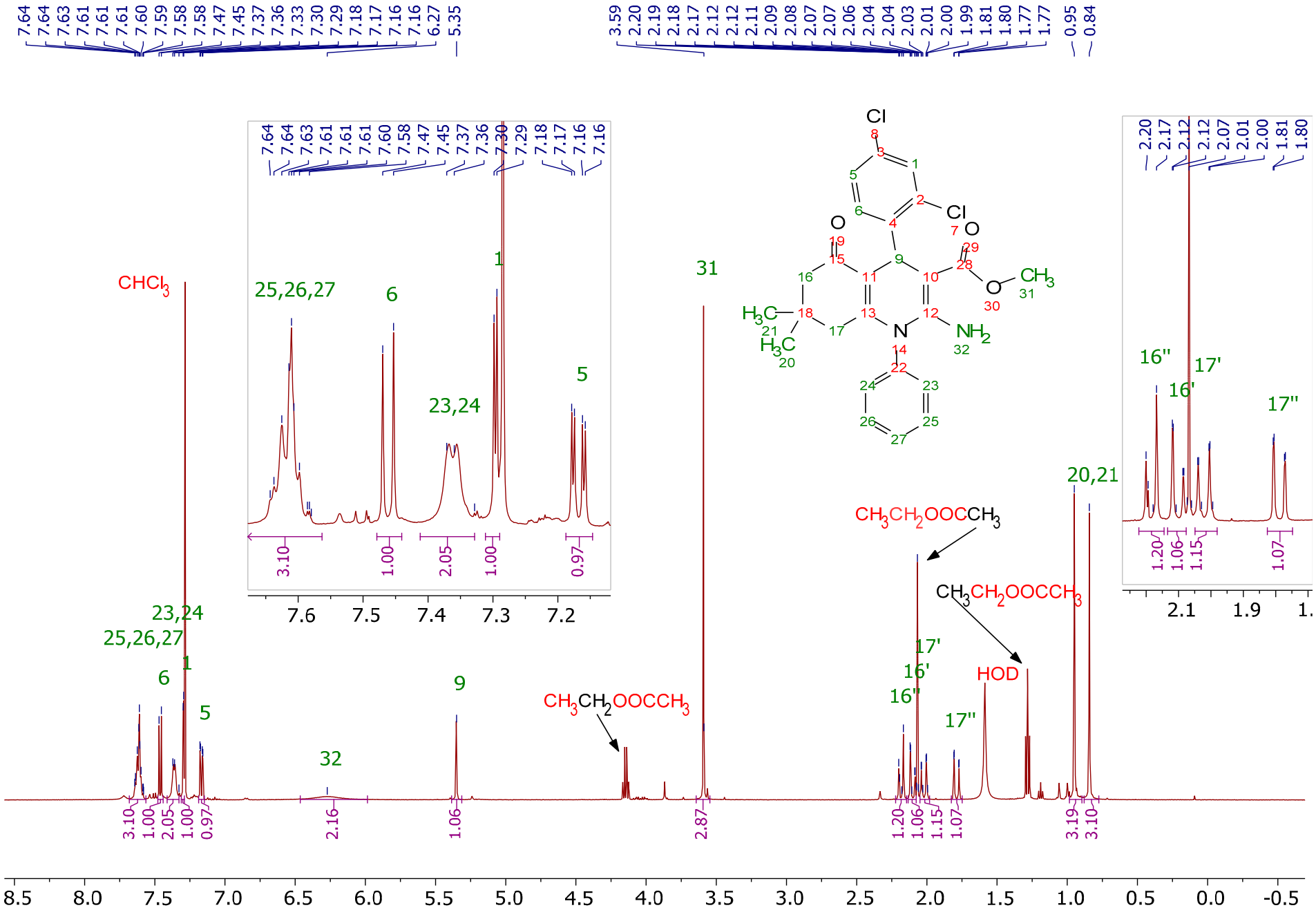


Figure S23 - ^{1}H NMR spectrum of 5i

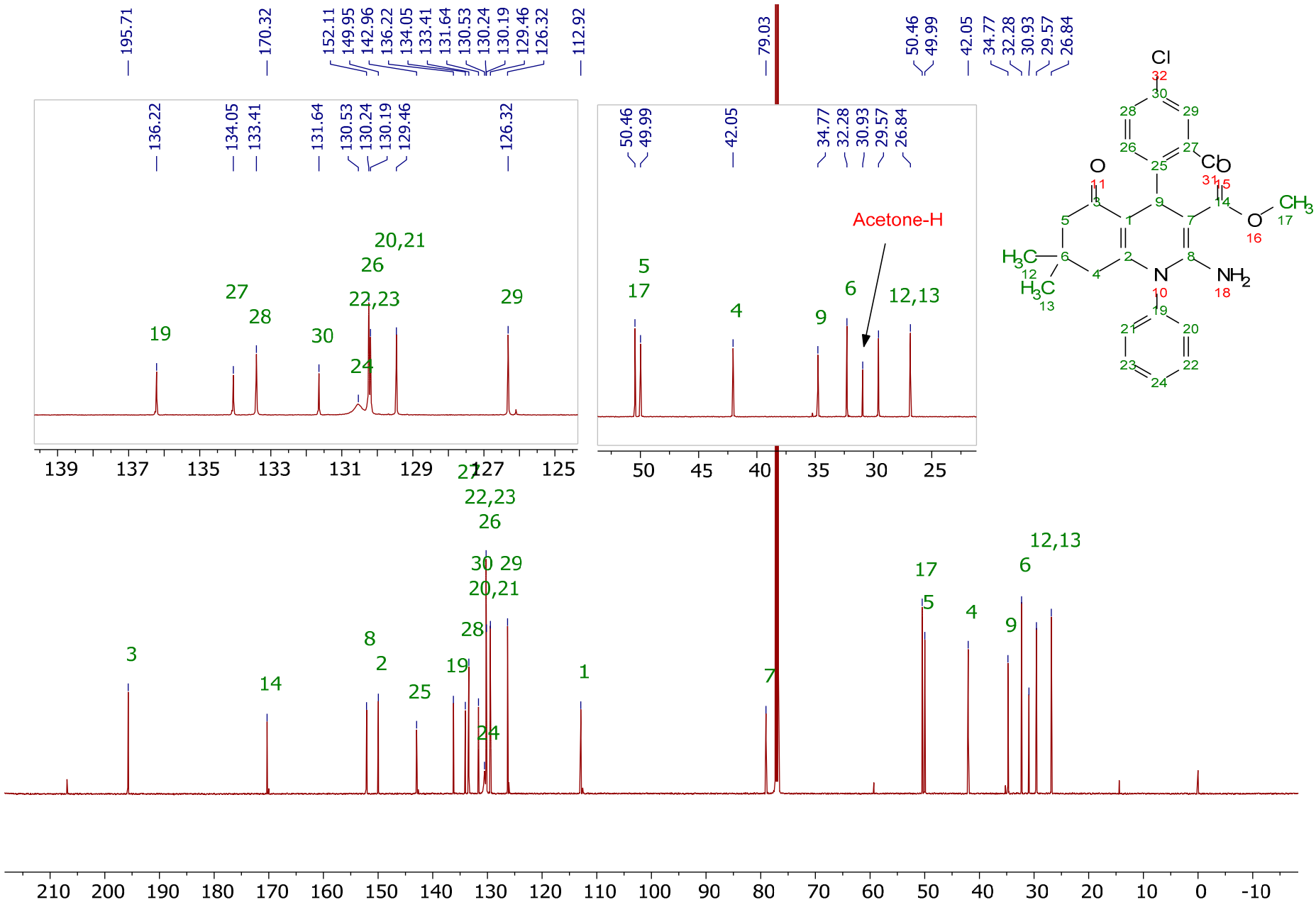


Figure S24 – ^{13}C NMR spectrum of 5i

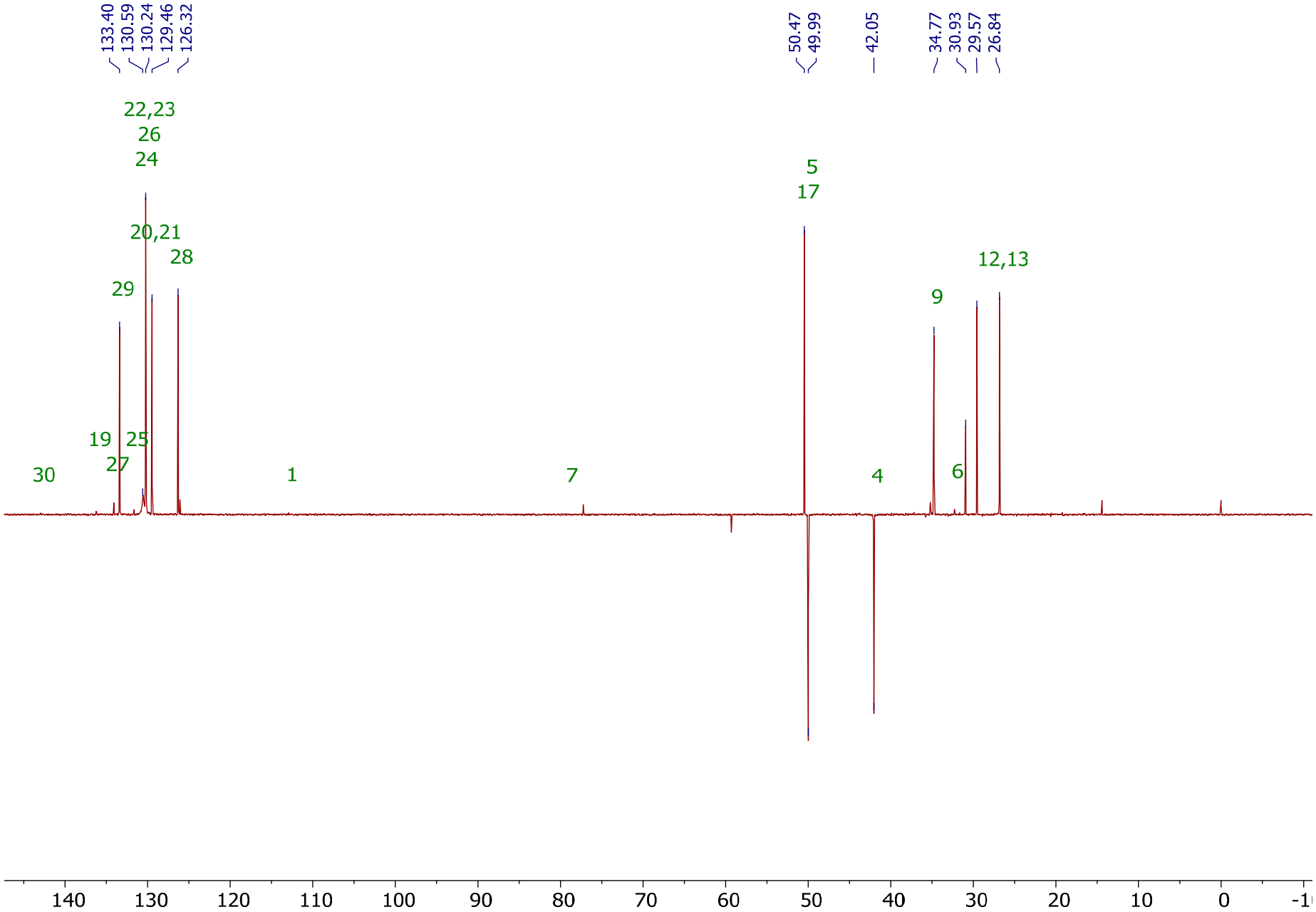


Figure S25 - DEPT spectrum of 5i

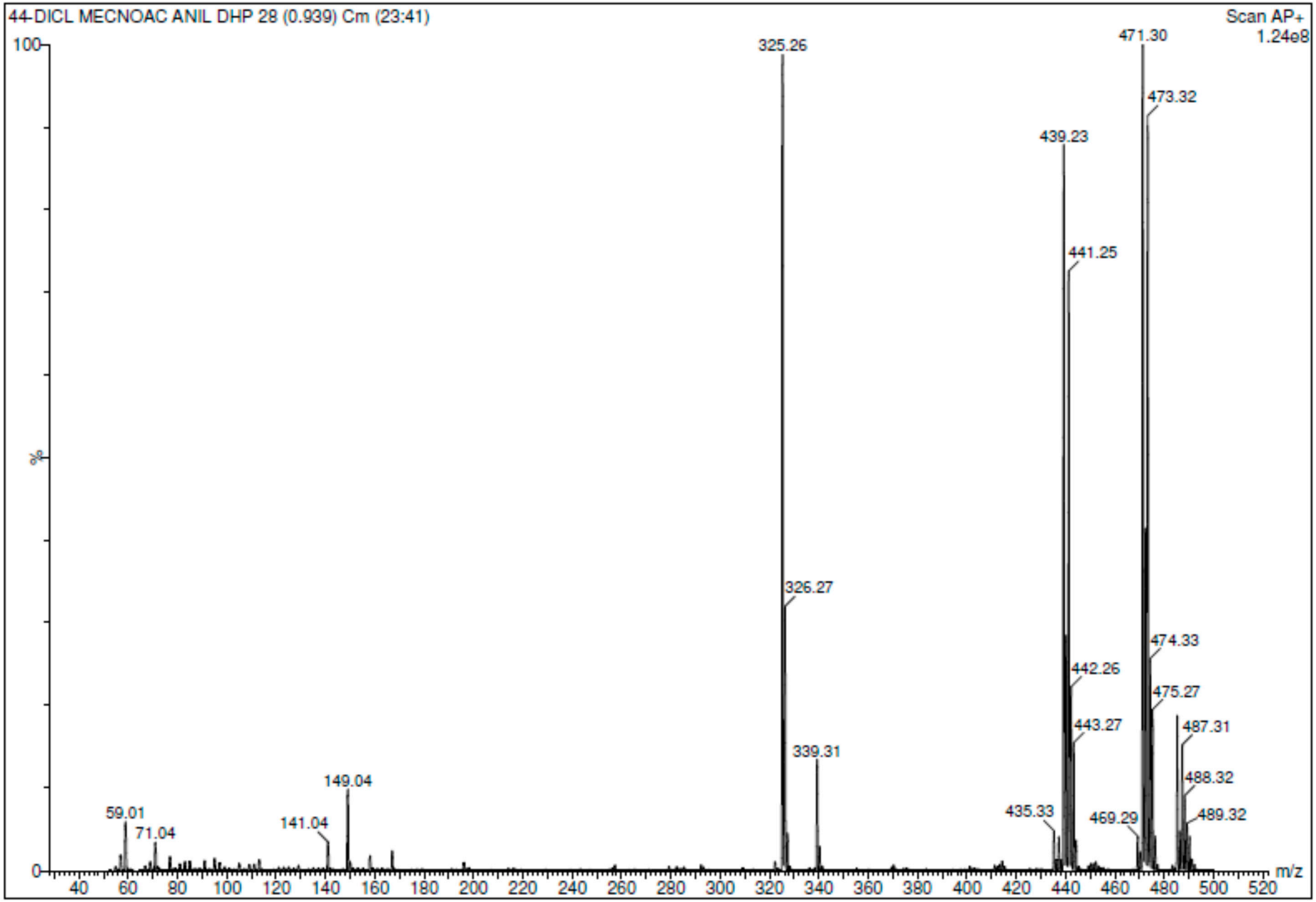
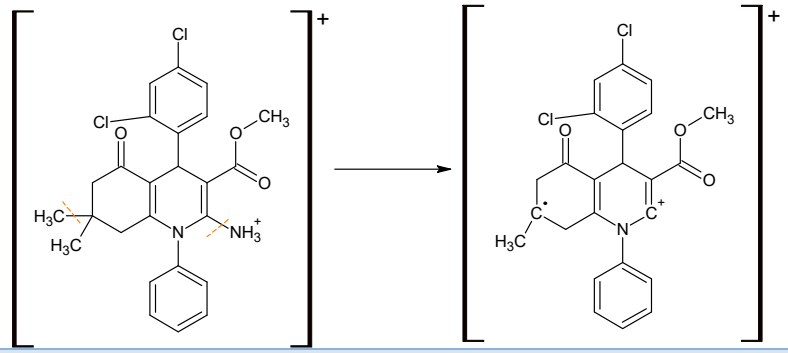
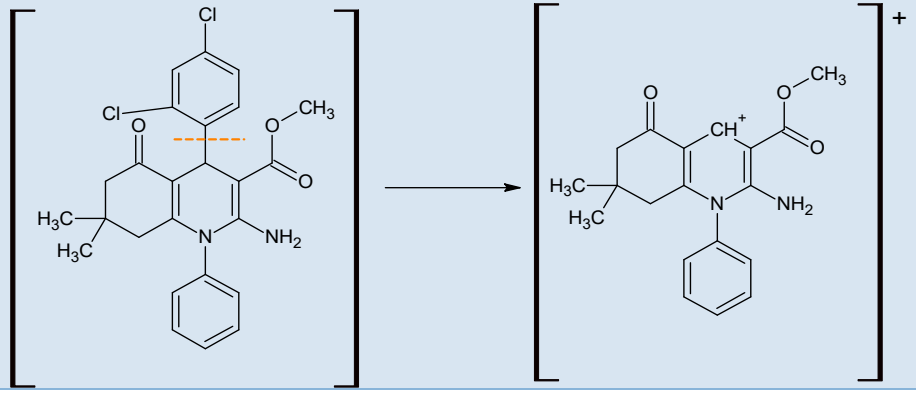
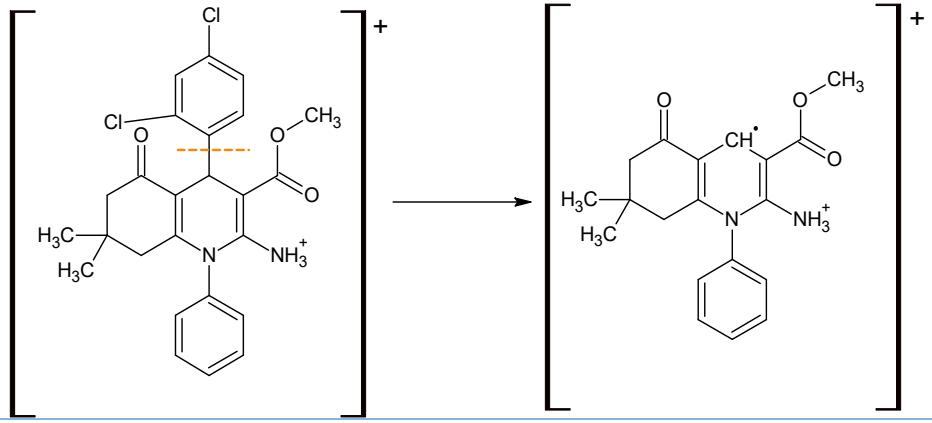


Figure S26 - MS spectrum of 29i

Table S2 - Fragmentation position for peaks in MS spectrum of 29i

m/z	Fragmentation position and structure
471.30 – 475.27	$[M+H]^+$ Multiple isotopes
439.23 – 443.27	
325.26	
326.27	

1.10. Product 5j: 2-amino-7,8-dimethyl-5-oxo-1-(3-methylphenyl)-4-(3-nitrophenyl)-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

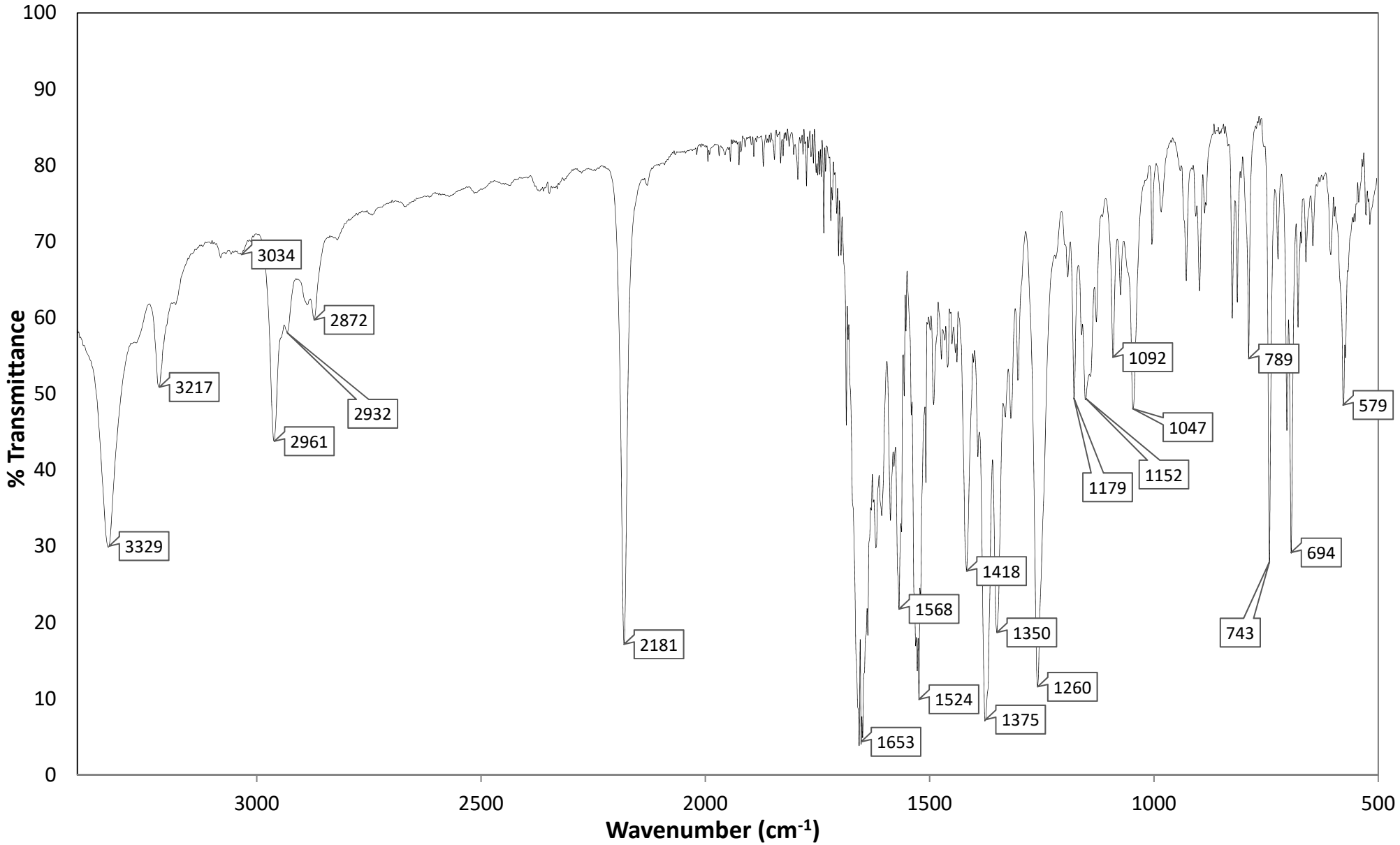


Figure S27 - IR spectrum of 5j

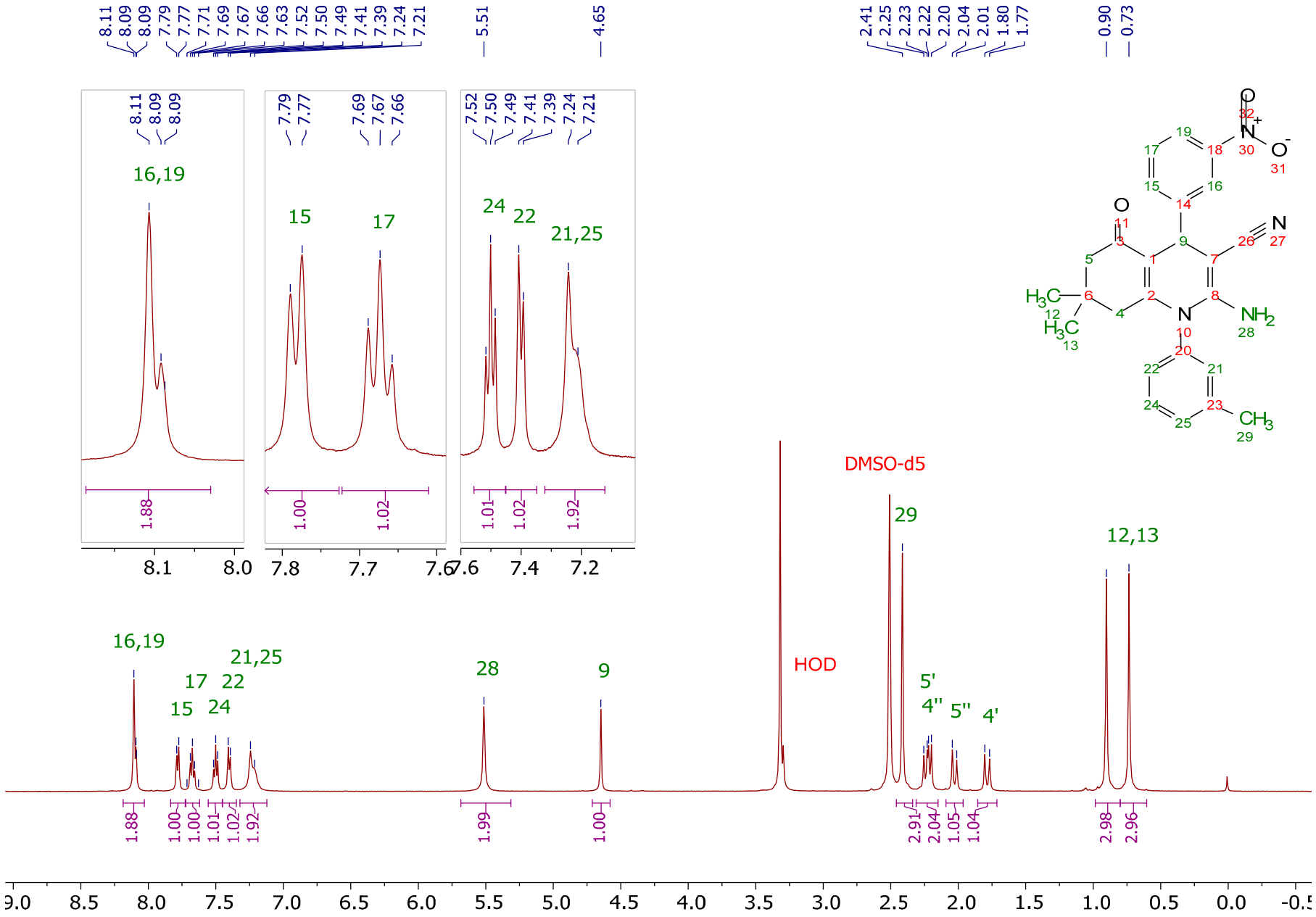


Figure S28 - ^{1}H NMR spectrum of 5j

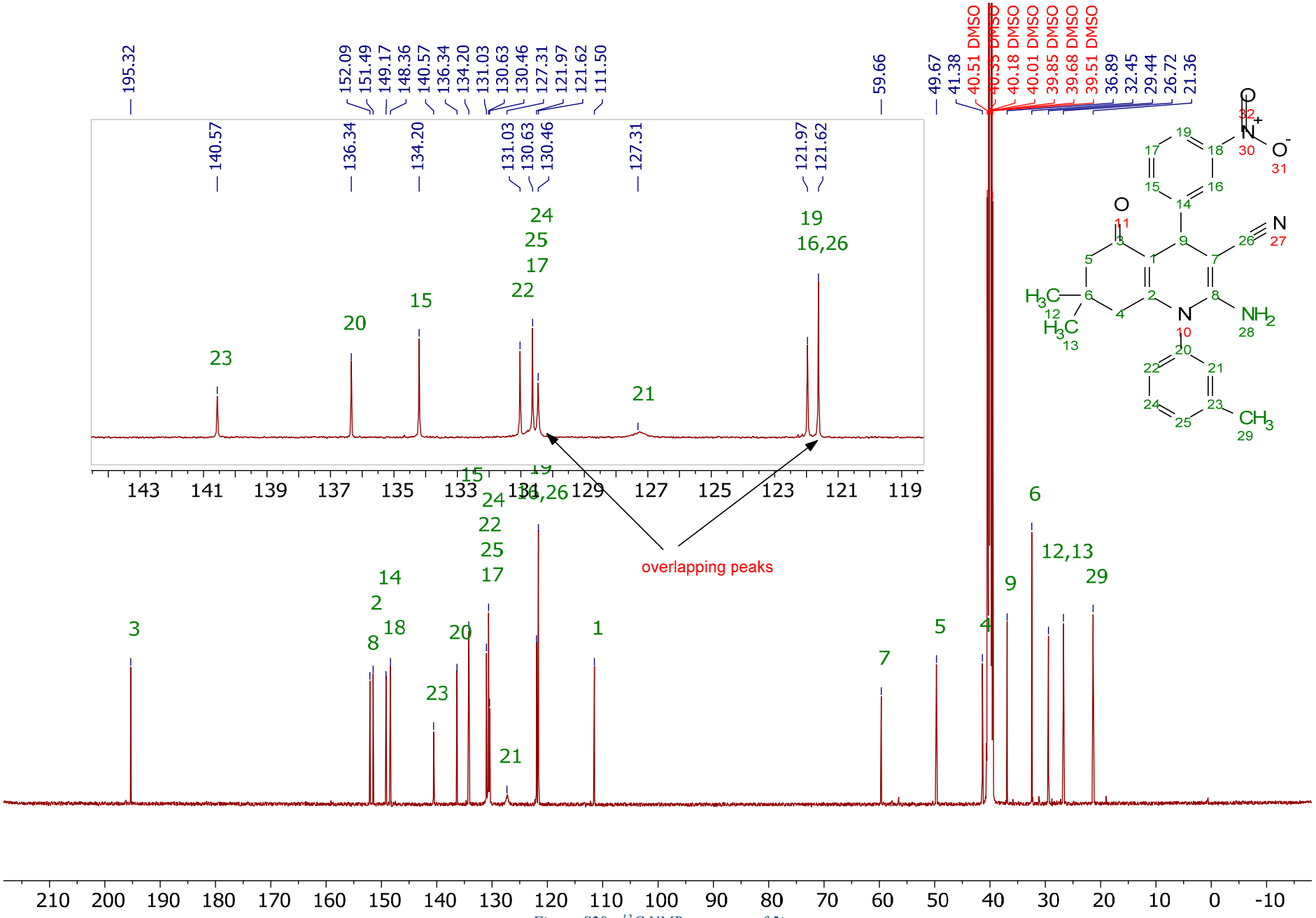


Figure S29 - ^{13}C NMR spectrum of 5j

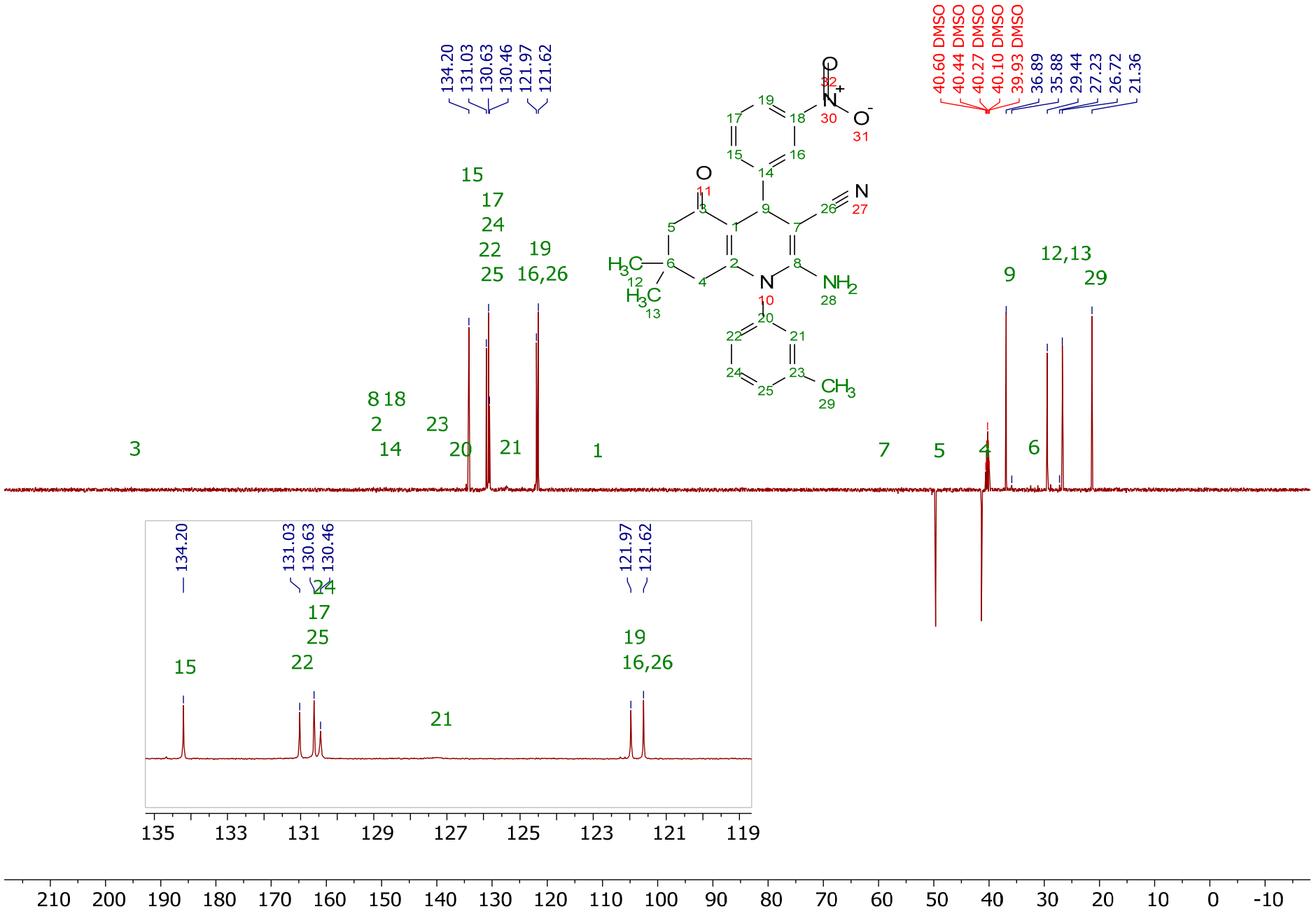


Figure S30 - DEPT spectrum of 5j

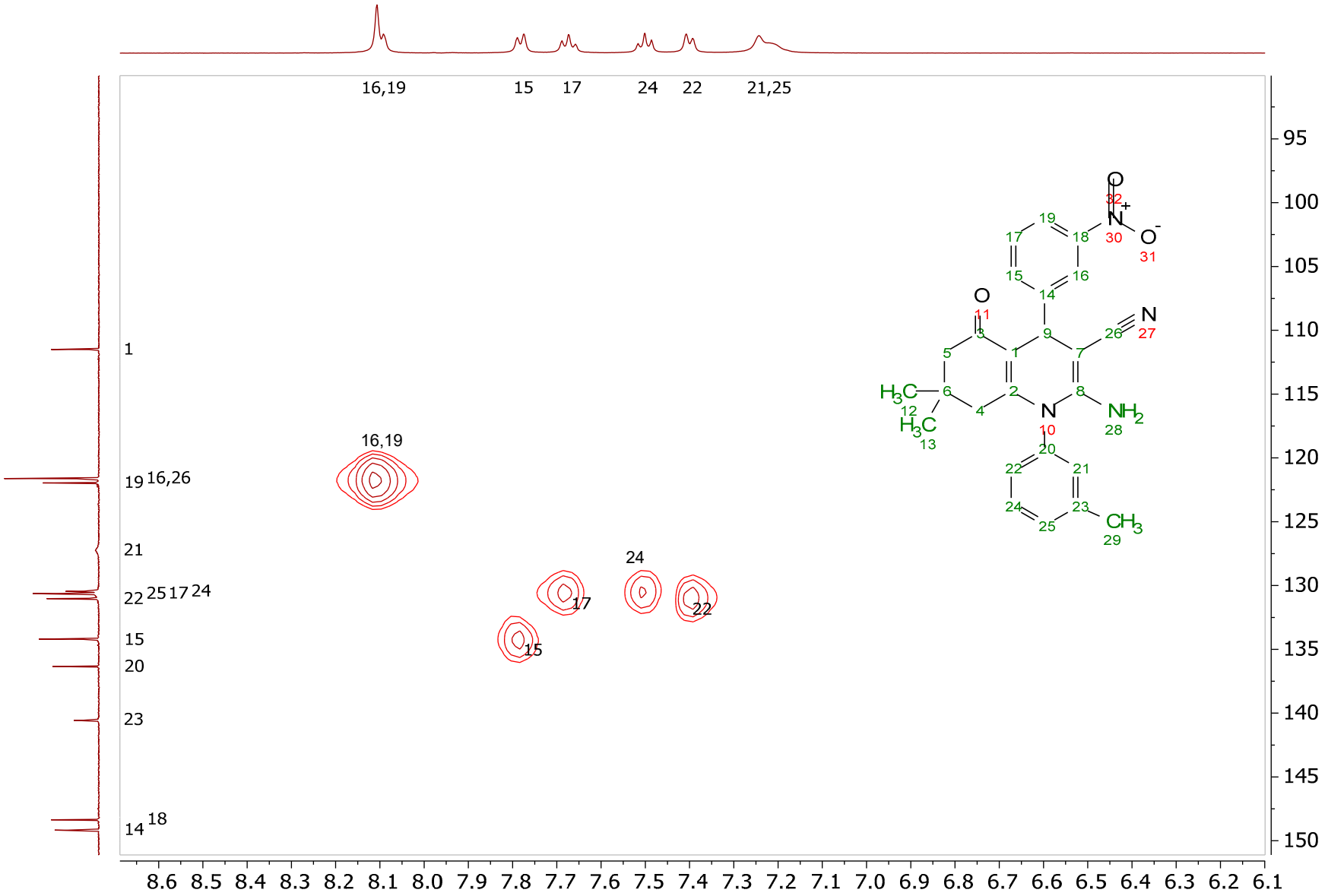


Figure S31 – Downfield region of HSQC NMR spectrum of 5j

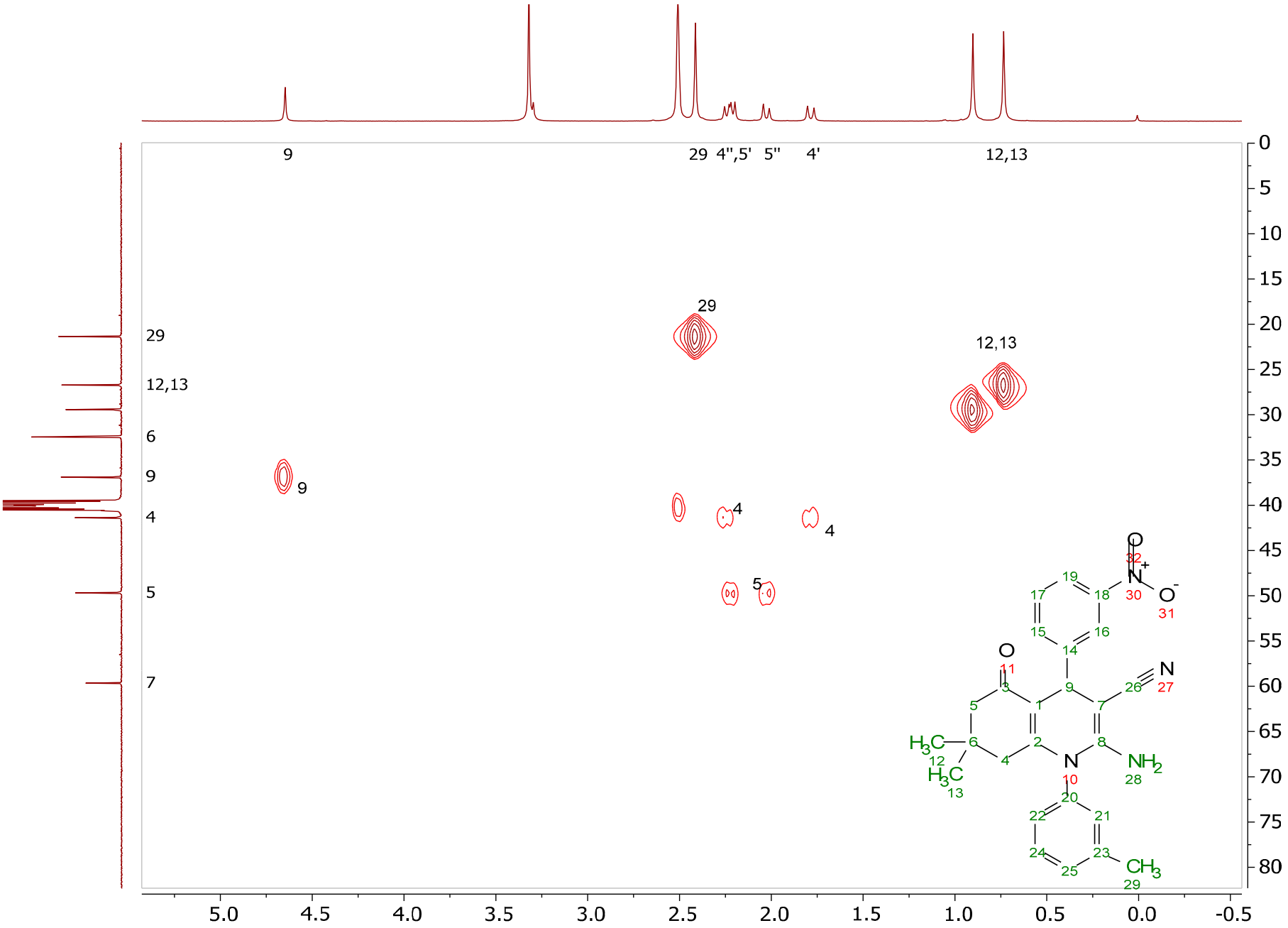


Figure S32 - Upfield region of HSQC spectrum of 5j

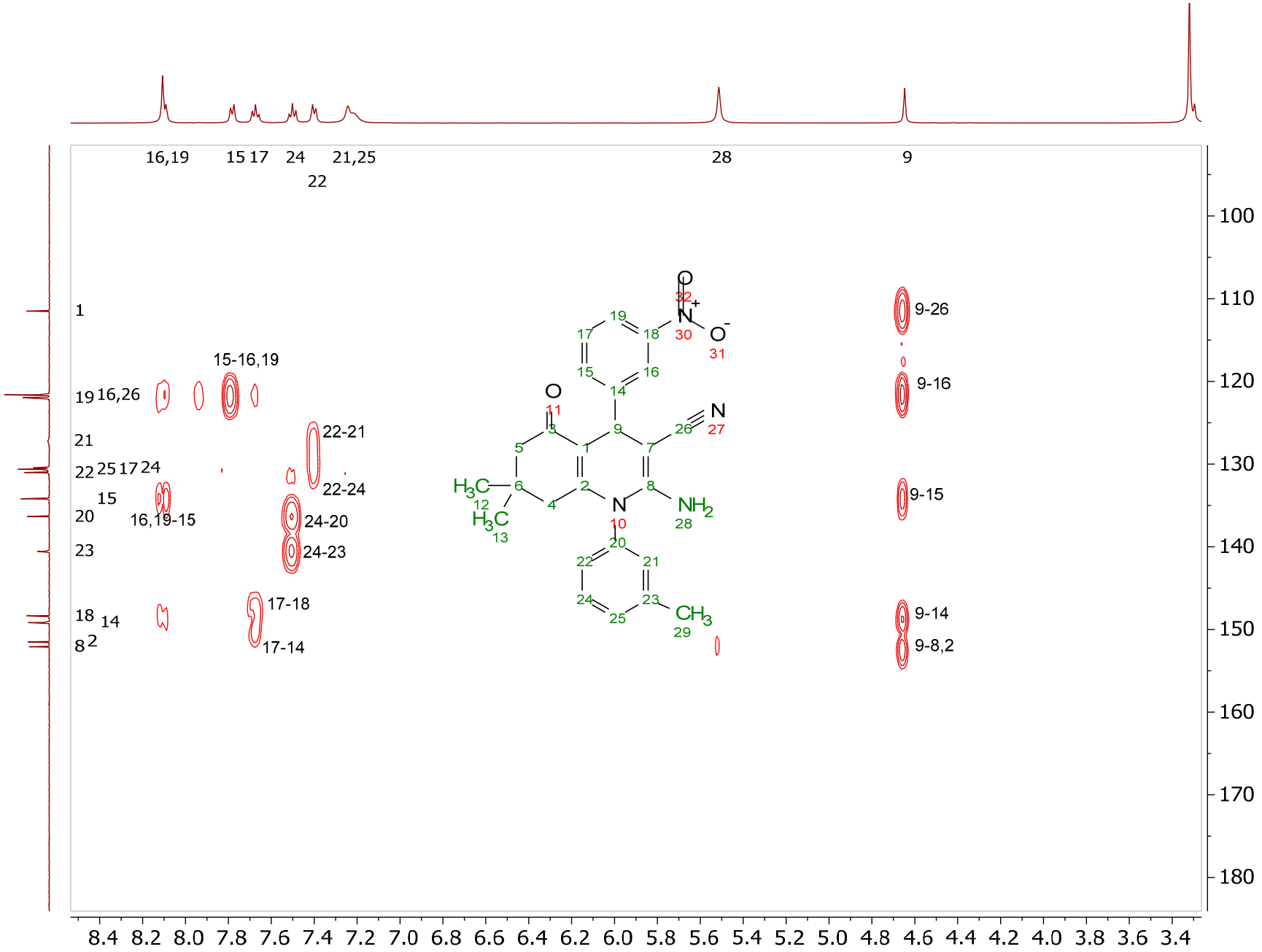


Figure S33 - Downfield region of HMBC spectrum of 5j

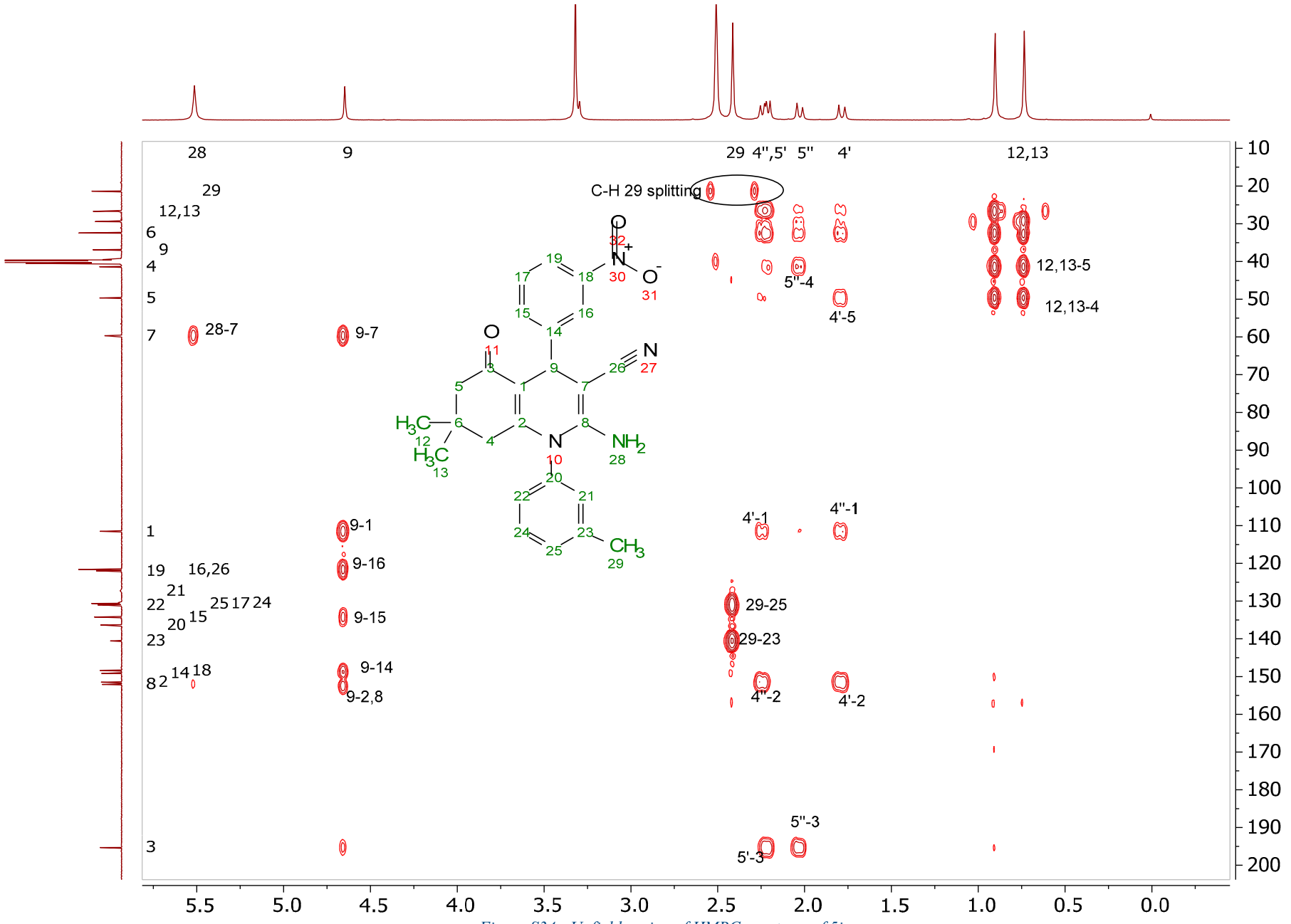


Figure S34 - Upfield region of HMBC spectrum of 5j

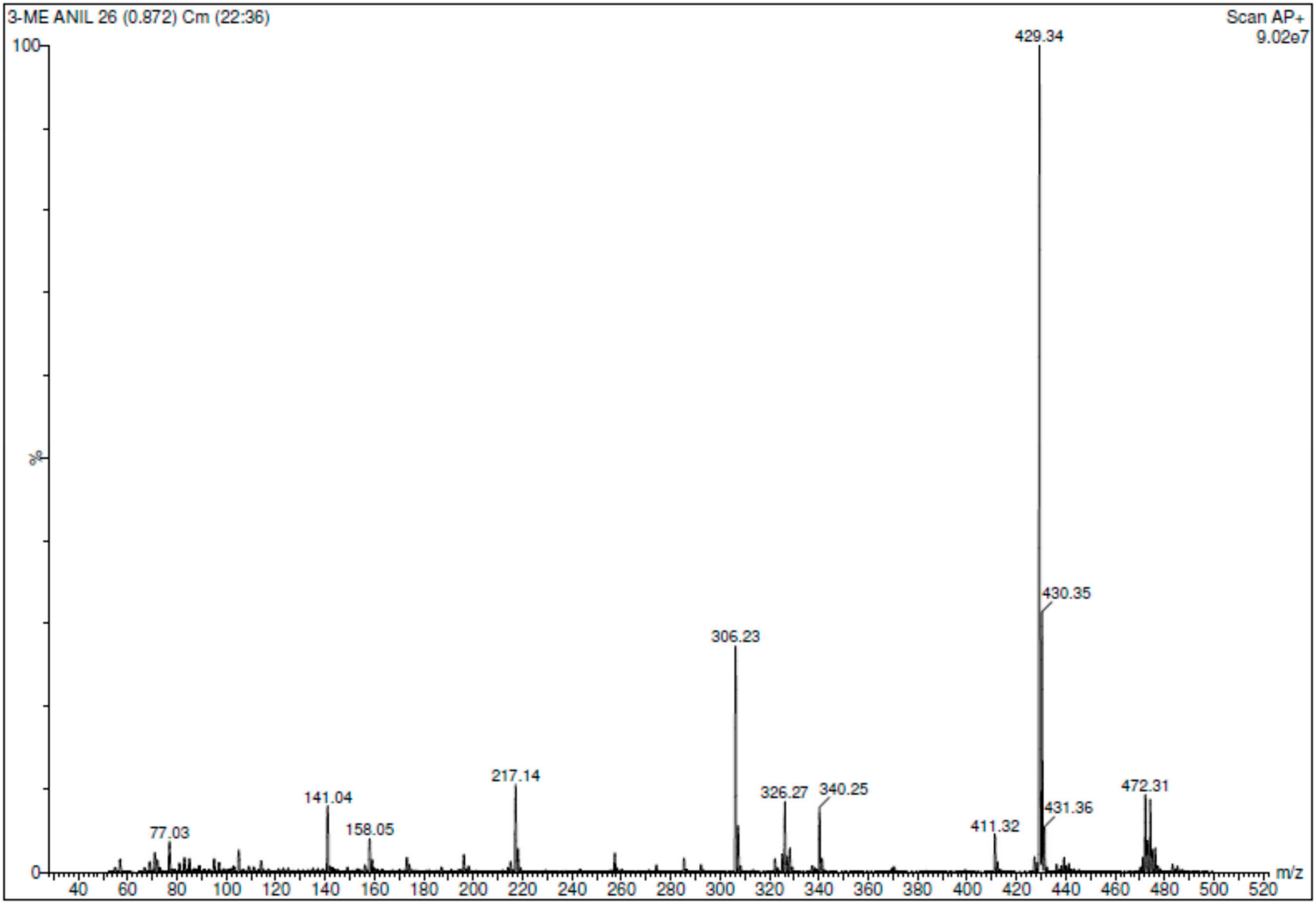
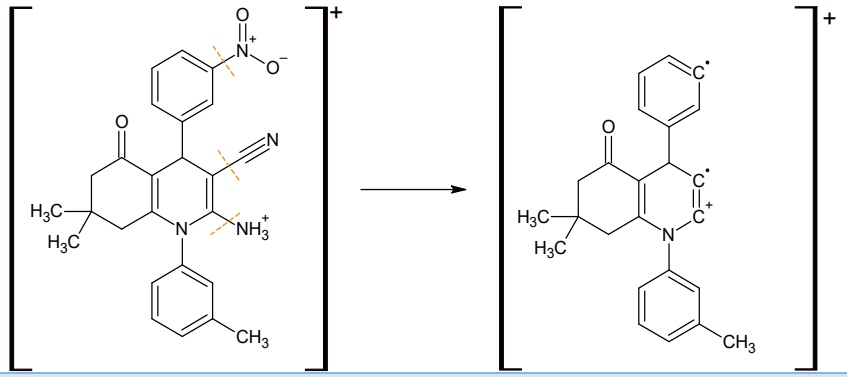
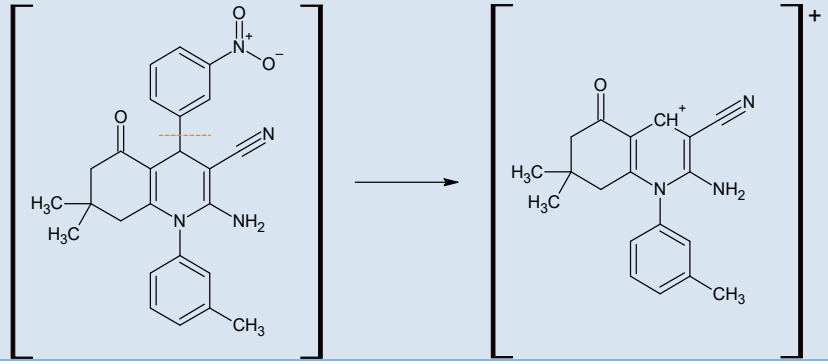


Figure S35 - MS spectrum of 5j

Table S3 - Fragmentation positions for peaks in MS spectrum of 5j

m/z	Fragmentation position and structure
429.34	$[M+H]^+$
340.25	
306.23	

1.11. Product 5k: 2-amino-1-(3-chlorophenyl)-4-(2,4-dichlorophenyl)-7,8-dimethyl-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

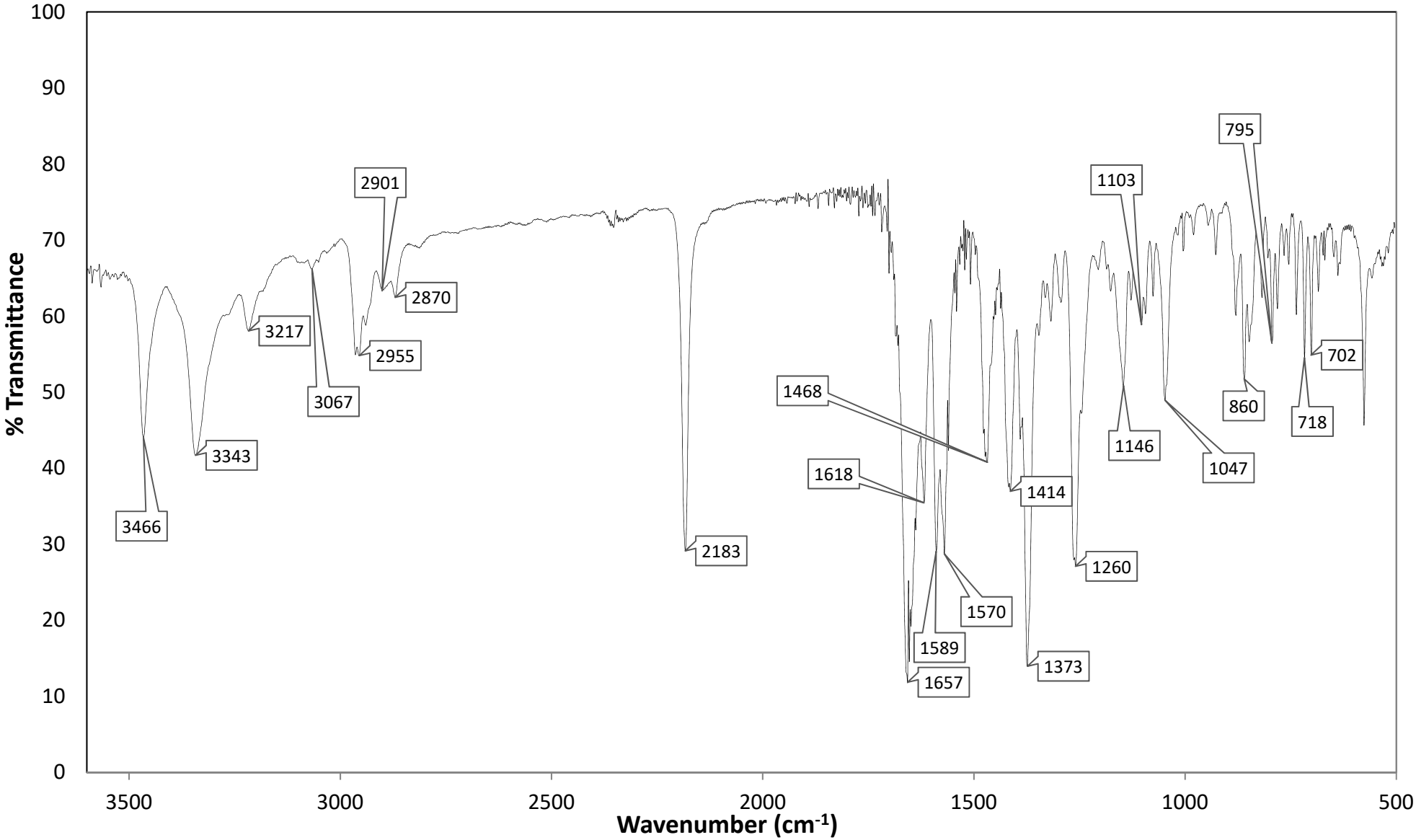


Figure S36 - IR spectrum of 5k

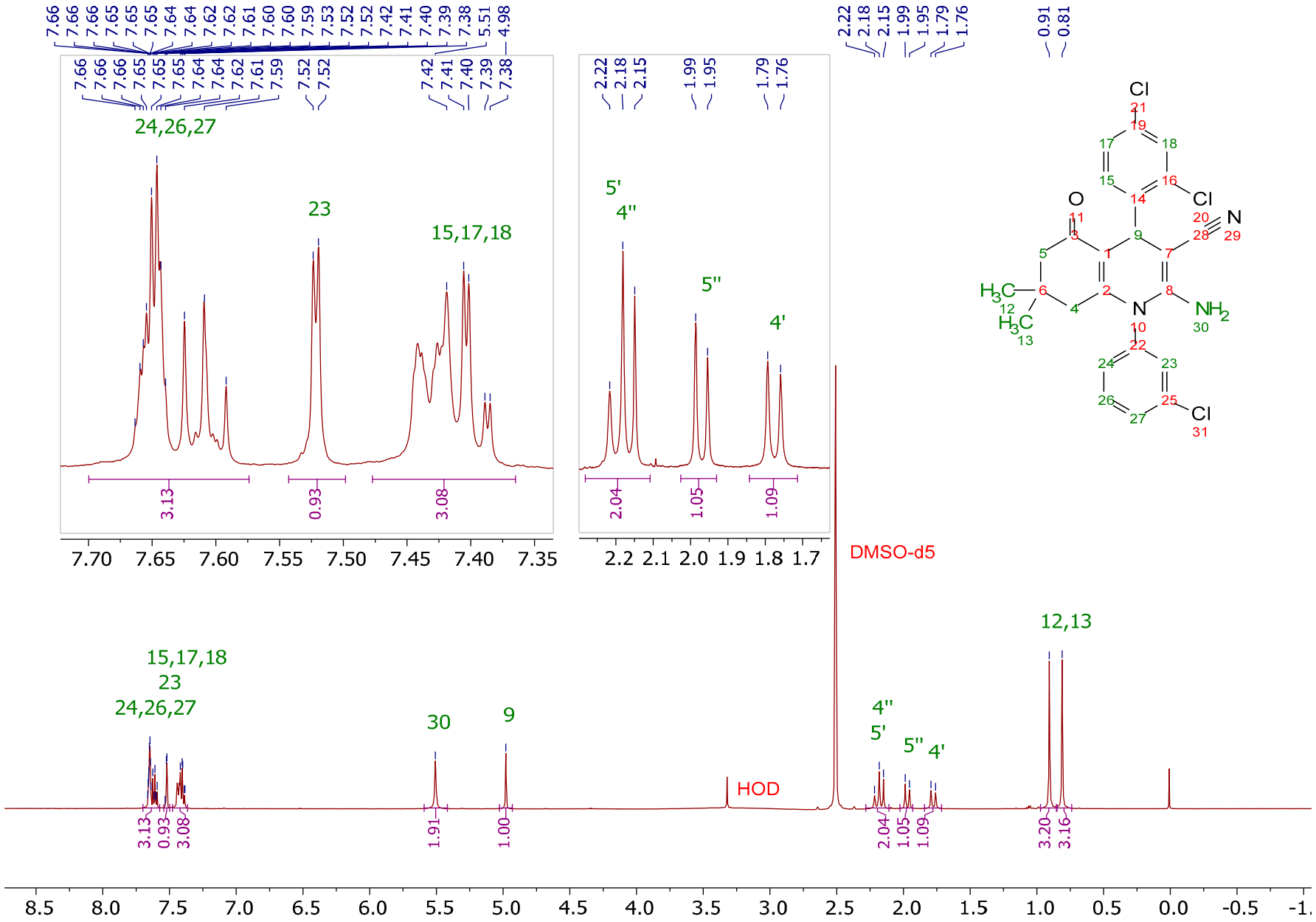


Figure S37 - ^{1}H NMR spectrum of 5k

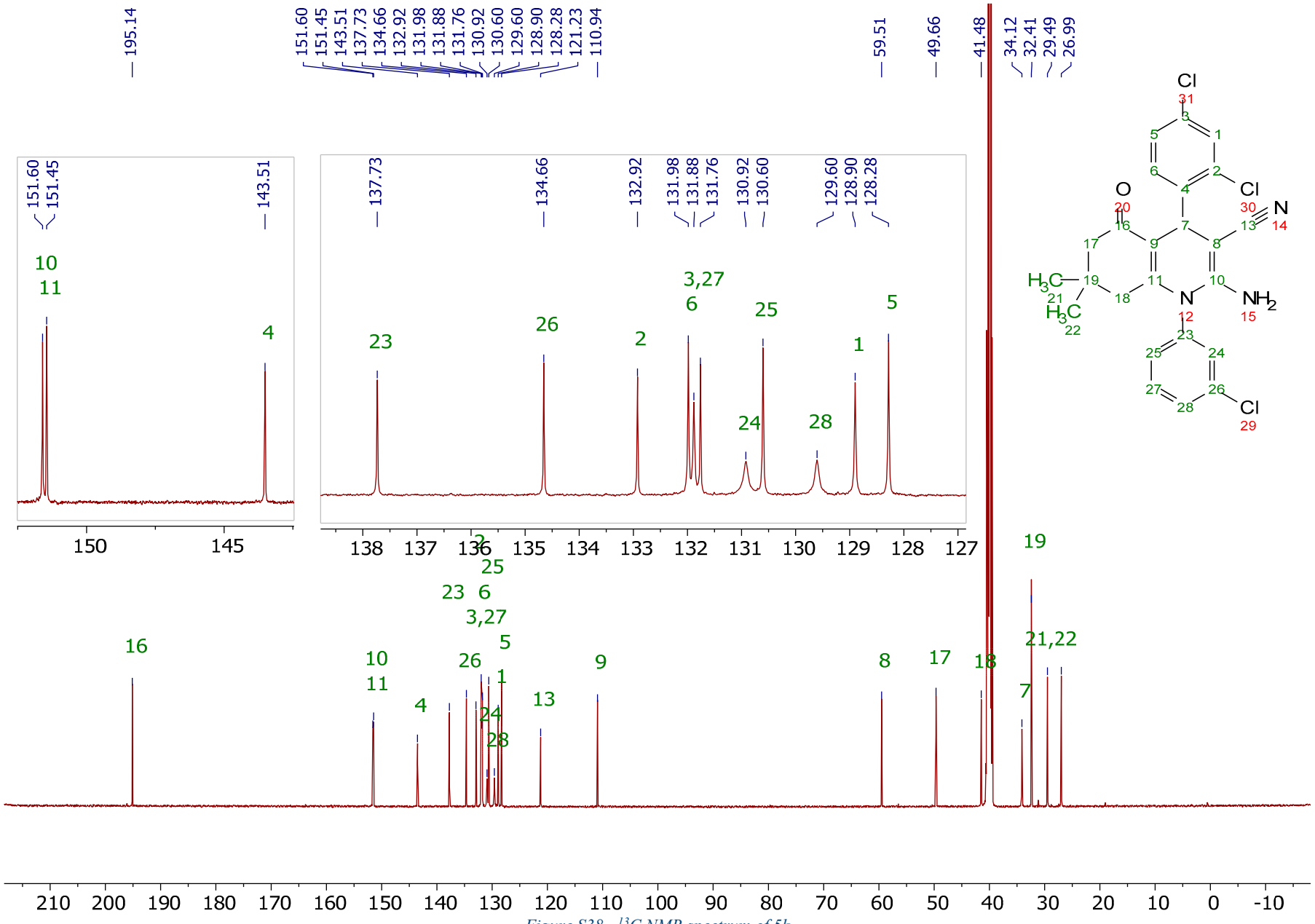


Figure S38 - ^{13}C NMR spectrum of 5k

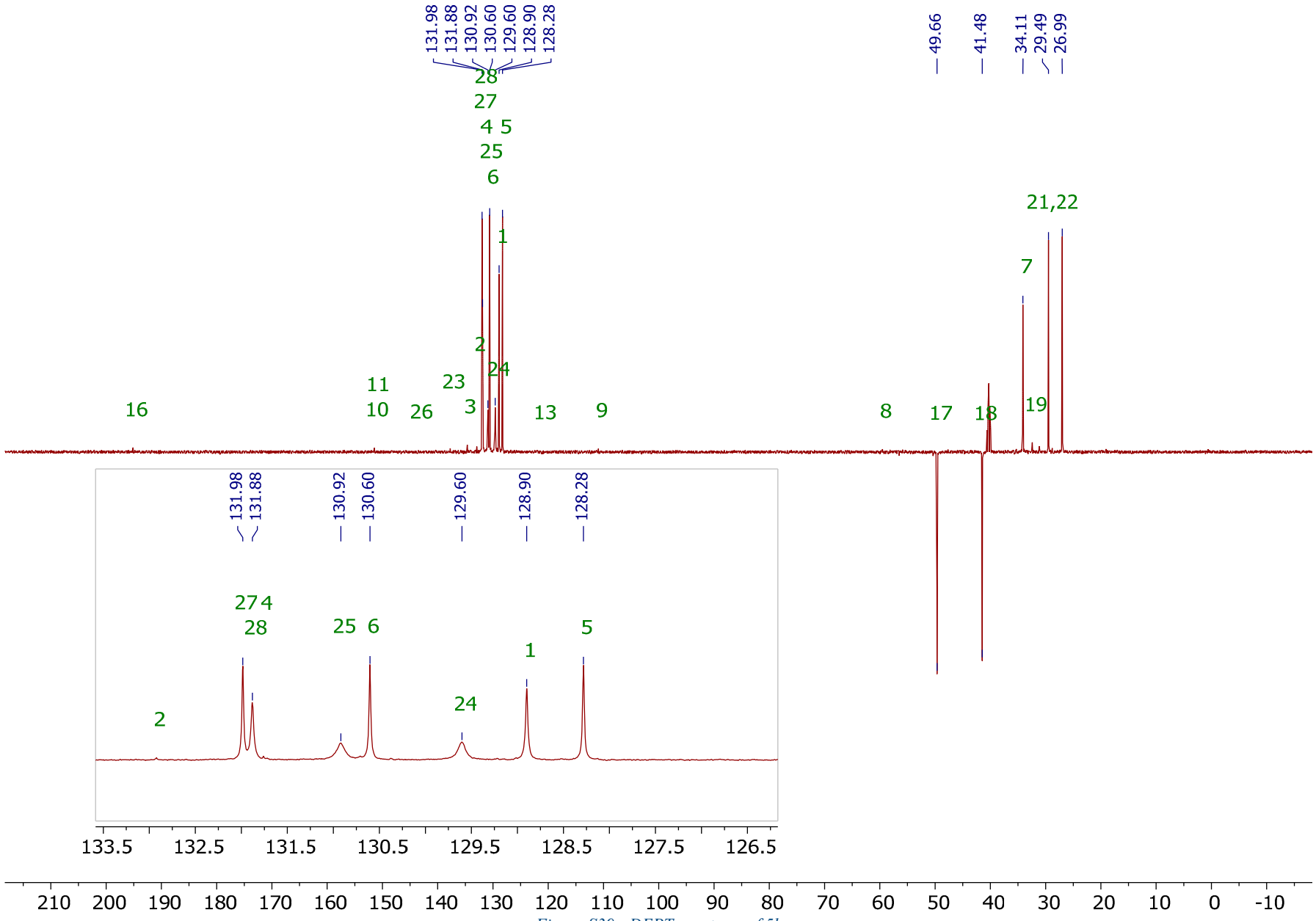


Figure S39 - DEPT spectrum of 5k

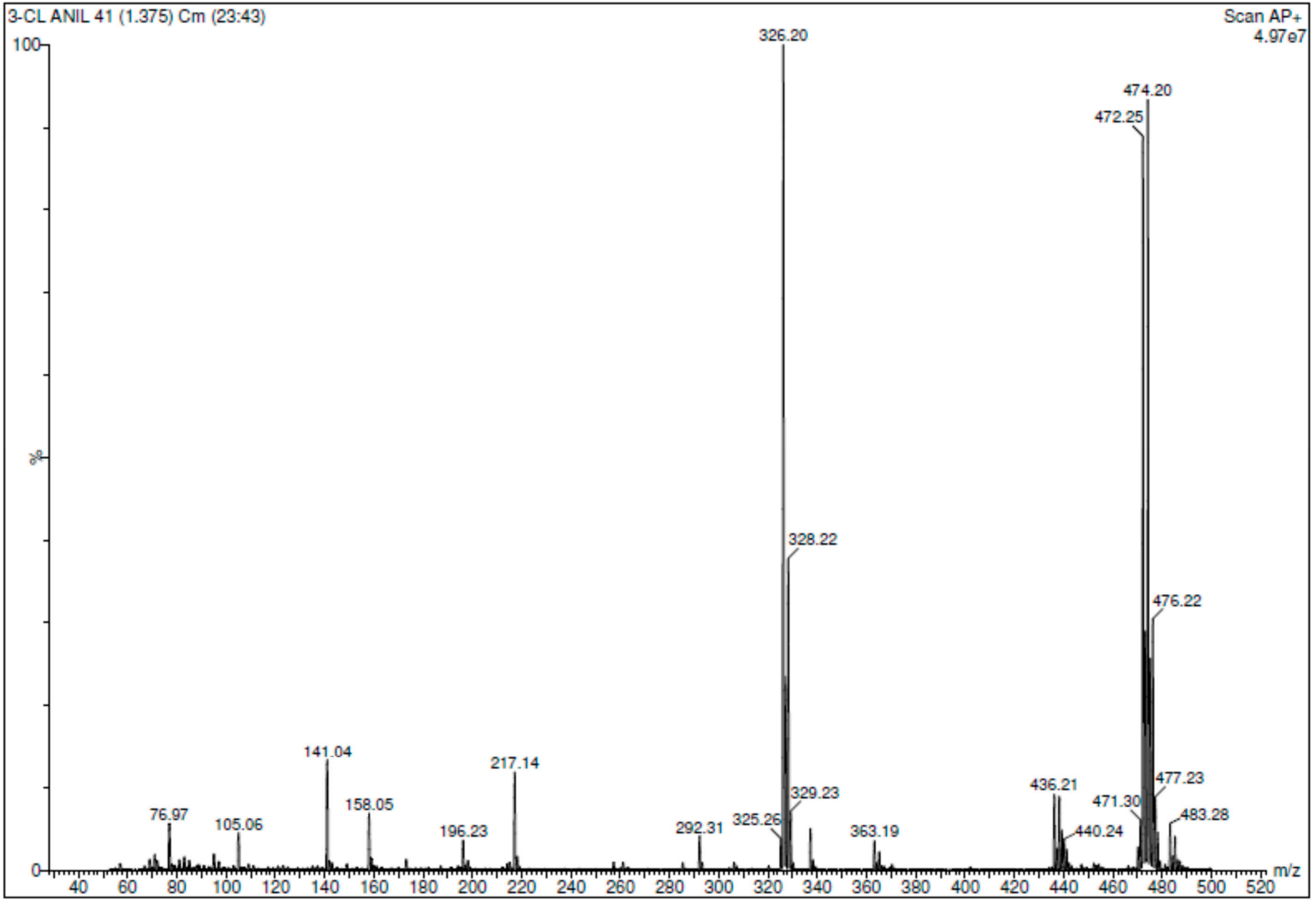
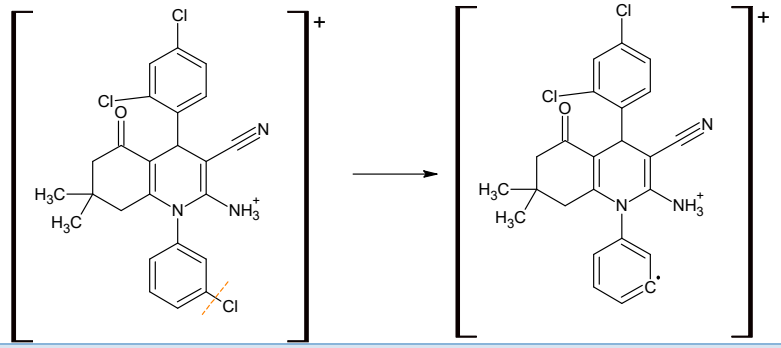
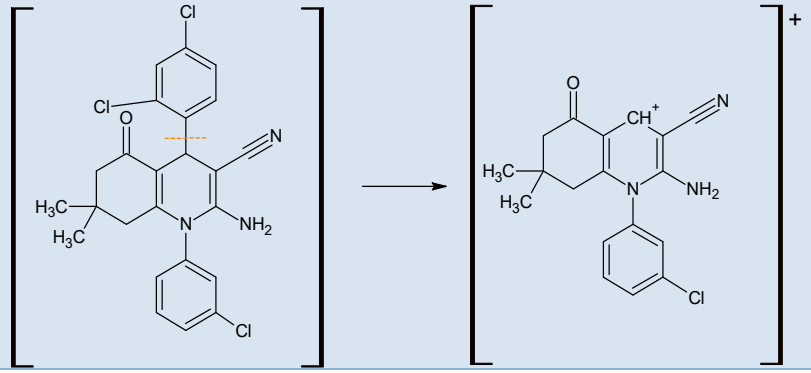


Figure S40 - MS spectrum of 5k

Table S4 - Fragmentation positions for peaks n MS spectrum of 5k

m/z	Fragmentation position and structure
471.30 – 477.23	$[M+H]^+$ Multiple isotopes
436.22-440.24	
326.20 – 328.22	

1.12. Product 5l: 2-amino-4-(2,4-dichlorophenyl)-7,8-dimethyl-1-(3-nitrophenyl)-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

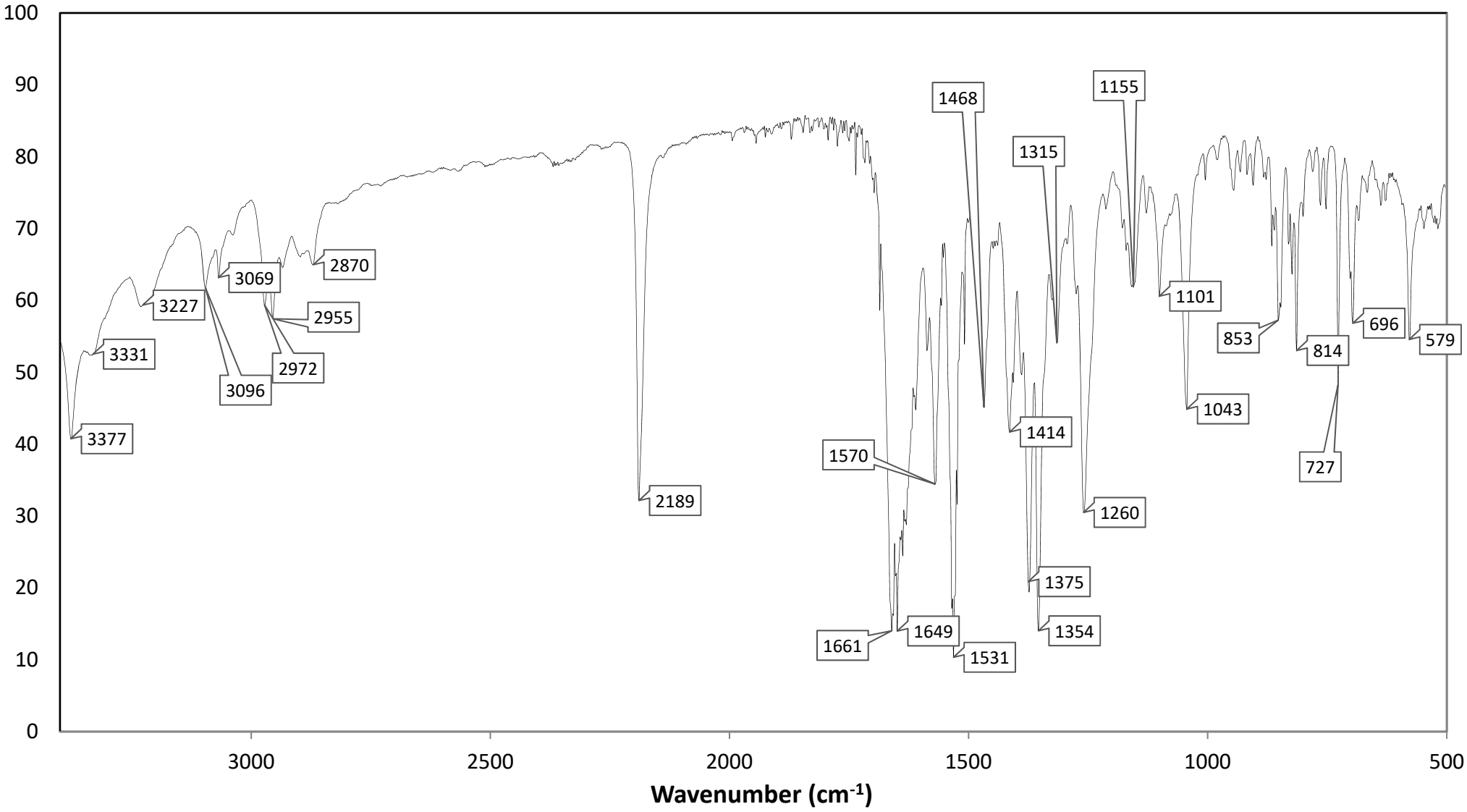


Figure S41 - IR spectrum of 5l

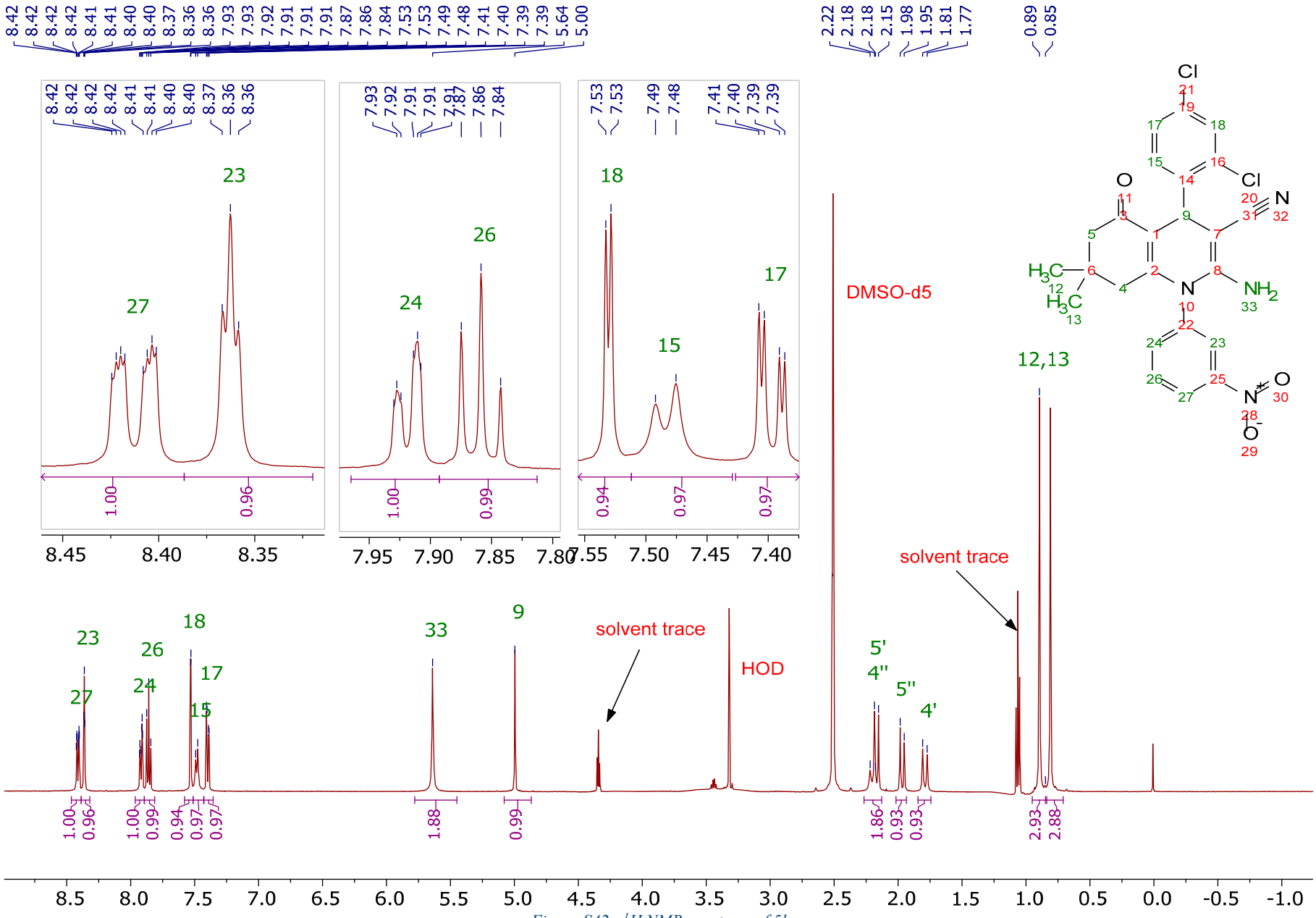


Figure S42 - ^{1}H NMR spectrum of 5l

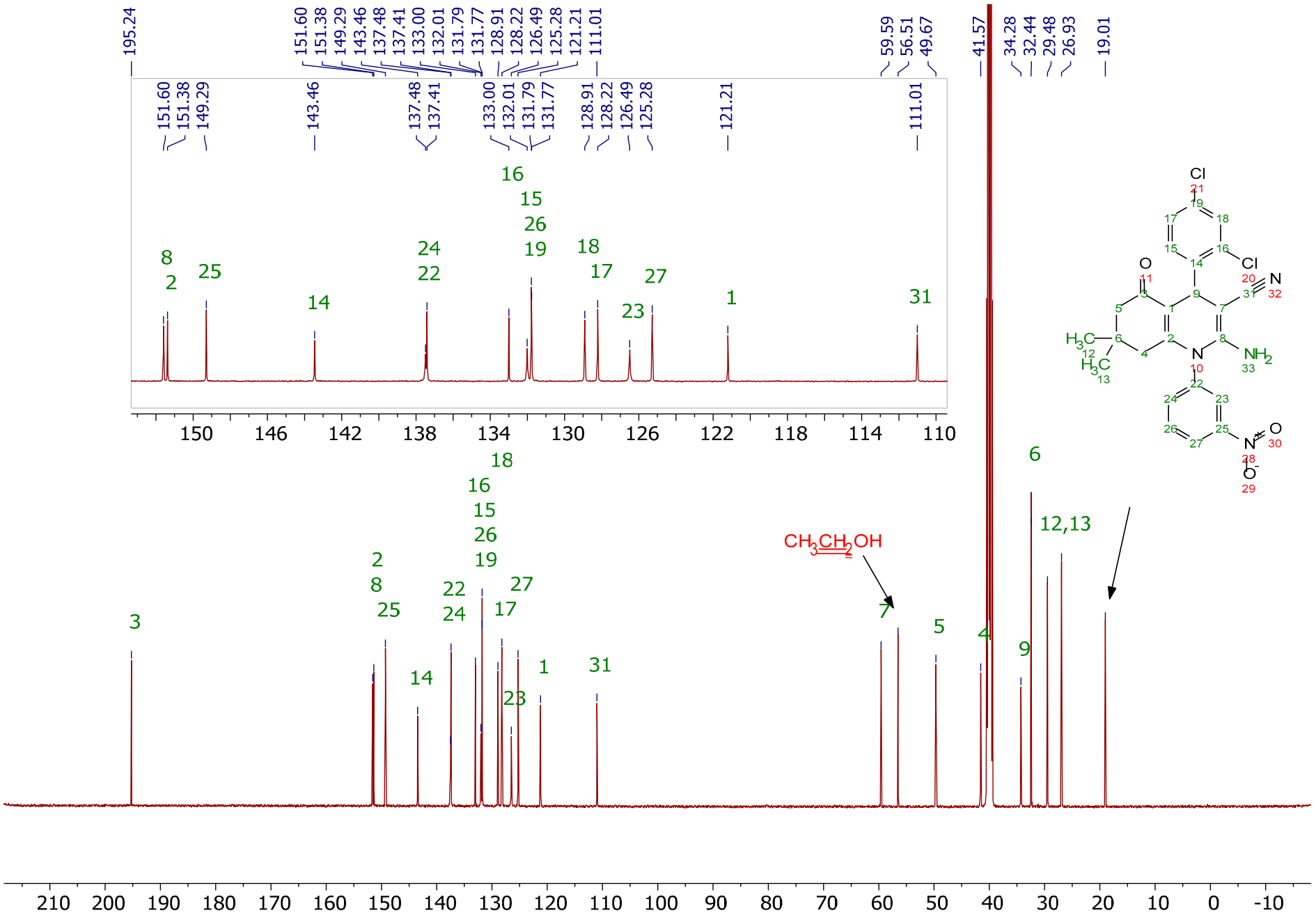


Figure S43 - ^{13}C NMR spectrum of 5l

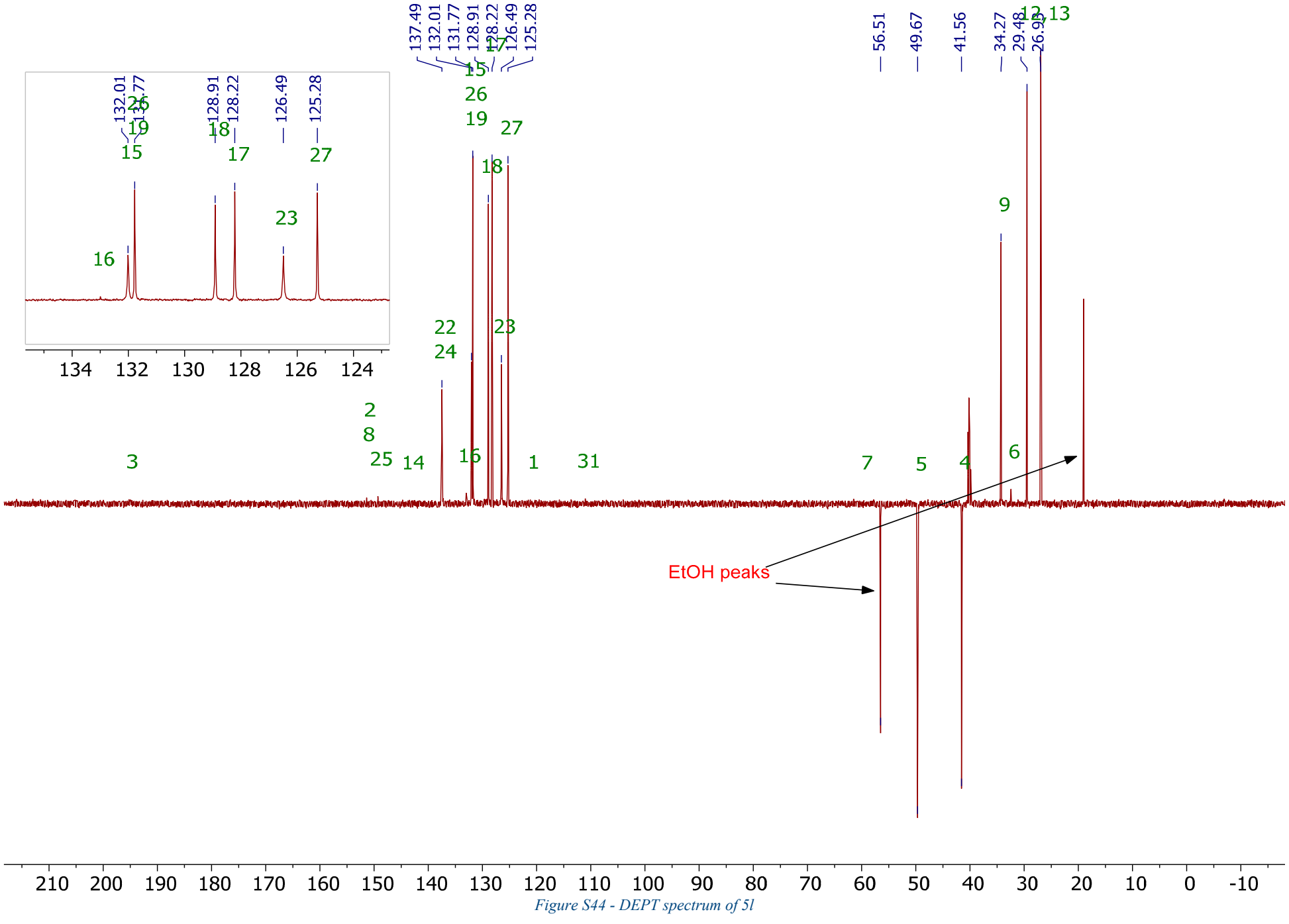


Figure S44 - DEPT spectrum of 5l

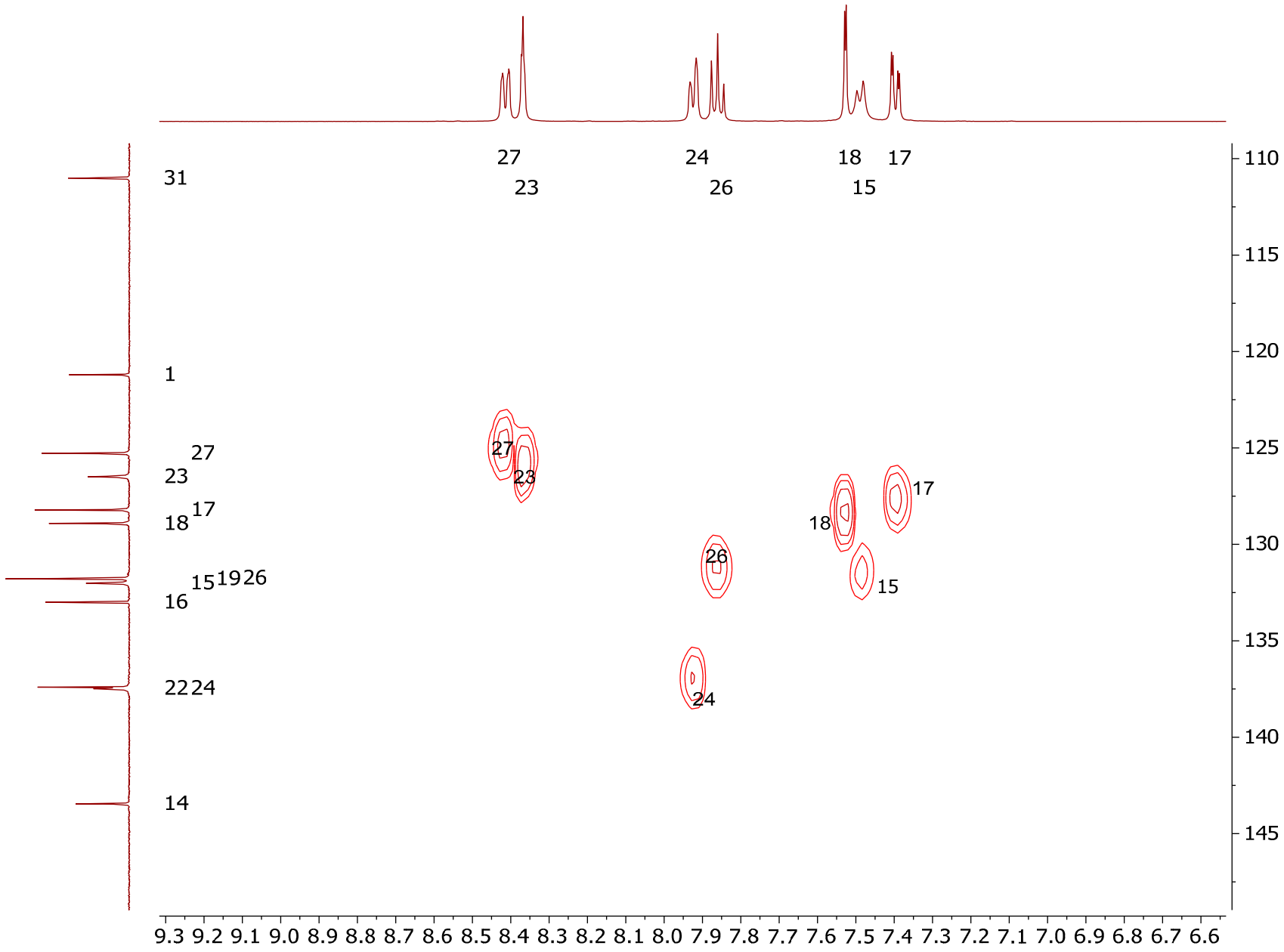


Figure S45 - HSQC spectrum of 5l

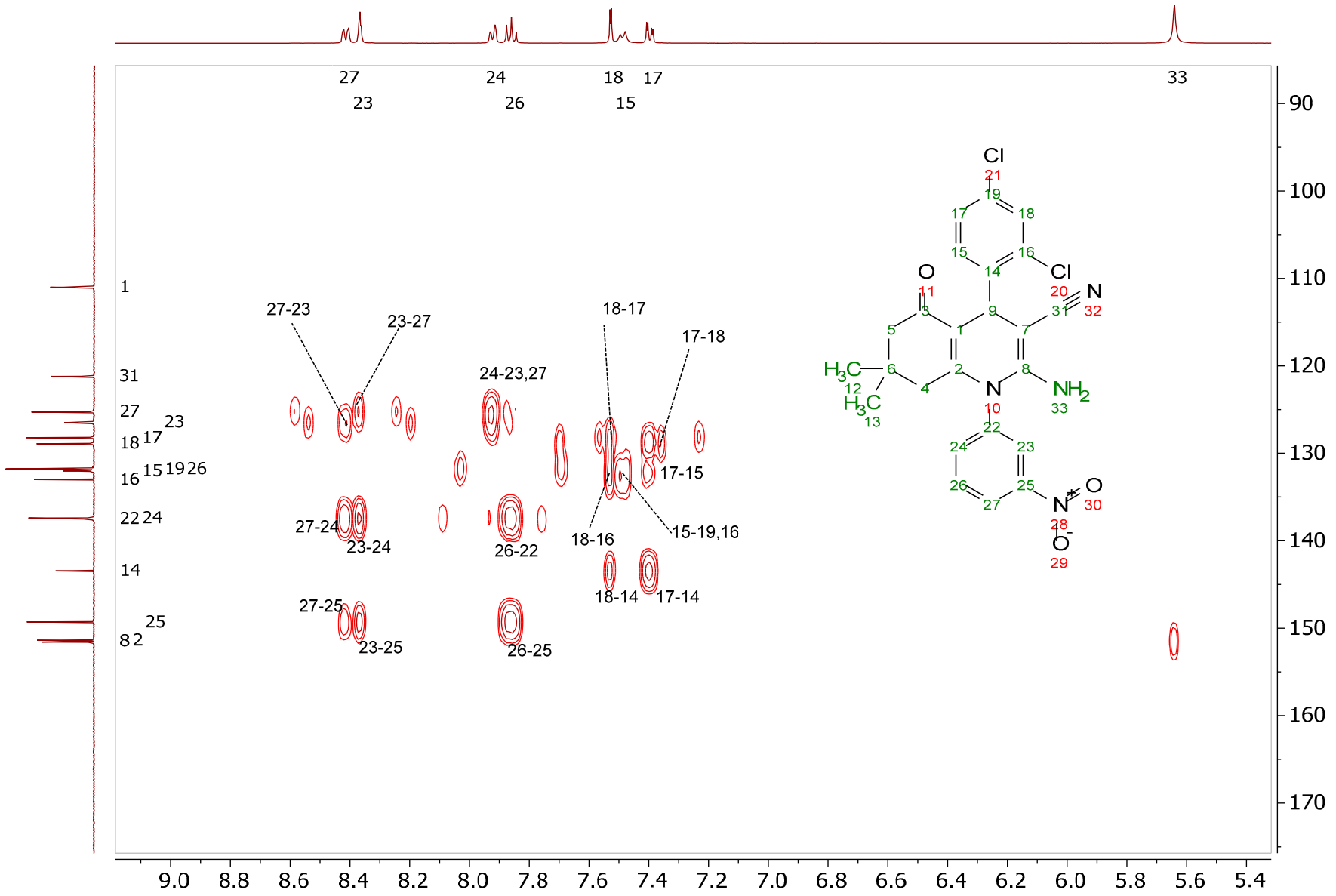


Figure S46 – Downfield region of HMBC spectrum of 5l

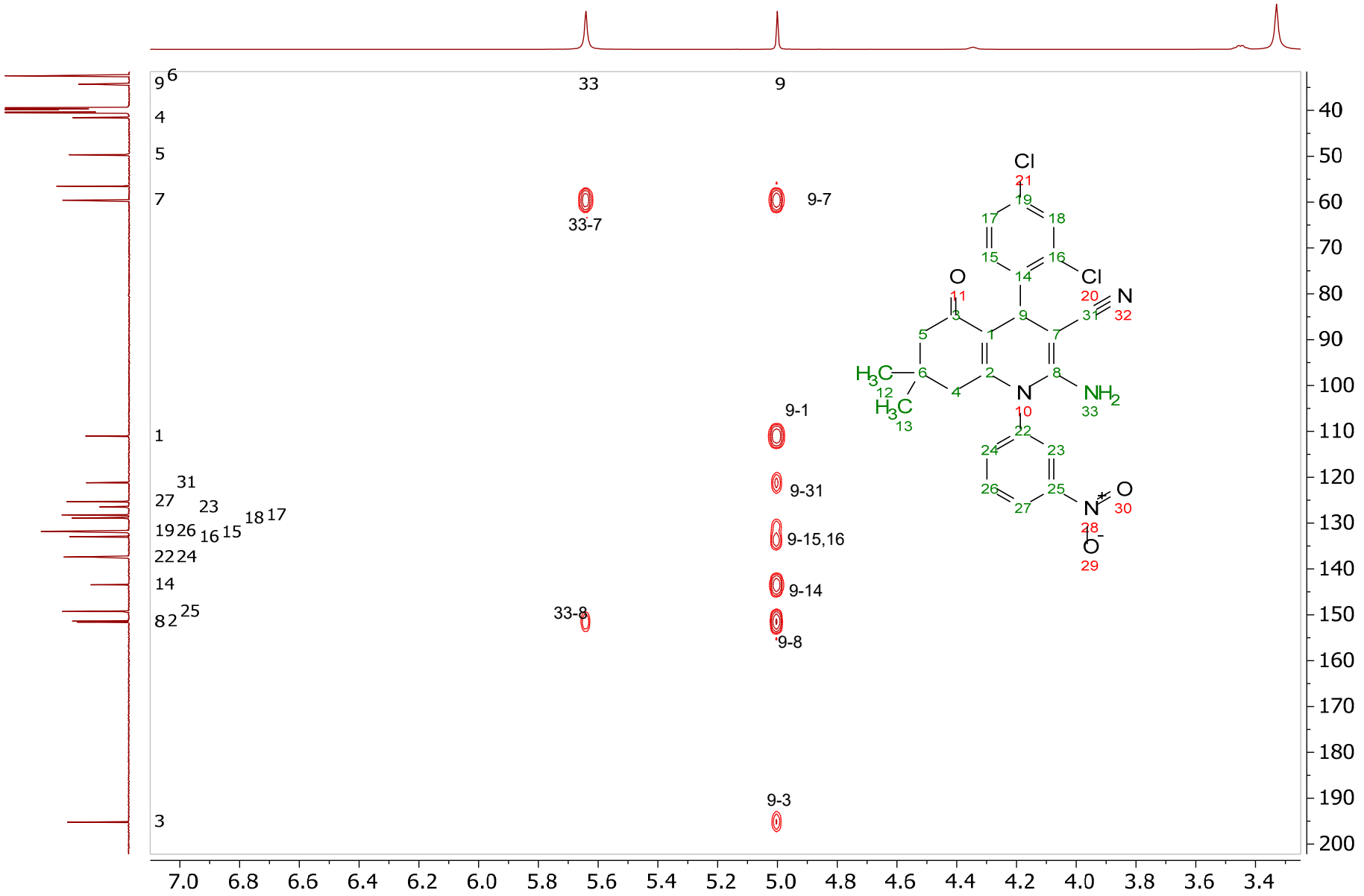


Figure S47 - Upfield region of HMBC spectrum of 5l

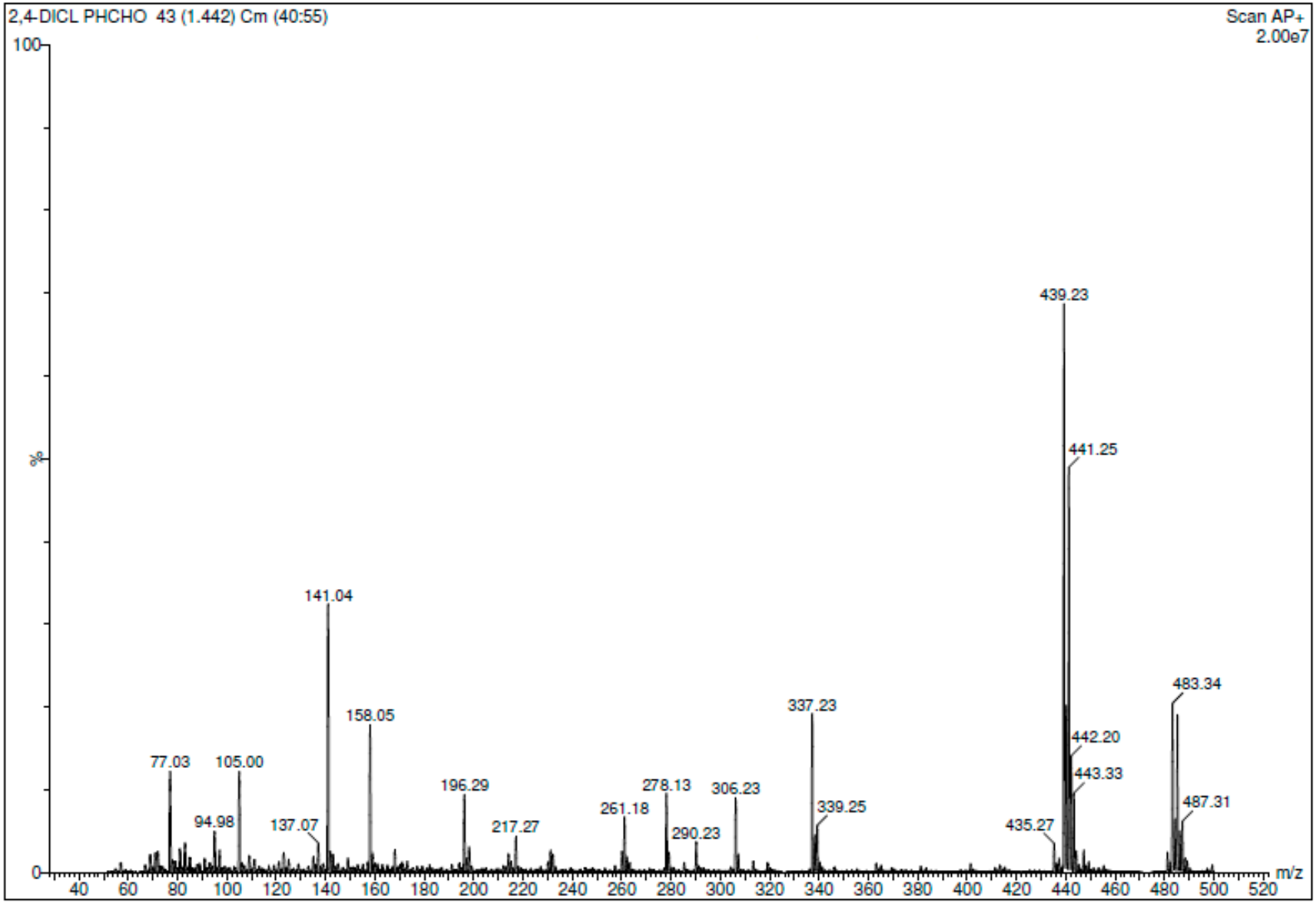
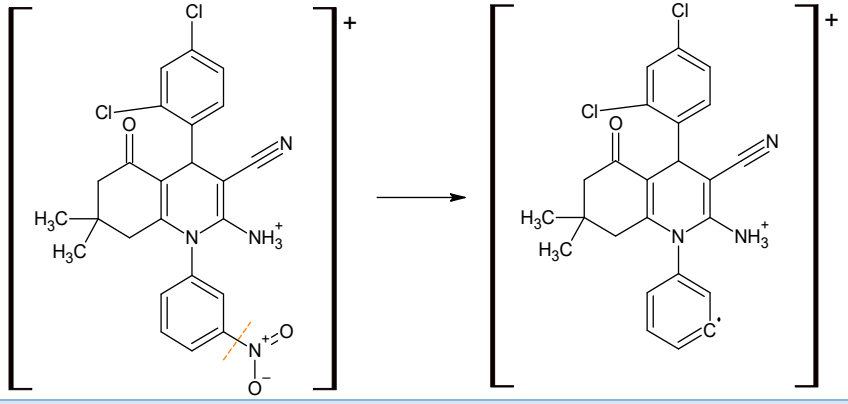
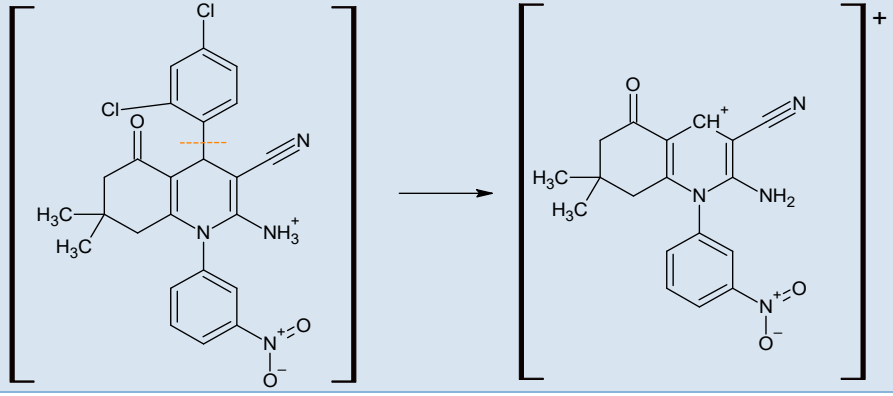
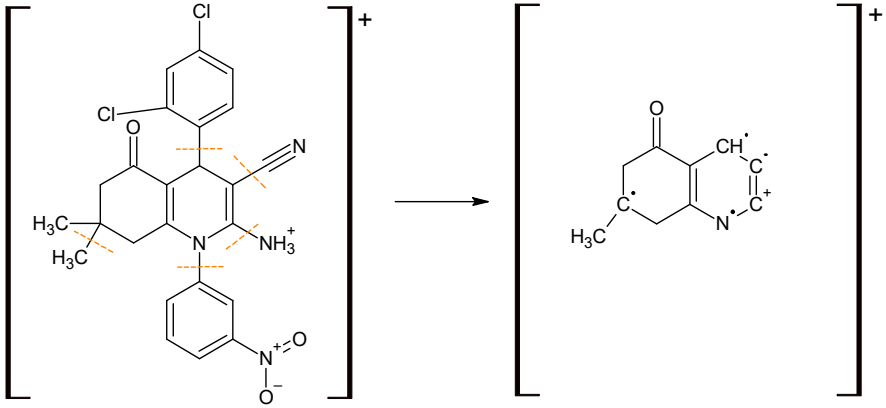


Figure S48 - MS spectrum of 5l

Table S5 – Peaks and fragmentation positions for 5l

m/z	Fragmentation position and structure
483.34-487.31	$[M+H]^+$ Multiple isotopes
439.23	
337.23	
158.05	

1.13. Product 5m: 2-amino-1-(4-methoxyphenyl)-7,8-dimethyl-4-(3-methylphenyl)-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

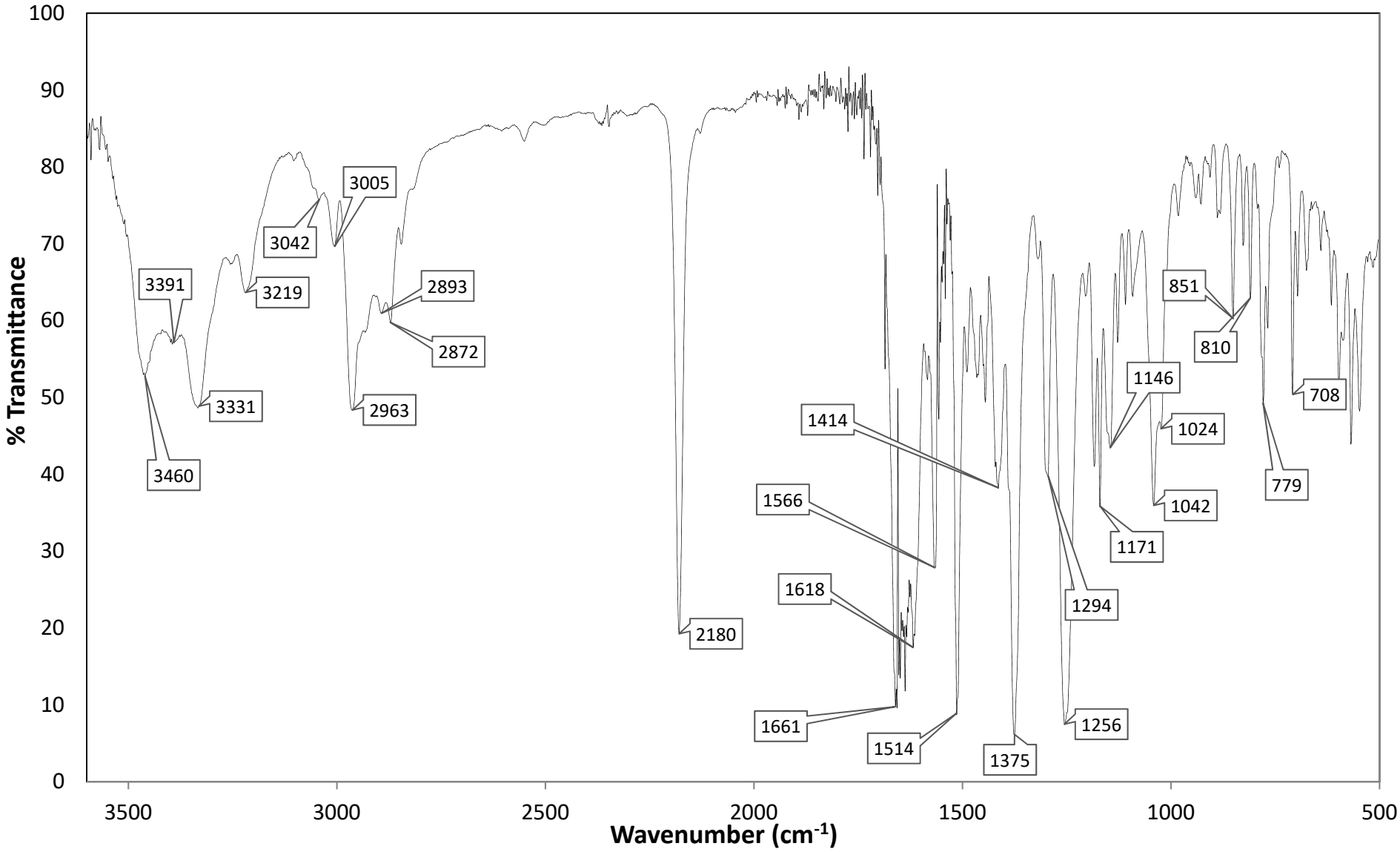


Figure S49 - IR spectrum of 5m

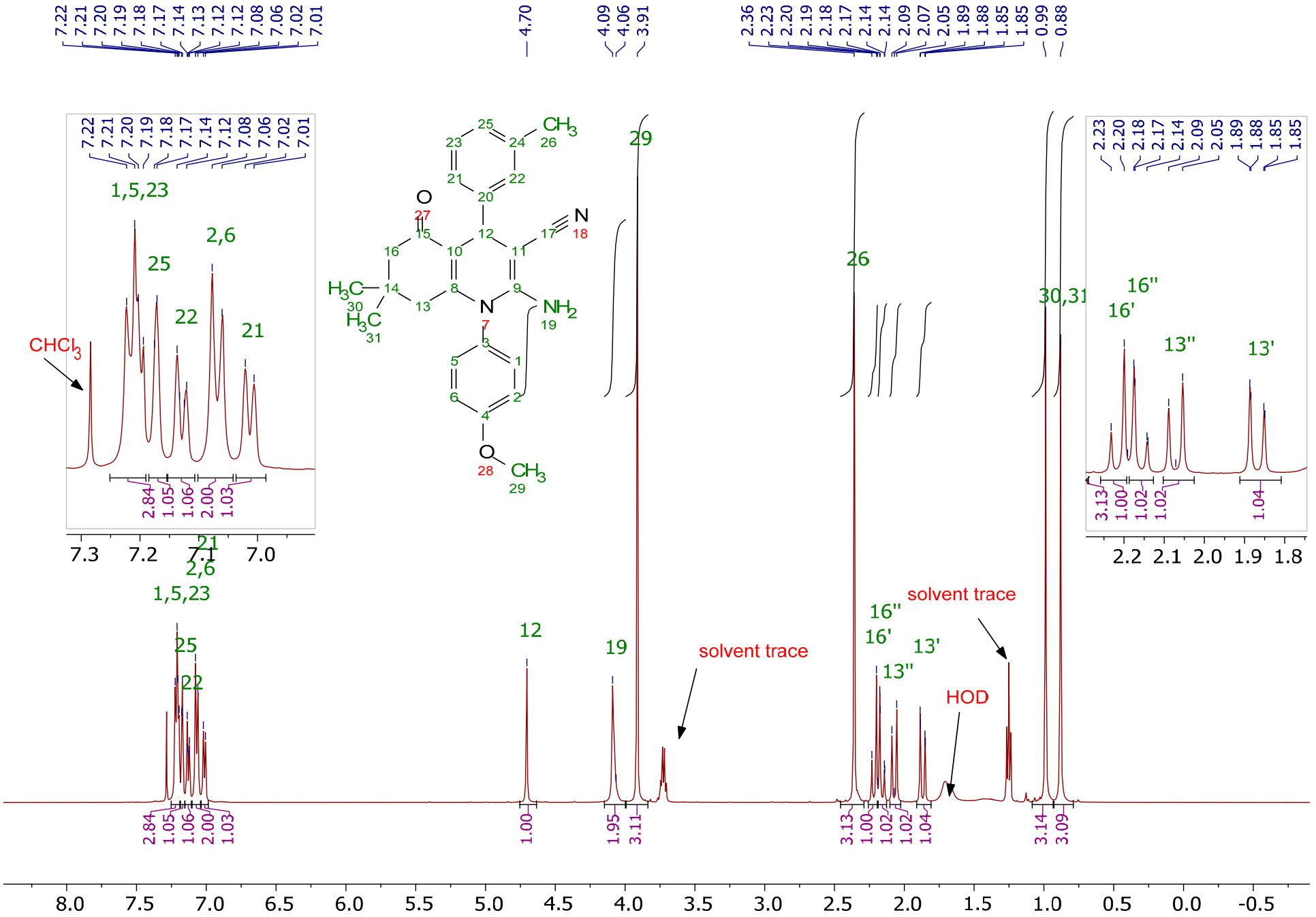


Figure S50 - ^{1}H NMR spectrum of 5m

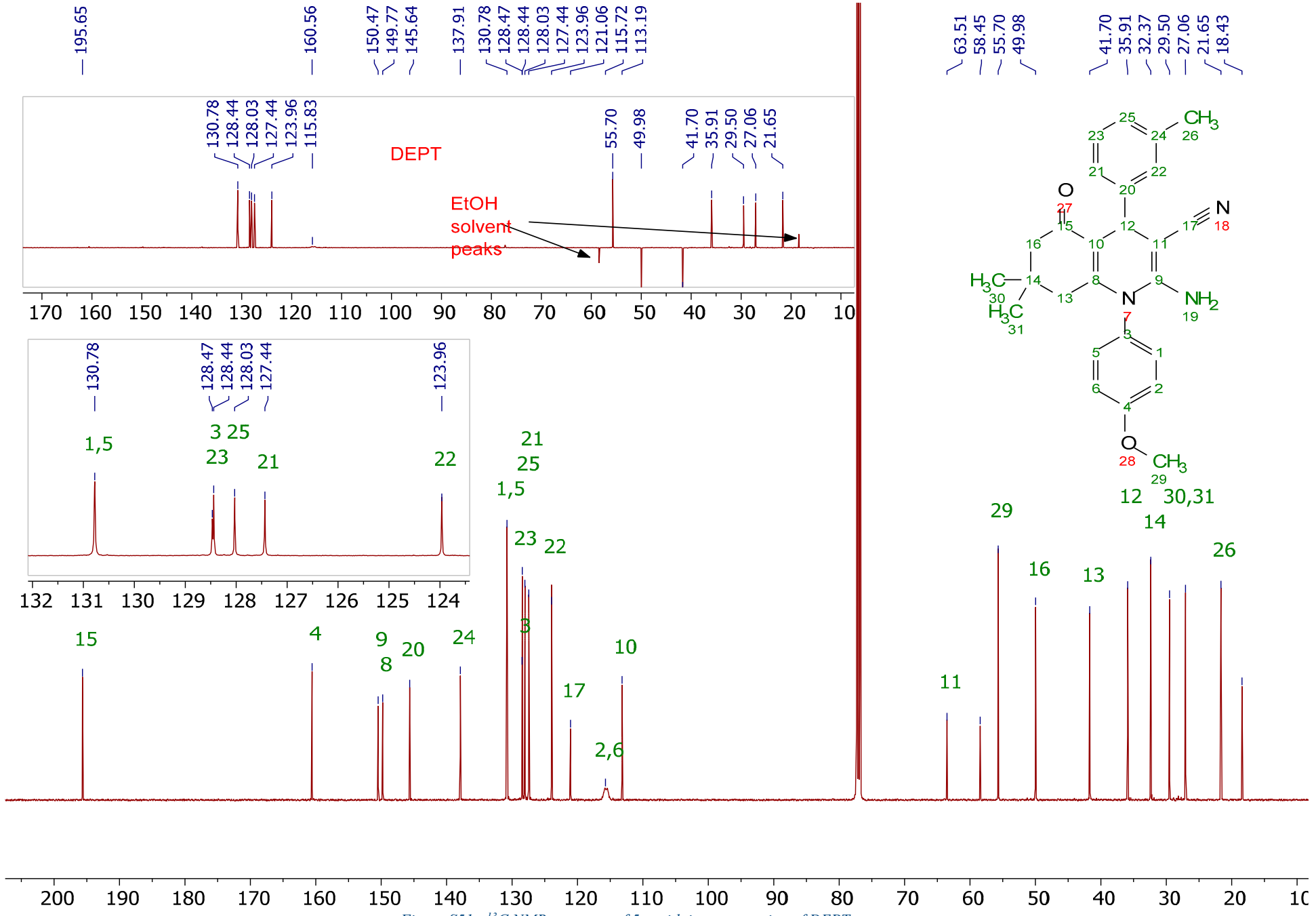


Figure S51 - ^{13}C NMR spectrum of 5m with inset expansion of DEPT spectrum

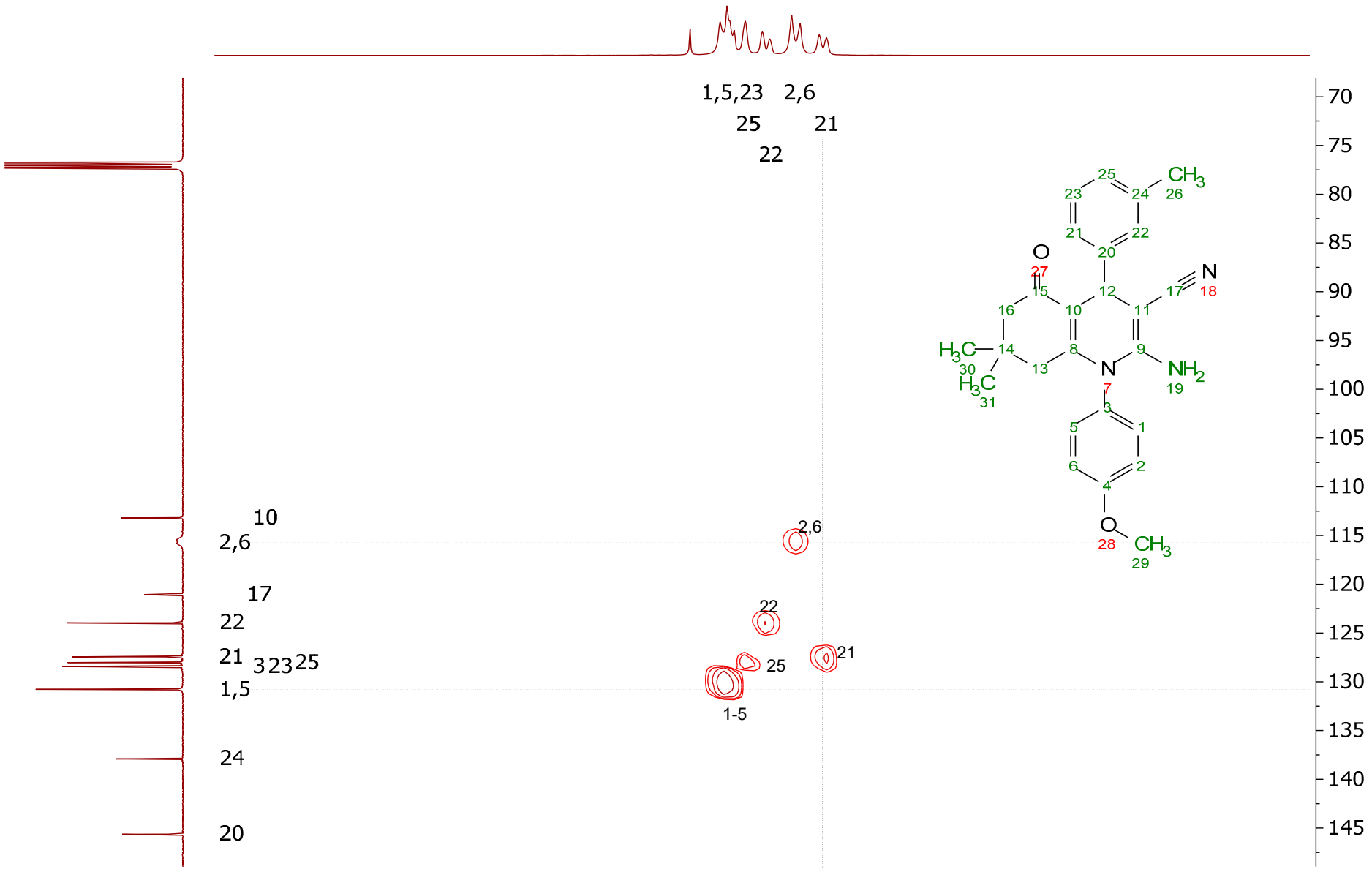


Figure S52 – Downfield region of HSQC NMR spectrum of 5m

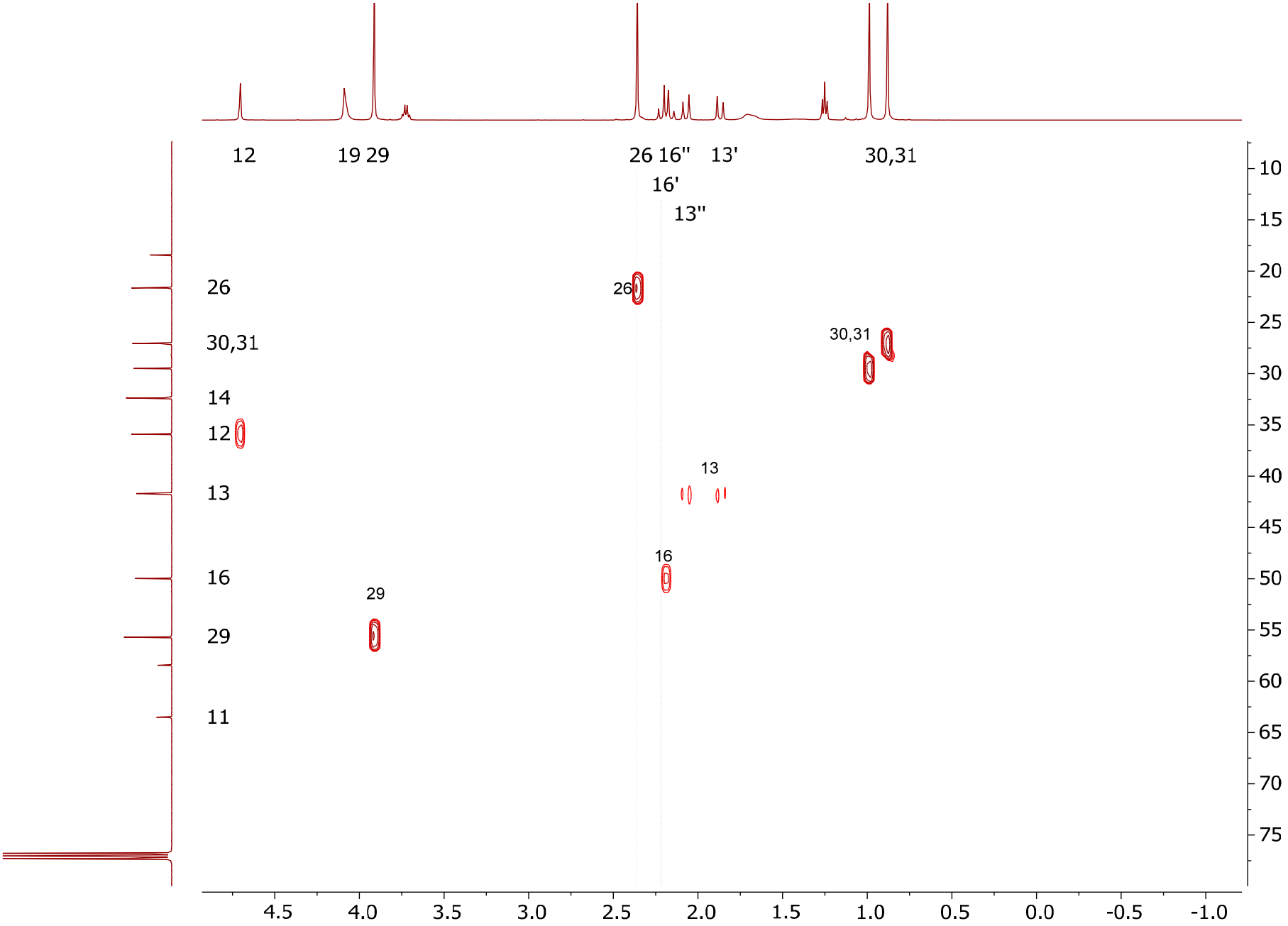


Figure S53 - Upfield region of HSQC spectrum of 5m

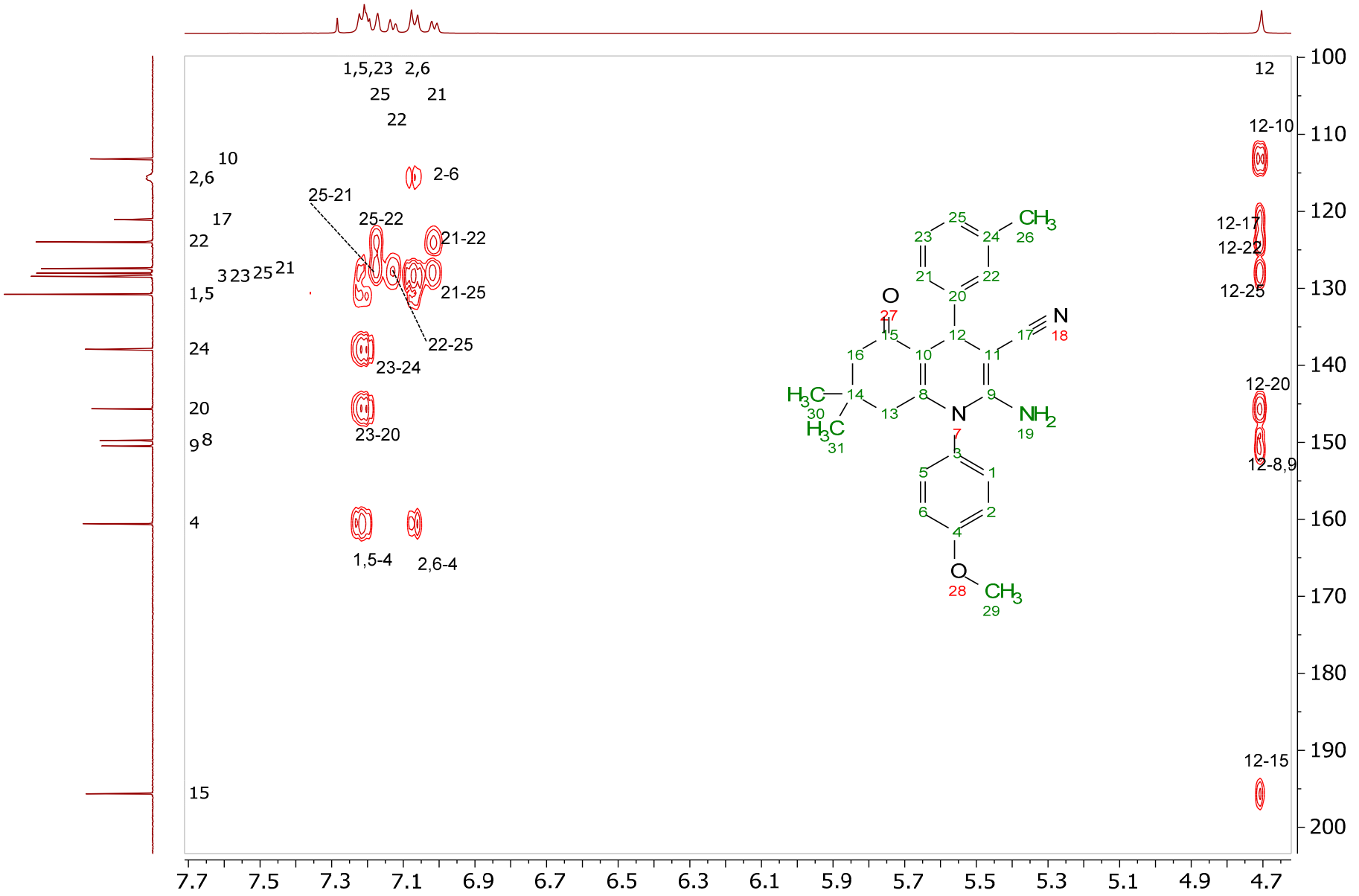


Figure S54 - Downfield region of HMBC NMR spectrum of 5m

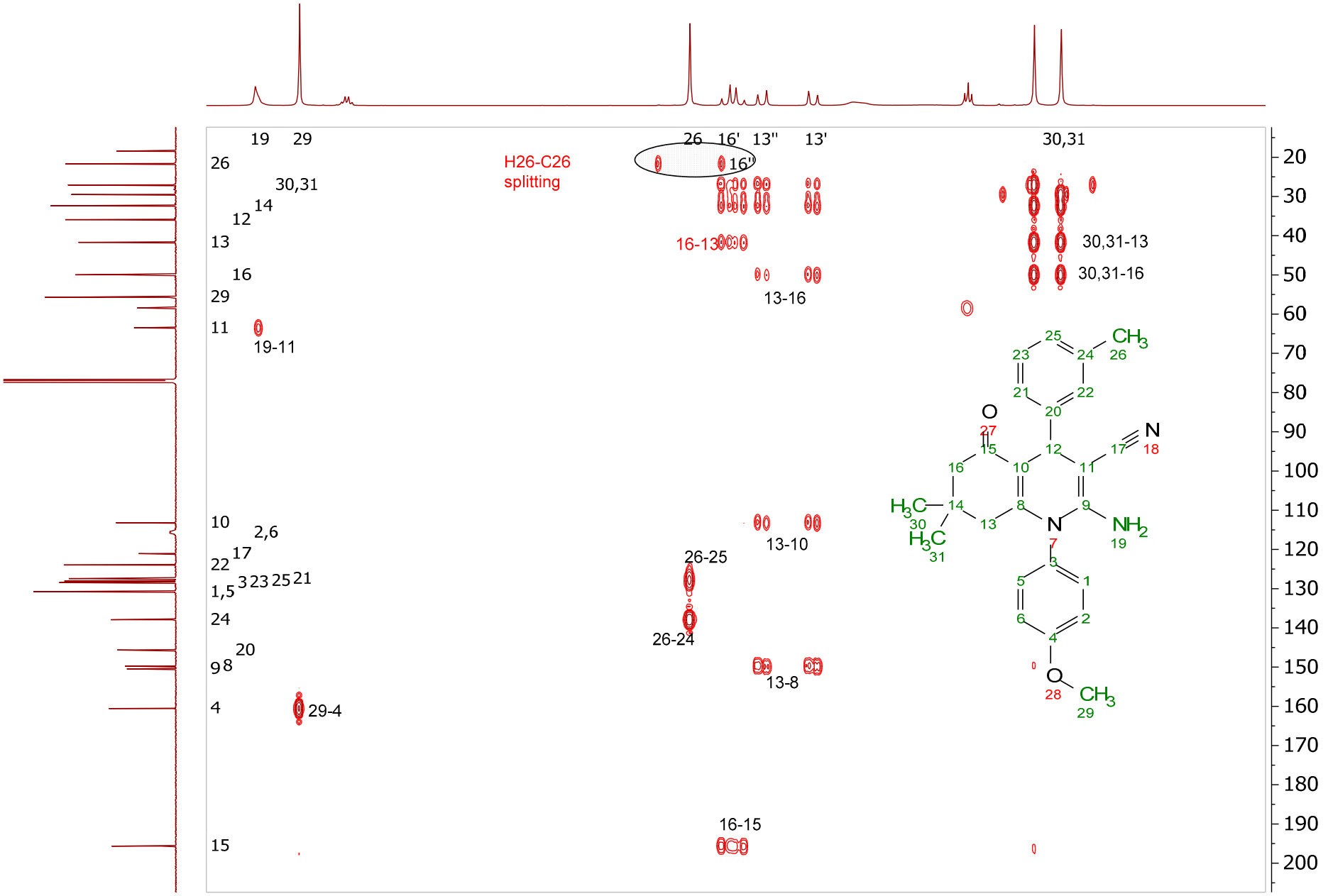


Figure S55 - Upfield region of HMBC NMR spectrum of 5m

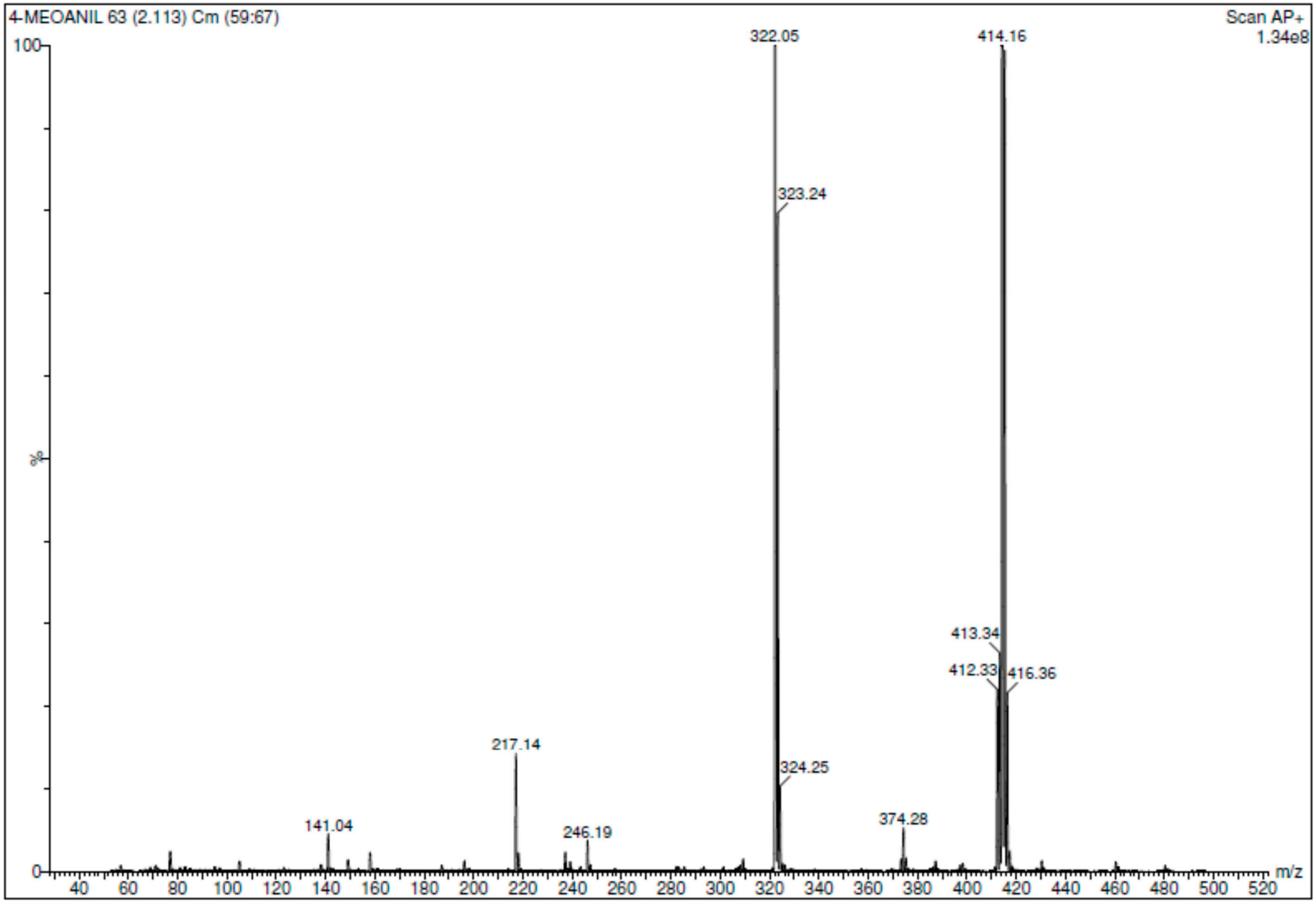
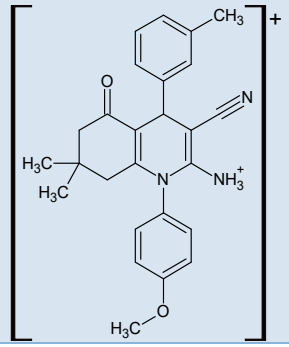
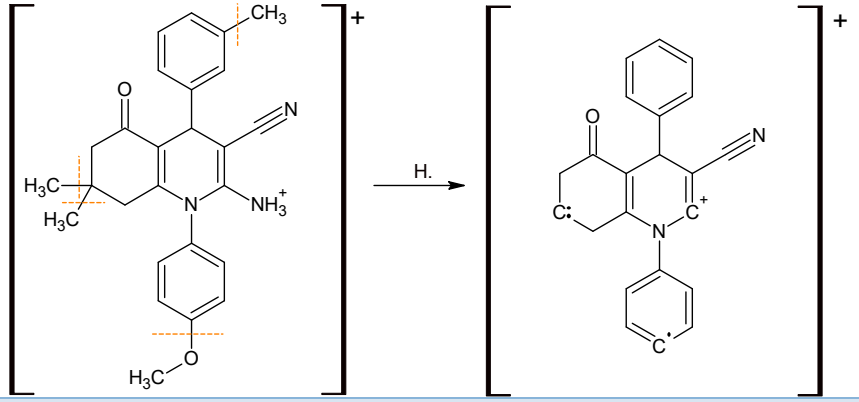
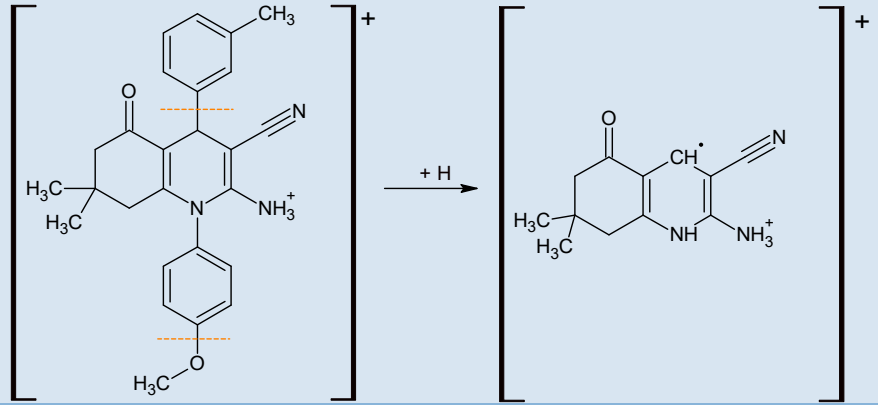


Figure S56 - MS spectrum of 5m

Table S6 - Fragmentation positions for peaks in MS spectrum of 29m

m/z	Fragmentation position and structure
414.16	
322.05	
217.14	

1.14. Product 5n: 2-amino-4-(2,4-dichlorophenyl)-7,8-dimethyl-1-(methylphenyl)-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

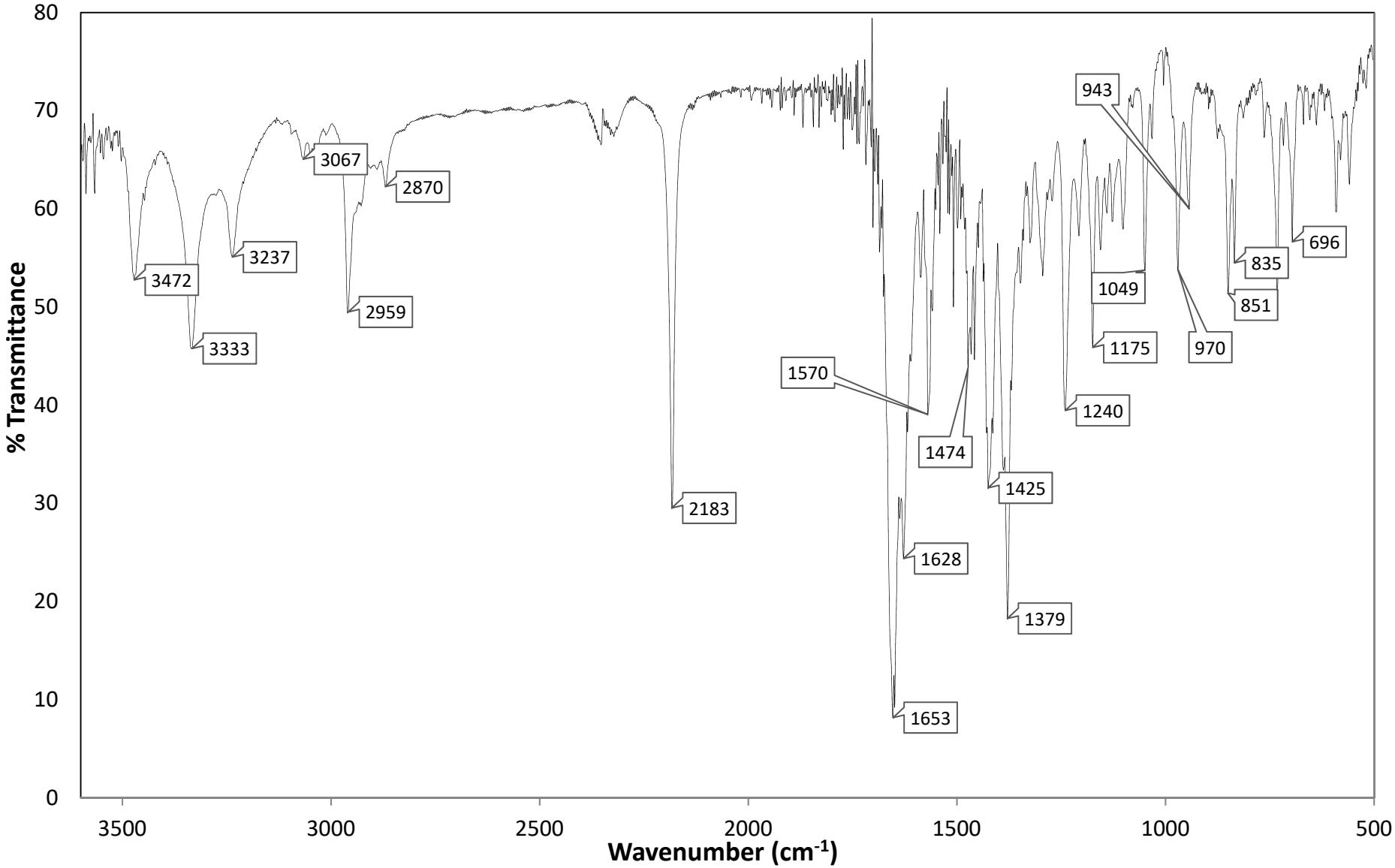


Figure S57 - IR spectrum of 5n

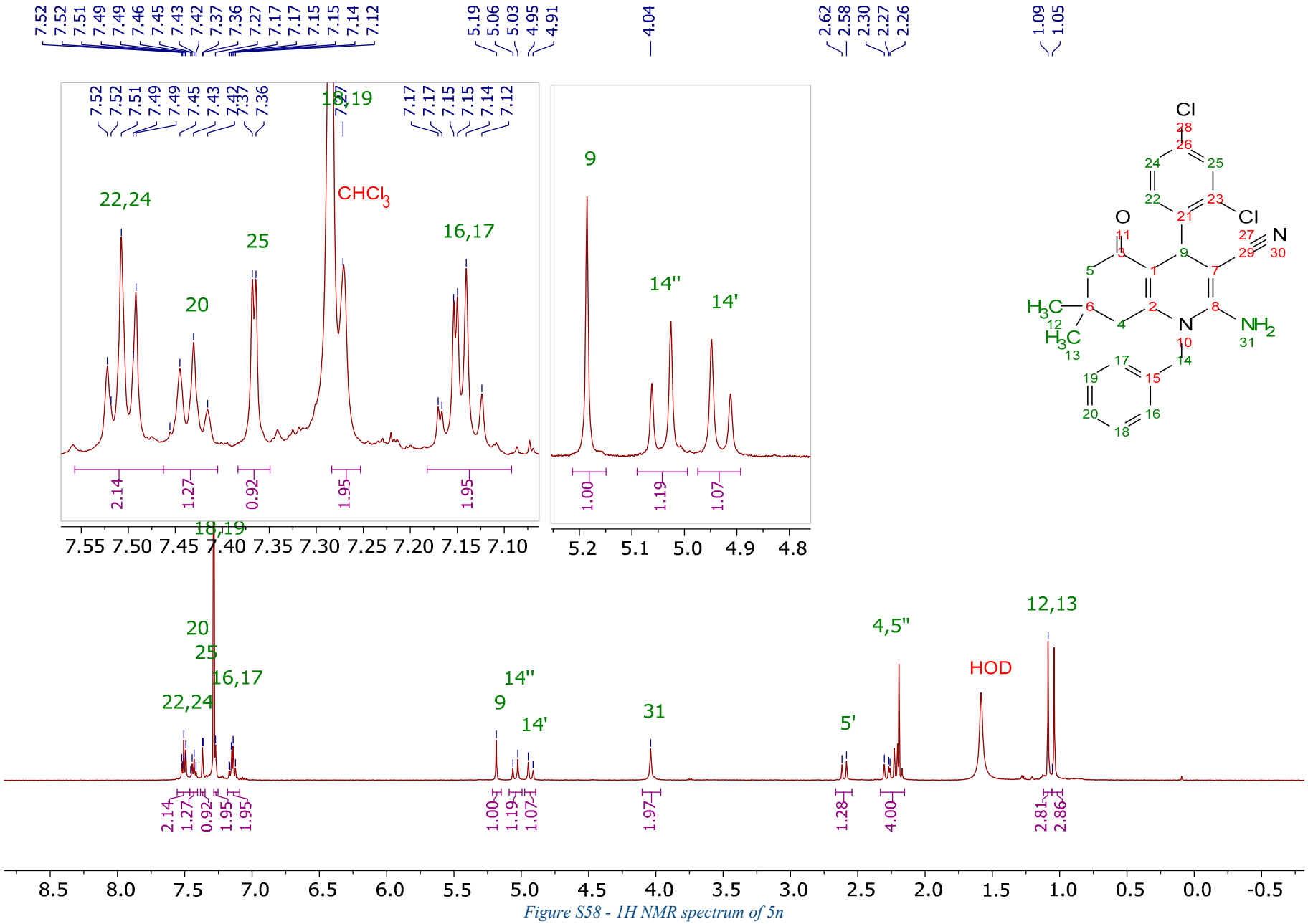


Figure S58 - 1H NMR spectrum of 5n

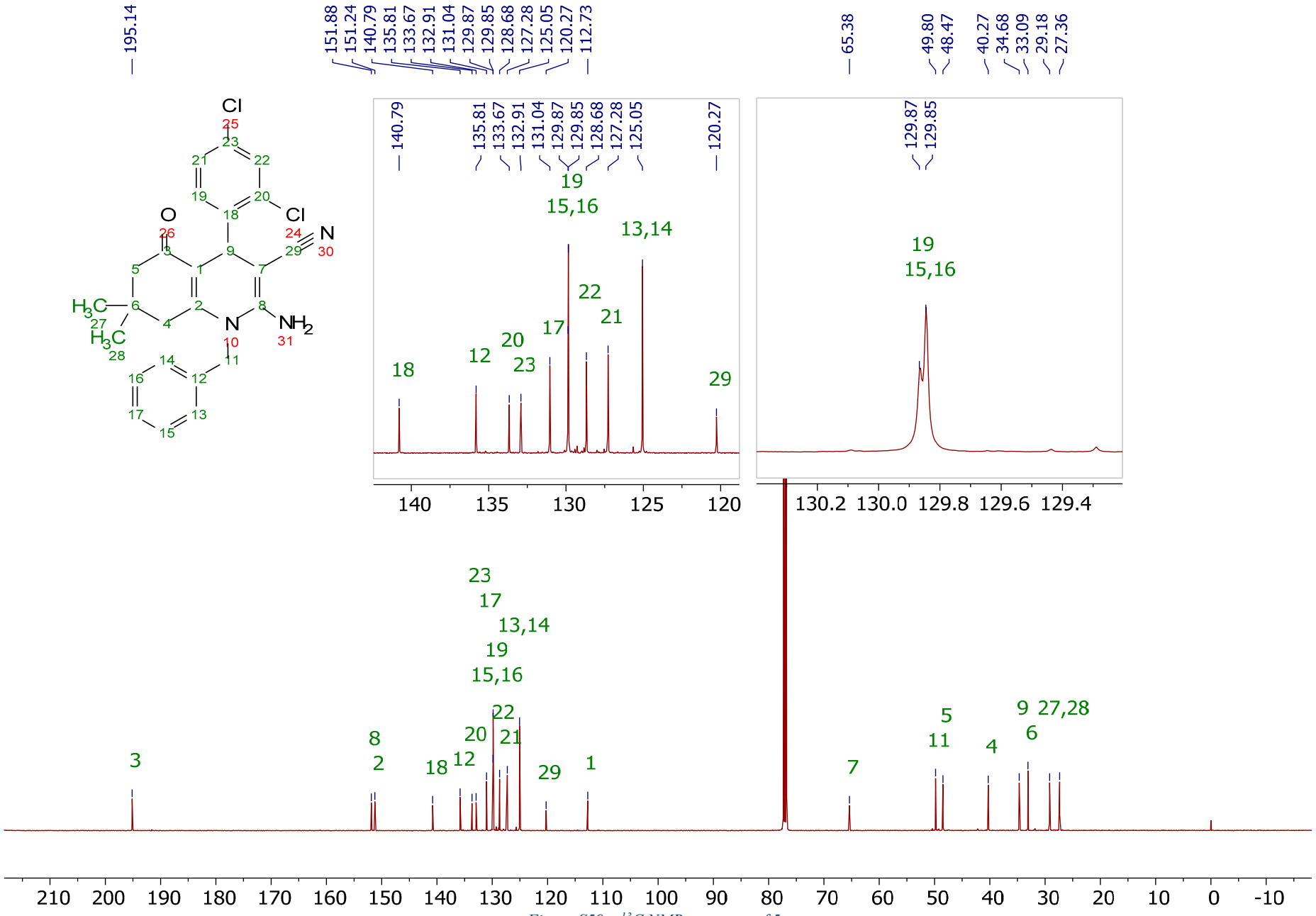


Figure S59 – ^{13}C NMR spectrum of 5n

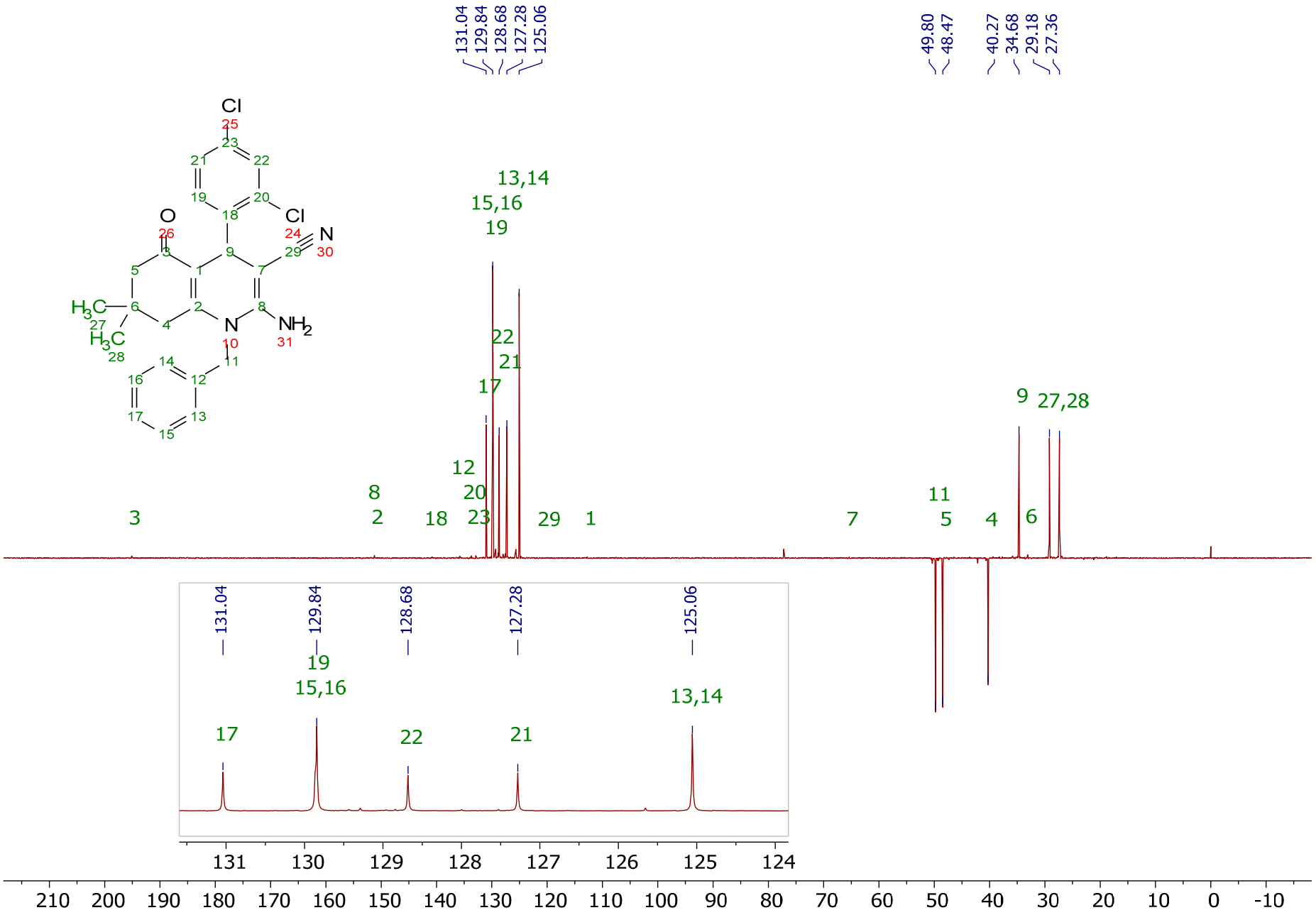


Figure S60 - DEPT spectrum of 5n

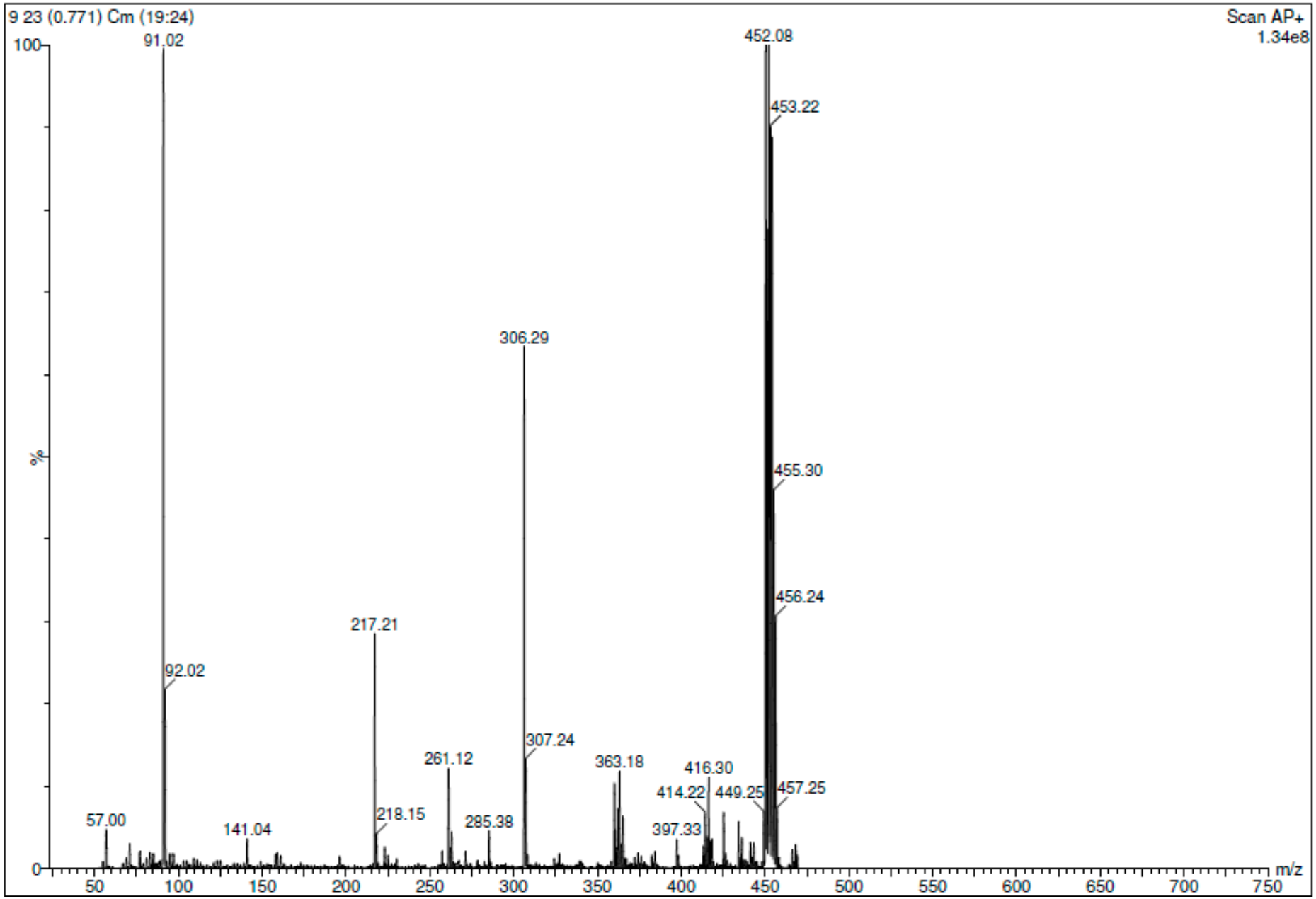
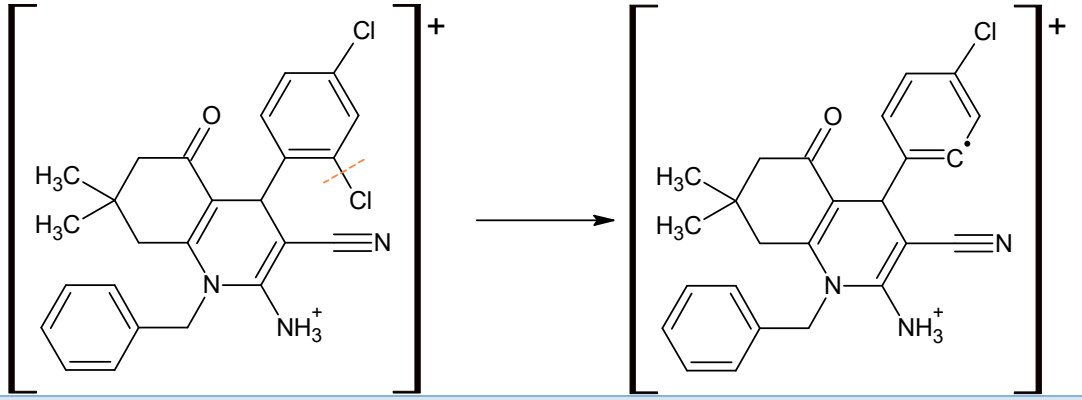
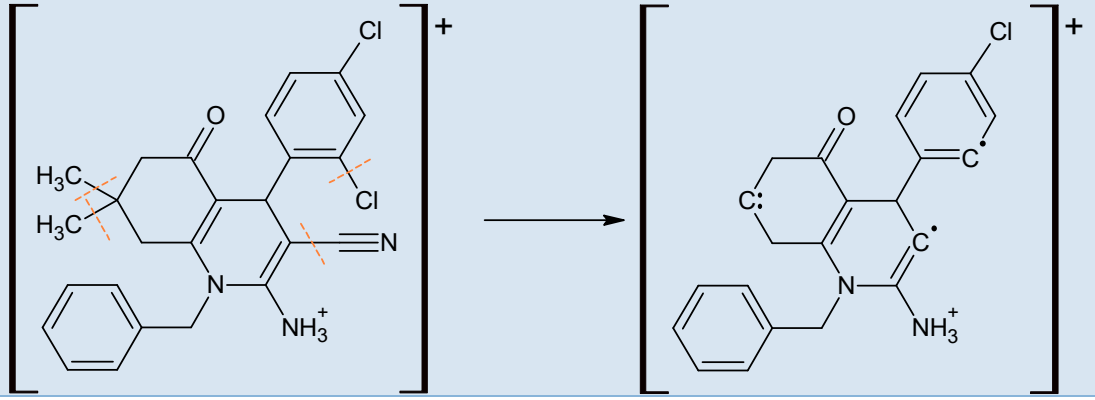
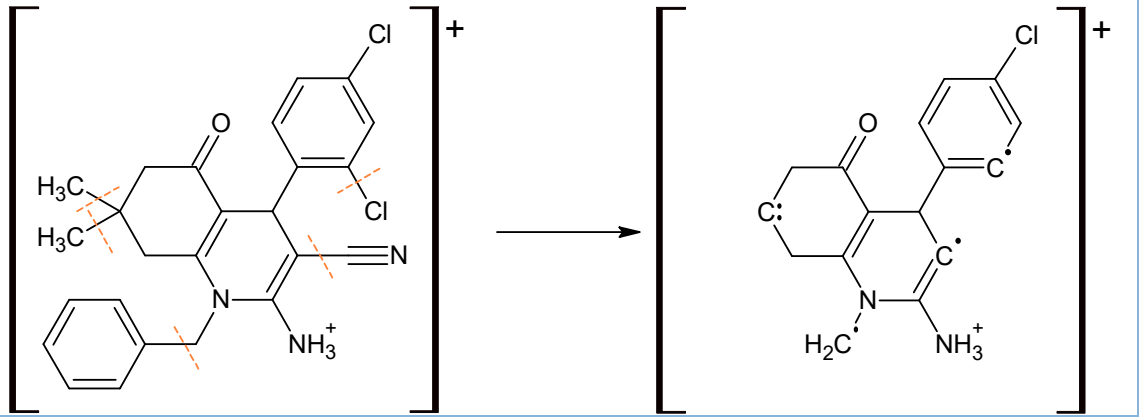
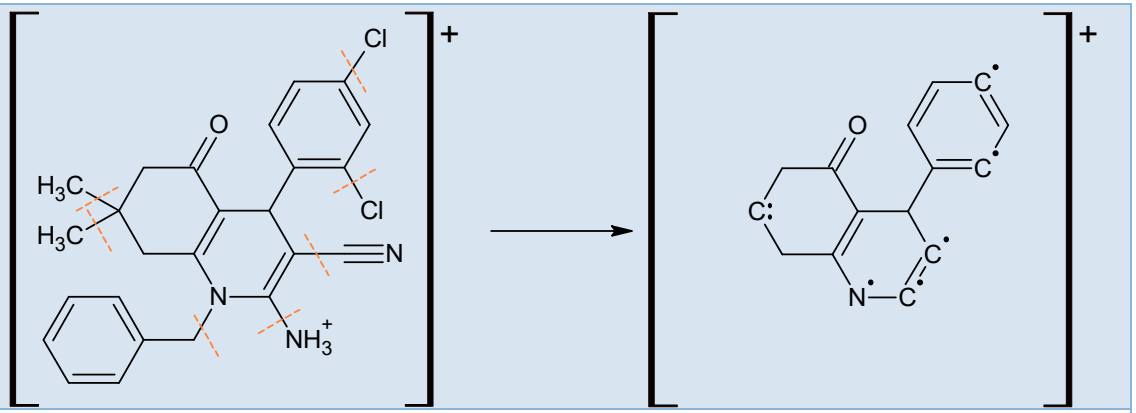
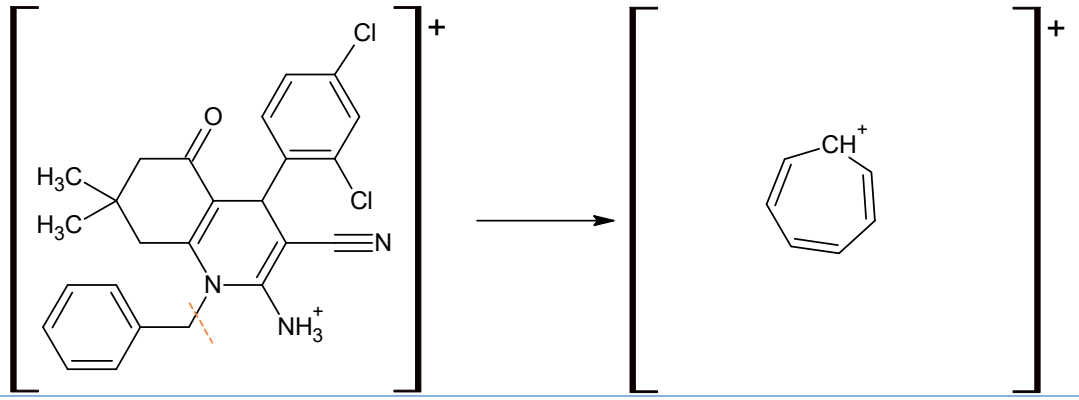


Figure S61 - MS spectrum of 5n

Table S7 - Fragmentation positions for peaks in MS spectrum of 5n

m/z	Fragmentation position
452.08 – 457.25 (isotopes)	$[M+H]^+$
414.22-416.30 (isotopes)	
361.18 - 363.18 (isotopes)	
285.38	

218.15	
91.02	

1.15. Product 5o: 2-amino-1-cyclohexyl-7,8-dimethyl-5-oxo-4-(3-nitrophenyl)-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

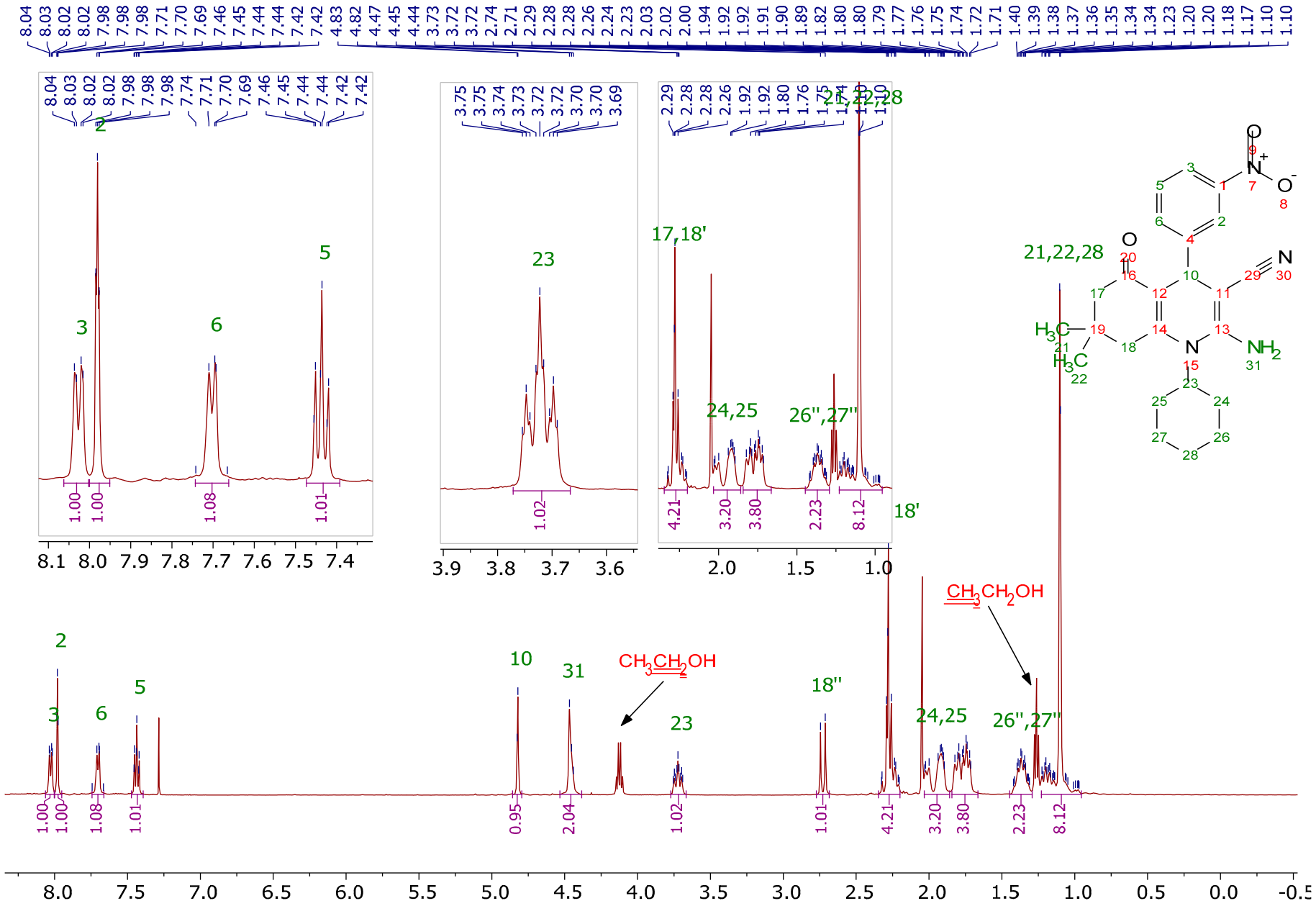


Figure S62 - ^{1}H NMR spectrum of 5o

1.16. Product 5p: 2-amino-4-(4-cyanophenyl)-7,8-dimethyl-1-(4-(phenylazo)phenyl)-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

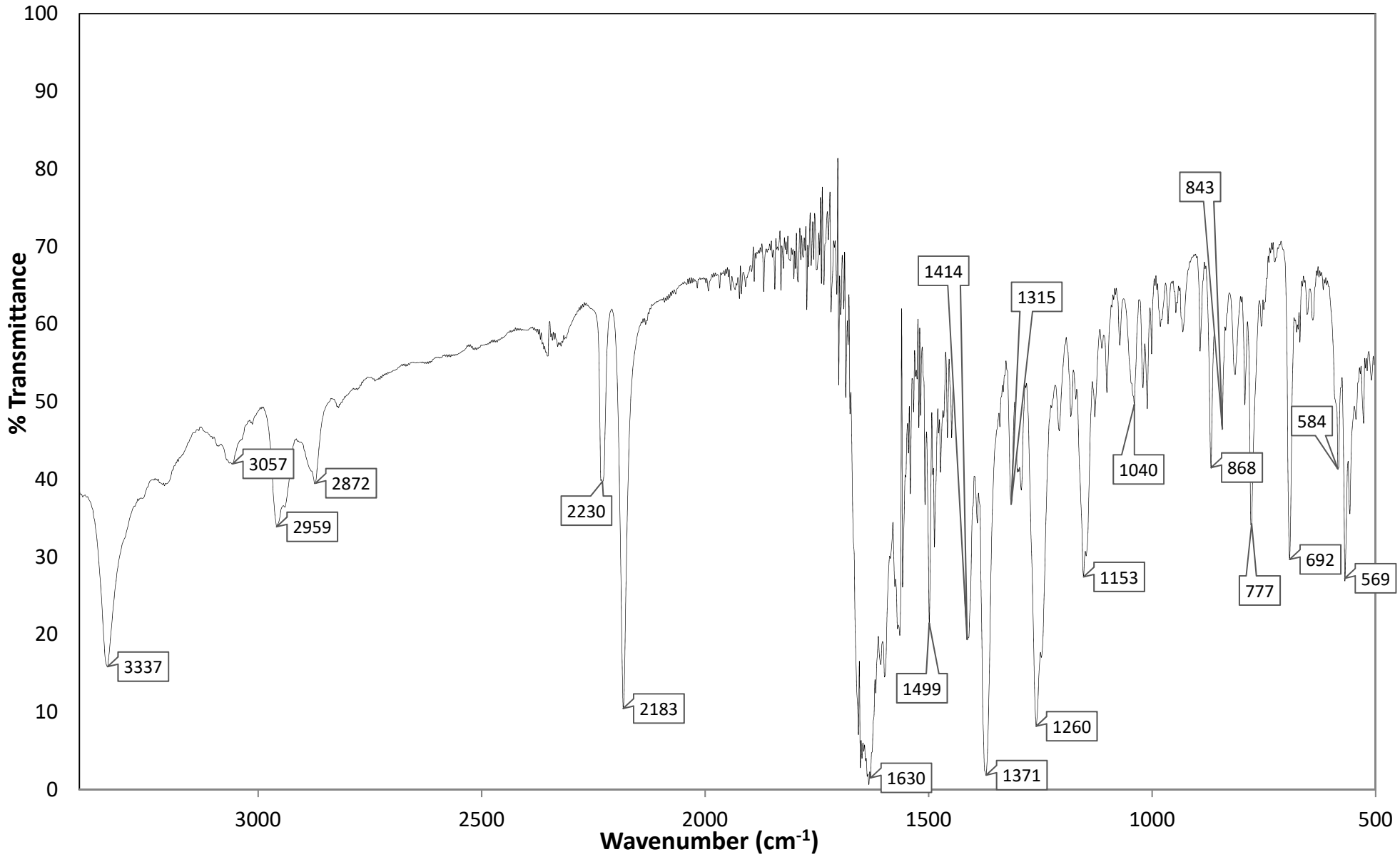


Figure S63 - IR spectrum of 5p

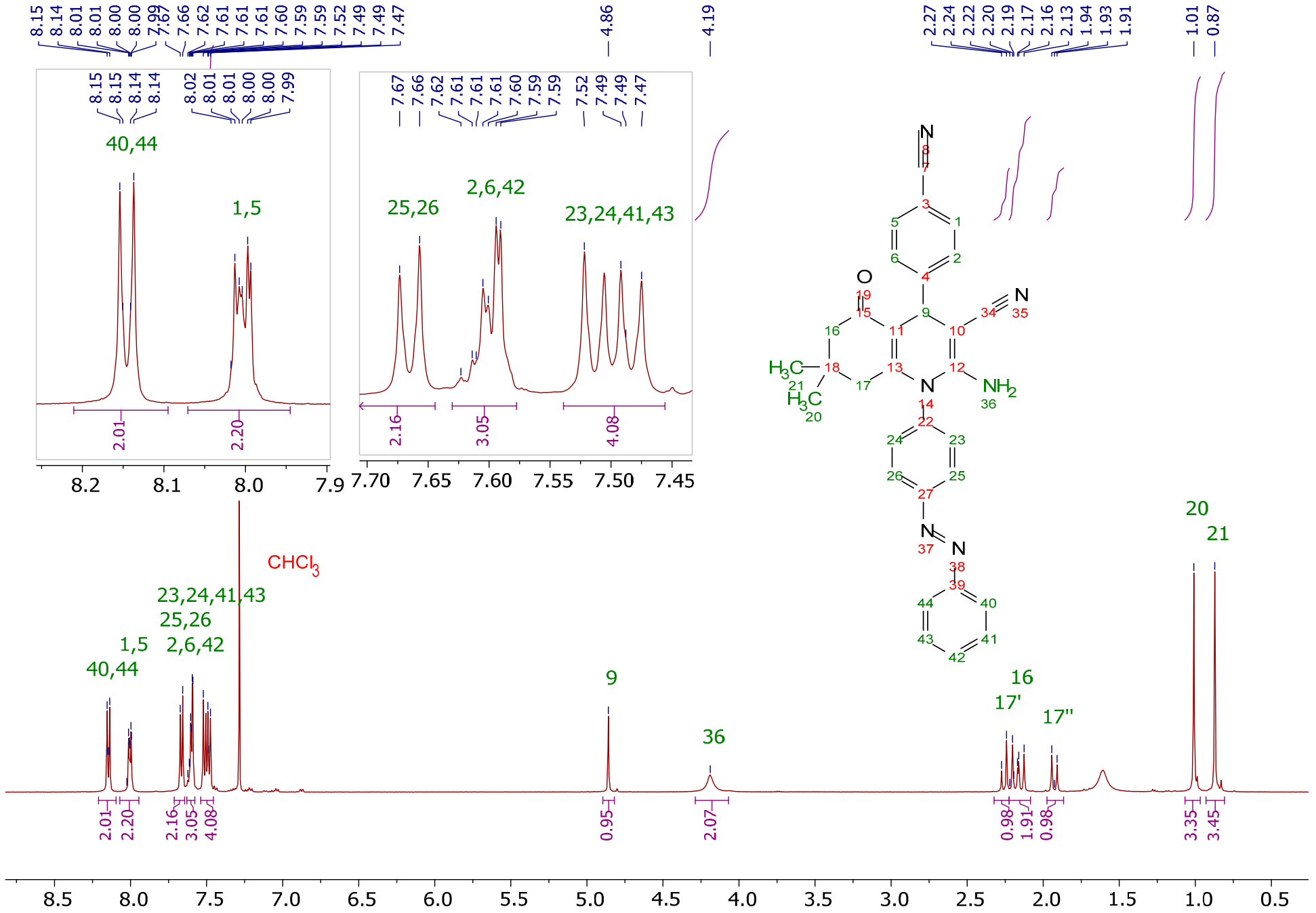


Figure S64 - ^{1}H NMR spectrum of 5p

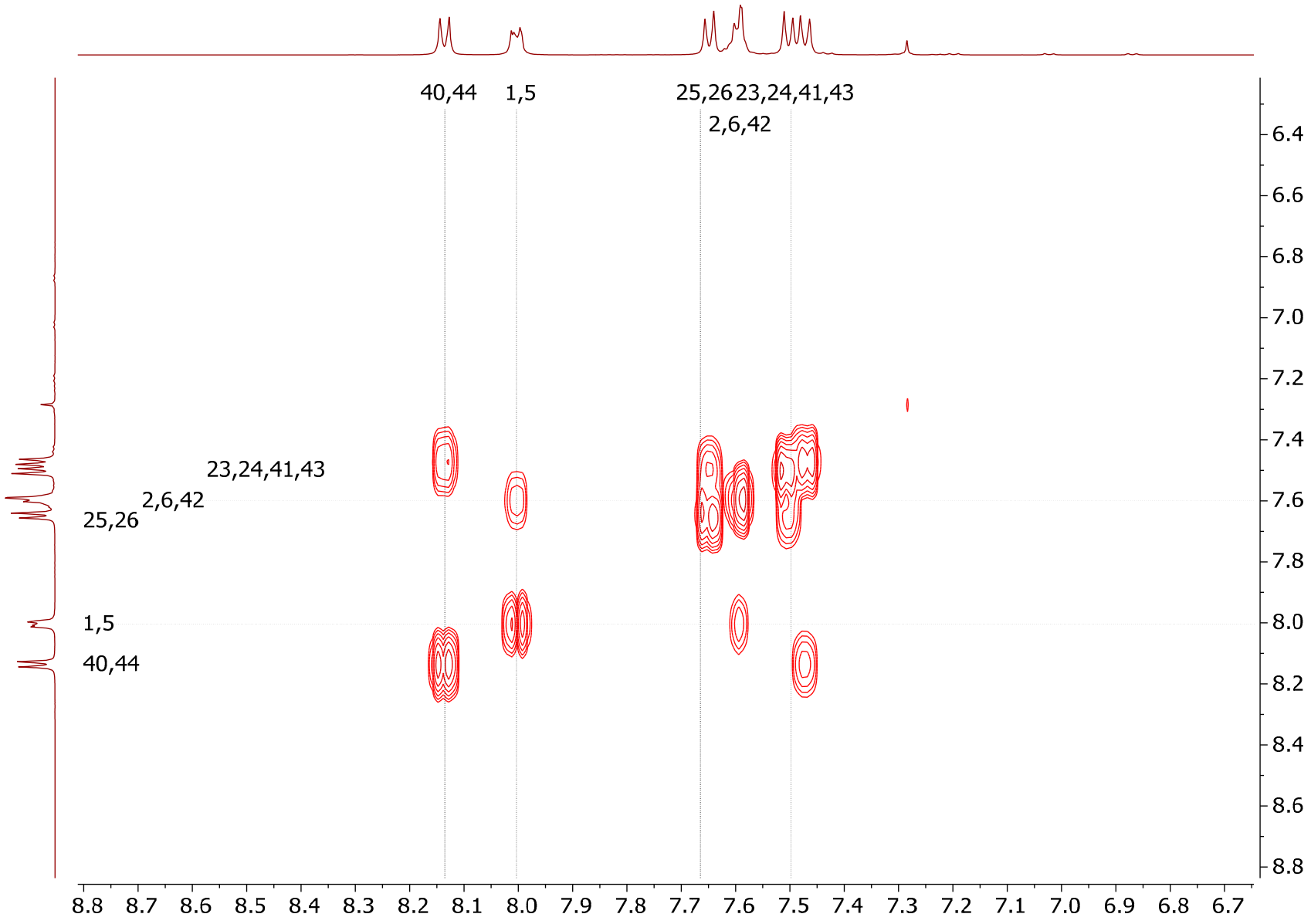


Figure S65 - ^{1}H-^{1}H COSY of 5p

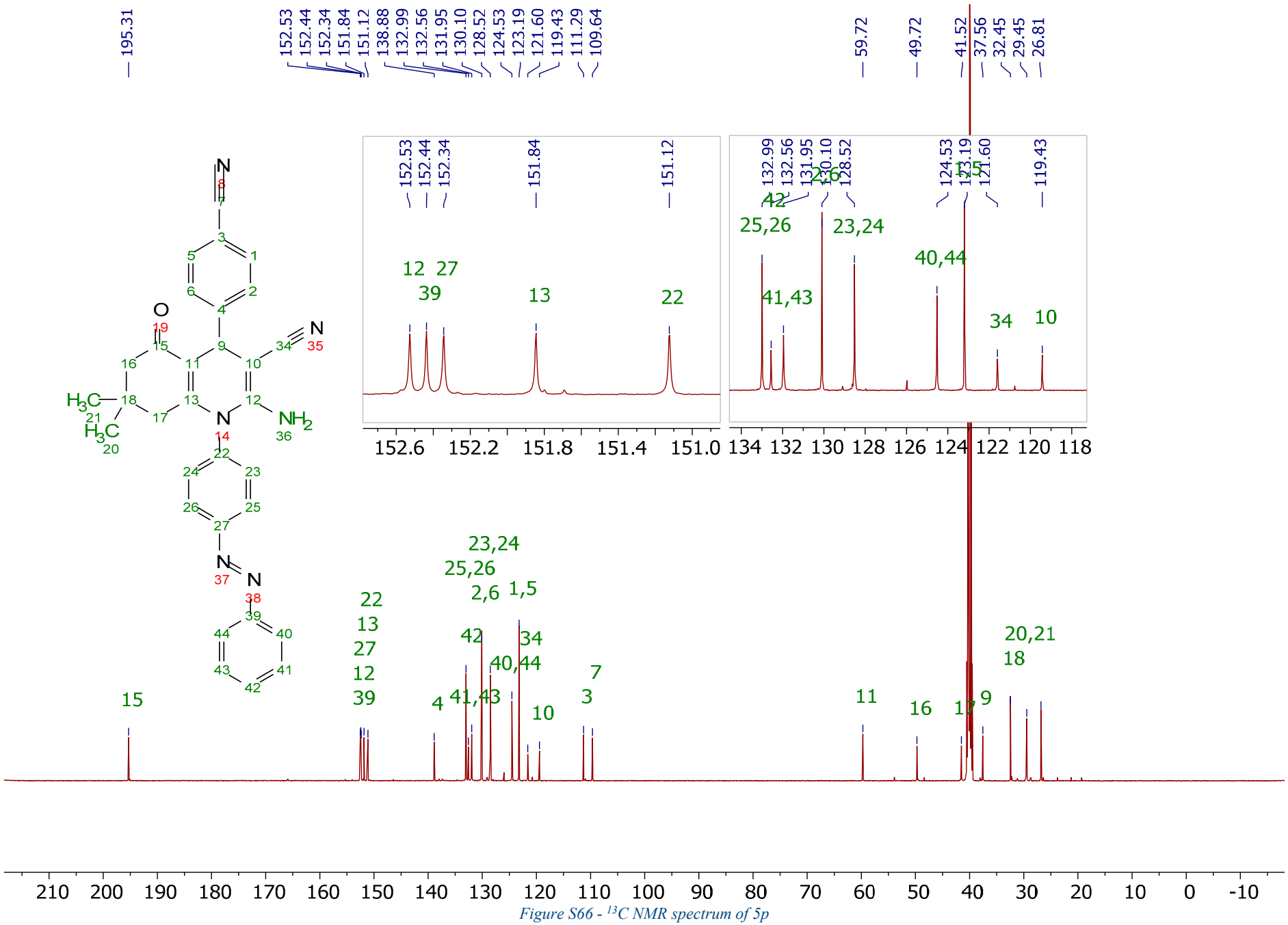


Figure S66 - ^{13}C NMR spectrum of 5p

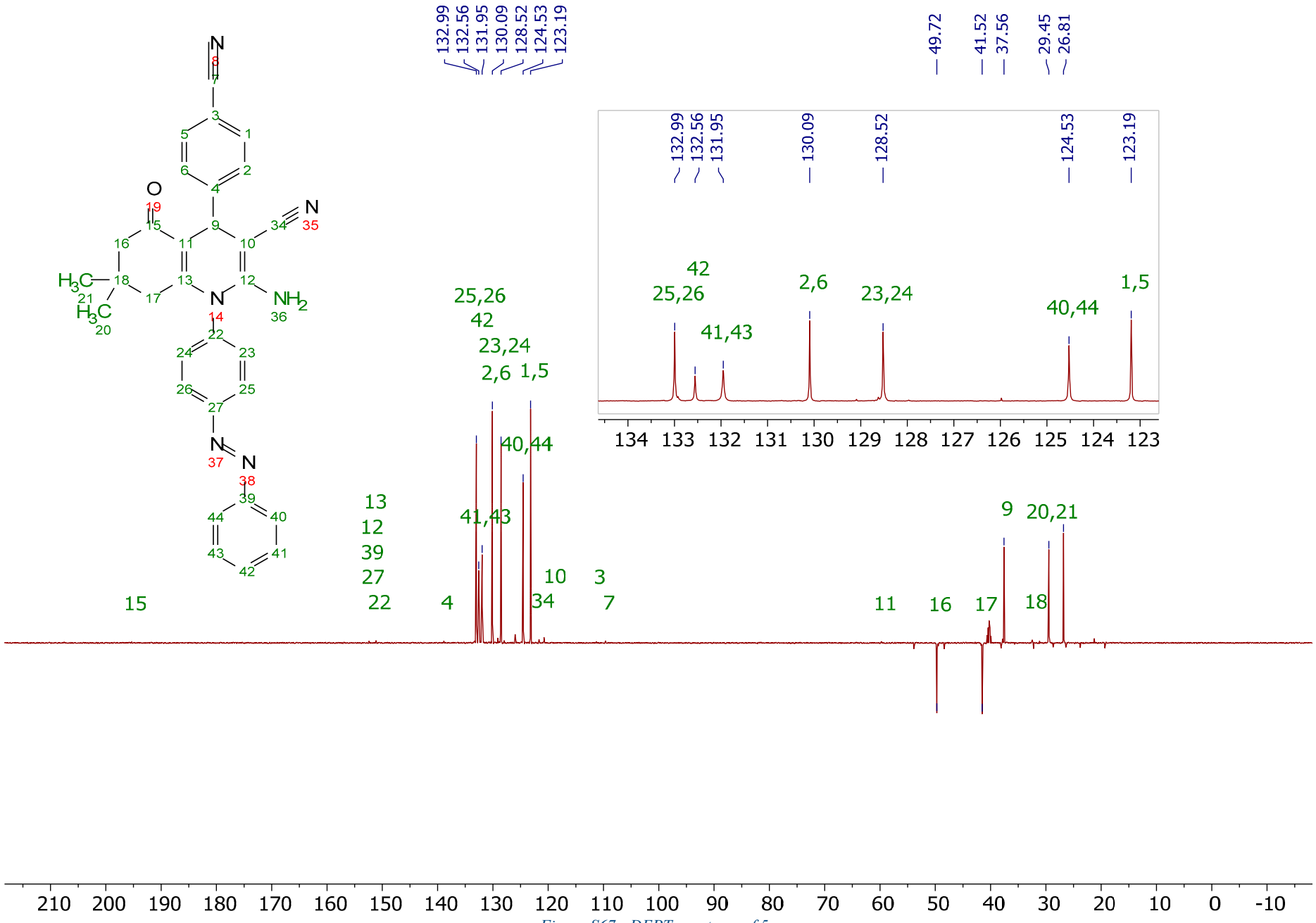


Figure S67 - DEPT spectrum of 5p

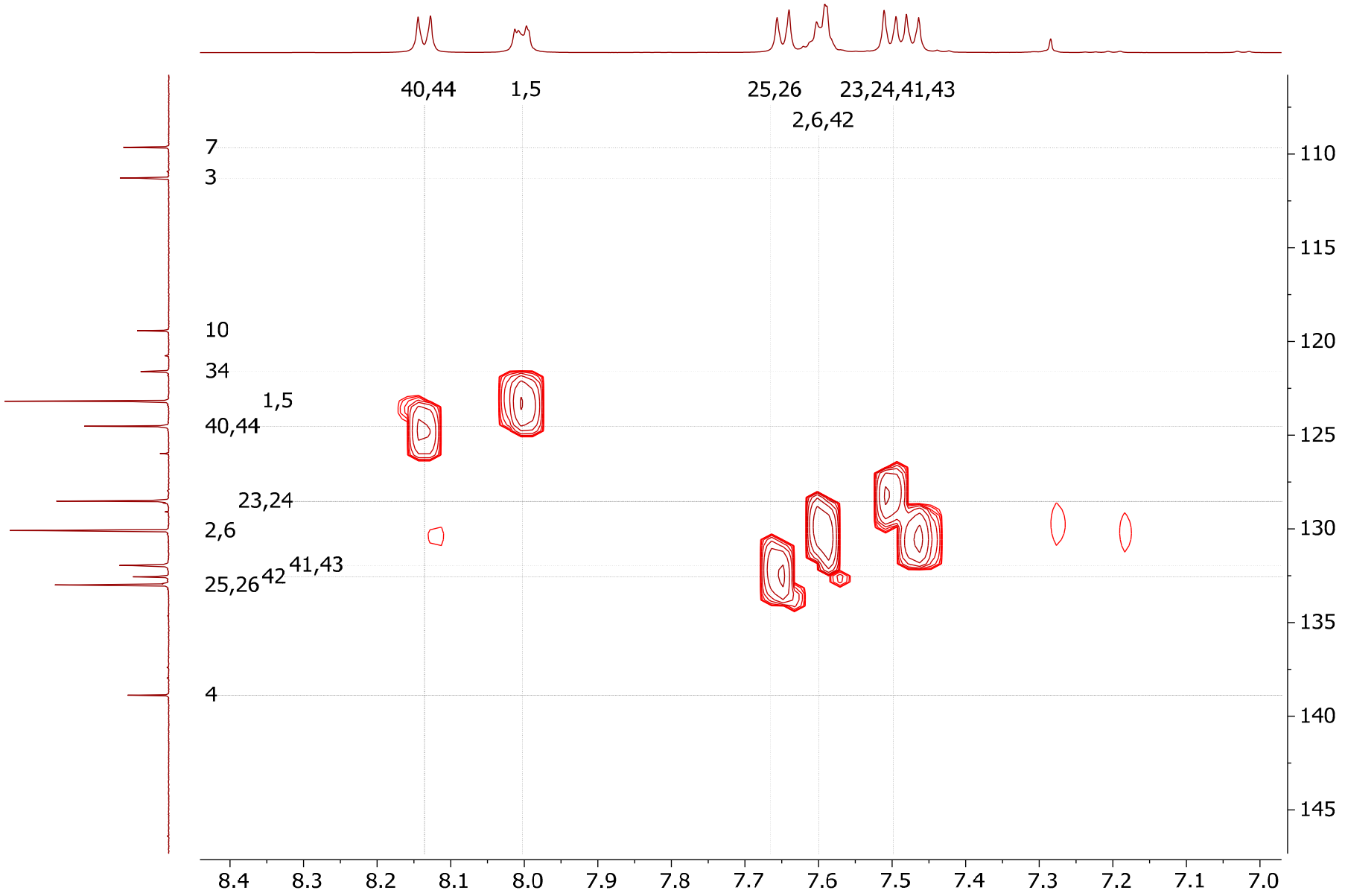


Figure S68 - HSQC spectrum of 5p

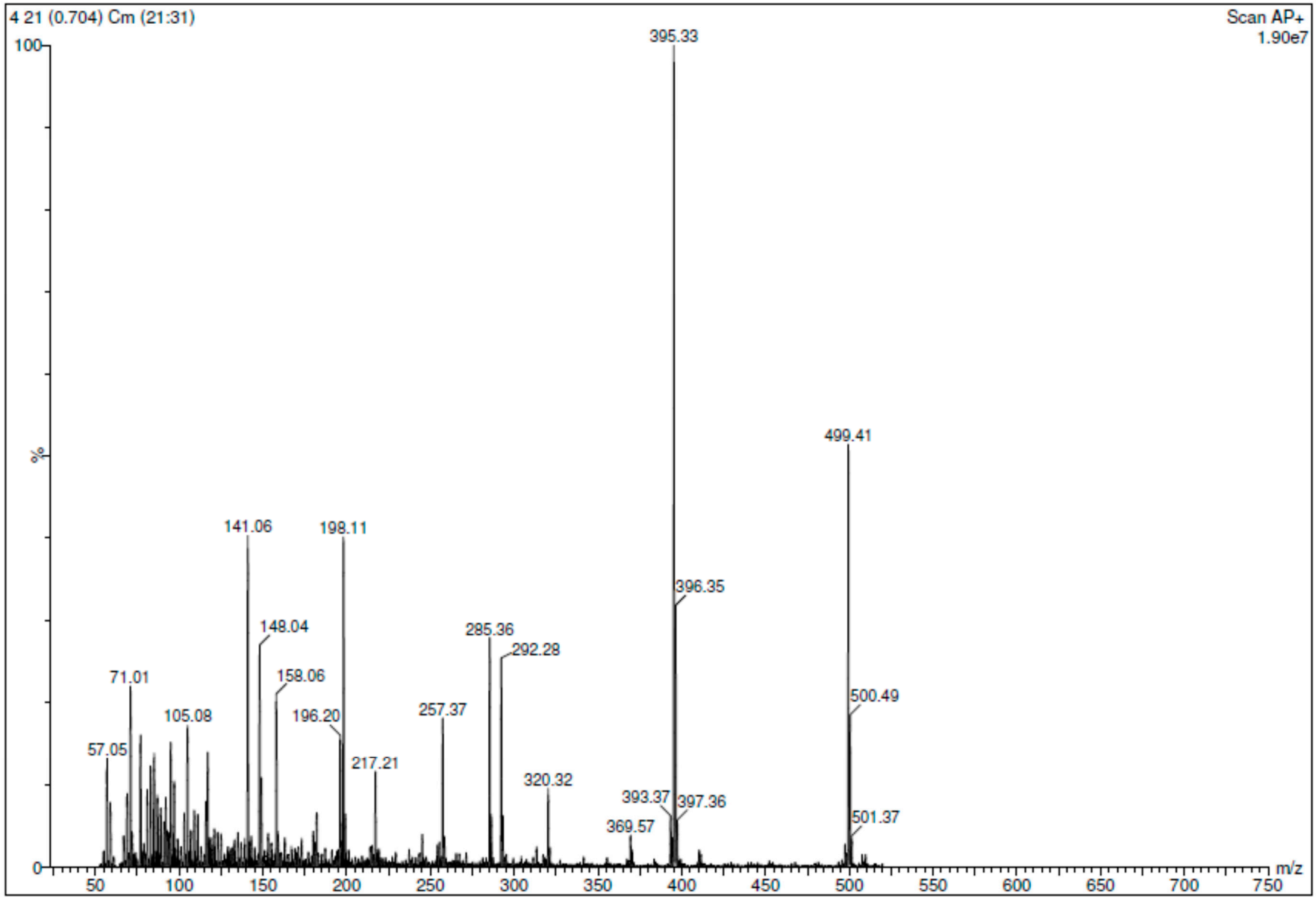
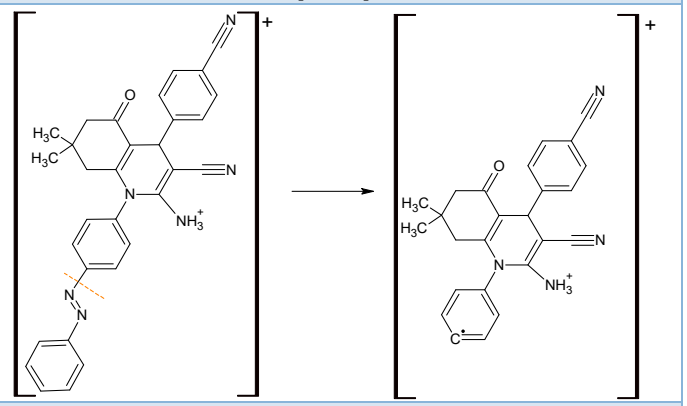
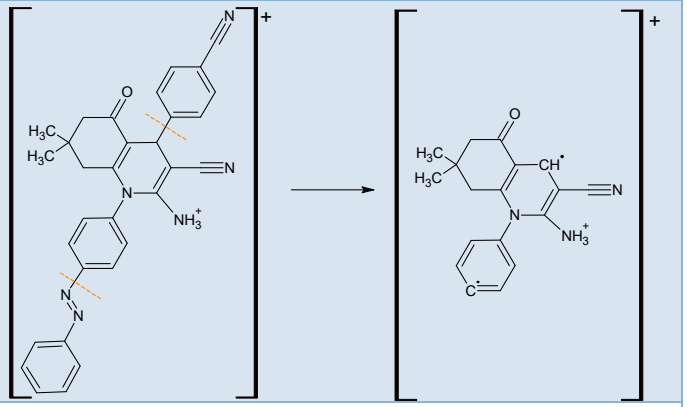
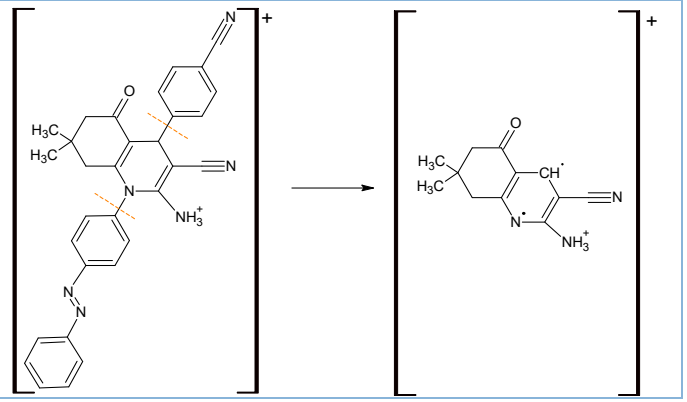


Figure S69 - MS spectrum of 5p

Table S8 - Fragmentation positions for peaks in MS spectrum of 5p

m/z	Fragmentation position
499.41	$[M+H]^+$
395.33	
292.28	
217.21	

1.17. Product 5q: 2-amino-4-(2,4-dichlorophenyl)-7,8-dimethyl-1-(4-(3-methylphenylazo)-3-methylphenyl)-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

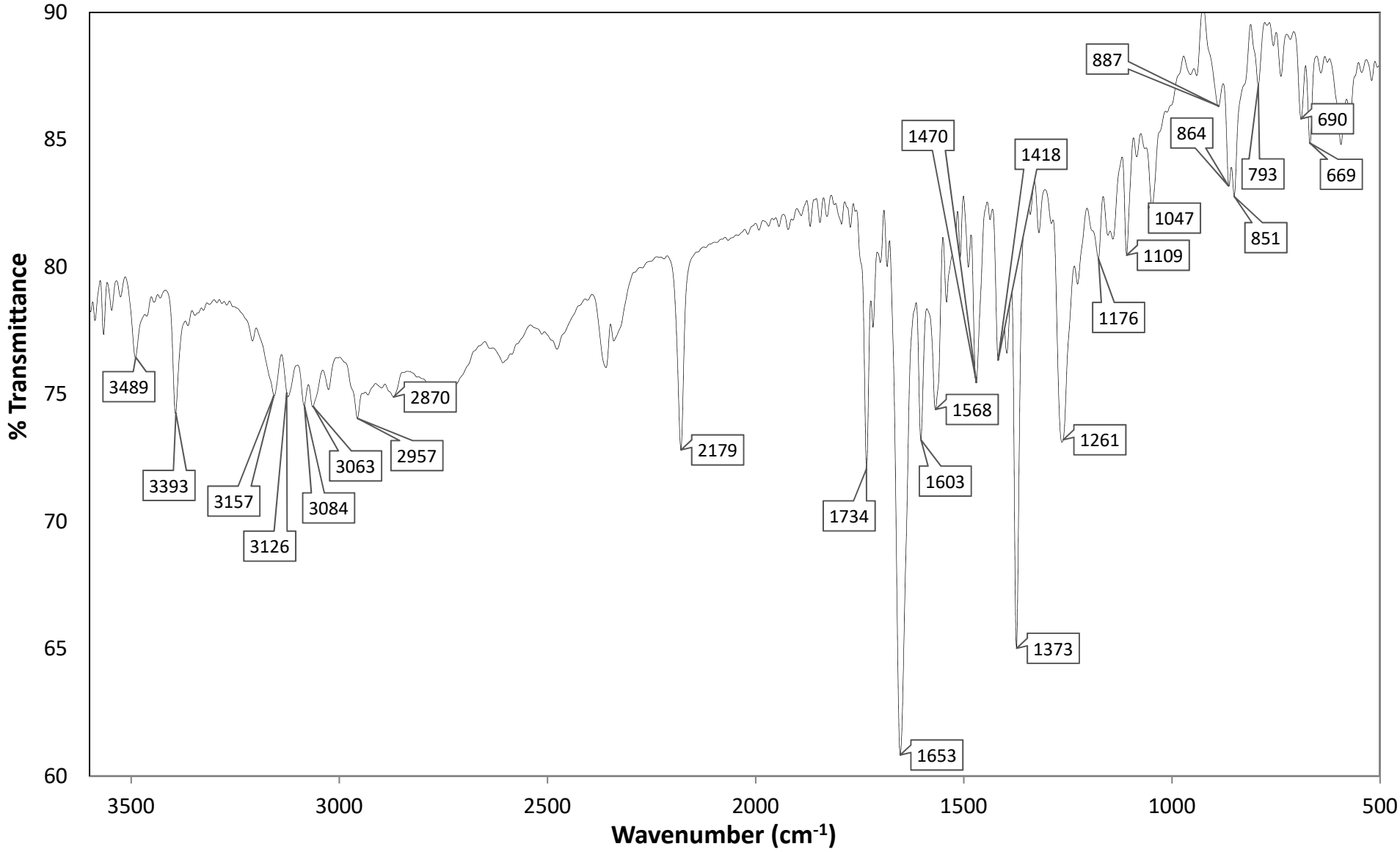


Figure S70 - IR spectrum of 29q

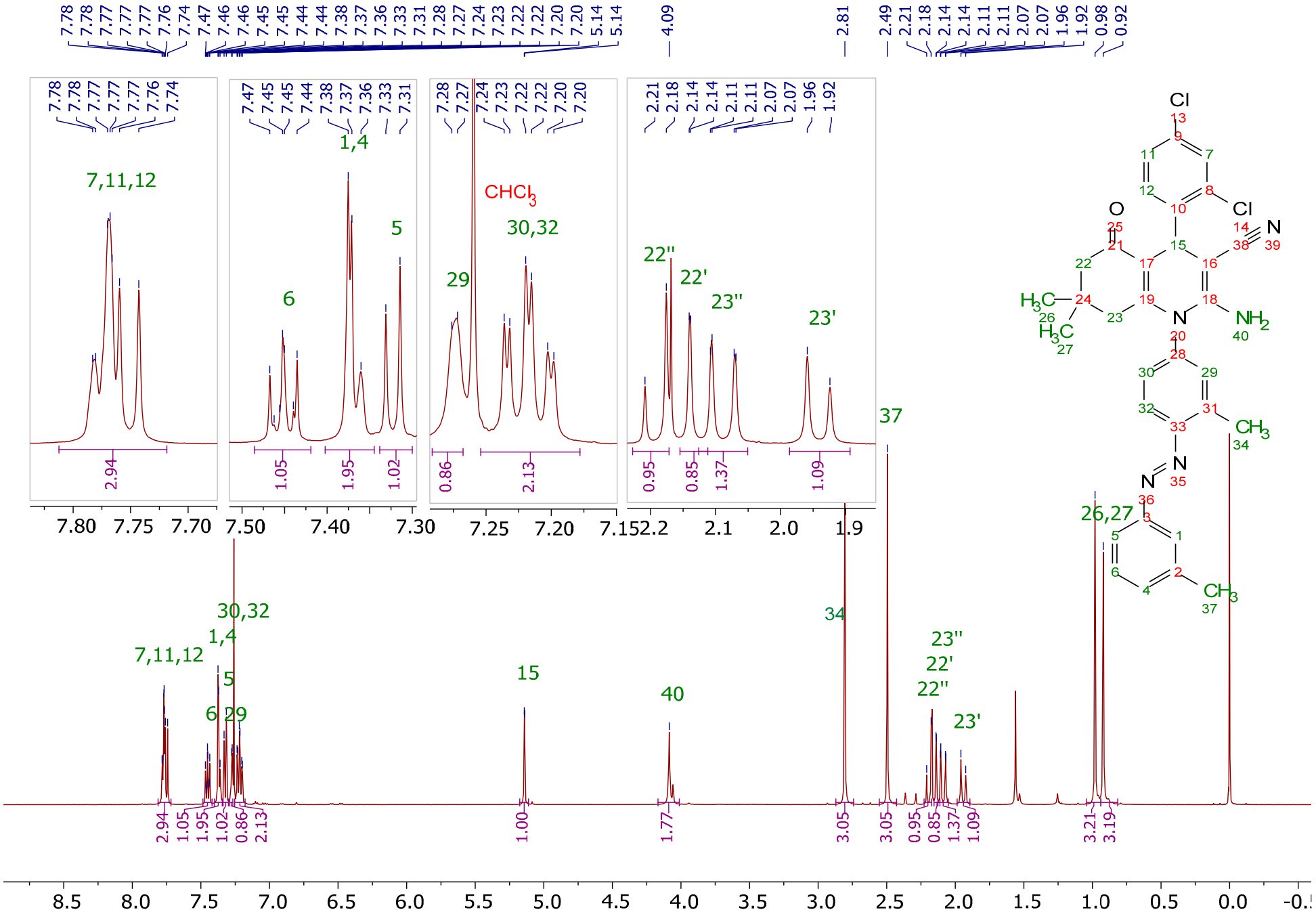


Figure S71 - ^{1}H NMR spectrum of 5q

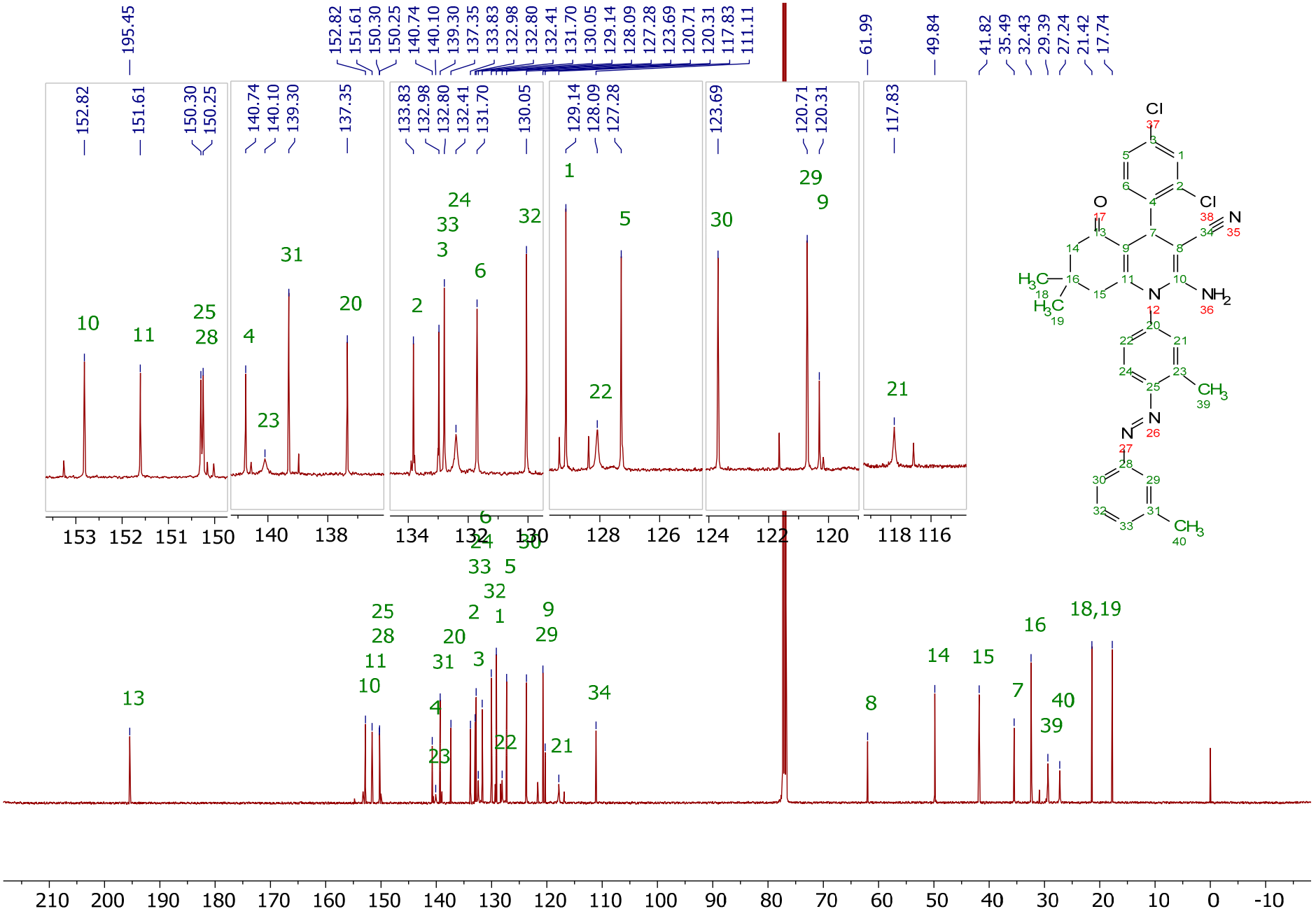


Figure S72 - ^{13}C NMR spectrum of 5q

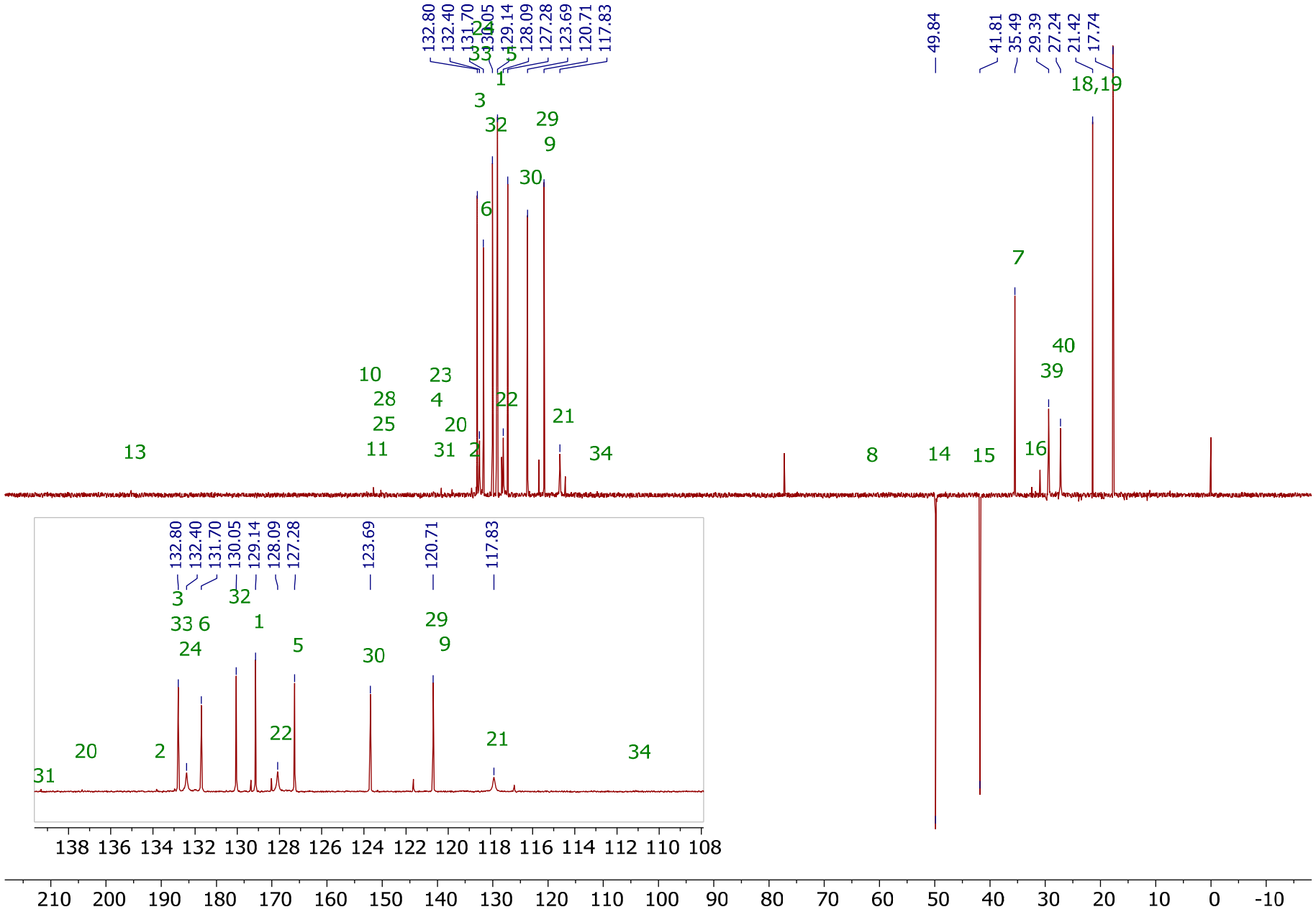


Figure S73 - DEPT spectrum of 5q

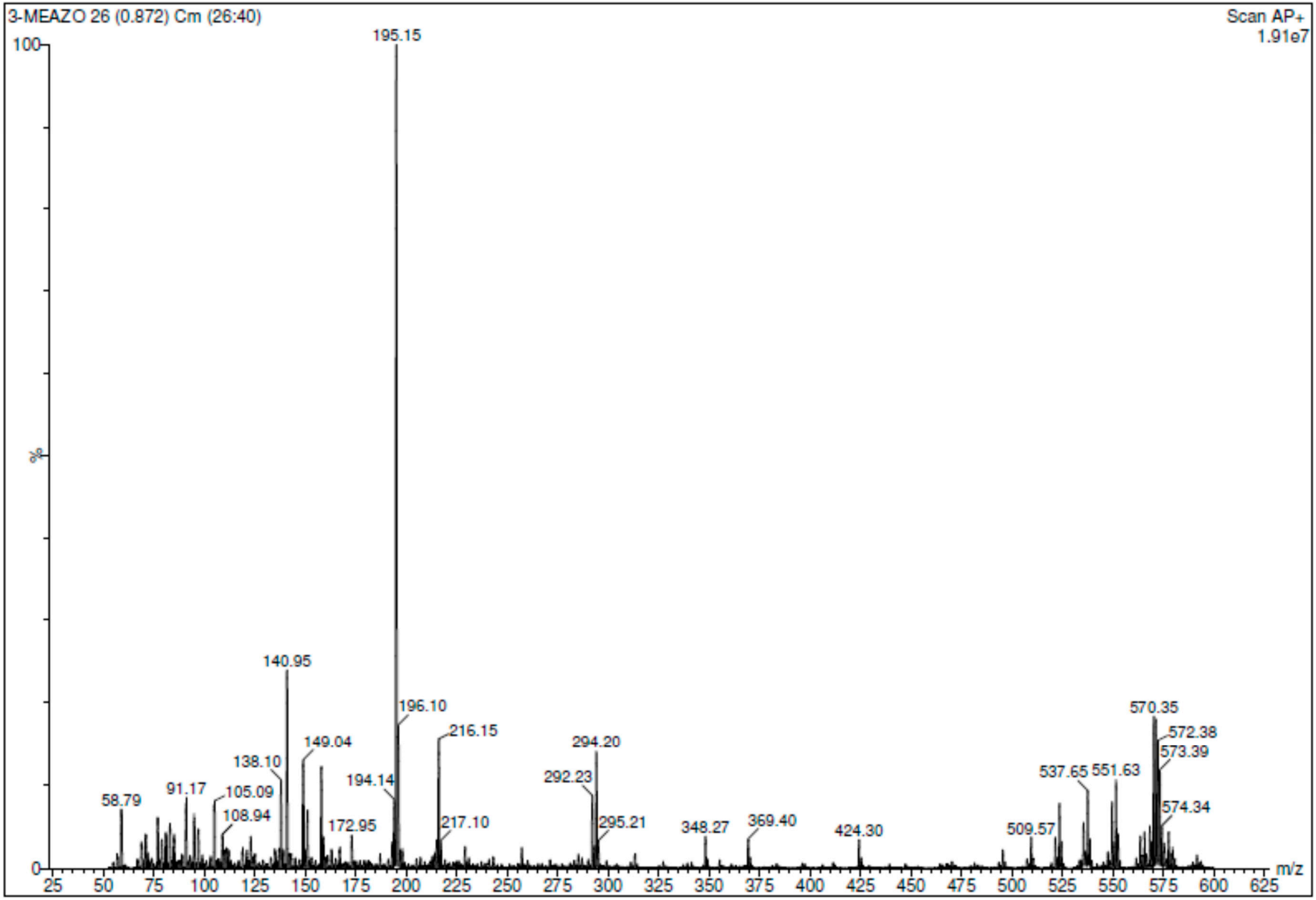
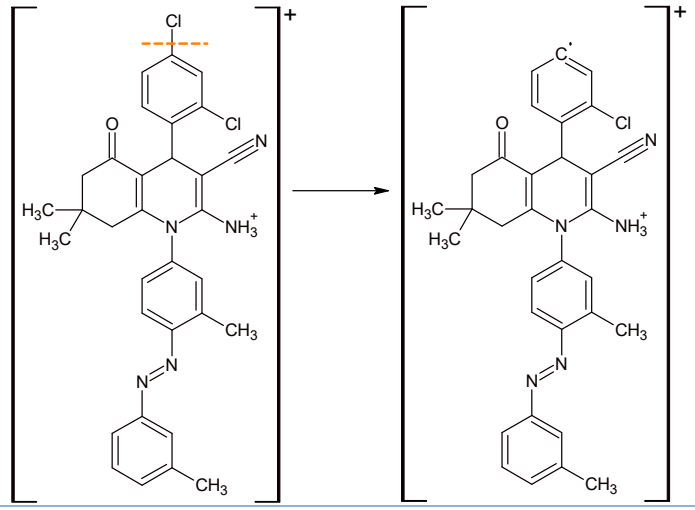
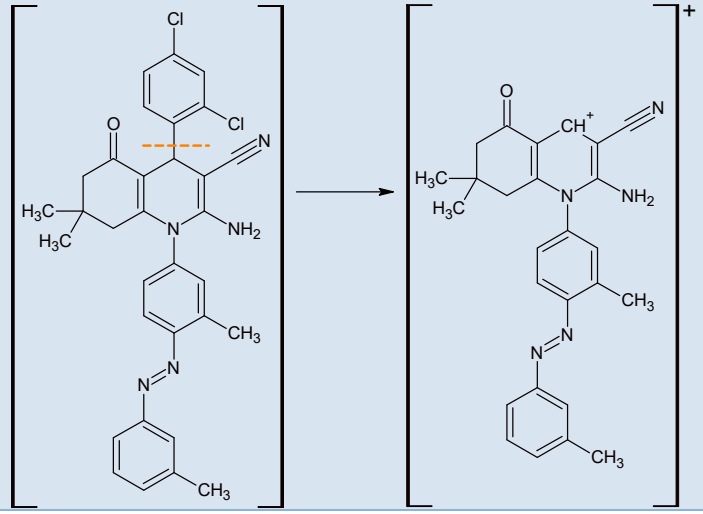
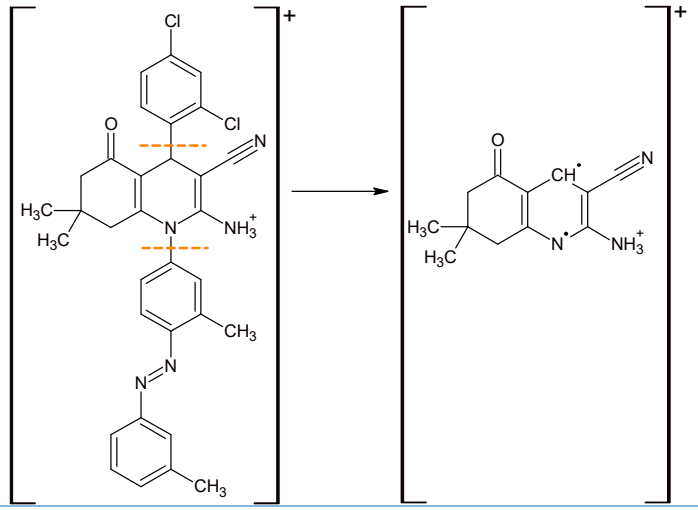
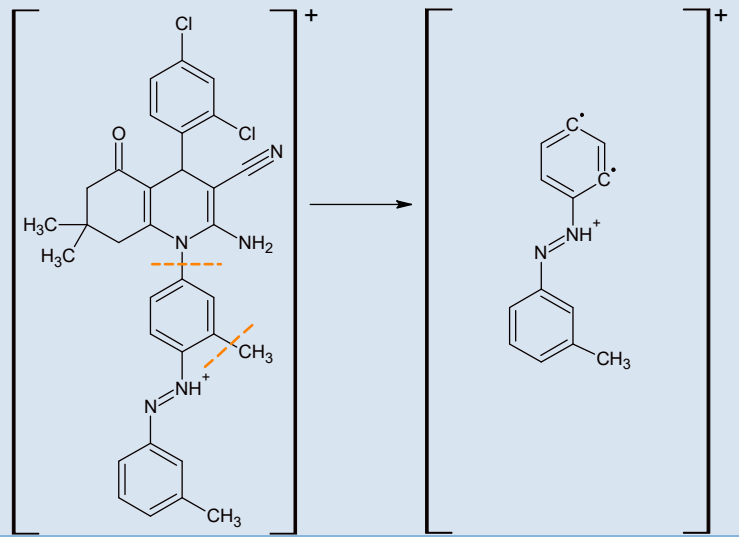


Figure S74 - MS spectrum of 5q

Table S9 - Fragmentation positions for peaks in MS spectrum of 29q

m/z	Fragmentation position
570.35 – 574.34	$[M+H]^+$ (isotopes)
537.65	
424.30	
216.15	

195.15				

1.18. Product 5s: 2-amino-4-(4-methylphenyl)-5-oxo-1-phenyl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

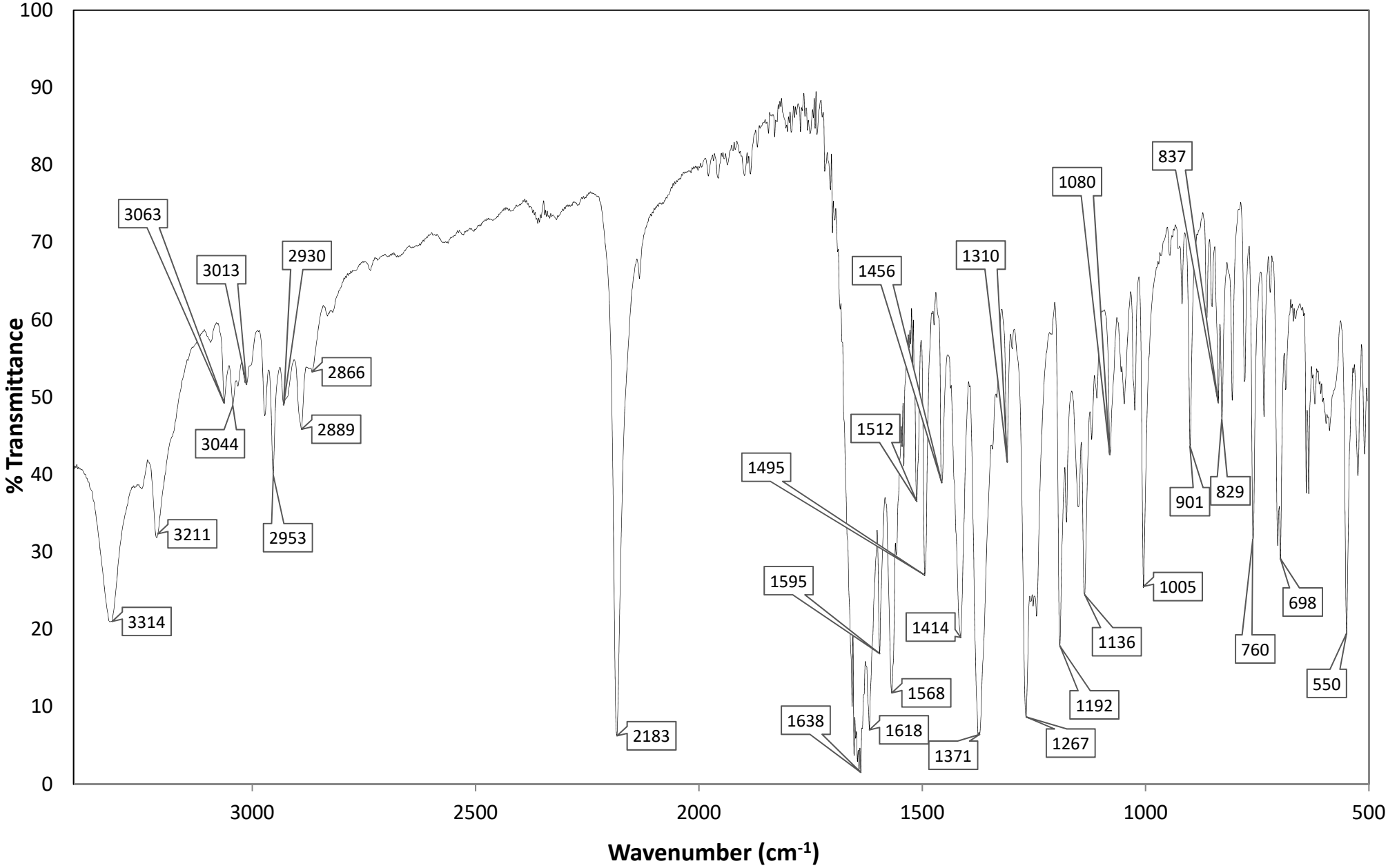


Figure S75 - IR spectrum of 5s

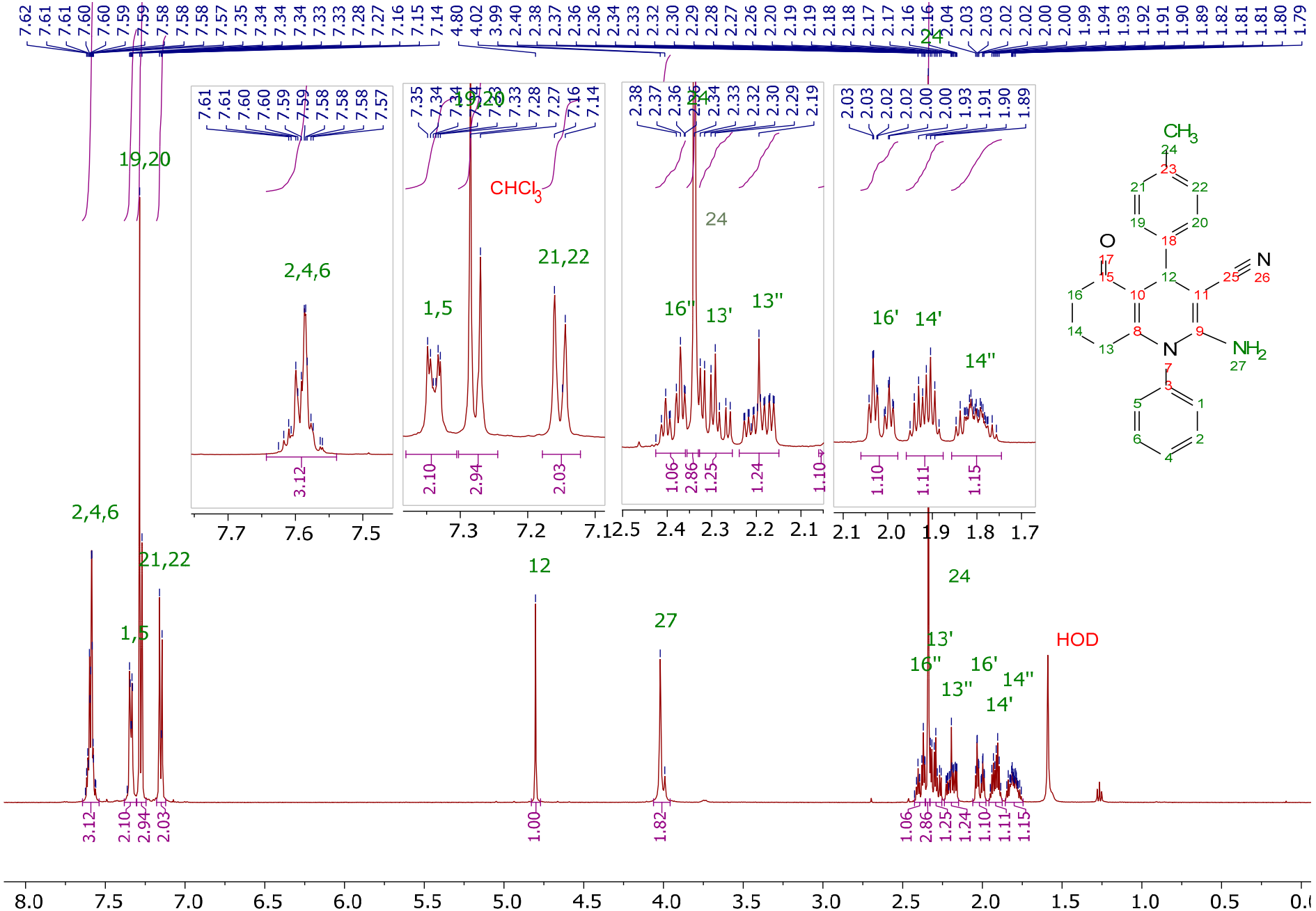


Figure S76 - ^{1}H NMR spectrum of 5s

1.19. Product 5t: 2-amino-4-(4-chlorophenyl)-5-oxo-1-phenyl-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile

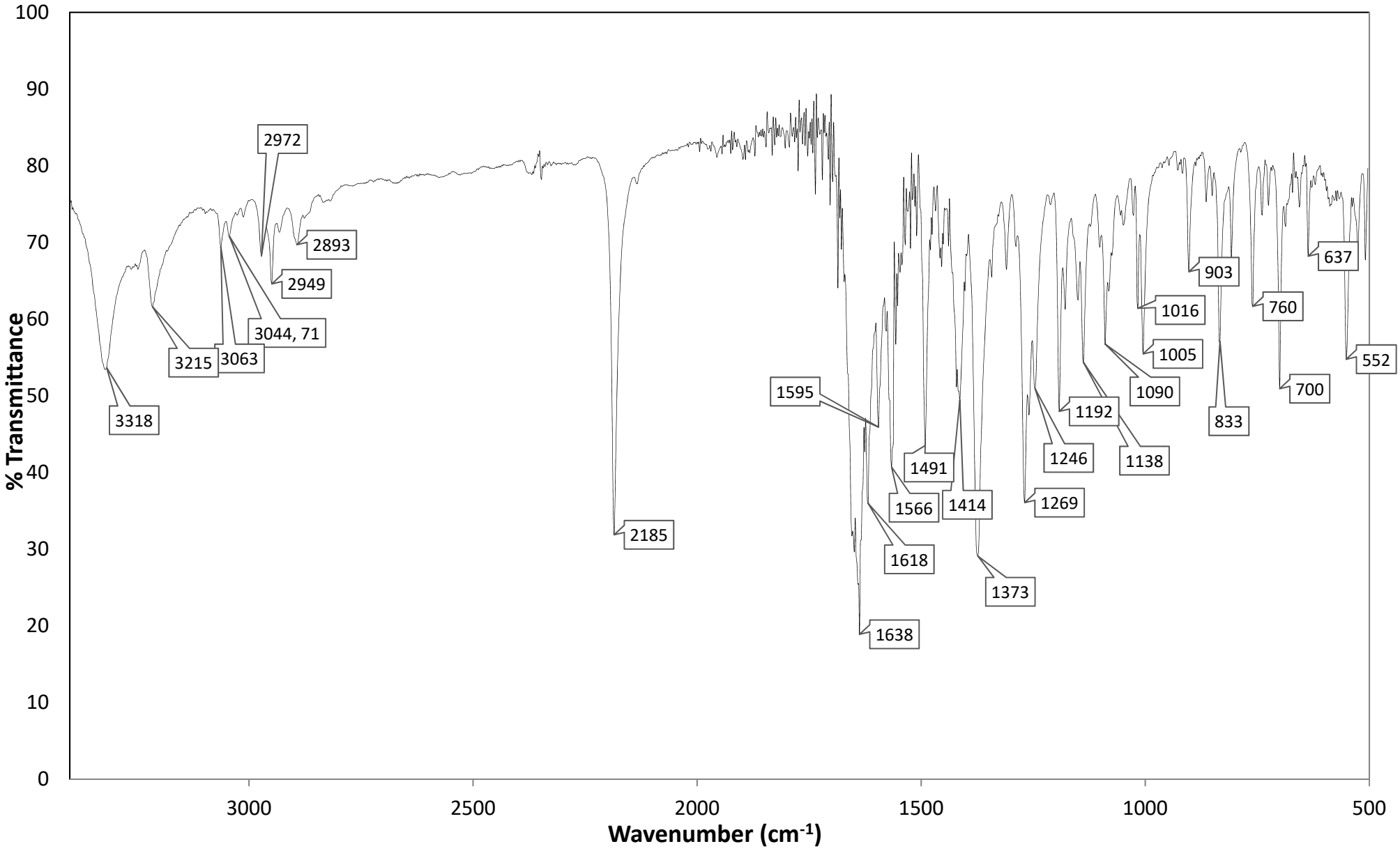


Figure S77 – IR spectrum of 5t

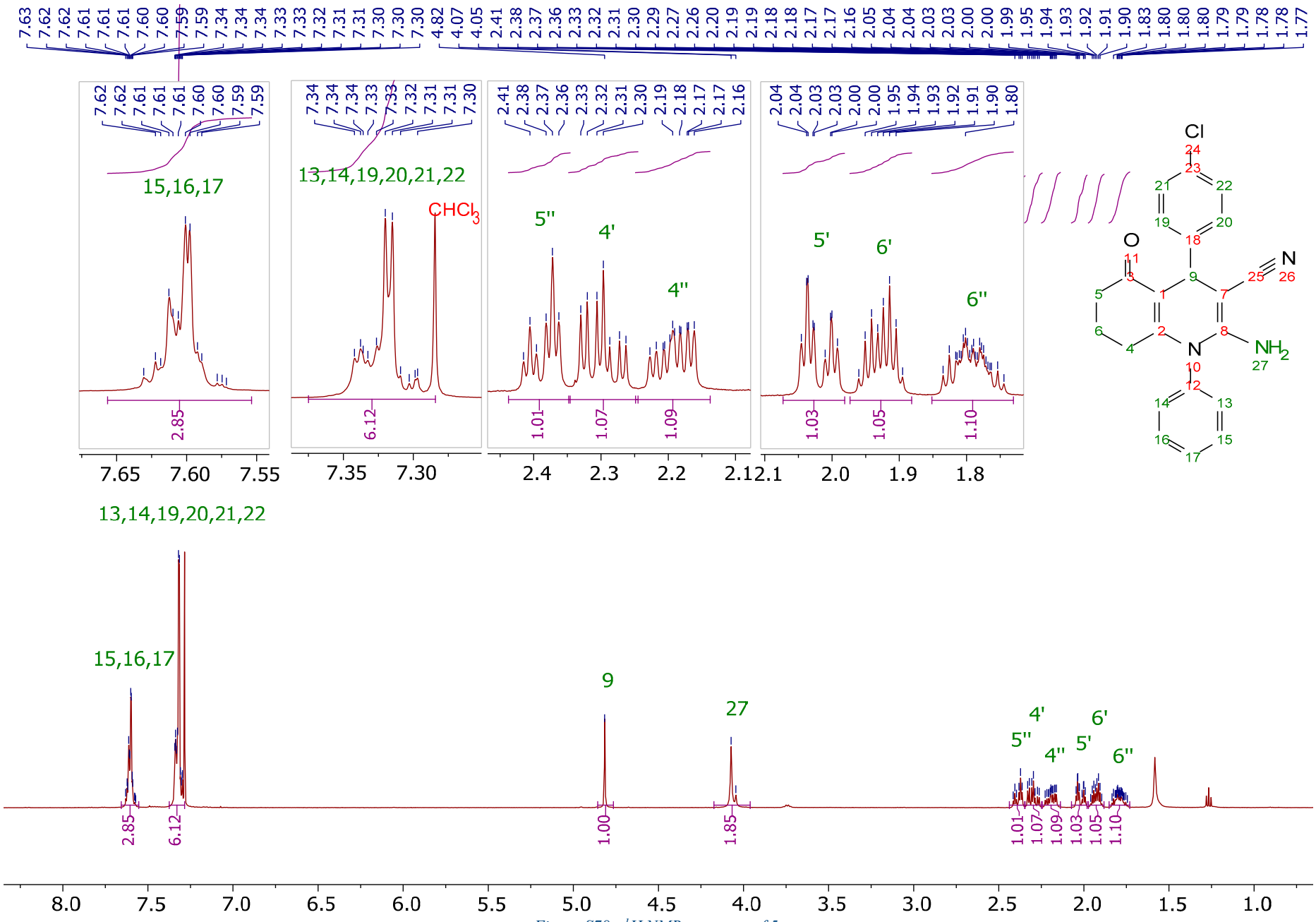


Figure S78 - ^{1}H NMR spectrum of 5t

1.20. Product 7a: 2′-Amino-7′,7′-dimethyl-2,5′-dioxo-1′-phenyl-5′,6′,7′,8′-tetrahydro-1*H*-spiro[indoline-3,4′-quinoline]-3′-carbonitrile

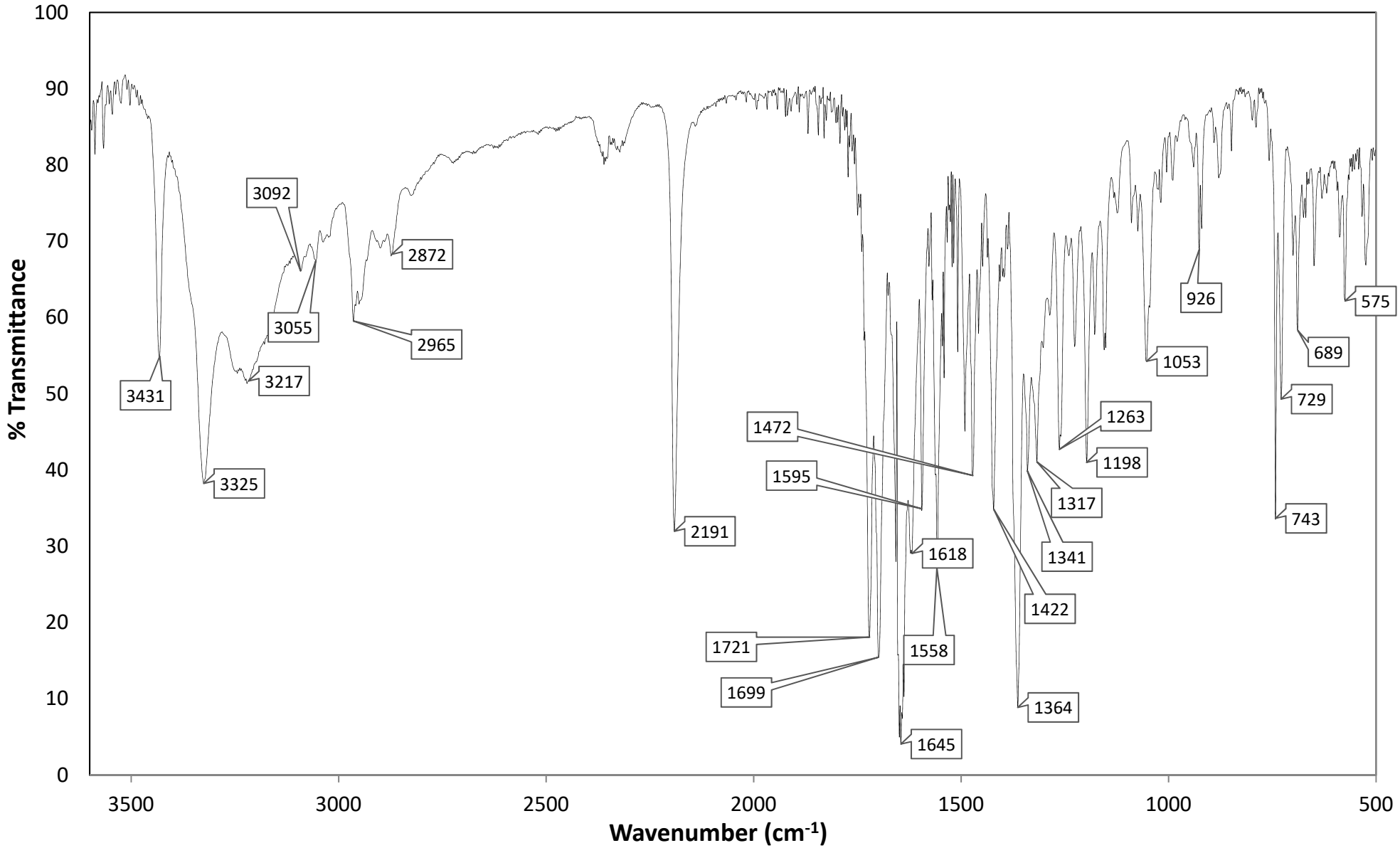


Figure S79 - IR spectrum of 7a

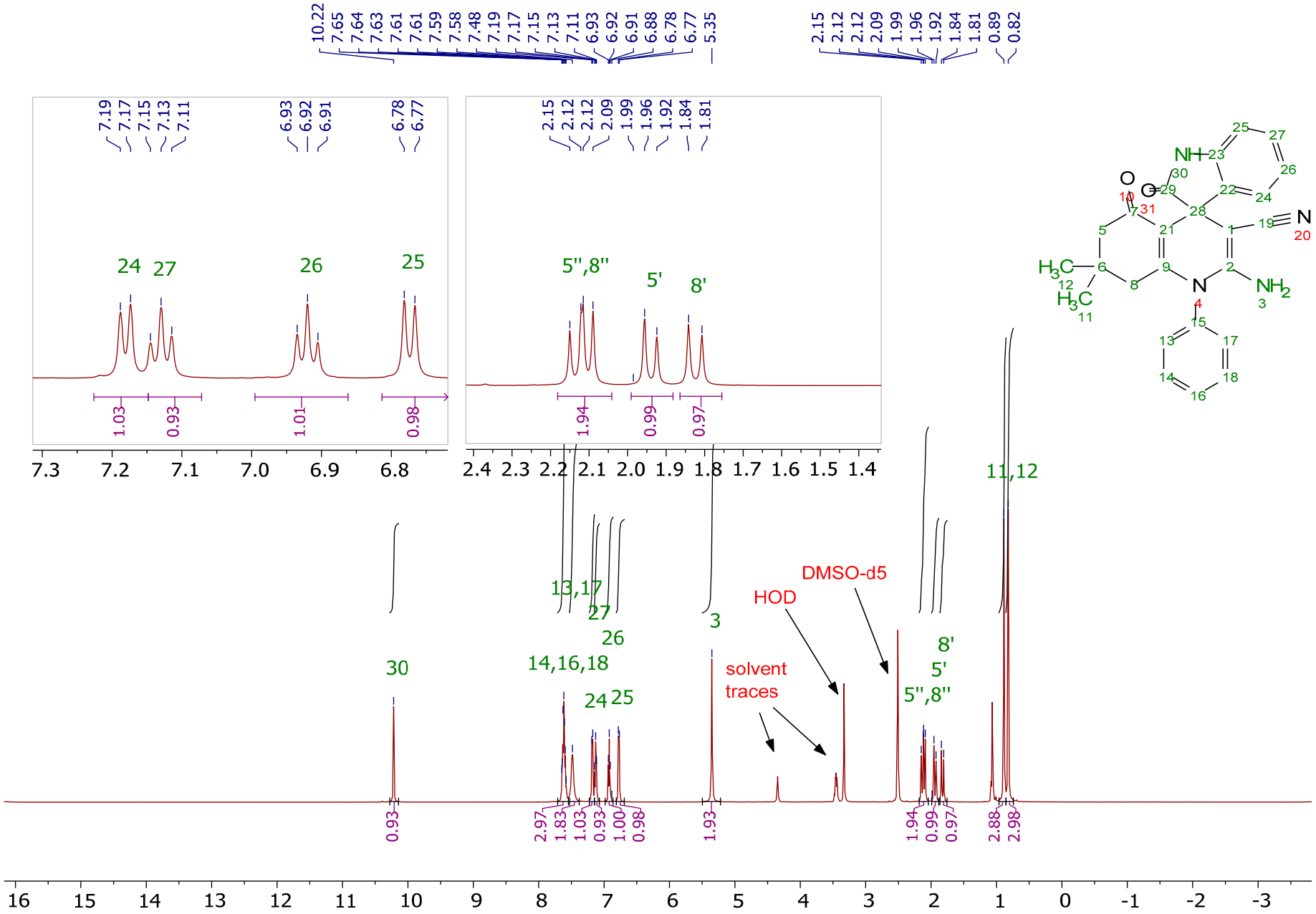


Figure S80 - ^{1}H NMR spectrum of 7a

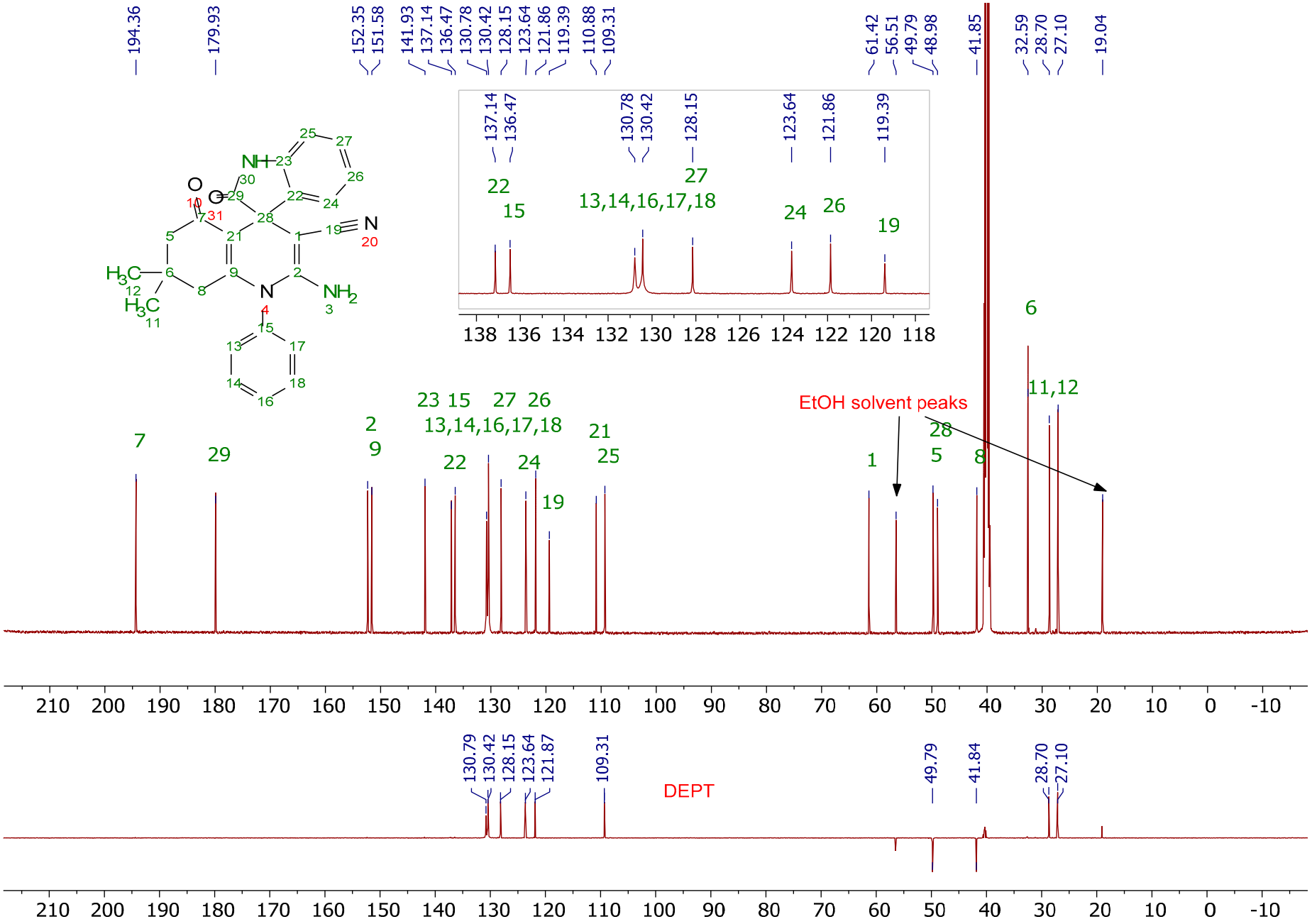


Figure S81 - ^{13}C NMR and DEPT spectra of 7a

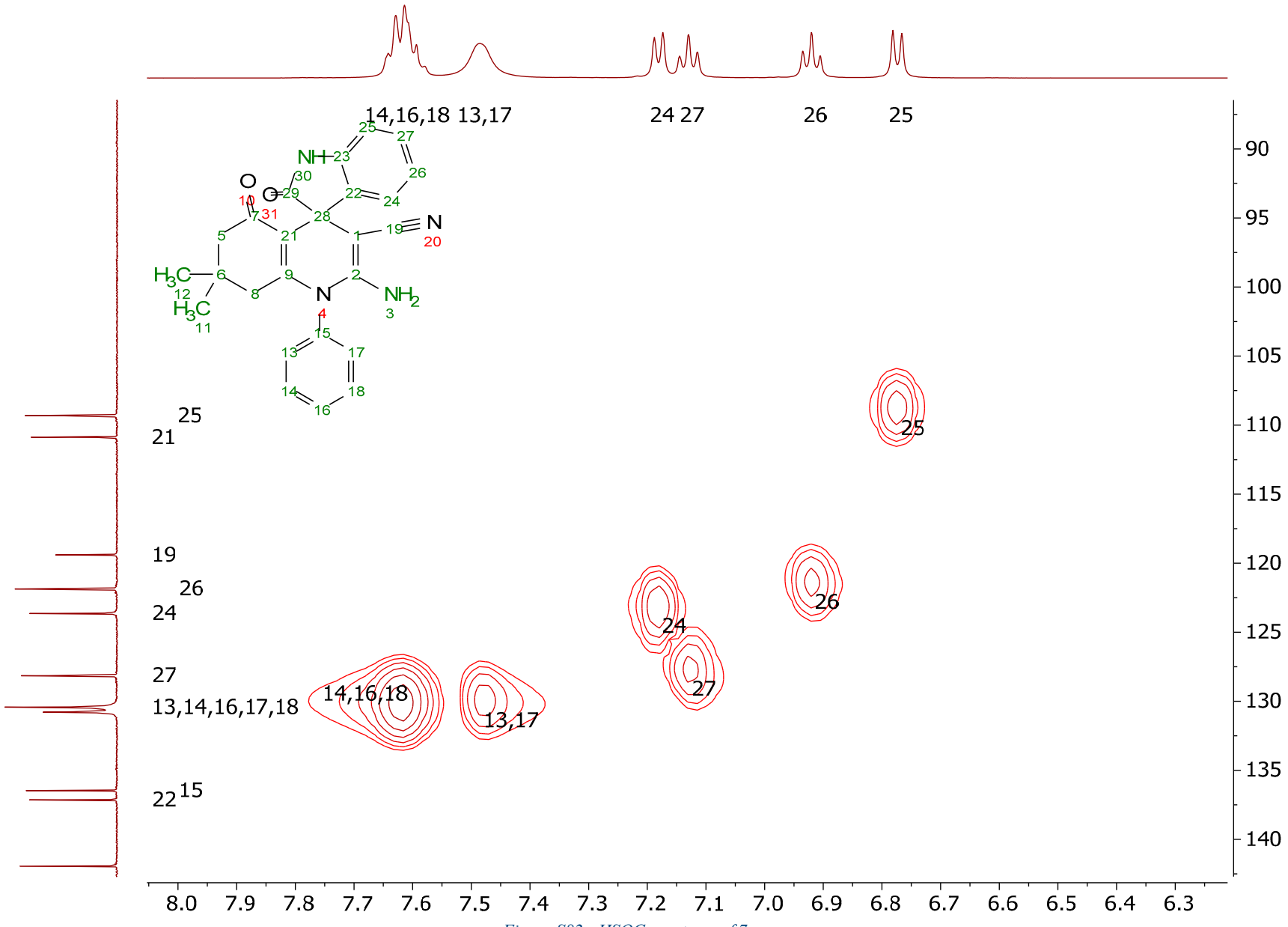


Figure S82 - HSQC spectrum of 7a

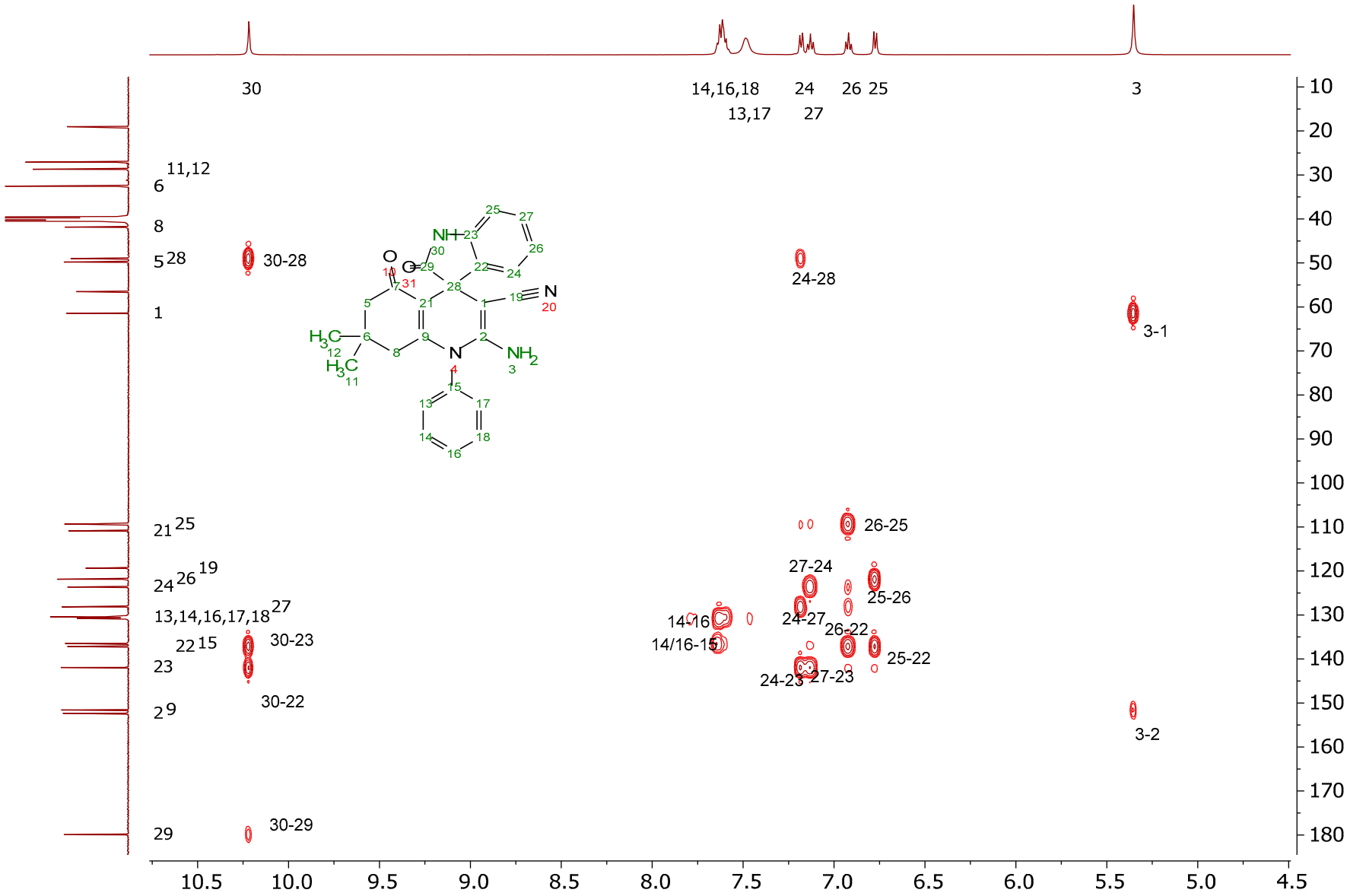


Figure S83 - Downfield region of HMBC spectrum of 7a

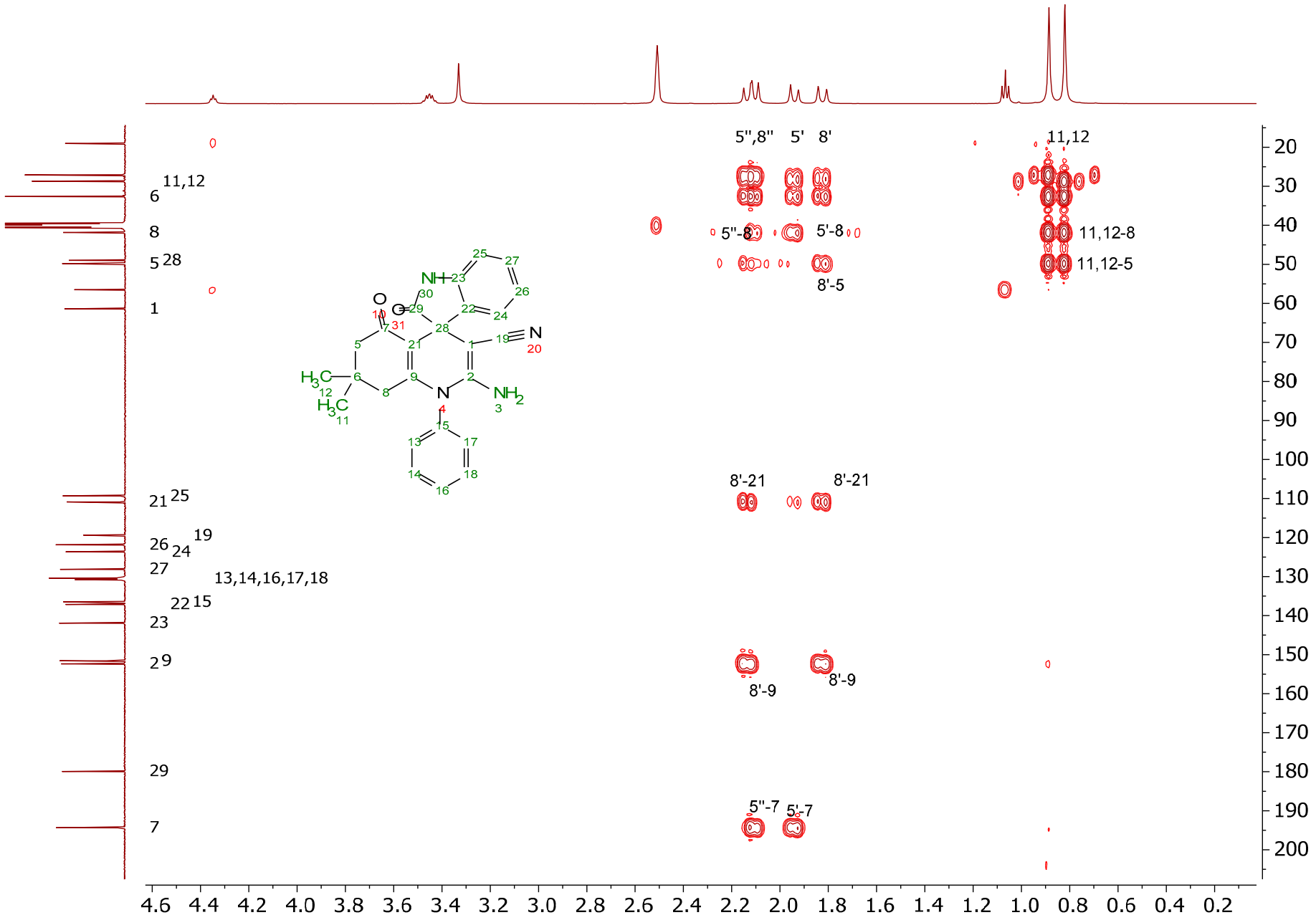


Figure S84 - Upfield region of HMBC spectrum of 7a

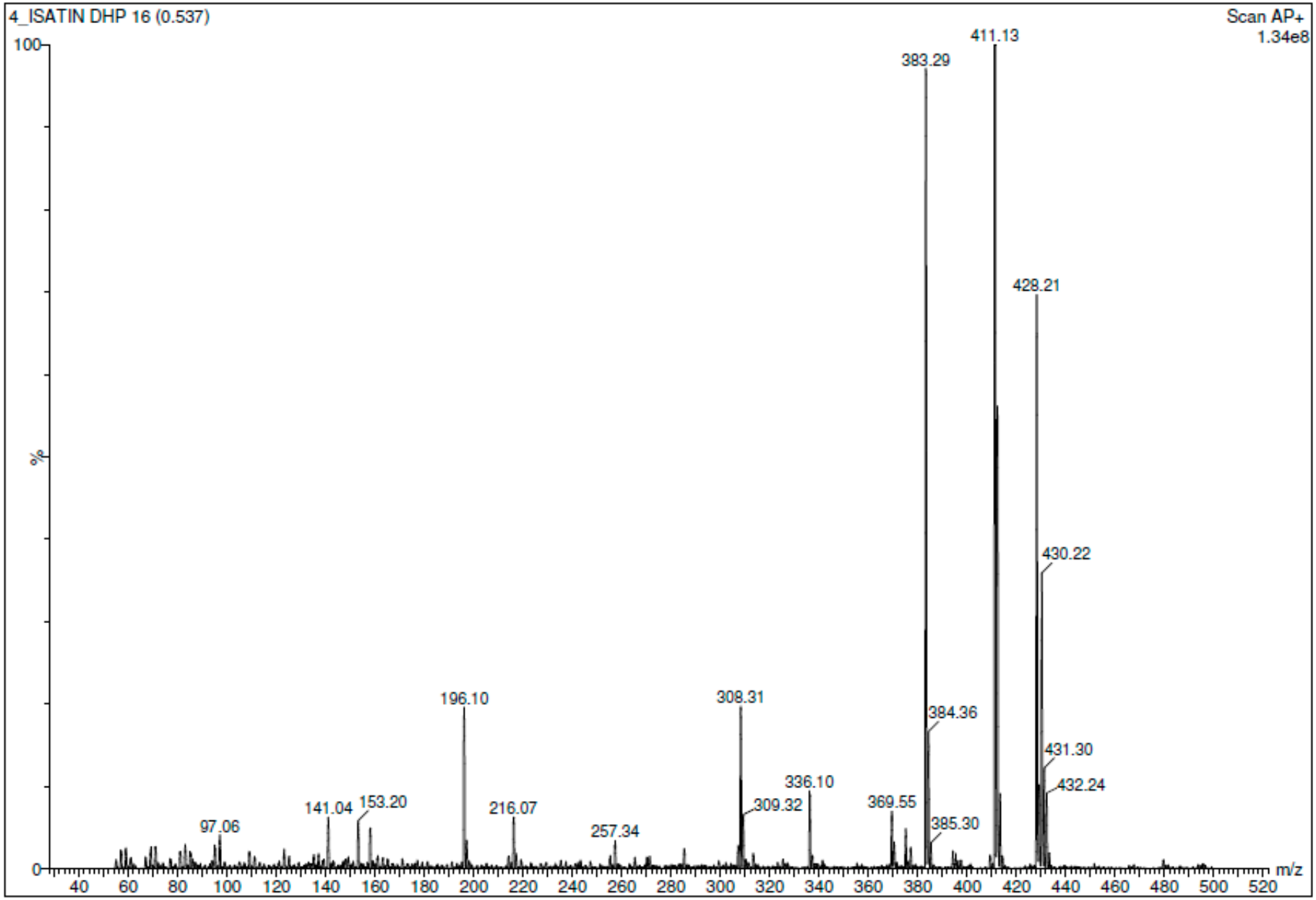
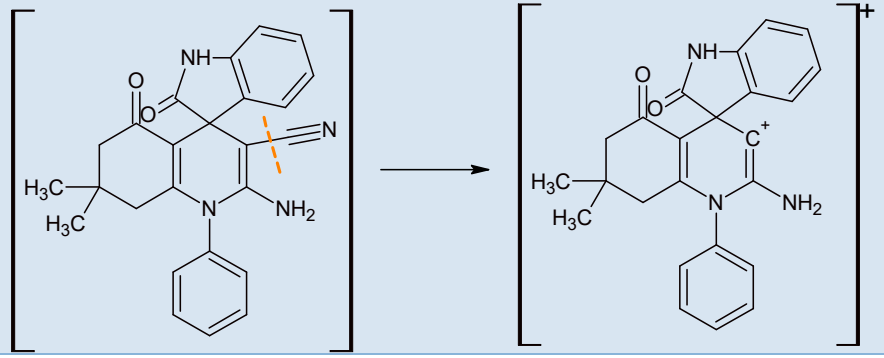
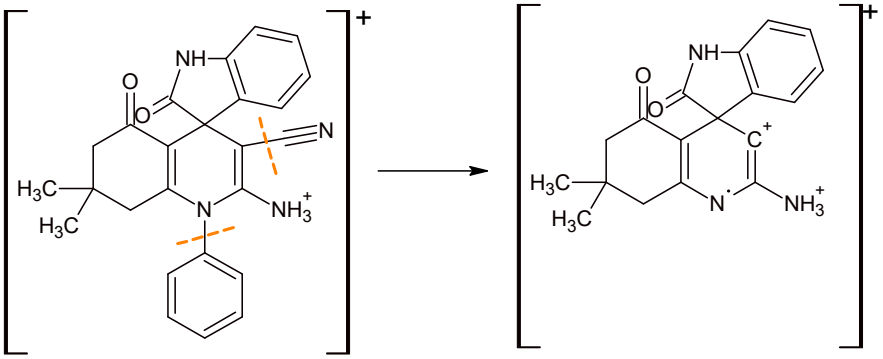


Figure S85 - MS spectrum of 7a

Table S10 - Peaks and fragmentation positions for MS spectrum of 7a

m/z	Fragmentation position and structure
428.21	$[M+H_2O]^+$
411.13	$[M+H]^+$
384.36	
308.31	

1.21. Product 7b: 2′-Amino-7′,7′-dimethyl-1′-(3-methylphenyl)-2,5′-dioxo--5′,6′,7′,8′-tetrahydro-1*H*-spiro[indoline-3,4′-quinoline]-3′-carbonitrile

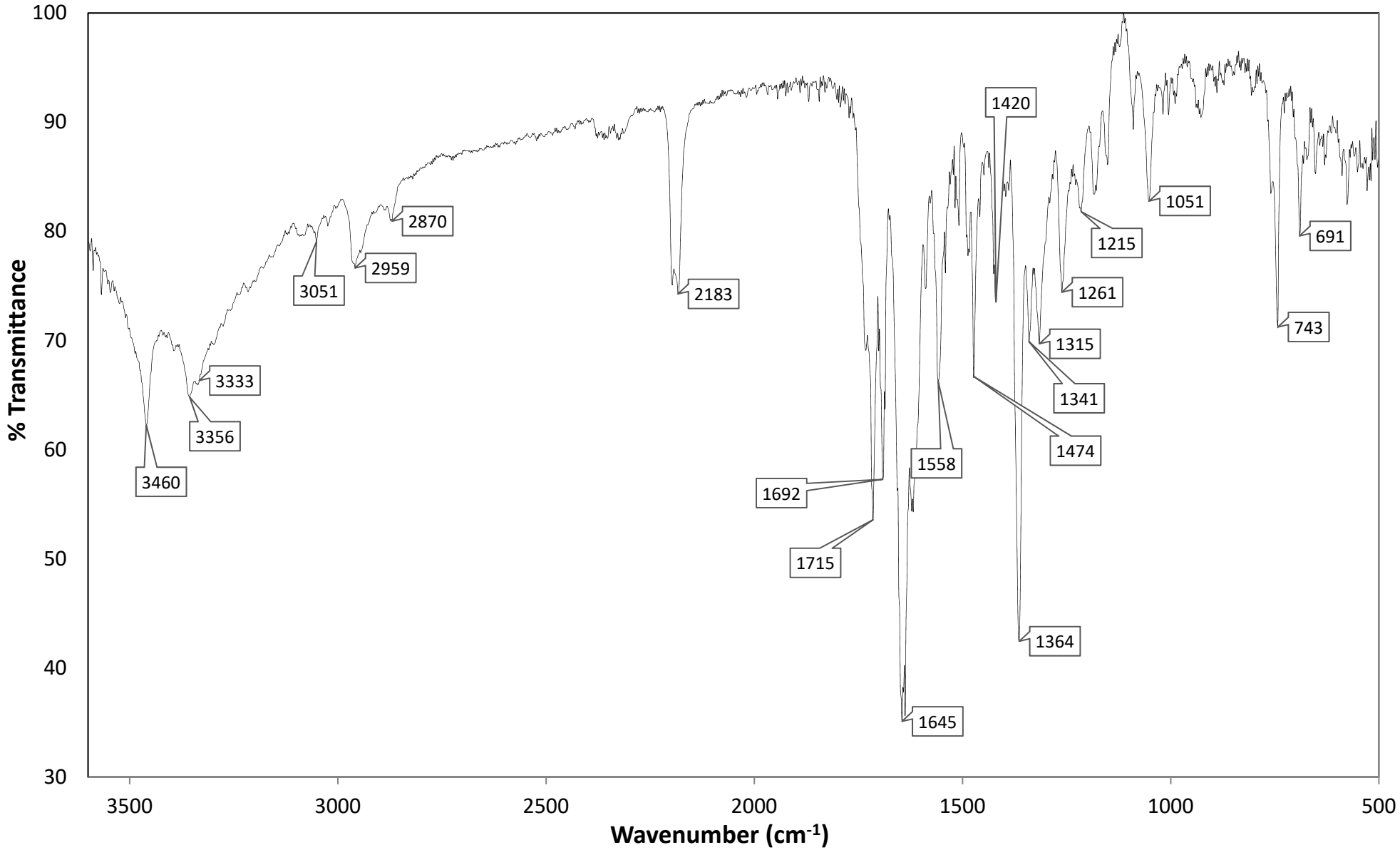


Figure S86 - IR spectrum of 7b

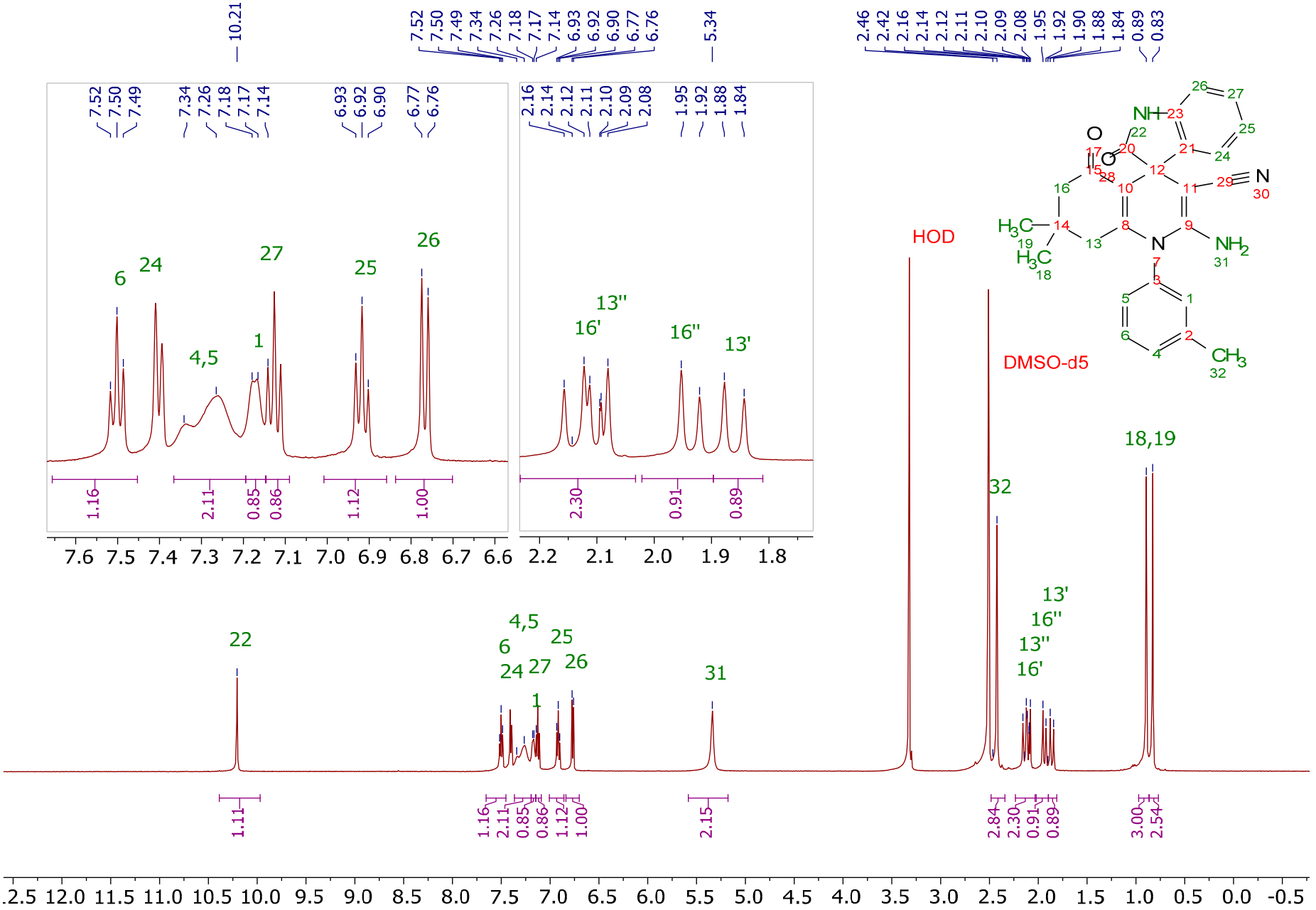


Figure S87 - ^{1}H NMR spectrum of 7b

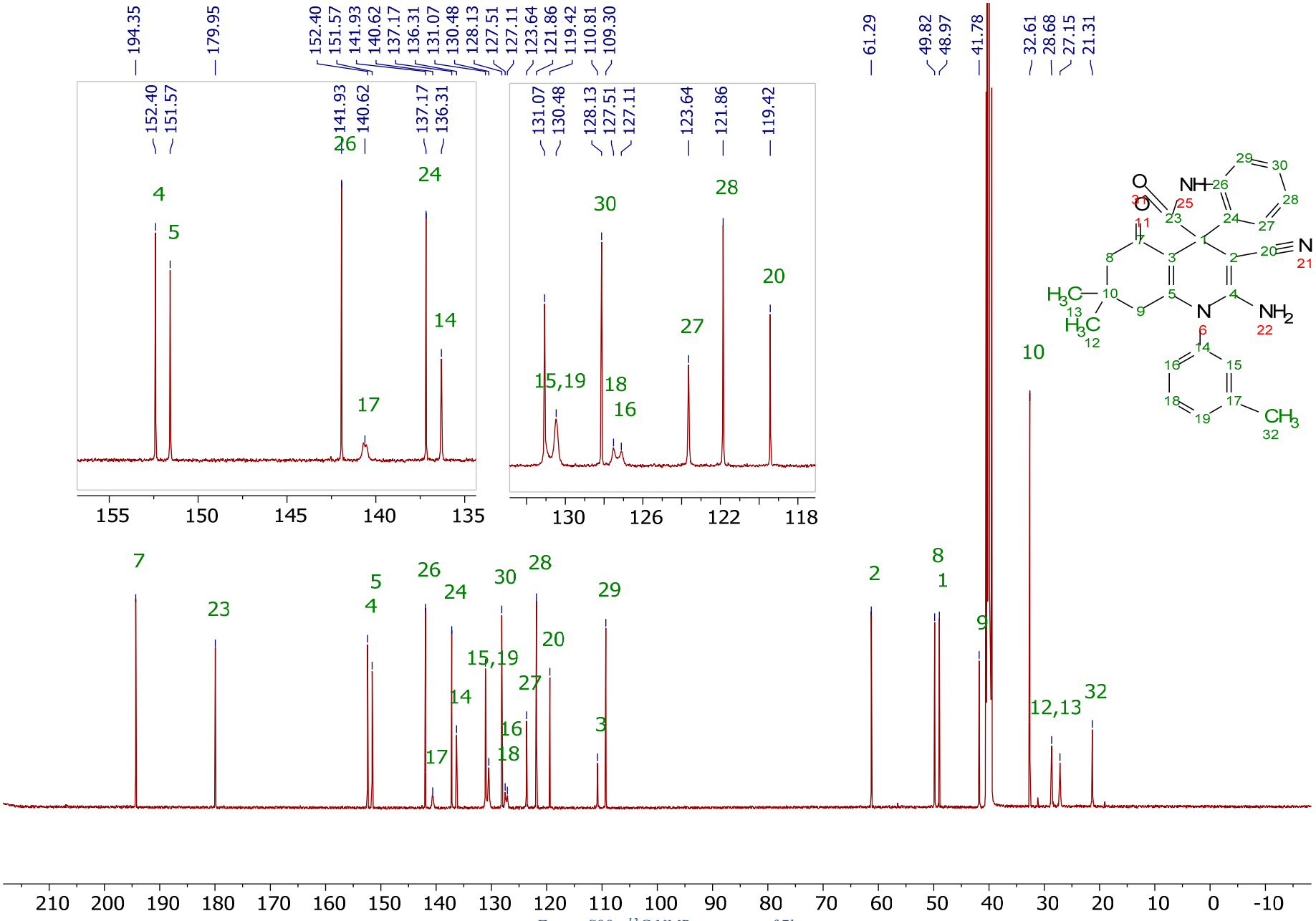


Figure S88 - ^{13}C NMR spectrum of 7b

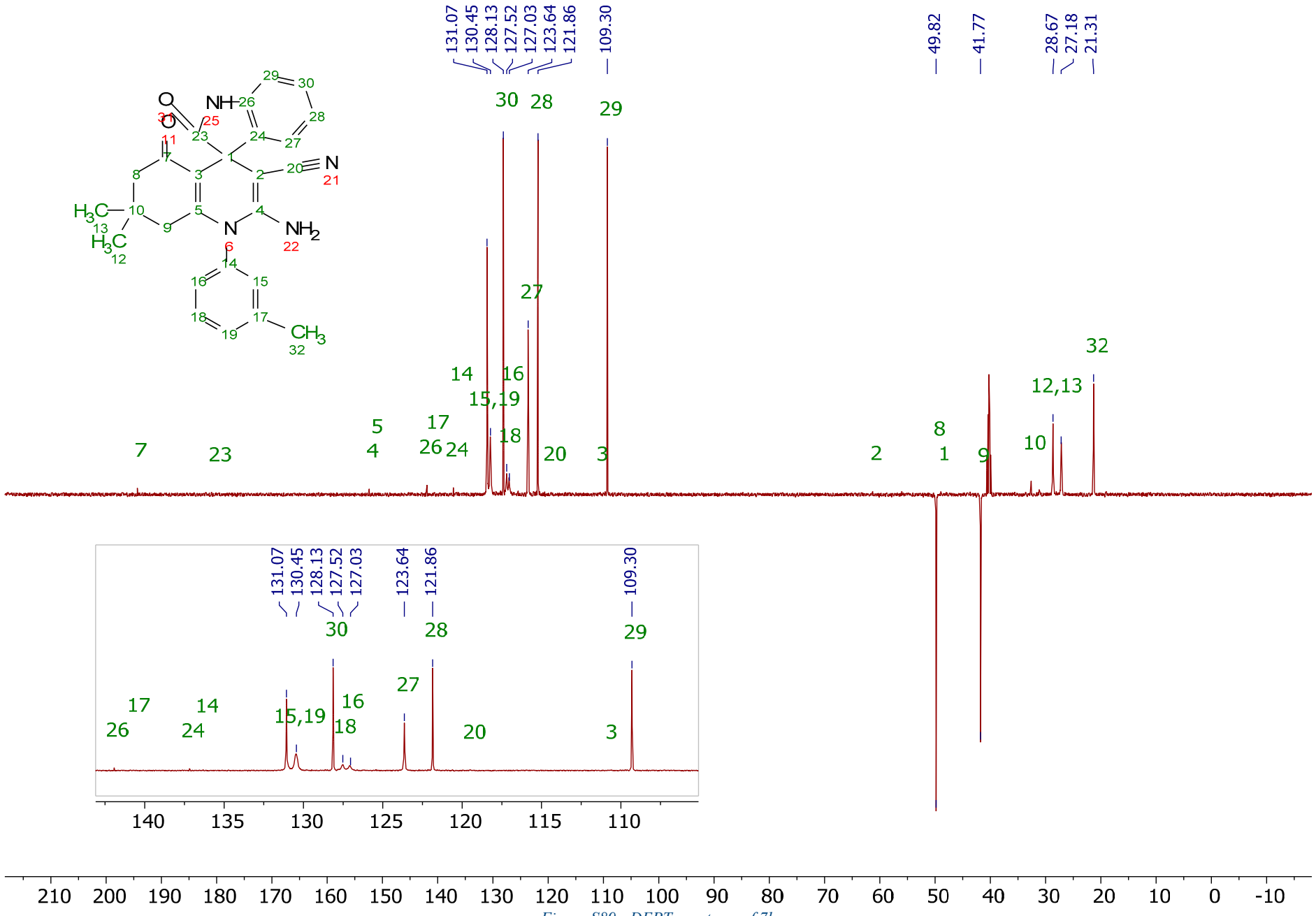


Figure S89 - DEPT spectrum of 7b

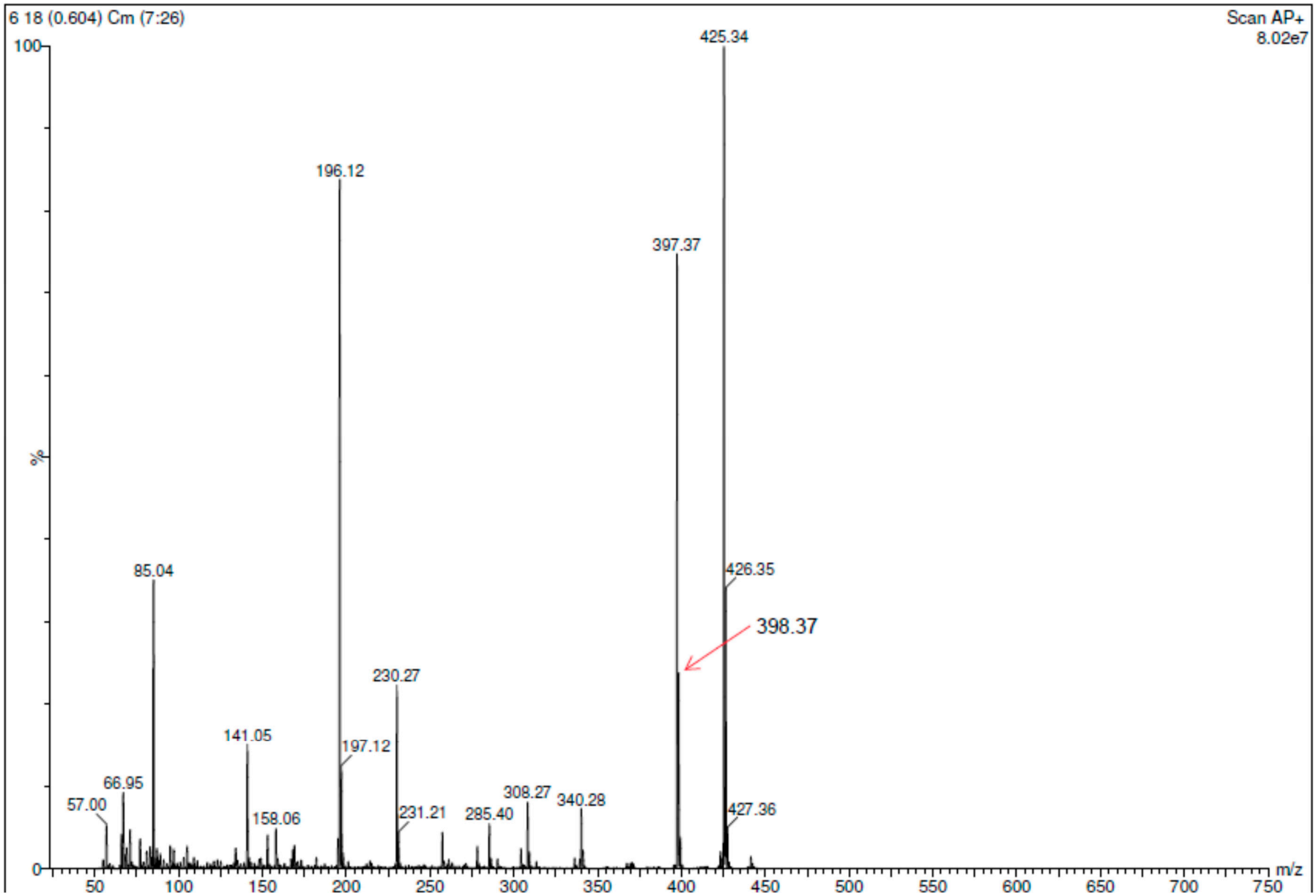
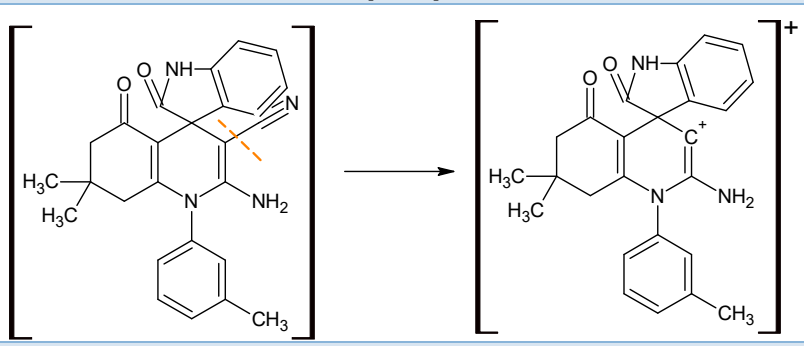
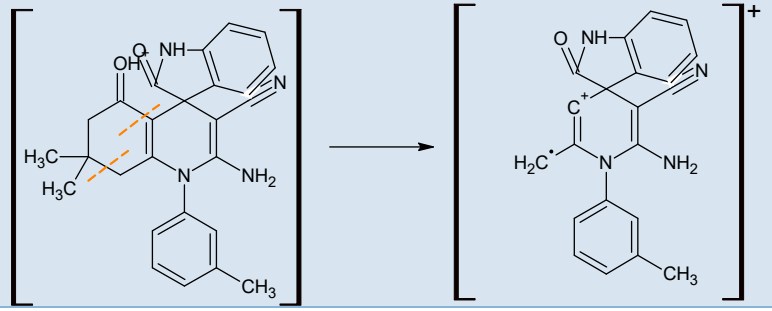
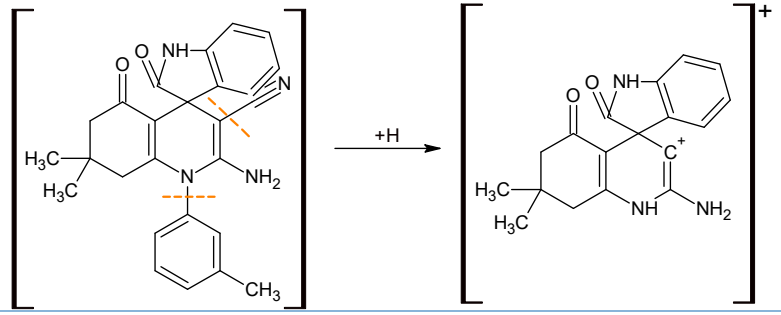


Figure S90 - MS spectrum of 7b

Table S11 - Fragmentation positions for peaks in MS spectrum of 7b

m/z	Fragmentation position
426.35	$[M+H]^+$
398.37	
340.28	
308.27	

1.22. Product 7c: 2′-Amino-7′,7′-dimethyl-1′-(3-nitrophenyl)-2,5′-dioxo-5′,6′,7′,8′-tetrahydro-1*H*-spiro[indoline-3,4′-quinoline]-3′-carbonitrile

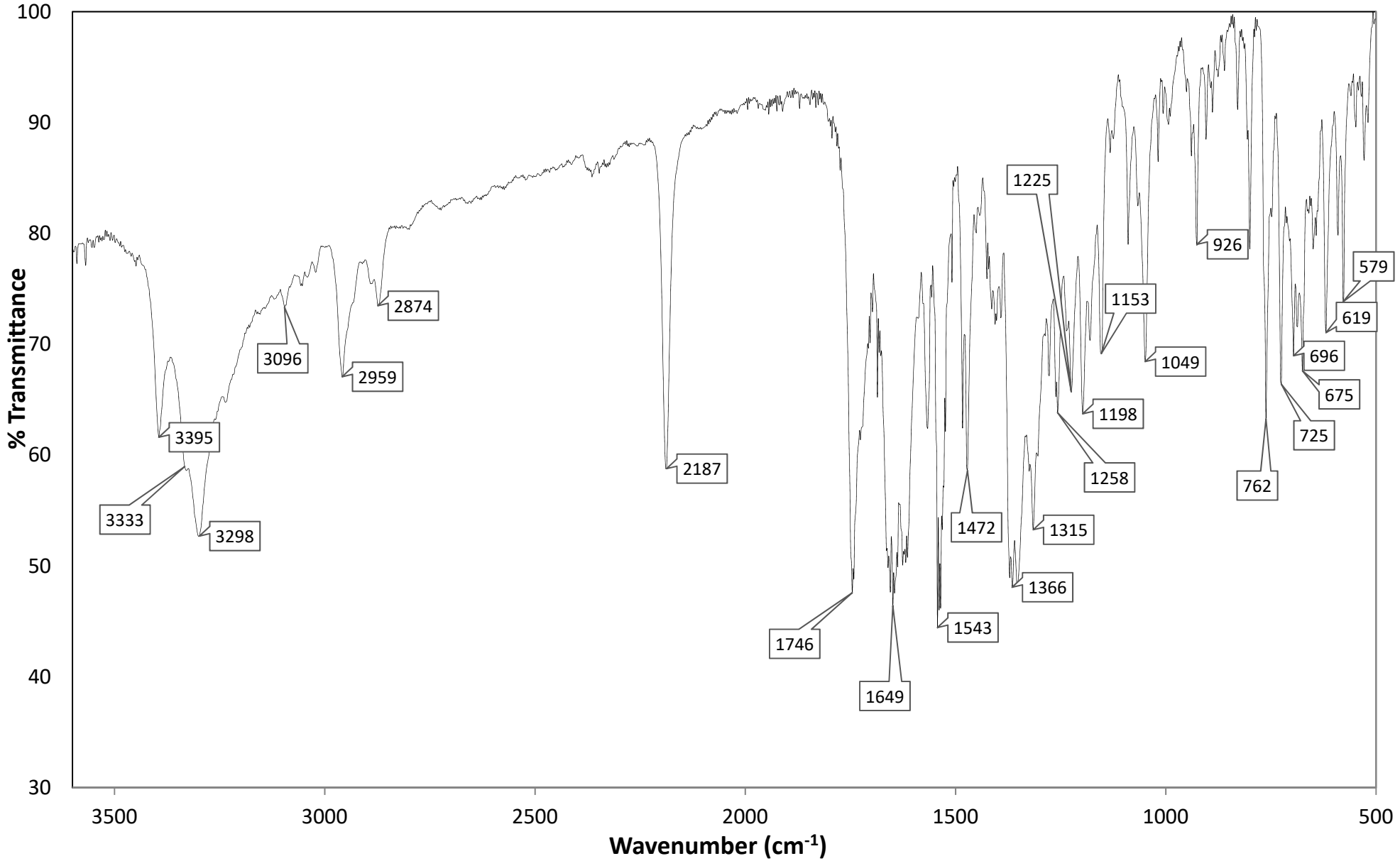


Figure S91 - IR spectrum of 7c

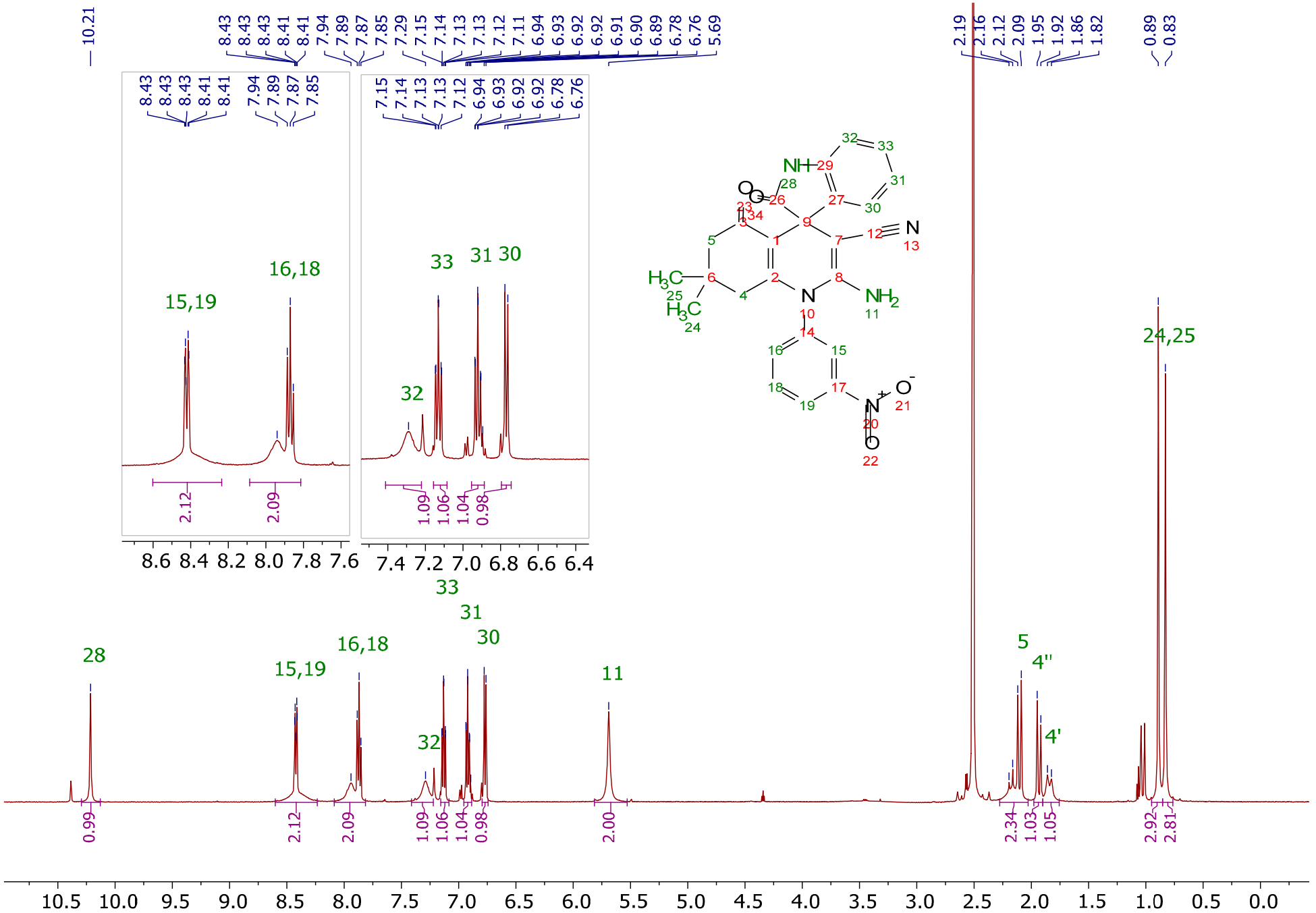


Figure S92 - ^{1}H NMR spectrum of 7c

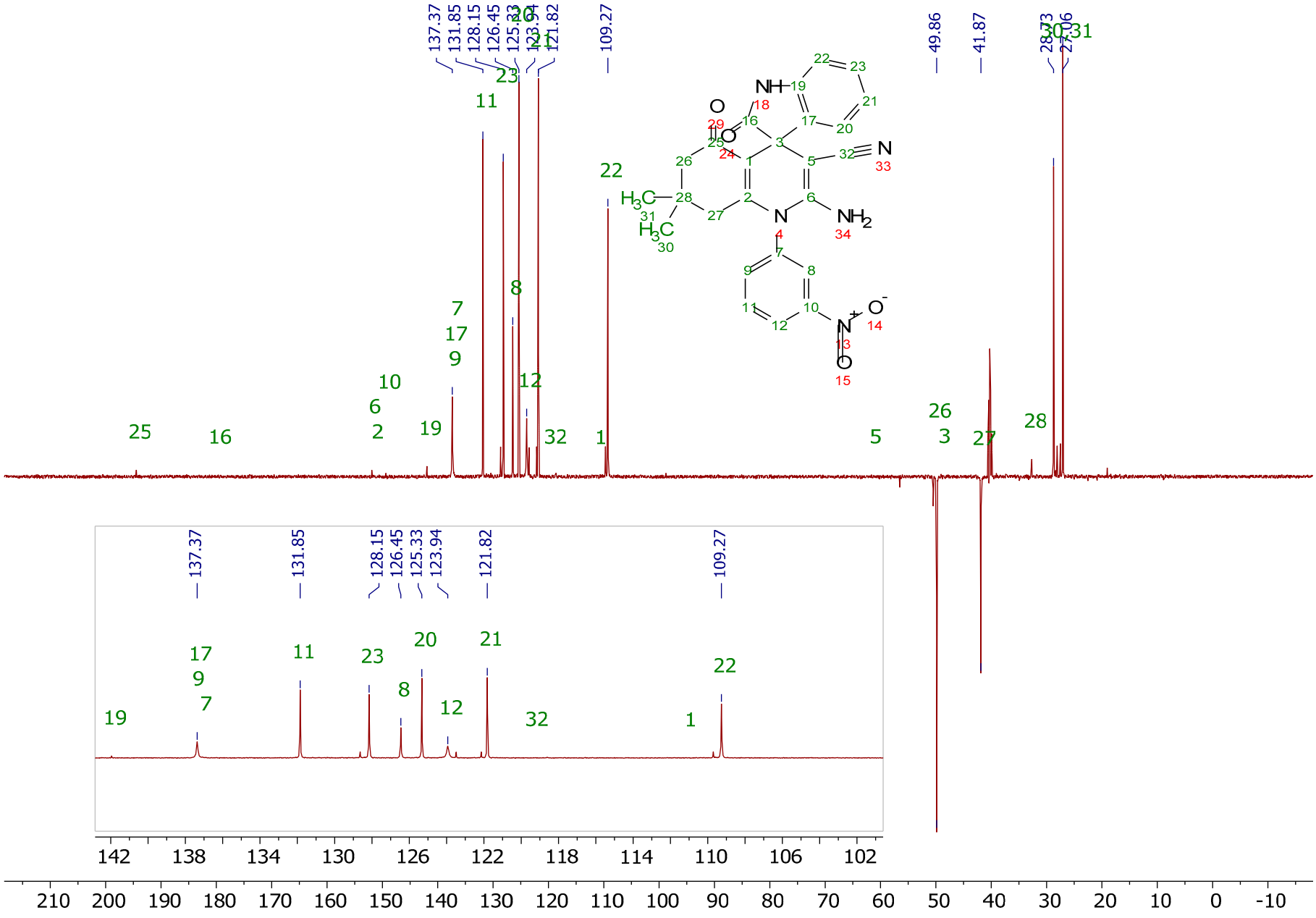


Figure S93 - ^{13}C NMR spectrum of 7c

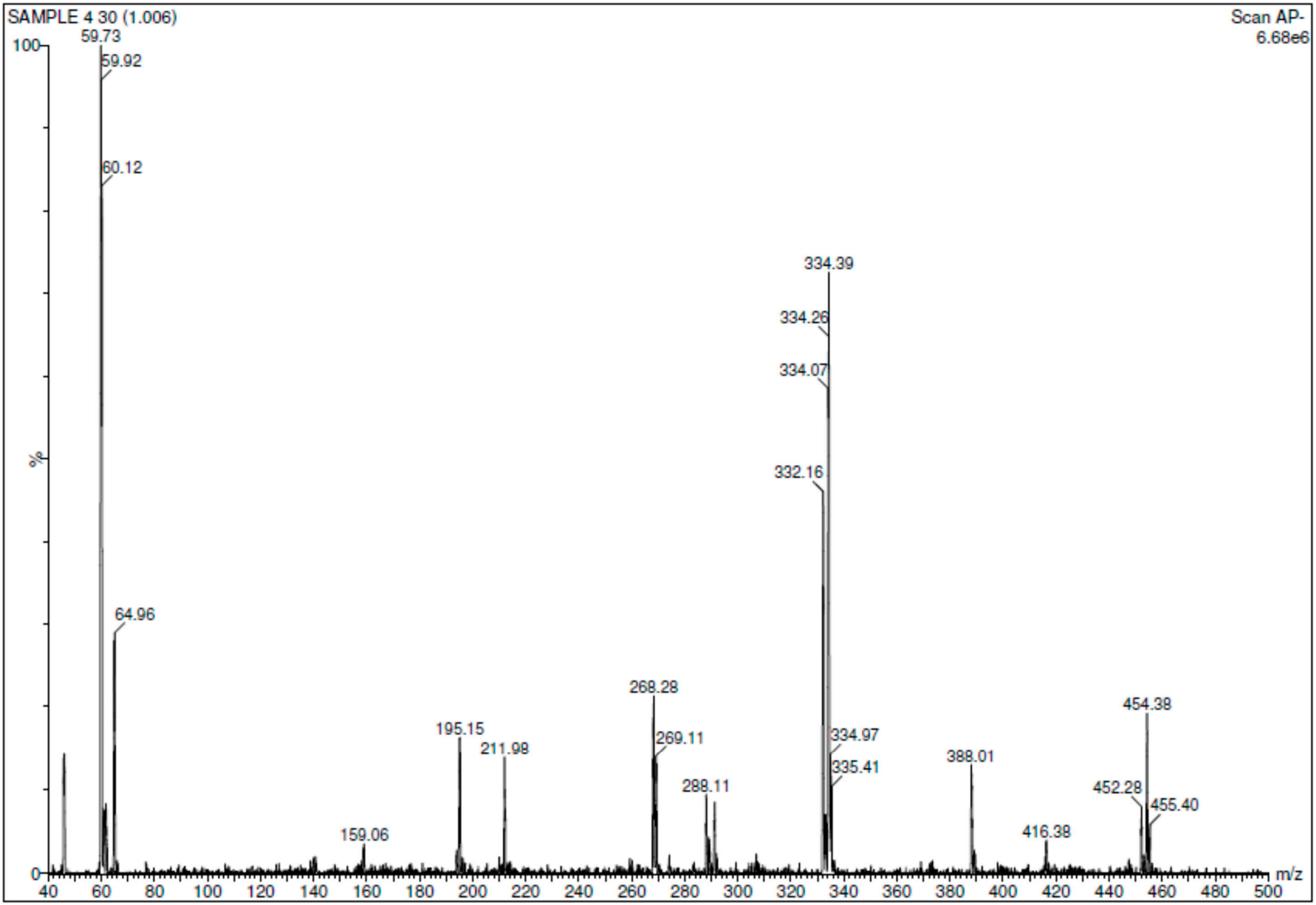
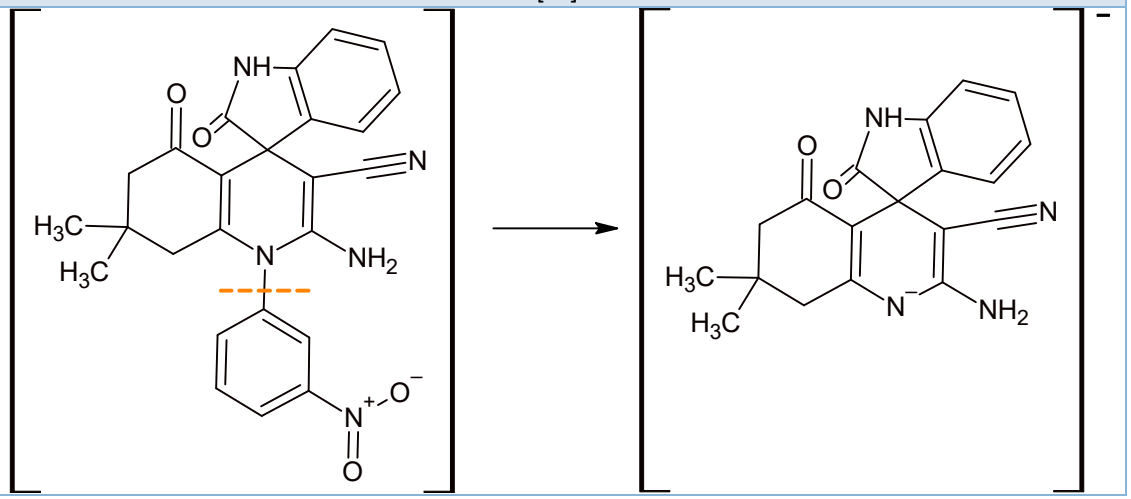


Figure S94 - Mass spectrum of 7c

Table S12 - Fragmentation positions for peaks in mass spectrum of 7c

m/z	Fragmentation position
455.40	$[M]^-$
334.07	

1.23. Product 7d: 2′-Amino-1′-(4-methoxyphenyl)-7′,7′-dimethyl-2,5′-dioxo-5′,6′,7′,8′-tetrahydro-1*H*-spiro[indoline-3,4′-quinoline]-3′-carbonitrile

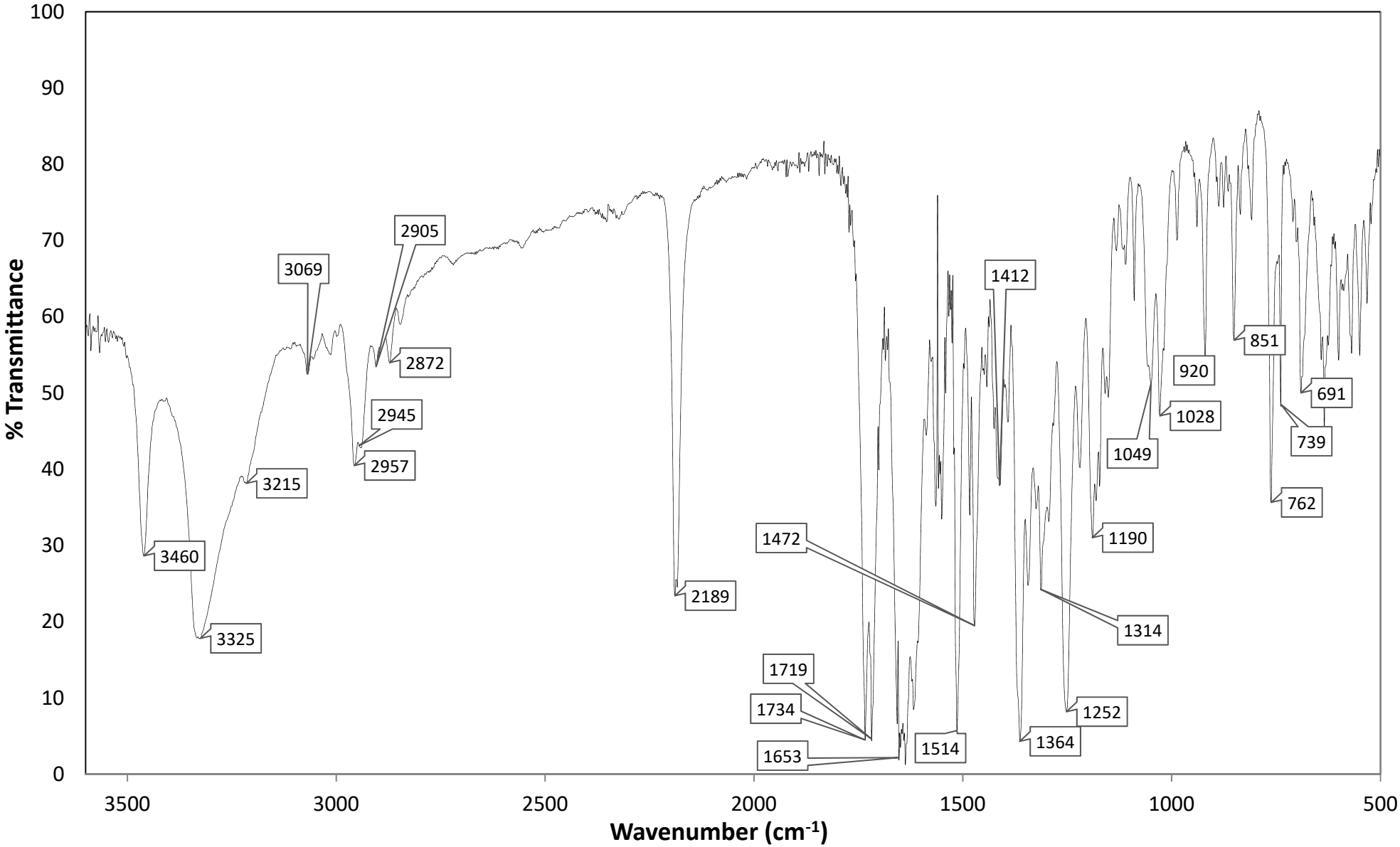


Figure S95 - IR spectrum of 7d

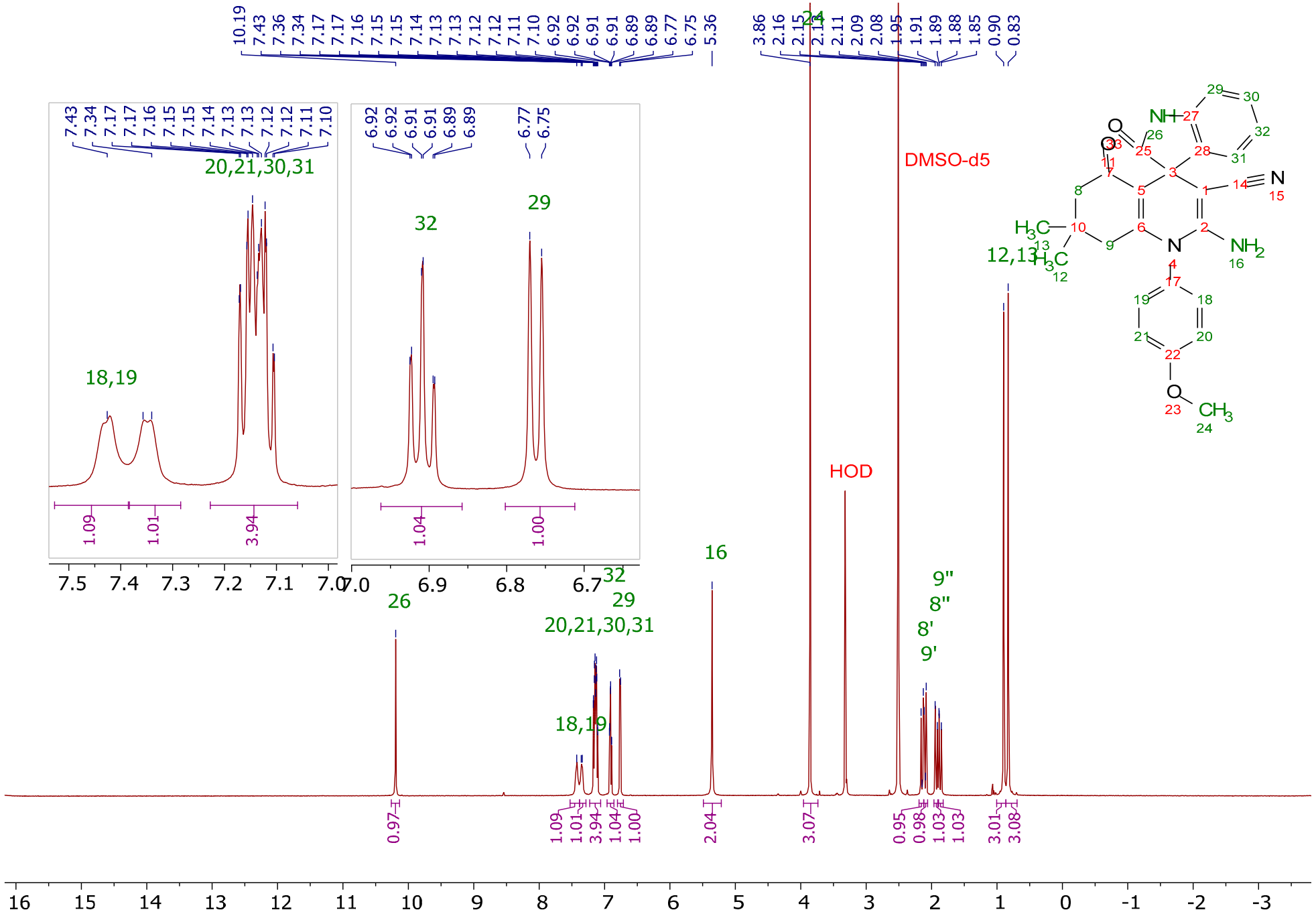


Figure S96 - ^{1}H NMR spectrum of 7d

1.24. Product 7e: 2′-Amino-1′-(4-chlorophenyl)-7′,7′-dimethyl-2,5′-dioxo-5′,6′,7′,8′-tetrahydro-1*H*-spiro[indoline-3,4′-quinoline]-3′-carbonitrile

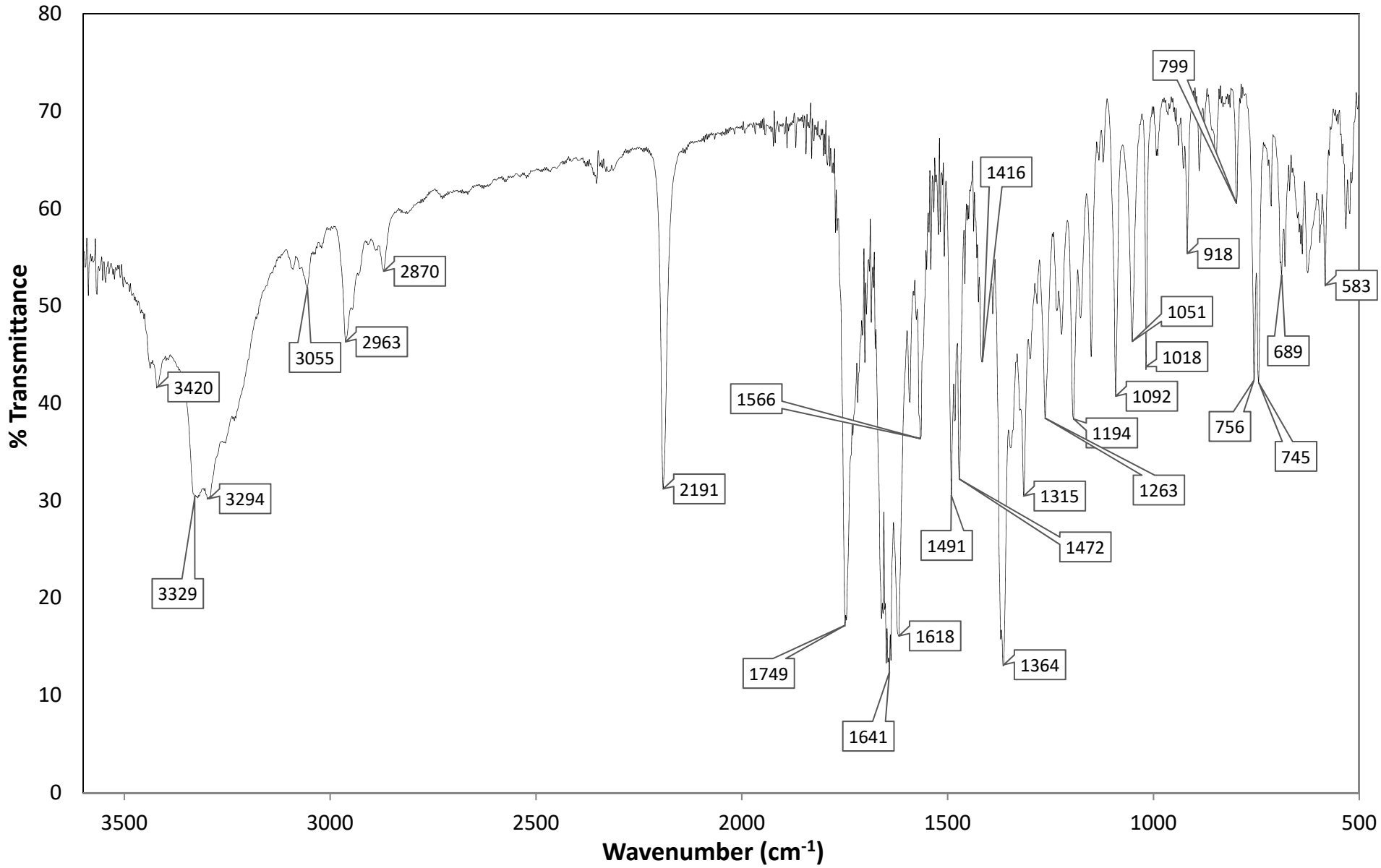


Figure S97 - IR spectrum of 7e

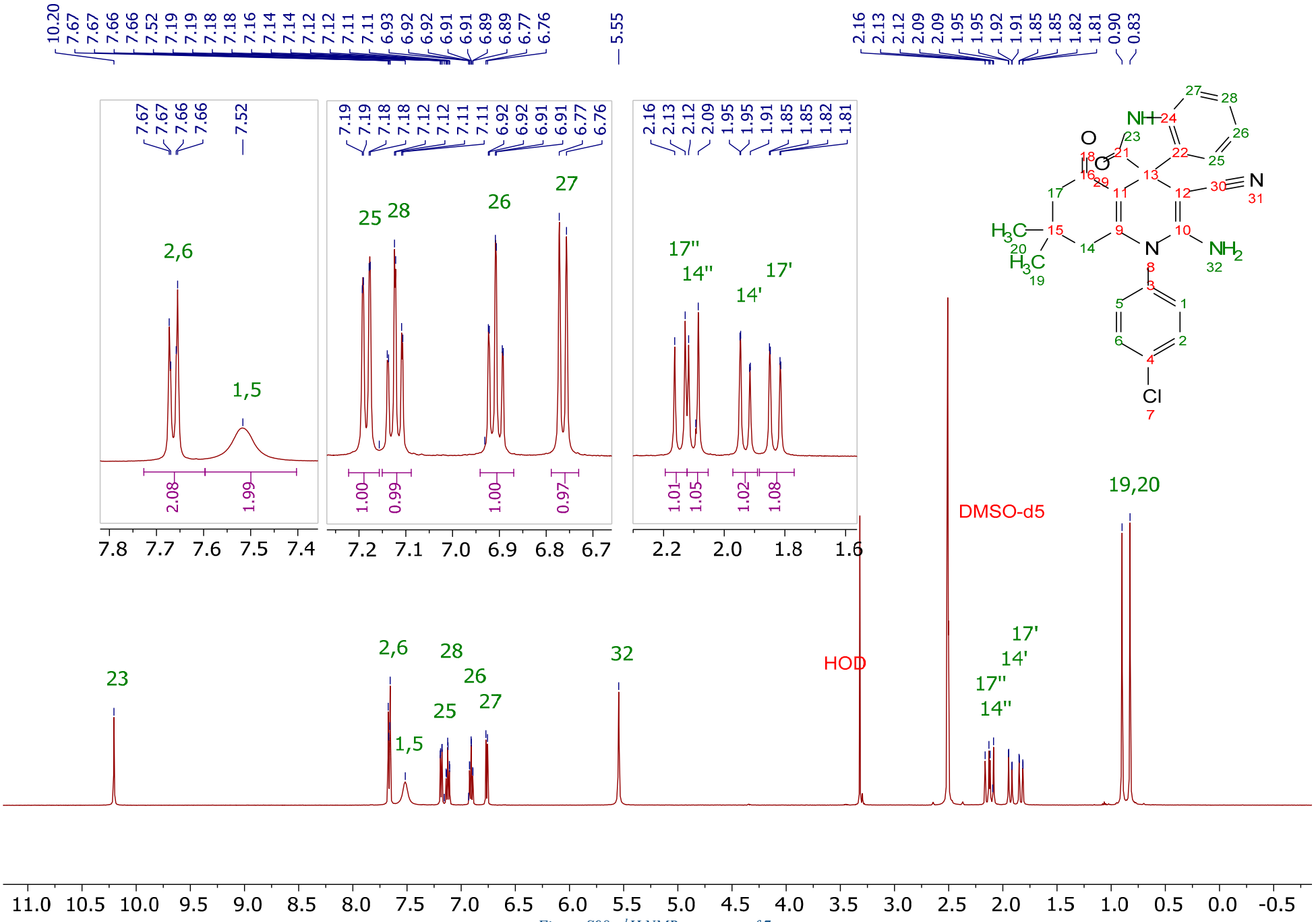


Figure S98 - ^{1}H NMR spectrum of 7e

1.25. Product 7f: 2′-Amino-1′-(4-bromophenyl)-7′,7′-dimethyl-2,5′-dioxo-5′,6′,7′,8′-tetrahydro-1*H*-spiro[indoline-3,4′-quinoline]-3′-carbonitrile

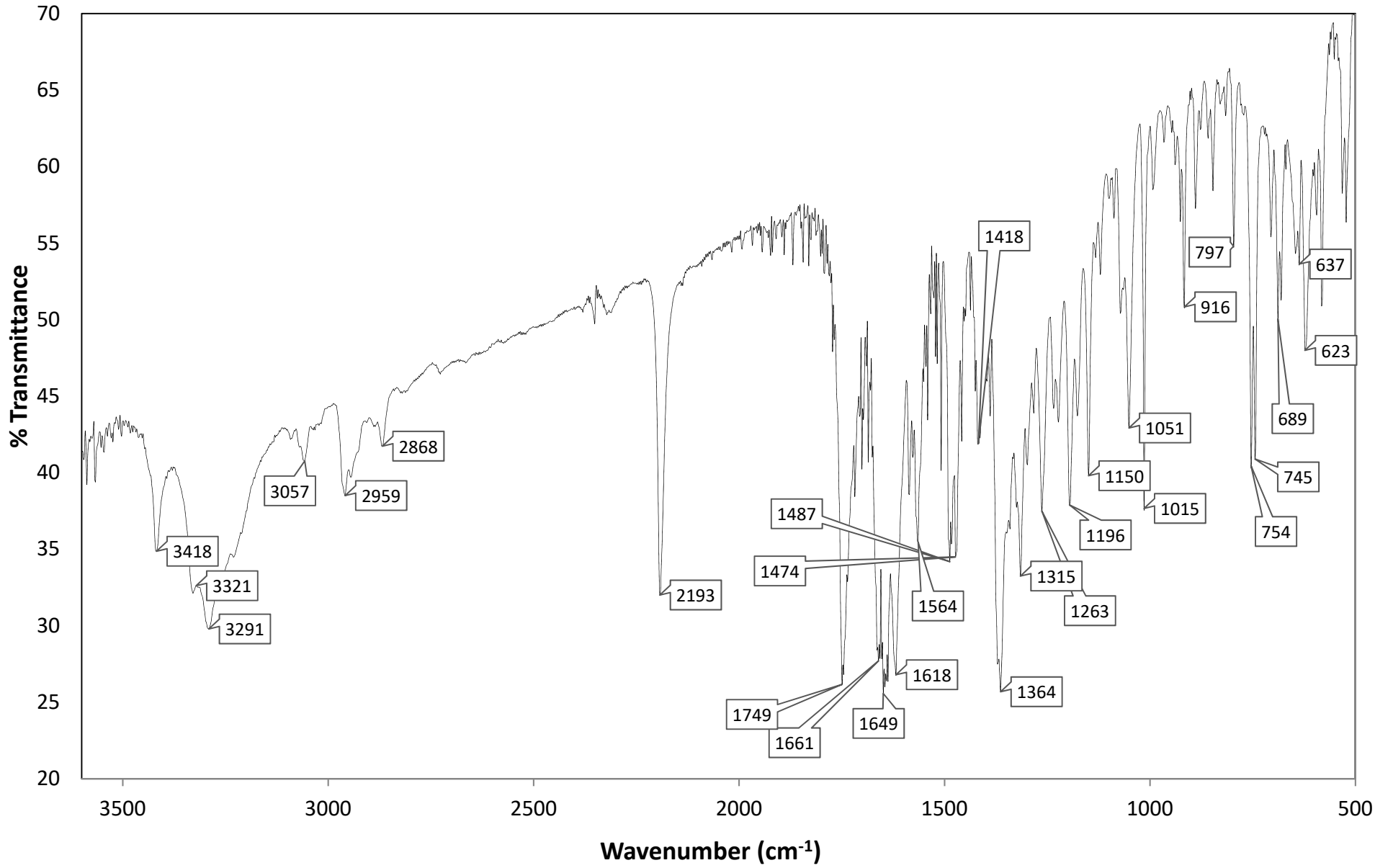


Figure S99 - IR spectrum of 7f

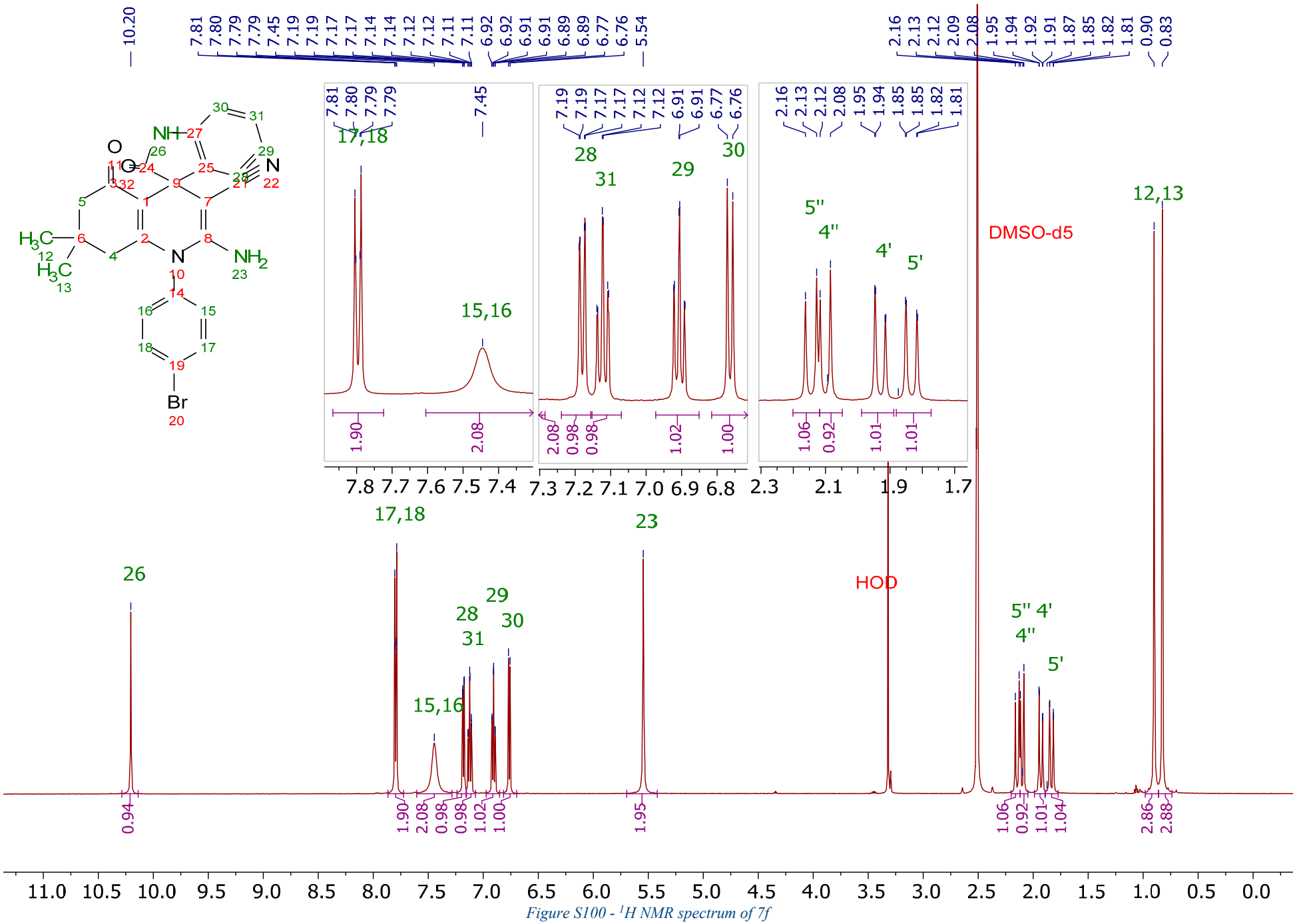


Figure S100 - ^{1}H NMR spectrum of 7f

1.26. Product 7g: 2′-Amino-7′,7′-dimethyl-1′-(4-methylphenyl)-2,5′-dioxo-5′,6′,7′,8′-tetrahydro-1*H*-spiro[indoline-3,4′-quinoline]-3′-carbonitrile

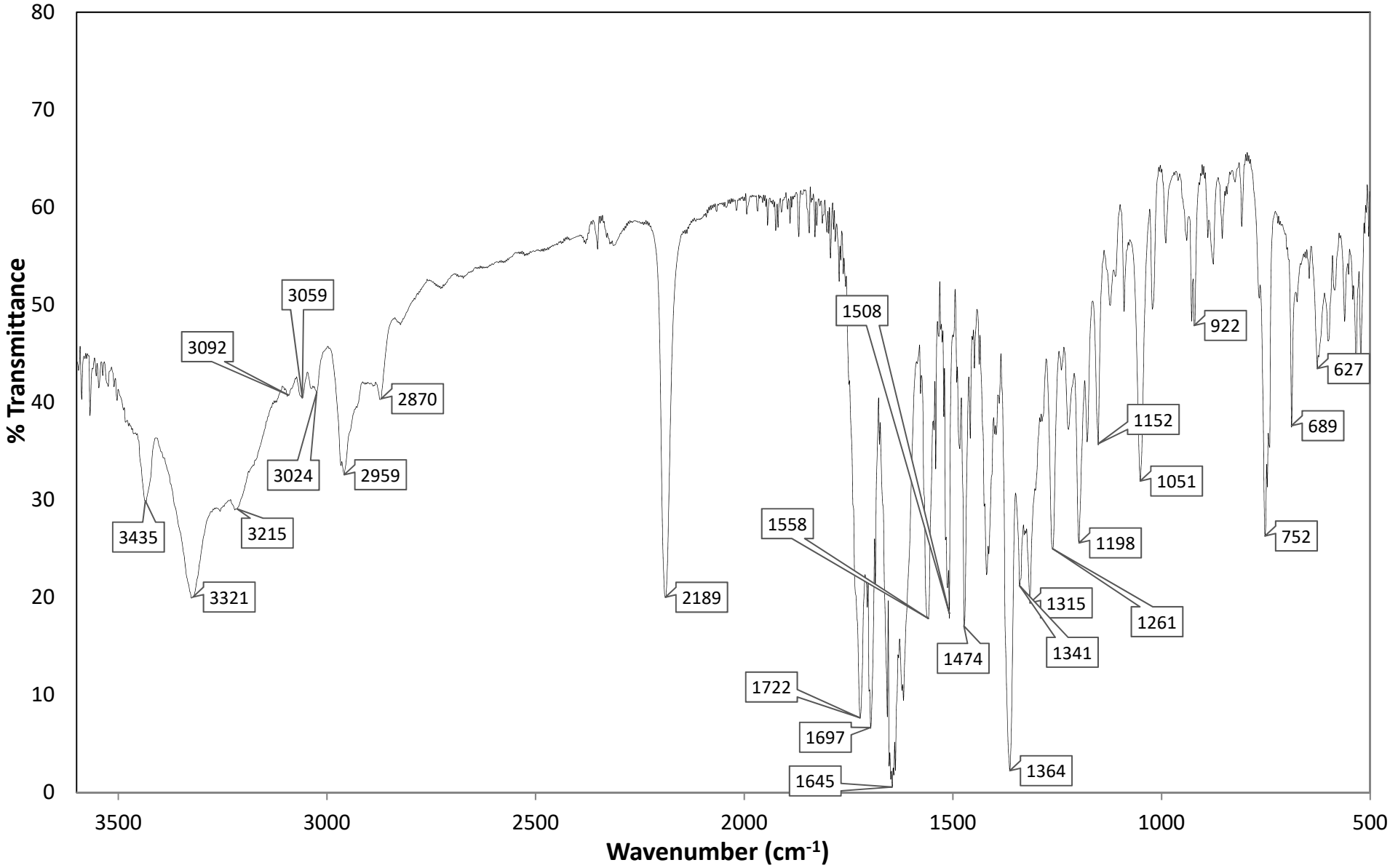


Figure S101 - IR spectrum of 7g

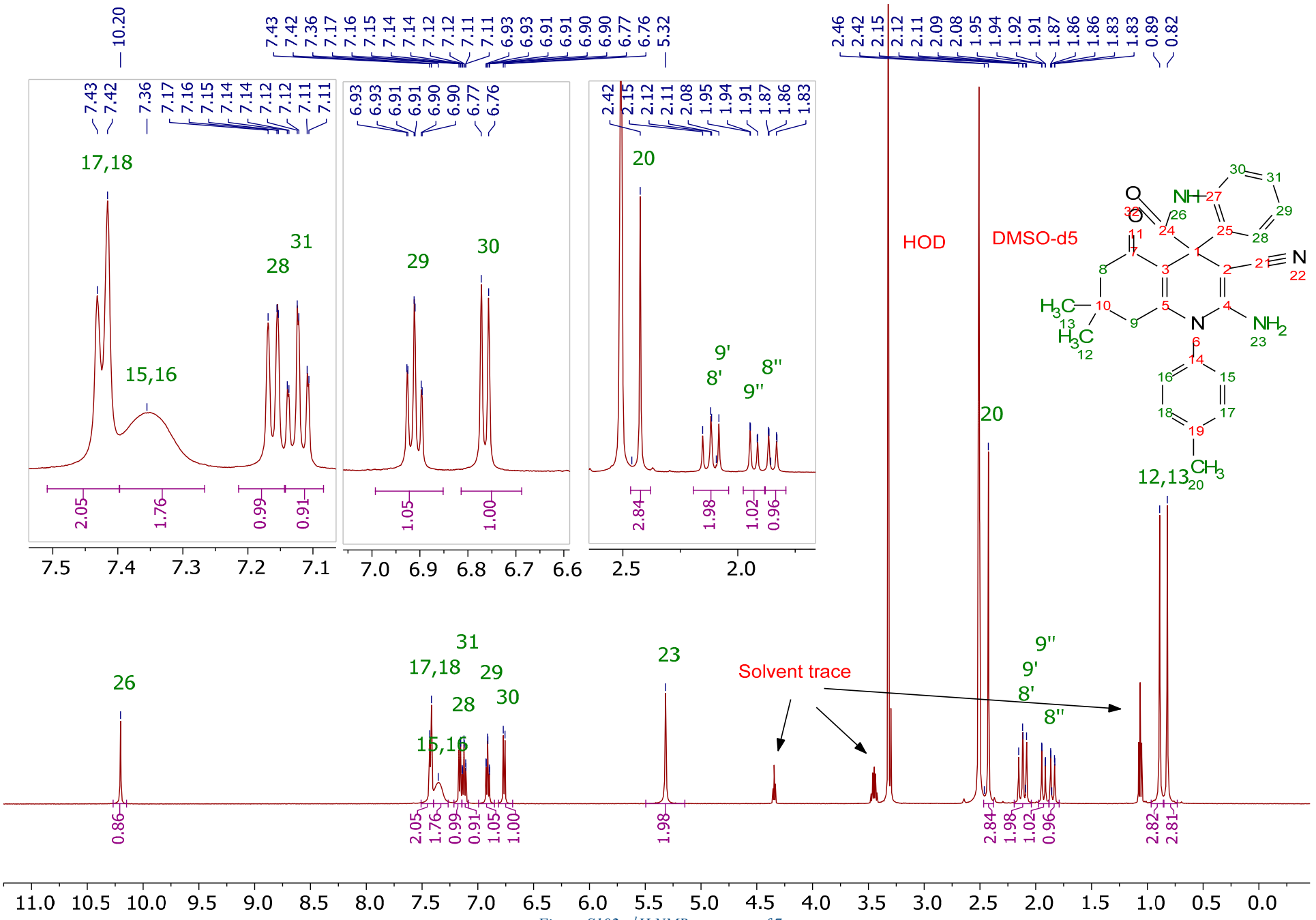


Figure S102 - ^{1}H NMR spectrum of 7g

1.27. Product 7i: 2′-Amino-1′-(3-chlorophenyl)-7′,7′-dimethyl-1′-2,5′-dioxo-5′,6′,7′,8′-tetrahydro-1*H*-spiro[indoline-3,4′-quinoline]-3′-carbonitrile

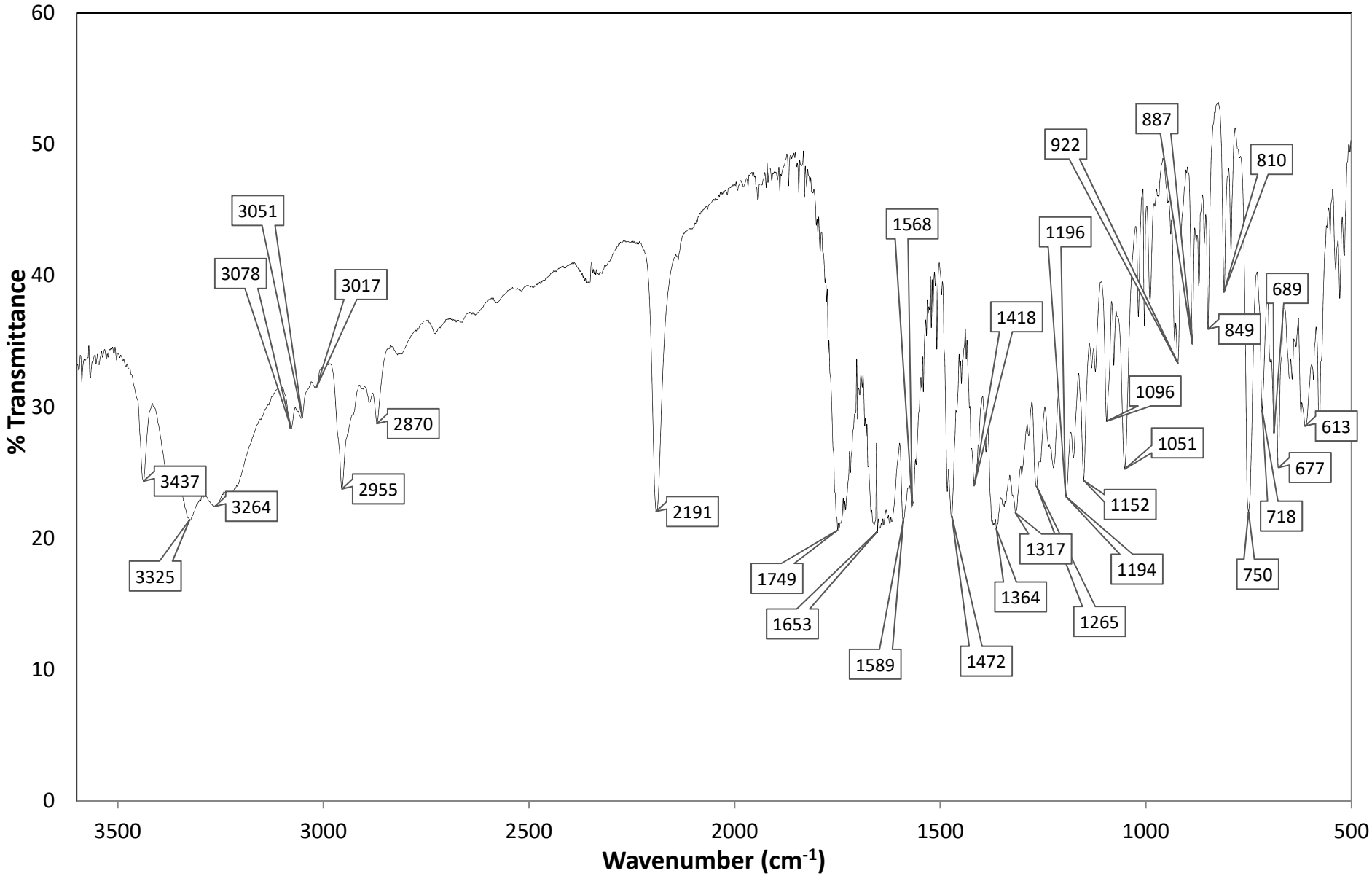


Figure S103 - IR spectrum of 7i

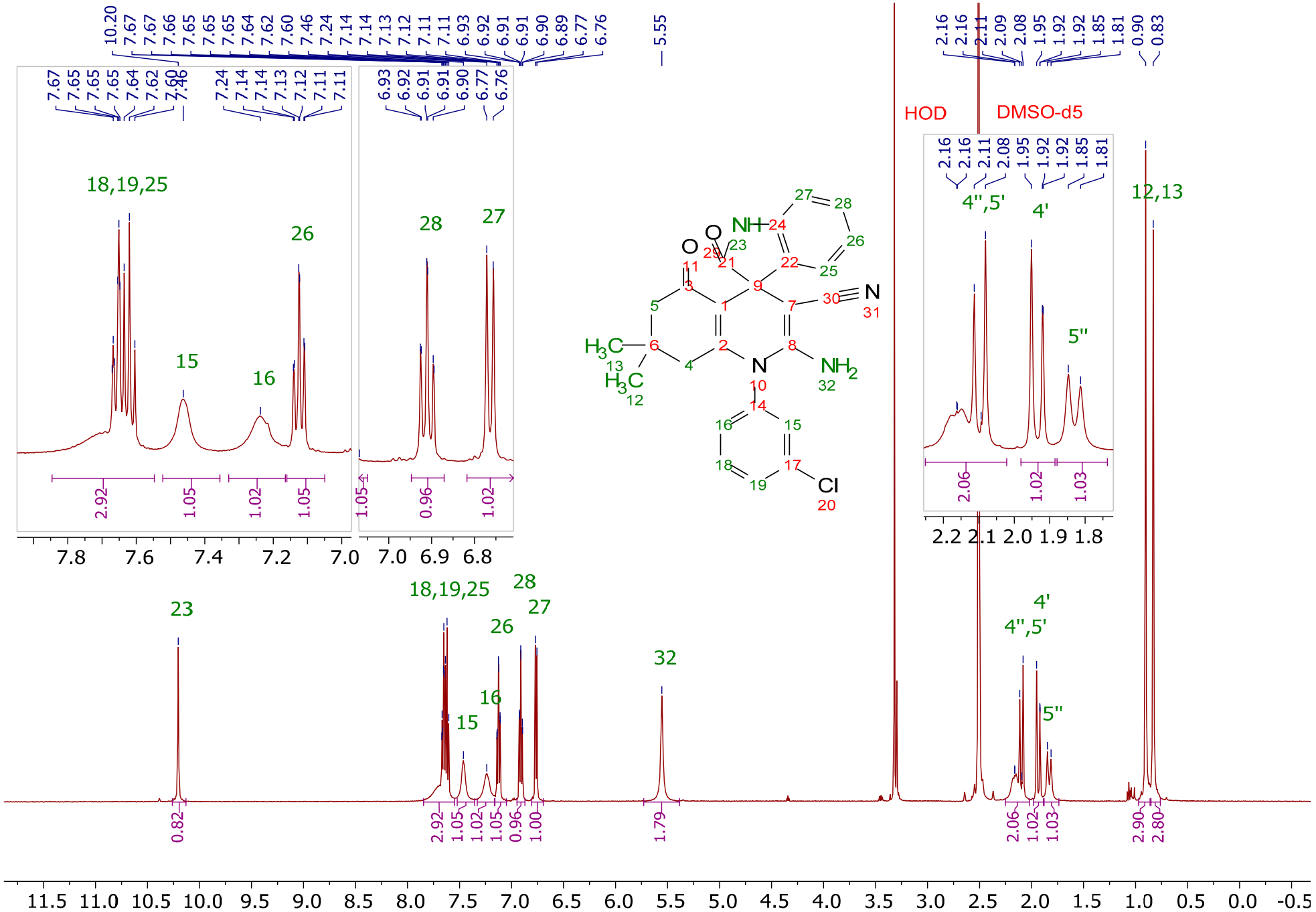


Figure S104 - ^{1}H NMR spectrum of 7i

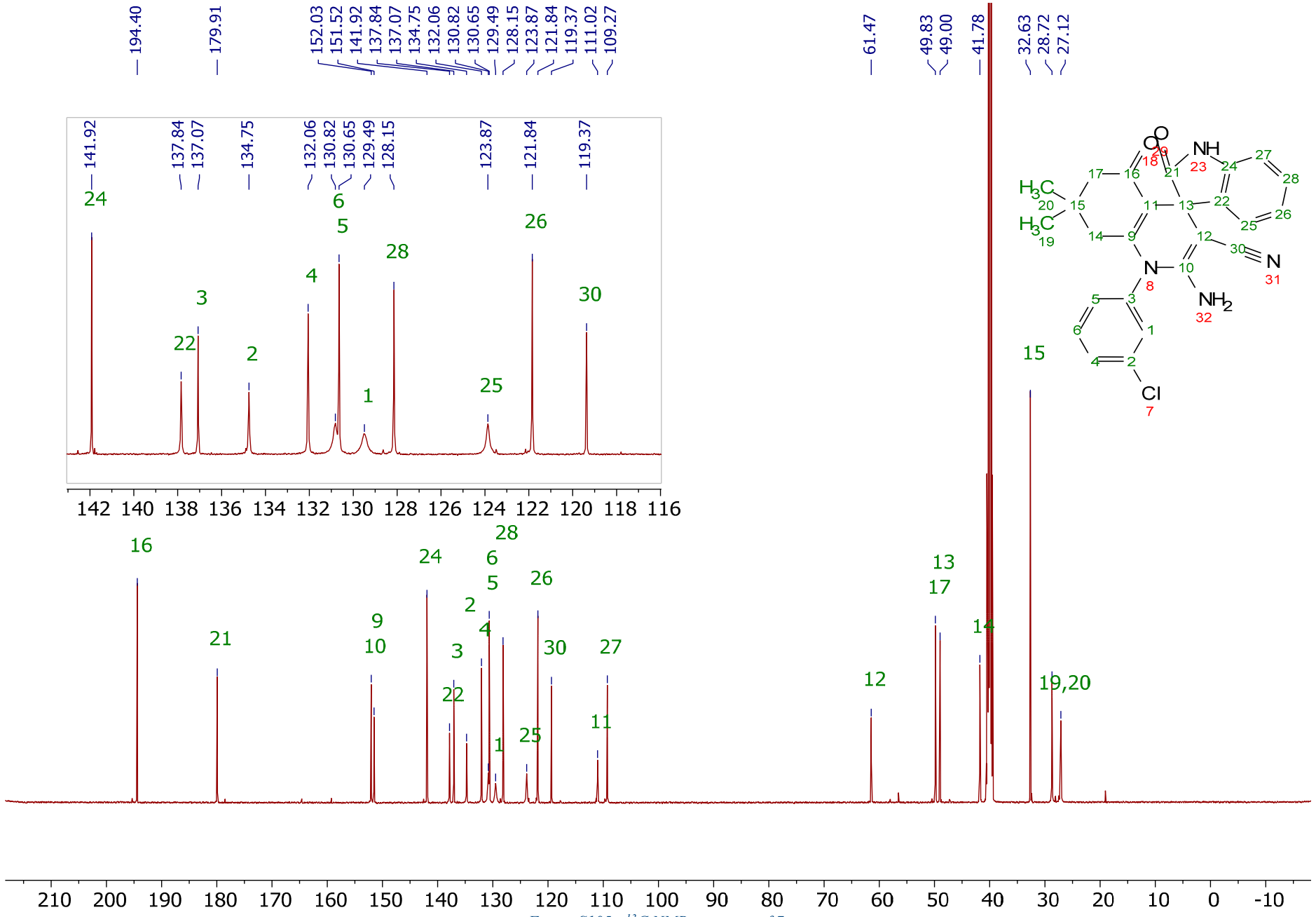


Figure S105 - ^{13}C NMR spectrum of 7i

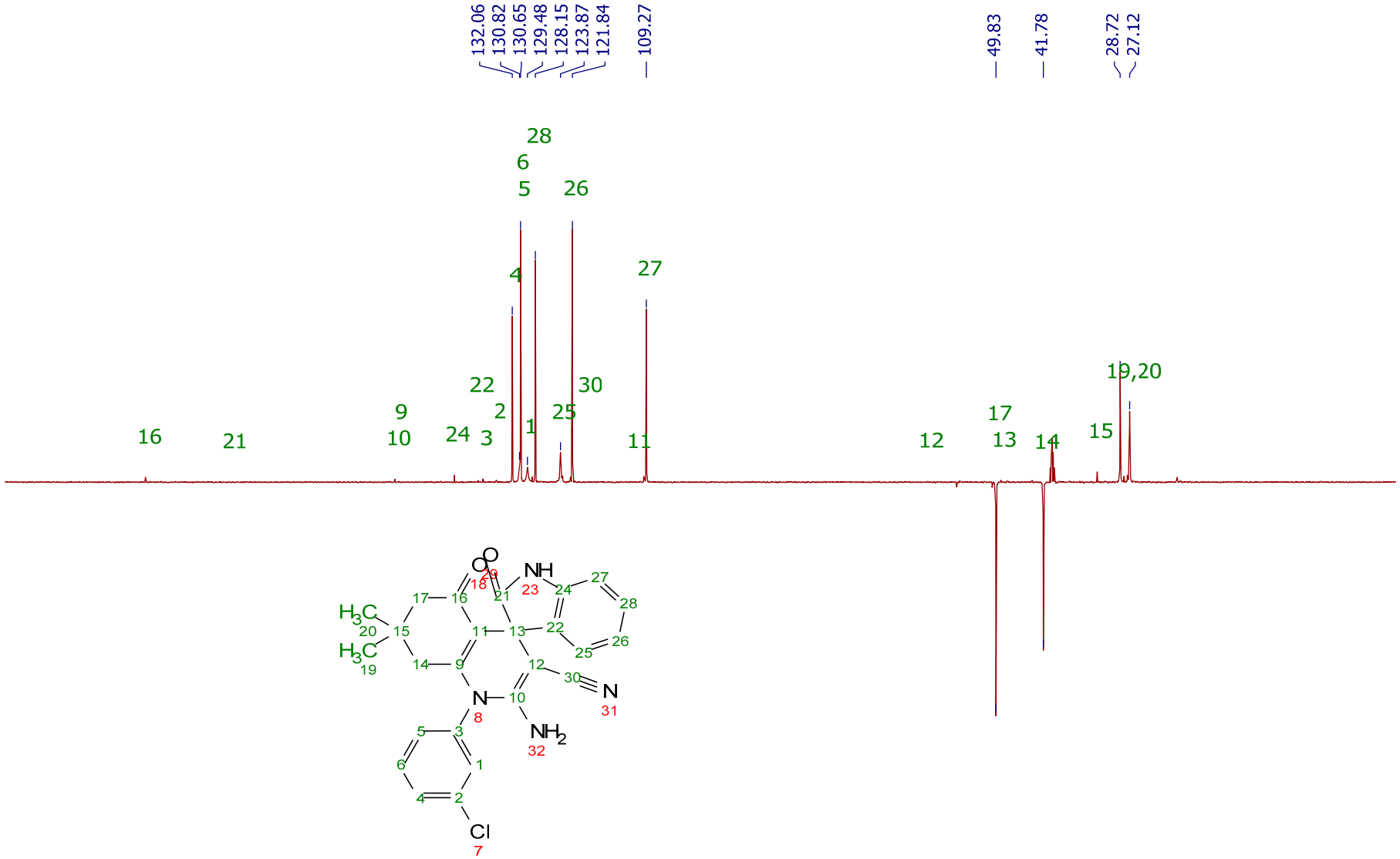


Figure S106 - DEPT spectrum of 7i

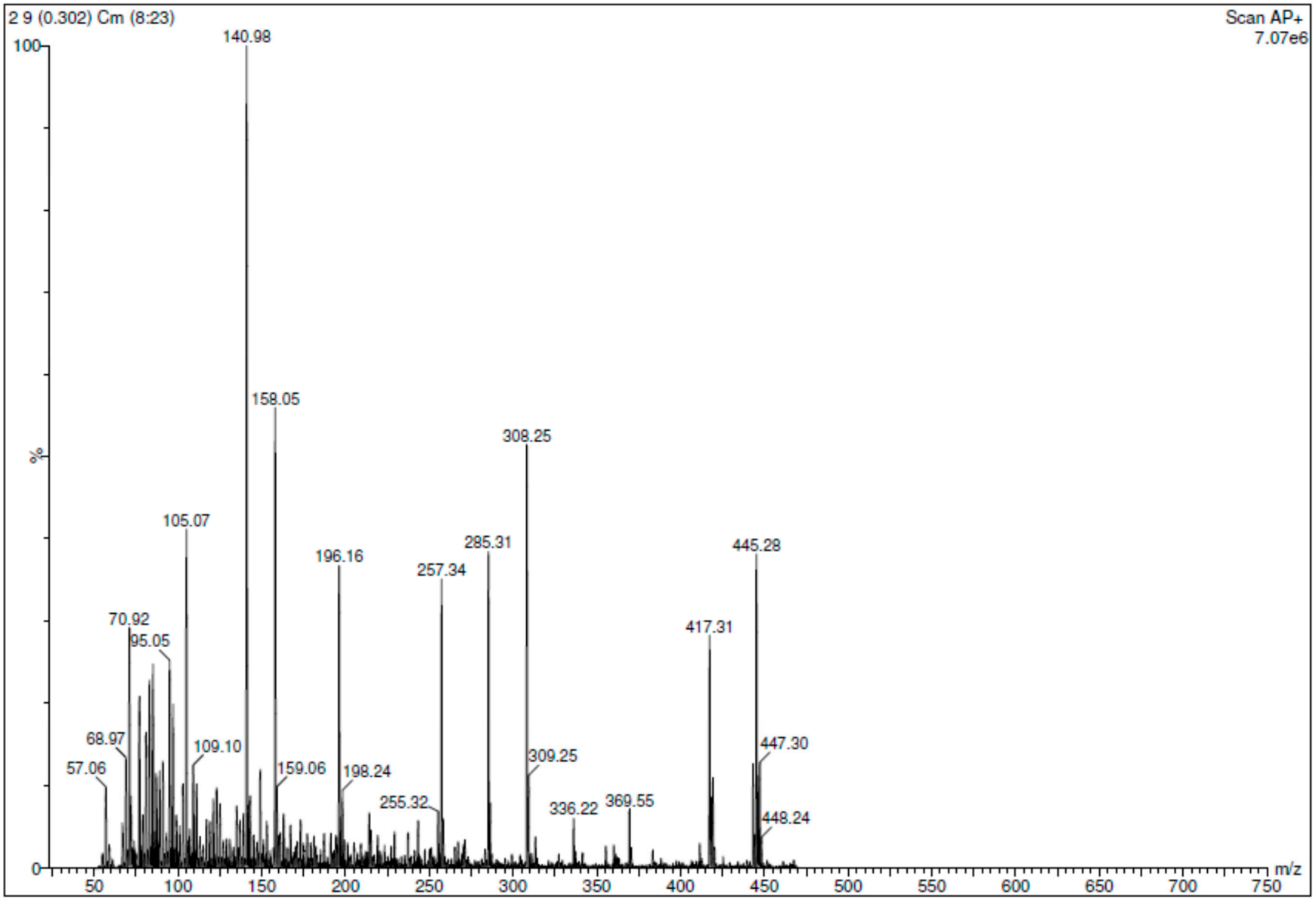
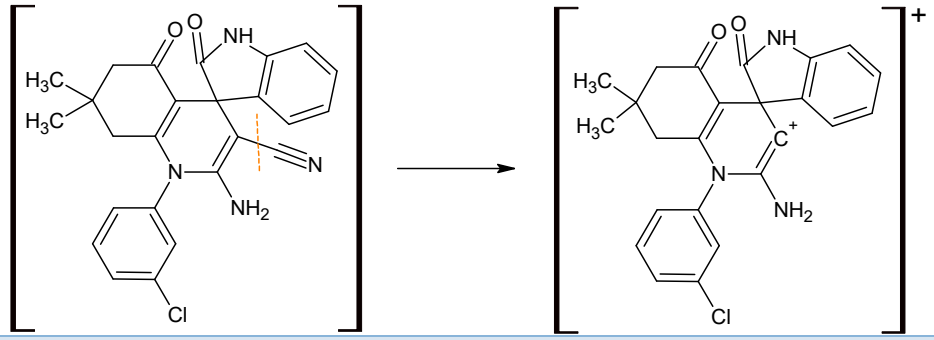
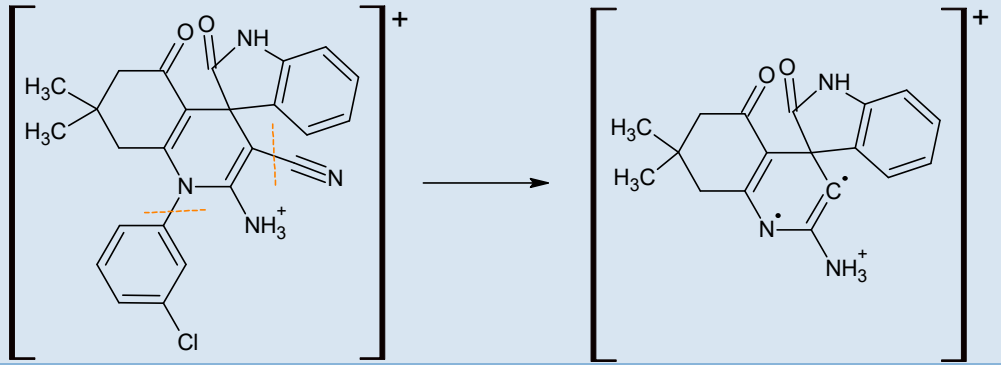
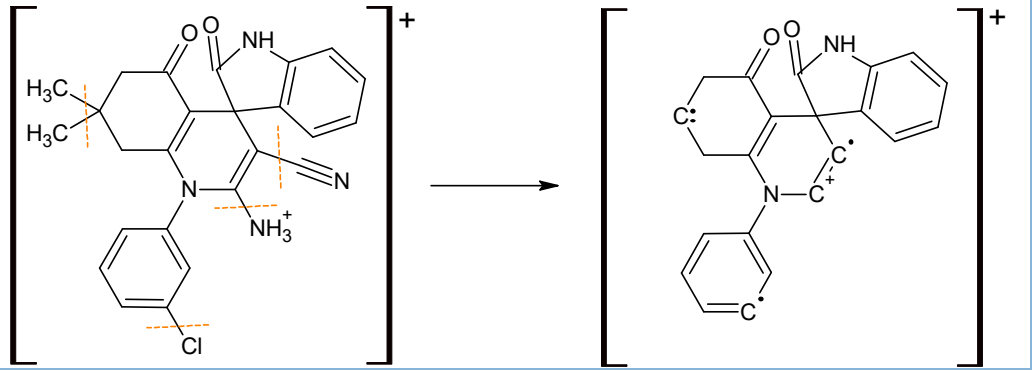


Figure S107 - MS spectrum of 7i

Table S13 - Fragmentation positions for peaks in MS spectrum of 7i

m/z	Fragmentation position
445.28 – 448.24 (isotopes)	$[M+H]^+$
417.31	
308.25	
336.22	

1.28. Product 12: 1-[4-[(4-bromophenyl)(1*H*-indol-3-yl)methyl]phenyl]-7,7-trimethyl-4-(3-nitrophenyl)-4,6,7,8-tetrahydroquinoline-2,5(1*H*,3*H*)-dione

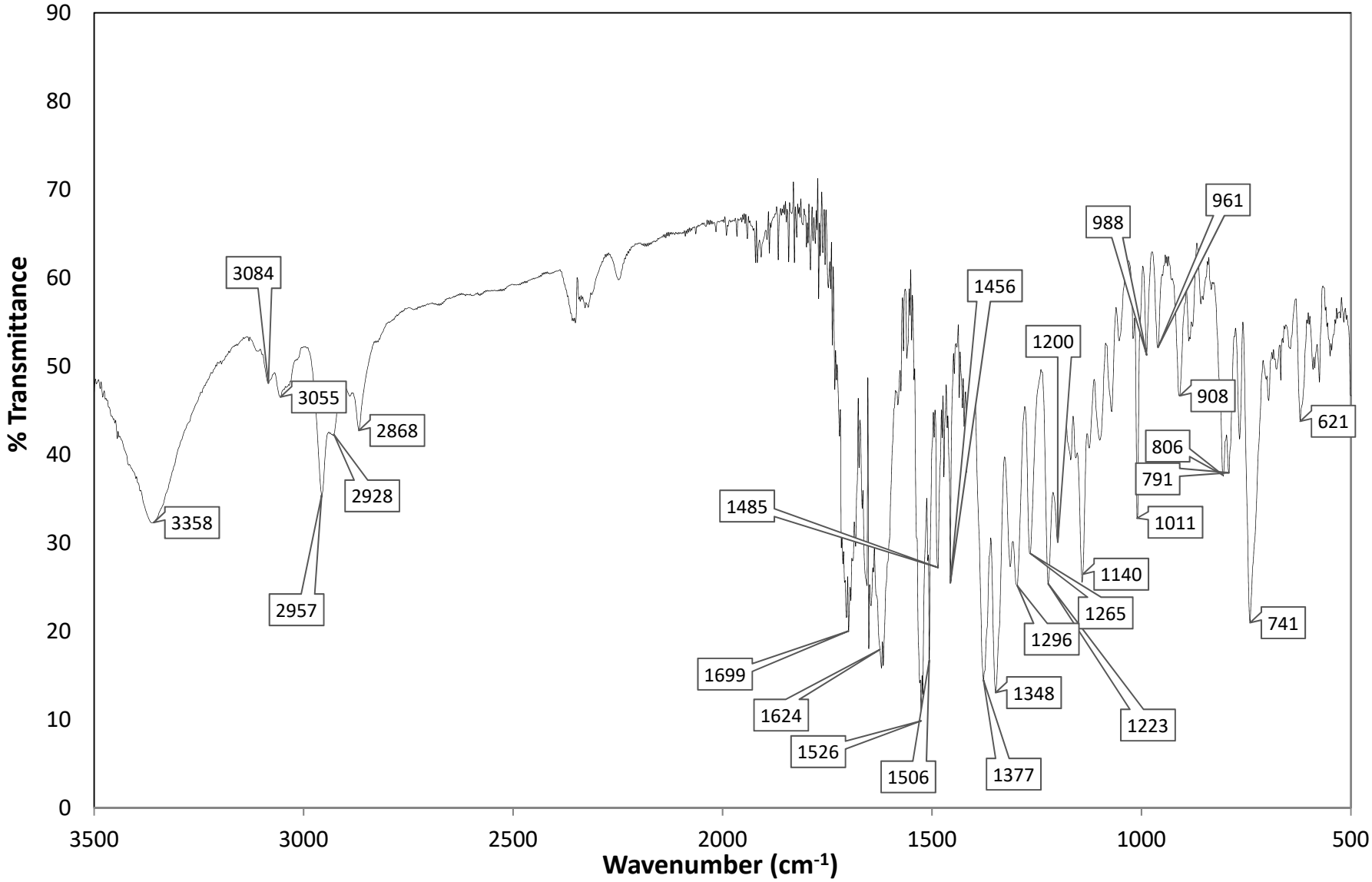


Figure S108 - IR spectrum of 12

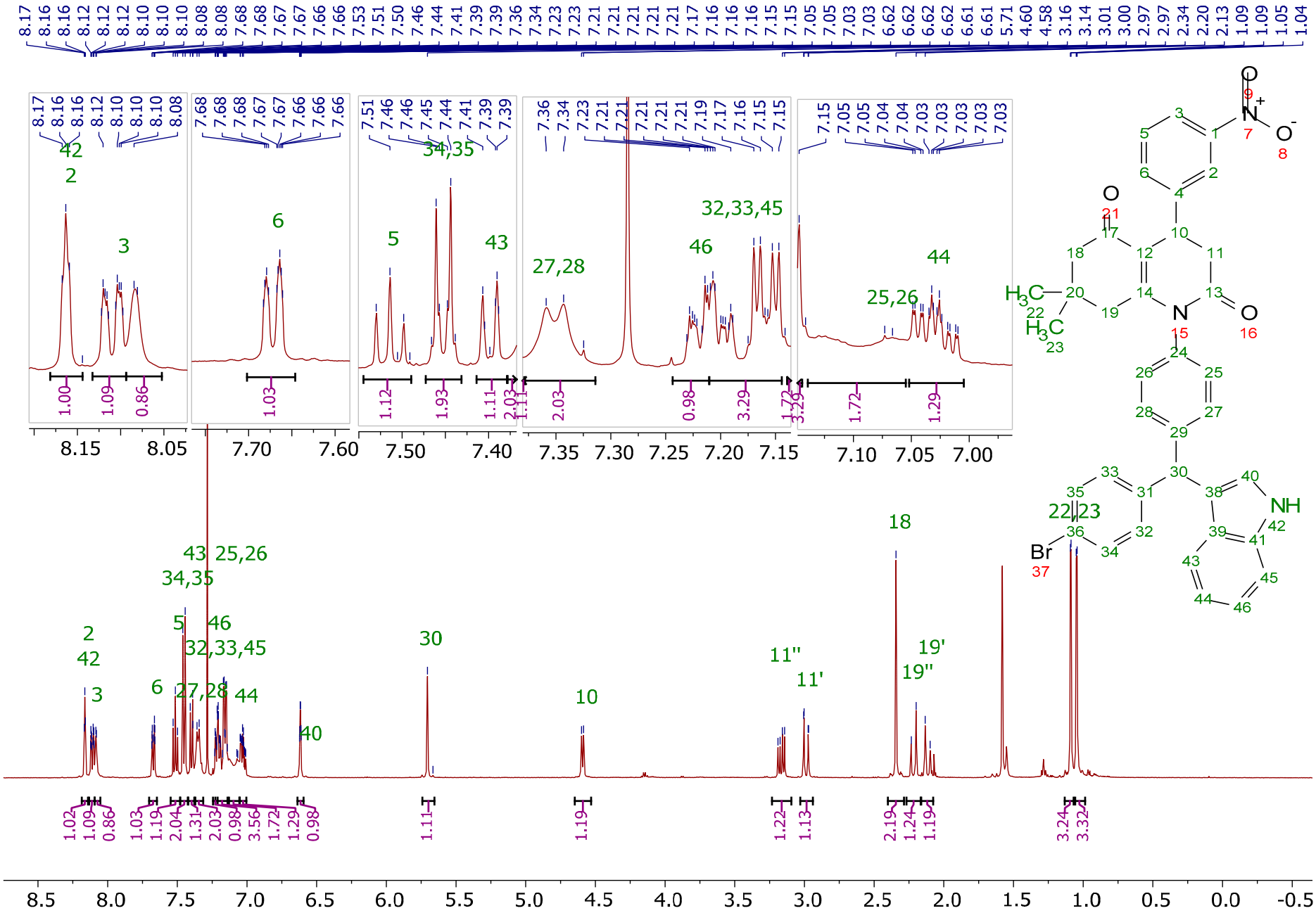


Figure S109 - ^{1}H NMR spectrum of 12

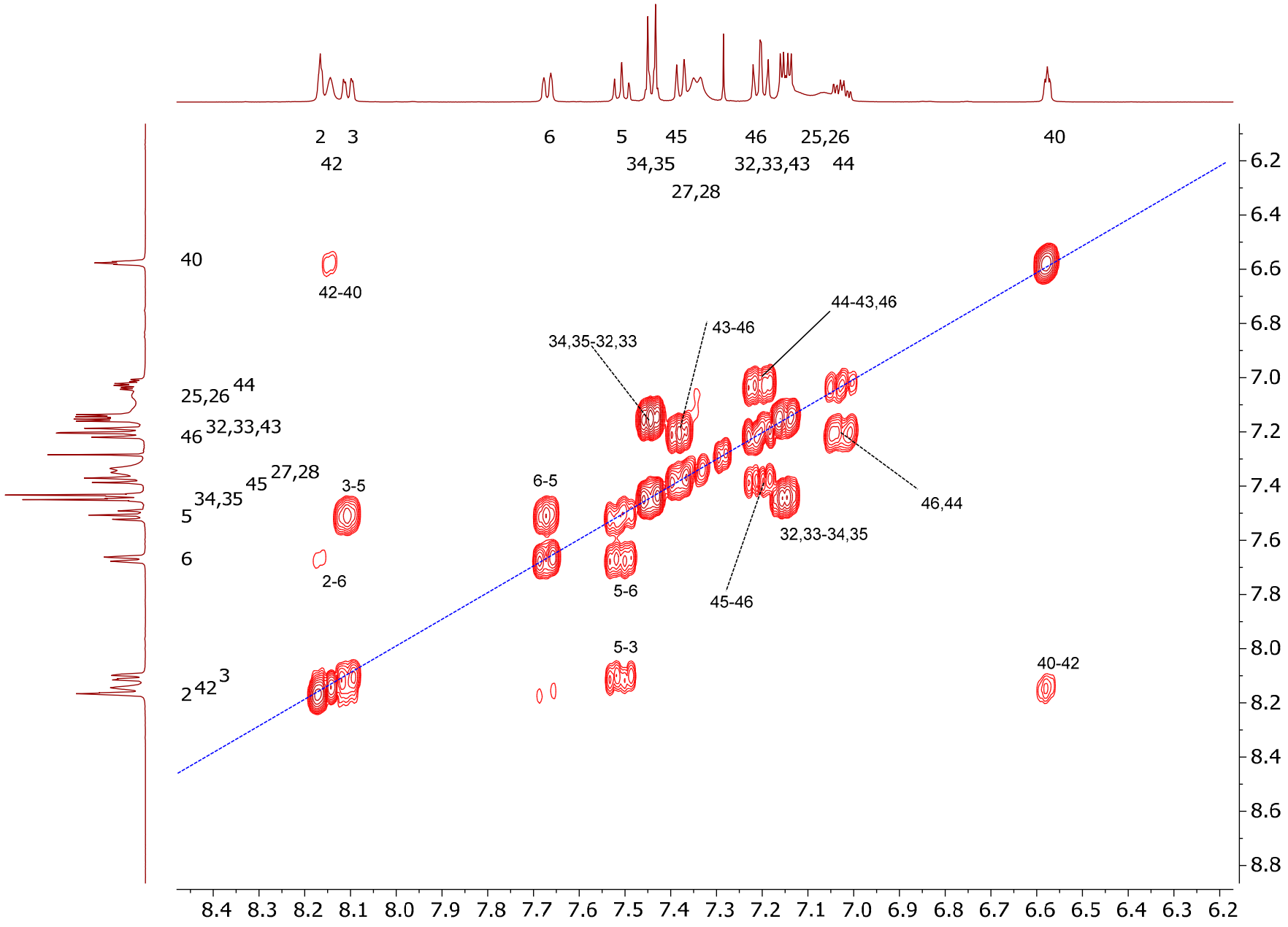


Figure S110 - ^{1}H-^{1}H COSY spectrum of 12

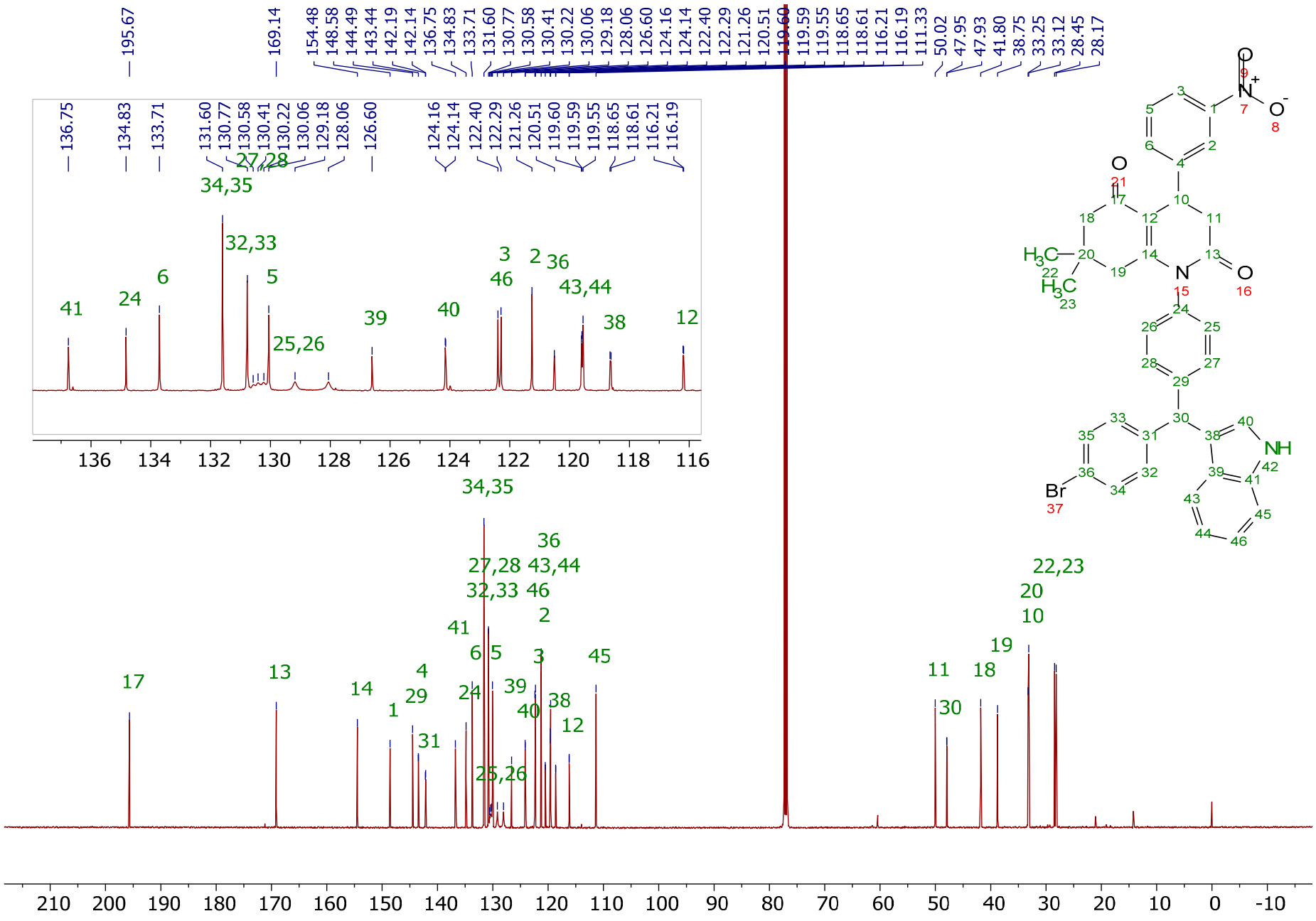


Figure S111 - ^{13}C NMR spectrum of 12

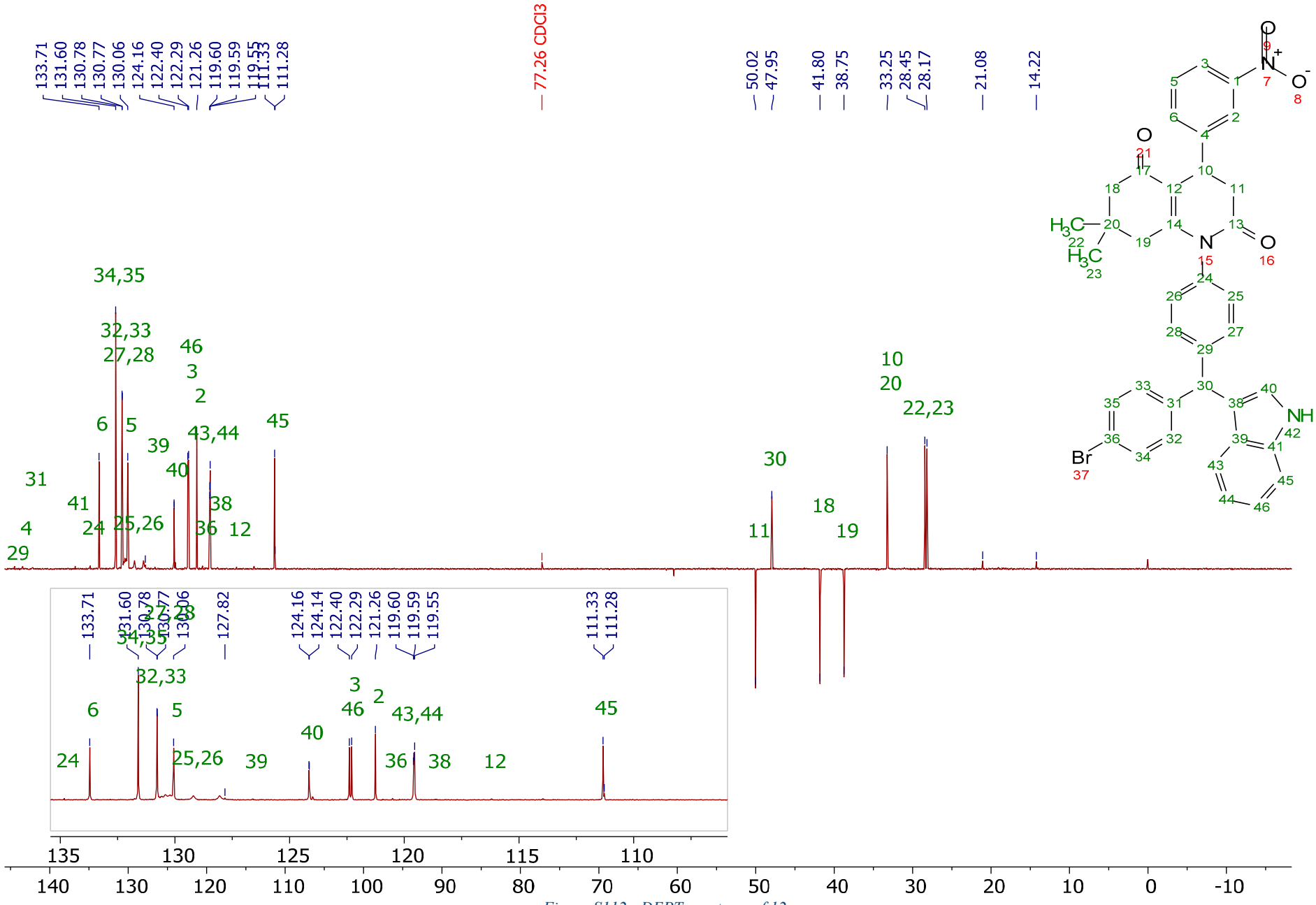


Figure S112 - DEPT spectrum of 12

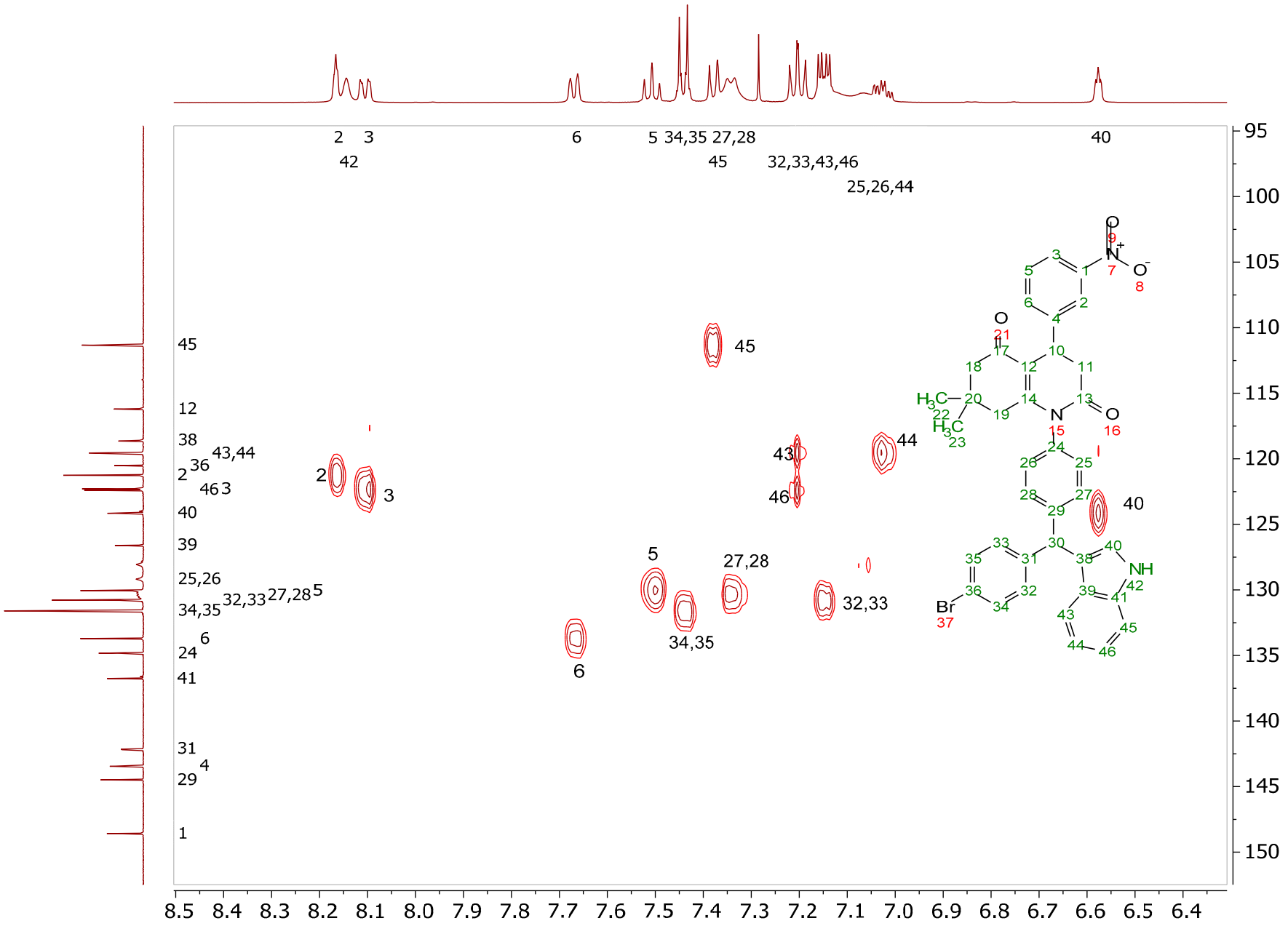


Figure S113 - HSQC spectrum of 12

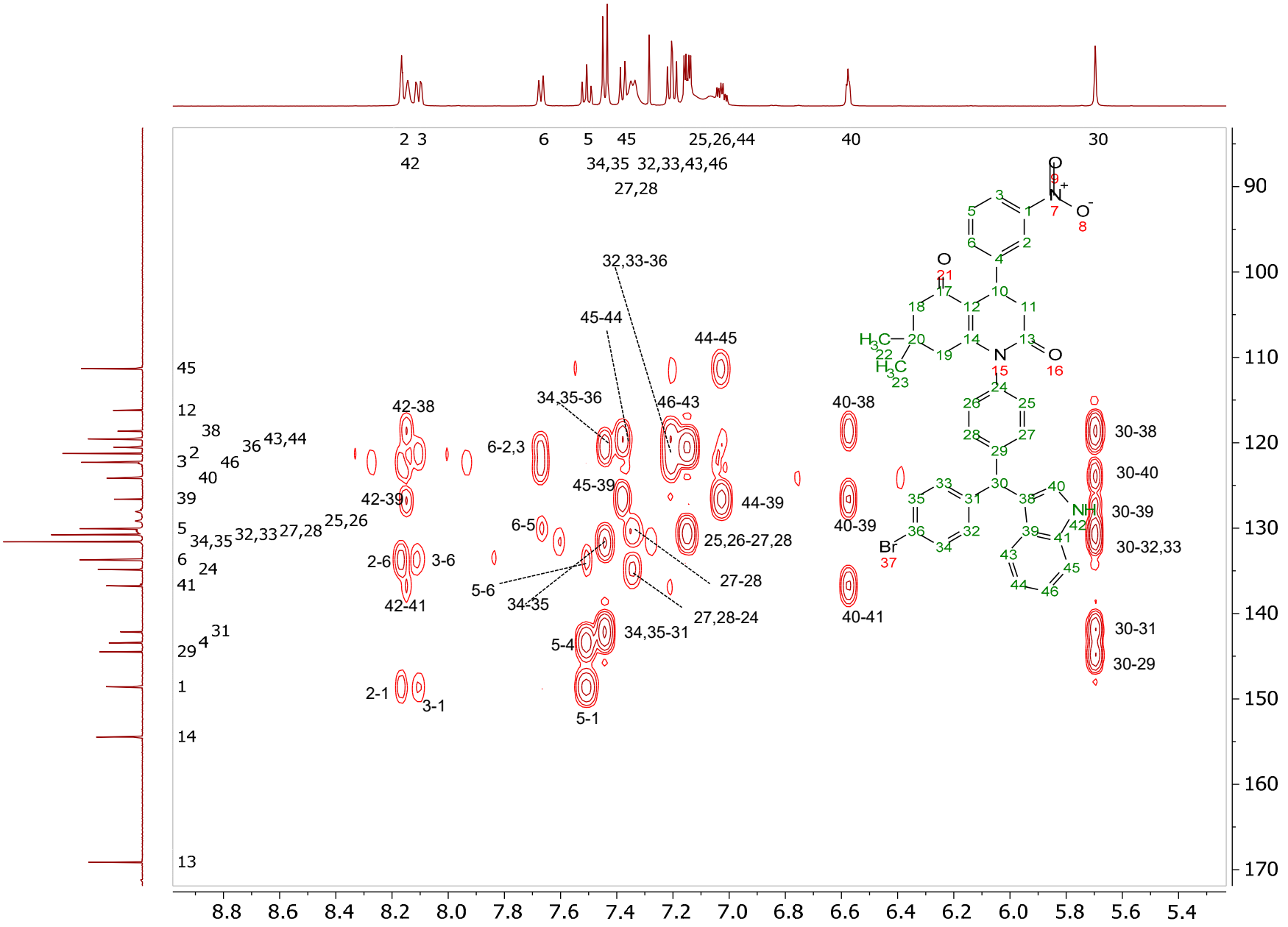


Figure S114 - Downfield region of HMBC spectrum of 12

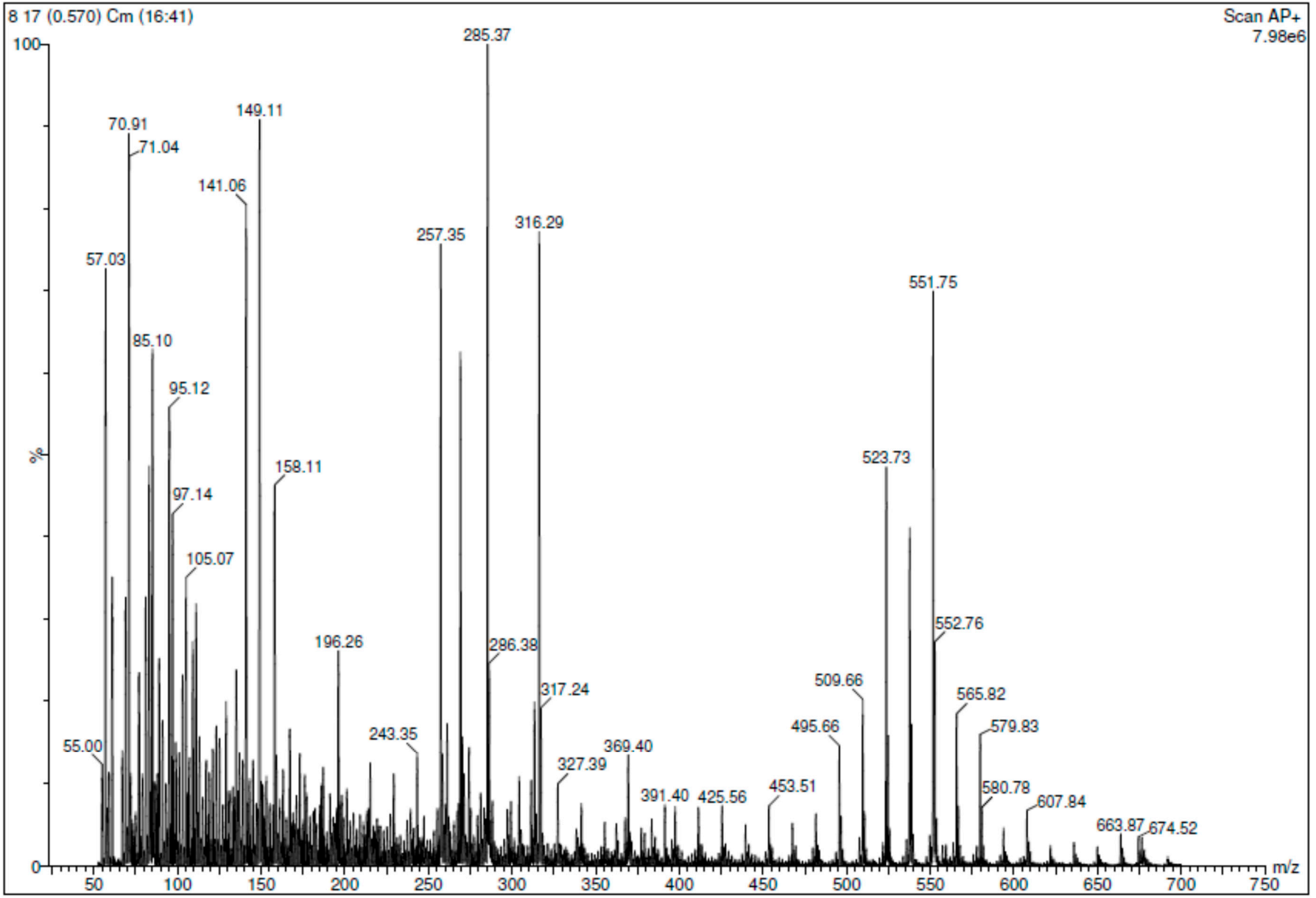
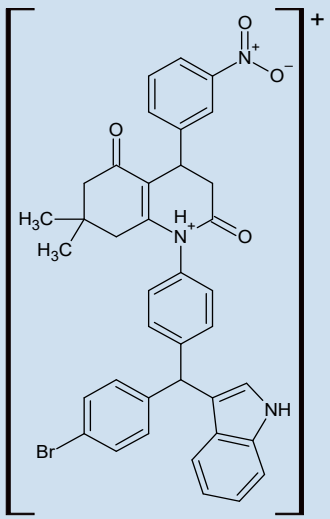
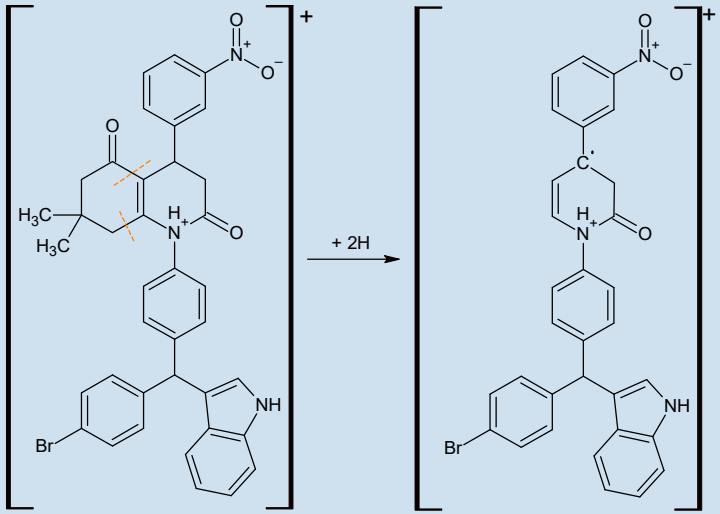
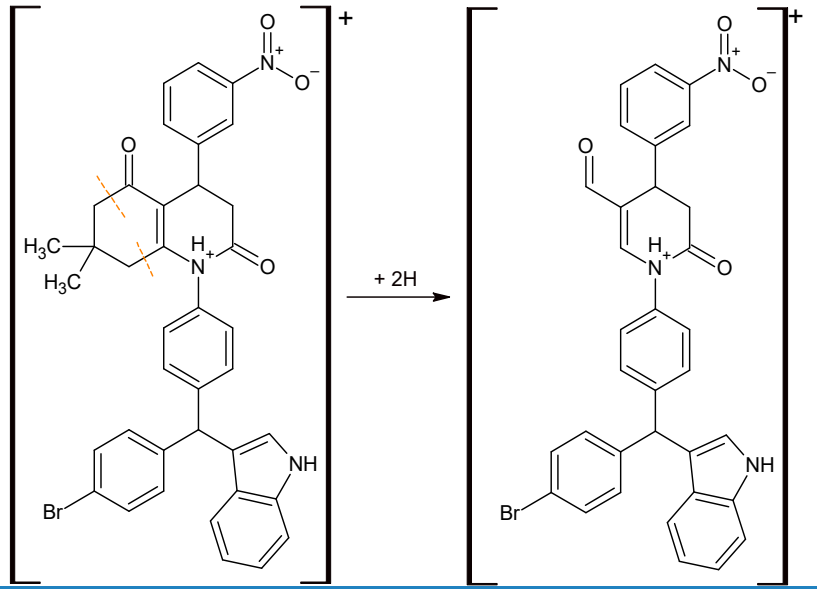
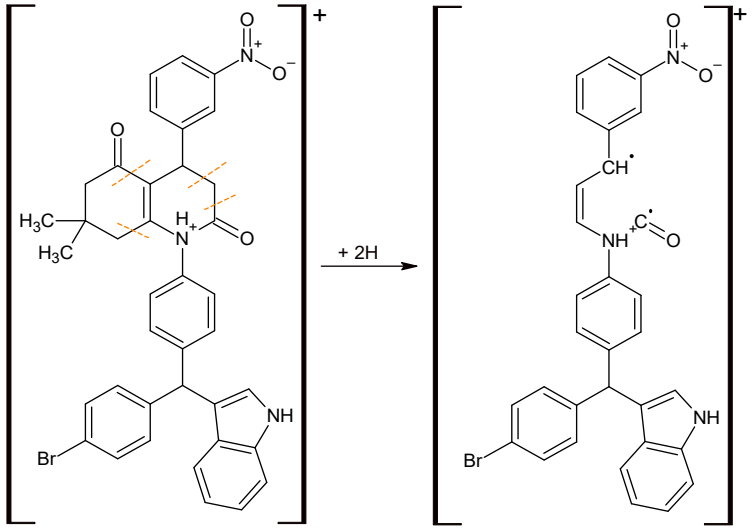
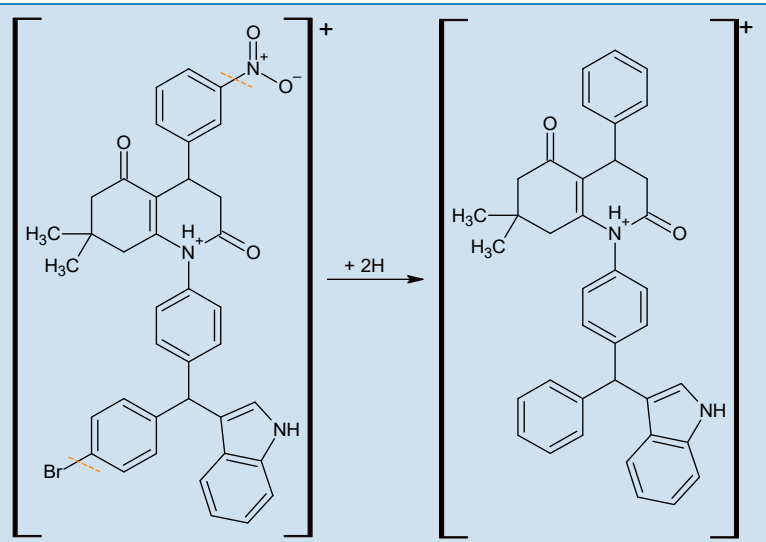
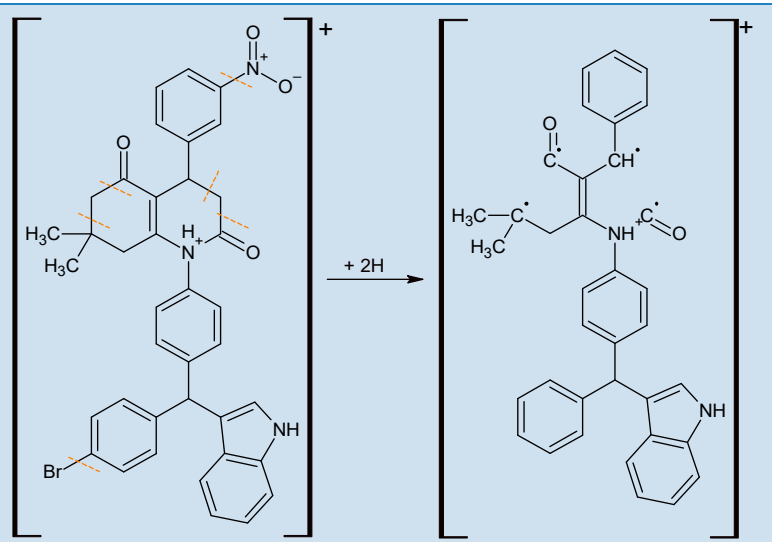
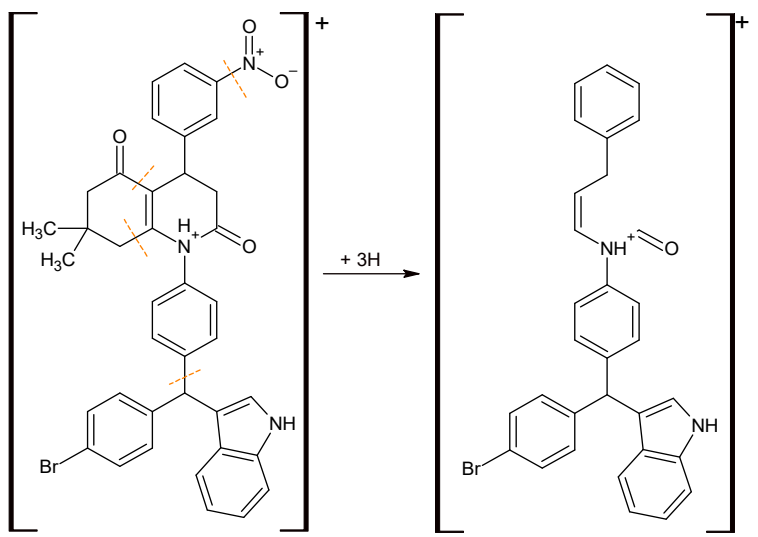
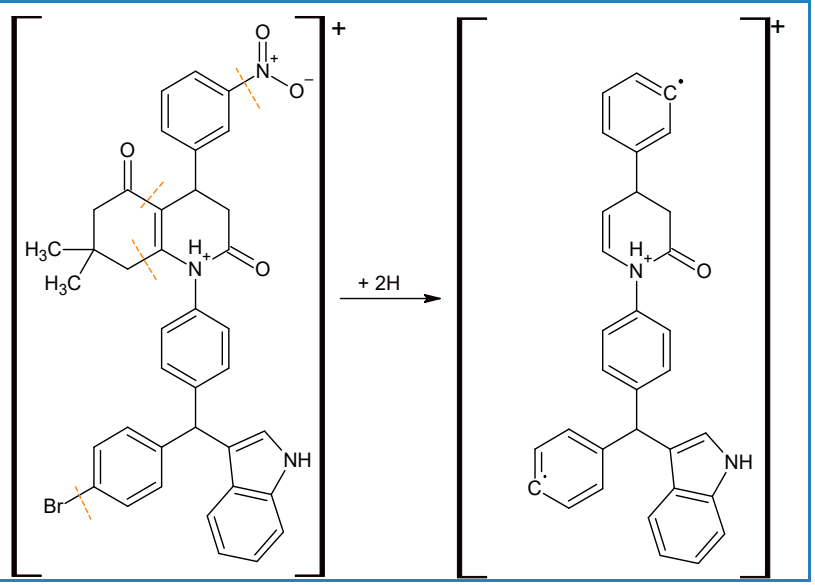
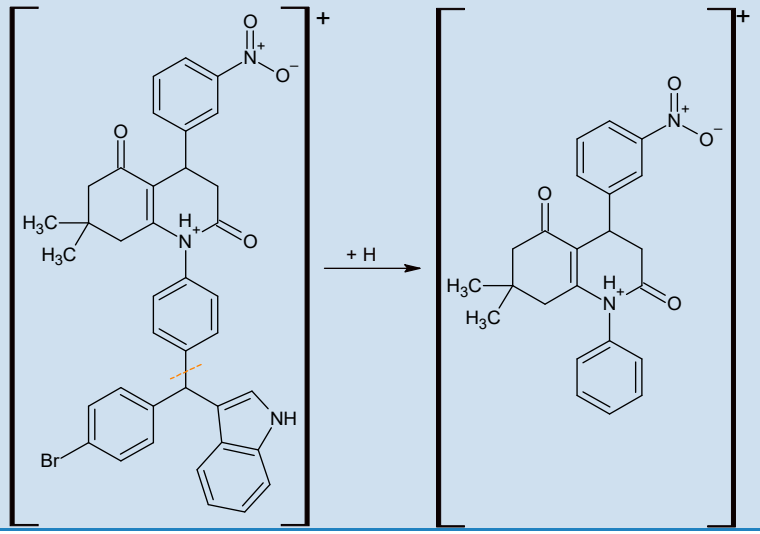


Figure S115 - MS spectrum of 12

Table S14 - Fragmentation positions for peaks in MS spectrum of 12

m/z	Fragmentation position
674.52	
579.83	
607.84	
565.82	

551.75		**523.73**	
534.46		**453.51**	

391.40				

1.29. Pip-Agar catalyst

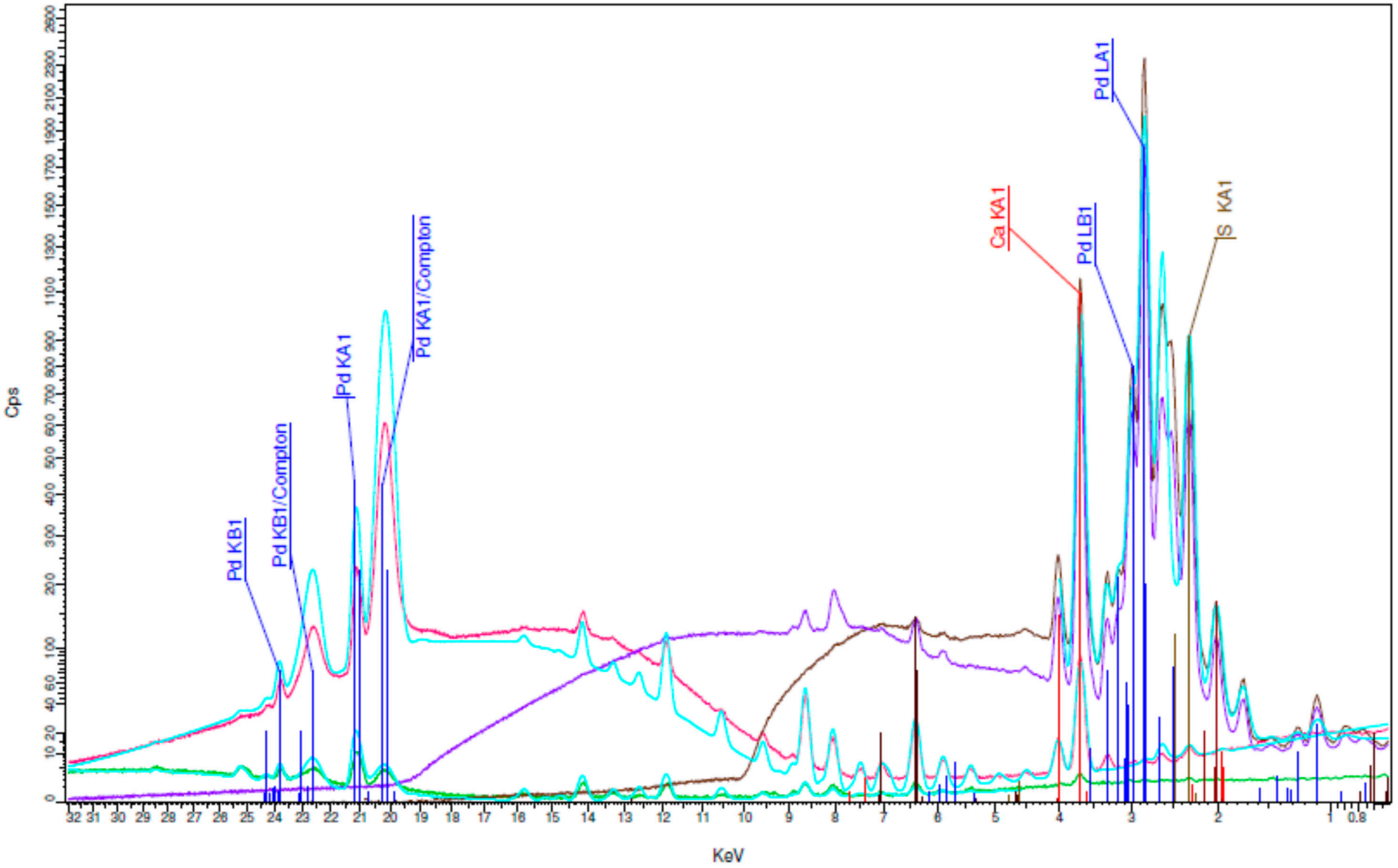


Figure S116 - XRF spectrum of Pip-Agar catalyst

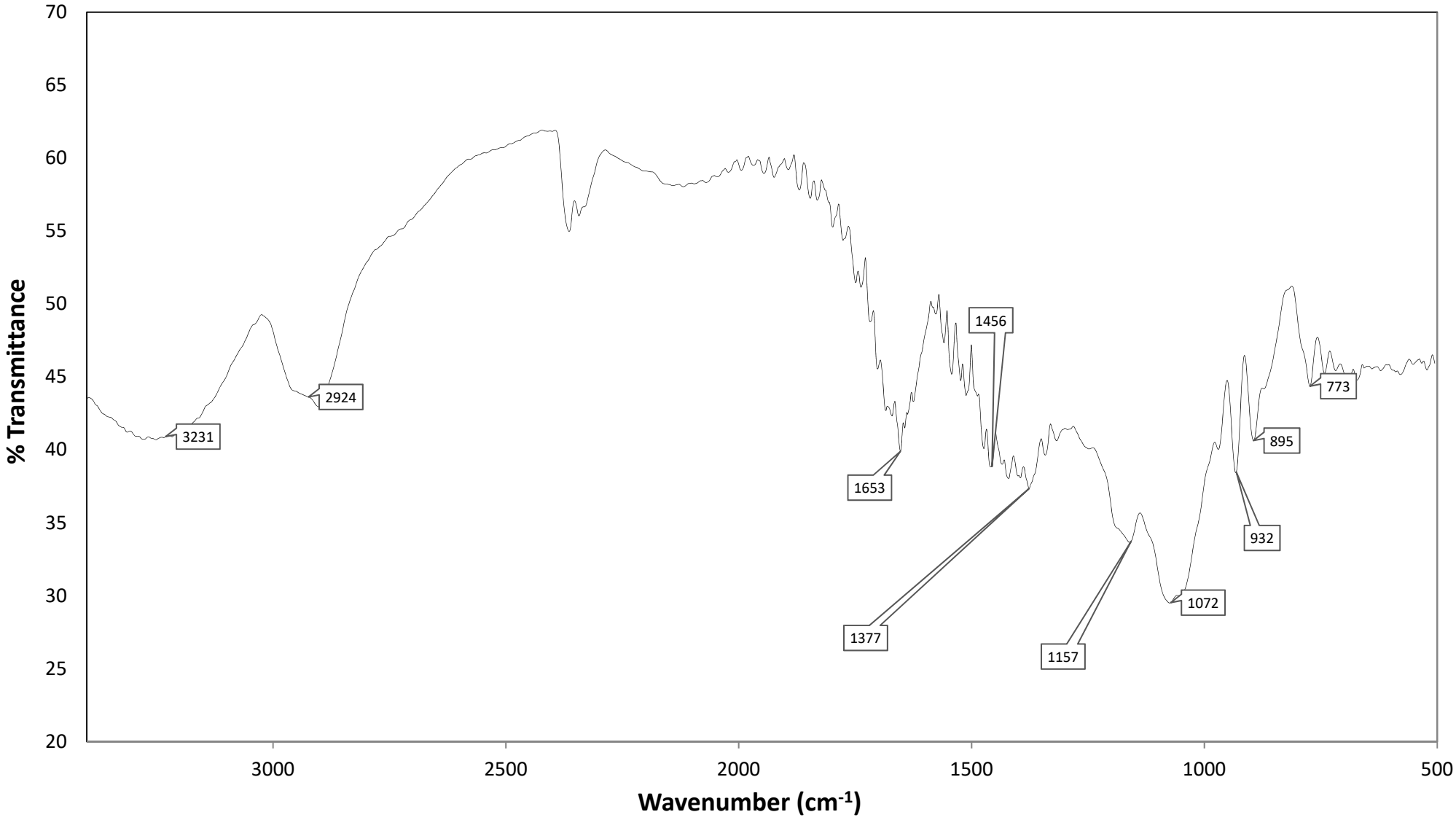


Figure S117 - IR spectrum of agar

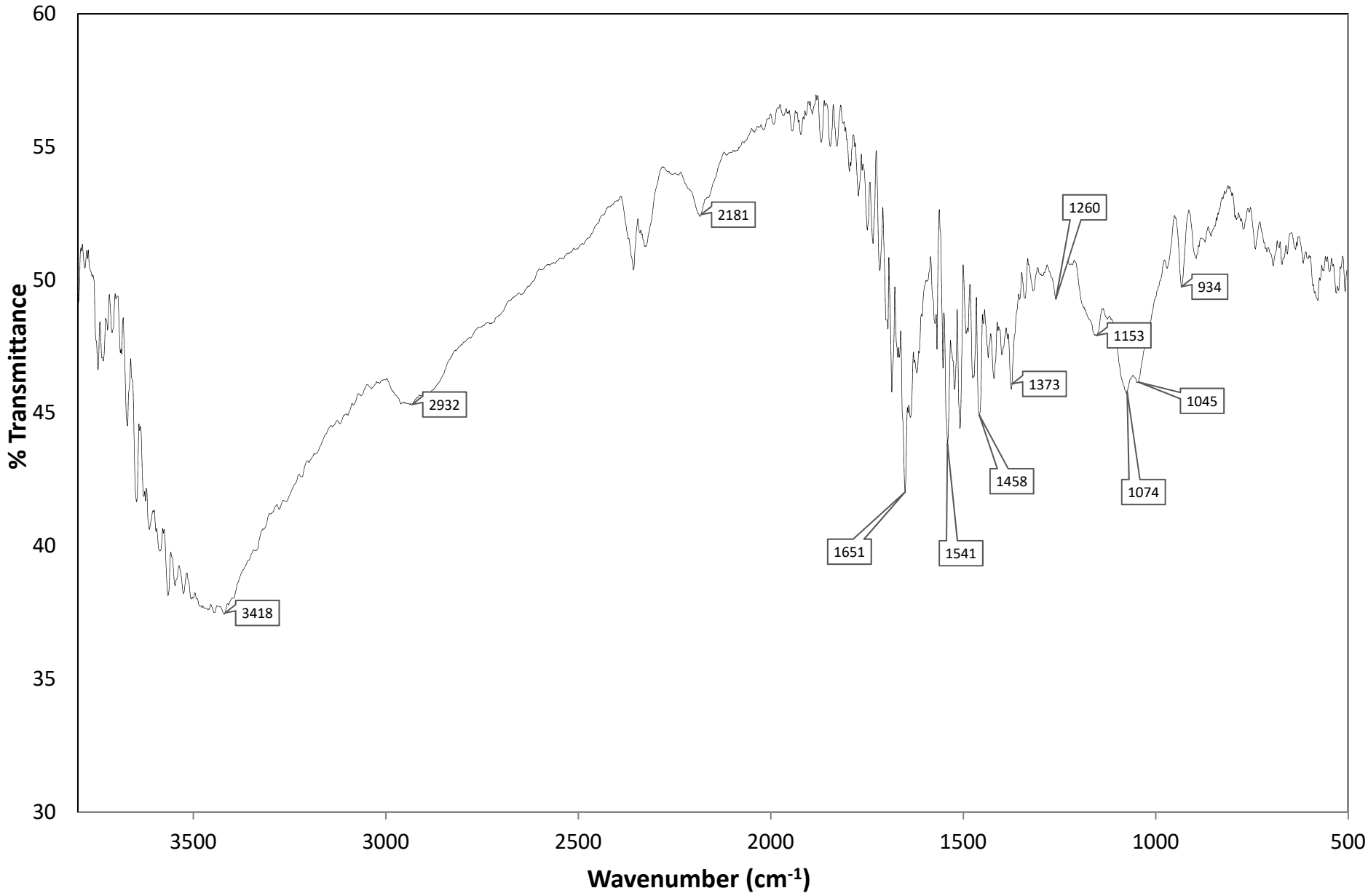


Figure S118 - IR spectrum of fresh Pip-Agar