Electronic Supplementary Information

Regioselective construction of 1,3-diazaheterocycle fused [1,2-*a*][1,8]naphthyridine derivatives *via* cascade reaction of quinolines with heterocyclic ketene aminals: a joint experimental–computational approach†

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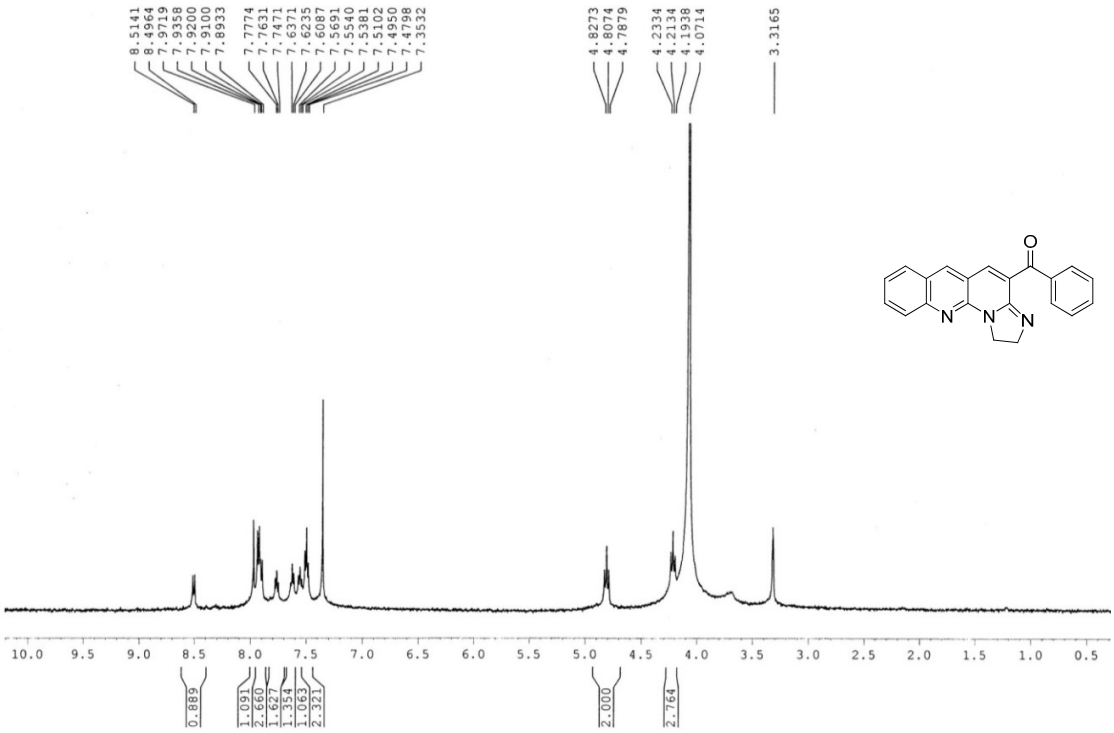


Fig. S1 ^{1}H NMR spectrum (500 MHz, CD_3OD+$CDCl_3$) of compound **3a**

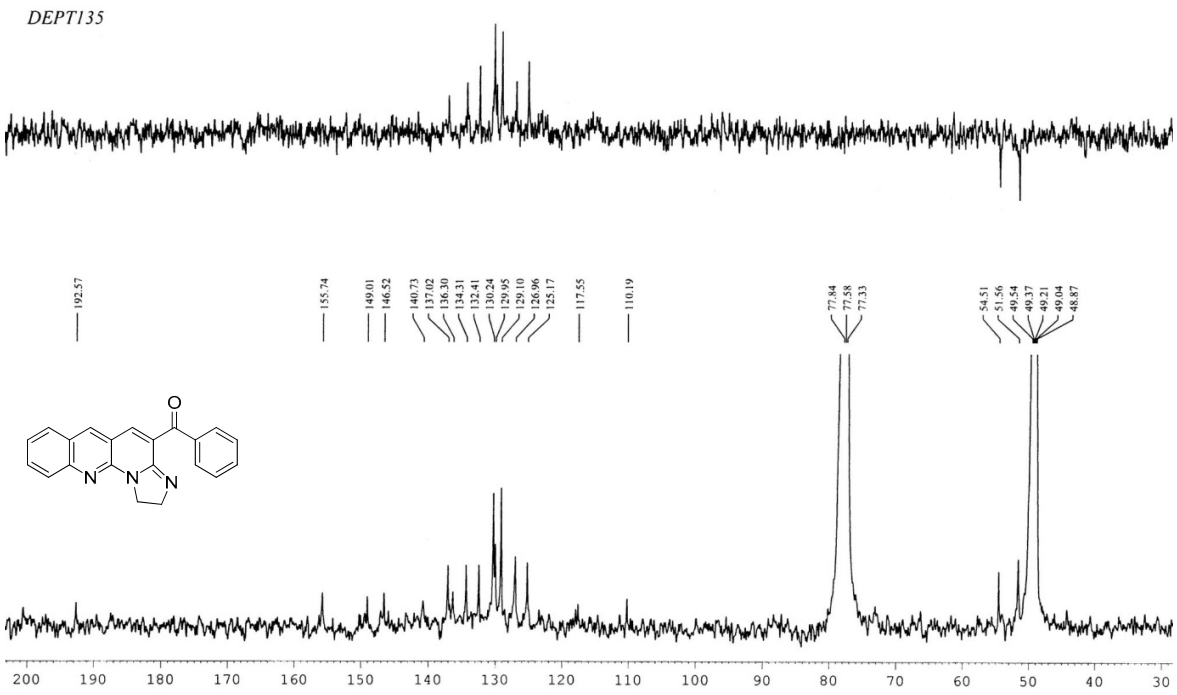


Fig. S2 ^{13}C NMR spectrum (125 MHz, CD_3OD+$CDCl_3$) of compound **3a**

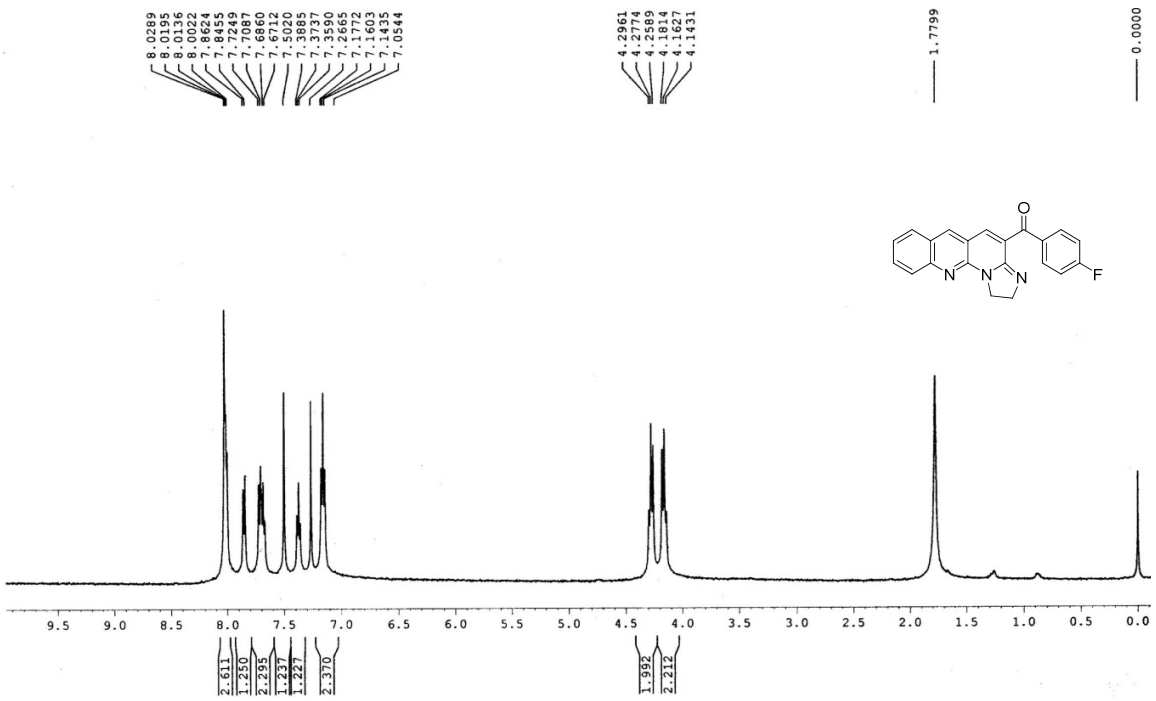


Fig. S3 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of compound **3b**

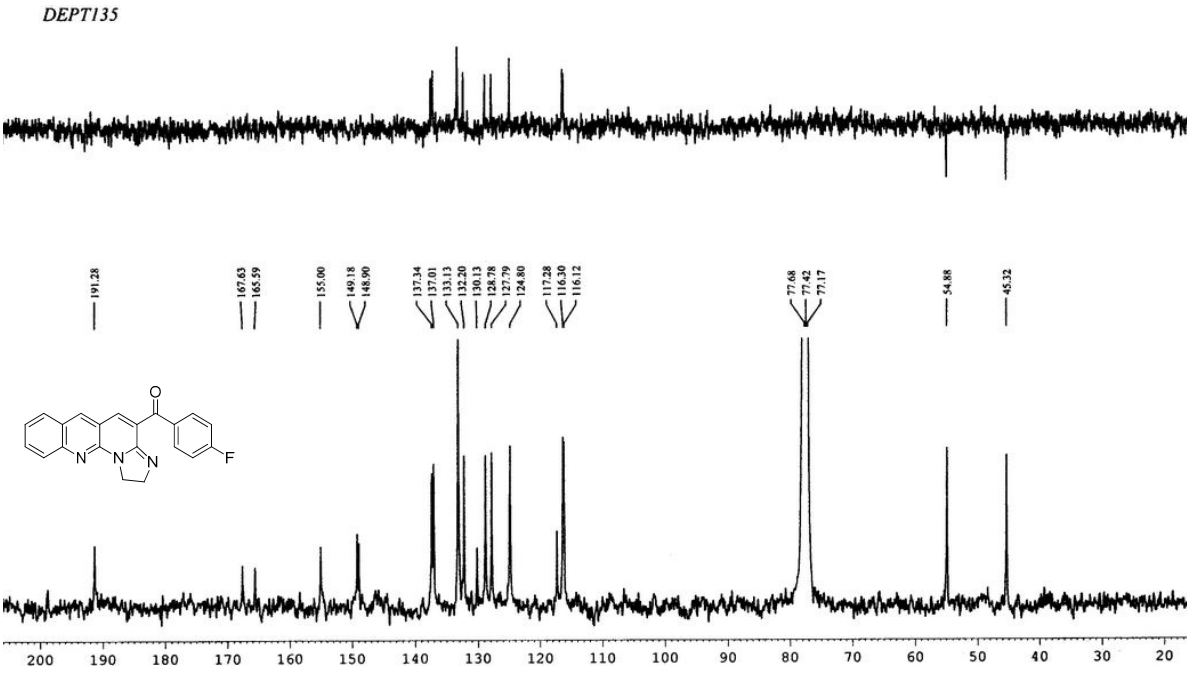


Fig. S4 ^{13}C NMR spectrum (125 MHz, $CDCl_3$) of compound **3b**

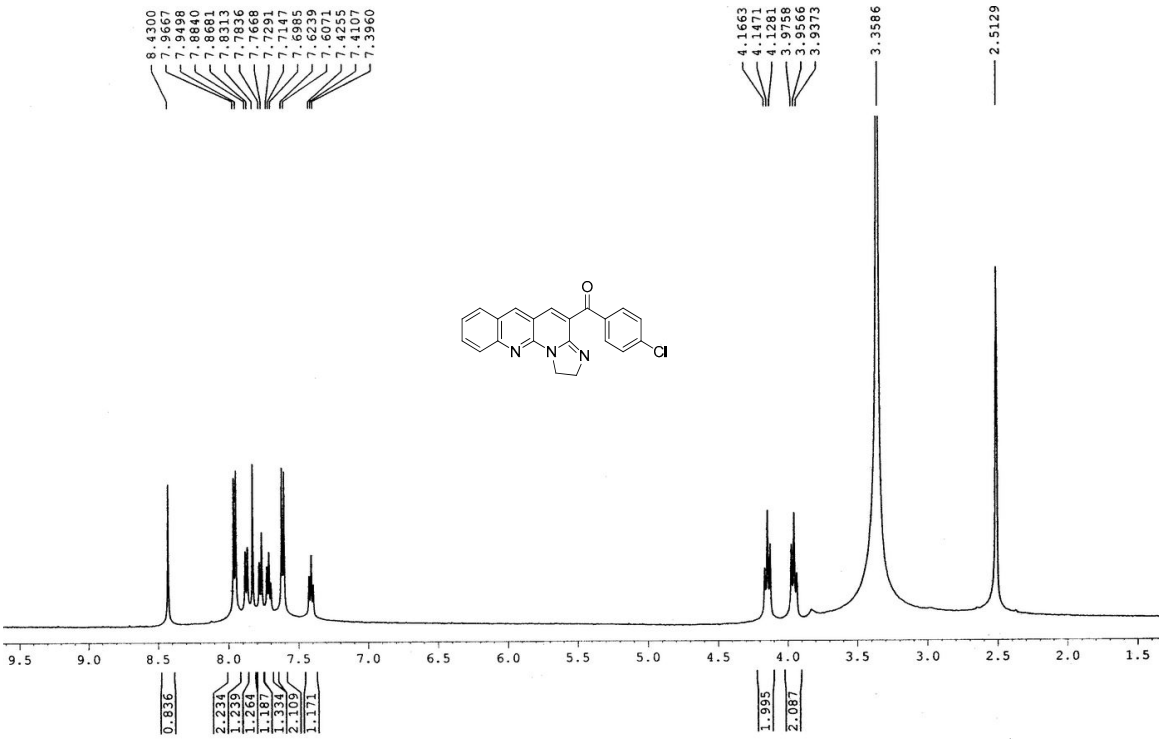


Fig. S5 ^{1}H NMR spectrum (500 MHz, DMSO-d_6) of compound **3c**

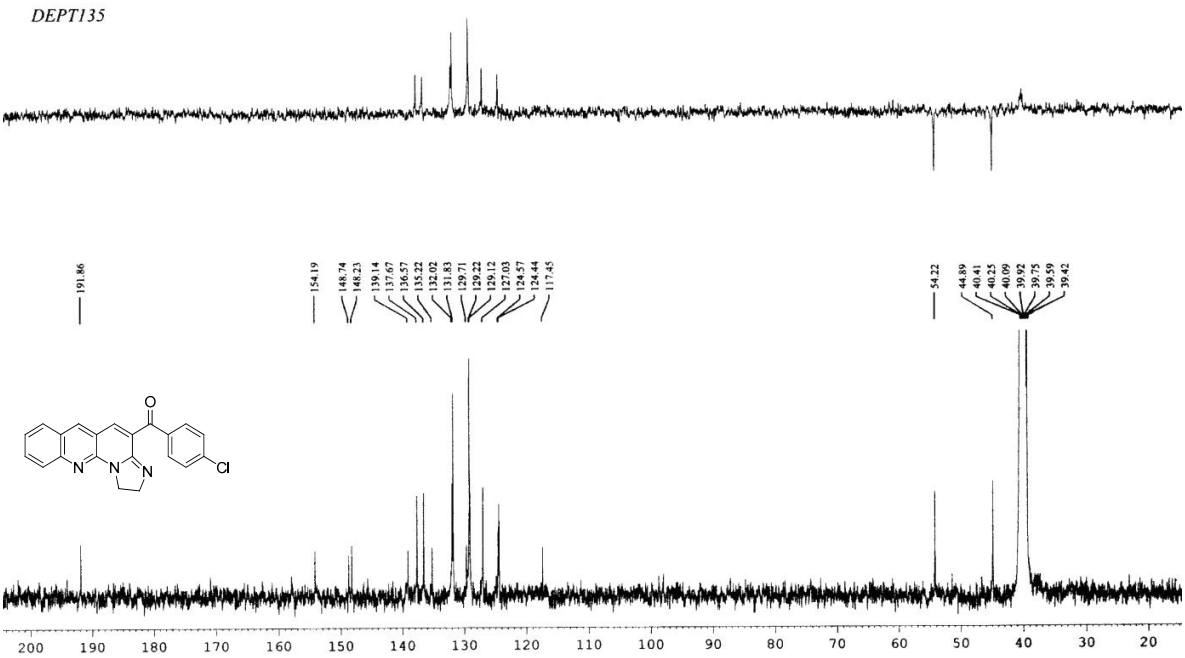


Fig. S6 ^{13}C NMR spectrum (125 MHz, DMSO-d_6) of compound **3c**

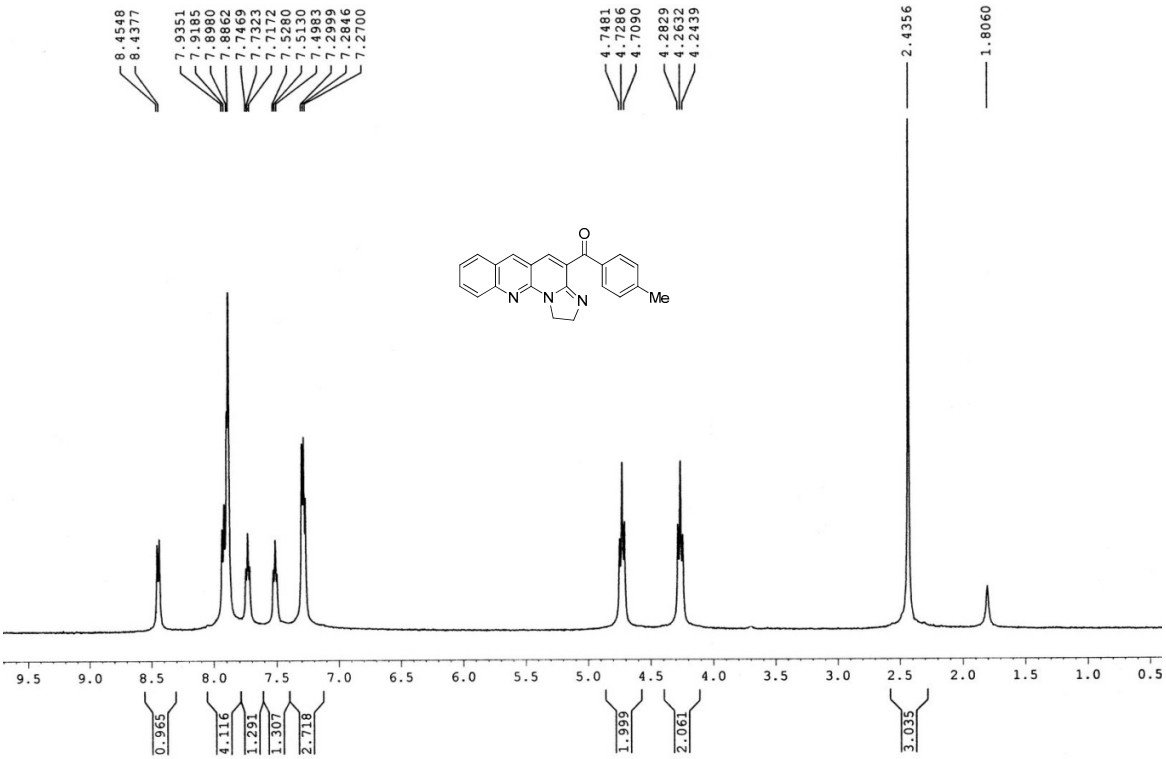


Fig. S7 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of compound **3d**

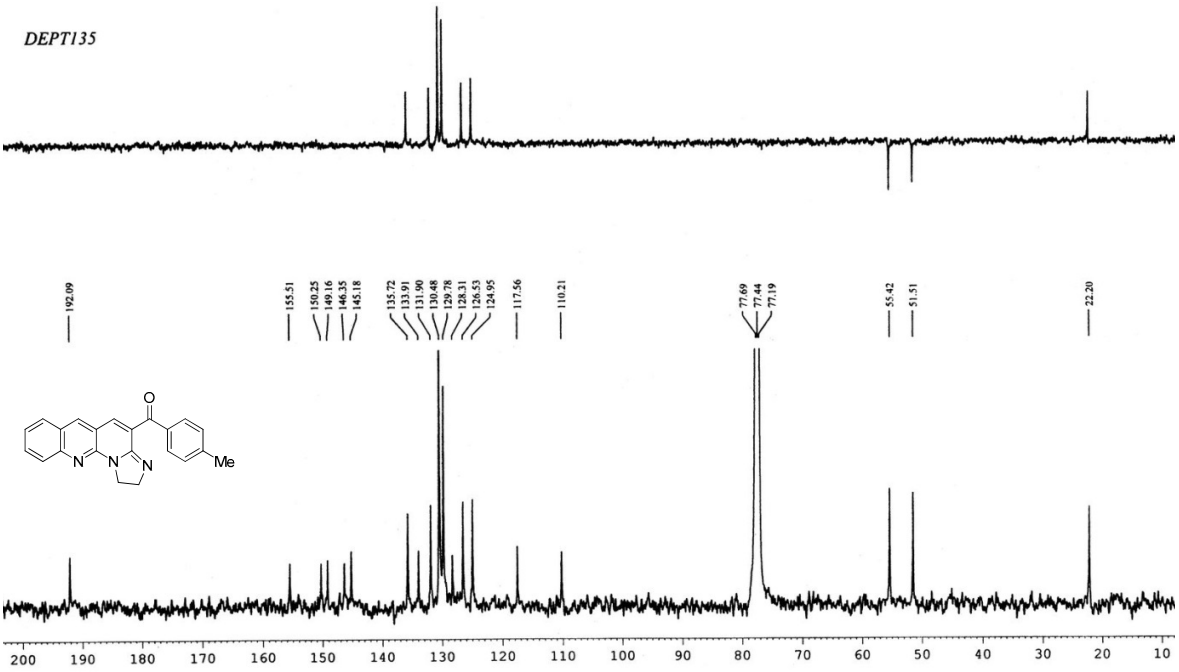


Fig. S8 ^{13}C NMR spectrum (125 MHz, $CDCl_3$) of compound **3d**

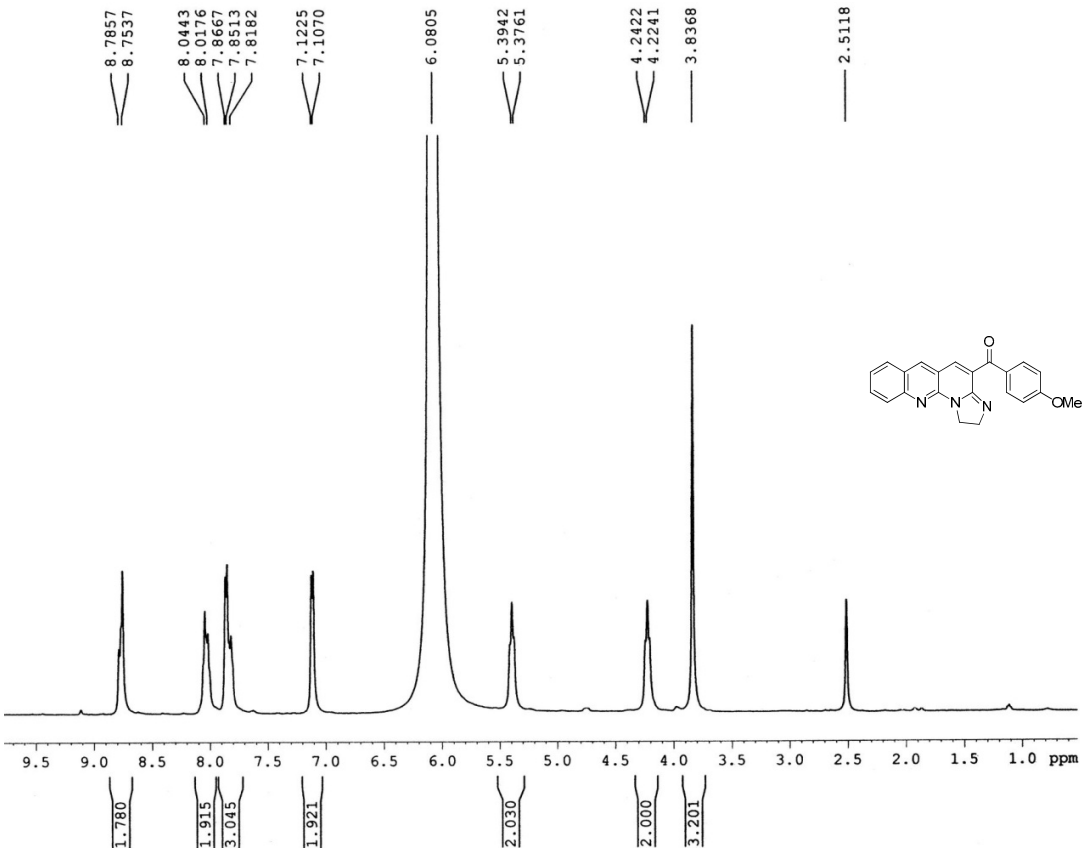


Fig. S9 ^{1}H NMR spectrum (500 MHz, DMSO-d_6+$HClO_4$) of compound **3e**

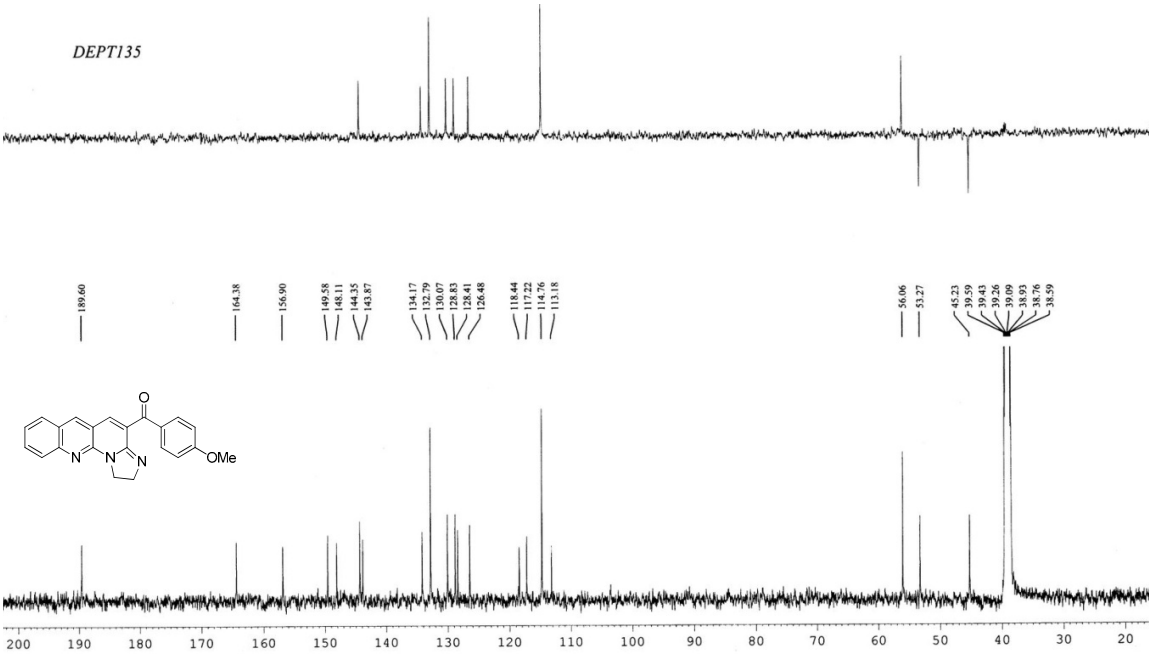


Fig. S10 ^{13}C NMR spectrum (125 MHz, DMSO-d_6+$HClO_4$) of compound **3e**

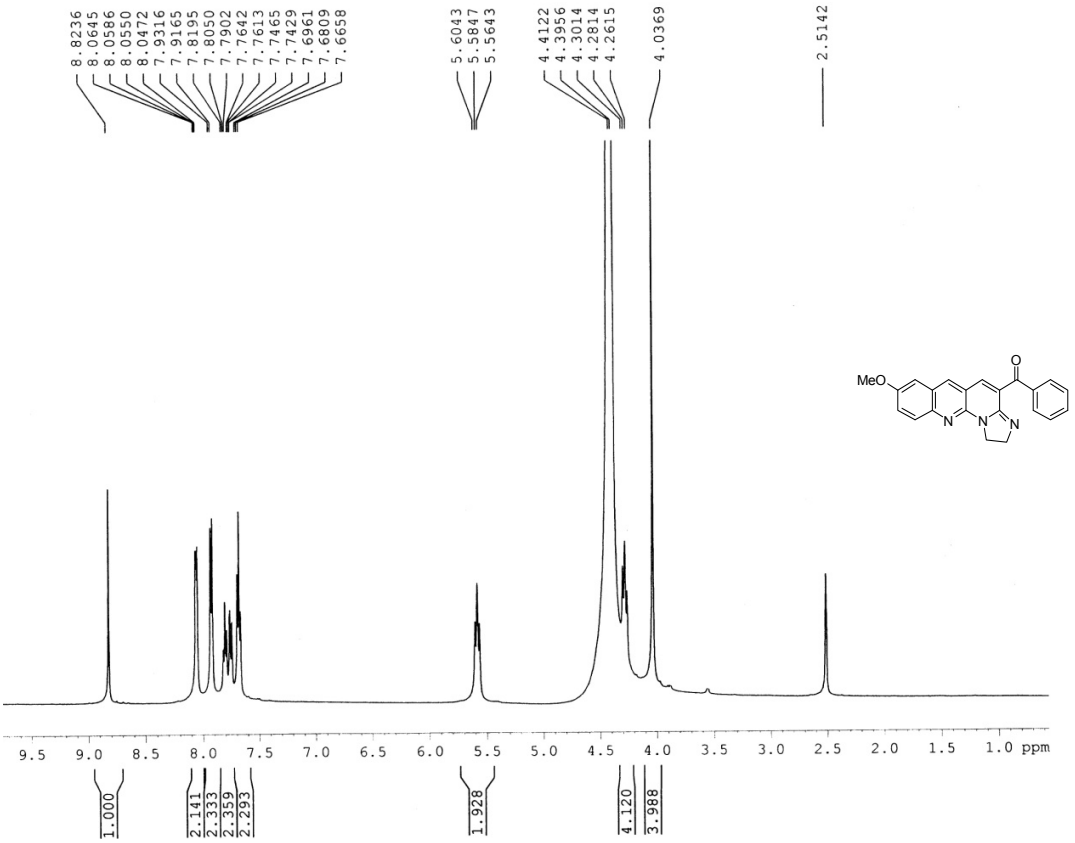


Fig. S11 ^{1}H NMR spectrum (500 MHz, DMSO-d_6+$HClO_4$) of compound **3f**

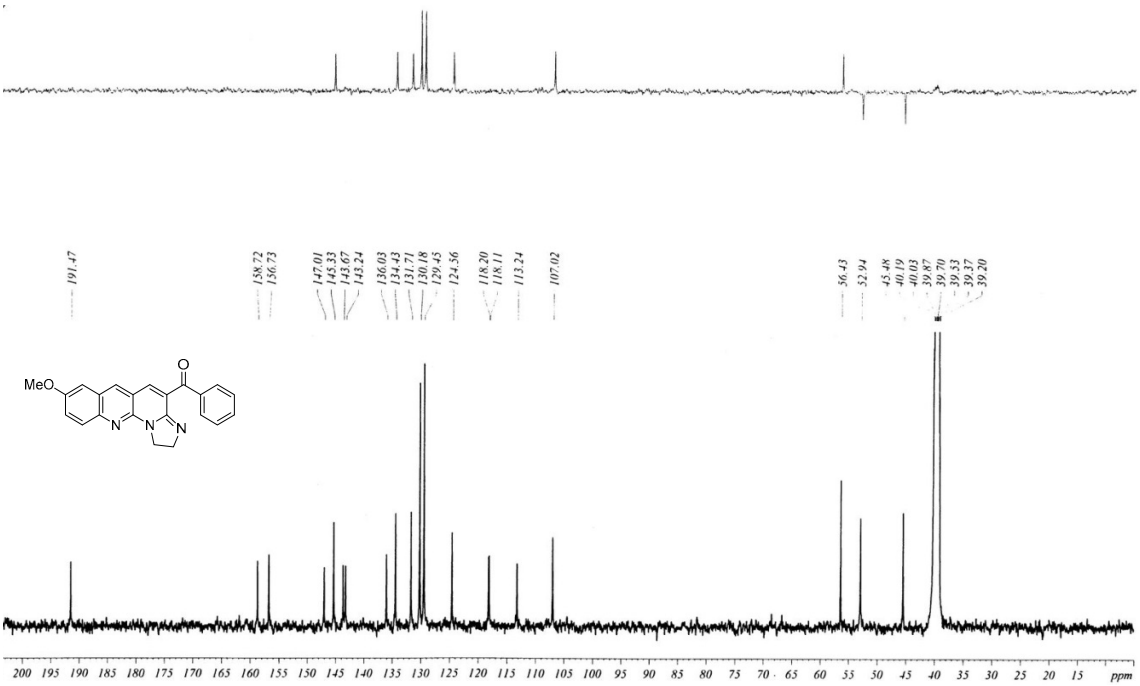


Fig. S12 ^{13}C NMR spectrum (125 MHz, DMSO-d_6+$HClO_4$) of compound **3f**

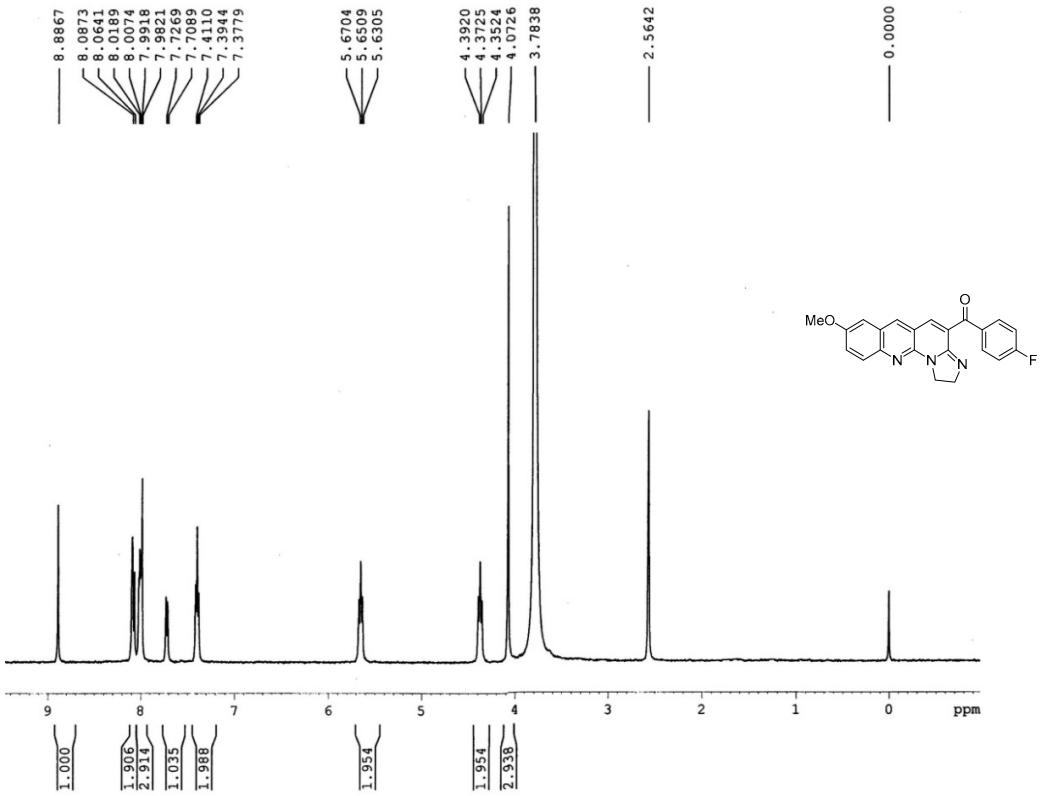


Fig. S13 ^{1}H NMR spectrum (500 MHz, $CDCl_3$+DMSO-d_6+$HClO_4$) of compound **3g**

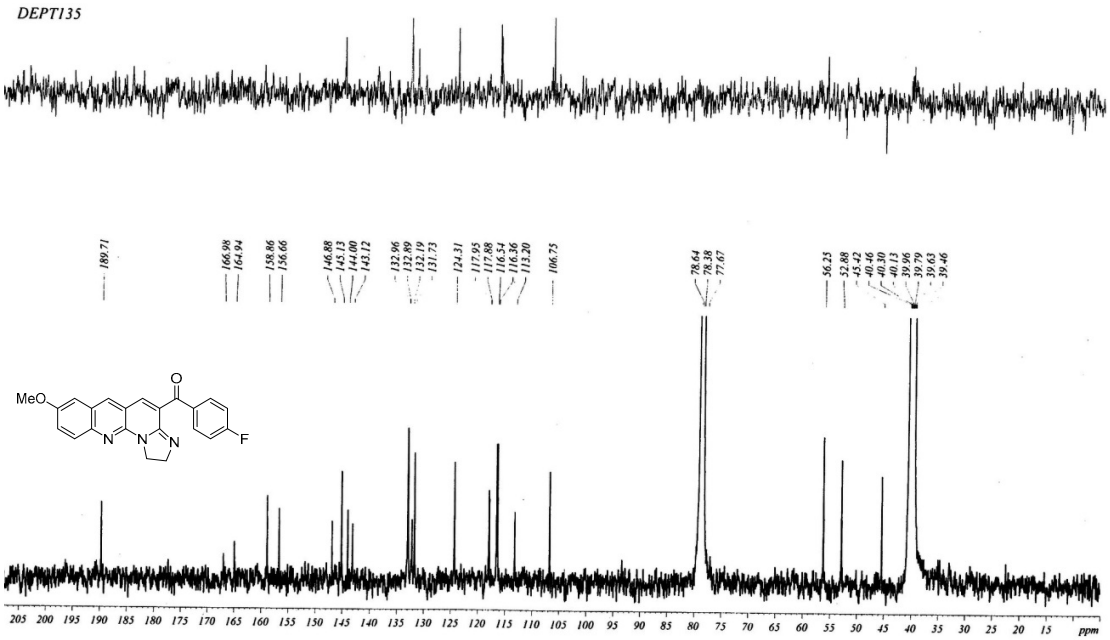


Fig. S14 ^{13}C NMR spectrum (125 MHz, $CDCl_3$+DMSO-d_6+$HClO_4$) of compound **3g**

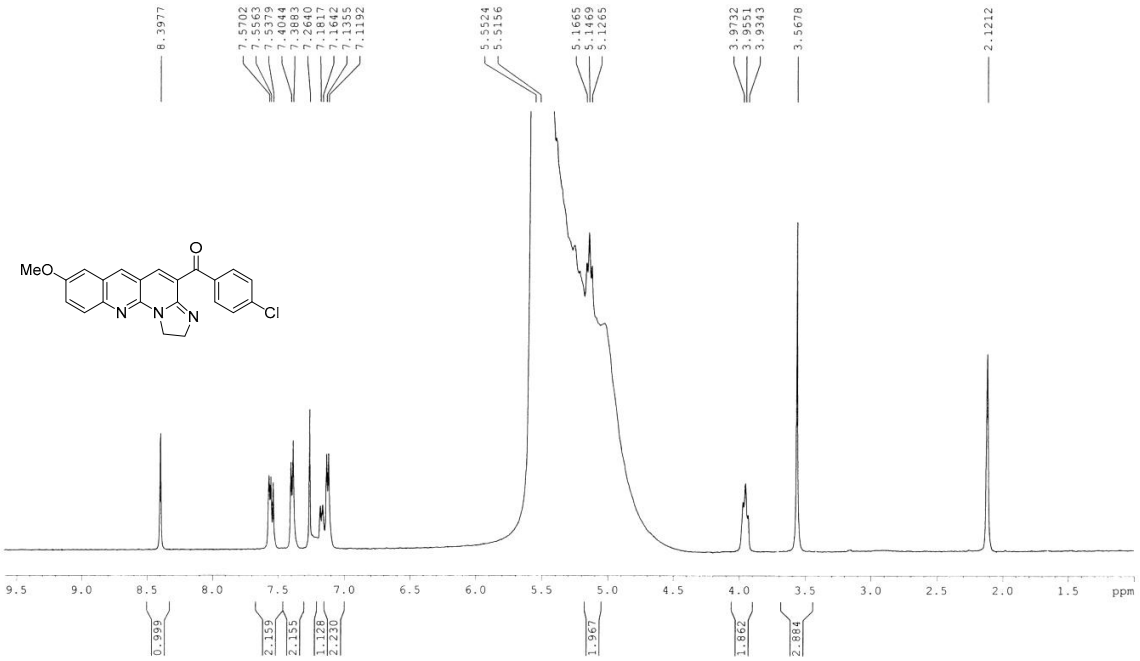


Fig. S15 ^{1}H NMR spectrum (500 MHz, $CDCl_3$+DMSO-d_6+$HClO_4$) of compound **3h**

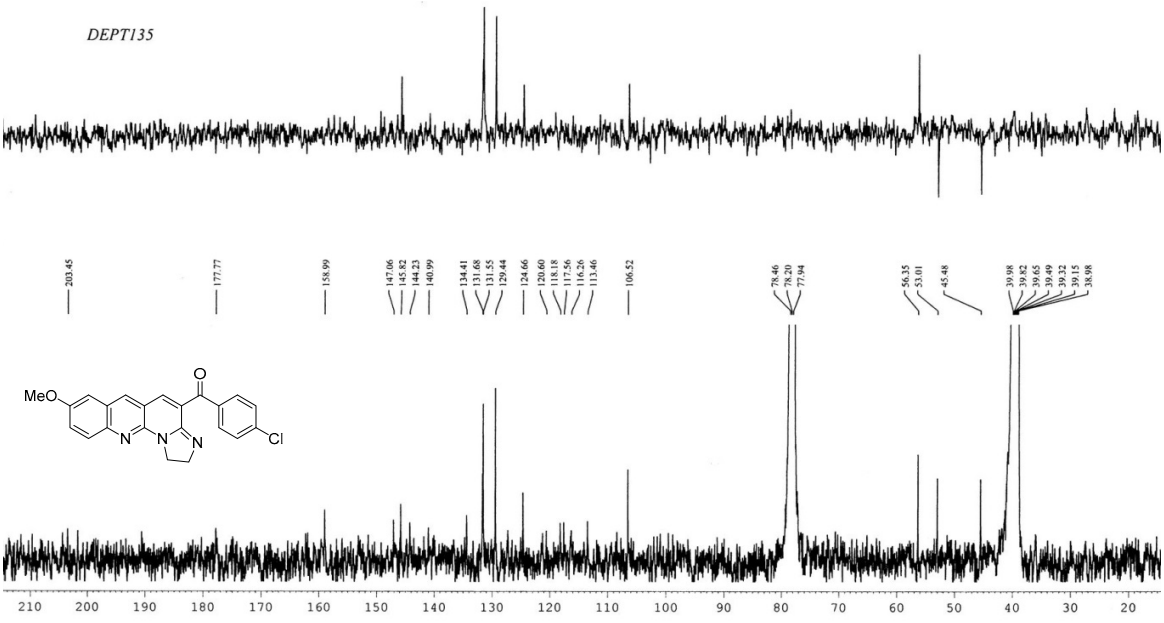


Fig. S16 ^{13}C NMR spectrum (125 MHz, $CDCl_3$+DMSO-d_6+$HClO_4$) of compound **3h**

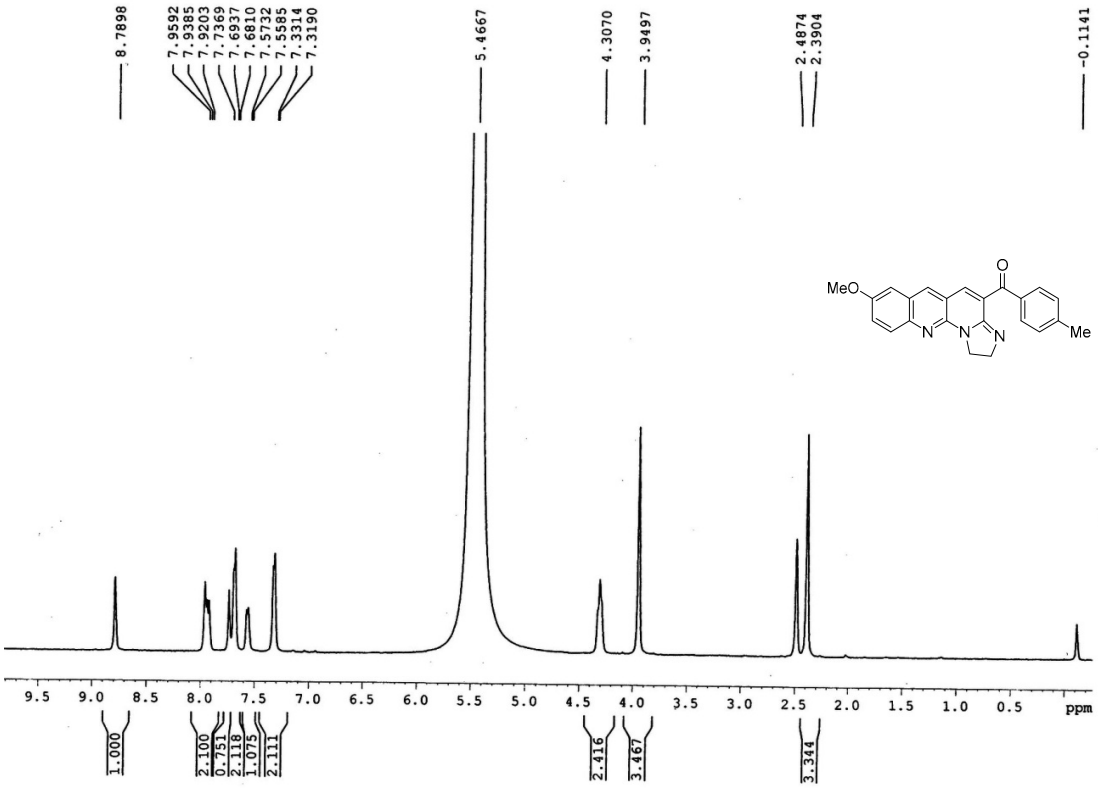


Fig. S17 ^{1}H NMR spectrum (500 MHz, $CDCl_3$+DMSO-d_6+$HClO_4$) of compound **3i**

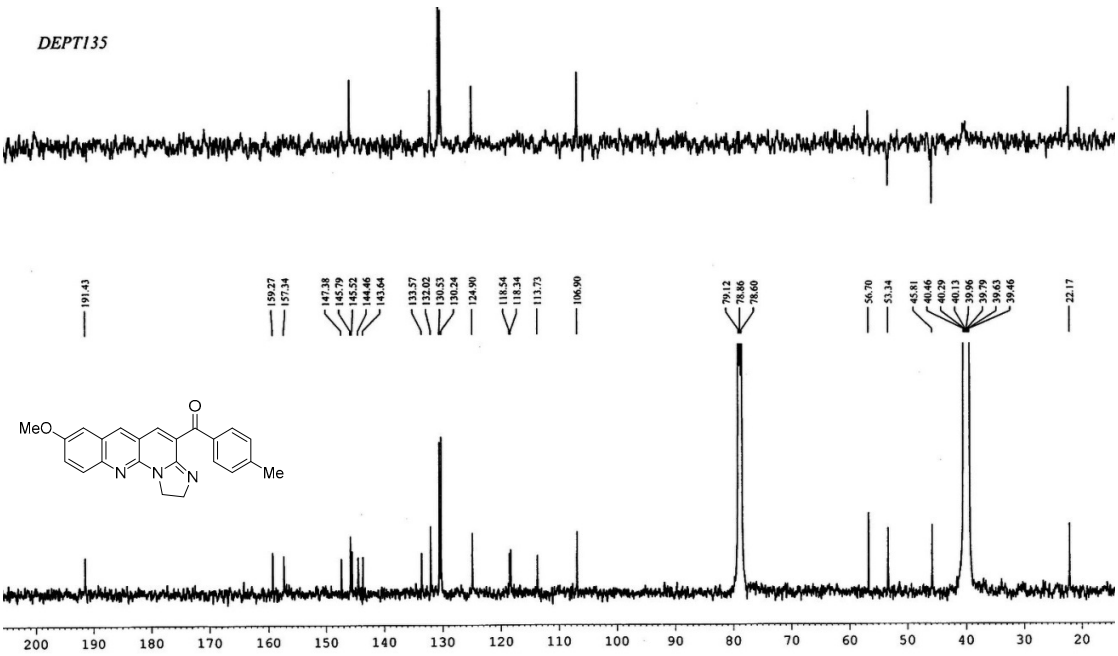


Fig. S18 ^{13}C NMR spectrum (125 MHz, $CDCl_3$+DMSO-d_6+$HClO_4$) of compound **3i**

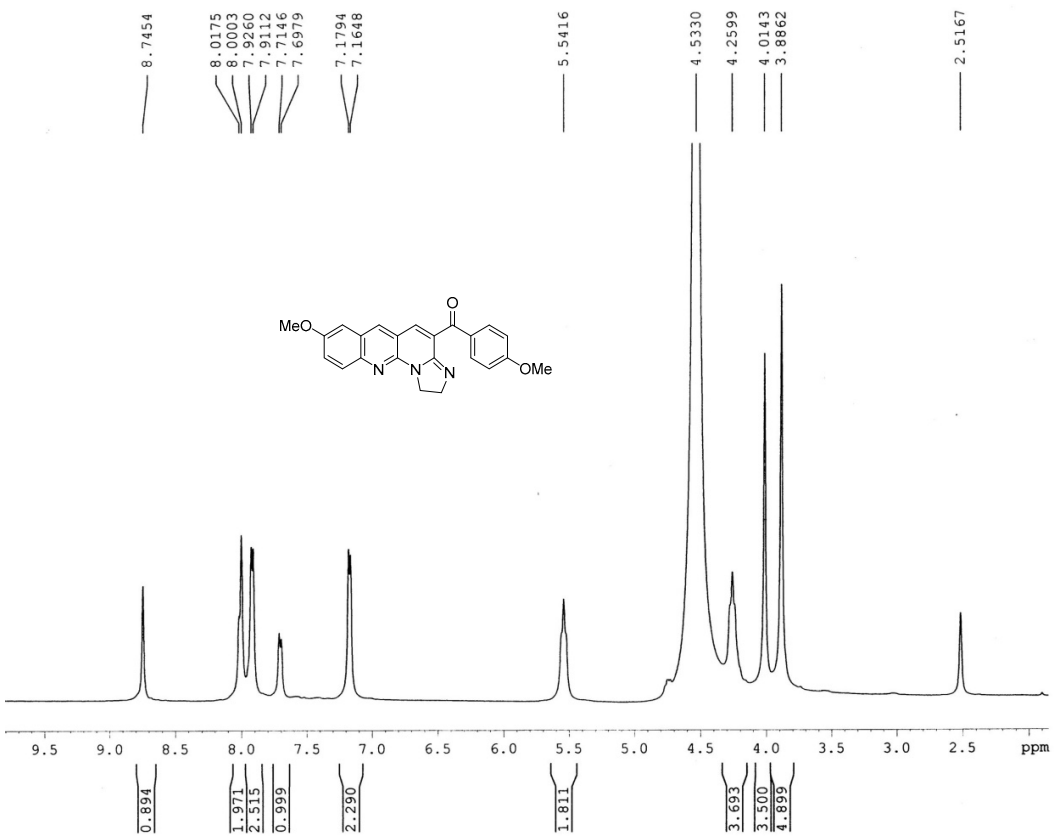


Fig. S19 ^{1}H NMR spectrum (500 MHz, DMSO-d_6+$HClO_4$) of compound **3j**

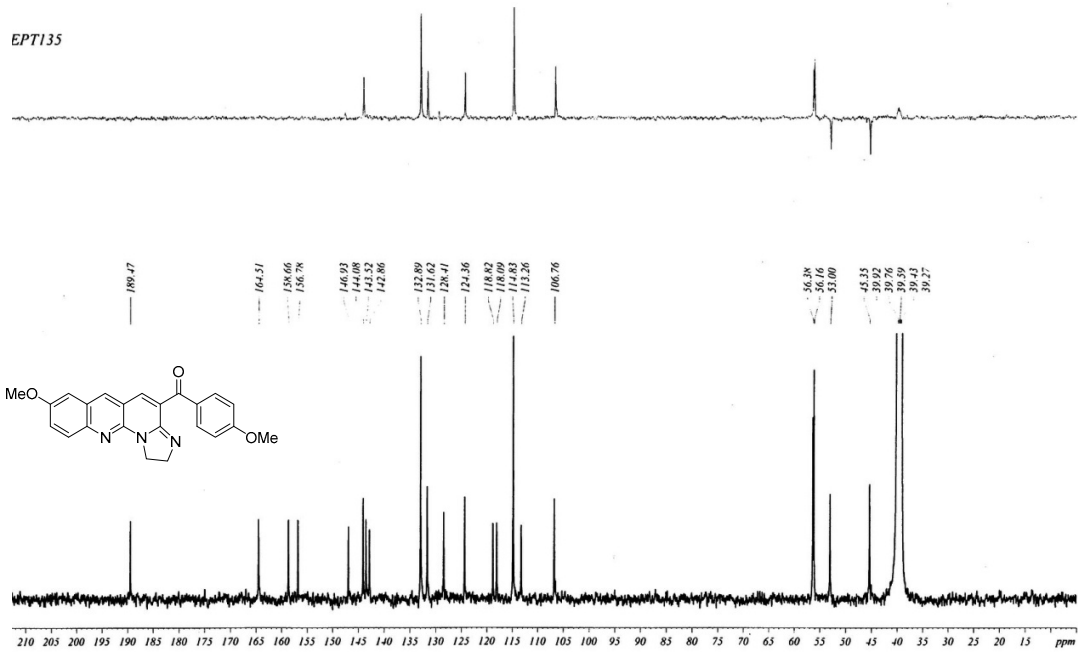


Fig. S20 ^{13}C NMR spectrum (125 MHz, DMSO-d_6+$HClO_4$) of compound **3j**

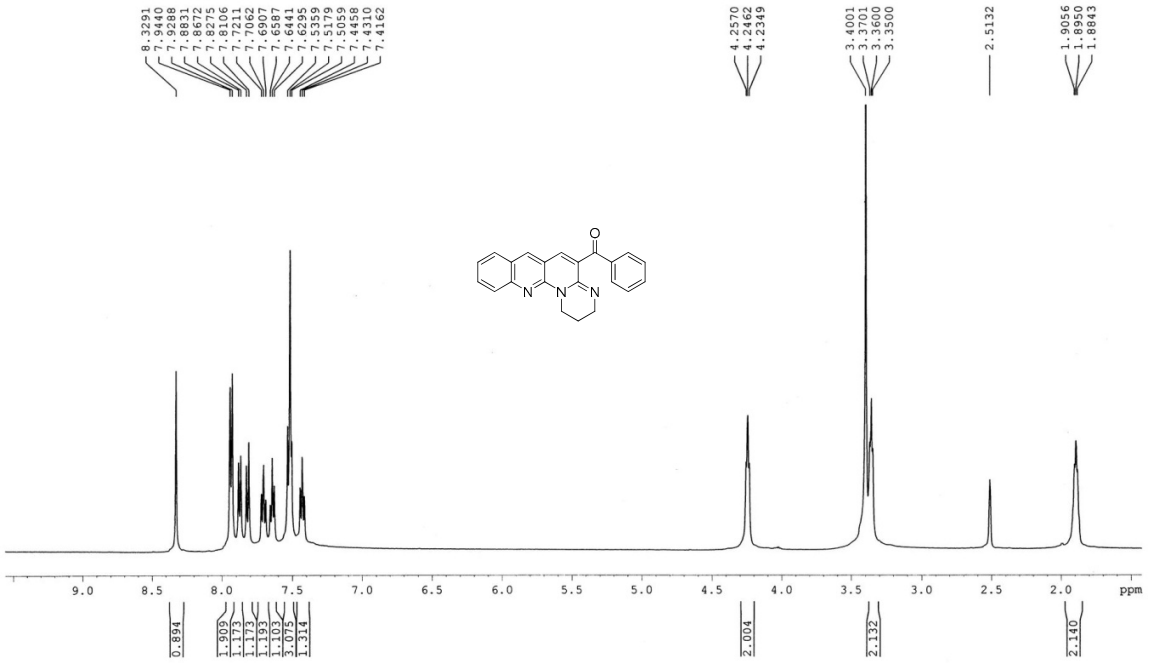


Fig. S21 ^{1}H NMR spectrum (500 MHz, DMSO-d_6) of compound **3k**

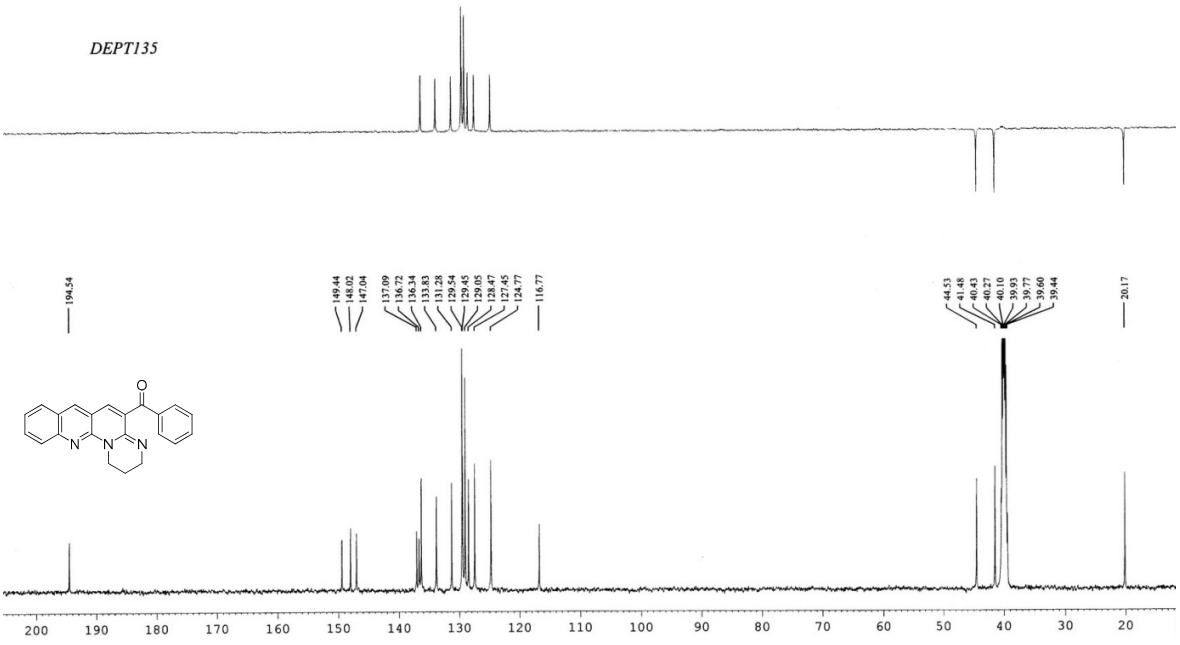


Fig. S22 ^{13}C NMR spectrum (125 MHz, DMSO-d_6) of compound **3k**

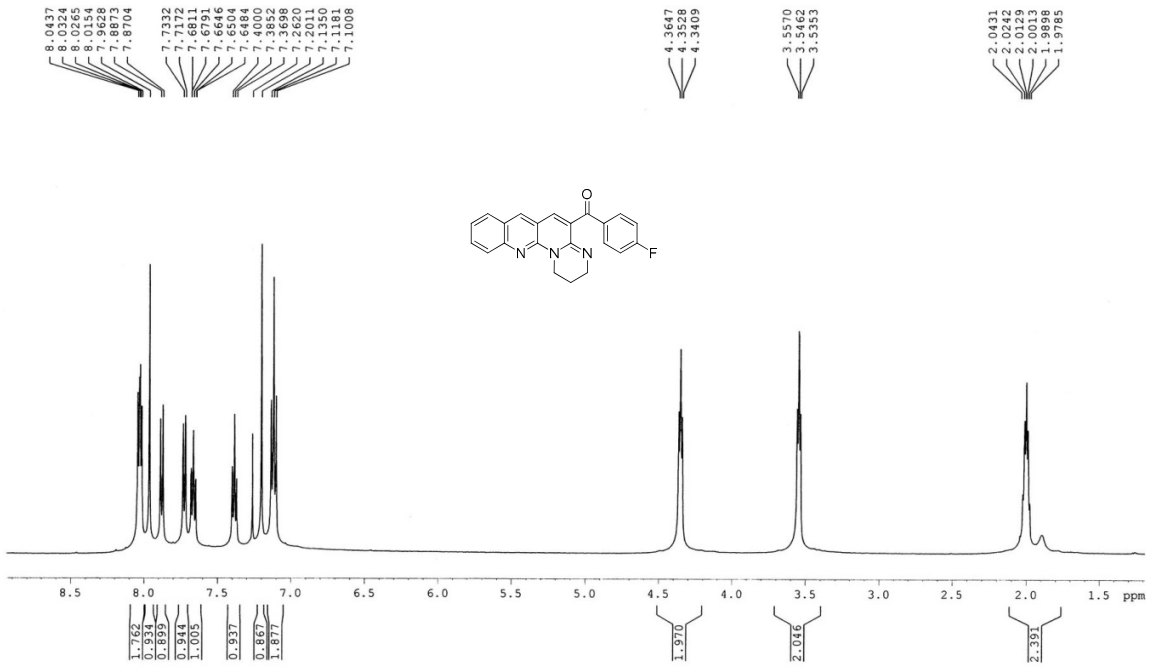


Fig. S23 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of compound **3l**

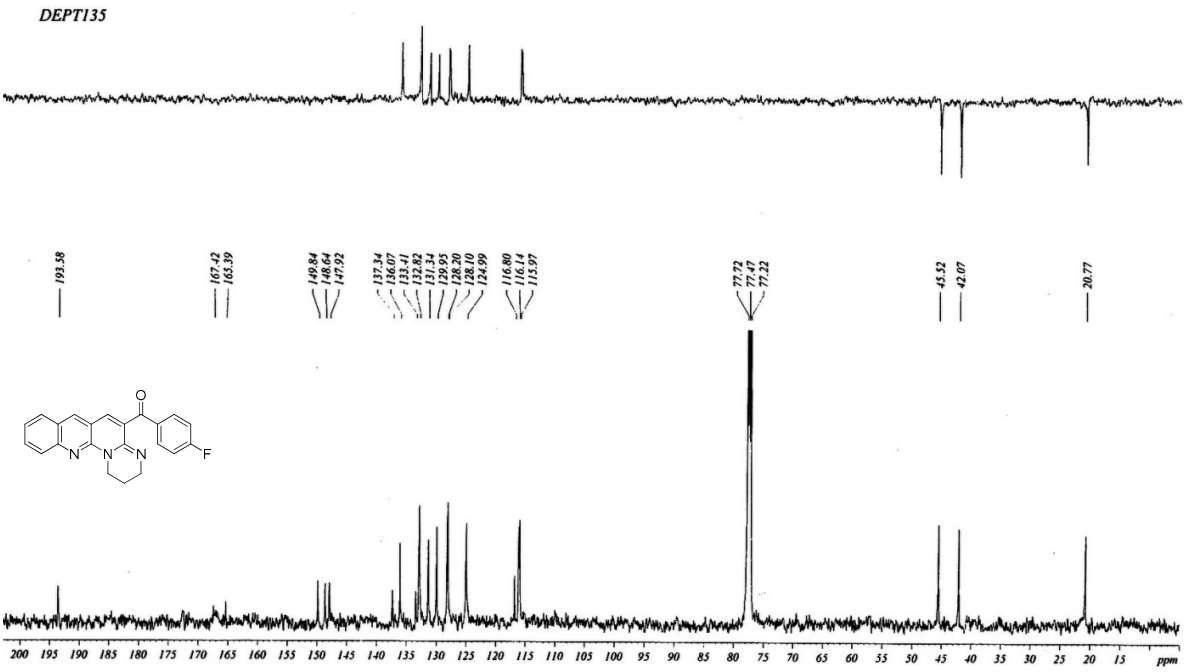


Fig. S24 ^{13}C NMR spectrum (125 MHz, $CDCl_3$) of compound **3l**

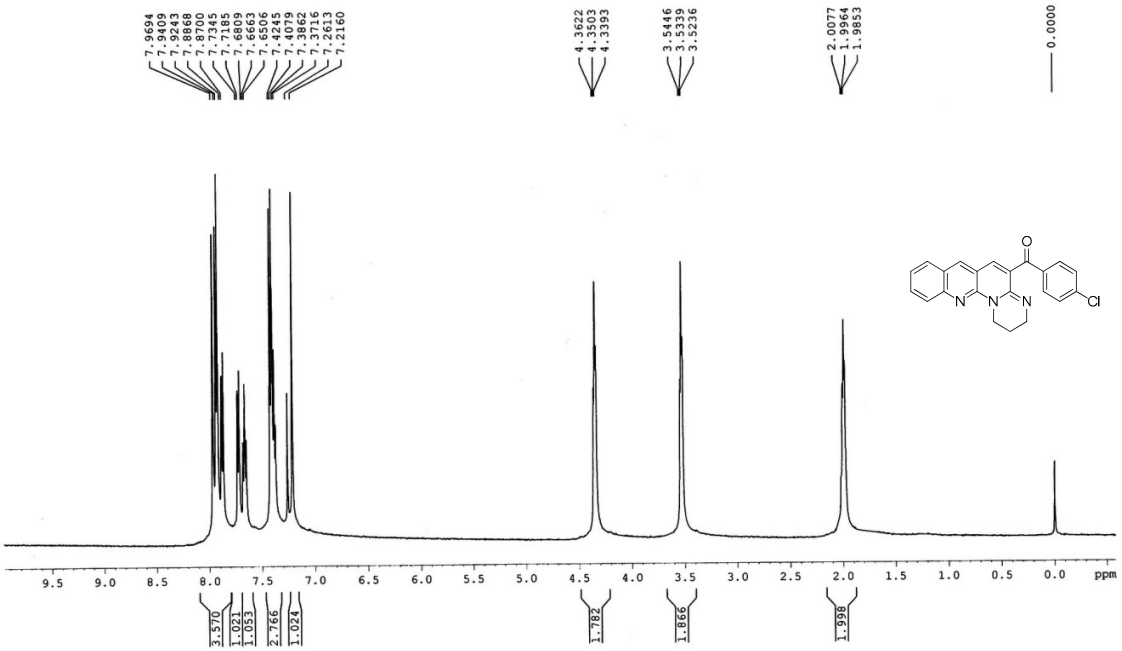


Fig. S25 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of compound **3m**

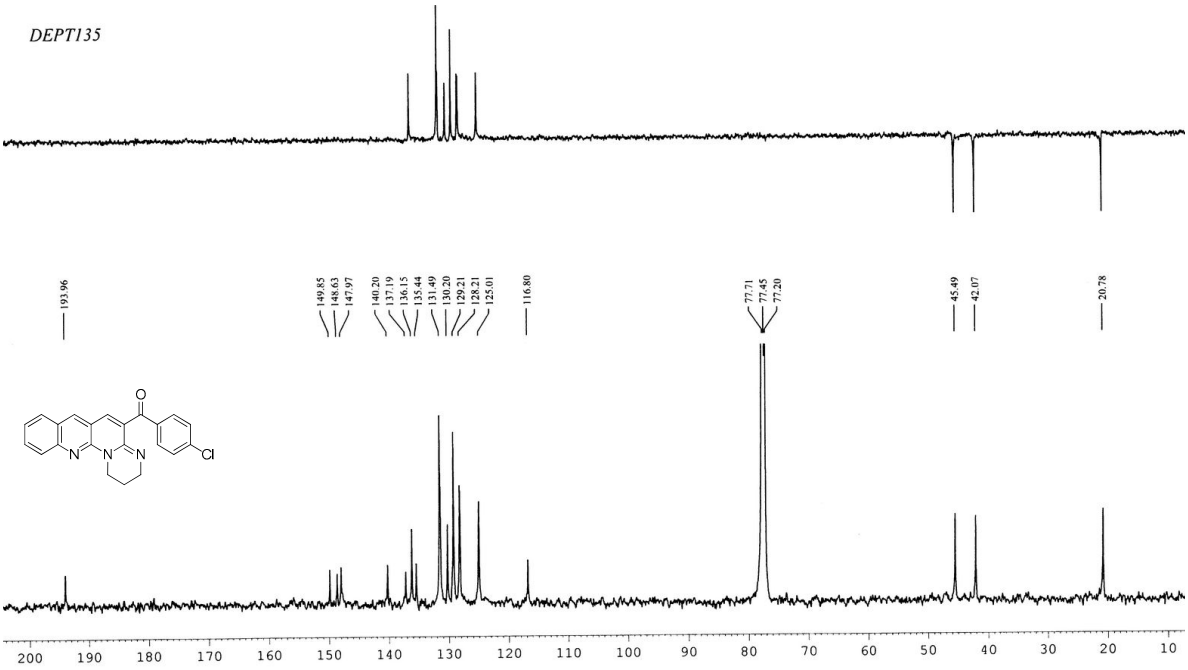


Fig. S26 ^{13}C NMR spectrum (125 MHz, $CDCl_3$) of compound **3m**

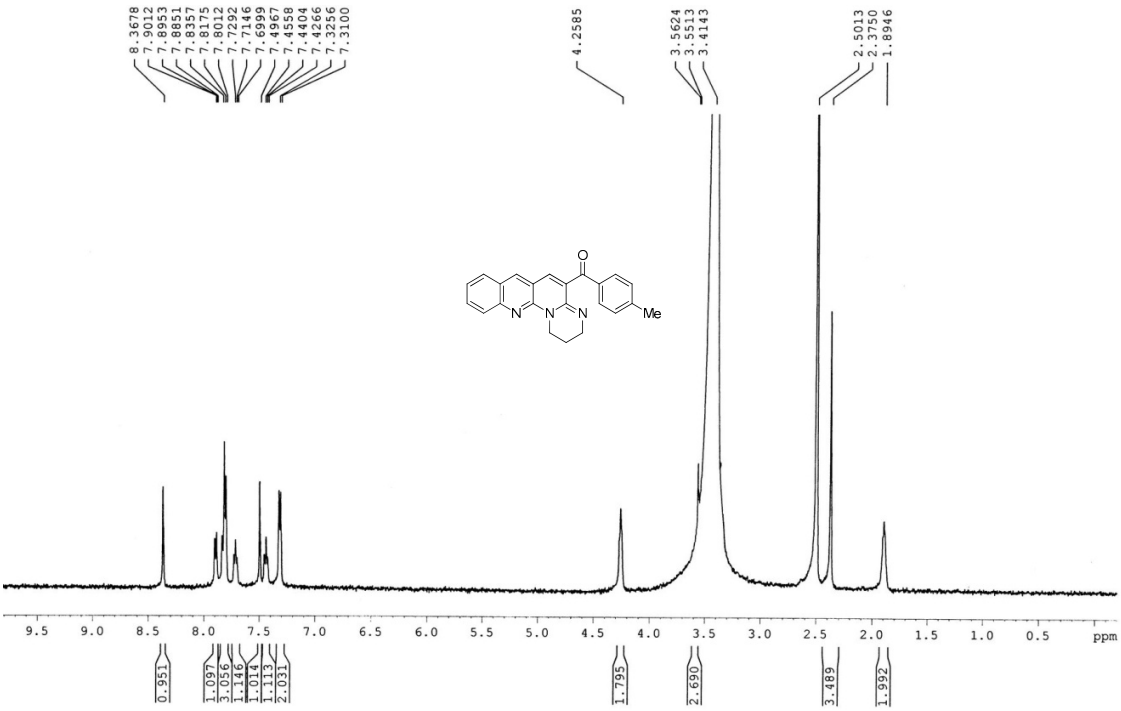


Fig. S27 ^{1}H NMR spectrum (500 MHz, DMSO-d_6) of compound **3n**

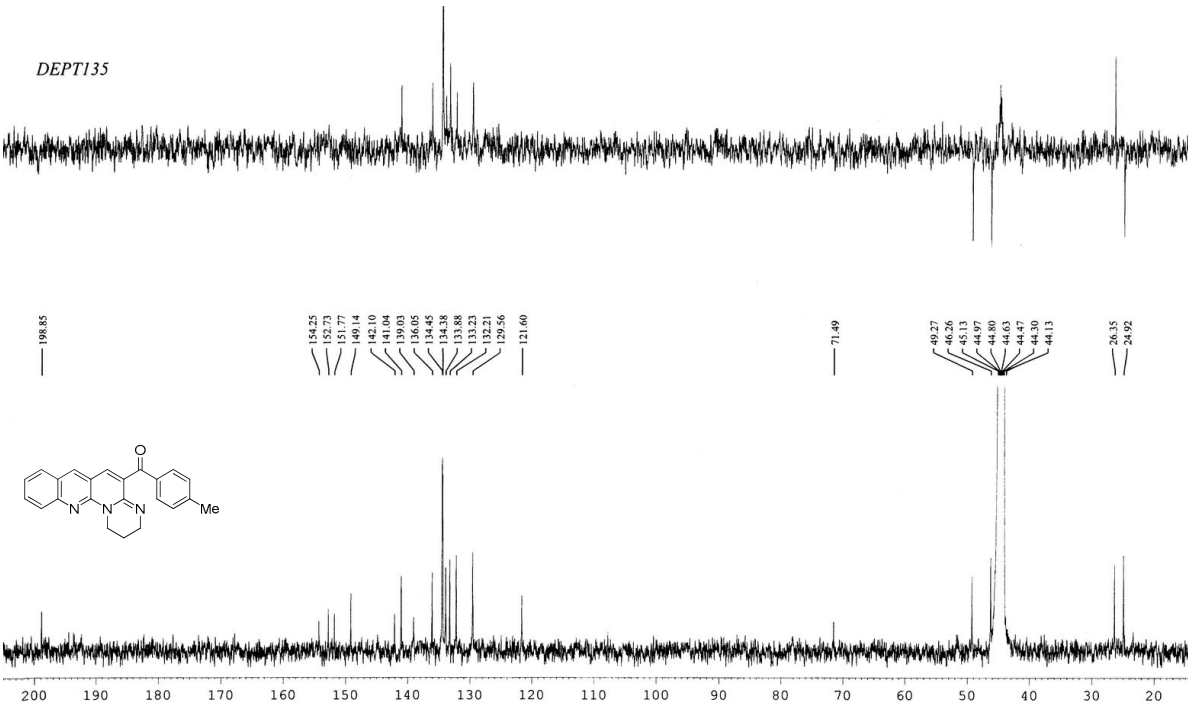


Fig. S28 ^{13}C NMR spectrum (125 MHz, DMSO-d_6) of compound **3n**

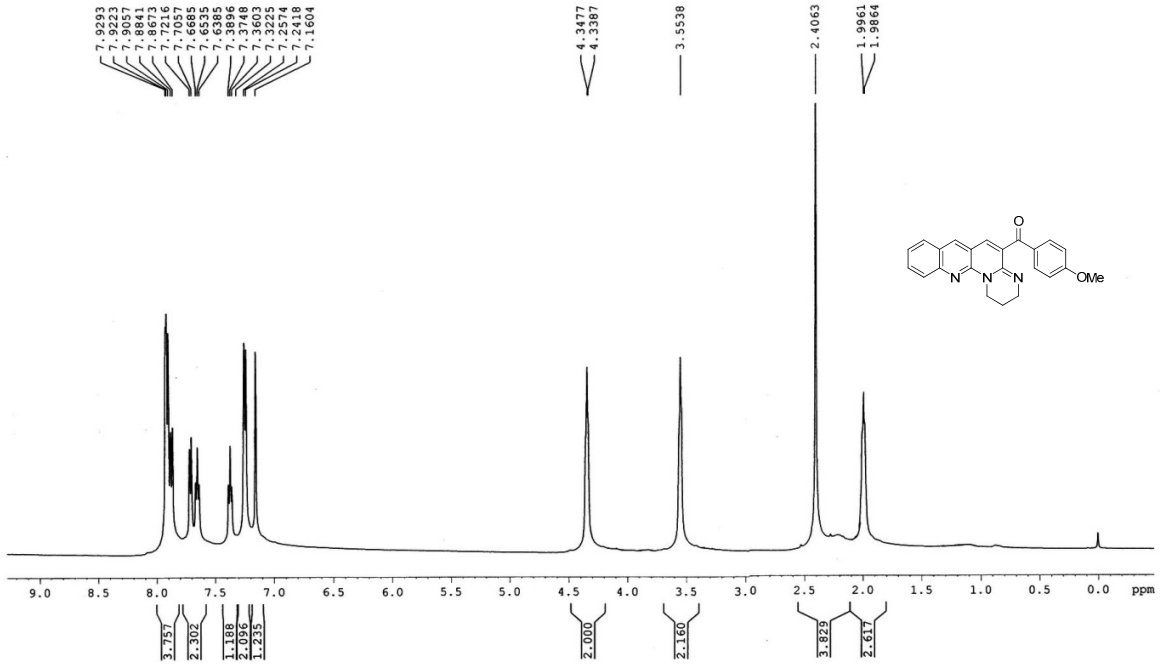


Fig. S29 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of compound **3o**

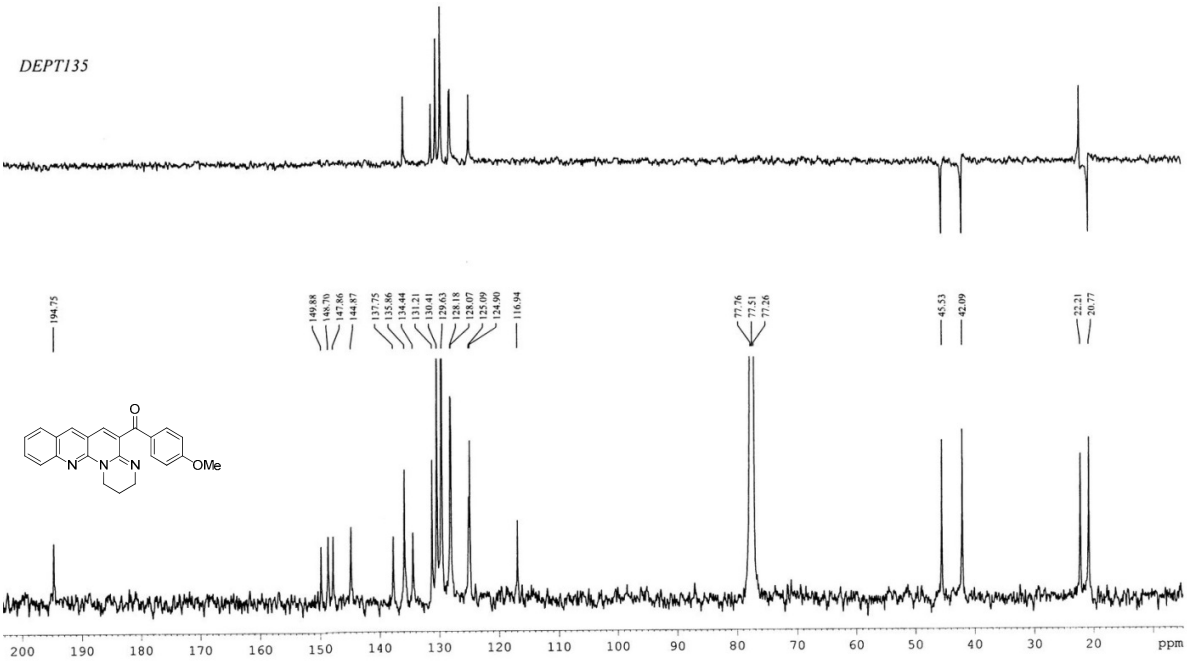


Fig. S30 ^{13}C NMR spectrum (125 MHz, $CDCl_3$) of compound **3o**

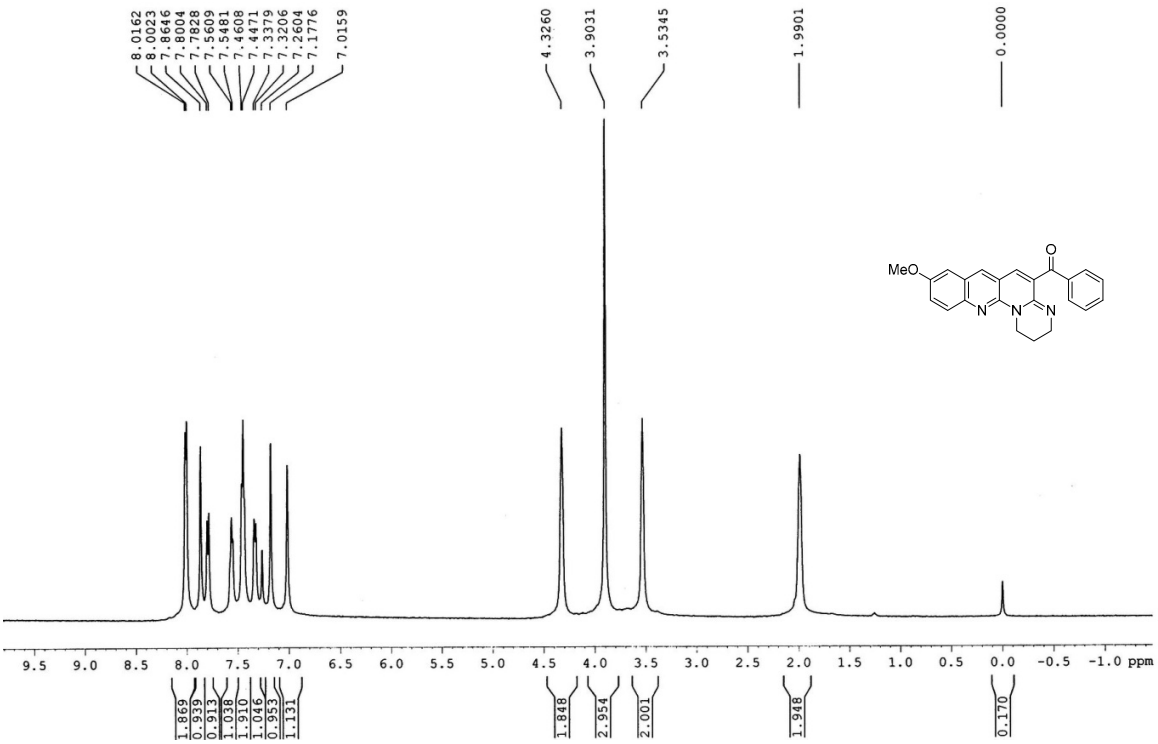


Fig. S31 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of compound **3p**

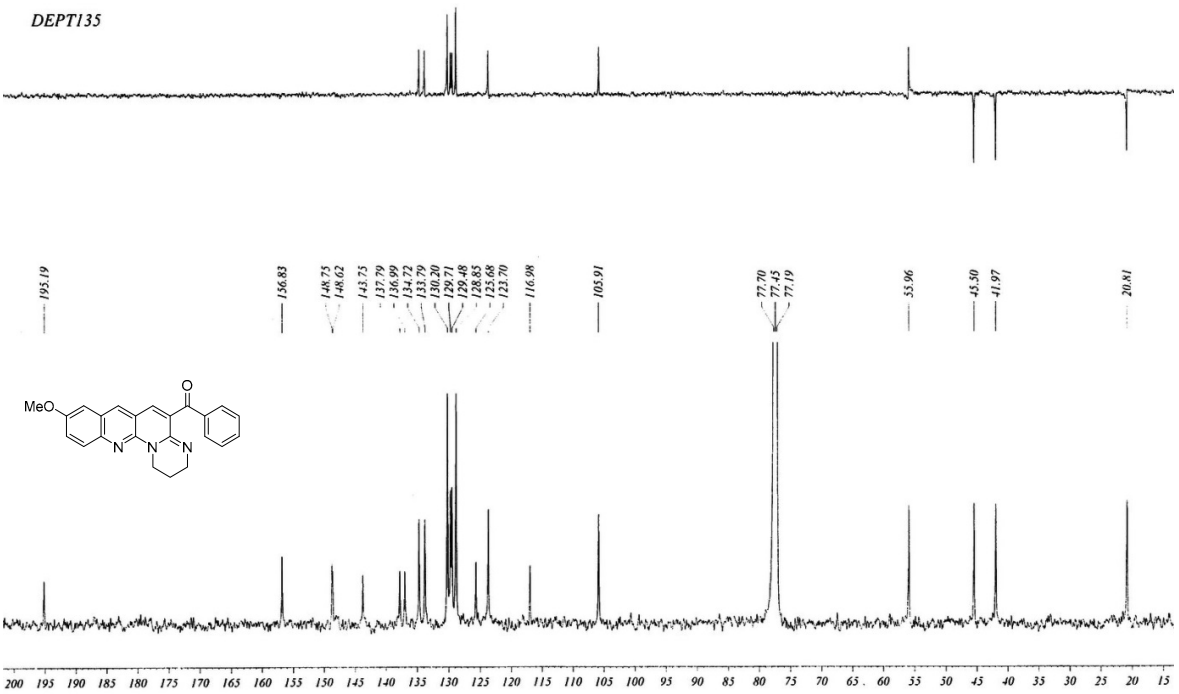


Fig. S32 ^{13}C NMR spectrum (125 MHz, $CDCl_3$) of compound **3p**

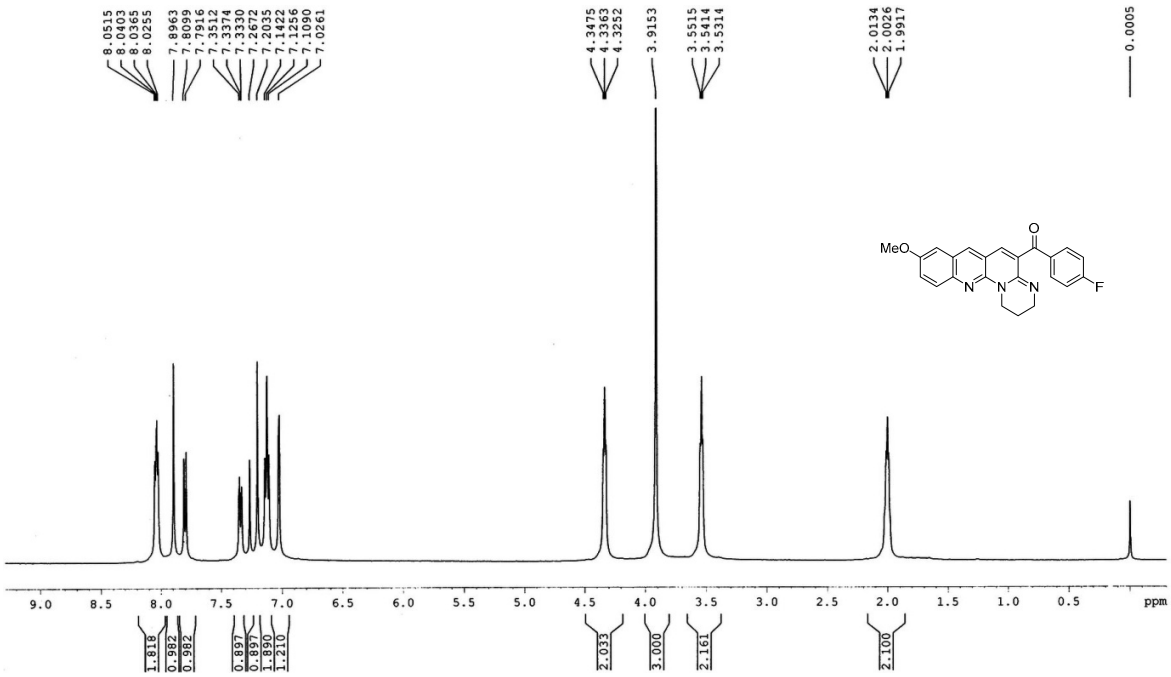


Fig. S33 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of compound **3q**

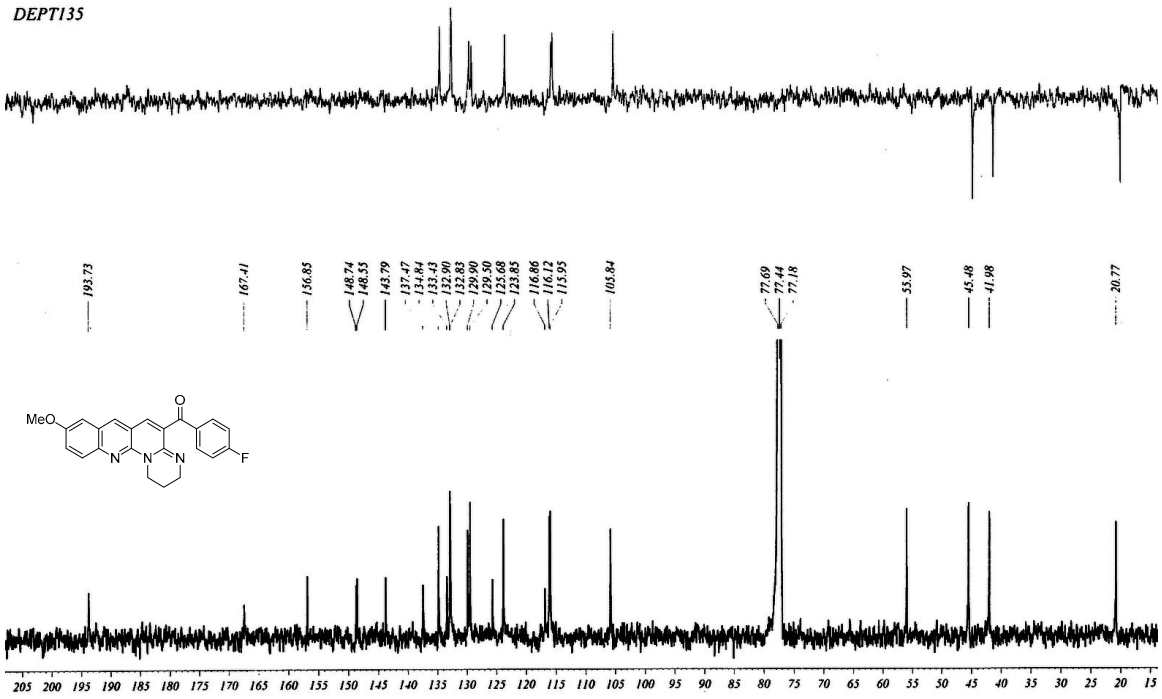


Fig. S34 ^{13}C NMR spectrum (125 MHz, $CDCl_3$) of compound **3q**

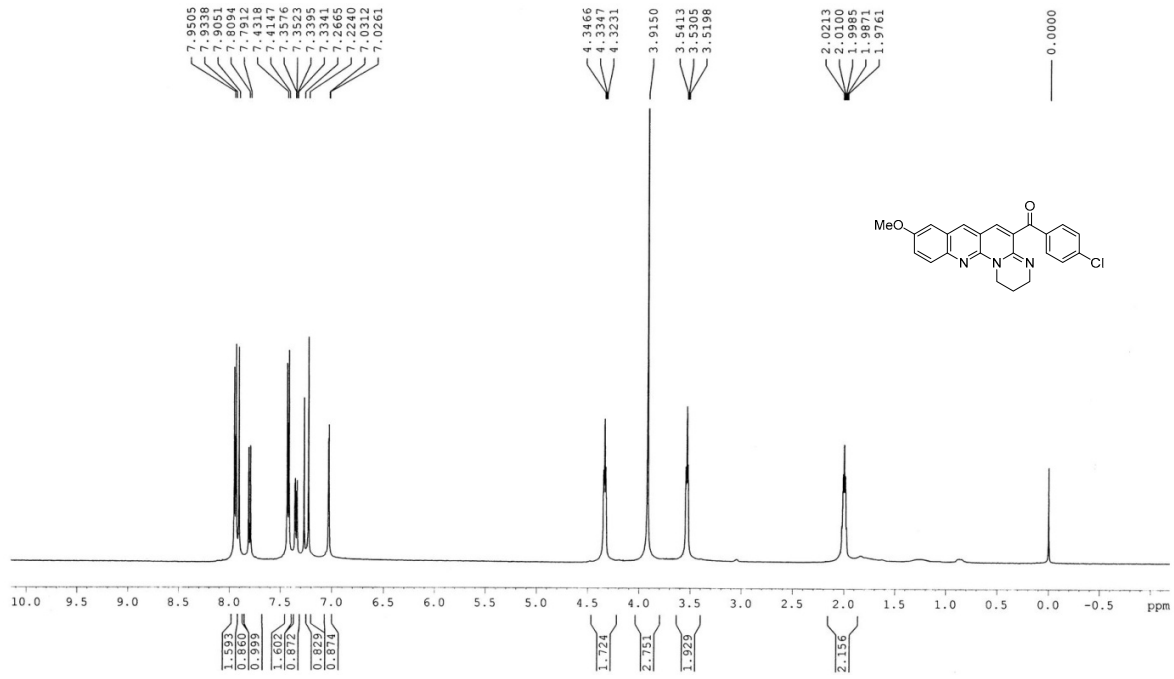


Fig. S35 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of compound **3r**

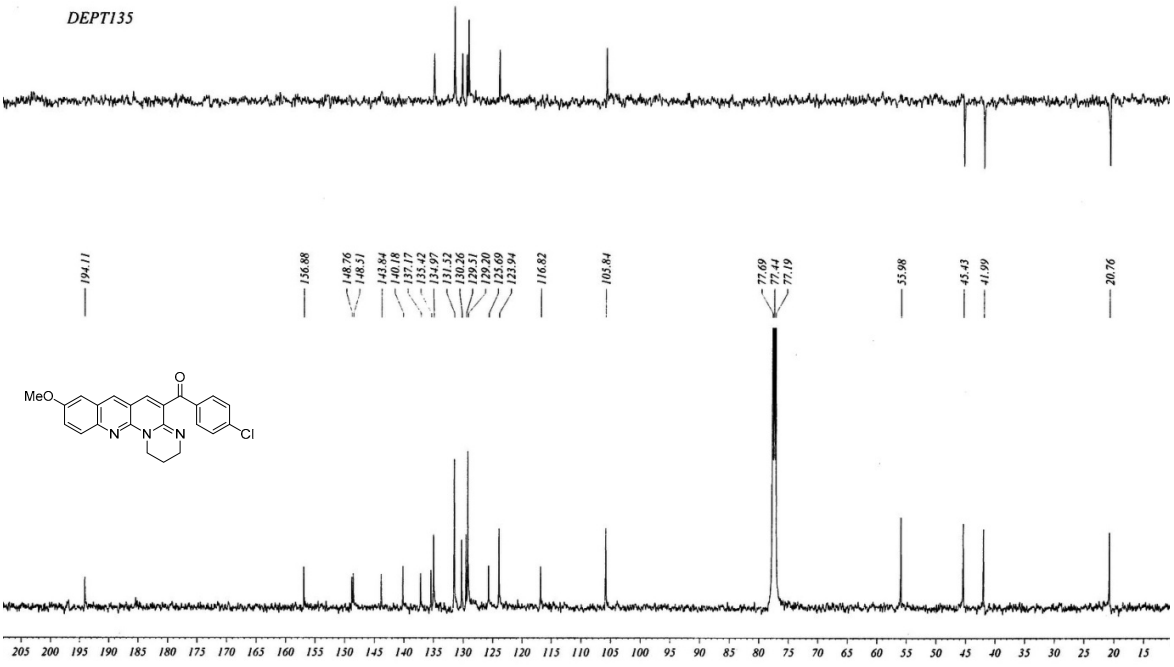


Fig. S36 ^{13}C NMR spectrum (125 MHz, $CDCl_3$) of compound **3r**

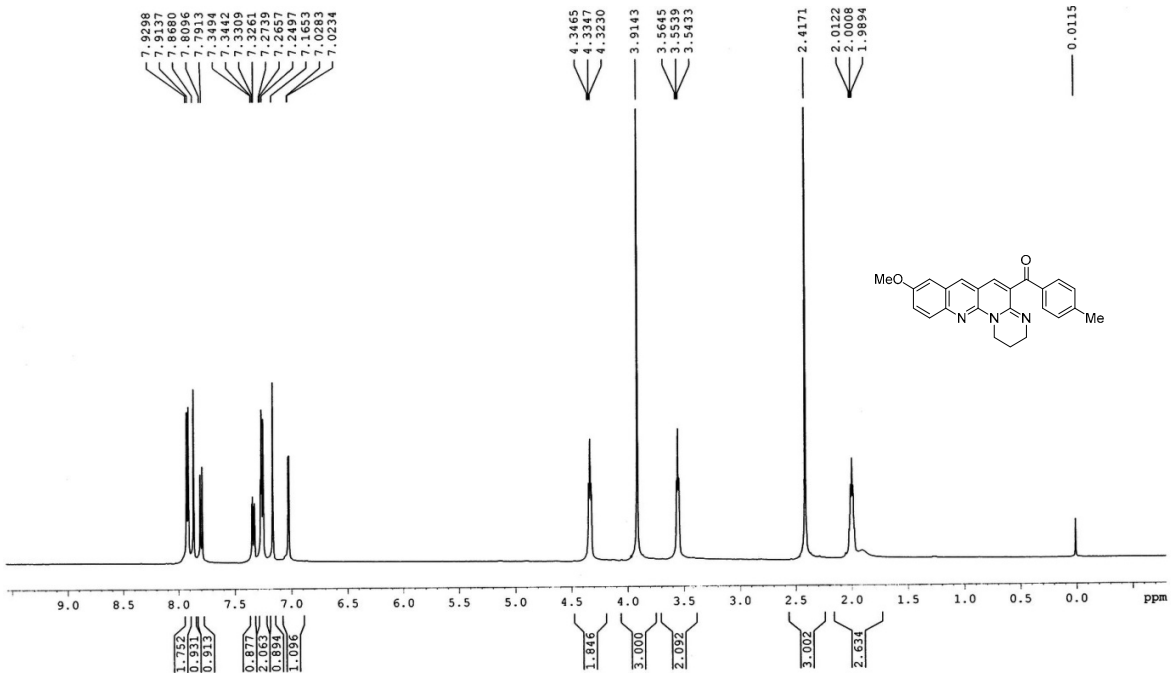


Fig. S37 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of compound **3s**

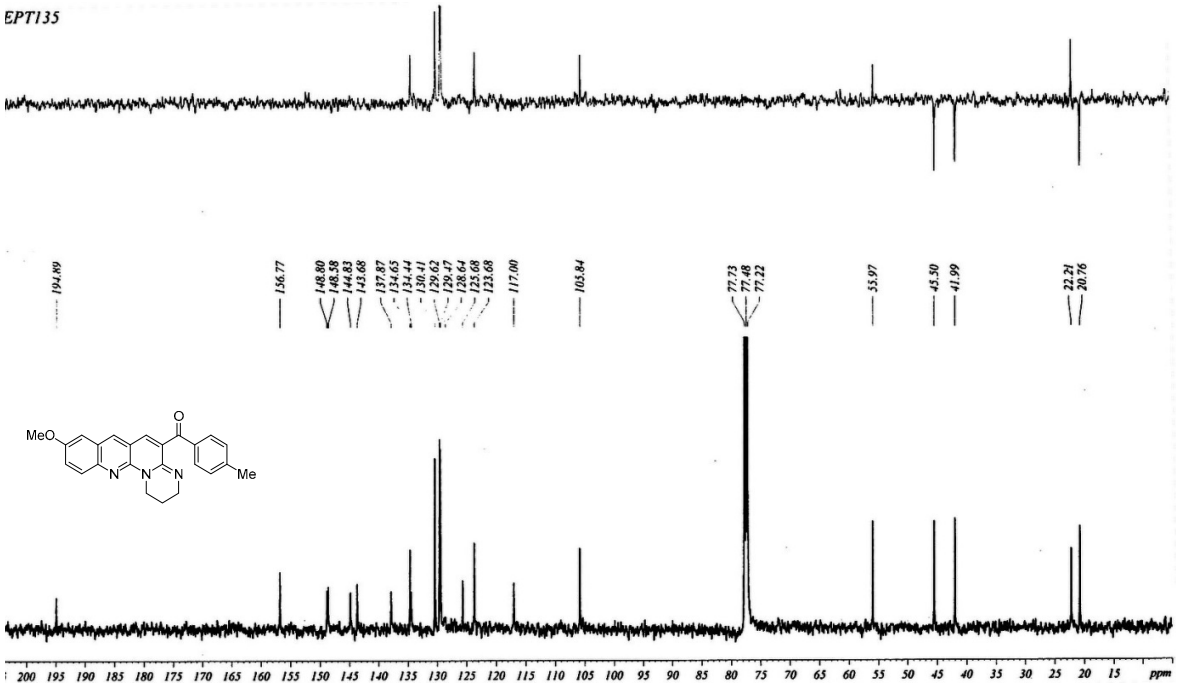


Fig. S38 ^{13}C NMR spectrum (125 MHz, $CDCl_3$) of compound **3s**

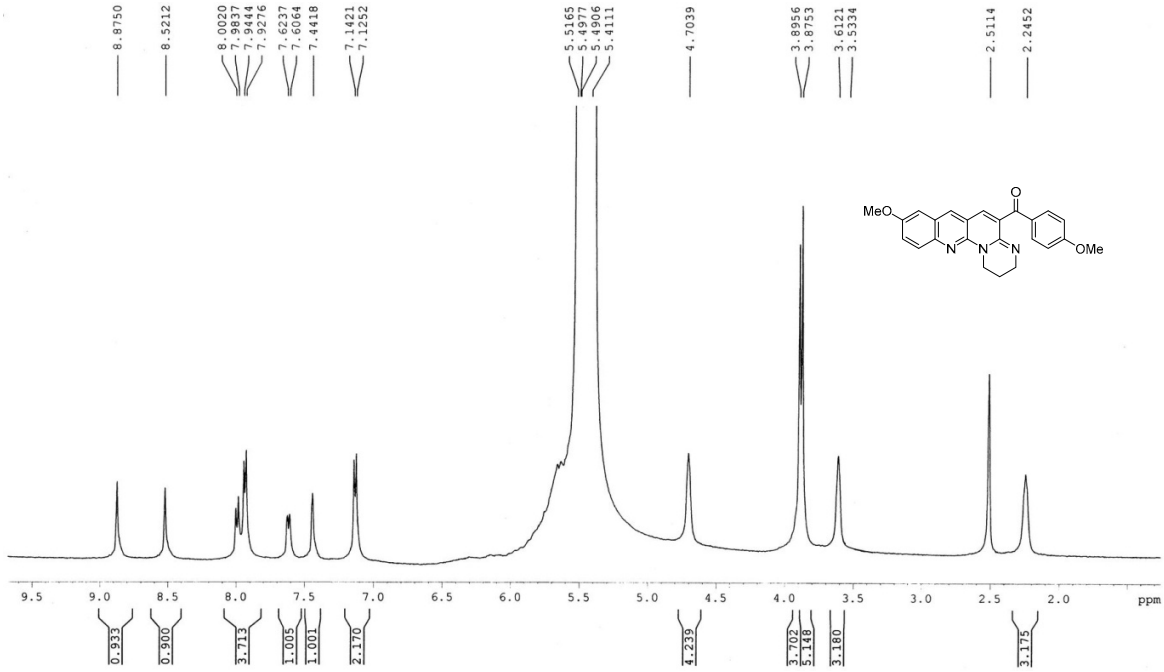


Fig. S39 ^{1}H NMR spectrum (500 MHz, DMSO-d_6+$HClO_4$) of compound **3t**

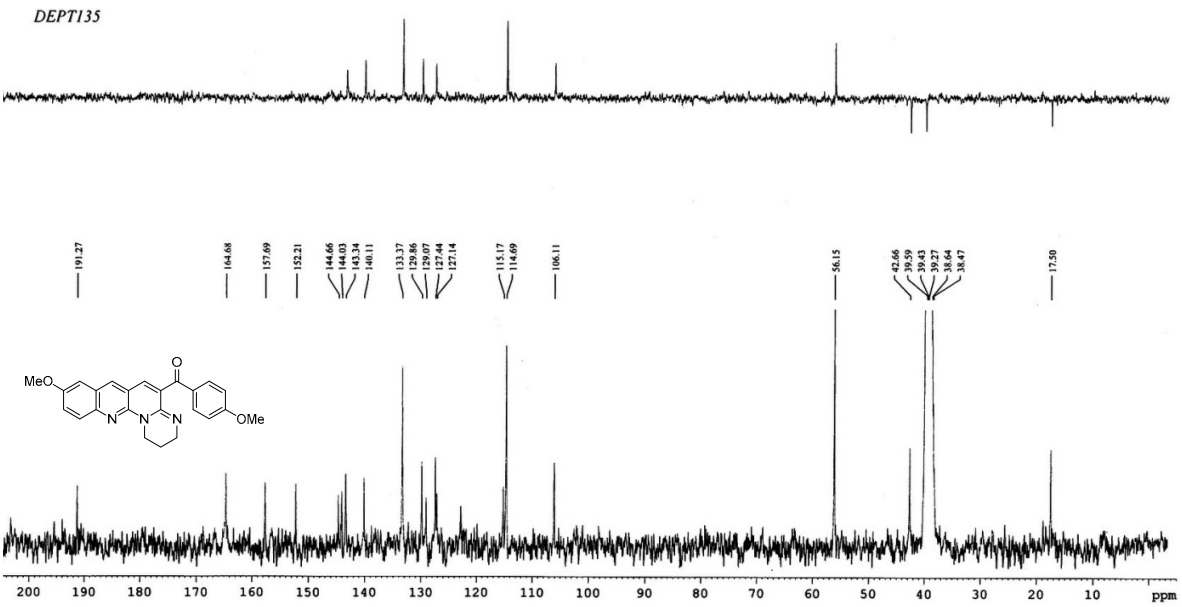


Fig. S40 ^{13}C NMR spectrum (125 MHz, DMSO-d_6+$HClO_4$) of compound **3t**

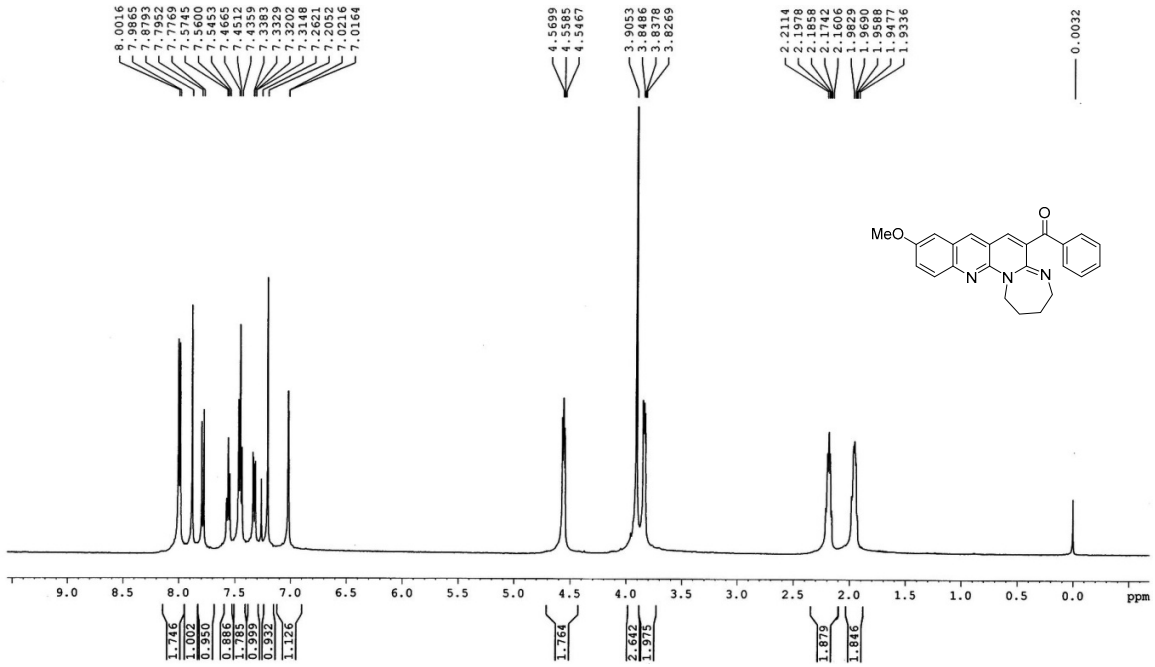


Fig. S41 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of compound **3u**

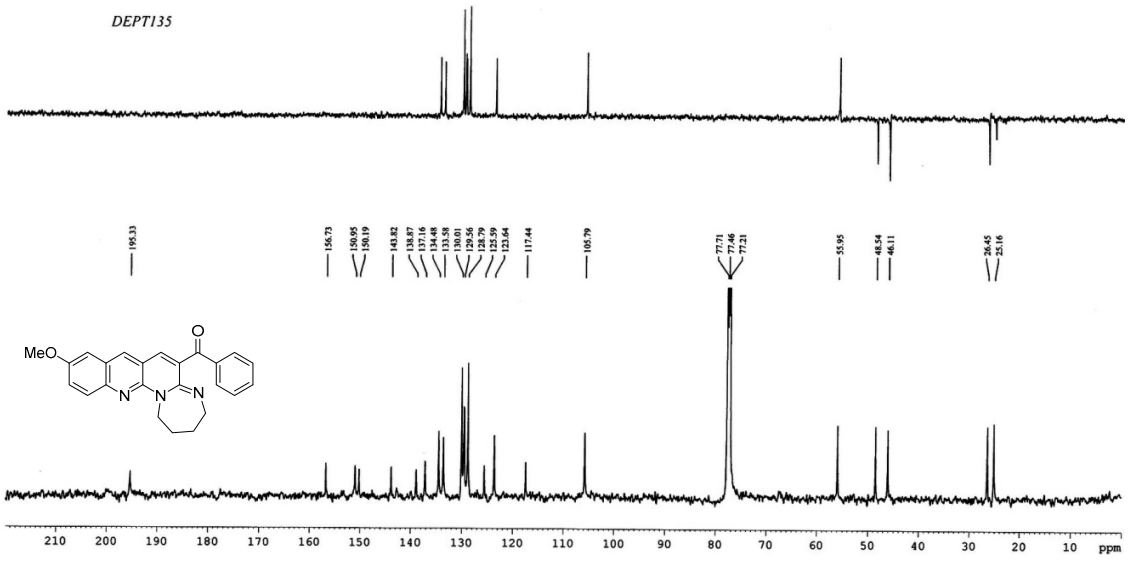


Fig. S42 ^{13}C NMR spectrum (125 MHz, $CDCl_3$) of compound **3u**

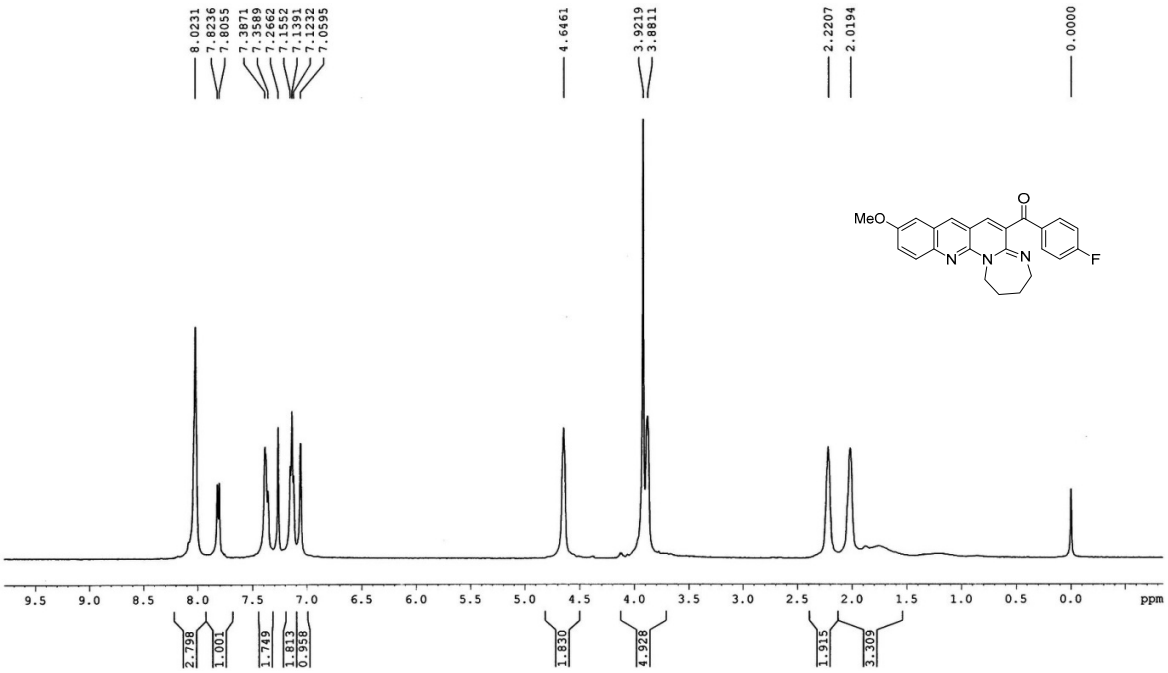


Fig. S43 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of compound **3v**

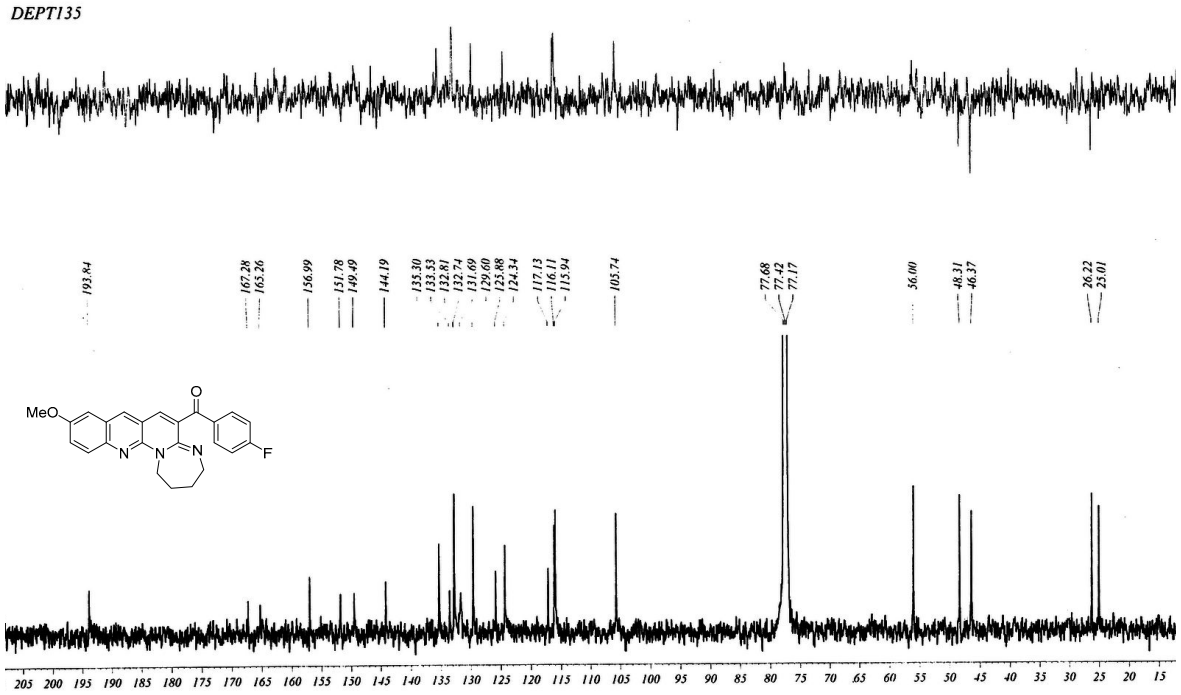


Fig. S44 ^{13}C NMR spectrum (125 MHz, $CDCl_3$) of compound **3v**

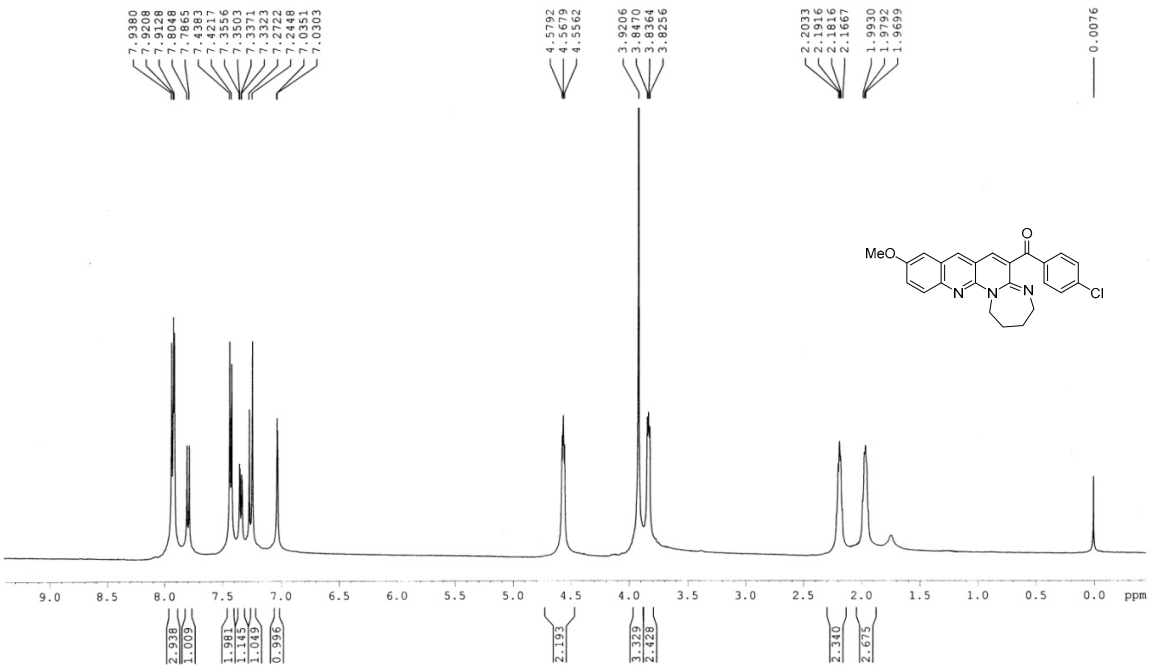


Fig. S45 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of compound **3w**

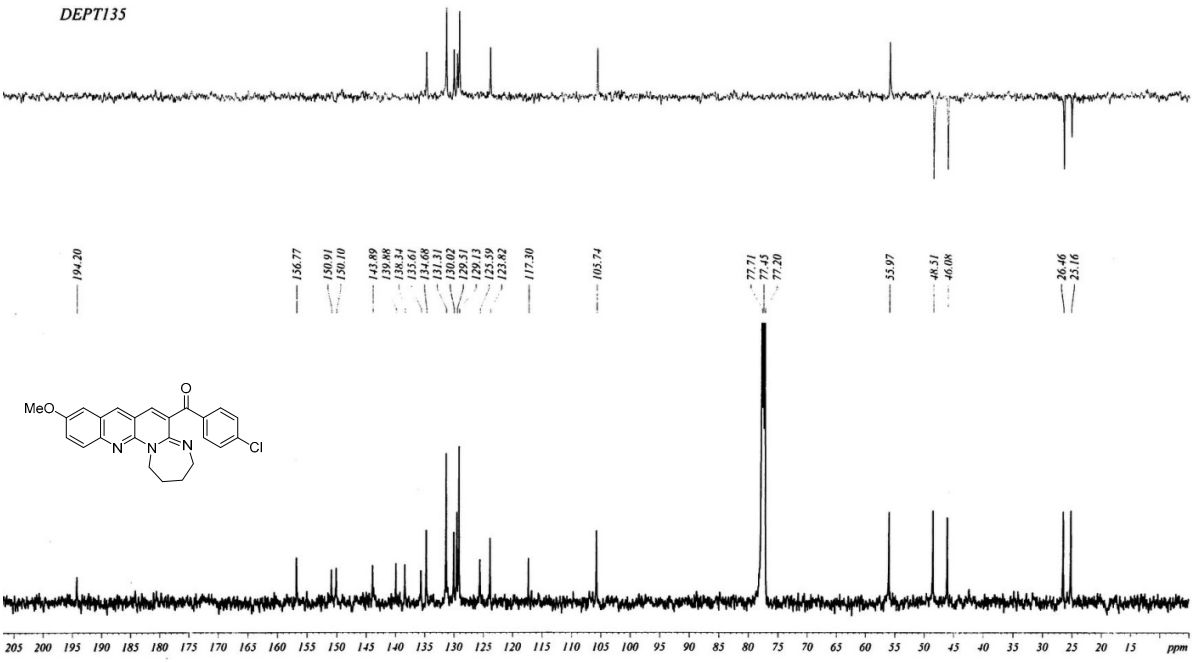


Fig. S46 ^{13}C NMR spectrum (125 MHz, $CDCl_3$) of compound **3w**

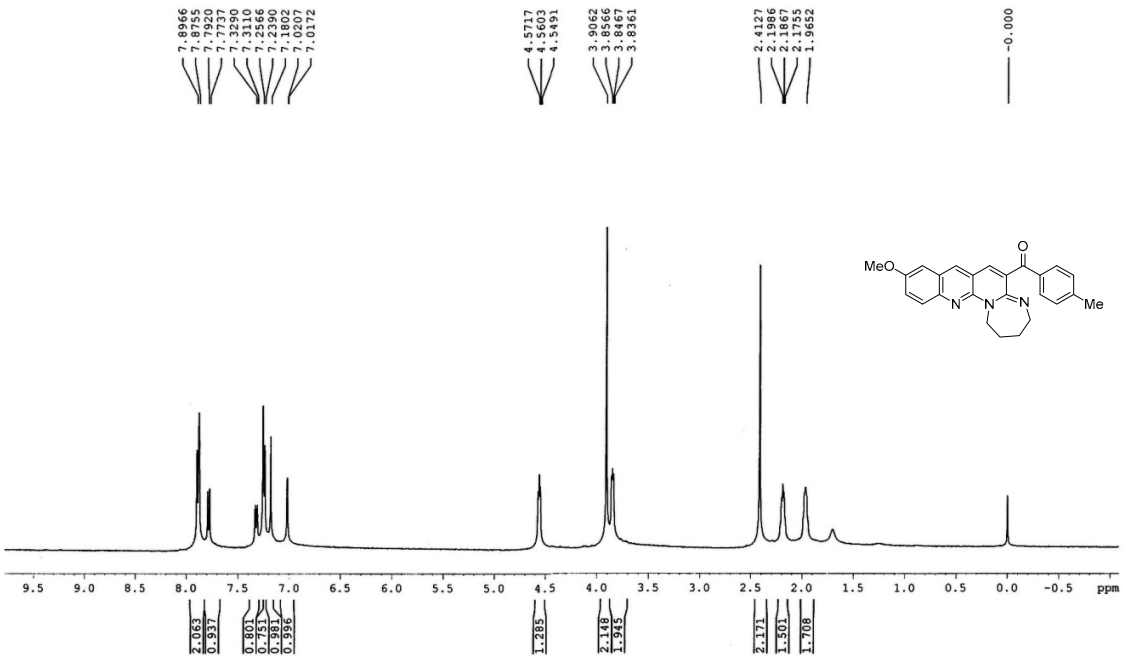


Fig. S47 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of compound **3x**

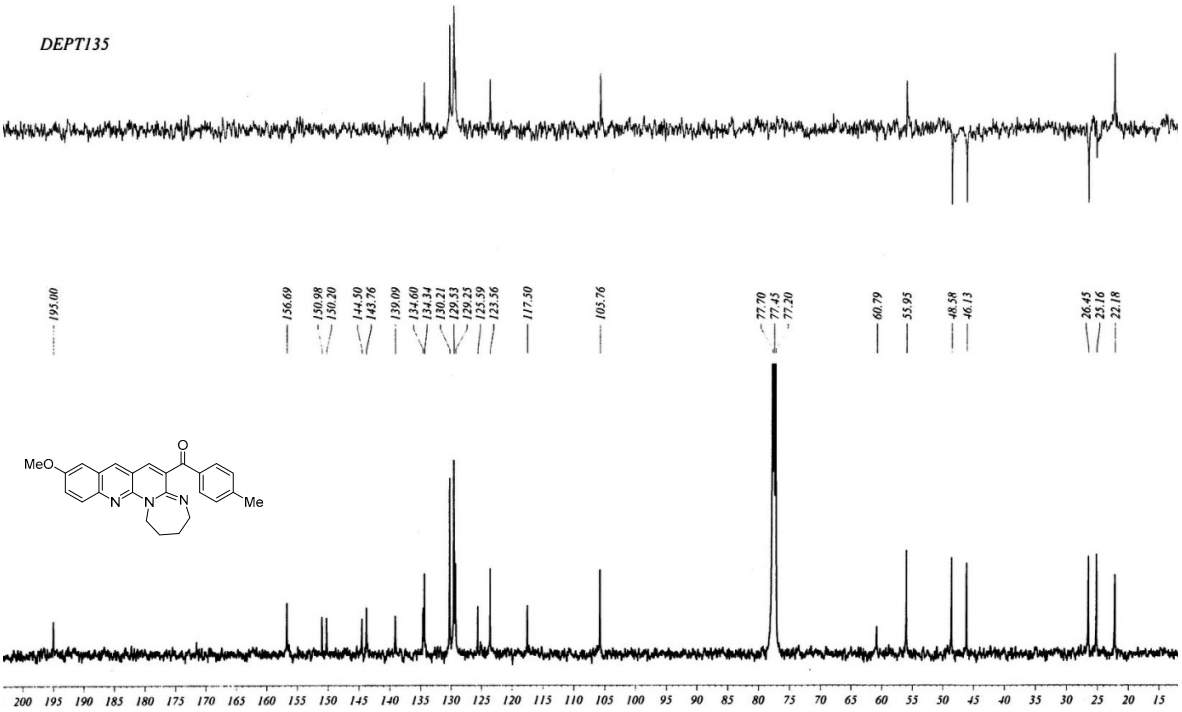


Fig. S48 ^{13}C NMR spectrum (125 MHz, $CDCl_3$) of compound **3x**

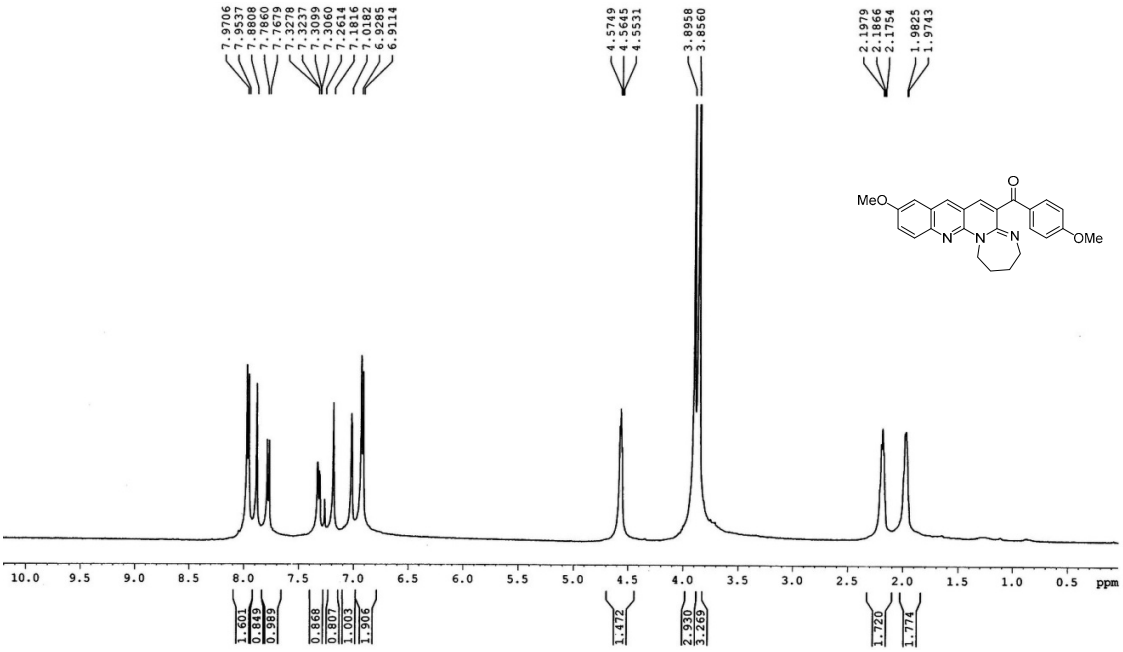


Fig. S49 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of compound **3y**

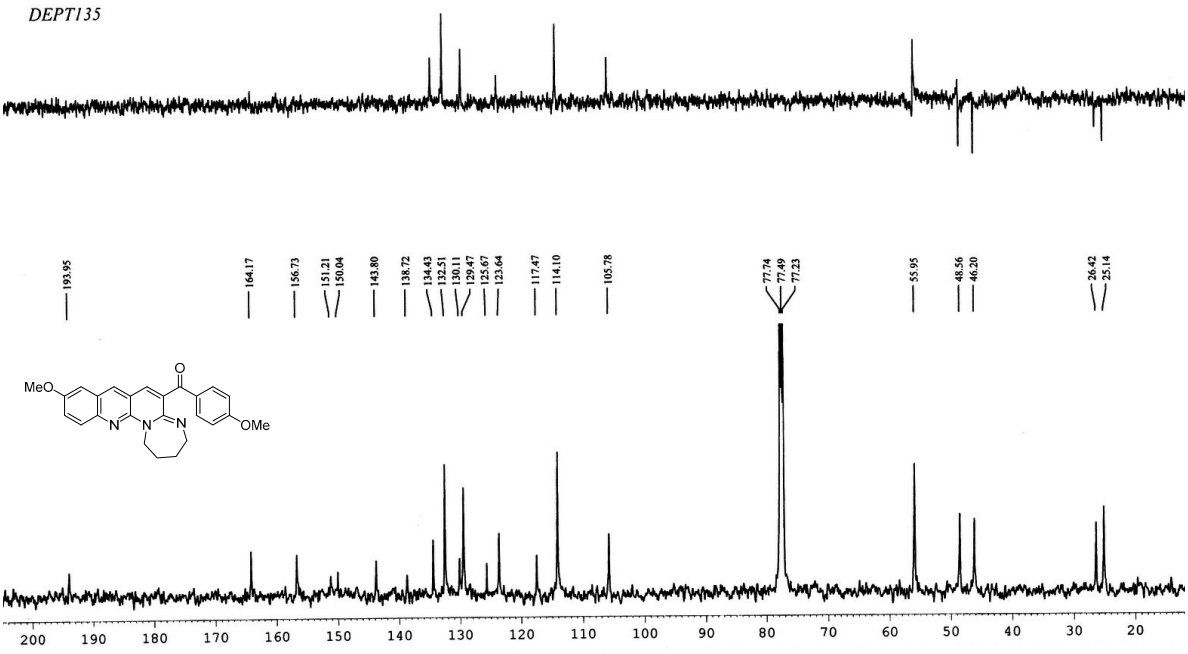


Fig. S50 ^{13}C NMR spectrum (125 MHz, $CDCl_3$) of compound **3y**

2 Geometries, energies and graphics of frontier molecular orbital for all optimized synthons and intermediates[1]

Single Point Energy (E_{B3LYP}, in Hartree)

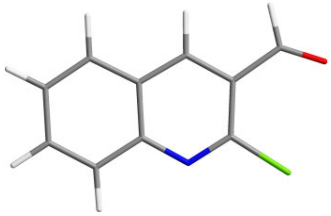
Zero-point correction (E_0, in Hartree)

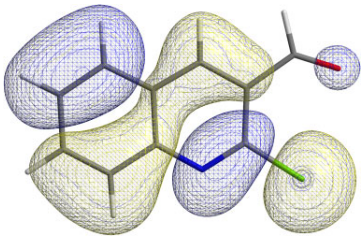
Thermal correction to Enthalpy (H, in Hartree)

Thermal correction to Gibbs Free Energy ($TCGFE$, in Hartree)

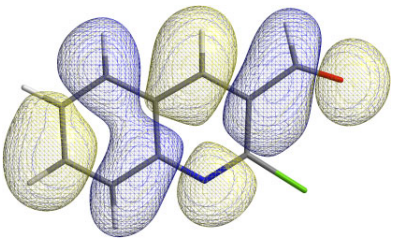
Table S1. Cartesian coordinates, optimized geometry, and energies of compound **1a**

2-Chloroquinoline-3-carbaldehyde (1a)			
E_{B3LYP}	E_0	H	$TCGFE$
-974.9948565	0.134526	0.145440	0.098169





HOMO



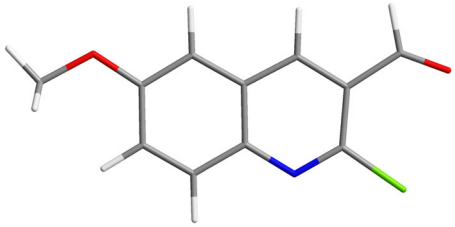
LUMO

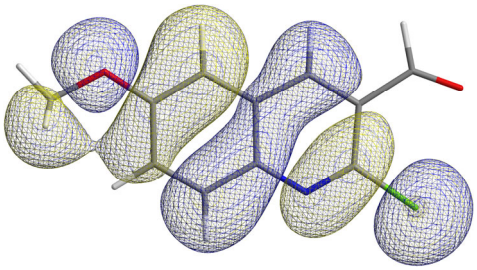
Atoms	x	y	z
C	-3.684814	0.636347	0.000005
C	-3.601097	-0.776043	0.000020
C	-2.381181	-1.411137	0.000019
C	-1.188228	-0.651591	0.000003
C	-1.270267	0.773099	-0.000011
C	-2.541957	1.398571	-0.000010
N	0.009338	-1.304226	0.000002
C	1.107071	-0.615205	-0.000013
C	1.163260	0.818243	-0.000027
C	-0.049021	1.477222	-0.000022
Cl	2.591102	-1.548839	-0.000013
C	2.383544	1.670219	-0.000051
H	2.127655	2.753706	0.000109
O	3.532035	1.310010	0.000076
H	-4.657650	1.113690	0.000006
H	-4.513381	-1.361560	0.000033
H	-2.294779	-2.490405	0.000030
H	-2.598244	2.481887	-0.000021
H	-0.057837	2.564105	-0.000030

Table S2. Cartesian coordinates, optimized geometry, and energies of compound **1b**

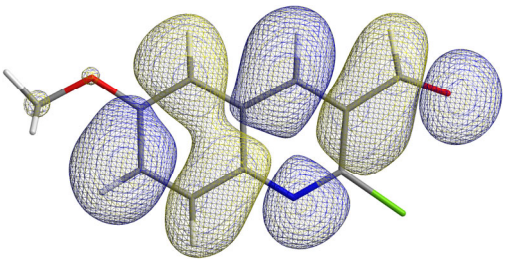
2-Chloro-6-methoxyquinoline-3-carbaldehyde (1b)

E_{B3LYP}	E_0	H	$TCGFE$
-1089.5477113	0.166616	0.180247	0.126714





HOMO



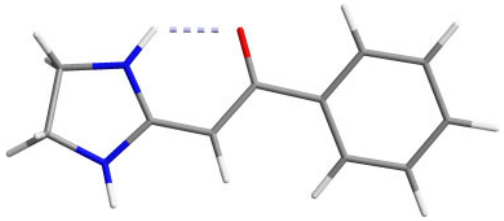
LUMO

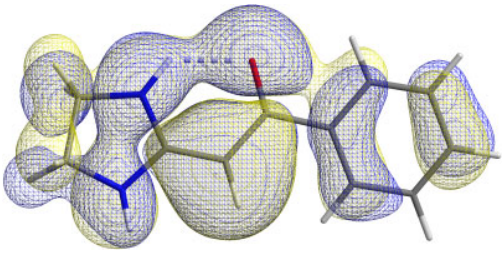
Atoms	*x*	*y*	*z*
C	2.933152	0.281679	0.000001
C	2.733864	-1.123411	0.000032
C	1.462827	-1.651000	0.000033
C	0.331718	-0.807653	0.000003
C	0.534168	0.605886	-0.000028
C	1.841801	1.130459	-0.000029
N	-0.912629	-1.364340	0.000005
C	-1.951896	-0.591765	-0.000019
C	-1.890412	0.842300	-0.000054
C	-0.631446	1.404726	-0.000050
Cl	-3.508107	-1.403755	-0.000027
C	-3.038201	1.790147	-0.000100
H	-2.695605	2.849451	0.000152
O	-4.212303	1.524393	0.000170
O	4.157312	0.871905	-0.000002
H	3.583031	-1.793275	0.000054
H	1.298992	-2.721285	0.000056
H	2.009263	2.201056	-0.000053
H	-0.538028	2.487449	-0.000073
C	5.323622	0.055338	0.000022
H	6.163886	0.746883	0.000011
H	5.374721	-0.573361	-0.895221
H	5.374713	-0.573321	0.895293

Table S3. Cartesian coordinates, optimized geometry, and energies of compound **2a**

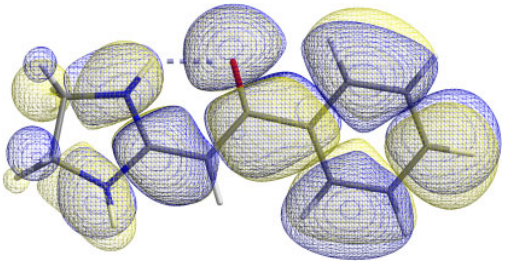
2-(Imidazolidin-2-ylidene)-1-phenylethan-1-one (2a)

E_{B3LYP}	E_0	H	$TCGFE$
-611.284314	0.213689	0.226396	0.174478





HOMO



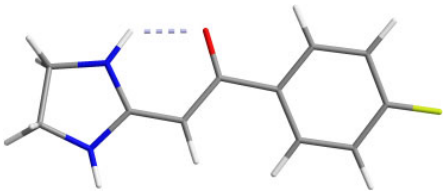
LUMO

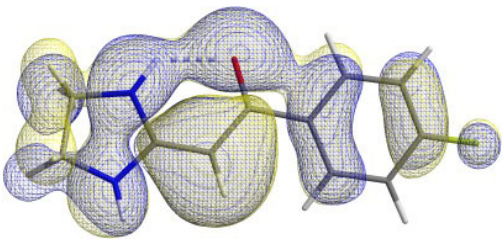
Atoms	x	y	z
N	2.618914	-0.863830	-0.003120
N	3.076600	1.304268	0.000268
O	0.079173	-1.625601	-0.131799
C	4.055546	-0.791075	-0.232127
H	4.302360	-0.902564	-1.296127
H	4.594788	-1.552404	0.333472
C	4.358630	0.636727	0.258746
H	4.607654	0.635158	1.328041
H	5.171860	1.108684	-0.293977
C	2.061360	0.367757	0.025840
C	0.704892	0.638280	0.049013
H	0.395160	1.669273	0.126689
C	-0.249417	-0.424613	-0.042604
C	-1.722323	-0.096282	-0.017424
C	-2.236780	1.197445	-0.168090
H	-1.571459	2.037374	-0.325237
C	-3.610594	1.423445	-0.142925
H	-3.992977	2.430695	-0.267358
C	-4.492172	0.359705	0.037145
C	-3.991291	-0.933476	0.183080
H	-4.671976	-1.766741	0.319831
C	-2.619919	-1.158082	0.150708
H	-2.208809	-2.154691	0.250121
H	1.988928	-1.642262	-0.188627
H	2.896795	2.203258	0.420051
H	-5.561895	0.536969	0.059299

Table S4. Cartesian coordinates, optimized geometry, and energies of compound **2b**

1-(4-Fluorophenyl)-2-(imidazolidin-2-ylidene)ethan-1-one (2b)

E_{B3LYP}	E_0	H	$TCGFE$
-611.284314	0.213689	0.226396	0.174478





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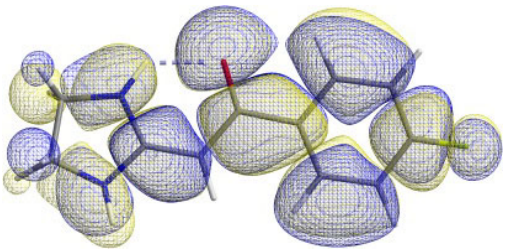


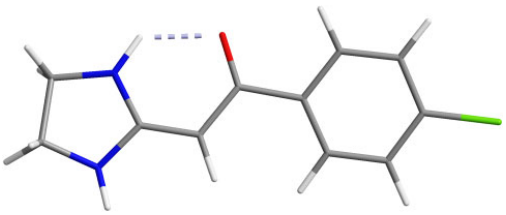
LUMO

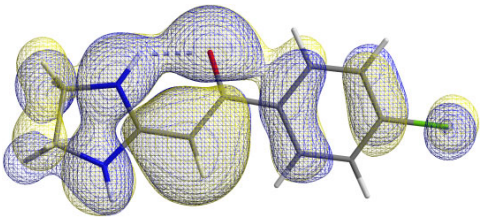
Atoms	*x*	*y*	*z*
N	-3.069639	0.835232	0.022340
N	-3.479147	-1.341230	-0.036419
O	-0.546036	1.654039	-0.089054
C	-4.504889	0.737614	-0.208310
H	-4.754495	0.875529	-1.268435
H	-5.060021	1.469679	0.379954
C	-4.776172	-0.710633	0.239553
H	-5.026027	-0.746908	1.307956
H	-5.577909	-1.184093	-0.328353
C	-2.485498	-0.383868	0.016057
C	-1.123034	-0.626333	0.031729
H	-0.794206	-1.653421	0.074401
C	-0.192013	0.458222	-0.030522
C	1.287167	0.164358	-0.016130
C	1.834750	-1.119326	-0.132965
H	1.193305	-1.983171	-0.250605
C	3.212260	-1.31689	-0.119478
H	3.647491	-2.304012	-0.214056
C	4.040964	-0.213588	0.014862
C	3.537724	1.074106	0.129102
H	4.220815	1.908637	0.229483
C	2.160458	1.252915	0.108835
H	1.725792	2.241130	0.186271
H	-2.457955	1.632471	-0.142428
H	-3.279533	-2.248675	0.355251
F	5.380247	-0.399662	0.031682

Table S5. Cartesian coordinates, optimized geometry, and energies of compound **2c**

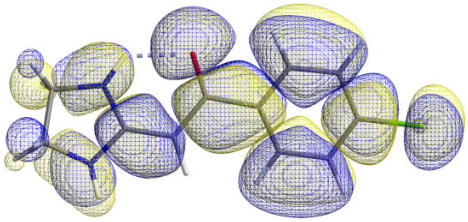
1-(4-Chlorophenyl)-2-(imidazolidin-2-ylidene)ethan-1-one (2c)

E_{B3LYP}	E_0	H	$TCGFE$
-1070.9071328	0.204050	0.218019	0.162612





HOMO



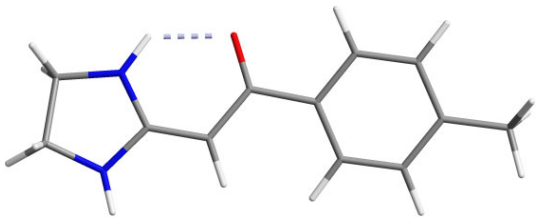
LUMO

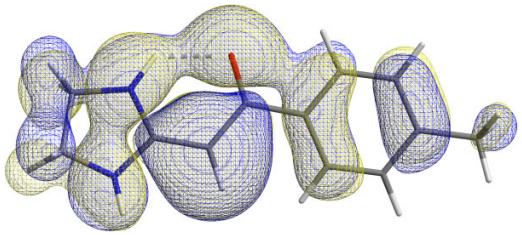
Atoms	x	y	z
Cl	5.369971	-0.326450	0.033232
N	-3.517365	0.807651	0.011582
N	-3.885576	-1.375998	-0.013283
O	-1.007834	1.673740	-0.111910
C	-4.951914	0.680517	-0.210320
H	-5.208677	0.801314	-1.270712
H	-5.517406	1.409341	0.371964
C	-5.194318	-0.767137	0.256406
H	-5.441761	-0.795121	1.325528
H	-5.987299	-1.263316	-0.304259
C	-2.910971	-0.399837	0.021930
C	-1.543262	-0.616014	0.037534
H	-1.193638	-1.635394	0.097461
C	-0.634574	0.484777	-0.040620
C	0.850737	0.217093	-0.025479
C	1.421805	-1.052858	-0.165735
H	0.797230	-1.926006	-0.305229
C	2.802512	-1.226333	-0.151321
H	3.238918	-2.210155	-0.265902
C	3.621658	-0.114511	0.008534
C	3.084493	1.161616	0.145036
H	3.738471	2.015852	0.264957
C	1.704355	1.316829	0.122321
H	1.254792	2.297213	0.215288
H	-2.922403	1.615223	-0.162824
H	-3.667615	-2.275646	0.386264

Table S6. Cartesian coordinates, optimized geometry, and energies of compound **2d**

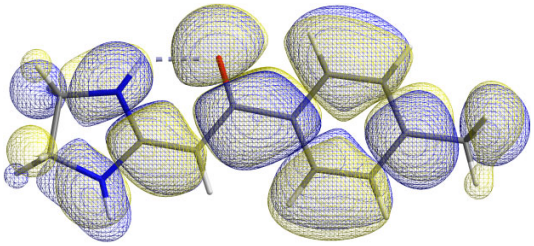
2-(Imidazolidin-2-ylidene)-1-(*p*-tolyl)ethanone (2d)

E_{B3LYP}	E_0	H	$TCGFE$
-650.6121139	0.240754	0.255415	0.197849





HOMO



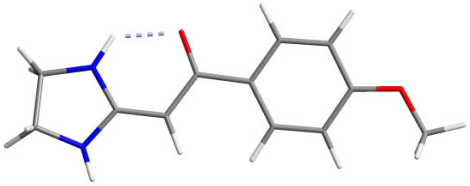
LUMO

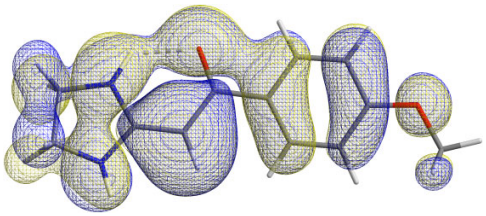
Atoms	*x*	*y*	*z*
N	3.104584	0.829310	-0.025829
N	3.506576	-1.349466	0.040972
O	0.585390	1.657436	0.089837
C	4.538740	0.726262	0.206457
H	4.788907	0.863106	1.266791
H	5.097765	1.455667	-0.381630
C	4.804269	-0.723187	-0.240581
H	5.050626	-0.760577	-1.309921
H	5.606917	-1.198282	0.324871
C	2.514948	-0.388176	-0.014275
C	1.152821	-0.625227	-0.027990
H	0.818963	-1.650907	-0.066037
C	0.224129	0.463578	0.032349
C	-1.255014	0.174236	0.017443
C	-1.809049	-1.105237	0.129955
H	-1.171589	-1.973228	0.244648
C	-3.188888	-1.290048	0.111368
H	-3.593414	-2.293349	0.202127
C	-4.062024	-0.207392	-0.021204
C	-3.504851	1.072738	-0.132036
H	-4.160365	1.931961	-0.238488
C	-2.130311	1.260837	-0.108700
H	-1.697627	2.250204	-0.187680
H	2.495015	1.628023	0.140797
H	3.303110	-2.255120	-0.352991
C	-5.558419	-0.402738	-0.021875
H	-5.825501	-1.446378	-0.200934
H	-5.993482	-0.110524	0.940184
H	-6.038674	0.207128	-0.791911

Table S7. Cartesian coordinates, optimized geometry, and energies of compound **2e**

2-(Imidazolidin-2-ylidene)-1-(4-methoxyphenyl)ethan-1-one (2e)

E_{B3LYP}	E_0	H	$TCGFE$
-725.8385691	0.245850	0.261233	0.202148





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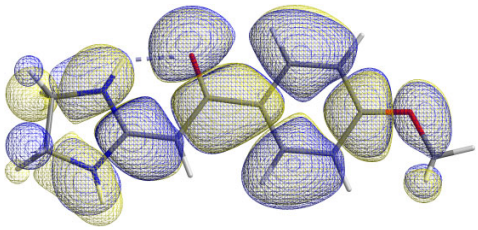


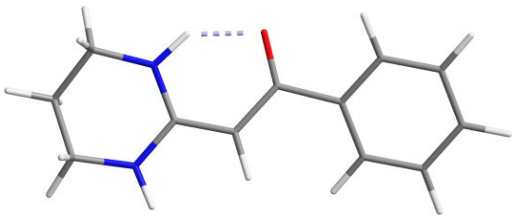
LUMO

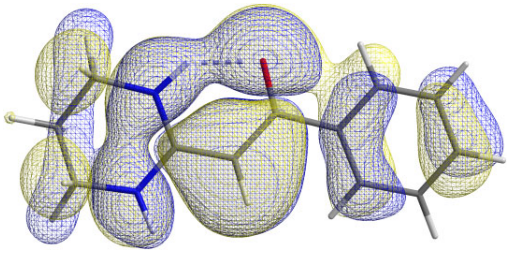
Atoms	x	y	z
N	3.555670	0.743621	-0.038775
N	3.833961	-1.453872	0.064677
O	1.085975	1.711778	0.063010
C	4.980223	0.562780	0.202995
H	5.233364	0.699042	1.262819
H	5.582674	1.251255	-0.391613
C	5.164857	-0.905066	-0.223471
H	5.409715	-0.970338	-1.291896
H	5.939338	-1.416468	0.349624
C	2.897141	-0.438688	-0.009093
C	1.524995	-0.599994	-0.022733
H	1.136881	-1.607103	-0.040722
C	0.656125	0.539883	0.020259
C	-0.833622	0.336323	0.008574
C	-1.462722	-0.907600	0.091851
H	-0.877878	-1.814630	0.179530
C	-2.851978	-1.026573	0.079919
H	-3.299797	-2.008711	0.150934
C	-3.643019	0.121129	-0.019517
C	-3.027208	1.377733	-0.099349
H	-3.657434	2.255975	-0.173496
C	-1.648847	1.476893	-0.082206
H	-1.160167	2.441283	-0.137648
H	2.992702	1.577493	0.118756
H	3.581171	-2.350647	-0.321041
O	-5.005209	0.124436	-0.044324
C	-5.688823	-1.118245	0.025579
H	-6.749686	-0.876695	-0.017309
H	-5.431425	-1.766893	-0.819308
H	-5.475737	-1.642967	0.963712

Table S8. Cartesian coordinates, optimized geometry, and energies of compound **2f**

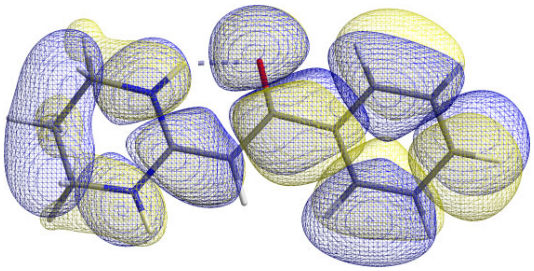
1-Phenyl-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethan-1-one (2f)

E_{B3LYP}	E_0	H	$TCGFE$
-650.6134471	0.242711	0.256594	0.202164





HOMO



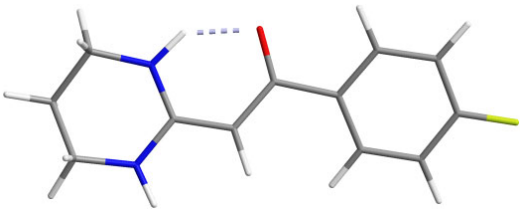
LUMO

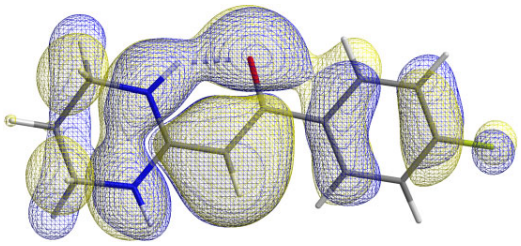
Atoms	*x*	*y*	*z*
N	2.224022	-0.901758	-0.093815
C	3.630810	-1.257458	-0.004276
C	4.505479	-0.066634	-0.393063
C	4.054609	1.169027	0.384120
N	2.629785	1.376915	0.162374
C	1.736812	0.344691	0.054939
C	0.358557	0.591908	0.061464
C	-0.599409	-0.452253	-0.056376
C	-2.067284	-0.103449	-0.029393
C	-2.977217	-1.137350	0.222853
C	-4.344765	-0.889515	0.260207
C	-4.829497	0.397740	0.031421
C	-3.935444	1.432074	-0.237601
C	-2.565549	1.183655	-0.265927
O	-0.295257	-1.665093	-0.157988
H	0.039733	1.615399	0.194556
H	1.477185	-1.605760	-0.147853
H	3.815400	-2.103727	-0.670510
H	3.882204	-1.587070	1.013957
H	4.410168	0.127032	-1.465205
H	5.554777	-0.283576	-0.179508
H	4.590265	2.057286	0.040953
H	4.279556	1.039654	1.453215
H	2.238301	2.272040	0.404176
H	-2.579898	-2.132052	0.380762
H	-5.035141	-1.700828	0.464304
H	-5.896143	0.592427	0.055851
H	-4.305463	2.433091	-0.431250
H	-1.888156	1.996117	-0.499662

Table S9. Cartesian coordinates, optimized geometry, and energies of compound **2g**

1-(4-Fluorophenyl)-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethan-1-one (2g)

E_{B3LYP}	E_0	H	$TCGFE$
-749.87872	0.234466	0.249209	0.192619





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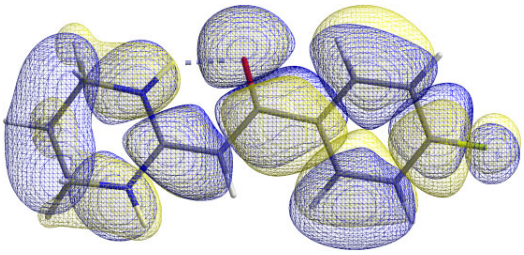


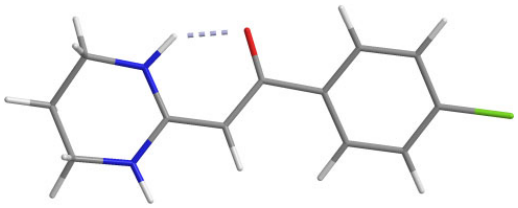
LUMO

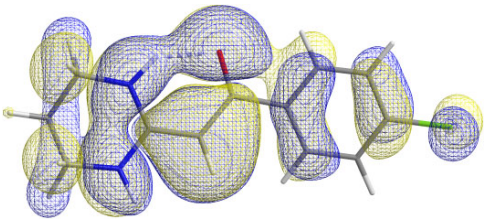
Atoms	x	y	z
N	-2.668807	0.883942	-0.078816
C	-4.083808	1.207634	0.007918
C	-4.454682	-1.231769	0.368999
N	-3.024459	-1.405080	0.151720
C	-2.154902	-0.353000	0.054956
C	-0.770832	-0.570764	0.058146
C	0.163556	0.494728	-0.048590
C	1.638461	0.180716	-0.029749
C	2.524412	1.241830	0.196591
C	3.898787	1.039984	0.225174
C	4.385758	-0.241006	0.010965
C	3.544039	-1.315463	-0.232199
C	2.169677	-1.096090	-0.249623
O	-0.166200	1.702149	-0.135939
H	-0.431795	-1.589232	0.179137
F	5.722638	-0.448624	0.033067
C	-4.930088	0.001778	-0.396722
H	-1.938406	1.605003	-0.126459
H	-4.283961	2.056240	-0.650725
H	-4.346083	1.521448	1.028254
H	-4.968456	-2.128101	0.013745
H	-4.686482	-1.119553	1.438422
H	-2.614388	-2.295476	0.379602
H	2.103337	2.228168	0.342912
H	4.591857	1.852521	0.405859
H	3.967150	-2.296547	-0.409308
H	1.515277	-1.932262	-0.461721
H	-4.826935	-0.178209	-1.470511
H	-5.984678	0.192917	-0.184829

Table S10. Cartesian coordinates, optimized geometry, and energies of compound **2h**

1-(4-Chlorophenyl)-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethan-1-one (2h)

E_{B3LYP}	E_0	H	$TCGFE$
-1110.2364377	0.233066	0.248225	0.190276





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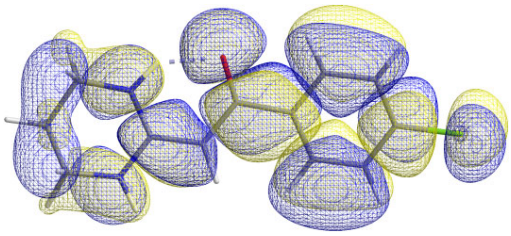


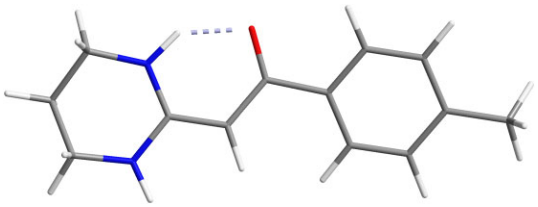
LUMO

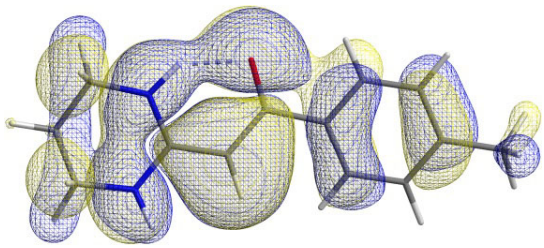
Atoms	*x*	*y*	*z*
N	-3.111440	0.864242	-0.077530
C	-4.532862	1.161176	0.004174
C	-4.857511	-1.283224	0.378076
N	-3.423222	-1.429106	0.167257
C	-2.574874	-0.361946	0.060323
C	-1.185946	-0.553367	0.059426
C	-0.274118	0.529328	-0.054631
C	1.207132	0.242261	-0.036907
C	2.073733	1.314814	0.203206
C	3.451445	1.137061	0.231888
C	3.972343	-0.132301	0.002757
C	3.140068	-1.215383	-0.255986
C	1.762130	-1.020602	-0.272304
O	-0.624263	1.730462	-0.146887
H	-0.826099	-1.564098	0.185176
Cl	5.718052	-0.371814	0.031397
C	-5.354639	-0.063166	-0.395458
H	-2.395161	1.598755	-0.133047
H	-4.747479	2.001949	-0.659837
H	-4.802851	1.475794	1.022088
H	-5.351889	-2.191392	0.025505
H	-5.094842	-1.170717	1.446098
H	-2.996365	-2.312222	0.392339
H	1.637615	2.293057	0.360639
H	4.115742	1.969508	0.425735
H	3.564503	-2.192533	-0.447308
H	1.124331	-1.866566	-0.496645
H	-5.245924	-0.246937	-1.468051
H	-6.413139	0.108679	-0.186820

Table S11. Cartesian coordinates, optimized geometry, and energies of compound **2i**

2-(Tetrahydropyrimidin-2(1*H*)-ylidene)-1-(*p*-tolyl)ethan-1-one (2i)

E_{B3LYP}	E_0	H	$TCGFE$
-689.9411536	0.269816	0.285615	0.226211





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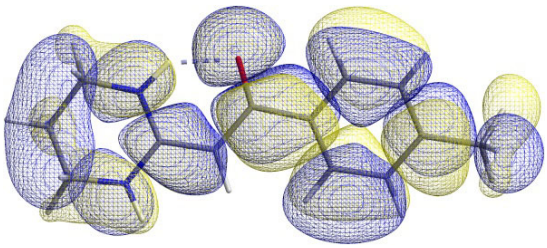


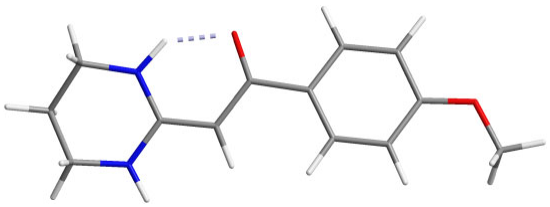
LUMO

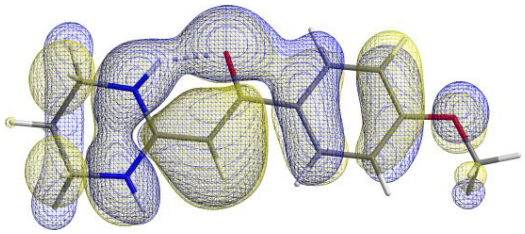
Atoms	*x*	*y*	*z*
N	-2.701907	0.877971	-0.086792
C	-4.116575	1.196479	0.015523
C	-4.473835	-1.243581	0.383864
N	-3.046819	-1.413921	0.145263
C	-2.181018	-0.356855	0.047621
C	-0.797357	-0.567508	0.048194
C	0.133449	0.503227	-0.058940
C	1.608351	0.194485	-0.038547
C	2.495766	1.251744	0.198106
C	3.867438	1.040799	0.226122
C	4.408528	-0.232054	0.007482
C	3.523138	-1.282557	-0.247749
C	2.146427	-1.075976	-0.268356
O	-0.204731	1.708456	-0.146167
H	-0.451586	-1.584070	0.166637
C	-4.963222	-0.012951	-0.377698
C	5.899521	-0.460255	0.061607
H	-1.973510	1.601641	-0.134449
H	-4.327782	2.043504	-0.641866
H	-4.369495	1.510689	1.038361
H	-4.990882	-2.141934	0.038370
H	-4.691058	-1.128809	1.456338
H	-2.630717	-2.299204	0.382426
H	2.076633	2.238519	0.349989
H	4.533332	1.877400	0.415921
H	3.916128	-2.276163	-0.440021
H	1.496142	-1.912761	-0.493777
H	-4.871859	-0.194187	-1.452362
H	-6.016029	0.174551	-0.153586
H	6.444591	0.345839	-0.436933
H	6.254307	-0.498534	1.097576
H	6.177041	-1.402467	-0.415954

Table S12. Cartesian coordinates, optimized geometry, and energies of compound **2j**

1-(4-Methoxyphenyl)-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethan-1-one (2j)

E_{B3LYP}	E_0	H	$TCGFE$
-765.1674207	0.274930	0.291458	0.230841





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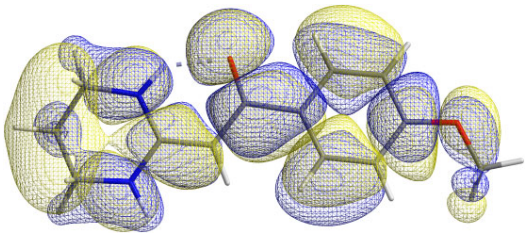


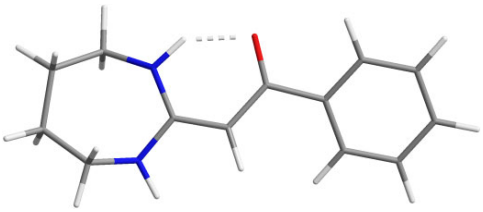
LUMO

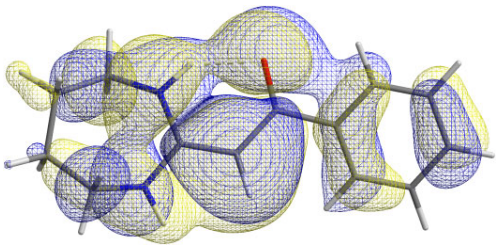
Atoms	x	y	z
N	-3.146980	0.820326	0.110758
C	-4.573314	1.027780	0.296051
C	-4.804926	-1.444335	0.071399
N	-3.371451	-1.479599	-0.186195
C	-2.561379	-0.383915	-0.036183
C	-1.169815	-0.517878	-0.079738
C	-0.294056	0.597810	0.050099
C	1.191936	0.369725	0.018029
C	2.025059	1.484566	-0.169268
C	3.401191	1.355894	-0.208450
C	3.995082	0.096973	-0.046844
C	3.185720	-1.023554	0.157786
C	1.799166	-0.876113	0.185954
O	-0.696847	1.779627	0.178877
H	-0.774332	-1.506496	-0.263174
C	-5.362352	-0.094374	-0.376661
O	5.357390	0.070985	-0.099260
C	6.017996	-1.176201	0.052865
H	-2.457703	1.577198	0.201492
H	-4.838726	1.996495	-0.134035
H	-4.824801	1.069270	1.365769
H	-5.275756	-2.259043	-0.484199
H	-5.025586	-1.601560	1.137917
H	-2.908428	-2.372115	-0.140232
H	1.554415	2.453272	-0.279605
H	4.045977	2.213264	-0.360849
H	3.617232	-2.005113	0.301584
H	1.195603	-1.757115	0.367504
H	-5.271539	-0.008429	-1.463170
H	-6.421227	-0.020650	-0.117445
H	7.082196	-0.961657	-0.032175
H	5.817170	-1.621772	1.033800
H	5.728335	-1.883796	-0.732303

Table S13. Cartesian coordinates, optimized geometry, and energies of compound **2k**

2-(1,3-Diazepan-2-ylidene)-1-phenylethan-1-one (2k)

E_{B3LYP}	E_0	H	$TCGFE$
-689.9353596	0.272233	0.286965	0.230764





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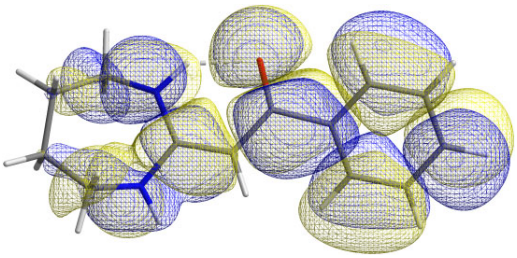


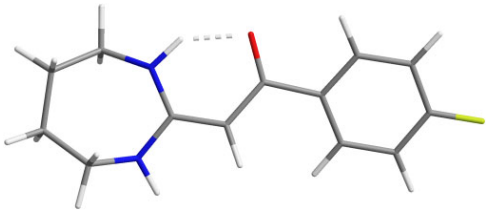
LUMO

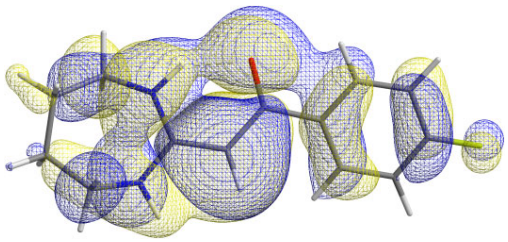
Atoms	x	y	z
O	0.646990	-1.690792	-0.021446
N	-1.908006	-0.912592	-0.009322
H	-1.150877	-1.603395	-0.050919
N	-2.281047	1.393803	0.114218
H	-1.806303	2.283037	0.153961
C	3.331747	-1.110596	-0.351495
H	2.948265	-2.088847	-0.612692
C	4.696402	-0.846197	-0.349114
H	5.398195	-1.625619	-0.625181
C	5.163606	0.417617	0.010843
C	4.256189	1.411808	0.370790
H	4.613584	2.392151	0.666421
C	2.888981	1.147956	0.359822
H	2.198310	1.924597	0.665293
C	2.408874	-0.115145	-0.007507
C	0.945531	-0.477260	-0.023592
C	-0.024475	0.573687	-0.070633
H	0.300915	1.595506	-0.205176
C	-1.395671	0.340890	0.004732
C	-3.542230	1.436640	-0.643013
H	-3.376337	1.107896	-1.677998
H	-3.840149	2.486651	-0.680579
C	-4.673390	0.619015	-0.016227
H	-5.597840	0.846519	-0.557966
H	-4.815622	0.955728	1.016791
C	-4.425832	-0.890189	-0.041621
H	-5.267331	-1.401556	0.437844
H	-4.381041	-1.246827	-1.076614
C	-3.138620	-1.306471	0.676037
H	-3.105188	-2.394697	0.753349
H	-3.145963	-0.908411	1.699857
H	6.228153	0.624604	0.016763

Table S14. Cartesian coordinates, optimized geometry, and energies of compound **2l**

2-(1,3-Diazepan-2-ylidene)-1-(4-fluorophenyl)ethan-1-one (2l)

E_{B3LYP}	E_0	H	$TCGFE$
-789.200519	0.263973	0.279561	0.221221





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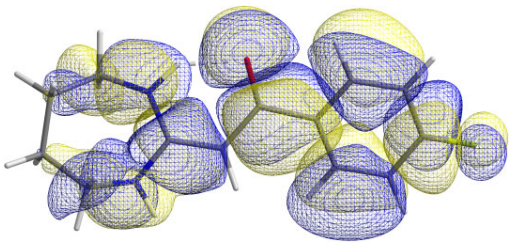


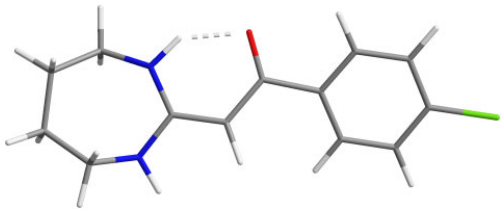
LUMO

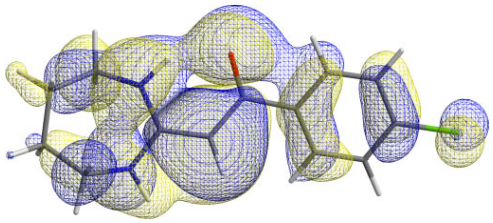
Atoms	x	y	z
O	-0.192594	1.732344	-0.037325
N	2.345810	0.901777	-0.016686
H	1.603898	1.608071	-0.066007
N	2.670593	-1.410216	0.125255
H	2.178460	-2.289456	0.173793
C	-2.885201	1.222952	-0.332432
H	-2.478015	2.195806	-0.576110
C	-4.256843	1.004161	-0.328938
H	-4.961309	1.787623	-0.579871
C	-4.726454	-0.256731	0.009848
C	-3.870693	-1.294629	0.345677
H	-4.281340	-2.258725	0.618922
C	-2.499048	-1.059713	0.329819
H	-1.831216	-1.862898	0.614807
C	-1.985924	0.196970	-0.014264
C	-0.515744	0.524958	-0.031507
C	0.431685	-0.545773	-0.069199
H	0.087370	-1.562134	-0.197714
C	1.807994	-0.340294	0.006580
C	3.932953	-1.485477	-0.627782
H	3.776146	-1.161560	-1.665631
H	4.208641	-2.541790	-0.656169
C	5.079278	-0.686542	-0.004719
H	6.000118	-0.938197	-0.541836
H	5.211706	-1.016892	1.031618
C	4.863430	0.827184	-0.044441
H	5.714237	1.325265	0.432479
H	4.828649	1.175237	-1.082753
C	3.583824	1.276812	0.666608
H	3.572468	2.366170	0.733200
H	3.580954	0.888730	1.694258
F	-6.060043	-0.479799	0.019348

Table S15. Cartesian coordinates, optimized geometry, and energies of compound **2m**

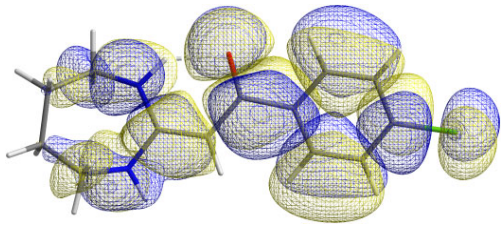
1-(4-Chlorophenyl)-2-(1,3-diazepan-2-ylidene)ethan-1-one (2m)

E_{B3LYP}	E_0	H	$TCGFE$
-1149.5580754	0.262570	0.278572	0.218892





HOMO



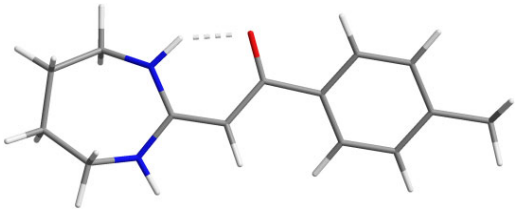
LUMO

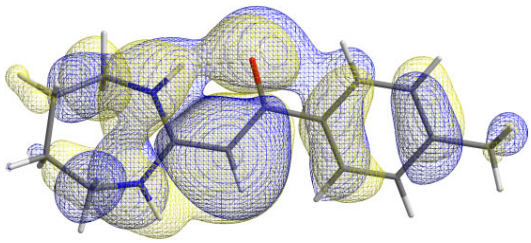
Atoms	x	y	z
O	0.259490	1.766846	-0.029028
N	2.782812	0.889682	-0.013113
H	2.053863	1.609390	-0.060742
N	3.065906	-1.428011	0.124014
H	2.557873	-2.298095	0.173453
C	-2.440055	1.303043	-0.335268
H	-2.017043	2.268911	-0.580491
C	-3.815483	1.108782	-0.336416
H	-4.490928	1.914235	-0.594807
C	-4.319964	-0.142607	0.003793
C	-3.473770	-1.191305	0.346054
H	-3.886001	-2.153029	0.622950
C	-2.098097	-0.981348	0.334381
H	-1.447731	-1.797067	0.624613
C	-1.560347	0.263422	-0.011617
C	-0.083149	0.564961	-0.025612
C	0.842915	-0.522629	-0.064149
H	0.479518	-1.532266	-0.192932
C	2.223893	-0.342318	0.009137
C	4.326590	-1.526526	-0.629349
H	4.174714	-1.200076	-1.667081
H	4.582537	-2.587740	-0.657529
C	5.487660	-0.748342	-0.007742
H	6.403204	-1.017482	-0.545349
H	5.614950	-1.079750	1.028872
C	5.299398	0.768995	-0.049498
H	6.160245	1.252314	0.424472
H	5.268491	1.115946	-1.088308
C	4.030442	1.243183	0.664564
H	4.038950	2.332637	0.729138
H	4.023634	0.857058	1.692894
Cl	-6.061966	-0.402077	0.009242

Table S16. Cartesian coordinates, optimized geometry, and energies of compound **2n**

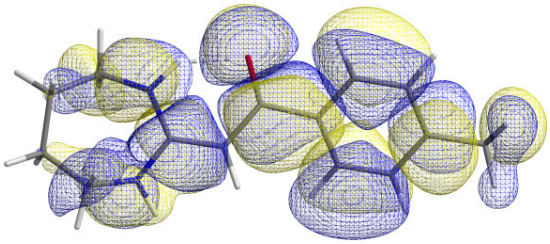
2-(1,3-Diazepan-2-ylidene)-1-(*p*-tolyl)ethan-1-one (2n)

E_{B3LYP}	E_0	H	$TCGFE$
-729.2631458	0.299254	0.315943	0.254335





HOMO



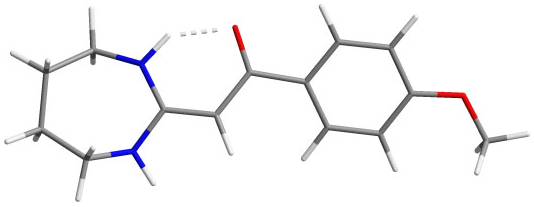
LUMO

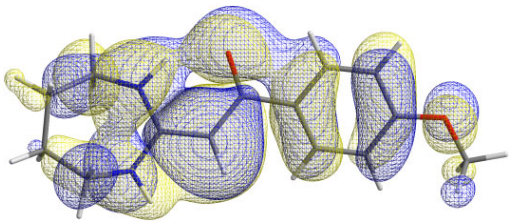
Atoms	x	y	z
O	0.156929	-1.738712	-0.039091
N	-2.377078	-0.899008	-0.018170
H	-1.636378	-1.607178	-0.066168
N	-2.693720	1.414285	0.128554
H	-2.198058	2.291652	0.175656
C	2.858782	-1.232676	-0.336577
H	2.453820	-2.205846	-0.584427
C	4.227987	-1.004163	-0.333851
H	4.905514	-1.810937	-0.596542
C	4.751272	0.250203	0.002580
C	3.851080	1.267182	0.333298
H	4.230325	2.247222	0.605957
C	2.477381	1.043988	0.323701
H	1.813249	1.851887	0.606437
C	1.957706	-0.209783	-0.016473
C	0.487525	-0.533189	-0.033342
C	-0.457736	0.541699	-0.070063
H	-0.108647	1.556817	-0.195533
C	-1.833316	0.341487	0.006923
C	-3.953799	1.493117	-0.627381
H	-3.796918	1.166662	-1.664506
H	-4.226156	2.550319	-0.658210
C	-5.103902	0.699440	-0.004131
H	-6.023532	0.952962	-0.542613
H	-5.235929	1.032731	1.031347
C	-4.893544	-0.815291	-0.039729
H	-5.745649	-1.308800	0.439821
H	-4.861581	-1.166180	-1.077203
C	-3.613893	-1.268019	0.669742
H	-3.606918	-2.357356	0.738618
H	-3.607749	-0.877931	1.696726
C	6.241571	0.485345	0.037609
H	6.482526	1.543870	-0.083138
H	6.668043	0.158267	0.992545
H	6.751506	-0.071446	-0.752564

Table S17. Cartesian coordinates, optimized geometry, and energies of compound **2o**

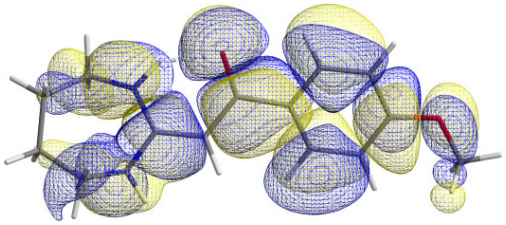
2-(1,3-Diazepan-2-ylidene)-1-(4-methoxyphenyl)ethan-1-one (2o)

E_{B3LYP}	E_0	H	$TCGFE$
-804.4896206	0.304396	0.321783	0.259399





HOMO



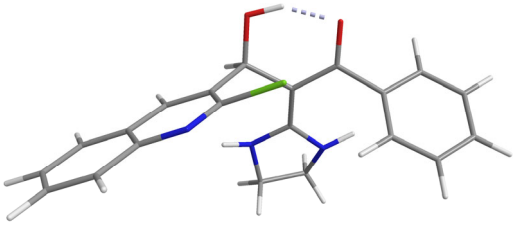
LUMO

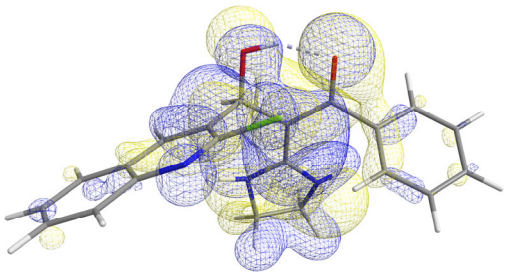
Atoms	x	y	z
O	0.326570	1.826004	-0.025772
N	2.815571	0.865792	0.000473
H	2.109179	1.608751	-0.040624
N	3.020719	-1.461814	0.112840
H	2.483704	-2.315281	0.142011
C	-2.395715	1.470804	-0.326365
H	-1.940379	2.430664	-0.534876
C	-3.769697	1.321140	-0.336412
H	-4.428391	2.151141	-0.562539
C	-4.343853	0.075279	-0.047632
C	-3.516887	-1.010297	0.253753
H	-3.933549	-1.978826	0.495647
C	-2.132576	-0.843094	0.251607
H	-1.513980	-1.693723	0.510631
C	-1.545575	0.389372	-0.042074
C	-0.064635	0.638276	-0.042248
C	0.830019	-0.480774	-0.089212
H	0.435580	-1.476238	-0.236786
C	2.212005	-0.347455	0.002024
C	4.279114	-1.587357	-0.639467
H	4.143826	-1.233666	-1.670660
H	4.500708	-2.655667	-0.689610
C	5.462364	-0.861861	0.004590
H	6.372179	-1.148766	-0.533901
H	5.572016	-1.220663	1.034142
C	5.324963	0.661728	-0.002947
H	6.195741	1.104448	0.492451
H	5.318757	1.033290	-1.033710
C	4.062222	1.162704	0.704997
H	4.107251	2.249757	0.793443
H	4.028864	0.755425	1.724793
O	-5.704856	0.026925	-0.081328
C	-6.347829	-1.207214	0.202151
H	-7.415525	-1.013607	0.112224
H	-6.059720	-1.984501	-0.514499
H	-6.127517	-1.551325	1.218871

Table S18. Cartesian coordinates, optimized geometry, and energies of **Int 2a**

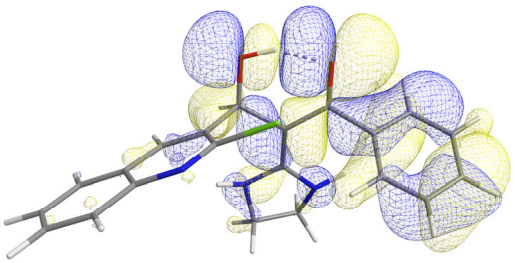
3-(2-Chloro-6-methylquinolin-3-yl)-3-hydroxy-2-(imidazolidin-2-ylidene)-1-(*p*-tolyl) propan-1-one (Int 2a)

E_{B3LYP}	E_0	H	$TCGFE$
-1586.2693512	0.352743	0.376069	0.299358

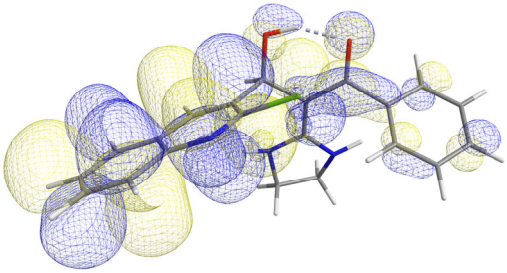




HOMO

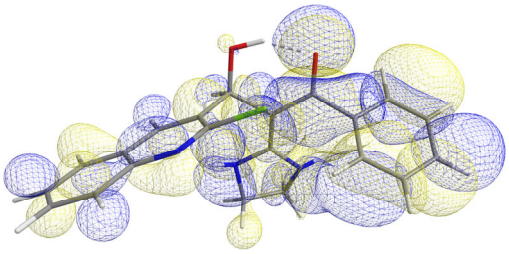


HOMO-1



LUMO

Atoms	x	y	z
C	-6.060117	0.772791	-0.435400
C	-6.016799	-0.294117	-1.362350
C	-4.884931	-1.064833	-1.491286
C	-3.747967	-0.793215	-0.694057
C	-3.785819	0.285565	0.233768
C	-4.966744	1.058202	0.348772
N	-2.642346	-1.581372	-0.823468
C	-1.483736	-0.257703	0.848388
C	-2.612828	0.515150	0.995814
C	1.126535	1.559662	0.354447
C	1.038020	0.312296	0.958755
N	2.201534	2.159313	-0.255826
C	2.163284	-0.596620	1.045500
C	3.298571	-0.554899	0.050744
C	4.611803	-0.645211	0.527633
C	5.682000	-0.696370	-0.360994
C	5.447593	-0.697298	-1.735741
C	4.140682	-0.641183	-2.217552
C	3.070899	-0.560088	-1.330098
O	2.252256	-1.475395	1.918225
C	1.960747	3.587829	-0.448948
C	0.430711	3.616149	-0.517579
H	-6.963390	1.365034	-0.343779
H	-6.887844	-0.506180	-1.971894
H	-4.828924	-1.891999	-2.188063
H	-4.997739	1.873926	1.063714
H	-2.612610	1.315752	1.731998
H	4.777191	-0.689996	1.597526
H	6.696448	-0.753552	0.017535
H	6.279812	-0.753920	-2.428578
H	3.953940	-0.666132	-3.285332
H	2.054410	-0.521375	-1.702223
H	2.433326	3.951194	-1.362373



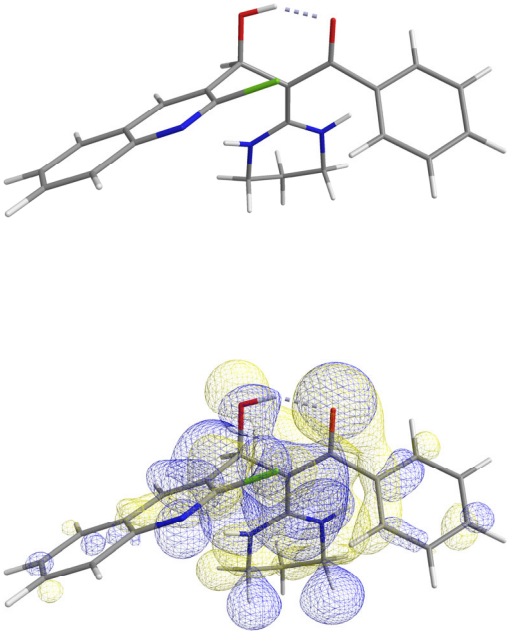
LUMO+1

H	2.326406	4.179206	0.400708
H	0.005758	4.541736	-0.127562
H	0.082353	3.469630	-1.547449
C	-0.262947	0.023764	1.740743
H	3.130971	1.817602	-0.070712
N	0.087746	2.470513	0.330940
O	-0.171756	-0.999842	2.701798
H	0.721357	-1.395733	2.620814
H	-0.520039	0.954891	2.269909
Cl	-0.250486	-2.443076	-0.303780
C	-1.601708	-1.328916	-0.094546
H	-0.849192	2.099205	0.272219

Table S19. Cartesian coordinates, optimized geometry, and energies of **Int 2b**

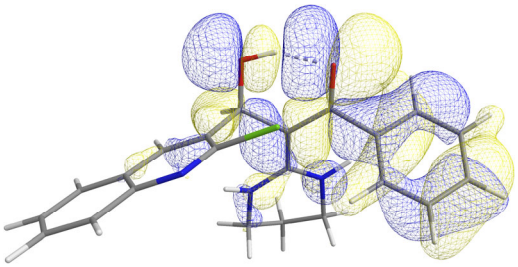
3-(2-Chloro-6-methylquinolin-3-yl)-3-hydroxy-2-(tetrahydropyrimidin-2(1*H*)-ylidene)-1-(*p*-tolyl)propan-1-one (Int 2b)

E_{B3LYP}	E_0	*H*	*TCGFE*
-1586.2693512	0.352743	0.376069	0.299358

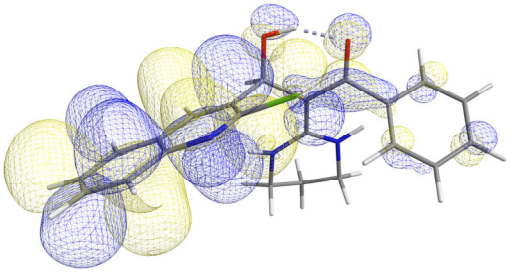


HOMO

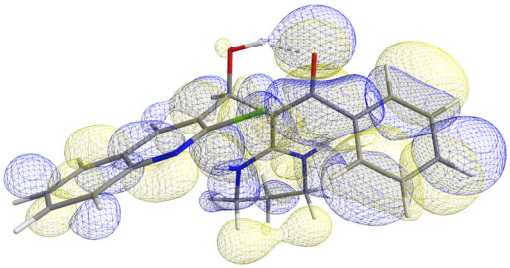
Atoms	*x*	*y*	*z*
C	-6.118268	0.783900	-0.440086
C	-6.100964	-0.281882	-1.369309
C	-4.987122	-1.077755	-1.502139
C	-3.842752	-0.833762	-0.706418
C	-5.017132	1.042753	0.342410
N	-2.755543	-1.645887	-0.839611
C	-1.563849	-0.355250	0.836540
C	-2.675579	0.443697	0.984427
C	0.989700	0.150517	1.002354
C	2.037045	-0.840477	1.049848
C	3.170221	-0.854168	0.045434
C	4.458475	-1.163568	0.497794
C	5.517078	-1.267008	-0.400422
C	5.293233	-1.098337	-1.766132
C	4.006537	-0.824637	-2.228807
C	2.951955	-0.696234	-1.328658
O	2.054320	-1.778454	1.869120
H	4.609686	-1.336225	1.556628
H	6.512923	-1.495360	-0.037176



HOMO-1



LUMO



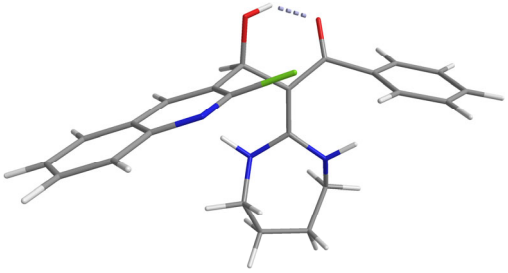
LUMO+1

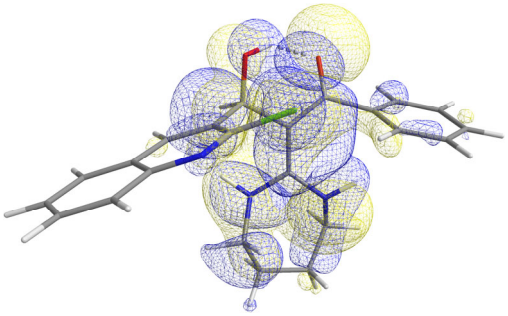
H	6.114126	-1.194770	-2.467982
H	3.822450	-0.723421	-3.292718
H	1.948412	-0.501034	-1.688117
C	-0.342187	-0.101156	1.743551
Cl	-0.384716	-2.563479	-0.328013
C	-1.707981	-1.420281	-0.110703
C	-3.854407	0.243382	0.223736
H	-7.007753	1.396236	-0.345502
H	-6.977870	-0.472958	-1.977419
H	-4.951207	-1.904550	-2.200695
H	-5.028077	1.857350	1.059250
H	-2.655113	1.244875	1.719502
O	-0.294464	-1.116345	2.716718
H	0.558510	-1.588677	2.598241
C	1.182834	1.433456	0.458668
N	0.113971	2.258725	0.244267
N	2.409256	1.981015	0.229582
C	0.175238	3.583577	-0.373826
C	2.611752	3.172764	-0.583253
C	1.556391	4.205001	-0.198939
H	-0.058985	3.506584	-1.444158
H	2.551993	2.943543	-1.657116
H	1.638967	5.097811	-0.823017
H	3.204271	1.388943	0.403217
H	-0.779404	1.794709	0.186189
H	-0.592899	4.213820	0.082329
H	3.617155	3.546356	-0.383267
H	1.708236	4.502574	0.842086
H	-0.589197	0.837559	2.263158

Table S20. Cartesian coordinates, optimized geometry, and energies of **Int 2c**

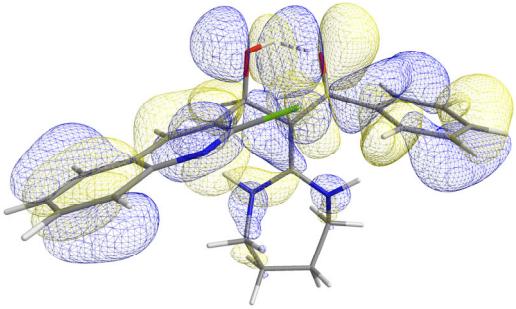
3-(2-Chloroquinolin-3-yl)-2-(1,3-diazepan-2-ylidene)-3-hydroxy-1-phenylpropan-1-one (Int 2c)

E_{B3LYP}	E_0	H	$TCGFE$
-1664.9124809	0.410608	0.436129	0.353866

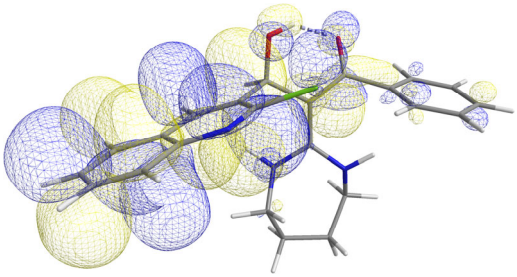




HOMO

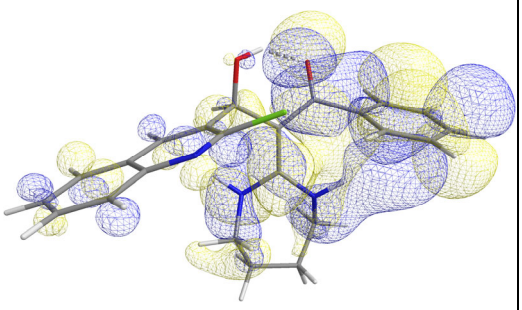


HOMO-1



LUMO

Atoms	x	y	z
C	-6.232741	0.431168	0.244242
C	-6.213802	-0.087673	-1.071308
C	-5.069711	-0.650761	-1.586234
C	-3.895004	-0.713895	-0.799483
C	-3.908744	-0.184370	0.521621
C	-5.103430	0.383677	1.027523
N	-2.777613	-1.296255	-1.319359
C	-1.557487	-0.811547	0.718628
C	-2.698562	-0.262292	1.253334
C	0.980435	-0.304526	0.930859
C	2.221449	-1.039709	1.110037
C	3.428067	-0.854080	0.221417
C	4.691427	-0.718064	0.809207
C	5.832121	-0.650964	0.016862
C	5.729523	-0.758184	-1.371014
C	4.480181	-0.928621	-1.962102
C	3.331703	-0.969636	-1.171323
O	2.357366	-1.897343	1.999406
H	-7.146136	0.865367	0.634359
H	-7.113574	-0.043310	-1.674354
H	-5.031637	-1.062183	-2.587278
H	-5.115657	0.777449	2.038603
H	-2.677542	0.113940	2.273184
H	4.764301	-0.678438	1.889292
H	6.804812	-0.529840	0.480530
H	6.621425	-0.720306	-1.986316
H	4.396486	-1.037995	-3.037554
H	2.358205	-1.108985	-1.627748
C	-0.295423	-0.911266	1.578255
O	-0.189516	-2.276827	1.907305
H	0.735257	-2.414742	2.193396
H	-0.519747	-0.338734	2.499876
Cl	-0.347815	-2.197149	-1.354797



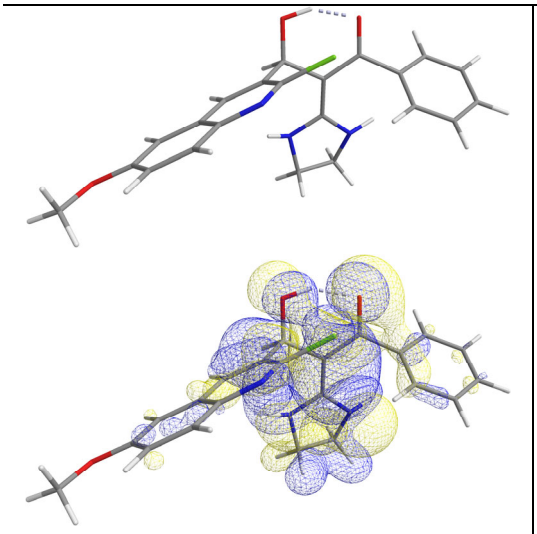
LUMO+1

C	-1.700973	-1.349040	-0.600316
C	0.941490	1.021451	0.488529
C	2.341379	3.031367	0.035587
C	-0.666702	2.842618	-0.113802
C	1.488388	4.105055	-0.646525
H	2.357191	3.200315	1.119135
H	3.371160	3.107513	-0.319496
C	0.066490	4.186846	-0.089295
H	-0.671843	2.437006	-1.131801
H	-1.707870	2.989150	0.180473
H	1.457625	3.893158	-1.720914
H	1.986135	5.073135	-0.525108
H	0.089559	4.553229	0.943209
H	-0.515352	4.908792	-0.672103
N	-0.111480	1.843897	0.811833
N	1.908357	1.655297	-0.250248
H	2.640047	1.053321	-0.590138
H	-0.788080	1.441142	1.434825

Table S21. Cartesian coordinates, optimized geometry, and energies of **Int 2d**

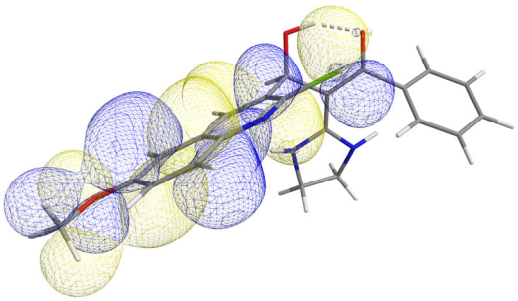
3-(2-Chloro-6-methoxyquinolin-3-yl)-3-hydroxy-2-(imidazolidin-2-ylidene)-1-phenyl propan-1-one (Int 2d)

E_{B3LYP}	E_0	H	$TCGFE$
-1700.8205993	0.384097	0.410489	0.326885

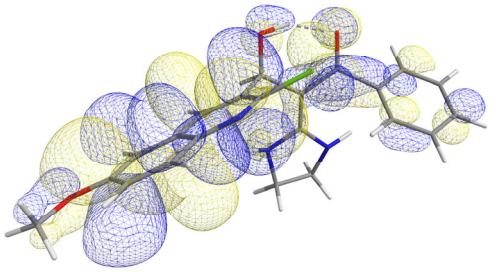


HOMO

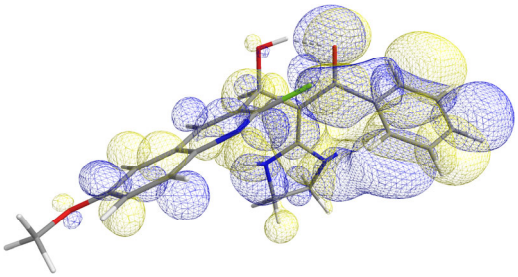
Atoms	x	y	z
C	-5.496968	0.326352	-0.215715
C	-5.413526	-0.713850	-1.175444
C	-4.236944	-1.408359	-1.345987
C	-3.096899	-1.098680	-0.574386
C	-3.181391	-0.049677	0.385687
C	-4.392619	0.648958	0.552271
N	-1.950962	-1.815428	-0.755463
C	-0.835001	-0.474407	0.928834
C	-2.002278	0.224846	1.128593
C	1.617119	1.545747	0.426124
C	1.638424	0.271984	0.979305
N	2.624001	2.242954	-0.197895
C	2.827955	-0.555864	0.987411



HOMO-1



LUMO



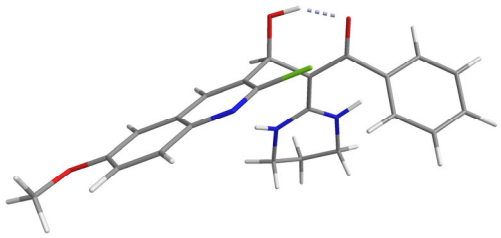
LUMO+1

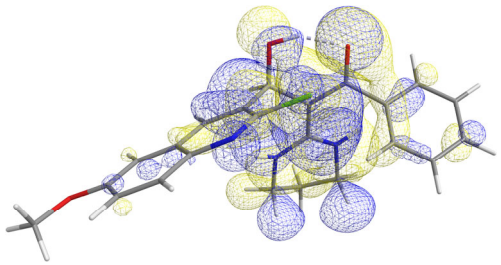
C	3.913468	-0.395246	-0.049913
C	5.249290	-0.401481	0.368836
C	6.280493	-0.341807	-0.564434
C	5.986921	-0.314533	-1.927516
C	4.659569	-0.341512	-2.352591
C	3.626650	-0.371150	-1.419442
O	3.016564	-1.455494	1.821949
C	2.279972	3.658749	-0.314704
C	0.750227	3.583774	-0.327070
H	-6.273583	-0.972925	-1.777917
H	-4.158274	-2.209694	-2.070227
H	-4.478561	1.443898	1.284002
H	-2.030594	0.999622	1.891161
H	5.464245	-0.469264	1.428675
H	7.311984	-0.334625	-0.230234
H	6.789942	-0.284914	-2.655599
H	4.428461	-0.344892	-3.411964
H	2.594720	-0.397409	-1.747657
H	2.690959	4.094578	-1.226212
H	2.637315	4.235576	0.548455
H	0.278747	4.460373	0.118689
H	0.372995	3.456530	-1.349347
C	0.391979	-0.140361	1.793626
H	3.580887	1.959047	-0.061012
N	0.519589	2.382890	0.481425
O	0.592381	-1.190122	2.708976
H	1.508389	-1.514659	2.583565
H	0.088454	0.748426	2.368767
Cl	0.503940	-2.543975	-0.323321
C	-0.908784	-1.522629	-0.046126
O	-6.624092	1.057390	0.011846
C	-7.800648	0.761017	-0.729516
H	-8.144008	-0.263358	-0.549079
H	-8.556078	1.459165	-0.373133
H	-7.649743	0.912238	-1.804026
H	-0.392527	1.951543	0.443855

Table S22. Cartesian coordinates, optimized geometry, and energies of **Int 2e**

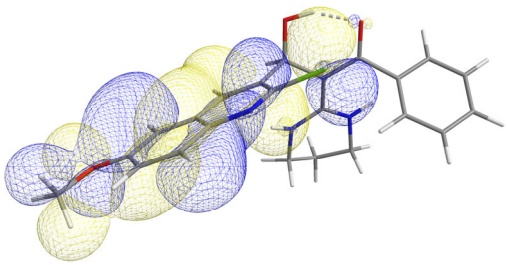
3-(2-Chloro-6-methoxyquinolin-3-yl)-3-hydroxy-1-phenyl-2-(tetrahydropyrimidin-2(1*H*)-ylidene)propan-1-one (Int 2e)

E_{B3LYP}	E_0	H	$TCGFE$
-1740.1465617	0.413817	0.441075	0.355373



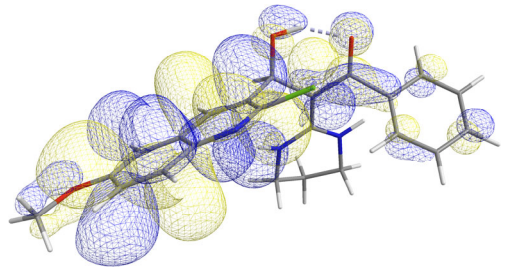


HOMO

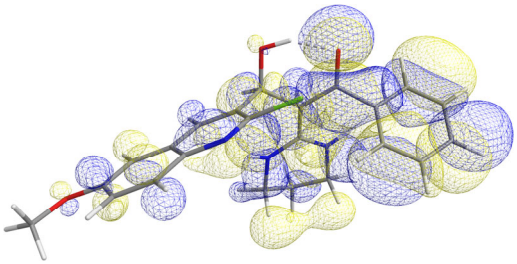


HOMO-1

Atoms	x	y	z
C	-5.574124	0.293459	-0.225993
C	-5.508033	-0.756482	-1.176484
C	-4.342204	-1.469909	-1.342794
C	-3.196376	-1.170579	-0.575564
C	-4.463624	0.606737	0.536797
N	-2.062005	-1.906235	-0.751043
C	-0.922374	-0.569213	0.922322
C	-2.079277	0.150914	1.113595
C	1.585155	0.129732	1.021049
C	2.706974	-0.777838	1.000895
C	3.790753	-0.680647	-0.052676
C	5.118765	-0.899082	0.332554
C	6.138671	-0.898015	-0.615202
C	5.838223	-0.715411	-1.964337
C	4.513836	-0.532519	-2.360898
C	3.496323	-0.507874	-1.410657
O	2.833039	-1.728296	1.795413
H	5.332458	-1.084299	1.378437
H	7.165295	-1.055834	-0.303805
H	6.630138	-0.730456	-2.704913
H	4.272546	-0.420859	-3.412268
H	2.464717	-0.382911	-1.718015
C	0.304452	-0.245028	1.799303
O	0.466023	-1.278034	2.740974
H	1.348656	-1.677483	2.582460
H	0.003792	0.657443	2.353461
Cl	0.378799	-2.671571	-0.314292
C	-1.014380	-1.624812	-0.044075
C	-3.263292	-0.111788	0.374817
H	-6.373035	-1.008294	-1.774961
H	-4.276772	-2.278973	-2.059740
H	-4.536268	1.409367	1.261548
H	-2.093722	0.936292	1.865399



LUMO



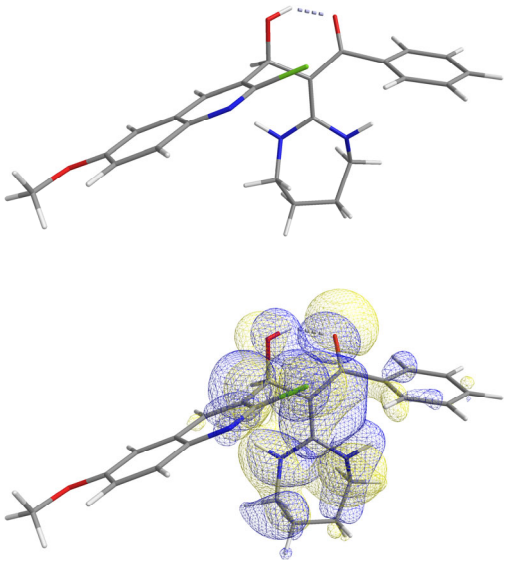
LUMO+1

O	-6.690129	1.043276	-0.003035
C	-7.873502	0.754396	-0.736222
H	-8.229370	-0.263466	-0.543461
H	-8.618369	1.466045	-0.384256
H	-7.725474	0.892094	-1.812984
C	1.659748	1.437035	0.507736
N	0.524955	2.184284	0.355578
N	2.832804	2.082309	0.251763
C	0.463885	3.521150	-0.235621
C	2.914957	3.303469	-0.537954
C	1.798908	4.243416	-0.092441
H	0.200787	3.446886	-1.299564
H	2.833606	3.094126	-1.614521
H	1.791332	5.153561	-0.696660
H	3.674723	1.544664	0.374830
H	-0.333160	1.655639	0.317555
H	3.895787	3.748643	-0.363679
H	1.964705	4.528387	0.949996
H	-0.334466	4.081396	0.258325

Table S23. Cartesian coordinates, optimized geometry, and energies of **Int 2f**

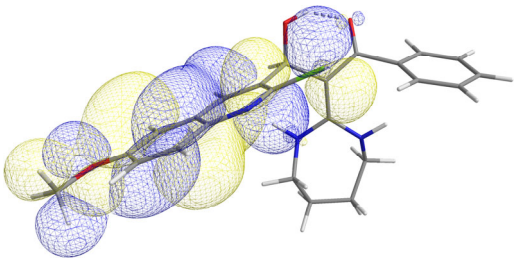
3-(2-Chloro-6-methoxyquinolin-3-yl)-2-(1,3-diazepan-2-ylidene)-3-hydroxy-1-phenyl propan-1-one (Int 2f)

E_{B3LYP}	E_0	H	$TCGFE$
-1779.4650962	0.442756	0.470991	0.382768

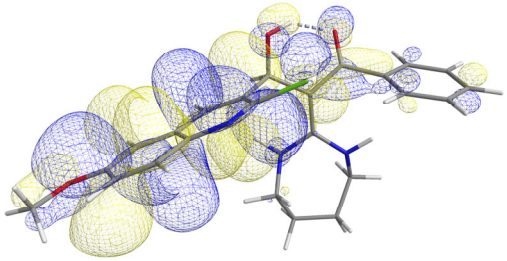


HOMO

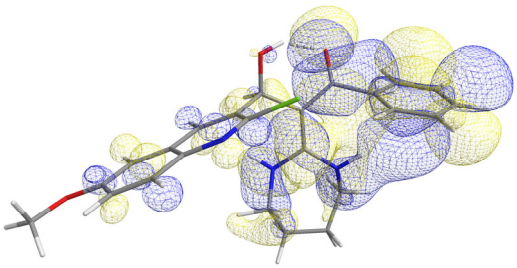
Atoms	x	y	z
C	-5.684489	0.100770	0.116732
C	-5.622645	-0.487289	-1.172080
C	-4.440983	-1.018953	-1.636872
C	-3.273760	-0.988228	-0.844062
C	-3.336774	-0.392376	0.448170
C	-4.553701	0.144137	0.911954
N	-2.124280	-1.541883	-1.323105
C	-0.962644	-0.907886	0.705870
C	-2.132311	-0.382949	1.198963
C	1.543237	-0.272656	0.937114
C	2.813170	-0.951322	1.135696
C	4.013893	-0.735311	0.245718
C	5.269410	-0.539173	0.833475
C	6.408767	-0.441031	0.042427
C	6.314158	-0.576522	-1.343561



HOMO-1



LUMO



LUMO+1

C	5.074104	-0.806821	-1.933937
C	3.925989	-0.879587	-1.144978
O	2.982475	-1.782796	2.043988
H	-6.503701	-0.526610	-1.798141
H	-4.378726	-1.477542	-2.615986
H	-4.623239	0.592741	1.896153
H	-2.142651	0.039156	2.200804
H	5.337586	-0.477750	1.912859
H	7.374630	-0.273289	0.505874
H	7.205505	-0.513987	-1.957686
H	4.997745	-0.938619	-3.007427
H	2.960090	-1.066164	-1.600539
C	0.287699	-0.917275	1.588463
O	0.452472	-2.262873	1.970349
H	1.383062	-2.351631	2.257177
H	0.021731	-0.323235	2.484974
Cl	0.348167	-2.324289	-1.288546
C	-1.057342	-1.510278	-0.590779
C	1.451594	1.041300	0.467793
C	2.776949	3.097996	0.001384
C	-0.218289	2.787334	-0.183782
C	1.890311	4.131459	-0.699885
H	2.777878	3.279303	1.083129
H	3.806038	3.209404	-0.346467
C	0.458653	4.160595	-0.161864
H	-0.187204	2.371413	-1.197170
H	-1.269908	2.893305	0.090035
H	1.881672	3.909571	-1.772736
H	2.347697	5.119570	-0.580804
H	0.452579	4.536817	0.867287
H	-0.143772	4.853127	-0.759355
N	0.360542	1.822828	0.762799
N	2.398931	1.703100	-0.273262
H	3.159370	1.125834	-0.592226
O	-6.815456	0.645654	0.646200
C	-8.018850	0.611037	-0.110572
H	-8.771956	1.089055	0.513425
H	-7.922430	1.168884	-1.048461
H	-8.330242	-0.416665	-0.326489
H	-0.309757	1.400226	1.379423

3 Crystal X-ray structure and packing diagrams of compound 3k

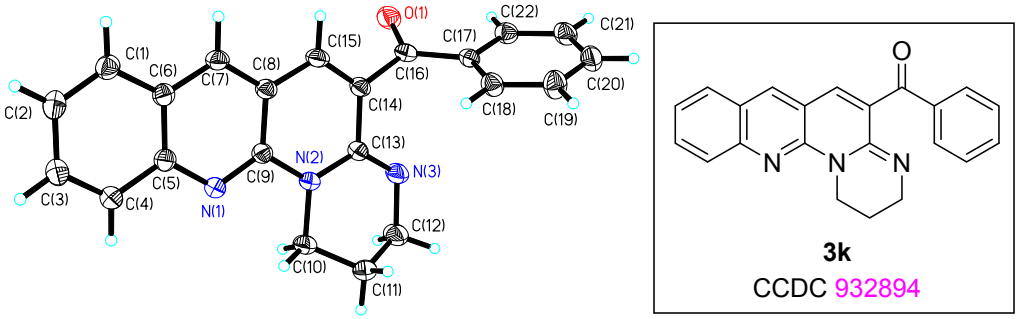


Fig. S51 ORTEP view of the molecular structure of **3k**,[2] thermal ellipsoids are drawn at 30% probability

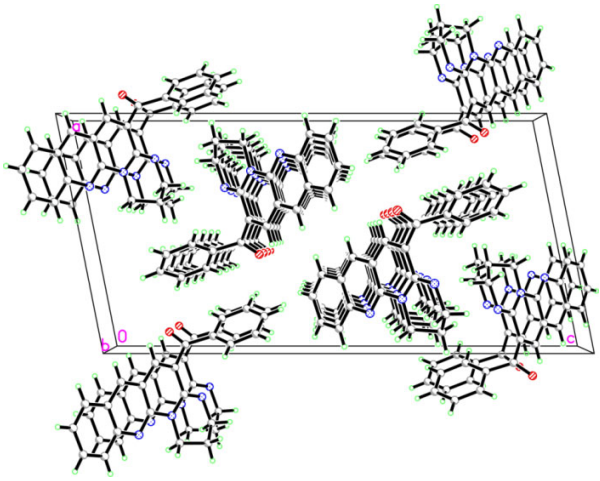


Fig. S52 Packing in the crystal structure of **3k**, viewed along the *b* axis

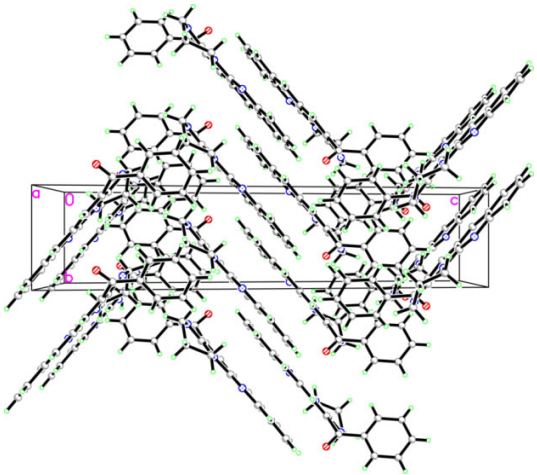


Fig. S53 Packing in the crystal structure of **3k**, viewed along the *a* axis.

Table S24. Crystal data, data collection, and structure refinement for compound **3k**

(2,3-Dihydro-1*H*-benzo[*g*]pyrimido[1,2-*a*][1,8]naphthyridin-5-yl)(phenyl)methanone (3k)	
Crystal data	
$C_{22}H_{17}N_3O$	$Z = 4$
$M_r = 339.39$	$F(000) = 712$
Monoclinic, $P2_1/n$	$D_x = 1.368$ Mg m^{-3}
Hall symbol: -P 2yn	Mo $K\alpha$ radiation, $\lambda = 0.71073$ Å
$a = 12.3869(16)$ Å	Cell parameters from 1628 reflections
$b = 5.4576(7)$ Å	$\theta = 1.72$–$24.99°$
$c = 24.843(3)$ Å	$\mu = 0.086$ mm^{-1}
$\alpha = 90°$	$T = 298(2)$ K
$\beta = 101.217(2)°$	Block, yellow
$\gamma = 90°$	0.27 × 0.20 × 0.17 mm
$V = 1647.4(4)$ Å^3	
Data collection	
Bruker apex II diffractometer	8916 measured reflections
Radiation source: fine-focus sealed tube	2901 independent reflections
Graphite monochromator	1992 reflections with $I > 2\sigma(I)$
Absorption correction: multi-scan (SADABS; Bruker, **2000**)	Rint = 0.0321
	$\theta_{max} = 24.99°$, $\theta_{min} = 1.72°$
ω scans	$h = -14 \rightarrow 14$
$T_{min} = 0.9771$, $T_{max} = 0.9855$	$k = -5 \rightarrow 6$
	$l = -25 \rightarrow 29$
Refinement	
Refinement on F^2	Primary atom site location: structure-invariant direct methods
Least-squares matrix: full	Hydrogen site location: inferred from neighboring sites
$R[F^2 > 2\sigma(F^2)] = 0.0593$	$w = 1/[\sigma^2(F_o^2) + (0.0629P)^2 + 0.2149P]$ where $P=(F_o^2 + 2F_c^2)/3$
$wR(F^2) = 0.1190$	$(\Delta/\sigma)_{max} < 0.001$
$S = 1.062$	$\Delta\rho_{max} = 0.160$ e Å^{-3}
2901 reflections	$\Delta\rho_{min} = -0.157$ e Å^{-3}
236 parameters	
0 restraints	

Table S25. Geometric parameters of compound **3k**

Bond lengths (Å)

N(1)-C(9)	1.314(2)	C(10)-H(10A)	0.9700
N(1)-C(5)	1.372(3)	C(10)-H(10B)	0.9700
N(2)-C(9)	1.395(2)	C(11)-C(12)	1.497(3)
N(2)-C(13)	1.408(2)	C(11)-H(11A)	0.9700
N(2)-C(10)	1.470(2)	C(11)-H(11B)	0.9700
N(3)-C(13)	1.279(2)	C(12)-H(12A)	0.9700
N(3)-C(12)	1.462(3)	C(12)-H(12B)	0.9700
O(1)-C(16)	1.225(2)	C(13)-C(14)	1.472(3)
C(1)-C(2)	1.358(3)	C(14)-C(15)	1.344(3)
C(1)-C(6)	1.411(3)	C(14)-C(16)	1.509(3)
C(1)-H(1)	0.9300	C(15)-H(15)	0.9300
C(2)-C(3)	1.401(3)	C(16)-C(17)	1.481(3)
C(2)-H(2)	0.9300	C(17)-C(18)	1.380(3)
C(3)-C(4)	1.362(3)	C(17)-C(22)	1.388(3)
C(3)-H(3)	0.9300	C(18)-C(19)	1.380(3)
C(4)-C(5)	1.406(3)	C(18)-H(18)	0.9300
C(4)-H(4)	0.9300	C(19)-C(20)	1.373(3)
C(5)-C(6)	1.416(3)	C(19)-H(19)	0.9300
C(6)-C(7)	1.409(3)	C(20)-C(21)	1.382(3)
C(7)-C(8)	1.370(3)	C(20)-H(20)	0.9300
C(7)-H(7)	0.9300	C(21)-C(22)	1.372(3)
C(8)-C(9)	1.427(3)	C(21)-H(21)	0.9300
C(8)-C(15)	1.434(3)	C(22)-H(22)	0.9300
C(10)-C(11)	1.495(3)		

Bond angles (°)

C(9)-N(1)-C(5)	117.73(16)	C(12)-C(11)-H(11A)	109.7
C(9)-N(2)-C(13)	123.76(16)	C(10)-C(11)-H(11B)	109.7
C(9)-N(2)-C(10)	118.24(16)	C(12)-C(11)-H(11B)	109.7
C(13)-N(2)-C(10)	117.82(16)	H(11A)-C(11)-H(11B)	108.2
C(13)-N(3)-C(12)	118.56(18)	N(3)-C(12)-C(11)	113.62(19)
C(2)-C(1)-C(6)	121.0(2)	N(3)-C(12)-H(12A)	108.8
C(2)-C(1)-H(1)	119.5	C(11)-C(12)-H(12A)	108.8
C(6)-C(1)-H(1)	119.5	N(3)-C(12)-H(12B)	108.8
C(1)-C(2)-C(3)	119.7(2)	C(11)-C(12)-H(12B)	108.8

C(1)-C(2)-H(2)	120.2	H(12A)-C(12)-H(12B)	107.7
C(3)-C(2)-H(2)	120.2	N(3)-C(13)-N(2)	125.75(17)
C(4)-C(3)-C(2)	121.1(2)	N(3)-C(13)-C(14)	118.23(18)
C(4)-C(3)-H(3)	119.5	N(2)-C(13)-C(14)	116.00(17)
C(2)-C(3)-H(3)	119.5	C(15)-C(14)-C(13)	120.98(18)
C(3)-C(4)-C(5)	120.5(2)	C(15)-C(14)-C(16)	119.37(17)
C(3)-C(4)-H(4)	119.8	C(13)-C(14)-C(16)	119.45(17)
C(5)-C(4)-H(4)	119.8	C(14)-C(15)-C(8)	121.90(18)
N(1)-C(5)-C(4)	118.61(18)	C(14)-C(15)-H(15)	119.1
N(1)-C(5)-C(6)	122.69(18)	C(8)-C(15)-H(15)	119.1
C(4)-C(5)-C(6)	118.69(19)	O(1)-C(16)-C(17)	120.50(18)
C(1)-C(6)-C(7)	123.84(18)	O(1)-C(16)-C(14)	117.25(19)
C(1)-C(6)-C(5)	119.08(19)	C(17)-C(16)-C(14)	122.00(17)
C(7)-C(6)-C(5)	117.07(18)	C(18)-C(17)-C(22)	119.49(19)
C(8)-C(7)-C(6)	120.81(18)	C(18)-C(17)-C(16)	121.76(18)
C(8)-C(7)-H(7)	119.6	C(22)-C(17)-C(16)	118.58(18)
C(6)-C(7)-H(7)	119.6	C(19)-C(18)-C(17)	119.7(2)
C(7)-C(8)-C(9)	117.38(18)	C(19)-C(18)-H(18)	120.1
C(7)-C(8)-C(15)	123.81(18)	C(17)-C(18)-H(18)	120.1
C(9)-C(8)-C(15)	118.79(18)	C(20)-C(19)-C(18)	120.7(2)
N(1)-C(9)-N(2)	117.26(16)	C(20)-C(19)-H(19)	119.6
N(1)-C(9)-C(8)	124.29(18)	C(18)-C(19)-H(19)	119.6
N(2)-C(9)-C(8)	118.45(17)	C(19)-C(20)-C(21)	119.6(2)
N(2)-C(10)-C(11)	110.14(17)	C(19)-C(20)-H(20)	120.2
N(2)-C(10)-H(10A)	109.6	C(21)-C(20)-H(20)	120.2
C(11)-C(10)-H(10A)	109.6	C(22)-C(21)-C(20)	120.1(2)
N(2)-C(10)-H(10B)	109.6	C(22)-C(21)-H(21)	119.9
C(11)-C(10)-H(10B)	109.6	C(20)-C(21)-H(21)	119.9
H(10A)-C(10)-H(10B)	108.1	C(21)-C(22)-C(17)	120.3(2)
C(10)-C(11)-C(12)	109.88(19)	C(21)-C(22)-H(22)	119.8
C(10)-C(11)-H(11A)	109.7	C(17)-C(22)-H(22)	119.8

Torsion angles (°)

C(6)-C(1)-C(2)-C(3)	-0.9(3)	C(10)-C(11)-C(12)-N(3)	51.5(3)
C(1)-C(2)-C(3)-C(4)	-0.6(3)	C(12)-N(3)-C(13)-N(2)	-3.1(3)
C(2)-C(3)-C(4)-C(5)	2.0(3)	C(12)-N(3)-C(13)-C(14)	178.6(2)
C(9)-N(1)-C(5)-C(4)	-178.82(18)	C(9)-N(2)-C(13)-N(3)	-175.6(2)
C(9)-N(1)-C(5)-C(6)	0.2(3)	C(10)-N(2)-C(13)-N(3)	-0.6(3)
C(3)-C(4)-C(5)-N(1)	177.0(2)	C(9)-N(2)-C(13)-C(14)	2.7(3)
C(3)-C(4)-C(5)-C(6)	-2.1(3)	C(10)-N(2)-C(13)-C(14)	177.75(17)

C(2)-C(1)-C(6)-C(7)	-178.4(2)	N(3)-C(13)-C(14)-C(15)	178.5(2)
C(2)-C(1)-C(6)-C(5)	0.8(3)	N(2)-C(13)-C(14)-C(15)	0.1(3)
N(1)-C(5)-C(6)-C(1)	-178.31(19)	N(3)-C(13)-C(14)-C(16)	3.6(3)
C(4)-C(5)-C(6)-C(1)	0.7(3)	N(2)-C(13)-C(14)-C(16)	-174.85(17)
N(1)-C(5)-C(6)-C(7)	0.9(3)	C(13)-C(14)-C(15)-C(8)	-1.0(3)
C(4)-C(5)-C(6)-C(7)	179.91(18)	C(16)-C(14)-C(15)-C(8)	173.94(17)
C(1)-C(6)-C(7)-C(8)	178.53(19)	C(7)-C(8)-C(15)-C(14)	-179.0(2)
C(5)-C(6)-C(7)-C(8)	-0.7(3)	C(9)-C(8)-C(15)-C(14)	-0.7(3)
C(6)-C(7)-C(8)-C(9)	-0.6(3)	C(15)-C(14)-C(16)-O(1)	-56.0(3)
C(6)-C(7)-C(8)-C(15)	177.71(18)	C(13)-C(14)-C(16)-O(1)	119.0(2)
C(5)-N(1)-C(9)-N(2)	178.57(17)	C(15)-C(14)-C(16)-C(17)	118.2(2)
C(5)-N(1)-C(9)-C(8)	-1.6(3)	C(13)-C(14)-C(16)-C(17)	-66.8(2)
C(13)-N(2)-C(9)-N(1)	175.48(17)	O(1)-C(16)-C(17)-C(18)	159.20(19)
C(10)-N(2)-C(9)-N(1)	0.4(3)	C(14)-C(16)-C(17)-C(18)	-14.9(3)
C(13)-N(2)-C(9)-C(8)	-4.4(3)	O(1)-C(16)-C(17)-C(22)	-16.0(3)
C(10)-N(2)-C(9)-C(8)	-179.44(18)	C(14)-C(16)-C(17)-C(22)	169.89(17)
C(7)-C(8)-C(9)-N(1)	1.8(3)	C(22)-C(17)-C(18)-C(19)	0.6(3)
C(15)-C(8)-C(9)-N(1)	-176.57(19)	C(16)-C(17)-C(18)-C(19)	-174.61(19)
C(7)-C(8)-C(9)-N(2)	-178.35(18)	C(17)-C(18)-C(19)-C(20)	-0.8(3)
C(15)-C(8)-C(9)-N(2)	3.3(3)	C(18)-C(19)-C(20)-C(21)	-0.1(3)
C(9)-N(2)-C(10)-C(11)	-154.99(19)	C(19)-C(20)-C(21)-C(22)	1.2(3)
C(13)-N(2)-C(10)-C(11)	29.7(3)	C(20)-C(21)-C(22)-C(17)	-1.4(3)
N(2)-C(10)-C(11)-C(12)	-53.4(2)	C(18)-C(17)-C(22)-C(21)	0.5(3)
C(13)-N(3)-C(12)-C(11)	-23.3(3)	C(16)-C(17)-C(22)-C(21)	175.86(19)

4 Notes and References

(1) Full reference (22) for Gaussian03: M. J. Frisch, G. W. Trucks, H. B. Schlegel, G. E. Scuseria, M. A. Robb, J. R. Cheeseman, J. A. Montgomery, Jr., T. Vreven, K. N. Kudin, J. C. Burant, J. M. Millam, S. S. Iyengar, J. Tomasi, V. Barone, B. Mennucci, M. Cossi, G. Scalmani, N. Rega, G. A. Petersson, H. Nakatsuji, M. Hada, M. Ehara, K. Toyota, R. Fukuda, J. Hasegawa, M. Ishida, T. Nakajima, Y. Honda, O. Kitao, H. Nakai, M. Klene, X. Li, J. E. Knox, H. P. Hratchian, J. B. Cross, V. Bakken, C. Adamo, J. Jaramillo, R. Gomperts, R. E. Stratmann, O. Yazyev, A. J. Austin, R. Cammi, C. Pomelli, J. W. Ochterski, P. Y. Ayala, K. Morokuma, G. A. Voth, P. Salvador, J. J. Dannenberg, V. G. Zakrzewski, S. Dapprich, A. D. Daniels, M. C. Strain, O. Farkas, D. K. Malick, A. D. Rabuck, K. Raghavachari, J. B. Foresman, J. V. Ortiz, Q. Cui, A. G. Baboul, S. Clifford, J. Cioslowski, B. B. Stefanov, G. Liu, A. Liashenko, P. Piskorz, I. Komaromi, R. L. Martin, D. J. Fox, T. Keith, M. A. Al-Laham, C. Y. Peng, A. Nanayakkara, M. Challacombe, P. M. W. Gill, B. Johnson, W. Chen, M. W. Wong, C. Gonzalez and J. A. Pople, *Gaussian03, Revision D.01*, Gaussian, Inc., Wallingford CT, 2004.

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