Green Synthesis, SC-XRD Exploration and Non-Covalent Interactive Potential via DFT-Investigation of the Functionalized Pyridine-based Novel Hydrazone

Supplementary Data

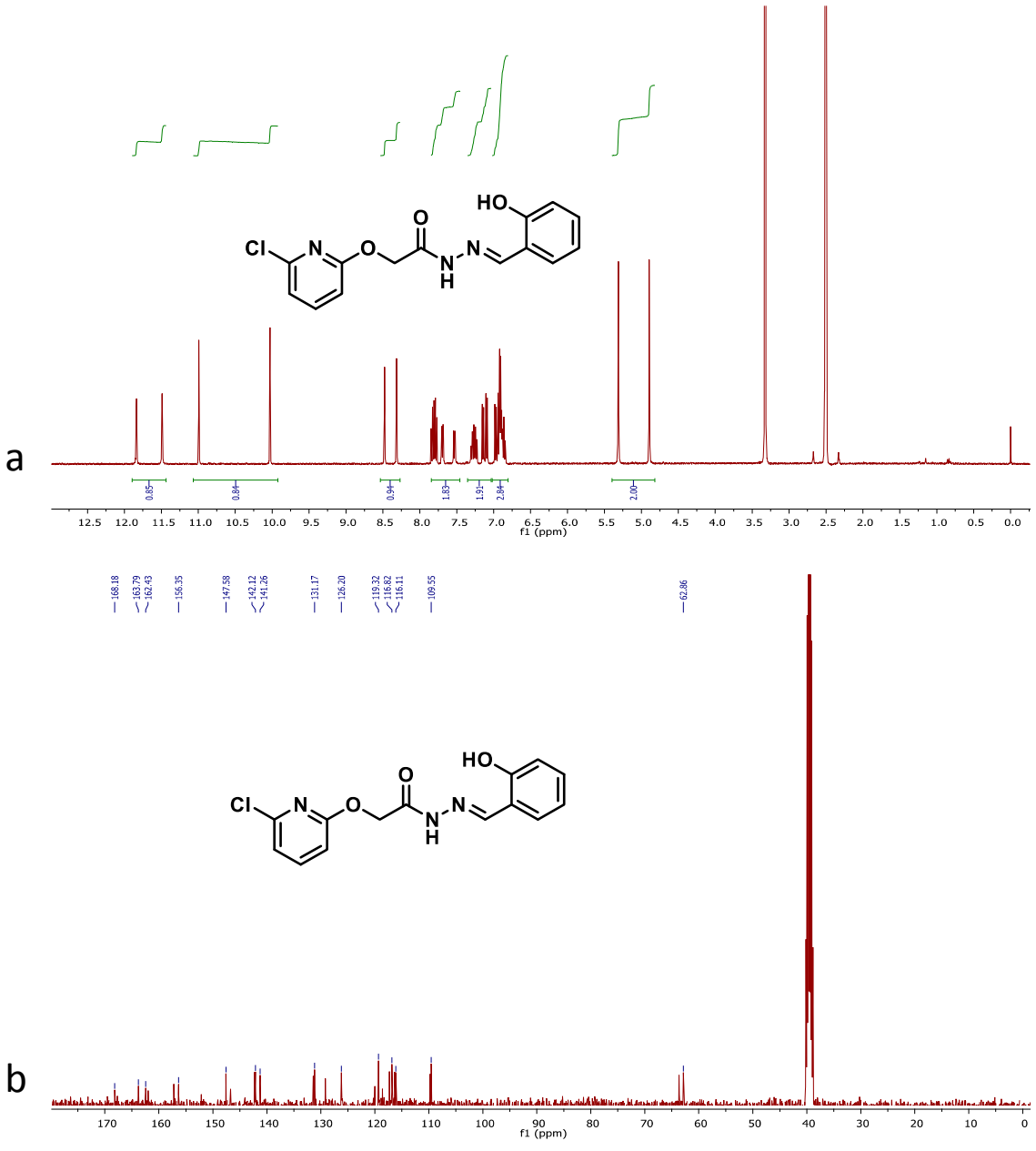


Figure S1: **(a)** ^{1}H and (b) ^{13}C NMR spectra of **HBPAH**

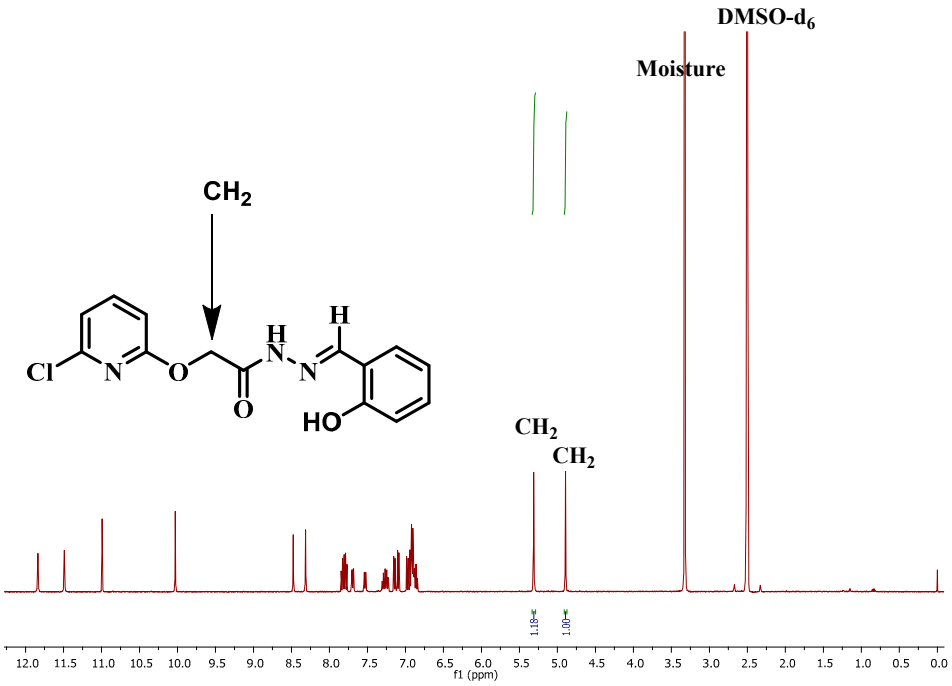


Figure S2: The ^{1}H-NMR of the title compound in $(CD_3)_2$ SO-d6 showing the integration of methylenic (CH_2) ^{1}Hs, which has been used as a tool to calculate the ratio of two isomers (A and B).

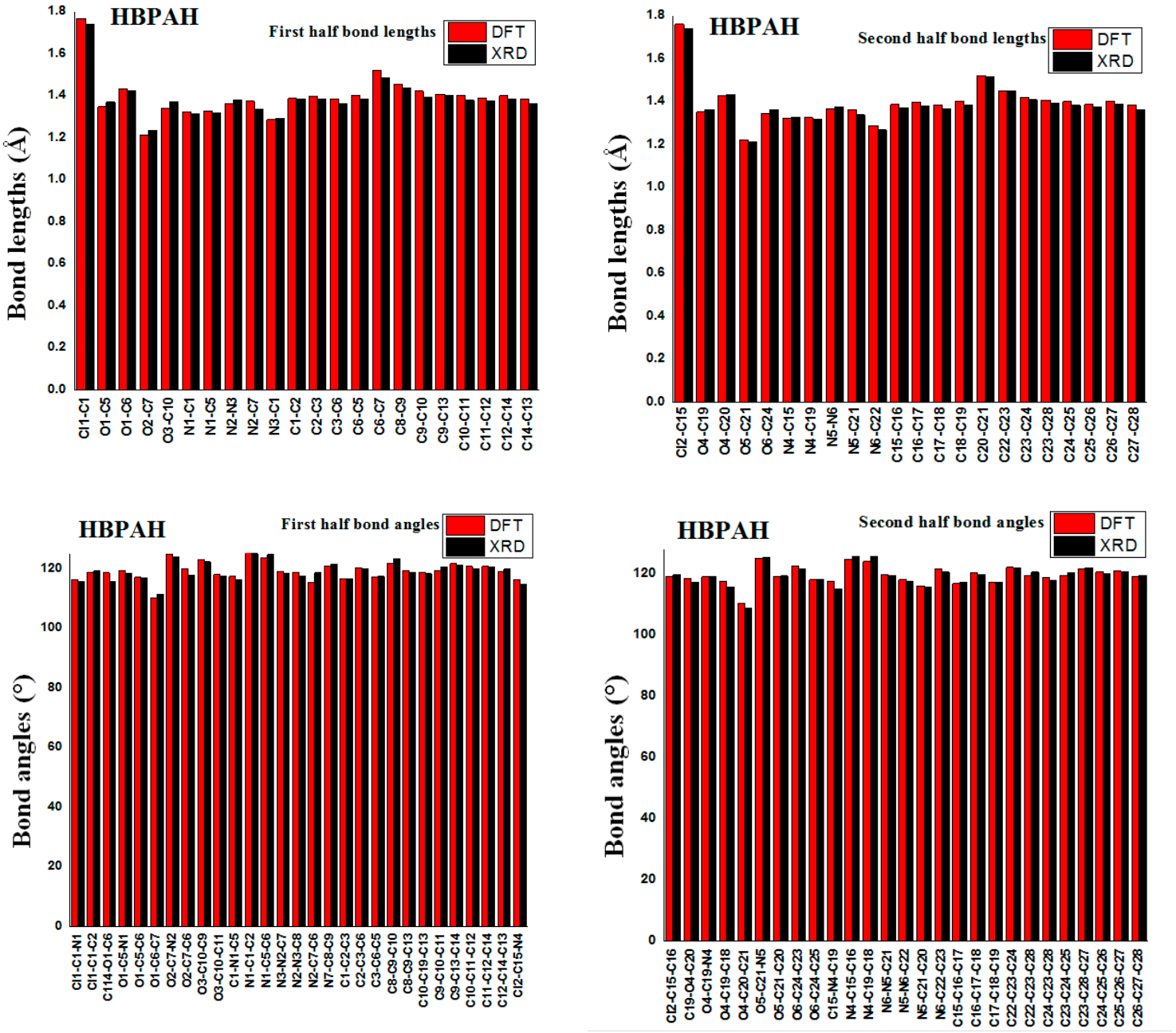


Figure S3: The geometrical parameters (bond lengths (Å) and bond angles (°) of entitled compound calculated through XRD and at DFT/ B3LYP/6-311G (d,p) level of theory.

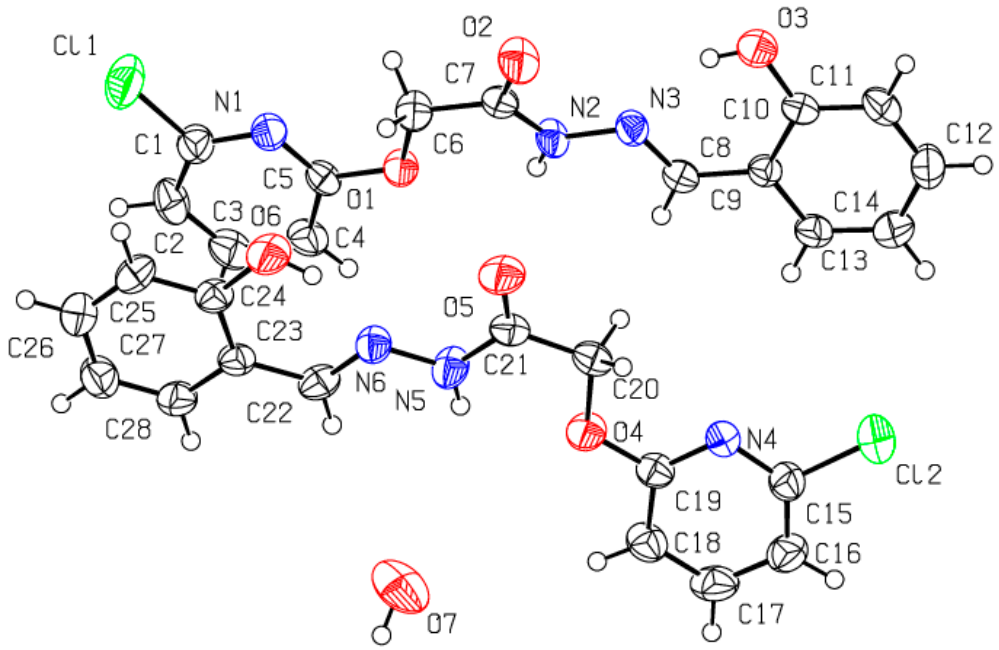
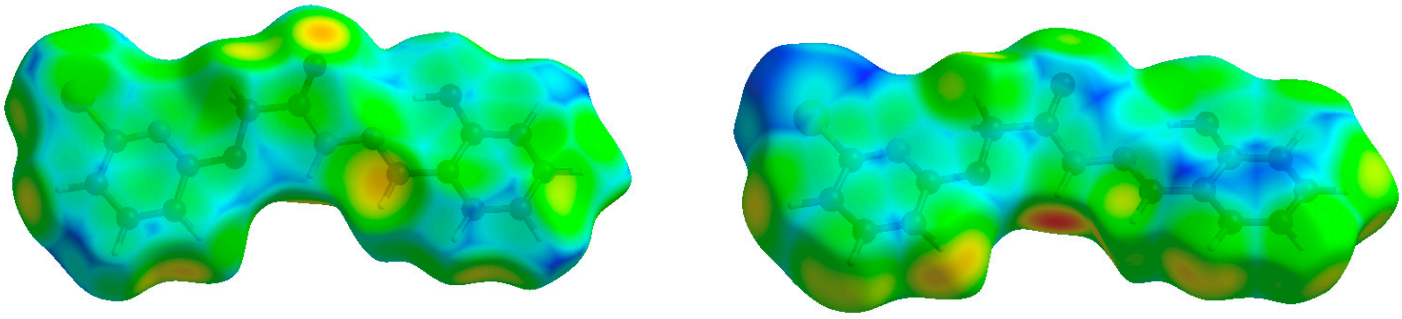
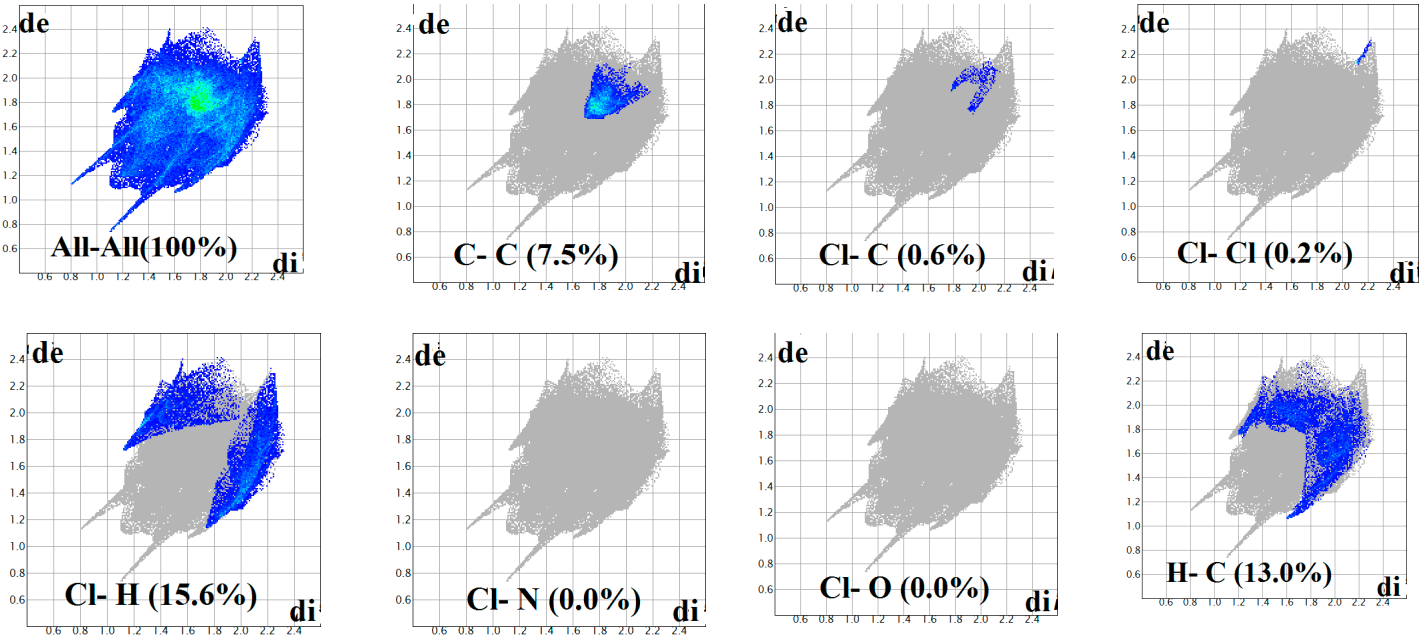


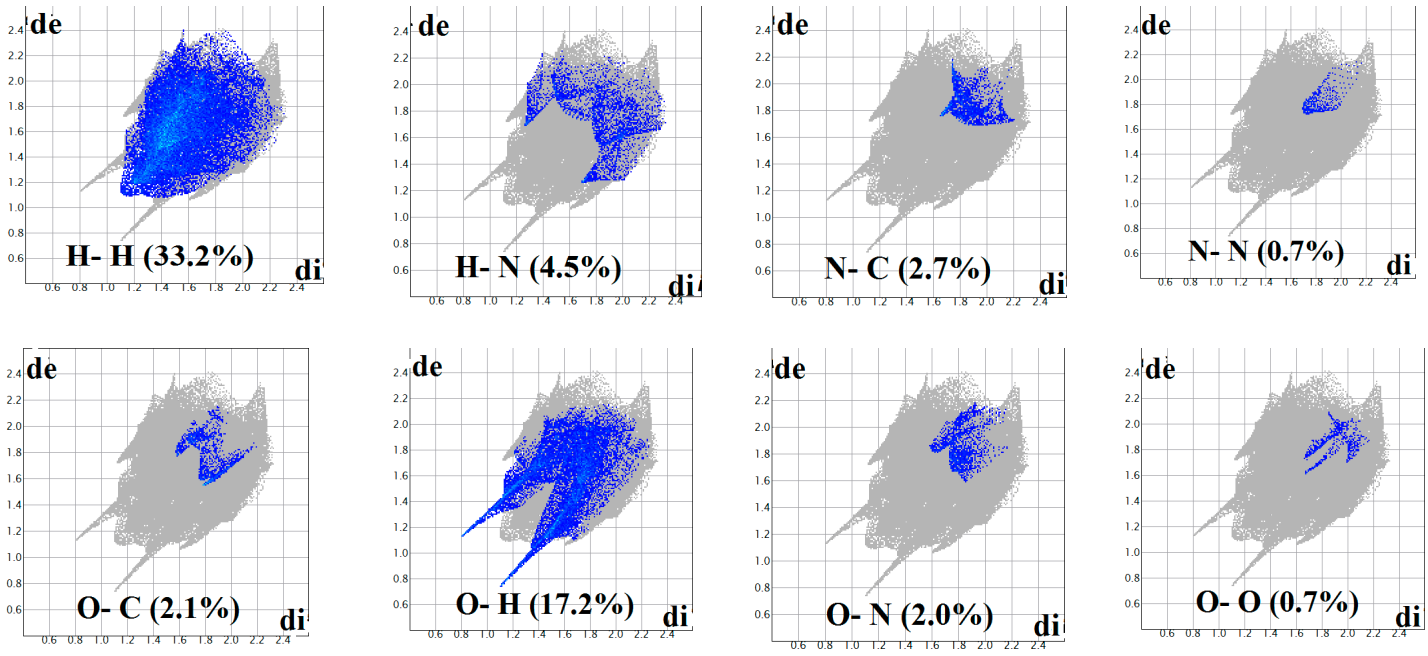
Figure S4: ORTEP diagram of **HBPAH** compound



(a) d_e (HBPAH) **(b) d_i (HBPAH)**

Figure S5: Hirshfeld surfaces of the entitled compound mapped over, (a) d_e and (b) d_i for **HBPAH** (1 a.u. of electron density = 6.748 e.Å^{-3}).





HBPAH

Figure S6: 2-D Fingerprint plots for individual contributions in **HBPAH**

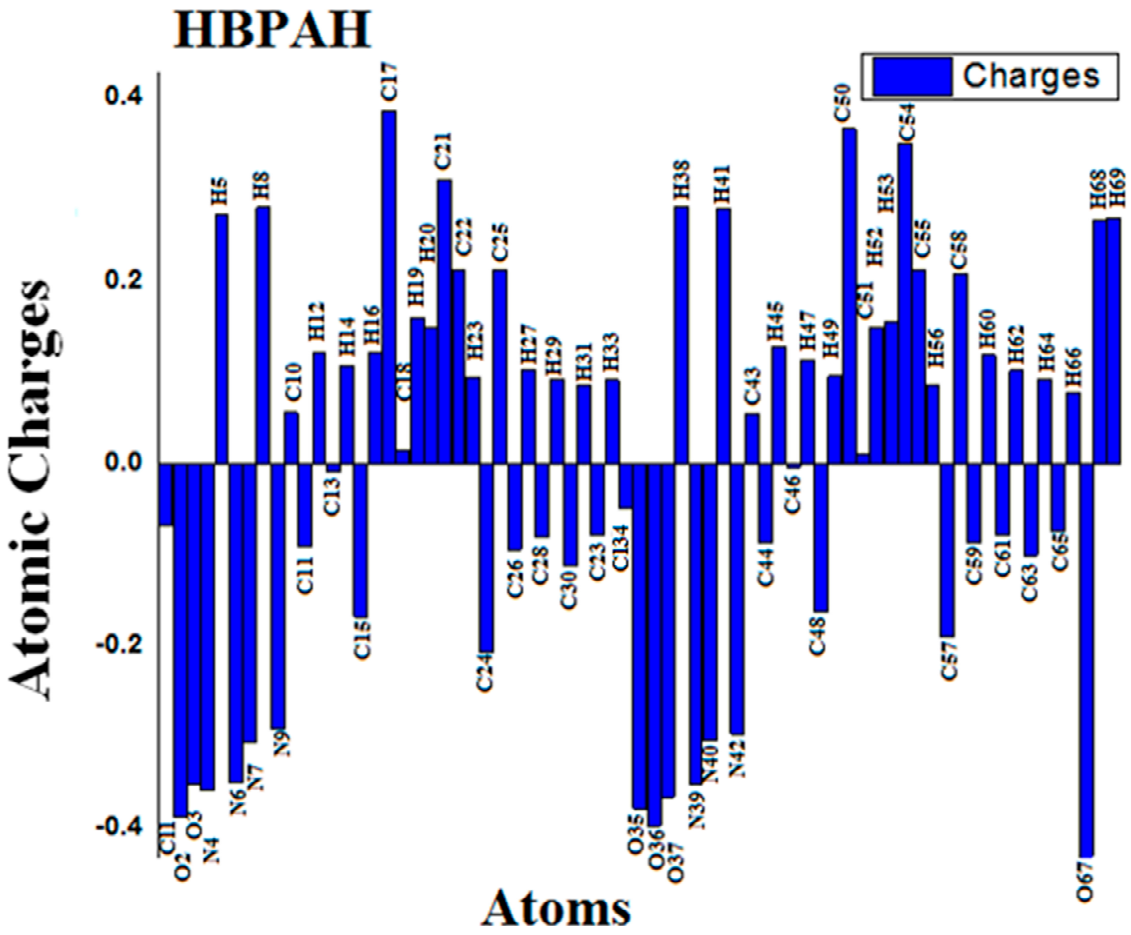


Figure S7: Natural population analysis (NPA) of entitled compound

Table S1: Comparison of XRD and DFT values of bond length (Å) and bond angle (°) of **HBPAH**

Bond length			**Bond angle**		
	DFT	**XRD**		**DFT**	**XRD**
CL1-C1	1.767	1.741	CL1-C1-N1	116.2	115.6
O1-C5	1.347	1.369	CL1-C1-C2	118.8	119.2
O1-C6	1.434	1.421	C114-O1-C6	118.6	115.6
O2-C7	1.212	1.236	O1-C5-N1	119.3	118.4
O3-C10	1.341	1.369	O1-C5-C6	117.1	116.9
N1-C1	1.321	1.311	O1-C6-C7	110.1	111.5
N1-C5	1.327	1.318	O2-C7-N2	124.8	123.8
N2-N3	1.359	1.379	O2-C7-C6	119.9	117.6
N2-C7	1.374	1.335	O3 -C10-C9	122.9	122.2
N3-C1	1.286	1.291	O3-C10-C11	117.9	117.4
C1-C2	1.386	1.384	C1-N1-C5	117.4	116.2
C2-C3	1.398	1.385	N1-C1-C2	125.1	125.2
C3-C6	1.383	1.363	N1-C5-C6	123.6	124.7
C6-C5	1.402	1.384	N3-N2-C7	119.1	118.3
C6-C7	1.522	1.486	N2-N3-C8	118.8	117.4
C8-C9	1.453	1.435	N2-C7-C6	115.3	118.6
C9-C10	1.421	1.394	N7-C8-C9	120.8	121.4
C9-C13	1.406	1.401	C1-C2-C3	116.5	116.6
C10-C11	1.401	1.378	C2-C3-C6	120.2	119.9
C11-C12	1.387	1.376	C3-C6-C5	117.3	117.4
C12-C14	1.399	1.383	C8-C9-C10	121.8	123.3
C14-C13	1.385	1.36	C8-C9-C13	119.4	118.5

CL2-C15	1.761	1.741	C10-C19-C13	118.7	118.2
O4-C19	1.352	1.363	C9-C10-C11	119.2	120.4
O4-C20	1.429	1.434	C9-C13-C14	121.7	121.1
O5-C21	1.222	1.21	C10-C11-C12	120.7	120
O6-C24	1.344	1.363	C11-C12-C14	120.7	120.4
N4-C15	1.323	1.326	C12-C14-C13	119	119.8
N4-C19	1.325	1.316	CL2-C15-N4	116.2	114.7
N5-N6	1.367	1.373	CL2-C15-C16	119.1	119.6
N5-C21	1.361	1.338	C19-O4-C20	118.3	117.2
N6-C22	1.288	1.268	O4-C19-N4	118.9	119
C15-C16	1.387	1.371	O4-C19-C18	117.3	115.5
C16-C17	1.397	1.381	O4-C20-C21	110.3	108.8
C17-C18	1.384	1.365	O5-C21-N5	125.1	125.3
C18-C19	1.4	1.384	O5-C21-C20	119	119.2
C20-C21	1.521	1.515	O6-C24-C23	122.6	121.5
C22-C23	1.45	1.45	O6-C24-C25	118	118.2
C23-C24	1.419	1.408	C15-N4-C19	117.5	115
C23-C28	1.407	1.394	N4-C15-C16	124.7	125.7
C24-C25	1.399	1.382	N4-C19-C18	123.9	125.5
C25-C26	1.386	1.373	N6-N5-C21	119.6	119.3
C26-C27	1.4	1.388	N5-N6-C22	118	117.3
C27-C28	1.384	1.36	N5-C21-C20	115.9	115.5

Table S1 (Continued): Comparison of XRD and DFT values of bond length (Å) and bond angle (°) of **HBPAH.**

	Bond angle	
	DFT	**XRD**
N6-C22-C23	121.4	120.5
C15-C16-C17	116.7	117
C16-C17-C18	120.2	119.8
C17-C18-C19	117.1	117.1
C22-C23-C24	122.1	121.8
C22-C23-C28	119.2	120.4
C24-C23-C28	118.7	117.8
C23-C24-C25	119.4	120.3
C23-C28-C27	121.5	121.9
C24-C25-C26	120.5	120
C25-C26-C27	120.8	120.7
C26-C27-C28	119.1	119.4

Table S2: AIM properties of **HBPAH**; Electronic density (ρ), Laplacian of density ($\nabla^2\rho$), ellipticity (ε) and density of potential energy (V).

BCP	Bonds	ρ (e/a^3)	$\nabla^2\rho$ (e/a^5)	ε	V (hartree.e/a^3)
1	C18 - C21	+0.256150	-0.631048	+0.106844	-0.266796
2	O4 - H5	+0.337054	-2.364523	+0.017247	-0.724624
3	O2 - C17	+0.296642	-0.392039	+0.047328	-0.758694
4	O2 - H8	+0.018129	+0.087823	+0.584732	-0.014400
5	H8 - O36	+0.012699	+0.048764	+0.033907	-0.008095
6	N7 - C21	+0.310673	-0.864750	+0.124981	-0.625161
7	O3 - C21	+0.412226	-0.235561	+0.079615	-1.332231
8	H5 - N9	+0.043584	+0.115749	+0.039706	-0.039206
9	N7 - N9	+0.355628	-0.682239	+0.095534	-0.507189
10	N7 - H8	+0.332853	-1.709316	+0.053177	-0.522020
11	N6 - C10	+0.352575	-1.039557	+0.178872	-0.757180
12	Cl1 - C10	+0.191036	-0.262179	+0.082707	-0.192295
13	N6 - C17	+0.349990	-1.053924	+0.164724	-0.734599
14	C15 - C17	+0.308174	-0.865372	+0.229241	-0.415808
15	C10 - C11	+0.313641	-0.877542	+0.257377	-0.439904
16	C11 - H12	+0.283368	-0.980422	+0.028534	-0.321160
17	C11 - C13	+0.305226	-0.842764	+0.196293	-0.407745
18	H16 - O36	+0.002956	+0.010713	+0.086271	-0.001659
19	C13 - C15	+0.314245	-0.886661	+0.221691	-0.433708
20	C13 - H14	+0.282903	-0.982422	+0.008366	-0.318796
21	C15 - H16	+0.283001	-0.978504	+0.024170	-0.318832
22	O2 - C18	+0.238396	-0.312278	+0.047978	-0.538370

23	C18 - H19	+0.284752	-0.987963	+0.040621	-0.316088
24	C18 - H20	+0.284026	-0.982065	+0.042746	-0.316028
25	N9 - C22	+0.365994	-0.695932	+0.233352	-0.971223
26	H23 - O36	+0.009085	+0.027944	+0.080827	-0.005420
27	C22 - H23	+0.280199	-0.958565	+0.021502	-0.310360
28	C22 - C24	+0.278876	-0.726000	+0.143605	-0.329411
29	O4 - C25	+0.303383	-0.419447	+0.006025	-0.781383
30	C24 - C25	+0.297063	-0.798893	+0.218033	-0.380171
31	C25 - C26	+0.309429	-0.873233	+0.229222	-0.414021
32	C26 - H27	+0.281064	-0.963912	+0.023771	-0.318348
33	C24 - C32	+0.301583	-0.819491	+0.206909	-0.396356
34	C26 - C28	+0.311514	-0.872607	+0.219379	-0.425355
35	C28 - H29	+0.281602	-0.970502	+0.014434	-0.319544
36	C28 - C30	+0.305586	-0.846899	+0.196481	-0.406955
37	C30 - C32	+0.312703	-0.878961	+0.224578	-0.429156
38	C30 - H31	+0.280769	-0.961312	+0.025949	-0.320708
39	C32 - H33	+0.279684	-0.954197	+0.017783	-0.318272
40	C51 - C54	+0.256757	-0.634351	+0.108928	-0.267950
41	O35 - C51	+0.242820	-0.342036	+0.045939	-0.546966
42	O2 - O37	+0.006587	+0.026723	+0.210189	-0.005195
43	H19 - O37	+0.007303	+0.026324	+0.156120	-0.004683
44	O37 - C58	+0.300777	-0.402184	+0.001754	-0.775111
45	O37 - H38	+0.339405	-2.384347	+0.017758	-0.730163
46	C51 - H53	+0.282851	-0.973342	+0.041063	-0.314073
47	Cl34 - C43	+0.194126	-0.271979	+0.083250	-0.196910

48	N39 - C50	+0.351454	-1.059682	+0.170207	-0.738827
49	O35 - H41	+0.017972	+0.086835	+0.617149	-0.014266
50	N40 - C54	+0.318938	-0.885930	+0.140694	-0.663325
51	H38 - N42	+0.042093	+0.114702	+0.043341	-0.037454
52	N40 - N42	+0.349340	-0.655572	+0.089154	-0.491942
53	N42 - C55	+0.365300	-0.713564	+0.213213	-0.962592
54	N40 - H41	+0.331242	-1.705362	+0.051644	-0.519825
55	H41 - O67	+0.014836	+0.055332	+0.131673	-0.009688
56	N39 - C43	+0.350518	-1.030229	+0.175088	-0.751517
57	O35 - C50	+0.292370	-0.387414	+0.046412	-0.741478
58	C43 - C44	+0.313411	-0.877396	+0.252862	-0.438487
59	C44 - H45	+0.283902	-0.985173	+0.026979	-0.320898
60	C46 - C48	+0.313499	-0.883281	+0.218376	-0.431262
61	C48 - C50	+0.308785	-0.867873	+0.233595	-0.419094
62	C44 - C46	+0.305975	-0.846908	+0.195718	-0.409639
63	C46 - H47	+0.283412	-0.986414	+0.007806	-0.318814
64	C48 - H49	+0.282885	-0.976511	+0.025057	-0.319160
65	H49 - O67	+0.006265	+0.018167	+0.063784	-0.003543
66	H33 - H53	+0.001564	+0.005249	+0.186851	-0.000659
67	O36 - C54	+0.402650	-0.283414	+0.066006	-1.274264
68	C51 - H52	+0.283387	-0.978039	+0.041584	-0.314713
69	C57 - C58	+0.298032	-0.804271	+0.220579	-0.382791
70	H56 - O67	+0.011411	+0.032495	+0.098034	-0.006780
71	C55 - C57	+0.280341	-0.733613	+0.144263	-0.334047
72	C57 - C65	+0.300885	-0.816760	+0.203995	-0.394304

73	C55 - H56	+0.281037	-0.964670	+0.018695	-0.310965
74	C58 - C59	+0.310139	-0.877798	+0.228086	-0.417094
75	C61 - C63	+0.305367	-0.847051	+0.190886	-0.405726
76	C59 - C61	+0.312160	-0.876939	+0.216092	-0.426772
77	C59 - H60	+0.281827	-0.970169	+0.022459	-0.318187
78	C61 - H62	+0.282591	-0.978085	+0.012698	-0.319611
79	C63 - C65	+0.313867	-0.885293	+0.225582	-0.432590
80	C63 - H64	+0.281337	-0.965704	+0.024681	-0.320748
81	C65 - H66	+0.280070	-0.957612	+0.016952	-0.318845
82	O67 - H68	+0.364861	-2.486667	+0.025918	-0.769719
83	O67 - H69	+0.364635	-2.486851	+0.025758	-0.769273

Natural bonding orbital (NBO) analysis

Table S3: NBO analysis of **HBPAH** at DFT/ B3LYP/6-311G (d,p) level of theory

Donor(i)	**Type**	**Acceptor(j)**	**Type**	***E*(2)[a] (kcal/mol)**	**E(*j*)E(*i*)[b](a.u.)**	**F(*i*,*j*)[c](a.u.)**
C13-H14	∂	C11-C13	∂*	0.51	1.09	0.021
C13-C15	π	C13-C15	π*	0.57	0.29	0.011
C46-C48	π	N39-C50	π*	30.35	0.26	0.082
C13-C15	π	N6-C17	π*	29.62	0.26	0.082
N39-C50	π	C43-C44	π*	28.32	0.33	0.088
C10-C11	π	C13-C15	π*	22.05	0.3	0.073
C43-C44	π	C45-C48	π*	22.45	0.30	0.074
C30-C32	π	C26-C28	π*	21.46	0.29	0.071
C63-C65	π	C59-C61	π*	21.43	0.29	0.071
C26-C28	π	C30-C32	π*	16.92	0.28	0.062
C46-C48	π	C43-C44	π*	16.12	0.28	0.060
C13-C15	π	C10-C11	π*	16.09	0.27	0.060
C43-C44	π	N39-C50	π*	14.78	0.27	0.058
C10-C11	π	N6-C17	π*	14.31	0.28	0.058

N6-C17	π	C13-C15	π*	9.99	0.34	0.052
N39-C50	π	C46-C48	π*	9.89	0.34	0.052
C18-H19	∂	O3-C21	π*	4.57	0.54	0.048
O3-C21	π	O3-C21	π*	1.00	0.39	0.019
C10-C11	π	C10-C11	π*	1.61	0.29	0.019
C15-C17	∂	N6-C10	∂*	0.55	1.25	0.024
C11-C13	∂	Cl1-C10	∂*	5.65	0.84	0.062
O4-H5	∂	C25-C26	∂*	4.91	1.29	0.071
C11-C13	∂	C10- C11	∂*	3.60	1.26	0.060
C26-H27	∂	C28-C30	∂*	3.96	1.08	0.059
C11-C13	∂	C13-C15	∂*	2.78	1.28	0.054
C11-C13	∂	C15-H16	∂*	2.60	1.15	0.049
C28-C30	∂	C26-C28	∂*	2.97	1.28	0.055
N40	LP(1)	O36-C54	π*	62.83	0.29	0.121
N7	LP(1)	O3-C21	π*	56.48	0.29	0.117
O35	LP(2)	N39-C50	π*	35.80	0.32	0.103
N7	LP(1)	N9-C22	π*	28.03	0.27	0.080
N9	LP(1)	O4-H5	∂*	18.25	0.82	0.111
N9	LP(1)	C22-H23	∂*	8.97	0.80	0.077
Cl1	LP(2)	C11-C13	∂*	0.50	0.87	0.019
N9	LP(1)	N7-H8	∂*	7.04	0.81	0.069
O2	LP(1)	N6-C17	∂*	6.61	1.08	0.076
O36	LP(1)	N7-H8	∂*	2.22	1.13	0.045
Cl1	LP(2)	N6-C10	∂*	5.79	0.85	0.063
Cl1	LP(2)	C10-C11	∂*	3.69	0.87	0.051

Table S4: Ionization potential (IP), electron affinity (EA), electronegativity (X), global hardness (η), chemical potential (μ), global electrophilicity index (ω) and global softness (σ)

Compounds	***IP***	***EA***	***X***	***η***	***μ***	***ω***	***σ***
HBPAH	5.6	1.966	3.783	1.817	-3.783	3.938109	0.275179

Units in *eV*