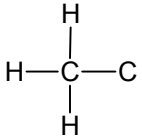
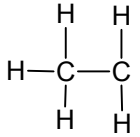
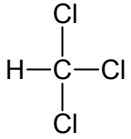
TABLE SI: X-RAY AND NEUTRON DIFFRACTION DATA

H—D	C≡N C≡N⋯H—D (R_E(A) = 1.37)		Pyridine PyN⋯H—D (R_E(A) = 1.30)		C—O C—O⋯H—D (C) (R_E(A) = 1.30)		C=O C=O⋯H—D (R_E(A) = 1.32)	
	$R_{H.....N}$	R_H(HD)	$R_{H.....N}$	R_H(HD)	$R_{H.....O}$	R_H(HD)	$R_{H.....O}$	R_H(HD)
O—H	1.98 ± 0.15	0.61± 0.15	1.86 ± 0.15	0.56± 0.15	1.98 ± 0.15	0.68± 0.15	1.85 ± 0.15	0.53± 0.15
H—N	2.20 ± 0.15	0.83± 0.15	2.10 ± 0.15	0.80± 0.15	2.05 ± 0.13	0.75± 0.13	1.98 ± 0.15	0.66± 0.15
H—C	2.79	1.42	_	_	2.32	1.02	_	_
H—C(=O)	_	_	_	_	2.52	1.22	2.44 ± 0.18	1.12± 0.18
H—C≡C	_	_	_	_	_	_	2.36 ± 0.10	1.04± 0.10
H—C(Cl)(Cl)Cl	_	_	2.37	1.07	2.32	1.02	2.16 ±0.16	0.84± 0.16

TABLE S2: ENTRIES IN X-RAY AND NEUTRON DIFFRACTION DATA

H—D	C≡N⋯H—D		PyN⋯H—D		C—O⋯H—D (C)		C=O⋯H—D	
	T	E	T	E	T	E	T	E
H—O	1830	315	2729	778	8430	2026	10000	4359
H—N	3602	1054	1187	439	4629	655	10000	4129
H—C	10000	0	10000	0	10000	39	10000	0
H—S	0	0	0	0	6	0	19	0
H—C≡C	8	0	12	0	76	0	119	47
F H—C F	3	0	1	0	2	0	15	0

H—D	C≡N⋯H—D		PyN⋯H—D		C—O⋯H—D (C)		C=O⋯H—D	
	T	E	T	E	T	E	T	E
	10000	0	4523	0	10000	0	10000	0
	2857	0	629	31	4497	0	10000	0
	99	0	19	0	159	18	1198	102
	41	0	48	17	73	21	207	70

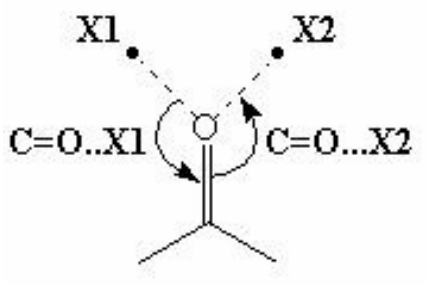
NOTE:
T – Total number of entries. E – Number of entries below the peak.

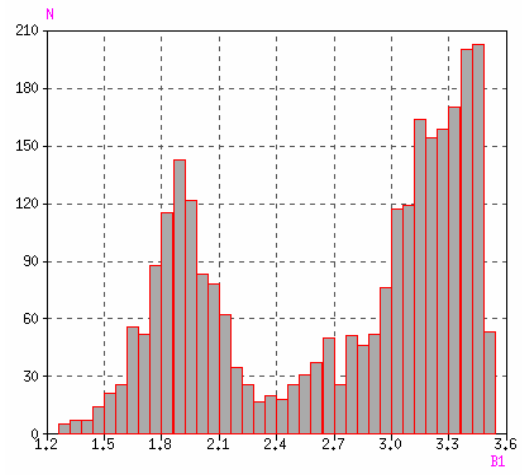
Table S3: Topographical and geometric parameters for substituted pyridines (Py) at HF/6-31+g(d) level of theory.

Substrate	d(N..X) (Å)	MESP at X ($kcal.mol^{-1}$)
Py	1.27	-67.82
Py-CHO(p)	1.27	-59.14
Py-CHO(m)	1.26	-58.17
Py-NH_2(m)	1.26	-68.97
Py-OMe(m)-OMe(m)-NMe_2(p)	1.26	-67.73
Py-OMe(m)-OMe(m)	1.26	-66.27
Py-CH_3 (p)	1.25	-70.18
Py-CH_3 (m)	1.25	-69.09
Py-NH_2(p)	1.25	-76.29
Py-OMe(m)-OMe(m)-NH_2(p)	1.25	-72.79
Py-NO_2(m)-NO_2(m)-COOH(p)	1.30	-34.26
Py-NO_2(m)-NO_2(m)-CN(p)	1.31	-27.65

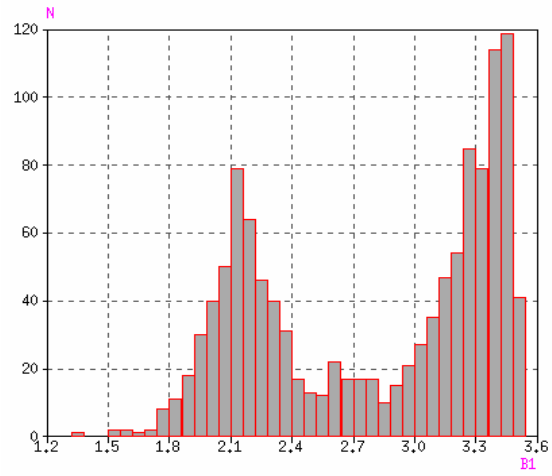
Table S4. Topographical parameters for substituted carbonyl molecules at HF/6-31+g(d) level of theory. Distances in Å and MESP values in kcal.mol^{-1} See Figure below for notation and details.

Carbonyl compound	d(O..X1) (Å)	d(O..X2) (Å)	∠COX1	∠COX2	MESP at X1	MESP at X2
H-CO-Cl	1.30	1.30	136	131	-31.75	-35.33
CH_3-CO-Cl	1.28	1.28	139	129	-35.96	-41.67
CH_3-CO-F	1.26	1.26	137	127	-42.36	-47.00
H-CO-H	1.26	1.26	129	129	-49.01	-49.01
CH_3-CO-H	1.25	1.24	130	128	-53.72	-54.72
CH_3-CO-OMe	1.21	1.22	129	136	-57.04	-53.21
CH_3-CO-CH_3	1.20	1.20	129	129	-58.58	-58.70
C_2H_5-CO-CH_3	1.20	1.20	129	128	-58.67	-59.05
H-CO-NH_2	1.20	1.20	128	134	-65.45	-61.94
H-CO-NO_2	1.36	1.41	142	121	-17.98	-25.29

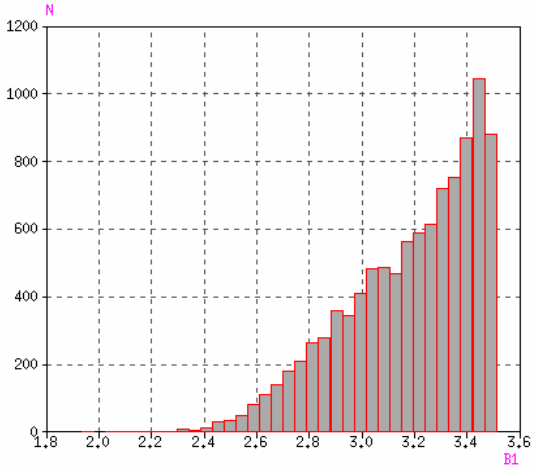




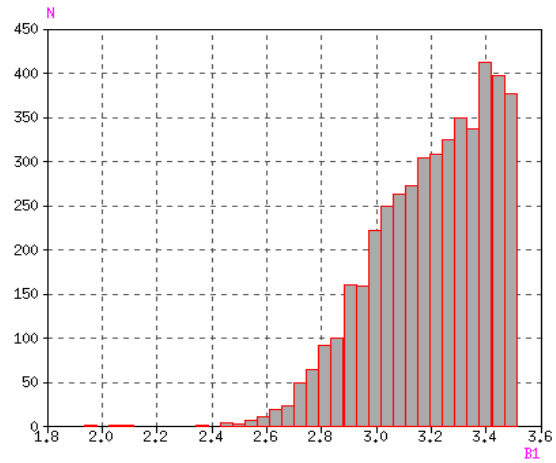
PyN⋯H—O



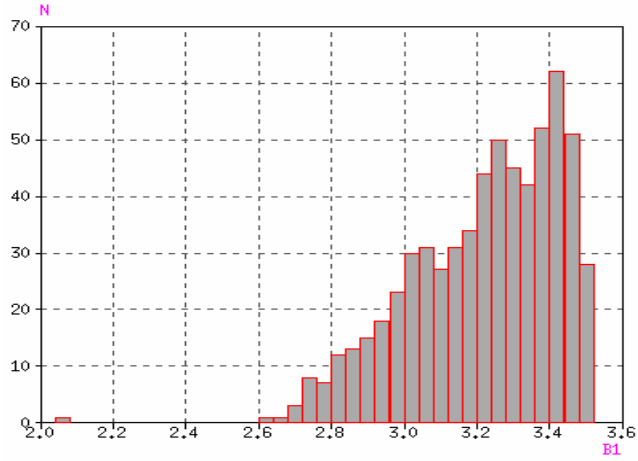
PyN⋯H—N



PyN⋯H—C

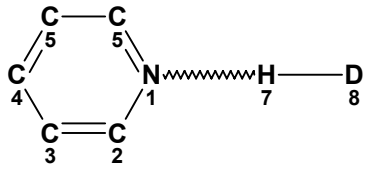


PyN⋯H—CH_2—C



PyN⋯H—CH_2—CH_2

Lakshmi et al. Figure S1



Printout of the JNL file used for searching C=O----H-N contacts from CSD.

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AT2 O 1                         :XY   410   496
AT3 H 1                         :XY   590   505
AT4 N 1                         :XY   797   511
BO 1 2 2
BO 3 4 1
GEOM
DEFINE B1 2 3
DEFINE B2 3 4
DEFINE A1 1 2 3
DEFINE A2 2 3 4
DEFINE T1 1 2 3 4
CONTACT INTER 2 A 3 A 0.5 3.5
C Search for ALL unique crystal fragments
NFRAG -99
C Overlap of crystal fragments permitted
C Rejection of symmetry-equivalent crystal fragments is ON
SYMCHK ON
ENANT NOIN
END
T2 *RFACTOR .LE. 4.0000
COMMENT Turning ON "INSIST-ON-COORDS"
SCREEN  153
COMMENT Turning ON "INSIST-NO-DISORDER"
SCREEN   35
COMMENT Turning ON "INSIST-ERROR-FREE"
SCREEN   33
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QUEST T1.AND.T2