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Supporting information for article:

Insights on structure and interactions of 2-amino-4-methoxy-6-methyl-pyrimidinium salts with 4-aminosalicylate and 5-chlorosalicylate: a combined experimental and theoretical charge–density analysis

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Computing details

Program(s) used to refine structure: *SHELXL97* (Sheldrick, 1997); molecular graphics: *ORTEP* for Windows (Farrugia,2012); software used to prepare material for publication: *WinGX* publication routines (Farrugia,2012).

Table (S1): ***2-amino 4-methoxy 6-methyl pyrimidine – 4-amino salicylic acid***

Crystal data	$F(000) = 616$
$C_{13}H_{16}N_4O_4$	$D_x = 1.392$ Mg m^{-3}
$M_r = 292.3$	Mo $K\alpha$ radiation, $\lambda = 0.71073$ Å
Monoclinic, $P2_1/n$	Cell parameters from 6906 reflections
$a = 8.8829$ (2) Å	$\theta = 2.7–28.2°$
$b = 15.1248$ (3) Å	$\mu = 0.11$ mm^{-1}
$c = 10.4933$ (3) Å	$T = 293$ K
$\beta = 98.392$ (1)°	Block
$V = 1394.70$ (6) Å^3	0.28 × 0.23 × 0.18 mm
$Z = 4$	
Data collected	
Bruker D8 Quest eco diffractometer	$R_{int} = 0.019$
8088 measured reflections	$\theta_{max} = 25.9°$, $\theta_{min} = 2.7°$
2690 independent reflections	$h = -8\rightarrow10$
2573 reflections with $I > 2s(I)$	$k = -18\rightarrow17$
Refinement	$l = -12\rightarrow12$
Refinement on F^2	
Least-squares matrix: full	1 restraints
$R[F^2 > 2\sigma(F^2)] = 0.051$	Primary atom site location: structure-invariant direct methods
$wR(F^2) = 0.126$	Secondary atom site location: difference Fourier map
$S = 1.17$	Hydrogen site location: mixed
2690 reflections	$w = 1/[\sigma^2(F_o^2) + (0.0578P)^2 + 0.2501P]$ where $P = (F_o^2 + 2F_c^2)/3$
228 parameters	
H atoms treated by a mixture of independent and constrained refinement	$\Delta\rho_{max} = 0.19$ e Å^{-3} $\Delta\rho_{min} = -0.14$ e Å^{-3}

Special details

Geometry: Allesds(expect the esd in the dihedral angle between two l.s planes) are estimated using the full convariance matrix. The cell esds are taken into account individually in the estimation of esds in distances, angles and torsion angles; correlations between esds in cell parameters are only used when they are defined by crystal symmetry. An approximate (isotropic) treatment of cell esds is used for estimating esds involving l.s. planes.

Table (S2): ***Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (Å^2)***

Atom	*x*	*y*	*z*	U_{iso}*/U_{eq}
H8	0.557 (2)	0.2155 (14)	0.9816 (19)	0.062 (6)*
H11	0.119 (2)	0.3848 (12)	0.8608 (16)	0.043 (4)*
H4	0.509 (3)	0.307 (2)	0.678 (3)	0.111 (9)*
H3	0.473 (2)	0.5151 (13)	0.8772 (19)	0.058 (6)*
H3A	0.882 (2)	0.5480 (14)	0.540 (2)	0.059 (6)*
H13	0.646 (2)	0.5910 (14)	0.566 (2)	0.067 (6)*
H4B	0.463 (3)	0.1857 (16)	1.177 (2)	0.066 (7)*
H3B	0.984 (3)	0.4953 (15)	0.651 (2)	0.065 (6)*
H4A	0.314 (2)	0.2248 (13)	1.206 (2)	0.054 (6)*
O2	0.16222 (13)	0.41781 (8)	0.63413 (11)	0.0499 (3)
N2	0.80749 (15)	0.47671 (9)	0.80428 (12)	0.0423 (3)
C12	0.31316 (17)	0.33716 (10)	0.79760 (14)	0.0357 (4)
O1	0.70149 (14)	0.43536 (9)	0.98058 (11)	0.0527 (4)
O3	0.37952 (14)	0.36188 (9)	0.59087 (11)	0.0529 (4)
N1	0.65577 (15)	0.56046 (9)	0.64433 (13)	0.0420 (3)
C13	0.28234 (18)	0.37514 (11)	0.66724 (15)	0.0387 (4)
O4	0.54959 (14)	0.27302 (10)	0.75865 (13)	0.0610 (4)
C11	0.21193 (18)	0.35023 (11)	0.88628 (15)	0.0378 (4)
C4	0.69030 (19)	0.47687 (11)	0.86762 (15)	0.0405 (4)
C9	0.36391 (18)	0.26076 (11)	1.04421 (15)	0.0418 (4)
N3	0.90016 (19)	0.52269 (12)	0.62232 (16)	0.0593 (5)
N4	0.3878 (2)	0.22401 (13)	1.16520 (16)	0.0608 (5)
C10	0.23558 (18)	0.31375 (11)	1.00685 (15)	0.0410 (4)
H10	0.1667	0.3241	1.0639	0.049*
C2	0.53537 (18)	0.56017 (11)	0.71129 (16)	0.0403 (4)
C3	0.55101 (19)	0.51842 (12)	0.82589 (16)	0.0445 (4)
C7	0.44378 (18)	0.28625 (11)	0.83790 (16)	0.0411 (4)
C1	0.78794 (19)	0.51967 (11)	0.69082 (15)	0.0419 (4)
C8	0.4677 (2)	0.24902 (12)	0.95922 (17)	0.0470 (4)
C6	0.8393 (3)	0.38770 (16)	1.0263 (2)	0.0688 (6)
H6A	0.8568	0.3438	0.9639	0.103*
H6B	0.8294	0.3594	1.1065	0.103*
H6C	0.9234	0.4281	1.0388	0.103*
C5	0.3953 (2)	0.60699 (14)	0.65024 (19)	0.0548 (5)
H5A	0.3104	0.5889	0.6911	0.082*

H5B	0.3758	0.5925	0.5603	0.082*
H5C	0.4096	0.6697	0.6601	0.082*

Table (S3): ***Atomic displacement parameters (Å^2)***

Atom	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
O2	0.0450 (7)	0.0616 (8)	0.0434 (7)	0.0130 (6)	0.0079 (5)	0.0123 (6)
N2	0.0459 (8)	0.0471 (8)	0.0362 (7)	0.0073 (6)	0.0133 (6)	0.0057 (6)
C12	0.0357 (8)	0.0357 (8)	0.0355 (8)	-0.0021 (6)	0.0047 (6)	0.0008 (6)
O1	0.0562 (7)	0.0644 (8)	0.0409 (7)	0.0102 (6)	0.0184 (6)	0.0144 (6)
O3	0.0505 (7)	0.0701 (9)	0.0411 (7)	0.0132 (6)	0.0166 (5)	0.0125 (6)
N1	0.0437 (8)	0.0464 (8)	0.0376 (7)	0.0072 (6)	0.0119 (6)	0.0054 (6)
C13	0.0390 (8)	0.0387 (8)	0.0384 (8)	-0.0009 (7)	0.0056 (6)	0.0015 (7)
O4	0.0494 (7)	0.0853 (10)	0.0517 (8)	0.0247 (7)	0.0187 (6)	0.0140 (7)
C11	0.0359 (8)	0.0394 (9)	0.0381 (8)	-0.0009 (7)	0.0048 (6)	-0.0005 (6)
C4	0.0468 (9)	0.0417 (9)	0.0351 (8)	-0.0007 (7)	0.0128 (7)	0.0006 (7)
C9	0.0456 (9)	0.0424 (9)	0.0363 (8)	-0.0064 (7)	0.0018 (7)	0.0030 (7)
N3	0.0516 (9)	0.0817 (13)	0.0494 (9)	0.0243 (9)	0.0236 (7)	0.0279 (9)
N4	0.0611 (11)	0.0755 (12)	0.0458 (9)	0.0092 (9)	0.0084 (8)	0.0211 (8)
C10	0.0415 (9)	0.0460 (9)	0.0366 (8)	-0.0029 (7)	0.0094 (7)	-0.0011 (7)
C2	0.0408 (8)	0.0390 (8)	0.0422 (9)	0.0008 (7)	0.0094 (7)	-0.0042 (7)
C3	0.0425 (9)	0.0509 (10)	0.0431 (9)	0.0009 (7)	0.0160 (7)	-0.0001 (7)
C7	0.0383 (8)	0.0445 (9)	0.0412 (9)	0.0032 (7)	0.0086 (7)	0.0020 (7)
C1	0.0448 (9)	0.0445 (9)	0.0384 (8)	0.0068 (7)	0.0130 (7)	0.0040 (7)
C8	0.0430 (9)	0.0499 (10)	0.0469 (10)	0.0080 (8)	0.0025 (7)	0.0068 (8)
C6	0.0699 (13)	0.0839 (15)	0.0553 (12)	0.0228 (11)	0.0178 (10)	0.0276 (11)
C5	0.0458 (10)	0.0592 (12)	0.0596 (11)	0.0089 (8)	0.0081 (8)	0.0032 (9)

Table (S4): ***Geometric parameters (Å, º)***

O2—C13	1.2521 (19)	C9—C10	1.402 (2)
N2—C4	1.314 (2)	N3—C1	1.312 (2)
N2—C1	1.345 (2)	N3—H3A	0.94 (2)
C12—C11	1.399 (2)	N3—H3B	0.87 (2)
C12—C7	1.405 (2)	N4—H4B	0.88 (2)
C12—C13	1.472 (2)	N4—H4A	0.83 (2)
O1—C4	1.3322 (19)	C10—H10	0.93
O1—C6	1.441 (2)	C2—C3	1.347 (2)
O3—C13	1.2765 (19)	C2—C5	1.493 (2)
N1—C1	1.353 (2)	C3—H3	0.94 (2)
N1—C2	1.363 (2)	C7—C8	1.380 (2)
N1—H13	0.94 (2)	C8—H8	0.94 (2)
O4—C7	1.3577 (19)	C6—H6A	0.96
O4—H4	1.01 (3)	C6—H6B	0.96
C11—C10	1.368 (2)	C6—H6C	0.96
C11—H11	0.981 (18)	C5—H5A	0.96

C4—C3	1.400 (2)	C5—H5B	0.96
C9—N4	1.374 (2)	C5—H5C	0.96
C9—C8	1.385 (2)		
C4—N2—C1	115.93 (14)	C9—C10—H10	120.1
C11—C12—C7	117.38 (14)	C3—C2—N1	118.38 (15)
C11—C12—C13	120.83 (14)	C3—C2—C5	125.20 (15)
C7—C12—C13	121.79 (14)	N1—C2—C5	116.42 (15)
C4—O1—C6	118.77 (14)	C2—C3—C4	117.45 (15)
C1—N1—C2	121.18 (14)	C2—C3—H3	123.3 (12)
C1—N1—H13	120.2 (13)	C4—C3—H3	119.2 (12)
C2—N1—H13	118.6 (13)	O4—C7—C8	118.53 (15)
O2—C13—O3	122.26 (14)	O4—C7—C12	120.76 (14)
O2—C13—C12	119.63 (14)	C8—C7—C12	120.71 (15)
O3—C13—C12	118.10 (14)	N3—C1—N2	119.43 (15)
C7—O4—H4	104.1 (16)	N3—C1—N1	118.38 (15)
C10—C11—C12	122.08 (15)	N2—C1—N1	122.19 (14)
C10—C11—H11	118.5 (10)	C7—C8—C9	120.94 (16)
C12—C11—H11	119.4 (10)	C7—C8—H8	117.5 (12)
N2—C4—O1	119.17 (15)	C9—C8—H8	121.6 (12)
N2—C4—C3	124.88 (15)	O1—C6—H6A	109.5
O1—C4—C3	115.96 (14)	O1—C6—H6B	109.5
N4—C9—C8	121.03 (17)	H6A—C6—H6B	109.5
N4—C9—C10	119.97 (17)	O1—C6—H6C	109.5
C8—C9—C10	118.95 (15)	H6A—C6—H6C	109.5
C1—N3—H3A	118.4 (12)	H6B—C6—H6C	109.5
C1—N3—H3B	118.2 (14)	C2—C5—H5A	109.5
H3A—N3—H3B	122.9 (19)	C2—C5—H5B	109.5
C9—N4—H4B	114.4 (14)	H5A—C5—H5B	109.5
C9—N4—H4A	116.3 (14)	C2—C5—H5C	109.5
H4B—N4—H4A	125 (2)	H5A—C5—H5C	109.5
C11—C10—C9	119.88 (15)	H5B—C5—H5C	109.5
C11—C10—H10	120.1		
C11—C12—C13—O2	-1.8 (2)	C5—C2—C3—C4	179.16 (16)
C7—C12—C13—O2	177.82 (15)	N1—C4—C3—C2	1.1 (3)
C11—C12—C13—O3	179.25 (15)	O1—C4—C3—C2	-179.28 (15)
C7—C12—C13—O3	-1.1 (2)	C11—C12—C7—O4	-177.92 (15)
C7—C12—C11—C10	-1.4 (2)	C13—C12—C7—O4	2.4 (2)
C13—C12—C11—C10	178.20 (15)	C11—C12—C7—C8	1.7 (2)
C1—N2—C4—O1	179.81 (14)	C13—C12—C7—C8	-177.93 (15)
C1—N2—C4—C3	-0.5 (3)	C4—N2—C1—N3	179.71 (17)
C6—O1—C4—N2	-3.2 (3)	C4—N2—C1—N1	-0.1 (2)
C6—O1—C4—C3	177.10 (17)	C2—N1—C1—N3	-179.58 (17)
C12—C11—C10—C9	-0.6 (2)	C2—N1—C1—N2	0.3 (3)
N4—C9—C10—C11	-179.95 (16)	O4—C7—C8—C9	179.68 (16)

C8—C9—C10—C11	2.3 (2)	C12—C7—C8—C9	0.0 (3)
C1—N1—C2—C3	0.3 (2)	N4—C9—C8—C7	-179.76 (17)
C1—N1—C2—C5	-179.75 (16)	C10—C9—C8—C7	-2.1 (3)
N1—C2—C3—C4	-0.9 (2)		

Table (S5): 2-amino 4-methoxy 6-methyl pyrimidine – 5-chloro salicylic acid

Crystal data	
$C_{13}H_{14}N_3O_4Cl$	$F(000) = 324$
$M_r = 311.72$	$D_x = 1.483$ Mg m^{-3}
Triclinic, $P\bar{1}$	Mo $K\alpha$ radiation, l = 0.71076 Å
$a = 8.329$ (6) Å	Cell parameters from 3294 reflections
$b = 9.962$ (8) Å	θ = 3.1–28.3°
$c = 9.998$ (7) Å	μ = 0.29 mm^{-1}
α = 111.66 (2)°	$T = 293$ K
β = 111.263 (13)°	Block
γ = 94.052 (18)°	0.33 × 0.16 × 0.10 mm
$V = 698.1$ (9) Å^3	
$Z = 2$	
Data collection	
Bruker D8 Quest Eco	$R_{int} = 0.030$
Absorption correction: multi-scan	$\theta_{max} = 28.3°$, $\theta_{min} = 2.5°$
9507 measured reflections	$h = -11 \rightarrow 11$
3433 independent reflections	$k = -13 \rightarrow 13$
2483 reflections with $I > 2\sigma(I)$	$l = -13 \rightarrow 13$
Refinement	
Refinement on F^2	
Least-squares matrix: full	1 restraints
$R[F^2 > 2\sigma(F^2)] = 0.064$	Primary atom site location: structure-invariant direct methods
$wR(F^2) = 0.162$	Secondary atom site location: difference Fourier map
$S = 1.08$	Hydrogen site location: mixed
3433 reflections	$w = 1/[\sigma^2(F_o^2) + (0.0738P)^2 + 0.2537P]$ where $P = (F_o^2 + 2F_c^2)/3$
221 parameters	
H atoms treated by a mixture of independent and constrained refinement	$\Delta\rho_{max} = 0.42$ e Å^{-3} $\Delta\rho_{min} = -0.27$ e Å^{-3}

Special details

***Geometry*.** All esds (except the esd in the dihedral angle between two l.s. planes) are estimated using the full covariance matrix. The cell esds are taken into account individually in the estimation of esds in distances, angles and torsion angles; correlations between esds in cell parameters are only used when they are defined by crystal symmetry. An approximate (isotropic) treatment of cell esds is used for estimating esds involving l.s. planes.

Table (S6): Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (Å^2)

	x	*y*	*z*	U_{iso}*/U_{eq}
H9	0.526 (4)	0.902 (3)	0.910 (3)	0.059 (8)*
H11	0.697 (4)	0.677 (3)	0.590 (3)	0.049 (8)*
H3	1.176 (3)	0.438 (3)	0.092 (3)	0.044 (7)*
H8	0.750 (4)	1.120 (3)	1.005 (3)	0.048 (7)*
H13	1.074 (4)	0.724 (3)	0.438 (4)	0.059 (8)*
H4	1.033 (5)	1.111 (4)	0.832 (4)	0.077 (11)*
Cl1	0.42523 (9)	0.62173 (8)	0.68319 (10)	0.0615 (3)
N1	1.1392 (2)	0.7017 (2)	0.3749 (2)	0.0354 (5)
N2	1.3851 (2)	0.7841 (2)	0.3361 (2)	0.0341 (4)
C4	1.3439 (3)	0.6505 (3)	0.2202 (3)	0.0380 (5)
C12	0.8394 (3)	0.8872 (3)	0.7207 (3)	0.0360 (5)
O3	1.0850 (2)	0.9862 (2)	0.6925 (2)	0.0553 (5)
O2	0.9380 (3)	0.7520 (2)	0.5297 (2)	0.0568 (5)
O1	1.4390 (2)	0.61839 (19)	0.1345 (2)	0.0483 (5)
N3	1.3071 (3)	0.9393 (2)	0.5279 (2)	0.0424 (5)
H3A	1.3943	1.0097	0.5548	0.051*
H3B	1.2388	0.9544	0.5764	0.051*
C3	1.2024 (3)	0.5346 (3)	0.1761 (3)	0.0450 (6)
C2	1.1004 (3)	0.5644 (3)	0.2587 (3)	0.0386 (5)
O4	0.9775 (3)	1.1457 (2)	0.8921 (3)	0.0530 (5)
C13	0.9628 (3)	0.8736 (3)	0.6413 (3)	0.0406 (6)
C1	1.2784 (3)	0.8089 (2)	0.4130 (3)	0.0321 (5)
C11	0.7070 (3)	0.7642 (3)	0.6725 (3)	0.0391 (6)
C10	0.5941 (3)	0.7747 (3)	0.7457 (3)	0.0408 (6)
C7	0.8553 (3)	1.0214 (3)	0.8433 (3)	0.0401 (6)

C8	0.7394 (4)	1.0286 (3)	0.9164 (4)	0.0520 (7)
C5	0.9474 (4)	0.4539 (3)	0.2308 (4)	0.0554 (7)
H5A	0.9724	0.4364	0.3238	0.083*
H5B	0.9286	0.3623	0.1425	0.083*
H5C	0.8427	0.4919	0.2083	0.083*
C9	0.6111 (4)	0.9056 (3)	0.8679 (4)	0.0529 (7)
C6	1.5901 (4)	0.7267 (3)	0.1751 (4)	0.0539 (7)
H6A	1.5546	0.8150	0.1701	0.081*
H6B	1.6425	0.6880	0.1020	0.081*
H6C	1.6750	0.7502	0.2807	0.081*

Table (S7): Atomic displacement parameters (Å^2)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
Cl1	0.0509 (4)	0.0591 (5)	0.0830 (6)	0.0011 (3)	0.0410 (4)	0.0283 (4)
N1	0.0358 (10)	0.0348 (10)	0.0385 (11)	0.0022 (8)	0.0238 (9)	0.0117 (9)
N2	0.0324 (9)	0.0351 (10)	0.0371 (11)	0.0058 (7)	0.0210 (8)	0.0118 (9)
C4	0.0383 (12)	0.0388 (12)	0.0415 (13)	0.0087 (9)	0.0238 (11)	0.0148 (11)
C12	0.0364 (12)	0.0413 (13)	0.0388 (13)	0.0106 (9)	0.0225 (10)	0.0188 (11)
O3	0.0514 (11)	0.0548 (12)	0.0598 (12)	-0.0031 (8)	0.0364 (10)	0.0145 (10)
O2	0.0606 (11)	0.0522 (11)	0.0616 (12)	0.0024 (9)	0.0472 (10)	0.0092 (10)
O1	0.0524 (10)	0.0436 (10)	0.0495 (11)	0.0061 (8)	0.0370 (9)	0.0061 (8)
N3	0.0412 (11)	0.0359 (10)	0.0466 (12)	-0.0031 (8)	0.0304 (10)	0.0043 (9)
C3	0.0505 (14)	0.0340 (13)	0.0473 (15)	0.0026 (10)	0.0282 (12)	0.0076 (12)
C2	0.0395 (12)	0.0335 (12)	0.0412 (13)	0.0023 (9)	0.0195 (11)	0.0129 (11)
O4	0.0582 (12)	0.0372 (10)	0.0613 (13)	0.0039 (8)	0.0333 (10)	0.0116 (9)
C13	0.0399 (13)	0.0461 (14)	0.0413 (14)	0.0065 (10)	0.0248 (11)	0.0171 (12)
C1	0.0322 (11)	0.0336 (11)	0.0338 (12)	0.0055 (8)	0.0180 (9)	0.0143 (10)
C11	0.0427 (13)	0.0369 (13)	0.0417 (14)	0.0078 (10)	0.0265 (11)	0.0126 (11)
C10	0.0387 (12)	0.0428 (13)	0.0506 (15)	0.0084 (10)	0.0266 (11)	0.0224 (12)
C7	0.0427 (13)	0.0388 (13)	0.0428 (14)	0.0099 (10)	0.0220 (11)	0.0175 (11)
C8	0.0658 (18)	0.0435 (15)	0.0551 (17)	0.0177 (13)	0.0427 (15)	0.0116 (13)
C5	0.0552 (16)	0.0378 (14)	0.0685 (19)	-0.0056 (11)	0.0352 (14)	0.0115 (13)
C9	0.0592 (16)	0.0543 (16)	0.0637 (18)	0.0174 (13)	0.0463 (15)	0.0234 (14)
C6	0.0512 (15)	0.0532 (16)	0.0588 (17)	0.0066 (12)	0.0394 (14)	0.0104 (14)

Table (S8): Geometric parameters (Å, º)

Cl1—C10	1.746 (3)	C3—H3	0.96 (3)
N1—C2	1.348 (3)	C2—C5	1.494 (3)
N1—C1	1.354 (3)	O4—C7	1.350 (3)
N1—H13	0.95 (3)	O4—H4	0.87 (4)
N2—C4	1.319 (3)	C11—C10	1.374 (3)
N2—C1	1.353 (3)	C11—H11	0.92 (3)
C4—O1	1.338 (3)	C10—C9	1.373 (4)
C4—C3	1.404 (3)	C7—C8	1.398 (4)
C12—C11	1.392 (3)	C8—C9	1.371 (4)
C12—C7	1.399 (4)	C8—H8	0.98 (3)
C12—C13	1.496 (3)	C5—H5A	0.9600
O3—C13	1.262 (3)	C5—H5B	0.9600
O2—C13	1.247 (3)	C5—H5C	0.9600
O1—C6	1.423 (3)	C9—H9	0.95 (3)
N3—C1	1.311 (3)	C6—H6A	0.9600
N3—H3A	0.8600	C6—H6B	0.9600
N3—H3B	0.8600	C6—H6C	0.9600
C3—C2	1.362 (3)		
C2—N1—C1	121.7 (2)	C10—C11—C12	120.2 (2)
C2—N1—H13	120.7 (19)	C10—C11—H11	121.7 (17)
C1—N1—H13	117.5 (19)	C12—C11—H11	118.1 (17)
C4—N2—C1	116.17 (19)	C9—C10—C11	120.7 (2)
N2—C4—O1	120.0 (2)	C9—C10—Cl1	118.95 (19)
N2—C4—C3	124.6 (2)	C11—C10—Cl1	120.3 (2)
O1—C4—C3	115.4 (2)	O4—C7—C8	118.7 (2)
C11—C12—C7	119.3 (2)	O4—C7—C12	122.1 (2)
C11—C12—C13	119.6 (2)	C8—C7—C12	119.2 (2)
C7—C12—C13	121.1 (2)	C9—C8—C7	120.4 (3)
C4—O1—C6	119.38 (19)	C9—C8—H8	118.6 (17)
C1—N3—H3A	120.0	C7—C8—H8	121.0 (17)
C1—N3—H3B	120.0	C2—C5—H5A	109.5

H3A—N3—H3B	120.0	C2—C5—H5B	109.5
C2—C3—C4	116.9 (2)	H5A—C5—H5B	109.5
C2—C3—H3	120.2 (16)	C2—C5—H5C	109.5
C4—C3—H3	122.8 (16)	H5A—C5—H5C	109.5
N1—C2—C3	118.8 (2)	H5B—C5—H5C	109.5
N1—C2—C5	116.9 (2)	C8—C9—C10	120.2 (2)
C3—C2—C5	124.4 (2)	C8—C9—H9	125.2 (18)
C7—O4—H4	100 (2)	C10—C9—H9	114.6 (18)
O2—C13—O3	124.7 (2)	O1—C6—H6A	109.5
O2—C13—C12	117.8 (2)	O1—C6—H6B	109.5
O3—C13—C12	117.5 (2)	H6A—C6—H6B	109.5
N3—C1—N2	120.42 (19)	O1—C6—H6C	109.5
N3—C1—N1	117.8 (2)	H6A—C6—H6C	109.5
N2—C1—N1	121.8 (2)	H6B—C6—H6C	109.5
C1—N2—C4—O1	177.7 (2)	C2—N1—C1—N3	-179.8 (2)
C1—N2—C4—C3	-1.8 (4)	C2—N1—C1—N2	0.4 (4)
N2—C4—O1—C6	3.4 (4)	C7—C12—C11—C10	-0.5 (4)
C3—C4—O1—C6	-177.1 (2)	C13—C12—C11—C10	179.3 (2)
N2—C4—C3—C2	0.9 (4)	C12—C11—C10—C9	-0.7 (4)
O1—C4—C3—C2	-178.7 (2)	C12—C11—C10—Cl1	178.82 (19)
C1—N1—C2—C3	-1.4 (4)	C11—C12—C7—O4	-177.9 (2)
C1—N1—C2—C5	178.0 (2)	C13—C12—C7—O4	2.3 (4)
C4—C3—C2—N1	0.8 (4)	C11—C12—C7—C8	0.9 (4)
C4—C3—C2—C5	-178.5 (3)	C13—C12—C7—C8	-178.8 (3)
C11—C12—C13—O2	2.4 (4)	O4—C7—C8—C9	178.6 (3)
C7—C12—C13—O2	-177.9 (2)	C12—C7—C8—C9	-0.3 (4)
C11—C12—C13—O3	-177.5 (2)	C7—C8—C9—C10	-0.8 (5)
C7—C12—C13—O3	2.2 (4)	C11—C10—C9—C8	1.3 (5)
C4—N2—C1—N3	-178.7 (2)	Cl1—C10—C9—C8	-178.2 (2)
C4—N2—C1—N1	1.2 (3)		

Table (S9) : ***Acidity of complexes 2A4M6MP base and aromatic carboxylic acids.***

Compounds	**pKa**	**ΔpKa**	**Crystalline form**

4-amino salicylic acid (4ASA)	3.68	2.09	Salt
5-chloro salicylic acid (5CLSA)	2.59	3.18	Salt

* The pKa value of 2-amino 4-methoxy 6-methyl pyrimidine (2A4M6MP) is 5.77, which was calculated using ChemAxon calculator, Marvin 15.2.16.0 (http://www.chemaxon.com)

Table (S10): Interaction Energies (kJ/mol) of Salt-I
R is the distance between molecular centroids (mean atomic position) in Å.

Total energies, only reported for two benchmarked energy models, are the sum of the four energy components, scaled appropriately (see the scale factor table below)

	N	Symop	R	Electron Density	E_ele	E_pol	E_dis	E_rep	E_tot
	0	-	4.32	B3LYP/6-31G(d,p)	-5.6	-4.9	-29.6	15.8	-25.5
	0	-	7.59	B3LYP/6-31G(d,p)	0.6	-0.7	-6.5	1.9	-4.4
	0	-	9.51	B3LYP/6-31G(d,p)	-3.0	-0.3	-0.6	0.0	-4.0
	0	-x, -y, -z	6.06	B3LYP/6-31G(d,p)	-9.0	-4.8	-19.0	29.7	-11.4
	0	-	7.10	B3LYP/6-31G(d,p)	-96.8	-22.6	-16.1	130.1	-52.7
	0	-	6.47	B3LYP/6-31G(d,p)	-41.0	-8.9	-15.4	40.2	-38.5
	0	-	11.27	B3LYP/6-31G(d,p)	0.7	-0.1	-0.2	0.0	0.6
	0	-x, -y, -z	6.39	B3LYP/6-31G(d,p)	-9.0	-2.0	-14.1	4.5	-20.5
	0	x, y, z	8.88	B3LYP/6-31G(d,p)	0.6	-0.3	-2.0	0.2	-1.2
	0	-	6.95	B3LYP/6-31G(d,p)	-7.0	-4.1	-10.6	3.6	-17.4
	0	-x+1/2, y+1/2, -z+1/2	10.82	B3LYP/6-31G(d,p)	-0.3	-0.0	-0.3	0.0	-0.6
	0	-	4.82	B3LYP/6-31G(d,p)	-1.4	-3.7	-26.3	13.8	-18.5
	0	-	6.67	B3LYP/6-31G(d,p)	3.3	-0.9	-13.0	9.8	-2.4
	0	-x+1/2, y+1/2, -z+1/2	7.68	B3LYP/6-31G(d,p)	-1.7	-0.3	-2.0	0.1	-3.8
	0	x+1/2, -y+1/2, z+1/2	10.67	B3LYP/6-31G(d,p)	0.0	-0.0	-0.2	0.0	-0.2
	0	x+1/2, -y+1/2, z+1/2	10.01	B3LYP/6-31G(d,p)	0.7	-0.0	-0.4	0.0	0.3
	0	-	6.48	B3LYP/6-31G(d,p)	14.1	-2.2	-7.8	2.5	8.0
	0	-x, -y, -z	8.67	B3LYP/6-31G(d,p)	10.9	-2.3	-6.4	3.2	6.2
	0	-	10.87	B3LYP/6-31G(d,p)	1.8	-0.3	-0.4	0.0	1.3

	0	x+1/2, -y+1/2, z+1/2	9.75	B3LYP/6-31G(d,p)	0.9	-0.1	-0.5	0.0	0.4
	0	-	10.87	B3LYP/6-31G(d,p)	0.7	-0.1	-0.3	0.0	0.3
	0	-x+1/2, y+1/2, -z+1/2	12.30	B3LYP/6-31G(d,p)	0.3	-0.0	-0.1	0.0	0.2
	0	-	10.55	B3LYP/6-31G(d,p)	-2.7	-0.3	-0.5	0.0	-3.5
	0	-x, -y, -z	6.62	B3LYP/6-31G(d,p)	-4.3	-1.4	-11.0	5.3	-11.9
	0	-x, -y, -z	13.69	B3LYP/6-31G(d,p)	0.2	-0.0	-0.1	0.0	0.2
	0	-	12.38	B3LYP/6-31G(d,p)	-0.4	-0.0	-0.1	0.0	-0.5
	0	x+1/2, -y+1/2, z+1/2	10.43	B3LYP/6-31G(d,p)	-0.3	-0.1	-0.3	0.0	-0.6
	0	-	12.32	B3LYP/6-31G(d,p)	2.6	-0.1	-0.1	0.0	2.5
	0	x, y, z	10.49	B3LYP/6-31G(d,p)	-1.9	-0.1	-0.4	0.0	-2.4
	0	-	10.28	B3LYP/6-31G(d,p)	-1.2	-0.1	-0.4	0.0	-1.7
	0	-	11.50	B3LYP/6-31G(d,p)	1.9	-0.1	-0.1	0.0	1.9
	0	-x, -y, -z	12.82	B3LYP/6-31G(d,p)	-0.5	-0.0	-0.1	0.0	-0.6
	0	-	12.16	B3LYP/6-31G(d,p)	1.1	-0.2	-0.2	0.0	0.9
	0	-	14.60	B3LYP/6-31G(d,p)	-0.8	-0.0	-0.0	0.0	-0.9
	0	x+1/2, -y+1/2, z+1/2	15.63	B3LYP/6-31G(d,p)	0.2	-0.0	-0.0	0.0	0.2
	0	-	13.89	B3LYP/6-31G(d,p)	-1.7	-0.0	-0.0	0.0	-1.9
	0	-	12.97	B3LYP/6-31G(d,p)	-1.1	-0.1	-0.1	0.0	-1.3
	0	-	12.80	B3LYP/6-31G(d,p)	0.3	-0.0	-0.1	0.0	0.2
	0	x+1/2, -y+1/2, z+1/2	16.89	B3LYP/6-31G(d,p)	-0.2	-0.0	-0.0	0.0	-0.2
	0	-x+1/2, y+1/2, -z+1/2	15.39	B3LYP/6-31G(d,p)	0.4	-0.0	-0.0	0.0	0.4
	0	-x+1/2, y+1/2, -z+1/2	14.76	B3LYP/6-31G(d,p)	-0.3	-0.0	-0.0	0.0	-0.4
	0	-x+1/2, y+1/2, -z+1/2	13.67	B3LYP/6-31G(d,p)	0.2	-0.0	-0.1	0.0	0.1
	0	-	11.88	B3LYP/6-31G(d,p)	-2.5	-0.2	-0.2	0.0	-2.9
	0	-	12.09	B3LYP/6-31G(d,p)	-0.9	-0.0	-0.1	0.0	-1.0
	0	-	17.71	B3LYP/6-31G(d,p)	0.4	-0.0	-0.0	0.0	0.5
	0	-	14.31	B3LYP/6-31G(d,p)	0.5	-0.0	-0.0	0.0	0.5
	0	-	12.50	B3LYP/6-31G(d,p)	1.3	-0.1	-0.1	0.0	1.2
	0	-	17.10	B3LYP/6-31G(d,p)	0.5	-0.0	-0.0	0.0	0.5

	1	-	4.32	B3LYP/6-31G(d,p)	-5.6	-4.9	-29.6	15.8	-25.5
	2	x+1/2, -y+1/2, z+1/2	6.65	B3LYP/6-31G(d,p)	-10.8	-8.2	-16.2	17.1	-21.1
	1	-	4.82	B3LYP/6-31G(d,p)	-1.4	-3.7	-26.3	13.8	-18.5
	1	-x, -y, -z	9.86	B3LYP/6-31G(d,p)	6.6	-0.5	-0.5	0.0	6.2
	2	x, y, z	8.88	B3LYP/6-31G(d,p)	6.6	-0.3	-0.7	0.0	6.2
	2	x+1/2, -y+1/2, z+1/2	7.60	B3LYP/6-31G(d,p)	-12.3	-3.6	-8.5	8.1	-18.1
	1	-	7.10	B3LYP/6-31G(d,p)	-96.8	-22.6	-16.1	130.1	-52.7
	1	-	6.47	B3LYP/6-31G(d,p)	-41.0	-8.9	-15.4	40.2	-38.5
	1	-	6.67	B3LYP/6-31G(d,p)	3.3	-0.9	-13.0	9.8	-2.4
	1	-x, -y, -z	6.86	B3LYP/6-31G(d,p)	-7.6	-0.4	-1.5	0.0	-9.6
	2	-x+1/2, y+1/2, -z+1/2	7.95	B3LYP/6-31G(d,p)	-9.3	-0.7	-1.1	0.0	-11.3
	1	-	6.48	B3LYP/6-31G(d,p)	14.1	-2.2	-7.8	2.5	8.0
	1	-	6.95	B3LYP/6-31G(d,p)	-7.0	-4.1	-10.6	3.6	-17.4
	1	-	9.51	B3LYP/6-31G(d,p)	-3.0	-0.3	-0.6	0.0	-4.0
	1	-x+1/2, y+1/2, -z+1/2	10.79	B3LYP/6-31G(d,p)	-2.7	-0.1	-0.2	0.0	-3.1
	1	-	11.85	B3LYP/6-31G(d,p)	-1.5	-0.1	-0.1	0.0	-1.8
	1	-x, -y, -z	10.45	B3LYP/6-31G(d,p)	8.7	-0.6	-0.5	0.0	8.4
	1	-	7.59	B3LYP/6-31G(d,p)	0.6	-0.7	-6.5	1.9	-4.4
	1	-	13.16	B3LYP/6-31G(d,p)	-0.9	-0.0	-0.1	0.0	-1.0
	1	-	11.85	B3LYP/6-31G(d,p)	0.1	-0.1	-0.1	0.0	0.0
	1	-x, -y, -z	9.29	B3LYP/6-31G(d,p)	-7.1	-0.3	-0.6	0.0	-8.3
	1	-	10.55	B3LYP/6-31G(d,p)	-2.7	-0.3	-0.5	0.0	-3.5
	1	-	12.38	B3LYP/6-31G(d,p)	-0.4	-0.0	-0.1	0.0	-0.5
	1	-x+1/2, y+1/2, -z+1/2	12.76	B3LYP/6-31G(d,p)	-0.5	-0.1	-0.1	0.0	-0.6
	1	-	9.28	B3LYP/6-31G(d,p)	2.3	-0.1	-0.4	0.0	2.0
	1	-	10.87	B3LYP/6-31G(d,p)	0.7	-0.1	-0.3	0.0	0.3
	1	-	11.50	B3LYP/6-31G(d,p)	1.9	-0.1	-0.1	0.0	1.9
	1	-	11.39	B3LYP/6-31G(d,p)	0.9	-0.1	-0.1	0.0	0.8
	1	-	14.48	B3LYP/6-31G(d,p)	0.2	-0.0	-0.0	0.0	0.1

	2	x, y, z	10.49	B3LYP/6-31G(d,p)	-7.7	-0.7	-0.7	0.0	-9.2
	1	-	12.32	B3LYP/6-31G(d,p)	2.6	-0.1	-0.1	0.0	2.5
	1	-	11.44	B3LYP/6-31G(d,p)	-1.8	-0.0	-0.1	0.0	-2.1
	1	-	10.87	B3LYP/6-31G(d,p)	1.8	-0.3	-0.4	0.0	1.3
	1	-	11.27	B3LYP/6-31G(d,p)	0.7	-0.1	-0.2	0.0	0.6
	1	-	10.28	B3LYP/6-31G(d,p)	-1.2	-0.1	-0.4	0.0	-1.7
	2	x, y, z	12.72	B3LYP/6-31G(d,p)	-0.4	-0.1	-0.1	0.0	-0.6
	1	-	12.09	B3LYP/6-31G(d,p)	-0.9	-0.0	-0.1	0.0	-1.0
	1	-x+1/2, y+1/2, -z+1/2	17.04	B3LYP/6-31G(d,p)	0.8	-0.0	-0.0	0.0	0.8
	1	-x+1/2, y+1/2, -z+1/2	14.49	B3LYP/6-31G(d,p)	1.9	-0.0	-0.0	0.0	2.0
	1	x+1/2, -y+1/2, z+1/2	15.14	B3LYP/6-31G(d,p)	0.4	-0.0	-0.0	0.0	0.4
	1	-	12.80	B3LYP/6-31G(d,p)	0.3	-0.0	-0.1	0.0	0.2
	1	-	11.88	B3LYP/6-31G(d,p)	-2.5	-0.2	-0.2	0.0	-2.9
	1	-	12.97	B3LYP/6-31G(d,p)	-1.1	-0.1	-0.1	0.0	-1.3
	1	-	13.89	B3LYP/6-31G(d,p)	-1.7	-0.0	-0.0	0.0	-1.9
	1	-x+1/2, y+1/2, -z+1/2	11.69	B3LYP/6-31G(d,p)	1.3	-0.2	-0.2	0.0	1.0
	1	x+1/2, -y+1/2, z+1/2	13.72	B3LYP/6-31G(d,p)	1.2	-0.0	-0.0	0.0	1.3
	2	x, y, z	14.70	B3LYP/6-31G(d,p)	-1.0	-0.0	-0.0	0.0	-1.1
	2	-x+1/2, y+1/2, -z+1/2	12.95	B3LYP/6-31G(d,p)	-2.6	-0.0	-0.1	0.0	-2.8

Energy Model	k_ele	k_pol	k_disp	k_rep
CE-HF ... HF/3-21G electron densities	1.019	0.651	0.901	0.811
CE-B3LYP ... B3LYP/6-31G(d,p) electron densities	1.057	0.740	0.871	0.618

Table (S11): Interaction Energies (kJ/mol) of Salt-II
R is the distance between molecular centroids (mean atomic position) in Å.

Total energies, only reported for two benchmarked energy models, are the sum of the

four energy components, scaled appropriately (see the scale factor table below)

	N	Symop	R	Electron Density	E_ele	E_pol	E_dis	E_rep	E_tot
	1	-	7.17	B3LYP/6-31G(d,p)	-96.0	-19.6	-16.1	151.9	-36.2
	0	-x, -y, -z	5.50	B3LYP/6-31G(d,p)	-0.9	-0.8	-19.0	7.6	-13.4
	0	-	7.79	B3LYP/6-31G(d,p)	-0.5	-2.2	-9.6	9.9	-4.4
	0	-x, -y, -z	8.71	B3LYP/6-31G(d,p)	-2.2	-2.7	-3.5	0.7	-7.0
	0	-	4.70	B3LYP/6-31G(d,p)	-14.3	-3.3	-29.5	15.8	-33.5
	0	-	9.70	B3LYP/6-31G(d,p)	-1.2	-0.2	-4.8	4.8	-2.7
	0	-	7.17	B3LYP/6-31G(d,p)	-96.0	-2.6	-16.1	151.9	-23.6
	0	x, y, z	8.33	B3LYP/6-31G(d,p)	-0.5	-0.5	-5.4	1.7	-4.5
	0	-	8.45	B3LYP/6-31G(d,p)	-1.5	-1.5	-5.0	1.2	-6.2
	0	-x, -y, -z	7.86	B3LYP/6-31G(d,p)	3.0	-1.1	-8.7	4.6	-2.4
	0	-	4.41	B3LYP/6-31G(d,p)	-0.8	-1.1	-26.4	15.8	-14.8
	0	-	5.27	B3LYP/6-31G(d,p)	-8.1	-2.2	-20.3	12.8	-19.9
	0	-	9.10	B3LYP/6-31G(d,p)	0.2	-1.1	-5.1	2.2	-3.7
	0	-x, -y, -z	6.13	B3LYP/6-31G(d,p)	-65.2	-19.0	-29.8	65.9	-68.3
	1	-x, -y, -z	7.82	B3LYP/6-31G(d,p)	9.1	-2.7	-10.3	7.8	3.4
	1	-x, -y, -z	6.90	B3LYP/6-31G(d,p)	-7.2	-0.8	-9.8	10.2	-10.4
	2	x, y, z	11.21	B3LYP/6-31G(d,p)	0.1	-0.1	-0.2	0.0	-0.2
	1	-	7.79	B3LYP/6-31G(d,p)	-0.5	-1.1	-9.6	9.9	-3.7
	1	-	5.27	B3LYP/6-31G(d,p)	-8.1	-2.2	-20.3	12.8	-19.9
	1	-	9.70	B3LYP/6-31G(d,p)	-1.2	-0.2	-4.8	4.8	-2.7
	1	-	9.10	B3LYP/6-31G(d,p)	0.2	-1.1	-5.1	2.2	-3.7
	1	-x, -y, -z	8.30	B3LYP/6-31G(d,p)	1.5	-1.5	-3.3	0.3	-2.2
	1	-x, -y, -z	9.09	B3LYP/6-31G(d,p)	-1.0	-0.3	-0.6	0.0	-1.8
	1	-	4.41	B3LYP/6-31G(d,p)	-0.8	-3.1	-26.4	15.8	-16.3
	1	-	4.70	B3LYP/6-31G(d,p)	-14.3	-3.3	-29.5	15.8	-33.5

	2	x, y, z	8.33	B3LYP/6-31G(d,p)	-3.2	-0.9	-2.8	0.1	-6.5
	1	-x, -y, -z	4.91	B3LYP/6-31G(d,p)	-9.7	-2.6	-27.1	11.7	-28.5
	1	-x, -y, -z	15.20	B3LYP/6-31G(d,p)	2.1	-0.0	-0.0	0.0	2.1
	1	-	9.52	B3LYP/6-31G(d,p)	2.4	-0.2	-0.5	0.0	2.0
	1	x, y, z	10.00	B3LYP/6-31G(d,p)	-0.7	-0.1	-0.3	0.0	-1.1
	1	-	14.90	B3LYP/6-31G(d,p)	-1.1	-0.0	-0.0	0.0	-1.2
	1	-x, -y, -z	6.76	B3LYP/6-31G(d,p)	-3.1	-0.2	-1.6	0.0	-4.8
	1	-	6.56	B3LYP/6-31G(d,p)	1.4	-0.3	-1.9	0.0	-0.3
	1	-	10.01	B3LYP/6-31G(d,p)	1.2	-0.1	-0.3	0.0	0.9
	1	-	9.26	B3LYP/6-31G(d,p)	3.8	-0.5	-1.1	0.0	2.7
	1	-	8.45	B3LYP/6-31G(d,p)	-1.5	-0.5	-5.0	1.2	-5.5
	1	-	10.72	B3LYP/6-31G(d,p)	1.5	-0.1	-0.3	0.0	1.2
	2	x, y, z	9.96	B3LYP/6-31G(d,p)	1.8	-0.0	-0.3	0.0	1.6
	2	x, y, z	12.53	B3LYP/6-31G(d,p)	-0.2	-0.0	-0.2	0.0	-0.4
	1	-	11.94	B3LYP/6-31G(d,p)	0.9	-0.0	-0.2	0.0	0.8
	1	-	12.60	B3LYP/6-31G(d,p)	0.1	-0.0	-0.1	0.0	0.0
	1	x, y, z	15.16	B3LYP/6-31G(d,p)	-1.4	-0.0	-0.0	0.0	-1.5
	1	-	10.04	B3LYP/6-31G(d,p)	-2.1	-0.1	-0.5	0.0	-2.8
	1	x, y, z	16.35	B3LYP/6-31G(d,p)	-0.8	-0.0	-0.0	0.0	-0.8
	1	-x, -y, -z	11.21	B3LYP/6-31G(d,p)	-0.9	-0.1	-0.2	0.0	-1.1
	1	-	9.27	B3LYP/6-31G(d,p)	-1.7	-0.1	-0.5	0.0	-2.3
	1	-	17.37	B3LYP/6-31G(d,p)	-0.1	-0.0	-0.0	0.0	-0.1
	1	-	16.80	B3LYP/6-31G(d,p)	0.2	-0.0	-0.0	0.0	0.1
	2	x, y, z	11.09	B3LYP/6-31G(d,p)	1.5	-0.0	-0.2	0.0	1.4
	1	-	17.13	B3LYP/6-31G(d,p)	-0.9	-0.0	-0.0	0.0	-0.9
	1	-	14.49	B3LYP/6-31G(d,p)	0.4	-0.0	-0.0	0.0	0.4
	1	-x, -y, -z	12.30	B3LYP/6-31G(d,p)	0.3	-0.0	-0.2	0.0	0.1
	1	-x, -y, -z	15.01	B3LYP/6-31G(d,p)	-0.3	-0.0	-0.0	0.0	-0.4
	2	x, y, z	18.80	B3LYP/6-31G(d,p)	-0.3	-0.0	-0.0	0.0	-0.4

	1	-	13.91	B3LYP/6-31G(d,p)	-0.8	-0.0	-0.1	0.0	-0.9
	1	-x, -y, -z	11.63	B3LYP/6-31G(d,p)	-1.6	-0.0	-0.2	0.0	-1.9
	1	-	12.62	B3LYP/6-31G(d,p)	0.9	-0.0	-0.2	0.0	0.7
	1	-	13.76	B3LYP/6-31G(d,p)	-0.2	-0.0	-0.0	0.0	-0.2
	1	-	14.97	B3LYP/6-31G(d,p)	1.2	-0.0	-0.0	0.0	1.2
	1	-	14.68	B3LYP/6-31G(d,p)	-0.4	-0.0	-0.0	0.0	-0.5
	1	-	14.95	B3LYP/6-31G(d,p)	1.0	-0.0	-0.0	0.0	1.0
	1	-	14.88	B3LYP/6-31G(d,p)	0.0	-0.0	-0.0	0.0	0.0
	1	-x, -y, -z	16.89	B3LYP/6-31G(d,p)	-0.4	-0.0	-0.0	0.0	-0.4
	1	-	15.45	B3LYP/6-31G(d,p)	-0.3	-0.0	-0.0	0.0	-0.4
	1	-	24.48	B3LYP/6-31G(d,p)	-0.1	-0.0	-0.0	0.0	-0.1
	1	x, y, z	22.32	B3LYP/6-31G(d,p)	-0.4	-0.0	-0.0	0.0	-0.5

Energy Model	k_ele	k_pol	k_disp	k_rep
CE-HF ... HF/3-21G electron densities	1.019	0.651	0.901	0.811
CE-B3LYP ... B3LYP/6-31G(d,p) electron densities	1.057	0.740	0.871	0.618

Table(S12) : Topological properties of salt I (1st row) and II (2nd row)

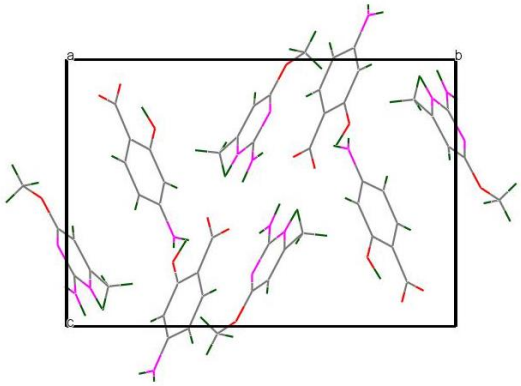
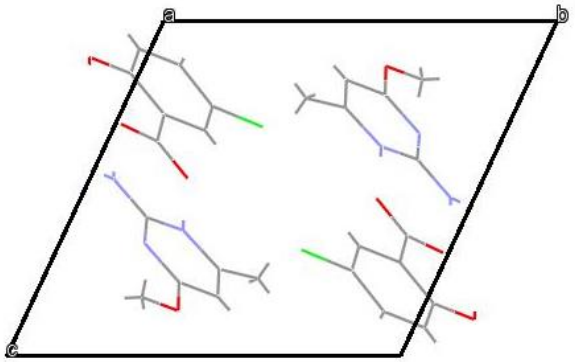
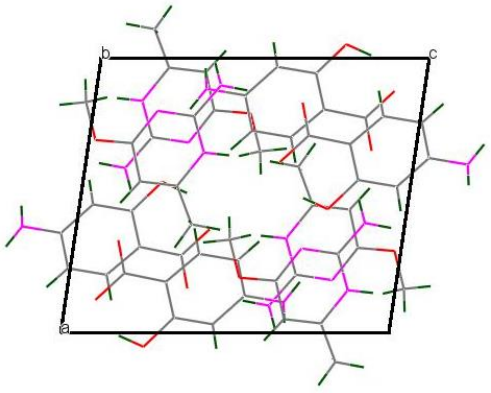
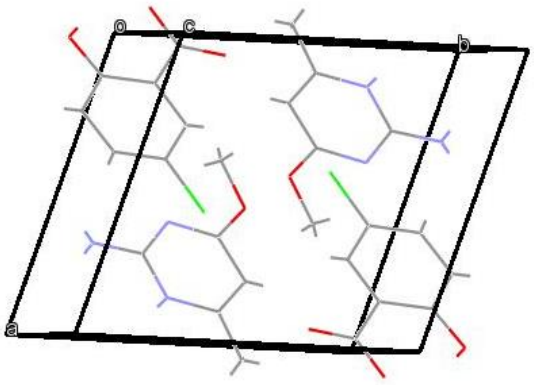
bonds	**rho**	**del2rho**
C2-C3	2.283	-24.492
	2.223	-23.213
C3-C4	2.091	-21.417

	2.074	-20.992
C5-C2	1.735	-15.147
	1.731	-15.077
C7-C12	2.07	-20.485
	2.099	-21.05
C7-C8	2.158	-22.311
	2.1	-21.273
C8-C9	2.122	-21.428
	2.156	-22.112
C9-C10	2.066	-20.572
	2.181	-22.783
C10-C11	2.176	-22.421
	2.165	-22.198
C11-C12	2.059	-20.198
	2.081	-20.562
C12-C13	1.854	-17.024
	1.785	-15.843
C1-N1	2.228	-24.023
	2.233	-24.41
C2-N1	2.124	-19.74
	2.177	-19.133
C1-N2	2.326	-26.348
	2.295	-25.561
C4-N2	2.452	-27.332
	2.425	-27.058
C1-N3	2.441	-27.461
	2.432	-27.008
N4-C9	2.089	-21.651
	-	-
C4-O1	2.093	-10.067
	2.07	-10.709
C6-O1	1.545	-3.969
	1.598	-2.723
C13-O2	2.555	-10.365
	2.56	-8.302
C13-O3	2.388	-9.885
	2.492	-10.543
C7-O4	1.991	-11.744
	2.025	-11.455
C10-Cl1	-	-
	1.321	-7.05

C3-H3	2.575	-41.019
	2.466	-38.068
C5-H5A	2.442	-36.817
	2.443	-36.969
C5-H5B	2.428	-36.455
	2.445	-36.881
C5-H5C	2.453	-37.266
	2.435	-36.708
C6-H6A	2.509	-39.247
	2.513	-39.334
C6-H6B	2.513	-39.563
	2.514	-39.564
C6-H6C	2.519	-39.536
	2.51	-39.245
C8-H8	2.526	-39.186
	2.367	-35.372
C9-H9	-	-
	2.523	-39.871
C10-H10	2.592	-40.734
	-	-
C11-H11	2.385	-36.097
	2.695	-44.873
O2-H3A	0.207	2.514
	-	-
O3-H3B	-	-
	0.162	2.244
O3-H13	0.267	3.146
	-	-
O2-H13	-	-
	0.343	3.676
H4-O3	0.426	3.78
	0.32	3.969
O4-H4	2.125	-49.829
	3.124	-90.162
H13-N1	2.725	-69.288
	2.61	-64.943
H3A-N3	2.702	-66.798
	3.342	-88.004
N3-H3B	3.28	-85.224
	3.325	-91.496
N4-H4A	3.567	-95.364

	-	-
H4B-N4	3.167	-76.645
	-	-

Figure (S13):Molecular packing view of along (a) a-axis (b) b- axis (c) c-axis for salt (I & II)

SALT I	**SALT II**
	
(a)	
	
(b)	

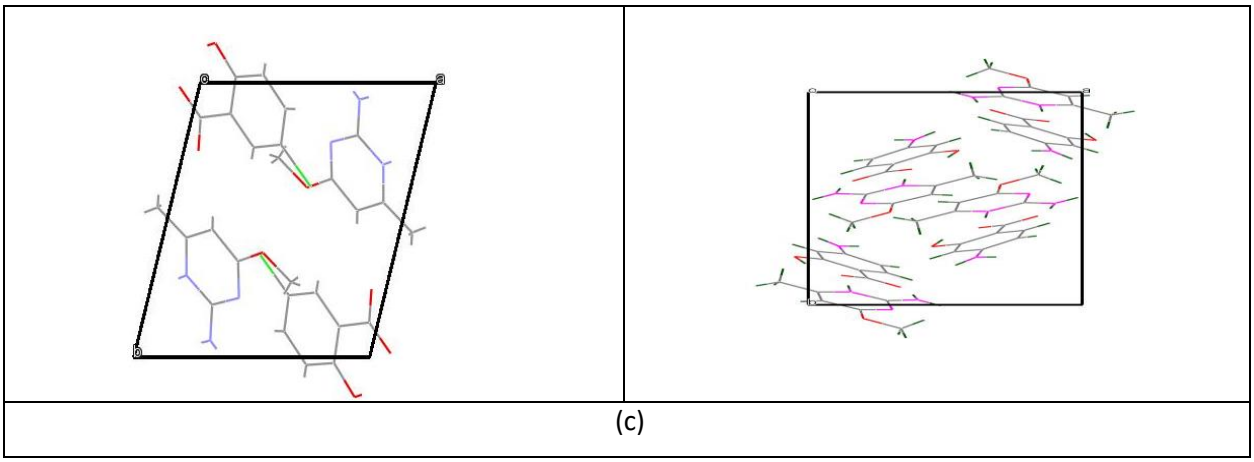


Figure (S14): Hydrogen bonding in the crystal of salt (I) showing the $R_2^2(8)$ and centrosymmetric $R_4^2(8)$ ring motifs.(salt – I)

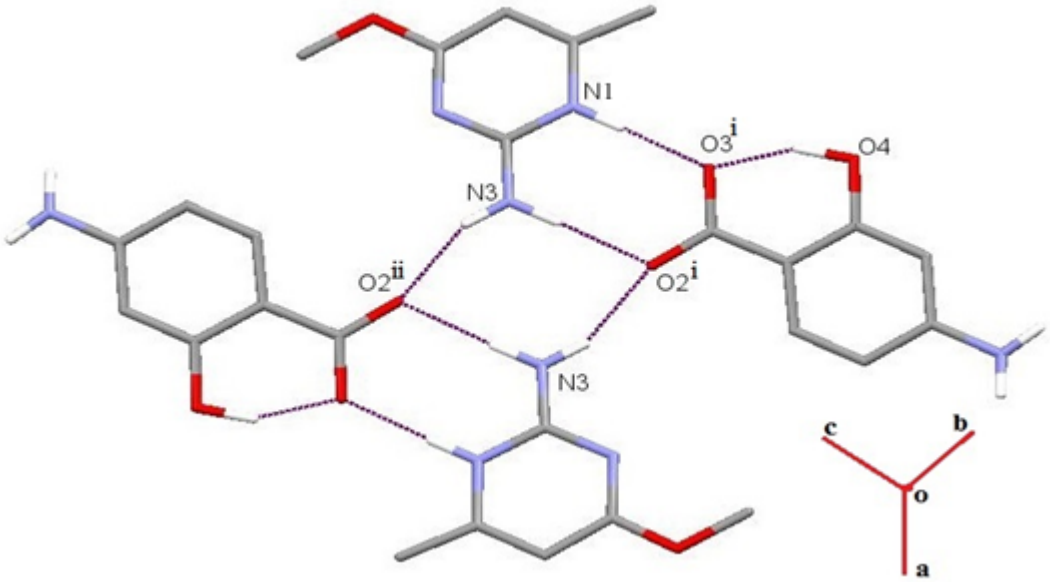


Figure (S15): A view of supramolecular chain formed in (I) via N−H···O interactions with dashed lines representing hydrogen bonds. (salt – I)

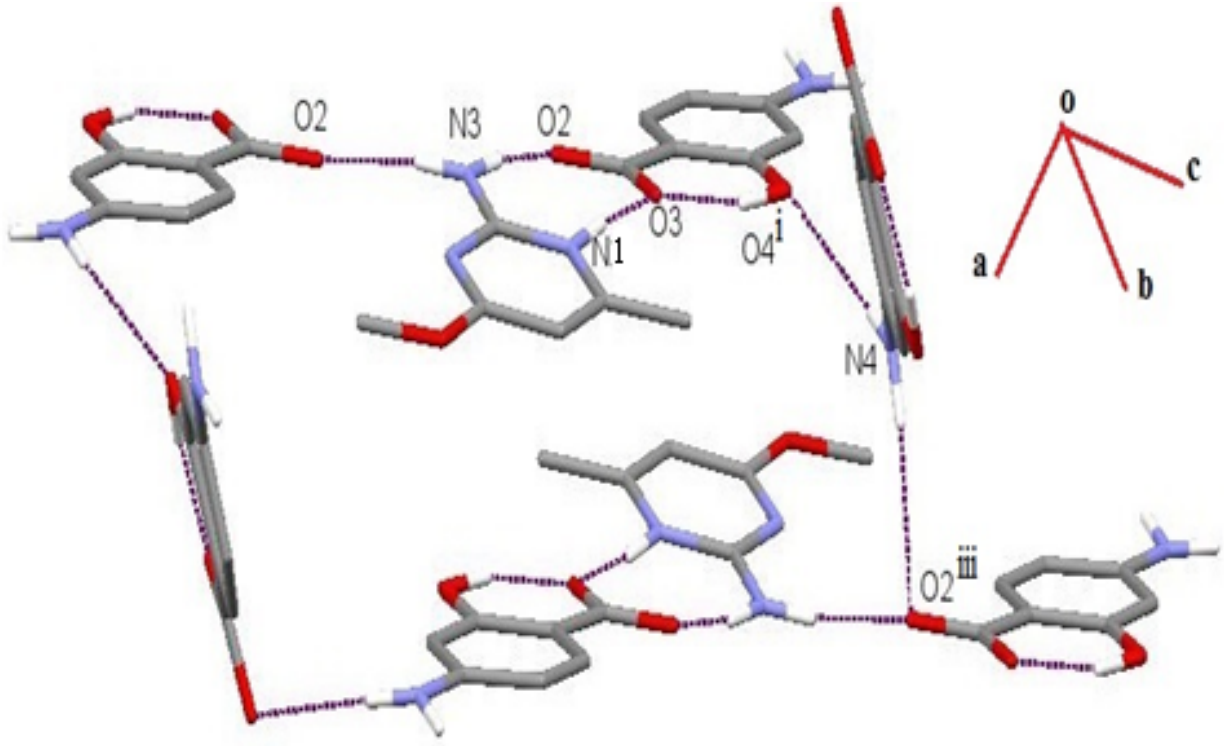


Figure S16: A molecular packing view of the supramolecular sheet. (salt – I)

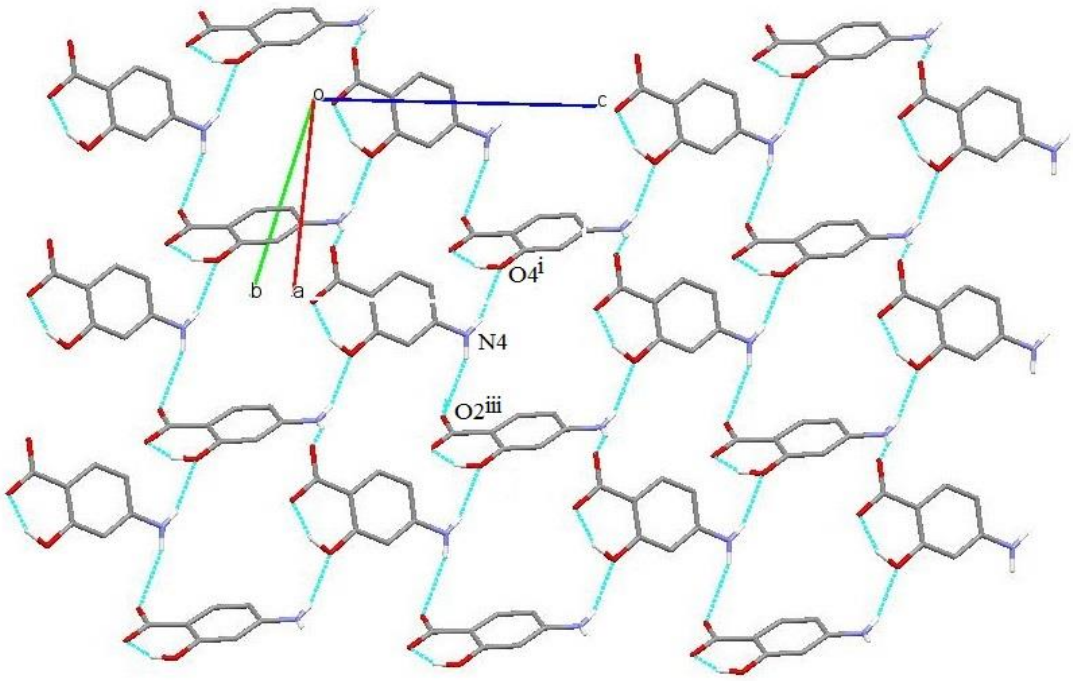


Figure S17: A view of the supramolecular structures. (salt – I)

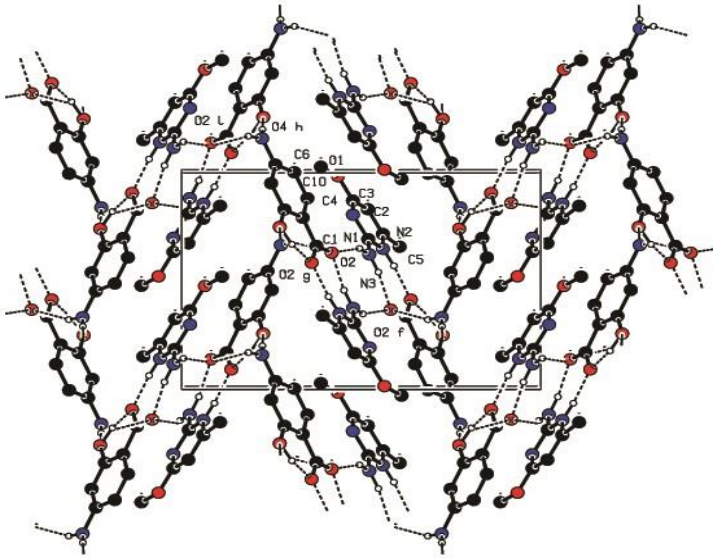


Figure (S18) : A molecular packing view of the cyclic heterotetramer. (salt – I)

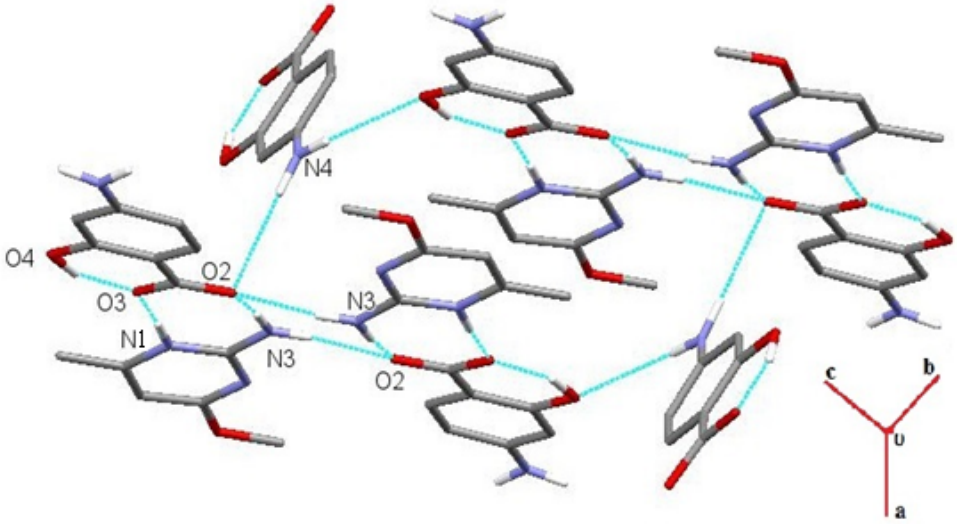


Figure (S19): A view of the C−H···π stacking interactions. (salt – I)

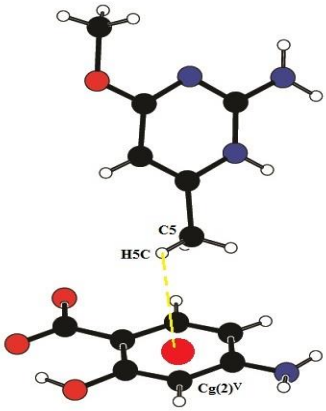


Figure (S20): A molecular packing view of linear heterotetramer. (salt – II)

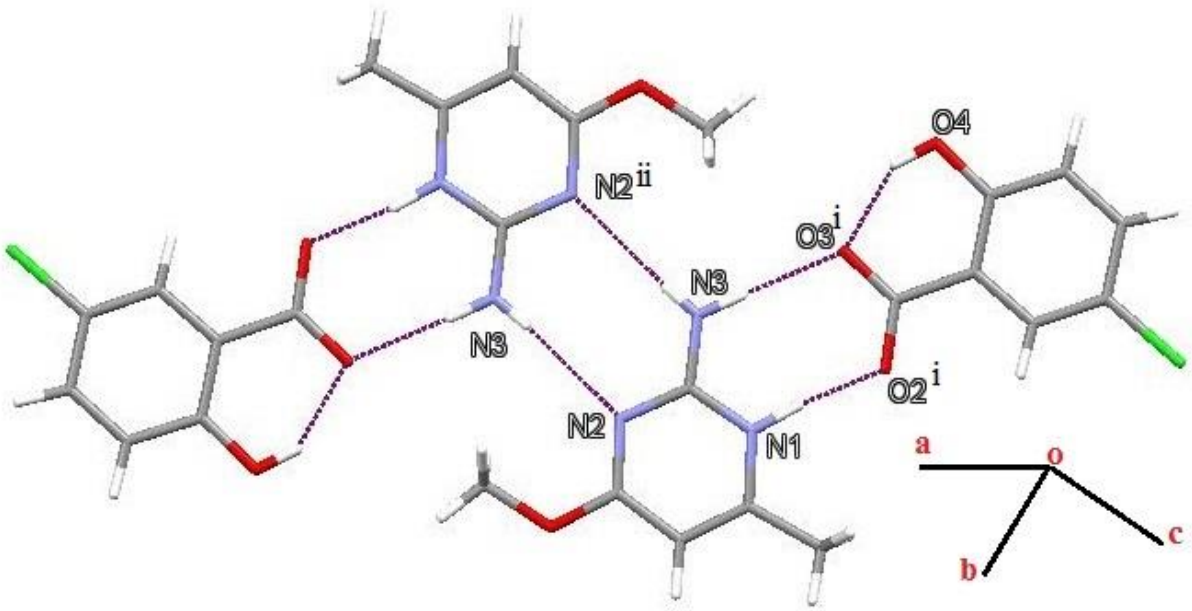


Figure (S21): A view of the supramolecular chain formed in (II) via N−H···N interactions in different planes. (salt – II)

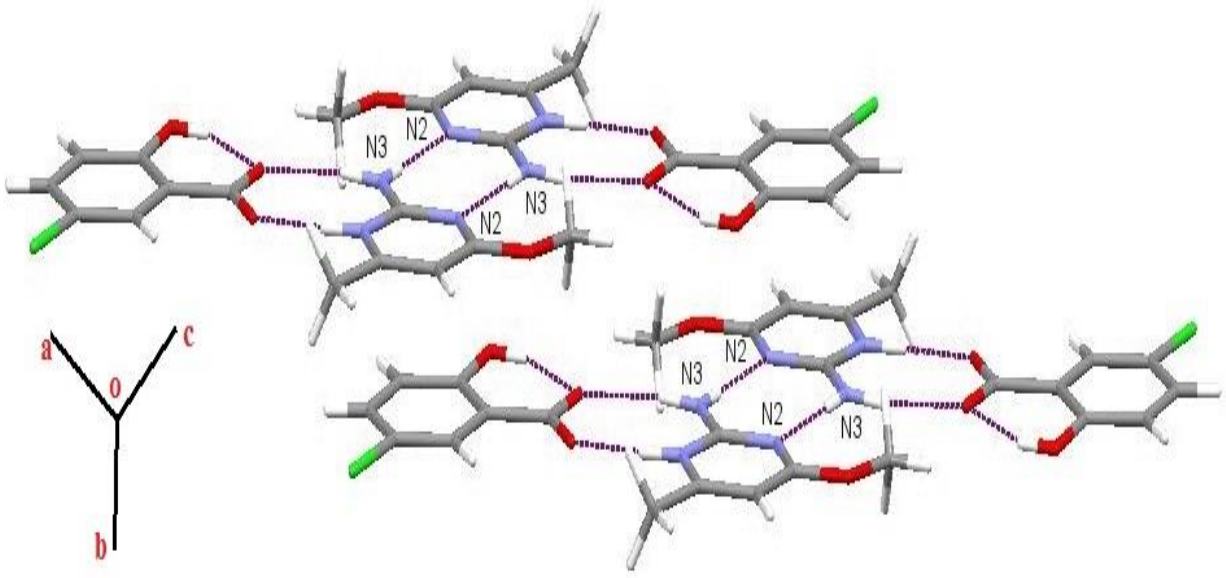


Figure (S22): A view of the C−Cl···π stacking interactions (salt – II)

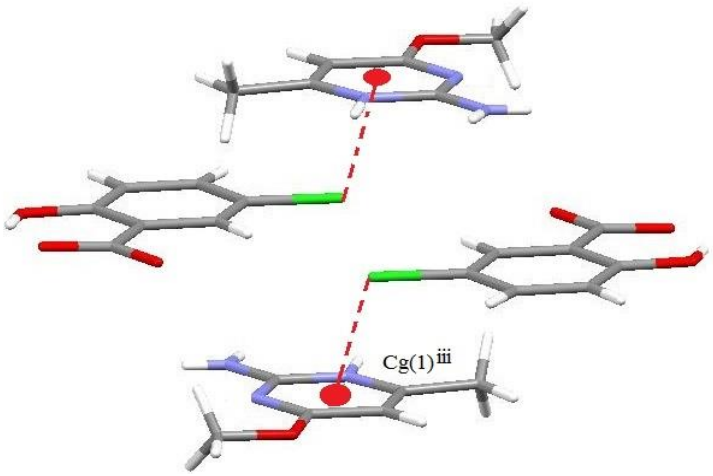


Figure (S23): Showing the dipole moment vector orientation of the salt (I & II) molecules. The origin is at the center of mass of the molecule.

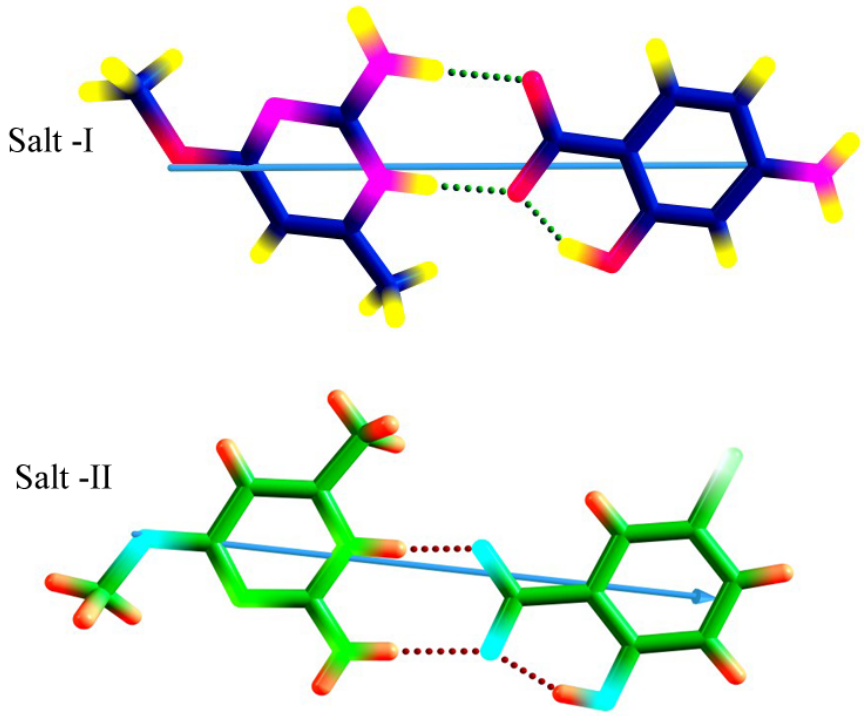


Figure (S24): *Three dimensional Hirshfeld surface of Salt (I) & (II): (a) di, (b) de (c) curvedness (d) shape index*

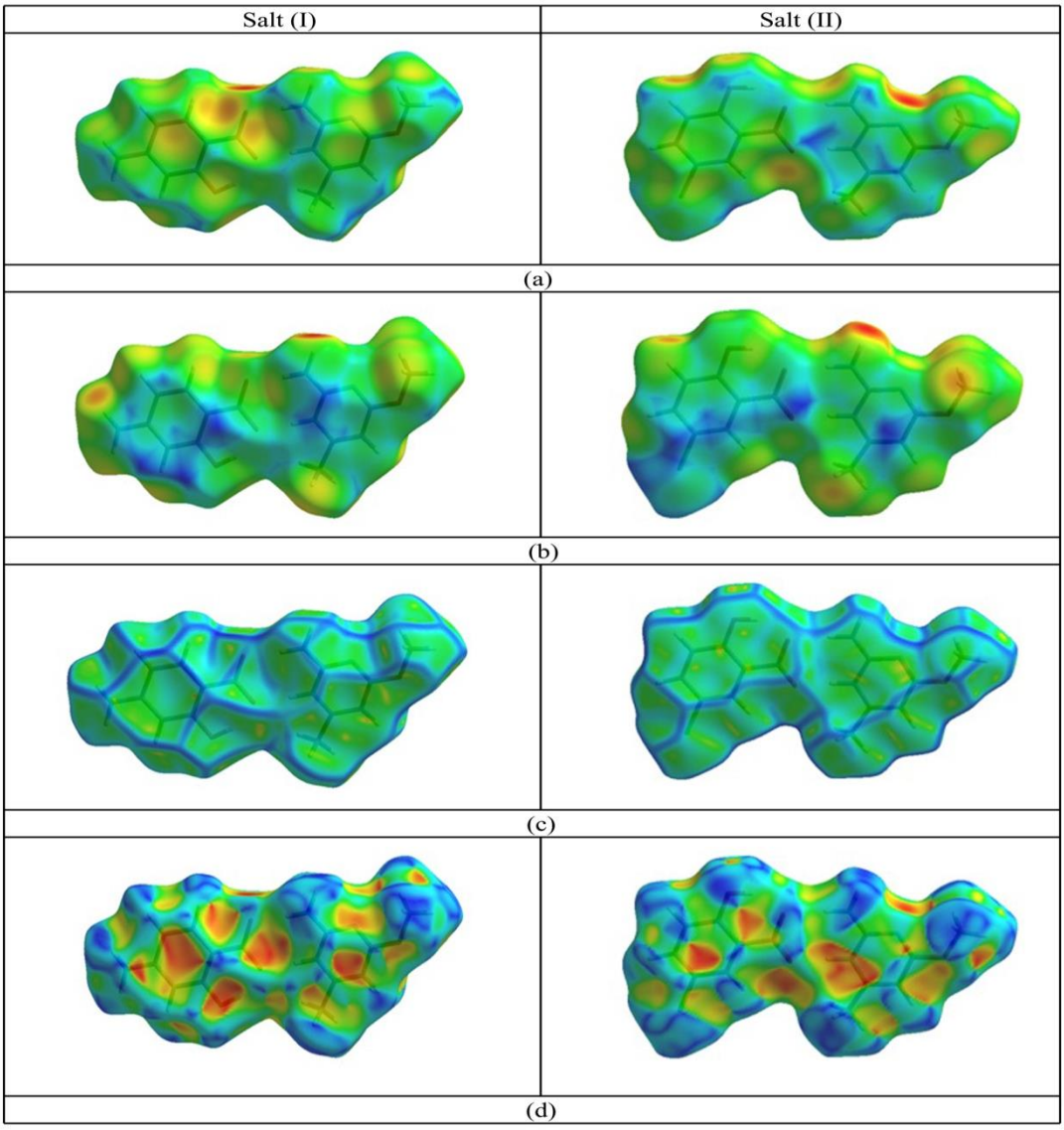
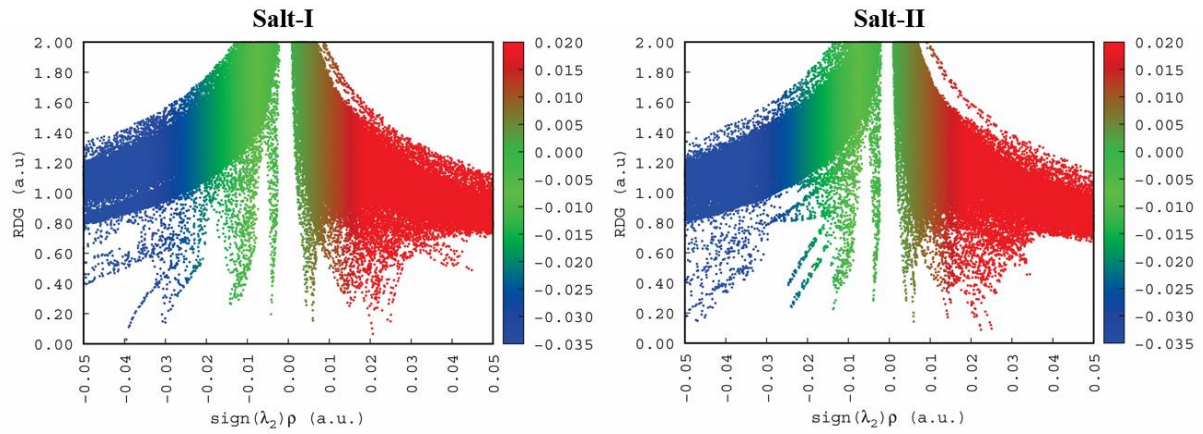


Figure (S25) The overall plot of the reduced density gradient versus the electron density times the sign of the second Hessian eigenvalue.



Salt-I

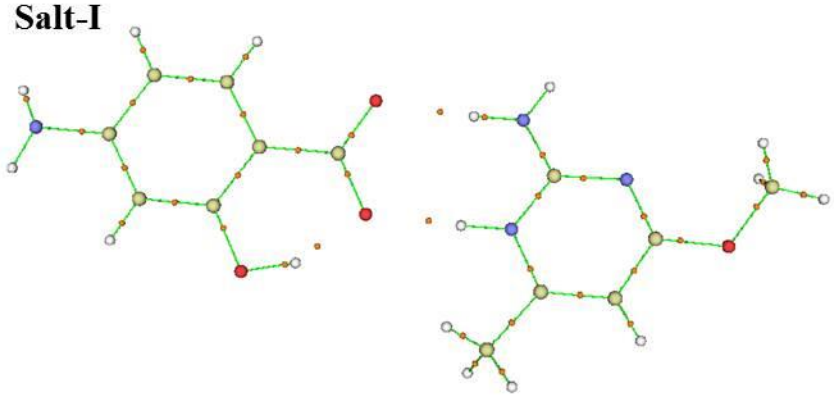
Counterpoise corrected energy = -1025.015290987964
BSSE energy = 0.004536300468
sum of monomers = -1024.846532788945
complexation energy = -108.74 kcal/mole (raw)
complexation energy = **-105.90 kcal/mole** (corrected)

Salt-II

Counterpoise corrected energy = -1429.312889110770
BSSE energy = 0.001566736969
sum of monomers = -1429.131205986360
complexation energy = -114.99 kcal/mole (raw)
complexation energy = **-114.01 kcal/mole** (corrected)

Figure (S26) The salt molecular graph of salt (I & II) showing (3,-1) and (3,+1) critical points.

Salt-I



Salt-II

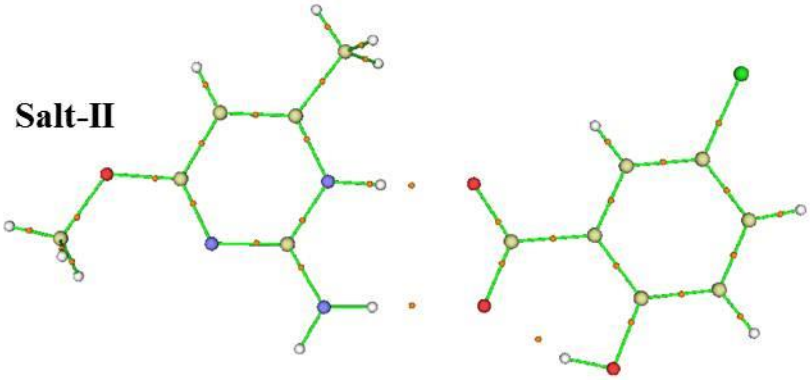


Figure (S27) Electrostatic potential map of salt molecules (I&II) drawn for the values +0.7e. (e)

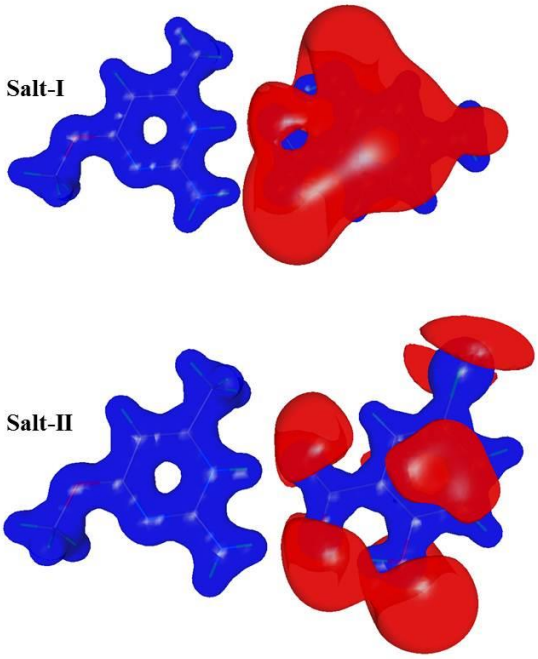


Figure (S28) The salt (1& II) graph map of Gradient vector Field.

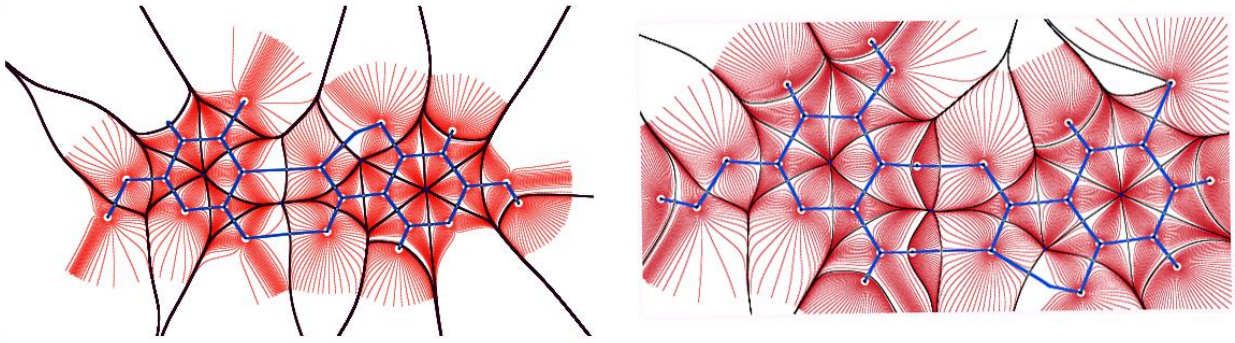
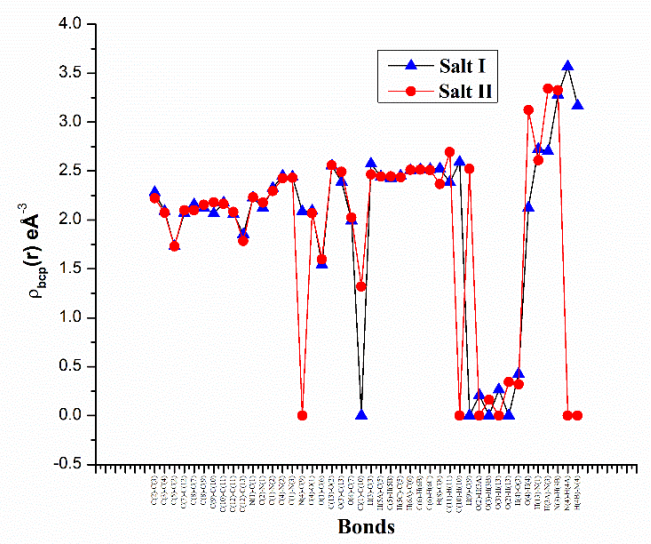
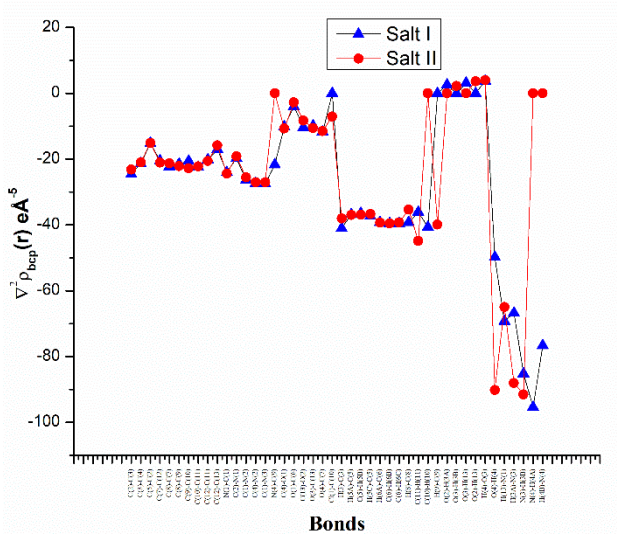


Figure (S29) The difference of (a) electron density $\rho_{bcp}(r)$ eÅ^{-3} and (b) the Laplacian of electron density $\rho_{bcp}(r)$ eÅ$^{\pm 5}$



(a)



(b)