# Supporting information

# Proposal for Crystallization of 3-Amino-4-halo-5-methylisoxazoles: An Energetic and Topological Approach

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**Table S1**. QTAIM data and  $G_{AI}$  of dimers of compound **1**.

Dimer	Inter.	ρ <sub>ιντ</sub>	<b>∇2</b> ρ	G	V	BPL	Ellipticity	G <sub>AI</sub> <sup>b</sup>	
M1•••M2	N•••H-N	0.015326	+0.064098	+0.013455	-0.010885	4.251078	0.129167	-4.17	
	N-H•••N	0.015326	+0.064098	+0.013455	-0.010885	4.251078	0.129167	-4.17	
M1•••M3	O•••H-N	0.008197	+0.039956	+0.007819	-0.005649	4.527889	0.097784	-1.66	
	0•••Cl	0.002279	+0.008790	+0.001713	-0.001228	7.221419	3.010133	-0.46	
	N•••Cl	0.002483	+0.008521	+0.001649	-0.001169	7.300750	0.393319	-0.50	
M1•••M4	C-H•••Cl	0.006021	+0.025268	+0.004847	-0.003378	5.416969	0.094680	-0.37	
M1•••M5	CI•••N	0.001028	+0.003606	+0.000668	-0.000435	8.429618	0.156814	-0.42	
	C-H•••N	0.002989	+0.010214	+0.001997	-0.001441	5.870038	0.128641	-1.24	
M1•••M6	C-H•••Cl	0.006020	+0.025265	+0.004847	-0.003377	5.417108	0.094694	-0.37	
M1•••M7	O•••H-N	0.008197	+0.039958	+0.007819	-0.005649	4.527896	0.097793	-1.66	
	0•••Cl	0.002279	+0.008790	+0.001713	-0.001228	7.221421	3.011512	-0.46	
	N•••Cl	0.002483	+0.008521	+0.001649	-0.001169	7.300748	0.393299	-0.50	
M1•••M8	C-H•••Cl	0.002202	+0.008392	+0.001518	-0.000938	6.764440	0.410496	-0.69	
M1•••M9	C-H•••Cl	0.002203	+0.008393	+0.001518	-0.000938	6.764275	0.410382	-0.69	
M1•••M10	С-Н•••п	0.005263	+0.020647	+0.004021	-0.002881	6.836096	0.855462	-1.07	
	C-H•••N	0.004368	+0.018990	+0.003719	-0.002691	5.734348	2.746002	-0.89	
	CI•••N	0.002123	+0.006810	+0.001306	-0.000910	7.605826	0.106768	-0.43	
	CI•••CI	0.001739	+0.005592	+0.001044	-0.000691	8.081175	0.103875	-0.35	
M1•••M11	С-Н•••п	0.005263	+0.020647	+0.004021	-0.002881	6.836096	0.855462	-1.07	
	C-H•••N	0.004368	+0.018990	+0.003719	-0.002691	5.734348	2.746002	-0.89	
	CI•••N	0.001739	+0.005592	+0.001044	-0.000691	8.081175	0.103875	-0.35	
	CI•••CI	0.002123	+0.006810	+0.001306	-0.000910	7.605826	0.106768	-0.43	
M1•••M12	CI•••N	0.002518	+0.008267	+0.001572	-0.001078	7.357650	0.300825	-0.60	
M1••••M13	N•••Cl	0.002518	+0.008267	+0.001572	-0.001078	7.357751	0.300809	-0.60	
M1•••M14	N•••Cl	0 001028	+0 003606	+0 000668	-0 000435	8 429618	0 156814	-0 42	
	N•••H-C	0.002989	+0.010214	+0.001997	-0.001441	5.870038	0.128641	-1.24	
M1•••M15	C-H•••N	0.003899	+0.014744	+0.002846	-0.002006	5.959126	0.305849	-1.02	
	N•••C	0.003618	+0.012906	+0.002485	-0.001744	7.006365	0.916383	-0.95	
	C•••O	0.003681	+0.013956	+0.002744	-0.001999	6.685321	0.323654	-0.96	
	0•••C	0.003681	+0.013956	+0.002744	-0.001999	6.685321	0.323654	-0.96	
	N•••C	0.003618	+0.012906	+0.002485	-0.001744	7.006365	0.916383	-0.95	
	N•••H-C	0.003899	+0.014744	+0.002846	-0.002006	5.959126	0.305849	-1.02	

<sup>a</sup> Atom...atom interaction.  $G_{AI}$  = Atom...atom interaction energy (kcal·mol<sup>-1</sup>).

 $\label{eq:compound} \textbf{Table S2}. \ \text{QTAIM} \ \text{data and} \ G_{\text{AI}} \ \text{of dimers of compound} \ \textbf{2}.$ 

Dimer	Inter. <sup>a</sup>	$\rho_{\text{INT}}$	<b>∇2</b> ρ	G	V	BPL	Ellipticity	$G_{AI}{}^{b}$
M1•••M2	N•••H-N	0.014235	+0.059791	+0.012428	-0.009908	4.304799	0.111969	-4.25
	N-H•••N	0.014235	+0.059791	+0.012428	-0.009908	4.304799	0.111969	-4.25
M1•••M3	C-Br•••O	0.001118	+0.005077	+0.000948	-0.000627	6.537782	0.106920	-1.53
N11000N14		0 002202	+0.000257	+0.001720	0.001120	E 252005	0 220020	0.01
101 1 000 1014	П	0.002393	+0.009357	+0.001739	-0.001139	5.252665	0.220920	-0.01
M1•••M5	C-H•••Br	0 002415	+0 009191	+0 001646	-0 000995	7 074769	0.390169	-0 49
	H•••H	0 001549	+0.007183	+0.001230	-0.000665	6 814131	12 52159	-0.32
	C-H•••Br	0.002415	+0.009192	+0.001646	-0.000995	7.074641	0.390012	-0.49
M1•••M6	C-Br•••O	0.005928	+0.025483	+0.005003	-0.003635	6.247424	0.065919	-1.51
M1•••M7	N•••Br	0.003856	+0.014164	+0.002699	-0.001857	7.280892	0.529749	-0.47
	H•••H	0.006959	+0.028133	+0.005699	-0.004364	4.596268	0.682365	-0.85
	N•••Br	0.003856	+0.014162	+0.002699	-0.001857	7.280999	0.529563	-0.47
M1M0	ClimPr	0 002020	0.012762	10.000610	0.001704	6 202005	0.070296	1 10
		0.003930	+0.013703	+0.002012	-0.001764	0.293993	0.079300	-1.12
	N-11000	0.001110	+0.003077	+0.000940	-0.000027	0.557762	0.100920	-0.52
M1•••M9	C-H•••Br	0.003938	+0.013764	+0.002612	-0.001784	6.294001	0.079421	-1.11
	N-H•••O	0.001119	+0.005078	+0.000948	-0.000627	6.537775	0.106939	-0.32
M1•••M10	C-H•••N	0.005617	+0.019871	+0.003919	-0.002870	5.267387	0.163343	-1.41
	Br•••N	0.003177	+0.010190	+0.001953	-0.001359	7.462650	0.292583	-0.79
	<b>.</b>							
M1•••M11	C-H•••N	0.005617	+0.019871	+0.003919	-0.002870	5.267387	0.163343	-1.41
	Br•••N	0.003177	+0.010190	+0.001953	-0.001359	7.462650	0.292583	-0.79
M1M12	BreeeBr	0 000082	+0.002613	+0 000472	-0 000200	9 31/000	0 228/72	-0.42
	DISSIDI	0.000302	10.002013	10.000472	-0.000230	3.314000	0.220472	-0.42
M1•••M13	Br•••Br	0 004107	+0 013920	+0 002644	-0 001807	7 448000	0 201361	-0.87
M1•••M14	C-H•••N	0.002839	+0.011958	+0.002235	-0.001480	6.590872	0.264485	-1.03
	C•••N	0.005018	+0.016430	+0.003198	-0.002289	7.586456	1.028788	-1.82
	N•••C	0.005018	+0.016430	+0.003198	-0.002289	7.586456	1.028788	-1.82
	C-H•••N	0.002839	+0.011958	+0.002235	-0.001480	6.590872	0.264485	-1.03
•••	·							
M1•••M15	C-H•••N	0.003340	+0.012825	+0.002462	-0.001718	5.838272	1.193427	-1.01
	Br•••π	0.003474	+0.012581	+0.002458	-0.001770	7.147323	0.653462	-1.05
	C•••C	0.003773	+0.011159	+0.002099	-0.001408	7.854917	1.573783	-1.14
	Br•••π	0.003474	+0.012581	+0.002458	-0.001770	7.147320	0.653497	-1.05
	C-H•••N	0.003341	+0.012827	+0.002463	-0.001718	5.838079	1.192675	-1.01

<sup>a</sup> Atom…atom interaction.  $G_{AI}$  = Atom…atom interaction energy (kcal·mol<sup>-1</sup>).

Table S3. QTAIM data and  $G_{\text{AI}}$  of dimers of compound 3.

M1•••M2         N•••H-N         0.014716         +0.055657         +0.011841         -0.009770         4.312157         0.089828         4.37           M1•••M3         C-I•••O         0.008488         +0.031085         +0.006405         -0.005039         6.206098         0.071883         -2.27           M1•••M4         H•••H         0.002189         +0.008409         +0.001567         -0.001031         5.226553         0.127831         -0.72           M1•••M5         C-H•••I         0.002925         +0.009542         +0.001784         -0.001183         7.275380         0.375193         -0.56           M1•••M6         C-I•••I         0.002925         +0.009543         +0.00663         -0.001183         7.275380         0.375193         -0.56           M1•••M6         C-I•••O         0.008487         +0.031082         +0.006644         -0.005039         6.206134         0.07187         -2.25           M1•••M7         N•••I         0.004541         +0.014614         +0.002913         -0.002172         7.512974         0.570255         -0.43           N•••I         0.002930         +0.008858         +0.001689         -0.001164         7.003412         4.468030         0.570187         -0.33           M1•••M9 <t< th=""><th>Dimer</th><th>Inter.<sup>a</sup></th><th>ριντ</th><th><b>∇2</b>ρ</th><th>G</th><th>V</th><th>BPL</th><th>Ellipticity</th><th><math>G_{AI}^{b}</math></th></t<>	Dimer	Inter. <sup>a</sup>	ριντ	<b>∇2</b> ρ	G	V	BPL	Ellipticity	$G_{AI}^{b}$
M1•••M2         N•+++N         0.014716         +0.055649         +0.011841         -0.009771         4.312185         0.089814         -4.36           M1•••M3         C-I•••O         0.008488         +0.031085         +0.006405         -0.005039         6.206098         0.071883         -2.27           M1•••M3         C-I•••O         0.008488         +0.031085         +0.001567         -0.01031         5.226553         0.127831         -0.72           M1•••M4         H•••H         0.002189         +0.009542         +0.001784         -0.001183         7.275435         0.375193         -0.56           M1•••M5         C-H•••I         0.002925         +0.009542         +0.001784         -0.001183         7.275435         0.375181         -0.56           M1•••M6         C-I•••O         0.008487         +0.031082         +0.006404         -0.005039         6.206134         0.071879         -2.25           M1•••M7         N•••I         0.004541         +0.014614         +0.002813         -0.002172         7.512974         0.570255         -0.43           M1•••M8         C-H•••I         0.002930         +0.008858         +0.001689         -0.001164         7.003450         0.126747         -1.19           M1•••M9									
N-H····N         0.014717         +0.055657         +0.011842         -0.009771         4.312157         0.089828         -4.37           M1···M3         C-I···O         0.008488         +0.031085         +0.006405         -0.005039         6.206098         0.071883         -2.27           M1···M4         H····H         0.002189         +0.008409         +0.001567         -0.001031         5.226553         0.127831         -0.72           M1···M5         C-H···I         0.002925         +0.009542         +0.001784         -0.001183         7.275435         0.375181         -0.56           M1···M6         C-I···0         0.008487         +0.031082         +0.006404         -0.0005039         6.206134         0.071879         -2.25           M1···M6         C-I···0         0.004541         +0.014614         +0.002913         -0.002172         7.512974         0.570255         -0.43           M1···M8         C-H···1         0.00230         +0.008858         +0.001689         -0.001164         7.003455         0.126799         -1.22           M1···M9         C-H···1         0.002930         +0.008858         +0.001689         -0.001164         7.003455         0.126799         -1.22           M1···M10         C-H····N	M1•••M2	N•••H-N	0.014716	+0.055649	+0.011841	-0.009770	4.312185	0.089814	-4.36
M1•••M3         C-I••O         0.008488         +0.031085         +0.006405         -0.005039         6.206098         0.071883         -2.27           M1•••M4         H•••H         0.002189         +0.008409         +0.001567         -0.001031         5.226553         0.127831         -0.72           M1•••M5         C-H•••1         0.002925         +0.009642         +0.001784         -0.00183         7.275435         0.375193         -0.56           M1•••M6         C-I•••0         0.008487         +0.031082         +0.006404         -0.001784         -0.001183         7.275435         0.375181         -0.56           M1•••M6         C-I•••0         0.008487         +0.031082         +0.006404         -0.002172         7.512974         0.570255         -0.43           M1•••M7         N•••I         0.004541         +0.014614         +0.002913         -0.002172         7.512974         0.570187         -0.53           M1•••M8         C-H•••I         0.002903         +0.008858         +0.001689         -0.001164         7.003430         0.126747         -1.19           M1•••M9         C-H•••I         0.002937         +0.01689         -0.001661         7.601744         0.291482         -0.98           M1•••M10		N-H•••N	0.014717	+0.055657	+0.011842	-0.009771	4.312157	0.089828	-4.37
M1***M3         C-I***O         0.008488         ±0.031085         ±0.006405         -0.005039         6.206098         0.071883         -2.27           M1***M4         H***H         0.002189         ±0.008409         ±0.001567         -0.001031         5.226553         0.127831         -0.72           M1***M5         C-H***I         0.002925         ±0.009542         ±0.001784         -0.001183         7.275435         0.375193         -0.56           M1***M6         C-I***O         0.002925         ±0.009543         ±0.001784         -0.001183         7.275380         0.375181         -0.56           M1***M6         C-I***O         0.004847         ±0.031082         ±0.006404         -0.005039         6.206134         0.071879         -2.25           M1***M6         C-I***O         0.004541         ±0.014614         ±0.002913         -0.002172         7.512974         0.570255         -0.43           M1***M8         C-H***I         0.002930         ±0.008858         ±0.001689         -0.001164         7.003455         0.126799         -1.22           M1***M9         C-H***I         0.002930         ±0.008858         ±0.001689         -0.001164         7.003430         0.126747         -1.19           M1***M10		<u>.</u>	0 000 400			0.00-000		0.074000	0 0 <del>7</del>
M1•••M4         H••·H         0.002189         +0.008409         +0.001567         -0.001031         5.226553         0.127831         -0.72           M1••·M5         C-H••·I         0.002925         +0.009542         +0.001784         -0.001183         7.275435         0.375193         -0.56           M1••·M6         C-H••·I         0.002925         +0.009543         +0.001784         -0.001183         7.275380         0.375193         -0.56           M1••·M6         C-I••·O         0.008487         +0.031082         +0.006404         -0.002172         7.512974         0.570255         -0.43           M1••·M7         N••·I         0.004541         +0.014614         +0.002913         -0.002172         7.512974         0.570255         -0.43           M1••·M8         C-H•··I         0.002930         +0.008858         +0.001689         -0.001164         7.003455         0.126799         -1.22           M1••·M9         C-H•··I         0.002930         +0.008858         +0.001689         -0.001164         7.003430         0.126747         -1.19           M1••·M10         C-H•··N         0.004995         +0.016123         +0.003238         -0.002446         5.392469         0.141335         -1.23         -0.98	M1•••M3	C-I•••O	0.008488	+0.031085	+0.006405	-0.005039	6.206098	0.071883	-2.27
M1***M4         H***H         0.002189         +0.00409         +0.00137         -0.00131         5.22833         0.127831         -0.72           M1***M5         C-H***I         0.002925         +0.009464         +0.001784         -0.001183         7.275335         0.375193         -0.56           M1***M5         C-H***I         0.002925         +0.009543         +0.001784         -0.001183         7.275380         0.375193         -0.56           M1***M6         C-I***O         0.008487         +0.031082         +0.006404         -0.001183         7.275380         0.375181         -0.56           M1***M6         C-I***O         0.004541         +0.014614         +0.002913         -0.002172         7.512974         0.570255         -0.43           M1***M7         N***I         0.002930         +0.008858         +0.001689         -0.001164         7.003455         0.126747         -1.19           M1***M8         C-H***I         0.002930         +0.008858         +0.001689         -0.001164         7.003430         0.126747         -1.19           M1***M10         C-H***N         0.004995         +0.016123         +0.003238         -0.002446         5.392469         0.141335         -1.23           M1***M11	M1	Und	0.000190	10.009400	0.001567	0.001021	E 006550	0 107001	0 70
M1•••M5         C-H•••I H•••H C-H•••I         0.002925 0.002925         +0.009542 +0.009643         +0.001784 +0.000863 +0.001784         -0.001183 -0.00485         7.275435 6.698287         0.375193 0.375181         -0.56 -0.24 -0.56           M1•••M6         C-I•••O         0.008487         +0.031082         +0.006404         -0.000485         6.698287         1.060076         -0.24           M1•••M6         C-I•••O         0.008487         +0.031082         +0.006404         -0.005039         6.206134         0.071879         -2.25           M1•••M7         N•••I         0.004541         +0.014614         +0.002913         -0.002172         7.512974         0.570255         -0.43           M1•••M8         C-H•••I         0.002930         +0.008858         +0.001689         -0.001164         7.003455         0.126799         -1.22           M1•••M9         C-H•••I         0.002930         +0.008858         +0.001689         -0.001164         7.003430         0.126747         -1.19           M1•••M10         C-H•••N         0.004995         +0.016123         +0.003238         -0.002446         5.392469         0.141335         -1.23           M1•••M11         C-H•••N         0.0029376         +0.016123         +0.003238         -0.002446         5.392469	101 1 •••• 1014	П•••П	0.002169	+0.006409	+0.001567	-0.001031	5.220555	0.127031	-0.72
Min         Min <th>M1•••M5</th> <th>C-H•••I</th> <th>0 002925</th> <th>+0 009542</th> <th>+0 001784</th> <th>-0 001183</th> <th>7 275435</th> <th>0.375193</th> <th>-0 56</th>	M1•••M5	C-H•••I	0 002925	+0 009542	+0 001784	-0 001183	7 275435	0.375193	-0 56
C-H····I         0.002925         +0.009543         +0.001784         -0.001183         7.275380         0.375181         -0.56           M1···M6         C-I···O         0.008487         +0.031082         +0.006404         -0.005039         6.206134         0.071879         -2.25           M1···M7         N···I         0.004541         +0.014614         +0.002913         -0.002172         7.512974         0.570255         -0.43           M1···M8         C-H···I         0.004541         +0.014613         +0.002913         -0.002172         7.512974         0.570255         -0.43           M1···M8         C-H···I         0.002930         +0.008858         +0.001689         -0.001164         7.003455         0.126799         -1.22           M1···M9         C-H···I         0.002930         +0.008858         +0.001689         -0.001164         7.003430         0.126747         -1.19           M1···M10         C-H···N         0.004995         +0.016123         +0.003238         -0.002466         5.392469         0.141335         -1.23           M1···M11         C-H···N         0.002976         +0.016123         +0.003238         -0.002466         5.392469         0.141335         -1.23           M1···M11         C-H···N		H•••H	0.001234	+0.000042	+0.000863	-0.000485	6 698287	1 060076	-0.24
M1•••M6         C-I••·O         0.008487         +0.031082         +0.006404         -0.005039         6.206134         0.071879         -2.25           M1•••M7         N•••I         0.004541         +0.014614         +0.002913         -0.002172         7.512974         0.570255         -0.43           M1•••M7         N•••I         0.004541         +0.014613         +0.002913         -0.002172         7.512974         0.570255         -0.43           M1•••M8         C-H•••I         0.002930         +0.008858         +0.001689         -0.001164         7.003455         0.126799         -1.22           M1•••M9         C-H•••I         0.002930         +0.008858         +0.001689         -0.001164         7.003430         0.126747         -1.19           M1•••M10         C-H•••N         0.004995         +0.016123         +0.003238         -0.002466         5.392469         0.141335         -1.23           M1•••M11         C-H•••N         0.003976         +0.01746         +0.002169         -0.001651         7.601744         0.291482         -0.98           M1•••M11         C-H•••N         0.002931         +0.01626         +0.002169         -0.001651         7.601744         0.291482         -0.98           M1•••M13		C-H•••I	0.002925	+0.009543	+0.001784	-0.001183	7.275380	0.375181	-0.56
M1•••M6C-I•••O0.008487+0.031082+0.006404-0.0050396.2061340.071879-2.25M1•••M7N•••I0.004541+0.014614+0.002913-0.0021727.5129740.570255-0.43M1•••M8C-H•••I0.002930+0.008858+0.001689-0.0011647.0034550.126799-1.22M1•••M9C-H•••I0.002930+0.008858+0.001689-0.0011647.0034300.126747-1.19M1•••M10C-H•••I0.002930+0.008858+0.001689-0.0016517.6017440.291482-0.98M1•••M10C-H•••N0.004995+0.016123+0.003238-0.0024465.3924690.141335-1.23M1•••M11C-H•••N0.002937+0.016123+0.003238-0.0024465.3924690.141335-1.23M1•••M11C-H•••N0.002614+0.006366+0.001226-0.008608.9400000.207405-0.82M1•••M12I•••I0.002531+0.014624+0.003088-0.0021465.3322490.141335-1.23M1•••M12I•••I0.002614+0.014824+0.003080-0.0022146.9859062.004273-1.88M1•••M14C-H•••N0.003291+0.014613+0.002438-0.0017116.3322190.183240-1.27M1•••M14C-H•••N0.002891+0.015451+0.002020-0.0017116.3320490.183141-1.88M1•••M15C-H•••N0.002898+0.01372+0.002020-0.0014476.985									
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	M1•••M6	C-I•••O	0.008487	+0.031082	+0.006404	-0.005039	6.206134	0.071879	-2.25
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$									
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	M1•••M7	N•••I	0.004541	+0.014614	+0.002913	-0.002172	7.512974	0.570255	-0.43
N++I         0.004541         +0.014613         +0.002913         -0.002172         7.512914         0.570187         -0.43           M1++M8         C-H++I         0.002930         +0.008858         +0.001689         -0.001164         7.003455         0.126799         -1.22           M1++M9         C-H++I         0.002930         +0.008858         +0.001689         -0.001164         7.003430         0.126747         -1.19           M1++M10         C-H+++N         0.004995         +0.016123         +0.003238         -0.002446         5.392469         0.141335         -1.23           M1+++M11         C-H+++N         0.004995         +0.016123         +0.003238         -0.002446         5.392469         0.141335         -1.23           M1+++M11         C-H+++N         0.004995         +0.016123         +0.003238         -0.002446         5.392469         0.141335         -1.23           M1+++M12         I+++N         0.002614         +0.006366         +0.001226         -0.000860         8.940000         0.207405         -0.82           M1+++M13         I+++I         0.005753         +0.014824         +0.003080         -0.002455         7.760000         0.204166         -1.26           M1+++M14         C-H+++N		H•••H	0.005562	+0.020604	+0.004281	-0.003412	4.468403	0.436664	-0.53
M1•••M8C-H•••I0.002930+0.008858+0.001689-0.0011647.0034550.126799-1.22M1•••M9C-H•••I0.002930+0.008858+0.001689-0.0011647.0034300.126747-1.19M1•••M10C-H•••N0.004995+0.016123+0.003238-0.0024465.3924690.141335-1.23M1•••M11C-H•••N0.004995+0.016123+0.003238-0.0024465.3924690.141335-1.23M1•••M11C-H•••N0.004995+0.016123+0.003238-0.0024465.3924690.141335-1.23M1•••M12I•••I0.002614+0.010746+0.002169-0.0016517.6017440.291482-0.98M1•••M13I•••I0.002614+0.006366+0.001226-0.0008608.9400000.207405-0.82M1•••M13I•••I0.003291+0.012660+0.002438-0.0017116.3322190.183240-1.27M1•••M14C-H•••N0.003291+0.012661+0.002438-0.0021466.9863962.004273-1.88M1•••M15C-H•••N0.002898+0.010372+0.002020-0.0014476.0642901.983599-0.95M1•••M15C-H•••N0.002898+0.010372+0.002020-0.0014476.0642901.983599-0.95M1•••M15C-H•••N0.002898+0.010372+0.002020-0.0014476.0642901.983599-0.95M1•••M15C-H•••N0.002898+0.010372+0.002020-0.0014476		N•••I	0.004541	+0.014613	+0.002913	-0.002172	7.512914	0.570187	-0.43
$\begin{array}{c c c c c c c c c c c c c c c c c c c $		<u></u>				0.004404		0.400-00	4 00
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M1•••M8	C-H•••I	0.002930	+0.008858	+0.001689	-0.001164	7.003455	0.126799	-1.22
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	MisseMO		0 002030	+0 008858	+0.001680	0.001164	7 003430	0 126747	1 10
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	10110001019	0-11-001	0.002930	+0.000050	+0.001009	-0.001104	7.003430	0.120747	-1.19
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	M1•••M10	C-H•••N	0.004995	+0.016123	+0.003238	-0.002446	5.392469	0.141335	-1.23
M1•••M11         C-H•••N         0.004995         +0.016123         +0.003238         -0.002446         5.392469         0.141335         -1.23           M1•••M12         I•••N         0.002614         +0.006366         +0.001226         -0.000860         8.940000         0.207405         -0.82           M1•••M13         I•••I         0.005753         +0.014824         +0.003080         -0.002455         7.760000         0.207405         -0.82           M1•••M14         C-H•••N         0.003291         +0.012660         +0.002438         -0.001711         6.332219         0.183240         -1.27           M1•••M14         C-H•••N         0.004861         +0.015451         +0.003038         -0.002214         6.985906         2.004273         -1.88           N•••C         0.004861         +0.015451         +0.003038         -0.002214         6.986396         2.005260         -1.88           C-H••N         0.003292         +0.012661         +0.002439         -0.001712         6.332049         0.183141         -1.27           M1•••M15         C-H••N         0.002898         +0.010372         +0.002020         -0.001712         6.332049         0.183141         -1.27           M1•••M15         C-H••N         0.002898		I•••N	0.003976	+0.010746	+0.002169	-0.001651	7.601744	0.291482	-0.98
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$									
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	M1•••M11	C-H•••N	0.004995	+0.016123	+0.003238	-0.002446	5.392469	0.141335	-1.23
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$		I•••N	0.003976	+0.010746	+0.002169	-0.001651	7.601744	0.291482	-0.98
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$									
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	M1•••M12	•••	0.002614	+0.006366	+0.001226	-0.000860	8.940000	0.207405	-0.82
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$									
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	M1•••M13	•••	0.005753	+0.014824	+0.003080	-0.002455	7.760000	0.204166	-1.26
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	N11N11.1		0.003201	+0.012660	+0 002438	0 001711	6 332210	0 183240	1 27
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	101100010114		0.003291	+0.012000	+0.002438	0.001711	6 085006	2 004273	1 99
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$		Nee	0.004001	+0.015451	+0.003038	0.002214	6.086306	2.004273	1 99
M1•••M15       C-H•••N       0.002898       +0.010372       +0.002020       -0.001447       6.064290       1.983599       -0.95         I•••π       0.004347       +0.013152       +0.002696       -0.002103       7.338056       1.337403       -1.42         C•••C       0.003293       +0.009261       +0.001768       -0.001221       7.801040       5.714340       -1.08		C-H•••N	0.004001	+0.013451	+0.003038	-0.002214	6 332040	0.1831/1	-1.00
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$		C-Heern	0.003292	+0.012001	+0.002439	-0.001712	0.332049	0.105141	-1.27
$\begin{matrix} \textbf{I} \bullet \bullet \pi \\ \textbf{C} \bullet \bullet \bullet \textbf{C} \end{matrix} 0.004347 + 0.013152 + 0.002696 - 0.002103 7.338056 1.337403 - 1.42 \\ \textbf{C} \bullet \bullet \bullet \textbf{C} \end{matrix} 0.003293 + 0.009261 + 0.001768 - 0.001221 7.801040 5.714340 - 1.08 \\ 0.001221 7.801040 5.71440 5.71440 - 1.08 \\ 0.001221 7.801040 5.71400 - 1.08 \\ 0.001221 7.80$	M1•••M15	C-H•••N	0.002898	+0.010372	+0.002020	-0.001447	6.064290	1,983599	-0.95
C····C 0.003293 +0.009261 +0.001768 -0.001221 7.801040 5.714340 -1.08		•••π	0.004347	+0.013152	+0.002696	-0.002103	7.338056	1.337403	-1.42
		C•••C	0.003293	+0.009261	+0.001768	-0.001221	7.801040	5.714340	-1.08
$1^{\bullet\bullet\bullet\pi}$ 0.004347 +0.013152 +0.002695 -0.002103 7.338067 1.338053 -1.42		<b>Ι•••</b> Π	0.004347	+0.013152	+0.002695	-0.002103	7.338067	1.338053	-1.42
C-H•••N 0.002899 +0.010373 +0.002020 -0.001447 6.064046 1.980064 -0.95		C-H•••N	0.002899	+0.010373	+0.002020	-0.001447	6.064046	1.980064	-0.95

<sup>a</sup> Atom...atom interaction.  $G_{AI}$  = Atom...atom interaction energy (kcal·mol<sup>-1</sup>).

Interaction type	d <sub>(XY)</sub> range (Å)	Angle <sub>(XY)</sub> range (°)
N-H···N	3.03-3.08	159-163
N-H <sup>…</sup> O	3.20	165
	3.96	131
C-H <sup>…</sup> N	3.64-4.08	110-170
C-H <sup></sup> X	3.56-4.15	92-135
	3.99-4.30	117-130
C-XO	3.28-3.82	102-160
C-X <sup>…</sup> N	3.76-4.36	98-103
	3.85-4.01	95-117
XX	4.27-4.73	102-120
	3.93-4.01	138-141
Χ…π	3.77-3.86	90-91
C-H <sup>…</sup> π	3.63	159
$\pi\cdots\pi$	3.43-3.57	92-100
	3.52-4.66	126-127
CHHC	4.17-4.30	-
	4.08-4.30	-
NH…HN	3.40-3.59	-

Table S4. Geometrical parameters of intermolecular interaction of compounds 1–3.

 Table S5. Interaction energies<sup>a</sup> of the dimers present in the clusters of compounds 1-3, with and without BSSE correction.

Dimer			G <sub>M1</sub> <sub>MN</sub> (	kcal⋅mol⁻¹)		
-	Wit	th BSSE Corre	ection	Withc	out BSSE Corr	ection
-	1	2	3	1	2	3
M1…M2	-8.33	-8.50	-8.73	-9,95	-10,06	-10,28
M1…M3	-2.63	-1.53	-2.27	-3,68	-2,21	-3,11
M1…M4	-0.37	-0.81	-0.72	-0,63	-1,04	-0,89
M1…M5	-1.66	-1.30	-1.35	-2,25	-1,81	-1,89
M1…M6	-0.37	-1.51	-2.25	-0,63	-2,20	-3,09
M1…M7	-2.63	-1.80	-1.40	-3,68	-3,52	-3,05
M1…M8	-0.69	-1.44	-1.22	-0,95	-2,15	-1,71
M1…M9	-0.69	-1.43	-1.19	-0,95	-2,14	-1,68
M1…M10	-1.66	-2.20	-2.21	-2,25	-3,17	-3,22
M1…M11	-0.60	-2.20	-2.21	-1,06	-3,17	-3,22
M1…M12	-0.60	-0.42	-0.82	-1,06	-0,50	-1,01
M1…M13	-2.75	-0.87	-1.26	-3,91	-1,16	-1,63
M1…M14	-2.75	-5.69	-6.29	-3,91	-8,43	-9,18
M1…M15	-5.87	-5.25	-5.81	-8,23	-7,64	-8,27
GCluster	-31.61	-34.94	-37.71	-43,15	-49,19	-52,22

<sup>a</sup> Calculated at the MP2/cc-pVTZ level of theory.

#### Figures



**Figure S1.** Correlation between the sum of the electron densities of all atoms...atom interactions and sum of energy dimers from the supramolecular cluster of compounds **1-3**.



**Figure S2**.<sup>1</sup>H NMR chemical shift changes of signals due to 3-NH<sub>2</sub> (K<sub>ass</sub> = 8.0,  $\Delta\delta_{dimer}$ = 1.78) and 5-CH<sub>3</sub> (K<sub>ass</sub> = 0.6,  $\Delta\delta_{dimer}$ = -0.03) of compound **1** in CDCI<sub>3</sub>.



Figure S3.<sup>1</sup>H NMR chemical shift changes of signals due to 3-NH<sub>2</sub> (K<sub>ass</sub> = 12.8,  $\Delta \delta_{dimer}$ = 1.02) and 5-CH<sub>3</sub> (K<sub>ass</sub> = 2.1,  $\Delta \delta_{dimer}$ = -0.03) of compound **3** in CDCI<sub>3</sub>.



**Figure S4**. Supramolecular clusters showing central, superior, and inferior layer and each dimer separately for compound **1**.



**Figure S5**. Supramolecular clusters showing central, superior, and inferior layer and each dimer separately for compound **2**.



**Figure S6**. Supramolecular clusters showing central, superior, and inferior layer and each dimer separately for compound **3**.