Supplementary information

Table 1. Characteristic data of compounds 2–10

Compounds	M.p., ^{0}C; (Solvent)*	Molecular Formula	Data of elemental analysis, (Calculated / Found) %			Yield, %
			C	H	N	
2a	211-212 [a]	$C_{12}H_{14}ClN_3O_2$	53.84/53.42	5.27/5.16	15.70/15.54	90.2
2b	109-110 [a]	$C_{12}H_{14}ClN_3O_2$	53.84/54.3	5.27/5.08	15.70/13.59	76.8
3a	159-160 [c]	$C_{15}H_{18}ClN_3O_2$	58.54/58.51	5.89/6.0	13.56/13.32	57
3b	138-139 [a]	$C_{15}H_{18}ClN_3O_2$	58.54/59.01	5.89/6.13	13.56/13.22	48
4a	119-120 [a]	$C_{16}H_{20}ClN_3O_2$	59.72/59.67	6.26/6.55	13.06/12.9	64.6
4b	111-112 [a]	$C_{16}H_{20}ClN_3O_2$	59.72/59.67	6.26/5.85	13.06/13.16	60
5a	238-239[a]	$C_{19}H_{17}ClBrN_3O_2$	52.50/52.16	3.94/3.76	9.67/9.49	96.9
5b	186-187 [b]	$C_{19}H_{17}ClBrN_3O_2$	52.50/52.28	3.94/3.64	9.67/9.53	79.1
6a	267-268 [b]	$C_{19}H_{17}ClN_4O_4$	56.93/57.18	4.28/3.99	13.98/13.67	97.7
6b	211-212 [b]	$C_{19}H_{17}ClN_4O_4$	56.93/56.55	4.28/4.14	13.98/13.67	89.3
7a	213-214 [b]	$C_{19}H_{18}ClN_3O_3$	61.38/60.7	4.88/4.6	11.30/11.54	61.5
7b	277-278 [a]	$C_{19}H_{18}ClN_3O_3$	61.38/61.67	4.88/4.53	11.30/11.16	82.7
8a	235-236 [b]	$C_{21}H_{23}ClN_4O_2$	63.23/63.46	5.81/5.6	14.05/14.02	83.2
8b	179-180 [a]	$C_{21}H_{23}ClN_4O_2$	63.23/63.68	5.81/5.44	14.05/14.24	60.7
9a	141 [b], dec.	$C_{27}H_{25}ClN_4O_2$	68.56/68.4	5.33/5.65	11.85/11.51	76.3
9b	160 [a], dec.	$C_{27}H_{25}ClN_4O_2$	68.56/68.83	5.33/5.38	11.85/12.03	94.6
10a	136-137 [a]	$C_{17}H_{18}ClN_3O_2$	61.54/61.22	5.47/5.6	12.66/12.38	60.5
10b	139-140 [d]	$C_{17}H_{18}ClN_3O_2$	61.54/61.37	5.47/5.73	12.66/12.69	66.5

Solvents for crystallization - [a] ***2a***,***b***, ***3b***, ***4a***,***b***, ***5b***, ***7 – 9 b***, ***10a*** *– 2-propanol, [b]* ***5 – 9 a***, ***6b*** *– ethanol, [c]* ***3a*** *– acetone, [d]* ***10b*** *– toluene.*

Table 2. ^{13}C- NMR chemical shifts of compounds **2a,b, 3a,b** (d_6-DMSO), **3a*** ($CDCl_3$)

Carbons	Chemical shifts (δ, in ppm) of compounds						
	2a	2b	3a*	3a		3b	
				E	*Z*	*E*	*Z*
C-1	138.22	138.85	137.78	138.28		138.91	
C-2	119.35	130.25	120.39	119.41	119.31	130.25	
C-3	133.10	134.74	134.33	133.09		134.73	134.66
C-4	131.08	127.39	132.18	131.08		127.37	127.33
C-5	130.72	132.21	130.82	130.68		132.20	
C-6	117.68	126.64	118.23	117.77	117.65	126.61	126.56
C-2’	172.27	171.82	172.34	172.38	172.28	171.93	
C-3’	35.73	35.28	35.24	35.82	34.95	35.63	34.26
C-4’	33.97	34.26	33.63	34.19	32.81,	34.26	33.38
C-5’	50.57	52.25	50.39	50.69	50.06	52.48	51.94
CONH	171.45	171.82	173.65	173.55	168.48	173.77	168.80
C=N			150.98	156.24	151.28	156.17	151.24
$(CH_3)_2$ *cis/trans*			16.32 / 25.52	17.62 / 25.25	17.13 / 24.94	17.61 / 25.25	17.12 / 24.98
Others	4-CH_3 18.95	2-CH_3 17.06	4-CH_3 19.46	4-CH_3 18.93		2-CH_3	
						17.08	17.04

Table 3. ^{13}C- NMR chemical shifts of compounds **4a*** ($CDCl_3$), **4a, 4b** (d_6-DMSO)

Carbons	Chemical shifts (δ, in ppm) of compounds				
	4a*	4a		4b	
		E	*Z*	*E*	*Z*
C-1	137.78	138.27	138.22	138.89	
C-2	120.38	119.38	119.30	130.22	
C-3	134.31	133.05		134.71	134.63
C-4	132.14	131.05		127.32	
C-5	130.80	130.66		132.22	
C-6	118.16	117.72	117.65	126.58	
C-2’	172.37	172.35	172.24	171.93	171.83
C-3’	35.19	35.78	34.89	35.62	34.45
C-4’	33.68	34.23	33.03	34.38	33.28
C-5’	50.31	50.70	49.95	52.43	51.83
CONH	173.88	173.58	168.48	173.83	168.84
C=N	154.88	159.75	154.54	159.56	154.46
CH_2CH_3	32.11, 10.37	31.50, 10.73	31.41, 10.32	31.55, 10.77	31.43, 10.36
CH_3	15.39	16.02	15.67	16.08	16.02
Others	4-CH_3 19.44	4-CH_3 18.89		2-CH_3 17.02	

Table 4. ^{13}C- NMR chemical shifts (δ, in ppm) of compounds **6a, 7b, 10a,b** (in d_6-DMSO), and **8a*** (in $CDCl_3$)

Carbons	Chemical shifts (δ, in ppm) of compounds						
	6a		7b		8a*	10a	10b
	E	*Z*	*E*	*Z*			
C-1	138.24	138.19	138.87		138.41	138.22	138.65
C-2	119.49	119.42	130.23		121.02	119.56	130.26
C-3	133.11	132.05	134.68		134.68	133.09	134.71
C-4	131.21	131.12	127.33		132.55	131.11	127.41
C-5	130.83	130.78	132.17		131.14	130.91	132.17
C-6	117.88	117.77	126.62		118.55	117.94	126.57
C-2’	172.22		171.85	171.72	172.60	172.52	172.69
C-3’	34.89		35.97	34.21	35.59	35.25	36.61
C-4’	32.75		34.03	33.35	34.16	35.05	33.54
C-5’	49.87		52.17	51.78	50.63	50.03	51.74
CONH	173.90		173.28	168.49	173.34	171.79	171.23
N=C	144.63	141.42	147.24	143.87	145.91	152.15	152.07
C-1’’	140.45	140.41	125.11		120.82		
C-2’’	128.03	127.89	128.83	128.56	128.96		
C-3’’	124.07	124.03	115.67		112.05		
C-4’’	147.73		159.43	159.22	152.16		
Others	4-CH_3 18.97		2-CH_3 17.08		$N(CH_3)_2$ 40.44 4-CH_3 19.76	C-d 111.94 C-e 143.89 c/e-CH_3 13.59 or 14.05 4-CH_3 18.96	C-d 111.56 C-e 143.85 c/e-CH_3 13.54 or 14.09 2-CH_3 17.07

Table 5. Mass, IR, and 1H- NMR spectroscopic data of compounds 2–10

Cpd.	MS (m/z), %	IR, ν (cm^{-1})	^{1}H- NMR (δ, in ppm), J (Hz)
2a	268.2$[M+H]^+$ (100), 270.5 $[M+2+H]^+$ (33).	3316 (NH_2), 3283 (NH), 683 (C=O), 1637 (C=O), 642-579 (Ar).	2.51 (s, 3H, 4-CH_3), 2.59-2.78 (m, 2H, CH_2CO), 3.12-3.23 (m, 1H, CH), 3.81-4.01 (m, 2H, CH_2N), 7.33-7.87 (m, 3H, Ar), 8.59, 9.31 (2s, 1H, NH)
2b	268.2$[M+H]^+$ (100), 270.5 $[M+2+H]^+$ (33).	3320 (NH_2, NH), 1703(C=O), 1672 (C=O), 868-652(Ar).	2.14 (s, 3H, 2-CH_3), 3.18-3.30 (m, 2H, CH_2CO), 3.20-3.30 (m, 1H, CH), 3.66-3.91 (m, 2H, CH_2N), 7.31-7.37 (m, 3H, Ar), 9.45 (br. s, 1H, NH)
3a*	308.5$[M+H]^+$, (100), 310.5 $[M+2+H]^+$, (33).	3184 (NH), 1707 (C=O), 1678 (C=O), 882-650 (Ar).	1.92, 2.05 (2s, 6H, N=CCH_3), 2.37 (s, 3H, 4-CH_3), 2.82-3.10 (m, 2H, CH_2CO), 4.02-4.12 (m, 3H, CH+CH_2N), 7.22-7.65 (m, 3H, Ar), 8.73, 9.05 (2s, 1H, NH).
3a			1.89, 1.90 (2s, 3H, N=CCH_3), 1.95 (s, 3H, N=C$\underline{CH_3}$), 2.31 (s, 3H, 4-CH_3), 2.63-2.83 (m, 2H, CH_2CO), 3.39-3.49 (m, 0.5H, CH), 3.85-4.10 (m, 2H, CH_2N + 0.5H, CH), 7.33–7.87 (m, 3H, Ar), 10.24, 10.33 (2s, 1H, NH)
3b	308.5[M+H]+, (100), 310.5 [M+2+H]+, (33).	3251 (NH), 1703 (C=O), 1664 (C=O), 812-728 (Ar).	1.87, 1.88 (2s, 3H, N=C$\underline{CH_3}$), 1.93, 1.95 (2s, 3H, N=C$\underline{CH_3}$), 2.14, 2.16 (2s, 3H, 2-CH_3), 2.51-2.75 (m, 2H, CH_2CO), 3.20-4.02 (m, 3H, CH_2N+ CH), 7.30-7.38 (m, 3H, Ar), 10.21, 10.30 (2s, 1H, NH)
4a*	322.5 $[M+H]^+$, (100), 324.5 $[M+2+H]^+$, (33).	3194 (NH), 1709 (C=O), 1679 (C=O). 803-697 (Ar).	1.13 (t, J = 7.4, 3H, N=C$CH_2\underline{CH_3}$), 1.91, 2.01 (2s, 3H, N=CCH_3), 2.30, 2.33 (2k, J = 7.4, 2H, N=C$\underline{CH_2}CH_3$), 2.36 (s, 3H, 4-CH_3), 2.82-3.10 (m, 2H, CH_2CO), 4.02-4.12 (m, 3H, CH_2N+CH), 7.21-7.66 (m, 3H, Ar), 9.32, 9.34 (2s, 1H, NH)
4a			1.00, 1.04, 1.05, (3t, J = 7.4, 3H, N=C$CH_2\underline{CH_3}$), 1.87, 1.90 (2s, 3H, N=CCH_3), 1.87, 1.90 (2s, 3H, 4-CH_3), 2.22-2.36 (m, 2H, N=C$\underline{CH_2}CH_3$), 2.63-2.82 (m, 2H, CH_2CO), 3.40-3.52 (m, 0.4H, CH), 3.85-4.10 (m, 2H, CH_2N + 0.6H, CH), 7.33-7.89 (m, 3H, Ar), 10.21, 10.27, 10.35, 10.42 (4s, 1H, NH).

4b	322.5 $[M+H]^+$, (100), 324.5 $[M+2+H]^+$, (33).	3239 (NH), 1694 (C=O), 1652 (C=O), 823-650 (Ar).	0.99, 1.02, 1.03, 1.04 (4t, J=7.4, 3H, $N=CCH_2\underline{CH_3}$), 1.86, 1.88, 1.92, 1.93 (4s, 3H, $N=CCH_3$), 2.13, 2.15, 2.16 (3s, 3H, 2-CH_3), 2.20-2.35 (m, 2H, $N=C\underline{CH_2}CH_3$), 2.55-2.75 (m, 2H, CH_2CO), 3.45-3.55 (m, 0.4H, CH), 3.66-4.06 (m, 2H, CH_2N + 0.6H, CH), 7.32-7.39 (m, 3H, Ar), 10.19, 10.26, 10.36, 10.44 (4s, 1H, NH).
5a	436.3 $[M+H]^+$, (100), 438.3 $[M+2+H]^+$, (33).	3133 (NH), 1689 (C=O), 1661 (C=O), 879-689 (Ar).	2.31 (s, 3H, 4-CH_3), 2.70-2.91 (m, 2H, CH_2CO), 3.28-3.42 (m, 0.3H, CH), 3.94-4.18 (m, 2H, CH_2N + 0.7H, CH), 7.33-7.89 (m, 7H, Ar), 8.02, 8.21 (s, 1H, CH=N), 11.68, 11.74 (2s, 1H, NH).
5b	436.4 $[M+H]^+$, (100), 438.3 $[M+2+H]^+$, (33).	3127 (NH), 1691 (C=O), 1670 (C=O), 876-648 (Ar).	2.16, 2.17 (2s, 3H, 2-CH_3), 2.62-2.84 (m, 2H, CH_2CO), 3.37-3.47 (m, 0.3H, CH), 3.77-4.21 (m, 2H, CH_2N + 0.7H, CH), 7.32-7.86 (m, 7H, Ar), 8.02, 8.20 (s, 1H, CH=N), 11.65, 11.72 (2s, 1H, NH).
6a	401.4 $[M+H]^+$, (100), 403.4$[M+2+H]^+$, (33).	3130 (NH), 1690 (C=O), 1656 (C=O), 874-689 (Ar).	2.31 (s, 3H, 4-CH_3), 2.72-2.95 (m, 2H, CH_2CO), 3.30-3.42 (m, 0.2H, CH), 3.96-4.22 (m, 2H, CH_2N + 0.8H, CH), 7.34-8.34 (m, 7H, Ar), 8.15, 8.29 (s, 1H, CH=N), 11.90, 11.97 (2s, 1H, NH).
6b	401.4 $[M+H]^+$, (100), 403.4 $[M+2+H]^+$, (33).	3012 (NH), 1686 (C=O), 1663 (C=O), 852-692 (Ar).	2.16, 2.17 (2s, 3H, 2-CH_3), 2.63-2.87 (m, 2H, CH_2CO), 3.42-3.44 (m, 0.2H, CH), 3.79-4.25 (m, 2H, CH_2N +0.8H, CH), 7.32-8.32 (m, 7H, Ar), 8.14, 8.29 (s, 1H, CH=N), 11.88, 11.94 (2s, 1H, NH).
7a	372.4 $[M+H]^+$, (100), 374.4 $[M+2+H]^+$, (33).	3201 (NH), 3073 (OH), 1659 (C=O, 834-692 (Ar).	2.31 (s, 3H, 4-CH_3), 2.69-2.90 (m, 2H, CH_2CO), 3.26-3.39 (m, 0.3H, CH), 3.92-4.17 (m, 2H, CH_2N + 0.7H, CH), 6.81-7.90 (m, 7H, Ar), 7.95, 8.11 (s, 1H, CH=N), 9.93, 9.96 (2s, 1H, OH), 11.41, 11.47 (2s, 1H, NH).
7b	372.4 $[M+H]^+$, (100), 374.4 $[M+2+H]^+$, (33).	3308 (NH), 3189(OH), 1663 (C=O), 1610 (C=O), 836-648 (Ar).	2.16, 2.17 (2s, 3H, 2'-CH_3), 2.60-2.83 (m, 2H, CH_2CO), 3.36-3.46 (m, 0.2H, CH), 3.76-4.17 (m, 2H, CH_2N + 0.8H, CH), 6.81-7.56 (m, 7H, Ar), 7.94, 8.11 (2s, 1H, CH=N), 9.91, 9.95 (2s, 1H, OH), 11.37, 11.42 (2s, 1H, NH).
8a*	399.5 $[M+H]^+$, (100), 401.5 $[M+2+H]^+$, (33).	2904 (NH), 1699 (C=O), 1664 (C=O), 848-640 (Ar).	2.37 (s, 3H, 4-CH_3), 2.77-3.18 (m, 2H, CH_2CO), 3.06 (s, 6H, $N(CH_3)_2$), 3.91-4.18 (m, 3H, CH_2N + 1H, CH), 6.70-7.71 (m, 7H, Ar), 7.75 (s, 1H, CH=N), 9.32 (s, 1H, NH).

8a			2.31 (s, 3H, 4-CH_3), 2.59-2.86 (m, 2H, CH_2CO), 2.96, 2.97 (2s, 6H, $N(CH_3)_2$), 3.12-3.30 (m, 0.3H, CH), 3.81-4.17 (m, 2H, CH_2N + 0.7H, CH), 6.72-7.91 (m, 7H, Ar), 7.92, 8.08 (2s, 1H, CH=N), 11.33, 11.37 (2s, 1H, NH).
8b	399.5 $[M+H]^+$, (100), 401.5 $[M+2+H]^+$, (33).	2913 (NH), 1691 (C=O), 1669 (C=O), 812-678 (Ar).	2.16, 2.17 (2s, 3H, 2-CH_3), 2.59-2.79 (m, 2H, CH_2CO), 2.97, 2.98, 3.01 (3s, 6H, $N(CH_3)_2$), 3.32-3.42 (m, 0.3H, CH), 3.75-4.16 (m, 2H, CH_2N + 0.7H, CH), 6.72-7.68 (m, 7H, Ar), 7.91, 8.06 (2s, 1H, CH=N), 11.30, 11.33 (2s, 1H, NH).
9a	473.4 $[M+H]^+$, (100), 475.5 $[M+2+H]^+$, (33).	2973 (NH), 1689 (C=O), 1667 (C=O), 805-695 (Ar).	1.33, 1.34 (2t, 3H, J = 7.0, $NCH_2\underline{CH_3}$), 2.30, 2.31 (2s, 3H, 4-CH_3), 2.74-2.98 (m, 2H, CH_2CO), 3.32-3.42 (m, 0.4H, CH), 3.91-4.26 (m, 2H, CH_2N + 0.6H, CH), 4.47 (k, 2H, J = 7.0, $N\underline{CH_2}CH_3$), 7.22-8.47 (m, 10H, Ar), 8.23, 8.39 (2s, 1H, CH=N), 11.56, 11.61 (2s, 1H, NH).
9b	473.4 $[M+H]^+$, (100), 475.5 $[M+2+H]^+$, (33).	2974 (NH), 1670 (C=O, 810-728(Ar).	1.33, 1.36 (2t, 3H, J = 7.0, $NCH_2\underline{CH_3}$), 2.19 (s, 3H, 2-CH_3), 2.66-2.91 (m, 2H, CH_2CO), 3.38-3.48 (m, 0.4H, CH), 3.74-4.30 (m, 2H, CH_2N + 0.6H, CH), 4.50, 4.60 (2k, 2H, J = 7.0, $N\underline{CH_2}CH_3$), 7.23-8.69 (m, 10H, Ar), 8.22, 8.39 (2s, 1H, CH=N), 11.54, 11.61 (2s, 1H, NH).
10a	332.5 $[M+H]^+$, (100), 334.5 $[M+2+H]^+$, (33).	1735 (C=O), 1693 (C=O), 884-694 (Ar).	2.23, 2.31 (2s, 6H, c'-or e'-CH_3), 2.50 (s, 3H, 4-CH_3), 2.81-2.98 (m, 2H, CH_2CO), 4.01-4.24 (m, 2H, CH_2N), 4.44-4.55 (m, 1H, CH), 6.26 (s, 1H, CH=C), 7.35-7.87 (m, 3H, Ar).
10b	332.5 $[M+H]^+$, (100), 334.5 $[M+2+H]^+$, (33).	1722 (C=O), 1699 (C=O), 843-686 (Ar).	2.15, 2.21 (2s, 6H, c'or e'-CH_3), 2.52 (s, 3H, 2-CH_3), 2.72-2.90 (m, 2H, CH_2CO), 3.79-4.14 (m, 2H, CH_2N), 4.50-4.59 (m, H, CH), 6.25 (s, 1H, CH=C), 7.32-7.41(m, 3H, Ar).

** The compound was dissolved in $CDCl_3$.*

Figure 1. ^{1}H- NMR spectral region of NH resonances of compounds **3a**, **4a** and **8a**