Monatsh Chem (2010) 141:1123–1133
DOI 10.1007/s00706-010-0366-5

ORIGINAL PAPER

Synthesis of novel 6-(substituted amino)-4-(4-ethoxyphenyl)-1-phenyl-2(1*H*)-pyridinones via azo coupling

N. B. Patel · A. I. Gandhi · R. D. Sharma

Received: 2 August 2009 / Accepted: 1 July 2010 / Published online: 5 August 2010

Abstract 6-(Substituted amino)-4-(4-ethoxyphenyl)-1-phenyl-2(1*H*)-pyridinones were prepared from *β*-aryl glutaconic acid, which, on fusion with aniline, results in 4-(4-ethoxyphenyl)-1-phenylpyridine-2,6(1*H*,5*H*)-dione. This, on further treatment with phosphorus oxychloride gave 6-chloro-4-(4-ethoxyphenyl)-1-phenyl-2(1*H*)-pyridinone, and further treatment with secondary amines yielded 6-(substituted amino)-4-(4-ethoxyphenyl)-1-phenyl-2(1*H*)-pyridinones. These were subjected to azo coupling with different aryldiazonium chlorides furnishing two isomers, which were separated by column chromatography. All compounds were characterized by elemental analysis, and use of IR and NMR spectral data, and were evaluated for antimicrobial activity.

Keywords 2-Pyridinone · Secondary amines · Aryldiazonium chlorides · Antimicrobial activity

Introduction

2-Pyridinones are an important type of heterocycles with a variety of biological activity. In particular 2-pyridinones containing an H-bond acceptor substituent in position 5 are a relatively new class of specific phosphodiesterase 3 (PDE3) inhibitors [1]. Some of these, e.g. amrinone [2] and milrinone [3], are good alternatives to classic digitalis glycosides for acute treatment of congestive heart failure (CHF). Substituted pyridinones and their dihydro/tetrahydro derivatives are found in a wide variety of naturally occurring alkaloids, and compounds with these structural motifs have been shown to have significant pharmacological properties [4].

Pyridinones have been reported to have non-nucleoside HIV type I specific reverse transcriptase inhibitor [5] and anti-inflammatory [6] activity, besides a wide range of pharmacological activity. They are well known for their diverse biological activity [7], for example fungicidal, bactericidal, insecticidal, herbicidal, virucidal, anti-tubercular, and cardiovascular. 2-Pyridinones have also been reported to be tissue factor VIIa inhibitors [8]. Cyclopenta[*b*]pyridin-2,5-dione is an interesting building block for access to 2-cyclopenta[*b*]pyridin-5-ones, seco analogues of 8-azasteroids [9], and it also has antiviral activity [10, 11].

Orally administrated piperazine is almost devoid of pharmacological activity. Intravenous administration results in a transient fall in blood pressure. Lethal doses cause convulsions and respiratory depression. A large number of substituted piperazine derivatives have anathematic activity. Various derivatives of piperazine have actual or potential use as antihistamines, anaesthetics, analgesics, anticonvulsants, antispasmodics, and for treatment of hypertension, epilepsy, burn shock, and haemorrhagic shock. Other, non-medical, applications of piperazine and its derivatives are as rubber antioxidants, corrosion inhibitors, wetting and emulsifying additives, in the cosmetics and dyeing industries, in resins, polymers, and synthetic fibres, and as analytical reagents [12–14].

Keeping these applications of 2-pyridinones and piperazine in mind, and in continuation of our earlier work, we decided to synthesize 6-amino-substituted 2(1*H*)-pyridinones from *β*-aryl glutaconic acid [15]. Substituted 2-pyridinones have been prepared by nucleophilic substitution reactions using heterocyclic groups, for example methyl/ethyl piperazinyl, etc. under basic conditions. These

N. B. Patel (✉) · A. I. Gandhi · R. D. Sharma
Department of Chemistry, Veer Narmad South Gujarat University, Udhna Magdalla Road, Surat 395007, Gujarat, India
e-mail: drnavin@satyam.net.in

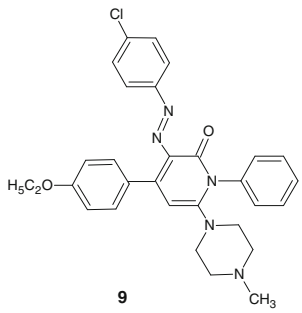
substituted pyridinones have two nucleophilic positions, 3 and 5 [16–18]. To check the nucleophilicity of these positions they were subjected for electrophilic substitution and then characterized by use of spectral data [19, 20]. The purpose of this work was to observe any variation in antimicrobial properties after structural changes and SAR studies.

Result and discussion

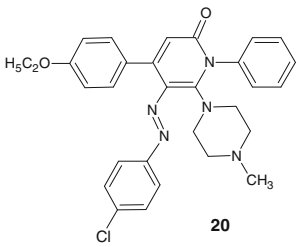
Synthesis

Compounds **4** and **5** were subjected to azo coupling. Two azo-coupled products were obtained, which were separated by column chromatography on activated silica gel using ethyl acetate–chloroform (60:40) as mobile phase. The compounds were characterized as 6-(substituted amino)-3-(arylazo)-4-(4-ethoxyphenyl)-1-phenyl-2(1*H*)-pyridinones **6–16** and **30–40** and their 5-(arylazo) isomers **17–27** and **41–51** (Scheme 1, Table 1) on the basis of spectral data.

In the ^{1}H NMR spectrum of 3-(4-chlorophenylazo)-4-(4-ethoxyphenyl)-6-(4-methylpiperazinyl)-1-phenyl-2(1*H*)-pyridinone (**9**), a proton singlet was observed at 5.9 ppm because of the proton at the C-5 position, whereas for 5-(4-chlorophenylazo)-4-(4-ethoxyphenyl)-6-(4-methylpiperazinyl)-1-phenyl-2(1*H*)-pyridinone (**20**), a proton singlet was observed at 6.5 ppm because of the proton at C-3 position.



9



20

After having checked the nucleophilicity at positions 3 and 5, a Mannich reaction was also carried out using secondary amines, for example 1-methylpiperazine and 1-ethylpiperazine. The resulting compounds **28**, **29**, **52**, and **53** prove that position 3 is more nucleophilic than position 5 [21].

Antimicrobial activity

Antibacterial activity against two Gram-positive (*S. aureus* and *B. subtilius*) and two Gram-negative (*E. coli* and *P. vulgarius*) bacteria at two concentrations (200 and 400 μg/cm^3) was tested by use of the diffusion method. This experimental testing of antibacterial activity did not produce results comparable with those from the standard drug, cefoperazone, even when the substituents at position C-6 of the pyridinones were changed to methylpiperazine and ethylpiperazine [14, 22].

Antifungal activity was tested against *C. albicans* by use of the diffusion method. This showed that 6-(substituted amino)-2(1*H*)-pyridinones with a methylpiperazine and ethylpiperazine moiety at the C-6 position of the pyridinones were inactive. These derivatives were subjected to azo coupling and the products obtained were tested for the antifungal activity in the same way. Mild activity was observed; ethylpiperazine derivatives had no activity. Therefore it can be concluded that introduction of a diazo group to either the C-3 or C-5 position of the pyridinone its antifungal activity is lost [20, 22, 23].

Experimental

Melting points were determined by means of the open capillary method. IR spectra (KBr pellets) were recorded on a Perkin–Elmer spectrophotometer. NMR spectra were recorded in $CDCl_3$ on a Bruker Avance II 400 MHz instrument using TMS as internal standard. Elemental analysis was performed by means of a Heraeus CHN rapid analyser; results agreed satisfactorily with calculated values. For TLC analysis, silica gel coated Al plates (Merck) were used. 6-Chloro-4-(4-ethoxyphenyl)-1-phenyl-2(1*H*)-pyridinone (**3**) was synthesized from 1,3-acetonedicarboxylic acid via **1** and **2** according to a procedure published elsewhere [15].

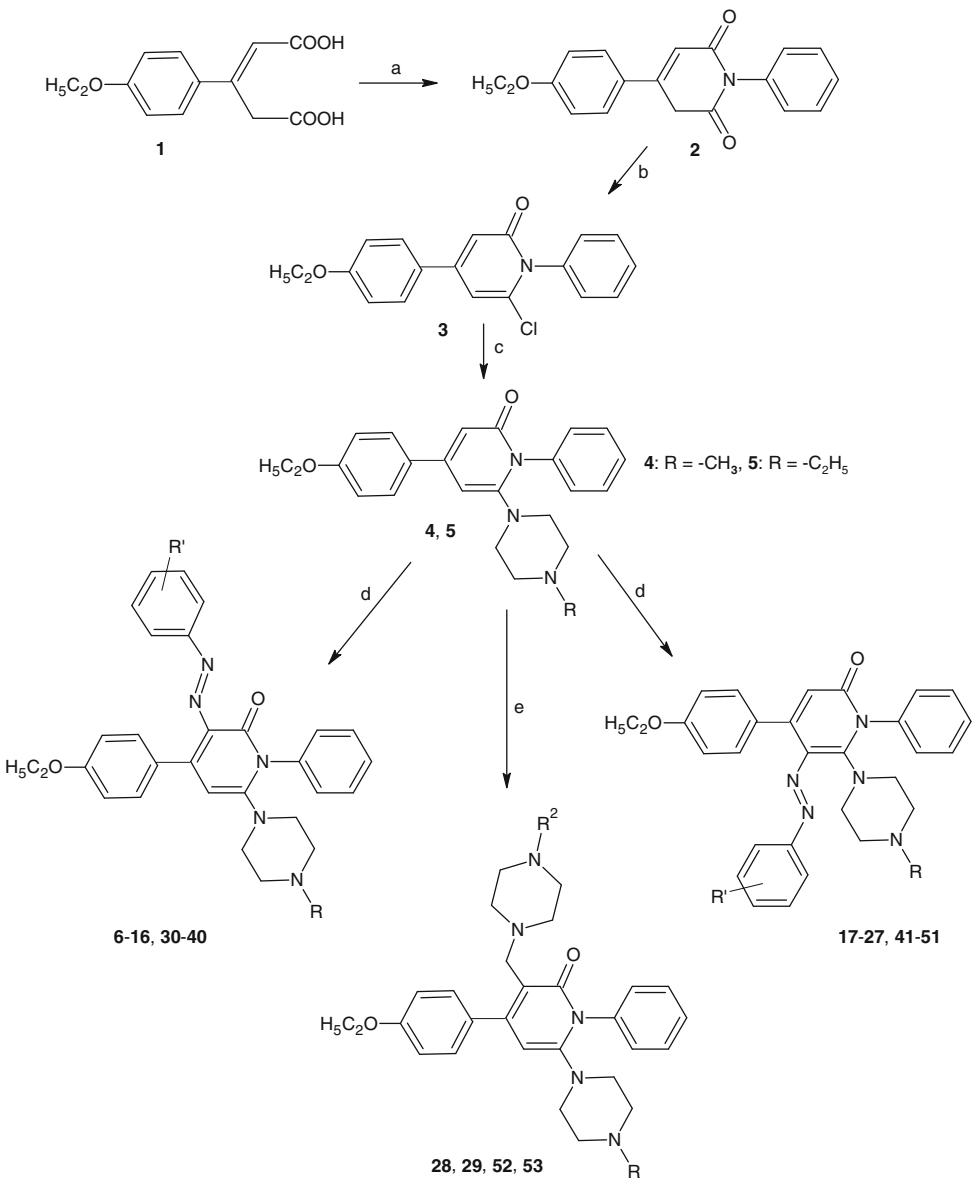
4-(4-Ethoxyphenyl)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone (**4**, $C_{24}H_{27}N_3O_2$)

A mixture of 3.255 g (0.01 mol) **3** and 2.0 g (0.01 mol) 1-methylpiperazine was heated under reflux for 12 h. The reaction mass was then poured into acidic crushed ice and the resulting solid was filtered, washed with water, and crystallized from acetone to furnish 3.3 g (84.8%) **4**. M.p.: 199–201 °C; IR (KBr): $\bar{\nu}$ = 3,020, 2,920 (C–H), 1,700, 1,660 (C=O), 1,245 (C–O–C), 1,430 (C–N) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.5 (s, 3H, NCH_3), 2.7–3.4 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.0 (d, 1H, C_5–H), 6.6 (d, 1H, C3–H), 7.0–7.7 (m, 9H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.9, 55.3, 64.7, 74.9, 114.4, 114.4, 118.9, 120.4, 121.6, 121.6, 124.4, 129.0, 129.0, 129.4, 129.4, 132.8, 138.8, 156.7, 157.5, 160.7 ppm; MS (EI): *m*/*z* = 389; HRMS (EI): *m*/*z* calcd $[M]^+$ 389.2103, found 389.2101.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-1-phenyl-2(1H)-pyridinone (**5**, $C_{25}H_{29}N_3O_2$)

Compound **5** was prepared similarly to **4** but by reaction of **3** with 1-ethylpiperazine. Yield 3.2 g (79.4%); m.p.:

Scheme 1



207–209 °C; IR (KBr): $\bar{\nu}$ = 3,020, 2,920 (C–H), 1,700, 1,660 (C=O), 1,245 (C–O–C), 1,430 (C–N) cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (m, 6H, CH_3), 2.5 (q, 2H, Pip–NCH_2), 2.7–3.4 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.0 (d, 1H, C_5–H), 6.6 (d, 1H, C_3–H), 7.0–7.7 (m, 9H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 49.2, 49.2, 49.2, 52.8, 52.8, 64.7, 74.9, 114.4, 114.4, 118.9, 120.4, 121.6, 121.6, 124.4, 129.0, 129.4, 129.4, 132.8, 138.8, 156.7, 157.5, 160.7 ppm; MS (EI): *m*/*z* = 403; HRMS (EI): *m*/*z* calcd $[M]^+$ 403.2260, found 403.2260.

General procedure for 4-(4-ethoxyphenyl)-6-(4-methyl/ethylpiperazinyl)-1-phenyl-3/5-(substituted phenylazo)-2(1H)-pyridinones ***6–27*** *and* ***30–51***

To a precooled (0–5 °C) solution of **4** or **5** (0.01 mol) in experimental; after 100 cm^3 acetone, a precooled (0–5 °C) solution of benzenediazonium chloride was added (prepared by diazotizing substituted aniline (0.01 mol) in 6 cm^3 1:1 HCl with 0.69 g (0.01 mol) $NaNO_2$ in 10 cm^3 water). The reaction mixture was poured into 200 cm^3 water and then made alkaline by addition of dilute NaOH.

Table 1 Characterization data for compounds **6–53**

Compound number	R	R^2	R′	M.p. (°C)	Yield (%)
6	CH_3		H	172–174	36
7	CH_3		*o*-Cl	183–185	35
8	CH_3		*m*-Cl	179–181	36
9	CH_3		*p*-Cl	189–191	35
10	CH_3		*o*-CH_3	163–165	34
11	CH_3		*m*-CH_3	170–172	35
12	CH_3		*p*-CH_3	155–157	35
13	CH_3		*o*-NO_2	199–201	35
14	CH_3		*m*-NO_2	186–188	35
15	CH_3		*p*-NO_2	179–181	35
16	CH_3		*o*-OCH_3	160–162	35
17	CH_3		H	148–150	35
18	CH_3		*o*-Cl	161–163	35
19	CH_3		*m*-Cl	182–184	34
20	CH_3		*p*-Cl	168–170	34
21	CH_3		*o*-CH_3	150–152	34
22	CH_3		*m*-CH_3	144–146	35
23	CH_3		*p*-CH_3	149–151	35
24	CH_3		*o*-NO_2	202–204	33
25	CH_3		*m*-NO_2	207–209	36
26	CH_3		*p*-NO_2	189–191	34
27	CH_3		*o*-OCH_3	152–154	36
28	CH_3	CH_3	–	182–184	70
29	CH_3	C_2H_5	–	205–207	65
30	C_2H_5		H	169–171	36
31	C_2H_5		*o*-Cl	185–187	34
32	C_2H_5		*m*-Cl	173–175	32
33	C_2H_5		*p*-Cl	193–195	35
34	C_2H_5		*o*-CH_3	142–144	35
35	C_2H_5		*m*-CH_3	159–161	36
36	C_2H_5		*p*-CH_3	152–154	36
37	C_2H_5		*o*-NO_2	172–174	36
38	C_2H_5		*m*-NO_2	149–151	35
39	C_2H_5		*p*-NO_2	168–170	36
40	C_2H_5		*o*-OCH_3	177–179	36
41	C_2H_5		H	170–172	34
42	C_2H_5		*o*-Cl	175–177	34
43	C_2H_5		*m*-Cl	188–190	34
44	C_2H_5		*p*-Cl	179–181	33
45	C_2H_5		*o*-CH_3	152–154	35
46	C_2H_5		*m*-CH_3	160–162	33
47	C_2H_5		*p*-CH_3	146–148	34
48	C_2H_5		*o*-NO_2	166–168	34
49	C_2H_5		*m*-NO_2	175–177	34
50	C_2H_5		*p*-NO_2	181–183	34
51	C_2H_5		*o*-OCH_3	192–194	35
52	C_2H_5	CH_3	–	182–184	70
53	C_2H_5	C_2H_5	–	195–197	65

The coloured product was filtered, washed with water, and dried. Two azo-coupled products were obtained; these were separated by column chromatography on activated silica gel using ethyl acetate–chloroform (60:40) as mobile phase.

General procedure for 4-(4-ethoxyphenyl)-6-(4-methyl/ethylpiperazinyl)-3-[(4-methyl/ethyl-1-piperazinyl)-methyl]-1-phenyl-2(1H)-pyridinones ***28, 29, 52, 53***

A mixture of 0.3 g (0.01 mol) paraformaldehyde and 1-methyl or 1-ethylpiperazine (0.01 mol) was heated under reflux for 45 min in 50 cm^3 chlorobenzene. Compound **4** or **5** (0.01 mol) was then added and the reaction mixture was heated under reflux for 22 h. The reaction mass was subjected to steam distillation to remove chlorobenzene. The solid product was filtered, washed, and recrystallized from acetone.

4-(4-Ethoxyphenyl)-6-(4-methylpiperazinyl)-1-phenyl-3-(phenylazo)-2(1H)-pyridinone (**6**, $C_{30}H_{31}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,260 (C–O–C), 1,650 (>C=O), 1,570 (–N=N–), 1,435 (C–N), 3,010, 2,920 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.4 (s, 3H, NCH_3), 2.7–3.5 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.0 (d, 1H, C_5–H), 7.0–7.7 (m, 14H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.9, 48.9, 55.3, 55.3, 64.7, 74.9, 114.4, 114.4, 120.4, 121.6, 121.6, 124.0, 124.4, 128.7, 128.8, 128.8, 128.9, 128.8, 128.9, 129.0, 129.0, 129.4, 129.4, 132.8, 146.5, 156.7, 157.5, 158.4 ppm; MS (EI): *m*/*z* = 493; HRMS (EI): *m*/*z* calcd $[M]^+$ 493.2478, found 493.2475.

3-(2-Chlorophenylazo)-4-(4-ethoxyphenyl)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone
(**7**, $C_{30}H_{30}ClN_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,250 (C–O–C), 1,660 (>C=O), 1,575 (–N=N–), 1,425 (C–N), 3,020, 2,910 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.5 (s, 3H, NCH_3), 2.7–3.5 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 5.9 (d, 1H, C_5–H), 7.0–7.7 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.7, 48.9, 55.4, 55.3, 64.7, 74.9, 114.4, 114.4, 120.3, 121.6, 121.6, 124.0, 124.4, 126.9, 129.0, 129.0, 129.1, 129.4, 129.4, 130.2, 130.2, 132.8, 134.3, 146.5, 156.7, 157.5, 158.4 ppm; MS (EI): *m*/*z* = 527 $[M]^+$, 529 $[M + 2]^+$; HRMS (EI): *m*/*z* calcd $[M]^+$ 527.2088, found 527.2092.

3-(3-Chlorophenylazo)-4-(4-ethoxyphenyl)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone
(**8**, $C_{30}H_{30}ClN_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,250 (C–O–C), 1,670 (>C=O), 1,575 (–N=N–), 1,420 (C–N), 3,020, 2,915 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.4 (t, 3H, CH_3), 2.4 (s, 3H, NCH_3), 2.7–3.5 (m, 8H, Pip–H), 4.1 (q, 2H,

OCH_2), 6.0 (d, 1H, C_5–H), 7.0–7.7 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.9, 48.9, 55.3, 55.3, 64.7, 74.9, 114.2, 114.4, 120.4, 121.6, 121.6, 124.0, 124.4, 126.9, 128.9, 129.0, 129.0, 129.1, 129.2, 129.4, 130.1, 130.2, 132.8, 134.3, 146.4, 156.7, 157.5, 158.4 ppm; MS (EI): *m/z* = 527 $[M]^+$, 529 $[M + 2]^+$; HRMS (EI): *m/z* calcd $[M]^+$ 527.2088, found 527.2086.

3-(4-Chlorophenylazo)-4-(4-ethoxyphenyl)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone
(**9**, $C_{30}H_{30}ClN_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,265 (C–O–C), 1,665 (>C=O), 1,560 (–N=N–), 1,435 (C–N), 3,025, 2,910 (C–H) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.5 (s, 3H, NCH_3), 2.7–3.5 (m, 8H, Pip–H), 3.9–4.0 (q, 2H, OCH_2), 5.9 (s, 1H, C_5–H), 7.0–7.7 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.9, 48.9, 55.2, 55.3, 64.7, 74.9, 114.4, 114.4, 120.4, 121.6, 121.6, 124.0, 124.4, 126.8, 128.7, 128.9, 129.0, 129.1, 129.4, 129.4, 130.2, 130.2, 132.8, 134.3, 146.5, 156.7, 157.5, 158.4 ppm; MS (EI): *m/z* = 527 $[M]^+$, 529 $[M + 2]^+$; HRMS (EI): *m/z* calcd $[M]^+$ 527.2088, found 527.2089.

4-(4-Ethoxyphenyl)-3-(2-methylphenylazo)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone
(**10**, $C_{31}H_{33}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,255 (C–O–C), 1,655 (>C=O), 1,570 (–N=N–), 1,435 (C–N), 3,030, 2,910 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.4 (s, 6H, CH_3), 2.7–3.5 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.0 (d, 1H, C_5–H), 7.0–7.7 (m, 14H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 15.2, 43.1, 48.9, 48.9, 55.3, 55.3, 64.7, 74.9, 114.3, 114.4, 120.4, 121.6, 121.6, 124.0, 124.4, 125.8, 128.1, 128.7, 128.7, 129.0, 129.0, 129.1, 129.4, 129.4, 132.8, 137.1, 146.5, 156.7, 157.5, 158.4 ppm; MS (EI): *m/z* = 507; HRMS (EI): *m/z* calcd $[M]^+$ 507.2634, found 507.2629.

4-(4-Ethoxyphenyl)-3-(3-methylphenylazo)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone
(**11**, $C_{31}H_{33}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,240 (C–O–C), 1,660 (>C=O), 1,565 (–N=N–), 1,420 (C–N), 3,025, 2,915 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.3 (s, 6H, CH_3), 2.7–3.5 (m, 8H, Pip–H), 3.9–4.1 (q, 2H, OCH_2), 6.0 (d, 1H, C_5–H), 7.0–7.7 (m, 14H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 24.3, 43.1, 48.9, 48.9, 55.3, 55.3, 64.7, 74.9, 114.4, 114.4, 120.4, 121.6, 121.6, 124.0, 124.4, 125.8, 128.6, 128.7, 129.0, 129.0, 129.1, 129.4, 130.4, 132.7, 138.4, 146.5, 156.7 ppm; MS (EI): *m/z* = 507; HRMS (EI): *m/z* calcd $[M]^+$ 507.2634, found 507.2630.

4-(4-Ethoxyphenyl)-3-(4-methylphenylazo)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone
(**12**, $C_{31}H_{33}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,250 (C–O–C), 1,650 (>C=O), 1,560 (–N=N–), 1,425 (C–N), 3,010, 2,910 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.4 (s, 6H, CH_3), 2.6–3.4 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 5.9 (d, 1H, C_5-H), 7.0–7.7 (m, 14H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 24.3, 43.1, 48.9, 48.9, 55.3, 55.3, 64.7, 74.9, 113.0, 114.3, 114.3, 121.6, 121.6, 124.2, 124.4, 125.7, 127.3, 127.0, 128.7, 128.7, 129.0, 129.1, 129.1, 129.1, 132.8, 138.4, 155.0, 156.7, 156.7, 158.4 ppm; MS (EI): *m/z* = 507; HRMS (EI): *m/z* calcd $[M]^+$ 507.2634, found 507.2635.

4-(4-Ethoxyphenyl)-6-(4-methylpiperazinyl)-3-(2-nitrophenylazo)-1-phenyl-2(1H)-pyridinone
(**13**, $C_{30}H_{30}N_6O_4$)
IR (KBr): $\bar{\nu}$ = 1,245 (C–O–C), 1,660 (>C=O), 1,570 (–N=N–), 1,435 (C–N), 3,020, 2,910 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.4 (s, 3H, NCH_3), 2.7–3.5 (m, 8H, Pip–H), 3.9–4.0 (q, 2H, OCH_2), 6.0 (d, 1H, C_5–H), 7.0–7.7 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.9, 48.9, 55.3, 55.3, 64.7, 74.9, 113.0, 114.3, 114.3, 121.1, 121.6, 121.6, 124.0, 124.2, 127.0, 127.0, 129.0, 129.7, 129.7, 132.8, 134.9, 148.0, 155.0, 156.7, 156.7, 156.7, 158.4 ppm; MS (EI): *m/z* = 538; HRMS (EI): *m/z* calcd $[M]^+$ 538.2329, found 538.2326.

4-(4-Ethoxyphenyl)-6-(4-methylpiperazinyl)-3-(3-nitrophenylazo)-1-phenyl-2(1H)-pyridinone
(**14**, $C_{30}H_{30}N_6O_4$)
IR (KBr): $\bar{\nu}$ = 1,260 (C–O–C), 1,655 (>C=O), 1,570 (–N=N–), 1,430 (C–N), 3,020, 2,920 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.4 (t, 3H, CH_3), 2.4 (s, 3H, NCH_3), 2.7–3.4 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.0 (d, 1H, C_5–H), 7.0–7.6 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.7, 48.9, 55.3, 55.3, 64.7, 74.9, 112.9, 114.3, 114.3, 121.1, 121.6, 121.6, 124.0, 124.2, 124.4, 127.0, 127.1, 129.0, 129.0, 129.6, 129.7, 132.8, 134.9, 148.0, 155.0, 156.7, 156.8, 158.4 ppm; MS (EI): *m/z* = 538; HRMS (EI): *m/z* calcd $[M]^+$ 538.2329, found 538.2325.

4-(4-Ethoxyphenyl)-6-(4-methylpiperazinyl)-3-(4-nitrophenylazo)-1-phenyl-2(1H)-pyridinone
(**15**, $C_{30}H_{30}N_6O_4$)
IR (KBr): $\bar{\nu}$ = 1,250 (C–O–C), 1,650 (>C=O), 1,575 (–N=N–), 1,435 (C–N), 3,010, 2,925 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.5 (s, 3H, NCH_3), 2.7–3.5 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.0 (d, 1H, C_5–H), 6.9–7.7 (m, 13H, Ar–H) ppm;

^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.9, 48.9, 55.3, 55.3, 64.7, 74.9, 113.0, 114. 3, 114.3, 121.1, 121.1, 121.6, 121.6, 124.2, 124.4, 126.9, 127.0, 129.0, 129.0, 129.7, 129.7, 132.8, 134.8, 148.0, 155.0, 156.6, 156.7, 158.4 ppm; MS (EI): *m/z* = 538; HRMS (EI): *m/z* calcd $[M]^+$ 538.2329, found 538.2334.

4-(4-Ethoxyphenyl)-3-(2-methoxyphenylazo)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone (**16**, $C_{31}H_{33}N_5O_3$)
IR (KBr): $\bar{\nu}$ = 1,255, 1,240 (C–O–C), 1,670 (>C=O), 1,565 (–N=N–), 1,425 (C–N), 3,020, 2,915 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.3 (s, 3H, CH_3), 2.6–3.3 (m, 8H, Pip–H), 3.6 (s, 3H, OCH_3), 4.2 (q, 2H, NCH_2), 6.0 (s, 1H, C_5–H), 7.0–7.8 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.9, 48.9, 55.3, 55.3, 55.9, 64.7, 74.1, 99.6, 113.0, 114.3, 114.3, 114.3, 114.3, 121.1, 121.6, 121.6, 124.2, 124.4, 127.0, 127.0, 129.0, 129.2, 129.7, 129.8, 132.8, 155.0, 156.6, 156.7, 158.4, 159.1 ppm; MS (EI): *m/z* = 523; HRMS (EI): *m/z* calcd $[M]^+$ 523.2583, found 523.2576.

4-(4-Ethoxyphenyl)-6-(4-methylpiperazinyl)-1-phenyl-5-(phenylazo)-2(1H)-pyridinone (**17**, $C_{30}H_{31}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,250 (C–O–C), 1,660 (>C=O), 1,565 (–N=N–), 1,430 (C–N), 3,020, 2,910 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.5 (s, 3H, NCH_3), 2.7–3.5 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C_3–H), 7.0–7.7 (m, 14H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.9, 49.0, 55.3, 55.3, 64.7, 75.0, 114.4, 114.4, 118.9, 120.4, 121.6, 121.6, 124.4, 128.7, 128.8, 128.8, 128.8, 128.9, 128.9, 129.0, 129.0, 129.4, 129.4, 132.8, 146.5, 157.0, 157.5, 160.7 ppm; MS (EI): *m/z* = 493; HRMS (EI): *m/z* calcd $[M]^+$ 493.2478, found 493.2479.

5-(2-Chlorophenylazo)-4-(4-ethoxyphenyl)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone (**18**, $C_{30}H_{30}ClN_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,255 (C–O–C), 1,650 (>C=O), 1,570 (–N=N–), 1,420 (C–N), 3,025, 2,920 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.4 (s, 3H, NCH_3), 2.7–3.5 (m, 8H, Pip–H), 3.9–4.0 (q, 2H, OCH_2), 6.5 (d, 1H, C3–H), 7.0–7.7 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.9, 48.9, 55.2, 55.3, 64.7, 75.0, 114.3, 114.4, 118.9, 120.4, 121.6, 121.6, 124.4, 126.9, 128.9, 129.0, 129.0, 129.1, 129.4, 129.4, 130.2, 130.2, 132.8, 134.3, 146.5, 157.1, 157.5, 160.7 ppm; MS (EI): *m/z* = 527 $[M]^+$, 529 $[M + 2]^+$; HRMS (EI): *m/z* calcd $[M]^+$ 527.2088, found 527.2083.

5-(3-Chlorophenylazo)-4-(4-ethoxyphenyl)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone (**19**, $C_{30}H_{30}ClN_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,240 (C–O–C), 1,655 (>C=O), 1,560 (–N=N–), 1,425 (C–N), 3,025, 2,910 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.4 (t, 3H, CH_3), 2.4 (s, 3H, NCH_3), 2.7–3.4 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C3–H), 7.0–7.6 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.2, 48.9, 48.9, 55.3, 55.3, 64.7, 75.0, 114.4, 114.4, 118.9, 120.4, 121.6, 121.6, 124.4, 126.9, 128.9, 129.0, 129.0, 129.1, 129.4, 129.4, 130.1, 130.1, 132.8, 134.3, 146.5, 157.0, 157.5, 160.7 ppm; MS (EI): *m/z* = 527 $[M]^+$, 529 $[M + 2]^+$; HRMS (EI): *m/z* calcd $[M]^+$ 527.2088, found 527.2085.

5-(4-Chlorophenylazo)-4-(4-ethoxyphenyl)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone (**20**, $C_{30}H_{30}ClN_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,245 (C–O–C), 1,660 (>C=O), 1,565 (–N=N–), 1,430 (C–N), 3,020, 2,915 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.5 (s, 3H, NCH_3), 2.7–3.4 (m, 8H, Pip–H), 3.9–4.0 (q, 2H, OCH_2), 6.6 (s, 1H, C3–H), 6.9–7.6 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.9, 48.9, 55.3, 55.3, 64.6, 75.0, 114.4, 114.4, 118.9, 120.4, 121.6, 121.6, 124.4, 126.8, 128.9, 128.9, 129.0, 129.1, 129.4, 129.4, 130.2, 130.2, 132.8, 134.3, 146.5, 157.1, 157.5, 160.7 ppm; MS (EI): *m/z* = 527 $[M]^+$, 529 $[M + 2]^+$; HRMS (EI): *m/z* calcd $[M]^+$ 527.2088, found 527.2087.

4-(4-Ethoxyphenyl)-5-(2-methylphenylazo)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone (**21**, $C_{31}H_{33}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,245 (C–O–C), 1,650 (>C=O), 1,575 (–N=N–), 1,445 (C–N), 3,020, 2,920 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.5 (s, 6H, CH_3), 2.7–3.4 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C3–H), 6.9–7.6 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 15.2, 43.1, 48.9, 48.9, 55.3, 55.3, 64.7, 75.0, 114.4, 114.4, 118.9, 120.4, 121.6, 121.6, 124.4, 125.8, 128.1, 128.7, 128.7, 129.0, 129.0, 129.1, 129.4, 129.4, 132.8, 137.1, 146.5, 157.0, 157.5, 160.7 ppm; MS (EI): *m/z* = 507; HRMS (EI): *m/z* calcd $[M]^+$ 507.2634, found 507.2631.

4-(4-Ethoxyphenyl)-5-(3-methylphenylazo)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone (**22**, $C_{31}H_{33}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,265 (C–O–C), 1,665 (>C=O), 1,570 (–N=N–), 1,430 (C–N), 3,020, 2,910 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.4 (s, 6H, CH_3), 2.7–3.4 (m, 8H, Pip–H), 4.0–4.1 (q, 2H,

OCH_2), 6.5 (d, 1H, C3–H), 7.0–7.6 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 24.3, 43.1, 48.9, 48.8, 55.3, 55.3, 64.7, 75.0, 114.3, 114.4, 118.9, 120.4, 121.6, 121.6, 124.4, 125.8, 128.6, 128.7, 129.0, 129.1, 129.1, 129.4, 129.4, 130.4, 132.8, 138.4, 146.4, 157.0, 157.5, 160.7 ppm; MS (EI): *m/z* = 507; HRMS (EI): *m/z* calcd $[M]^+$ 507.2634, found 507.2634.

4-(4-Ethoxyphenyl)-5-(4-methylphenylazo)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone
(**23**, $C_{31}H_{33}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,255 (C–O–C), 1,660 (>C=O), 1,565 (–N=N–), 1,435 (C–N), 3,020, 2,920 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.5 (s, 6H, CH_3), 2.7–3.4 (m, 8H, Pip–H), 3.9–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C3–H), 6.9–7.5 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 24.3, 43.1, 48.9, 48.9, 55.3, 55.3, 64.7, 75.0, 114.4, 114.4, 118.9, 120.4, 121.6, 121.6, 124.4, 125.7, 128.7, 128.7, 128.8, 129.0, 129.1, 129.1, 129.4, 129.4, 132.8, 138.4, 146.6, 157.0, 157.5, 160.7 ppm; MS (EI): *m/z* = 507; HRMS (EI): *m/z* calcd $[M]^+$ 507.2634, found 507.2629.

4-(4-Ethoxyphenyl)-6-(4-methylpiperazinyl)-5-(2-nitrophenylazo)-1-phenyl-2(1H)-pyridinone
(**24**, $C_{30}H_{30}N_6O_4$)
IR (KBr): $\bar{\nu}$ = 1,240 (C–O–C), 1,665 (>C=O), 1,575 (–N=N–), 1,430 (C–N), 3,015, 2,915 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.4 (s, 3H, NCH_3), 2.8–3.6 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C3–H), 6.9–7.6 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.9, 48.9, 55.2, 55.3, 64.7, 75.0, 114.4, 114.4, 118.9, 120.4, 121.1, 121.6, 121.6, 124.0, 124.4, 129.0, 129.0, 129.4, 129.4, 129.7, 129.7, 132.8, 134.9, 146.5, 147.9, 157.0, 157.5, 160.7 ppm; MS (EI): *m/z* = 538; HRMS (EI): *m/z* calcd $[M]^+$ 538.2329, found 538.2335.

4-(4-Ethoxyphenyl)-6-(4-methylpiperazinyl)-5-(3-nitrophenylazo)-1-phenyl-2(1H)-pyridinone
(**25**, $C_{30}H_{30}N_6O_4$)
IR (KBr): $\bar{\nu}$ = 1,250 (C–O–C), 1,650 (>C=O), 1,560 (–N=N–), 1,425 (C–N), 3,010, 2,910 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.4 (s, 3H, NCH_3), 2.7–3.5 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C3–H), 7.0–7.7 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.9, 48.9, 55.3, 55.3, 64.7, 75.0, 114.4, 114.4, 118.9, 120.4, 121.1, 121.6, 121.6, 124.0, 124.4, 129.0, 129.1, 129.4, 129.4, 129.6, 129.7, 132.8, 134.9, 146.5, 148.0, 157.0, 157.5, 160.7 ppm; MS (EI): *m/z* = 538; HRMS (EI): *m/z* calcd $[M]^+$ 538.2329, found 538.2327.

4-(4-Ethoxyphenyl)-6-(4-methylpiperazinyl)-5-(4-nitrophenylazo)-1-phenyl-2(1H)-pyridinone
(**26**, $C_{30}H_{30}N_6O_4$)
IR (KBr): $\bar{\nu}$ = 1,255 (C–O–C), 1,655 (>C=O), 1,570 (–N=N–), 1,430 (C–N), 3,020, 2,920 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.4 (t, 3H, CH_3), 2.5 (s, 3H, NCH_3), 2.6–3.4 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.5 (d, 1H, C3–H), 7.0–7.7 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.9, 48.9, 55.1, 55.3, 64.7, 75.0, 114.4, 114.5, 118.9, 120.4, 121.1, 121.1, 121.6, 121.6, 124.4, 129.0, 129.0, 129.4, 129.4, 129.7, 129.8, 132.8, 134.8, 146.5, 147.8, 157.0, 157.5 ppm; MS (EI): *m/z* = 538; HRMS (EI): *m/z* calcd $[M]^+$ 538.2329, found 538.2330.

4-(4-Ethoxyphenyl)-5-(2-methoxyphenylazo)-6-(4-methylpiperazinyl)-1-phenyl-2(1H)-pyridinone
(**27**, $C_{31}H_{33}N_5O_3$)
IR (KBr): $\bar{\nu}$ = 1,260, 1,250 (C–O–C), 1,660 (>C=O), 1,570 (–N=N–), 1,420 (C–N), 3,015, 2,910 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.5 (t, 3H, CH_3), 2.4 (s, 3H, CH_3), 2.6–3.5 (m, 8H, Pip–H), 3.6 (s, 3H, OCH_3), 4.2 (q, 2H, NCH_2), 6.6 (s, 1H, C3–H), 7.0–7.8 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 48.8, 48.9, 55.3, 55.3, 55.9, 64.7, 75.0, 99.6, 114.3, 114.4, 114.4, 118.9, 120.4, 121.1, 121.6, 121.7, 124.4, 129.0, 129.1, 129.4, 129.4, 129.8, 129.8, 132.8, 146.5, 157.0, 157.5, 159.1, 160.7 ppm; MS (EI): *m/z* = 523; HRMS (EI): *m/z* calcd $[M]^+$ 523.2583, found 523.2577.

4-(4-Ethoxyphenyl)-6-(4-methylpiperazinyl)-3-[(4-methyl-piperazinyl)methyl]-1-phenyl-2(1H)-pyridinone
(**28**, $C_{30}H_{39}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,260 (C–O–C), 1,650 (>C=O), 1,570 (–N=N–), 1,435 (C–N), 3,010, 2,920 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4 (t, 3H, CH_3), 2.4 (s, 4H, NCH_3), 2.6 (s, 2H, CH_2), 2.3 (q, 2H, NCH_2), 2.7–3.4 (m, 16H, Pip–H), 3.5 (s, 2H, OCH_2), 5.9 (d, 1H, C_5–H), 6.7–7.6 (m, 9H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.8, 43.1, 43.1, 48.9, 48.9, 52.4, 52.4, 55.2, 55.3, 55.3, 55.3, 57.9, 64.7, 74.9, 114.4, 114.4, 120.4, 121.6, 121.6, 124.4, 129.0, 129.0, 129.4, 129.4, 132.8, 134.8, 135.3, 156.7, 157.5, 158.3 ppm; MS (EI): *m/z* = 501; HRMS (EI): *m/z* calcd $[M]^+$ 501.3104, found 501.3105.

4-(4-Ethoxyphenyl)-6-(4-methylpiperazinyl)-3-[(4-ethyl-piperazinyl)methyl]-1-phenyl-2(1H)-pyridinone
(**29**, $C_{31}H_{41}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,260 (C–O–C), 1,650 (>C=O), 1,570 (–N=N–), 1,435 (C–N), 3,010, 2,920 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.5 (t, 3H, CH_3), 2.3 (s, 2H, NCH_3), 2.4 (s, 2H, NCH_3), 2.6 (s, 4H, CH_2), 2.3 (q,

2H, NCH_2), 2.7–3.3 (m, 16H, Pip–H), 3.5 (s, 2H, OCH_2), 6.0 (d, 1H, C_5-H), 6.7–7.6 (m, 9H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 43.1, 48.9, 48.9, 49.2, 52.7, 57.8, 52.8, 52.8, 55.3, 55.3, 57.9, 64.7, 74.9, 114.4, 114.5, 120.4, 121.6, 121.6, 124.4, 129.0, 129.0, 129.4, 129.5, 132.8, 134.8, 135.3, 156.8, 157.5, 158.3 ppm; MS (EI): *m/z* = 515; HRMS (EI): *m/z* calcd $[M]^+$ 515.3260, found 515.3257.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-1-phenyl-3-(phenylazo)-2(1H)-pyridinone (**30**, $C_{31}H_{33}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,255 (C–O–C), 1,665 (>C=O), 1,575 (–N=N–), 1,425 (C–N), 3,020, 2,915 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.4 (m, 6H, CH_3), 2.6 (q, 2H, NCH_2), 2.7–3.4 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.0 (d, 1H, C_5-H), 7.0–7.6 (m, 14H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 49.1, 49.2, 49.2, 52.8, 52.8, 64.7, 74.9, 114.4, 114.4, 120.4, 121.6, 121.6, 124.1, 124.4, 128.7, 128.8, 128.8, 128.8, 128.9, 128.9, 129.0, 129.0, 129.4, 129.4, 132.8, 146.5, 156.7, 157.5, 158.4 ppm; MS (EI): *m/z* = 507; HRMS (EI): *m/z* calcd $[M]^+$ 507.2634, found 507.2631.

3-(2-Chlorophenylazo)-4-(4-ethoxyphenyl)-6-(4-ethylpiperazinyl)-1-phenyl-2(1H)-pyridinone
(**31**, $C_{31}H_{32}ClN_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,250 (C–O–C), 1,655 (>C=O), 1,570 (–N=N–), 1,420 (C–N), 3,010, 2,920 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.4 (m, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.3 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.1 (d, 1H, C_5-H), 7.0–7.6 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.9, 49.2, 49.2, 49.2, 52.8, 52.6, 64.7, 74.9, 114.3, 114.4, 120.4, 121.5, 121.6, 124.0, 124.4, 126.9, 128.9, 129.0, 129.0, 129.1, 129.4, 129.4, 130.2, 130.2, 132.8, 134.3, 146.5, 156.7, 157.5, 158.5 ppm; MS (EI): *m/z* = 541 $[M]^+$, 543 $[M + 2]^+$; HRMS (EI): *m/z* calcd $[M]^+$ 541.2245, found 541.2245.

3-(3-Chlorophenylazo)-4-(4-ethoxyphenyl)-6-(4-ethylpiperazinyl)-1-phenyl-2(1H)-pyridinone
(**32**, $C_{31}H_{32}ClN_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,260 (C–O–C), 1,660 (>C=O), 1,570 (–N=N–), 1,425 (C–N), 3,020, 2,915 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.5 (m, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.6–3.3 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.0 (d, 1H, C_5-H), 7.0–7.7 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 49.1, 49.2, 49.2, 52.8, 52.8, 64.7, 74.9, 114.4, 114.4, 120.4, 121.6, 121.6, 124.1, 124.4, 126.9, 128.9, 129.0, 129.0, 129.1, 129.4, 129.4, 130.1, 130.2, 132.8, 134.3, 146.4, 156.8, 157.5, 158.4 ppm; MS (EI): *m/z* = 541 $[M]^+$, 543 $[M + 2]^+$; HRMS (EI): *m/z* calcd $[M]^+$ 541.2245, found 541.2243.

3-(4-Chlorophenylazo)-4-(4-ethoxyphenyl)-6-(4-ethylpiperazinyl)-1-phenyl-2(1H)-pyridinone
(**33**, $C_{31}H_{32}ClN_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,260 (C–O–C), 1,660 (>C=O), 1,565 (–N=N–), 1,430 (C–N), 3,025, 2,920 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.31.5 (m, 6H, CH_3), 2.4 (q, 2H, NCH_2), 2.7–3.3 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.1 (d, 1H, C_5-H), 7.0–7.6 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 29.4, 49.1, 49.2, 49.2, 52.8, 52.8, 64.7, 74.9, 114.4, 114.4, 120.4, 121.6, 121.6, 124.0, 124.4, 126.8, 128.9, 128.9, 129.0, 129.0, 129.4, 130.2, 130.2, 132.8, 134.3, 146.5, 156.7, 157.5, 158.6 ppm; MS (EI): *m/z* = 541 $[M]^+$, 543 $[M + 2]^+$; HRMS (EI): *m/z* calcd $[M]^+$ 541.2245, found 541.2247.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-3-(2-methylphenylazo)-1-phenyl-2(1H)-pyridinone
(**34**, $C_{32}H_{35}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,250 (C–O–C), 1,650 (>C=O), 1,575 (–N=N–), 1,445 (C–N), 3,025, 2,905 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.4 (s, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.5 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.0 (d, 1H, C_5-H), 7.0–7.7 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 15.2, 49.2, 49.2, 49.2, 52.8, 52.9, 64.7, 74.9, 114.4, 114.4, 120.4, 121.6, 121.6, 124.1, 124.4, 125.8, 128.1, 128.7, 128.8, 129.0, 129.1, 129.1, 129.4, 129.4, 132.8, 137.1, 146.5, 156.7, 157.5, 158.4 ppm; MS (EI): *m/z* = 521; HRMS (EI): *m/z* calcd $[M]^+$ 521.2791, found 521.2786.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-3-(3-methylphenylazo)-1-phenyl-2(1H)-pyridinone
(**35**, $C_{32}H_{35}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,245 (C–O–C), 1,655 (>C=O), 1,570 (–N=N–), 1,430 (C–N), 3,020, 2,910 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.6 (t, 3H, CH_3), 2.3 (s, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.5 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 5.9 (d, 1H, C_5-H), 7.0–7.7 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 41.7, 24.3, 49.2, 49.2, 49.2, 52.8, 52.8, 64.7, 74.9, 114.4, 114.5, 120.4, 121.6, 121.7, 124.0, 124.4, 125.8, 128.6, 128.8, 129.0, 129.0, 129.1, 129.4, 130.4, 132.8, 138.4, 146.5, 156.7, 157.5, 158.5 ppm; MS (EI): *m/z* = 521; HRMS (EI): *m/z* calcd $[M]^+$ 521.2791, found 521.2793.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-3-(4-methylphenylazo)-1-phenyl-2(1H)-pyridinone
(**36**, $C_{32}H_{35}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,260 (C–O–C), 1,650 (>C=O), 1,565 (–N=N–), 1,435 (C–N), 3,015, 2,905 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.6 (t, 3H, CH_3), 2.4 (s, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.5 (m, 8H,

Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.0 (d, 1H, C_5-H), 7.0–7.6 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 24.3, 49.2, 49.2, 49.2, 52.8, 52.8, 64.5, 74.9, 114.4, 114.4, 120.4, 121.6, 121.6, 124.0, 124.4, 125.7, 128.7, 128.7, 129.0, 129.0, 129.1, 129.1, 129.4, 129.4, 132.8, 138.2, 146.5, 156.7, 157.5, 158.4 ppm; MS (EI): *m/z* = 521; HRMS (EI): *m/z* calcd $[M]^+$ 521.2791, found 521.2790.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-3-(2-nitrophenylazo)-1-phenyl-2(1H)-pyridinone
(**37**, $C_{31}H_{32}N_6O_4$)
IR (KBr): $\bar{\nu}$ = 1,265 (C–O–C), 1,660 (>C=O), 1,575 (–N=N–), 1,430 (C–N), 3,025, 2,905 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.5 (m, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.3 (m, 8H, Pip–H), 4.0–4.2 (q, 2H, OCH_2), 6.0 (d, 1H, C_5-H), 7.0–7.6 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 49.2, 49.2, 49.2, 52.8, 52.9, 64.7, 74.9, 114.4, 114.4, 120.5, 121.1, 121.4, 121.6, 124.0, 124.0, 124.4, 129.0, 129.0, 129.4, 129.4, 129.7, 129.7, 132.8, 134.9, 146.5, 148.1, 156.7, 157.5, 158.4 ppm; MS (EI): *m/z* = 552; HRMS (EI): *m/z* calcd $[M]^+$ 552.2485, found 552.2481.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-3-(3-nitrophenylazo)-1-phenyl-2(1H)-pyridinone
(**38**, $C_{31}H_{32}N_6O_4$)
IR (KBr): $\bar{\nu}$ = 1,255 (C–O–C), 1,650 (>C=O), 1,570 (–N=N–), 1,430 (C–N), 3,025, 2,915 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.4 (m, 6H, CH_3), 2.6 (q, 2H, NCH_2), 2.7–3.3 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 5.9 (d, 1H, C_5-H), 7.0–7.6 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 49.2, 49.2, 49.3, 52.8, 52.8, 64.7, 74.9, 114.2, 114.4, 120.4, 121.1, 121.6, 121.6, 124.0, 124.0, 124.4, 129.0, 129.1, 129.4, 129.4, 129.6, 129.7, 132.8, 134.9, 146.5, 148.0, 156.7, 157.5, 158.3 ppm; MS (EI): *m/z* = 552; HRMS (EI): *m/z* calcd $[M]^+$ 552.2485, found 552.2487.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-3-(4-nitrophenylazo)-1-phenyl-2(1H)-pyridinone
(**39**, $C_{31}H_{32}N_6O_4$)
IR (KBr): $\bar{\nu}$ = 1,255 (C–O–C), 1,655 (>C=O), 1,570 (–N=N–), 1,430 (C–N), 3,020, 2,920 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.4 (m, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.3 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.0 (d, 1H, C_5-H), 7.0–7.6 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 49.2, 49.2, 49.2, 52.8, 52.8, 64.7, 74.9, 114.4, 114.4, 120.4, 121.1, 121.2, 121.6, 121.6, 124.0, 124.4, 129.0, 129.0, 129.4, 129.4, 129.7, 129.7, 132.8, 134.8, 146.5, 148.1, 156.7, 157.5, 158.4 ppm; MS (EI): *m/z* = 552; HRMS (EI): *m/z* calcd $[M]^+$ 552.2485, found 552.2481.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-3-(2-methoxyphenylazo)-1-phenyl-2(1H)-pyridinone
(**40**, $C_{32}H_{35}N_5O_3$)
IR (KBr): $\bar{\nu}$ = 1,250, 1,245 (C–O–C), 1,660 (>C=O), 1,575 (–N=N–), 1,435 (C–N), 3,020, 2,915 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.5 (t, 3H, CH_3), 2.4 (s, 6H, CH_3), 2.6–3.5 (m, 8H, Pip–H), 3.6 (s, 3H, OCH_3), 4.2 (q, 2H, NCH_2), 6.0 (s, 1H, C_5-H), 7.0–7.8 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 49.2, 49.2, 49.3, 52.8, 52.8, 55.9, 64.7, 74.7, 99.6, 114.3, 114.4, 114.4, 120.4, 121.1, 121.6, 121.6, 124.0, 124.4, 129.0, 129.1, 129.4, 129.8, 129.9, 132.8, 146.5, 156.7, 157.5, 158.4, 159.1 ppm; MS (EI): *m/z* = 537; HRMS (EI): *m/z* calcd $[M]^+$ 537.2740, found 537.2738.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-1-phenyl-5-(phenylazo)-2(1H)-pyridinone
(**41**, $C_{31}H_{33}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,250 (C–O–C), 1,660 (>C=O), 1,570 (–N=N–), 1,430 (C–N), 3,025, 2,910 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.4 (m, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.3 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C3–H), 7.0–7.6 (m, 14H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 14.8, 49.2, 49.2, 49.3, 52.8, 52.8, 64.7, 75.0, 114.4, 114.4, 118.9, 120.4, 121.6, 121.6, 124.4, 128.7, 128.8, 128.8, 128.9, 129.0, 129.1, 129.4, 132.8, 146.5, 157.0, 157.5, 160.7 ppm; MS (EI): *m/z* = 507; HRMS (EI): *m/z* calcd $[M]^+$ 507.2634, found 507.2637.

5-(2-Chlorophenylazo)-4-(4-ethoxyphenyl)-6-(4-ethylpiperazinyl)-1-phenyl-2(1H)-pyridinone
(**42**, $C_{31}H_{32}ClN_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,255 (C–O–C), 1,670 (>C=O), 1,575 (–N=N–), 1,425 (C–N), 3,020, 2,910 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.4 (m, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.3 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C3–H), 7.0–7.6 (m, 14H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.9, 49.2, 49.2, 49.2, 52.8, 52.6, 64.7, 75.0, 114.4, 114.4, 118.9, 120.4, 121.6, 124.4, 126.9, 128.9, 129.0, 129.0, 129.1, 129.4, 129.4, 130.2, 130.2, 132.8, 134.3, 146.5, 157.1, 157.5, 160.7 ppm; MS (EI): *m/z* = 541 $[M]^+$, 543 $[M + 2]^+$; HRMS (EI): *m/z* calcd $[M]^+$ 541.2245, found 541.2241.

5-(3-Chlorophenylazo)-4-(4-ethoxyphenyl)-6-(4-ethylpiperazinyl)-1-phenyl-2(1H)-pyridinone
(**43**, $C_{31}H_{32}ClN_5O_2$)
IR (KBr): $\bar{\nu}$ = 1,250 (C–O–C), 1,660 (>C=O), 1,575 (–N=N–), 1,430 (C–N), 3,010, 2,910 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.4 (m, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.3 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C3–H), 7.0–7.6 (m, 14H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 49.1, 49.2,

49.2, 52.8, 52.8, 64.7, 75.0, 114.4, 114.3, 118.9, 120.4, 121.6, 121.6, 124.4, 126.9, 128.9, 129.0, 129.0, 129.1, 129.4, 129.4, 130.1, 130.2, 132.8, 134.3, 146.5, 157.0, 157.5, 160.7 ppm; MS (EI): $m/z = 541$ $[M]^+$, 543 $[M + 2]^+$; HRMS (EI): m/z calcd $[M]^+$ 541.2245, found 541.2246.

5-(4-Chlorophenylazo)-4-(4-ethoxyphenyl)-6-(4-ethylpiperazinyl)-1-phenyl-2(1H)-pyridinone
(**44**, $C_{31}H_{32}ClN_5O_2$)
IR (KBr): $\bar{\nu} = 1{,}255$ (C–O–C), 1,665 (>C=O), 1,570 (–N=N–), 1,435 (C–N), 3,030, 2,905 (C–H str) cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): $\delta = 1.3$–1.4 (m, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.3 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C3–H), 7.0–7.6 (m, 14H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): $\delta = 13.4$, 14.8, 49.2, 49.2, 49.2, 52.6, 52.7, 64.7, 75.0, 114.4, 114.4, 118.9, 120.4, 121.6, 121.6, 124.5, 126.8, 128.9, 128.9, 129.0, 129.1, 129.4, 129.4, 130.2, 130.2, 132.7, 134.3, 146.5, 157.0, 157.5, 160.7 ppm; MS (EI): $m/z = 541$ $[M]^+$, 543 $[M + 2]^+$; HRMS (EI): m/z calcd $[M]^+$ 541.2245, found 541.2245.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-5-(2-methylphenylazo)-1-phenyl-2(1H)-pyridinone
(**45**, $C_{32}H_{35}N_5O_2$)
IR (KBr): $\bar{\nu} = 1{,}240$ (C–O–C), 1,640 (>C=O), 1,570 (–N=N–), 1,420 (C–N), 3,010, 2,915 (C–H str) cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): $\delta = 1.4$–1.6 (t, 3H, CH_3), 2.3 (s, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.5 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C3–H), 7.0–7.7 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): $\delta = 13.4$, 14.8, 15.2, 49.1, 49.2, 49.2, 52.8, 52.8, 64.7, 75.0, 114.4, 114.4, 118.9, 120.4, 121.6, 121.6, 124.4, 125.8, 128.1, 128.7, 128.7, 129.0, 129.0, 129.1, 129.4, 129.4, 132.8, 137.1, 146.5, 157.0, 157.5, 160.7 ppm; MS (EI): $m/z = 521$; HRMS (EI): m/z calcd $[M]^+$ 521.2791, found 521.2787.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-5-(3-methylphenylazo)-1-phenyl-2(1H)-pyridinone
(**46**, $C_{32}H_{35}N_5O_2$)
IR (KBr): $\bar{\nu} = 1{,}240$ (C–O–C), 1,645 (>C=O), 1,575 (–N=N–), 1,425 (C–N), 3,010, 2,905 (C–H str) cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): $\delta = 1.4$–1.5 (t, 3H, CH_3), 2.4 (s, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.6 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C3–H), 7.0–7.7 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): $\delta = 13.4$, 14.8, 24.3, 49.2, 49.2, 49.3, 52.8, 52.9, 64.7, 75.0, 114.4, 114.5, 118.9, 120.4, 121.4, 121.6, 124.4, 125.8, 128.6, 128.7, 129.0, 129.0, 129.1, 129.4, 129.4, 130.4, 132.8, 138.4, 146.5, 157.1, 157.5, 160.7 ppm; MS (EI): $m/z = 521$; HRMS (EI): m/z calcd $[M]^+$ 521.2791, found 521.2793.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-5-(4-methylphenylazo)-1-phenyl-2(1H)-pyridinone
(**47**, $C_{32}H_{35}N_5O_2$)
IR (KBr): $\bar{\nu} = 1{,}265$ (C–O–C), 1,650 (>C=O), 1,560 (–N=N–), 1,420 (C–N), 3,015, 2,900 (C–H str) cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): $\delta = 1.4$–1.6 (t, 3H, CH_3), 2.3 (s, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.4 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.5 (d, 1H, C3–H), 6.9–7.6 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): $\delta = 13.4$, 14.8, 24.3, 49.1, 49.2, 49.2, 52.8, 52.8, 64.7, 75.0, 114.4, 114.2, 118.9, 120.4, 121.3, 121.6, 124.4, 125.7, 128.7, 128.7, 129.0, 129.0, 129.1, 129.1, 129.4, 129.4, 132.8, 138.4, 146.5, 157.0, 157.5, 160.7 ppm; MS (EI): $m/z = 521$; HRMS (EI): m/z calcd $[M]^+$ 521.2791, found 521.2793.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-5-(2-nitrophenylazo)-1-phenyl-2(1H)-pyridinone
(**48**, $C_{31}H_{32}N_6O_4$)
IR (KBr): $\bar{\nu} = 1{,}260$ (C–O–C), 1,665 (>C=O), 1,570 (–N=N–), 1,435 (C–N), 3,030, 2,920 (C–H str) cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): $\delta = 1.3$–1.4 (m, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.3 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C3–H), 7.0–7.6 (m, 14H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): $\delta = 13.4$, 14.8, 49.2, 49.2, 49.2, 52.8, 52.8, 64.7, 75.1, 114.4, 114.4, 118.9, 120.4, 121.1, 121.6, 121.6, 124.0, 124.4, 129.0, 129.0, 129.4, 129.4, 129.7, 129.7, 132.8, 134.9, 146.5, 148.0, 157.0, 157.5, 160.0 ppm; MS (EI): $m/z = 552$; HRMS (EI): m/z calcd $[M]^+$ 552.2485, found 552.2481.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-5-(3-nitrophenylazo)-1-phenyl-2(1H)-pyridinone
(**49**, $C_{31}H_{32}N_6O_4$)
IR (KBr): $\bar{\nu} = 1{,}250$ (C–O–C), 1,645 (>C=O), 1,575 (–N=N–), 1,420 (C–N), 3,025, 2,910 (C–H str) cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): $\delta = 1.3$–1.4 (m, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.3 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C3–H), 7.0–7.6 (m, 14H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): $\delta = 13.4$, 14.8, 49.2, 49.2, 49.2, 52.8, 52.8, 64.7, 75.1, 114.3, 114.4, 118.9, 120.4, 121.1, 121.6, 121.6, 123.4, 124.4, 129.0, 128.7, 129.4, 129.4, 129.6, 129.7, 132.6, 134.9, 146.5, 148.0, 157.0, 157.5, 160.7 ppm; MS (EI): $m/z = 552$; HRMS (EI): m/z calcd $[M]^+$ 552.2485, found 552.2479.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-5-(4-nitrophenylazo)-1-phenyl-2(1H)-pyridinone
(**50**, $C_{31}H_{32}N_6O_4$)
IR (KBr): $\bar{\nu} = 1{,}240$ (C–O–C), 1,650 (>C=O), 1,575 (–N=N–), 1,425 (C–N), 3,030, 2,920 (C–H str) cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): $\delta = 1.3$–1.4 (m, 6H, CH_3), 2.5 (q, 2H, NCH_2), 2.7–3.3 (m, 8H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 6.6 (d, 1H, C3–H), 7.0–7.6 (m, 14H, Ar–H) ppm;

^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 49.2, 49.2, 49.2, 52.8, 52.8, 64.7, 74.8, 114.4, 114.4, 118.9, 120.4, 121.1, 121.1, 121.6, 121.6, 124.4, 129.0, 129.0, 129.4, 129.4, 129.7, 129.7, 132.8, 134.8, 146.3, 148.0, 156.7, 157.5, 160.5 ppm; MS (EI): *m/z* = 552; HRMS (EI): *m/z* calcd $[M]^+$ 552.2485, found 552.2483.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-5-(2-methoxyphenylazo)-1-phenyl-2(1H)-pyridinone
(**51**, $C_{32}H_{35}N_5O_3$)
IR (KBr): $\bar{\nu}$ = 1,260 (C–O–C), 1,650 (>C=O), 1,570 (–N=N–), 1,435 (C–N), 3,010, 2,920 (C–H str) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.3–1.5 (t, 3H, CH_3), 2.4 (s, 6H, CH_3), 2.6–3.5 (m, 8H, Pip–H), 3.6 (s, 3H, OCH_3), 4.2 (q, 2H, NCH_2), 6.6 (s, 1H, C3–H), 7.0–7.8 (m, 13H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 49.2, 49.2, 49.3, 52.6, 52.8, 55.9, 64.7, 75.5, 99.6, 114.3, 114.4, 114.4, 118.9, 120.4, 121.1, 121.6, 121.6, 124.4, 129.0, 129.1, 129.4, 129.4, 129.6, 129.8, 132.8, 146.5, 157.0, 157.5, 159.1, 160.7 ppm; MS (EI): *m/z* = 537; HRMS (EI): *m/z* calcd $[M]^+$ 537.2740, found 537.27437.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-3-[(4-methyl-piperazinyl)methyl]-1-phenyl-2(1H)-pyridinone
(**52**, $C_{31}H_{41}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 3,035, 2,925 (C–H), 1,660 (>C=O), 1,245 (C–O–C), 1,430 (C = N) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.3 (q, 6H, CH_3), 2.4 (s, 2H, NCH_2), 2.6 (s, 4H, CH_2), 2.7–3.4 (m, 16H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 5.9 (d, 1H, C_5-H), 6.9–7.6 (m, 9H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 14.8, 43.1, 49.2, 49.2, 49.3, 52.4, 52.4, 52.8, 52.8, 55.3, 55.3, 57.9, 64.7, 74.9, 114.4, 114.4, 120.4, 121.6, 121.6, 124.4, 129.0, 129.0, 129.4, 129.4, 132.8, 134.8, 135.5, 156.7, 157.5, 158.3 ppm; MS (EI): *m/z* = 515; HRMS (EI): *m/z* calcd $[M]^+$ 515.3260, found 515.3263.

4-(4-Ethoxyphenyl)-6-(4-ethylpiperazinyl)-3-[(4-ethyl-piperazinyl)methyl]-1-phenyl-2(1H)-pyridinone
(**53**, $C_{32}H_{43}N_5O_2$)
IR (KBr): $\bar{\nu}$ = 3,035, 2,925 (C–H), 1,660 (>C=O), 1,245 (C–O–C), 1,430 (C=N) cm^{-1}; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.4–1.5 (t, 3H, CH_3), 2.3 (q, 6H, CH_3), 2.4 (s, 4H, NCH_2), 2.5 (s, 2H, CH_2), 2.7–3.4 (m, 16H, Pip–H), 4.0–4.1 (q, 2H, OCH_2), 5.9 (d, 1H, C_5-H), 7.0–7.6 (m, 9H, Ar–H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ = 13.4, 13.4, 14.8, 49.2, 49.2, 49.2, 49.2, 52.7, 52.7, 52.8, 52.8, 52.8, 52.8, 57.9, 64.7, 74.9, 114.4, 114.5, 120.4, 121.6, 121.7, 124.4, 129.0, 129.0, 129.4, 129.4, 132.8, 134.8, 135.3, 156.7, 157.5, 158.3 ppm; MS (EI): *m/z* = 529; HRMS (EI): *m/z* calcd $[M]^+$ 529.3417, found 529.3411.

Acknowledgments The authors are grateful to the Head of the Chemistry Department, Veer Narmad South Gujarat University, Surat, Gujarat, for providing laboratory facilities, and to the SAIF, Punjab University, Chandigarh, for NMR spectra.

References

1. Dorigo P, Gaion RM, Belluco P, Fraccarollo D, Maragno I, Bobieri G, Benetollo F, Mosti L, Orsini F (1993) J Med Chem 36:2475
2. Farah AE, Alousi AA (1978) Life Sci 22:1139
3. Alousi AA, Canter JM, Monterano MJ, Fort DJ, Ferrari RA (1983) J Cardiovasc Pharmacol 5:792
4. Paulvannan K, Chen T (2000) J Org Chem 65:6160
5. Rodgers JD, Wang H, Patel M, Arvanitis A, Cocuzza AJ (2005) US Patent 6,969,721 B2. Bristol-Myers Squibb Pharma. Princeton
6. Flynn DL, Petillo PA (2007) US Patent 7,202,257 B2. Deciphera Pharma, LLC. Lawrence
7. Pemberton N (2007) Synthesis and functionalization of ring fused 2-pyridones - targeting pilli formation in *E. Coli*. Akademik av-handling. Department of Chemistry, Umea University, Sweden
8. Prakash HS, Senthilkumar SP (2004) Curr Org Chem 8:1521
9. Misic-Vukovic M, Radojkovic-Velickovic M (1998) J Serb Chem Soc 63:585
10. Uscumlic G, Mijin D, Valentic N, Vajs V, Susic B (2004) Chem Phys Lett 397:148
11. Forlani L, Gristoni G, Boga C, Todesco PE, Del Vechio E, Salva S, Monari M (2002) Arkivoc 11:198
12. Kirk-Othmer (1968) Encyclopedia of chemical technology, 2nd rev edn. Wiley Interscience Publication, Wiley
13. Armarego WLF (1977) In: Taylor EC, Weisberger A (eds) Stereochemistry of heterocyclic compounds. Wiley, New York
14. Martin WB, Martell AE (1948) J Am Chem Soc 70:1817
15. Patel NB, Patel AK (2004) Indian J Chem 45B:1774
16. Kubo K, Murakami M, Itoh N, Arima H, Sozu I, Homma H (1979) Kokai 77, 77,066. Yamanouchi Pharmaceutical Co. Ltd., Japan
17. Kubo K, Murakami M, Itoh N, Arima H, Sozu I, Homma H (1979) Chem Abstr 87:201328h
18. Kubo K, Murakami M, Itoh N, Arima H, Sozu I, Homma H (1979) Yakuguku Zasshi 99:558
19. Silverstein RM, Webster FX (1998) Spectrometric identification of organic compounds, vol 6. Wiley, New York
20. Dyer JR, Cazes J, Marcel D, Khadatare AM, Jitkar SC, Malcolm R, Thomas N, Febiger L (1987) Application of absorption spectroscopy of organic compounds, vol 6. Lea & Febiger, Philadelphia
21. Matoba K, Fukushima A, Arai H, Yamazaki T (1979) Chem Bull 27:242
22. Edward DI (1980) Antimicrobial drug action, vol 1. Macmillan Press, London and Basingstoke, p 8
23. Barry AL (1976) The antimicrobial susceptibility test, principle and practices. Lea & Febiger, Philadelphia