Tetrahedron 64 (2008) 5933–5943

Contents lists available at ScienceDirect

Tetrahedron

journal homepage: www.elsevier.com/locate/tet

# A synthesis of 5-hetaryl-3-(2-hydroxybenzoyl)pyrroles

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ARTICLE INFO

*Article history:*
Received 9 January 2008
Received in revised form 20 March 2008
Accepted 10 April 2008
Available online 12 April 2008

*Keywords:*
3-Formylchromone
Pyrroles
Hetarylmethylamines
Recyclization
Chlorotrimethylsilane

ABSTRACT

A facile and versatile procedure for the synthesis of 5-hetaryl-3-(2-hydroxybenzoyl)-1*H*-pyrroles and 4-(2-hydroxybenzoyl)-1*H*-pyrrole-2-carboxylic acid derivatives was established on the basis of TMSCl promoted recyclization of 3-formylchromone with various hetarylmethylamines and glycine derivatives.


## 1. Introduction

The pyrrole structural unit is widely found in natural biomolecules (porphins, aminoacids, etc.) and a variety of pharmacologically active compounds.[1] Pyrroles have been employed as antioxidants,[2a] antibacterial,[2b,c] ionotropic,[2d,e] antitumor,[2f] anti-inflammatory,[2g,h] and antifungal agents,[2i] P38 kinase,[3a] prolyl-4-hydroxylase,[3b] poly(ADP-ribose) polymerase inhibitors,[3c] estrogen receptor β selective ligands,[3d] $AT_1$-selective angiotensin II receptor antagonists,[3e] and minor groove recognition elements.[3f,g] At present, a variety of synthetic approaches to substituted pyrroles exist. However, a search of the literature for effective methods for the synthesis of pyrrole libraries to be employed in high-throughput screening remains a challenge of medicinal chemistry.

The Paal–Knorr reactions of 1,4-diketones and primary amines have been used extensively for the synthesis of various pyrrole derivatives.[1] Another, method is based on the reaction of 1,3-*CCC*-bielectrophiles with 1,2-*CN*-binucleophiles.[4–6] Acid and base catalyzed recyclization reactions of 3-formylchromone[7] with primary and secondary amines have been used for the synthesis of 3(2-hydroxybenzoyl)pyrroles containing chromyl and carboxyl groups in position 5 of the pyrrole ring although in rather low yield (10–30%).[4] Apparently, these synthetic drawbacks hamper combinatorial synthesis of drug like 3(2-hydroxybenzoyl)pyrroles.

The primary objective of the present study was to develop a facile and versatile method for the synthesis of previously unknown 5-hetaryl-2-hydroxybenzoylpyrroles through the recyclization reaction of 3-formylchromone **1** with various hetarylmethylamines.

## 2. Results and discussion

We have demonstrated that chlorotrimethylsilane (TMSCl) is a convenient reagent for the condensation reactions of carbonyl compounds[9] and recyclizations of 3-formylchromones.[8] Thus, TMSCl was considered as a potential promoter and water scavenger in the reaction of 3-formylchromone with hetarylmethylamines and glycine derivatives.

### 2.1. Reaction with primary hetarylmethylamines

3-Formylchromone **1** reacted with hetarylmethylamines **2** (DMF, 100 °C, molar ratio 1:2) in the presence of 4 molar equiv of TMSCl to give 5-hetaryl-1*H*-pyrrol-3-yl](2-hydroxyphenyl)methanones **3** in 68–91% yields (Scheme 1). The reaction between **1** and **2** strongly depends on their molar ratio, such that at a 2:1 ratio, fused chromonepyrroles **4** were formed exclusively in moderate yields. The reaction of **1** with representative amines **2c**,**d**, **2f**–**h**, and **2l** at a 1:1 initial molar ratio gave mixtures of the corresponding pyrroles **3** and chromonopyrroles **4**.

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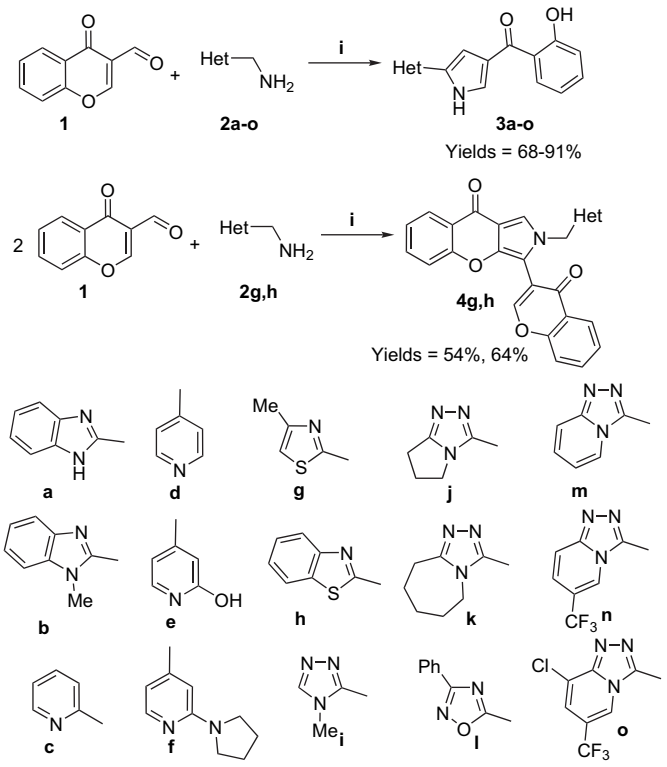
doi:10.1016/j.tet.2008.04.041



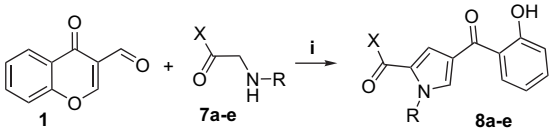

**Scheme 1.** Reagents and conditions: (i) 4 equiv $Me_3SiCl$, DMF, 100 °C, 6–15 h.

## 2.2. Reaction with secondary hetarylmethylamines

The reactions of formylchromone **1** with secondary hetarylamines **5** (DMF, TMSCl, 100 °C) led to [5-hetaryl-1-alkyl-1*H*-pyrrol-3-yl](2-hydroxyphenyl)methanones **6** (Scheme 2) independent of the molar ratio of reagents since the formation of compounds **4** is not possible in this case.

## 2.3. Reaction with glycine derivatives

The TMSCl mediated [3+3] cyclocondensation of **1** with glycine derivatives **7** afforded pyrrole derivatives **8** in moderate yields (Scheme 3). It should be noted that the use of TMSCl in the reaction of glycine **7a** avoided the previously described decarboxylation.[4a–d] Compounds **8a**,**c**,**d** were purified by precipitation of the crude product with water followed by simple recrystallization, whereas preparative HPLC was used for purification of pyrroles **8b** and **8e**.




**a**: X = OH, R = H (55%) **b**: X = OMe, R = Me (43%) **c**: X = $NMe_2$, R = H (68%)
**d**: X = $N(CH_2)_4$, R = H (61%) **e**: X = $NEt_2$, R = Me (45%)

**Scheme 3.** Reagents and conditions: (i) 4 equiv $Me_3SiCl$, DMF, 100 °C, 12–15 h.

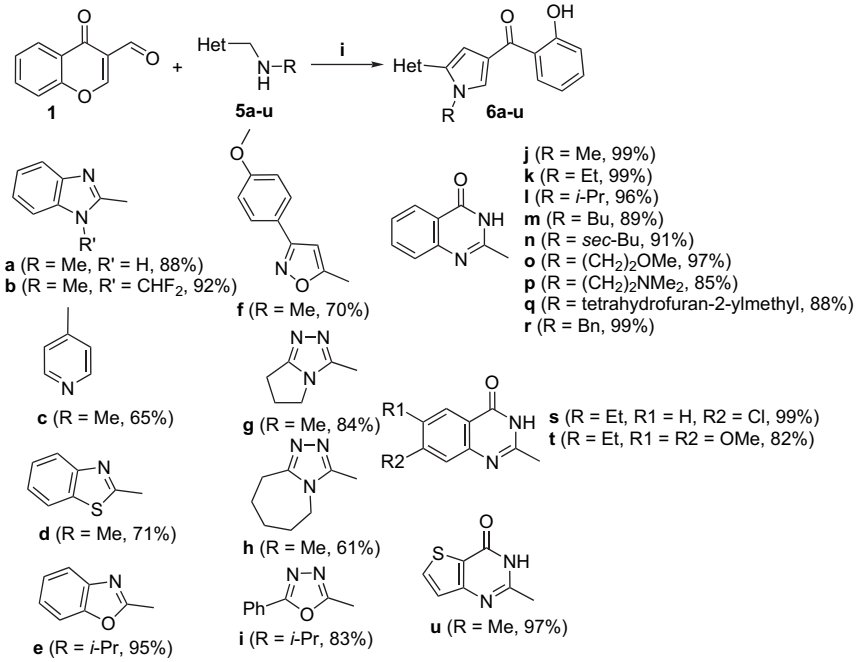
The reaction of formylchromone **1** with $N^1$,$N^2$-dimethylglycinamide or L-prolinamide proceeds as a [4+1] recyclization[10] and results in **9a** (79%) and **9b** (84%), respectively (Scheme 4).

The TMSCl mediated pyrrole synthesis is applicable to the fusion of the pyrrole and the dihydroquinoxaline rings. The reaction of 3,4-dihydroquinoxalin-2(1*H*)-one **10a** or its sulfamide derivative **10b** gave the previously unknown 2-(2-hydroxybenzoyl)pyrrolo[1,2-*a*]quinoxalin-4(5*H*)-ones **11a**,**b** in 52 and 58% yields, respectively (Scheme 4).

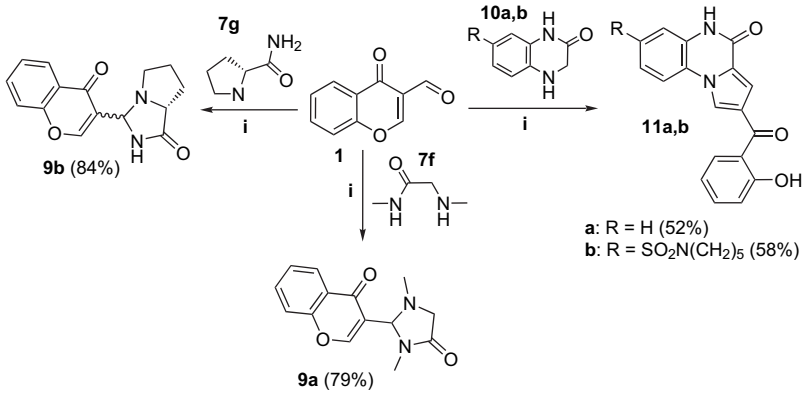
## 2.4. Structure determination

The composition and structure of all the compounds obtained were determined by LCMS, elemental analysis, $^1H$, $^{13}C$ NMR and IR spectroscopy. The signals in $^1H$ and $^{13}C$ NMR spectra were assigned on the basis of $^1H$–$^{13}C$ HMQC and HMBC 2D NMR spectra of representative compounds.

The $^1H$ NMR spectra of compounds **3** contain a characteristic set of signals for the protons of *ortho*-acylated phenol ring and two




**Scheme 2.** Reagents and conditions: (i) 4 equiv $Me_3SiCl$, DMF, 100 °C, 6–15 h.




**Scheme 4.** Reagents and conditions: (i) 4 equiv $Me_3SiCl$, DMF, 100 °C, 15 h.

doublets ($^4J_{HH}$=0.9–2.0 Hz) or broadened singlets for the protons of the pyrrole fragment. The signals of the OH group appear in the range of 10.9–11.3 ppm (DMSO-$d_6$) and are magnetically deshielded due to the formation of the stable intramolecular hydrogen bond to the adjacent carbonyl group. The NH protons of the pyrrole ring emerge at 12.3–12.9 ppm (DMSO-$d_6$). The $^1$H NMR spectra of *N*-substituted pyrroles **6**, **8**, and **11** are similar to those of compounds **3** except they do not contain the signals of the NH protons.

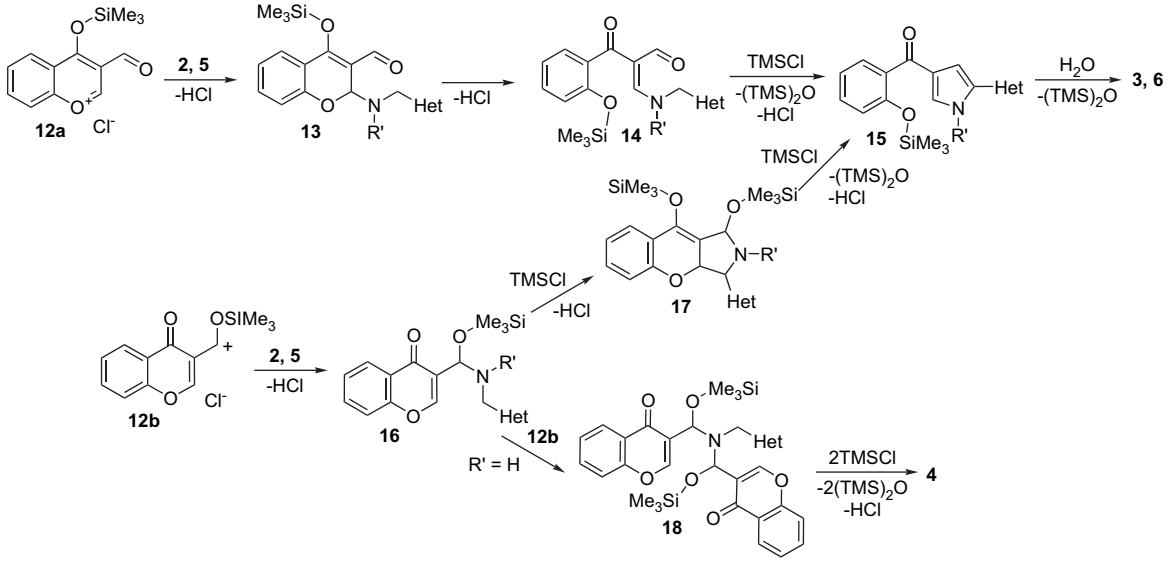
The $^1$H NMR spectra of compounds **4** contain a double set of signals for the *ortho*-substituted aryl rings of the non-equivalent chromone residues, a singlet for the methylene protons (3.85, 3.98 ppm) and two singlets of the pyrrole and pyranone fragments (at 8.47, 8.62 and 8.76, 8.89 ppm, respectively) whereas no signal was detected for the phenolic OH group. The structure of compound **4** was unambiguously proved by the characteristic $^1$H–$^{13}$C HMQC and HMBC correlations. Furthermore NOESY correlations between the methylene protons and the proton of the pyrrole ring are also in keeping with the proposed structure of compounds **4**.

### 2.5. A possible mechanism

A possible mechanism for the TMSCl mediated formation of pyrroles **3**, **4**, and **6** is outlined in Scheme 5. TMSCl can activate 3-formylchromone **1** through addition to the C=O bonds. The addition to the ketone bond may result in *O*-trimethylsilyl-3-formylchromone chloride **12a**, which has previously been characterized by Iwasaki et al.[11] Alkylation of the amine by **12a** is likely to result in compound **13**, which can undergo *retro*-Michael reaction to give enaminal **14**. The TMSCl mediated intramolecular Knoevenagel condensation[9d] of **14** to give **15** followed by hydrolysis would likely give pyrroles **3** and **6**. Another possible reaction pathway involves the activation of aldehyde group of **1** with TMSCl to give intermediate **12b**. Reaction of amines with **12b** could give rise to intermediate **16**, which can be transformed into compound **17** through the activation of the keto group with TMSCl followed by *retro*-Michael reaction and elimination of TMSOH. The formation of compounds **4** may proceed through the alkylation of the primary amine with 2 mol of **12b** leading to intermediate **18**. Elimination of TMSOH and intramolecular ring closure of the pyrrole ring can lead to **4**.

## 3. Conclusions

In conclusion, we have elaborated an efficient methodology for the preparation of 5-hetaryl-3-(2-hydroxybenzoyl)pyrroles and 4-(2-hydroxybenzoyl)-1*H*-pyrrole-2-carboxylic acid derivatives from 3-formylchromone, hetarylmethylamines, and glycine derivatives using $Me_3SiCl$ as a promoter and water scavenger. This allows simple insertion of hetaryl residues at position 2(5) of the pyrrole ring, which previously has been achieved through complicated multistep procedures. The methodology is applicable to a wide variety of hetarylmethylamines and affords target compounds in preparative yields using simple purification procedure.




**Scheme 5.**

This synthetic methodology can also be used for the generation of highly diverse combinatorial sets of novel pyrrole derivatives, which exhibit a range of biological activities.

## 4. Experimental

### 4.1. General

All chemicals were obtained from commercially available sources and used without further purification (Aldrich, Fluka, 'Enamine Ltd'). DMF for the reactions was freshly distilled and dried by standard methods, monitoring of water content in solvents (all solvents have <0.05%, usually 0.02% of water) was performed using Mettler Toledo DL31 KF Titrator. All solvents for the crystallizations were used without additional purification.

Melting points were measured with a Buchi melting points apparatus and are uncorrected. The $^1$H NMR spectra (400 and 500 MHz) were recorded on a Varian Mercury-400 spectrometer and Bruker Avance DRX 500 spectrometer with TMS as an internal standard. $^{13}$C NMR spectra (125 MHz) and 2D NMR experiments were recorded on a Bruker Avance DRX 500 spectrometer with TMS as an internal standard. $^{19}$F NMR spectra (470 MHz) were recorded on a Bruker Avance DRX 500 spectrometer with $CFCl_3$ as an internal standard. NMR experiments were recorded on a Bruker Avance DRX 500 spectrometer. LC/MS spectra were recorded using chromatography/mass spectrometric system that consists of high-performance liquid chromatograph 'Agilent 1100 Series' equipped with diode-matrix and mass-selective detector 'Aligent LC/MSD SL'. According to HPLC–MS and $^1$H NMR data all the synthesized compounds have purity >95%. IR spectra were recorded on a Nexus-470 spectrometer for samples in KBr discs. Microanalyses were performed in the Microanalytical Laboratory of the Institute of Organic Chemistry, National Academy of Sciences of Ukraine.

Commercially unavailable hetarylmethylamines **2g**,[12a] **2i** and **5i**,[12b] **2m**–**o**,[12c] **5a**,**b**,**e**,[12d] **5f**,[12e] **5j**–**t**[12f] were prepared according to literature.

### 4.2. General procedure for the synthesis of pyrroles 3, 6, 8, and 11

Hetarylmethylamines **2a**–**o**, **5a**–**u** or glycine derivatives **7a**–**e** and **10a**,**b** (**2a**–**o**, **7a**,**c**,**d**—4 mmol, **5a**–**u**, **10a**,**b**—2 mmol, **7b**,**e**—2.6 mmol) and 3-formylchromone **1** (348 mg, 2 mmol) were placed in 15 mL pressure tube and dissolved in DMF (2–4 mL). Chlorotrimethylsilane (869 mg, 8 mmol) was added dropwise to the solution. The tube was thoroughly sealed and heated on a water-bath for 6–15 h. After cooling the flask was opened (*caution! excessive pressure inside*) and the reaction mixture was poured into water (15 mL) and allowed to stand at 20 °C in an ultrasonic bath for 1 h. The precipitate formed was filtered and washed with a small amount of *i*-PrOH. Recrystallization from an appropriate solvent yielded targeted compounds **3**, **6**, **8**, and **11**. Compounds **8b** and **8e** were purified by preparative HPLC.

Compounds **4g**,**h** and **9a**,**b** were obtained by the same procedure.

#### 4.2.1. *[5-(1H-Benzimidazol-2-yl)-1H-pyrrol-3-yl](2-hydroxyphenyl)methanone (**3a**)*

Gray solid (503 mg, 83%); mp 224–225 °C (EtOH/DMF). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=6.96 (t, $^3J_{HH}$=8.2 Hz, 1H, 5-$H_{Ar}$), 7.01 (d, $^3J_{HH}$=8.2 Hz, 1H, 3-$H_{Ar}$), 7.43–7.49 (m, 3H, 4-$H_{Ar}$, 5,6-$H_{BIm}$), 7.62 (d, $^3J_{HH}$=8.2 Hz, 1H, 6-$H_{Ar}$), 7.71 (s, 1H, 2-$H_{pyrrole}$), 7.74 (m, 2H, 4,7-$H_{BIm}$), 7.90 (s, 1H, 4-$H_{pyrrole}$), 8.52 (br s, 1H, NH), 10.88 (br s, 1H, OH), 13.00 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=114.1 (4,7-$C_{BIm}$), 116.5 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 118.3 (3-$C_{pyrrole}$), 119.5 (5-$C_{Ar}$), 124.6 (1-$C_{Ar}$), 125.8 (5,6-$C_{BIm}$), 126.5 (2-$C_{pyrrole}$), 130.7 (6-$C_{Ar}$), 131.5 (5-$C_{pyrrole}$), 132.2 (3a,7a-$C_{BIm}$), 134.1 (4-$C_{Ar}$), 142.9 (2-$C_{BIm}$), 158.5 (2-$C_{Ar}$), 191.4 (C=O). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3367 (br, OH, NH), 3122, 3101, 3057, 2970, 2881, 1639 (C=O), 1589, 1464, 1377, 1342, 1244, 1219, 1149, 893, 752, 708. APSI MS: $M^+$+1=304. Anal. Calcd for $C_{18}H_{13}N_3O_2$: C, 71.28; H, 4.32; N, 13.85. Found: C, 71.45; H, 4.21; N, 13.79.

#### 4.2.2. *(2-Hydroxyphenyl)[5-(1-methyl-1H-benzimidazol-2-yl)-1H-pyrrol-3-yl]methanone (**3b**)*

Light green solid (577 mg, 91%); mp 261–262 °C (EtOH/DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=4.01 (s, 3H, $NCH_3$), 6.92–7.03 (m, 2H, 3,5-$H_{Ar}$), 7.25 (m, 3H, 5,7-$H_{BIm}$, 2-$H_{pyrrole}$), 7.45 (t, $^3J_{HH}$=7.9 Hz, 1H, 4-$H_{Ar}$), 7.53 (s, 1H, 4-$H_{pyrrole}$), 7.63 (d+t, $^3J_{HH}$=7.9 Hz, 2H, 6-$H_{Ar}$, 6-$H_{BIm}$), 7.73 (d, $^3J_{HH}$=7.9 Hz, 1H, 4-$H_{BIm}$), 11.13 (s, 1H, OH), 12.64 (br s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=31.9 ($CH_3$), 110.8 (7-$C_{BIm}$), 111.2 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 118.9 (4-$C_{BIm}$), 119.5 (5-$C_{Ar}$), 122.5 (6-$C_{BIm}$), 122.7 (5-$C_{BIm}$), 123.8 (3-$C_{pyrrole}$), 124.3 (1-$C_{Ar}$), 125.5 (2-$C_{pyrrole}$), 129.2 (5-$C_{pyrrole}$), 131.0 (6-$C_{Ar}$), 134.0 (4-$C_{Ar}$), 136.9 (7a-$C_{BIm}$), 142.7 (2-$C_{BIm}$), 146.3 (3a-$C_{BIm}$), 159.0 (2-$C_{Ar}$), 191.9 (C=O). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3650–3100 (br, OH, NH), 3057, 3034, 2956, 1614 (C=O), 1578, 1487, 1441, 1385, 1342, 1271, 1213, 1134, 949, 891, 818, 798, 760, 741, 706, 677. APSI MS: $M^+$+1=318. Anal. Calcd for $C_{19}H_{15}N_3O_2$: C, 71.91; H, 4.76; N, 13.24. Found: C, 71.75; H, 4.87; N, 13.19.

#### 4.2.3. *(2-Hydroxyphenyl)(5-pyridin-2-yl-1H-pyrrol-3-yl)methanone hydrochloride (**3c**)*

Light green solid (529 mg, 88%); mp 146–147 °C (MeCN). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=6.93–6.99 (m, 2H, 3,5-$H_{Ar}$), 7.24 (m, 1H, 5-$H_{Py}$), 7.27 (s, 1H, 2-$H_{pyrrole}$), 7.44 (m, 2H, 4-$H_{pyrrole}$, 4-$H_{Ar}$), 7.72 (d, $^3J_{HH}$=7.9 Hz, 1H, 6-$H_{Ar}$), 7.81 (t, $^3J_{HH}$=7.9 Hz, 1H, 4-$H_{Py}$), 7.88 (d, $^3J_{HH}$=7.9 Hz, 1H, 3-$H_{Py}$), 8.55 (d, $^3J_{HH}$=4.6 Hz, 1H, 6-$H_{Py}$), 11.22 (s, 1H, OH), 12.35 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=109.2 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.3 (3-$C_{Py}$), 119.4 (5-$C_{Ar}$), 122.2 (3-$C_{pyrrole}$), 123.9 (1-$C_{Ar}$), 125.4 (5-$C_{Py}$), 128.6 (2-$C_{pyrrole}$), 131.0 (6-$C_{Ar}$), 133.3 (5-$C_{pyrrole}$), 134.0 (4-$C_{Ar}$), 137.7 (4-$C_{Py}$), 149.3 (6-$C_{Py}$), 150.0 (2-$C_{Py}$), 159.3 (2-$C_{Ar}$), 192.2 (C=O). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3226 (br, OH, NH), 3138, 3051, 3007, 2983, 1618 (C=O), 1593, 1568, 1481, 1462, 1392, 1282, 1238, 1205, 1147, 893, 789, 754, 706, 673. APSI MS: $M^+$+1=265. Anal. Calcd for $C_{16}H_{13}ClN_2O_2$: C, 63.90; H, 4.36; Cl, 11.79; N, 9.31. Found: C, 63.97; H, 4.30; Cl, 11.83; N, 9.34.

#### 4.2.4. *(2-Hydroxyphenyl)(5-pyridin-4-yl-1H-pyrrol-3-yl)methanone hydrochloride (**3d**)*

Yellow solid (427 mg, 71%); mp 184–185 °C (MeCN). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=6.95 (t, $^3J_{HH}$=8.3 Hz, 1H, 5-$H_{Ar}$), 7.01 (d, $^3J_{HH}$=8.3 Hz, 1H, 3-$H_{Ar}$), 7.45 (td, $^3J_{HH}$=8.3 Hz, $^4J_{HH}$=1.5 Hz, 1H, 4-$H_{Ar}$), 7.66 (dd, $^3J_{HH}$=8.3 Hz, $^4J_{HH}$=1.5 Hz, 1H, 6-$H_{Ar}$), 7.74 (m, 1H, 2-$H_{pyrrole}$), 7.80 (m, 1H, 4-$H_{pyrrole}$), 8.35 (d, $^3J_{HH}$=6.7 Hz, 2H, 3,5-$H_{Py}$), 8.76 (d, $^3J_{HH}$=6.7 Hz, 2H, 2,6-$H_{Py}$), 11.04 (s, 1H, OH), 13.33 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=115.8 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 120.2 (3,5-$C_{Py}$), 124.3 (1-$C_{Ar}$), 126.9 (3-$C_{pyrrole}$), 128.8 (2-$C_{pyrrole}$), 131.0 (6-$C_{Ar}$), 132.3 (5-$C_{pyrrole}$), 134.1 (4-$C_{Ar}$), 142.3 (2,6-$C_{Py}$), 146.6 (4-$C_{Py}$), 158.8 (2-$C_{Ar}$), 191.8 (C=O). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3650–3280 (br, OH, NH), 3145, 3051, 3043, 3032, 2991, 2935, 2900, 1633 (C=O), 1595, 1572, 1475, 1446, 1379, 1298, 1213, 1194, 1161, 893, 812, 766, 706, 690. APSI MS: $M^+$+1=265. Anal. Calcd for $C_{16}H_{13}ClN_2O_2$: C, 63.90; H, 4.36; Cl, 11.79; N, 9.31. Found: C, 63.86; H, 4.42; Cl, 11.80; N, 9.29.

#### 4.2.5. *(2-Hydroxyphenyl)[5-(2-hydroxypyridin-4-yl)-1H-pyrrol-3-yl]methanone (**3e**)*

Pale yellow solid (459 mg, 82%); mp 292 °C (EtOH/DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=6.57 (d, $^3J_{HH}$=7.0 Hz, 1H, 5-$H_{Py}$), 6.67 (s, 1H, 3-$H_{Py}$), 6.91–7.01 (m, 2H, 3,5-$H_{Ar}$), 7.18 (s, 1H, 2-$H_{pyrrole}$), 7.34 (d, $^3J_{HH}$=7.0 Hz, 1H, 6-$H_{Py}$), 7.45 (t, $^3J_{HH}$=7.7 Hz, 1H, 4-$H_{Ar}$), 7.59 (s, 1H, 4-$H_{pyrrole}$), 7.73 (d, $^3J_{HH}$=7.7 Hz, 1H, 6-$H_{Ar}$), 11.24 (s, 1H, OH),

11.35 (br s, 1H, OH), 12.30 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=102.5 (5-$C_{Py}$), 111.1 (3-$C_{Py}$), 112.0 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 123.7 (1-$C_{Ar}$), 125.5 (3-$C_{pyrrole}$), 129.6 (2-$C_{pyrrole}$), 130.7 (5-$C_{pyrrole}$), 131.2 (6-$C_{Ar}$), 134.2 (4-$C_{Ar}$), 135.9 (4-$C_{Py}$), 142.9 (6-$C_{Py}$), 159.4 (2-$C_{Ar}$), 163.2 (2-$C_{Py}$), 192.2 (C=O). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3650–3320 (br, OH), 3269 (NH), 3134, 2974, 2848, 1632 (C=O), 1595, 1564, 1524, 1487, 1443, 1421, 1336, 1267, 1213, 1555, 1136, 895, 841, 816, 766. APSI MS: M$^+$+1=281. Anal. Calcd for $C_{16}H_{13}ClN_2O_3$: C, 60.67; H, 4.14; Cl, 11.19; N, 8.84. Found: C, 60.80; H, 4.08; Cl, 11.15; N, 8.81.

#### 4.2.6. (2-Hydroxyphenyl)[5-(2-pyrrolidin-1-ylpyridin-4-yl)-1H-pyrrol-3-yl]methanone dihydrochloride (**3f**)

Light green solid (601 mg, 74%); mp 227–228 °C (MeCN). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=2.03 (m, 4H, 2$CH_2$), 3.59 (m, 4H, 2$NCH_2$), 6.96 (t, $^3J_{HH}$=8.2 Hz, 1H, 5-$H_{Ar}$), 7.00 (d, $^3J_{HH}$=8.2 Hz, 1H, 3-$H_{Ar}$), 7.31 (d, $^3J_{HH}$=7.6 Hz, 1H, 5-$H_{Py}$), 7.45 (m, 2H, 6-$H_{Py}$, 4-$H_{Ar}$), 7.55 (d, $^4J_{HH}$=1.3 Hz, 1H, 2-$H_{pyrrole}$), 7.68 (dd, $^3J_{HH}$=7.6 Hz, $^4J_{HH}$=1.3 Hz, 1H, 6-$H_{Py}$), 7.74 (d, $^4J_{HH}$=1.3 Hz, 1H, 4-$H_{pyrrole}$), 7.86 (d, $^3J_{HH}$=8.2 Hz, 1H, 6-$H_{Ar}$), 11.10 (s, 1H, OH), 13.13 (s, 1H, NH), 13.19 (br s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=25.3, 49.0, 104.6 (3-$C_{Py}$), 107.9 (5-$C_{Py}$), 114.0 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 124.1 (1-$C_{Ar}$), 126.1 (3-$C_{pyrrole}$), 129.5 (2-$C_{pyrrole}$), 130.9 (5-$C_{pyrrole}$), 131.0 (6-$C_{Ar}$), 134.1 (4-$C_{Ar}$), 136.6 (4-$C_{Py}$), 144.2 (6-$C_{Py}$), 150.3 (2-$C_{Py}$), 159.0 (2-$C_{Ar}$), 191.9 (C=O). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3640–3150 (br, OH, NH), 2926, 1653 (C=O), 1616, 1595, 1487, 1456, 1377, 1354, 1230, 1155, 941, 895. APSI MS: M$^+$+1=334. Anal. Calcd for $C_{20}H_{21}Cl_2N_3O_2$: C, 59.12; H, 5.21; Cl, 17.45; N, 10.34. Found: C, 59.20; H, 5.16; Cl, 17.40; N, 10.37.

#### 4.2.7. (2-Hydroxyphenyl)[5-(4-methyl-1,3-thiazol-2-yl)-1H-pyrrol-3-yl]methanone (**3g**)

Light green solid (471 mg, 83%); mp 194–195 °C (EtOH). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=2.39 (s, 3H, $CH_3$), 6.92–6.99 (m, 2H, 3,5-$H_{Ar}$), 7.01 (s, 1H, 5-$H_{Tz}$), 7.21 (s, 1H, 2-$H_{pyrrole}$), 7.43 (m, 2H, 4-$H_{Ar}$, 4-$H_{pyrrole}$), 7.67 (d, $^3J_{HH}$=7.5 Hz, 1H, 6-$H_{Ar}$), 11.09 (s, 1H, OH), 12.60 (s, 1H, NH). $^1$H NMR (500 MHz, $CDCl_3$): $\delta$=2.45 (s, 3H, $CH_3$), 6.81 (s, 1H, 5-$H_{Tz}$), 6.94 (t, $^3J_{HH}$=8.1 Hz, 1H, 5-$H_{Ar}$), 7.05 (d, $^3J_{HH}$=8.1 Hz, 1H, 3-$H_{Ar}$), 7.15 (s, 1H, 2-$H_{pyrrole}$), 7.49 (t, $^3J_{HH}$=8.1 Hz, 1H, 4-$H_{Ar}$), 7.50 (s, 1H, 4-$H_{pyrrole}$), 7.94 (d, $^3J_{HH}$=8.1 Hz, 1H, 6-$H_{Ar}$), 10.36 (br s, 1H, NH), 12.07 (s, 1H, OH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=17.3 ($CH_3$), 110.0 (4-$C_{pyrrole}$), 113.5 (5-$C_{Tz}$), 117.6 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 124.1 (1-$C_{Ar}$), 125.3 (3-$C_{pyrrole}$), 128.3 (5-$C_{pyrrole}$), 128.6 (2-$C_{pyrrole}$), 130.9 (6-$C_{Ar}$), 134.0 (4-$C_{Ar}$), 152.9 (4-$C_{Tz}$), 158.7 (2-$C_{Tz}$), 158.9 (2-$C_{Ar}$), 191.8 (C=O). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3650–3310 (br, OH, NH), 3115, 3014, 2918, 1622 (C=O), 1593, 1512, 1483, 1454, 1390, 1362, 1273, 1238, 1190, 1161, 893, 814, 769, 719, 671. APSI MS: M$^+$+1=285. Anal. Calcd for $C_{15}H_{12}N_2O_2S$: C, 63.36; H, 4.25; N, 9.85; S, 11.28. Found: C, 63.44; H, 4.34; N, 9.81; S, 11.31.

#### 4.2.8. [5-(1,3-Benzothiazol-2-yl)-1H-pyrrol-3-yl](2-hydroxyphenyl)methanone (**3h**)

Brown solid (499 mg, 78%); mp 170–171 °C (EtOH/DMF). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=6.93–7.01 (m, 2H, 3,5-$H_{Ar}$), 7.25 (m, 1H, 2-$H_{pyrrole}$), 7.38–7.48 (m, 2H, 5,6-$H_{BTz}$), 7.51 (t, $^3J_{HH}$=7.8 Hz, 1H, 4-$H_{Ar}$), 7.58 (m, 1H, 4-$H_{pyrrole}$), 7.68 (d, $^3J_{HH}$=7.8 Hz, 1H, 6-$H_{Ar}$), 7.96 (d, $^3J_{HH}$=8.1 Hz, 1H, 7-$H_{BTz}$), 8.09 (d, $^3J_{HH}$=8.1 Hz, 1H, 4-$H_{BTz}$), 11.05 (s, 1H, OH), 12.95 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=113.0 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 122.5 (7-$C_{BTz}$), 122.8 (4-$C_{BTz}$), 124.2 (1-$C_{Ar}$), 125.6 (6-$C_{BTz}$), 125.8 (3-$C_{pyrrole}$), 127.1 (5-$C_{BTz}$), 127.8 (2-$C_{pyrrole}$), 129.9 (5-$C_{pyrrole}$), 130.9 (6-$C_{Ar}$), 134.1 (4-$C_{Ar}$), 134.3 (7a-$C_{BTz}$), 153.7 (3a-$C_{BTz}$), 158.9 (2-$C_{BTz}$), 159.3 (2-$C_{Ar}$), 191.7 (C=O). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3650–3350 (br, OH), 3250 (br, NH), 3061, 2926, 1620 (C=O), 1585, 1481, 1466, 1437, 1369, 1232, 1163, 922, 893, 758, 727, 706, 677. APSI MS: M$^+$+1=321. Anal. Calcd for $C_{18}H_{12}N_2O_2S$: C, 67.48; H, 3.78; N, 8.74; S, 10.01. Found: C, 67.31; H, 3.89; N, 8.81; S, 9.90.

#### 4.2.9. (2-Hydroxyphenyl)[5-(4-methyl-4H-1,2,4-triazol-3-yl)-1H-pyrrol-3-yl]methanone (**3i**)

Yellow solid (418 mg, 78%); mp 239–240 °C (EtOH). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=3.96 (s, 3H, $NCH_3$), 6.94 (t, $^3J_{HH}$=8.1 Hz, 1H, 5-$H_{Ar}$), 7.01 (d, $^3J_{HH}$=8.1 Hz, 1H, 3-$H_{Ar}$), 7.27 (s, 1H, 2-$H_{pyrrole}$), 7.43 (t, $^3J_{HH}$=8.1 Hz, 1H, 4-$H_{Ar}$), 7.63 (d, $^3J_{HH}$=8.1 Hz, 1H, 6-$H_{Ar}$), 7.64 (s, 1H, 4-$H_{pyrrole}$), 9.38 (s, 1H, 5-$H_{triazole}$), 10.99 (br s, 1H, OH), 12.90 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=33.9 ($CH_3$), 113.7 (4-$C_{pyrrole}$), 117.1 (3-$C_{pyrrole}$), 117.8 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 124.1 (1-$C_{Ar}$), 125.8 (5-$C_{pyrrole}$), 130.0 (2-$C_{pyrrole}$), 131.0 (6-$C_{Ar}$), 134.1 (4-$C_{Ar}$), 145.5 (5-$C_{triazole}$), 146.9 (3-$C_{triazole}$), 159.1 (2-$C_{Ar}$), 191.8 (C=O). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3410 (br, OH, NH), 3093, 2920, 1620 (C=O), 1595, 1485, 1444, 1363, 1269, 1209, 1151, 1065, 895, 812, 773. APSI MS: M$^+$+1=269. Anal. Calcd for $C_{14}H_{12}N_4O_2$: C, 62.68; H, 4.51; N, 20.88. Found: C, 62.85; H, 4.38; N, 20.80.

#### 4.2.10. [5-(6,7-Dihydro-5H-pyrrolo[2,1-c][1,2,4]triazol-3-yl)-1H-pyrrol-3-yl](2-hydroxyphenyl)methanone (**3j**)

Pale yellow solid (470 mg, 80%); mp 275–276 °C (MeOH). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=2.73 (m, 2H, $NCH_2CH_2$), 2.88 (t, $^3J_{HH}$=7.2 Hz, 2H, $CH_2$), 4.19 (t, $^3J_{HH}$=7.2 Hz, 2H, $NCH_2$), 6.92–7.00 (m, 3H, 3,5-$H_{Ar}$, 2-$H_{pyrrole}$), 7.45 (m, 2H, 4-$H_{pyrrole}$, 4-$H_{Ar}$), 7.70 (d, $^3J_{HH}$=7.5 Hz, 1H, 6-$H_{Ar}$), 11.12 (s, 1H, OH), 12.59 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=20.7 ($NCH_2CH_2$), 28.6 ($CH_2$), 44.0 ($NCH_2$), 108.8 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 121.5 (3-$C_{pyrrole}$), 124.0 (1-$C_{Ar}$), 125.2 (5-$C_{pyrrole}$), 128.8 (2-$C_{pyrrole}$), 131.0 (6-$C_{Ar}$), 134.0 (4-$C_{Ar}$), 143.9 (3-$C_{triazole}$), 159.1 (2-$C_{Ar}$), 162.9 (5-$C_{triazole}$), 191.9 (C=O). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3650–3290 (br, OH, NH), 3130, 3055, 2966, 1614 (C=O), 1578, 1485, 1466, 1346, 1215, 1173, 891, 814, 756, 704. APSI MS: M$^+$+1=295. Anal. Calcd for $C_{16}H_{14}N_4O_2$: C, 65.30; H, 4.79; N, 19.04. Found: C, 65.41; H, 4.72; N, 19.01.

#### 4.2.11. (2-Hydroxyphenyl)[5-(6,7,8,9-tetrahydro-5H-[1,2,4]triazolo-[4,3-a]azepin-3-yl)-1H-pyrrol-3-yl]methanone (**3k**)

Pale yellow solid (438 mg, 68%); mp 216–217 °C (EtOH). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=1.63 (m, 2H, 5-$H_{PHAz}$), 1.73 (m, 2H, 4-$H_{PHAz}$), 1.82 (m, 2H, 6-$H_{PHAz}$), 2.95 (m, 2H, 3-$H_{PHAz}$), 4.17 (m, 2H, 7-$H_{PHAz}$), 6.89 (s, 1H, 2-$H_{pyrrole}$), 6.95 (m, 2H, 3,5-$H_{Ar}$), 7.43 (t, $^3J_{HH}$=8.0 Hz, 1H, 4-$H_{Ar}$), 7.50 (s, 1H, 4-$H_{pyrrole}$), 7.68 (d, $^3J_{HH}$=8.0 Hz, 1H, 6-$H_{Ar}$), 11.10 (s, 1H, OH), 12.49 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=25.6 (4-$C_{PHAz}$), 26.3 (6-$C_{PHAz}$), 28.4 (5-$C_{PHAz}$), 30.2 (3-$C_{PHAz}$), 45.6 (7-$C_{PHAz}$), 110.4 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 120.6 (3-$C_{pyrrole}$), 124.1 (1-$C_{Ar}$), 125.1 (5-$C_{pyrrole}$), 128.6 (2-$C_{pyrrole}$), 131.0 (6-$C_{Ar}$), 134.0 (4-$C_{Ar}$), 147.7 (3-$C_{Triazole}$), 157.9 (5-$C_{Triazole}$), 159.0 (2-$C_{Ar}$), 191.9 (C=O). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3650–3300 (br, OH, NH), 3107, 3039, 2941, 2916, 2899, 1622 (C=O), 1581, 1531, 1487, 1444, 1350, 1275, 1223, 1157, 897, 856, 816, 760, 706. APSI MS: M$^+$+1=323. Anal. Calcd for $C_{18}H_{18}N_4O_2$: C, 67.07; H, 5.63; N, 17.38. Found: C, 67.00; H, 5.67; N, 17.33.

#### 4.2.12. (2-Hydroxyphenyl)[5-(3-phenyl-1,2,4-oxadiazol-5-yl)-1H-pyrrol-3-yl]methanone (**3l**)

Beige solid (477 mg, 72%); mp 207–209 °C (EtOH). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=6.99 (m, 2H, 3,5-$H_{Ar}$), 7.43 (d, $^4J_{HH}$=1.7 Hz, 1H, 2-$H_{pyrrole}$), 7.47 (t, $^3J_{HH}$=8.0 Hz, 1H, 4-$H_{Ar}$), 7.61 (m, 3H, 3,4,5-$H_{Ph}$), 7.66 (d, $^3J_{HH}$=8.0 Hz, 1H, 6-$H_{Ar}$), 7.76 (d, $^4J_{HH}$=1.7 Hz, 1H, 2-$H_{pyrrole}$), 8.08 (d, $^3J_{HH}$=7.8 Hz, 2H, 2,6-$H_{Ph}$), 10.94 (s, 1H, OH), 13.24 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=115.8 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 118.0 (3-$C_{pyrrole}$), 119.6 (5-$C_{Ar}$), 124.4 (1-$C_{Ar}$), 126.3 (2-$C_{pyrrole}$), 126.6 (1-$C_{Ph}$), 127.6 (3,5-$C_{Ph}$), 129.7 (2,6-$C_{Ph}$), 130.9 (6-$C_{Ar}$), 131.1 (4-$C_{Ph}$), 132.1 (5-$C_{pyrrole}$), 134.1 (4-$C_{Ar}$), 158.6 (2-$C_{Ar}$), 168.4 (5-$C_{oxadiazole}$), 169.6 (3-$C_{oxadiazole}$), 191.5 (C=O). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3650–3320 (br, OH), 3240 (br, NH), 3128, 3062, 2926, 1637 (C=O), 1593, 1485, 1444, 1352, 1217, 1155, 897, 742, 690. APSI MS: M$^+$+1=332. Anal. Calcd for $C_{19}H_{13}N_3O_3$: C, 68.88; H, 3.95; N, 12.68. Found: C, 69.02; H, 3.83; N, 12.63.

*4.2.13. (2-Hydroxyphenyl)(5-[1,2,4]triazolo[4,3-a]pyridin-3-yl-1H-pyrrol-3-yl)methanone (**3m**)*

Light brown solid (547 mg, 90%); mp 283–284 °C (EtOH). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=6.93–7.01 (m, 2H, 3,5-H$_{Ar}$), 7.08 (t, $^3J_{HH}$=8.6 Hz, 1H, 6-H$_{Het}$), 7.37 (s, 1H, 2-H$_{pyrrole}$), 7.45 (m, 2H, 4-H$_{Ar}$, 7-H$_{Het}$), 7.60 (s, 1H, 4-H$_{pyrrole}$), 7.75 (d, $^3J_{HH}$=7.8 Hz, 1H, 6-H$_{Ar}$), 7.85 (d, $^3J_{HH}$=8.6 Hz, 1H, 8-H$_{Het}$), 8.71 (d, $^3J_{HH}$=7.8 Hz, 1H, 5-H$_{Het}$), 11.18 (s, 1H, OH), 12.80 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=109.6 (8-C$_{Het}$), 115.1 (6-C$_{Het}$), 116.0 (4-C$_{pyrrole}$), 117.6 (3-C$_{Ar}$), 119.5 (5-C$_{Ar}$), 120.1 (3-C$_{pyrrole}$), 124.0 (1-C$_{Ar}$), 125.0 (2-C$_{pyrrole}$), 125.5 (5-C$_{pyrrole}$), 128.4 (5-C$_{Het}$), 129.1 (7-C$_{Het}$), 131.1 (6-C$_{Ar}$), 134.1 (4-C$_{Ar}$), 140.4 (3-C$_{Het}$), 150.1 (8a-C$_{Het}$), 159.1 (2-C$_{Ar}$), 192.0 (C=O). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3650–3300 (br, OH, NH), 3134, 3059, 2900, 1635 (C=O), 1622, 1589, 1489, 1466, 1356, 1242, 1211, 1146, 1068, 893, 816, 750. APSI MS: M$^+$+1=305. Anal. Calcd for $C_{17}H_{12}N_4O_2$: C, 67.10; H, 3.97; N, 18.41. Found: C, 66.97; H, 4.09; N, 18.35.

*4.2.14. (2-Hydroxyphenyl){5-[6-(trifluoromethyl)[1,2,4]triazolo-[4,3-a]pyridin-3-yl]-1H-pyrrol-3-yl}methanone (**3n**)*

Light brown solid (603 mg, 81%); mp 238–239 °C (EtOH). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=6.90–7.03 (m, 2H, 3,5-H$_{Ar}$), 7.47 (m, 2H, 4-H$_{Ar}$, 2-H$_{pyrrole}$, 6-H$_{Ar}$), 7.65 (m, 2H, 4-H$_{pyrrole}$), 7.76 (d, $^3J_{HH}$=7.8 Hz, 1H, 7-H$_{Het}$), 8.05 (d, $^3J_{HH}$=7.8 Hz, 1H, 8-H$_{Het}$), 8.91 (m, 1H, 5-H$_{Het}$), 11.16 (s, 1H, OH), 12.80 (br s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=111.2 (8-C$_{Het}$), 117.0 (q, $^2J_{CF}$=33.6 Hz, 6-C$_{Het}$), 117.5 (4-C$_{pyrrole}$), 117.6 (3-C$_{Ar}$), 119.1 (3-C$_{pyrrole}$), 119.5 (5-C$_{Ar}$), 123.8 (q, $^1J_{CF}$=271.1 Hz, CF$_3$), 123.9 (1-C$_{Ar}$), 124.0 (5-C$_{pyrrole}$), 125.3 (q, $^3J_{CF}$=6.7 Hz, 7-C$_{Het}$), 125.5 (2-C$_{pyrrole}$), 129.5 (3-C$_{Het}$), 131.1 (6-C$_{Ar}$), 134.1 (4-C$_{Ar}$), 142.0 (8a-C$_{Het}$), 149.6 (5-C$_{Het}$), 159.1 (2-C$_{Ar}$), 192.0 (C=O). $^{19}$F NMR (470 MHz, DMSO-$d_6$): $\delta$=−61.6 (CF$_3$). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3650–3300 (br, OH, NH), 3126, 3041, 2980, 2875, 1655 (C=O), 1620, 1589, 1487, 1354, 1327, 1300, 1240 (C–F), 1174, 1144, 1061, 895, 810, 764, 710. APSI MS: M$^+$+1=373. Anal. Calcd for $C_{18}H_{11}F_3N_4O_2$: C, 58.07; H, 2.98; N, 15.05. Found: C, 58.01; H, 3.09; N, 14.96.

*4.2.15. {5-[8-Chloro-6-(trifluoromethyl)[1,2,4]triazolo[4,3-a]-pyridin-3-yl]-1H-pyrrol-3-yl}(2-hydroxyphenyl)methanone (**3o**)*

Light brown solid (626 mg, 77%); mp 297–298 °C (EtOH). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=6.98 (m, 2H, 3,5-H$_{Ar}$), 7.46 (m, 1H, 4-H$_{Ar}$), 7.51 (s, 1H, 2-H$_{pyrrole}$), 7.66 (s, 1H, 4-H$_{pyrrole}$), 7.75 (m, 1H, 6-H$_{Ar}$), 8.01 (s, 1H, 7-H$_{Het}$), 8.90 (s, 1H, 5-H$_{Het}$), 11.14 (s, 1H, OH), 12.86 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=111.7 (8-C$_{Het}$), 117.1(q, $^2J_{CF}$=34.5 Hz, 6-C$_{Het}$), 117.6 (3-C$_{Ar}$), 118.7 (4-C$_{pyrrole}$), 119.5 (5-C$_{Ar}$), 122.3 (3-C$_{pyrrole}$), 123.0 (5-C$_{pyrrole}$), 123.2 (q, $^1J_{CF}$=271.5 Hz, CF$_3$), 124.1 (1-C$_{Ar}$), 124.7 (q, $^3J_{CF}$=6.7 Hz, 7-C$_{Het}$), 125.6 (2-C$_{pyrrole}$), 129.7 (3-C$_{Het}$), 131.1 (6-C$_{Ar}$), 134.1 (4-C$_{Ar}$), 143.7 (8a-C$_{Het}$), 147.7 (5-C$_{Het}$), 159.1 (2-C$_{Ar}$), 192.0 (C=O). $^{19}$F NMR (470 MHz, DMSO-$d_6$): $\delta$=−61.3 (CF$_3$). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3650–3340 (br, OH), 3169 (br, NH), 3089, 3051, 2916, 1664 (C=O), 1624, 1589, 1487, 1466, 1356, 1315, 1298, 1242 (C–F), 1132, 893, 810, 752, 704, 677. APSI MS: M$^+$+1=407. Anal. Calcd for $C_{18}H_{10}ClF_3N_4O_2$: C, 53.15; H, 2.48; Cl, 8.72; N, 13.77. Found: C, 53.01; H, 2.61; Cl, 8.80; N, 13.82.

*4.2.16. 2-[(4-Methyl-1,3-thiazol-2-yl)methyl]-3-(4-oxo-4H-chromen-3-yl)chromeno[2,3-c]pyrrol-9(2H)-one (**4g**)*

Light green solid (475 mg, 54%); mp 244–245 °C (EtOH/DMSO). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=2.21 (s, 3H, CH$_3$), 3.98 (s, 2H, CH$_2$), 7.15 (s, 1H, 5-H$_{Tz}$), 7.50 (t, $^3J_{HH}$=8.4 Hz, 1H, 6-H$_{Chr'}$), 7.61 (t, $^3J_{HH}$=8.4 Hz, 1H, 6-H$_{Chr}$), 7.69 (d, $^3J_{HH}$=8.4 Hz, 1H, 8-H$_{Chr'}$), 7.82 (m, 2H, 8-H$_{Chr}$, 7-H$_{Chr'}$), 7.93 (t, $^3J_{HH}$=8.4 Hz, 1H, 7-H$_{Chr}$), 8.06 (d, $^3J_{HH}$=8.4 Hz, 1H, 5-H$_{Chr'}$), 8.13 (d, $^3J_{HH}$=8.4 Hz, 1H, 5-H$_{Chr}$), 8.62 (s, 1H, 2-H$_{pyrrole}$), 8.89 (s, 1H, 2-H$_{Chr'}$). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=12.8 (CH$_3$), 23.7 (CH$_2$), 114.2 (5-C$_{Tz}$), 115.87 (3-C$_{Chr}$), 115.93 (3-C$_{Chr'}$), 119.0 (8-C$_{Chr'}$), 119.3 (8-C$_{Chr}$), 121.1 (5-C$_{pyrrole}$), 123.59 (4a-C$_{Chr}$), 123.63 (4a-C$_{Chr'}$), 125.6 (6-C$_{Chr}$), 125.9 (6-C$_{Chr'}$), 126.0 (5-C$_{Chr'}$), 126.9 (5-C$_{Chr}$), 128.0 (2-C$_{Chr}$), 128.8 (4-C$_{Tz}$), 129.7 (2-C$_{Tz}$), 134.7 (7-C$_{Chr'}$), 135.6 (7-C$_{Chr}$), 155.4 (2-C$_{pyrrole}$), 156.3 (8a-C$_{Chr'}$), 156.5 (8a-C$_{Chr}$), 159.1 (2-C$_{Chr'}$), 176.6 (C=O$_{Chr'}$), 176.7 (C=O$_{Chr}$). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3082, 2956, 2922, 1645 (C=O), 1616, 1574, 1466, 1398, 1350, 1319, 1217, 1153, 1103, 1045, 891, 760. APSI MS: M$^+$+1=441. Anal. Calcd for $C_{25}H_{16}N_2O_4S$: C, 68.17; H, 3.66; N, 6.36; S, 7.28. Found: C, 68.31; H, 3.55; N, 6.32; S, 7.37.

*4.2.17. 2-(1,3-Benzothiazol-2-ylmethyl)-3-(4-oxo-4H-chromen-3-yl)chromeno[2,3-c]pyrrol-9(2H)-one (**4h**)*

Green solid (609 mg, 64%); mp 298–299 °C (EtOH/DMSO). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=3.85 (s, 2H, CH$_2$), 7.27 (m, 3H, 5,6,7-H$_{BTz}$), 7.50 (t, $^3J_{HH}$=8.3 Hz, 1H, 6-H$_{Chr'}$), 7.59 (t, $^3J_{HH}$=8.3 Hz, 1H, 6-H$_{Chr}$), 7.67 (d, $^3J_{HH}$=8.3 Hz, 1H, 8-H$_{Chr'}$), 7.75–7.85 (m, 3H, 4-H$_{BTz}$, 8-H$_{Chr}$, 7-H$_{Chr'}$), 7.91 (td, $^3J_{HH}$=8.1 Hz, $^4J_{HH}$=1.7 Hz, 1H, 7-H$_{Chr}$), 8.10 (dd, $^3J_{HH}$=8.1 Hz, $^4J_{HH}$=1.7 Hz, 1H, 5-H$_{Chr'}$), 8.16 (dd, $^3J_{HH}$=8.1 Hz, $^4J_{HH}$=1.7 Hz, 1H, 5-H$_{Chr}$), 8.47 (s, 1H, 2-H$_{pyrrole}$), 8.76 (s, 1H, 2-H$_{Chr'}$). $^1$H NMR (500 MHz, CF$_3$COOD): $\delta$=4.32 (s, 2H, CH$_2$), 7.52 (t, $^3J_{HH}$=7.6 Hz, 1H, 5-H$_{BTz}$), 7.62–7.72 (m, 3H, 6-H$_{BTz}$, 6-H$_{Ch'}$, 6-H$_{Chr}$), 7.78 (m, 2H, 7-H$_{BTz}$, 8-H$_{Chr'}$), 7.91 (m, 2H, 4-H$_{BTz}$, 8-H$_{Chr}$), 7.97 (t, $^3J_{HH}$=7.6 Hz, 1H, 7-H$_{Chr'}$), 8.12 (t, $^3J_{HH}$=7.6 Hz, 1H, 7-H$_{Chr}$), 8.21 (d, $^3J_{HH}$=7.6 Hz, 1H, 5-H$_{Chr}$), 8.41 (d, $^3J_{HH}$=7.6 Hz, 1H, 5-H$_{Chr'}$), 8.61 (s, 1H, 2-H$_{pyrrole}$), 8.97 (s, 1H, 2-H$_{Chr'}$). $^{13}$C NMR (125 MHz, CF$_3$COOD): $\delta$=21.9 (CH$_2$), 109.1 (3-C$_{Chr'}$), 115.7 (7-C$_{BTz}$), 118.5 (3-C$_{Chr}$), 118.6 (8-C$_{Chr'}$), 118.7 (8-C$_{Chr}$), 121.7 (5-C$_{pyrrole}$), 122.0 (4a-C$_{Chr}$), 122.1 (4a-C$_{Chr'}$), 124.6 (4,6-C$_{BTz}$), 125.7 (6-C$_{Chr}$), 126.8 (6-C$_{Chr'}$), 127.3 (5-C$_{BTz}$), 128.0 (5-C$_{Chr'}$), 128.2 (2-C$_{Chr}$), 129.6 (5-C$_{Chr}$), 130.3 (7a-C$_{BTz}$), 131.2 (3a-C$_{BTz}$), 132.7 (2-C$_{BTz}$), 136.2 (7-C$_{Chr'}$), 137.3 (7-C$_{Chr}$), 156.9 (2-C$_{pyrrole}$), 157.2 (8a-C$_{Chr'}$), 157.6 (8a-C$_{Chr}$), 162.0 (2-C$_{Chr'}$), 177.4 (C=O$_{Chr'}$), 181.1 (C=O$_{Chr}$). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3082, 3062, 3041, 1647 (C=O), 1608, 1572, 1466, 1400, 1346, 1321, 1119, 891, 760, 744. APSI MS: M$^+$+1=477. Anal. Calcd for $C_{28}H_{16}N_2O_4S$: C, 70.58; H, 3.38; N, 5.88; S, 6.73. Found: C, 70.46; H, 3.51; N, 5.95; S, 6.67.

*4.2.18. [5-(1H-Benzimidazol-2-yl)-1-methyl-1H-pyrrol-3-yl](2-hydroxyphenyl)methanone (**6a**)*

Light green solid (558 mg, 88%); mp 211–212 °C (EtOH/DMF). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=4.17 (s, 3H, NCH$_3$), 6.96 (t, $^3J_{HH}$=8.3 Hz, 1H, 5-H$_{Ar}$), 7.01 (d, $^3J_{HH}$=8.3 Hz, 1H, 3-H$_{Ar}$), 7.41–7.50 (m, 3H, 5,6-H$_{BIm}$, 4-H$_{Ar}$), 7.56 (d, $^4J_{HH}$=1.4 Hz, 1H, 2-H$_{pyrrole}$), 7.68 (d, $^3J_{HH}$=8.3 Hz, 1H, 6-H$_{Ar}$), 7.77 (m, 2H, 4,7-H$_{BIm}$), 7.90 (d, $^4J_{HH}$=1.4 Hz, 1H, 4-H$_{pyrrole}$), 8.77 (br s, 1H, NH), 10.98 (br s, 1H, OH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=37.4 (CH$_3$), 114.5 (4,7-C$_{BIm}$), 117.8 (3-C$_{Ar}$), 118.1 (4-C$_{pyrrole}$), 119.5 (5-C$_{Ar}$), 120.3 (3-C$_{pyrrole}$), 123.7 (5-C$_{pyrrole}$), 124.3 (1-C$_{Ar}$), 125.4 (5,6-C$_{BIm}$), 126.5 (2-C$_{pyrrole}$), 131.0 (6-C$_{Ar}$), 133.6 (3a,7a-C$_{BIm}$), 134.3 (4-C$_{Ar}$), 142.1 (2-C$_{BIm}$), 159.4 (2-C$_{Ar}$), 191.3 (C=O). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3413 (br, OH, NH), 3097, 3045, 2926, 1622 (C=O), 1595, 1481, 1442, 1389, 1354, 1246, 1200, 1159, 895, 750. APSI MS: M$^+$+1=318. Anal. Calcd for $C_{19}H_{15}N_3O_2$: C, 71.91; H, 4.76; N, 13.24. Found: C, 71.73; H, 4.89; N, 13.31.

*4.2.19. {5-[1-(Difluoromethyl)-1H-benzimidazol-2-yl]-1-methyl-1H-pyrrol-3-yl}(2-hydroxyphenyl)methanone (**6b**)*

Pale yellow solid (675 mg, 92%); mp 161–162 °C (EtOH/DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=3.95 (s, 3H, NCH$_3$), 6.94–7.02 (m, 3H, 3,5-H$_{Ar}$, 2-H$_{pyrrole}$), 7.38–7.49 (m, 3H, 5,6-H$_{BIm}$, 4-H$_{Ar}$), 7.72 (d, $^3J_{HH}$=8.0 Hz, 1H, 6-H$_{Ar}$), 7.76–7.84 (m, 2H, 4,7-H$_{BIm}$), 7.87 (s, 1H, 4-H$_{pyrrole}$), 8.02 (t, $^2J_{HF}$=58.0 Hz, 1H, CHF$_2$), 11.04 (s, 1H, OH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=36.7 (CH$_3$), 111.0 (t, $^1J_{CF}$=247.3 Hz, CHF$_2$), 112.9 (7-C$_{BIm}$), 115.3 (4-C$_{pyrrole}$), 117.7 (3-C$_{Ar}$), 119.5 (5-C$_{Ar}$), 120.5 (4-C$_{BIm}$), 122.2 (3-C$_{pyrrole}$), 123.6 (5-C$_{pyrrole}$), 124.0 (1-C$_{Ar}$), 124.7 (5-C$_{BIm}$), 125.1 (6-C$_{BIm}$), 131.0 (6-C$_{Ar}$), 131.4 (7a-C$_{BIm}$), 134.1 (2-C$_{pyrrole}$), 134.4 (4-C$_{Ar}$), 143.0 (2-C$_{BIm}$), 143.9 (3a-C$_{BIm}$), 159.1 (2-C$_{Ar}$), 191.3 (C=O). $^{19}$F NMR (470 MHz, DMSO-$d_6$): $\delta$=−95.2 (d, $^2J_{HF}$=58.0 Hz, CHF$_2$). IR (KBr), $\nu_{max}$ (cm$^{-1}$): 3640–3310 (br, OH), 3126, 3099, 3055, 2954, 1620 (C=O), 1585, 1529, 1487, 1417, 1398,

1338, 1286, 1217, 1157, 1120, 1093, 1038, 924, 897, 850, 766, 750, 710. APSI MS: $M^+$+1=368. Anal. Calcd for $C_{20}H_{15}F_2N_3O_2$: C, 65.39; H, 4.12; N, 11.44. Found: C, 65.52; H, 4.01; N, 11.50.

*4.2.20. (2-Hydroxyphenyl)(1-methyl-5-pyridin-4-yl-1H-pyrrol-3-yl)methanone (**6c**)*

Pale yellow solid (361 mg, 65%); mp 130–131 °C (EtOH). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=3.82 (s, 3H, $NCH_3$), 6.91–7.00 (m, 3H, 3,5-$H_{Ar}$, 2-$H_{pyrrole}$), 7.45 (t, $^3J_{HH}$=7.8 Hz, 1H, 4-$H_{Ar}$), 7.58 (dd, $^3J_{HH}$=4.8 Hz, $^4J_{HH}$=1.7 Hz, 2H, 3,5-$H_{Py}$), 7.70 (s, 1H, 4-$H_{pyrrole}$), 7.73 (d, $^3J_{HH}$=7.8 Hz, 1H, 6-$H_{Ar}$), 8.61 (dd, $^3J_{HH}$=4.8 Hz, $^4J_{HH}$=1.7 Hz, 2H, 2,6-$H_{Py}$), 11.21 (s, 1H, OH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=36.5 ($CH_3$), 112.2 (4-$C_{pyrrole}$), 117.7 (3-$C_{Ar}$), 119.4 (5-$C_{Ar}$), 122.6 (3,5-$C_{Py}$), 123.4 (3-$C_{pyrrole}$), 123.7 (1-$C_{Ar}$), 131.0 (6-$C_{Ar}$), 133.1 (5-$C_{pyrrole}$), 133.8 (2-$C_{pyrrole}$), 134.1 (4-$C_{Ar}$), 139.1 (4-$C_{Py}$), 150.4 (2,6-$C_{Py}$), 159.4 (2-$C_{Ar}$), 191.8 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3160 (br, OH), 3109, 3034, 2924, 1624 (C=O), 1599, 1581, 1541, 1477, 1441, 1396, 1344, 1284, 1250, 1205, 1157, 893, 835, 818, 760, 704, 669. APSI MS: $M^+$+1=279. Anal. Calcd for $C_{17}H_{14}N_2O_2$: C, 73.37; H, 5.07; N, 10.07. Found: C, 73.44; H, 5.01; N, 10.04.

*4.2.21. [5-(1,3-Benzothiazol-2-yl)-1-methyl-1H-pyrrol-3-yl](2-hydroxyphenyl)methanone (**6d**)*

Light green solid (474 mg, 71%); mp 155–156 °C (EtOH). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=4.14 (s, 3H, $NCH_3$), 6.98 (m, 2H, 3,5-$H_{Ar}$), 7.24 (d, $^4J_{HH}$=1.7 Hz, 1H, 2-$H_{pyrrole}$), 7.45 (m, 2H, 5,6-$H_{BTz}$), 7.52 (t, $^3J_{HH}$=8.1 Hz, 1H, 4-$H_{Ar}$), 7.67 (t, $^3J_{HH}$=8.1 Hz, 1H, 6-$H_{Ar}$), 7.81 (d, $^4J_{HH}$=1.7 Hz, 1H, 4-$H_{pyrrole}$), 8.00 (d, $^3J_{HH}$=8.1 Hz, 1H, 7-$H_{BTz}$), 8.10 (d, $^3J_{HH}$=8.1 Hz, 1H, 4-$H_{BTz}$), 11.02 (br s, 1H, OH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=38.0 ($CH_3$), 115.6 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 122.4 (7-$C_{BTz}$), 123.0 (2-$C_{BTz}$), 123.9 (3-$C_{pyrrole}$), 124.2 (1-$C_{Ar}$), 125.9 (6-$C_{BTz}$), 127.1 (5-$C_{BTz}$), 127.7 (2-$C_{pyrrole}$), 130.8 (6-$C_{Ar}$), 133.9 (5-$C_{pyrrole}$), 134.0 (4-$C_{Ar}$), 135.2 (7a-$C_{BTz}$), 153.9 (3a-$C_{BTz}$), 158.8 (2-$C_{Ar}$), 159.4 (2-$C_{BTz}$), 191.2 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3150 (br, OH), 3062, 2926, 1645, 1620 (C=O), 1587, 1564, 1489, 1342, 1211, 1157, 955, 895, 750, 702. APSI MS: $M^+$+1=335. Anal. Calcd for $C_{19}H_{14}N_2O_2S$: C, 68.25; H, 4.22; N, 8.38; S, 9.59. Found: C, 68.39; H, 4.10; N, 8.45; S, 9.47.

*4.2.22. [5-(1,3-Benzoxazol-2-yl)-1-isopropyl-1H-pyrrol-3-yl](2-hydroxyphenyl)methanone (**6e**)*

Light green solid (657 mg, 95%); mp 135–136 °C (EtOH/MeCN). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=1.53 (d, $^3J_{HH}$=6.6 Hz, 6H, $CH(CH_3)_2$), 5.76 (m, 1H, $CH(CH_3)_2$), 6.99 (m, 2H, 3,5-$H_{Ar}$), 7.33 (m, 1H, 2-$H_{pyrrole}$), 7.39 (m, 2H, 5,6-$H_{BOx}$), 7.46 (t, $^3J_{HH}$=8.0 Hz, 1H, 4-$H_{Ar}$), 7.66 (d, $^3J_{HH}$=8.0 Hz, 1H, 6-$H_{Ar}$), 7.74 (d, $^3J_{HH}$=8.0 Hz, 1H, 7-$H_{BOx}$), 7.78 (d, $^3J_{HH}$=8.0 Hz, 1H, 4-$H_{BOx}$), 8.02 (d, $^4J_{HH}$=1.4 Hz, 1H, 4-$H_{pyrrole}$), 10.97 (s, 1H, OH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=23.5 ($CH(CH_3)_2$), 50.2 ($CH(CH_3)_2$), 111.0 (7-$C_{BOx}$), 116.5 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.6 (5-$C_{Ar}$), 120.0 (4-$C_{BOx}$), 120.9 (3-$C_{pyrrole}$), 124.3 (1-$C_{Ar}$), 124.6 (5-$C_{pyrrole}$), 125.3 (6-$C_{BOx}$), 125.8 (5-$C_{BOx}$), 129.8 (2-$C_{pyrrole}$), 130.9 (6-$C_{Ar}$), 134.1 (4-$C_{Ar}$), 141.8 (3a-$C_{BOx}$), 149.4 (2-$C_{BOx}$), 156.5 (7a-$C_{BOx}$), 158.7 (2-$C_{Ar}$), 191.3 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3100 (br, OH), 2966, 2924, 1626 (C=O), 1583, 1458, 1392, 1242, 1215, 1153, 1034, 748. APSI MS: $M^+$+1=347. Anal. Calcd for $C_{21}H_{18}N_2O_3$: C, 72.82; H, 5.24; N, 8.09. Found: C, 72.70; H, 5.20; N, 8.17.

*4.2.23. (2-Hydroxyphenyl){5-[3-(4-methoxyphenyl)isoxazol-5-yl]-1-methyl-1H-pyrrol-3-yl}methanone (**6f**)*

Pale yellow solid (524 mg, 70%); mp 134–135 °C (EtOH). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=3.82 (s, 3H, $OCH_3$), 3.95 (s, 3H, $NCH_3$), 6.96 (m, 2H, 3,5-$H_{Ar}$), 7.08 (d, $^3J_{HH}$=8.3 Hz, 2H, 3,5-$H_{Ar'}$), 7.12 (1H, s, 2-$H_{pyrrole}$), 7.33 (1H, s, 4-$H_{isoxazole}$), 7.45 (t, $^3J_{HH}$=7.8 Hz, 1H, 4-$H_{Ar}$), 7.67 (d, $^3J_{HH}$=7.8 Hz, 1H, 6-$H_{Ar}$), 7.76 (1H, s, 4-$H_{pyrrole}$), 7.87 (d, $^3J_{HH}$=8.3 Hz, 2H, 2,6-$H_{Ar'}$), 11.04 (s, 1H, OH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=37.1 ($NCH_3$), 55.8 ($OCH_3$), 99.0 (4-$C_{isoxazole}$), 112.9 (4-$C_{pyrrole}$), 115.0 (3,5-$C_{Ar'}$), 117.7 (3-$C_{Ar}$), 119.4 (5-$C_{Ar}$), 121.2 (1-$C_{Ar'}$), 123.2 (3-$C_{pyrrole}$), 123.7 (5-$C_{pyrrole}$), 124.1 (1-$C_{Ar}$), 128.7 (2,6-$C_{Ar'}$), 130.8 (6-$C_{Ar}$), 133.8 (4-$C_{Ar}$), 134.1 (2-$C_{pyrrole}$), 159.0 (2-$C_{Ar}$), 161.3 (5-$C_{isoxazole}$), 162.4 (3-$C_{isoxazole}$), 162.6 (4-$C_{Ar'}$), 191.5 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3180 (br, OH), 3143, 3134, 3012, 2972, 2922, 1622 (C=O), 1583, 1487, 1433, 1390, 1344, 1248, 1225, 1176, 1161, 1028, 943, 891, 837, 785, 758, 704, 669. APSI MS: $M^+$+1=375. Anal. Calcd for $C_{22}H_{18}N_2O_4$: C, 70.58; H, 4.85; N, 7.48. Found: C, 70.74; H, 4.76; N, 7.41.

*4.2.24. [5-(6,7-Dihydro-5H-pyrrolo[2,1-c][1,2,4]triazol-3-yl)-1-methyl-1H-pyrrol-3-yl](2-hydroxyphenyl)methanone (**6g**)*

Light green solid (517 mg, 84%); mp 164–165 °C (EtOH). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=2.79 (m, 2H, $NCH_2CH_2$), 3.14 (t, $^3J_{HH}$=7.1 Hz, 2H, $CH_2$), 3.95 (s, 3H, $NCH_3$), 4.32 (t, $^3J_{HH}$=7.1 Hz, 2H, $NCH_2$), 6.94 (t, $^3J_{HH}$=8.3 Hz, 1H, 5-$H_{Ar}$), 7.00 (d, $^3J_{HH}$=8.3 Hz, 1H, 3-$H_{Ar}$), 7.16 (d, $^4J_{HH}$=1.7 Hz, 1H, 2-$H_{pyrrole}$), 7.43 (t, $^3J_{HH}$=8.3 Hz, 1H, 4-$H_{Ar}$), 7.61 (d, $^3J_{HH}$=8.3 Hz, 1H, 6-$H_{Ar}$), 7.79 (d, $^4J_{HH}$=1.7 Hz, 1H, 4-$H_{pyrrole}$), 10.94 (br s, 1H, OH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=21.8 ($NCH_2CH_2$), 28.4 ($CH_2$), 37.5 ($NCH_3$), 46.7 ($NCH_2$), 114.2 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.1 (3-$C_{pyrrole}$), 119.5 (5-$C_{Ar}$), 124.1 (1-$C_{Ar}$), 124.6 (5-$C_{pyrrole}$), 130.6 (6-$C_{Ar}$), 133.8 (4-$C_{Ar}$), 135.0 (2-$C_{pyrrole}$), 143.4 (3-$C_{triazole}$), 158.4 (2-$C_{Ar}$), 162.0 (5-$C_{triazole}$), 191.0 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3520 (OH), 3196 (br, OH), 3138, 3113, 2966, 2939, 1603 (C=O), 1572, 1481, 1390, 1369, 1335, 1261, 1227, 1194, 1157, 1101, 989, 891, 810, 771, 710, 669. APSI MS: $M^+$+1=309. Anal. Calcd for $C_{17}H_{16}N_4O_2$: C, 66.22; H, 5.23; N, 18.17. Found: C, 66.30; H, 5.16; N, 10.40.

*4.2.25. (2-Hydroxyphenyl)[1-methyl-5-(6,7,8,9-tetrahydro-5H-[1,2,4]triazolo[4,3-a]azepin-3-yl)-1H-pyrrol-3-yl]methanone (**6h**)*

Light green solid (410 mg, 61%); mp 175–176 °C (*i*-PrOH). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=1.65 (m, 2H, 5-$H_{PHAz}$), 1.71 (m, 2H, 4-$H_{PHAz}$), 1.81 (m, 2H, 6-$H_{PHAz}$), 2.96 (m, 2H, 3-$H_{PHAz}$), 3.73 (s, 3H, $NCH_3$), 4.02 (m, 2H, 7-$H_{PHAz}$), 6.79 (d, $^4J_{HH}$=1.2 Hz, 1H, 2-$H_{pyrrole}$), 6.92–7.00 (m, 2H, 3,5-$H_{Ar}$), 7.44 (t, $^3J_{HH}$=7.8 Hz, 1H, 4-$H_{Ar}$), 7.68–7.74 (m, 2H, 6-$H_{Ar}$, 4-$H_{pyrrole}$), 11.09 (s, 1H, OH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=25.5 (4-$C_{PHAz}$), 26.4 (6-$C_{PHAz}$), 28.3 (5-$C_{PHAz}$), 30.2 (3-$C_{PHAz}$), 36.1 ($NCH_3$), 45.6 (7-$C_{PHAz}$), 113.3 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.4 (5-$C_{Ar}$), 121.6 (3-$C_{pyrrole}$), 123.4 (5-$C_{pyrrole}$), 124.1 (1-$C_{Ar}$), 130.9 (6-$C_{Ar}$), 133.0 (2-$C_{pyrrole}$), 134.0 (4-$C_{Ar}$), 146.7 (3-$C_{triazole}$), 157.6 (5-$C_{triazole}$), 159.0 (2-$C_{Ar}$), 191.5 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3300 (br, OH), 3109, 2939, 2918, 1620 (C=O), 1578, 1525, 1483, 1443, 1396, 1342, 1275, 1242, 1213, 1151, 893, 814, 756, 704, 669. APSI MS: $M^+$+1=337. Anal. Calcd for $C_{19}H_{20}N_4O_2$: C, 67.84; H, 5.99; N, 16.65. Found: C, 67.96; H, 5.88; N, 16.58.

*4.2.26. (2-Hydroxyphenyl)[1-isopropyl-5-(5-phenyl-1,3,4-oxadiazol-2-yl)-1H-pyrrol-3-yl]methanone (**6i**)*

Beige solid (619 mg, 83%); mp 117–118 °C (EtOH/MeCN). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=1.53 (d, $^3J_{HH}$=6.8 Hz, 6H, $CH(CH_3)_2$), 5.45 (septet, $^3J_{HH}$=6.8 Hz, 1H, $CH(CH_3)_2$), 6.99 (m, 2H, 3,5-$H_{Ar}$), 7.38 (d, $^4J_{HH}$=1.5 Hz, 1H, 2-$H_{pyrrole}$), 7.47 (t, $^3J_{HH}$=8.1 Hz, 1H, 4-$H_{Ar}$), 7.58–7.66 (m, 3H, 3,4,5-$H_{Ph}$), 7.68 (d, $^3J_{HH}$=8.1 Hz, 1H, 6-$H_{Ar}$), 7.99 (d, $^4J_{HH}$=1.5 Hz, 1H, 4-$H_{pyrrole}$), 8.11 (d, $^3J_{HH}$=8.1 Hz, 2H, 2,6-$H_{Ph}$), 11.00 (s, 1H, OH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=23.4 ($CH(CH_3)_2$), 50.7 ($CH(CH_3)_2$), 115.8 (4-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 118.2 (3-$C_{pyrrole}$), 119.6 (5-$C_{Ar}$), 123.6 (1-$C_{Ph}$), 124.1 (1-$C_{Ar}$), 124.8 (5-$C_{pyrrole}$), 127.2 (2,6-$C_{Ph}$), 129.7 (4-$C_{Ph}$), 129.9 (3,5-$C_{Ph}$), 131.0 (6-$C_{Ar}$), 132.5 (2-$C_{pyrrole}$), 134.2 (4-$C_{Ar}$), 158.6 (2-$C_{oxadiazole}$), 158.9 (2-$C_{Ar}$), 163.1 (5-$C_{oxadiazole}$), 191.4 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3160 (br, OH), 3136, 3062, 2989, 2918, 1622 (C=O), 1606, 1589, 1487, 1446, 1394, 1348, 1236, 1205, 1053, 895, 758, 723, 694. APSI MS: $M^+$+1=374. Anal. Calcd for $C_{22}H_{19}N_3O_3$: C, 70.76; H, 5.13; N, 11.25. Found: C, 70.59; H, 5.25; N, 11.31.

*4.2.27. 2-[4-(2-Hydroxybenzoyl)-1-methyl-1H-pyrrol-2-yl]quinazolin-4(3H)-one (**6j**)*

Pale yellow solid (683 mg, 99%); mp 236–237 °C (EtOH/DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=4.14 (s, 3H, $NCH_3$), 6.98 (m, 2H,

3,5-$H_{Ar}$), 7.48 (m, 2H, 4-$H_{Ar}$, 6-$H_{Qnz}$), 7.68 (d, $^3J_{HH}$=8.0 Hz, 1H, 6-$H_{Ar}$), 7.69 (d, $^4J_{HH}$=1.2 Hz, 1H, 2-$H_{pyrrole}$), 7.76–7.82 (m, 2H, 7,8-$H_{Qnz}$), 7.82 (d, $^4J_{HH}$=1.2 Hz, 1H, 4-$H_{pyrrole}$), 8.12 (d, $^3J_{HH}$=8.0 Hz, 1H, 5-$H_{Qnz}$), 11.26 (s, 1H, OH), 12.38 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=38.7 (qd, $^1J_{CH}$=141.6 Hz, $^3J_{CH}$=2.0 Hz, $CH_3$), 116.4 (dd, $^1J_{CH}$=176.4 Hz, $^3J_{CH}$=5.5 Hz, 4-$C_{pyrrole}$), 117.7 (ddd, $^1J_{CH}$=163.1 Hz, $^2J_{CH}$=6.9 Hz, $^3J_{CH}$=5.5 Hz, 3-$C_{Ar}$), 119.4 (dd, $^1J_{CH}$=163.6 Hz, $^2J_{CH}$=7.8 Hz, 5-$C_{Ar}$), 121.3 (s, 4a-$C_{Qnz}$), 122.7 (dd, $^2J_{CH}$=6.4 Hz, $^2J_{CH}$=5.1 Hz, 3-$C_{pyrrole}$), 123.4 (d, $^2J_{CH}$=6.0 Hz, 1-$C_{Ar}$), 126.3 (dd, $^1J_{CH}$=163.1 Hz, $^2J_{CH}$=8.3 Hz, 5-$C_{Qnz}$), 126.4 (dd, $^2J_{CH}$=7.3 Hz, $^3J_{CH}$=5.1 Hz, 5-$C_{pyrrole}$), 126.8 (dd, $^1J_{CH}$=163.5 Hz, $^2J_{CH}$=7.4 Hz, 6-$C_{Qnz}$), 127.6 (dd, $^1J_{CH}$=162.7 Hz, $^2J_{CH}$=6.4 Hz, 8-$C_{Qnz}$), 131.3 (dd, $^1J_{CH}$=159.5 Hz, $^2J_{CH}$=8.3 Hz, 6-$C_{Ar}$), 134.4 (dd, $^1J_{CH}$=159.9 Hz, $^2J_{CH}$=8.7 Hz, 4-$C_{Ar}$), 135.0 (dd, $^1J_{CH}$=161.2 Hz, $^2J_{CH}$=9.2 Hz, 7-$C_{Qnz}$), 135.8 (dq, $^1J_{CH}$=187.0 Hz, $^3J_{CH}$=2.0 Hz, 2-$C_{pyrrole}$), 146.7 (s, 2-$C_{Qnz}$), 148.8 (dd, $^2J_{CH}$=8.2 Hz, $^2J_{CH}$=7.4 Hz, 8a-$C_{Qnz}$), 159.7 (m, 2-$C_{Ar}$), 162.3 (s, C=$O_{Qnz}$), 191.7 (s, C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3300 (br, OH, NH), 3178, 3124, 3039, 2958, 2926, 1726, 1672 (C=$O_{Qnz}$), 1614 (C=O), 1587, 1485, 1446, 1234, 1209, 1159, 893, 766. APSI MS: $M^+$+1=346. Anal. Calcd for $C_{20}H_{15}N_3O_3$: C, 69.56; H, 4.38; N, 12.17. Found: C, 69.49; H, 4.45; N, 12.20.

#### 4.2.28. 2-[1-Ethyl-4-(2-hydroxybenzoyl)-1H-pyrrol-2-yl]quinazolin-4(3H)-one (**6k**)

Pale yellow solid (711 mg, 99%); mp 258–260 °C (EtOH/DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=1.41 (t, $^3J_{HH}$=6.8 Hz, 3H, $CH_2CH_3$), 4.68 (q, $^3J_{HH}$=6.8 Hz, 2H, $CH_2CH_3$), 6.99 (m, 2H, 3,5-$H_{Ar}$), 7.49 (t, $^3J_{HH}$=8.0 Hz, 2H, 4-$H_{Ar}$, 6-$H_{Qnz}$), 7.65 (d, $^3J_{HH}$=8.0 Hz, 1H, 6-$H_{Ar}$), 7.71 (d, $^4J_{HH}$=1.6 Hz, 1H, 2-$H_{pyrrole}$), 7.81 (m, 2H, 7,8-$H_{Qnz}$), 7.89 (d, $^4J_{HH}$=1.6 Hz, 1H, 4-$H_{pyrrole}$), 8.12 (d, $^3J_{HH}$=8.0 Hz, 1H, 5-$H_{Qnz}$), 11.29 (s, 1H, OH), 12.37 (br s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=17.3 ($CH_3$), 45.3 ($CH_2$), 116.8 (4-$C_{pyrrole}$), 117.7 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 121.3 (4a-$C_{Qnz}$), 123.0 (3-$C_{pyrrole}$), 123.4 (1-$C_{Ar}$), 125.4 (5-$C_{Qnz}$), 126.3 (5-$C_{pyrrole}$), 126.8 (6-$C_{Qnz}$), 127.6 (8-$C_{Qnz}$), 131.3 (6-$C_{Ar}$), 134.4 (4-$C_{Ar}$), 134.7 (7-$C_{Qnz}$), 135.0 (2-$C_{pyrrole}$), 146.4 (2-$C_{Qnz}$), 148.8 (8a-$C_{Qnz}$), 159.6 (2-$C_{Ar}$), 162.3 (C=$O_{Qnz}$), 191.6 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3220 (br, OH, NH), 3113, 2981, 2931, 1672 (C=$O_{Qnz}$), 1612 (C=O), 1583, 1566, 1487, 1443, 1346, 1306, 1230, 1157, 933, 895, 771, 756, 710. APSI MS: $M^+$+1=360. Anal. Calcd for $C_{21}H_{17}N_3O_3$: C, 70.18; H, 4.77; N, 11.69. Found: C, 70.05; H, 4.88; N, 11.74.

#### 4.2.29. 2-[4-(2-Hydroxybenzoyl)-1-isopropyl-1H-pyrrol-2-yl]quinazolin-4(3H)-one (**6l**)

Pale yellow solid (716 mg, 96%); mp 231–232 °C (EtOH/DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=1.51 (d, $^3J_{HH}$=6.8 Hz, 6H, CH($CH_3$)$_2$), 5.80 (septet, $^3J_{HH}$=6.8 Hz, 1H, $CH$($CH_3$)$_2$), 7.00 (m, 2H, 3,5-$H_{Ar}$), 7.49 (t, $^3J_{HH}$=8.1 Hz, 2H, 4-$H_{Ar}$, 6-$H_{Qnz}$), 7.56 (d, $^4J_{HH}$=1.8 Hz, 1H, 2-$H_{pyrrole}$), 7.65 (d, $^3J_{HH}$=8.1 Hz, 1H, 6-$H_{Ar}$), 7.82 (m, 2H, 7,8-$H_{Qnz}$), 7.97 (d, $^4J_{HH}$=1.8 Hz, 1H, 4-$H_{pyrrole}$), 8.13 (d, $^3J_{HH}$=8.1 Hz, 1H, 5-$H_{Qnz}$), 11.36 (s, 1H, OH), 12.39 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=23.8 (CH($CH_3$)$_2$), 50.1 ($CH$($CH_3$)$_2$), 116.8 (4-$C_{pyrrole}$), 117.8 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 121.3 (4a-$C_{Qnz}$), 123.1 (3-$C_{pyrrole}$), 123.3 (1-$C_{Ar}$), 125.9 (5-$C_{Qnz}$), 126.3 (5-$C_{pyrrole}$), 126.9 (6-$C_{Qnz}$), 127.7 (8-$C_{Qnz}$), 130.2 (2-$C_{pyrrole}$), 131.5 (6-$C_{Ar}$), 134.5 (4-$C_{Ar}$), 135.0 (7-$C_{Qnz}$), 146.7 (2-$C_{Qnz}$), 148.7 (8a-$C_{Qnz}$), 159.8 (2-$C_{Ar}$), 162.4 (C=$O_{Qnz}$), 191.8 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3300 (br, OH, NH), 3163, 3138, 3057, 2999, 2960, 2922, 1672 (C=$O_{Qnz}$), 1621 (C=O), 1597, 1581, 1485, 1443, 1346, 1232, 1153, 895, 812, 771, 748, 704. APSI MS: $M^+$+1=374. Anal. Calcd for $C_{22}H_{19}N_3O_3$: C, 70.76; H, 5.13; N, 11.25. Found: C, 70.61; H, 5.25; N, 11.32.

#### 4.2.30. 2-[1-Butyl-4-(2-hydroxybenzoyl)-1H-pyrrol-2-yl]quinazolin-4(3H)-one (**6m**)

Pale yellow solid (689 mg, 89%); mp 246–247 °C (EtOH/DMF). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=0.89 (t, $^3J_{HH}$=7.2 Hz, 3H, $CH_2CH_3$), 1.31 (m, 2H, $CH_2CH_3$), 1.75 (m, 2H, $NCH_2CH_2$), 4.65 (t, $^3J_{HH}$=7.3 Hz, 2H, $NCH_2$), 6.98 (m, 2H, 3,5-$H_{Ar}$), 7.48 (m, 2H, 4-$H_{Ar}$, 6-$H_{Qnz}$), 7.62 (d, $^3J_{HH}$=8.1 Hz, 1H, 6-$H_{Ar}$), 7.72 (d, $^4J_{HH}$=1.5 Hz, 1H, 2-$H_{pyrrole}$), 7.78 (d, $^3J_{HH}$=8.1 Hz, 1H, 8-$H_{Qnz}$), 7.81 (t, $^3J_{HH}$=8.1 Hz, 1H, 7-$H_{Qnz}$), 7.86 (d, $^4J_{HH}$=1.5 Hz, 1H, 4-$H_{pyrrole}$), 8.11 (d, $^3J_{HH}$=8.1 Hz, 1H, 5-$H_{Qnz}$), 11.25 (s, 1H, OH), 12.36 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=11.2 ($CH_3$), 24.7 ($CH_2CH_3$), 39.6 ($NCH_2CH_2$), 51.8 ($NCH_2$), 117.5 (3-$C_{Ar}$), 117.8 (4-$C_{pyrrole}$), 119.4 (5-$C_{Ar}$), 120.1 (4a-$C_{Qnz}$), 122.8 (3-$C_{pyrrole}$), 123.3 (1-$C_{Ar}$), 125.1 (5-$C_{Qnz}$), 126.5 (5-$C_{pyrrole}$), 126.9 (6-$C_{Qnz}$), 128.4 (8-$C_{Qnz}$), 131.3 (6-$C_{Ar}$), 134.4 (4-$C_{Ar}$), 135.6 (7-$C_{Qnz}$), 139.6 (2-$C_{pyrrole}$), 147.8 (2-$C_{Qnz}$), 150.0 (8a-$C_{Qnz}$), 159.7 (2-$C_{Ar}$), 161.7 (C=$O_{Qnz}$), 191.6 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3300 (br, OH, NH), 3120, 3028, 2956, 1676 (C=$O_{Qnz}$), 1622 (C=O), 1579, 1483, 1227, 1157, 895, 766, 748, 704. APSI MS: $M^+$+1=388. Anal. Calcd for $C_{23}H_{22}N_3O_3$: C, 71.30; H, 5.46; N, 10.85. Found: C, 71.17; H, 5.53; N, 10.94.

#### 4.2.31. 2-[1-sec-Butyl-4-(2-hydroxybenzoyl)-1H-pyrrol-2-yl]-quinazolin-4(3H)-one (**6n**)

Pale yellow solid (704 mg, 91%); mp 187–188 °C (EtOH/DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=0.82 (t, $^3J_{HH}$=7.0 Hz, 3H, $CH_2CH_3$), 1.52 (d, $^3J_{HH}$=6.8 Hz, 3H, $CHCH_3$), 1.78 (m, 1H, $CH_3CH_{2(A)}$), 1.91 (m, 1H, $CH_3CH_{2(B)}$), 5.63 (m, 1H, $CHCH_3$), 7.00 (m, 2H, 3,5-$H_{Ar}$), 7.49 (t, $^3J_{HH}$=7.9 Hz, 2H, 4-$H_{Ar}$, 6-$H_{Qnz}$), 7.56 (d, $^4J_{HH}$=1.5 Hz, 1H, 2-$H_{pyrrole}$), 7.64 (d, $^3J_{HH}$=7.9 Hz, 1H, 6-$H_{Ar}$), 7.82 (m, 2H, 7,8-$H_{Qnz}$), 7.94 (d, $^4J_{HH}$=1.5 Hz, 1H, 4-$H_{pyrrole}$), 8.13 (d, $^3J_{HH}$=7.9 Hz, 1H, 5-$H_{Qnz}$), 11.34 (s, 1H, OH), 12.38 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=11.0 ($CH_2CH_3$), 21.3 ($CHCH_3$), 30.8 ($CH_2CH_3$), 55.3 ($CHCH_3$), 116.5 (4-$C_{pyrrole}$), 117.8 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 121.3 (4a-$C_{Qnz}$), 123.3 (3-$C_{pyrrole}$), 123.5 (1-$C_{Ar}$), 125.8 (5-$C_{Qnz}$), 126.3 (5-$C_{pyrrole}$), 126.9 (6-$C_{Qnz}$), 127.6 (8-$C_{Qnz}$), 130.4 (7-$C_{Qnz}$), 131.5 (6-$C_{Ar}$), 134.5 (4-$C_{Ar}$), 135.0 (2-$C_{pyrrole}$), 146.8 (2-$C_{Qnz}$), 148.7 (8a-$C_{Qnz}$), 159.7 (2-$C_{Ar}$), 163.9 (C=$O_{Qnz}$), 191.8 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3240 (br, OH, NH), 3136, 3066, 2974, 2924, 1678 (C=$O_{Qnz}$), 1613 (C=O), 1597, 1583, 1485, 1254, 1232, 1153, 893, 812, 766, 752, 710. APSI MS: $M^+$+1=388. Anal. Calcd for $C_{23}H_{21}N_3O_3$: C, 71.30; H, 5.46; N, 10.85. Found: C, 71.21; H, 5.59; N, 10.89.

#### 4.2.32. 2-[4-(2-Hydroxybenzoyl)-1-(2-methoxyethyl)-1H-pyrrol-2-yl]quinazolin-4(3H)-one (**6o**)

Pale yellow solid (755 mg, 97%); mp 241–242 °C (EtOH/DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=3.22 (s, 3H, $OCH_3$), 3.73 (t, $^3J_{HH}$=5.4 Hz, 2H, $OCH_2$), 4.84 (t, $^3J_{HH}$=5.4 Hz, 2H, $NCH_2$), 6.99 (m, 2H, 3,5-$H_{Ar}$), 7.48 (t, $^3J_{HH}$=7.8 Hz, 2H, 4-$H_{Ar}$, 6-$H_{Qnz}$), 7.63 (d, $^3J_{HH}$=8.1 Hz, 1H, 6-$H_{Ar}$), 7.71 (d, $^4J_{HH}$=1.6 Hz, 1H, 2-$H_{pyrrole}$), 7.75–7.85 (m, 3H, 4-$H_{pyrrole}$, 7,8-$H_{Qnz}$), 8.12 (d, $^3J_{HH}$=7.8 Hz, 1H, 5-$H_{Qnz}$), 11.25 (s, 1H, OH), 12.39 (br s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=49.6 ($NCH_2$), 58.6 ($OCH_3$), 71.9 ($OCH_2$), 116.8 (4-$C_{pyrrole}$), 117.7 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 121.3 (4a-$C_{Qnz}$), 122.8 (3-$C_{pyrrole}$), 123.4 (1-$C_{Ar}$), 125.6 (5-$C_{Qnz}$), 126.3 (5-$C_{pyrrole}$), 126.8 (6-$C_{Qnz}$), 127.5 (8-$C_{Qnz}$), 131.2 (6-$C_{Ar}$), 134.4 (4-$C_{Ar}$), 135.0 (7-$C_{Qnz}$), 135.9 (2-$C_{pyrrole}$), 146.6 (2-$C_{Qnz}$), 148.7 (8a-$C_{Qnz}$), 159.6 (2-$C_{Ar}$), 162.3 (C=$O_{Qnz}$), 191.6 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3630–3300 (br, OH, NH), 3174, 3124, 2980, 2926, 1676 (C=$O_{Qnz}$), 1612 (C=O), 1597, 1579, 1483, 1346, 1302, 1275, 1230, 1203, 1161, 1099, 928, 895, 822, 766, 754, 706. APSI MS: $M^+$+1=390. Anal. Calcd for $C_{22}H_{19}N_3O_4$: C, 67.86; H, 4.92; N, 10.79. Found: C, 67.99; H, 4.83; N, 10.73.

#### 4.2.33. 2-[1-[2-(Dimethylamino)ethyl]-4-(2-hydroxybenzoyl)-1H-pyrrol-2-yl]quinazolin-4(3H)-one hydrochloride (**6p**)

Pale yellow solid (745 mg, 85%); mp 257–258 °C (EtOH/DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=2.82 (s, 6H, N($CH_3$)$_2$), 3.60 (t, $^3J_{HH}$=7.1 Hz, 2H, $CH_2$N($CH_3$)$_2$), 5.08 (t, $^3J_{HH}$=7.1 Hz, 2H, $NCH_2$), 6.99 (t, $^3J_{HH}$=8.3 Hz, 1H, 5-$H_{Ar}$), 7.02 (d, $^3J_{HH}$=8.3 Hz, 1H, 3-$H_{Ar}$), 7.50 (m, 2H, 4-$H_{Ar}$, 6-$H_{Qnz}$), 7.77 (d, $^3J_{HH}$=8.0 Hz, 1H, 6-$H_{Ar}$), 7.83 (m, 3H, 2-$H_{pyrrole}$, 7,8-$H_{Qnz}$), 7.97 (d, $^4J_{HH}$=1.2 Hz, 1H, 4-$H_{pyrrole}$), 8.13 (d,

$^3J_{HH}$=8.0 Hz, 1H, 5-$H_{Qnz}$), 10.77 (br s, 1H, NH), 11.13 (s, 1H, OH), 12.41 (br s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=43.1 ($N(CH_3)_2$), 45.0 ($NCH_2$), 56.8 ($CH_2N(CH_3)_2$), 117.0 (4-$C_{pyrrole}$), 117.7 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 121.4 (4a-$C_{Qnz}$), 123.7 (3-$C_{pyrrole}$), 123.8 (1-$C_{Ar}$), 125.5 (5-$C_{Qnz}$), 126.2 (5-$C_{pyrrole}$), 127.0 (6-$C_{Qnz}$), 128.0 (8-$C_{Qnz}$), 131.1 (6-$C_{Ar}$), 134.3 (4-$C_{Ar}$), 135.0 (7-$C_{Qnz}$), 135.9 (2-$C_{pyrrole}$), 146.3 (2-$C_{Qnz}$), 148.7 (8a-$C_{Qnz}$), 159.2 (2-$C_{Ar}$), 162.3 (C=$O_{Qnz}$), 191.4 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3466 (br, OH, NH), 3180, 3138, 3097, 3045, 2933, 1682 (C=$O_{Qnz}$), 1620 (C=O), 1593, 1568, 1487, 1464, 1381, 1236, 1157, 893, 756. APSI MS: $M^+$+1=403. Anal. Calcd for $C_{23}H_{23}ClN_4O_3$: C, 62.94; H, 5.28; Cl, 8.08; N, 12.76. Found: C, 62.80; H, 5.42; Cl, 8.15; N, 12.71.

#### 4.2.34. 2-[4-(2-Hydroxybenzoyl)-1-(tetrahydrofuran-2-ylmethyl)-1H-pyrrol-2-yl]quinazolin-4(3H)-one (**6q**)

Pale yellow solid (730 mg, 88%); mp 245–246 °C (EtOH/DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=1.58 (m, 1H, 4-$H_{THF(A)}$), 1.78 (m, 2H, 4-$H_{THF(B)}$, 3-$H_{THF(A)}$), 1.96 (m, 1H, 3-$H_{THF(B)}$), 3.58 (m, 1H, 2-$H_{THF(A)}$), 3.66 (m, 1H, 2-$H_{THF(B)}$), 4.23 (m, 1H, $NCH_{2(A)}$), 4.59 (m, 1H, $NCH_{2(B)}$), 4.89 (m, 1H, 2-$H_{THF}$), 6.99 (m, 2H, 3,5-$H_{Ar}$), 7.48 (t, $^3J_{HH}$=8.1 Hz, 2H, 4-$H_{Ar}$, 6-$H_{Qnz}$), 7.63 (d, $^3J_{HH}$=8.1 Hz, 1H, 6-$H_{Ar}$), 7.67 (d, $^4J_{HH}$=1.5 Hz, 1H, 2-$H_{pyrrole}$), 7.74–7.84 (m, 3H, 4-$H_{pyrrole}$, 7,8-$H_{Qnz}$), 8.12 (d, $^3J_{HH}$=8.1 Hz, 1H, 5-$H_{Qnz}$), 11.25 (br s, 1H, OH), 12.44 (br s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=25.5 (4-$C_{THF}$), 28.7 (3-$C_{THF}$), 53.4 ($NCH_2$), 67.7 (5-$C_{THF}$), 78.3 (2-$C_{THF}$), 116.6 (4-$C_{pyrrole}$), 117.7 (3-$C_{Ar}$), 119.4 (5-$C_{Ar}$), 121.2 (4a-$C_{Qnz}$), 122.8 (3-$C_{pyrrole}$), 123.5 (1-$C_{Ar}$), 125.9 (5-$C_{Qnz}$), 126.3 (5-$C_{pyrrole}$), 126.8 (6-$C_{Qnz}$), 127.5 (8-$C_{Qnz}$), 131.2 (6-$C_{Ar}$), 134.3 (4-$C_{Ar}$), 135.1 (7-$C_{Qnz}$), 135.8 (2-$C_{pyrrole}$), 146.8 (2-$C_{Qnz}$), 148.7 (8a-$C_{Qnz}$), 159.5 (2-$C_{Ar}$), 162.3 (C=$O_{Qnz}$), 191.6 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3290 (br, OH, NH), 3169, 3113, 3064, 3032, 2974, 2953, 1672 (C=$O_{Qnz}$), 1610 (C=O), 1574, 1539, 1487, 1448, 1344, 1298, 1252, 1228, 1076, 893, 818, 775, 762. APSI MS: $M^+$+1=416. Anal. Calcd for $C_{24}H_{21}N_3O_4$: C, 69.39; H, 5.10; N, 10.11. Found: C, 69.55; H, 4.98; N, 10.04.

#### 4.2.35. 2-[1-Benzyl-4-(2-hydroxybenzoyl)-1H-pyrrol-2-yl]quinazolin-4(3H)-one (**6r**)

Pale yellow solid (834 mg, 99%); mp 259 °C (DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=5.98 (s, 2H, $CH_2$), 6.99 (m, 2H, 3,5-$H_{Ar}$), 7.18 (m, 3H, 2,4,6-$H_{Ph}$), 7.25 (t, $^3J_{HH}$=7.6 Hz, 2H, 3,5-$H_{Ph}$), 7.47 (m, 2H, 4-$H_{Ar}$, 6-$H_{Qnz}$), 7.66 (d, $^3J_{HH}$=8.0 Hz, 1H, 6-$H_{Ar}$), 7.75–7.82 (m, 3H, 2-$H_{pyrrole}$, 7,8-$H_{Qnz}$), 8.05 (d, $^4J_{HH}$=2.0 Hz, 1H, 4-$H_{pyrrole}$), 8.07 (d, $^3J_{HH}$=8.0 Hz, 1H, 5-$H_{Qnz}$), 11.17 (s, 1H, OH), 12.33 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=53.1 ($NCH_2$), 117.2 (4-$C_{pyrrole}$), 117.7 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 121.2 (4a-$C_{Qnz}$), 123.3 (3-$C_{pyrrole}$), 123.8 (1-$C_{Ar}$), 125.5 (5-$C_{Qnz}$), 126.3 (5-$C_{pyrrole}$), 126.8 (6-$C_{Qnz}$), 127.3 (3,5-$C_{Ph}$), 127.5 (8-$C_{Qnz}$), 127.8 (4-$C_{Ph}$), 128.9 (2,6-$C_{Ph}$), 131.2 (6-$C_{Ar}$), 134.3 (4-$C_{Ar}$), 135.1 (7-$C_{Qnz}$), 135.9 (2-$C_{pyrrole}$), 138.9 (1-$C_{Ph}$), 146.2 (2-$C_{Qnz}$), 148.6 (8a-$C_{Qnz}$), 159.3 (2-$C_{Ar}$), 162.2 (C=$O_{Qnz}$), 191.5 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3600–3300 (br, OH, NH), 3176, 3142, 3111, 3037, 2941, 1682 (C=$O_{Qnz}$), 1614 (C=O), 1585, 1547, 1498, 1487, 1462, 1443, 1342, 1302, 1228, 1211, 1155, 893, 812, 756, 725. APSI MS: $M^+$+1=422. Anal. Calcd for $C_{26}H_{19}N_3O_3$: C, 74.10; H, 4.54; N, 9.97. Found: C, 73.94; H, 4.68; N, 10.02.

#### 4.2.36. 7-Chloro-2-[1-ethyl-4-(2-hydroxybenzoyl)-1H-pyrrol-2-yl]quinazolin-4(3H)-one (**6s**)

Pale yellow solid (779 mg, 99%); mp 276 °C (DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=1.39 (t, $^3J_{HH}$=7.2 Hz, 3H, $CH_2CH_3$), 4.65 (q, $^3J_{HH}$=7.2 Hz, 2H, $CH_2CH_3$), 6.98 (m, 2H, 3,5-$H_{Ar}$), 7.45–7.51 (m, 2H, 4,6-$H_{Ar}$), 7.69 (d, $^4J_{HH}$=2.0 Hz, 1H, 8-$H_{Qnz}$), 7.73 (d, $^4J_{HH}$=2.0 Hz, 1H, 2-$H_{pyrrole}$), 7.77 (d, $^3J_{HH}$=8.5 Hz, 1H, 6-$H_{Qnz}$), 7.89 (d, $^4J_{HH}$=2.0 Hz, 1H, 4-$H_{pyrrole}$), 8.09 (d, $^3J_{HH}$=8.5 Hz, 1H, 5-$H_{Qnz}$), 11.25 (s, 1H, OH), 12.41 (br s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=17.4 ($CH_3$), 45.5 ($CH_2$), 117.4 (4-$C_{pyrrole}$), 117.7 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 120.1 (4a-$C_{Qnz}$), 123.1 (3-$C_{pyrrole}$), 123.5 (1-$C_{Ar}$), 125.0 (5-$C_{Qnz}$), 126.7 (5-$C_{pyrrole}$), 126.9 (6-$C_{Qnz}$), 128.4 (8-$C_{Qnz}$), 131.2 (6-$C_{Ar}$), 134.4 (4-$C_{Ar}$), 135.1 (2-$C_{pyrrole}$), 139.6 (7-$C_{Qnz}$), 147.6 (2-$C_{Qnz}$), 150.1 (8a-$C_{Qnz}$), 159.5 (2-$C_{Ar}$), 161.7 (C=$O_{Qnz}$), 191.5 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3280 (br, OH, NH), 3116, 3028, 2985, 2926, 1672 (C=$O_{Qnz}$), 1624 (C=O), 1608, 1578, 1487, 1446, 1225, 1159, 891, 760. APSI MS: $M^+$+1=394. Anal. Calcd for $C_{21}H_{16}ClN_3O_3$: C, 64.05; H, 4.10; Cl, 9.00; N, 10.67. Found: C, 64.18; H, 3.98; Cl, 9.07; N, 10.62.

#### 4.2.37. 2-[1-Ethyl-4-(2-hydroxybenzoyl)-1H-pyrrol-2-yl]-6,7-dimethoxyquinazolin-4(3H)-one (**6t**)

Beige solid (687 mg, 82%); mp 287 °C (EtOH/DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=1.39 (t, $^3J_{HH}$=7.0 Hz, 3H, $CH_2CH_3$), 3.88 (s, 3H, $OCH_3$), 3.92 (s, 3H, $OCH_3$), 4.68 (q, $^3J_{HH}$=7.0 Hz, 2H, $CH_2CH_3$), 6.98 (m, 2H, 3,5-$H_{Ar}$), 7.11 (s, 1H, 8-$H_{Qnz}$), 7.46 (s, 1H, 5-$H_{Qnz}$), 7.48 (t, $^3J_{HH}$=7.8 Hz, 1H, 4-$H_{Ar}$), 7.64 (d, $^4J_{HH}$=1.3 Hz, 1H, 2-$H_{pyrrole}$), 7.82 (d, $^3J_{HH}$=7.8 Hz, 1H, 6-$H_{Ar}$), 7.85 (d, $^4J_{HH}$=1.3 Hz, 1H, 4-$H_{pyrrole}$), 11.34 (br s, 1H, OH), 12.31 (br s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=17.4 ($CH_2CH_3$), 45.1 ($CH_2$), 56.2 ($OCH_3$), 56.5 ($OCH_3$), 105.6 (4a-$C_{Qnz}$), 108.4 (8-$C_{Qnz}$), 114.2 (5-$C_{Qnz}$), 116.1 (4-$C_{pyrrole}$), 117.7 (3-$C_{Ar}$), 119.4 (5-$C_{Ar}$), 122.9 (3-$C_{pyrrole}$), 123.4 (1-$C_{Ar}$), 125.6 (5-$C_{pyrrole}$), 131.3 (6-$C_{Ar}$), 134.2 (2-$C_{pyrrole}$), 134.4 (4-$C_{Ar}$), 136.5 (2-$C_{Qnz}$), 144.9 (6-$C_{Qnz}$), 149.0 (8a-$C_{Qnz}$), 155.2 (7-$C_{Qnz}$), 159.6 (2-$C_{Ar}$), 162.3 (C=$O_{Qnz}$), 191.6 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3300 (br, OH, NH), 3130, 3082, 3062, 3012, 2958, 1670 (C=$O_{Qnz}$), 1633 (C=O), 1608, 1497, 1431, 1390, 1281, 1238, 1221, 1159, 1101, 1003, 893, 814, 766. APSI MS: $M^+$+1=420. Anal. Calcd for $C_{23}H_{21}N_3O_5$: C, 65.86; H, 5.05; N, 10.02. Found: C, 65.99; H, 4.90; N, 9.96.

#### 4.2.38. 2-[4-(2-Hydroxybenzoyl)-1-methyl-1H-pyrrol-2-yl]-thieno[3,2-d]pyrimidin-4(3H)-one (**6u**)

Beige solid (681 mg, 97%); mp 275–276 °C (DMF). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=4.08 (s, 3H, $NCH_3$), 6.97 (t, $^3J_{HH}$=8.4 Hz, 1H, 5-$H_{Ar}$), 6.98 (d, $^3J_{HH}$=8.4 Hz, 1H, 3-$H_{Ar}$), 7.39 (d, $^3J_{HH}$=5.2 Hz, 1H, 4-$H_{thiophene}$), 7.46 (t, $^3J_{HH}$=8.4 Hz, 1H, 4-$H_{Ar}$), 7.62 (d, $^4J_{HH}$=1.4 Hz, 1H, 2-$H_{pyrrole}$), 7.78 (d, $^3J_{HH}$=8.4 Hz, 1H, 6-$H_{Ar}$), 7.79 (d, $^4J_{HH}$=1.4 Hz, 1H, 4-$H_{pyrrole}$), 8.18 (d, $^3J_{HH}$=5.2 Hz, 1H, 5-$H_{thiophene}$), 11.26 (s, 1H, OH), 12.54 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=38.5 ($CH_3$), 116.3 (4-$C_{pyrrole}$), 117.7 (3-$C_{Ar}$), 119.4 (5-$C_{Ar}$), 121.1 (4a-$C_{ThP}$), 122.7 (3-$C_{pyrrole}$), 123.4 (1-$C_{Ar}$), 125.8 (5-$C_{pyrrole}$), 126.2 (7-$C_{ThP}$), 131.2 (6-$C_{Ar}$), 134.3 (4-$C_{Ar}$), 135.6 (6-$C_{ThP}$), 135.7 (2-$C_{pyrrole}$), 148.4 (2-$C_{ThP}$), 157.8 (7a-$C_{ThP}$), 158.7 (C=$O_{ThP}$), 159.5 (2-$C_{Ar}$), 191.6 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3280 (br, OH), 3126, 3088, 3034, 3003, 2951, 1655 (C=$O_{ThP}$), 1626 (C=O), 1583, 1500, 1485, 1290, 1248, 1213, 1159, 889, 812, 789, 756, 735, 669. APSI MS: $M^+$+1=352. Anal. Calcd for $C_{18}H_{13}N_3O_3S$: C, 61.53; H, 3.73; N, 11.96; S, 9.12. Found: C, 61.65; H, 3.64; N, 11.89; S, 9.20.

#### 4.2.39. 4-(2-Hydroxybenzoyl)-1H-pyrrole-2-carboxylic acid (**8a**)

Green solid (254 mg, 55%); mp 251–252 °C (EtOH). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=6.91–6.99 (m, 2H, 3,5-$H_{Ar}$), 7.08 (s, 1H, 5-$H_{pyrrole}$), 7.43 (td, $^3J_{HH}$=7.8 Hz, $^4J_{HH}$=1.6 Hz, 1H, 4-$H_{Ar}$), 7.50 (s, 1H, 3-$H_{pyrrole}$), 7.61 (dd, $^3J_{HH}$=7.8 Hz, $^4J_{HH}$=1.6 Hz, 1H, 6-$H_{Ar}$), 10.99 (1H, s, OH), 12.54 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=116.0 (3-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 124.2 (1-$C_{Ar}$), 125.1 (4-$C_{pyrrole}$), 125.3 (2-$C_{pyrrole}$), 129.5 (5-$C_{pyrrole}$), 130.9 (6-$C_{Ar}$), 134.0 (4-$C_{Ar}$), 158.8 (2-$C_{Ar}$), 162.0 (COOH), 192.0 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3250 (br, OH), 3269 (br, OH), 3124, 2993, 1714 (C=$O_{acid}$), 1645 (C=O), 1595, 1556, 1487, 1340, 1288, 1259, 1209, 1190, 1157, 895, 812, 758, 704, 675. APSI MS: $M^+$+1=232, $M^+$−1=230. Anal. Calcd for $C_{12}H_9NO_4$: C, 62.34; H, 3.92; N, 6.06. Found: C, 62.21; H, 4.05; N, 6.15.

#### 4.2.40. Methyl 4-(2-hydroxybenzoyl)-1-methyl-1H-pyrrole-2-carboxylate (**8b**)

Pale yellow crystals (223 mg, 43%); mp 116–117 °C (*i*-PrOH/hexane). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=3.76 (s, 3H, $OCH_3$), 3.91 (s, 3H, $NCH_3$), 6.89–7.00 (m, 2H, 3,5-$H_{Ar}$), 7.16 (d, $^4J_{HH}$=1.2 Hz, 1H,

5-$H_{pyrrole}$), 7.43 (t, $^3J_{HH}$=7.9 Hz, 1H, 4-$H_{Ar}$), 7.58 (d, $^3J_{HH}$=7.9 Hz, 1H, 6-$H_{Ar}$), 7.77 (d, $^4J_{HH}$=1.2 Hz, 1H, 3-$H_{pyrrole}$), 10.91 (s, 1H, OH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=37.5 ($NCH_3$), 51.9 ($OCH_3$), 117.6 (3-$C_{Ar}$), 118.7 (3-$C_{pyrrole}$), 119.5 (5-$C_{Ar}$), 122.9 (4-$C_{pyrrole}$), 123.7 (2-$C_{pyrrole}$), 124.2 (1-$C_{Ar}$), 130.7 (6-$C_{Ar}$), 134.0 (4-$C_{Ar}$), 135.5 (5-$C_{pyrrole}$), 158.7 (2-$C_{Ar}$), 161.0 ($COOCH_3$), 191.4 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3300 (br, OH), 3126, 2999, 2956, 1707 (C=$O_{ester}$), 1622 (C=O), 1581, 1537, 1489, 1446, 1294, 1250, 1198, 1157, 1099, 893, 823, 758, 704, 667. APSI MS: $M^+$+1=260. Anal. Calcd for $C_{14}H_{13}NO_4$: C, 64.86; H, 5.05; N, 5.40. Found: C, 64.95; H, 4.98; N, 5.47.

#### 4.2.41. 4-(2-Hydroxybenzoyl)-N,N-dimethyl-1H-pyrrole-2-carboxamide (**8c**)

Light green solid (351 mg, 68%); mp 146–147 °C (EtOH/MeCN). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=3.10 (m, 6H, $N(CH_3)_2$), 6.89–6.98 (m, 3H, 3,5-$H_{Ar}$, 5-$H_{pyrrole}$), 7.40 (d, $^4J_{HH}$=1.7 Hz, 1H, 3-$H_{pyrrole}$), 7.42 (t, $^3J_{HH}$=7.8 Hz, 1H, 4-$H_{Ar}$), 7.64 (d, $^3J_{HH}$=7.8 Hz, 1H, 6-$H_{Ar}$), 11.06 (s, 1H, OH), 12.21 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=36.3 $(NCH_3)_2$, 113.2 (3-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 124.0 (1-$C_{Ar}$), 124.5 (4-$C_{pyrrole}$), 127.0 (2-$C_{pyrrole}$), 128.1 (5-$C_{pyrrole}$), 131.0 (6-$C_{Ar}$), 134.0 (4-$C_{Ar}$), 159.1 (2-$C_{Ar}$), 161.8 (CON), 192.2 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3300 (br, OH, NH), 3170, 3145, 3041, 2981, 2935, 1622 (C=O), 1601, 1587, 1564, 1481, 1385, 1352, 1271, 1163, 901, 760, 696, 669. APSI MS: $M^+$+1=259. Anal. Calcd for $C_{14}H_{14}N_2O_3$: C, 65.11; H, 5.46; N, 10.85. Found: C, 65.25; H, 5.36; N, 10.79.

#### 4.2.42. (2-Hydroxyphenyl)[5-(pyrrolidin-1-ylcarbonyl)-1H-pyrrol-3-yl]methanone (**8d**)

Light green solid (346 mg, 61%); mp 179–180 °C (EtOH/MeCN). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=1.82 (m, 2H, $CH_2$), 1.95 (m, 2H, $CH_2$), 3.49 (m, 2H, $NCH_2$), 3.72 (m, 2H, $NCH_2$), 6.88–7.03 (m, 3H, 3,5-$H_{Ar}$, 5-$H_{pyrrole}$), 7.40 (d, $^4J_{HH}$=1.5 Hz, 1H, 3-$H_{pyrrole}$), 7.43 (t, $^3J_{HH}$=8.0 Hz, 1H, 4-$H_{Ar}$), 7.63 (d, $^3J_{HH}$=8.0 Hz, 1H, 6-$H_{Ar}$), 11.04 (s, 1H, OH), 12.23 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=23.9 ($CH_2$), 26.7 ($CH_2$), 47.3 ($NCH_2$), 48.2 ($NCH_2$), 112.8 (3-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.5 (5-$C_{Ar}$), 124.2 (1-$C_{Ar}$), 124.9 (4-$C_{pyrrole}$), 127.9 (2-$C_{pyrrole}$), 128.3 (5-$C_{pyrrole}$), 130.9 (6-$C_{Ar}$), 133.9 (4-$C_{Ar}$), 158.9 (2-$C_{Ar}$), 159.5 (CON), 192.1 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3415 (br, OH), 3209 (br, NH), 2981, 2968, 1626 (C=O), 1589 (C=O), 1552, 1487, 1454, 1385, 1354, 1213, 1146, 895, 812, 754, 704. APSI MS: $M^+$+1=285. Anal. Calcd for $C_{16}H_{16}N_2O_3$: C, 67.59; H, 5.67; N, 9.85. Found: C, 67.73; H, 5.55; N, 9.78.

#### 4.2.43. N,N-Diethyl-4-(2-hydroxybenzoyl)-1-methyl-1H-pyrrole-2-carboxamide (**8e**)

Pale yellow solid (270 mg, 45%); mp 84 °C (*i*-PrOH/hexane). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=1.14 (t, $^3J_{HH}$=7.0 Hz, 6H, $2CH_2\mathit{CH}_3$), 3.45 (q, $^3J_{HH}$=7.0 Hz, 4H, $2\mathit{CH}_2CH_3$), 3.69 (s, 3H, $NCH_3$), 6.69 (d, $^4J_{HH}$=1.4 Hz, 1H, 5-$H_{pyrrole}$), 6.91–7.00 (m, 2H, 3,5-$H_{Ar}$), 7.43 (t, $^3J_{HH}$=8.0 Hz, 1H, 4-$H_{Ar}$), 7.58 (d, $^4J_{HH}$=1.4 Hz, 1H, 3-$H_{pyrrole}$), 7.68 (d, $^3J_{HH}$=8.0 Hz, 1H, 6-$H_{Ar}$), 11.10 (s, 1H, OH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=14.1 ($CH_2\mathit{CH}_3$), 36.0 ($NCH_3$), 39.5 ($\mathit{CH}_2CH_3$), 110.9 (3-$C_{pyrrole}$), 117.6 (3-$C_{Ar}$), 119.4 (5-$C_{Ar}$), 122.1 (4-$C_{pyrrole}$), 124.0 (1-$C_{Ar}$), 128.6 (5-$C_{pyrrole}$), 130.9 (6-$C_{Ar}$), 132.5 (2-$C_{pyrrole}$), 134.0 (4-$C_{Ar}$), 159.1 (2-$C_{Ar}$), 162.3 (CON), 191.6 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3200 (br, OH), 3140, 2970, 2933, 1624 (C=O), 1591 (C=O), 1549, 1483, 1425, 1282, 1219, 1157, 897, 766. APSI MS: $M^+$+1=301. Anal. Calcd for $C_{17}H_{20}N_2O_3$: C, 67.98; H, 6.71; N, 9.33. Found: C, 68.10; H, 6.60; N, 9.27.

#### 4.2.44. 1,3-Dimethyl-2-(4-oxo-4H-chromen-3-yl)imidazolidin-4-one (**9a**)

Colorless crystals (408 mg, 79%); mp 195–196 °C (EtOH). $^1$H NMR (500 MHz, DMSO-$d_6$): $\delta$=2.49 (s, 3H, $NCH_3$), 2.63 (s, 3H, $CONCH_3$), 3.54 (m, 1H, $CH_{2(A)}$), 3.84 (m, 1H, $CH_{2(B)}$), 5.47 (m, 1H, CH), 7.55 (t, $^3J_{HH}$=8.5 Hz, 1H, 6-$H_{Chr}$), 7.73 (d, $^3J_{HH}$=8.5 Hz, 1H, 8-$H_{Chr}$), 7.87 (t, $^3J_{HH}$=8.5 Hz, 1H, 7-$H_{Chr}$), 8.09 (d, $^3J_{HH}$=8.5 Hz, 1H, 5-$H_{Chr}$), 8.61 (s, 1H, 2-$H_{Chr}$). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=27.4 ($CONCH_3$), 40.5 ($NCH_3$), 54.8 ($CH_2$), 77.3 (CH), 114.2 (3-$C_{Chr}$), 119.2 (8-$C_{Chr}$), 123.8 (4a-$C_{Chr}$), 125.6 (5-$C_{Chr}$), 126.8 (6-$C_{Chr}$), 135.6 (7-$C_{Chr}$), 156.3 (2-$C_{Chr}$), 160.9 (8a-$C_{Chr}$), 166.7 ($CONCH_3$), 176.3 (C=$O_{Chr}$). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3055, 3022, 2980, 2933, 1726 (C=O), 1647 (C=$O_{Chr}$), 1572, 1464, 1396, 1363, 1317, 1128, 1012, 939, 885, 862, 758, 698. APSI MS: $M^+$+1=259. Anal. Calcd for $C_{14}H_{14}N_2O_3$: C, 65.11; H, 5.46; N, 10.85. Found: C, 65.22; H, 5.33; N, 10.89.

#### 4.2.45. (7aS)-3-(4-Oxo-4H-chromen-3-yl)hexahydro-1H-pyrrolo[1,2-c]imidazol-1-one hydrochloride (**9b**)

Beige solid (515 mg, 84%); mp 232–233 °C (MeCN). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=1.97 (m, 2H, 7-H), 2.10 (m, 1H, 6-$H_A$), 2.21 (m, 1H, 6-$H_B$), 3.66 (m, 2H, 5-H), 4.57 (m, 1H, 7a-H), 5.88 (m, 1H, 3-H), 7.59 (t, $^3J_{HH}$=8.6 Hz, 1H, 6-$H_{Chr}$), 7.76 (d, $^3J_{HH}$=8.6 Hz, 1H, 8-$H_{Chr}$), 7.90 (t, $^3J_{HH}$=8.6 Hz, 1H, 7-$H_{Chr}$), 8.11 (d, $^3J_{HH}$=8.6 Hz, 1H, 5-$H_{Chr}$), 8.61 (s, 1H, 2-$H_{Chr}$), 9.30 (s, 1H, NH), 12.37 (br s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$, *signals of minor diastereomer): $\delta$=24.6* and 24.9 (6-C), 26.8* and 27.7 (7-C), 57.0* and 57.7 (5-C), 64.3 and 65.4* (7a-C), 71.4* and 72.9 (3-C), 115.0* and 115.1 (3-$C_{Chr}$), 119.1 and 119.2* (8-$C_{Chr}$), 123.7 and 123.8* (4a-$C_{Chr}$), 125.5* and 125.6 (5-$C_{Chr}$), 126.8 and 126.9* (6-$C_{Chr}$), 135.6 and 135.8* (7-$C_{Chr}$), 156.2 and 156.4* (2-$C_{Chr}$), 157.8 and 157.9* (8a-$C_{Chr}$), 170.8* and 170.9 (CON), 175.7* and 175.9 (C=$O_{Chr}$). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3448 (br, NH), 3055, 2989, 2943, 1732 (C=O), 1645 (C=$O_{Chr}$), 1624, 1608, 1460, 1402, 1350, 1309, 1182, 1165, 914, 795, 773. APSI MS: $M^+$+1=271. Anal. Calcd for $C_{15}H_{15}ClN_2O_3$: C, 58.73; H, 4.93; Cl, 11.56; N, 9.13. Found: C, 58.87; H, 4.80; Cl, 11.48; N, 9.19.

#### 4.2.46. 2-(2-Hydroxybenzoyl)pyrrolo[1,2-a]quinoxalin-4(5H)-one (**11a**)

Beige solid (316 mg, 52%); mp>300 °C (EtOH+DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=7.00 (m, 2H, 3,5-$H_{Ar}$), 7.22 (t, $^3J_{HH}$=8.3 Hz, 1H, 7-$H_{Qnx}$), 7.29 (d, $^4J_{HH}$=1.5 Hz, 1H, 5-$H_{pyrrole}$), 7.30–7.39 (m, 2H, 6,8-$H_{Qnx}$), 7.49 (t, $^3J_{HH}$=7.6 Hz, 1H, 4-$H_{Ar}$), 7.69 (d, $^3J_{HH}$=7.6 Hz, 1H, 6-$H_{Ar}$), 8.26 (d, $^3J_{HH}$=8.3 Hz, 1H, 5-$H_{Qnx}$), 8.75 (d, $^4J_{HH}$=1.5 Hz, 1H, 3-$H_{pyrrole}$), 10.90 (s, 1H, OH), 11.47 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=112.8 (8-$C_{Qnx}$), 116.5 (3-$C_{pyrrole}$), 117.2 (5-$C_{Qnx}$), 117.7 (3-$C_{Ar}$), 119.7 (5-$C_{Ar}$), 122.47 (4-$C_{pyrrole}$), 122.49 (8a-$C_{Qnx}$), 123.3 (5-$C_{pyrrole}$), 124.3 (1-$C_{Ar}$), 124.7 (4a-$C_{Qnx}$), 127.0 (2-$C_{pyrrole}$), 127.5 (6-$C_{Qnx}$), 129.6 (7-$C_{Qnx}$), 131.3 (6-$C_{Ar}$), 134.4 (4-$C_{Ar}$), 155.4 (CONH), 158.8 (2-$C_{Ar}$), 192.3 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3650–3320 (br, OH, NH), 3132, 3035, 2983, 2924, 2900, 1674 (C=O), 1626 (C=O), 1595, 1545, 1516, 1483, 1379, 1338, 1298, 1252, 1223, 1157, 895, 816, 754, 739. APSI MS: $M^+$+1=305. Anal. Calcd for $C_{18}H_{12}N_2O_3$: C, 71.05; H, 3.97; N, 9.21. Found: C, 71.18; H, 3.86; N, 9.15.

#### 4.2.47. 2-(2-Hydroxybenzoyl)-7-(piperidin-1-ylsulfonyl)pyrrolo[1,2-a]quinoxalin-4(5H)-one (**11b**)

White solid (523 mg, 58%); mp>300 °C (EtOH+DMF). $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$=1.36 (m, 2H, $CH_2$), 1.55 (m, 4H, $2NCH_2\mathit{CH}_2$), 2.83 (m, 1H, NCH), 2.92 (m, 3H, NCH), 7.01 (m, 2H, 3,5-$H_{Ar}$), 7.34 (d, $^4J_{HH}$=1.3 Hz, 1H, 5-$H_{pyrrole}$), 7.50 (m, 2H, 4-$H_{Ar}$, 8-$H_{Qnx}$), 7.67 (m, 2H, 6-$H_{Ar}$, 6-$H_{Qnx}$), 8.51 (d, $^3J_{HH}$=8.3 Hz, 1H, 5-$H_{Qnx}$), 8.84 (d, $^4J_{HH}$=1.3 Hz, 1H, 3-$H_{pyrrole}$), 10.84 (s, 1H, OH), 11.63 (s, 1H, NH). $^{13}$C NMR (125 MHz, DMSO-$d_6$): $\delta$=23.3 ($CH_2$), 25.2 ($NCH_2\mathit{CH}_2$), 47.1 ($NCH_2$), 113.5 (8-$C_{Qnx}$), 116.3 (3-$C_{pyrrole}$), 117.5 (5-$C_{Qnx}$), 117.7 (3-$C_{Ar}$), 119.7 (5-$C_{Ar}$), 121.9 (4-$C_{pyrrole}$), 123.4 (5-$C_{pyrrole}$), 124.3 (1-$C_{Ar}$), 124.8 (6-$C_{Qnx}$), 125.6 (8a-$C_{Qnx}$), 127.7 (2-$C_{pyrrole}$), 130.2 (7-$C_{Qnx}$), 131.3 (6-$C_{Ar}$), 134.1 (4a-$C_{Qnx}$), 134.5 (4-$C_{Ar}$), 155.1 (CONH), 158.7 (2-$C_{Ar}$), 192.1 (C=O). IR (KBr), $\nu_{max}$ ($cm^{-1}$): 3670–3300 (br, OH, NH), 3142, 3074, 2941, 1701 (C=O), 1668, 1620 (C=O), 1587, 1485, 1360, 1333 ($SO_2$), 1163 ($SO_2$), 1092, 1053, 931, 893, 816, 758, 715, 604. APSI MS:

$M^+$+1=452. Anal. Calcd for $C_{23}H_{21}N_3O_5S$: C, 61.19; H, 4.69; N, 9.31; S, 7.10. Found: C, 61.37; H, 4.57; N, 9.24; S, 7.18.

## Acknowledgements

The authors acknowledge V.V. Polovinko (Enamine Ltd) and Dr. S.A. Alekseev (Kyiv National Taras Shevchenko University) for spectral measurements.

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