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Syntheses of indolo[1,2-*a*]quinazolinone derivatives *via* palladium catalyzed intramolecular C–H amidation†

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The synthesis of indolo[1,2-*a*]quinazolinone starting from 2-iodobenzamide and indole derivatives is reported. In this two-step procedure, indole is *N*-arylated with 2-iodobenzamide derivatives *via* Ullman coupling, followed by an intramolecular C–H amidation using a palladium catalyst.

Introduction

Quinazolinone,[1] and indole,[2] two important structural motifs that are found in many natural and synthetic compounds, exhibit a wide variety of biological activities. Indole fused quinazolinone compounds such as indoloquinazolinone derivatives show particularly promising biological activities, including antibacterial, anticancer and antifungal activities.[3] In addition, certain types of indolo[1,2-*a*]quinazolinone structures are found in selective protein kinase CK2 inhibitors.[4] Indoloquinazolinone derivatives are generally classified into three types, based on structural arrangements namely indolo[1,2-*a*]quinazolinone (A), indolo[1,2-*b*]quinazolinone (B) and indolo[1,2-*c*]quinazolinone (C) derivatives (Fig. 1). Many synthetic protocols available for the synthesis of indolo[1,2-*b*]quinazolinone,[5] indolo[1,2-*c*]-quinazolinone derivatives,[6] but only a very few methods are available for the synthesis of indolo[1,2-*a*]quinazolinone derivatives (Scheme 1).[4]

Furet and co-workers reported on the synthesis of such derivatives by the rearrangement of a substituted 11-oxo-10,11-dihydro-5*H*-dibenzo[*b*,*f*]azepine-10-carbonitrile under basic conditions.[4a] Moore *et al.* reported a synthesis of indolo[1,2-*a*]-quinazolinone derivatives in three steps starting from anthranilamide (Scheme 1).[4c] Both of the above cited methods involve the use of complex starting materials or involve multi-steps. Hence, a great demand exists for a straightforward, convenient procedure for the synthesis of indolo[1,2-*a*]quinazolinone derivatives.

A copper catalyzed C–N bond forming reaction is frequently used in the synthesis of various N-heterocycles.[7] In our recent research, we focused our attention on copper catalyzed C–C and C–N bond formation reactions of 2-iodobenzamide derivatives for use in the construction of various heterocycles, including 2-aminophenylbenzoxazoles,[8a] isocoumarin derivatives[8b] and fused sultam derivatives.[8c] In a continuation of this effort, we wish to report on the synthesis of indolo[1,2-*a*]quinazolinone derivatives from 2-iodobenzamide derivatives. Our strategy for this synthesis involves the *N*-arylation of indole derivatives under Ullman conditions to give 2-indolylbenzamide derivatives followed by C–H amidation. In fact, many well-developed methods are available for the *N*-arylation of indoles and most of them utilized catalytic amount of Cu.[9] However, majority of the reported procedures utilize either ligands, higher temperatures or strong base to achieve this transformation. Moreover, we recently found that the *N*-arylation of indole with a substrate containing an *N*-phenylbenzamide group proceeded in the

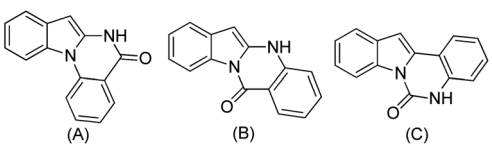
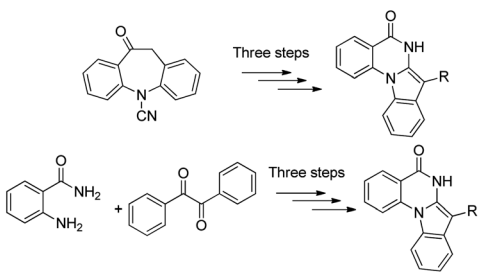


Fig. 1 Types of indoloquinazolinone derivatives.



Scheme 1 Previously reported procedures for the synthesis of indolo [1,2-*a*]quinazolinone derivatives.

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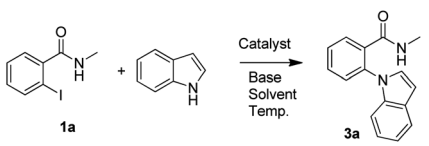
absence of a ligand in the presence of a catalytic amount of copper iodide. In addition, the reaction proceeded at comparatively lower temperature than the literature procedures.[8a] The reason for this efficient reaction in absence of a ligand can be attributed to the ortho effect caused by the *N*-phenylamide.[10] On the other hand, the Fu and Bao groups reported on the synthesis of azoquinazolinone derivatives by Cu(I)-catalyzed C–N bond coupling/C–H activation/C–N bond forming reactions in an atmosphere of O_2.[11] Furthermore, Thasana and coworkers have reported an interesting intramolecular palladium catalyzed C–H amidation reaction for the synthesis of indolobenzazocin-8-one derivatives.[12] Based on these observations, we envisioned that an indolo[1,2-*a*]quinazolinone derivative could be generated from the reaction of 2-iodobenzamide derivatives and indole derivatives in the presence of a copper(I) catalyst and a suitable oxidant.

Results and discussion

To test our assumption, we initially investigated the reaction of 2-iodo-*N*-methylbenzamide with indole in the presence of copper iodide (20 mol%) and potassium carbonate (2 equiv.) in DMSO under an oxygen atmosphere at 80 °C.[9] Under these conditions, we obtained 2-(1*H*-indol-1-yl)-*N*-methylbenzamide exclusively and no trace of indoloquinazolinone derivative was found in the reaction mixture. The course of the reaction was the same, even when it was carried out at higher temperatures (*i.e.* at 100 °C and 120 °C). As Fu and co-workers achieved indoloimidazole derivatives in one-pot using copper iodide in the presence of L-proline, we utilized the same condition in our reaction. Under these conditions, the reaction produced 2-(1*H*-indol-1-yl)-*N*-methylbenzamide only. Also, we utilized Bao and co-workers conditions for this reaction using copper iodide, 1,10-phenanthroline and sodium ethoxide. Under these conditions, the reaction produced mixture of products. Since we failed to obtain the indolo[1,2-*a*]quinazolinone derivative in one pot reactions in the presence of copper catalysts, we further pursued the reaction in the presence palladium catalysts like palladium acetate and palladium chloride. To our disappointment, the reactions produce mixture of unidentified products in both these cases. Hence, we decided to synthesize the indolo-[1,2-*a*]quinazolinone derivatives in two step process by isolating

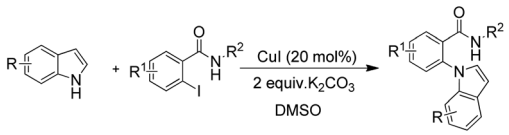
Table 1 Optimization of the formation of 2-(1*H*-indol-1-yl)-*N*-methylbenzamide derivatives

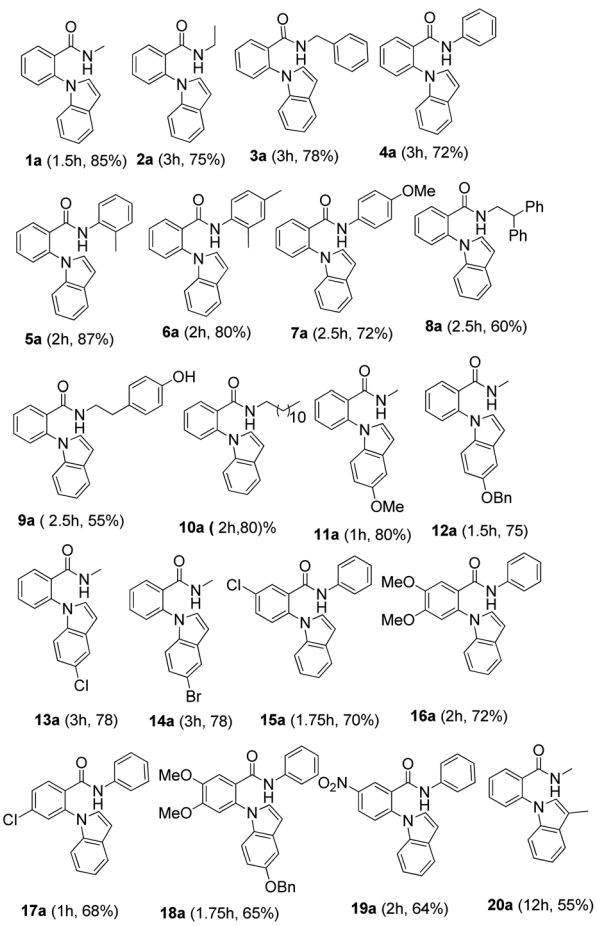


Entry[a]	Catalyst	Solvent	Base	Temp °C	Time (h)	Yield[b] (%)
1[c]	CuI	DMSO	K_2CO_3	80	1.5	82
2[c]	CuI	DMSO	K_2CO_3	100	1	80
3[c]	CuI	DMSO	K_2CO_3	120	0.5	76
4[c]	CuI	DMSO	Cs_2CO_3	120	24	60
5[c,d]	CuI	DMF	NaOEt	120	24	—
6[c,d]	$PdCl_2$	DMF	NaOEt	120	24	—
7[c,d]	$Pd(OAc)_2$	DMF	NaOEt	120	24	—
8	CuI	DMSO	K_2CO_3	80	1.5	85
9	CuI	DMSO	K_2CO_3	80	1.5	85
10	CuBr	DMSO	K_2CO_3	80	2	83
11	CuCl	DMSO	K_2CO_3	80	2	84
12	CuI	Toluene	K_2CO_3	80	24	36
13	CuI	CH_3CN	K_2CO_3	80	5	76
14	CuI	Dioxane	K_2CO_3	80	8	45
15	CuI	EtOH	K_2CO_3	80	24	50
16	CuI	DMSO	K_3PO_4	80	24	70
17	CuI	DMSO	Cs_2CO_3	80	1.5	86

[a] All reactions were carried out with **1a** (0.5 mmol), base (2 equiv.) DMSO (1.5 mL) and 20 mol% copper(I) salt. [b] Isolated yields. [c] Reactions performed in O_2 atmosphere. [d] Mixture of unidentified products.

Table 2 Synthesis of 2-(1*H*-indol-1-yl)-*N*-substituted benzamide derivatives[a]





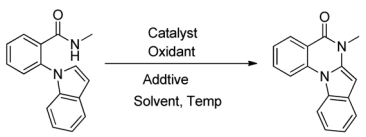
[a] All reactions were carried out on a 1 mmol scale. The yields reported here are corresponds to isolated.

2-(1*H*-indol-1-yl)-*N*-substituted benzamide derivatives, followed by applying suitable conditions for C–N bond formation through C–H activation, in a second step.

To accomplish this, we first synthesized various substituted 2-(1*H*-indol-1-yl)-*N*-substituted benzamide derivatives using copper catalysis. In our earlier experiments, we found that DMSO, copper iodide and potassium carbonate at 80 °C gave the best results for *N*-arylation.[8a] Hence, we investigated the reaction of 2-iodo-*N*-methylbenzamide and indole in the presence of copper iodide and potassium carbonate in DMSO at 80 °C. Under these conditions, the desired 2-(1*H*-indol-1-yl)-*N*-substituted benzamide was produced in 85% yield. To test whether any other solvent could be used in this reaction, we ran the reaction in different solvents including toluene, 1,4-dioxane and acetonitrile. However, the reaction was incomplete, when toluene and 1,4-dioxane used as solvents and a longer reaction time was needed when acetonitrile was used as the solvent. We also examined different bases such as cesium carbonate, potassium carbonate and potassium phosphate. The reaction was equally efficient, when cesium carbonate or potassium carbonates were used but the use of potassium phosphate resulted in a low product yield. The use of a copper iodide catalyst in DMSO at 80 °C in the presence of potassium carbonate or cesium carbonate gave the best results. However, considering the cost of cesium carbonate, we used potassium carbonate as the base in this reaction (Table 1).

With the optimized reaction condition in hand, we synthesized a diverse array of 2-(1*H*-indol-1-yl)-*N*-substituted benzamide derivatives *via* the reaction of various 2-iodo-*N*-substituted benzamides and indole derivatives using the optimized reaction conditions. The results were summarized in Table 2. As depicted in Table 2, we first synthesized various 2-(1*H*-indol-1-yl)-*N*-substituted benzamide derivatives by reacting various 2-iodo-benzamide generated from 2-iodo-benzoylchloride and various amines with unsubstituted indoles. The reaction of 2-iodo-*N*-methylbenzamide with indole furnished the corresponding 2-(1*H*-indol-1-yl)-*N*-methylbenzamide (**1a**) in excellent yield. However, when ethyl, benzyl amides were used as substrates, the product yields (**2a**, **3a**) were decreased slightly. On the other hand, the reaction of indole with 2-iodo-*N*-phenylbenzamide derivatives provided the desired products (**4a–7a**) in good yields. Further, the reaction of 2-iodo-*N*-substituted benzamide derived from sterically hindered amine such as 2,2-diphenylethylamine and indole also provided the corresponding product (**8a**). However, the yield of product moderate (60%) in this case. Moreover, the reaction of 2-iodo-*N*-susbtituted benzamide generated from tyramine and indole produced poor yield of corresponding product (**9a**). We next investigated the reactions of 2-iodo-*N*-methylbenzamide and substituted indole derivatives to synthesize the corresponding 2-(1*H*-indol-1-yl)-*N*-methylbenzamide derivatives. It is noteworthy that the reaction of 2-iodo-*N*-methylbenzamide with an indole that contained electron-donating groups produced good yields of the corresponding 2-(1*H*-indol-1-yl)-*N*-methylbenzamide derivatives (**11a** and **12a**) in a relatively short period of time. Indoles bearing electron withdrawing groups, however, gave the corresponding product in moderate yields (**13a**, **14a**). Next, we investigated the reactions of various substituted 2-iodo-*N*-phenylbenzamide and indole by using the optimized reaction conditions. As seen in Table 2, the reaction of 2-iodobenzamide derivatives that contain both electron withdrawing and electron-donating groups reacted with equal ease to produce the corresponding 2-(1*H*-indol-1-yl)-*N*-phenylbenzamides (**15a**,

Table 3 Optimization studies for the synthesis of 2-(1*H*-indol-1-yl)-*N*-methylbenzamide derivatives



Entry[a]	Catalyst	Oxidant	Additives	Solvents	Temp (°C)	Time (h)	Yield[b] (%)
1	$Pd(OAc)_2$	$Cu(OAc)_2$	Acetic acid	Toluene	120	16	30
2	$Pd(OAc)_2$	$Cu(OAc)_2$	Acetic acid	Toluene	100	24	10
3	$Pd(OAc)_2$	$Cu(OAc)_2$	PTSA	Toluene	100	24	44
4	$Pd(OAc)_2$	$Cu(OAc)_2$	TFA	Toluene	100	8	54
5	$Pd(OAc)_2$	$Mn(OAc)_4$	TFA	Toluene	100	24	25
6	$Pd(OAc)_2$	$PhI(OAc)_2$	TFA	Toluene	100	24	20
7	$Pd(OAc)_2$	BQ	TFA	Toluene	100	36	30
8	$Pd(OAc)_2$	AgOAc	TFA	Toluene	100	8	54
9	$PdCl_2$	AgOAc	TFA	Toluene	100	20	48
10	$PdCl_2(PPh_3)_2$	AgOAc	TFA	1,2-Dichloroethane	80	2.30	35
11	$PdCl_2(PPh_3)_2$	AgOAc	TFA	1,4-Dioxane	100	12	56
12	$PdCl_2(PPh_3)_2$	AgOAc	TFA	DMSO	100	12	NR
13	$PdCl_2(PPh_3)_2$	AgOAc	TFA	Toluene	100	6	80
14[c]	$PdCl_2(PPh_3)_2$	AgOac	TFA	Toluene	100	6	88

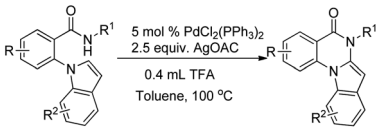
[a] All reactions were conducted at 0.5 mmol scale by using 10 mol% catalyst, 2.5 equiv. oxidant, 3 mL solvent, 0.4 mL additive. [b] Isolated yields. [c] 5 mol% catalyst was used.

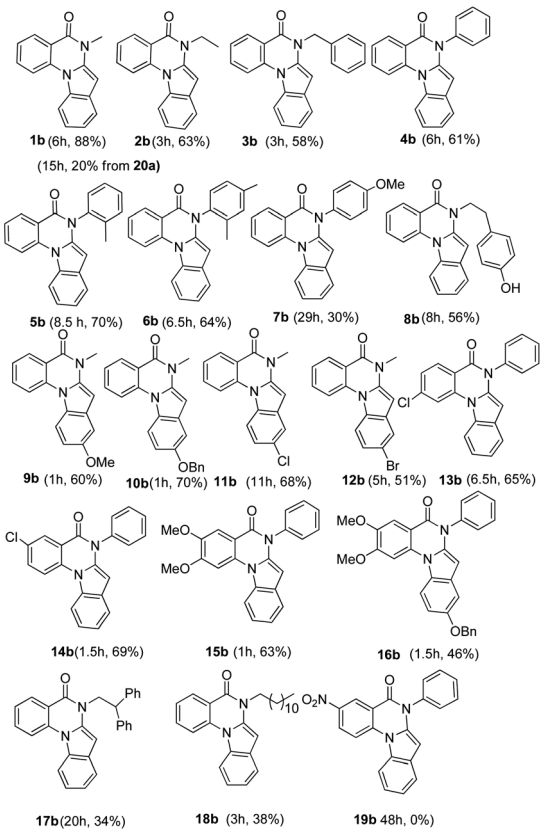
16a, **17a**, **18a** and **19a**) in good to moderate yields. Further, the reaction of 3-methylindole with 2-(1*H*-indol-1-yl)-*N*-methylbenzamide gave desired product in moderate yield.

After synthesizing various structurally diverse 2-(1*H*-indol-1-yl)-*N*-substituted benzamide derivatives, we focused our attention on the synthesis of indolo[1,2-*a*]quinazolinone derivatives by C–N arylation *via* C–H activation. To determine suitable conditions for this reaction, we initially treated 2-(1*H*-indol-1-yl)-*N*-methylbenzamide with a combination of palladium acetate, copper acetate and acetic acid. Under these conditions, the indolo[1,2-*a*]quinazolinone product was produced in 30% yield. Encouraged by this initial result, we screened the reaction using different acid additives including pivalic acid, *p*-toluene sulfonic acid (PTSA) and trifluoroacetic acid (TFA). The use of pivalic acid gave trace of the desired product. A slight improvement in the yield of the desired product was observed in the presence of PTSA. However, the desired product was produced in 54% yield when TFA was present as an additive. We next screened the reactions using different oxidants such as manganese acetate $Mn(OAc)_4$, silver acetate AgOAc, diacetoxy iodobenzene ($PhI(OAc)_2$) and benzoquinone. Among these oxidants, silver acetate proved to be the best choice for this reaction. We also screened the reaction using different palladium catalysts such as palladium chloride ($PdCl_2$), bistriphenylphosphinepalladium(II) dichloride ($Pd(PPh_3)_2Cl_2$) and palladium acetate $Pd(OAc)_2$. Of the catalysts tested, the use of bistriphenylphosphinepalladium(II) dichloride, silver acetate and trifluoroacetic acid resulted in the best yield (Table 3).

The optimal conditions were next employed for the synthesis of various structurally diverse indolo[1,2-*a*]quinazolinone derivatives. As described in Table 4, under optimized reaction conditions, 2-(1*H*-indol-1-yl)-*N*-methylbenzamide was converted into the corresponding indolo[1,2-*a*]quinazolinone derivative (**1b**) in excellent yield. However, when the reactant was 2-(1*H*-indol-1-yl)-*N*-ethylbenzamide, the yield of the desired indolo [1,2-*a*]quinazolinone derivative (**2b**) dropped slightly. Further, the reaction of 2-(1*H*-indol-1-yl)-*N*-benzylbenzamide furnished the expected product (**3b**) in moderate yield. In contrast, using the above optimized conditions, the reaction of *N*-phenyl indolylbenzamide derivatives gave good yields of the corresponding indolo[1,2-*a*]quinazolinone derivatives (**4b**, **5b** and **6b**). On the other hand, the reaction of an *N*-phenyl indolylbenzamide containing a strong electron donating group reacted slowly, providing the desired product (**7b**) in poor yield. We next investigated the reaction of 2-(1*H*-indol-1-yl)-*N*-methylbenzamide derived from various indoles. It should be noted that the reaction of 2-(1*H*-indol-1-yl)-*N*-methylbenzamides that contain electron releasing groups, such as OMe, OBn as well as moderate electron withdrawing groups, such as Cl, Br in the indole moiety furnished the desired products (**9b**, **10b**, **11b** and **12b**) in good yield. We also investigated the reactions of 2-(1*H*-indol-1-yl)-*N*-phenylbenzamide with substituents on the benzene ring. As can be seen in Table 4, substrates that contained both electron withdrawing and electron-donating groups on the benzene ring of the benzamide group produced the expected products (**13b**, **14b** and **15b**) in good yield. However, the substrate possessing NO_2 is *para*-positioned to the nitrogen

Table 4 Synthesis of indolo[1,2-*a*]quinazolinone derivatives[a]





[a] All reactions were conducted on a 0.5 mmol scale. The yields reported here are corresponds to isolated.

of the indole ring is failed produce its corresponding product and starting material was recovered in this case. The reason may be due to the presence of nitro group at *para* position to the nitrogen of the indole ring might reduce the electron density on indole nitrogen and as a result the carbon the 2-position of the indole ring will be less reactive. Furthermore, when the substrates having bulky group like 2,2-diphenylethyl and long chain alkyl group treated under the present reaction conditions, the corresponding indazoloquinazolinone derivatives (**17b** and **18b**) were obtained in poor yields. Steric factors may be the reason for the low yields in these two cases. Surprisingly, the reaction of **20a** under the present reaction conditions produced demethylated product **1b** in 20% yield.

Conclusion

In summary, a new method for the synthesis of indolo[1,2-*a*]-quinazolinone derivatives is reported. The procedure involves two steps, starting from 2-iodobenzamide and indole derivatives. The first step involves the *N*-arylation of indole by 2-

iodobenzamide derivatives *via* Ullman coupling and the second step involves intramolecular C–H amidation using a palladium catalyst. This procedure offers an easy, convenient and alternative method to existing methodologies for the synthesis of indolo[1,2-*a*]quinazolinone derivatives.

Experimental section

General information

Reagents and solvents were purchased from various commercial sources and were used directly without any further purification, unless otherwise stated. Column chromatography was performed with 63–200 mesh silica gel. ^{1}H and ^{13}C NMR spectra were recorded at 400 and 100 MHz, respectively. Chemical shifts are reported in parts per million (δ) using TMS and Chloroform as internal standards and coupling constants are expressed in Hertz. IR spectra were recorded on an FT-IR spectrometer and are reported in cm^{-1}. Melting points were recorded using an electro thermal capillary melting point apparatus and are uncorrected. HRMS spectra were recorded using ESI-TOF or EI$^+$ mode.

General procedure for the *N*-arylation of indoles (1a–19a)

Copper iodide (20 mol%) and potassium carbonate (2 equiv.) were added to a solution of 2-iodo-*N*-substituted benzamide (1 mmol) and the indole derivative (1.2 mmol) in DMSO. The reaction mixture then heated at 80 °C until the 2-iodo-*N*-substituted benzamide was completely converted into the corresponding *N*-arylated indole product as evidenced by monitoring with TLC. The reaction mixture was then extracted with ethyl acetate and the ethyl acetate layer was washed with a brine solution. The organic layer was separated, dried over anhydrous $MgSO_4$ and concentrated under vacuum to give the crude product. The crude product was then further purified by column chromatography.

General procedure for C–H amidation (1b–19b)

To a suspension of silver acetate (2.5 equiv.) in toluene, trifluoroacetic acid (0.4 mL) was added and the suspension stirred for 10 min. To this was added, 2-indolylbenzamide (**1a**) (0.5 mmol) and 5 mol% bistriphenylphosphinepalladium(II) dichloride. The reaction mixture was heated at 100 °C until the starting material (**1a**) was consumed, as indicated by TLC. The reaction mixture was then allowed to cool down to room temperature and 10 mL of ethyl acetate was added to this mixture. The suspension was passed through a celite pad and the solids washed with 2 × 10 mL ethyl acetate. The combined organic layer was washed with a $NaHCO_3$ solution and then with a brine solution. The organic layer was separated, dried over anhydrous $MgSO_4$ and concentrated under a vacuum to give the crude product. The crude product was further purified by column chromatography.

Spectral data

2-(1*H*-Indol-1-yl)-*N*-methylbenzamide (1a). Yield: (212 mg) 85%; white solid; m.p: 155–157 °C. FT-IR (KBr) (ν/cm^{-1}): 3315 (–NH–), 1644 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.94–7.92 (m, 1H), 7.69–7.67 (m, 1H), 7.60–7.56 (m, 1H)), 7.50 (t, J = 7.5 Hz, 1H), 7.43 (d, J = 7.8 Hz, 1H, 7.24–7.16 (m, 4H), 6.70 (d, J = 3.16 Hz, 1H), 5.14 (brs, 1H), 2.49 (d, J = 4.88 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 167.2, 136.9, 136.3, 133.3, 131.6, 131.0, 128.9, 128.8, 128.4, 127.9, 123.1, 121.4, 120.9, 110.3, 104.5, 26.8. LRMS (EI) (*m*/*z*) (relative intensity): 250.1 (M$^+$, 100), 220.1 (52), 191.0 (39); HRMS calcd for $C_{16}H_{14}ON_2$ (M$^+$): 250.1106, found 250.1103.

***N*-Ethyl-2-(1*H*-indol-1-yl)benzamide (2a).** Yield: (198 mg) 75%; white solid; m.p: 106–108 °C. FT-IR (KBr) (ν/cm^{-1}): 3292, (–NH–), 1645 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.96 (dd, J = 7.66, 1.4 Hz, 1H), 7.69–7.67 (m, 1H), 7.58–7.56 (m, 1H), 7.54–7.50 (m, 1H), 7.42 (dd, J = 7.82, 0.78 Hz, 1H), 7.26–7.16 (m, 4H), 6.72 (d, J = 3.16 Hz, 1H), 5.01 (brs, 1H), 2.97 (q, J = 7.2, 5.76 Hz, 2H), 0.49 (t, J = 7.28 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 166.2, 137.1, 136.2, 133.8, 131.6, 131.0, 128.8, 128.6, 128.6, 128.1, 123.1, 121.3, 120.9, 110.3, 104.4, 34.7, 13.8. LRMS (EI) (*m*/*z*) (relative intensity): 264.1 (M$^+$, 100), 220.1 (64), 193.1 (46), 191.1 (37). HRMS calcd for $C_{17}H_{16}ON_2$ (M$^+$): 264.1264, found 264.1268.

***N*-Benzyl-2-(1*H*-indol-1-yl)benzamide (3a).** Yield: (254 mg) 78%; white solid; m.p: 113–115 °C. FT-IR (KBr) (ν/cm^{-1}): 3282 (–NH–), 1645 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.98 (d, J = 7.64 Hz, 1H), 7.69–7.66 (m, 1H), 7.60–7.51 (m, 2H), 7.40 (d, J = 7.64 Hz, 1H), 7.25–7.18 (m, 4H), 7.17–7.08 (m, 3H), 6.66–6.62 (m, 3H), 5.44 (brs, 1H), 4.13 (d, J = 5.36 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 166.3, 137.1, 137.1, 136.3, 133.5, 131.7, 131.0, 128.9, 128.6, 128.6, 128.3, 127.7, 127.4, 123.2, 121.4, 121.0, 110.3, 104.6, 44.3. LRMS (EI) (*m*/*z*) (relative intensity): 326.1 (M$^+$, 100), 220.1 (49), 191.1 (54); HRMS calcd for $C_{22}H_{18}ON_2$ (M$^+$): 326.1419, found 326.1417.

2-(1*H*-Indol-1-yl)-*N*-phenylbenzamide (4a). Yield: (224 mg) 72%; white solid; m.p: 151–153 °C. FT-IR (KBr) (ν/cm^{-1}): 3285 (–NH–), 1659 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.13 (d, J = 7.64 Hz, 1H), 7.73–7.72 (m, 1H), 7.68–7.63 (m, 1H), 7.61–7.58 (m, 1H), 7.50 (d, J = 7.8 Hz, 1H), 7.29–7.26 (m, 2H), 7.24–7.20 (m, 2H), 7.12 (t, J = 7.88 Hz, 2H), 6.98 (t, J = 7.38 Hz, 1H), 6.85 (d, J = 7.64 Hz, 3H), 6.76 (d, J = 3.04 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 164.1, 137.3, 137.1, 136.2, 133.4, 132.3, 131.7, 129.0, 128.8, 128.8, 128.6, 124.7, 123.5, 121.6, 121.3, 120.4, 110.3, 105.1. LRMS (EI) (*m*/*z*) (relative intensity): 312.1 (M$^+$, 100), 220.1 (94.5) 191.0 (37); HRMS calcd for $C_{21}H_{16}ON_2$ (M$^+$): 312.1263, found 312.1262.

2-(1*H*-Indol-1-yl)-*N*-*o*-tolylbenzamide (5a). Yield: (283 mg) 87%; white solid; m.p: 123–125 °C. FT-IR (KBr) (ν/cm^{-1}): 3277 (–NH–), 1663 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.13 (d, J = 7.28 Hz, 1H), 7.68–7.57 (m, 4H), 7.46 (d, J = 7.6 Hz, 1H), 7.34–7.30 (m, 2H), 7.24–7.17 (m, 2H), 7.11–7.07 (m, 1H), 6.93 (d, J = 4.0 Hz, 2H), 6.90 (brs, 1H), 6.72 (d, J = 2.96 Hz, 1H), 1.40 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 164.6, 137.3, 136.2, 135.5, 133.5, 132.1, 131.7, 130.4, 129.2, 129.1, 128.9, 128.7, 128.4, 126.7, 125.3, 123.4, 122.6, 121.6, 121.3, 110.4, 105.3, 16.7. LRMS (EI) (*m*/*z*) (relative intensity): 326.1 (M$^+$, 91), 220.1 (100), 191.0 (49); HRMS calcd for $C_{22}H_{18}ON_2$ (M$^+$): 326.1419, found 326.1410.

***N*-(2,4-Dimethylphenyl)-2-(1*H*-indol-1-yl)benzamide (6a).** Yield: (272 mg) 80%; white solid; m.p: 100–102 °C. FT-IR (KBr) (ν/cm^{-1}): 3282 (–NH–), 1657 (–CONH–); ^{1}H NMR (400 MHz,

$CDCl_3$) δ 8.12 (dd, J = 7.56, 1.2 Hz, 1H), 7.68–7.56 (m, 3H), 7.45 (d, J = 6.76 Hz, 1H), 7.38–7.30 (m, 3H), 7.23–7.17 (m, 2H), 6.88 (d, J = 8.16 Hz, 1H), 6.83 (brs, 1H), 6.77 (s, 1H), 6.72 (d, J = 3.0 Hz, 1H), 2.20 (s, 3H), 1.41 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 164.6, 137.3, 136.2, 135.0, 133.6, 132.8, 132.0, 131.6, 131.0, 129.5, 129.2, 128.8, 128.7, 128.4, 127.2, 123.4, 122.9, 121.5, 121.2, 110.4, 105.2, 20.9, 16.7.

2-(1*H*-Indol-1-yl)-*N*-(4-methoxyphenyl)benzamide (7a). Yield: (246 mg) 72%; white solid; m.p: 121–123 °C. FT-IR (KBr) (ν/cm^{-1}): 3277 (–NH–), 1652 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.12–8.11 (m, 1H), 7.73–7.71 (m, 1H), 7.64–7.57 (m, 2H), 7.49 (d, J = 7.68 Hz, 1H), 7.29–7.25 (m, 2H), 7.23–7.21 (m, 2H), 6.76–6.72 (m, 4H), 6.66 (d, J = 8.96 Hz, 2H), 3.70 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 164.0, 156.7, 137.1, 136.2, 133.5, 132.1, 131.7, 130.3, 128.9, 128.8, 128.5, 123.5, 122.4, 121.5, 121.3, 114.0, 110.3, 105.0, 55.5. LRMS (EI) (*m*/*z*) (relative intensity): 342.1 (M^+, 100), 220.1 (100), 191.1 (30); HRMS calcd for $C_{22}H_{18}O_2N_2$ (M^+): 342.1368, found 342.1366.

***N*-(2,2-Diphenylethyl)-2-(1*H*-indol-1-yl)benzamide (8a).** Yield: (249 mg) 60%; white solid; m.p: 116–118 °C. FT-IR (KBr) (ν/cm^{-1}): 3297 (–NH–), 1645 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.85 (dd, J = 7.7, 1.62 Hz, 1H), 7.77–7.75 (m, 1H), 7.56–7.51 (m, 1H), 7.48–7.44 (m, 1H), 7.39–7.37 (dd, J = 7.75, 1.12 Hz, 1H), 7.28–7.20 (m, 3H), 7.16–7.11 (m, 7H), 6.86–6.84 (m, 4H), 6.55 (d, J = 3.28 Hz, 1H), 5.17 (brs, 1H), 3.68–3.65 (m, 2H), 3.44 (t, J = 7.94 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 166.7, 141.5, 136.9, 136.3, 133.2, 131.6, 130.8, 129.0, 128.8, 128.7, 128.2, 127.8, 127.8, 126.8, 123.1, 121.4, 121.0, 110.5, 104.5, 50.1, 44.3. LRMS (EI) (*m*/*z*) (relative intensity): 416.2 (M^+, 37.5), 220.1 (100), 191.1 (30); HRMS calcd for $C_{29}H_{24}ON_2$ (M^+): 416.1889, found 416.1878.

***N*-(4-Hydroxyphenethyl)-2-(1*H*-indol-1-yl)benzamide (9a).** Yield: (195 mg) 55%; white solid; m.p: 195–197 °C. FT-IR (KBr) (ν/cm^{-1}): 3277 (–NH–), 1640 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.23 (brs, 1H), 6.98–6.91 (m, 3H), 6.83–6.80 (m, 3H), 6.64–6.61 (m, 2H), 6.51–6.44 (m, 2H), 6.13–6.11 (m, 2H), 5.99–5.97 (m, 3H), 2.53 (q, J = 13.8, 6.76 Hz, 2H), 1.66 (t, J = 7.3 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 165.6, 154.4, 135.4, 135.0, 133.4, 129.3, 128.1, 128.0, 127.9, 127.4, 126.1, 126.1, 120.8, 119.5, 118.9, 114.0, 109.2, 101.9, 39.9, 32.8; LRMS (EI) (*m*/*z*) (relative intensity): 356.1 (M^+, 41), 236.1 (99), 220.1 (100), 191.1 (46); HRMS calcd for $C_{23}H_{20}O_2N_2$ (M^+): 356.1525, found 356.1528.

***N*-Dodecyl-2-(1*H*-indol-1-yl)benzamide (10a).** Yield: (323 mg) 80%; white solid; m.p: 65–67 °C. FT-IR (KBr) (ν/cm^{-1}): 3277 (–NH–), 1640 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.97 (dd, J = 7.6, 1.6, 1H), 7.68–7.66 (m, 1H), 7.59–7.49 (m, 2H), 7.40 (dd, J = 7.68, 1.04, 1H), 7.22–7.15 (m, 4H), 6.71 (d, J = 3.2, 1H), 5.14 (brs, 1H), 2.94 (q, J = 12.4, 6.56, 2H), 1.32–1.17 (m, 12H), 1.11–1.07 (m, 2H), 1.04–0.98 (m, 2H), 0.89 (t, J = 6.8, 3H), 0.85–0.77 (m, 4H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 166.2, 137.1, 136.2, 133.7, 131.5, 131.1, 128.8, 128.6, 128.2, 123.1, 121.3, 120.9, 110.3, 104.5, 39.9, 32.1, 29.8, 29.7, 29.5, 29.3, 28.8, 26.6, 22.8, 14.2; LRMS (EI) (*m*/*z*) (relative intensity): 404.3 (M^+, 46), 220.1 (100), 191.1 (42); HRMS calcd for $C_{27}H_{36}ON_2$ (M^+): 404.2828, found 404.2823.

2-(5-Methoxy-1*H*-indol-1-yl)-*N*-methylbenzamide (11a). Yield: (224 mg) 80%; white solid; m.p: 128–130 °C. FT-IR (KBr) (ν/cm^{-1}): 3285 (–NH–), 1644 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.92 (dd, J = 7.82, 1.06 Hz, 1H), 7.58–7.54 (m, 1H), 7.50–7.46 (m, 1H), 7.41 (dd, J = 7.88, 0.92 Hz, 1H), 7.18 (d, J = 3.24 Hz, 1H), 7.14–7.12 (m, 2H), 6.86–6.84 (m, 1H), 6.62 (d, J = 3.04 Hz, 1H), 5.16 (brs, 1H), 3.85 (s, 3H), 2.51 (d, J = 4.92 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 167.2, 155.0, 136.5, 133.1, 132.2, 1316, 131.0, 129.5, 129.3, 128.2, 127.8, 113.2, 111.1, 104.2, 103.0, 55.9, 26.8. LRMS (EI) (*m*/*z*) (relative intensity): 280.1 (M^+, 100), 265.1 (34), 236.0 (9); HRMS calcd for $C_{17}H_{16}O_2N_2$ (M^+): 280.1212, found 280.1219.

2-(5-(Benzyloxy)-1*H*-indol-1-yl)-*N*-methylbenzamide (12a). Yield: (267 mg) 75%; white solid; m.p: 140–142 °C. FT-IR (KBr) (ν/cm^{-1}): 3297 (–NH–), 1645 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.93 (dd, J = 7.62, 1.5 Hz, 1H), 7.59–7.54 (m, 1H), 7.51–7.47 (m, 3H), 7.42–7.37 (m, 3H), 7.34–7.30 (m, 1H), 7.20 (dd, J = 8.04, 2.76 Hz, 2H), 7.15 (d, J = 8.92 Hz, 1H), 6.95 (dd, J = 8.92, 2.36 Hz, 1H), 6.62 (d, J = 3.08 Hz, 1H), 5.12 (s, 3H), 2.52 (d, J = 4.88 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 167.3, 154.2, 137.6, 136.4, 133.1, 132.4, 131.6, 131.0, 129.5, 129.4, 128.7, 128.3, 128.0, 127.8, 127.7, 113.9, 111.2, 104.6, 104.3, 71.0, 26.9. LRMS (EI) (*m*/*z*) (relative intensity): 356.1 (M^+, 49), 265 (100); HRMS calcd for $C_{23}H_{20}O_2N_2$ (M^+): 356.1525, found 356.1523.

2-(5-Chloro-1*H*-indol-1-yl)-*N*-methylbenzamide (13a). Yield: (221 mg) 78% white solid; m.p: 182–184 °C. FT-IR (KBr) (ν/cm^{-1}): 3315 (–NH–), 1636 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.91–7.89 (m, 1H), 7.64 (s, 1H), 7.60–7.56 (m, 1H), 7.54–7.50 (m, 1H), 7.42–7.40 (m, 1H), 7.25 (s, 1H), 7.15 (s, 2H), 6.64 (d, J = 3.16 Hz, 1H), 5.08 (brs, 1H), 2.52 (d, J = 4.92 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 167.1, 135.9, 135.4, 133.5, 131.7, 130.9, 130.1, 129.9, 128.7, 127.9, 126.7, 123.4, 120.8, 111.4, 104.0, 26.9. LRMS (EI) (*m*/*z*) (relative intensity): 284.0 (M^+, 100), 254 (33), 219.1 (31.5); HRMS calcd for $C_{16}H_{13}ON_2Cl$ (M^+): 284.0716, found 284.0718.

2-(5-Bromo-1*H*-indol-1-yl)-*N*-methylbenzamide (14a). Yield: (255 mg) 78%; white solid; m.p: 181–183 °C. FT-IR (KBr) (ν/cm^{-1}): 3285 (–NH–), 1640 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.85 (dd, J = 7.7, 1.46 Hz, 1H), 7.79 (d, J = 1.64 Hz, 1H), 7.59–7.55 (m, 1H), 7.52–7.48 (m, 1H), 7.39 (dd, J = 7.78, 0.86 Hz, 1H), 7.35–7.25 (m, 1H), 7.22 (d, J = 3.28 Hz, 1H), 7.09 (d, J = 3.28 Hz, 1H), 6.63 (d, J = 3.04 Hz, 1H), 5.18 (brs, 1H), 2.51 (d, J = 4.88 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 167.0, 135.4, 135.1, 134.9, 131.0, 130.6, 130.2, 128.8, 127.8, 127.4, 124.1, 122.7, 112.3, 112.2, 102.3, 25.9. LRMS (EI) (*m*/*z*) (relative intensity): 328.0 (M^+, 100), 220 (29), 219 (67); HRMS calcd for $C_{16}H_{13}ON_2Br$ (M^+): 328.0211, found 328.0205.

5-Chloro-2-(1*H*-indol-1-yl)-*N*-phenylbenzamide (15a). Yield: (242 mg) 70%; white solid; m.p: 126–128 °C. FT-IR (KBr) (ν/cm^{-1}): 3285 (–NH–), 1655 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.10 (d, J = 8.4 Hz, 1H), 7.71 (d, J = 6.76 Hz, 1H), 7.57 (d, J = 8.44 Hz, 1H), 7.50 (d, J = 1.8 Hz, 1H), 7.27–7.22 (m, 4H), 7.13 (t, J = 3.92 Hz, 2H), 6.99 (t, J = 3.64 Hz, 1H), 6.84 (d, J = 7.84 Hz, 3H), 6.78 (d, J = 3.16 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 163.0, 138.0, 137.3, 137.1, 136.9, 133.1, 131.6, 129.2, 129.0, 128.9, 128.7, 128.4, 124.9, 123.9, 121.7, 121.7, 120.4, 110.2, 105.8. LRMS (EI) (*m*/*z*) (relative intensity): 346.1 (M^+, 31), 254.0 (44), 117.0 (100); HRMS calcd for $C_{21}H_{15}ON_2Cl$ (M^+): 346.0873, found 346.0869.

2-(1*H*-Indol-1-yl)-4,5-dimethoxy-*N*-phenylbenzamide (16a). Yield: (268 mg) 72%; white solid; m.p: 176–177 °C. FT-IR (KBr)

(ν/cm^{-1}): 3297 (–NH–), 1692 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.77–7.76 (m, 2H), 7.28–7.23 (m, 4H), 7.12 (t, J = 7.8 Hz, 2H), 6.97 (t, J = 7.38 Hz, 1H), 6.89–6.87 (m, 2H), 6.84–6.79 (m, 3H), 4.05 (s, 3H), 3.92 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 163.1, 151.8, 149.4, 137.6, 137.3, 129.6, 129.0, 128.8, 128.8, 125.4, 124.4, 123.7, 121.6, 121.4, 120.1, 113.5, 111.5, 110.4, 105.0, 56.5, 56.5. LRMS (ESI) (m/z) (relative intensity): 395.1 ((M + Na)$^+$, 37) 373.1 (80), 372.1 (100); HRMS calcd for $C_{23}H_{20}N_2O_3$ (M + Na)$^+$ 395.1372, found 395.1384.

4-Chloro-2-(1*H*-indol-1-yl)-*N*-phenylbenzamide (17a). Yield: (235 mg) 68%; white solid; m.p: 144–146 °C. FT-IR (KBr) (ν/cm^{-1}): 3282 (–NH–), 1652 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.08 (d, J = 8.36 Hz, 1H), 7.73 (d, J = 6.8 Hz, 1H), 7.56 (d, J = 8.44 Hz, 1H), 7.50 (d, J = 1.44 Hz, 1H), 7.29–7.21 (m, 4H), 7.12 (t, J = 7.74 Hz, 2H), 6.99 (t, J = 7.32 Hz, 1H), 6.84 (d, J = 7.88 Hz, 3H), 6.78 (d, J = 3.0 Hz, 1H), 5.29 (brs, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 163.0, 138.0, 137.3, 137.1, 136.9, 133.0, 131.6, 129.2, 129.0, 128.9, 128.6, 128.4, 124.8, 123.9, 121.7, 121.7, 120.4, 110.2, 105.8. LRMS (ESI) (m/z) (relative intensity): 369.0 ((M + Na)$^+$, 15), 349.0 (36), 348 (43), 347.0 (100); HRMS calcd for $C_{21}H_{15}ClN_2O$ (M + Na)$^+$ 369.0771, found 369.0780.

2-(5-(Benzyloxy)-1*H*-indol-1-yl)-4,5-dimethoxy-*N*-phenylbenzamide (18a). Yield: (311 mg) 65%; white solid; m.p: 80–82 °C. FT-IR (KBr) (ν/cm^{-1}): 3297 (–NH–), 1657 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.73 (s, 1H), 7.48–7.46 (m, 2H), 7.41–7.37 (m, 3H), 7.33 (d, J = 7.12 Hz, 1H), 7.22 (d, J = 3.16, 1H), 7.15–7.10 (m, 3H), 6.99–6.97 (m, 2H), 6.90–6.86 (m, 4H), 6.69 (d, J = 3.04 Hz, 1H), 5.13 (s, 2H), 4.02 (s, 3H), 3.90 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 163.2, 154.5, 151.8, 149.3, 137.6, 137.5, 132.7, 129.7, 129.6, 129.3, 128.8, 128.7, 128.0, 127.6, 125.2, 124.4, 120.1, 114.6, 113.4, 111.4, 111.2, 104.8, 104.7, 71.0, 56.5, 56.5; LRMS (EI) (m/z) (relative intensity): 478.2 (M$^+$, 53), 378.2 (100); HRMS calcd for $C_{30}H_{26}O_4N_2$ (M$^+$): 478.1893, found 478.1887.

2-(1H-Indol-1-yl)-5-nitro-*N*-phenylbenzamide (19a). Yield: (232 mg) 65%; yellow solid; m.p: 85–87 °C. FT-IR (KBr) (ν/cm^{-1}): 3289 (–NH–), 1653 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.95 (d, J = 2.6 Hz, 1H), 8.48 (dd, J = 8.66, 0.2.66 Hz, 1H), 7.73–7.71 (m, 2H), 7.32–7.30 (m, 2H), 7.28–7.23 (m, 3H), 7.17 (t, J = 7.8 Hz, 2H), 7.04 (t, J = 7.38 Hz, 1H), 6.92 (d, J = 7.92 Hz, 3H), 6.83 (d, J = 3.28 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 162.1, 147.1, 141.6, 136.6, 136.5, 134.0, 129.4, 129.0, 129.0, 128.2, 127.4, 126.8, 125.4, 124.3, 122.2, 122.0, 120.7, 110.1, 106.9; LRMS (EI) (m/z) (relative intensity): 357 (M$^+$, 90), 264 (22), 219 (57), 191 (25), 93 (100).

***N*-Methyl-2-(3-methyl-1*H*-indol-1-yl)benzamide (20a).** Yield: (145 mg) 55%; white solid; m.p: 195–197 °C. FT-IR (KBr) (ν/cm^{-1}): 3338 (–NH–), 1640 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.94 (dd, J = 7.7, 1.46 Hz, 1H), 7.63–7.61 (m, 1H), 7.56 (td, J = 7.64, 1.6 Hz, 1H), (td, J = 7.48, 1 Hz, 1H), 7.39 (dd, J = 7.68, 0.72 Hz, 1H), 7.32–7.16 (m, 3H), 7.00 (d, J = 0.56 Hz, 1H), 5.23 (brs, 1H), 2.51 (d, J = 4.92 Hz, 3H), 2.38 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 167.4, 137.2, 136.6, 132.9, 131.6, 131.0, 129.5, 128.0, 127.8, 126.2, 123.1, 120.4, 119.4, 113.9, 110.3, 26.8, 9.76; LRMS (EI) (m/z) (relative intensity): 264 (M$^+$, 100), 235 (7), 234 (10), 206 (15), 204 (40), 191 (7), 130 (12), 77 (5).

C–H activation

6-Methylindolo[1,2-*a*]quinazolin-5(6*H*)-one (1b). Yield: (109 mg) 88%; yellow green solid; m.p: 181–183 °C. FT-IR (KBr) (ν/cm^{-1}): 1660, (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.40 (d, J = 7.88 Hz, 1H), 8.26 (dd, J = 8.44, 2.32, 1H), 8.10–8.07 (m, 1H), 7.76 (t, J = 7.80 Hz, 1H), 7.64–7.62 (m, 1H), 7.34 (t, J = 7.5 Hz, 1H), 7.32–7.28 (m, 2H), 6.05 (d, J = 2 Hz, 1H), 3.62 (d, J = 2.12 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.7, 139.0, 138.4, 134.5, 131.6, 130.1, 130.1, 123.6, 122.7, 121.5, 120.2, 116.7, 114.7, 113.2, 84.3, 30.6. LRMS (EI) (m/z) (relative intensity): 248.1 (M$^+$, 100), 205.1 (23); HRMS calcd for $C_{16}H_{12}ON_2$ (M$^+$): 248.0950 found 248.0950.

6-Ethylindolo[1,2-*a*]quinazolin-5(6*H*)-one (2b). Yield: (83 mg) 63%; yellow green solid; m.p: 108–110 oCFT-IR (KBr) (ν/cm^{-1}): 1664 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.41–8.38 (m, 1H), 8.26 (d, J = 8.48 Hz, 1H), 8.09 (d, J = 5.48 Hz, 1H), 7.77–7.73 (m, 1H), 7.64–7.62 (m, 1H), 7.34 (t, J = 7.56 Hz, 1H), 7.31–7.26 (m, 2H), 6.08 (s, 1H), 4.21 (q, J = 14.34, 7.14 Hz, 2H), 1.44 (t, J = 7.14 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.2, 139.0, 137.4, 134.4, 131.4, 130.1, 130.0, 123.6, 122.6, 121.3, 120.0, 116.9, 114.7, 113.2, 84.0, 39.5, 12.0; LRMS (EI) (m/z) (relative intensity): 262.1 (M$^+$, 97), 234.1 (100), 205.1 (48); HRMS calcd for $C_{17}H_{14}ON_2$ (M$^+$): 262.1106, found 262.1107.

6-Benzylindolo[1,2-*a*]quinazolin-5(6*H*)-one (3b). Yield: (94 mg) 58%; yellow green solid; m.p: 157–159 °C. FT-IR (KBr) (ν/cm^{-1}): 1664 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.45 (dd, J = 7.88, 1.44 Hz, 1H), 8.26–8.23 (m, 1H), 8.06–8.04 (m, 1H), 7.78–7.74 (m, 1H), 7.55–7.52 (m, 1H), 7.41 (d, J = 7.4 Hz, 2H), 7.37–7.30 (m, 3H), 7.28–7.24 (m, 3H), 6.00 (s, 1H), 5.35 (s, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.9, 139.1, 137.5, 135.8, 134.7, 131.3, 130.3, 130.0, 128.9, 127.8, 127.5, 123.6, 122.6, 121.5, 120.2, 116.5, 114.7, 113.1, 85.5, 47.7; LRMS (EI) (m/z) (relative intensity): 324.1 (M$^+$, 100), 205.1 (51); HRMS calcd for $C_{22}H_{16}ON_2$ (M$^+$): 324.1263, found 324.1258.

6-Phenylindolo[1,2-*a*]quinazolin-5(6*H*)-one (4b). Yield: (94 mg) 61%; yellow green solid; m.p: 226–228 °C. FT-IR (KBr) (ν/cm^{-1}): 1667 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.43 (dd, J = 7.82, 1.5 Hz, 1H), 8.34 (d, J = 8.48 Hz, 1H), 8.14 (d, J = 8.12 Hz, 1H), 7.84–7.80 (m, 1H), 7.63–7.59 (m, 2H), 7.56–7.54 (m, 1H), 7.48–7.46 (m, 3H), 7.37 (t, J = 7.54 Hz, 1H), 7.31–7.23 (m, 2H), 5.50 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.7, 139.4, 139.4, 137.2, 134.9, 131.4, 130.4, 130.3, 129.8, 129.4, 128.6, 123.7, 122.7, 121.5, 120.2, 117.2, 114.8, 113.2, 86.3; LRMS (EI) (m/z) (relative intensity): 310.1 (M$^+$, 78), 220 (31), 197.1 (62), 105.0 (100); HRMS calcd for $C_{21}H_{14}ON_2$ (M$^+$): 310.1106, found 310.1097.

6-(*o*-Tolyl)indolo[1,2-*a*]quinazolin-5(6*H*)-one (5b). Yield: (113 mg) 70%; yellow green solid; m.p: 198–200 °C. FT-IR (KBr) (ν/cm^{-1}): 1670 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.46 (dd, J = 7.86, 1.18 Hz, 1H), 8.36 (d, J = 8.48 Hz, 1H), 8.15 (d, J = 8.12 Hz, 1H), 7.86–7.81 (m, 1H), 7.49–7.35 (m, 6H), 7.32–7.24 (m, 2H), 5.41 (s, 1H), 2.21 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.2, 139.5, 138.6, 136.5, 136.1, 134.9, 131.9, 131.5, 130.5, 130.0, 129.7, 128.7, 127.9, 123.7, 122.7, 121.5, 120.2, 117.0, 114.8, 113.2, 86.0, 17.5; LRMS (EI) (m/z) (relative intensity): 324.1 (M$^+$, 73), 211.1 (56), 105.0 (100); HRMS calcd for $C_{22}H_{16}ON_2$ (M$^+$): 324.1263, found 324.1258.

6-(2,4-Dimethylphenyl)indolo[1,2-*a*]quinazolin-5(6*H*)-one (6b). Yield: (108 mg) 64%; yellow green solid; m.p: 211–212 °C. FT-IR (KBr) (ν/cm^{-1}): 1670 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.45 (dd, J = 7.82, 1.46 Hz, 1H), 8.35 (d, J = 8.44 Hz, 1H), 8.15 (d, J = 8.16 Hz, 1H), 7.85–7.80 (m, 1H), 7.48 (dd, J = 8.26 Hz, 0.98, 1H), 7.38 (t, J = 7.66 Hz, 1H), 7.32–7.22 (m, 5H), 5.43 (s, 1H), 2.44 (s, 3H), 2.16 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.3, 139.6, 139.5, 138.8, 136.0, 134.8, 133.5, 132.6, 131.6, 130.5, 130.0, 128.6, 128.4, 123.6, 122.7, 121.4, 120.2, 117.1, 114.8, 113.2, 86.0, 21.4, 17.4; LRMS (EI) (m/z) (relative intensity): 338.1 (M^+, 100), 225.1 (17), 105.0 (26); HRMS calcd for $C_{23}H_{18}ON_2$ (M^+): 338.1419, found 338.1424.

6-(4-Methoxyphenyl)indolo[1,2-*a*]quinazolin-5(6*H*)-one (7b). Yield: (51 mg) 30%; yellow green solid; m.p: 262–264 °C. FT-IR (KBr) (ν/cm^{-1}): 1667 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.43 (dd, J = 7.84, 1.56 Hz, 1H), 8.33 (d, J = 8.44 Hz, 1H), 8.13 (d, J = 8.04 Hz, 1H), 7.83–7.79 (m, 1H), 7.49–7.47 (m, 1H), 7.40–7.35 (m, 3H), 7.31–7.23 (m, 2H), 7.12–7.08 (m, 2H), 5.52 (s, 1H), 3.90 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 160.1, 139.7, 139.4, 134.9, 130.5, 129.9, 129.8, 129.6, 123.7, 122.7, 121.5, 120.2, 115.5, 114.8, 113.2, 86.3, 55.8; LRMS (EI) (m/z) (relative intensity): 340.1 (M^+, 100); HRMS calcd for $C_{22}H_{16}O_2N_2$ (M^+): 340.1212, found 340.1220.

6-(4-Hydroxyphenethyl)indolo[1,2-*a*]quinazolin-5(6*H*)-one (8b). Yield: (99 mg) 56%; yellow green solid; m.p: 240–243 °C. FT-IR (KBr) (ν/cm^{-1}): 1640 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.26 (bs, 1H), 8.43 (d, J = 8.48 Hz, 1H), 8.27 (d, J = 8.08 Hz, 1H), 8.21–8.19 (m, 1H), 7.85 (t, J = 7.44 Hz, 1H), 7.65–7.63 (m, 1H), 7.39 (t, J = 7.56 Hz, 1H), 7.28–7.23 (m, 2H), 7.15 (d, J = 8.36 Hz, 2H), 6.73 (d, J = 8.4 Hz, 2H), 6.39 (s, 1H), 4.20 (t, J = 8.02 Hz, 2H), 2.93 (t, J = 7.98 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 157.1, 155.9, 138.0, 136.8, 134.9, 130.5, 129.7, 129.5, 129.0, 128.2, 123.5, 122.3, 121.1, 119.6, 115.8, 115.2, 114.9, 113.0, 84.2, 45.2, 31.2; LRMS (EI) (m/z) (relative intensity): 354.1 (M^+, 79), 235.1 (69.5); HRMS calcd for $C_{23}H_{18}O_2N_2$ (M^+): 354.1368, found 354.1371.

9-Methoxy-6-methylindolo[1,2-*a*]quinazolin-5(6*H*)-one (9b). Yield: (83 mg) 60%; yellow green solid; m.p: 198–200 °C. FT-IR (KBr) (ν/cm^{-1}): 1659 (–CONH–) ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.37 (dd, J = 6.68 Hz, 1H), 8.14 (d, J = 8.4 Hz, 1H), 7.93 (d, J = 9.08 Hz, 1H), 7.73 (t, J = 7.88 Hz, 1H), 7.31 (t, J = 7.6 Hz, 1H), 7.06 (d, J = 2.24 Hz, 1H), 6.87 (dd, J = 9.08, 2.4 Hz, 1H), 5.96 (s, 1H), 3.89 (s, 3H), 3.59 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.6, 155.8, 138.8, 134.5, 134.4, 131.22, 130.0, 126.3, 123.3, 116.3, 114.2, 113.9, 110.0, 102.8, 84.2, 55.8, 30.5; LRMS (EI) (m/z) (relative intensity): 278.1 (M^+, 100), 235.1 (49); HRMS calcd for $C_{17}H_{14}O_2N_2$ (M^+): 278.1055, found 278.1048.

9-(Benzyloxy)-6-methylindolo[1,2-*a*]quinazolin-5(6*H*)-one (10b). Yield: (124 mg) 70%; yellow green solid; m.p: 173–175 °C. FT-IR (KBr) (ν/cm^{-1}): 1664 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.36 (d, J = 7.2 Hz, 1H), 8.11 (s, 1H), 7.91 (s, 1H), 7.71 (s, 1H), 7.49 (d, J = 7.24 Hz, 2H), 7.41 (t, J = 7.28 Hz, 2H), 7.36–7.30 (m, 2H), 7.14 (s, 1H), 6.95 (d, J = 8.6 Hz, 1H), 5.92 (d, J = 2.68 Hz, 1H), 5.14 (s, 2H), 3.56 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.6, 154.9, 138.8, 138.7, 137.5, 134.5, 134.4, 131.1, 130.0, 130.0, 128.8, 128.1, 127.7, 126.4, 123.3, 123.3, 116.3, 114.2, 113.9, 110.8, 104.1, 84.2, 70.7, 30.5; LRMS (EI) (m/z) (relative intensity): 354.1 (M^+, 100), 263.1 (37.5), 235.1 (94); HRMS calcd for $C_{23}H_{18}O_2N_2$ (M^+): 354.1368, found 354.1360.

9-Chloro-6-methylindolo[1,2-*a*]quinazolin-5(6*H*)-one (11b). Yield: (96 mg) 68%; yellow green solid; m.p: 216–218 °C. FT-IR (KBr) (ν/cm^{-1}): 1660 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.38 (d, J = 7.64 Hz, 1H), 8.10 (d, J = 8.36 Hz, 1H), 7.92 (d, J = 8.8 Hz, 1H), 7.74 (t, J = 7.68 Hz, 1H), 7.54 (s, 1H), 7.35 (t, J = 7.46 Hz, 1H), 7.19 (d, J = 8.68 Hz, 1H), 5.94 (s, 1H), 3.58 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.5, 139.3, 138.5, 134.6, 131.3, 130.2, 129.8, 128.2, 124.0, 121.4, 119.6, 116.7, 114.5, 113.9, 83.8, 30.6; LRMS (EI) (m/z) (relative intensity): 282.0 (M^+, 100), 239.0 (24); HRMS calcd for $C_{16}H_{11}ON_2$ Cl (M^+): 282.0560, found 282.0564.

9-Bromo-6-methylindolo[1,2-*a*]quinazolin-5(6*H*)-one (12b). Yield: (83 mg) 51%; yellow green solid; m.p: 225–227 °C. FT-IR (KBr) (ν/cm^{-1}): 1660 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.38 (dd, J = 7.88, 1.16 Hz, 1H), 8.12 (d, J = 8.44 Hz, 1H), 7.90 (d, J = 8.84 Hz, 1H), 7.77–7.73 (m, 1H), 7.71 (d, J = 1.76 Hz, 1H), 7.38–7.32 (m, 2H), 5.96 (s, 1H), 3.59 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.6, 139.2, 138.5, 134.6, 131.8, 130.2, 124.1, 124.0, 122.6, 116.7, 115.9, 114.6, 114.3, 83.7, 30.6; LRMS (EI) (m/z) (relative intensity): 326.0 (M^+, 100); HRMS calcd for $C_{16}H_{11}ON_2$ Br (M^+): 326.0055, found 326.0064.

2-Chloro-6-phenylindolo[1,2-*a*]quinazolin-5(6*H*)-one (13b). Yield: (112 mg) 65%; yellow green solid; m.p: 168–170 °C; FT-IR (KBr) (ν/cm^{-1}): 1660 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.34 (d, J = 8.44 Hz, 1H), 8.29 (d, J = 168 Hz, 1H), 8.04 (d, J = 8.24 Hz, 1H), 7.63–7.59 (m, 2H), 7.57–7.55 (m, 1H), 7.48–7.45 (m, 3H), 7.33–7.29 (m, 2H), 7.28 (d, J = 0.76 Hz, 1H), 5.50 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.0, 141.3, 140.0, 139.3, 137.0, 131.8, 131.3, 130.3, 129.9, 129.5, 128.5, 124.0, 123.2, 122.0, 120.4, 115.6, 115.0, 113.00, 86.9; LRMS (EI) (m/z) (relative intensity): 344.0 (M^+, 100); HRMS calcd for $C_{21}H_{13}ON_2Cl$ (M^+): 344.0716 found 344.0712.

3-Chloro-6-phenylindolo[1,2-*a*]quinazolin-5(6*H*)-one (14b). Yield: (118 mg) 69%; yellow green solid; m.p; 223–231 °C. FT-IR (KBr) (ν/cm^{-1}): 1663 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.35 (d, J = 8.44 Hz, 1H), 8.30 (d, J = 1.68 Hz, 1H), 8.06 (d, J = 8.32 Hz, 1H), 7.63–7.59 (m, 2H), 7.56–7.55 (m, 1H), 7.48–7.45 (m, 3H), 7.34–7.25 (m, 3H), 5.50 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.0, 141.3, 140.0, 139.3, 137.0, 131.8, 131.3, 130.3, 129.9, 129.5, 128.5, 124.0, 123.2, 122.0, 120.4, 115.7, 115.0, 113.0, 87.0; LRMS (EI) (m/z) (relative intensity): 344.0 (M^+, 100), 254.0 (82), 231.0 (41), 139.0 (100); HRMS calcd for $C_{21}H_{13}ON_2Cl$ (M^+): 344.0716 found 344.0720.

2,3-Dimethoxy-6-phenylindolo[1,2-*a*]quinazolin-5(6*H*)-one (15b). Yield: (116 mg) 63%; yellow green solid; m.p: 200–202 °C. FT-IR (KBr) (ν/cm^{-1}): 1659 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.03 (d, J = 7.88 Hz, 1H), 7.81 (d, J = 9.48 Hz, 2H), 7.60 (t, J = 7.44 Hz, 2H), 7.54 (d, J = 7.12 Hz, 1H), 7.52–7.45 (m, 3H), 7.28–7.23 (m, 2H), 5.48 (s, 1H), 4.15 (s, 3H), 3.98 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.5, 154.7, 145.8, 139.6, 137.4, 134.9, 130.8, 130.2, 129.8, 129.3, 128.7, 122.5, 121.1, 120.2, 112.4, 110.9, 109.7, 98.3, 85.6, 56.7, 56.5; LRMS (EI) (m/z) (relative intensity): 370.1 (M^+, 100); HRMS calcd for $C_{23}H_{18}O_3N_2$ (M^+): 370.1317, found 370.1312.

9-(Benzyloxy)-2,3-dimethoxy-6-phenylindolo[1,2-*a*]quinazolin-5(6*H*)-one (16b). Yield: (109 mg) 46%; yellow green solid; mp: 238–240 °C. FT-IR (KBr) (ν/cm^{-1}): 1693 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.90 (d, J = 9.04 Hz, 1H), 7.81 (s, 1H), 7.70 (s, 1H), 7.59 (t, J = 7.46 Hz, 2H), 7.54 (d, J = 7.28 Hz, 1H), 7.45 (d, J = 7.8 Hz, 4H), 7.37 (t, J = 7.4 Hz, 2H), 7.32 (d, J = 7.16 Hz, 1H), 7.01 (d, J = 2.36 Hz, 1H), 6.95 (dd, J = 9.02, 2.46 Hz, 1H), 5.39 (s, 1H), 5.10 (s, 2H), 4.15 (s, 3H), 3.98 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.5, 154.9, 154.9, 145.7, 140.2, 137.5, 137.4, 134.8, 130.9, 130.2, 129.3, 128.7, 128.7, 128.1, 127.6, 125.9, 113.1, 110.9, 110.7, 109.4, 104.2, 97.9, 85.5, 70.7, 56.7, 56.5. LRMS (EI) (m/z) (relative intensity): 476.1 (M^+, 51), 385.1 (19), 357.1 (100); HRMS calcd for $C_{30}H_{24}O_4N_2$ (M^+): 476.1736, found 476.1743.

6-(2,2-Diphenylethyl)indolo[1,2-*a*]quinazolin-5(6*H*)-one (17b). Yield: (70 mg) 34%; yellow green solid; m.p: 192–194 °C. FT-IR (KBr) (ν/cm^{-1}): 1659 (–CONH); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.21 (t, J = 7.24 Hz, 2H), 8.07 (d, J = 8.04 Hz, 1H), 7.70 (t, J = 7.84 Hz, 1H), 7.62 (t, J = 4.34 Hz, 1H), 7.33 (d, J = 7.4 Hz, 4H), 7.30–7.24 (m, 7H), 7.17 (t, J = 7.22 Hz, 2H), 6.07 (s, 1H), 4.84 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.5, 141.6, 138.8, 137.5, 134.4, 131.3, 130.1, 130.0, 128.7, 128.4, 127.1, 123.5, 122.6, 121.4, 120.1, 116.6, 114.6, 113.2, 84.9, 48.3, 47.6; LRMS (EI) (m/z) (relative intensity): 414.2 (M^+, 42), 234.1 (100), 219.1 (22); HRMS calcd for $C_{29}H_{22}ON_2$ (M^+): 414.1732, found 414.1736.

6-Dodecylindolo[1,2-*a*]quinazolin-5(6*H*)-one (18b). Yield: (76 mg) 38%; yellow green solid; m.p: 72–74 °C. FT-IR (KBr) (ν/cm^{-1}); 1667 (–CONH–); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.39 (dd, J = 7.86, 1.18 Hz, 1H), 8.26 (d, J = 8.44 Hz, 1H), 8.09 (d, J = 8.56 Hz, 1H), 7.75 (t, J = 7.90 Hz, 1H), 7.64–7.61 (m, 1H), 7.36–7.31 (m, 1H), 7.29–7.26 (m, 2H), 6.06 (s, 1H), 4.13 (t, J = 7.7 Hz, 2H), 1.87–1.84 (m, 2H), 1.60–1.43 (m, 2H), 1.39–1.31 (m, 2H), 1.26 (s, 14H), 0.88 (t, J = 6.8 Hz, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.4, 139.0, 137.8, 134.4, 131.4, 130.1, 130.1, 123.6, 122.6, 121.3, 120.0, 116.9, 114.7, 113.2, 84.2, 44.6, 32.1, 29.8, 29.8, 29.8, 29.7, 29.5, 29.5, 27.3, 26.7, 22.9, 14.3. LRMS (EI) (m/z) (relative intensity): 402.3 (M^+, 100), 263.1 (92), 234.1 (66); HRMS calcd for $C_{27}H_{34}ON_2$ (M^+): 402.2671, found 402.2673.

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