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Synthesis of Spiro[indoline-3,2′-quinoline] Derivatives through a Four-Component Reaction

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An efficient synthetic procedure for the synthesis of functionalized tetrahydrospiro[indoline-3,2′-quinoline] derivatives was successfully developed and involves the four-component reaction of arylamines, dimethyl acetylenedicarboxylate, isatins, and a cyclohexane-1,3-dione in acetic acid. The advantages of this reaction include the use of common starting materials and mild reaction conditions and it is operationally simple.

Introduction

The spirooxindole system is the core structure of many pharmacological agents and natural alkaloids.[1,2] Spirooxindoles, especially those spiroannulated with heterocycles at the 3-position, have shown good biological activities.[3] These potential properties have prompted many efforts toward the synthesis of spirooxindole-fused heterocycles, and numerous impressive successes have been obtained for the synthesis of diversely structured spirocyclic oxindoles.[4,5] Isatin is probably one of the most widely used reagents for the construction of spirooxindoles and has been used in many reactions such as 1,3-dipolar cycloadditions,[6–8] Morita–Baylis–Hillman reactions,[9,10] and other condensation reactions.[11] In the past few years, multicomponent reactions based on the versatile reactivity of isatins have become efficient methods for the synthesis of various spirooxindoles.[12–16] On the other hand, the diverse reactivity of Huisgen 1,4-dipoles, which are easily formed from the addition reaction of nitrogen heterocycles to electron-deficient alkynes, has been widely used in multicomponent reactions to develop a number of carbon–carbon bond formation reactions and heterocyclic constructions.[17,18] Recently, several multicomponent reactions based on the sequential reactions of the initially formed Huisgen's 1,4-dipoles with isatins[19] or benzofuran-2,3-diones[20] have become efficient synthetic procedures for constructing versatile spirooxindole systems. Perumal and co-workers successfully reported the synthesis of spiro[indole-3,4′-pyridine] derivatives through a four-component reaction of arylamine, acetylenedicarboxylate, an aromatic aldehyde, and isatin.[21] As part of our program on the study of the reactivity of Huisgen's 1,4-dipoles derived from the reaction of electron-deficient alkynes with primary arylamines and developing new multicomponent reactions for the synthesis of heterocyclic compounds,[22,23] herein we wish to report the efficient synthesis of functionalized spiro[indoline-3,2′-quinoline] derivatives through the four-component reaction of arylamines, acetylenedicarboxylate, isatins, and cyclohexane-1,3-dione.

Results and Discussion

In an initial study, *p*-methoxyaniline was treated with dimethyl acetylenedicarboxylate in acetic acid to give the active intermediate β-enamino ester according to our previously established procedure.[22] Then, isatin and dimedone were added, and the mixture was stirred at ambient temperature overnight. After workup, a yellow solid was obtained in about 40% yield after separation by preparative thin-layer chromatography. Its structure was assigned as unusual tetrahydrospiro[indoline-3,2′-quinoline]-2,5′-dione (**1a**) and not the expected normal spiro[indoline-3,4′-quinoline] derivative (Scheme 1) according to the results of the reported multicomponent reactions containing isatin.[15,21] From the molecular structure of spiro compound **1a** it is clear that the β-enamino ester is absent in the final product, whereas the amino group of the arylamine is bridged between the isatin and dimedone molecules. In a second experiment, the three-component reaction of dimethyl acetylenedicarboxylate, dimedone, and isatin imine, which was prepared in situ from the reaction of *p*-methoxyaniline with isatin, was performed (Scheme 1). This three-component reaction also resulted in the formation of **1a** in a similar yield. In the next experiment, the four-component mixture of *p*-methoxyaniline, dimethyl acetylenedicarboxylate, isatin, and dimedone in acetic acid was run at ambient temperature overnight to give spiro compound **1a** in 46% yield. These results indicate that a one-pot four-component reaction could be established for the preparation of functionalized tetrahydrospiro[indoline-3,2′-quinoline]-2,5′-diones.

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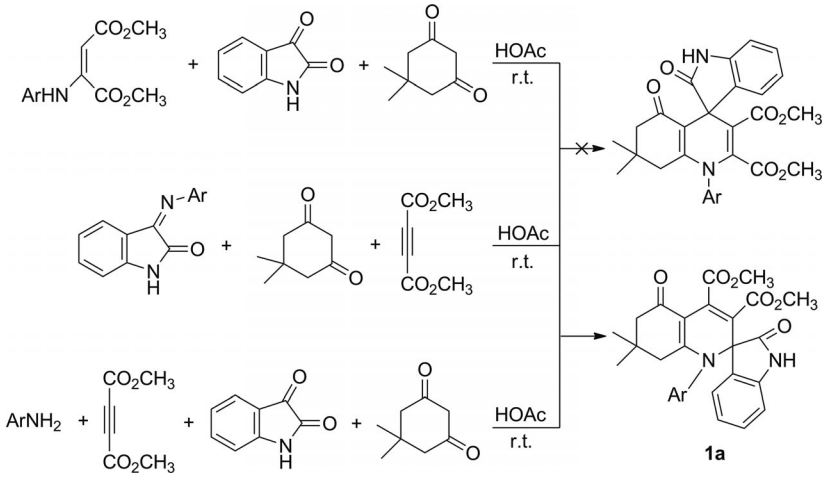
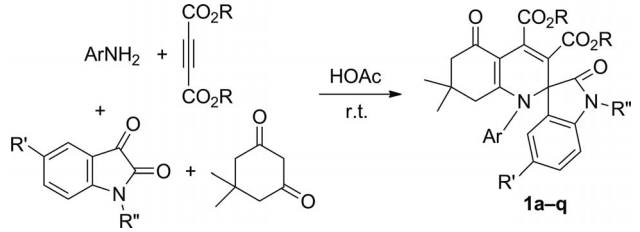


Scheme 1. Synthetic routes to tetrahydrospiro[indoline-3,2′-quinoline] (**1a**).

Having achieved inspiring results from the four-component reactions, we explored its cope and limitations. Under similar reaction conditions, various arylamines and isatins bearing a 5-methyl, 5-chloro, 1-benzyl, or 1-*n*-butyl group and diethyl acetylenedicarboxylate were used in the four-component reaction. All reactions proceeded smoothly at room temperature, and the corresponding spiro derivatives **1b**–**q** were prepared in moderate yields (Table 1). Products **1a**–**j** were fully characterized by elemental analysis; ^{1}H NMR, ^{13}C NMR, and IR spectroscopy; and mass spectrometry, and the structures were further confirmed by single-crystal X-ray diffraction studies performed for compounds **1e** (Figure 1) and **1j** (Supporting Information, Figure S1).

Table 1. Synthesis of spiro[indoline-3,2′-quinoline]s **1a**–**q** through a four-component reaction.



Entry	1	Ar	R	R′	R″	Yield [%]
1	**1a**	*p*-$CH_3OC_6H_4$	CH_3	H	H	46
2	**1b**	C_6H_5	CH_3	H	H	42
3	**1c**	*p*-ClC_6H_4	CH_3	H	H	43
4	**1d**	*p*-$CH_3OC_6H_4$	CH_3	CH_3	H	52
5	**1e**	*p*-$CH_3C_6H_4$	CH_3	CH_3	H	49
6	**1f**	*m*-$CH_3C_6H_4$	CH_3	CH_3	H	42
7	**1g**	C_6H_5	CH_3	CH_3	H	42
8	**1h**	*p*-ClC_6H_4	CH_3	CH_3	H	53
9	**1i**	*p*-$CH_3OC_6H_4$	CH_3	Cl	H	55
10	**1j**	C_6H_5	CH_3	Cl	H	40
11	**1k**	*p*-$CH_3C_6H_4$	CH_2CH_3	H	H	50
12	**1l**	*p*-$CH_3OC_6H_4$	CH_2CH_3	Cl	H	60
13	**1m**	*p*-ClC_6H_4	CH_2CH_3	H	H	55
14	**1n**	*p*-$CH_3C_6H_4$	CH_3	H	$CH_2C_6H_5$	61
15	**1o**	*p*-$CH_3OC_6H_4$	CH_2CH_3	CH_3	$CH_2C_6H_5$	67
16	**1p**	*p*-$CH_3C_6H_4$	CH_3	Cl	*n*-C_4H_9	65
17	**1q**	*p*-ClC_6H_4	CH_3	CH_3	*n*-C_4H_9	70

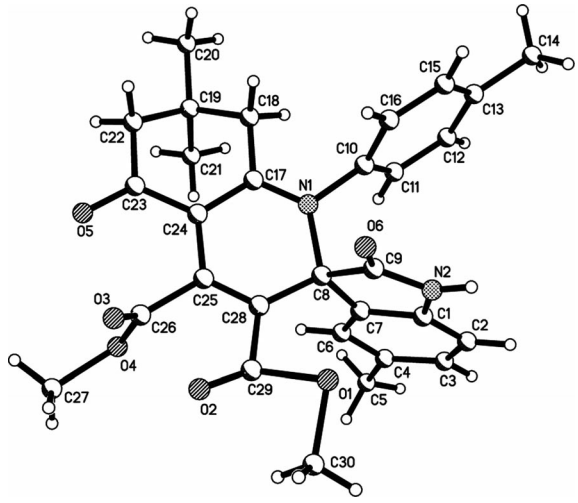
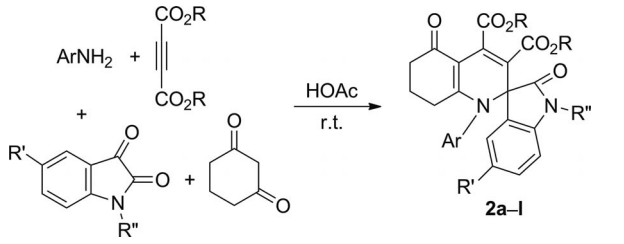


Figure 1. Molecular structure of spiro compound **1e**.

The preparation of tetrahydrospiro[indoline-3,2′-quinoline]-2,5′-diones is very exciting. In the literature, there are several multicomponent reactions containing isatin, cyclic 1,3-diketones, and aminoheterocycles, which give spiro[indoline-3,4′-quinoline] derivatives as main products.[15,21] However, to the best of our knowledge, there are very few synthetic methods for the preparation of the isomeric spiro[indoline-3,2′-quinoline] system.[16] Thus, our present protocol provides an efficient synthetic procedure for the preparation of spiro[indoline-3,2′-quinoline] derivatives. To broaden the scope of this four-component reaction, similar reactions containing cyclohexane-1,3-dione were also carried out and spiro[indoline-3,2′-quinoline] derivatives **2a**–**l** were prepared in good yields (Table 2). Their structures were established on the basis of spectroscopic data and single-crystal structure determination of spiro compounds **2e** (Figure 2) and **2k** (Supporting Information, Figure S2).

Table 2. Synthesis of spiro[indoline-3,2′-quinoline] **2a–l** through a four-component reaction.



Entry	2	Ar	R	R′	R′′	Yield [%]
1	**2a**	*p*-$CH_3OC_6H_4$	CH_3	CH_3	$CH_2C_6H_5$	72
2	**2b**	*p*-$CH_3C_6H_4$	CH_3	H	$CH_2C_6H_5$	62
3	**2c**	C_6H_5	CH_3	CH_3	$CH_2C_6H_5$	53
4	**2d**	*p*-ClC_6H_4	CH_3	H	$CH_2C_6H_5$	60
5	**2e**	*p*-$CH_3OC_6H_4$	CH_3	CH_3	*n*-C_4H_9	67
6	**2f**	*p*-$CH_3C_6H_4$	CH_3	CH_3	*n*-C_4H_9	61
7	**2g**	*p*-$CH_3OC_6H_4$	CH_3	F	*n*-C_4H_9	70
8	**2h**	*p*-$CH_3C_6H_4$	CH_3	Cl	*n*-C_4H_9	64
9	**2i**	*p*-ClC_6H_4	CH_3	CH_3	*n*-C_4H_9	59
10	**2j**	*m*-$NO_2C_6H_4$	CH_3	CH_3	*n*-C_4H_9	72
11	**2k**	*p*-ClC_6H_4	CH_2CH_3	CH_3	$CH_2C_6H_5$	51
12	**2l**	*p*-$CH_3C_6H_4$	CH_2CH_3	CH_3	H	78

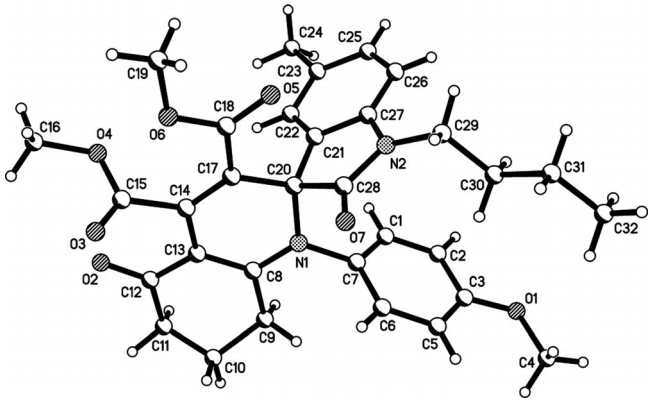
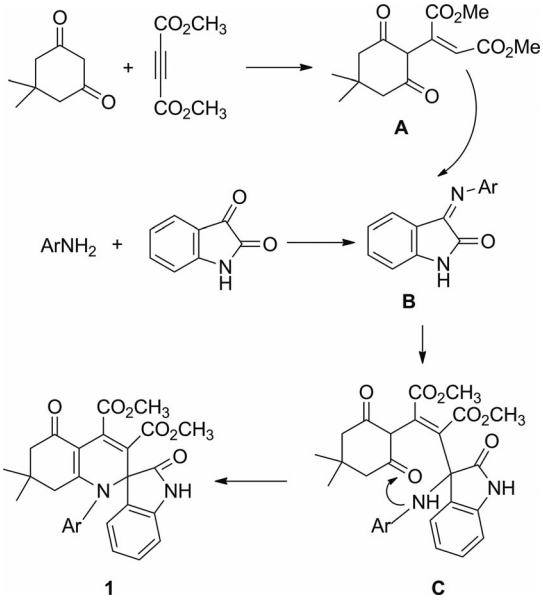


Figure 2. Molecular structure of spiro compound **2e**.

Although the exact mechanism of this reaction is not very clear at present, a plausible mechanism for the formation of spiro[indoline-3,2′-quinoline]s (Scheme 2) is presented on the basis of the analysis of similar multicomponent reactions of isatin[15,19] and our recently reported preparation of fused 1,2-dihydropyridines through a domino reaction involving arylamines, acetylenedicarboxylate, aromatic aldehydes, and dimedone.[23] Firstly, reaction of dimedone with dimethyl acetylenedicarboxylate in acetic acid forms adduct **A**, which has been described in several reports.[24] Secondly, the reaction of isatin with an arylamine in acetic acid forms isatin imine **B**. Then, Michael addition of intermediate **A** to imine **B** affords adduct **C**. Thirdly, intramolecular condensation of the amino group with the carbonyl group in intermediate **C** affords tetrahydrospiro[indoline-3,2′-quinoline]-2,5′-dione **1** after dehydration. This mechanism was partially supported by the two one-pot three-component reactions described in Scheme 1. In situ formed isatin imine **B** reacted with dimedone and dimethyl acetylenedicarboxylate to result in spiro compound **1**. On the other hand, if the β-enamino ester derived from the reaction of acetylenendicarboxylate and the arylamine reacted further with isatin and dimedone, the main product was still spiro compound **1**, because the β-enamino ester was in equilibrium with acetylenendicarboxylate and the arylamine in solution. Attempts to isolate intermediate **A** were not successful, and **A** was quickly converted into hexahydro-1-benzofuran-2,3-dicarboxylate through intramolecular Michael *O*-alkylation of the enolate to the C=C bond.[24] Alternatively, a reaction mechanism involving an imino-Diels–Alder reaction[25] might also be possible, in which intermediate **A** is converted into its enol form, and then the Diels–Alder reaction takes place between the enol and imine **B**. It is a pity that our results cannot distinguish between these two mechanisms at this time.



Scheme 2. Mechanism for the formation of tetrahydrospiro[indoline-3,2′-quinoline]-2,5′-dione.

Conclusions

In summary we have developed a four-component reaction for the efficient synthesis of functionalized tetrahydrospiro[indoline-3,2′-quinoline]s. The available diversity of each of the components used in the four-component reactions enables a wide range of substrates to be prepared with a broad range of physical properties. Furthermore, this reaction has several advantages including the use of common starting materials and mild reaction conditions and it is operationally simple. Potential uses of the reaction in synthetic and medicinal chemistry might be quite significant. Further expansion of the reaction scope and synthetic applications of this methodology are in progress in our laboratory.

Experimental Section

General Procedure for the Preparation of Spiro[indoline-3,2′-quinoline]-2,5′-diones 1a–q and 2a–l: A solution of arylamine (2.0 mmol), acetylenedicarboxylate (2.0 mmol), isatin (2.0 mmol), and cyclic 1,3-diketone (2.0 mmol) in acetic acid (5.0 mL) was stirred at room temperature for about 24 h. The resulting precipitates were collected by filtration. This crude product was subjected to preparative thin-layer chromatography (SiO_2; ethyl acetate/light petroleum, 1:3) and washed with ethanol to give the pure product for analysis.

1a: Yellow solid, yield: 46%, m.p. 270–271 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.81 (s, 1 H, NH), 7.53 (s, 1 H, ArH), 7.25 (br. s, 1 H, ArH), 7.19–7.18 (m, 1 H, ArH), 7.06 (br. s, 1 H, ArH), 6.85–6.84 (m, 1 H, ArH), 6.63 (br. s, 1 H, ArH), 6.53–6.52 (m, 1 H, ArH), 6.12–6.11 (m, 1 H, ArH), 3.90 (s, 3 H, OCH_3), 3.76 (s, 3 H, OCH_3), 3.50 (s, 3 H, OCH_3), 2.26 (s, 2 H, CH_2), 2.03–2.00 (m, 1 H, CH), 1.88 (d, J = 16.8 Hz, 1 H, CH), 0.96 (s, 6 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 164.0, 159.8, 140.0, 131.3, 131.1, 130.9, 130.1, 123.4, 114.4, 114.0, 110.1, 55.4, 52.4, 52.0, 49.9, 42.7, 32.0, 28.4, 28.0 ppm. IR (KBr): $\tilde{\nu}$ = 2953, 1744, 1610, 1502, 1429, 1246, 1206, 1133, 1105, 1019, 933, 856, 753 cm^{-1}. MS: m/z (%) = 515.31 (100) $[M - 1]^+$. $C_{29}H_{28}N_2O_7$ (516.55): calcd. C 67.43, H 5.46, N 5.42; found C 67.56, H 5.71, N 5.15.

1b: Yellow solid, yield: 42%, m.p. ~162–164 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.93 (s, 1 H, NH), 7.56–7.55 (m, 1 H, ArH), 7.38 (t, J = 7.2 Hz, 1 H, ArH), 7.32 (t, J = 7.2 Hz, 1 H, ArH), 7.30–7.28 (m, 1 H, ArH), 7.26–7.24 (m, 1 H, ArH), 7.10–7.05 (m, 2 H, ArH), 6.62–6.61 (m, 1 H, ArH), 6.23 (d, J = 7.2 Hz, 1 H, ArH), 3.91 (s, 3 H, OCH_3), 3.52 (s, 3 H, OCH_3), 2.28 (s, 2 H, CH_2), 2.01 (d, J = 16.8 Hz, 1 H, CH), 1.86 (d, J = 16.8 Hz, 1 H, CH), 0.96 (s, 6 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 176.0, 164.0, 139.8, 137.4, 131.2, 131.0, 130.0, 129.8, 129.6, 128.7, 123.6, 110.2, 52.4, 52.1, 49.8, 42.8, 28.0, 20.7 ppm. IR (KBr): $\tilde{\nu}$ = 2965, 1742, 1693, 1626, 1596, 1482, 1428, 1344, 1281, 1214, 1132, 1099, 1021, 890, 826, 765 cm^{-1}. MS: m/z (%) = 485.34 (100) $[M - 1]^+$. $C_{28}H_{26}N_2O_6$ (486.52): calcd. C 69.12, H 5.39, N 5.76; found C 68.75, H 5.53, N 5.68.

1c: Yellow solid, yield: 43%, m.p. 286–287 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.71 (s, 1 H, NH), 7.55 (br. s, 1 H, ArH), 7.39–7.37 (m, 1 H, ArH), 7.31–7.24 (m, 2 H, ArH), 7.12–7.04 (m, 2 H, ArH), 6.67 (br. s, 1 H, ArH), 6.18 (br. s, 1 H, ArH), 3.91 (s, 3 H, OCH_3), 3.54 (s, 3 H, OCH_3), 2.30–2.28 (m, 2 H, CH_2), 2.00 (br. s, 1 H, CH), 1.86–1.84 (m, 1 H, CH), 0.97 (s, 6 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 164.0, 139.8, 135.9, 135.7, 135.6, 131.7, 131.6, 131.2, 130.9, 130.1, 129.0, 123.7, 110.3, 52.5, 52.2, 49.8, 42.8, 32.1, 32.0, 28.4, 28.3, 28.2, 28.1 ppm. IR (KBr): $\tilde{\nu}$ = 3083, 2991, 2821, 1742, 1710, 1596, 1494, 1414, 1317, 1277, 1238, 1202, 1135, 1104, 1014, 856, 824, 777 cm^{-1}. MS: m/z (%) = 519.32 (100) $[M - 1]^+$, 521.28 (38) $[M - 1]^+$. $C_{28}H_{25}ClN_2O_6$ (520.97): calcd. C 64.55, H 4.84, N 5.38; found C 64.29, H 5.10, N 5.23.

1d: Yellow solid, yield: 52%, m.p. 270–272 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.35 (s, 1 H, ArH), 7.21 (d, J = 8.4 Hz, 1 H, ArH), 7.07 (d, J = 7.8 Hz, 1 H, ArH), 6.87 (d, J = 8.4 Hz, 1 H, ArH), 6.56–6.52 (m, 2 H, ArH), 6.13–6.12 (m, 1 H, ArH), 3.93 (s, 3 H, OCH_3), 3.78 (s, 3 H, OCH_3), 3.56 (s, 3 H, OCH_3), 2.39–2.32 (m, 5 H, CH_3, CH_2), 2.04 (d, J = 17.4 Hz, 1 H, CH), 1.90 (d, J = 17.4 Hz, 1 H, CH), 0.97 (s, 6 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 164.1, 159.8, 137.5, 133.1, 131.3, 131.2, 130.1, 114.3, 114.0, 109.9, 70.8, 55.4, 52.3, 51.9, 49.9, 42.7, 28.3, 28.1, 21.1 ppm. IR (KBr): $\tilde{\nu}$ = 2990, 1747, 1607, 1498, 1427, 1246, 1208, 1154, 1117, 1016, 864, 825 cm^{-1}. MS: m/z (%) = 529.36 (100) $[M - 1]^+$. $C_{30}H_{30}N_2O_7$ (530.58): calcd. C 67.91, H 5.70, N 5.28; found C 67.74, H 5.48, N 5.03.

1e: Yellow solid, yield: 49%, m.p. 272–274 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.35 (s, 1 H, ArH), 7.16 (br. s, 2 H, ArH), 7.05 (d, J = 7.2 Hz, 1 H, ArH), 6.84 (d, J = 7.2 Hz, 1 H, ArH), 6.51–6.50 (m, 1 H, ArH), 6.11–6.09 (m, 1 H, ArH), 3.91 (s, 3 H, OCH_3), 3.54 (s, 3 H, OCH_3), 2.33–2.31 (m, 6 H, CH_3), 2.27 (br. s, 2 H, CH_2), 2.02–2.00 (m, 1 H, CH), 1.87 (d, J = 16.8 Hz, 1 H, CH), 0.96 (s, 6 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 164.0, 139.6, 137.5, 134.9, 133.0, 131.4, 131.3, 130.3, 129.7, 129.3, 109.9, 70.7, 52.3, 51.9, 49.9, 42.7, 32.0, 28.3, 28.2, 28.1, 21.2, 21.1 ppm. IR (KBr): $\tilde{\nu}$ = 2950, 1748, 1707, 1601, 1493, 1431, 1380, 1302, 1197, 1153, 1122, 1017, 855, 822 cm^{-1}. MS: m/z (%) = 513.39 (100) $[M - 1]^+$. $C_{30}H_{30}N_2O_6$ (514.58): calcd. C 70.02, H 5.88, N 5.44; found C 69.84, H 6.15, N 5.37.

1f: Yellow solid, yield: 42%, m.p. 274–276 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.67 (d, J = 10.8 Hz, 1 H, NH), 7.34 (br. s, 1 H, ArH), 7.23 (t, J = 7.8 Hz, 1 H, ArH), 7.12–7.10 (m, 1 H, ArH), 7.05–7.04 (m, 2 H, ArH), 6.51 (br. s, 1 H, ArH), 5.99 (s, 1 H, ArH), 3.90 (s, 3 H, OCH_3), 3.50 (s, 3 H, OCH_3), 2.33–2.32 (m, 6 H, CH_3), 2.27 (s, 2 H, CH_2), 1.93–1.91 (m, 1 H, CH), 1.89–1.86 (m, 1 H, CH), 0.96 (s, 6 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 164.1, 139.8, 138.7, 137.6, 137.5, 137.4, 137.3, 133.1, 133.0, 131.5, 131.3, 131.2, 130.4, 130.2, 130.1, 129.3, 128.4, 126.9, 109.8, 109.7, 52.4, 51.9, 49.9, 42.8, 42.7, 32.0, 28.3, 28.2, 28.1, 28.0, 21.3, 21.1, 21.0 ppm. IR (KBr): $\tilde{\nu}$ = 2951, 1746, 1707, 1625, 1600, 1492, 1431, 1373, 1313, 1193, 1150, 1110, 1047, 1018, 831, 797 cm^{-1}. MS: m/z (%) = 513.34 (100) $[M - 1]^+$. $C_{30}H_{30}N_2O_6$ (514.58): calcd. C 70.02, H 5.88, N 5.44; found C 70.16, H 6.23, N 5.16.

1g: Yellow solid, yield: 42%, m.p. 266–268 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.80 (s, 1 H, NH), 7.40–7.37 (m, 2 H, ArH), 7.34–7.32 (m, 1 H, ArH), 7.29–7.27 (m, 1 H, ArH), 7.08–7.05 (m, 2 H, ArH), 6.51 (br. s, 2 H, ArH), 6.23 (d, J = 7.8 Hz, 1 H, ArH), 3.92 (s, 3 H, OCH_3), 3.53 (s, 3 H, OCH_3), 2.33 (s, 3 H, CH_3), 2.28 (s, 2 H, CH_2), 2.01 (d, J = 17.4 Hz, 1 H, CH), 1.87 (d, J = 17.4 Hz, 1 H, CH), 0.96 (s, 6 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 176.0, 164.0, 137.5, 137.3, 133.4, 131.4, 131.3, 130.1, 129.7, 129.5, 128.7, 109.9, 52.4, 52.0, 49.8, 42.8, 28.1, 21.1, 20.7 ppm. IR (KBr): $\tilde{\nu}$ = 2953, 1742, 1697, 1624, 1597, 1488, 1430, 1385, 1342, 1298, 1273, 1213, 1153, 1020, 828, 792, 761 cm^{-1}. MS: m/z (%) = 499.40 (100) $[M - 1]^+$. $C_{29}H_{28}N_2O_6$ (500.55): calcd. C 69.59, H 5.64, N 5.60; found C 69.27, H 5.92, N 5.40.

1h: Yellow solid, yield: 53%, m.p. 294–297 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.37–7.34 (m, 3 H, NH, ArH), 7.25–7.23 (m, 1 H, ArH), 7.07–7.04 (m, 2 H, ArH), 6.54–6.53 (m, 1 H, ArH), 6.18–6.17 (m, 1 H, ArH), 3.91 (s, 3 H, OCH_3), 3.55 (s, 3 H, OCH_3), 2.33 (s, 3 H, CH_3), 2.28 (s, 2 H, CH_2), 1.99 (d, J = 17.4 Hz, 1 H, CH), 1.86 (d, J = 17.4 Hz, 1 H, CH), 0.97 (s, 6 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 164.0, 137.2, 136.0, 135.7, 133.5, 131.7, 131.6, 131.0, 130.0, 128.9, 109.9, 52.4, 52.1, 49.9, 42.9, 28.2, 21.1 ppm. IR (KBr): $\tilde{\nu}$ = 3105, 2989, 1742, 1707, 1599, 1493, 1419, 1313, 1271, 1207, 1153, 1111, 1083, 1045, 1015, 857, 820 cm^{-1}. MS: m/z (%) = 533.29 (100) $[M - 1]^+$, 535.31 (35) $[M - 1]^+$. $C_{29}H_{27}ClN_2O_6$ (535.00): calcd. C 65.11, H 5.09, N 5.24; found C 64.83, H 5.46, N 5.47.

1i: Yellow solid, yield: 55%, m.p. 248–250 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.59 (s, 1 H, NH), 7.50 (s, 3 H, ArH), 7.23–7.22 (m, 1 H, ArH), 7.18 (d, J = 8.4 Hz, 1 H, ArH), 6.86 (d, J = 8.4 Hz, 1 H, ArH), 6.57 (d, J = 8.4 Hz, 2 H, ArH), 6.19 (d, J = 7.8 Hz, 1 H, ArH), 3.91 (s, 3 H, OCH_3), 3.77 (s, 3 H, OCH_3), 3.54 (s, 3 H, OCH_3), 2.27 (s, 2 H, CH_2), 2.00 (d, J = 17.1 Hz, 1 H, CH), 1.90 (d, J = 17.5 Hz, 1 H, CH), 0.96 (s, 6 H, CH_3) ppm. ^{13}C NMR

(150 MHz, $CDCl_3$): δ = 164.0, 160.0, 138.5, 132.8, 130.9, 129.8, 128.6, 114.6, 114.1, 111.1, 55.4, 52.4, 52.2, 49.8, 42.7, 32.0, 28.3, 28.2 ppm. IR (KBr): $\tilde{\nu}$ = 2949, 1741, 1714, 1604, 1493, 1430, 1377, 1344, 1292, 1256, 1214, 1177, 1136, 1021, 958, 883, 826, 784 cm^{-1}. MS: *m*/*z* (%) = 549.32 (100) $[M - 1]^+$, 551.30 (38) $[M - 1]^+$. $C_{29}H_{27}ClN_2O_7$ (550.99): calcd. C 63.22, H 4.94, N 5.08; found C 62.92, H 5.43, N 4.86.

1j: Yellow solid, yield: 40%, m.p. 252–254 °C. ^1H NMR (600 MHz, $CDCl_3$): δ = 7.86 (s, 1 H, NH), 7.52 (br. s, 1 H, ArH), 7.40 (t, *J* = 7.2 Hz, 1 H, ArH), 7.35 (t, *J* = 7.2 Hz, 1 H, ArH), 7.28 (s, 1 H, ArH), 7.23–7.22 (m, 1 H, ArH), 7.11 (t, *J* = 7.2 Hz, 1 H, ArH), 6.56 (br. s, 1 H, ArH), 6.31–6.30 (m, 1 H, ArH), 3.92 (s, 3 H, OCH_3), 3.55 (s, 3 H, OCH_3), 2.28 (s, 2 H, CH_2), 1.99 (d, *J* = 17.4 Hz, 1 H, CH), 1.89 (d, *J* = 17.4 Hz, 1 H, CH), 0.96 (s, 6 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 175.6, 164.0, 137.3, 132.6, 130.1, 130.0, 129.7, 128.9, 128.7, 111.2, 52.5, 52.2, 49.8, 42.8, 32.0, 28.2, 28.1, 20.7 ppm. IR (KBr): $\tilde{\nu}$ = 2954, 1746, 1597, 1485, 1430, 1343, 1278, 1215, 1136, 1019, 885, 830, 784, 757 cm^{-1}. MS: *m*/*z* (%) = 519.30 (100) $[M - 1]^+$, 521.28 (18) $[M - 1]^+$. $C_{28}H_{25}ClN_2O_6$ (520.97): calcd. C 64.55, H 4.84, N 5.38; found C 64.22, H 5.10, N 5.24.

1k: Yellow solid, yield: 50%, m.p.230–233 °C. ^1H NMR (600 MHz, $CDCl_3$): δ = 7.57 (d, *J* = 7.2 Hz, 1 H, NH), 7.32 (s, 1 H, ArH), 7.24 (br. s, 1 H, ArH), 7.16 (s, 2 H, ArH), 7.08–7.07 (m, 1 H, ArH), 6.83 (d, *J* = 7.8 Hz, 1 H, ArH), 6.60 (br. s, 1 H, ArH), 6.10 (d, *J* = 7.8 Hz, 1 H, ArH), 4.36 (br. s, 2 H, CH_2), 4.01–3.95 (m, 2 H, CH_2), 2.30 (s, 3 H, CH_3), 2.25 (br. s, 2 H, CH_2), 1.99 (d, *J* = 17.4 Hz, 1 H, CH), 1.86 (d, *J* = 17.4 Hz, 1 H, CH), 1.37 (br. s, 3 H, CH_3), 1.08 (br. s, 3 H, CH_3), 0.95 (s, 6 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 163.7, 139.5, 135.0, 130.7, 130.3, 129.3, 123.4, 109.8, 61.3, 61.1, 50.0, 42.8, 28.8, 28.2, 21.2, 13.9 ppm. IR (KBr): $\tilde{\nu}$ = 3165, 2942, 1735, 1605, 1491, 1424, 1261, 1200, 1128, 1029, 934, 866, 824, 765 cm^{-1}. MS: *m*/*z* (%) = 527.42 (100) $[M - 1]^+$. $C_{31}H_{32}N_2O_6$ (528.60): calcd. C 70.44, H 6.10, N 5.30; found C 70.25, H 6.52, N 5.01.

1l: Yellow solid, yield: 60%, m.p. 268–270 °C. ^1H NMR (600 MHz, $CDCl_3$): δ = 7.54 (br. s, 1 H, NH), 7.23 (d, *J* = 6.6 Hz, 1 H, ArH), 7.19 (d, *J* = 8.4 Hz, 1 H, ArH), 7.13 (br. s, 1 H, ArH), 6.87 (d, *J* = 7.8 Hz, 1 H, ArH), 6.58–6.55 (m, 2 H, ArH), 6.19 (d, *J* = 7.2 Hz, 1 H, ArH), 4.41–4.35 (m, 2 H, CH_2), 4.05–3.99 (m, 2 H, CH_2), 3.78 (s, 3 H, OCH_3), 2.27 (s, 2 H, CH_2), 1.99 (d, *J* = 17.4 Hz, 1 H, CH), 1.90 (d, *J* = 17.4 Hz, 1 H, CH), 1.38 (br. s, 3 H, CH_3), 1.14–1.12 (m, 3 H, CH_3), 0.97 (s, 6 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 163.7, 159.9, 130.8, 130.0, 114.6, 114.1, 110.9, 61.5, 61.3, 55.4, 50.0, 42.8, 28.4, 13.9 ppm. IR (KBr): $\tilde{\nu}$ = 3107, 2965, 1739, 1703, 1605, 1495, 1423, 1372, 1291, 1256, 1198, 1132, 1025, 875, 825, 780 cm^{-1}. MS: *m*/*z* (%) = 577.58 (100) $[M - 1]^+$. $C_{31}H_{31}ClN_2O_7$ (579.05): calcd. C 64.30, H 5.40, N 4.84; found C 64.57, H 5.62, N 4.59.

1m: Yellow solid, yield: 55%, m.p. 292–293 °C. ^1H NMR (600 MHz, $CDCl_3$): δ = 7.60 (br. s, 1 H, NH), 7.56 (d, *J* = 7.2 Hz, 1 H, ArH), 7.37–7.36 (m, 1 H, ArH), 7.26–7.24 (m, 2 H, ArH), 7.08 (t, *J* = 7.2 Hz, 1 H, ArH), 7.04–7.03 (m, 1 H, ArH), 6.64–6.63 (m, 1 H, ArH), 6.18 (d, *J* = 7.8 Hz, 1 H, ArH), 4.36 (br. s, 2 H, CH_2), 4.01–3.95 (m, 2 H, CH_2), 2.26 (s, 2 H, CH_2), 1.98 (d, *J* = 17.4 Hz, 1 H, CH), 1.84 (d, *J* = 17.4 Hz, 1 H, CH), 1.37 (br. s, 3 H, CH_3), 1.09–1.07 (m, 3 H, CH_3), 0.96 (s, 6 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 167.6, 163.6, 139.7, 136.1, 135.6, 131.0, 130.0, 128.9, 123.6, 110.1, 61.4, 61.2, 49.9, 42.9, 28.2, 28.1, 13.9 ppm. IR (KBr): $\tilde{\nu}$ = 3081, 2987, 2881, 2814, 1740, 1703, 1598, 1492, 1413, 1309, 1274, 1239, 1199, 1136, 1097, 1023, 919, 859, 826 cm^{-1}. MS: *m*/*z* (%) = 547.37 (100) $[M - 1]^+$. $C_{30}H_{29}ClN_2O_6$ (549.02): calcd. C 65.63, H 5.32, N 5.10; found C 65.44, H 5.50, N 4.87.

1n: Yellow solid, yield: 61%, m.p. 225–227 °C. ^1H NMR (600 MHz, $CDCl_3$): δ = 7.59 (br. s, 1 H, ArH), 7.20–7.14 (m, 6 H, ArH), 7.07 (t, *J* = 7.8 Hz, 1 H, ArH), 6.84 (d, *J* = 7.8 Hz, 1 H, ArH), 6.71 (d, *J* = 7.2 Hz, 2 H, ArH), 6.42 (d, *J* = 6.6 Hz, 1 H, ArH), 6.09–6.08 (m, 1 H, ArH), 4.79 (d, *J* = 16.2 Hz, 1 H, CH), 4.30 (d, *J* = 16.2 Hz, 1 H, CH), 3.92 (s, 3 H, OCH_3), 3.51 (s, 3 H, OCH_3), 2.36 (s, 3 H, CH_3), 2.26 (s, 2 H, CH_2), 1.98 (d, *J* = 17.4 Hz, 1 H, CH), 1.83 (d, *J* = 17.4 Hz, 1 H, CH), 0.96 (s, 3 H, CH_3), 0.95 (s, 3 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 163.9, 142.1, 139.7, 135.2, 130.8, 130.5, 129.5, 128.4, 127.3, 127.2, 123.6, 109.4, 52.3, 52.0, 49.9, 44.5, 42.7, 28.1, 21.3 ppm. IR (KBr): $\tilde{\nu}$ = 2953, 1729, 1642, 1605, 1498, 1429, 1366, 1306, 1251, 1192, 1117, 1026, 826 cm^{-1}. MS: *m*/*z* (%) = 589.56 (100) $[M - 1]^+$. $C_{36}H_{34}N_2O_6$ (590.67): calcd. C 73.20, H 5.80, N 4.74; found C 72.86, H 6.23, N 4.49.

1o: Yellow solid, yield: 67%, m.p. 246–249 °C. ^1H NMR (600 MHz, $CDCl_3$): δ = 7.39–7.38 (m, 1 H, ArH), 7.25–7.23 (m, 1 H, ArH), 7.20–7.17 (m, 1 H, ArH), 7.16–7.13 (m, 2 H, ArH), 6.99 (d, *J* = 7.8 Hz, 1 H, ArH), 6.86 (d, *J* = 7.2 Hz, 1 H, ArH), 6.67 (d, *J* = 7.2 Hz, 2 H, ArH), 6.54–6.52 (m, 1 H, ArH), 6.33 (d, *J* = 7.2 Hz, 1 H, ArH), 6.11–6.10 (m, 1 H, ArH), 4.81 (d, *J* = 16.2 Hz, 1 H, CH), 4.27 (d, *J* = 16.2 Hz, 1 H, CH), 3.93 (s, 3 H, OCH_3), 3.79 (s, 3 H, OCH_3), 3.52 (s, 3 H, OCH_3), 2.31 (s, 3 H, CH_3), 2.28 (s, 2 H, CH_2), 1.99 (d, *J* = 17.4 Hz, 1 H, CH), 1.86 (d, *J* = 17.4 Hz, 1 H, CH), 0.97 (s, 3 H, CH_3), 0.96 (s, 3 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 164.0, 160.0, 139.8, 135.2, 133.3, 131.5, 131.3, 130.4, 128.3, 127.3, 127.1, 114.4, 109.2, 55.4, 52.3, 52.1, 49.9, 44.4, 42.7, 28.2, 21.1 ppm. IR (KBr): $\tilde{\nu}$ = 2952, 1734, 1606, 1494, 1434, 1367, 1300, 1233, 1186, 1136, 1024, 979, 943, 873, 824, 763 cm^{-1}. MS: *m*/*z* (%) = 619.42 (100) $[M - 1]^+$. $C_{37}H_{36}N_2O_7$ (620.70): calcd. C 71.60, H 5.85, N 4.51; found C 71.42, H 6.15, N 4.48.

1p: Yellow solid, yield: 65%, m.p. 274–275 °C. ^1H NMR (600 MHz, $CDCl_3$): δ = 7.52 (br. s, 1 H, ArH), 7.29 (br. s, 1 H, ArH), 7.19 (d, *J* = 7.8 Hz, 1 H, ArH), 7.14–7.13 (m, 1 H, ArH), 6.86 (d, *J* = 7.2 Hz, 1 H, ArH), 6.52–6.51 (m, 1 H, ArH), 6.10 (d, *J* = 7.2 Hz, 1 H, ArH), 3.93 (s, 3 H, OCH_3), 3.54 (s, 3 H, OCH_3), 3.47–3.42 (m, 1 H, CH), 3.08 (br. s, 1 H, CH), 2.31 (s, 3 H, CH_3), 2.28 (br. s, 2 H, CH_2), 1.99 (d, *J* = 17.4 Hz, 1 H, CH), 1.87 (d, *J* = 17.4 Hz, 1 H, CH), 1.04 (br. s, 3 H, CH, CH_2), 0.97 (s, 3 H, CH_3), 0.96 (s, 3 H, CH_3), 0.80 (t, *J* = 7.2 Hz, 3 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 190.4, 189.4, 163.8, 163.7, 141.0, 139.9, 134.8, 132.4, 130.8, 130.3, 130.1, 130.0, 129.3, 128.4, 109.3, 52.4, 52.1, 49.9, 42.7, 40.1, 32.0, 31.9, 28.9, 28.2, 21.1, 20.1, 13.8 ppm. IR (KBr): $\tilde{\nu}$ = 2952, 1738, 1689, 1642, 1600, 1487, 1428, 1295, 1227, 1189, 1136, 1012, 879, 820 cm^{-1}. MS: *m*/*z* (%) = 613.92 (100) $[M + Na]^+$. $C_{33}H_{35}ClN_2O_6$ (591.10): calcd. C 67.05, H 5.97, N 4.74; found C 66.71, H 6.16, N 4.66.

1q: Yellow solid, yield: 70%, m.p. 250–251 °C. ^1H NMR (600 MHz, $CDCl_3$): δ = 7.37 (d, *J* = 7.2 Hz, 2 H, ArH), 7.24 (d, *J* = 8.4 Hz, 1 H, ArH), 7.12 (d, *J* = 7.8 Hz, 1 H, ArH), 7.03–7.02 (m, 1 H, ArH), 6.50 (d, *J* = 7.8 Hz, 1 H, ArH), 6.10 (d, *J* = 8.4 Hz, 1 H, ArH), 3.92 (s, 3 H, OCH_3), 3.52 (s, 3 H, OCH_3), 3.43–3.42 (m, 1 H, CH), 3.09 (br. s, 1 H, CH), 2.34 (s, 3 H, CH_3), 2.28 (br. s, 2 H, CH_2), 1.97 (d, *J* = 17.4 Hz, 1 H, CH), 1.83 (d, *J* = 17.4 Hz, 1 H, CH), 1.12–1.05 (m, 4 H, CH_2), 0.97 (s, 3 H, CH_3), 0.96 (s, 3 H, CH_3), 0.84 (t, *J* = 7.2 Hz, 3 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 163.7, 139.8, 136.1, 135.7, 133.2, 132.0, 131.5, 130.6, 129.8, 128.8, 108.3, 70.2, 52.4, 52.0, 49.9, 42.8, 40.0, 32.1, 29.1, 28.3, 28.2, 21.1, 20.1, 13.8 ppm. IR (KBr): $\tilde{\nu}$ = 2949, 1736, 1697, 1637, 1599, 1493, 1425, 1360, 1295, 1187, 1105, 1011, 817 cm^{-1}. MS: *m*/*z* (%)

= 613.90 (100) [M + Na]$^+$. $C_{33}H_{35}ClN_2O_6$ (591.10): calcd. C 67.05, H 5.97, N 4.74; found C 67.34, H 6.19, N 4.46.

2a: Yellow solid, yield: 72%, m.p. 236–238 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.41 (br. s, 1 H, ArH), 7.27 (br. s, 1 H, ArH), 7.19–7.17 (m, 1 H, ArH), 7.14 (t, J = 7.2 Hz, 2 H, ArH), 6.99 (d, J = 7.8 Hz, 1 H, ArH), 6.84–6.83 (m, 1 H, ArH), 6.66 (d, J = 7.2 Hz, 2 H, ArH), 6.54–6.52 (m, 1 H, ArH), 6.31 (d, J = 7.8 Hz, 1 H, ArH), 6.15 (d, J = 7.8 Hz, 1 H, ArH), 4.82 (d, J = 16.2 Hz, 1 H, CH), 4.28 (d, J = 16.2 Hz, 1 H, CH), 3.92 (s, 3 H, OCH_3), 3.77 (s, 3 H, OCH_3), 3.51 (s, 3 H, OCH_3), 2.40 (br. s, 2 H, CH_2), 2.32 (s, 3 H, CH_3), 2.15–2.11 (m, 1 H, CH), 2.04–1.99 (m, 1 H, CH), 1.86–1.84 (m, 2 H, CH_2) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 190.9, 174.2, 168.5, 163.9, 160.0, 139.7, 135.2, 133.2, 131.4, 131.3, 130.7, 130.3, 130.2, 128.4, 127.3, 127.0, 125.2, 114.4, 114.2, 109.1, 70.5, 55.4, 52.3, 44.3, 36.3, 29.5, 21.1, 20.4 ppm. IR (KBr): $\tilde{\nu}$ = 2949, 1731, 1606, 1498, 1422, 1366, 1335, 1293, 1247, 1189, 1115, 1026, 994, 948, 844, 759 cm^{-1}. MS: m/z (%) = 615.78 (100) [M + Na]$^+$. $C_{35}H_{32}N_2O_7$ (592.65): calcd. C 70.93, H 5.44, N 4.73; found C 70.68, H 5.70, N 4.65.

2b: Yellow solid, yield: 62%, m.p. 259–260 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.62 (d, J = 7.2 Hz, 1 H, ArH), 7.24–7.14 (m, 6 H, ArH), 7.07 (t, J = 7.8 Hz, 1 H, ArH), 6.84 (d, J = 8.4 Hz, 1 H, ArH), 6.71 (d, J = 7.2 Hz, 2 H, ArH), 6.41 (d, J = 7.8 Hz, 1 H, ArH), 6.13 (d, J = 7.8 Hz, 1 H, ArH), 4.79 (d, J = 16.2 Hz, 1 H, CH), 4.32 (d, J = 16.2 Hz, 1 H, CH), 3.92 (s, 3 H, OCH_3), 3.51 (s, 3 H, OCH_3), 2.40 (br. s, 2 H, CH_2), 2.34 (s, 3 H, CH_3), 2.13–2.08 (m, 1 H, CH), 2.01–1.96 (m, 1 H, CH), 1.88–1.81 (m, 2 H, CH_2) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 190.9, 174.3, 163.9, 142.1, 139.6, 135.1, 135.0, 130.8, 130.4, 129.9, 129.4, 129.3, 128.4, 127.3, 127.1, 123.5, 109.4, 70.2, 60.4, 52.4, 52.1, 44.4, 36.3, 29.6, 21.3, 21.0, 20.4, 14.2 ppm. IR (KBr): $\tilde{\nu}$ = 2950, 1730, 1645, 1609, 1500, 1413, 1368, 1265, 1231, 1188, 1140, 1003, 937, 751 cm^{-1}. MS: m/z (%) = 585.63 (100) [M + Na]$^+$. $C_{34}H_{30}N_2O_6$ (562.62): calcd. C 72.58, H 5.37, N 4.98; found C 72.33, H 5.62, N 4.67.

2c: Yellow solid, yield: 53%, m.p. 254–255 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.42 (br. s, 1 H, ArH), 7.37 (br. s, 3 H, ArH), 7.19–7.13 (m, 1 H, ArH), 7.15–7.13 (m, 2 H, ArH), 7.07–7.04 (m, 1 H, ArH), 6.98 (d, J = 7.8 Hz, 1 H, ArH), 6.67 (d, J = 7.8 Hz, 2 H, ArH), 6.29–6.26 (m, 2 H, ArH), 4.66 (d, J = 16.2 Hz, 1 H, CH), 4.32 (d, J = 16.2 Hz, 1 H, CH), 3.93 (s, 3 H, OCH_3), 3.50 (s, 3 H, OCH_3), 2.41 (br. s, 2 H, CH_2), 2.33 (s, 3 H, CH_3), 2.14–2.10 (m, 1 H, CH), 2.02–1.99 (m, 1 H, CH), 1.86–1.83 (m, 2 H, CH_2) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 174.1, 163.9, 139.7, 137.7, 135.2, 133.3, 131.3, 130.3, 129.8, 129.5, 128.8, 128.5, 127.3, 127.0, 109.2, 109.2, 70.4, 52.4, 52.1, 44.4, 36.3, 29.6, 21.1, 20.5 ppm. IR (KBr): $\tilde{\nu}$ = 2949, 1738, 1701, 1640, 1603, 1494, 1423, 1365, 1297, 1237, 1187, 1111, 993, 818, 756 cm^{-1}. MS: m/z (%) = 585.75 (100) [M + Na]$^+$. $C_{34}H_{30}N_2O_6$ (562.62): calcd. C 72.58, H 5.37, N 4.98; found C 72.20, H 5.77, N 4.82.

2d: Yellow solid, yield: 60%, m.p. 244–247 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.61 (d, J = 7.2 Hz, 1 H, ArH), 7.30 (s, 2 H, ArH), 7.24–7.21 (m, 4 H, ArH), 7.08 (t, J = 7.8 Hz, 1 H, ArH), 7.01–6.99 (m, 1 H, ArH), 6.72–6.71 (m, 2 H, ArH), 6.48 (d, J = 7.8 Hz, 1 H, ArH), 6.18 (d, J = 8.4 Hz, 1 H, ArH), 4.84 (d, J = 16.2 Hz, 1 H, CH), 4.32 (d, J = 16.2 Hz, 1 H, CH), 3.92 (s, 3 H, OCH_3), 3.51 (s, 3 H, OCH_3), 2.41–2.36 (m, 2 H, CH_2), 2.10–2.07 (m, 1 H, CH), 2.00–1.95 (m, 1 H, CH), 1.89–1.85 (m, 2 H, CH_2) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 190.9, 174.1, 168.2, 142.0, 136.1, 135.7, 134.9, 131.8, 130.1, 129.1, 128.6, 127.6, 127.1, 123.7, 109.5, 70.2, 58.3, 52.4, 52.1, 44.5, 36.2, 29.6, 20.4 ppm. IR (KBr): $\tilde{\nu}$ = 2950, 1735, 1640, 1604, 1495, 1412, 1367, 1285, 1226, 1180, 1139, 1004, 935, 863 cm^{-1}. MS: m/z (%) = 605.96 (100) [M + Na]$^+$. $C_{33}H_{27}ClN_2O_6$ (583.04): calcd. C 67.98, H 4.67, N 4.80; found C 67.70, H 4.76, N 4.43.

2e: Yellow solid, yield: 67%, m.p. 222–225 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.38 (s, 1 H, ArH), 7.23–7.22 (m, 1 H, ArH), 7.10 (d, J = 7.8 Hz, 1 H, ArH), 6.86–6.84 (m, 1 H, ArH), 6.52–7.50 (m, 1 H, ArH), 6.47 (d, J = 7.8 Hz, 1 H, ArH), 6.10–6.08 (m, 1 H, ArH), 3.92 (s, 3 H, OCH_3), 3.74 (s, 3 H, OCH_3), 3.51 (s, 3 H, OCH_3), 3.44–3.41 (m, 1 H, CH), 3.11–3.08 (m, 1 H, CH), 2.38 (br. s, 2 H, CH_2), 2.35 (s, 3 H, CH_3), 2.15–2.11 (m, 1 H, CH), 2.03–1.98 (m, 1 H, CH), 1.87–1.82 (m, 2 H, CH_2), 1.07–1.05 (m, 3 H, CH, CH_2), 0.96 (br. s, 1 H, CH), 0.80 (t, J = 7.2 Hz, 3 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 190.9, 190.8, 173.6, 163.9, 159.9, 139.9, 132.8, 131.4, 131.2, 130.1, 114.1, 113.7, 108.1, 70.2, 55.4, 52.2, 51.8, 39.9, 36.2, 29.5, 29.2, 21.0, 20.4, 20.1, 13.7 ppm. IR (KBr): $\tilde{\nu}$ = 2947, 1718, 1600, 1488, 1429, 1368, 1336, 1293, 1248, 1192, 1019, 844, 798, 750 cm^{-1}. MS: m/z (%) = 581.88 (100) [M + Na]$^+$. $C_{32}H_{34}N_2O_7$ (558.63): calcd. C 68.80, H 6.13, N 5.01; found C 68.53, H 6.45, N 4.79.

2f: Yellow solid, yield: 61%, m.p. 215–218 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.39 (s, 1 H, ArH), 7.19 (d, J = 7.8 Hz, 1 H, ArH), 7.16 (d, J = 7.8 Hz, 1 H, ArH), 7.09 (d, J = 7.8 Hz, 1 H, ArH), 6.81 (d, J = 7.8 Hz, 1 H, ArH), 6.46 (d, J = 7.8 Hz, 1 H, ArH), 6.06 (d, J = 7.8 Hz, 1 H, ArH), 3.92 (s, 3 H, OCH_3), 3.51 (s, 3 H, OCH_3), 3.45–3.40 (m, 1 H, CH), 3.09–3.04 (m, 1 H, CH), 2.39 (br. s, 2 H, CH_2), 2.35 (s, 3 H, CH_3), 2.28 (s, 3 H, CH_3), 2.15–2.11 (m, 1 H, CH), 2.02–1.98 (m, 1 H, CH), 1.85–1.81 (m, 2 H, CH_2), 1.04–1.00 (m, 3 H, CH, CH_2), 0.89–0.88 (m, 1 H, CH), 0.79 (t, J = 6.6 Hz, 3 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 190.9, 190.8, 173.5, 163.8, 139.9, 139.5, 134.9, 132.8, 131.2, 130.0, 129.0, 128.8, 108.0, 70.1, 52.3, 51.9, 39.9, 36.3, 29.5, 29.0, 21.1, 20.4, 20.1, 13.8 ppm. IR (KBr): $\tilde{\nu}$ = 2951, 1735, 1689, 1636, 1601, 1494, 1426, 1365, 1297, 1236, 1191, 1147, 1027, 980, 815 cm^{-1}. MS: m/z (%) = 565.92 (100) [M + Na]$^+$. $C_{32}H_{34}N_2O_6$ (542.63): calcd. C 70.83, H 6.32, N 5.16; found C 70.72, H 6.56, N 4.85.

2g: Yellow solid, yield: 70%, m.p. 234–235 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.34 (br. s, 1 H, ArH), 7.23 (d, J = 8.4 Hz, 1 H, ArH), 7.01 (t, J = 7.8 Hz, 1 H, ArH), 6.86 (d, J = 6.6 Hz, 1 H, ArH), 6.54–6.51 (m, 2 H, ArH), 6.16 (d, J = 7.2 Hz, 1 H, ArH), 3.92 (s, 3 H, OCH_3), 3.75 (s, 3 H, OCH_3), 3.53 (s, 3 H, OCH_3), 3.46–3.42 (m, 1 H, CH), 3.13 (br. s, 1 H, CH), 2.39 (br. s, 2 H, CH_2), 2.15–2.12 (m, 1 H, CH), 2.03–2.00 (m, 1 H, CH), 1.86 (m, 2 H, CH_2), 1.08–1.04 (m, 3 H, CH, CH_2), 1.00–0.99 (m, 1 H, CH), 0.81 (t, J = 7.2 Hz, 3 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 190.9, 190.8, 173.6, 163.8, 160.1, 158.5, 138.5, 131.4, 130.0, 117.3, 117.2, 114.3, 113.9, 109.0, 108.9, 70.2, 55.4, 52.4, 52.0, 40.1, 36.3, 29.5, 29.1, 20.4, 20.1, 13.7 ppm. IR (KBr): $\tilde{\nu}$ = 2956, 1737, 1687, 1633, 1600, 1491, 1433, 1363, 1299, 1249, 1187, 1144, 1031, 991, 857, 819 cm^{-1}. MS: m/z (%) = 585.83 (100) [M + Na]$^+$. $C_{31}H_{31}FN_2O_7$ (562.59): calcd. C 66.18, H 5.55, N 4.98; found C 65.80, H 5.79, N 4.86.

2h: Yellow solid, yield: 64%, m.p. 252–253 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.54 (br. s, 1 H, ArH), 7.28 (br. s, 1 H, ArH), 7.17 (br. s, 2 H, ArH), 6.85 (d, J = 7.8 Hz, 1 H, ArH), 6.50 (d, J = 8.4 Hz, 1 H, ArH), 6.13 (d, J = 8.4 Hz, 1 H, ArH), 3.93 (s, 3 H, OCH_3), 3.53 (s, 3 H, OCH_3), 3.46–3.43 (m, 1 H, CH), 3.08 (br. s, 1 H, CH), 2.40 (br. s, 2 H, CH_2), 2.29 (s, 3 H, CH_3), 2.15–2.09 (m, 1 H, CH), 2.03–1.99 (m, 1 H, CH), 1.86–1.85 (m, 2 H, CH_2), 1.04–1.03 (m, 3 H, CH, CH_2), 0.90 (br. s, 1 H, CH), 0.80 (t, J = 7.2 Hz, 3 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 190.9, 190.8, 173.4, 168.2, 163.7, 141.0, 139.9, 134.7, 130.8, 130.2, 130.0, 129.2, 128.7, 128.3, 109.3, 69.9, 52.3, 52.1, 40.1, 36.2, 29.5, 28.9, 21.1, 20.4, 20.1, 13.8 ppm. IR (KBr): $\tilde{\nu}$ = 2956, 2871, 1737, 1686, 1638, 1602, 1489,

1427, 1365, 1299, 1227, 1188, 1141, 1028, 989, 937, 894, 821, 789 cm^{-1}. MS: *m/z* (%) = 585.93 (100) [M + Na]$^+$. $C_{31}H_{31}ClN_2O_6$ (563.05): calcd. C 66.13, H 5.55, N 4.98; found C 66.37, H 5.94, N 4.70.

2i: Yellow solid, yield: 59%, m.p. 241–244 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.38–7.35 (m, 2 H, ArH), 7.29 (d, J = 8.4 Hz, 1 H, ArH), 7.12 (d, J = 7.8 Hz, 1 H, ArH), 7.02–7.00 (m, 1 H, ArH), 6.49 (d, J = 7.8 Hz, 1 H, ArH), 6.15–6.13 (m, 1 H, ArH), 3.92 (s, 3 H, OCH_3), 3.52 (s, 3 H, OCH_3), 3.45–3.41 (m, 1 H, CH), 3.11–3.09 (m, 1 H, CH), 2.40 (br. s, 2 H, CH_2), 2.35 (s, 3 H, CH_3), 2.13–2.09 (m, 1 H, CH), 2.01–1.97 (m, 1 H, CH), 1.88–1.86 (m, 2 H, CH_2), 1.08–1.05 (m, 3 H, CH, CH_2), 0.94 (br. s, 1 H, CH), 0.84 (t, J = 7.2 Hz, 3 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 190.9, 173.4, 168.3, 163.7, 163.6, 163.5, 139.8, 136.0, 135.6, 133.1, 131.9, 131.5, 130.5, 129.7, 128.7, 125.3, 125.2, 108.3, 70.1, 52.4, 52.0, 40.0, 36.2, 29.6, 29.1, 21.1, 20.4, 20.1, 13.8 ppm. IR (KBr): $\tilde{\nu}$ = 2946, 1733, 1698, 1643, 1603, 1494, 1422, 1365, 1296, 1236, 1191, 1100, 987, 815 cm^{-1}. MS: *m/z* (%) = 585.97 (100) [M + Na]$^+$. $C_{31}H_{31}ClN_2O_6$ (563.05): calcd. C 66.13, H 5.55, N 4.98; found C 65.82, H 5.675, N 4.80.

2j: Yellow solid, yield: 72%, m.p. 235–236 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 8.18 (d, J = 7.2 Hz, 1 H, ArH), 7.70 (d, J = 6.6 Hz, 1 H, ArH), 7.60 (t, J = 7.8 Hz, 1 H, ArH), 7.45 (s, 1 H, ArH), 7.25 (br. s, 1 H, ArH), 7.15 (d, J = 7.2 Hz, 1 H, ArH), 7.10 (br. s, 1 H, ArH), 3.93 (s, 3 H, OCH_3), 3.53 (s, 3 H, OCH_3), 3.38–3.36 (m, 1 H, CH), 3.14–3.12 (m, 1 H, CH), 2.43–2.42 (m, 2 H, CH_2), 2.39 (s, 3 H, CH_3), 2.12–2.09 (m, 1 H, CH), 2.03–1.90 (m, 3 H, CH_2, CH), 1.07–0.95 (m, 4 H, CH_2), 0.94 (br. s, 1 H, CH), 0.80–079 (m, 3 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 190.9, 173.2, 168.1, 163.5, 148.5, 14.7, 139.8, 139.4, 138.6, 138.4, 137.0, 135.7, 133.8, 133.3, 132.0, 131.7, 130.3, 130.0, 129.5, 125.9, 125.4, 125.3, 125.2, 124.7, 124.2, 108.5, 108.4, 70.2, 70.1, 52.4, 52.1, 40.0, 39.9, 36.2, 29.8, 29.1, 29.0, 21.1, 20.5, 20.0, 19.9, 13.6 ppm. IR (KBr): $\tilde{\nu}$ = 2950, 1728, 1644, 1607, 1499, 1418, 1350, 1292, 1189, 994, 901, 810 cm^{-1}. MS: *m/z* (%) = 596.92 (100) [M + Na]$^+$. $C_{31}H_{31}N_3O_8$ (573.60): calcd. C 64.91, H 5.45, N 7.33; found C 64.78, H 5.81, N 7.05.

2k: Yellow solid, yield: 51%, m.p. 242–243 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 7.42 (s, 1 H, ArH), 7.29 (br. s, 2 H, ArH), 7.22 (br. s, 3 H, ArH), 7.01 (d, J = 7.8 Hz, 2 H, ArH), 6.69–6.68 (m, 2 H, ArH), 6.36 (d, J = 7.8 Hz, 1 H, ArH), 6.20 (d, J = 8.4 Hz, 1 H, ArH), 4.85 (d, J = 16.2 Hz, 1 H, CH), 4.38 (br. s, 2 H, CH_2), 4.26 (d, J = 16.2 Hz, 1 H, CH), 4.07–4.04 (m, 1 H, CH), 3.94 (br. s, 1 H, CH), 2.40–2.39 (m, 2 H, CH_2), 2.33 (s, 3 H, CH_3), 2.09–2.05 (m, 1 H, CH), 1.99–1.97 (m, 1 H, CH), 1.87–1.85 (m, 2 H, CH_2), 1.39 (t, J = 7.2 Hz, 3 H, CH_3), 1.08 (br. s, 3 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 190.9, 190.8, 174.1, 167.7, 163.5, 139.7, 136.2, 135.6, 135.0, 133.4, 131.9, 131.4, 130.8, 130.0, 129.0, 128.5, 127.5, 127.1, 125.3, 109.2, 70.3, 61.4, 61.1, 44.5, 36.3, 29.8, 21.1, 20.4, 18.4, 13.9, 13.7 ppm. IR (KBr): $\tilde{\nu}$ = 2969, 1731, 1644, 1596, 1489, 1419, 1369, 1331, 1291, 1251, 1186, 1099, 1019, 801 cm^{-1}. MS: *m/z* (%) = 647.99 (100) [M + Na]$^+$. $C_{36}H_{33}ClN_2O_6$ (625.12): calcd. C 69.17, H 5.32, N 4.48; found C 68.82, H 5.77, N 4.23.

2l: Yellow solid, yield: 78%, m.p. >250 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ = 10.11 (s, 1 H,NH), 7.30 (s, 1 H, ArH), 7.22 (d, J = 8.4 Hz, 1 H, ArH), 7.13–7.12 (m, 1 H, ArH), 7.04 (d, J = 7.8 Hz, 1 H, ArH), 6.92 (d, J = 7.8 Hz, 1 H, ArH), 6.46 (d, J = 7.8 Hz, 1 H, ArH), 6.24–6.22 (m, 1 H, ArH), 4.21–4.12 (m, 2 H, CH_2), 3.82–3.79 (m, 2 H, CH_2), 2.35 (br. s, 1 H, CH), 2.29 (s, 3 H, CH_3), 2.24–2.18 (m, 5 H, CH_2, CH_3), 2.04–2.03 (m, 2 H, CH_2), 1.76–1.73 (m, 1 H, CH), 1.72–1.68 (m, 1 H, CH), 1.25 (t, J = 7.2 Hz, 3 H, CH_3), 0.92 (t, J = 7.2 Hz, 3 H, CH_3) ppm. ^{13}C NMR (150 MHz, $CDCl_3$): δ = 189.8, 175.2, 166.9, 163.0, 138.8, 134.8, 131.3, 131.0, 130.9, 129.7, 129.4, 129.1, 128.9, 125.0, 109.4, 70.1, 60.3, 60.2, 35.8, 29.0, 20.6, 20.5, 20.0, 13.7, 13.1 ppm. IR (KBr): $\tilde{\nu}$ = 2947, 1733, 1604, 1492, 1427, 1311, 1275, 1199, 1115, 1022, 971, 823 cm^{-1}. MS: *m/z* (%) = 537.88 (100) [M + Na]$^+$. $C_{30}H_{30}N_2O_6$ (514.58): calcd. C 70.02, H 5.88, N 5.44; found C 69.67, H 6.21, N 5.19.

CCDC-842844 (for **1e**), -842847 (for **1j**), -850310 (for **2e**), and -850311 (for **2k**) contain the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.

Supporting Information (see footnote on the first page of this article): Experimental details and spectroscopic data for all new compounds.

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