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SYNTHESIS OF 6,7-DIHYDRO-9,10-DIMETHOXYDIBENZO[*a*,*f*]QUINOLIZINIUM SALT AND ITS D-RING SUBSTITUTED DERIVATIVES

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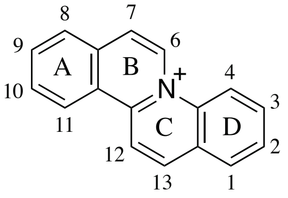
Abstract – Synthesis of 6,7-dihydro-9,10-dimethoxydibenzo[*a*,*f*]quinolizinium salt (**2a**) and its derivatives (**2b**–**g**) substituted at the D ring has been achieved from 3-(2-bromoaryl)propanoic acid (**3**) in four steps for future examination of the inhibitory activity of these compounds against topoisomerase. The dihydrocarbostyrils (**5a**–**g**), key intermediates in this synthetic scheme, were prepared from the amides (**4a**–**g**) *via* intramolecular aryl amidation reactions.

The dibenzo[*a*,*f*]quinolizinium skeleton (**1**) has long been known as one of the seven theoretically possible dibenzoquinolizinium compounds.[1] Although the Bischler–Napieralski cyclization of an *N*-arylethylated carbostyril or dihydrocarbostyril moiety has been mainly employed to form the B ring of this skeleton,[1] several alternative routes have also been reported.[2] The synthesis of 6,7-dihydro-9,10-dimethoxydibenzo[*a*,*f*]quinolizinium iodide (**2a**), a prototype of this ring system, was performed by Fujii and co-workers *via* mercuric acetate–edetic acid oxidation of a tetrahydroquinoline and subsequent cyclization of the resulting dihydrocarbostyril.[3] On preliminary examination of the inhibitory activity against topoisomerase I, the quinolizinium iodide (**2a**) showed a moderate effect (IC_{50} = 13.9 μM).[4] Therefore, the synthesis of 6,7-dihydro-9,10-dimethoxydibenzo[*a*,*f*]quinolizinium salts (**2a**–**g**) bearing different substituents on the D ring was undertaken for future structure–activity relationship (SAR) studies.



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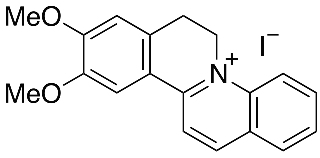
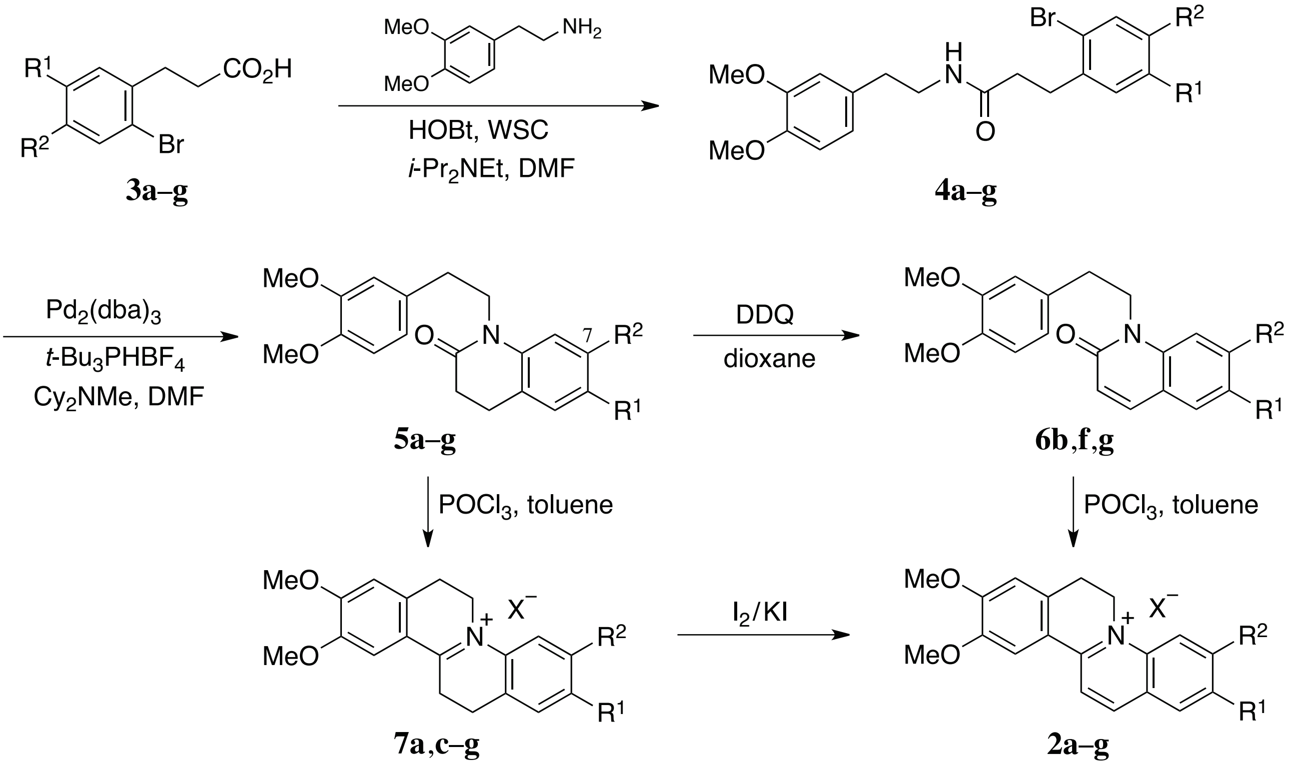


2a

We envisioned the dihydrocarbostyril (**5**) as a key intermediate in the synthesis of various dibenzoquinolizinium salts (**2**), as the lactam structure such as **5** would be available through the intramolecular aryl amidation reaction[5] of the amide (**4**), and also because conversion of **5** into the iminium salt (**7**) is feasible *via* the Bischler–Napieralski reaction described above. Thus, the amides (**4a–g**) required for the preparation of **5** were obtained from a variety of 3-(2-bromoaryl)propanoic acids (**3a–g**) in good yields through condensation with homoveratrylamine.



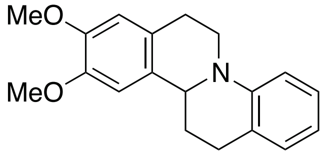
a: $R^1 = R^2 = H$
b: $R^1 = H$, $R^2 = MeO$
c: $R^1 = MeO$, $R^2 = H$
d: $R^1 = H$, $R^2 = F$
e: $R^1 = Cl$, $R^2 = H$
f: $R^1 = R^2 = MeO$
g: R^1–$R^2 = OCH_2O$

The intramolecular aryl amidation reaction[5] of the amide (**4**) was first investigated using the method of Buchwald.[5a] Thus, treatment of **4a** with $Pd_2(dba)_3$, P(*o*-tolyl)$_3$, and K_2CO_3 in toluene at 100 °C provided the dihydrocarbostyril (**5a**)[3] in 50% yield. In contrast, the intramolecular *N*-arylation of **4a** using copper (CuI, *N*,*N*′-dimethylethylenediamine, and K_2CO_3 in toluene at 110 °C) instead of palladium as a transition metal catalyst, an application of intermolecular version,[6] proved less effective. Best results were obtained when the amide (**4a**) was heated with $Pd_2(dba)_3$, t-Bu_3PHBF_4, and *N*,*N*-dicyclohexylmethylamine in DMF at 100 °C;[7] under these conditions, the desired product (**5a**) was produced in 77% yield. In a similar fashion, other dihydrocarbostyrils (**5b–g**) were prepared in moderate yields from the amides (**4b–g**), respectively.

For the conversion of **5** into the dibenzoquinolizinium salts (**2a–g**), we initially examined the oxidation of

5 followed by the Bischler–Napieralski cyclization of the resulting carbostyril (**6**), because a few precedents for the cyclization of carbostyrils had been found in the literature.[1c,f,h] Oxidation of **5b** with 2,3-dichloro-5,6-dicyano-1,4-benzoquinone (DDQ), a versatile reagent for the dehydrogenation of carbonyl compounds to give α,β-unsaturated congeners,[8] proceeded smoothly at 80 °C to provide **6b** in 79% yield. However, similar treatment of **5c** failed to give the desired carbostyril (**6c**). The mechanism of dehydrogenation by DDQ involves an initial rate-determining transfer of a hydride ion to the quinone oxygen, followed by a rapid proton transfer to the hydroquinone ion.[8] It is likely that the incipient benzylic carbonium ion derived from **5b** is stabilized effectively by the 7-methoxy group, whereas such stabilization is unlikely in the carbonium ion arising from **5c**. Indeed, oxidation of **5f** and **5g** bearing electron-donating substituents at the 7-position proceeded as expected, providing **6f** and **6g** in 81% and 74% yields, respectively. In addition, conversion of **6b** into the dibenzoquinolizinium chloride (**2b**: X = Cl) in 46% yield was achieved in refluxing $POCl_3$ by means of the Bischler–Napieralski cyclization.

As the oxidation of **5** was found to depend on the substitution pattern of its dihydrocarbostyril ring, we next investigated the cyclization of **5** and subsequent oxidation of the resulting iminium salt (**7**). The Fujii group previously reported that the cyclization of **5a** with $POCl_3$ followed by treatment of crude **7a** with KI afforded **2a** (X = I), instead of **7a** (X = I), in 38% yield accompanying the tetracyclic base (**8**) due to the disproportionation of **7a** (X = Cl or I).[3] Therefore, to obtain the desired ring system (**2**) alone, the iminium salt (**7a**), derived from **5a** *via* cyclization with $POCl_3$ in refluxing toluene, was treated with 0.5 M aqueous I_2/KI at 0 °C for 30 min to give the 6,7-dihydrodibenzo[*a,f*]quinolizinium triiodide (**2a**: X = I_3) in 87% yield from **5a**. On similar treatment of the dihydrocarbostyrils (**5c–g**), the corresponding dibenzoquinolizinium triiodides (**2c–g**: X = I_3) were prepared in good yields.



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In conclusion, 6,7-dihydro-9,10-dimethoxydibenzo[*a,f*]quinolizinium salt (**2a**) and its derivatives (**2b–g**) bearing a range of different substituents on the D ring were successively prepared from the dihydrocarbostyrils (**5a–g**) through either a cyclization–dehydrogenation route (**2a**,**c–g**) or a dehydrogenation–cyclization route (**2b**). The inhibitory activities of **2a–g** against topoisomerase will be reported elsewhere.

EXPERIMENTAL

General Notes. All melting points were taken on a Yamato MP-21 capillary melting point apparatus. Flash chromatography was carried out by using Merck silica gel 60 (No. 9385). The ratios of solvents in mixtures are shown in v/v. Spectra reported herein were recorded on a JEOL JMS-700 mass spectrometer, a Perkin-Elmer Spectrum 100 FT-IR spectrometer, or a JEOL JNM-ECA500 (^{1}H 500 MHz, ^{13}C 125 MHz) NMR spectrometer. Chemical shifts are reported in δ values relative to internal Me_4Si.

2-Bromo-*N*-[2-(3,4-dimethoxyphenyl)ethyl]benzenepropanamide (4a). A mixture of 2-bromobenzenepropanoic acid (**3a**)[9] (4.22 g, 18.4 mmol), homoveratrylamine (3.6 mL, 21 mmol), 1-ethyl-3-(3-dimethylaminopropyl)carbodiimide hydrochloride (4.2 g, 22 mmol), 1-hydroxybenzotriazole (3.0 g, 22 mmol), and *N*-ethyldiisopropylamine (6.3 mL, 37 mmol) in DMF (75 mL) was stirred at rt for 19 h. The reaction mixture was poured into H_2O (200 mL) and extracted with AcOEt. The AcOEt extracts were washed successively with 10% aqueous citric acid, H_2O, and brine, dried over anhydrous Na_2SO_4, and concentrated *in vacuo*. Purification of the residual solid by flash chromatography [AcOEt–hexane (1 : 1.2)] gave **4a** (6.59 g, 91%) as a slightly yellow solid, mp 94–95 °C; MS (FAB) *m*/*z*: 392, 394 (M^++1); IR (ATR) ν, cm^{-1}: 3293 (NH), 1637 (CO) ^{1}H-NMR ($CDCl_3$) δ: 2.44 (2H, t, *J* = 7.7 Hz, $COCH_2C\underline{H}_2$), 2.70 (2H, t, *J* = 6.9 Hz, $NCH_2C\underline{H}_2$), 3.06 (2H, t, *J* = 7.7 Hz, $COCH_2$), 3.47 (2H, dt, *J* = 6.9, 5.9 Hz, NCH_2), 3.860 and 3.863 (3H each, s, two MeO's), 5.39 (1H, br, NH), 6.64 [1H, dd, *J* = 8.1, 2.0 Hz, C(6)-H], 6.67 [1H, d, *J* = 2.0 Hz, C(2)-H], 6.78 [1H, d, *J* = 8.1 Hz, C(5)-H], 7.07 [1H, ddd, *J* = 8.0, 7.0, 2.0 Hz, C(4′)-H], 7.22 [1H, ddd, *J* = 7.8, 7.0, 1.2 Hz, C(5′)-H], 7.26 [1H, dd, *J* = 7.8, 2.0 Hz, C(6′)-H], 7.52 [1H, dd, *J* = 8.0, 1.2 Hz, C(3′)-H];[10] ^{13}C-NMR ($CDCl_3$) δ: 32.2 (t), 35.3 (t), 36.5 (t), 40.8 (t), 55.89 (q), 55.95 (q), 111.3 (d), 111.8 (d), 120.7 (d), 124.3 (s), 127.7 (d), 128.1 (d), 130.8 (d), 131.4 (s), 132.8 (d), 140.1 (s), 147.7 (s), 149.0 (s), 171.8 (s).

2-Bromo-*N*-[2-(3,4-dimethoxyphenyl)ethyl]-4-methoxybenzenepropanamide (4b). Prepared from 2-bromo-4-methoxybenzenepropanoic acid (**3b**)[11] (1.17 g, 4.5 mmol) according to the procedure described for the preparation of **4a**. Purification by flash chromatography [AcOEt–hexane (2 : 1)] provided **4b** (1.81 g, 95%) as a pale yellow solid, mp 99–100 °C; MS (EI) *m*/*z*: 421, 423 (M^+); IR (ATR) ν, cm^{-1}: 3305 (NH), 1638 (CO); ^{1}H-NMR ($CDCl_3$) δ: 2.40 (2H, t, *J* = 7.7 Hz, $COCH_2C\underline{H}_2$), 2.70 (2H, t, *J* = 6.9 Hz, $NCH_2C\underline{H}_2$), 2.99 (2H, t, *J* = 7.7 Hz, $COCH_2$), 3.46 (2H, dt, *J* = 6.9, 5.9 Hz, NCH_2), 3.77, 3.860, and 3.862 (3H each, s, three MeO's), 5.39 (1H, br, NH), 6.64 [1H, dd, *J* = 8.1, 1.8 Hz, C(6)-H], 6.68 [1H, d, *J* = 1.8 Hz, C(2)-H], 6.78 [1H, dd, *J* = 8.5, 2.6 Hz, C(5′)-H], 6.79 [1H, d, *J* = 8.1 Hz, C(5)-H], 7.08 [1H, d, *J* = 2.6 Hz, C(3′)-H], 7.15 [1H, d, *J* = 8.5 Hz, C(6′)-H];[10] ^{13}C-NMR ($CDCl_3$) δ: 31.3 (t), 35.3 (t), 36.9 (t), 40.7 (t), 55.5 (q), 55.88 (q), 55.93 (q), 111.3 (d), 111.8 (d), 113.6 (d), 118.0 (d), 120.7 (d), 124.3 (s), 131.1 (d), 131.3 (s), 131.9 (s), 147.7 (s), 149.1 (s), 158.7 (s), 171.8 (s).

2-Bromo-*N*-[2-(3,4-dimethoxyphenyl)ethyl]-5-methoxybenzenepropanamide (4c). Synthesized from 2-bromo-5-methoxybenzenepropanoic acid (**3c**)[12] (3.32 g, 12.8 mmol) according to the procedure described for the preparation of **4a**. Purification by flash chromatography [AcOEt–hexane (2 : 1)] gave **4c** (5.09 g, 94%) as a pale yellow solid, mp 99–100 °C; MS (EI) *m*/*z*: 421, 423 (M^+); IR (ATR) ν, cm^{-1}: 3296 (NH), 1635 (CO); ^{1}H-NMR ($CDCl_3$) δ: 2.43 (2H, t, *J* = 7.8 Hz, $COCH_2C\underline{H}_2$), 2.71 (2H, t, *J* = 6.9 Hz, $NCH_2C\underline{H}_2$), 3.02 (2H, t, *J* = 7.8 Hz, $COCH_2$), 3.47 (2H, dt, *J* = 6.9, 5.9 Hz, NCH_2), 3.76 (3H, s) and 3.86 (6H, s, three MeO's), 5.39 (1H, br, NH), 6.64 [1H, dd, *J* = 8.7, 3.1 Hz, C(4′)-H], 6.65 [1H, dd, *J* = 8.1, 1.9

Hz, C(6)-H], 6.68 [1H, d, J = 1.9 Hz, C(2)-H], 6.79 [1H, d, J = 8.1 Hz, C(5)-H], 6.82 [1H, d, J = 3.1 Hz, C(6′)-H], 7.39 [1H, d, J = 8.7 Hz, C(3′)-H];[10] ^{13}C-NMR (CDCl$_3$) δ: 32.5 (t), 35.3 (t), 36.6 (t), 40.7 (t), 55.5 (q), 55.87 (q), 55.92 (q), 111.3 (d), 111.8 (d), 113.9 (d), 114.5 (s), 116.1 (d), 120.6 (d), 131.3 (s), 133.3 (d), 141.0 (s), 147.7 (s), 149.1 (s), 159.0 (s), 171.6 (s).

2-Bromo-*N*-[2-(3,4-dimethoxyphenyl)ethyl]-4-fluorobenzenepropanamide (4d). Prepared from 2-bromo-4-fluorobenzenepropanoic acid (**3d**)[13] (4.00 g, 16.2 mmol) according to the method described for the preparation of **4a**. Purification by flash chromatography [AcOEt–hexane (1 : 2)] furnished **4d** (5.08 g, 76%) as a pale yellow solid, mp 110–111.5 °C; MS (EI) *m*/*z*: 409, 411 (M$^+$); IR (ATR) ν, cm^{-1}: 3307 (NH), 1639 (CO); ^{1}H-NMR (CDCl$_3$) δ: 2.41 (2H, t, J = 7.6 Hz, $\mathrm{COCH_2C\underline{H}_2}$), 2.71 (2H, t, J = 6.9 Hz, $\mathrm{NCH_2C\underline{H}_2}$), 3.03 (2H, t, J = 7.6 Hz, COCH$_2$), 3.47 (2H, dt, J = 6.9, 6.0 Hz, NCH$_2$), 3.86 and 3.87 (3H each, s, two MeO’s), 5.40 (1H, br, NH), 6.64 [1H, dd, J = 8.1, 1.9 Hz, C(6)-H], 6.67 [1H, d, J = 1.9 Hz, C(2)-H], 6.79 [1H, d, J = 8.1 Hz, C(5)-H], 6.95 [1H, ddd, J = 8.5, 8.2, 2.6 Hz, C(5′)-H], 7.23 [1H, dd, J = 8.5, 6.1 Hz, C(6′)-H], 7.27 [1H, dd, J = 8.2, 2.6 Hz, C(3′)-H];[10] ^{13}C-NMR (CDCl$_3$) δ: 31.3 (t), 35.2 (t), 36.5 (t), 40.7 (t), 55.86 (q), 55.90 (q), 111.2 (d), 111.7 (d), 114.7 (d), 119.9 (d), 120.6 (d), 123.9 (s), 131.2 (d), 131.5 (s), 135.9 (s), 147.7 (s), 149.0 (s), 161.0 (s), 171.5 (s).

2-Bromo-*N*-[2-(3,4-dimethoxyphenyl)ethyl]-5-chlorobenzenepropanamide (4e). Synthesized from 2-bromo-5-chlorobenzenepropanoic acid (**3e**)[14] (5.50 g, 20.9 mmol) according to the procedure described for the preparation of **4a**. Purification by flash chromatography [AcOEt–hexane (1 : 1)] gave **4e** (8.70 g, 98%) as a colorless solid, mp 107–108 °C; MS (EI) *m*/*z*: 425 (M$^+$); IR (ATR) ν, cm^{-1}: 3301 (NH), 1632 (CO); ^{1}H-NMR (CDCl$_3$) δ: 2.42 (2H, t, J = 7.7 Hz, $\mathrm{COCH_2C\underline{H}_2}$), 2.72 (2H, t, J = 7.0 Hz, $\mathrm{NCH_2C\underline{H}_2}$), 3.03 (2H, t, J = 7.7 Hz, COCH$_2$), 3.48 (2H, dt, J = 7.0, 5.9 Hz, NCH$_2$), 3.86 (6H, s, two MeO’s), 5.40 (1H, br, NH), 6.65 [1H, dd, J = 8.0, 2.0 Hz, C(6)-H], 6.68 [1H, d, J = 2.0 Hz, C(2)-H], 6.79 [1H, d, J = 8.0 Hz, C(5)-H], 7.06 [1H, dd, J = 8.5, 2.6 Hz, C(4′)-H], 7.25 [1H, d, J = 2.6 Hz, C(6′)-H], 7.44 [1H, d, J = 8.5 Hz, C(3′)-H];[10] ^{13}C-NMR (CDCl$_3$) δ: 32.0 (t), 35.2 (t), 36.1 (t), 40.7 (t), 55.87 (q), 55.92 (q), 111.3 (d), 111.7 (d), 120.6 (d), 122.1 (s), 128.1 (d), 130.6 (d), 131.2 (s), 133.4 (s), 133.8 (d), 141.8 (s), 147.7 (s), 149.1 (s), 171.2 (s).

2-Bromo-*N*-[2-(3,4-dimethoxyphenyl)ethyl]-4,5-dimethoxybenzenepropanamide (4f). Obtained from 2-bromo-4,5-dimethoxybenzenepropanoic acid (**3f**)[15] (6.45 g, 22.3 mmol) according to the method described for the preparation of **4a**. Purification by flash chromatography [AcOEt–hexane (2 : 1)] afforded **4b** (9.79 g, 97%) as a colorless solid, mp 124–125 °C (lit.[15a,b] mp 123–125 °C); MS (FAB) *m*/*z*: 452, 454 (M$^+$+1); IR (ATR) ν, cm^{-1}: 3307 (NH), 1641 (CO); ^{1}H-NMR (CDCl$_3$) δ: 2.41 (2H, t, J = 7.7 Hz, $\mathrm{COCH_2C\underline{H}_2}$), 2.71 (2H, t, J = 6.9 Hz, $\mathrm{NCH_2C\underline{H}_2}$), 2.99 (2H, t, J = 7.7 Hz, COCH$_2$), 3.47 (2H, dt, J = 6.9, 5.9 Hz, NCH$_2$), 3.84 (3H, s), 3.85 (3H, s), and 3.86 (6H, s, four MeO’s), 5.39 (1H, br, NH), 6.65 [1H, dd, *J* = 8.0, 1.8 Hz, C(6)-H], 6.67 [1H, d, *J* = 1.8 Hz, C(2)-H], 6.787 [1H, s, C(6′)-H], 6.788 [1H, d, *J* = 8.0 Hz, C(5)-H], 6.98 [1H, s, C(3′)-H];[10] ^{13}C-NMR ($CDCl_3$) δ: 31.9 (t), 35.3 (t), 36.9 (t), 40.8 (t), 55.8 (q), 55.9 (q), 56.0 (q), 56.1 (q), 111.3 (d), 111.8 (d), 113.3 (d), 113.8 (s), 115.5 (d), 120.6 (d), 131.4 (s), 132.1 (s), 147.6 (s), 148.1 (s), 148.4 (s), 149.0 (s), 171.9 (s).

6-Bromo-*N*-[2-(3,4-dimethoxyphenyl)ethyl]-1,3-benzodioxole-5-propanamide (4g). Synthesized from 6-bromo-1,3-benzodioxole-5-propanoic acid (**3g**)[16] (7.72 g, 28.3 mmol) according to the method described for the preparation of **4a**. Purification by flash chromatography [AcOEt–hexane (2 : 1)] furnished **4g** (11.2 g, 91%) as a colorless solid, mp 140–141 °C; MS (EI) *m*/*z*: 435, 437 (M^+); IR (ATR) ν, cm^{-1}: 3268 (NH), 1634 (CO); ^{1}H-NMR ($CDCl_3$) δ: 2.38 (2H, t, *J* = 7.7 Hz, $COCH_2C\underline{H}_2$), 2.72 (2H, t, *J* = 6.9 Hz, $NCH_2C\underline{H}_2$), 2.96 (2H, t, *J* = 7.7 Hz, $COCH_2$), 3.48 (2H, dt, *J* = 6.9, 5.9 Hz, NCH_2), 3.87 (6H, s, two MeO’s), 5.39 (1H, br, NH), 5.94 (2H, s, OCH_2O), 6.66 [1H, dd, *J* = 8.1, 1.9 Hz, C(6)-H], 6.69 [1H, d, *J* = 1.9 Hz, C(2)-H], 6.75 [1H, s, C(6′)-H], 6.79 [1H, d, *J* = 8.1 Hz, C(5)-H], 6.97 [1H, s, C(3′)-H];[10] ^{13}C-NMR ($CDCl_3$) δ: 32.1 (t), 35.3 (t), 36.7 (t), 40.8 (t), 55.89 (q), 55.94 (q), 101.7 (t), 110.3 (d), 111.3 (d), 111.8 (d), 112.7 (d), 114.2 (s), 120.7 (d), 131.4 (s), 133.1 (s), 147.0 (s), 147.4 (s), 147.7 (s), 149.1 (s), 171.7 (s).

1-[2-(3,4-Dimethoxyphenyl)ethyl]-3,4-dihydro-2(1*H*)-quinolinone (5a). A stirred solution of **4a** (6.01 g, 15.3 mmol), tris(dibenzylideneacetone)dipalladium (1.33 g, 1.5 mmol), tri-*t*-butylphosphonium tetrafluoroborate (1.05 g, 3.6 mmol), *N*,*N*-dicyclohexylmethylamine (4.2 mL, 20 mmol) in DMF (46 mL) was heated in an atmosphere of Ar at 100 °C for 137 h. The reaction mixture was poured into H_2O (100 mL) and extracted with AcOEt. The AcOEt extracts were washed successively with 1 M aqueous HCl, H_2O and brine, dried over anhydrous Na_2SO_4, and concentrated *in vacuo* to leave a brown oil. Purification by flash chromatography [AcOEt–hexane (2 : 3)] gave **5a** (3.66 g, 77%) as a slightly yellow solid, mp 62–63 °C (lit.[3] mp 64.5–65.5 °C). ^{1}H-NMR and IR spectral data for this sample were in agreement with those reported in the literature.[3]

1-[2-(3,4-Dimethoxyphenyl)ethyl]-3,4-dihydro-7-methoxy-2(1*H*)-quinolinone (5b). A mixture of **4b** (3.61 g, 8.5 mmol), tris(dibenzylideneacetone)dipalladium (381 mg, 0.42 mmol), tri-*t*-butylphosphonium tetrafluoroborate (743 mg, 2.6 mmol), *N*,*N*-dicyclohexylmethylamine (2.2 mL, 10 mmol), and DMF (50 mL) was stirred in an atmosphere of Ar at 110 °C for 92 h. The reaction mixture was treated with 1 M aqueous HCl (100 mL) and extracted with AcOEt. The AcOEt extracts were washed successively with H_2O and brine, dried over anhydrous Na_2SO_4, and concentrated *in vacuo*. Purification of the residual oil by flash chromatography [AcOEt–hexane (1 : 1)] provided **5b** (2.29 g, 78%) as a pale yellow solid, mp 90–91 °C; MS (EI) *m*/*z*: 341 (M^+); IR (ATR) ν: 1662 cm^{-1} (CO); ^{1}H-NMR ($CDCl_3$) δ: 2.60 and 2.76 [2H each, t, *J* = 7.3 Hz, C(3)-H’s, C(4)-H’s], 2.89 (2H, t, *J* = 7.9 Hz, $NCH_2C\underline{H}_2$), 3.81 (3H, s) and 3.86 (6H, s, three MeO’s), 4.12 (2H, t, *J* = 7.9 Hz, NCH_2), 6.55 [1H, dd, *J* = 8.2, 2.4 Hz, C(6)-H], 6.61 [1H, d, *J* = 2.4

Hz, C(8)-H], 6.75–6.82 [3H, m, $(\mathrm{MeO})_2\mathrm{C}_6\underline{\mathrm{H}}_3$], 7.07 [1H, d, *J* = 8.2 Hz, C(5)-H]; ^{13}C-NMR (CDCl_3) δ: 24.7 (t), 32.3 (t), 33.0 (t), 43.8 (t), 55.4 (q), 55.87 (q), 55.92 (q), 102.7 (d), 106.5 (d), 111.3 (d), 112.1 (d), 119.0 (s), 120.7 (d), 128.5 (d), 131.2 (s), 140.5 (s), 147.7 (s), 148.9 (s), 159.2 (s), 170.3 (s).

1-[2-(3,4-Dimethoxyphenyl)ethyl]-3,4-dihydro-6-methoxy-2(1*H*)-quinolinone (5c). Prepared from **4c** (7.53 g, 17.8 mmol) according to the procedure described for the preparation of **5b**. Purification by flash chromatography [AcOEt–hexane (1 : 1)] afforded **5c** (3.09 g, 51%) as a pale yellow solid, mp 72–73 °C; MS (EI) *m*/*z*: 341 (M^+); IR (ATR) ν: 1660 cm^{-1} (CO); ^{1}H-NMR (CDCl_3) δ: 2.60 and 2.78 [2H each, t, *J* = 7.3 Hz, C(3)-H's, C(4)-H's], 2.88 (2H, t, *J* = 7.8 Hz, $\mathrm{NCH}_2\mathrm{C}\underline{\mathrm{H}}_2$), 3.80 (3H, s) and 3.86 (6H, s, three MeO's), 4.13 (2H, t, *J* = 7.8 Hz, NCH_2), 6.73–6.81 [5H, m, $(\mathrm{MeO})_2\mathrm{C}_6\underline{\mathrm{H}}_3$, C(5)-H, C(7)-H], 6.95 [1H, d, *J* = 8.8 Hz, C(8)-H]; ^{13}C-NMR (CDCl_3) δ: 25.8 (t), 31.9 (t), 33.0 (t), 43.6 (t), 55.6 (q), 55.89 (q), 55.92 (q), 111.2 (d), 112.0 (d), 112.1 (d), 114.0 (d), 115.2 (d), 120.7 (d), 128.4 (s), 131.2 (s), 133.0 (s), 147.6 (s), 148.9 (s), 155.2 (s), 169.8 (s).

1-[2-(3,4-Dimethoxyphenyl)ethyl]-3,4-dihydro-7-fluoro-2(1*H*)-quinolinone (5d). Synthesized from **4d** (4.70 g, 11.5 mmol) according to the method described for the preparation of **5b**. Purification by flash chromatography [AcOEt–hexane (1 : 1)] afforded **5d** (1.86 g, 49%) as a pale yellow solid, mp 107–108 °C, MS (EI) *m*/*z*: 329 (M^+); IR (ATR) ν: 1680 cm^{-1} (CO); ^{1}H-NMR (CDCl_3) δ: 2.61 and 2.79 [2H each, t, *J* = 7.3 Hz, C(3)-H's, C(4)-H's], 2.88 (2H, t, *J* = 7.9 Hz, $\mathrm{NCH}_2\mathrm{C}\underline{\mathrm{H}}_2$), 3.863 and 3.865 (3H each, s, two MeO's), 4.11 (2H, t, *J* = 7.9 Hz, NCH_2), 6.71 [1H, ddd, *J* = 8.2, 8.2, 2.4 Hz, C(6)-H], 6.74–6.82 [4H, m, $(\mathrm{MeO})_2\mathrm{C}_6\underline{\mathrm{H}}_3$, C(8)-H], 7.10 [1H, dd, *J* = 8.2, 6.4 Hz, C(5)-H]; ^{13}C-NMR (CDCl_3) δ: 24.9 (t), 31.9 (t), 32.9 (t), 43.7 (t), 55.87 (q), 55.91 (q), 102.9 (d), 109.0 (d), 111.2 (d), 112.0 (d), 120.7 (d), 122.2 (s), 128.9 (d), 130.8 (s), 140.8 (s), 147.7 (s), 148.9 (s), 162.2 (s), 170.0 (s).

1-[2-(3,4-Dimethoxyphenyl)ethyl]-3,4-dihydro-6-chloro-2(1*H*)-quinolinone (5e). Obtained from **4e** (8.50 g, 19.9 mmol) according to the procedure described for the preparation of **5b**. Purification by flash chromatography [AcOEt–hexane (1 : 1)] gave **5e** (3.98 g, 58%) as a pale yellow solid, mp 96–97 °C, MS (EI) *m*/*z*: 345, 347 (M^+); IR (ATR) ν: 1669 cm^{-1} (CO); ^{1}H-NMR (CDCl_3) δ: 2.60 and 2.78 [2H each, t, *J* = 7.3 Hz, C(3)-H's, C(4)-H's], 2.87 (2H, t, *J* = 7.8 Hz, $\mathrm{NCH}_2\mathrm{C}\underline{\mathrm{H}}_2$), 3.855 and 3.859 (3H each, s, two MeO's), 4.13 (2H, t, *J* = 7.8 Hz, NCH_2), 6.72–6.80 [3H, m, $(\mathrm{MeO})_2\mathrm{C}_6\underline{\mathrm{H}}_3$], 6.94 [1H, d, *J* = 8.7 Hz, C(8)-H], 7.14 [1H, d, *J* = 2.4 Hz, C(5)-H], 7.22 [1H, dd, *J* = 8.7, 2.4 Hz, C(7)-H]; ^{13}C-NMR (CDCl_3) δ: 25.3 (t), 31.6 (t), 32.9 (t), 43.5 (t), 55.88 (q), 55.90 (q), 111.2 (d), 112.0 (d), 116.1 (d), 120.7 (d), 127.3 (d), 127.9 (s), 128.0 (d), 128.6 (s), 130.8 (s), 138.1 (s), 147.7 (s), 148.9 (s), 169.7 (s).

1-[2-(3,4-Dimethoxyphenyl)ethyl]-3,4-dihydro-6,7-dimethoxy-2(1*H*)-quinolinone (5f). Prepared from **4f** (4.52 g, 10.0 mmol) according to the procedure described for the preparation of **5b**. Purification by flash chromatography [AcOEt–hexane (1 : 1)] afforded **5f** (1.37 g, 37%) as a pale yellow solid, mp 132–133 °C, MS (EI) *m*/*z*: 371 (M^+); IR (ATR) ν: 1642 cm^{-1} (CO); ^{1}H-NMR (CDCl_3) δ: 2.60 and 2.75

[2H each, t, J = 7.3 Hz, C(3)-H's, C(4)-H's], 2.91 (2H, t, J = 7.6 Hz, $NCH_2C\underline{H}_2$), 3.84 (6H, s), 3.85 (3H, s), and 3.87 (3H, s, four MeO's), 4.13 (2H, t, J = 7.6 Hz, NCH_2), 6.53 and 6.69 [1H each, s, C(5)-H, C(8)-H], 6.73–6.80 [3H, m, $(MeO)_2C_6\underline{H}_3$]; ^{13}C-NMR ($CDCl_3$) δ: 25.2 (t), 32.3 (t), 33.3 (t), 44.0 (t), 55.8 (q), 55.9 (q), 56.29 (q), 56.31 (q), 100.9 (d), 111.3 (d), 111.7 (s), 112.1 (d), 118.7 (s), 120.7 (d), 131.3 (s), 132.9 (s), 144.5 (s), 147.7 (s), 148.0 (s), 149.0 (s), 169.9 (s).

7,8-Dihydro-5-[2-(3,4-dimethoxyphenyl)ethyl]-1,3-dioxolo[4,5-*g*]quinolin-6(5*H*)-one (5g). Prepared from **4g** (3.55 g, 8.1 mmol) according to the procedure described for the preparation of **5b**. Purification by flash chromatography [AcOEt–hexane (3 : 1)] provided **5g** (1.62 g, 56%) as a pale yellow solid, mp 108–109 °C; MS (EI) *m*/*z*: 355 (M^+); IR (ATR) ν: 1663 cm^{-1} (CO); ^{1}H-NMR ($CDCl_3$) δ: 2.56 and 2.69 [2H each, t, J = 7.2 Hz, C(3)-H's, C(4)-H's], 2.87 (2H, t, J = 7.9 Hz, $NCH_2C\underline{H}_2$), 3.86 and 3.87 (3H each, s, two MeO's), 4.09 (2H, t, J = 7.9 Hz, NCH_2), 5.96 (2H, s, OCH_2O), 6.63 and 6.65 [1H each, s, C(5)-H, C(8)-H], 6.74–6.81 [3H, m, $(MeO)_2C_6\underline{H}_3$]; ^{13}C-NMR ($CDCl_3$) δ: 25.5 (t), 32.2 (t), 33.1 (t), 43.9 (t), 55.89 (q), 55.92 (q), 97.8 (d), 101.3 (t), 108.2 (d), 111.3 (d), 112.1 (d), 119.8 (s), 120.8 (d), 131.0 (s), 133.7 (s), 142.8 (s), 146.9 (s), 147.7 (s), 148.9 (s), 170.0 (s).

1-[2-(3,4-Dimethoxyphenyl)ethyl]-7-methoxy-2(1*H*)-quinolinone (6b). A solution of **5b** (171 mg, 0.5 mmol) and DDQ (136 mg, 0.6 mmol) in dioxane (5 mL) was heated with stirring at 80 °C for 2 h. After the mixture had been concentrated *in vacuo*, the residue was partitioned between $CHCl_3$ and 5% aqueous K_2CO_3. The $CHCl_3$ extracts were washed successively with H_2O and brine, dried over anhydrous $MgSO_4$, and concentrated *in vacuo*. The residual oil was purified by flash chromatography [AcOEt–hexane (2 : 1)] to give **6b** (134 mg, 79%) as a pale yellow solid, mp 125–126 °C; MS (FAB) *m*/*z*: 340 (M^++1): IR (ATR) ν: 1646 cm^{-1} (CO); ^{1}H-NMR ($CDCl_3$) δ: 2.98 (2H, t, J = 8.1 Hz, $ArC\underline{H}_2$), 3.86, 3.87, and 3.89 (3H each, s, three MeO's), 4.44 (2H, t, J = 8.1 Hz, NCH_2), 6.57 [1H, d, J = 9.4 Hz, C(3)-H], 6.81–6.88 [5H, m, $(MeO)_2C_6\underline{H}_3$, C(6)-H, C(8)-H], 7.48 [1H, d, J = 8.4 Hz, C(5)-H], 7.62 [1H, d, J = 9.4 Hz, C(4)-H]; ^{13}C-NMR ($CDCl_3$) δ: 33.1 (t), 44.2 (t), 55.5 (q), 55.9 (q), 56.0 (q), 98.8 (d), 109.4 (d), 111.4 (d), 112.1 (d), 115.2 (s), 118.6 (d), 120.6 (d), 130.3 (d), 131.1 (s), 139.0 (d), 140.9 (s), 147.8 (s), 149.1 (s), 161.8 (s), 162.4 (s).

1-[2-(3,4-Dimethoxyphenyl)ethyl]-6,7-dimethoxy-2(1*H*)-quinolinone (6f). A mixture of **5f** (371 mg, 1.0 mmol), DDQ (318 mg, 1.4 mmol), and dioxane (10 mL) was stirred at 80 °C for 30 min. The reaction mixture was then worked up as described above for the preparation of **6b**. Purification by flash chromatography [AcOEt–hexane (10 : 1)] afforded **6f** (299 mg, 81%) as a pale brown solid, mp 147–148 °C; MS (FAB) *m*/*z*: 370 (M^++1): IR (ATR) ν: 1645 cm^{-1} (CO); ^{1}H-NMR ($CDCl_3$) δ: 3.02 (2H, t, J = 7.8 Hz, $ArC\underline{H}_2$), 3.82, 3.86, 3.928, and 3.934 (3H each, s, four MeO's), 4.48 (2H, t, J = 7.8 Hz, NCH_2), 6.62 [1H, d, J = 9.4 Hz, C(3)-H], 6.72 and 6.95 [1H each, s, C(5)-H, C(8)-H], 6.78–6.84 [3H, m, $(MeO)_2C_6\underline{H}_3$], 7.59 [1H, d, J = 9.4 Hz, C(4)-H]; ^{13}C-NMR ($CDCl_3$) δ: 33.4 (t), 44.5 (t), 55.8 (q), 55.9 (q), 56.0 (q), 56.2 (q), 97.3 (d), 109.4 (d), 111.4 (d), 112.1 (d), 114.2 (s), 119.1 (d), 120.5 (d), 131.2 (s), 134.9 (s), 138.5 (d), 145.0 (s), 147.9 (s), 149.1 (s), 152.0 (s), 162.0 (s).

5-[2-(3,4-Dimethoxyphenyl)ethyl]-1,3-dioxolo[4,5-*g*]quinolin-6(5*H*)-one (6g). Prepared from **5g** (1.13 g, 3.2 mmol) according to the method described for the preparation of **6f**. Purification by flash chromatography [AcOEt–hexane (5 : 1)] gave **6g** (834 mg, 74%) as a slightly yellow solid, mp 137–139 °C; MS (FAB) *m*/*z*: 354 (M^++1): IR (ATR) ν: 1650 cm^{-1} (CO); ^{1}H-NMR ($CDCl_3$) δ: 2.96 (2H, t, J = 8.2 Hz, Ar$C\underline{H_2}$), 3.877 and 3.879 (3H each, s, two MeO's), 4.42 (2H, t, J = 8.2 Hz, NCH_2), 6.07 (2H, s, OCH_2O), 6.60 [1H, d, J = 9.5 Hz, C(3)-H], 6.83–6.89 [3H, m, $(MeO)_2C_6\underline{H_3}$], 6.88 and 6.94 [1H each, s, C(5)-H, C(8)-H], 7.55 [1H, d, J = 9.5 Hz, C(4)-H]; ^{13}C-NMR ($CDCl_3$) δ: 33.2 (t), 44.7 (t), 55.92 (q), 55.94 (q), 95.0 (d), 101.9 (t), 106.5 (d), 111.4 (d), 112.1 (d), 115.4 (s), 119.0 (d), 120.7 (d), 130.9 (s), 136.3 (s), 138.7 (d), 143.3 (s), 147.9 (s), 149.1 (s), 151.0 (s), 161.9 (s).

6,7-Dihydro-9,10-dimethoxydibenzo[*a*,*f*]quinolizinium Triiodide (2a: X = I_3). A mixture of **5a** (311 mg, 1.0 mmol) and $POCl_3$ (1 mL, 11 mmol) in toluene (5 mL) was heated under reflux with stirring for 6 h. The solvent and excess $POCl_3$ were distilled off *in vacuo* to leave an orange solid, which was then triturated with Et_2O. The insoluble material was collected by filtration and dissolved in hot H_2O (15 mL). The solution was then cooled and treated with 0.5 M aqueous I_2/KI (4 mL) at 0 °C for 30 min. The precipitate that resulted was collected by filtration and dried to afford **2a** (549 mg, 87%) as a dark brown solid. Recrystallization from MeOH gave an orange solid, mp 190–191 °C; ^{1}H-NMR (DMSO-d_6) δ: 3.33 [2H, t, J = 7.1 Hz, C(7)-H's], 3.94 and 3.95 (3H each, s, two MeO's), 5.09 [2H, t, J = 7.1 Hz, C(6)-H's], 7.26 [1H, s, C(8)-H], 7.82 [1H, s, C(11)-H], 7.94 [1H, dd, J = 8.1, 7.1 Hz, C(2)-H], 8.20 [1H, ddd, J = 9.0, 7.1, 1.5 Hz, C(3)-H], 8.38 [1H, dd, J = 8.1, 1.5 Hz, C(1)-H], 8.64 [1H, d, J = 9.0 Hz, C(4)-H], 8.76 [1H, d, J = 9.0 Hz, C(12)-H], 9.12 [1H, d, J = 9.0 Hz, C(13)-H];[17] ^{13}C-NMR (DMSO-d_6) δ: 24.0 (t), 45.5 (t), 55.0 (q), 55.1 (q), 109.7 (d), 110.2 (d), 117.1 (d), 117.6 (s), 119.0 (d), 126.3 (s), 127.4 (d), 128.9 (d), 130.7 (s), 133.5 (d), 137.3 (s), 143.6 (d), 147.3 (s), 150.2 (s), 153.0 (s). *Anal*. Calcd for $C_{19}H_{18}I_3NO_2$: C, 33.91; H, 2.70; N, 2.08. Found: C, 33.70; H, 2.73; N, 2.04.

6,7-Dihydro-3,9,10-trimethoxydibenzo[*a*,*f*]quinolizinium Chloride (2b: X = Cl). A stirred mixture of **6b** (170 mg, 0.5 mmol) and $POCl_3$ (1 mL, 11 mmol) was heated under reflux for 28 h. After cooling, the reaction mixture was treated with cold H_2O, and the insoluble material was collected by filtration and dried to give **2b** (82.1 mg, 46%) as a yellow solid, mp 294–295 °C; ^{1}H-NMR (DMSO-d_6) δ: 3.31 [2H, t, J = 7.0 Hz, C(7)-H's], 3.937, 3.943, and 4.11 (3H each, s, three MeO's), 5.03 [2H, t, J = 7.0 Hz, C(6)-H's], 7.25 [1H, s, C(8)-H], 7.60 [1H, dd, J = 9.0, 1.9 Hz, C(2)-H], 7.79 [1H, s, C(11)-H], 7.86 [1H, d, J = 1.9 Hz, C(4)-H], 8.31 [1H, d, J = 9.0 Hz, C(1)-H], 8.58 [1H, d, J = 8.8 Hz, C(12)-H], 9.02 [1H, d, J = 8.8 Hz, C(13)-H]; ^{13}C-NMR (DMSO-d_6) δ: 25.3 (t), 46.6 (t), 56.1 (q), 56.2 (q), 56.7 (q), 99.1 (d), 110.9 (d), 111.2

(d), 117.3 (d), 119.1 (s), 120.3 (d), 123.1 (s), 131.5 (s), 131.9 (d), 141.0 (s), 144.3 (d), 148.6 (s), 150.8 (s), 153.9 (s), 164.5 (s).

6,7-Dihydro-2,9,10-trimethoxydibenzo[*a*,*f*]quinolizinium Triiodide (2c: X = I_3). Cyclization of **5c** (341 mg, 1.1 mmol) was carried out as described above for the preparation of **2a**, giving the iminium salt (360 mg) as an orange solid. To a solution of the solid in DMSO (1 mL) was added 0.5 M aqueous I_2/KI (2 mL), and the mixture was stirred at 0 °C for 30 min. After addition of H_2O (10 mL), the precipitate that resulted was collected by filtration and dried to afford **2c** (620 mg, 88%) as a dark brown solid, mp 274–275 °C; ^{1}H-NMR (DMSO-d_6) δ: 3.31 [2H, t, J = 7.0 Hz, C(7)-H's], 3.936, 3.943, and 4.01 (3H each, s, three MeO's), 5.06 [2H, t, J = 7.0 Hz, C(6)-H's], 7.22 [1H, s, C(8)-H], 7.79 [1H, s, C(11)-H], 7.81 [1H, dd, J = 9.5, 2.9 Hz, C(3)-H], 7.84 [1H, d, J = 2.9 Hz, C(1)-H], 8.56 [1H, d, J = 9.5 Hz, C(4)-H], 8.70 [1H, d, J = 9.1 Hz, C(12)-H], 8.98 [1H, d, J = 9.1 Hz, C(13)-H]; ^{13}C-NMR (DMSO-d_6) δ: 24.0 (t), 45.8 (t), 54.88 (q), 54.92 (q), 55.0 (q), 107.4 (d), 109.8 (d), 110.1 (d), 117.7 (s), 118.9 (d), 119.2 (d), 124.7 (d), 128.3 (s), 130.0 (s), 132.7 (s), 142.2 (d), 147.4 (s), 147.8 (s), 152.7 (s), 157.2 (s).

3-Fluoro-6,7-dihydro-9,10-dimethoxydibenzo[*a*,*f*]quinolizinium Triiodide (2d: X = I_3). Cyclization of **5d** (160 mg, 0.49 mmol) and subsequent oxidation were performed as described for the synthesis of **2c** to afford **2d** (270 mg, 80%) as a dark brown solid, mp 200–201 °C; ^{1}H-NMR (DMSO-d_6) δ: 3.33 [2H, t, J = 7.1 Hz, C(7)-H's], 3.95 (6H, s, two MeO's), 5.01 [2H, t, J = 7.1 Hz, C(6)-H's], 7.24 [1H, s, C(8)-H], 7.82 [1H, s, C(11)-H], 7.90 [1H, ddd, J = 9.0, 8.2, 2.0 Hz, C(2)-H], 8.49 [1H, dd, J = 9.0, 6.3 Hz, C(1)-H], 8.56 [1H, dd, J = 11.8, 2.0 Hz, C(4)-H], 8.73 [1H, d, J = 9.0 Hz, C(12)-H], 9.12 [1H, d, J = 9.0 Hz, C(13)-H]; ^{13}C-NMR (DMSO-d_6) δ: 23.9 (t), 46.0 (t), 55.0 (q), 55.1 (q), 103.6 (d), 109.8 (d), 110.5 (d), 117.2 (d), 117.5 (s), 118.4 (d), 123.7 (s), 131.0 (s), 132.1 (d), 139.0 (s), 143.5 (d), 147.5 (s), 151.0 (s), 153.4 (s), 164.0 (s). *Anal*. Calcd for $C_{19}H_{17}FI_3NO_2$: C, 33.02; H, 2.48; N, 2.03. Found: C, 33.20; H, 2.40; N, 2.08.

2-Chloro-6,7-dihydro-9,10-dimethoxydibenzo[*a*,*f*]quinolizinium Triiodide (2e: X = I_3). Prepared from **5e** (170 mg, 0.49 mmol) according to the procedure described for the preparation of **2c** to give **2e** (225 mg, 77%) as a dark brown solid, mp 287–288 °C; ^{1}H-NMR (DMSO-d_6) δ: 3.34 [2H, t, J = 7.1 Hz, C(7)-H's], 3.951 and 3.954 (3H each, s, two MeO's), 5.08 [2H, t, J = 7.1 Hz, C(6)-H's], 7.26 [1H, s, C(8)-H], 7.82 [1H, s, C(11)-H], 8.21 [1H, dd, J = 9.5, 2.5 Hz, C(3)-H], 8.53 [1H, d, J = 2.5 Hz, C(1)-H], 8.66 [1H, d, J = 9.5 Hz, C(4)-H], 8.81 [1H, d, J = 9.1 Hz, C(12)-H], 9.04 [1H, d, J = 9.1 Hz, C(13)-H]; ^{13}C-NMR (DMSO-d_6) δ: 23.9 (t), 46.0 (t), 55.0 (q), 55.1 (q), 109.9 (d), 110.5 (d), 117.4 (s), 119.6 (d), 120.3 (d), 127.3 (s), 127.4 (d), 131.0 (s), 131.7 (s), 133.2 (d), 136.1 (s), 142.6 (d), 147.5 (s), 150.6 (s), 153.5 (s). *Anal*. Calcd for $C_{19}H_{17}ClI_3NO_2$: C, 32.26; H, 2.42; N, 1.98. Found: C, 32.25; H, 2.61; N, 1.92.

6,7-Dihydro-2,3,9,10-tetramethoxydibenzo[*a*,*f*]quinolizinium Triiodide (2f: X = I_3). Synthesized from **5f** (130 mg, 0.35 mmol) according to the method described for the preparation of **2c** to furnish **2f** (248 mg, 97%) as a brown solid, mp 214–216 °C; ^{1}H-NMR (DMSO-d_6) δ: 3.30 [2H, t, J = 6.9 Hz, C(7)-H's], 3.927, 3.933, 4.01, and 4.14 (3H each, s, four MeO's), 5.04 [2H, t, J = 6.9 Hz, C(6)-H's], 7.22 [1H, s, C(8)-H], 7.74, 7.80, and 7.82 [1H each, s, C(1)-H, C(4)-H, C(11)-H], 8.53 [1H, d, J = 8.8 Hz, C(12)-H], 8.87 [1H, d, J = 8.8 Hz, C(13)-H]; ^{13}C-NMR (DMSO-d_6) δ: 24.2 (t), 45.8 (t), 54.8 (q), 55.0 (q), 55.1 (q), 55.9 (q), 97.7 (d), 106.7 (d), 109.6 (d), 109.7 (d), 116.4 (d), 118.0 (s), 122.9 (s), 129.3 (s), 134.8 (s), 141.1 (d), 146.7 (s), 147.4 (s), 148.9 (s), 152.2 (s), 154.8 (s). *Anal.* Calcd for $C_{21}H_{22}I_3NO_4$: C, 34.41; H, 3.02; N, 1.91. Found: C, 34.71; H, 3.08; N, 2.03.

13,14-Dihydro-2,3-dimethoxybenzo[*a*][1,3]benzodioxolo[5,6-*f*]quinolizinium Triiodide (2g: X = I_3). Prepared from **5g** (180 mg, 0.51 mmol) according to the procedure described for the preparation of **2c** to provide **2g** (350 mg, 96%) as a brown solid, mp 226–227 °C; ^{1}H-NMR (DMSO-d_6) δ: 3.27 [2H, t, J = 6.9 Hz, C(7)-H's], 3.92 and 3.93 (3H each, s, two MeO's), 4.94 [2H, t, J = 6.9 Hz, C(6)-H's], 6.44 (2H, s, OCH_2O), 7.19 [1H, s, C(8)-H], 7.72, 7.74, and 8.16 [1H each, s, C(1)-H, C(4)-H, C(11)-H], 8.52 [1H, d, J = 8.9 Hz, C(12)-H], 8.83 [1H, d, J = 8.9 Hz, C(13)-H]; ^{13}C-NMR (DMSO-d_6) δ: 24.1 (t), 46.4 (t), 54.8 (q), 55.0 (q), 95.6 (d), 102.9 (t), 103.5 (d), 109.67 (d), 109.72 (d), 116.7 (d), 117.8 (s), 124.5 (s), 129.4 (s), 136.7 (s), 141.7 (d), 147.2 (s), 147.4 (s), 147.5 (s), 152.3 (s), 153.9 (s). *Anal.* Calcd for $C_{20}H_{18}I_3NO_4$: C, 33.50; H, 2.53; N, 1.95. Found: C, 33.69; H, 2.55; N, 1.92.

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