A direct, two-step synthetic pathway to novel dibenzo[*a,c*]phenanthridines

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General methods

For general experimental details, see ref 1. X-ray single-crystal analysis was taken on a Enraf Nonius Cad4 diffractometer with Mo-K α radiation (λ 0.71073 Å).² The crystallographic data reduction was carried out by Xtal3.2³ and the structure was solved by SHELXS97 and refined by SHELXL97 programs.⁴

General procedure for the Ritter-type heterocyclization of triarylethanones 3. To a solution of triarylethanone **3** (1 mmol) in dry acetonitrile (40 mL) was added P₂O₅ (33 mmol) in portions at room temperature under argon. The reaction mixture was stirred at room temperature for 16 h, the solvent was removed *in vacuo*, and water (50 mL) and CH₂Cl₂ (20 mL) were added. The resulting solution was basified with NaOH (20 mL of 10% solution in water) and extracted with CH₂Cl₂ (3 x 30 mL). The combined organic extracts were dried over anhydrous sodium sulfate and evaporated *in vacuo* to give a residue which was purified by flash chromatography on silicagel using 0-30% EtOAc/CH₂Cl₂ as eluent.

6,7-Dimethoxy-3-(3,4-dimethoxyphenyl)-1-methyl-4-phenylisoquinoline (2a). 94%. White powder: mp 199-200°C (MeOH); *R*_f 0.54 (20% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 2.99 (3H, s), 3.55 (3H, s), 3.76 (3H, s), 3.82 (3H, s), 4.06 (3H, s), 6.74 (1H, d, *J* 8.3 Hz), 6.76 (1H, d, *J* 1.9 Hz), 6.90 (1H, s), 7.07 (1H, dd, *J* 8.3, 1.9 Hz), 7.23-7.26 (2H, m), 7.31-7.41 (4H, m); ¹³C NMR (63 MHz, CDCl₃) δ 22.7, 55.3, 55.6, 55.9, 103.6, 104.4, 110.3, 113.5, 121.8, 122.8, 127.0, 127.8, 128.4, 131.0, 132.6, 133.5, 138.3, 147.5, 147.7, 149.2, 152.2, 154.8; FTIR (neat film, cm⁻¹): 1618, 1255, 1028; EIMS (*m/z*, %) 415 (M⁺, 100), 398 (19); HRMS calcd for C₂₆H₂₅NO₄ 415.1784, found 415.1774. Anal. calcd for C₂₆H₂₅NO₄: C, 75.16; H, 6.06; N, 3.37. Found: C, 75.12; H, 6.04; N, 3.45.

6,7-Dimethoxy-3-(3,4-dimethoxyphenyl)-4-(3-methoxyphenyl)-1-methylisoquinoline (2b). 74%.

Beige powder: mp 140-141°C (MeOH); R_f 0.57 (30% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 2.96 (3H, s), 3.57 (3H, s), 3.66 (3H, s), 3.75 (3H, s), 3.80 (3H, s), 4.02 (3H, s), 6.71-6.73 (2H, m), 6.82-6.88 (2H, m), 6.93 (1H, s), 7.06 (1H, dd, J 8.3, 1.6 Hz), 7.28 (1H, dd, J 8.3, 2.0 Hz), 7.30 (1H, s); ¹³C NMR (63 MHz, CDCl₃) δ 22.6, 55.0, 55.2, 55.5, 55.8, 103.5, 104.3, 110.3, 112.6, 113.2, 116.4, 121.6, 122.5, 123.5, 127.5, 129.5, 132.3, 133.6, 139.6, 147.5, 147.6, 149.1, 152.0, 154.8, 159.5; FTIR (neat film, cm⁻¹): 1598, 1257, 1024; EIMS (m/z, %) 445 (M⁺, 100); HRMS calcd for C₂₇H₂₇NO₅ 445.1889, found 445.1885. Anal. calcd for C₂₇H₂₇NO₅: C, 72.79; H, 6.11; N, 3.14. Found: C, 72.71; H, 6.13; N, 3.12.

6,7-Dimethoxy-3-(3,4-dimethoxyphenyl)-1-methyl-4-(4-nitrophenyl)isoquinoline (2c). 51%.

Yellow powder: mp 202-203°C (MeOH); R_f 0.43 (20% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 3.00 (3H, s), 3.68 (3H, s), 3.77 (3H, s), 3.82 (3H, s), 4.07 (3H, s), 6.66 (1H, d, J 8.3 Hz), 6.74 (1H, s), 6.78 (1H, dd, J 8.3, 1.2 Hz), 6.86 (1H, d, J 1.6 Hz), 7.36 (1H, s), 7.44 (2H, d, J 7.9 Hz), 8.23 (2H, d, J 8.3 Hz); ¹³C NMR (63 MHz, CDCl₃) δ 22.8, 55.6, 55.7, 55.8, 56.0, 103.4, 103.9, 110.4, 113.3, 121.8, 122.9, 123.6, 125.7, 131.6, 132.2, 132.9, 146.1, 146.7, 148.1, 148.2, 148.3, 149.5, 152.7, 156.1; FTIR (neat film, cm⁻¹): 1604, 1508, 1345, 1259, 1026; EIMS (m/z, %) 460 (M⁺, 100), 233 (17); HRMS calcd for C₂₆H₂₄N₂O₄ 460.1634, found 460.1626. Anal. calcd for C₂₆H₂₄N₂O₄: C, 67.82; H, 5.25; N, 6.08. Found: C, 67.80; H, 5.19; N, 6.11.

3-(3,4-Dimethoxyphenyl)-7-fluoro-4-(4-fluorophenyl)-1-methylisoquinoline (2e). 75%. Yellow powder: mp 172-173°C (MeOH); R_f 0.43 (7% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 3.02 (3H, s), 3.64 (3H, s), 3.84 (3H, s), 6.74 (1H, d, J 8.3 Hz), 6.81 (1H, d, J 1.9 Hz), 6.99 (1H, dd, J 8.3, 1.9 Hz), 7.08 (2H, ddd, J 8.7, 8.3, 2.4 Hz), 7.20 (2H, ddd, J 8.7, 5.5, 2.0 Hz), 7.36 (1H, ddd, J 9.5, 8.3, 2.4 Hz), 7.63 (1H, dd, J 9.5, 5.6 Hz), 7.77 (1H, dd, J 9.5, 2.4 Hz); ¹³C NMR (63 MHz, CDCl₃) δ 22.7, 55.4, 55.7, 109.0 (d, J_{C-F} 21.5 Hz), 110.4, 113.4, 115.4 (d, J_{C-F} 21.5 Hz), 120.1 (d, J_{C-F} 25.1 Hz),

122.9, 126.8 (d, $J_{\text{C-F}}$ 7.2 Hz), 127.3, 128.6 (d, $J_{\text{C-F}}$ 9.0 Hz), 132.8 (d, $J_{\text{C-F}}$ 7.2 Hz), 132.9, 133.0 (d, $J_{\text{C-F}}$ 3.4 Hz), 133.5 (d, $J_{\text{C-F}}$ 3.6 Hz), 147.9, 148.1, 148.6, 157.1, 160.2 (d, $J_{\text{C-F}}$ 249.5 Hz), 161.9 (d, $J_{\text{C-F}}$ 247.7 Hz); FTIR (neat film, cm^{-1}): 1610, 1260, 1027; EIMS (m/z , %) 390 (M^+ , 100), 374 (23), 304 (11); HRMS calcd for $\text{C}_{24}\text{H}_{19}\text{F}_2\text{NO}_2$ 391.1384, found 391.1379. Anal. calcd for $\text{C}_{24}\text{H}_{19}\text{F}_2\text{NO}_2$: C, 73.65; H, 4.89; F, 9.71; N, 3.58. Found: C, 73.60; H, 4.94; F, 9.78; N, 3.52.

3-(3,4-Dimethoxyphenyl)-1-methyl-4-phenylisoquinoline (2f). 78%. Beige powder: mp 78-79°C (MeOH); R_f 0.46 (15% EtOAc/ CH_2Cl_2); ^1H NMR (250 MHz, CDCl_3) δ 3.07 (3H, s), 3.57 (3H, s), 3.83 (3H, s), 6.77 (1H, d, J 8.3 Hz), 6.82 (1H, d, J 1.9 Hz), 7.14 (1H, dd, J 8.3, 1.9 Hz), 7.24-7.27 (2H, m), 7.33-7.42 (3H, m), 7.54-7.59 (2H, m), 7.65 (1H, ddd, J 3.9, 1.9, 0.8 Hz), 8.17 (1H, dd, J 6.7, 2.8 Hz); ^{13}C NMR (63 MHz, CDCl_3) δ 22.4, 55.1, 55.3, 110.2, 113.8, 122.8, 125.1, 125.6, 125.7, 126.0, 126.8, 128.1, 128.2, 129.5, 131.0, 133.1, 135.7, 137.7, 147.4, 147.7, 148.3, 157.2; FTIR (neat film, cm^{-1}): 1600, 1257, 1027; EIMS (m/z , %) 355 (M^+ , 94), 354 ($\text{M}-1$, 100), 338 (17); HRMS calcd for $\text{C}_{24}\text{H}_{21}\text{NO}_2$ 355.1572, found 355.1563. Anal. calcd for $\text{C}_{24}\text{H}_{21}\text{NO}_2$: C, 81.10; H, 5.96; N, 3.94. Found: C, 81.16; H, 5.91; N, 3.99.

3,4-Diphenyl-1-methylisoquinoline (2g). 77%. Colorless prisms: mp 155-156°C (MeOH) (Lit.⁵ 154-156°C (hexane)); R_f 0.42 (CH_2Cl_2); ^{13}C NMR (63 MHz, CDCl_3) δ 22.5, 125.2, 125.8, 125.9, 126.2, 126.6, 126.8, 127.3, 127.9, 128.8, 129.6, 130.0, 131.1, 135.6, 137.2, 140.7, 149.0, 157.4; FTIR (neat film, cm^{-1}): 1613.

1-Methyl-3-(4-methylphenyl)-4-phenylisoquinoline (2h). 78%. Colorless crystals: mp 122-123°C (MeOH) (Lit.⁶ 126-128°C (EtOH)); R_f 0.38 (CH_2Cl_2); ^1H NMR (250 MHz, CDCl_3) δ 2.32 (3H, s), 3.15 (3H, s), 7.06 (2H, d, J 7.9 Hz), 7.29 (2H, dd, J 7.9, 2.0 Hz), 7.32 (2H, d, J 7.9 Hz), 7.36-7.39 (3H, m), 7.58-7.61 (2H, m), 7.71 (1H, dd, J 7.9, 2.3 Hz), 8.21 (1H, dd, J 6.3, 2.3 Hz); ^{13}C NMR (63 MHz, CDCl_3) δ 21.0, 22.6, 125.3, 125.8, 126.0, 126.2, 126.9, 128.1, 128.2, 128.7, 129.7, 130.0, 131.2,

135.8, 136.4, 137.6, 137.9, 149.1, 157.5; FTIR (neat film, cm^{-1}): 1610; EIMS (m/z , %) 309 (M^+ , 72), 308 ($M-1$, 100).

3,4-Diphenyl-1-methyl-6,7-methylenedioxyisoquinoline (2i). 75%. Colorless crystals: mp 190-191°C (MeOH); R_f 0.70 (15% EtOAc/ CH_2Cl_2); ^1H NMR (250 MHz, CDCl_3) δ 2.95 (3H, s), 6.00 (2H, s), 6.92 (1H, s), 7.17-7.22 (5H, m), 7.32-7.37 (5H, m), 7.40 (1H, s); ^{13}C NMR (63 MHz, CDCl_3) δ 22.8, 101.3, 101.4, 102.3, 123.0, 126.6, 126.9, 127.4, 128.1, 129.9, 131.1, 128.9, 134.1, 137.8, 140.9, 147.5, 148.7, 149.1, 155.2; FTIR (neat film, cm^{-1}): 1615, 1245, 1039; EIMS (m/z , %) 339 (M^+ , 87), 338 ($M-1$, 100); HRMS calcd for $\text{C}_{23}\text{H}_{17}\text{NO}_2$ 339.1259, found 339.1248. Anal. calcd for $\text{C}_{23}\text{H}_{17}\text{NO}_2$: C, 81.40; H, 5.05; N, 4.13. Found: C, 81.32; H, 5.09; N, 4.08.

6-Methoxy-4-(3-methoxyphenyl)-1-methyl-3-phenylisoquinoline (2j). 43%. Yellow powder: mp 114-115°C (MeOH); R_f 0.43 (5% EtOAc/ CH_2Cl_2); ^1H NMR (250 MHz, CDCl_3) δ 3.01 (3H, s), 3.68 (3H, s), 3.74 (3H, s), 6.74 (1H, m), 6.82-6.86 (2H, m), 6.97 (1H, d, J 2.4 Hz), 7.15-7.29 (5H, m), 7.34-7.38 (2H, m), 8.10 (1H, d, J 9.1 Hz); ^{13}C NMR (63 MHz, CDCl_3) δ 22.6, 55.2, 55.2, 104.4, 112.9, 116.6, 118.7, 121.8, 123.8, 126.9, 127.4, 127.5, 128.4, 129.2, 130.0, 137.9, 139.1, 141.1, 150.0, 157.0, 159.4, 160.5; FTIR (neat film, cm^{-1}): 1625, 1223, 1031; EIMS (m/z , %) 354 ($M-1$, 100), 311 (13); HRMS calcd for $\text{C}_{24}\text{H}_{21}\text{NO}_2$ 355.1572, found 355.1561. Anal. calcd for $\text{C}_{24}\text{H}_{21}\text{NO}_2$: C, 81.10; H, 5.96; N, 3.94. Found: C, 81.17; H, 6.01; N, 3.85.

6-Methoxy-4-(3-methoxyphenyl)-1-methyl-3-(4-methylphenyl)isoquinoline (2k). 45%. Yellow powder: mp 108-109°C (MeOH); R_f 0.42 (30% EtOAc/ CH_2Cl_2); ^1H NMR (250 MHz, CDCl_3) δ 2.28 (3H, s), 3.00 (3H, s), 3.70 (3H, s), 3.74 (3H, s), 6.77 (1H, m), 6.84-6.88 (2H, m), 6.95 (1H, d, J 2.0 Hz), 7.00 (2H, d, J 7.9 Hz), 7.13-7.30 (4H, m), 8.09 (1H, d, J 9.1 Hz); ^{13}C NMR (63 MHz, CDCl_3) δ 21.1, 22.6, 55.2, 104.3, 112.8, 115.0, 116.6, 118.5, 121.7, 123.8, 127.3, 128.3, 128.5, 129.2, 129.8, 136.4, 137.9, 138.2, 139.3, 149.9, 156.9, 159.4, 160.4; FTIR (neat film, cm^{-1}): 1615, 1267, 1048;

EIMS (m/z, %) 368 (M-1, 100), 325 (12); HRMS calcd for C₂₅H₂₃NO₂ 369.1792, found 369.1799.

Anal. calcd for C₂₅H₂₃NO₂: C, 81.27; H, 6.27; N, 3.79. Found: C, 81.29; H, 6.24; N, 3.91.

The same procedure on triarylethanone **3d** provided the following products:

3-(3,4-Dimethoxyphenyl)-6-methoxy-1-methyl-4-(3-methoxyphenyl)isoquinoline (2d) (60%) as beige powder: mp 142-143°C (MeOH); *R_f* 0.35 (15% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 2.98 (3H, s), 3.59 (3H, s), 3.68 (3H, s), 3.71 (3H, s), 3.81 (3H, s), 6.72-6.76 (2H, m), 6.84-6.93 (4H, m), 7.09 (1H, dd, *J* 8.3, 1.6 Hz), 7.16 (1H, dd, *J* 9.1, 2.3 Hz), 7.28 (1H, dd, *J* 7.9, 7.5 Hz), 8.05 (1H, d, *J* 9.1 Hz); ¹³C NMR (63 MHz, CDCl₃) δ 22.4, 55.0, 55.3, 55.6, 104.2, 110.3, 112.6, 113.3, 116.5, 118.3, 121.4, 122.7, 123.5, 127.2, 127.7, 129.2, 133.4, 137.8, 139.5, 147.5, 147.8, 149.0, 156.8, 159.5, 160.3; FTIR (neat film, cm⁻¹): 1617, 1257, 1027; EIMS (m/z, %) 414 (M-1, 100), 398 (11); HRMS calcd for C₂₆H₂₅NO₄ 415.1784, found 415.1774. Anal. calcd for C₂₆H₂₅NO₄: C, 75.16; H, 6.06; N, 3.37. Found: C, 75.11; H, 6.09; N, 3.41.

3-(3,4-Dimethoxyphenyl)-8-methoxy-1-methyl-4-(3-methoxyphenyl)isoquinoline (2l) (15%) as beige powder: mp 95-96°C (MeOH); *R_f* 0.62 (15% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 3.19 (3H, s), 3.59 (3H, s), 3.70 (3H, s), 3.82 (3H, s), 3.99 (3H, s), 6.73-6.77 (2H, m), 6.82-6.88 (4H, m), 7.14 (1H, dd, *J* 8.3, 1.9 Hz), 7.18 (1H, d, *J* 8.3 Hz), 7.28 (1H, dd, *J* 7.9, 7.7 Hz), 7.42 (1H, dd, *J* 8.3, 8.3 Hz); ¹³C NMR (63 MHz, CDCl₃) δ 27.2, 55.1, 55.4, 55.6, 105.6, 110.3, 112.5, 113.3, 116.7, 118.1, 118.7, 122.8, 123.8, 127.4, 129.2, 129.9, 133.3, 136.7, 139.9, 147.6, 147.9, 148.4, 157.3, 157.9, 159.5; FTIR (neat film, cm⁻¹): 1605, 1259, 1027; EIMS (m/z, %) 415 (M⁺, 100), 398 (11); HRMS calcd for C₂₆H₂₅NO₄ 415.1784, found 415.1778. Anal. calcd for C₂₆H₂₅NO₄: C, 75.16; H, 6.06; N, 3.37. Found: C, 75.25; H, 5.99; N, 3.33.

The same procedure on triarylethanone **3l** provided *9-(3',4'-dimethoxybenzylidene)-9H-2,3,6,7-tetramethoxyfluorene (4a)* (77%) as beige powder: mp 184-185°C (MeOH); *R_f* 0.30 (40% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 3.83 (3H, s), 3.90 (3H, s), 3.98 (3H, s), 4.02 (3H, s),

4.13 (6H, s), 6.99-7.12 (3H, m), 7.23 (1H, d, J 8.7 Hz), 7.34 (1H, s), 7.50 (1H, s), 7.81 (1H, s), 7.86 (1H, s); ^{13}C NMR (63 MHz, CDCl_3) δ 55.7, 55.9, 56.0, 102.6, 102.9, 106.9, 108.1, 110.1, 113.0, 122.0, 123.8, 124.8, 124.9, 125.3, 126.0, 133.9, 136.0, 148.1, 148.3, 148.5, 148.9, 149.1; FTIR (neat film, cm^{-1}): 1244, 1096; EIMS (m/z , %) 434 (M^+ , 100), 419 (17); HRMS calcd for $\text{C}_{26}\text{H}_{26}\text{O}_6$ 434.1729, found 434.1715. Anal. calcd for $\text{C}_{26}\text{H}_{26}\text{O}_6$: C, 71.87; H, 6.03. Found: C, 71.94; H, 6.05.

The same procedure on triarylethanone **3m** provided 9-(4'-methylbenzylidene)-9H-2,3,6,7-tetramethoxyfluorene (**4b**) (75%) as beige powder: mp 158-160°C (MeOH); R_f 0.36 (CH_2Cl_2); ^1H NMR (250 MHz, CDCl_3) δ 2.47 (3H, s), 3.84 (3H, s), 4.02 (3H, s), 4.14 (6H, s), 7.21 (1H, s), 7.30-7.34 (3H, m), 7.45 (2H, d, J 7.6 Hz), 7.49 (1H, s), 7.82 (1H, s), 7.86 (1H, s); ^{13}C NMR (63 MHz, CDCl_3) δ 21.2, 55.6, 55.8, 56.0, 102.6, 102.8, 107.0, 108.2, 123.7, 124.7, 124.9, 125.2, 126.1, 129.0, 129.7, 136.2, 136.7, 138.2, 148.3, 148.8, 148.8, 148.9; FTIR (neat film, cm^{-1}): 1251, 1092; EIMS (m/z , %) 388 (M^+ , 100), 373 (17), 345 (14), 330 (11), 314 (10); HRMS calcd for $\text{C}_{25}\text{H}_{24}\text{O}_4$ 388.1675, found 388.1664. Anal. calcd for $\text{C}_{25}\text{H}_{24}\text{O}_4$: C, 77.30; H, 6.23. Found: C, 77.36; H, 6.15.

The same procedure on triarylethanone **3n** provided 9-benzylidene-9H-2,3,6,7-tetramethoxyfluorene (**4c**)⁷ (71%) as beige powder: mp 43-44°C (MeOH); R_f 0.35 (CH_2Cl_2); ^1H NMR (250 MHz, CDCl_3) δ 3.82 (3H, s), 4.03 (3H, s), 4.14 (6H, s), 7.22 (1H, s), 7.30 (1H, s), 7.46-7.58 (6H, m), 7.82 (1H, s), 7.87 (1H, s); ^{13}C NMR (63 MHz, CDCl_3) δ 55.6, 55.8, 56.0, 58.1, 102.6, 102.9, 106.4, 108.2, 123.9, 124.8, 125.0, 125.2, 126.1, 127.1, 128.3, 129.8, 136.3, 141.2, 148.4, 148.9, 148.9, 149.2; FTIR (neat film, cm^{-1}): 1254, 1042; EIMS (m/z , %) 374 (M^+ , 100), 359 (10), 316 (10), 300 (10).

The same procedure on triarylethanone **3o** provided 9-(3',4'-dimethoxybenzylidene)-9H-2,3-dimethoxy-6,7-methylenedioxyfluorene (**4d**) (50%) as mixture of *E/Z* isomers. Orange powder: mp 197-200°C (MeOH); R_f 0.28 (CH_2Cl_2); ^1H NMR (250 MHz, CDCl_3) δ 3.83 (3H, s), 3.86 (3H, s), 3.91 (6H, s), 3.98 (3H, s), 4.02 (3H, s), 4.11 (3H, s), 4.14 (3H, s), 6.06 (2H, s), 6.10 (2H, s), 6.99-7.12 (6H, m),

7.19 (1H, s), 7.20 (1H, s), 7.32 (1H, s), 7.33 (1H, s), 7.46 (1H, s), 7.48 (1H, s), 7.81-7.92 (4H, m); ^{13}C NMR (63 MHz, CDCl_3) δ 55.7, 55.9, 56.0, 56.1, 100.1, 101.1, 101.2, 102.7, 103.0, 105.6, 106.7, 106.9, 108.2, 110.4, 111.0, 113.0, 113.2, 122.0, 122.1, 123.3, 123.9, 124.2, 125.2, 125.3, 125.4, 126.0, 127.2, 133.8, 133.9, 136.0, 136.1, 146.7, 146.8, 147.0, 147.8, 147.9, 148.2, 148.5, 148.6, 148.9, 149.1; FTIR (neat film, cm^{-1}): 1268, 1236, 1039; EIMS (m/z , %) 418 (M^+ , 100), 388 (13), 372 (11); HRMS calcd for $\text{C}_{25}\text{H}_{22}\text{O}_6$ 418.1416, found 418.1409. Anal. calcd for $\text{C}_{25}\text{H}_{22}\text{O}_6$: C, 71.76; H, 5.30. Found: C, 71.73; H, 5.37.

The same procedure on triarylethanone **3p** provided 9-(3',4'-dimethoxybenzylidene)-9H-2,3,6-trimethoxyfluorene (**4e**) (71%) as mixture of *E/Z* isomers. Beige powder: mp 74-77°C (MeOH); R_f 0.26 (CH_2Cl_2); ^1H NMR (250 MHz, CDCl_3) δ 3.84 (3H, s), 3.84 (3H, s), 3.91 (3H, s), 3.94 (3H, s), 3.98 (3H, s), 4.02 (3H, s), 4.04 (3H, s), 4.13 (3H, s), 4.14 (6H, s), 7.00-7.13 (5H, m), 7.19-7.24 (3H, m), 7.32 (1H, s), 7.35 (1H, s), 7.46-7.54 (3H, m), 7.78 (1H, s), 7.82-7.89 (3H, m), 7.96 (1H, s); ^{13}C NMR (63 MHz, CDCl_3) δ 55.2, 55.5, 55.7, 55.8, 55.9, 56.0, 102.6, 102.9, 103.4, 103.7, 106.9, 108.1, 111.0, 113.0, 113.7, 115.6, 122.0, 123.7, 124.6, 124.8, 124.9, 125.4, 125.8, 126.1, 126.5, 130.1, 130.4, 130.9, 133.5, 133.8, 135.3, 135.9, 148.1, 148.3, 148.5, 148.7, 148.9, 149.1, 149.1, 158.1, 158.7; FTIR (neat film, cm^{-1}): 1247, 1030; EIMS (m/z , %) 404 (M^+ , 100), 389 (15), 374 (10), 187 (11), 179 (15) and 404 (M^+ , 100), 330 (12), 281 (14), 202 (13), 191 (13); HRMS calcd for $\text{C}_{25}\text{H}_{24}\text{O}_5$ 404.1624, found 404.1612. Anal. calcd for $\text{C}_{25}\text{H}_{24}\text{O}_5$: C, 74.24; H, 5.98. Found: C, 74.35; H, 5.92.

N-Benzyl-N-[1,2,2-tris(3,4-dimethoxyphenyl)ethyl]amine (7). To a stirred mixture of triarylethanone **3l** (0.33 mmol), *N*-benzylamine (1.65 mmol) and Et_3N (1.98 mmol) in dry THF (3 mL) at -78°C and under argon, TiCl_4 (0.33 mmol, 0.33 mL of a 1M solution in CH_2Cl_2) was added dropwise. After 5 min of stirring at -78°C, the resulting suspension was allowed to warm to -20°C, and stirred for 1.5 h. To this solution, a methanolic solution (1.5 mL) of NaBH_4 (1.48 mmol) was added.

The resulting mixture was stirred for 3 h, quenched with saturated aqueous K_2CO_3 (5 mL) and water (10 mL), and extracted with CH_2Cl_2 (3 x 5 mL). The combined organic extracts were dried over anhydrous sodium sulfate and evaporated *in vacuo* to give a residue which was purified by flash chromatography on silicagel using 10% EtOAc/ CH_2Cl_2 as eluent affording benzylamine **7** (24%) as colorless oil; R_f 0.40 (30% hexane/EtOAc); ^1H NMR (250 MHz, CDCl_3) δ 1.81 (1H, bs), 3.42 (1H, d, J 13.5 Hz), 3.62 (3H, s), 3.66 (1H, d, J 13.5 Hz), 3.73 (3H, s), 3.80 (6H, s), 3.83 (3H, s), 3.87 (3H, s), 3.93 (1H, d, J 9.5 Hz), 4.10 (1H, d, J 9.5 Hz), 6.37 (1H, s), 6.48-6.58 (2H, m), 6.63-6.69 (2H, m), 6.77-6.85 (4H, m), 7.08 (2H, d, J 6.7 Hz), 7.24-7.28 (3H, m); ^{13}C NMR (63 MHz, CDCl_3) δ 51.6, 55.6, 55.7, 55.8, 58.9, 65.3, 110.2, 110.4, 110.7, 111.2, 111.5, 112.0, 120.0, 120.2, 121.1, 126.8, 128.2, 134.0, 134.1, 134.9, 140.2, 147.0, 147.7, 147.8, 147.9, 148.6, 148.9; FTIR (neat film, cm^{-1}): 3350, 1602, 1262, 1028; EIMS (m/z , %) 544 ($M+1$, 46), 527 (11), 437 (61), 286 (100), 91 (25); HRMS calcd for $\text{C}_{33}\text{H}_{37}\text{NO}_6$ 543.2621, found 543.2609. Anal. calcd for $\text{C}_{33}\text{H}_{37}\text{NO}_6$: C, 72.91; H, 6.86; N, 2.58. Found: C, 72.87; H, 6.83; N, 2.66.

2,2-Bis(3-methoxyphenyl)-1-phenylethanone oxime (8a). A solution of triarylethanone **3j** (0.30 mmol) and hydroxylamine hydrochloride (1.8 mmol) in dry pyridine (6 mL) was refluxed for 2.5 h. The reaction mixture was allowed to cool to room temperature and evaporated to dryness *in vacuo*, and the residue was partitioned between water (4 mL) and CH_2Cl_2 (4 mL). The organic extract was washed with water (2 x 4 mL) and the aqueous layer was extracted with CH_2Cl_2 (2 x 4 mL). The combined organic extracts were dried over anhydrous sodium sulfate and evaporated *in vacuo* to give a residue which was purified by flash chromatography on silicagel using 30% hexane/ Et_2O as eluent affording oxime **8a** (96%) as separable *E/Z* 1.2:1 mixture of diastereoisomers. *E*-2,2-Bis(3-methoxyphenyl)-1-phenylethanone oxime **8a**: 52%. White powder: mp 136-137°C (MeOH); R_f 0.47 (50% hexane/ Et_2O); ^1H NMR (250 MHz, CDCl_3) δ 3.70 (6H, s), 6.23 (1H, s), 6.75-6.82 (6H, m), 7.15-7.30 (7H, m), 8.93

(1H, bs); ^{13}C NMR (63 MHz, CDCl_3) δ 49.5, 55.1, 112.1, 115.1, 121.8, 127.9, 128.1, 128.2, 128.6, 129.2, 129.3, 135.8, 141.0, 159.5, 159.7; FTIR (neat film, cm^{-1}): 3311, 1597, 1263, 1048; EIMS (m/z , %) 347 (M^+ , 46), 329 (100), 242 (10), 227 (22), 211 (12), 196 (12), 152 (14), 103 (17), 77 (39); HRMS calcd for $\text{C}_{22}\text{H}_{21}\text{NO}_3$ 347.1521, found 347.1516. Anal. calcd for $\text{C}_{22}\text{H}_{21}\text{NO}_3$: C, 76.06; H, 6.09; N, 4.03. Found: C, 76.16; H, 6.02; N, 4.00. Z-2,2-Bis(3-methoxyphenyl)-1-phenylethanone oxime **8a'**: 44%. Yellow oil; R_f 0.46 (50% hexane/ Et_2O); ^1H NMR (250 MHz, CDCl_3) δ 3.73 (6H, s), 5.31 (1H, s), 6.76-6.82 (4H, m), 6.90 (2H, d, J 7.9 Hz), 7.24 (2H, d, J 7.9 Hz), 7.31-7.40 (5H, m), 8.73 (1H, bs); ^{13}C NMR (63 MHz, CDCl_3) δ 55.1, 57.3, 112.0, 115.2, 121.7, 127.9, 128.7, 129.3, 133.9, 141.4, 159.3, 159.6; FTIR (neat film, cm^{-1}): 3281, 1594, 1261, 1051; EIMS (m/z , %) 347 (M^+ , 46), 329 (100), 242 (12), 227 (22), 211 (11), 196 (12), 152 (17), 77 (44); HRMS calcd for $\text{C}_{22}\text{H}_{21}\text{NO}_3$ 347.1521, found 347.1509. Anal. calcd for $\text{C}_{22}\text{H}_{21}\text{NO}_3$: C, 76.06; H, 6.09; N, 4.03. Found: C, 76.14; H, 6.11; N, 3.95.

General procedure for reductive amination of triarylethanones **3 via oximes. Synthesis of triarylethylamines **9**.** A solution of triarylethanone **3** (0.90 mmol) and hydroxylamine hydrochloride (5.4 mmol) in dry pyridine (18 mL) was refluxed for 2-2.5 h. The reaction mixture was allowed to cool to room temperature and evaporated to dryness *in vacuo*, and the residue was partitioned between water (10 mL) and CH_2Cl_2 (10 mL). The organic extract was washed with water (2 x 10 mL) and the aqueous layer was extracted with CH_2Cl_2 (2 x 10 mL). The combined organic extracts were dried over anhydrous sodium sulfate and evaporated *in vacuo*. Without performing further purification, the resulting mixture was dissolved in the smallest amount of THF, 70% aqueous formic acid (17 mL) was added, and after cooling the solution with an ice bath, was added zinc powder (26.7 mmol) portionwise over 30 min. The reaction mixture was stirred for 17-19 h at room temperature, filtered on sand and washed with EtOAc. The filtrate was neutralized with concentrated ammonia solution to pH 8 and then

extracted with EtOAc (3 x 30 mL). The organic phase was washed with water (50 mL), dried over anhydrous sodium sulfate and evaporated *in vacuo* to give a residue which was purified by flash chromatography on silicagel using 10-40% EtOAc/CH₂Cl₂ or 2-5% MeOH/CH₂Cl₂ as eluent.

2,2-Bis(3-methoxyphenyl)-1-phenylethylamine (9a). 90%. Colorless oil; *R_f* 0.48 (2% MeOH/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 1.81 (2H, bs), 3.63 (3H, s), 3.80 (3H, s), 4.07 (1H, d, *J* 10.3 Hz), 4.68 (1H, d, *J* 10.3 Hz), 6.56 (1H, dd, *J* 8.3, 2.0 Hz), 6.62 (1H, dd, *J* 2.4, 2.0 Hz), 6.70 (1H, d, *J* 7.9 Hz), 6.78 (1H, dd, *J* 7.9, 2.4 Hz), 6.97-7.03 (2H, m), 7.08 (1H, d, *J* 8.3 Hz), 7.14-7.31 (6H, m); ¹³C NMR (63 MHz, CDCl₃) δ 54.9, 55.1, 59.7, 60.7, 111.1, 111.7, 114.4, 114.5, 120.6, 120.7, 127.0, 127.5, 128.0, 128.9, 129.7, 143.5, 143.6, 143.8, 159.0, 159.8; FTIR (neat film, cm⁻¹): 3382-3290, 1600, 1262, 1048; EIMS (*m/z*, %) 331 (M-2, 3), 106 (100); HRMS calcd for C₂₂H₂₃NO₂ 333.1729, found 333.1723. Anal. calcd for C₂₂H₂₃NO₂: C, 79.25; H, 6.95; N, 4.20. Found: C, 79.32; H, 6.91; N, 4.22.

2,2-Bis(3-methoxyphenyl)-1-(4-methyl)phenylethylamine (9b). 61%. Yellowish oil; *R_f* 0.44 (2% MeOH/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 1.57 (2H, bs), 2.29 (3H, s), 3.64 (3H, s), 3.80 (3H, s), 4.13 (1H, d, *J* 9.9 Hz), 4.72 (1H, d, *J* 9.9 Hz), 6.56 (1H, dd, *J* 8.3, 2.0 Hz), 6.64 (1H, dd, *J* 2.0, 2.0 Hz), 6.72 (1H, d, *J* 7.9 Hz), 6.78 (1H, dd, *J* 7.9, 2.0 Hz), 6.99 (2H, d, *J* 8.3 Hz), 7.00-7.06 (2H, m), 7.08 (1H, d, *J* 7.5 Hz), 7.17 (2H, d, *J* 8.3 Hz), 7.27 (1H, dd, *J* 7.9, 7.9 Hz); ¹³C NMR (63 MHz, CDCl₃) δ 20.8, 54.7, 54.8, 59.2, 60.5, 110.8, 111.5, 114.3, 114.4, 120.6, 127.2, 128.6, 128.7, 129.5, 136.2, 140.6, 143.7, 143.9, 158.9, 159.6; FTIR (neat film, cm⁻¹): 3360, 3280, 1600, 1263, 1050; EIMS (*m/z*, %) 345 (M-2, 7), 120 (100), 93 (11); HRMS calcd for C₂₃H₂₅NO₂ 347.1885, found 347.1892. Anal. calcd for C₂₃H₂₅NO₂: C, 79.51; H, 7.25; N, 4.03. Found: C, 79.43; H, 7.29; N, 4.11.

1,2,2-Tris(3,4-dimethoxyphenyl)ethylamine (9c). 52%. Yellowish oil; *R_f* 0.40 (8% MeOH/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 1.61 (2H, bs), 3.65 (3H, s), 3.72 (3H, s), 3.76 (3H, s), 3.78 (3H, s), 3.83

(3H, s), 3.85 (3H, s), 3.91 (1H, d, *J* 9.9 Hz), 4.53 (1H, d, *J* 9.9 Hz), 6.47 (1H, d, *J* 2.0 Hz), 6.59 (2H, m), 6.64 (1H, d, *J* 8.3 Hz), 6.68 (1H, dd, *J* 8.3, 1.6 Hz), 6.79 (1H, d, *J* 1.6 Hz), 6.85 (1H, d, *J* 8.3 Hz), 6.88 (1H, d, *J* 1.6 Hz), 7.00 (1H, dd, *J* 8.3, 2.0 Hz); ¹³C NMR (63 MHz, CDCl₃) δ 55.4, 55.5, 55.5, 59.7, 59.8, 110.2, 110.3, 110.5, 111.2, 111.8, 111.9, 119.6, 119.7, 120.1, 134.7, 135.0, 136.3, 146.9, 147.6, 147.7, 148.0, 148.4, 148.8; FTIR (neat film, cm⁻¹): 3352, 3292, 1593, 1266, 1027; EIMS (m/z, %) 166 (100); HRMS calcd for C₂₆H₃₁NO₆ 453.2151, found 453.2139. Anal. calcd for C₂₆H₃₁NO₆: C, 68.86; H, 6.89; N, 3.09. Found: C, 68.96; H, 6.93; N, 2.96.

2,2-Bis(3,4-dimethoxyphenyl)-1-(4-methyl)phenylethylamine (9d). 50%. White powder: mp 69-70°C (Et₂O); *R*_f 0.46 (8% MeOH/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 1.77 (2H, bs), 2.25 (3H, s), 3.64 (3H, s), 3.74 (3H, s), 3.86 (3H, s), 3.87 (3H, s), 3.99 (1H, d, *J* 9.9 Hz), 4.54 (1H, d, *J* 9.9 Hz), 6.43 (1H, d, *J* 2.0 Hz), 6.59-6.68 (2H, m), 6.86 (1H, d, *J* 8.3 Hz), 6.89 (1H, d, *J* 2.0 Hz), 6.98-7.01 (3H, m), 7.11 (2H, d, *J* 7.9 Hz); ¹³C NMR (63 MHz, CDCl₃) δ 20.9, 55.4, 55.5, 55.8, 59.5, 59.8, 110.5, 111.3, 112.1, 119.9, 120.0, 127.3, 128.7, 134.9, 135.0, 136.5, 140.8, 146.9, 147.6, 147.9, 148.9; FTIR (neat film, cm⁻¹): 3360, 3290, 1592, 1268, 1029; EIMS (m/z, %) 287 (10), 120 (100); HRMS calcd for C₂₅H₂₉NO₄ 407.2097, found 407.2088. Anal. calcd for C₂₅H₂₉NO₄: C, 73.68; H, 7.17; N, 3.44. Found: C, 73.61; H, 7.10; N, 3.51.

(3*R,4*R**)-6-Methoxy-4-(3-methoxyphenyl)-3-phenyl-1,2,3,4-tetrahydroisoquinoline (5).**

Formaldehyde (0.1 mmol, 0.02 mL of a 38% solution in water) was added to a stirred solution of amine **9a** (0.29 mmol) in the smallest amount of THF and HCl (5.6 mL of 1M solution in water) under argon at room temperature. The resultant stirred mixture was heated to 60°C for 4 days. After cooling, the suspension was basified with concentrated ammonia solution to pH 8 and then extracted with CH₂Cl₂ (3 x 5 mL). The organic phase was dried over anhydrous sodium sulfate and evaporated *in vacuo* to give a residue which was purified by flash chromatography on silicagel using 35% EtOAc/Et₂O as

eluent affording tetrahydroisoquinoline **5** (31%) as white powder: mp 138-139°C (Et₂O); *R*_f 0.40 (35% EtOAc/Et₂O); ¹H NMR (250 MHz, CDCl₃) δ 1.79 (1H, bs), 3.63 (3H, s), 3.65 (3H, s), 3.98 (1H, d, *J* 9.2 Hz), 4.12 (1H, d, *J* 14.6 Hz), 4.15 (1H, d, *J* 9.2, Hz), 4.32 (1H, d, *J* 14.6 Hz), 6.36-6.47 (2H, m), 6.49 (1H, d, *J* 7.9 Hz), 6.67 (1H, dd, *J* 8.3, 2.7 Hz), 6.73 (1H, dd, *J* 8.7, 2.7 Hz), 7.02-7.20 (7H, m); ¹³C NMR (63 MHz, CDCl₃) δ 48.7, 53.4, 55.1, 55.3, 67.1, 111.6, 112.2, 114.8, 115.5, 122.1, 126.8, 127.2, 127.4, 127.7, 128.1, 128.8, 139.5, 142.1, 144.7, 157.9; FTIR (neat film, cm⁻¹): 3312, 1607, 1260, 1034; EIMS (*m/z*, %) 345 (M⁺, 20), 340 (36), 238 (68), 224 (22), 209 (100), 195 (24), 165 (38), 152 (25), 132 (22), 127 (10), 115 (10), 106 (12), 91 (16); HRMS calcd for C₂₃H₂₃NO₂ 345.1729, found 345.1718. Anal. calcd for C₂₃H₂₃NO₂: C, 79.97; H, 6.71; N, 4.05. Found: C, 79.92; H, 6.77; N, 4.11.

13-Phenyl-7,12-dihydro-2,10-dimethoxy-5*H*-6,12-methano-dibenzo[*c,f*]azocine (10).

Formaldehyde (4.4 mmol, 0.88 mL of a 38% solution in water) was added to a stirred solution of amine **9a** (0.30 mmol) in the smallest amount of THF and HCl (5.6 mL of 1M solution in water) under argon at room temperature. The resultant stirred mixture was heated to 60°C for 3 days. After cooling, the suspension was basified with concentrated ammonia solution to pH 8 and then extracted with CH₂Cl₂ (3 x 5 mL). The organic phase was dried over anhydrous sodium sulfate and evaporated *in vacuo* to give a residue which was purified by flash chromatography on silicagel using 10% EtOAc/CH₂Cl₂ as eluent affording dibenzoazocine **10** (63%) as white powder: mp 146-147°C (Et₂O); *R*_f 0.48 (35% EtOAc/Et₂O); ¹H NMR (250 MHz, CDCl₃) δ 3.69 (1H, d, *J* 17.8 Hz), 3.77 (3H, s), 3.79 (3H, s), 4.11 (1H, d, *J* 17.4 Hz), 4.12 (1H, d, *J* 17.4 Hz), 4.20 (1H, s), 4.49 (1H, s), 4.77 (1H, d, *J* 17.8 Hz), 6.54 (1H, dd, *J* 8.3, 2.4 Hz), 6.68 (1H, dd, *J* 8.3, 2.7 Hz), 6.70 (1H, d, *J* 8.3 Hz), 6.86 (1H, d, *J* 2.7 Hz), 6.89 (1H, d, *J* 2.4 Hz), 6.98 (1H, d, *J* 8.3 Hz), 7.16-7.27 (3H, m), 7.44 (2H, d, *J* 7.1 Hz); ¹³C NMR (63 MHz, CDCl₃) δ 39.2, 52.1, 55.1, 55.3, 58.9, 59.4, 111.4, 112.0, 112.8, 112.9, 126.3, 126.6, 126.7,

126.9, 127.5, 128.1, 139.7, 139.9, 142.3, 157.7; FTIR (neat film, cm^{-1}): 1611, 1258, 1034; EIMS (m/z , %) 357 (M^+ , 70), 358 (14), 356 (100), 265 (12), 253 (12); HRMS calcd for $\text{C}_{24}\text{H}_{23}\text{NO}_2$ 357.1729, found 357.1722. Anal. calcd for $\text{C}_{24}\text{H}_{23}\text{NO}_2$: C, 80.64; H, 6.49; N, 3.92. Found: C, 80.69; H, 6.53; N, 3.87.

General procedure for the synthesis of *N*-triarylethylacetamides 11. Triarylethylamine **9** (1.38 mmol) was treated with acetic anhydride (9.68 mmol) at reflux for 1 h. After cooling, the excess of anhydride was evaporated *in vacuo*, and the residue was purified by flash chromatography on silicagel using 0-40% hexane/EtOAc as eluent.

***N*-[2,2-Bis(3-methoxyphenyl)-1-phenylethyl]acetamide (11a).** 65%. Beige oil; R_f 0.36 (30% hexane/EtOAc); ^1H NMR (250 MHz, CDCl_3) δ 1.78 (3H, s), 3.64 (3H, s), 3.75 (3H, s), 4.23 (1H, d, J 9.9 Hz), 5.79 (1H, dd, J 9.9, 8.7 Hz), 6.02 (1H, d, J 8.7 Hz), 6.61 (1H, d, J 8.7 Hz), 6.62 (1H, s), 6.71 (1H, dd, J 8.7, 7.5 Hz), 6.79 (1H, d, J 7.5 Hz), 6.81 (1H, s), 7.05 (1H, dd, J 7.5, 7.5 Hz), 7.16-7.27 (6H, m); ^{13}C NMR (63 MHz, CDCl_3) δ 23.1, 54.9, 55.1, 55.6, 57.4, 111.7, 112.2, 114.2, 120.6, 127.1, 127.2, 128.1, 129.1, 129.5, 141.2, 142.5, 142.8, 159.2, 159.6, 169.1; FTIR (neat film, cm^{-1}): 3380, 1648, 1600, 1261, 1042; EIMS (m/z , %) 148 (70), 106 (100), 79 (12); HRMS calcd for $\text{C}_{24}\text{H}_{25}\text{NO}_3$ 375.1834, found 375.1827. Anal. calcd for $\text{C}_{24}\text{H}_{25}\text{NO}_3$: C, 76.77; H, 6.71; N, 3.73. Found: C, 76.70; H, 6.77; N, 3.64.

***N*-[2,2-Bis(3-methoxyphenyl)-1-(4-methyl)phenylethyl]acetamide (11b).** 72%. Yellowish oil; R_f 0.38 (30% hexane/EtOAc); ^1H NMR (250 MHz, CDCl_3) δ 1.65 (3H, s), 2.25 (3H, s), 3.61 (3H, s), 3.72 (3H, s), 4.30 (1H, d, J 9.9 Hz), 5.80 (1H, dd, J 9.9, 9.5 Hz), 6.59 (1H, d, J 8.3 Hz), 6.68-6.75 (3H, m), 6.95-7.24 (8H, m); ^{13}C NMR (63 MHz, CDCl_3) δ 20.7, 22.6, 54.7, 54.8, 55.4, 57.2, 111.3, 111.8, 113.9, 114.2, 120.4, 120.6, 127.1, 128.6, 128.9, 129.2, 136.2, 138.3, 143.0, 159.3, 169.1; FTIR (neat

film, cm^{-1}): 3280, 1648, 1604, 1263, 1041; EIMS (m/z , %) 390 ($M+1$, 8), 162 (66), 120 (100), 93 (13); HRMS calcd for $\text{C}_{25}\text{H}_{27}\text{NO}_3$ 389.1991, found 389.1982. Anal. calcd for $\text{C}_{25}\text{H}_{27}\text{NO}_3$: C, 77.09; H, 6.99; N, 3.60. Found: C, 77.21; H, 6.92; N, 3.64.

***N*-[1,2,2-Tris(3,4-dimethoxyphenyl)ethyl]acetamide (11c).** 67%. White powder: mp 156-157°C (Et_2O); R_f 0.44 (5% $\text{MeOH}/\text{CH}_2\text{Cl}_2$); ^1H NMR (250 MHz, CDCl_3) δ 1.74 (3H, s), 2.22 (3H, s), 3.63 (3H, s), 3.72 (3H, s), 3.76 (3H, s), 3.79 (3H, s), 4.15 (1H, d, J 9.9 Hz), 5.67 (1H, dd, J 9.9, 9.1 Hz), 6.39 (1H, d, J 9.1 Hz), 6.46 (1H, s), 6.61-6.84 (5H, m), 6.97-7.11 (4H, m); ^{13}C NMR (63 MHz, CDCl_3) δ 20.6, 22.7, 55.3, 55.4, 56.2, 110.5, 110.7, 111.3, 111.5, 120.0, 126.9, 128.5, 133.8, 134.2, 136.1, 138.3, 146.9, 147.2, 147.9, 148.3, 168.8; FTIR (neat film, cm^{-1}): 3380, 1647, 1590, 1262, 1027; EIMS (m/z , %) 287 (100), 120 (19); HRMS calcd for $\text{C}_{27}\text{H}_{31}\text{NO}_5$ 449.2202, found 449.2198. Anal. calcd for $\text{C}_{27}\text{H}_{31}\text{NO}_5$: C, 67.86; H, 6.71; N, 2.83. Found: C, 67.81; H, 6.78; N, 2.76.

***N*-[2,2-Bis(3,4-dimethoxyphenyl)-1-(4-methyl)phenylethyl]acetamide (11d).** 60%. White powder: mp 153-154°C (Et_2O); R_f 0.56 (5% $\text{MeOH}/\text{CH}_2\text{Cl}_2$); ^1H NMR (250 MHz, CDCl_3) δ 1.82 (3H, s), 3.67 (3H, s), 3.69 (3H, s), 3.75 (3H, s), 3.78 (3H, s), 3.79 (3H, s), 3.82 (3H, s), 4.13 (1H, d, J 9.1 Hz), 5.66 (1H, dd, J 9.1, 9.1 Hz), 5.89 (1H, d, J 8.7 Hz), 6.53 (1H, d, J 1.6 Hz), 6.55 (1H, d, J 1.6 Hz), 6.59-6.68 (4H, m), 6.71-6.77 (3H, m); ^{13}C NMR (63 MHz, CDCl_3) δ 23.3, 55.4, 55.7, 56.6, 110.6, 110.7, 110.8, 110.9, 111.4, 111.5, 118.8, 120.4, 133.5, 133.9, 134.1, 147.4, 147.6, 147.9, 148.4, 148.6, 168.9; FTIR (neat film, cm^{-1}): 3282, 1646, 1592, 1260, 1026; EIMS (m/z , %) 287 (100), 208 (99), 166 (75); HRMS calcd for $\text{C}_{28}\text{H}_{33}\text{NO}_7$ 495.2257, found 495.2265. Anal. calcd for $\text{C}_{28}\text{H}_{33}\text{NO}_7$: C, 72.14; H, 6.95; N, 3.12. Found: C, 72.07; H, 6.87; N, 3.20.

General procedure for the Bischler-Napieralski heterocyclization of triarylethylacetamides 11.
Synthesis of 3,4-diaryl-3,4-dihydroisoquinolines 6. To a stirred solution of acetamide **11** (0.63 mmol) in dry acetonitrile (30 mL), POCl_3 (5.1 mmol) was added dropwise at room temperature, under

strictly anhydrous conditions. The resulting solution was refluxed for 2-3 h and after cooling, the solvent was removed *in vacuo* and the residue treated with NaOH (15 mL of 10% solution in water). The aqueous layer was extracted with CH₂Cl₂ (3 x 15 mL) and combined organic extracts were dried over anhydrous sodium sulfate and evaporated *in vacuo* to give a residue which was purified by flash chromatography on silicagel using 10% EtOAc/CH₂Cl₂ and 5% MeOH/CH₂Cl₂ as eluent. By the use of this procedure the compounds **6a** and **6b** were prepared. However, the preparation of 3,4-dihydroisoquinolines **6c** and **6d** required the addition of 6.3 mmol of P₂O₅.

(3*R,4*R**)-6-Methoxy-4-(3-methoxyphenyl)-1-methyl-3-phenyl-3,4-dihydroisoquinoline (6a).** 57%. Yellow oil; *R*_f 0.57 (10% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 2.51 (3H, s), 3.69 (3H, s), 3.71 (3H, s), 4.06 (1H, d, *J* 10.3 Hz), 4.87 (1H, dd, *J* 9.9, 1.6 Hz), 6.42 (1H, d, *J* 2.4 Hz), 6.55 (1H, s), 6.65 (1H, d, *J* 7.5 Hz), 6.71 (1H, dd, *J* 8.1, 2.4 Hz), 6.82 (1H, dd, *J* 8.3, 2.4 Hz), 7.11-7.20 (6H, m), 7.57 (1H, d, *J* 8.3 Hz); ¹³C NMR (63 MHz, CDCl₃) δ 23.3, 50.5, 54.9, 55.1, 67.4, 111.8, 113.8, 114.9, 121.4, 122.4, 126.5, 127.1, 127.5, 127.9, 129.2, 141.0, 142.8, 143.1, 159.2, 161.5, 163.4; FTIR (neat film, cm⁻¹): 1604, 1252, 1031; EIMS (*m/z*, %) 357 (M⁺, 100), 266 (12); HRMS calcd for C₂₄H₂₃NO₂ 357.1729, found 357.1727. Anal. calcd for C₂₄H₂₃NO₂: C, 80.64; H, 6.49; N, 3.92. Found: C, 80.58; H, 6.41; N, 3.96.

(3*R,4*R**)-6-Methoxy-4-(3-methoxyphenyl)-1-methyl-3-(4-methylphenyl)-3,4-dihydroisoquinoline (6b).** 67%. Orange oil; *R*_f 0.59 (10% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 2.25 (3H, s), 2.51 (3H, s), 3.67 (3H, s), 3.69 (3H, s), 4.09 (1H, d, *J* 9.9 Hz), 4.92 (1H, d, *J* 9.9 Hz), 6.42 (1H, d, *J* 2.4 Hz), 6.61 (1H, d, *J* 2.4 Hz), 6.69 (1H, d, *J* 7.9 Hz), 6.71 (1H, d, *J* 7.5 Hz), 6.81 (1H, dd, *J* 8.3, 2.4 Hz), 6.97-7.05 (4H, m), 7.14 (1H, dd, *J* 7.9, 7.9 Hz), 7.57 (1H, d, *J* 8.3 Hz); ¹³C NMR (63 MHz, CDCl₃) δ 20.9, 23.3, 50.3, 54.9, 55.1, 67.0, 111.6, 111.8, 113.9, 114.8, 121.3, 122.5, 127.0, 127.3, 128.6, 129.2, 135.9, 139.8, 140.9, 143.4, 159.4, 161.5, 162.9; FTIR (neat film,

cm⁻¹): 1608, 1256, 1041; EIMS (m/z, %) 371 (M⁺, 100), 266 (21), 264 (16), 239 (11), 211 (14), 140 (18), 105 (13); HRMS calcd for C₂₅H₂₅NO₂ 371.1885, found 371.1883. Anal. calcd for C₂₅H₂₅NO₂: C, 80.83; H, 6.78; N, 3.77. Found: C, 80.69; H, 6.72; N, 3.71.

(3*R,4*R**)-6,7-Dimethoxy-3,4-bis(3,4-dimethoxyphenyl)-1-methyl-3,4-dihydroisoquinoline (6c).** 81%. Beige powder: mp 104-105°C (MeOH); *R*_f 0.34 (10% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 2.51 (3H, s), 3.69 (3H, s), 3.74 (6H, s), 3.80 (3H, s), 3.83 (3H, s), 3.95 (3H, s), 3.96 (1H, d, *J* 12.3 Hz), 4.67 (1H, d, *J* 11.5 Hz), 6.42 (1H, s), 6.51-6.69 (3H, m), 6.92-6.97 (4H, m), 7.10 (1H, s); ¹³C NMR (63 MHz, CDCl₃) δ 23.6, 49.8, 55.8, 55.9, 56.2, 67.8, 108.6, 110.7, 110.8, 110.9, 112.0, 119.9, 121.7, 122.2, 133.3, 134.1, 135.9, 147.7, 148.3, 148.7, 151.2, 163.1; FTIR (neat film, cm⁻¹): 1630, 1260, 1029; EIMS (m/z, %) 477 (M⁺, 100), 462 (41), 340 (14), 326 (11), 287 (23), 170 (14), 151 (16); HRMS calcd for C₂₈H₃₁NO₆ 477.2151, found 477.2142. Anal. calcd for C₂₈H₃₁NO₆: C, 70.42; H, 6.54; N, 2.93. Found: C, 70.51; H, 6.59; N, 2.85.

(3*R,4*R**)-6,7-Dimethoxy-4-(3,4-dimethoxyphenyl)-1-methyl-3-(4-methylphenyl)-3,4-dihydroisoquinoline (6d).** 76%. Yellow powder: mp 73-74°C (MeOH); *R*_f 0.47 (10% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 2.20 (3H, s), 2.48 (3H, s), 3.65 (3H, s), 3.69 (3H, s), 3.78 (3H, s), 3.90 (3H, s), 3.96 (1H, d, *J* 10.7 Hz), 4.75 (1H, d, *J* 10.3 Hz), 6.39 (1H, s), 6.51-6.54 (2H, m), 6.67 (1H, d, *J* 8.3 Hz), 6.93-6.97 (4H, m), 7.07 (1H, s); ¹³C NMR (63 MHz, CDCl₃) δ 20.7, 23.3, 49.2, 55.3, 55.4, 55.9, 67.6, 108.3, 110.6, 110.7, 111.8, 121.0, 121.9, 127.2, 128.4, 132.7, 134.3, 135.7, 139.8, 147.3, 148.4, 150.9, 162.6; FTIR (neat film, cm⁻¹): 1631, 1261, 1028; EIMS (m/z, %) 431 (M⁺, 100), 416 (13), 326 (15), 241 (26); HRMS calcd for C₂₇H₂₉NO₄ 431.2097, found 431.2088. Anal. calcd for C₂₇H₂₉NO₄: C, 75.15; H, 6.77; N, 3.25. Found: C, 75.08; H, 6.72; N, 3.31.

General procedure for PIFA-mediated oxidative coupling reaction of isoquinolines 2 and 6.
Synthesis of dibenzo[*a,c*]phenanthridines 1. A solution of PIFA (0.11 mmol) in dry dichloromethane

(1 mL) was added to a stirred solution of isoquinoline **2** or **6** (0.1 mmol) in dry dichloromethane (1.5 mL) at -40°C under argon. After adding BF₃·Et₂O (0.12 mmol), the resulting blue solution was stirred for 2 h at -40°C and quenched with Na₂CO₃ (3 mL of 10% solution in water). The aqueous layer was extracted with CH₂Cl₂ (3 x 2 mL) and the combined organic extracts were dried over anhydrous sodium sulfate and evaporated *in vacuo* to give a residue which was purified by flash chromatography on silicagel using 10% EtOAc/CH₂Cl₂ as eluent.

10-Methyl-6,7,12,13-tetramethoxydibenzo[*a,c*]phenanthridine (1a). 75%. Beige powder: mp 215-216°C (MeOH); *R*_f 0.70 (10% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 3.03 (3H, s), 4.05 (6H, s), 4.12 (3H, s), 4.18 (3H, s), 7.38 (1H, s), 7.56 (1H, ddd, *J* 8.3, 7.9, 1.2 Hz), 7.62 (1H, ddd, *J* 8.3, 7.9, 1.2 Hz), 7.91 (1H, s), 8.24 (1H, s), 8.54 (1H, d, *J* 7.9 Hz), 8.76 (1H, s), 8.80 (1H, d, *J* 7.9 Hz); ¹³C NMR (63 MHz, CDCl₃) δ 23.3, 55.9, 56.0, 103.2, 104.9, 105.5, 106.8, 117.0, 122.5, 123.1, 124.6, 125.1, 126.0, 126.1, 127.7, 128.3, 128.9, 130.1, 139.5, 148.5, 149.5, 149.8, 151.8, 155.0; FTIR (neat film, cm⁻¹): 1617, 1260, 1027; EIMS (*m/z*, %) 413 (M⁺, 100), 398 (13), 367 (12); HRMS calcd for C₂₆H₂₃NO₄ 413.1627, found 413.1619. Anal. calcd for C₂₆H₂₃NO₄: C, 75.53; H, 5.61; N, 3.39. Found: C, 75.58; H, 5.57; N, 3.37.

10-Methyl-2,6,7,12,13-pentamethoxydibenzo[*a,c*]phenanthridine (1b). 86%. Beige powder: mp 185-186°C (MeOH); *R*_f 0.61 (10% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 3.07 (3H, s), 3.96 (3H, s), 4.07 (6H, s), 4.13 (3H, s), 4.18 (3H, s), 7.27 (1H, dd, *J* 9.1, 2.4 Hz), 7.43 (1H, s), 7.84 (1H, s), 8.31 (1H, d, *J* 2.4 Hz), 8.32 (1H, s), 8.47 (1H, d, *J* 9.1 Hz), 8.76 (1H, s); ¹³C NMR (63 MHz, CDCl₃) δ 23.3, 55.4, 55.9, 56.0, 102.8, 105.0, 105.6, 106.3, 110.0, 115.1, 116.7, 122.4, 124.3, 124.6, 124.7, 124.8, 129.0, 129.5, 140.0, 148.3, 148.8, 149.9, 151.8, 155.1, 157.3; FTIR (neat film, cm⁻¹): 1617, 1261, 1026; EIMS (*m/z*, %) 443 (M⁺, 100), 428 (11), 400 (10), 214 (10); HRMS calcd for C₂₇H₂₅NO₅ 443.1733, found 443.1725. Anal. calcd for C₂₇H₂₅NO₅: C, 73.12; H, 5.68; N, 3.16. Found: C, 73.16; H, 5.61; N, 3.12.

10-Methyl-3-nitro-6,7,12,13-tetramethoxydibenzo[*a,c*]phenanthridine (1c). 42%. Orange powder: mp 231-232°C (MeOH); R_f 0.47 (10% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 3.09 (3H, s), 4.06 (3H, s), 4.11 (3H, s), 4.14 (3H, s), 4.18 (3H, s), 7.49 (1H, s), 7.83 (1H, s), 8.07 (1H, s), 8.22 (1H, d, J 9.1 Hz), 8.75 (1H, s), 8.80 (1H, d, J 9.1 Hz), 9.28 (1H, s); ¹³C NMR (63 MHz, CDCl₃) δ 23.4, 56.1, 56.2, 103.0, 105.2, 105.6, 106.3, 115.9, 118.5, 118.9, 122.6, 124.1, 126.5, 128.3, 128.7, 129.8, 132.6, 141.1, 144.9, 148.9, 150.5, 152.5, 157.2; FTIR (neat film, cm⁻¹): 1617, 1508, 1347, 1265, 1045; EIMS (m/z, %) 458 (M⁺, 100), 443 (10), 428 (14), 412 (12); HRMS calcd for C₂₆H₂₂N₂O₆ 458.1478, found 458.1466. Anal. calcd for C₂₆H₂₂N₂O₆: C, 68.11; H, 4.84; N, 6.11. Found: C, 68.19; H, 4.81; N, 6.16.

10-Methyl-2,6,7,13-tetramethoxydibenzo[*a,c*]phenanthridine (1d). 61%. Beige powder: mp 118-119°C (MeOH); R_f 0.32 (CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 3.03 (3H, s), 3.92 (3H, s), 3.94 (3H, s), 4.11 (3H, s), 4.18 (3H, s), 7.13-7.23 (2H, m), 7.78 (1H, s), 8.07 (1H, d, J 9.1 Hz), 8.24 (1H, d, J 1.9 Hz), 8.26 (1H, d, J 2.4 Hz), 8.39 (1H, d, J 9.1 Hz), 8.75 (1H, s); ¹³C NMR (63 MHz, CDCl₃) δ 22.9, 55.3, 55.7, 55.8, 102.6, 105.7, 106.4, 109.9, 114.9, 116.4, 116.5, 121.8, 124.0, 124.3, 124.6, 125.2, 127.8, 129.4, 134.6, 140.9, 148.6, 150.5, 156.4, 157.1, 160.4; FTIR (neat film, cm⁻¹): 1614, 1242, 1049; EIMS (m/z, %) 413 (M⁺, 100), 398 (10), 370 (10); HRMS calcd for C₂₆H₂₃NO₄ 413.1627, found 413.1621. Anal. calcd for C₂₆H₂₃NO₄: C, 75.53; H, 5.61; N, 3.39. Found: C, 75.61; H, 5.58; N, 3.27.

10-Methyl-2,6,7,11-tetramethoxydibenzo[*a,c*]phenanthridine (1e). 80%. Beige powder: mp 140-140.5°C (MeOH); R_f 0.57 (CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 3.28 (3H, s), 3.96 (3H, s), 4.02 (3H, s), 4.13 (3H, s), 4.18 (3H, s), 6.96 (1H, d, J 7.9 Hz), 7.26 (1H, dd, J 9.1, 2.4 Hz), 7.59 (1H, dd, J 8.3, 7.9 Hz), 7.85 (1H, s), 8.29 (1H, d, J 2.4 Hz), 8.46 (1H, d, J 9.1 Hz), 8.50 (1H, d, J 8.7 Hz), 8.75 (1H, s); ¹³C NMR (63 MHz, CDCl₃) δ 29.8, 55.5, 55.9, 56.0, 102.8, 105.7, 105.8, 111.3, 114.9, 116.3, 118.7, 119.0, 124.1, 124.3, 124.4, 125.4, 129.5, 129.8, 135.7, 140.2, 148.8, 150.1, 156.7, 157.2, 158.5;

FTIR (neat film, cm^{-1}): 1615, 1260, 1043; EIMS (m/z , %) 413 (M^+ , 100); HRMS calcd for $\text{C}_{26}\text{H}_{23}\text{NO}_4$ 413.1627, found 413.1619. Anal. calcd for $\text{C}_{26}\text{H}_{23}\text{NO}_4$: C, 75.53; H, 5.61; N, 3.39. Found: C, 75.47; H, 5.68; N, 3.33.

3,12-Difluoro-6,7-dimethoxy-10-methyldibenzo[*a,c*]phenanthridine (1f). 75%. Yellow powder: mp 228-229°C (MeOH); R_f 0.56 (30% EtOAc/ CH_2Cl_2); ^1H NMR (250 MHz, CDCl_3) δ 3.07 (3H, s), 4.13 (3H, s), 4.18 (3H, s), 7.27 (1H, ddd, J 9.5, 7.9, 2.4 Hz), 7.49 (1H, ddd, J 9.1, 7.9, 2.8 Hz), 7.72 (1H, s), 7.84 (1H, dd, J 9.9, 2.8 Hz), 8.10 (1H, dd, J 11.1, 2.4 Hz), 8.60 (1H, dd, J 9.1, 5.9 Hz), 8.69 (1H, dd, J 9.5, 5.6 Hz), 8.72 (1H, s); ^{13}C NMR (63 MHz, CDCl_3) δ 23.1, 55.9, 56.0, 103.2, 105.6, 108.2, 110.0 (d, $J_{\text{C-F}}$ 21.5 Hz), 113.7 (d, $J_{\text{C-F}}$ 23.3 Hz), 116.8 (d, $J_{\text{C-F}}$ 3.6 Hz), 119.1 (d, $J_{\text{C-F}}$ 23.3 Hz), 124.2, 125.9, 127.8 (d, $J_{\text{C-F}}$ 7.9 Hz), 129.3, 129.4, 130.5 (d, $J_{\text{C-F}}$ 7.2 Hz), 132.1 (d, $J_{\text{C-F}}$ 7.2 Hz), 139.0 (d, $J_{\text{C-F}}$ 3.4 Hz), 150.0, 150.1, 155.9, 156.0, 160.1 (d, $J_{\text{C-F}}$ 247.8 Hz), 161.3 (d, $J_{\text{C-F}}$ 245.9 Hz); FTIR (neat film, cm^{-1}): 1616, 1262, 1022; EIMS (m/z , %) 389 (M^+ , 100), 374 (13), 346 (24), 303 (24), 194 (13), 187 (19), 152 (16); HRMS calcd for $\text{C}_{24}\text{H}_{17}\text{F}_2\text{NO}_2$ 389.1227, found 389.1225. Anal. calcd for $\text{C}_{24}\text{H}_{17}\text{F}_2\text{NO}_2$: C, 74.03; H, 4.40; F, 9.76; N, 3.60. Found: C, 74.11; H, 4.44; F, 9.71; N, 3.62.

6,7-Dimethoxy-10-methyldibenzo[*a,c*]phenanthridine (1g). 72%. Beige powder: mp 145-146°C (MeOH); R_f 0.39 (40% hexane/EtOAc); ^1H NMR (250 MHz, CDCl_3) δ 3.15 (3H, s), 4.15 (3H, s), 4.20 (3H, s), 7.56-7.69 (3H, m), 7.77 (1H, ddd, J 8.3, 7.1, 1.2 Hz), 7.96 (1H, s), 8.27 (1H, dd, J 8.3, 1.6 Hz), 8.57 (1H, dd, J 7.9, 1.6 Hz), 8.82-8.85 (2H, m), 8.90 (1H, d, J 8.3 Hz); ^{13}C NMR (63 MHz, CDCl_3) δ 23.1, 55.9, 56.0, 103.1, 105.6, 117.2, 122.8, 125.1, 125.2, 125.7, 125.8, 126.0, 126.2, 126.7, 127.1, 128.0, 128.7, 129.6, 130.0, 132.7, 139.8, 149.5, 150.0, 157.1; FTIR (neat film, cm^{-1}): 1616, 1267, 1026; EIMS (m/z , %) 353 (M^+ , 100), 310 (17); HRMS calcd for $\text{C}_{24}\text{H}_{19}\text{NO}_2$ 353.1416, found 353.1406. Anal. calcd for $\text{C}_{24}\text{H}_{19}\text{NO}_2$: C, 81.56; H, 5.42; N, 3.96. Found: C, 81.62; H, 5.47; N, 3.92.

2,3,6,7,12,13-Hexamethoxy-10-methyldibenzo[*a,c*]phenanthridine (1h). 72%. Yellow powder: mp 232-233°C (MeOH); R_f 0.37 (10% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 3.11 (3H, s), 4.07 (3H, s), 4.09 (3H, s), 4.11 (3H, s), 4.16 (3H, s), 4.17 (3H, s), 4.19 (3H, s), 7.52 (1H, s), 7.80 (1H, s), 7.91 (1H, s), 8.33 (1H, s), 8.34 (1H, s), 8.83 (1H, s); ¹³C NMR (63 MHz, CDCl₃) δ 23.3, 56.0, 102.9, 104.4, 105.2, 105.6, 106.3, 109.3, 116.9, 122.4, 122.6, 124.4, 124.9, 125.3, 128.7, 138.9, 147.8, 148.4, 148.5, 149.0, 149.8, 151.7, 154.2; FTIR (neat film, cm⁻¹): 1627, 1252, 1068; EIMS (m/z, %) 473 (M⁺, 100), 430 (12), 237 (21), 229 (11); HRMS calcd for C₂₈H₂₇NO₆ 473.1838, found 473.1831. Anal. calcd for C₂₈H₂₇NO₆: C, 71.02; H, 5.75; N, 2.96. Found: C, 71.11; H, 5.73; N, 2.91.

6,10-Dimethyl-2,3,12,13-tetramethoxydibenzo[*a,c*]phenanthridine (1i). 75%. Yellow powder: mp 223-224°C (MeOH); R_f 0.42 (5% EtOAc/CH₂Cl₂); ¹H NMR (250 MHz, CDCl₃) δ 2.66 (3H, s), 3.10 (3H, s), 4.07 (3H, s), 4.09 (3H, s), 4.12 (3H, s), 4.18 (3H, s), 7.52 (1H, s), 8.09 (1H, s), 8.26 (1H, s), 8.31 (1H, d, *J* 2.4 Hz), 8.33 (1H, s), 9.22 (1H, dd, *J* 8.4, 2.4 Hz), 9.25 (1H, d, *J* 8.4 Hz); ¹³C NMR (63 MHz, CDCl₃) δ 21.1, 23.2, 55.9, 56.0, 104.6, 105.2, 106.4, 109.3, 116.8, 122.4, 122.7, 123.4, 125.5, 126.6, 128.2, 128.9, 129.2, 132.3, 137.2, 139.8, 147.8, 148.4, 148.5, 149.0, 156.1; FTIR (neat film, cm⁻¹): 1616, 1242, 1054; EIMS (m/z, %) 427 (M⁺, 100), 384 (13), 368 (21); HRMS calcd for C₂₇H₂₅NO₄ 427.1784, found 427.1777. Anal. calcd for C₂₇H₂₅NO₄: C, 75.86; H, 5.89; N, 3.28. Found: C, 75.81; H, 5.83; N, 3.30.

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