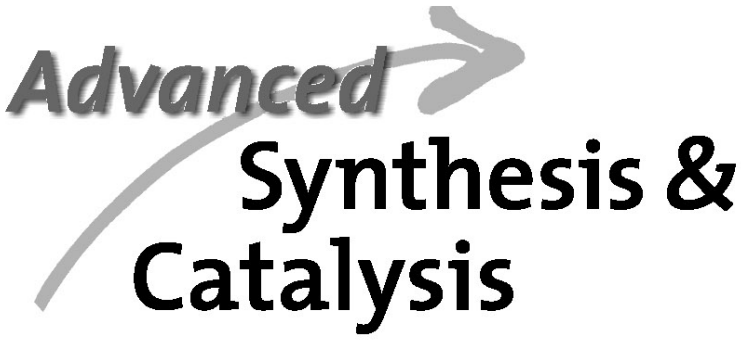

Supporting Information

Supporting Information

Palladium-Catalyzed *N*-Arylation of *N,N*-Dialkylhydrazines with Aryl Chlorides

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Melting points were determined with a Büchi B-545 apparatus and are uncorrected. All of the reagents, catalysts, and solvents are commercially available and were used as purchased, without further purification. Reaction products were purified on axially compressed columns, packed with SiO_2 25-40 μm (Macherey Nagel), connected to a Jasco RI-031 Plus solvent delivery system and to a Jasco PU-2087 Plus refractive index detector, and eluting with *n*-hexane/ethyl acetate mixtures. ^{1}H NMR (400 MHz), ^{13}C NMR (100.6 MHz) and ^{19}F NMR (376.5 MHz) spectra were recorded with a Bruker Avance 400 spectrometer. Splitting patterns are designed as s (singlet), d (doublet), t (triplet), q (quartet), qp (quintuplet), m (multiplet), or bs (broad singlet). IR spectra were recorded with a Jasco FT/IR-430 spectrometer. MS spectra were recorded with a Shimadzu GCMS-QP2010S.

Typical procedure for the synthesis of *N,N*-dialkyl-*N'*-arylhydrazines (3) from *N,N*-dialkylhydrazines (1) and aryl chlorides (2) - Preparation of (3a): In a Carousel Tube Reactor (Radley Discovery), $Pd_2(dba)_3$ (0.0129 g, 0.014 mmol) and Xphos (0.0134, 0,028 mmol) were stirred at room temperature under argon in 1.0 mL of dioxane for ten minutes. Then 4-chloroanisole (0.080 g, 0.563 mmol), *N,N*-dimethylhydrazine (0.086 mL, 1.126 mmol), 1 mL of dioxane and NaO-*t*-Bu (0.0757 g, 0.788 mmol) were added. The reaction mixture was stirred at 120 °C under argon for 6 h. After cooling, the reaction mixture was diluted with ethyl acetate, washed with $NaHCO_3$, dried over Na_2SO_4 and concentrated under reduced pressure. The residue was purified by chromatography (silica gel, 35 g; *n*-hexane/ethylacetate 80/20 v/v) to give 0.083 g (71%) of **3a**; IR (neat) 3218, 2978 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 6.89-6.80 (m, 4H), 4.01 (bs, 1H), 3.78 (s, 3H), 2.53 (s, 6H); ^{13}C NMR ($CDCl_3$) δ 153.6, 141.7, 115.5, 114.7, 55.8, 47.9; MS calculated for $C_9H_{14}N_2O$ = 166.22, MS (*m/z*) (relative intensity) 166 (M^+, 35), 122 (100), 151 (14), 42 (57).

3b: oil; IR (neat) 3409, 2950, 1508 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 7.12 (t, J = 8.0 Hz, 1H), 6.54 (t, J = 2.0 Hz, 1H), 6.46 (dd, J_1 = 8.0 Hz, J_2 = 1.1 Hz, 1H), 6.38 (dd, J_1 = 8.0 Hz, J_2 = 1.8 Hz, 1H), 4.26 (bs, 1H), 3.81 (s, 3H), 2.55 (s, 6H); ^{13}C NMR ($CDCl_3$) δ 161.0, 149.4, 130.0, 106.3, 105.0, 99.4, 55.2, 47.8. MS calculated for $C_9H_{14}N_2O$ = 166.22; MS (*m/z*) (relative intensity) 166 (M^+, 83), 122 (57), 107 (37), 95 (69), 42 (100).

3c: oil; IR (neat) 3254, 2949 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.26-7.18 (m, 2H), 7.08(d, *J* = 7.3 Hz, 1H), 6.79 (t, *J* = 7.1 Hz, 1H), 4.14 (bs, 1H), 2.60 (s, 6H), 2.17 (s, 3H); ^{13}C NMR ($CDCl_3$) δ 145.4, 130.2, 127.2, 121.6, 118.9, 113.1, 48.1, 17.4. MS calculated for $C_9H_{14}N_2$ = 150.22; MS (*m/z*) (relative intensity) 150 (M^+, 36), 106 (100), 77 (45), 42 (83).

3d: wax; IR (neat) 3280, 2980 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.31 (dd, J_1 = 6.7 Hz, J_2 = 1.9 Hz, 2H), 6.8 (dd, J_1 = 6.7 Hz, J_2 = 1.9 Hz, 2H), 4.17 (bs, 1H), 4.04-3.97 (m, 2H), 3.82-3.75 (m, 2H), 2.54 (s, 6H), 1.65 (s, 3H); ^{13}C NMR ($CDCl_3$) δ 147.4, 134.1, 126.3, 113.1, 109.0, 64.4, 47.8, 27.6; MS calculated for $C_{12}H_{18}N_2O_2$ = 222.28, MS (*m/z*) (relative intensity) 222 (M^+, 36), 207 (88), 163 (43), 59 (45), 43 (100).

3e: oil; IR (neat) 3441, 2862, 1602 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.24 (t, *J* = 7.9 Hz, 1H), 6.91 (d, *J* = 7.9 Hz, 1H), 6.82 (t, *J* = 7.8 Hz, 1H), 4.16 (bs, 1H), 2.57 (s, 6H); ^{13}C NMR ($CDCl_3$) δ 147.8, 129.2, 119.5, 113.7, 47.9; MS calculated for $C_8H_{12}N_2$ = 136.19, MS (*m/z*) (relative intensity) 136 (M^+, 62), 121(40), 92 (86), 42 (100).

3f: wax; IR (neat) 3398, 2972 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.91-7.86 (m, 2H), 7.52-7.31 (m, 5H), 4.86 (s, 1H), 2.70 (s, 6H); ^{13}C NMR ($CDCl_3$) δ 142.3, 134.4, 128.7, 126.8, 125.7, 124.8, 123.1, 120.2, 119.4, 108.2, 47.9. MS calculated for $C_{12}H_{14}N_2$ = 186.25; MS (*m/z*) (relative intensity) 186 (M^+, 50), 142 (44), 115 (92), 42 (100).

3g: wax; IR (KBr) 3249, 2949 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.29 (t, *J* = 7.8 Hz, 1 H), 7.17 (s, 1H), 7.05-6.98 (m, 2H), 4.42 (bs, 1H), 2.55 (s, 6H); ^{13}C NMR ($CDCl_3$) δ 148.2, 131.6 (q, *J* = 31.9 Hz), 129.6, 124.4 (q, *J* = 272.2 Hz), 116.4, 115.7 (q, *J* = 3.9 Hz), 109.9 (q, J = 3.9 Hz), 47.7; ^{19}F NMR ($CDCl_3$) δ -62.7; MS calculated for $C_9H_{11}N_2F_3$ = 204.09, MS (*m/z*) (relative intensity) 204 (M^+, 68), 189(42), 160 (57), 42 (100).

3h: mp: 65-69 °C; IR (KBr) 3156, 2847 cm^{-1}; ^1H NMR ($CDCl_3$) δ 8.19 (d, *J* = 2.3 Hz, 1H), 8.04 (dd, J_1 = 4.6 Hz, J_2 = 1.4 Hz, 1H), 7.21-7.18 (m, 1H), 7.13-7.08 (m, 1H), 4.31 (bs, 1H), 2.53 (s, 6H); ^{13}C NMR ($CDCl_3$) δ 143.8, 140.9, 136.4, 123.8, 120.3, 47.8. MS calculated for $C_7H_{11}N_3$ = 137.18; MS (*m/z*) (relative intensity) 137(M^+, 100), 42 (72), 93 (68), 122 (47).

3i: mp: 68-69 °C; IR (KBr) 3174, 2941, 2804 cm^{-1}; ^1H NMR ($CDCl_3$) δ 6.88-6.82 (m, 2H), 6.80-6.75 (m, 2H), 4.14 (bs, 1H), 3.75 (s, 3H), 2.72-2.51 (m, 8H), 2.32 (s, 3H); ^{13}C NMR ($CDCl_3$) δ 153.5, 141.1, 115.2, 114.5, 55.6, 55.5, 55.0, 45.6; MS calculated for $C_{12}H_{19}N_3O$ = 221.29, MS (*m/z*) (relative intensity) 221 (M^+, 21), 99 (49), 56 (95), 42 (100).

3j: mp: 117-118 °C; IR (KBr) 3206, 2995 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.29 (t, *J* = 8.6 Hz, 2H), 6.8 (d, *J* = 8.6 Hz, 2H), 4.41 (bs, 1H), 4.41- 3.98 (m, 2H), 3.81-3.75 (m, 2H), 2.98- 2.51 (m, 8H), 2.33 (s, 3H), 1.64 (s, 3H); ^{13}C NMR ($CDCl_3$) δ 147.2, 134.2, 126.3, 113.2, 109.0, 64.4, 55.8, 55.2, 45.9, 27.7; MS calculated for $C_{15}H_{23}N_3O_2$ = 277.36, MS (*m/z*) (relative intensity) 277 (M^+, 26), 86 (100), 56 (100).

3k: mp: 150-151 °C; IR (KBr) 3169, 3028, 2946 cm^{-1}; ^1H NMR (DMSO d_6) δ 7.42-7.26 (m, 5H), 7.05 (t, *J* = 7.8 Hz, 1H), 7.05 (s, 1H), 6.70 (t, *J* = 8.0 Hz, 2H),6,47 (s, 1H), 3.95 (s, 4H), 3.31 (bs, 1H), 2.69-2.38 (m, 7H), 2.18 (s, 3H); ^{13}C NMR (DMSO d_6) δ 148.5, 143.4, 143.3, 129.1, 128.5, 128.3, 126.2, 115.6, 112.4, 110.7, 109.3, 64.9, 55.3, 54.8, 46.0; MS calculated for $C_{15}H_{23}N_3O_2$ = 339.43, MS (*m/z*) (relative intensity) 339 (M^+, 26), 99 (100), 56 (76).

3l: mp: 121-122°C; IR (KBr), 3209, 3099 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.21-7.17 (m, 2H), 6.90-6.89 (m, 2H), 6.81-6.77 (m, 1H), 4.39 (bs, 1H), 2.88-2.52 (m, 8H), 2.36 (s, 3H); ^{13}C NMR ($CDCl_3$) δ 147.2, 129.0, 119.4, 113.5, 55.3, 55.0, 45.5; MS calculated for $C_{11}H_{17}N_3$ = 191.27 MS (*m/z*) (relative intensity) 191 (M^+, 16), 99 (47), 56 (69), 42 (100).

3m: mp: 78-79°C; IR (KBr) 3410, 2939, 2799 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.00 (d, *J* = 7.4 Hz, 2H), 6.82 (t, J_1 = 7.4 Hz, 1H), 4.20 (bs, 1H), 2.99-2.22 (m, 17H); ^{13}C NMR ($CDCl_3$) δ 143.1, 129.1, 128.8, 122.3, 56.0, 55.3, 45.7, 19.0; MS calculated for $C_{13}H_{21}N_3$ = 219.32, MS (*m/z*) (relative intensity) 219 (M^+, 43), 99(100), 56 (99), 42 (74).

3n: oil; IR (neat) 3304, 2929, cm^{-1}; ^1H NMR ($CDCl_3$) δ 6.81-6.72 (m, 4H), 4.05 (bs, 1H), 3.75 (s, 3H), 2.33 (bs, 2H), 1.68-1.58 (m, 3H), 1.41-1.27 (m, 3H), 1.05-1.01 (m, 6H); ^{13}CNMR ($CDCl_3$)

δ 151.8, 145.6, 114.3, 112.7, 63.1, 55.6, 34.8, 24.0, 20.9; MS calculated for $C_{14}H_{22}N_2O$ = 234.34, MS (*m/z*) (relative intensity) 234 (M^+, 37), 122 (100).

3o: oil; IR (neat) 3428, 3268 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.27 (d, J_1 = 8.7 Hz, 1H), 7.12 (t, J = 7.5, 1H), 7.02 (d, J = 7.5 Hz ,1H), 6.50 (t, J_1 = 7.3 Hz, 1H), 4.11 (bs, 1H), 2.40 (bs, 1H), 2.16 (s, 3H), 1,80-1,54 (m, 3H);1.52-1.30 (m, 3H), 1.10-0.92 (m, 6H); ^{13}CNMR ($CDCl_3$) δ 148.7, 129.8, 126.7, 118.0, 116.7, 112.8, 63.6, 35.1, 24.3, 20.8, 17.3; MS calculated for $C_{14}H_{22}N_2$ = 218.34, MS (*m/z*) (relative intensity) 218 (M^+, 59), 203 (32), 106 (100), 98 (42).

3p: oil; IR (neat) 3308, 2929 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.18-7.12 (m, 2H), 6.85 (d, J = 7.7 Hz, 2H), 6.67 (dt, J_1 =7.3 Hz, J_2 =1.2 Hz, 1H), 4.17 (bs, 1H), 2.33 (bs, 1 H), 1.75-1.64 (m, 2H); 1.45-1.39 (m, 3H), 1.07 (s, 6H); ^{13}C NMR ($CDCl_3$) δ 151.2, 128.7, 117.1, 111.7, 63.2, 34.9, 24.0, 21.0; MS calculated for $C_{13}H_{20}N_2$ = 204.31 MS (*m/z*) (relative intensity) 204 (M^+, 80), 189 (87), 66 (60), 92 (100).

3q: oil; IR (neat) 3442, 2969 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.21 (t, J =7.9 Hz , 1H), 7.10-6.95 (m, 2H), 6.90 (d, J = 7.5, 1H), 4.36 (bs, 1H), 2.36-2.33 (m, 1H), 1.75-1.62 (m, 2H); 1.45-1.25 (m, 4H), 1.05-1.03 (m, 6H); ^{13}C NMR (100.6 MHz) ($CDCl_3$) δ 152.8, 130.0 (q, J = 30.8 Hz), 129.9, 129.3, . 125.2 (q, J = 272.3 Hz), 114.8 (q, J = 3.8 Hz), 110.0 (q, J = 3.8 Hz); ^{19}F NMR ($CDCl_3$) δ -62.7; MS calculated for $C_{14}H_{19}N_2F_3$ = 272.31, MS (*m/z*) (relative intensity) 272 (M^+, 58), 257 (100), 98 (30), 55 (44).

3r: 209-211 °C; IR (neat) 3307, 2929 cm^{-1}; ^1H NMR ($CDCl_3$) δ 6.89 (d, J = 7.4 Hz, 2H), 6.57 (t, J_1 = 7.4 Hz, 1H), 4.06 (bs, 1H), 2.37-2.25 (m, 8H), 1.70-1.62 (m, 3H), 1.45-1.35 (m, 4H), 1.09-1.07 (m, 6H); ^{13}C NMR ($CDCl_3$) δ 146.0, 129.8, 122.9, 120.2, 65.0, 34.6, 24.3, 21.0; MS calculated for $C_{15}H_{24}N_2$ = 232.36, MS (*m/z*) (relative intensity) 232 (M^+, 27), 120 (100), 98 (53), 42 (54).

3s: oil; IR (neat) 3389, 2935, 2853, 1035 cm^{-1}; ^1H NMR ($CDCl_3$) δ 6.90-6.75 (m, 4H), 4.14 (bs, 1H), 3.76 (s, 3H), 2.62 (bs, 4H), 1.70 (bs, 4H), 1.45 (bs, 2H); ^{13}C NMR ($CDCl_3$) δ 153.4, 141.7, 115.2, 114.7, 57.5, 55.8, 26.1, 23.8; MS calculated for $C_{12}H_{18}N_2O$ = 206.28; MS (*m/z*) (relative intensity) 206 (M^+, 45), 122 (100).

3t: oil; IR (neat) 3255, 2936, 1038 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.11 (t, J = 8.0 Hz, 1H), 6.58 (t, J = 2.0 Hz, 1H), 6.49-6.45 (m, 1H), 6.38-6.34 (m, 1H), 4.41 (bs, 1H), 3.81 (s, 3H), 2.62 (bs, 4H), 1.70 (bs, 4H), 1.49 (bs, 2H);^{13}C NMR ($CDCl_3$) δ 160.9, 149.4, 129.9, 106.3, 104.4, 99.4, 57.4, 55.1, 26.1, 23.7; MS calculated for $C_{12}H_{18}N_2O$ = 206.28, MS (*m/z*) (relative intensity) 206 (100), 123 (75), 84 (56), 41 (65).

3u: oil; IR (neat) 3438, 2935, 1036 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.29-7.18 (m, 2H), 7.08 (d, J = 7.3 Hz, 1H), 6.79 (dt, J_1 = 7.3 Hz, J_2=1.3, 1H), 4.30 (bs, 1H), 2.72 (bs, 4H), 2.18 (s, 3H), 1.75 (qp, J = 5.7 Hz, 4H) 1.49 (bs, 2H); ^{13}C NMR ($CDCl_3$) δ 145.4, 130.2, 127.1, 121.5, 118.5, 112.9, 57.6, 26.2, 23.9, 17.4; MS calculated for $C_{12}H_{18}N_2$ = 190.28, MS (*m/z*) (relative intensity) 190 (M^+, 53), 106 (100), 84 (83), 77 (30).

3v: mp: 123-125 °C; IR (KBr) 3248, 2943, 1028 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.31 (t, J = 8.6 Hz, 2H),. 6.86 (d, J = 8.5 Hz, 2H), 4.41 (bs, 1H), 4.05-3.97 (m, 2H), 3.82-3.78 (m, 2H), 2,69 (bs, 4H), 1.73-1.60 (m, 7H), 1.44 (bs, 2H); ^{13}C NMR ($CDCl_3$) δ 147.5, 133.8, 126.2, 113.0, 109.1, 64.4, 57.5, 27.7, 26.1, 23.8; MS calculated for $C_{15}H_{22}N_2O_2$ = 262.35, MS (*m/z*) (relative intensity) 262 (M^+, 31), 247 (58), 207 (50), 44 (100).

3w: oil; IR (neat) 3250, 2936, 1036 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.25-7.21 (m, 2H), 6.92 (d, J = 7.7 Hz, 2H). 6.81 (t, J = 7.3 Hz, 1H), 4.39 (bs, 1H), 2,69 (bs, 4H), 1.75-1.70 (qp, J = 5.7 Hz, 4H), 1.47 (bs, 2H); ^{13}C NMR ($CDCl_3$) δ 147.9, 129.2, 119.2, 113.6, 57.5, 26.1, 23.8; MS calculated for $C_{11}H_{16}N_2$ = 176.26, MS (*m/z*) (relative intensity) 176 (M^+, 100), 93 (90), 77 (70), 41 (95).

3y: oil; IR (neat) 3335, 2935, 2853 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.85-7.82 (m, 2H), 7.48-7.29 (m, 5H), 5.04 (bs, 1H), 2.88-2.60 (m, 4H), 1.81-1.76 (m, 4H), 1.58-1.50 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 142.3, 134.4, 128.7, 126.8, 125.5, 124.7, 122.9, 120.1, 119.0, 107.7, 57.5, 26.2, 23.9; MS calculated for $C_{15}H_{18}N_2$ = 226.32, MS (*m/z*) (relative intensity) 226 (M^+, 62), 143(87), 115 (77), 44 (100).

3z: oil; IR (neat) 3270, 2940, 1614 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.28 (t, *J* = 8.0 Hz, 1H), 7.16 (s, 1H), 7.05-6.99 (m, 2H), 4.56 (bs, 1H), 2.67 (bs, 4H), 1.73 (qp, *J* = 5.7 Hz, 4H), 1.59-1.40 (m, 2H); ^{13}C NMR ($CDCl_3$) δ 148.2, 131.5 (q, *J* = 31.8 Hz), 129.5, 124.4 (q, *J* = 272.3 Hz), 116.3, 115.4 (q, *J* = 4.0 Hz), 109.8 (q, *J* = 4.0 Hz), 57.4, 26.1, 23.6; ^{19}F NMR ($CDCl_3$) δ -62.7; MS calculated for $C_{12}H_{15}N_2F_3$ = 244.26, MS (*m/z*) (relative intensity) 244 (M^+, 100), 161 (25), 145 (34), 42 (63).
3za: mp: 90-92 °C; IR (KBr) 3093, 2938, 1036 cm^{-1}; ^1H NMR ($CDCl_3$) δ 8.19 (d, *J* = 2.4 Hz, 1H), 8.04 (dd, J_1 = 4.1 Hz, J_2 = 1.2 Hz, 1H), 7.29-7.19 (m, 1H), 7.12-7.07 (m, 1H), 4.49 (bs, 1H), 2,66 (bs, 4H), 1.73-1.60 (m, 4H), 1.44 (bs, 2H); ^{13}C NMR ($CDCl_3$) δ 143.9, 140.6, 136.4, 123.8, 120.2, 57.4, 26.0, 23.6; MS calculated for $C_{10}H_{15}N_3$ = 177.25, MS (*m/z*) (relative intensity) 177 (M^+, 100), 84 (52), 55 (70), 42 (86).
3zb: oil; IR (neat) 3437, 2928, 1508 cm^{-1}; ^1H NMR ($CDCl_3$) δ 6.90-6.79 (m, 4H), 4.57 (bs, 1H), 3.78 (s, 3H), 2.93-2.88 (m, 4H), 1.73-1.67 (m, 8H); ^{13}C NMR ($CDCl_3$) δ 153.4, 142.2, 115.1, 114.7, 58.8, 55.8, 26.8, 26.7; MS calculated for $C_{13}H_{20}N_2O$ = 220.31, MS (*m/z*) (relative intensity) 220 (M^+, 46), 122 (100), 108 (35), 41 (59).
3zc: oil; IR (neat) 3428, 3268, 2927 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.28-7.25 (m, 1H), 7.17 (t, *J* = 7.7, 1H), 7.04 (d, *J* = 7.0 Hz ,1H), 6.73 (dt, J_1 = 7.3 Hz, J_2 = 1.2 Hz, 1H), 4.70 (bs, 1H), 2.97-2.91 (m, 4H), 2.14 (s, 3H), 1,76-1,67 (m, 8H); $^{13}CNMR$ ($CDCl_3$) δ 145.7, 130.1, 127.1, 121.2, 118.3, 112.9, 58.9, 26.9, 26.8, 17.3; MS calculated for $C_{13}H_{20}N_2$ = 204.31, MS (*m/z*) (relative intensity) 204 (M^+, 64), 106 (100), 98 (47), 77 (32), 42 (66).
3zd: oil; IR (neat) 3447, 3260, 2927 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.57-7.53 (m, 2H), 7.35-7.28 (m, 3H), 7.17 (t, *J* = 7.8 Hz, 4H), 7.01 (s, 1H), 7.00-6.91 (m, 2H), 4.82 (bs, 1H), 4.12-4.05 (m, 4H), 2.96-2.87 (m 4H), 1.35-1.26 (m, 8H); ^{13}C NMR ($CDCl_3$) δ 148.1, 143.0, 142.4, 129.1, 128.1, 128.0, 126.2, 117.0, 113.1, 111.4, 109.6, 64.9, 58.6, 26.9, 26.8; MS calculated for $C_{21}H_{26}N_2O_2$ = 338.44, MS (*m/z*) (relative intensity) 338 (M^+, 10), 207 (100), 149 (27), 77 (23).
3ze: oil; IR (neat) 3335, 2927 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.23-7.18 (m, 2H), 6.94-6.89 (m, 2H), 6.84 (tt, J_1 = 7.3 Hz, J_2 = 1.1 Hz, 1H), 4.79 (bs, 1H), 2.95-2.92 (m, 4H), 1.75-1.66 (m, 8H); ^{13}C NMR ($CDCl_3$) δ 148.3, 129.2, 119.1, 113.5, 58.8, 26.9, 26.8; MS calculated for $C_{12}H_{18}N_2$ = 190.28, MS (*m/z*) (relative intensity) 190 (M^+, 74), 93 (58), 77 (56), 42 (100).
3zf: oil; IR (neat) 3270, 2931, 2857 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.28 (t, *J* =7.9 Hz, 1H), 7.17 (t, *J* =7.9 Hz, 1H), 4.98 (bs, 1H), 2.95-2.90 (m, 4H), 1.76-1.67 (m, 8H); $^{13}CNMR$ ($CDCl_3$) δ 148.4, 131.4 (q, *J* = 31.7 Hz), 129.4, 124.3 (q, *J* = 272.3 Hz), 115.9, 115.1 (q, *J* = 3.9 Hz), 109.5 (q, *J* = 4.0 Hz), 58.6, 26.7, 26.5; ^{19}F NMR ($CDCl_3$) δ -62.7; MS calculated for $C_{13}H_{17}N_2F_3$ = 258.28, MS (*m/z*) (relative intensity) 258 (M^+, 63), 161 (15), 145 (20), 42 (100).
3zg: mp: 83-84 °C; lit. mp[1]: 83-84 °C
3zh: mp: 112-114 °C; IR (KBr) 3245, 2988, 2812cm^{-1}; ^1H NMR ($CDCl_3$), 7.11 (t, *J* = 8.1 Hz, 1H), 6.58-6.53 (m, 1H), 6.48 (dd, J_1 = 7.7 Hz, J_2 = 1.4 Hz, 1H), 6.37 (dd, J_1 = 7.8 Hz, J_2 = 2.4 Hz, 1H), 4.48 (bs, 1H), 3.85-3.78 (m, H), 2.75 (bs, 4H); ^{13}C NMR ($CDCl_3$) δ 160.7, 148.7, 129.8, 106.3, 104.7, 99.6, 67.0, 56.3, 55.0. MS calculated for $C_{12}H_{18}N_2O_2$ = 208.26, MS (*m/z*) (relative intensity) 208 (M^+, 84), 123 (44), 107 (67), 56 (100).
3zi: mp: 120-121 °C; IR (KBr) 3263, 3015, 2972, 1069 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.29-7.24 (m, 1H), 7.16 (t, J = 7.1 Hz, 1H), 7.08 (d, J = 7.0 Hz, 1H), 6.78 (t, J = 7.0 Hz, 1H), 4.35 (bs, 1H), 3.85 (t, *J* = 4.7 Hz, 4H), 2.79 (bs, 4H), 2.17 (s, 3H); ^{13}C NMR ($CDCl_3$) δ 144.7, 130.4, 127.1, 121.9, 119.2, 113.1, 67.2, 56.7, 17.3; MS calculated for $C_{11}H_{16}N_2O$ = 192.26, MS (*m/z*) (relative intensity) 192 (M^+, 31), 106 (100),77 (47), 56 (82).
3zl: mp: 121-123 °C; lit. mp[1]: 121-123 °C
3zm: mp: 70-71 °C; IR (KBr) 3252, 2964, 2859, 1064 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.30 (t, *J* =8.1 Hz, 1H), 7.18 (s, 1 H), 7.05 (d, *J* = 7.7, 2H), 4.64 (bs, 1H), 3.86-3.82 (m, 4H), 2.78 (bs, 4H); ^{13}C NMR ($CDCl_3$) δ 147.6, 131.7 (q, *J* = 31.8 Hz), 129.6, 124.3 (q, *J* =272.3), 116.5, 116.1 (q, *J* = 3.8 Hz), 110.0 (q, *J* = 3.8 Hz). 67.0, 56.4; ^{19}F NMR ($CDCl_3$) δ - 62.7; MS calculated for $C_{11}H_{13}N_2OF_3$ = 246.23, MS (*m/z*) (relative intensity) 246 (M^+, 83), 188 (28), 160 (51), 56 (100).

3zn: mp :130-131°C; IR (KBr) 3201 cm^{-1}; ^1H NMR ($CDCl_3$) δ 8.26 (d, J = 2.2 Hz, 1H), 8.06 (dd, J_1 = 4.6 Hz, J_2 = 1.4 Hz, 1H), 7.25-7.21 (m, 1H), 7.15-7.10 (m, 1H), 4.60 (bs, 1H), 3.80 (t, J =4.6 Hz, 4H), 2.77-2.70 (m, 4H); ^{13}C NMR(100.6 MHz) ($CDCl_3$) δ 143.2, 141.2, 136.5, 123.8, 120.4, 67.0, 56.4; MS calculated for $C_9H_{13}N_3O$ = 179.22, MS (*m/z*) (relative intensity) 179 (M^+, 100), 93 (66), 56 (66), 78 (60).

3zo: oil; IR (neat) 3272, 1024 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.01 (d, J = 7.5 Hz, 2H), 6.87 (t, J_1 = 7.5 Hz, 1H), 4.30 (bs, 1H), 3.74-3.72 (m, 4H), 2.78 (bs, 4H), 2.35 (m, 6H); ^{13}C NMR ($CDCl_3$) δ 142.7, 129.2, 128.8, 122.4, 67.1, 56.8, 18.9; MS calculated for $C_{12}H_{18}N_2O$ = 206.28, MS (*m/z*) (relative intensity) 206 (M^+, 27), 120 (100), 56 (43).

3zk: mp:106-107 °C; IR (KBr) 3241, 2956, 2850, 1068 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.29-7.21 (m, 2 H), 6.94 (d, J = 7.5 Hz, 2H), 6.83 (t, J =7.3 Hz, 1H), 4.46 (bs, 1H), 3.85-3.82 (m, 4H), 2.78 (bs, 4H); ^{13}C NMR ($CDCl_3$) δ 147.1, 129.3, 119.8, 113.8, 67.1, 56.5; MS calculated for $C_{10}H_{14}N_2O$ = 178.23; MS (*m/z*) (relative intensity) 178 (M^+, 55), 92 (80), 77 (54), 65 (100), 56 (80).

3zj: mp :134-135°C; IR (KBr) 3277 cm^{-1}; ^1H NMR ($CDCl_3$) δ 7.31 (d, J = 8.3 Hz, 2H), 6.90 (d, J = 8.3 Hz, 2H), 4.52 (bs, 1H), 4.21-4.01 (m, 2H), 3.83-3.78 (m, 6H), 2.75 (bs, 4H), 1.64 (bs, 4H); ^{13}C NMR ($CDCl_3$) δ 146.8, 134.5, 126.3, 113.2, 109.0, 67.1, 64.4, 56.5, 27.7; MS calculated for $C_{14}H_{20}N_2O_3$ = 264.32, MS (*m/z*) (relative intensity) 264 (M^+, 27), 249 (95), 56 (50), 43 (100).

[1] S. Cacchi, G. Fabrizi, A. Goggiamani, E. Licandro, S. Maiorana, D. Perdicchia *Org. Lett*. **2005**, *7*, 1497-1500