Fast and Efficient Synthesis of Pyrano[3,2-*c*]quinolines Catalyzed by Niobium(V) Chloride

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Abstract: A highly efficient two-step method for the synthesis of pyranoquinoline derivatives from imino-Diels–Alder reactions between aldimines and 3,4-dihydro-2*H*-pyran using niobium(V) chloride as catalyst under mild conditions is described.

Key words: Diels–Alder reactions, Lewis acids, catalysis, quinolines, Schiff bases

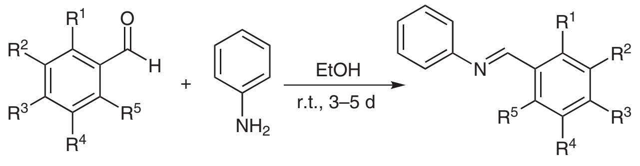
Pyranoquinoline derivatives are an important class of natural products that are present in many alkaloids.[1,2] They exhibit a wide spectrum of biological activity in various fields,[3–6] such as psychotropic, antiallergenic, anti-inflammatory, and estrogenic activity. In addition, these derivatives are also found to posses a vast range of pharmacological activities.[7] It is therefore not surprising that many synthetic methods have been developed for these compounds.[8–11] Among the various methods, the imino-Diels–Alder reaction between *N*-arylimines and nucleophilic alkenes is probably one of the most powerful synthetic tools for the construction of nitrogen-containing six-membered heterocyclic compounds.

Generally, Lewis acids[11–14] are employed to catalyze such reactions (there are also one-pot versions).[15–18] However, several Lewis acids are deactivated or decomposed by nitrogen-containing substrates.[19] Some of the Lewis acids are not easily available or are expensive, require longer reaction times, and form the products with poor yields. Therefore, developing simple and efficient synthetic methods for the preparation of this type of compound becomes more and more important.

Niobium(V) chloride, a low-cost and commercially available reagent, has been used by our group and other researchers as an effective catalyst for synthetic methodologies in a variety of reactions.[20–32] In the present report a highly efficient two-step method for the synthesis of pyranoquinolines using niobium(V) chloride as catalyst is described.

For the proposed studies, aldimines **3a–p** were prepared in good yields by treatment of the respective aromatic aldehydes **1a–p** with aniline (**2**) at room temperature (Table 1).

Table 1 Preparation of Aldimines **3a–p**



Starting aldehyde	R^1	R^2	R^3	R^4	R^5	Time (h)	Aldimine	Yield (%)
1a	H	H	H	H	H	72	**3a**	98
1b	H	H	Me	H	H	72	**3b**	95
1c	H	Me	H	H	H	72	**3c**	98
1d	Me	H	H	H	H	72	**3d**	96
1e	Me	H	Me	H	Me	72	**3e**	94
1f	H	NO_2	H	H	H	96	**3f**	92
1g	NO_2	H	H	H	H	96	**3g**	90
1h	H	OCH_2O		H	NO_2	120	**3h**	93
1i	H	OCH_2O		H	H	120	**3i**	94
1j	H	H	Cl	H	H	72	**3j**	96
1k	H	Cl	H	H	H	72	**3k**	97
1l	Cl	H	H	H	H	72	**3l**	96
1m	H	H	OMe	H	Me	96	**3m**	93
1n	H	OMe	H	H	H	96	**3n**	95
1o	OMe	H	H	H	H	96	**3o**	91
1p	H	OMe	OMe	OMe	H	120	**3p**	90

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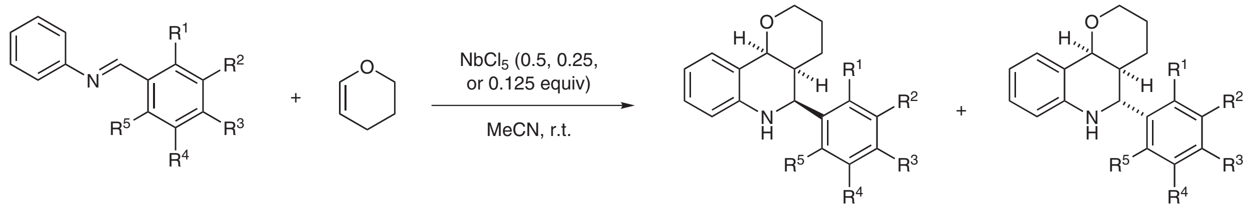

Scheme 1 Synthesis of pyranoquinoline derivatives from the imino-Diels–Alder reaction between aldimines **3a**–**p** and 3,4-dihydro-2*H*-pyran (**4**) catalyzed by niobium(V) chloride

Table 2 Niobium(V) Chloride Catalyzed Synthesis of Pyranoquinoline Derivatives **5a**–**p** (*endo*) and **6a**–**p** (*exo*)

Aldimine	$NbCl_5$ (equiv)	Time (min)	Yield (%)	Ratio **5a–p/6a–p** (*endo/exo*)
3a	0.500	1	92	44:56
	0.250	5	89	44:56
	0.125	15	90	33:67
3b	0.500	1	90	44:56
	0.250	5	90	39:41
	0.125	15	84	22:78
3c	0.500	1	85	31:69
	0.250	5	85	31:69
	0.125	15	81	22:78
3d	0.500	1	85	31:69
	0.250	10	87	28:72
	0.125	15	84	13:87
3e	0.500	30	80	0:100
	0.250	80	81	0:100
	0.125	150	75	0:100
3f	0.500	1	85	49:51
	0.250	5	82	48:52
	0.125	10	82	43:57
3g	0.500	5	79	50:50
	0.250	20	77	48:52
	0.125	50	74	30:70
3h	0.500	30	88	41:59
	0.250	100	85	41:59
	0.125	190	84	40:60
3i	0.500	15	75	46:54
	0.250	60	72	34:66
	0.125	150	73	18:82
3j	0.500	1	89	40:60
	0.250	5	90	42:58
	0.125	15	86	34:66
3k	0.500	1	87	42:58
	0.250	5	87	42:58
	0.125	15	85	35:65
3l	0.500	1	83	40:60
	0.250	5	84	44:56
	0.125	15	80	35:65
3m	0.500	5	87	40:60
	0.250	25	81	19:81
	0.125	75	78	0:100
3n	0.500	5	88	41:59
	0.250	35	86	37:63
	0.125	100	83	26:74
3o	0.500	10	70	36:64
	0.250	50	67	26:74
	0.125	180	68	23:77
3p	0.500	15	79	35:65
	0.250	50	80	20:80
	0.125	190	76	07:93

Imino-Diels–Alder reactions between aldimines **3a**–**p** and 3,4-dihydro-2*H*-pyran (**4**) in the presence of niobium(V) chloride in acetonitrile at room temperature afforded the corresponding pyranoquinoline derivatives **5a**–**p** and **6a**–**p** in high yields. In most of the cases, a mixture of *endo*-isomers **5** and *exo*-isomers **6** were obtained (Scheme 1, Table 2).

All products were isolated and characterized by spectroscopic and spectrometric methods (^{1}H NMR, ^{13}C NMR, IR, and mass spectra). The product ratios were determined by ^{1}H NMR analysis of the crude product. The relative stereochemistry of the Diels–Alder adducts was determined using the ^{1}H–^{1}H scalar coupling constant values between H1 and H2 ($J_{1,2}$) and comparison with literature data. Adducts **5a**–**p** (*endo*), show smaller coupling constants $J_{1,2}$ (5.5–5.7 Hz), typical for a gauche conformation. This is consistent with an orientation where the pyran ring and the phenyl group are on the same side (Figure 1). In adducts **6a**–**p** (*exo*), the coupling constants are significantly higher, $J_{1,2}$ between 8.1 and 11.1 Hz, indicative of the *anti* orientation of H1/H2, which is only possible when the pyran ring and the phenyl group are on opposite sides of the quinoline ring.

As can be observed from Table 2, in all cases the reactions proceed smoothly to give the pyranoquinoline derivatives, which could be separated by column chromatography. Substitution on the aromatic ring has a remarkable influence on reactivity: methylated **3e** or oxygenated **3p** aldimines, for example, are less reactive. The use of a lower molar ratio of niobium(V) chloride leads to an enhancement of diastereoselectivity, although the required reaction times are longer.

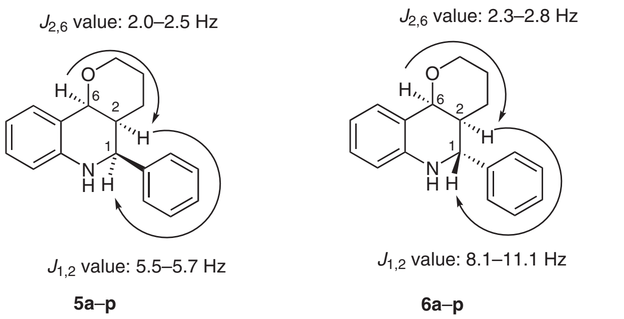

Figure 1 Coupling constant values used for determining stereochemistry

A remarkable aspect of this work is the higher efficiency of niobium(V) chloride, as compared to other Lewis acid catalysts. The strong activation of the aldimine system exerted by niobium(V) chloride is demonstrated by the shorter reaction times required. Table 3 shows some examples comparing our results for aldimine **3a** with literature data using other Lewis acids.

Table 3 Comparison of Imino Diels–Alder Reactions of Aldimine **3a** with 3,4-Dihydro-2*H*-pyran Catalyzed by Various Lewis Acids

Lewis acid	Time (min)	Yield (%)	Ratio *endo*/*exo*
$NbCl_5$	1	92	44:56
$InCl_3$[13]	30	80	41:59
$LiBF_4$[33]	120	88	15:85
K10/Fe^{3+} [34]	1020	86	42:58

In summary, this paper describes an efficient two-step method for the synthesis of pyranoquinoline derivatives from imino Diels–Alder reactions between aldimines and 3,4-dihydro-2*H*-pyran (**4**) using niobium(V) chloride as a catalyst. As compared to other Lewis acids (e.g., $InCl_3$),[13] niobium(V) chloride is more effective, requiring shorter reaction times, giving in most cases better yields and good diastereoselectivity, especially with lower molar concentrations of niobium(V) chloride. These notable features make this procedure an useful and attractive process for the synthesis of fused pyrano[3,2-*c*]quinolines of biological importance.

NMR spectra were measured using a Bruker DRX 400 instrument [400 MHz for ^{1}H NMR and 100 MHz for the ^{13}C NMR (proton decoupled)] using $CDCl_3$ as solvent and TMS as internal standard. IR spectra were measured with a Perkin Elmer Spectrum RX IFTIR System, and the most intense or representative bands are reported. EI-MS (70 eV) used a HP 5988-A spectrometer with fused silica capillary GC columns: HP-1 (25 m × 0.2 mm i.d. × 0.33 μm phase thickness), column temperature was 100 °C (3 min) ramped to 220 °C at 20 °C min^{-1}. Mass spectral analyses of new compounds were performed on a high-resolution q-TOF. ESI-MS were acquired on a UltrOTOF apparatus (Bruker Daltonics, Billerica, MA, USA), with flux 150 μL/h in MeCN, in positive mode. Column chromatography was performed with silica gel (60–230 mesh, Acros). Except where specified, all reagents were purchased from commercial sources and were used without further purification.

N-[(*E*)-Arylmethylene]anilines 3a–p; General Procedure[35]

Aniline (**2**, 2 mmol) and the appropriate aromatic aldehyde (2 mmol) were dissolved together in EtOH (10 mL) and left to stand overnight at r.t. The solvent was removed under vacuum, and the products were dissolved in EtOH (10 mL) and left to stand at r.t. for the time shown in Table 1. The yields were 90–98%.

N-[(*E*)-Phenylmethylene]aniline (3a)[36]

IR (film): 3028, 2863, 1700, 1627, 1484, 1191, 873, 767 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.44 (s, 1 H), 7.87–7.93 (m, 2 H), 7.44–7.49 (m, 3 H), 7.36–7.42 (m, 2 H), 7.19–7.20 (m, 3 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 160.4 (CH), 151.9 (C), 136.1 (C), 131.4 (CH), 129.1 (CH), 128.8 (CH), 128.7 (CH), 125.9 (CH), 120.9 (CH).

MS: *m*/*z* (%) = 181 (M)$^+$, 152, 149, 104, 89, 77, 63, 51.

N-[(*E*)-(4-Methylphenyl)methylene]aniline (3b)[36]

IR (film): 3039, 2869, 1623, 1484, 1173, 815, 753 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.40 (s, 1 H), 7.79 (d, *J* = 8.1 Hz, 2 H), 7.34–7.40 (m, 2 H), 7.26 (d, *J* = 8.1 Hz, 2 H), 7.18–7.23 (m, 3 H), 2.40 (s, 3 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 160.3 (CH), 152.2 (C), 141.8 (C), 133.7 (C), 129.5 (CH), 129.1 (CH), 128.8 (CH), 125.7 (CH), 120.9 (CH), 21.6 (CH_3).

MS: *m*/*z* = 195 (M)$^+$, 180, 152, 116, 104, 91, 77, 65, 51.

N-[(*E*)-(3-Methylphenyl)methylene]aniline (3c)[37]

IR (film): 3036, 2863, 1627, 1603, 1487, 1205, 1148, 786 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.36 (s, 1 H), 7.62 (s, 1 H), 7.63 (d, *J* = 7.5 Hz, 1 H), 7.33–7.39 (m, 2 H), 7.31 (t, *J* = 7.5 Hz, 1 H), 7.24 (d, *J* = 7.5 Hz, 1 H), 7.15–7.22 (m, 3 H), 2.37 (s, 3 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 160.6 (CH), 152.1 (C), 138.4 (C), 136.1 (C), 132.2 (CH), 129.1 (CH), 129.0 (CH), 128.6 (CH), 126.4 (CH), 125.8 (CH), 120.8 (CH), 21.2 (CH_3).

MS: *m*/*z* = 195 (M)$^+$, 180, 152, 116, 104, 91, 77, 65, 51

N-[(*E*)-(2-Methylphenyl)methylene]aniline (3d)[38]

IR (film): 3027, 2908, 1734, 1623, 1491, 1201, 763 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.74 (s, 1 H), 8.08 (dd, J_1 = 7.5 Hz, J_2 = 1.1 Hz, 1 H), 7.18–7.42 (m, 8 H), 2.58 (s, 3 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 159.0 (CH), 152.6 (C), 138.5 (C), 134.1 (C), 131.0 (CH), 129.1 (CH), 127.8 (CH), 126.3 (CH), 120.9 (CH), 19.3 (CH_3).

MS: *m*/*z* = 195 (M)$^+$, 178, 152, 115, 104, 91, 77, 65, 51

N-[(*E*)-Mesitylmethylene]aniline (3e)[39]

IR (film): 2953, 2917, 1588, 1450, 1194, 969, 853, 758 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.75 (s, 1 H), 7.35–7.41 (m, 2 H), 7.14–7.23 (m, 3 H), 6.90 (s, 2 H), 2.51 (s, 6 H), 2.29 (s, 3 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 160.8 (CH), 153.1 (C), 139.8 (C), 138.6 (C), 130.5 (C), 129.7 (CH), 129.1 (CH), 125.5 (CH), 120.7 (CH), 21.2 (CH_3), 21.0 (CH_3).

MS: *m*/*z* = 223 (M)$^+$, 206, 140, 131, 115, 103, 91, 77, 65, 51.

N-[(*E*)-(3-Nitrophenyl)methylene]aniline (3f)[39]

IR (film): 3075, 2880, 2355, 1528, 1352, 1191, 814, 764 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.62 (s, 1 H), 8.41 (s, 1 H), 8.20 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 8.14 (d, J = 8.1 Hz, 1 H), 7.54 (t, J = 8.1 Hz, 1 H), 7.33–7.40 (m, 2 H), 7.17–7.26 (m, 3 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 157.1 (CH), 150.7 (C), 148.5 (C), 137.7 (C), 134.1 (CH), 129.7 (CH), 129.2 (CH), 126.8 (CH), 125.4 (CH), 123.1 (CH), 120.9 (CH).

MS: m/z = 227 $(M)^+$, 179, 152, 104, 77, 63, 51.

N-[(E)-(2-Nitrophenyl)methylene]aniline (3g)[40]
IR (film): 3048, 2910, 2365, 1522, 1346, 1190, 858, 767 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.93 (s, 1 H), 8.30 (dd, J_1 = 7.6 Hz, J_2 = 1.3 Hz, 1 H), 8.06 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 7.72 (t, J = 7.6 Hz, 1 H), 7.60 (ddd, J_1 = 8.1, J_2 = 7.6 Hz, J_3 = 1.3 Hz, 1 H), 7.39–7.44 (m, 2 H), 7.25–7.31 (m, 3 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 155.8 (CH), 151.1 (C), 149.3 (C), 133.6 (CH), 131.2 (CH), 131.1 (C), 129.7 (CH), 129.3 (CH), 126.9 (CH), 124.5 (CH), 121.2 (CH).

MS: m/z = 227 $(M)^+$, 209, 195, 179, 167, 152, 77, 51.

N-[(E)-(6-Nitro-1,3-benzodioxol-5-yl)methylene]aniline (3h)[40]
IR (film): 3067, 2908, 2354, 1516, 1507, 1326, 1041, 924, 687 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.92 (s, 1 H), 7.73 (s, 1 H), 7.54 (s, 1 H), 7.39–7.44 (m, 2 H), 7.25–7.30 (m, 3 H), 6.18 (s, 2 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 155.7 (CH), 152.1 (C), 150.9 (C), 149.7 (C), 144.6 (C), 129.3 (CH), 128.3 (CH), 126.8 (CH), 121.2 (CH), 107.8 (CH), 105.1 (CH), 103.4 (CH_2).

MS: m/z = 271 $(M)^+$, 253, 222, 167, 139, 93, 77, 51.

N-[(E)-1,3-Benzodioxol-5-ylmethylene]aniline (3i)[41]
IR (film): 2879, 1584, 1436, 1264, 1042, 936, 767 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.32 (s, 1 H), 7.53 (d, J = 1.5 Hz, 1 H), 7.35–7.41 (m, 2 H), 7.27 (dd, J_1 = 7.8 Hz, J_2 = 1.5 Hz, 1 H), 7.15–7.23 (m, 3 H), 6.87 (d, J = 7.8 Hz, 1 H), 6.02 (s, 2 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 159.4 (CH), 152.6 (C), 149.9 (C), 147.7 (C), 134.1 (C), 129.1 (CH), 125.9 (CH), 125.8 (CH), 120.9 (CH), 108.2 (CH), 106.9 (CH), 101.6 (CH_2).

MS: m/z = 225 $(M)^+$, 194, 166, 139, 121, 104, 93, 77, 63, 51.

N-[(E)-(4-Chlorophenyl)methylene]aniline (3j)[36]
IR (film): 3056, 2869, 1623, 1490, 1405, 1088, 1013, 763 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.39 (s, 1 H), 7.82 (d, J = 8.3 Hz, 2 H), 7.43 (d, J = 8.3 Hz, 2 H), 7.35–7.41 (m, 2 H), 7.16–7.27 (m, 3 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 158.8 (CH), 151.6 (C), 137.3 (C), 134.7 (C), 129.9 (CH), 129.2 (CH), 129.1 (CH), 126.2 (CH), 120.8 (CH).

MS: m/z = 217 $(M + 2)^+$, 215 $(M)^+$, 178, 152, 137, 104, 89, 77, 63, 51.

N-[(E)-(3-Chlorophenyl)methylene]aniline (3k)[42]
IR (film): 3061, 2873, 1623, 1569, 1487, 1188, 1074, 763 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.27 (s, 1 H), 7.86 (s, 1 H), 7.64 (d, J = 7.8 Hz, 1 H), 7.31–7.37 (m, 3 H), 7.28 (t, J = 7.8 Hz, 1 H), 7.14–7.22 (m, 3 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 158.5 (CH), 151.3 (C), 137.8 (C), 134.8 (C), 131.1 (CH), 129.9 (CH), 129.1 (CH), 128.2 (CH), 127.1 (CH), 126.3 (CH), 120.8 (CH).

MS: m/z = 217 $(M + 2)^+$, 215 $(M)^+$, 196, 194, 180, 151, 112, 104, 89, 77, 63, 51.

N-[(E)-(2-Chlorophenyl)methylene]aniline (3l)[43]
IR (film): 3056, 2926, 1616, 1565, 1487, 1442, 1272, 1190, 1052, 763 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.90 (s, 1 H), 8.23 (dd, J_1 = 7.6 Hz, J_2 = 2.5 Hz, 1 H), 7.30–7.42 (m, 5 H), 7.21–7.27 (m, 3 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 156.8 (CH), 151.8 (C), 136.0 (C), 133.2 (C), 132.1 (CH), 129.9 (CH), 129.2 (CH), 128.5 (CH), 127.1 (CH), 126.3 (CH), 121.0 (CH).

MS: m/z = 217 $(M + 2)^+$, 215 $(M)^+$, 180, 152, 112, 104, 89, 77, 63, 51.

N-[(E)-(4-Methoxyphenyl)methylene]aniline (3m)[36]
IR (film): 3056, 2968, 2849, 1569, 1506, 1248, 1165, 1022, 843, 764 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.37 (s, 1 H), 7.84 (d, J = 8.8 Hz, 2 H), 7.34–7.40 (m, 2 H), 7.19 (d, J = 8.8 Hz, 2 H), 6.95–7.01 (m, 3 H), 3.85 (s, 3 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 162.3 (C), 159.8 (CH), 152.2 (C), 130.6 (CH), 129.1 (CH), 125.6 (CH), 120.9 (CH), 114.3 (C), 114.2 (CH), 55.4 (OCH_3).

MS: m/z = 211 $(M)^+$, 195, 167, 139, 104, 77, 63, 51.

N-[(E)-(3-Methoxyphenyl)methylene]aniline (3n)[37]
IR (film): 2946, 2834, 1580, 1487, 1268, 1213, 1152, 1041, 844, 787 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.38 (s, 1 H), 7.51 (dd, J_1 = 2.5 Hz, J_2 = 1.3 Hz, 1 H), 7.32–7.41 (m, 4 H), 7.17–7.24 (m, 3 H), 7.01 (ddd, J_1 = 8.0, J_2 = 2.8 Hz, J_3 = 1.3 Hz, 1 H), 3.83 (s, 3 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 160.2 (CH), 160.0 (C), 151.9 (C), 137.6 (C), 129.7 (CH), 129.1 (CH), 125.9 (CH), 122.3 (CH), 120.9 (CH), 118.3 (CH), 111.9 (CH), 55.3 (OCH_3).

MS: m/z = 211 $(M)^+$, 198, 181, 167, 139, 116, 104, 92, 77, 63, 51.

N-[(E)-(2-Methoxyphenyl)methylene]aniline (3o)[38]
IR (film): 2926, 2837, 1589, 14917, 1286, 1252, 1187, 1025, 758 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.91 (s, 1 H), 8.16 (dd, J_1 = 7.6 Hz, J_2 = 1.6 Hz, 1 H), 7.44 (ddd, J_1 = 8.5 Hz, J_2 = 7.6 Hz, J_3 = 1.8 Hz, 1 H), 7.35–7.40 (m, 2 H), 7.17–7.25 (m, 3 H), 7.01 (t, J = 7.6 Hz, 1 H), 6.94 (d, J = 8.5 Hz, 1 H), 3.87 (s, 3 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 159.5 (C), 156.6 (CH), 152.6 (C), 136.0 (CH), 132.8 (CH), 129.1 (CH), 127.6 (CH), 125.7 (CH), 124.6 (CH), 120.9 (CH), 111.1 (CH), 55.5 (OCH_3).

MS: m/z = 211 $(M)^+$, 198, 180, 167, 139, 119, 104, 93, 77, 63, 51.

N-[(E)-(3,4,5-Trimethoxyphenyl)methylene]aniline (3p)[44]
IR (film): 3067, 2908, 2354, 1516, 1507, 1326, 1041, 924, 687 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.34 (s, 1 H), 7.36–7.42 (m, 2 H), 7.19–7.24 (m, 3 H), 7.16 (s, 2 H), 3.93 (s, 6 H), 3.91 (s, 3 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 159.9 (CH), 153.5 (C), 151.6 (C), 141.1 (C), 131.5 (C), 129.2 (CH), 126.0 (CH), 120.9 (CH), 105.9 (CH), 61.0 (OCH_3), 56.3 (OCH_3).

MS: m/z = 271 $(M)^+$, 256, 225, 196, 153, 104, 93, 77, 66, 51.

5-Aryl-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinolines 5 and 6; General Procedure
To a soln of $NbCl_5$ (0.500, 0.250 or 0.125 mmol) in anhyd MeCN (1.0 mL), maintained at r.t., under N_2 atmosphere, was added a soln of the aldimine (1.0 mmol) and DHP (**4**, 2 mmol) in anhyd MeCN (2.0 mL). The mixture was quenched with 10% aq citric acid (2.0 mL). The mixture was diluted with H_2O (5.0 mL) and EtOAc (10.0 mL), the organic layer was separated and washed with 5% aq

$NaHCO_3$ (3 × 10.0 mL), sat. brine (2 × 10.0 mL), and dried (anhyd $MgSO_4$). The solvent was removed under vacuum and the products were purified by column chromatography (silica gel, hexane–EtOAc, 9.5:0.5).

rac-(4aS,5S,10bS)-5-Phenyl-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (5a)[11,45,46]
IR (film): 3312, 2941, 2865, 1608, 1486, 1317, 1265, 1069, 737 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.35–7.44 (m, 5 H), 7.30 (m, 1 H), 7.09 (td, J_1 = 7.7 Hz, J_2 = 0.8 Hz, 1 H), 6.79 (td, J_1 = 7.7 Hz, J_2 = 1.0 Hz, 1 H), 6.60 (dd, J_1 = 7.7 Hz, J_2 = 0.8 Hz, 1 H), 5.33 (d, J = 5.6 Hz, 1 H), 4.69 (d, J = 2.3 Hz, 1 H), 3.85 (NH, 1 H), 3.58 (m, 1 H), 3.43 (td, J_1 = 11.6 Hz, J_2 = 2.5 Hz, 1 H), 2.16 (m, 1 H), 1.47–1.58 (m, 2 H), 1.43 (m, 1 H), 1.31 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 145.6 (C), 141.5 (C), 129.2 (CH), 128.8 (CH), 128.7 (CH), 128.5 (CH), 128.0 (CH), 127.9 (CH), 127.2 (CH), 120.3 (C), 118.7 (CH), 114.8 (CH), 73.2 (CH), 61.0 (CH_2), 59.7 (CH), 39.3 (CH), 25.8 (CH_2), 18.4 (CH_2).

MS: *m/z* = 265 $(M)^+$, 234, 220, 194, 129, 117, 91, 77.

rac-(4aS,5R,10bS)-5-Phenyl-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (6a)[11,45,46]
IR (film): 3360, 2940, 2865, 1610, 1488, 1315, 1265, 1070, 737 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.30–7.44 (m, 5 H), 7.22 (dd, J_1 = 7.7 Hz, J_2 = 1.3 Hz, 1 H), 7.09 (td, J_1 = 7.7 Hz, J_2 = 1.3 Hz, 1 H), 6.71 (td, J_1 = 7.3 Hz, J_2 = 0.7 Hz, 1 H), 6.53 (dd, J_1 = 7.7 Hz, J_2 = 0.7 Hz, 1 H), 4.72 (d, J = 10.9 Hz, 1 H), 4.39 (d, J = 2.8 Hz, 1 H), 4.10 (dt, J_1 = 11.4 Hz, J_2 = 2.3 Hz, 1 H), 3.72 (td, J_1 = 11.4 Hz, J_2 = 2.5 Hz, 1 H), 2.11 (m, 1 H), 1.84 (tdt, J_1 = 13.4 Hz, J_2 = 12.4 Hz, J_3 = 4.5 Hz, 1 H), 1.65 (tt, J_1 = 13.4 Hz, J_2 = 4.5 Hz, 1 H), 1.47 (m, 1 H), 1.33 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 145.1 (C), 142.7 (C), 131.3 (CH), 129.8 (CH), 129.0 (CH), 128.3 (CH), 128.2 (CH), 121.0 (C), 117.9 (CH), 114.5 (CH), 74.9 (CH), 69.0 (CH_2), 55.2 (CH), 39.3 (CH), 24.5 (CH_2), 22.4 (CH_2).

MS: *m/z* = 265 $(M)^+$, 234, 220, 194, 129, 117, 91, 77.

rac-(4aS,5S,10bS)-5-(4-Methylphenyl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (5b)[47]
IR (film): 3307, 2926, 2861, 1608, 1505, 1318, 12645, 1071 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.41 (dt, J_1 = 7.5 Hz, J_2 = 1.0 Hz, 1 H), 7.29 (d, J = 7.8 Hz, 2 H), 7.18 (d, J = 7.8 Hz, 2 H), 7.08 (dddd, J_1 = 8.0 Hz, J_2 = 7.5, J_3 = 1.5 Hz, J_4 = 1.0 Hz, 1 H), 6.78 (td, J_1 = 7.5 Hz, J_2 = 1.0 Hz, 1 H), 6.58 (dd, J_1 = 8.0 Hz, J_2 = 1.0 Hz, 1 H), 5.31 (d, J = 5.7 Hz, 1 H), 4.65 (d, J = 2.4 Hz, 1 H), 3.93 (NH, 1 H), 3.58 (dtt, J_1 = 11.4, J_2 = 4.0 Hz, J_3 = 1.8 Hz, 1 H), 3.42 (td, J_1 = 11.4 Hz, J_2 = 2.7 Hz, 1 H), 2.36 (s, 3 H), 2.13 (dddd, J_1 = 11.9 Hz, J_2 = 5.7 Hz, J_3 = 4.3 Hz, J_4 = 2.7 Hz, 1 H), 1.46–1.56 (m, 2 H), 1.42 (m, 1 H), 1.32 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 145.5 (C), 140.3 (C), 139.4 (C), 131.3 (CH), 130.3 (CH), 129.9 (CH), 129.0 (CH), 122.1 (C), 120.4 (CH), 116.6 (CH), 75.0 (CH), 62.9 (CH_2), 61.3 (CH), 41.2 (CH), 27.7 (CH_2), 23.4 (CH_3), 20.3 (CH_2).

MS: *m/z* = 279 $(M)^+$, 249, 235, 221, 203, 188, 91, 77, 51.

rac-(4aS,5R,10bS)-5-(4-Methylphenyl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (6b)[47]
IR (film): 3328, 2942, 2850, 1611, 1486, 1366, 1080, 736 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.30 (d, J = 7.7 Hz, 2 H), 7.21 (d, J = 7.4 Hz, 1 H), 7.18 (d, J = 7.7Hz, 2 H), 7.07 (dd, J_1 = 7.6 Hz, J_2 = 7.4 Hz, 1 H), 6.69 (t, J = 7.4 Hz, 1 H), 6.51 (d, J = 7.6 Hz, 1 H), 4.68 (d, J = 10.9 Hz, 1 H), 4.39 (d, J = 2.3 Hz, 1 H), 4.10 (m, 1 H), 3.72 (td, J_1 = 11.6 Hz, J_2 = 1.8 Hz, 1 H), 2.36 (s, 3 H), 2.07 (m, 1 H), 1.83 (tdt, J_1 = 13.6 Hz, J_2 = 12.4 Hz, J_3 = 4.5 Hz, 1 H), 1.64 (tt, J_1 = 13.6 Hz, J_2 = 4.5 Hz, 1 H), 1.48 (m, 1 H), 1.32 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 143.7 (C), 138.1 (C), 136.5 (C), 129.8 (CH), 128.3 (CH), 128.2 (CH), 126.6 (CH), 119.6 (C), 116.3 (CH), 113.1 (CH), 73.5 (CH), 67.6 (CH_2), 53.4 (CH), 37.7 (CH), 23.0 (CH_2), 20.9 (CH_2), 20.1 (CH_3).

MS: *m/z* = 279 $(M)^+$, 249, 235, 221, 203, 188, 91, 77, 51.

rac-(4aS,5S,10bS)-5-(3-Methylphenyl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (5c)
IR (film): 3373, 2937, 2855, 1610, 1488, 1365, 1080, 914, 748 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.19–7.29 (m, 3 H), 7.42 (dt, J_1 = 7.7 Hz, J_2 = 1.3 Hz, 1 H), 7.11 (d, J = 6.8 Hz, 1 H), 7.08 (m, 1 H), 6.79 (td, J_1 = 7.6 Hz, J_2 = 1.0 Hz, 1 H), 6.60 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 5.32 (d, J = 5.6 Hz, 1 H), 4.65 (d, J = 2.5 Hz, 1 H), 3.94 (NH, 1 H), 3.59 (ddt, J_1 = 11.4, J_2 = 4.0 Hz, J_3 = 2.0 Hz, 1 H), 3.43 (td, J_1 = 11.4 Hz, J_2 = 2.5 Hz, 1 H), 2.36 (s, 3 H), 2.16 (m, 1 H), 1.47–1.59 (m, 2 H), 1.43 (m, 1 H), 1.32 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 146.4 (C), 142.2 (C), 139.2 (C), 129.4 (CH), 129.3 (CH), 129.2 (CH), 128.9 (CH), 128.7 (CH), 125.0 (CH), 121.1 (C), 119.4 (CH), 115.5 (CH), 73.9 (CH), 61.8 (CH_2), 60.4 (CH), 40.0 (CH), 26.6 (CH_2), 22.7 (CH_3), 19.2 (CH_2).

MS: *m/z* = 279 $(M)^+$, 249, 235, 221, 203, 188, 91, 77, 51.

HRMS (ESI): *m/z* [M + H] calcd for $C_{19}H_{22}NO$: 280.1695; found: 280.1699.

rac-(4aS,5R,10bS)-5-(3-Methylphenyl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (6c)
IR (film): 3374, 2940, 2865, 1607, 1488, 1365, 1088, 737 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.19–7.28 (m, 4 H), 7.13 (d, J = 7.3 Hz, 1 H), 7.08 (ddd, J_1 = 8.1, J_2 = 7.3 Hz, J_3 = 1.5 Hz, 1 H), 6.71 (t, J = 7.3 Hz, 1 H), 6.51 (d, J = 8.1 Hz, 1 H), 4.68 (d, J = 10.9 Hz, 1 H), 4.39 (d, J = 2.5 Hz, 1 H), 4.10 (m, 1 H), 3.71 (td, J_1 = 11.6 Hz, J_2 = 2.5 Hz, 1 H), 2.36 (s, 3 H), 2.09 (m, 1 H), 1.84 (tdt, J_1 = 13.4, J_2 = 12.4 Hz, J_3 = 4.3 Hz, 1 H), 1.65 (tt, J_1 = 13.4 Hz, J_2 = 4.5 Hz, 1 H), 1.49 (m, 1 H), 1.33 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 144.9 (C), 142.5 (C), 138.7 (C), 131.3 (CH), 129.8 (CH), 129.1 (CH), 128.9 (CH), 128.8 (CH), 125.4 (CH), 121.3 (C), 118.0 (CH), 114.7 (CH), 75.0 (CH), 69.1 (CH_2), 55.2 (CH), 39.1 (CH), 24.6 (CH_2), 22.4 (CH_2), 21.9 (CH_3).

MS: *m/z* = 279 $(M)^+$, 249, 235, 221, 203, 188, 91, 77, 51.

HRMS (ESI): *m/z* [M + H] calcd for $C_{19}H_{22}NO$: 280.1695; found: 280.1698.

rac-(4aS,5S,10bS)-5-(2-Methylphenyl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (5d)
IR (film): 3373, 2940, 2865, 1608, 1486, 1316, 1265, 1088, 736 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.62 (d, J = 6.6 Hz, 1 H), 7.45 (d, J = 7.3 Hz, 1 H), 7.19–7.25 (m, 3 H), 7.10 (t, J = 7.3 Hz, 1 H), 6.81 (t, J = 7.3 Hz, 1 H), 6.62 (d, J = 7.3 Hz, 1 H), 5.32 (d, J = 5.6 Hz, 1 H), 4.89 (d, J = 2.2 Hz, 1 H), 3.60 (ddt, J_1 = 11.5 Hz, J_2 = 4.1 Hz, J_3 = 2.0 Hz, 1 H), 3.43 (td, J_1 = 11.5 Hz, J_2 = 2.6 Hz, 1 H), 2.34 (s, 3 H), 2.18 (m, 1 H), 1.49–1.67 (m, 3 H), 1.34 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 138.6 (C), 135.02 (C), 130.7 (CH), 128.1 (CH), 127.7 (CH), 127.3 (C), 127.2 (CH), 126.5 (CH), 125.7 (CH), 118.3 (CH), 114.5 (CH), 99.6 (C), 72.8 (CH), 60.8 (CH_2), 55.4 (CH), 35.6 (CH), 25.5 (CH_2), 18.9 (CH_3), 18.3 (CH_2).

MS: *m/z* = 279 $(M)^+$, 249, 235, 221, 203, 188, 91, 77, 51.

HRMS (ESI): *m/z* [M + H] calcd for $C_{19}H_{22}NO$: 280.1695; found: 280.1699.

rac-(4aS,5R,10bS)-5-(2-Methylphenyl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (6d)
IR (film): 3374, 2936, 2865, 1609, 1485, 1320, 1264, 1088, 736 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.46 (d, *J* = 4.4 Hz, 1 H), 7.28 (d, *J* = 7.6 Hz, 1 H), 7.23 (s, 3 H), 7.12 (t, *J* = 7.6 Hz, 1 H), 6.75 (t, *J* = 7.6 Hz, 1 H), 6.54 (d, *J* = 8.1 Hz, 1 H), 5.00 (d, *J* = 10.3 Hz, 1 H), 4.49 (d, *J* = 2.6 Hz, 1 H), 4.10 (d, *J* = 10.9 Hz, 1 H), 3.74 (td, J_1 = 10.9 Hz, J_2 = 1.9 Hz, 1 H), 2.50 (s, 3 H), 2.29 (m, 1 H), 1.88–1.66 (m, 2 H), 1.56 (m, 1 H), 1.43 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 144.9 (C), 140.5 (C), 136.9 (C), 131.2 (CH), 131.1 (CH), 129.7 (CH), 128.2 (CH), 127.9 (CH), 126.9 (CH), 120.9 (C), 117.9 (CH), 114.5 (CH), 74.7 (CH), 68.4 (CH_2), 51.5 (CH), 38.1 (CH), 24.5 (CH_2), 23.3 (CH_2), 20.4 (CH_3).

MS: *m/z* = 279 $(M)^+$, 249, 235, 221, 203, 188, 91, 77, 51.

HRMS (ESI): *m/z* [M + H] calcd for $C_{19}H_{22}NO$: 280.1695; found: 280.1689.

rac-(4aS,5R,10bS)-5-Mesityl-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (5e)
IR (film): 3384, 2944, 2853, 1610, 1496, 1365, 1265, 1080, 736 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.22 (dd, J_1 = 7.3 Hz, J_2 = 1.5 Hz, 1 H), 7.07 (ddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, 1 H), 6.86 (s, 2 H), 6.68 (td, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 6.48 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 5.21 (d, *J* = 11.6 Hz, 1 H), 4.39 (d, *J* = 2.8 Hz, 1 H), 4.11 (dd, J_1 = 11.4 Hz, J_2 = 4.0 Hz, 1 H), 3.88 (NH, 1 H), 3.69 (td, J_1 = 11.4 Hz, J_2 = 2.2 Hz, 1 H), 2.64 (m, 1 H), 2.45 (s, 6 H), 2.27 (s, 3 H), 1.66–1.77 (m, 2 H), 1.47–1.60 (m, 2 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 145.4 (C), 136.9 (C), 133.5 (C), 131.3 (CH), 129.3 (CH), 120.9 (C), 117.1 (CH), 114.3 (CH), 75.4 (CH), 68.9 (CH_2), 50.0 (CH), 34.6 (CH), 23.9 (CH_2), 22.4 (CH_2), 21.3 (CH_3), 20.7 (2 CH_3).

MS: *m/z* = 307 $(M)^+$, 277, 263, 249, 231, 188, 119, 91, 77, 51.

HRMS (ESI): *m/z* [M + H] calcd for $C_{21}H_{26}NO$: 308.2008; found: 308.2024.

rac-(4aS,5S,10bS)-5-(3-Nitrophenyl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (5f)[47]
IR (film): 3369, 2950, 2857, 1606, 1528, 1349, 1087, 759 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.32 (t, *J* = 2.0 Hz, 1 H), 8.16 (ddd, J_1 = 8.0 Hz, J_2 = 2.0 Hz, J_3 = 1.0 Hz, 1 H), 7.76 (d, *J* = 7.8 Hz, 1 H), 7.56 (dd, J_1 = 8.0 Hz, J_2 = 7.8 Hz, 1 H), 7.42 (d, *J* = 7.3 Hz, 1 H), 7.12 (dd, J_1 = 7.8 Hz, J_2 = 7.3 Hz, 1 H), 6.84 (td, J_1 = 7.3 Hz, J_2 = 0.8 Hz, 1 H), 6.67 (dd, J_1 = 7.8 Hz, J_2 = 1.0 Hz, 1 H), 5.34 (d, *J* = 5.6 Hz, 1 H), 4.79 (d, *J* = 2.3 Hz, 1 H), 3.94 (NH, 1 H), 3.59 (m, 1 H), 3.43 (td, J_1 = 11.4 Hz, J_2 = 2.5 Hz, 1 H), 2.20 (m, 1 H), 1.46–1.63 (m, 2 H), 1.44 (m, 1 H), 1.19 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 148.4 (C), 144.5 (C), 143.5 (C), 133.0 (CH), 129.4 (CH), 128.3 (CH), 127.6 (CH), 122.6 (CH), 121.7 (CH), 119.9 (C), 119.1 (CH), 114.9 (CH), 72.4 (CH), 60.6 (CH_2), 58.8 (CH), 38.8 (CH), 25.26 (CH_2), 17.9 (CH_2).

MS: *m/z* = 310 $(M)^+$, 266, 251, 205, 179, 130, 115, 91, 77, 65, 51.

rac-(4aS,5R,10bS)-5-(3-Nitrophenyl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (6f)[47]
IR (film): 3386, 2943, 2828, 1608, 1524, 1482, 1344, 1059, 759 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.31 (t, *J* = 1.8 Hz, 1 H), 8.18 (ddd, J_1 = 8.1 Hz, J_2 = 2.3 Hz, J_3 = 1.0 Hz, 1 H), 7.76 (d, *J* = 7.8 Hz, 1 H), 7.55 (dd, J_1 = 8.0 Hz, J_2 = 7.8 Hz, 1 H), 7.23 (dd, J_1 = 7.5 Hz, J_2 = 1.4 Hz, 1 H), 7.12 (ddd, J_1 = 8.1 Hz, J_2 = 7.5 Hz, J_3 = 1.4 Hz, 1 H), 6.75 (td, J_1 = 7.5 Hz, J_2 = 1.0 Hz, 1 H), 6.57 (d, *J* = 8.1 Hz, 1 H), 4.83 (d, *J* = 10.6 Hz, 1 H), 4.40 (d, *J* = 2.8 Hz, 1 H), 4.11 (m, 1 H), 3.74 (td, J_1 = 11.5 Hz, J_2 = 2.0 Hz, 1 H), 2.11 (m, 1 H), 1.84 (m, 1 H), 1.70 (tt, J_1 = 13.8 Hz, J_2 = 5.0 Hz, 1 H), 1.40 (m, 2 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 148.6 (C), 144.8 (C), 144.7 (C), 134.0 (CH), 130.9 (CH), 129.6 (CH), 129.5 (CH), 123.0 (CH), 122.7 (CH), 120.7 (C), 118.2 (CH), 114.4 (CH), 74.1 (CH), 68.5 (CH_2), 54.5 (CH), 39.0 (CH), 24.1 (CH_2), 22.1 (CH_2).

MS: *m/z* = 310 $(M)^+$, 266, 251, 205, 179, 130, 115, 91, 77, 65, 51.

rac-(4aS,5S,10bS)-5-(2-Nitrophenyl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (5g)
IR (film): 3373, 2939, 2850, 1608, 1526, 1505, 1352, 1265, 1073, 736 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.97 (dd, J_1 = 7.6 Hz, J_2 = 1.0 Hz, 1 H), 7.95 (dd, J_1 = 8.0 Hz, J_2 = 1.3 Hz, 1 H), 7.64 (td, J_1 = 7.6 Hz, J_2 = 1.0 Hz, 1 H), 7.46 (ddd, J_1 = 8.0 Hz, J_2 = 7.6 Hz, J_3 = 1.0 Hz, 1 H), 7.44 (d, *J* = 7.6 Hz, 1 H), 7.10 (dd, J_1 = 8.1 Hz, J_2 = 7.6 Hz, 1 H), 6.83 (t, *J* = 7.6 Hz, 1 H), 6.61 (d, *J* = 8.1 Hz, 1 H), 5.34 (d, *J* = 5.5 Hz, 1 H), 5.15 (d, *J* = 2.0 Hz, 1 H), 3.60 (m, 1 H), 3.43 (td, J_1 = 11.6 Hz, J_2 = 2.5 Hz, 1 H), 2.52 (m, 1 H), 1.51–1.70 (m, 2 H), 1.46 (m, 1 H), 1.32 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 148.5 (C), 144.5 (C), 135.5 (C), 132.3 (CH), 129.1 (CH), 128.0 (CH), 127.8 (CH), 127.5 (CH), 124.5 (CH), 120.2 (C), 118.8 (CH), 114.6 (CH), 72.0 (CH), 60.2 (CH_2), 54.2 (CH), 36.0 (CH), 25.0 (CH_2), 18.2 (CH_2).

MS: *m/z* = 310 $(M)^+$, 266, 251, 217, 204, 188, 130, 115, 89, 77, 63, 51.

HRMS (ESI): *m/z* [M + H] calcd for $C_{18}H_{19}N_2O_3$: 311.1390; found: 311.1367.

rac-(4aS,5R,10bS)-5-(2-Nitrophenyl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (6g)
IR (film): 3373, 2939, 2850, 1608, 1526, 1505, 1352, 1265, 1073, 736 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.81 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 7.67 (dd, J_1 = 7.8 Hz, J_2 = 1.0 Hz, 1 H), 7.57 (td, J_1 = 7.8 Hz, J_2 = 1.5 Hz, 1 H), 7.43 (ddd, J_1 = 8.1 Hz, J_2 = 7.8 Hz, J_3 = 1.5 Hz, 1 H), 7.28 (dd, J_1 = 7.6 Hz, J_2 = 1.5 Hz, 1 H), 7.13 (ddd, J_1 = 8.1 Hz, J_2 = 7.6 Hz, J_3 = 1.5 Hz, 1 H), 6.76 (td, J_1 = 7.6 Hz, J_2 = 1.0 Hz, 1 H), 6.59 (d, *J* = 8.1 Hz, 1 H), 5.14 (d, *J* = 8.1 Hz, 1 H), 4.46 (d, *J* = 3.6 Hz, 1 H), 4.25 (NH, 1 H), 3.88 (m, 1 H), 3.64 (ddd, J_1 = 11.4 Hz, J_2 = 8.4 Hz, J_3 = 2.9 Hz, 1 H), 2.24 (m, 1 H), 1.77 (m, 1 H), 1.62 (m, 1 H), 1.41–1.50 (m, 2 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 149.9 (C), 143.9 (C), 137.2 (C), 132.9 (CH), 129.8 (CH), 129.4 (CH), 129.2 (CH), 128.4 (CH), 124.0 (CH), 119.8 (C), 119.0 (CH), 114.1 (CH), 72.2 (CH), 61.8 (CH_2), 50.8 (CH), 36.7 (CH), 24.5 (CH_2), 23.1 (CH_2).

MS: *m/z* = 310 $(M)^+$, 266, 251, 217, 204, 188, 130, 115, 89, 77, 63, 51.

HRMS (ESI): *m/z* [M + H] calcd for $C_{18}H_{19}N_2O_3$: 311.1390; found: 311.1390.

rac-(4aS,5S,10bS)-5-(6-Nitro-1,3-benzodioxol-5-yl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (5h)
IR (film): 3374, 2925, 1732, 1480, 1333, 1269, 1036, 737 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.53 (s, 1 H), 7.44 (dd, J_1 = 7.4 Hz, J_2 = 1.5 Hz, 1 H), 7.42 (s, 1 H), 7.09 (dddd, J_1 = 8.1 Hz, J_2 = 7.4 Hz, J_3 = 1.5 Hz, J_4 = 0.7 Hz, 1 H), 6.83 (td, J_1 = 7.4 Hz, J_2 = 1.0 Hz, 1 H), 6.60 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 6.14 (d, *J* = 1.2 Hz, 1 H), 6.12 (d, *J* = 1.2 Hz, 1 H), 5.33 (d, *J* = 5.6 Hz, 1 H), 5.18 (d, *J* = 2.0 Hz, 1 H), 3.60 (m, 1 H), 3.42 (td, J_1 = 11.4 Hz, J_2 = 2.0 Hz, 1 H), 2.53 (m, 1 H), 1.55–1.64 (m, 2 H), 1.48 (m, 1 H), 1.37 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 151.6 (C), 147.0 (C), 144.8 (C), 142.6 (C), 133.6 (C), 128.0 (CH), 127.8 (CH), 120.6 (C), 119.1 (CH), 115.0 (CH), 108.2 (CH), 105.9 (CH), 103.0 (CH_2), 72.3 (CH), 60.5 (CH_2), 54.7 (CH), 36.2 (CH), 25.3 (CH_2), 18.7 (CH_2).

MS: *m/z* = 354 $(M)^+$, 324, 310, 296, 279, 194, 188, 164, 91, 77, 51.

HRMS (ESI): *m/z* [M + H] calcd for $C_{19}H_{19}N_2O_5$: 355.1288; found: 355.1298.

rac-(4aS,5R,10bS)-5-(6-Nitro-1,3-benzodioxol-5-yl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (6h)
IR (film): 3375, 2930, 1729, 1479, 1331, 1271, 1036, 737 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.37 (s, 1 H), 7.27 (dd, J_1 = 7.6 Hz, J_2 = 1.5 Hz, 1 H), 7.12 (ddd, J_1 = 8.1 Hz, J_2 = 7.6 Hz, J_3 = 1.5 Hz, 1 H), 7.04 (s, 1 H), 6.76 (td, J_1 = 7.6 Hz, J_2 = 1.0 Hz, 1 H), 6.58 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 6.09 (d, *J* = 1.0 Hz, 1 H), 6.08 (d, *J* = 1.0 Hz, 1 H), 5.19 (d, *J* = 7.6 Hz, 1 H), 4.48 (d, *J* = 3.8 Hz, 1 H), 3.85 (m, 1 H), 3.63 (ddd, J_1 = 11.4 Hz, J_2 = 8.1 Hz, J_3 = 3.0 Hz, 1 H), 2.16 (m, 1 H), 1.78 (m, 1 H), 1.59 (m, 1 H), 1.43–1.52 (m, 2 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 151.8 (C), 147.1 (C), 143.8 (C), 142.9 (C), 134.7 (C), 129.7 (CH), 129.1 (CH), 119.7 (C), 118.0 (CH), 114.0 (CH), 107.8 (CH), 105.2 (CH), 102.9 (CH_2), 71.5 (CH), 60.4 (CH_2), 51.1 (CH), 37.8 (CH), 24.5 (CH_2), 23.3 (CH_2).

MS: *m/z* = 354 $(M)^+$, 324, 310, 296, 279, 194, 188, 164, 91, 77, 51.

HRMS (ESI): *m/z* [M + H] calcd for $C_{19}H_{19}N_2O_5$: 355.1288; found: 355.1265.

rac-(4aS,5S,10bS)-5-(1,3-Benzodioxol-5-yl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (5i)[15,48]
IR (film): 3380, 2925, 1732, 1480, 1333, 1269, 1036, 737 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.41 (dt, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 7.09 (dddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, J_4 = 0.8 Hz, 1 H), 6.92 (d, *J* = 1.8 Hz, 1 H), 6.88 (ddd, J_1 = 8.1, J_2 = 1.8 Hz, J_3 = 0.7 Hz, 1 H), 6.81 (d, *J* = 8.1 Hz, 1 H), 6.79 (td, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 6.58 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 5.70 (m, 2 H), 5.30 (d, *J* = 5.6 Hz, 1 H), 4.60 (d, *J* = 2.5 Hz, 1 H), 3.59 (m, 1 H), 3.42 (td, J_1 = 11.4 Hz, J_2 = 2.5 Hz, 1 H), 2.11 (m, 1 H), 1.43–1.48 (m, 3 H), 1.36 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 147.7 (C), 146.8 (C), 145.1 (C), 135.1 (C), 128.1 (CH), 127.6 (CH), 119.9 (C), 119.8 (CH), 118.4 (CH), 114.4 (CH), 108.1 (CH), 107.4 (CH), 101.1 (CH_2), 72.7 (CH), 60.7 (CH_2), 59.1 (CH), 39.1 (CH), 25.4 (CH_2), 18.1 (CH_2).

MS: *m/z* = 309 $(M)^+$, 279, 265, 225, 233, 218, 188, 121, 91, 77, 51.

rac-(4aS,5R,10bS)-5-(1,3-Benzodioxol-5-yl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (6i)[15,48]
IR (film): 3375, 2930, 1729, 1479, 1331, 1271, 1036, 737 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.21 (dd, J_1 = 7.3 Hz, J_2 = 1.5 Hz, 1 H), 7.09 (ddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, 1 H), 6.93 (d, *J* = 1.6 Hz, 1 H), 6.87 (dd, J_1 = 7.8 Hz, J_2 = 1.6 Hz, 1 H), 6.79 (d, *J* = 7.8 Hz, 1 H), 6.70 (td, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 6.52 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 5.97 (s, 2 H), 4.64 (d, *J* = 11.2 Hz, 1 H), 4.38 (d, *J* = 2.8 Hz, 1 H), 4.10 (ddt, J_1 = 11.4 Hz, J_2 = 4.3 Hz, J_3 = J_4 = 2.3 Hz, 1 H), 3.72 (td, J_1 = 11.4 Hz, J_2 = 2.5 Hz, 1 H), 2.01 (m, 1 H), 1.81 (tdt, J_1 = 13.4 Hz, J_2 = 11.9 Hz, J_3 = 4.3 Hz, 1 H), 1.66 (tt, J_1 = 13.6 Hz, J_2 = 4.6 Hz, 1 H), 1.52 (m, 1 H), 1.34 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 148.0 (C), 147.2 (C), 144.7 (C), 136.1 (C), 130.9 (CH), 129.4 (CH), 121.3 (CH), 120.7 (C), 117.6 (CH), 114.2 (CH), 108.1 (CH), 107.7 (CH), 74.6 (CH), 68.7 (CH_2), 54.5 (CH), 38.9 (CH), 24.1 (CH_2), 21.9 (CH_2).

MS: *m/z* = 309 $(M)^+$, 279, 265, 225, 233, 218, 188, 121, 91, 77, 51.

rac-(4aS,5S,10bS)-5-(4-Chlorophenyl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (5j)[49,50]
IR (film): 3387, 2940, 1604, 1486, 1276, 1085, 1014, 750 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.42 (dt, J_1 = 7.3 Hz, J_2 = 1.5 Hz, 1 H), 7.35 (s, 4 H), 7.09 (dddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, J_4 = 0.8 Hz, 1 H), 6.80 (td, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 6.60 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 5.30 (d, *J* = 5.6 Hz, 1 H), 4.65 (d, *J* = 2.5 Hz, 1 H), 3.80 (NH, 1 H), 3.59 (ddt, J_1 = 11.4 Hz, J_2 = 4.0 Hz, J_3 = 2.0 Hz, 1 H), 3.42 (td, J_1 = 11.4 Hz, J_2 = 2.5 Hz, 1 H), 2.12 (m, 1 H), 1.41–1.55 (m, 3 H), 1.26 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 144.9 (C), 139.6 (C), 133.1 (C), 128.5 (CH), 128.1 (CH), 127.6 (CH), 119.9 (C), 118.6 (CH), 114.5 (CH), 72.6 (CH), 60.6 (CH_2), 58.8 (CH), 38.9 (CH), 25.3 (CH_2), 17.9 (CH_2).

MS: *m/z* = 301 $(M + 2)^+$, 299 $(M)^+$, 268, 266, 240, 217, 154, 127, 115, 89, 77, 63, 51.

rac-(4aS,5R,10bS)-5-(4-Chlorophenyl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (6j)[49,50]
IR (film): 3364, 2935, 2851, 1609, 1486, 1262, 1050, 913, 750 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.35 (d, *J* = 1.4 Hz, 4 H), 7.22 (dd, J_1 = 7.3 Hz, J_2 = 1.5 Hz, 1 H), 7.09 (ddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, 1 H), 6.72 (td, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 6.53 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 4.69 (d, *J* = 10.6 Hz, 1 H), 4.38 (d, *J* = 2.8 Hz, 1 H), 4.09 (m, 1 H), 3.72 (td, J_1 = 11.4 Hz, J_2 = 2.3 Hz, 1 H), 2.04 (m, 1 H), 1.81 (m, 1 H), 1.66 (tt, J_1 = 13.7 Hz, J_2 = 4.5 Hz, 1 H), 1.44 (m, 1 H), 1.34 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 144.5 (C), 140.8 (C), 133.5 (C), 130.9 (CH), 129.4 (CH), 129.1 (CH), 128.8 (CH), 120.7 (C), 117.8 (CH), 114.2 (CH), 74.4 (CH), 60.6 (CH_2), 54.3 (CH), 38.9 (CH), 24.1 (CH_2), 21.9 (CH_2).

MS: *m/z* = 301 $(M + 2)^+$, 299 $(M)^+$, 268, 266, 240, 216, 188, 130, 115, 89, 77, 65, 51.

rac-(4aS,5S,10bS)-5-(3-Chlorophenyl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (5k)
IR (film): 3314, 2941, 2865, 1607, 1476, 1264, 1071, 737 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.40–7.44 (m, 2 H), 7.29 (m, 3 H), 7.10 (dddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, J_4 = 0.7 Hz, 1 H), 6.81 (td, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 6.61 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 5.31 (d, *J* = 5.6 Hz, 1 H), 4.65 (d, *J* = 2.5 Hz, 1 H), 3.83 (NH, 1 H), 3.59 (m, 1 H), 3.42 (td, J_1 = 11.4 Hz, J_2 = 2.5 Hz, 1 H), 2.15 (m, 1 H), 1.42–1.56 (m, 3 H), 1.27 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 144.8 (C), 143.3 (C), 134.4 (C), 129.7 (CH), 128.1 (CH), 127.7 (CH), 127.6 (CH), 126.9 (CH), 125.0 (CH), 119.9 (C), 118.6 (CH), 114.6 (CH), 72.6 (CH), 60.6 (CH_2), 58.9 (CH), 38.8 (CH), 25.3 (CH_2), 17.9 (CH_2).

MS: *m/z* = 301 $(M + 2)^+$, 299 $(M)^+$, 268, 266, 254, 240, 217, 144, 130, 115, 102, 89, 77, 63, 51.

HRMS (ESI): *m/z* [M + H] calcd for $C_{18}H_{19}ClNO$: 300.1149; found: 300.1125.

rac-(4aS,5R,10bS)-5-(3-Chlorophenyl)-3,4,4a,5,6,10b-hexahydro-2H-pyrano[3,2-c]quinoline (6k)
IR (film): 3356, 2925, 2839, 1609, 1490, 1368, 1260, 1053, 749 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.42 (s, 1 H), 7.29 (m, 3 H), 7.21 (dd, J_1 = 7.3 Hz, J_2 = 1.5 Hz, 1 H), 7.09 (ddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, 1 H), 6.71 (td, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 6.52 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 4.67 (d, *J* = 10.6 Hz, 1 H), 4.37 (d, *J* = 2.8 Hz, 1 H), 4.08 (m, 1 H), 3.71 (td, J_1 = 11.4 Hz, J_2 = 2.5

Hz, 1 H), 2.04 (m, 1 H), 1.81 (tdt, J_1 = 13.1 Hz, J_2 = 11.6 Hz, J_3 = 4.3 Hz, 1 H), 1.66 (tt, J_1 = 13.1 Hz, J_2 = 4.5 Hz, 1 H), 1.46 (m, 1 H), 1.36 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 144.5 (C), 144.4 (C), 134.5 (C), 130.9 (CH), 129.9 (CH), 129.4 (CH), 128.1 (CH), 127.8 (CH), 126.1 (CH), 120.6 (C), 117.7 (CH), 114.2 (CH), 74.3 (CH), 68.5 (CH_2), 54.5 (CH), 38.9 (CH), 24.1 (CH_2), 22.0 (CH_2).

MS: *m/z* = 301 (M + 2)$^+$, 299 (M)$^+$, 254, 240, 228, 144, 130, 115, 102, 89, 77, 65, 51.

HRMS (ESI): *m/z* [M + H] calcd for $C_{18}H_{19}ClNO$: 300.1149; found: 300.1154.

rac-(4a*S*,5*S*,10b*S*)-5-(2-Chlorophenyl)-3,4,4a,5,6,10b-hexahydro-2*H*-pyrano[3,2-*c*]quinoline (5l)[47]

IR (film): 3363, 2938, 2864, 1605, 1479, 1317, 1089, 928, 754 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.69 (dd, J_1 = 7.6 Hz, J_2 = 1.8 Hz, 2 H), 7.44 (dt, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 7.40 (dd, J_1 = 7.6 Hz, J_2 = 1.5 Hz, 1 H), 7.30 (td, J_1 = 7.6 Hz, J_2 = 1.5 Hz, 1 H), 7.24 (td, J_1 = 7.6 Hz, J_2 = 1.8 Hz, 1 H), 7.10 (dddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, J_4 = 0.8 Hz, 1 H), 6.82 (td, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 6.62 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 5.34 (d, *J* = 5.6 Hz, 1 H), 5.07 (d, *J* = 2.3 Hz, 1 H), 3.60 (m, 1 H), 3.43 (td, J_1 = 11.5 Hz, J_2 = 2.5 Hz, 1 H), 2.41 (m, 1 H), 1.52–1.60 (m, 2 H), 1.44 (m, 1 H), 1.22 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 145.2 (C), 138.0 (C), 132.7 (C), 129.9 (CH), 128.5 (CH), 128.3 (CH), 128.0 (CH), 127.7 (CH), 126.5 (CH), 120.2 (C), 118.6 (CH), 114.7 (CH), 72.4 (CH), 60.6 (CH_2), 55.7 (CH), 34.8 (CH), 25.4 (CH_2), 18.5 (CH_2).

MS: *m/z* = 301 (M + 2)$^+$, 299 (M)$^+$, 254, 240, 220, 204, 144, 130, 115, 102, 91, 77, 51.

rac-(4a*S*,5*R*,10b*S*)-5-(2-Chlorophenyl)-3,4,4a,5,6,10b-hexahydro-2*H*-pyrano[3,2-*c*]quinoline (6l)[47]

IR (film): 3362, 2939, 2858, 1609, 1464, 1264, 1083, 928, 736 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.48 (dd, J_1 = 7.6 Hz, J_2 = 1.8 Hz, 1 H), 7.37 (dd, J_1 = 7.6 Hz, J_2 = 1.5 Hz, 1 H), 7.22–7.28 (m, 2 H), 7.20 (dd, J_1 = 7.3 Hz, J_2 = 1.5 Hz, 1 H), 7.10 (ddd, J_1 = 7.8 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, 1 H), 6.71 (td, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 6.52 (d, *J* = 7.8 Hz, 1 H), 5.17 (d, *J* = 9.3 Hz, 1 H), 4.43 (d, *J* = 3.3 Hz, 1 H), 4.05 (NH, 1 H), 3.98 (m, 1 H), 3.67 (td, J_1 = 11.1 Hz, J_2 = 2.7 Hz, 1 H), 2.20 (m, 1 H), 1.92 (m, 1 H), 1.70 (m, 1 H), 1.38–1.51 (m, 2 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 144.4 (C), 140.1 (C), 133.8 (C), 130.3 (CH), 129.5 (CH), 129.2 (CH), 129.0 (CH), 128.7 (CH), 127.3 (CH), 120.1 (C), 117.5 (CH), 113.9 (CH), 73.3 (CH), 67.2 (CH_2), 51.4 (CH), 38.0 (CH), 24.3 (CH_2), 23.1 (CH_2).

MS: *m/z* = 301 (M + 2)$^+$, 299 (M)$^+$, 254, 240, 220, 204, 144, 130, 115, 102, 89, 77, 65, 41.

rac-(4a*S*,5*S*,10b*S*)-5-(4-Methoxyphenyl)-3,4,4a,5,6,10b-hexahydro-2*H*-pyrano[3,2-*c*]quinoline (5m)[15,47]

IR (film): 3375, 2939, 1700, 1607, 1512, 1245, 1176, 1033, 751 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.42 (dt, J_1 = 7.3 Hz, J_2 = 1.4 Hz, 1 H), 7.33 (d, *J* = 8.6 Hz, 2 H), 7.09 (dddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.4 Hz, J_4 = 0.8 Hz, 1 H), 6.92 (d, *J* = 8.6 Hz, 2 H), 6.79 (td, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 6.59 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 5.32 (d, *J* = 5.6 Hz, 1 H), 4.65 (d, *J* = 2.5 Hz, 1 H), 3.82 (s, 3 H), 3.59 (m, 1 H), 3.43 (td, J_1 = 11.4 Hz, J_2 = 2.6 Hz, 1 H), 2.12 (m, 1 H), 1.42–1.60 (m, 3 H), 1.34 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 159.0 (C), 143.3 (C), 133.1 (C), 128.0 (CH), 127.9 (CH), 127.6 (CH), 119.9 (C), 118.2 (CH), 114.3 (CH), 113.7 (CH), 72.8 (CH), 60.7 (CH_2), 58.8 (CH_3), 55.3 (CH), 39.0 (CH), 25.4 (CH_2), 18.0 (CH_2).

MS: *m/z* = 295 (M)$^+$, 276, 262, 248, 218, 204, 191, 130, 109, 89, 77, 63.

rac-(4a*S*,5*R*,10b*S*)-5-(4-Methoxyphenyl)-3,4,4a,5,6,10b-hexahydro-2*H*-pyrano[3,2-*c*]quinoline (6m)[15,47]

IR (film): 3384, 2939, 2853, 1610, 1513, 1249, 1174, 1080, 750 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.36 (d, *J* = 8.8 Hz, 2 H), 7.24 (dd, J_1 = 7.3 Hz, J_2 = 1.5 Hz, 1 H), 7.11 (ddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, 1 H), 6.93 (d, *J* = 8.8 Hz, 2 H), 6.72 (td, J_1 = 7.3 Hz, J_2 = 1.3 Hz, 1 H), 6.55 (dd, J_1 = 8.1 Hz, J_2 = 1.3 Hz, 1 H), 4.70 (d, *J* = 11.1 Hz, 1 H), 4.41 (d, *J* = 2.8 Hz, 1 H), 4.12 (m, 1 H), 3.84 (s, 3 H), 3.74 (td, J_1 = 11.6 Hz, J_2 = 2.5 Hz, 1 H), 2.08 (m, 1 H), 1.85 (m, 1 H), 1.67 (tt, J_1 = 13.4 Hz, J_2 = 4.6 Hz, 1 H), 1.52 (m, 1 H), 1.35 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 159.6 (C), 145.0 (C), 134.5 (C), 131.2 (CH), 129.6 (CH), 129.1 (CH), 121.1 (C), 117.8 (CH), 114.5 (CH), 114.3 (CH), 75.0 (CH), 69.0 (CH_2), 55.6 (CH_3), 54.4 (CH), 39.2 (CH), 24.4 (CH_2), 22.2 (CH_2).

MS: *m/z* = 295 (M)$^+$, 264, 250, 236, 224, 193, 167, 132, 1121, 91, 77, 65.

rac-(4a*S*,5*S*,10b*S*)-5-(3-Methoxyphenyl)-3,4,4a,5,6,10b-hexahydro-2*H*-pyrano[3,2-*c*]quinoline (5n)

IR (film): 3370, 2940, 2853, 1608, 1488, 1265, 1154, 1071, 736 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.42 (dt, J_1 = 7.3 Hz, J_2 = 1.3 Hz, 1 H), 7.29 (dd, J_1 = 8.1 Hz, J_2 = 7.6 Hz, 1 H), 7.09 (dddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, J_4 = 0.7 Hz, 1 H), 6.99 (d, *J* = 7.6 Hz, 1 H), 6.98 (s, 1 H), 6.84 (ddd, J_1 = 8.2 Hz, J_2 = 2.5 Hz, J_3 = 1.0 Hz, 1 H), 6.79 (td, J_1 = 7.3 Hz, J_2 = 1.3 Hz, 1 H), 6.60 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 5.32 (d, *J* = 5.6 Hz, 1 H), 4.66 (d, *J* = 2.3 Hz, 1 H), 3.82 (s, 3 H), 3.58 (ddt, J_1 = 11.6 Hz, J_2 = 4.0 Hz, J_3 = 1.7 Hz, 1 H), 3.43 (td, J_1 = 11.6 Hz, J_2 = 2.5 Hz, 1 H), 2.17 (m, 1 H), 1.47–1.57 (m, 2 H), 1.43 (m, 1 H), 1.33 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 159.6 (C), 145.1 (C), 142.9 (C), 124.4 (CH), 128.8 (CH), 127.6 (CH), 119.9 (C), 119.12 (CH), 118.3 (CH), 114.4 (CH), 112.6 (CH), 112.5 (CH), 72.7 (CH), 60.6 (CH_2), 59.3 (CH_3), 55.3 (CH), 38.9 (CH), 25.4 (CH_2), 18.1 (CH_2).

MS: *m/z* = 295 (M)$^+$, 264,, 250, 236, 224, 193, 167, 132, 121, 91, 77, 65.

HRMS (ESI): *m/z* [M + H] calcd for $C_{19}H_{22}NO_2$: 296.1645; found: 296.1651.

rac-(4a*S*,5*R*,10b*S*)-5-(3-Methoxyphenyl)-3,4,4a,5,6,10b-hexahydro-2*H*-pyrano[3,2-*c*]quinoline (6n)

IR (film): 3373, 2939, 2837, 1609, 1487, 1253, 1155, 1039, 750 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.28 (t, *J* = 7.6 Hz, 1 H), 7.21 (dd, J_1 = 7.1 Hz, J_2 = 1.3 Hz, 1 H), 7.09 (ddd, J_1 = 7.8 Hz, J_2 = 7.1 Hz, J_3 = 1.5 Hz, 1 H), 7.00 (d, *J* = 7.1 Hz, 1 H), 6.98 (s, 1 H), 6.86 (ddd, J_1 = 7.6 Hz, J_2 = 2.5 Hz, J_3 = 0.6 Hz, 1 H), 6.71 (t, J_1 = 7.1 Hz, 1 H), 6.53 (d, *J* = 7.6 Hz, 1 H), 4.68 (d, *J* = 10.9 Hz, 1 H), 4.38 (d, *J* = 2.8 Hz, 1 H), 4.10 (ddt, J_1 = 11.1 Hz, J_2 = 4.5 Hz, J_3 = 2.3 Hz, 1 H), 3.80 (s, 3 H), 3.72 (td, J_1 = 11.1 Hz, J_2 = 2.3 Hz, 1 H), 2.08 (m, 1 H), 1.84 (tdt, J_1 = 13.6 Hz, J_2 = 11.1 Hz, J_3 = 4.5 Hz, 1 H), 1.65 (tt, J_1 = 13.6 Hz, J_2 = 4.5 Hz, 1 H), 1.50 (m, 1 H), 1.34 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 159.9 (C), 144.5 (C), 143.8 (C), 130.9 (CH), 129.6 (CH), 129.4 (CH), 120.7 (C), 120.3 (CH), 117.6 (CH), 114.3 (CH), 113.2 (CH), 74.5 (CH), 68.6 (CH_2), 55.2 (CH_3), 54.8 (CH), 38.8 (CH), 24.1 (CH_2), 22.0 (CH_2).

MS: m/z = 295 (M)$^+$, 264, 250, 236, 224, 210, 193, 167, 144, 130, 115, 92, 77, 65.

HRMS (ESI): m/z [M + H] calcd for $C_{19}H_{22}NO_2$: 296.1645; found: 296.1647.

rac-(4a*S*,5*S*,10b*S*)-5-(2-Methoxyphenyl)-3,4,4a,5,6,10b-hexahydro-2*H*-pyrano[3,2-*c*]quinoline (5o)[51]
IR (film): 3373, 2937, 2865, 1602, 1488, 1241, 1090, 753 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.55 (dd, J_1 = 7.6 Hz, J_2 = 1.5 Hz, 1 H), 7.42 (dt, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 7.28 (td. J_1 = 8.1 Hz, J_2 = 7.6 Hz, J_3 = 1.5 Hz, 1 H), 7.08 (dddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, J_4 = 0.8 Hz, 1 H), 6.98 (td, J_1 = 7.6 Hz, J_2 = 1.0 Hz, 1 H), 6.90 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 6.78 (td, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 6.60 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 5.32 (d, J = 5.6 Hz, 1 H), 5.03 (d, J = 2.3 Hz, 1 H), 3.82 (s, 3 H), 3.58 (m, 1 H), 3.43 (td, J_1 = 11.6 Hz, J_2 = 2.4 Hz, 1 H), 2.33 (m, 1 H), 1.48–1.58 (m, 3 H), 1.42 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 156.4 (C), 145.7 (C), 129.1 (C), 128.1 (CH), 127.9 (CH), 127.7 (CH), 127.1 (C), 120.2 (CH), 120.1 (CH), 118.0 (CH), 114.5 (CH), 110.3 (CH), 72.8 (CH), 60.7 (CH_2), 55.3 (CH_3), 52.6 (CH), 35.2 (CH), 25.6 (CH_2), 18.5 (CH_2).

MS: m/z = 295 (M)$^+$, 264, 250, 236, 224, 209, 188, 130, 115, 91, 77, 65.

rac-(4a*S*,5*R*,10b*S*)-5-(2-Methoxyphenyl)-3,4,4a,5,6,10b-hexahydro-2*H*-pyrano[3,2-*c*]quinoline (6o)[51]
IR (film): 3362, 2938, 2837, 1609, 1491, 1244, 1079, 755 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.44 (dd, H, J_1 = 7.6 Hz, J_2 = 1.5 Hz, 1 H), 7.25 (ddd, J_1 = 8.1 Hz, J_2 = 7.6 Hz, J_3 = 1.5 Hz, 1 H), 7.23 (dd, J_1 = 7.3 Hz, J_2 = 1.5 Hz, 1 H), 7.07 (ddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, 1 H), 6.96 (td, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1 H), 7.84 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 6.68 (td, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 6.51 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 5.19 (d, J = 10.1 Hz, 1 H), 4.41 (d, J = 3.0 Hz, 1 H), 4.01 (m, 1 H), 3.84 (s, 3 H), 3.68 (td, J_1 = 11.1 Hz, J_2 = 2.8 Hz, 1 H), 2.13 (m, 1 H), 1.92 (m, 1 H), 1.65 (tt, J_1 = 13.6 Hz, J_2 = 4.1 Hz, 1 H), 1.49 (dqd, J_1 = 13.6 Hz, J_2 = 4.1 Hz, J_3 = 1.3 Hz, 1 H), 1.35 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 157.5 (C), 145.1 (C), 130.9 (C), 130.5 (CH), 129.1 (CH), 128.4 (CH), 128.1 (CH), 120.9 (CH), 120.4 (C), 117.1 (CH), 114.0 (CH), 110.4 (CH), 74.1 (CH), 67.7 (CH_2), 53.3 (CH_3), 47.6 (CH), 38.2 (CH), 24.5 (CH_2), 22.7 (CH_2).

MS: m/z = 295 (M)$^+$, 264, 250, 236, 224, 209, 167, 130, 115, 91, 77, 65.

rac-(4a*S*,5*S*,10b*S*)-5-(3,4,5-Trimethoxyphenyl)-3,4,4a,5,6,10b-hexahydro-2*H*-pyrano[3,2-*c*]quinoline (5p)
IR (film): 3353, 2937, 2838, 1591, 1465, 1328, 1235, 1126, 1089, 735 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.43 (dt, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 7.10 (dddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, J_4 = 0.8 Hz, 1 H), 6.81 (td, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 6.64 (s, 2 H), 6.63 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 5.32 (d, J = 5.6 Hz, 1 H), 4.61 (d, J = 2.5 Hz, 1 H), 3.89 (s, 6 H), 3.86 (s, 3 H), 3.60 (ddt, J_1 = 11.4 Hz, J_2 = 4.0 Hz, J_3 = 2.0 Hz, 1 H), 3.43 (td, J_1 = 11.4 Hz, J_2 = 2.2 Hz, 1 H), 2.15 (m, 1 H), 1.45–1.59 (m, 3 H), 1.38 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 153.2 (C), 145.0 (C), 137.2 (C), 136.9 (C), 128.1 (CH), 127.7 (CH), 120.1 (C), 118.5 (CH), 114.5 (CH), 106.7 (CH), 103.7 (CH), 72.7 (CH), 60.9 (CH_3), 60.6 (CH_2), 59.6 (CH), 56.3 (CH_3), 56.2 (CH_3), 39.1 (CH), 25.4 (CH_2), 18.3 (CH_2).

MS: m/z = 355 (M)$^+$, 325, 311, 297, 279, 264, 188, 167, 91, 77, 51.

HRMS (ESI): m/z [M + H] calcd for $C_{21}H_{26}NO_4$: 356.1856; found: 296.1874.

rac-(4a*S*,5*R*,10b*S*)-5-(3,4,5-Trimethoxyphenyl)-3,4,4a,5,6,10b-hexahydro-2*H*-pyrano[3,2-*c*]quinoline (6p)
IR (film): 3353, 2938, 2838, 1593, 1495, 1238, 1126, 1083, 749 cm^{-1}.

^{1}H NMR (400 MHz, $CDCl_3$): 7.22 (dd, J_1 = 7.3 Hz, J_2 = 1.5 Hz, 1 H), 7.10 (ddd, J_1 = 8.1 Hz, J_2 = 7.3 Hz, J_3 = 1.5 Hz, 1 H), 6.71 (td, J_1 = 7.3 Hz, J_2 = 1.0 Hz, 1 H), 6.65 (s, 2 H), 6.54 (dd, J_1 = 8.1 Hz, J_2 = 1.0 Hz, 1 H), 4.64 (d, J = 10.9 Hz, 1 H), 4.38 (d, J = 2.8 Hz, 1 H), 4.11 (m, 1 H), 3.86 (s, 9 H), 3.73 (td, J_1 = 11.6 Hz, J_2 = 2.5 Hz, 1 H), 2.04 (m, 1 H), 1.85 (tdt, J_1 = 13.4 Hz, J_2 = 12.1 Hz, J_3 = 4.5 Hz, 1 H), 1.67 (tt, J_1 = 13.4 Hz, J_2 = 4.8 Hz, 1 H), 1.53 (m, 1 H), 1.36 (m, 1 H).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 153.7 (C), 145.0 (C), 138.4 (C), 137.8 (C), 131.4 (CH), 129.8 (CH), 121.1 (C), 118.0 (CH), 114.6 (CH), 104.9 (CH), 75.0 (CH), 69.0 (CH_2), 61.3 (CH_3), 56.6 (CH_3), 55.5 (CH), 39.4 (CH), 24.5 (CH_2), 22.4 (CH_2).

MS: m/z = 355 (M)$^+$, 325, 311, 297, 279, 264, 188, 167, 91, 77, 51.

HRMS (ESI): m/z [M + H] calcd for $C_{21}H_{26}NO_4$: 356.1856; found: 296.1852.

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