

SUPPORTING INFORMATION

Title: **Total Synthesis of Natural and Unnatural Lamellarins with Saturated and Unsaturated D-rings**

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Supporting Information

General Experimental Methods

Unless otherwise noted: Reactions were run in oven-dried round-bottomed flasks. Tetrahydrofuran (THF) was distilled from sodium benzophenone ketyl while dichloromethane (DCM) from calcium hydride prior to use. Acetonitrile (CH_3CN) was stored over calcium hydride. All other compounds were used as received from the suppliers. The crude reaction mixtures were concentrated under reduced pressure by removing organic solvents on rotary evaporator. Column chromatography was performed using silica gel 60 (particle size 0.06-0.2 mm; 70-230 mesh ASTM). Analytical thin-layer chromatography (TLC) was performed with silica gel 60 F₂₅₄ aluminum sheets. Chemical shifts for ¹H nuclear magnetic resonance (NMR) spectra were reported in parts per million (ppm, δ) downfield from tetramethylsilane. Splitting patterns are described as singlet (s), doublet (d), triplet (t), quartet (q), multiplet (m), broad (br), and doublet of doublet (dd). Resonances for infrared (IR) spectra were reported in wavenumbers (cm^{-1}). Low resolution (LRMS) mass spectra were obtained either using electron ionization (EI) or time-of-flight (TOF) while high resolution (HRMS) mass spectra were obtained using either fast-atom-bombardment (FAB) or time-of-flight (TOF). Melting points were uncorrected.

Experimental Section

Ethyl 3-(3,4-Dimethoxyphenyl)-2-nitroacrylate (70). The material was obtained as a 1:1 mixture of *Z*- and *E*-isomers. IR (neat): ν_{max} 2967, 2940, 2841, 1719, 1683, 1633, 1596, 1514, 1464, 1423, 1381, 1260, 1233, 1197, 1146, 1072, 1018 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 1.34-1.40 (m, 6H), 3.84 (s, 3H), 3.86 (s, 3H), 3.91 (s, 3H), 3.93 (s, 3H), 4.30-4.49 (m, 2H), 6.89-6.93 (d, $J = 8.0$ Hz, 4H), 7.04 (s, 2H), 7.17 (d, $J = 8.0$ Hz, 2H), 7.44 (s, 1H), 8.02 (s, 1H). ^{13}C NMR (50 MHz, CDCl_3): δ 13.8, 14.0, 55.9, 56.0, 56.1, 62.8, 62.9, 111.4, 111.6, 112.3, 121.6, 125.3, 126.4, 132.8, 136.8, 149.5, 152.8, 153.1. LRMS (EI) m/z (rel intensity) 282 ($M + \text{H}^+$, 43), 281 (M^+ , 100), 162 (34). TOF-HRMS calcd for $\text{C}_{13}\text{H}_{16}\text{NO}_6$ ($M + \text{H}^+$) 282.0972, found 282.0971.

A. Characterization of Mi-RC Products 74-76 and 80-92.

Ethyl 1,2-Bis(3,4-dimethoxyphenyl)-8,9-dimethoxypyrrolo[2,1-*a*]isoquinoline-3-carboxylate (74). IR (neat): ν_{max} 2935, 2835, 1674, 1609, 1582, 1531, 1497, 1464, 1378, 1351, 1258, 1222, 1184, 1161, 1133, 1103, 1068, 1027 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 1.02 (t, $J = 7.6$ Hz, 3H), 3.43 (s, 3H), 3.70 (s, 6H), 3.85 (s, 3H), 3.86 (s, 3H), 3.86 (s, 3H), 4.15 (q, $J = 7.4$ Hz, 2H), 6.73-6.78 (m, 4H), 6.86 (s, 2H), 6.96 (d, $J = 7.2$ Hz, 1H), 7.02 (s, 1H), 7.15 (s, 1H), 9.33 (d, $J = 7.2$ Hz, 1H). ^{13}C NMR (50 MHz, CDCl_3): δ 13.9, 55.2, 55.6, 55.7, 55.80, 55.82, 55.9, 105.2, 107.0, 109.8, 111.0, 112.0, 112.2, 114.1, 114.8, 118.3, 119.5, 123.2, 123.57, 123.62, 124.1, 128.0, 128.5, 130.2, 135.8, 147.4, 147.5, 147.9, 148.7, 149.2, 162.2. LRMS (EI) m/z (rel intensity) 572 ($M + \text{H}^+$, 33), 571 (M^+ , 100), 499 (22). TOF-HRMS calcd for $\text{C}_{33}\text{H}_{34}\text{NO}_8$ ($M + \text{H}^+$) 572.2279, found 572.2287.

Ethyl 1,2-Bis(3,4-dimethoxyphenyl)-8,9-dimethoxy-5,6-dihydropyrrolo[2,1-*a*]isoquinoline-3-carboxylate (75). IR (neat): ν_{max} 2936, 2835, 1683, 1608, 1583, 1477, 1464, 1417, 1320, 1229, 1178, 1132, 1065, 1025 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 0.98 (t, $J = 7.4$ Hz, 3H), 3.03 (apparent t, $J = 6.6$ Hz, 2H), 3.31 (s, 3H), 3.60 (s, 3H), 3.65 (s, 3H),

3.80 (s, 3H), 3.81 (s, 3H), 3.85 (s, 3H), 4.07 (q, $J = 7.4$ Hz, 2H), 4.59 (apparent t, $J = 6.6$ Hz, 2H), 6.64-6.72 (m, 8H). ^{13}C NMR (50 MHz, CDCl_3): δ 13.9, 29.1, 42.9, 55.2, 55.7, 55.9 (2 carbons), 55.91, 55.93, 59.7, 108.9, 110.3, 110.9, 111.3, 114.6, 114.7, 118.4, 121.0, 121.5, 123.2, 123.6, 126.0, 128.22, 128.24, 131.0, 132.7, 147.4, 147.6, 147.78, 147.82, 148.2, 148.8, 162.0. LRMS (EI) m/z (rel intensity) 574 ($M + \text{H}^+$, 33), 573 (M^+ , 100), 558 (14). TOF-HRMS calcd for $\text{C}_{33}\text{H}_{36}\text{NO}_8$ ($M + \text{H}^+$) 574.2435, found 574.2436.

Ethyl 1-(3,4-Dimethoxyphenyl)-2-(2,4-dibenzylxyloxy-5-methoxyphenyl)-7,8,9-trimethoxy-5,6-dihydropyrrolo[2,1-a]isoquinoline-3-carboxylate (80). IR (neat): ν_{\max} 2935, 2835, 1685, 1605, 1582, 1531, 1464, 1421, 1404, 1316, 1333, 1255, 1174, 1131, 1107, 1026 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 0.84 (t, $J = 7.4$ Hz, 3H), 3.09 (apparent br t, $J = 6.6$ Hz, 2H), 3.32 (s, 3H), 3.54 (s, 3H), 3.65 (s, 3H), 3.84 (s, 3H), 3.86 (s, 3H), 3.90 (s, 3H), 4.01 (q, $J = 7.4$ Hz, 2H), 4.61 (br m, 2H), 4.76 (s, 2H), 5.03 (s, 2H), 6.43 (s, 1H), 6.55 (s, 1H), 6.58 (s, 1H), 6.67 (s, 1H), 6.75 (s, 2H), 7.07-7.11 (m, 2H), 7.21-7.33 (m, 8H). ^{13}C NMR (50 MHz, CDCl_3): δ 13.8, 22.3, 42.6, 55.3, 55.8, 55.9, 56.6, 59.6, 61.0, 71.4, 71.8, 103.5, 105.2, 111.1, 114.4, 116.5, 118.9, 119.3, 119.5, 122.5, 123.3, 124.1, 126.8, 127.3, 127.4, 127.8, 128.3, 128.4, 130.4, 137.3, 137.9, 141.2, 143.8, 147.3, 147.8, 148.7, 150.4, 150.8, 151.6, 162.0. LRMS (EI) m/z (rel intensity) 786 ($M + \text{H}^+$, 50), 785 (M^+ , 89), 695 (47), 694 (70), 648 (30), 604 (39), 603 (37), 558 (26), 530 (37), 526 (28), 484 (21), 91 (100), 65 (27), 55 (28). TOF-HRMS calcd for $\text{C}_{47}\text{H}_{48}\text{NO}_{10}$ ($M + \text{H}^+$) 786.3273, found 786.3264.

Ethyl 1-(4-Benzylxyloxy-3-methoxyphenyl)-2-(2,4-dibenzylxyloxy-5-methoxyphenyl)-7,8,9-trimethoxy-5,6-dihydropyrrolo[2,1-a]isoquinoline-3-carboxylate (81). IR (neat): ν_{\max} 2936, 2830, 1737, 1686, 1631, 1606, 1581, 1531, 1509, 1464, 1405, 1383, 1333, 1316, 1259, 1214, 1156, 1132, 1108, 1024 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 0.83 (t, $J = 7.4$ Hz, 3H), 3.07 (br apparent t, $J = 6.6$ Hz, 2H), 3.19 (s, 3H), 3.54 (s, 3H), 3.62 (s, 3H), 3.84 (s, 3H), 3.88 (s, 3H), 4.00 (q, $J = 7.4$ Hz, 2H), 4.59 (br m, 2H), 4.73 (s, 2H), 5.03 (s, 2H), 5.13 (s, 2H),

6.43 (s, 1H), 6.50 (s, 1H), 6.55-6.58 (m, 1H), 6.65-6.72 (m, 3H), 7.06-7.10 (m, 2H), 7.20-7.38 (m, 13H). ^{13}C NMR (50 MHz, CDCl_3) δ 13.8, 22.3, 42.6, 55.3, 55.9, 56.7, 59.6, 60.9, 61.0, 71.0, 71.5, 71.7, 103.5, 105.2, 114.0, 114.9, 116.6, 119.0, 119.3, 119.5, 122.5, 123.2, 124.1, 126.8, 127.1, 127.3, 127.4, 127.8, 128.3, 128.5, 128.6, 130.4, 137.3, 137.9, 141.2, 143.8, 146.7, 147.3, 149.4, 150.3, 150.7, 151.6, 162.0. TOF-LRMS m/z (rel intensity) 862 ($\text{M} + \text{H}^+$, 100). TOF-HRMS calcd for $\text{C}_{53}\text{H}_{52}\text{NO}_{10}$ ($\text{M} + \text{H}^+$) 862.3586, found 862.3579.

Ethyl 1-(3-Benzylxy-4-methoxyphenyl)-2-(2,4-dibenzylxy-5-methoxyphenyl)-7,8,9-trimethoxy-5,6-dihdropyrrolo[2,1-*a*]isoquinoline-3-carboxylate (82). IR (CHCl_3): ν_{\max} 2935, 2836, 1735, 1686, 1605, 1581, 1531, 1454, 1425, 1404, 1383, 1333, 1316, 1256, 1208, 1176, 1131, 1107, 1022 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 0.76 (t, $J = 7.4$ Hz, 3H), 3.01 (br apparent t, $J = 7.6$ Hz, 2H), 3.20 (s, 3H), 3.56 (s, 3H), 3.75 (s, 3H), 3.79 (s, 3H), 3.83 (s, 3H), 3.93 (q, $J = 7.4$ Hz, 2H), 4.51 (br m, 2H), 4.65 (s, 2H), 4.70 (s, 2H), 4.94 (s, 2H), 6.37 (s, 1H), 6.38 (s, 1H), 6.49 (s, 1H), 6.69 (s, 3H), 6.99-7.02 (m, 2H), 7.13-7.25 (m, 13H). ^{13}C NMR (50 MHz, CDCl_3): δ 13.7, 22.2, 42.6, 55.1, 56.0, 56.4, 59.6, 60.9, 61.0, 70.9, 71.1, 71.7, 103.0, 105.0, 111.5, 116.0, 116.6, 118.6, 119.2, 119.4, 122.3, 123.8, 124.0, 126.7, 127.17, 127.21, 127.4, 127.70, 127.73, 128.2, 128.3, 128.4, 130.4, 136.9, 137.1, 137.8, 141.1, 143.5, 147.1, 147.8, 148.3, 150.2, 150.5, 151.4, 161.9. LRMS (EI) m/z (rel intensity) 862 ($\text{M} + \text{H}^+$, 7), 861 (M^+ , 11), 91 (100), 65 (29). TOF-HRMS calcd for $\text{C}_{53}\text{H}_{52}\text{NO}_{10}$ ($\text{M} + \text{H}^+$) 862.3586, found 862.3574.

Ethyl 1-(3,4-Dimethoxyphenyl)-2-(2,4-dibenzylxy-5-methoxyphenyl)-7-benzylxy-8,9-dimethoxy-5,6-dihdropyrrolo[2,1-*a*]isoquinoline-3-carboxylate (83). IR (neat): ν_{\max} 2935, 2835, 1686, 1604, 1583, 1531, 1489, 1464, 1414, 1380, 1333, 1315, 1255, 1213, 1174, 1155, 1131, 1106, 1026 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 0.82 (t, $J = 7.4$ Hz, 3H), 2.93 (br apparent t, $J = 6.6$ Hz, 2H), 3.34 (s, 3H), 3.54 (s, 3H), 3.64 (s, 3H), 3.84 (s, 3H), 3.89 (s, 3H), 3.99 (q, $J = 7.4$ Hz, 2H), 4.42 (br m, 2H), 4.75 (s, 2H), 5.02 (s, 2H), 5.09 (s, 2H), 6.43

(s, 1H), 6.56 (s, 2H), 6.66 (s, 1H), 6.75 (s, 2H), 7.06-7.10 (m, 2H), 7.21-7.42 (m, 13H). ^{13}C NMR (50 MHz, CDCl_3): δ 13.7, 22.6, 42.5, 55.3, 55.6, 55.8, 56.4, 59.6, 61.0, 71.1, 71.6, 75.3, 102.8, 105.2, 110.7, 114.0, 116.0, 118.6, 119.4, 119.9, 122.4, 123.1, 124.0, 126.7, 127.2, 127.4, 127.8, 128.1, 128.2, 128.5, 128.6, 130.3, 137.1, 137.3, 137.8, 141.2, 143.4, 147.0, 147.5, 148.4, 148.8, 150.5, 151.5, 161.9. LRMS (EI) m/z (rel intensity) 862 ($\text{M} + \text{H}^+$, 5), 861 (M^+ , 7), 91 (100), 65 (35). TOF-HRMS calcd for $\text{C}_{53}\text{H}_{52}\text{NO}_{10}$ ($\text{M} + \text{H}^+$) 862.3586, found 862.3583.

Ethyl 1-(4-Benzylxyloxy-3-methoxyphenyl)-2-(2,4-dibenzylxyloxy-5-methoxyphenyl)-7-benzylxyloxy-8,9-dimethoxy-5,6-dihydropyrrolo[2,1-*a*]isoquinoline-3-carboxylate (84). IR (neat): ν_{max} 2934, 1685, 1508, 1457, 1417, 1216, 1206 cm^{-1} . ^1H NMR (400 MHz, CDCl_3): δ 0.83 (t, $J = 7.1$ Hz, 3H), 2.91 (br s, 2H), 3.22 (s, 3H), 3.54 (s, 3H), 3.63 (s, 3H), 3.87 (s, 3H), 3.99 (br q, $J = 7.1$ Hz, 2H), 4.40 (br s, 2H), 4.73 (s, 2H), 5.03 (s, 2H), 5.08 (s, 2H), 5.13 (s, 2H), 6.43 (s, 1H), 6.53 (s, 1H), 6.56-6.76 (m, 4H), 7.07-7.44 (m, 20H). ^{13}C NMR (100 MHz, CDCl_3): δ 13.7, 22.6, 29.7, 42.5, 55.2, 55.8, 56.5, 59.6, 61.0, 70.7, 71.2, 71.4, 75.4, 103.0, 105.2, 113.5, 114.5, 116.1, 118.6, 119.4, 119.9, 122.4, 123.1, 126.7, 127.0, 127.2, 127.4, 127.77, 127.79, 128.2, 128.47, 128.5, 128.6, 128.8, 130.3, 137.1, 137.2, 137.3, 137.8, 141.2, 143.5, 146.5, 147.1, 148.8, 149.1, 150.6, 151.5, 161.9. LRMS (EI) m/z (rel intensity) 938 ($\text{M} + \text{H}^+$, 4), 573 (92), 91 (100), 42 (90). HR-MS (FAB) calcd for $\text{C}_{59}\text{H}_{56}\text{NO}_{10}$ ($\text{M} + \text{H}^+$) 938.3904, found 938.3905. These spectroscopic data are identical to those reported previously.¹

Ethyl 1-(3-Benzylxyloxy-4-methoxyphenyl)-2-(2,4-dibenzylxyloxy-5-methoxyphenyl)-7-benzylxyloxy-8,9-dimethoxy-5,6-dihydropyrrolo[2,1-*a*]isoquinoline-3-carboxylate (85). IR (CHCl_3): ν_{max} 2935, 2900, 2833, 1737, 1686, 1604, 1531, 1496, 1455, 1415, 1379, 1333, 1315, 1256, 1213, 1177, 1156, 1131, 1024 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 0.82 (t, $J = 7.4$ Hz, 3H), 2.92 (br m, 2H), 3.29 (s, 3H), 3.63 (s, 3H), 3.83 (s, 3H), 3.90 (s, 3H), 3.98 (q, J

= 7.4 Hz, 2H), 4.40 (br m, 2H), 4.72 (s, 2H), 4.77 (s, 2H), 5.01 (s, 2H), 5.09 (s, 2H), 6.43 (s, 1H), 6.48 (s, 1H), 6.54 (s, 1H), 6.75-6.77 (m, 3H), 7.04-7.09 (m, 2H), 7.20-7.48 (m, 18H).

¹³C NMR (50 MHz, CDCl₃): δ 13.7, 22.7, 42.6, 55.3, 56.2, 56.7, 59.6, 61.0, 71.1, 71.4, 71.8, 75.4, 103.5, 105.4, 111.9, 116.6, 117.1, 119.0, 119.5, 119.8, 122.4, 123.97, 124.04, 126.8, 127.25, 127.31, 127.4, 127.7, 128.3, 128.37, 128.43, 128.5, 128.6, 130.5, 137.1, 137.3, 137.5, 137.9, 141.4, 143.8, 147.4, 148.1, 148.6, 148.9, 150.8, 151.6, 161.9. LRMS (EI) *m/z* (rel intensity) 938 (M + H⁺, 3), 937 (M⁺, 2), 91 (100) 65 (15). TOF-HRMS calcd for C₅₉H₅₆NO₁₀ (M + H⁺) 938.3899, found 938.3900.

Ethyl 1-(3,4-Dimethoxyphenyl)-2-(2,4-dibenzylxyloxy-5-methoxyphenyl)-8-benzylxyloxy-9-methoxy-5,6-dihydropyrrolo[2,1-*a*]isoquinoline-3-carboxylate (87). IR (neat): ν_{max} 2933, 2900, 2833, 1683, 1608, 1583, 1531, 1516, 1497, 1480, 1454, 1421, 1400, 1380, 1333, 1251, 1210, 1171, 1127, 1060, 1024 cm⁻¹. ¹H NMR (400 MHz, CDCl₃): δ 0.84 (t, *J* = 7.2 Hz, 3H), 3.01 (apparent t, *J* = 6.6 Hz, 2H), 3.37 (s, 3H), 3.54 (s, 3H), 3.66 (s, 3H), 3.84 (s, 3H), 3.98-4.03 (m, 2H), 4.61 (br m, 2H), 4.76 (s, 2H), 5.03 (s, 2H), 5.15 (s, 2H), 6.44 (s, 1H), 6.60 (s, 1H), 6.68 (s, 1H), 6.72-6.78 (m, 3H), 7.08-7.10 (m, 2H), 7.21-7.40 (m, 12H), 7.44-7.46 (m, 2H). ¹³C NMR (100 MHz, CDCl₃): δ 13.7, 29.0, 42.8, 55.3, 55.6, 55.8, 56.5, 59.6, 71.0, 71.1, 71.6, 103.0, 109.2, 110.8, 113.2, 114.0, 116.1, 118.8, 119.2, 121.6, 121.8, 123.1, 125.7, 126.7, 127.2, 127.4, 127.8, 127.9, 128.2, 128.4, 128.55, 128.62, 130.8, 137.1, 137.8, 143.5, 147.0, 147.8, 148.4, 150.5, 162.0. LRMS (EI) *m/z* (rel intensity) 831 (M⁺, 5), 91 (100), 65 (29). TOF-HRMS calcd for C₅₂H₅₀NO₉ (M + H⁺) 832.3480, found 832.3485.

Ethyl 1-(3-Benzylxyloxy-4-methoxyphenyl)-2-(2,4-dibenzylxyloxy-5-methoxyphenyl)-8-benzylxyloxy-9-methoxy-5,6-dihydropyrrolo[2,1-*a*]isoquinoline-3-carboxylate (89). IR (neat): ν_{max} 2933, 1685, 1498, 1381, 1254, 1212, 1174, 1106 cm⁻¹. ¹H NMR (400 MHz, CDCl₃): δ 0.82 (t, *J* = 7.1 Hz, 3H), 2.99 (br s, 2H), 3.31 (s, 3H), 3.64 (s, 3H), 3.82 (s, 3H), 3.99 (br q, *J* = 7.1 Hz, 2H), 4.59 (br s, 2H), 4.72 (s, 2H), 4.77 (s, 2H), 5.01 (s, 2H), 5.14 (s,

2H), 6.44 (s, 1H), 6.57 (s, 1H), 6.65 (s, 1H), 6.74 (s, 1H), 6.76 (s, 1H), 7.06-7.46 (m, 20H). ^{13}C NMR (100 MHz, CDCl_3): δ 13.7, 29.6, 42.8, 55.2, 56.0, 56.5, 59.6, 70.8, 71.0, 71.2, 71.7, 103.1, 109.1, 111.5, 113.1, 116.1, 116.5, 118.8, 119.1, 121.5, 121.7, 123.7, 125.6, 126.8, 127.2, 127.3, 127.4, 127.7, 127.9, 128.2, 128.4, 128.5, 128.6, 130.9, 136.95, 137.01, 137.1, 137.9, 143.5, 147.1, 147.8, 147.9, 148.2, 150.6, 162.0. LRMS (EI) m/z (rel intensity) 908 (M + H^+ , 6), 615 (100), 573 (84), 84 (38). HR-MS (FAB) calcd for $\text{C}_{59}\text{H}_{56}\text{NO}_{10}$ (M + H^+) 908.3799, found 908.3791. These spectroscopic data are identical to those reported previously.¹

Ethyl 1-(3-Benzylxy-4-methoxyphenyl)-2-(2,5-dibenzylxy-4-methoxyphenyl)-8-benzylxy-9-methoxy-5,6-dihydropyrrolo[2,1-a]isoquinoline-3-carboxylate (90). IR (neat): ν_{max} 3031, 2934, 2830, 1683, 1608, 1582, 1532, 1497, 1454, 1404, 1380, 1333, 1251, 1173, 1129, 1060, 1021 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 0.87 (t, $J = 7.4$ Hz, 3H), 2.98 (apparent t, $J = 6.6$ Hz, 2H), 3.30 (s, 3H), 3.75 (s, 3H), 3.81 (s, 3H), 3.97 (q, $J = 7.4$ Hz, 2H), 4.59 (apparent t, $J = 6.6$ Hz, 2H), 4.75 (s, 2H), 4.81 (s, 2H), 4.89 (s, 2H), 5.15 (s, 2H), 6.43 (s, 1H), 6.62 (s, 1H), 6.66 (s, 1H), 6.72-6.74 (m, 4H), 7.12-7.15 (m, 2H), 7.20-7.47 (m, 18H). ^{13}C NMR (50 MHz, CDCl_3): δ 13.8, 29.0, 42.8, 55.2, 55.95, 56.04, 59.5, 70.8, 70.9, 71.8, 100.7, 103.2, 109.0, 111.4, 113.0, 116.4, 118.3, 118.7, 119.0, 121.5, 121.8, 123.7, 125.6, 126.8, 127.2, 127.4, 127.6, 127.7, 127.9, 128.3, 128.6, 134.2, 136.9, 137.0, 137.4, 137.9, 142.0, 147.1, 147.4, 147.7, 147.8, 148.1, 149.1, 151.3, 162.0. TOF-LRMS m/z (rel intensity) 908 (M + H^+ , 100). TOF-HRMS calcd for $\text{C}_{58}\text{H}_{54}\text{NO}_9$ (M + H^+) 908.3793, found 908.3795.

Ethyl 1-(4-Benzylxy-3-methoxyphenyl)-2-(2,4-dibenzylxy-5-methoxyphenyl)-8-benzylxy-9-methoxy-5,6-dihydropyrrolo[2,1-a]isoquinoline-3-carboxylate (91). IR (neat): ν_{max} 3032, 2932, 1733, 1683, 1607, 1584, 1497, 1480, 1454, 1400, 1381, 1334, 1250, 1210, 1173, 1128, 1060, 1024 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 0.83 (t, $J = 7.4$ Hz, 3H), 2.98 (apparent t, $J = 6.6$ Hz, 2H), 3.23 (s, 3H), 3.53 (s, 3H), 3.63 (s, 3H), 3.99 (q, $J = 7.4$ Hz,

2H), 4.60 (br m, 2H), 4.73 (s, 2H), 5.03 (s, 2H), 5.13 (s, 4H), 6.43 (s, 1H), 6.57 (s, 1H), 6.66 (s, 1H), 6.70-6.73 (m, 4H), 7.05-7.10 (m, 2H), 7.20-7.47 (m, 18H). ^{13}C NMR (50 MHz, CDCl_3): δ 13.8, 29.1, 42.8, 55.3, 55.9, 56.6, 59.5, 71.0, 71.2, 71.5, 71.8, 103.5, 109.4, 113.5, 113.8, 114.0, 115.0, 116.7, 119.3, 121.8, 123.2, 125.7, 126.8, 127.1, 127.3, 127.8, 127.9, 128.2, 128.5, 130.8, 137.1, 137.4, 137.9, 143.9, 146.6, 147.3, 148.1, 149.4, 150.7, 162.0. TOF-LRMS m/z (rel intensity) 908 ($M + \text{H}^+$, 100). TOF-HRMS calcd for $\text{C}_{58}\text{H}_{54}\text{NO}_9$ ($M + \text{H}^+$) 908.3793, found 908.3790.

Ethyl 1-(3-Benzylxy-4-methoxyphenyl)-2-(2,4-dibenzylxy-5-methoxyphenyl)-9-benzylxy-8-methoxy-5,6-dihydropyrrolo[2,1-*a*]isoquinoline-3-carboxylate (92). IR (CHCl_3): ν_{max} 2934, 2900, 1683, 1608, 1582, 1497, 1454, 1424, 1382, 1333, 1251, 1204, 1174, 1129, 1061, 1022 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 0.82 (t, $J = 7.0$ Hz, 3H), 3.04 (apparent t, $J = 6.6$ Hz, 2H), 3.63 (s, 3H), 3.84 (s, 3H), 3.90 (s, 3H), 3.98 (q, $J = 7.0$ Hz, 2H) 4.57 (br m, 2H), 4.57 (s, 2H), 4.69 (s, 2H), 4.74 (s, 2H), 5.01 (s, 2H), 6.42 (s, 1H), 6.55 (s, 1H), 6.69 (d, $J = 3.6$ Hz, 2H), 6.74 (s, 3H), 7.02-7.12 (m, 2H), 7.18-7.34 (m, 18H). ^{13}C NMR (50 MHz, CDCl_3): δ 13.9, 29.4, 42.9, 56.3, 56.8, 59.7, 70.5, 71.2, 71.6, 72.0, 103.7, 107.7, 111.3, 111.5, 112.0, 115.0, 116.7, 116.8, 117.1, 119.3, 121.3, 121.8, 123.4, 123.9, 126.4, 126.9, 127.1, 127.2, 127.3, 127.4, 127.7, 127.8, 127.9, 128.4, 128.5, 128.6, 131.0, 137.1, 137.3, 137.4, 138.1, 143.9, 144.0, 145.0, 146.8, 147.5, 148.3, 148.6, 148.8, 150.9, 162.1. LRMS (EI) m/z (rel intensity) 908 ($M + \text{H}^+$, 5), 907 (M^+ , 5), 532 (11), 531 (23), 440 (17), 412 (12), 295 (14), 181 (12), 91 (100), 65 (11). TOF-HRMS calcd for $\text{C}_{58}\text{H}_{54}\text{NO}_9$ ($M + \text{H}^+$) 908.3793, found 908.3793.

B. Characterization of Saturated Lamellarins 3, 79 and 93-104.

14-(4-Hydroxy-3-methoxyphenyl)-3-hydroxy-2,10,11,12-tetramethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (3; Lamellarin C). mp (MeOH) 234–238 °C (lit.² 225–230 °C). IR (neat): ν_{max} 3410 (br), 3000, 2939, 2838, 1701, 1599,

1546, 1508, 1456, 1414, 1271, 1246, 1203, 1146, 1119, 1086, 1036 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ 3.05 (apparent t, *J* = 6.0 Hz, 2H), 3.27 (s, 3H), 3.34 (s, 3H), 3.73 (s, 3H), 3.74 (s, 3H), 3.78 (s, 3H), 4.55–4.65 (m, 2H), 6.56 (s, 1H), 6.61 (s, 1H), 6.79 (s, 1H), 6.87 (d, *J* = 6.6 Hz, 1H), 6.99 (s, 1H), 7.03 (s, 1H), 9.31 (s, 1H), 9.71 (s, 1H). ¹³C NMR (50 MHz, DMSO-*d*₆): δ 21.9, 42.1, 55.3, 55.5, 56.5, 60.9, 61.2, 104.1, 105.6, 109.1, 113.2, 115.1, 116.1, 116.8, 120.4, 123.0, 123.8, 125.7, 128.1, 135.2, 142.4, 145.0, 146.1, 147.1, 147.4, 149.0, 150.7, 151.8, 154.7. LRMS (EI) m/z (rel intensity) 546 (M + H⁺, 33), 545 (M⁺, 100), 531 (10), 530 (26). TOF-HRMS calcd for C₃₀H₂₈NO₉ (M + H⁺) 546.1759, found 546.1769. These spectroscopic data are identical to those reported previously.²

14-(3,4-Dimethoxyphenyl)-3-hydroxy-2,10,11,12-tetramethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (93; Lamellarin I). mp (MeOH) 243–247 °C (lit.³ 218–220 °C). IR (neat): ν_{max} 3407 (br), 2993, 2936, 2837, 1705, 1598, 1544, 1507, 1455, 1414, 1336, 1315, 1280, 1242, 1170, 1085, 1028 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 3.13 (apparent t, *J* = 6.6 Hz, 2H), 3.34 (s, 3H), 3.47 (s, 3H), 3.86 (s, 3H), 3.87 (s, 3H), 3.89 (s, 3H), 3.96 (s, 3H), 4.75 (apparent t, *J* = 6.6 Hz, 2H), 5.77 (s, 1H), 6.56 (s, 1H), 6.58 (s, 1H), 6.96 (s, 1H), 7.05 (s, 1H), 7.08 (s, 2H). ¹³C NMR (50 MHz, CDCl₃): δ 21.9, 42.2, 55.2, 55.6, 56.2, 56.3, 60.9, 61.0, 103.5, 104.1, 105.2, 110.2, 112.1, 114.0, 114.3, 115.4, 120.1, 123.0, 123.6, 128.1, 128.3, 135.3, 142.4, 143.3, 145.6, 146.5, 149.0, 150.0, 150.7, 151.9, 155.6. LRMS (EI) m/z (rel intensity) 560 (M + H⁺, 46), 559 (M⁺, 65), 248 (100). TOF-HRMS calcd for C₃₁H₃₀NO₉ (M + H⁺) 560.1915, found 560.1925. These spectroscopic data are identical to those reported previously.³

14-(3-Hydroxy-4-methoxyphenyl)-3-hydroxy-2,10,11,12-tetramethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (94; Lamellarin T). mp (MeOH) >250 °C (lit.⁴ 214–218 °C). IR (neat): ν_{max} 3376 (br), 2937, 2839, 1681, 1598, 1547, 1507, 1454, 1413, 1273, 1242, 1201, 1145, 1086, 1037 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ

3.05 (apparent t, $J = 6.6$ Hz, 2H), 3.27 (s, 3H), 3.37 (s, 3H), 3.73 (s, 3H), 3.78 (s, 3H), 3.81 (s, 3H), 4.50-4.72 (m, 2H), 6.58 (s, 1H), 6.63 (s, 1H), 6.79 (s, 1H), 6.87 (s, 1H), 6.88 (d, $J = 6.6$ Hz, 1H), 7.15 (d, $J = 8.8$ Hz, 1H), 9.34 (s, 1H), 9.72 (s, 1H). ^{13}C NMR (50 MHz, DMSO- d_6): δ 21.9, 42.1, 55.3, 55.6, 56.6, 60.9, 61.2, 104.1, 105.6, 109.0, 113.3, 114.1, 115.7, 118.3, 120.5, 122.0, 122.9, 127.7, 135.1, 142.4, 145.0, 146.1, 147.4, 148.1, 148.2, 150.7, 151.8, 154.7. LRMS (EI) m/z (rel intensity) 546 ($M + H^+$, 22), 545 (M^+ , 45), 530 (39), 272 (58), 241 (100). TOF-HRMS calcd for $\text{C}_{30}\text{H}_{28}\text{NO}_9$ ($M + H^+$) 546.1759, found 546.1768. These spectroscopic data are identical to those reported previously.⁴

14-(3,4-Dimethoxyphenyl)-3,10-dihydroxy-2,11,12-trimethoxy-8,9-dihydro-6H-chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (95; Lamellarin F). mp (MeOH) >250 °C (lit.⁵ 273–276 °C). IR (neat): ν_{\max} 3409 (br), 3001, 2938, 2837, 1698, 1596, 1547, 1511, 1479, 1456, 1408, 1340, 1260, 1242, 1170, 1143, 1121, 1084, 1035 cm⁻¹. ^1H NMR (200 MHz, DMSO- d_6): δ 3.00 (apparent br t, $J = 6.6$ Hz, 2H), 3.24 (s, 3H), 3.31 (s, 3H), 3.64 (s, 3H), 3.73 (s, 3H), 3.81 (s, 3H), 4.46-4.74 (m, 2H), 6.31 (s, 1H), 6.51 (s, 1H), 6.79 (s, 1H), 7.00 (d, $J = 8.0$ Hz, 1H), 7.08 (s, 1H), 7.18 (d, $J = 8.0$ Hz, 1H), 9.26 (s, 1H), 9.71 (s, 1H). ^{13}C NMR (50 MHz, DMSO- d_6): δ 21.7, 42.1, 55.1, 55.5, 56.4, 60.7, 101.5, 104.1, 105.5, 109.1, 113.1, 113.6, 114.8, 115.6, 122.8, 123.6, 127.8, 128.0, 135.7, 137.0, 145.0, 146.1, 147.4, 147.8, 149.3, 150.2, 150.3, 151.3, 154.7. LRMS (EI) m/z (rel intensity) 546 ($M + H^+$, 37), 545 (M^+ , 100), 531 (18), 530 (44). TOF-HRMS calcd for $\text{C}_{30}\text{H}_{28}\text{NO}_9$ ($M + H^+$) 546.1759, found 546.1760. These spectroscopic data are identical to those reported previously.⁵

14-(4-Hydroxy-3-methoxyphenyl)-3,10-dihydroxy-2,11,12-trimethoxy-8,9-dihydro-6H-chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (96; Lamellarin K). mp (MeOH) >250 °C. IR (KBr): ν_{\max} 3404 (br), 2936, 1712, 1544, 1510, 1457, 1265, 1118 cm⁻¹. ^1H NMR (400 MHz, CDCl₃): δ 3.04 (m, 2H), 3.28 (s, 3H), 3.39 (s, 3H), 3.75 (s, 3H), 3.78 (s, 3H), 3.78 (s, 3H), 4.08 (br s, 3H), 4.55 (m, 1H), 4.73 (m, 1H), 6.33 (s, 1H), 6.55 (s, 1H), 6.79

(s, 1H), 6.91 (dd, $J = 8.0, 1.8$ Hz, 1H), 6.94 (d, $J = 1.8$ Hz, 1H), 7.00 (d, $J = 8.0$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ 21.4, 42.0, 54.9, 55.3, 56.0, 60.7, 101.5, 103.5, 104.5, 109.8, 113.3, 113.75, 113.84, 115.6, 115.7, 123.1, 123.9, 126.8, 128.7, 135.9, 136.1, 144.4, 145.9, 146.0, 146.1, 146.7, 148.0, 150.7, 156.2. LRMS (EI) m/z (rel intensity) 532 ($M + \text{H}^+$, 38), 531 (M^+ , 100), 516 (52), 484 (23). TOF-HRMS calcd for $\text{C}_{29}\text{H}_{26}\text{NO}_9$ ($M + \text{H}^+$) 532.1602, found 532.1607. These spectroscopic data are identical to those reported previously.³

14-(3-Hydroxy-4-methoxyphenyl)-3,10-dihydroxy-2,11,12-trimethoxy-8,9-dihydro-6H-chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (97; Lamellarin E). mp (MeOH) >250 °C (lit.⁵ 228–232 °C). IR (neat): ν_{max} 3377 (br), 3000, 2931, 2850, 1682, 1591, 1549, 1516, 1485, 1433, 1407, 1246, 1203, 1159, 1143, 1026 cm^{-1} . ^1H NMR (200 MHz, $\text{DMSO}-d_6$): δ 3.00 (apparent t, $J = 6.0$ Hz, 2H), 3.26 (s, 3H), 3.37 (s, 3H), 3.64 (s, 3H), 3.81 (s, 3H), 4.46–4.72 (m, 2H), 6.34 (s, 1H), 6.62 (s, 1H), 6.78 (s, 1H), 6.86 (s, 1H), 6.88 (d, $J = 6.6$ Hz, 1H), 7.14 (d, $J = 8.8$ Hz, 1H), 9.25 (s, 1H), 9.32 (s, 1H), 9.71 (s, 1H). ^{13}C NMR (50 MHz, $\text{DMSO}-d_6$): δ 21.7, 42.1, 55.1, 55.5, 56.6, 60.9, 101.5, 104.1, 105.6, 109.1, 113.1, 114.1, 114.8, 115.6, 118.3, 122.0, 122.8, 127.9, 135.6, 137.0, 144.9, 146.1, 147.3, 147.7, 148.0, 148.1, 151.2, 154.7. LRMS (EI) m/z (rel intensity) 532 ($M + \text{H}^+$, 58), 531 (M^+ , 100), 516 (32), 221 (30). TOF-HRMS calcd for $\text{C}_{29}\text{H}_{26}\text{NO}_9$ ($M + \text{H}^+$) 532.1602 found 532.1616. These spectroscopic data are identical to those reported previously.⁵

14-(3,4-Dimethoxyphenyl)-3,11-dihydroxy-2,12-dimethoxy-8,9-dihydro-6H-chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (99; Lamellarin J). mp (MeOH) >250 °C (lit.³ 216–220 °C). IR (neat): ν_{max} 3406 (br), 2993, 2937, 2836, 1704, 1598, 1543, 1507, 1455, 1414, 1336, 1315, 1260, 1242, 1200, 1170, 1143, 1118, 1085, 1028, 1003 cm^{-1} . ^1H NMR (200 MHz, $\text{DMSO}-d_6$): δ 3.00 (apparent br t, $J = 6.6$ Hz, 2H), 3.23 (s, 3H), 3.32 (s, 3H), 3.73 (s, 3H), 3.82 (s, 3H), 4.50–4.66 (m, 2H), 6.55 (s, 1H), 6.62 (s, 1H), 6.74 (s, 1H), 6.78 (s, 1H), 7.01 (d, $J = 8.0$ Hz, 1H), 7.08 (s, 1H), 7.19 (d, $J = 8.0$ Hz, 1H), 9.46 (s, 1H), 9.69

(s, 1H). ^{13}C NMR (50 MHz, DMSO- d_6): δ 27.9, 42.4, 55.2, 55.5, 56.36, 56.42, 104.0, 105.6, 109.1, 109.8, 112.8, 113.6, 114.3, 115.0, 115.8, 118.5, 123.7, 127.6, 127.8, 136.3, 144.9, 146.1, 146.4, 147.3, 147.5, 149.3, 150.2, 154.7. LRMS (EI) m/z (rel intensity) 516 ($M + H^+$, 11), 515 (M^+ , 16), 41 (100). TOF-HRMS calcd for $\text{C}_{29}\text{H}_{26}\text{NO}_8$ ($M + H^+$) 516.1653, found 516.1652. These spectroscopic data are identical to those reported previously.³

14-(3-Hydroxy-4-methoxyphenyl)-3,11-dihydroxy-2,12-dimethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (101; Lamellarin L). mp (MeOH) >250 °C (lit.³ 285–287 °C). IR (KBr): ν_{max} 3629 (br), 3473 (br), 3266 (br), 2957, 1672, 1589, 1485, 1421, 1278 cm⁻¹. ^1H NMR (400 MHz, CDCl₃): δ 3.05 (apparent t, $J = 6.5$ Hz, 2H), 3.41 (s, 3H), 3.52 (s, 3H), 3.96 (s, 3H), 4.64–4.80 (m, 2H), 6.72 (s, 1H), 6.77 (s, 1H), 6.89 (s, 1H), 6.98 (dd, $J = 8.2, 2.0$ Hz, 1H), 7.05 (d, $J = 8.2$ Hz, 1H), 7.08 (d, $J = 2.0$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl₃): δ 28.3, 42.3, 55.1, 55.4, 56.2, 103.4, 104.5, 108.8, 109.9, 111.7, 113.2, 114.4, 117.6, 119.3, 122.8, 127.3, 128.5, 136.4, 143.9, 145.4, 145.8, 145.9, 146.1, 146.6, 146.8, 156.0. LRMS (EI) m/z (rel intensity) 501 (M^+ , 1), 251 (51), 42 (100). TOF-HRMS calcd for $\text{C}_{28}\text{H}_{24}\text{NO}_8$ ($M + H^+$) 502.1496, found 502.1501. These spectroscopic data are identical to those reported previously.³

14-(3-Hydroxy-4-methoxyphenyl)-2,11-dihydroxy-3,12-dimethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (102; Lamellarin G). mp (MeOH) >250 °C (lit.⁵ 263–265°C). IR (neat): ν_{max} 3384 (br), 2923, 2851, 1686, 1586, 1509, 1486, 1424, 1273, 1206, 1160, 1100, 1027 cm⁻¹. ^1H NMR (200 MHz, DMSO- d_6): δ 2.98 (apparent br t, $J = 6.6$ Hz, 2H), 3.25 (s, 3H), 3.79 (s, 3H), 3.85 (s, 3H), 4.46–4.72 (m, 2H), 6.47 (s, 1H), 6.65 (s, 1H), 6.72 (s, 1H), 6.80 (s, 1H), 6.82 (d, $J = 8.0$ Hz, 1H), 7.00 (s, 1H), 7.12 (d, $J = 8.0$ Hz, 1H), 9.00 (s, 1H), 9.29 (s, 1H), 9.43 (s, 1H). ^{13}C NMR (50 MHz, DMSO- d_6): δ 27.9, 42.4, 55.1, 56.3 (2C), 101.2, 108.8, 109.7, 110.6, 113.0, 113.9, 114.8, 115.7, 118.2, 118.5, 121.9, 127.3, 127.6, 127.8, 136.4, 143.3, 145.1, 146.4, 147.4, 147.9, 148.1, 148.3, 154.7.

LRMS (EI) m/z (rel intensity) 502 ($M + H^+$, 64), 501 (M^+ , 100). TOF-HRMS calcd for $C_{28}H_{24}NO_8$ ($M + H^+$) 502.1496, found 502.1500. These spectroscopic data are identical to those reported previously.⁵

14-(4-Hydroxy-3-methoxyphenyl)-3,11-dihydroxy-2,12-dimethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (103; Lamellarin χ). mp (MeOH) >250 °C. IR (neat): ν_{max} 3384 (br), 2935, 2850, 1682, 1596, 1554, 1515, 1485, 1427, 1337, 1280, 1250, 1208, 1163, 1045 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ 3.00 (apparent br t, *J* = 6.6 Hz, 2H), 3.26 (s, 3H), 3.34 (s, 3H), 3.73 (s, 3H), 4.59 (apparent br t, *J* = 6.6 Hz, 2H), 6.59 (s, 1H), 6.67 (s, 1H), 6.73 (s, 1H), 6.78 (s, 1H), 6.88 (d, *J* = 8.0 Hz, 1H), 7.00 (d, *J* = 8.0 Hz, 1H), 7.02 (s, 1H), 9.28 (s, 1H), 9.46 (s, 1H), 9.69 (s, 1H). ¹³C NMR (50 MHz, DMSO-*d*₆): δ 28.0, 42.4, 55.2, 55.6, 56.5, 104.0, 105.8, 109.3, 109.9, 112.7, 114.7, 115.3, 115.8, 116.8, 118.6, 123.9, 126.0, 127.6, 128.1, 136.4, 144.9, 146.2, 146.5, 147.0, 147.3, 147.5, 149.0, 154.7. LRMS (EI) m/z (rel intensity) 502 ($M + H^+$, 15), 501 (M^+ , 43), 368 (72), 41 (100). TOF-HRMS calcd for $C_{28}H_{24}NO_8$ ($M + H^+$) 502.1496, found 502.1501.

14-(3-Hydroxy-4-methoxyphenyl)-3,12-dihydroxy-2,11-dimethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (104; Lamellarin Y). mp (MeOH) >250 °C. IR (neat): ν_{max} 3378 (br), 2925, 2851, 1682, 1587, 1552, 1513, 1488, 1462, 1422, 1359, 1340, 1320, 1274, 1248, 1207, 1161, 1044, 1017 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ 3.04 (apparent br t, *J* = 6.0 Hz, 2H), 3.37 (s, 3H), 3.78 (s, 3H), 3.84 (s, 3H), 4.60 (apparent br t, *J* = 6.0 Hz, 2H), 6.47 (s, 1H), 6.64 (s, 1H), 6.77 (s, 1H), 6.81 (s, 1H), 6.82 (d, *J* = 6.6 Hz, 1H), 6.93 (s, 1H), 7.12 (d, *J* = 8.8 Hz, 1H), 8.84 (br s, 1H), 9.28 (br s, 1H), 9.66 (br s, 1H). ¹³C NMR (50 MHz, DMSO-*d*₆): δ 28.4, 42.5, 55.5, 56.1, 56.3, 104.0, 105.4, 109.1, 112.4, 112.9, 113.3, 113.7, 114.9, 118.2, 120.1, 121.9, 126.0, 127.6, 128.1, 135.7, 144.9, 145.2, 146.1, 147.2, 147.8, 148.1, 148.5, 154.7. LRMS (EI) m/z (rel intensity) 502 ($M + H^+$,

30), 501 (M^+ , 100), 468 (31). TOF-HRMS calcd for $C_{28}H_{24}NO_8$ ($M + H^+$) 502.1496, found 502.1502.

C. Characterization of Saturated Lamellarin Acetates 106-118.

14-(3,4-Dimethoxyphenyl)-3-acetoxy-2,10,11,12-tetramethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (106; Lamellarin I acetate). mp (MeOH) 232–235 °C (lit.³ 240–241 °C). IR (neat): ν_{max} 2933, 2851, 1765, 1713, 1607, 1582, 1541, 1505, 1462, 1413, 1369, 1315, 1259, 1195, 1177, 1136, 1083, 1022 cm^{-1} . ¹H NMR (200 MHz, CDCl_3): δ 2.31 (s, 3H), 3.15 (apparent t, $J = 6.6$ Hz, 2H), 3.34 (s, 3H), 3.40 (s, 3H), 3.87 (s, 3H), 3.88 (s, 3H), 3.90 (s, 3H), 3.96 (s, 3H), 4.78 (apparent t, $J = 6.6$ Hz, 2H), 6.56 (s, 1H), 6.72 (s, 1H), 7.04 (s, 1H), 7.09 (s, 3H). ¹³C NMR (50 MHz, CDCl_3): δ 20.5, 21.9, 42.3, 55.2, 55.5, 56.2, 56.3, 60.9, 61.1, 105.3, 105.8, 111.9, 112.1, 114.0, 114.6, 116.1, 116.3, 120.1, 122.9, 123.6, 127.3, 127.9, 135.5, 139.0, 142.5, 145.1, 147.6, 149.1, 150.1, 150.7, 152.0, 155.1, 168.6, 168.7. LRMS (EI) m/z (rel intensity) 602 ($M + H^+$, 18), 601 (M^+ , 20), 559 (19), 544 (12), 43 (100). TOF-HRMS calcd for $C_{33}H_{32}NO_{10}$ ($M + H^+$) 602.2021, found 602.2023. These spectroscopic data are identical to those reported previously.³

14-(4-Acetoxy-3-methoxyphenyl)-3-acetoxy-2,10,11,12-tetramethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (107; Lamellarin C diacetate). mp (MeOH) >250 °C. IR (neat): ν_{max} 2925, 2853, 1765, 1710, 1595, 1542, 1507, 1475, 1414, 1273, 1257, 1194, 1138, 1117, 1084, 1033, 1007 cm^{-1} . ¹H NMR (200 MHz, CDCl_3): δ 2.29 (s, 3H), 2.34 (s, 3H), 3.14 (apparent t, $J = 6.6$ Hz, 2H), 3.37 (s, 3H), 3.42 (s, 3H), 3.81 (s, 3H), 3.86 (s, 3H), 3.89 (s, 3H), 4.64–4.89 (m, 2H), 6.53 (s, 1H), 6.68 (s, 1H), 7.07 (s, 1H), 7.10 (s, 1H), 7.14 (d, $J = 1.6$ Hz, 1H), 7.22 (d, $J = 8.0$ Hz, 1H). ¹³C NMR (50 MHz, CDCl_3): δ 20.5, 21.9, 42.3, 55.5, 55.7, 56.3, 60.9, 61.0, 105.2, 105.6, 111.9, 114.7, 115.0, 115.6, 116.1, 119.9, 122.6, 123.3, 123.9, 127.1, 134.3, 135.4, 139.0, 140.1, 142.5, 145.0, 147.7, 150.7, 152.2, 152.3, 155.1, 168.58, 168.63. LRMS (EI) m/z (rel intensity) 630 ($M + H^+$, 31),

629 (M^+ , 29), 588 (26), 587 (25), 545 (14), 531 (15), 530 (30), 43 (100). TOF-HRMS calcd for $C_{34}H_{32}NO_{11}$ ($M + H^+$) 630.1970, found 630.1969.

14-(3-Acetoxy-4-methoxyphenyl)-3-acetoxy-2,10,11,12-tetramethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (108; Lamellarin T diacetate). mp (MeOH) >250 °C. IR (neat): ν_{max} 2930, 2851, 1767, 1713, 1602, 1545, 1506, 1454, 1414, 1370, 1293, 1263, 1199, 1140, 1083, 1030, 1017 cm^{-1} . 1H NMR (200 MHz, $CDCl_3$): δ 2.30 (s, 6H), 3.01-3.25 (m, 2H), 3.39 (s, 3H), 3.45 (s, 3H), 3.86 (s, 3H), 3.88 (s, 3H), 3.89 (s, 3H), 4.53-4.67 (m, 1H), 4.84-4.97 (m, 1H), 6.48 (s, 1H), 6.69 (s, 1H), 7.06 (s, 1H), 7.14 (d, $J = 8.0$ Hz, 1H), 7.23 (d, $J = 2.2$ Hz, 1H), 7.32 (dd, $J = 8.0, 2.2$ Hz, 1H). ^{13}C NMR (50 MHz, $CDCl_3$): δ 20.5, 20.6, 21.8, 42.2, 55.2, 55.5, 56.2, 60.9, 61.0, 105.1, 105.4, 111.8, 113.0, 114.5, 114.8, 116.1, 119.9, 122.6, 125.4, 127.2, 127.7, 129.5, 135.6, 138.7, 140.7, 142.3, 144.8, 147.5, 150.5, 151.3, 151.9, 155.1, 168.5, 168.8. LRMS (EI) m/z (rel intensity) 630 ($M + H^+$, 15), 629 (M^+ , 42), 588 (35), 587 (100), 546 (7), 545 (20). TOF-HRMS calcd for $C_{34}H_{32}NO_{11}$ ($M + H^+$) 630.1970, found 630.1966.

14-(3,4-Dimethoxyphenyl)-3,10-diacetoxy-2,11,12-trimethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (109; Lamellarin F diacetate). mp (MeOH) >250 °C. IR (neat): ν_{max} 2993, 2934, 1765, 1711, 1542, 1510, 1478, 1420, 1370, 1290, 1233, 1193, 1137, 1077, 1024 cm^{-1} . 1H NMR (200 MHz, $CDCl_3$): δ 2.30 (s, 3H), 2.40 (s, 3H), 2.97 (apparent t, $J = 6.6$ Hz, 2H), 3.34 (s, 3H), 3.38 (s, 3H), 3.81 (s, 3H), 3.89 (s, 3H), 3.96 (s, 3H), 4.72-4.81 (m, 2H), 6.70 (s, 2H), 7.03 (s, 1H), 7.08 (s, 3H). ^{13}C NMR (50 MHz, $CDCl_3$): δ 20.4, 20.5, 22.3, 41.9, 55.3, 55.5, 56.26, 56.32, 60.8, 105.8, 107.8, 111.9, 112.3, 114.0, 114.8, 116.2, 116.3, 119.5, 122.9, 123.6, 127.3, 127.7, 135.0, 139.1, 141.3, 141.7, 145.1, 147.6, 149.2, 150.1, 151.7, 155.1, 168.7. LRMS (EI) m/z (rel intensity) 630 ($M + H^+$, 24), 629 (M^+ , 64), 588 (35), 587 (100), 546 (17), 545 (50), 530 (28), 43 (19). TOF-HRMS calcd for $C_{34}H_{32}NO_{11}$ ($M + H^+$) 630.1970, found 630.1966.

14-(4-Acetoxy-3-methoxyphenyl)-3,10-diacetoxy-2,11,12-trimethoxy-8,9-dihydro-6H-chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (110; Lamellarin K triacetate). mp (MeOH) >250 °C (lit.³ 275–277 °C). IR (KBr): ν_{max} 2941, 2847, 1769, 1714, 1543, 1510, 1478, 1421, 1370, 1275, 1197, 1139, 1078, 1038, 1010 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 2.30 (s, 3H), 2.35 (s, 3H), 2.39 (s, 3H), 2.97 (br apparent t, *J* = 6.6 Hz, 2H), 3.38 (s, 3H), 3.42 (s, 3H), 3.80 (s, 3H), 3.83 (s, 3H), 4.60–4.74 (m, 1H), 4.80–4.93 (m, 1H), 6.67 (s, 2H), 7.10 (d, *J* = 8.0 Hz, 1H), 7.11 (s, 1H), 7.15 (s, 1H), 7.23 (d, *J* = 8.0 Hz, 1H). ¹³C NMR (50 MHz, CDCl₃): δ 20.5, 20.6, 22.2, 41.8, 55.4, 55.6, 56.2, 60.8, 105.4, 107.4, 111.9, 114.6, 114.7, 115.7, 116.0, 119.1, 122.6, 123.2, 123.9, 127.1, 134.0, 134.8, 138.8, 139.9, 141.1, 141.6, 144.8, 147.6, 151.8, 152.2, 155.1, 168.7, 168.8. LRMS (EI) *m/z* (rel intensity) 658 (M + H⁺, 16), 657 (M⁺, 25), 616 (47), 615 (100), 574 (22), 573 (52), 558 (23), 531 (28), 522 (15), 516 (18), 86 (10). TOF-HRMS calcd for C₃₅H₃₂NO₁₂ (M + H⁺) 658.1919, found 658.1906. These spectroscopic data are identical to those reported previously.³

14-(3-Acetoxy-4-methoxyphenyl)-3,10-diacetoxy-2,11,12-trimethoxy-8,9-dihydro-6H-chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (111; Lamellarin E triacetate). mp (MeOH) >250 °C. IR (neat): ν_{max} 2926, 2853, 1766, 1715, 1617, 1546, 1510, 1476, 1446, 1421, 1370, 1292, 1265, 1194, 1139, 1114, 1077, 1038, 1017 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 2.31 (s, 6H), 2.40 (s, 3H), 2.97 (apparent t, *J* = 6.6 Hz, 2H), 3.40 (s, 3H), 3.46 (s, 3H), 3.81 (s, 3H), 3.90 (s, 3H), 4.55–4.69 (m, 1H), 4.84–4.98 (m, 1H), 6.63 (s, 1H), 6.71 (s, 1H), 7.08 (s, 1H), 7.16 (d, *J* = 8.0 Hz, 1H), 7.24 (d, *J* = 2.2 Hz, 1H), 7.34 (dd, *J* = 8.0, 2.2 Hz, 1H). ¹³C NMR (50 MHz, CDCl₃): δ 20.57, 20.64, 22.5, 42.0, 55.5, 55.8, 56.5, 60.9, 105.8, 107.9, 112.0, 113.4, 114.9, 115.3, 116.3, 119.4, 122.8, 125.7, 127.5, 127.9, 129.8, 135.3, 139.2, 141.1, 141.4, 141.8, 145.2, 147.9, 151.7, 152.0, 155.2, 168.6, 168.7, 168.8. LRMS (EI) *m/z* (rel intensity) 657 (M⁺, 2), 531 (6), 43 (100). TOF-HRMS calcd for C₃₅H₃₂NO₁₂ (M + H⁺) 658.1919, found 658.1923. Only the ¹H NMR (DMSO-*d*₆) was previously reported.⁵

14-(3,4-Dimethoxyphenyl)-3-acetoxy-2,11,12-trimethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (112; Dihydrolamellarin η acetate). mp (EtOAc/hexanes) >250 °C. IR (neat): ν_{max} 2993, 2930, 2850, 1764, 1706, 1610, 1582, 1543, 1515, 1484, 1463, 1438, 1413, 1343, 1319, 1259, 1197, 1164, 1136, 1100, 1025 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 2.30 (s, 3H), 3.12 (apparent t, *J* = 6.6 Hz, 2H), 3.35 (s, 3H), 3.39 (s, 3H), 3.86 (s, 3H), 3.89 (s, 3H), 3.95 (s, 3H), 4.72-4.85 (m, 2H), 6.69 (s, 1H), 6.75 (s, 2H), 7.03 (s, 1H), 7.08 (s, 3H). ¹³C NMR (50 MHz, CDCl₃): δ 20.5, 28.7, 42.5, 55.2, 55.6, 56.0, 56.2, 56.3, 105.8, 108.9, 111.2, 111.9, 112.2, 114.2, 114.4, 115.4, 116.4, 120.0, 123.7, 126.7, 127.4, 127.9, 136.0, 139.0, 145.2, 147.6, 147.7, 149.1, 149.2, 150.0, 155.2, 168.7. LRMS (EI) *m/z* (rel intensity) 572 (M + H⁺, 11), 571 (M⁺, 31), 530 (31), 529 (100). TOF-HRMS calcd for C₃₂H₃₀NO₉ (M + H⁺) 572.1915, found 572.1916.

14-(3,4-Dimethoxyphenyl)-3,11-diacetoxy-2,12-dimethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (113; Lamellarin J diacetate). mp (MeOH) >250 °C. IR (neat): ν_{max} 2923, 2853, 1763, 1713, 1546, 1513, 1463, 1371, 1256, 1196, 1025 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 2.31 (s, 6H), 3.11 (apparent t, *J* = 6.6 Hz, 2H), 3.31 (s, 3H), 3.40 (s, 3H), 3.88 (s, 3H), 3.96 (s, 3H), 4.82 (apparent t, *J* = 6.6 Hz, 2H), 6.73 (s, 1H), 6.81 (s, 1H), 6.95 (s, 1H), 7.04 (s, 1H), 7.09 (apparent d, *J* = 1.4 Hz, 3H). ¹³C NMR (50 MHz, CDCl₃): δ 20.5, 28.1, 42.5, 55.3, 55.6, 56.2, 56.3, 105.8, 109.9, 111.9, 112.3, 114.0, 114.9, 116.2, 116.5, 122.6, 123.5, 125.9, 126.2, 127.3, 127.7, 135.2, 139.1, 139.5, 145.1, 147.6, 149.2, 149.8, 150.1, 155.1, 168.6, 168.8. LRMS (EI) *m/z* (rel intensity) 599 (M⁺, 6), 516 (44), 515 (100). TOF-HRMS calcd for C₃₃H₃₀NO₁₀ (M + H⁺) 600.1864, found 600.1858.

14-(3-Acetoxy-4-methoxyphenyl)-3,11-diacetoxy-2,12-dimethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (115; Lamellarin L triacetate). mp (MeOH) >250 °C (lit.³ >300 °C). IR (neat): ν_{max} 3080, 2939, 2840, 1766, 1716, 1549, 1512,

1479, 1439, 1419, 1368, 1296, 1282, 1192, 1149, 1130, 1033, 1014 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 2.29 (s, 6H), 2.31 (s, 3H), 3.01-3.15 (m, 2H), 3.36 (s, 3H), 3.45 (s, 3H), 3.88 (s, 3H), 4.57-4.71 (m, 1H), 4.91-5.04 (m, 1H), 6.72 (s, 1H), 6.73 (s, 1H), 6.95 (s, 1H), 7.09 (s, 1H), 7.14 (d, *J* = 8.0 Hz, 1H), 7.24 (d, *J* = 2.2 Hz, 1H), 7.32 (dd, *J* = 8.0, 2.2 Hz, 1H). ¹³C NMR (50 MHz, CDCl₃): δ 20.6, 28.2, 42.5, 55.4, 55.7, 56.3, 105.6, 109.9, 111.9, 113.3, 115.3, 116.1, 122.6, 125.6, 125.7, 126.0, 127.3, 127.7, 129.6, 135.4, 139.0, 139.5, 140.9, 145.0, 147.7, 149.9, 151.5, 155.1, 168.4, 168.6, 168.7. LRMS (EI) *m/z* (rel intensity) 628 (M + H⁺, 72), 586 (100), 544 (45), 529 (26) 501 (8). TOF-HRMS calcd for C₃₄H₃₀NO₁₁ (M + H⁺) 628.1813, found 628.1819. These spectroscopic data are identical to those reported previously.³

14-(3-Acetoxy-4-methoxyphenyl)-2,11-diacetoxy-3,12-dimethoxy-8,9-dihydro-6H-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (116; Lamellarin G triacetate). mp (MeOH) 241–244 °C. IR (neat): ν_{max} 2957, 2854, 2924, 1766, 1719, 1647, 1602, 1465, 1410, 1377, 1260, 1183, 1131, 1037, 1012 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 2.27 (s, 3H), 2.30 (s, 3H), 2.31 (s, 3H), 2.98-3.20 (m, 2H), 3.36 (s, 3H), 3.84 (s, 3H), 3.91 (s, 3H), 4.47-4.61 (m, 1H), 4.95-5.05 (m, 1H), 6.66 (s, 1H), 6.93 (d, *J* = 3.6 Hz, 2H), 6.97 (s, 1H), 7.02-7.38 (m, 3H). ¹³C NMR (50 MHz, CDCl₃): δ 20.6, 28.2, 42.5, 55.4, 56.15, 56.24, 101.4, 110.1, 110.8, 113.5, 114.2, 115.2, 116.9, 122.5, 125.6, 125.7, 126.1, 127.2, 127.3, 129.5, 135.6, 136.3, 139.5, 140.8, 149.9, 150.1, 150.9, 151.5, 155.1, 168.5, 168.9, 169.0. TOF-LRMS *m/z* (rel intensity) 628 (M + H⁺, 100). TOF-HRMS calcd for C₃₄H₃₀NO₁₁ (M + H⁺) 628.1813, found 628.1808. Only the ¹H NMR (DMSO-*d*₆) was previously reported.⁵

14-(4-Acetoxy-3-methoxyphenyl)-3,11-diacetoxy-2,12-dimethoxy-8,9-dihydro-6H-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (117; Lamellarin χ triacetate). mp (MeOH) >250 °C. IR (neat): ν_{max} 2938, 2847, 1765, 1713, 1617, 1593, 1546, 1513, 1482, 1439, 1418, 1401, 1275, 1196, 1150, 1120, 1035, 1016 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ

2.31 (s, 3H), 2.32 (s, 3H), 2.35 (s, 3H), 3.12 (apparent t, $J = 6.6$ Hz, 2H), 3.36 (s, 3H), 3.43 (s, 3H), 3.82 (s, 3H), 4.69-4.96 (m, 2H), 6.70 (s, 1H), 6.80 (s, 1H), 6.96 (s, 1H), 7.09 (s, 1H), 7.11 (s, 1H), 7.16 (d, $J = 1.4$ Hz, 1H), 7.23 (d, $J = 8.0$ Hz, 1H). ^{13}C NMR (50 MHz, CDCl_3): δ 20.6, 28.1, 42.5, 55.5, 55.7, 56.2, 105.5, 109.8, 112.0, 114.8, 115.0, 115.9, 116.0, 122.6, 123.2, 123.9, 125.7, 125.9, 127.1, 134.0, 135.0, 139.0, 139.5, 140.1, 145.0, 147.8, 149.9, 152.3, 155.1, 168.6, 168.7, 168.9. LRMS (EI) m/z (rel intensity) 628 ($M + \text{H}^+$, 5), 627 (M^+ , 10), 585 (24), 543 (36), 501 (7), 111 (100). TOF-HRMS calcd for $\text{C}_{34}\text{H}_{30}\text{NO}_{11}$ ($M + \text{H}^+$) 628.1813, found 628.1824. Only the ^1H NMR (CDCl_3), IR, FABMS and HRFABMS were previously reported.⁶

14-(3-Acetoxy-4-methoxyphenyl)-3,12-diacetoxy-2,11-dimethoxy-8,9-dihydro-6H-chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (118; Lamellarin Y triacetate). mp (MeOH) >250 °C. IR (neat): ν_{max} 2939, 2843, 1764, 1710, 1616, 1548, 1515, 1457, 1415, 1370, 1266, 1199, 1138, 1039, 1010 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 2.24 (s, 3H), 2.31 (s, 6H), 3.10-3.20 (m, 2H), 3.47 (s, 3H), 3.84 (s, 3H), 3.92 (s, 3H), 4.60-4.74 (m, 1H), 4.89-5.02 (m, 1H), 6.72 (s, 1H), 6.74 (s, 1H), 6.87 (s, 1H), 7.09 (d, $J = 8.2$ Hz, 1H), 7.15 (s, 1H), 7.20 (d, $J = 2.2$ Hz, 1H), 7.28 (dd, $J = 8.2, 2.2$ Hz, 1H). ^{13}C NMR (50 MHz, CDCl_3): δ 20.7, 29.5, 42.3, 55.9, 56.1, 56.4, 105.9, 112.0, 112.4, 113.5, 114.7, 115.0, 116.3, 120.4, 125.6, 127.3, 127.5, 129.7, 133.0, 135.4, 138.7, 139.2, 140.9, 145.1, 147.9, 151.4, 151.7, 155.3, 168.4, 168.8, 169.0. LRMS (EI) m/z (rel intensity) 628 ($M + \text{H}^+$, 15), 627 (M^+ , 47), 586 (30), 585 (95), 551 (15), 544 (31), 543 (100), 502 (11), 501 (41). TOF-HRMS calcd for $\text{C}_{34}\text{H}_{30}\text{NO}_{11}$ ($M + \text{H}^+$) 628.1813, found 628.1803.

D. Characterization of the Unsaturated Lamellarin Acetates 119-132.

14-(3,4-Dimethoxyphenyl)-3-acetoxy-2,10,11,12-tetramethoxy-6H-chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (119; Lamellarin ζ acetate). mp (EtOAc/hexanes) 223–225 °C. IR (neat): ν_{max} 3087, 3000, 2937, 2836, 1771, 1706, 1605,

1538, 1479, 1415, 1393, 1332, 1282, 1257, 1198, 1157, 1137, 1120, 1097, 1075, 1043, 1023, 1003 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 2.32 (s, 3H), 3.41 (s, 3H), 3.43 (s, 3H), 3.89 (s, 3H), 3.94 (s, 3H), 3.99 (s, 3H), 4.03 (s, 3H), 6.82 (s, 1H), 6.97 (s, 1H), 7.12-7.18 (m, 3H), 7.43 (d, *J* = 7.6 Hz, 1H), 9.22 (d, *J* = 7.6 Hz, 1H). ¹³C NMR (50 MHz, CDCl₃): δ 20.5, 55.3, 55.6, 56.3, 56.4, 61.1, 61.6, 101.7, 106.5, 107.6, 112.2, 112.4, 112.5, 114.5, 116.0, 119.5, 121.4, 122.8, 124.0, 128.2, 128.4, 133.9, 139.8, 142.5, 145.7, 147.7, 148.6, 149.4, 150.3, 153.5, 155.1, 168.6. LRMS (EI) *m/z* (rel intensity) 600 (M + H⁺, 27), 599 (M⁺, 65), 558 (33), 557 (100). TOF-HRMS calcd for C₃₃H₃₀NO₁₀ (M + H⁺) 600.1864, found 600.1865.

14-(4-Acetoxy-3-methoxyphenyl)-3-acetoxy-2,10,11,12-tetramethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (120; Lamellarin B diacetate). mp (EtOAc/hexanes) >250 °C. IR (neat): ν_{max} 3120, 3073, 1765, 1708, 1603, 1539, 1479, 1415, 1394, 1370, 1332, 1275, 1255, 1195, 1156, 1138, 1118, 1095, 1073, 1042, 1007 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 2.33 (s, 3H), 2.38 (s, 3H), 3.46 (s, 3H), 3.49 (s, 3H), 3.84 (s, 3H), 3.95 (s, 3H), 4.04 (s, 3H), 6.82 (s, 1H), 6.95 (s, 1H), 7.15 (s, 1H), 7.21 (s, 1H), 7.23 (d, *J* = 8.0 Hz, 1H), 7.31 (d, *J* = 8.0 Hz, 1H), 7.46 (d, *J* = 7.4 Hz, 1H), 9.22 (d, *J* = 7.4 Hz, 1H). ¹³C NMR (50 MHz, CDCl₃): δ 20.5, 55.6, 55.8, 56.3, 61.1, 61.6, 101.6, 106.3, 107.7, 108.9, 111.9, 112.2, 115.4, 115.8, 119.3, 121.2, 122.7, 123.8, 124.0, 128.2, 133.7, 134.6, 139.8, 140.4, 142.5, 145.6, 147.8, 148.6, 152.4, 153.7, 155.0, 168.6. LRMS (EI) *m/z* (rel intensity) 628 (M + H⁺, 32), 627 (M⁺, 42), 544 (20), 543 (66), 43 (100). TOF-HRMS calcd for C₃₄H₃₀NO₁₁ (M + H⁺) 628.1813, found 628.1799. Only the ¹H NMR (CDCl₃), MS, and HRMS were previously reported.²

14-(3-Acetoxy-4-methoxyphenyl)-3-acetoxy-2,10,11,12-tetramethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (121; Lamellarin W diacetate). mp (EtOAc/hexanes) 215–220 °C. IR (neat): ν_{max} 3120, 2938, 2849, 1769, 1712, 1605, 1540, 1479, 1418, 1395, 1370, 1333, 1284, 1266, 1205, 1156, 1140, 1123, 1073, 1043, 1020 cm⁻¹.

¹H NMR (200 MHz, CDCl₃): δ 2.32 (s, 6H), 3.48 (s, 3H), 3.52 (s, 3H), 3.94 (s, 6H), 4.03 (s, 3H), 6.78 (s, 1H), 6.94 (s, 1H), 7.14 (s, 1H), 7.24 (d, *J* = 8.0 Hz, 1H), 7.32 (d, *J* = 2.2 Hz, 1H), 7.41–7.48 (m, 2H), 9.20 (d, *J* = 7.4 Hz, 1H). ¹³C NMR (50 MHz, CDCl₃): δ 20.5, 20.6, 55.3, 55.6, 56.4, 61.1, 61.7, 101.5, 106.2, 107.6, 108.7, 111.3, 112.1, 113.1, 115.8, 119.3, 121.2, 122.6, 126.0, 128.0, 128.4, 130.1, 133.9, 139.6, 140.9, 142.3, 145.4, 147.6, 148.4, 151.6, 153.5, 155.1, 168.5, 168.7. LRMS (EI) *m/z* (rel intensity) 627 (M⁺, 12), 369 (68), 368 (95), 43 (49), 41 (100). TOF-HRMS calcd for C₃₄H₃₀NO₁₁ (M + H⁺) 628.1813, found 628.1821.

14-(3,4-Dimethoxyphenyl)-3,10-diacetoxy-2,11,12-trimethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (122; Lamellarin ε diacetate). mp (EtOAc/hexanes) 238–241 °C. IR (neat): ν_{max} 3000, 2840, 2837, 1771, 1710, 1615, 1539, 1483, 1420, 1396, 1371, 1333, 1313, 1255, 1199, 1139, 1117, 1069, 1045, 1009 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ 2.24 (s, 3H), 2.45 (s, 3H, superimposed with H₂O in DMSO), 3.32 (s, 6H, superimposed with H₂O in DMSO), 3.75 (s, 6H), 3.84 (s, 3H), 6.78 (s, 1H), 7.13 (s, 1H), 7.18 (s, 1H), 7.24–7.35 (m, 3H), 7.31 (d, *J* = 7.2 Hz, 1H), 9.05 (d, *J* = 7.2 Hz, 1H). ¹³C NMR (50 MHz, DMSO-*d*₆): δ 20.2, 55.0, 55.2, 56.1, 60.5, 103.8, 106.0, 106.9, 108.1, 112.1, 113.1, 113.4, 114.4, 115.1, 117.8, 120.5, 122.7, 123.3, 126.4, 127.7, 132.6, 138.8, 139.5, 144.9, 147.4, 149.4, 150.2, 152.7, 153.9, 168.2, 168.6. LRMS (EI) *m/z* (rel intensity) 627 (M⁺, 12), 544 (15), 543 (10), 255 (100). TOF-HRMS calcd for C₃₄H₃₀NO₁₁ (M + H⁺) 628.1813, found 628.1802.

14-(4-Acetoxy-3-methoxyphenyl)-3,10-diacetoxy-2,11,12-trimethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (123; Lamellarin M triacetate). mp (EtOAc/hexanes) >250 °C (lit.³ 265–267 °C). IR (neat): ν_{max} 3007, 2922, 2851, 1766, 1709, 1538, 1481, 1463, 1420, 1397, 1370, 1276, 1257, 1243, 1194, 1139, 1116, 1068, 1045, 1009 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 2.33 (s, 3H), 2.38 (s, 3H), 2.50 (s, 3H), 3.46 (s, 3H),

3.49 (s, 3H), 3.85 (s, 3H), 3.90 (s, 3H), 6.81 (s, 1H), 7.08 (d, $J = 7.4$ Hz, 1H), 7.10 (s, 1H), 7.15 (s, 1H), 7.21 (s, 1H), 7.23 (d, $J = 8.0$ Hz, 1H), 7.32 (d, $J = 8.0$ Hz, 1H), 9.22 (d, $J = 7.4$ Hz, 1H). ^{13}C NMR (50 MHz, CDCl_3): δ 20.5, 55.6, 55.8, 56.3, 60.8, 104.2, 106.3, 106.6, 109.1, 112.2, 115.3, 115.7, 118.3, 121.0, 123.4, 123.7, 124.1, 128.4, 133.3, 134.3, 139.1, 140.0, 140.4, 142.0, 145.6, 147.9, 152.5, 153.3, 155.0, 168.6, 168.7, 168.8. LRMS (EI) m/z (rel intensity) 656 ($M + \text{H}^+$, 44), 655 (M^+ , 90), 614 (32), 613 (81), 572 (34), 571 (100), 529 (44), 91 (56). TOF-HRMS calcd for $\text{C}_{35}\text{H}_{30}\text{NO}_{12}$ ($M + \text{H}^+$) 656.1763, found 656.1759. These spectroscopic data are those reported previously.³

14-(3-Acetoxy-4-methoxyphenyl)-3,10-diacetoxy-2,11,12-trimethoxy-8,9-dihydro-6H-chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (124; Lamellarin X triacetate). mp (EtOAc/hexanes) 232–234 °C. IR (neat): ν_{max} 2925, 2851, 1767, 1709, 1617, 1539, 1481, 1421, 1396, 1369, 1333, 1285, 1266, 1192, 1140, 1067, 1044, 1009 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 2.32 (s, 6H), 2.49 (s, 3H), 3.47 (s, 3H), 3.51 (s, 3H), 3.88 (s, 3H), 3.93 (s, 3H), 6.77 (s, 1H), 7.07 (d, $J = 7.4$ Hz, 1H), 7.09 (s, 1H), 7.15 (s, 1H), 7.23 (d, $J = 8.0$ Hz, 1H), 7.30 (d, $J = 1.4$ Hz, 1H), 7.43 (dd, $J = 7.4, 1.4$ Hz, 1H), 9.23 (d, $J = 8.0$ Hz, 1H). ^{13}C NMR (50 MHz, CDCl_3): δ 20.5, 55.4, 55.7, 56.4, 60.8, 104.3, 106.4, 106.6, 109.1, 111.6, 112.2, 113.4, 115.8, 118.4, 121.1, 123.5, 126.1, 128.0, 128.7, 130.1, 133.6, 139.1, 140.0, 141.2, 142.0, 145.6, 147.8, 151.9, 153.2, 155.0, 168.48, 168.54, 168.6. TOF-LRMS m/z (rel intensity) 656 ($M + \text{H}^+$, 100). TOF-HRMS calcd for $\text{C}_{35}\text{H}_{30}\text{NO}_{12}$ ($M + \text{H}^+$) 656.1763, found 656.1760.

14-(3,4-Dimethoxyphenyl)-3-acetoxy-2,11,12-trimethoxy-6H-chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (125; Lamellarin η acetate). mp (EtOAc/hexanes) >250 °C. IR (neat): ν_{max} 3073, 3001, 2937, 2836, 1766, 1704, 1615, 1582, 1539, 1511, 1479, 1416, 1370, 1317, 1260, 1223, 1185, 1166, 1136, 1103, 1087, 1041, 1007 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 2.32 (s, 3H), 3.42 (s, 3H), 3.46 (s, 3H), 3.89 (s, 3H),

3.99 (s, 6H), 6.86 (s, 1H), 7.08 (d, $J = 7.2$ Hz, 1H), 7.10 (s, 1H), 7.14 (s, 3H), 7.17 (s, 1H), 7.20 (d, $J = 1.4$ Hz, 1H), 9.25 (d, $J = 7.2$ Hz, 1H). ^{13}C NMR (50 MHz, CDCl_3): δ 20.6, 55.3, 55.6, 56.0, 56.3, 56.4, 105.4, 106.6, 107.6, 108.4, 111.6, 112.2, 112.4, 112.8, 114.6, 116.1, 119.2, 123.3, 124.1, 124.9, 128.2, 128.5, 134.4, 139.8, 145.8, 147.6, 149.4, 149.5, 150.2, 150.4, 155.1, 168.6. LRMS (EI) m/z (rel intensity) 570 ($\text{M} + \text{H}^+$, 16), 569 (M^+ , 33), 528 (31), 527 (100). TOF-HRMS calcd for $\text{C}_{32}\text{H}_{28}\text{NO}_9$ ($\text{M} + \text{H}^+$) 570.1759, found 570.1769.

14-(3,4-Dimethoxyphenyl)-2,3,11,12-tetramethoxy-6H-

chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (126; Dehydrolamellarin G trimethyl ether). mp ($\text{CH}_2\text{Cl}_2/\text{MeOH}$) >250 °C. IR (neat): ν_{max} 3102, 2998, 2935, 2836, 1701, 1616, 1535, 1512, 1491, 1429, 1415, 1269, 1225, 1167, 1045, 1011 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 3.40 (s, 3H), 3.41 (s, 3H), 3.76 (s, 3H), 3.84 (s, 3H), 3.91 (s, 3H), 3.92 (s, 3H), 6.66 (s, 1H), 6.78 (s, 1H), 6.93 (d, $J = 8.0$ Hz, 1H), 6.99 (s, 1H), 7.06-7.19 (m, 4H), 9.09 (d, $J = 8.0$ Hz, 1H). ^{13}C NMR (50 MHz, CDCl_3): δ 55.2, 55.5, 55.95, 56.03, 56.2, 56.4, 100.6, 105.3, 105.4, 107.5, 107.9, 109.9, 110.9, 112.3, 114.8, 119.1, 123.3, 124.2, 124.8, 128.4, 129.3, 134.4, 145.6, 146.7, 149.3, 149.7, 150.1, 150.2, 155.4. LRMS (EI) m/z (rel intensity) 542 ($\text{M} + \text{H}^+$, 34), 541 (M^+ , 100). TOF-HRMS calcd for $\text{C}_{31}\text{H}_{28}\text{NO}_8$ ($\text{M} + \text{H}^+$) 542.1809, found 542.1819.

14-(3,4-Dimethoxyphenyl)-3,11-diacetoxy-2,12-dimethoxy-6H-

chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (127; Dehydrolamellarin J diacetate). mp (EtOAc/hexanes) >250 °C. IR (neat): ν_{max} 2921, 2851, 1759, 1706, 1623, 1582, 1541, 1508, 1487, 1465, 1431, 1396, 1332, 1314, 1278, 1255, 1225, 1152, 1123, 1090, 1045, 1027 cm^{-1} . ^1H NMR (200 MHz, CDCl_3): δ 2.31 (s, 3H), 2.33 (s, 3H), 3.41 (s, 6H), 3.90 (s, 3H), 3.98 (s, 3H), 6.82 (s, 1H), 7.03 (d, $J = 7.4$ Hz, 1H), 7.13-7.18 (m, 3H), 7.24 (s, 2H), 7.38 (s, 1H), 9.21 (d, $J = 7.4$ Hz, 1H). ^{13}C NMR (50 MHz, CDCl_3): δ 20.5, 55.3, 55.6, 56.3, 56.4, 106.5, 106.6, 109.1, 112.2, 112.4, 112.7, 112.9, 114.3, 115.9, 120.7, 123.3, 123.9,

124.0, 127.9, 128.4, 133.7, 139.9, 140.9, 145.7, 147.7, 149.4, 150.3, 150.9, 155.1, 168.6, 168.7. LRMS (EI) m/z (rel intensity) 598 (11), 597 (53), 556 (17), 555 (46), 514 (31), 513 (100). TOF-HRMS calcd for $C_{33}H_{28}NO_{10}$ ($M + H^+$) 598.1708, found 598.1712.

14-(3-Acetoxy-4-methoxyphenyl)-3,11-diacetoxy-2,12-dimethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (129; Lamellarin N triacetate). mp (EtOAc/hexanes) >250 °C (lit.³ 283–285 °C). IR (neat): ν_{max} 2922, 2852, 1759, 1709, 1542, 1486, 1462, 1433, 1369, 1297, 1277, 1257, 1187, 1149, 1125, 1035, 1017 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 2.31 (s, 6H), 2.34 (s, 3H), 3.48 (s, 6H), 3.93 (s, 3H), 6.78 (s, 1H), 7.05 (d, *J* = 7.4 Hz, 1H), 7.14 (s, 1H), 7.21 (s, 2H), 7.32 (s, 1H), 7.38–7.46 (m, 2H), 9.23 (d, *J* = 7.4 Hz, 1H). ¹³C NMR (50 MHz, CDCl₃): δ 20.5, 20.6, 55.4, 55.7, 56.4, 106.4, 106.5, 109.1, 111.7, 112.2, 112.8, 113.4, 115.8, 120.7, 123.2, 123.8, 123.9, 126.0, 127.9, 128.6, 130.1, 133.8, 139.9, 141.0, 141.2, 145.6, 147.8, 151.1, 151.9, 155.1, 168.5, 168.6, 168.8. LRMS (EI) m/z (rel intensity) 626 ($M + H^+$, 72), 585 (35), 584 (100), 542 (45), 527 (26). TOF-HRMS calcd for $C_{34}H_{28}NO_{11}$ ($M + H^+$) 626.1657, found 626.1655. These spectroscopic data are identical to those reported previously.³

14-(3-Acetoxy-4-methoxyphenyl)-2,11-diacetoxy-3,12-dimethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (130; Dehydrolamellarin G triacetate). mp (MeOH) >250 °C. IR (neat): ν_{max} 3000, 2924, 2851, 1764, 1683, 1628, 1557, 1509, 1420, 1370, 1276, 1210, 1193, 1156, 1129, 1084, 1024 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 2.21 (s, 3H), 2.25 (s, 3H), 2.26 (s, 3H), 3.42 (s, 3H), 3.78 (s, 3H), 3.89 (s, 3H), 6.84 (s, 1H), 6.91 (s, 1H), 6.93 (d, *J* = 7.4 Hz, 1H), 7.08 (s, 1H), 7.15 (d, *J* = 8.0 Hz, 1H), 7.20 (s, 1H), 7.22 (d, *J* = 2.2 Hz, 1H), 7.33 (dd, *J* = 8.0, 2.2 Hz, 1H), 9.11 (d, *J* = 7.4 Hz, 1H). ¹³C NMR (50 MHz, CDCl₃): δ 20.6, 55.5, 56.2, 56.3, 101.5, 106.6, 108.4, 110.4, 111.6, 112.6, 113.5, 117.5, 120.6, 123.1, 123.7, 124.0, 126.0, 127.5, 128.7, 130.0, 134.0, 136.4, 141.0, 150.6, 151.0, 151.7, 151.8, 155.0, 168.4, 168.7, 168.9. LRMS (EI) m/z (rel intensity)

625 (M^+ , 10), 500 (17), 499 (100). TOF-HRMS calcd for $C_{34}H_{28}NO_{11}$ ($M + H^+$) 626.1657, found 626.1657.

14-(4-Acetoxy-3-methoxyphenyl)-3,11-diacetoxy-2,12-dimethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (131; Lamellarin D triacetate). mp (EtOAc/hexanes) >250 °C (lit.³ 288–290 °C; lit.⁷ >300 °C). IR (neat): ν_{max} 2919, 2850, 1762, 1709, 1623, 1593, 1542, 1509, 1485, 1465, 1431, 1370, 1277, 1257, 1196, 1151, 1042, 1012 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 2.33 (s, 3H), 2.35 (s, 3H), 2.37 (s, 3H), 3.46 (s, 6H), 3.84 (s, 3H), 6.82 (s, 1H), 7.08 (d, J = 7.6 Hz, 1H), 7.19 (d, J = 8.0 Hz, 1H), 7.22 (s, 1H), 7.25–7.33 (m, 3H), 7.41 (s, 1H), 9.25 (d, J = 7.6 Hz, 1H). ¹³C NMR (50 MHz, CDCl₃): δ 20.6, 55.7, 55.8, 56.3, 106.3, 106.5, 109.2, 112.3, 112.8, 115.2, 115.7, 120.8, 123.2, 123.7, 123.9, 124.1, 128.2, 133.6, 134.3, 139.9, 140.5, 141.0, 145.6, 147.9, 151.2, 152.5, 155.1, 168.6. LRMS (EI) *m/z* (rel intensity) 626 ($M + H^+$, 11), 625 (M^+ , 13), 555 (13), 255 (31), 254 (37), 213 (48), 212 (100), 55 (40). TOF-HRMS calcd for $C_{34}H_{28}NO_{11}$ ($M + H^+$) 626.1657, found 626.1653. These spectroscopic data are identical to those reported previously.³

14-(3-Acetoxy-4-methoxyphenyl)-3,12-diacetoxy-2,11-dimethoxy-8,9-dihydro-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (132; Dehydrolamellarin Y triacetate). mp (EtOAc/hexanes) >250 °C. IR (neat): ν_{max} 3080, 3007, 2940, 2843, 1764, 1709, 1639, 1617, 1542, 1505, 1476, 1438, 1417, 1395, 1369, 1336, 1293, 1270, 1183, 1159, 1139, 1099, 1082, 1040, 1010 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 2.28 (s, 3H), 2.31 (s, 6H), 3.48 (s, 3H), 3.93 (s, 3H), 3.95 (s, 3H), 6.75 (s, 1H), 7.08 (d, J = 7.2 Hz, 1H), 7.14 (s, 1H), 7.17–7.19 (m, 1H), 7.23–7.26 (m, 2H), 7.31 (s, 1H), 7.37 (dd, J = 8.0, 1.6 Hz, 1H), 9.30 (d, J = 7.2 Hz, 1H). ¹³C NMR (50 MHz, CDCl₃): δ 20.5, 55.7, 56.0, 56.3, 106.4, 108.8, 111.3, 112.1, 112.7, 113.6, 115.8, 118.4, 118.7, 120.3, 124.7, 125.8, 127.3, 128.9, 129.4, 129.9, 134.0, 139.9, 140.3, 141.0, 145.6, 147.8, 151.9, 155.1, 168.4, 168.6, 168.8. LRMS

(EI) *m/z* (rel intensity) 626 ($M + H^+$, 19), 625 (M^+ , 58), 583 (59), 541 (100), 500 (11), 499 (36). TOF-HRMS calcd for $C_{34}H_{28}NO_{11}$ ($M + H^+$) 626.1657, found 626.1654.

E. Characterization of Unsaturated Lamellarins 4, and 133-144.

14-(4-Hydroxy-3-methoxyphenyl)-3,11-dihydroxy-2,12-dimethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (4; Lamellarin D). mp ($CH_2Cl_2/MeOH$) >250 °C (lit.⁷ >300 °C). IR (neat): ν_{max} 3388 (br), 2922, 2852, 1672, 1635, 1596, 1547, 1509, 1490, 1455, 1433, 1405, 1367, 1274, 1218, 1155, 1121, 1082, 1040, 1016 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ 3.38 (s, 6H), 3.75 (s, 3H), 6.69 (s, 1H), 6.85 (s, 1H), 6.99 (d, *J* = 8.8 Hz, 1H), 7.06 (s, 1H), 7.12-7.21 (m, 4H), 8.98 (d, *J* = 7.4 Hz, 1H), 8.37 (br s, 1H), 9.81 (br s, 2H). ¹³C NMR (50 MHz, DMSO-*d*₆): δ 55.0, 55.6, 56.5, 104.2, 105.9, 106.4, 106.8, 108.8, 111.2, 112.0, 112.8, 115.7, 116.9, 118.0, 122.4, 124.3, 125.1, 125.9, 129.4, 134.5, 145.0, 146.8, 147.3, 148.3, 148.8, 149.0, 149.1, 154.8. LRMS (EI) *m/z* (rel intensity) 500 ($M + H^+$, 33), 499 (M^+ , 100), 466 (14), 438 (16). TOF-HRMS calcd for $C_{28}H_{22}NO_8$ ($M + H^+$) 500.1340, found 500.1349. Only ¹H NMR (acetone-*d*₆ and DMSO-*d*₆), MS, and HRMS were reported previously.^{2,7}

14-(3,4-Dimethoxyphenyl)-3-hydroxy-2,10,11,12-tetramethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (133; Lamellarin ζ). mp ($CH_2Cl_2/hexanes$) >250 °C (lit.⁶ 268–272 °C). IR (neat): ν_{max} 2923, 2853, 1700, 1603, 1538, 1479, 1425, 1332, 1285, 1260, 1235, 1203, 1177, 1120, 1075, 1044 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 3.43 (s, 3H), 3.49 (s, 3H), 3.90 (s, 3H), 3.94 (s, 3H), 4.00 (s, 3H), 4.02 (s, 3H), 6.67 (s, 1H), 6.98 (d, *J* = 2.2 Hz, 2H), 7.15-7.20 (m, 3H), 7.38 (d, *J* = 7.6 Hz, 1H), 9.19 (d, *J* = 7.6 Hz, 1H). ¹³C NMR (50 MHz, CDCl₃): δ 55.4, 55.8, 56.4, 56.5, 61.3, 61.8, 101.8, 103.8, 104.9, 107.1, 110.0, 111.8, 112.4, 114.7, 119.7, 121.5, 123.1, 124.3, 128.7, 129.5, 134.0, 142.5, 143.5, 146.6, 147.3, 148.7, 149.4, 150.3, 153.5. LRMS (EI) *m/z* (rel intensity) 558 ($M + H^+$, 39), 557 (M^+ , 100), 542 (20). TOF-HRMS calcd for $C_{31}H_{28}NO_9$ ($M + H^+$)

558.1759, found 558.1770. These spectroscopic data are identical to those reported previously.⁶

14-(4-Hydroxy-3-methoxyphenyl)-3-hydroxy-2,10,11,12-tetramethoxy-6H-chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (134; Lamellarin B). mp (CH₂Cl₂/MeOH) >250 °C (lit.² 258–259 °C). IR (neat): ν_{max} 3311 (br), 2934, 2827, 1702, 1677, 1603, 1594, 1541, 1479, 1425, 1377, 1267, 1238, 1148, 1117, 1077, 1034, 1014 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ 3.35 (s, 3H), 3.38 (s, 3H, superimposed with H₂O in DMSO), 3.74 (s, 3H), 3.79 (s, 3H), 3.91 (s, 3H), 6.64 (s, 1H), 6.81 (s, 1H), 6.93 (dd, *J* = 8.0, 2.2 Hz, 1H), 6.97 (s, 1H), 7.03 (d, *J* = 8.0 Hz, 1H), 7.12 (d, *J* = 2.2 Hz, 1H), 7.33 (d, *J* = 7.2 Hz, 1H), 8.96 (d, *J* = 7.2 Hz, 1H), 9.37 (s, 1H), 9.86 (s, 1H). ¹³C NMR (50 MHz, DMSO-*d*₆): δ 54.8, 55.1, 56.0, 60.7, 61.5, 101.5, 103.7, 105.8, 106.7, 107.0, 108.1, 112.2, 115.1, 116.4, 118.4, 120.6, 122.0, 123.7, 125.2, 128.9, 133.0, 141.8, 144.6, 146.3, 146.9, 147.9, 148.0, 148.7, 153.0, 154.3. LRMS (EI) *m/z* (rel intensity) 544 (M + H⁺, 31), 543 (M⁺, 100), 528 (15), 497 (11). TOF-HRMS calcd for C₃₀H₂₆NO₉ (M + H⁺) 544.1602, found 544.1603. Only ¹H NMR (acetone-*d*₆), MS, and HRMS were reported previously.²

14-(3-Hydroxy-4-methoxyphenyl)-3-hydroxy-2,10,11,12-tetramethoxy-6H-chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (135; Lamellarin W). mp (CH₂Cl₂/MeOH) >250 °C (lit.⁴ 224–228 °C). IR (neat): ν_{max} 3414, 3139 (br), 2980, 2940, 2840, 1667, 1607, 1481, 1421, 1334, 1274, 1241, 1207, 1155, 1114, 1075, 1046, 1021 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ 3.36 (s, 6H), 3.81 (s, 3H), 3.85 (s, 3H), 3.93 (s, 3H), 6.72 (s, 1H), 6.86 (s, 1H), 6.99 (m, 2H), 7.03 (s, 1H), 7.21 (d, *J* = 8.0 Hz, 1H), 7.38 (d, *J* = 7.4 Hz, 1H), 9.02 (d, *J* = 7.4 Hz, 1H), 9.42 (s, 1H), 9.90 (s, 1H). ¹³C NMR (50 MHz, DMSO-*d*₆): δ 55.3, 55.5, 56.6, 61.2, 62.0, 101.9, 104.2, 106.2, 107.3, 107.6, 108.5, 112.4, 114.2, 118.6, 118.9, 121.0, 122.4, 122.6, 127.6, 129.2, 133.3, 142.3, 145.1, 146.7, 148.2, 148.4, 148.5, 153.5, 154.8. LRMS (EI) *m/z* (rel intensity) 544 (M + H⁺, 36), 543 (M⁺, 100), 528 (19).

TOF-HRMS calcd for C₃₀H₂₆NO₉ (M + H⁺) 544.1602, found 544.1615. These spectroscopic data are identical to those reported previously.⁴

14-(3,4-Dimethoxyphenyl)-3,10-dihydroxy-2,11,12-trimethoxy-6H-chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (136; Lamellarin ε). mp (CH₂Cl₂/hexanes) >250 °C (lit.⁸ 271–275 °C). IR (neat): ν_{max} 3400 (br), 3007, 2938, 2837, 1694, 1618, 1596, 1543, 1507, 1481, 1428, 1403, 1257, 1230, 1203, 1143, 1073, 1042 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ 3.37 (s, 6H), 3.71 (s, 3H), 3.75 (s, 3H), 3.85 (s, 3H), 6.62 (s, 1H), 6.72 (s, 1H), 6.86 (s, 1H), 7.12 (d, *J* = 8.0 Hz, 1H), 7.20 (s, 1H), 7.25 (d, *J* = 8.0 Hz, 1H), 7.45 (d, *J* = 7.6 Hz, 1H), 8.99 (d, *J* = 7.6 Hz, 1H), 9.90 (br s, 1H), 9.94 (br s, 1H). ¹³C NMR (50 MHz, DMSO-*d*₆): δ 55.1, 55.5, 56.4, 56.5, 60.9, 97.8, 104.2, 106.2, 107.4, 107.9, 108.6, 112.2, 113.7, 115.3, 121.2, 121.4, 124.1, 127.8, 129.2, 133.7, 136.4, 145.1, 146.1, 146.7, 148.3, 149.6, 150.4, 153.3, 154.8. LRMS (EI) *m/z* (rel intensity) 544 (M + H⁺, 33), 543 (M⁺, 100), 528 (11), 499 (11), 497 (19), 496 (15). TOF-HRMS calcd for C₃₀H₂₆NO₉ (M + H⁺) 544.1602, found 544.1593. Only ¹H NMR (CDCl₃), IR (KBr), FABMS, and HRFABMS were reported previously.⁸

14-(4-Hydroxy-3-methoxyphenyl)-3,10-dihydroxy-2,11,12-trimethoxy-6H-chromeno[4',3':4,5]pyrrolo[2,1-a]isoquinolin-6-one (137; Lamellarin M). mp (CH₂Cl₂/MeOH) >250 °C. IR (neat): ν_{max} 3314 (br), 3000, 2941, 2835, 1683, 1617, 1597, 1548, 1508, 1480, 1455, 1429, 1405, 1376, 1267, 1224, 1143, 1119, 1067, 1038 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ 3.38 (s, 6H), 3.71 (s, 3H), 3.75 (s, 3H), 6.66 (s, 1H), 6.77 (s, 1H), 6.86 (s, 1H), 6.97 (d, *J* = 8.0 Hz, 1H), 7.05 (s, 1H), 7.11 (d, *J* = 8.0 Hz, 1H), 7.44 (d, *J* = 7.4 Hz, 1H), 8.98 (d, *J* = 7.4 Hz, 1H). ¹³C NMR (50 MHz, DMSO-*d*₆): δ 55.1, 55.5, 56.5, 60.9, 97.9, 104.2, 106.3, 107.3, 107.9, 108.7, 112.5, 115.2, 115.6, 116.9, 121.3, 121.4, 124.2, 125.8, 129.3, 133.8, 136.3, 145.1, 146.0, 146.7, 147.3, 148.3, 149.1, 153.2, 154.8. LRMS (EI) *m/z* (rel intensity) 530 (M + H⁺, 33), 529 (M⁺, 100), 514 (11), 483 (15), 482 (23), 290

(10), 92 (25), 91 (83), 65 (17). TOF-HRMS calcd for $C_{29}H_{24}NO_9$ ($M + H^+$) 530.1446, found 530.1451. These spectroscopic data are identical to those reported previously.³

14-(3-Hydroxy-4-methoxyphenyl)-3,10-dihydroxy-2,11,12-trimethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (138; Lamellarin X). mp (CH₂Cl₂/MeOH) >250 °C. IR (neat): ν_{max} 3400 (br), 2956, 2844, 1707, 1623, 1593, 1583, 1546, 1508, 1482, 1454, 1431, 1407, 1377, 1358, 1334, 1276, 1213, 1233, 1144, 1130, 1068, 1034, 1013 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ 3.40 (s, 6H), 3.71 (s, 3H), 3.85 (s, 3H), 6.71 (s, 1H), 6.79 (s, 1H), 6.86 (s, 1H), 6.98 (br m, 2H), 7.20 (d, *J* = 8.0 Hz, 1H), 7.44 (d, *J* = 7.4 Hz, 1H), 8.98 (d, *J* = 7.4 Hz, 1H), 9.44 (br s, 1H), 9.88 (br s, 2H). ¹³C NMR (50 MHz, DMSO-*d*₆): δ 55.2, 55.6, 56.7, 61.0, 97.9, 104.2, 106.3, 107.4, 107.9, 108.6, 112.2, 114.2, 115.2, 118.7, 121.2, 121.4, 122.5, 127.9, 129.1, 133.6, 136.4, 145.1, 146.1, 146.7, 148.2, 148.3, 148.5, 153.3, 154.8. TOF-LRMS *m/z* (rel intensity) 530 ($M + H^+$, 100). TOF-HRMS calcd for $C_{29}H_{24}NO_9$ ($M + H^+$) 530.1446, found 530.1445. Only ¹H NMR (DMSO-*d*₆), FABMS, and HRFABMS were reported previously.⁴

14-(3,4-Dimethoxyphenyl)-3-hydroxy-2,11,12-trimethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (139; Lamellarin η). mp (CH₂Cl₂/MeOH) >250 °C (lit.⁶ 265-269 °C). IR (neat): ν_{max} 3281 (br), 3100, 2999, 2933, 2834, 1690, 1595, 1536, 1513, 1491, 1461, 1431, 1364, 1263, 1243, 1226, 1165, 1140, 1046, 1023 cm⁻¹. ¹H NMR (200 MHz, CDCl₃): δ 3.46 (s, 3H), 3.50 (s, 3H), 3.88 (s, 3H), 3.98 (s, 3H), 3.99 (s, 3H), 5.83 (br s, 1H), 6.70 (s, 1H), 7.00 (s, 1H), 7.04 (d, *J* = 7.4 Hz, 1H), 7.08 (s, 1H), 7.13-7.20 (m, 4H), 9.22 (d, *J* = 7.4 Hz, 1H). ¹³C NMR (50 MHz, CDCl₃): δ 55.2, 55.7, 56.0, 56.3, 56.4, 103.7, 104.8, 105.5, 107.6, 108.0, 109.9, 110.8, 112.3, 114.7, 119.2, 123.4, 124.2, 124.9, 128.6, 129.5, 134.4, 143.4, 146.5, 147.2, 149.4, 149.5, 150.1, 150.3, 155.5. LRMS (EI) *m/z* (rel intensity) 528 ($M + H^+$, 36), 527 (M^+ , 100). TOF-HRMS calcd for

$C_{30}H_{26}NO_8$ ($M + H^+$) 528.1653, found 528.1662. These spectroscopic data are identical to those reported previously.⁶

14-(3,4-Dimethoxyphenyl)-3,11-dihydroxy-2,12-dimethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (140; Dehydrolamellarin J).

mp (CH₂Cl₂/MeOH) >250 °C. IR (neat): ν_{max} 3451, 3278 (br), 2995, 2951, 2835, 1679, 1491, 1426, 1387, 1277, 1241, 1222, 1202, 1164, 1142, 1051, 1032, 1018 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ 3.34 (s, 6H, superimposed with H₂O in DMSO), 3.74 (s, 3H), 3.84 (s, 3H), 6.65 (s, 1H), 6.85 (s, 1H), 7.07 (s, 1H), 7.12-7.29 (m, 4H), 7.23 (d, *J* = 5.0 Hz, 1H), 8.99 (d, *J* = 5.0 Hz, 1H), 9.84 (s, 1H), 9.94 (s, 1H). ¹³C NMR (50 MHz, DMSO-*d*₆): δ 53.1, 53.7, 54.5, 54.6, 102.3, 103.9, 104.3, 105.0, 106.8, 109.0, 110.1, 111.0, 111.8, 111.9, 113.3, 113.4, 143.2, 144.9, 146.4, 146.9, 147.1, 147.7, 148.5, 152.9. TOF-LRMS *m/z* (rel intensity) 514 (M⁺, 100). TOF-HRMS calcd for C₂₉H₂₄NO₈ ($M + H^+$) 514.1496, found 514.1494.

14-(3-Hydroxy-4-methoxyphenyl)-3,11-dihydroxy-2,12-dimethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (142; Lamellarin N).

mp (CH₂Cl₂/MeOH) >250 °C (lit.³ 271–275 °C). IR (neat): ν_{max} 3361 (br), 2922, 2852, 1660, 1633, 1466, 1378, 1277, 1135, 1041, 1010 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ 3.41 (s, 6H, superimposed with H₂O in DMSO), 3.86 (s, 3H), 6.75 (s, 1H), 6.86 (s, 1H), 7.01 (m, 2H), 7.16-7.25 (m, 4H), 8.99 (d, *J* = 7.2 Hz, 1H), 9.44 (s, 1H), 9.89 (s, 1H), 10.0 (s, 1H). ¹³C NMR (50 MHz, DMSO-*d*₆): δ 54.7, 55.2, 56.3, 103.9, 105.5, 106.0, 106.6, 108.4, 110.6, 111.7, 112.5, 114.0, 117.6, 118.4, 122.1, 122.2, 124.8, 127.6, 128.9, 134.0, 144.7, 146.4, 148.0, 148.1, 148.4, 148.7, 154.4. LRMS (EI) *m/z* (rel intensity) 499 (M⁺, 8), 91 (100), 65 (20). TOF-HRMS calcd for C₂₈H₂₂NO₈ ($M + H^+$) 500.1340, found 500.1344. These spectroscopic data are identical to those reported previously.³

14-(3-Hydroxy-4-methoxyphenyl)-2,11-dihydroxy-3,12-dimethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (143; Dehydrolamellarin G).

mp

(CH₂Cl₂/hexanes) >250 °C. IR (neat): ν_{\max} 3250 (br), 2924, 2847, 1717, 1463, 1417, 1377, 1276, 1261, 1030 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ 3.34 (s, 3H), 3.81 (s, 3H), 3.86 (s, 3H), 6.74 (s, 1H), 6.92-6.95 (m, 3H), 7.06 (s, 1H), 7.17-7.21 (m, 3H), 9.00 (d, *J* = 7.4 Hz, 1H), 9.12 (s, 1H), 9.54 (s, 1H), 10.1 (br s, 1H). ¹³C NMR (50 MHz, DMSO-*d*₆): δ 55.0, 56.4 (2 carbons), 101.4, 105.9, 107.1, 109.5, 110.2, 111.5, 112.1, 113.0, 114.2, 117.9, 118.6, 122.3, 122.4, 125.1, 127.8, 128.7, 134.7, 143.5, 145.7, 148.3, 148.5, 148.8, 149.0, 149.3, 154.9. LRMS (EI) *m/z* (rel intensity) 500 (M + H⁺, 30), 499 (M⁺, 100). TOF-HRMS calcd for C₂₈H₂₂NO₈ (M + H⁺) 500.1340, found 500.1349.

14-(3-Hydroxy-4-methoxyphenyl)-3,12-dihydroxy-2,11-dimethoxy-6*H*-chromeno[4',3':4,5]pyrrolo[2,1-*a*]isoquinolin-6-one (144; Dehydrolamellarin Y). mp (CH₂Cl₂/MeOH) >250 °C. IR (neat): ν_{\max} 3479, 3420 (br), 2999, 2922, 2844, 1714, 1682, 1553, 1514, 1491, 1479, 1434, 1421, 1271, 1219, 1163, 1177, 1082, 1041, 1020 cm⁻¹. ¹H NMR (200 MHz, DMSO-*d*₆): δ 3.36 (s, 3H), 3.89 (s, 6H), 6.49 (s, 1H), 6.84 (s, 1H), 6.91-6.95 (m, 2H), 7.17 (s, 1H), 7.18-7.24 (m, 1H), 7.26 (d, *J* = 7.4 Hz, 1H), 7.38 (s, 1H), 9.01 (d, *J* = 7.4 Hz, 1H), 9.37 (s, 1H), 9.66 (br s, 1H), 9.82 (br s, 1H). ¹³C NMR (50 MHz, DMSO-*d*₆): δ 55.4, 56.1, 56.3, 104.1, 106.1, 107.0, 108.7, 108.9, 109.5, 111.5, 113.3, 114.0, 118.4, 119.3, 121.8, 122.2, 124.0, 127.6, 129.4, 133.7, 145.0, 146.7, 147.6, 148.1, 148.4, 150.0, 154.7. TOF-LRMS *m/z* (rel intensity) 500 (M + H⁺, 100). TOF-HRMS calcd for C₂₈H₂₂NO₈ (M + H⁺) 500.1340, found 500.1357.

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