Synthesis and Properties of the [n](2,5)Pyridinophane Ring System[1)]

Tomoshige KOBAYASHI and Makoto NITTA*
Department of Chemistry, School of Science and Engineering, Waseda University, Shinjuku-ku, Tokyo 160
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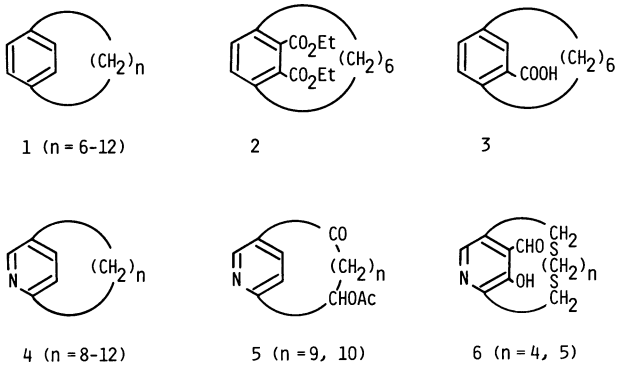
The synthesis and properties of a series of [n](2,5)pyridinophane derivatives (**13a—e**) (n=10, 8, 7, and 6) are described. These compounds are prepared in low yields by the [$Mo(CO)_6$]-induced cycloaddition reaction of 3-aryl-4,5-polymethylene-substituted isoxazoles (**11a—e**) with dimethyl acetylenedicarboxylate. The reaction pathways are suggested to involve the inclusion of the acetylenic ester across the C-4–C-5 bond of the N-complexed isoxazoles and the subsequent elimination of the oxygen atom from the resulting isoxazoline ring. The structures of **13a—e** are established on the basis of the ^{1}H-NMR, ^{13}C-NMR, and UV spectra. A significant strain for the pyridine ring in **13a—e** is reflected in the UV spectra. As the [n] decreases in **13—e**, there is a red shift in the absorption spectra. This feature is in good accordance with that observed in a series of [n]paracyclophane ring systems.

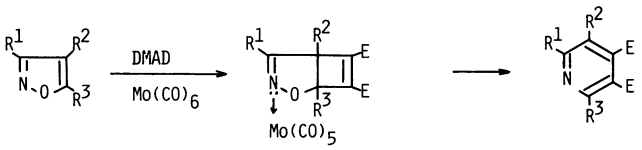
Much interest has been focussed on the chemistry of the cyclophanes.[2)] Especially, the [n]paracyclophane ring system (**1**)(n=6—12) has been one of the simplest models for clarifying the correlation of the aromaticity with the distortion of the benzene ring from planarity.[2)] The synthesis of "more bent" benzene ring than the previous example is though still an intriguing and challenging problem. The [n]paracyclophane ring system **1** has been synthesized for most values of [n], from 16 to 6.[2,3)] The [n]paracyclophane **1** (n=6), which has the smallest methylene chain known for this series, was first reported by Jones *et al.* in 1974.[3c)] However, it is only recently that new efficient routes to the [6]paracyclophane derivatives (**2**)[3g)] and (**3**),[3h)] both of which have substituents on the benzene ring, were developed. Their X-ray studies revealed that the benzene rings are highly deformed (*ca.* 20°) from planarity.[3g,h)]



1 (n = 6-12) 2 3

4 (n = 8-12) 5 (n = 9, 10) 6 (n = 4, 5)

In contrast to these studies, only a few examples of the aza analogue, the [n](2,5)pyridinophane ring system, have been known. The first example of [n](2,5)pyridinophane (**4**) (n=8—12) appeared in 1968.[4)] The functionalization of **4** (n=9 and 10) leading to **5** has also been studied.[5)] With a view toward a model for vitamin B_6-dependent enzyme, the chemistry of [n](2,5)pyridinophane (**6**)(n=4 and 5) containing pyridoxal has also been studied.[6)] Thus, the smallest value of [n] for [n](2,5)pyridinophane known so far is [8].[4)]



7 8 9

Scheme 1.

In our previous studies,[7)] [$Mo(CO)_6$] has been shown to effect a novel inclusion of the acetylenic ester across the C-4–C-5 bond of isoxazoles **7** and the subsequent elimination of an oxygen atom to give pyridine derivatives **9** *via* an intermediate **8** (Scheme 1). If this reaction is applicable to some 4,5-polymethylene-substituted isoxazoles (R^2-R^3=-$(CH_2)_n$- in **7**), [n](2,5)pyridinophanes can be expected as the products. We wish to describe here on the synthesis of [6](2,5)pyridinophane derivatives (**13d, e**), which might contain one of the most deformed pyridine rings, as well as a series of [10], [8], and [7] homologues, (**13a—c**).

Results and Discussion

The synthetic sequences of the [n](2,5)pyridinophane derivatives **13** are illustrated in Scheme 2. The syntheses of the desired 3-aryl-4,5-polymethylene-substituted isoxazoles (**12**) were achieved by method similar to that used in the preparation of ring-annelated isoxazoles.[8)] The 1,3-dipolar cycloaddition of benzonitrile oxide to 1-(1-pyrrolidinyl)-1-cycloalkenes (**10a—d**) afforded the corresponding cycloadducts, 4,5-polymethlene-subsituted 3-phenyl-5-(1-pyrrloidinyl)isoxazolines (**11a—d**). Similarly, the isoxazoline (**11e**) was obtained from the reaction of 4-chlorobenzonitrile oxide with **10d**. Compounds **11a, b** and **11d, e** were easily purified by recrystallization to give colorless crystals. On the other hand, oily **11c** decomposed on distillation [at *ca.* 120 °C (bath temp)/667 Pa]. Therefore, **11c** was purified by column chromatography on silica gel to give a

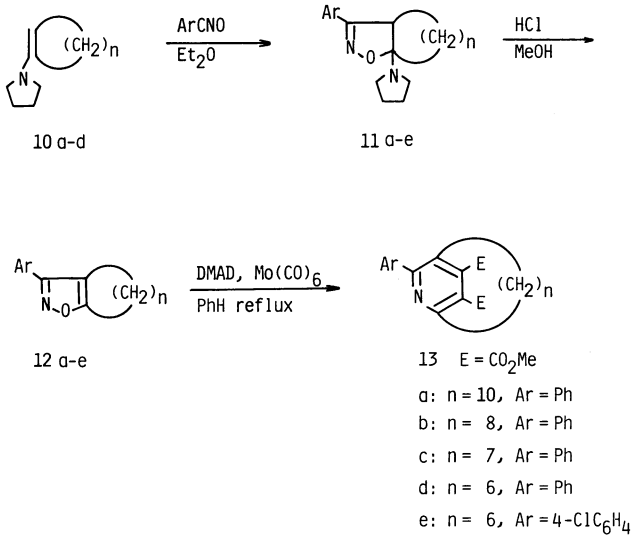
colorless oil and then submitted to the next step. On the acid-catalyzed elimination of the pyrrolidine, **11a—e** were converted to the desired 3-aryl-4,5-polymethylene-substituted isoxazoles (**12a—e**). The yields of the 2-isoxazolines **11a—e** and the isoxazoles **12a—e** are listed in Table 1. The structures of **11a—e** and the **12a—e** were unequivocally assigned on the basis of the physical data (see Experimental section). According to the procedure employed in our related studies,[7] the reaction of the isoxazoles **12** (1 mmol) with dimethyl acetylenedicarboxylate (DMAD, 2 mmol) in the presence of $[Mo(CO)_6]$ (1 mmol) afforded $[n]$(2,5)pyridinophane derivatives (**13a—e**). Although the yields of **13a—e** are only 5—11% (Table 1), no other product is isolated except for hexamethyl benzenehexacarboxylate (*ca.* a 10% yield based on the DMAD used), which results from the $[Mo(CO)_6]$-induced trimerization of DMAD.[7]

The formation of **13a—e** can be best explained by the reaction pathways shown in Scheme 3.[7] The initial stage of this reaction would be the coordination of $[Mo(CO)_5]$ on the nitrogen atom of the isoxazole **12**, leading to **14**. In **14**, there may be a delocalization of the d-electron from the molybdenum atom to the π^* (LUMO) of the isoxazole moiety.[9] In the presence of DMAD, the C-5-atom of **14** may connect with DMAD to give **15**, the following cyclization would afford the propellane-type[3] intermediate **16**. The facile N-O bond cleavage of the 2-isoxazoline ring has been shown previously.[7,10] Thus, the intermediate **16** undergoes the cleavage of both N-O and C-4-C-5 bonds to give the complexed vinylnitrene **17**.[11] The cyclization of **17** to lead to **18**[7,12] and the subsequent decomplexation eliminating CO_2 and $[Mo(CO)_4]$ result in the formation of the $[n]$(2,5)pyridinophane derivatives **13a—e**.

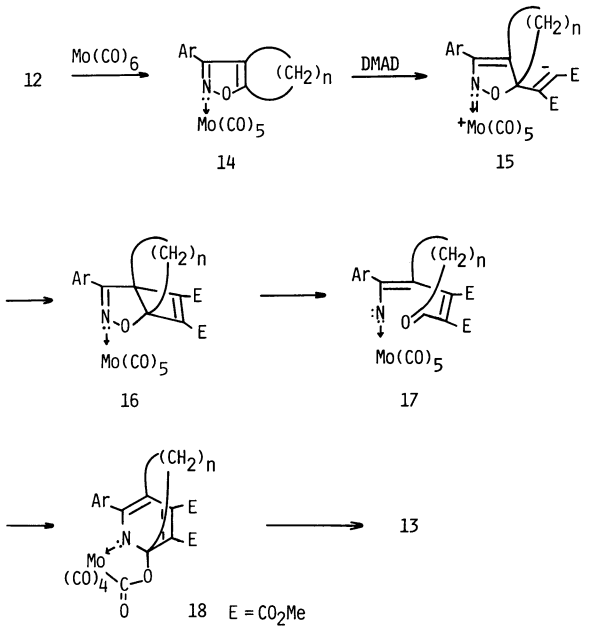
The structures of **13a—e** were determined on the basis of the elemental analyses and the mass, IR, ^{1}H-NMR, ^{13}C-NMR, and UV spectra (see Experimental section). Considering the ^{1}H-NMR spectra, several protons of the methylene chain appear at δ 0.60—0.95 (10H) for **13a**, at δ 0.10—2.10 (12H) for **13b**, at δ 0.20—2.20 (10H) for **13c**, at δ 0.45—0.98 (4H) for **13d**, and at δ 0.40—0.80 (4H) for **13e**. These protons located over the face of the corresponding pyridine ring. The shielding effect can be exactly attributed to the aromatic ring current of the pyridine ring. However, the degree of the shielding effect for the methylene chain in **13a** and **13b** seems to be smaller than that of $[n]$(2,5)pyridinopane **4**.[4] In compound **4**, the high-field signals appear at δ 0.2—2.2 (16H) for **4** (n=10) and at δ −0.40—1.85 (12H) for **4** (n=8). Thus, it can be suggested the two methoxycarbonyl groups on the pyridine ring of **13** reduce the ring current. Furthermore, **13d** exhibits a signal at δ 3.30 (1H, J_{jem}=12.0 Hz, J_{vic}=6.0 Hz). The signal could be assigned to the H_α proton (Fig. 1), which is located in the deshielding region of the phenyl group. A similar characteristic is observed in **13e**. This fact suggests that the [6](2,5)pyridinophane derivatives

TABLE 1. YIELDS OF ISOXAZOLINES **11**, ISOXAZOLES **12**, AND $[n]$(2,5)PYRIDINOPHANES **13**

			Yield/%		
	Ar	[n]	**11**	**12**	**13**
a	Ph	10	30	97	11
b	Ph	8	21	98	8
c	Ph	7	59	67	8
d	Ph	6	43	87	8
e	4-ClC_6H_4	6	56	57	5



Scheme 2.



Scheme 3.

Table 2. UV Data of the [n](2, 5)pyridinophanes **13a—d**, **4**, and the [n]paracyclophanes **1**

[n]	λ_{max} (log ε) (nm)									
	13[a]			**4**[b]			**1**[c]			
—	**19**:[d]	—	252, (4.33)	305 (4.24)	—	—	**20**:[c]	—	214,	265
[10]	**13a**:	—	253, (4.01)	302 (4.03)	216, (3.83)	271 (3.53)		—	223,	268
[9]		—	—	—	218, (3.79)	273 (3.48)		—	224,	271
[8]	**13b**:	—	266, (4.02)	313 (3.93)	223, (3.79)	278 (3.49)		—	230,[e]	276[e]
[7]	**13c**:	230, (4.12)	273, (3.97)	322 (3.87)	—	—		216,	245,	283
[6]	**13d**:	249, (4.12)	285, (3.90)	342 (3.80)	—	—		212,	253,	296

a) This work; b) Ref. (4); c) Ref. (3c); d) Ref. (7); e) Ref. (3d).

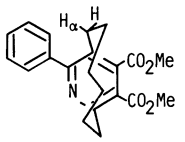


Fig. 1.

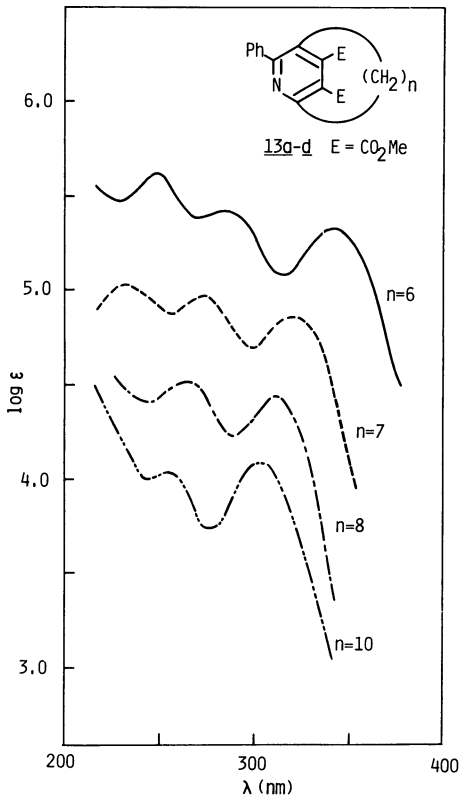


Fig. 2. UV spectra of the [n](2,5)pyridinophanes **13a—d** in absolute ethanol.
The curves are displayed upward successively by 0.5 log unit from the curve immediately below.

13d, **e** have rather rigid structures. The ^{13}C-NMR of the **13a—e** series are similar to each other, and the ring-current effect for the methylene chain seems to be negligibly small. The ring-current effect for the chemical shift of ^{13}C-NMR is not a predominant factor and often overwhelmed by a steric compression.[13]

The UV spectra of **13a—d** in ethanol are depicted in Fig. 2. The absorption maxima of [n](2,5)pyridinophane **13a—d**, **4**, and a series of [n]paracyclophane **1** along with the model compounds, 2-methyl-3,4-bis(methoxycarbonyl)-6-phenylpyridine (**19**)[7] and 1,4-diethylbenzene (**20**),[3c] are also summarized in Table 2. The significant ring strain of the [n](2,5)pyridinophane derivatives **13a—d** is reflected in the UV spectra. An increase in the red shift is observed for **13—d** as the value of [n] decreases. Since the absorption maximum of **13a** is very similar to that of **19**, **13a** seems to contain a planar and less-constrained pyridine ring. In contrast, the longest absorption maxima of **13d** and **13e**, both of which have the smallest value of [n] in the **13** series are exactly shifted to wavelength longer by 40 and 43 nm respectively as compared to **13a**. These features are in good agreement with those observed in the series of the [n]paracyclophanes **1**[3c,d] and the [n](2,5)pyridinophanes **4**.[4] Therefore, these can be no doubt that the [n](2,5)pyridinophanes **13d** and **13e** contain one of the most deformed pyridine ring thus far obtained.

Experimental

The IR spectra were recorded on a Shimadzu IR-400 spectrometer. The ^{1}H-NMR spectra were recorded on a Hitachi R-24 (60 MHz) spectrometer, and the chemical shifts are given in ppm (δ) relative to an internal $SiMe_4$ standard. The ^{13}C-NMR spectra were recorded on a JMN-FX90Q (22.5 MHz) spectrometer. The mass-spectral studies were conducted on a Shimadzu GCMS QP-1000 spectrom-

eter at 70 eV. All of the melting points are uncorrected. The solvents were dried and purified by the standard methods. The enamines, 1-(1-pyrrolidinyl)cyclododecene (**10a**),[14] 1-pyrrolidino-1-cyclodecene (**10b**),[14] 1-pyrrolidino-1-cyclononene (**10c**),[15] and 1-pyrrolidino-1-cyclooctene (**10d**),[15] were prepared by the method described in the literature. Benzohydroximoyl chloride and 4-chlorobenzohydroximoyl chloride were also obtained by the standard method.[16]

General Procedure for the Synthesis of 2-Isoxazolines ***11a—e.*** A solution of enamine **10** (30 mmol) and triethylamine (5 cm³) in ether (200 cm³) was cooled at 0 °C. To this solution we then slowly added a solution of benzohydroximoyl chloride (30 mmol) in ether (30 cm³). The reaction mixture was stirred at 0 °C for 6 h, and then it was poured into water (100 cm³). The ether layer was separated, washed with water, and dried over $MgSO_4$. The ether was removed *in vacuo* to give the 2-isoxazolines **11a—e**.

Compounds **11a**, **b**, **d**, and **11e** were purified by recrystallization to give colorless crystals. Compound **11c** was obtained as a colorless oil and decomposed on distillation [*ca.* 120 °C (bath temp)/667 Pa]. Therefore, **11c** was purified by column chromatography on silica gel using benzene as the eluent and submitted to the next step. The structures of **11a—e** were determined on the basis of the following physical data. For **11a**: mp 78—79 °C (from hexane); IR ($CHCl_3$), 2920, 1451, 1343, 1130 cm⁻¹; ¹H-NMR ($CDCl_3$) δ=1.20—2.45 (24H, m), 2.40—3.05 (4H, m), 3.10—3.55 (1H, m), 7.20—7.70 (5H, m). Found: C, 77.98; H, 9.75; N, 7.72%. Calcd for $C_{23}H_{34}N_2O$: C, 77.92; H, 9.67; N, 7.90%. For **11b**: mp 115—116 °C (from hexane); IR ($CHCl_3$), 2923, 1605, 1574, 1492, 1453, 1358, 1134 cm⁻¹; ¹H-NMR ($CDCl_3$) δ=1.10—2.50 (20H, m), 2.50—3.10 (4H, m), 3.12—3.41 (1H, m), 7.18—7.70 (5H, m). Found: C, 77.05; H, 8.99; N, 8.52%. Calcd for $C_{21}H_{30}N_2O$: C, 77.25; H, 9.26; N, 8.58%. For **11c**: colorless oil; IR ($CHCl_3$), 2923, 1605, 1574, 1492, 1453, 1358, 1132 cm⁻¹; ¹H-NMR ($CDCl_3$) δ=1.20—2.25 (18H, m), 2.25—3.00 (4H, m), 3.10—3.35 (1H, m), 7.20—7.80 (5H, m). For **11d**: mp 126—128 °C (from hexane); IR ($CHCl_3$), 2928, 2857, 1449, 1349, 1328, 1250 cm⁻¹; ¹H-NMR ($CDCl_3$), δ=1.40—2.40 (16H, m), 2.45—2.95 (4H, m), 3.10—3.35 (1H, m), 7.30—7.68 (5H, m). Found: C, 76.53; H, 8.85; N, 9.30%. Calcd for $C_{19}H_{26}N_2O$: C, 76.47; H, 8.78; N, 9.39%. For **11e**: mp 128—129 °C (from hexane); IR ($CHCl_3$), 2923, 2808, 1594, 1494, 1402, 1338, 1090, 1011 cm⁻¹; ¹H-NMR ($CDCl_3$), δ=1.10—2.40 (16H, m), 2.40—2.90 (4H, m), 3.10—3.30 (1H, m), 7.32 (2H, d, J=8.6 Hz), 7.60 (2H, d, J=8.6 Hz). Found: C, 68.41; H, 7.49; N, 8.42%. Calcd for $C_{19}H_{25}N_2OCl$: C, 68.56; H, 7.57; N. 8.42%.

General Procedure for the Synthesis of Isoxazoles ***12a—e.*** A solution of the 2-isoxazoline **11** (10 mmol) in methanol (25 cm³) and concentrated HCl (45 cm³) was refluxed for 1 h. The solution was then neutralized with aqueous NaOH and extracted with benzene. The benzene extract was washed with water and dried over Na_2SO_4. After the benzene had been evaporated, the residue was purified by recrystallization or distillation to give the isoxazoles **12a—e**. **12a**: mp 38—39 °C (from hexane); IR (CCl_4), 2915, 2865, 1620, 1468, 1445, 1410 cm⁻¹; ¹H-NMR ($CDCl_3$) δ=1.20—2.10 (16H, m), 2.50—3.00 (4H, m), 7.30—7.80 (5H, m); MS, *m/z* (rel intensity), 283 (M^+, 100). Found: C, 80.28; H, 8.79; N, 4.88%. Calcd for $C_{19}H_{25}NO$: C, 80.52; H, 8.89; N, 4.94%. **12b**: bp 128 °C/132 Pa; IR (film), 2923, 1618, 1582, 1473, 1447, 1414, 1335, 1273, 1131 cm⁻¹; ¹H-NMR ($CDCl_3$) δ=1.05—2.10 (12H, m), 2.45—3.00 (4H, m), 7.15—7.65 (5H, m); MS, *m/z* (rel intensity), 255 (M^+, 14), 77 (100). Found: C, 79.65; H, 8.38; N, 5.38%. Calcd for $C_{17}H_{21}NO$: C, 79.96; H, 8.29; N, 5.49%. **12c**: bp 120—125 °C/132 Pa; IR ($CHCl_3$), 2911, 1620, 1583, 1465, 1445, 1418, 1353, 1283, 1205 cm⁻¹; ¹H-NMR ($CDCl_3$) δ=1.18—2.05 (10H, m), 2.40—2.70 (2H, m), 2.73—3.00 (2H, m), 7.30—7.75 (5H, m); MS, *m/z* (rel intensity), 241 (M^+, 53), 77 (100). Found: C, 79.67; H, 7.78; N, 5.52%. Calcd for $C_{16}H_{19}NO$: C, 79.63; H, 7.94; N, 5.80%. **12d**: mp 54—55 °C (from hexane); IR ($CHCl_3$), 2932, 2857, 1594, 1584, 1449, 1418, 1351, 1261, 1126, 1071, 1010 cm⁻¹; ¹H-NMR ($CDCl_3$) δ=1.35—2.05 (8H, m), 2.40—2.75 (2H, m), 2.75—3.10 (2H, m), 7.22—7.70 (5H, m); MS *m/z* (rel intensity), 227 (M^+, 100). Found: C, 79.05; H, 7.69; N, 6.08%. Calcd for $C_{15}H_{17}NO$: C, 79.26; H, 7.54; N, 6.16%. **12e**: mp 97—98 °C (from hexane); IR ($CHCl_3$), 2941, 1620, 1441, 1143, 1081, 1020 cm⁻¹; ¹H-NMR ($CDCl_3$), δ=1.35—2.10 (8H, m), 2.40—2.75 (2H, m), 2.75—3.05 (2H, m), 7.40 (2H, d, J=9.0 Hz), 7.50 (2H, d, J=9.0 Hz); MS, *m/z* (rel intensity), 263 (M^+, 33), 261 (M^+, 100). Found: C, 68.72; H, 6.23; N, 5.47%. Calcd for $C_{15}H_{16}NOCl$: C, 68.83; H, 6.16; N, 5.35%.

General Procedure for the Synthesis of [n](2,5)Pyridinophanes ***13a—e.*** A solution of the isoxazole **12** (1 mmol), [$Mo(CO)_6$] (1 mmol), and DMAD (2 mmol) in benzene (10 cm³) was refluxed for 24 h under dry-nitrogen atmosphere. After hexane (10 cm³) was added to the reaction mixture, it was filtered through Celite to remove insoluble material. The filtrate was concentrated, and the resulting residue was separated by TLC on silica gel using benzene as the developer to give the [*n*](2,5)pyridinophane derivatives **13a—e** and hexamethyl benzenehexacarboxylate (*ca.* 10% yield in each case based on the DMAD used). The UV spectra of **13a—d** are displayed in Fig. 2, while absorption maxima are summarized in Table 3. **13a**: bp 150 °C (bath temp)/66 Pa; IR ($CHCl_3$), 1729 cm⁻¹; ¹H-NMR ($CDCl_3$) δ=0.60—0.95 (10H, m), 1.00—2.06 (6H, m), 2.83—3.35 (4H, m), 3.88 (3H, s), 3.90 (3H, s), 7.30—7.75 (5H, m); ¹³C-NMR ($CDCl_3$) δ=24.4, 25.9, 26.5, 26.6, 27.2, 27.4, 27.6, 27.7, 28.2, 35.2 (methylene chain), 52.4 (methoxyl groups), 128.4, 128.7, 129.7, 139.8, 142.4, 158.4, 161.3, 167.1, 167.7 (aromatic rings and carbonyls); MS, *m/z* (rel intensity), 409 (M^+, 38), 350 (100). Found: C, 73.12; H, 7.42; N; 3.19%. Calcd for $C_{25}H_{31}NO_4$: C, 73.32; H, 7.63; N, 3.42%. **13b**: bp 145 °C (bath temp)/132 Pa; IR ($CHCl_3$), 1721 cm⁻¹; ¹H-NMR ($CDCl_3$) δ=0.10—2.10 (12H, m), 2.10—3.10 (3H, m), 3.30—3.60 (1H, m), 3.87 (3H, s), 3.91 (3H, s), 7.30—7.62 (3H, m), 7.69—7.98 (2H, m); ¹³C-NMR ($CDCl_3$) δ=24.8, 25.3, 27.9, 28.8, 29.3, 30.1, 30.8, 36.2 (methylene chain), 52.4, 52.7 (methoxyl groups), 121.0, 128.6, 129.2, 129.3, 130.3, 138.8, 142.8, 159.6, 159.8, 166.7, 167.8 (aromatic rings and carbonyls); MS, *m/z* (rel intensity), 381 (M^+, 26), 380 (100). Found: C, 72.53; H, 7.15; N, 3.40%. Calcd for $C_{23}H_{27}NO_4$: C, 72.42; H, 7.13; N, 3.67%. **13c**: bp 130 °C (bath temp)/66 Pa; IR ($CHCl_3$), 1722 cm⁻¹; ¹H-NMR ($CDCl_3$) δ=0.20—2.10 (10H, m), 2.50—3.10 (3H, m), 3.53—3.93 (1H, m), 3.90 (3H, s), 3.97 (3H, s), 7.31—7.70 (3H, m), 7.84—8.07 (2H, m); ¹³C-NMR ($CDCl_3$) δ=25.6, 25.9, 26.1, 29.1, 29.9, 30.9, 36.5 (methylene chain), 52.4, 52.7 (methoxyl groups), 120.3, 127.9, 128.7, 129.5, 130.8, 138.4, 143.2, 158.8, 159.5, 166.5, 167.9 (aromatic rings and carbonyls); MS *m/z*

(rel intensity), 367 (M^+, 27), 366 (100). Found: C, 71.87; H, 6.93; N, 3.70%. Calcd for $C_{22}H_{25}NO_4$: C, 71.91; H, 6.86; N, 3.81%. **13d**: bp 120 °C (bath temp)/66 Pa; IR ($CHCl_3$), 1721 cm^{-1}; ^{1}H-NMR ($CDCl_3$) δ=0.45—0.98 (4H, m), 1.00—1.75 (4H, m), 2.50—3.05 (3H, m), 3.30 (1H, dxt, J=12.0, 6.0 Hz), 3.87 (3H, s), 3.93 (3H, s), 7.38—7.60 (3H, m), 7.90—8.10 (2H, m); ^{13}C-NMR ($CDCl_3$) δ=24.8, 25.3, 31.5, 33.0, 33.8, 37.4 (methylene chain), 52.2, 52.8 (methoxyl groups), 127.6, 128.7, 130.1, 130.4, 136.3, 136.7, 142.5, 158.1, 164.4, 166.3, 168.2 (aromatic rings and carbonyls); MS, m/z (rel intensity), 353 (M^+, 76), 352 (100). Found: C, 71.37; H, 6.56; N, 3.96%. Calcd for $C_{21}H_{23}NO_4$: C, 71.34; H, 6.71; N, 3.94%. **13e**: bp 150 °C (bath temp)/93 Pa; IR ($CHCl_3$), 1721 cm^{-1}; ^{1}H-NMR ($CDCl_3$) δ=0.40—0.80 (4H, m), 1.05—1.80 (4H, m), 2.40—3.05 (3H, m), 3.32 (1H, dxt, J=12.1, 6.2 Hz), 3.91 (3H, s), 4.00 (3H, s), 7.40 (2H, d, J=8.5 Hz), 7.90 (2H, d, J=8.5 Hz); ^{13}C-NMR ($CDCl_3$) δ=25.5, 25.9, 31.5, 32.9, 33.7, 37.4, (methylene chain), 52.3, 52.9, (methoxy groups), 127.6, 128.4, 129.0, 131.5, 136.4, 142.6, 156.9, 164.2, 165.9, 167.7 (aromatic rings and carbonyls); MS, m/z (rel intensity), 389 (M^+, 25), 387 (M^+, 75), 386 (100); UV (EtOH, log ε) 252 (4.23), 287 (4.01), 345 (3.98) nm. Found: C, 64.69; H, 5.82; N, 3.88%. Calcd for $C_{21}H_{22}NO_4Cl$: C, 65.03; H, 5.72; N, 3.61%.

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