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Dedicated to the memory of
Professor Mircea D. Banciu (1941–2005)

NEW PYRROLO[2,1-a]PHTHALAZINE DERIVATIVES BY 1,3-DIPOLAR CYCLOADDTION REACTIONS

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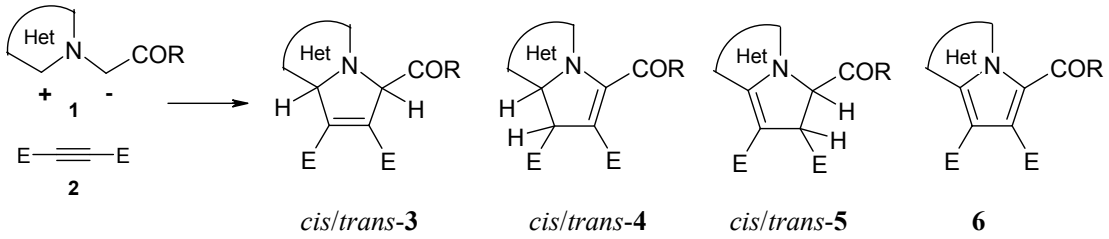
The esters of pyrrolo[2,1-a]phthalazine-1,2-dicarboxylic acid **13a-c** were obtained from pthalazinium bromide **9**, esters of acetylenedicarboxylic acid, triethylamine and tetrakis-pyridinocobalt(II) dichromate (TPCD) as oxidant. The synthesis of pyrrolo[2,1-a]phthalazine-1-carboxylic esters from **9**, acetylenemonocarboxylic acid esters and triethylamine was performed in the absence of TPCD. Structure proof for the new compunds is based on H-, C-NMR, COSY and Hetcor experiments.

INTRODUCTION

1,3-Dipolar cycloadditions are powerful tools for the synthesis of simple and highly fused heterocyclic compounds because their ability to create sets of bonds in a single operation. Among a variety of 1,3-dipoles, heteroaromatic *N*-ylides are some of the most important dipoles in the construction of *N*-heterocycles, raising at the same time interesting regio- and stereoselectivity problems.[1-13]

The 1,3-dipolar cycloaddition reactions between heteroaromatic *N*-ylides **1** and acetylenic dipolarophiles **2** give the primary cycloadducts **3** which rearrange to pyrrolines **4** and/or **5**. Usually, in the reaction conditions, intermediates **4** and **5** are too unstable due to their tendency to dehydrogenation with the formation of fused pyrroles **6** (Scheme 1).



Scheme 1

Pyrrolo[2,1-a]phthalazine derivatives have also been found to possess biological activity and interesting optical properties.[14-18]

Herein we describe the synthesis of new pyrrolo[2,1-a]phthalazines **13a-e** by 1,3-dipolar cycloaddition reactions between phthalazinium *N*-ylide **10** and activated alkynes.

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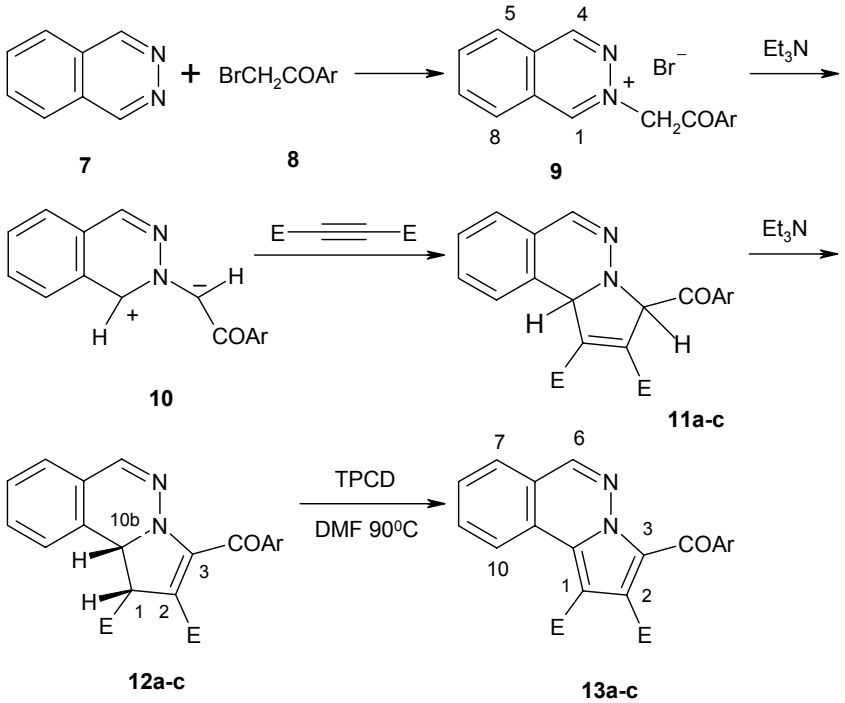
RESULTS AND DISCUSSION

The starting material, 2-[2-(3-nitrophenyl-2-oxoethyl)]phthalazinium bromide **9** was prepared by *N*-alkylation of phthalazine **7** with 2-bromo-3'-nitroacetophenone **8** (Scheme 2). The structure of the phthalazinium bromide **9** was confirmed by elemental analysis and NMR spectroscopy. In the ^{1}H-NMR spectrum of compound **9**, recorded in $CDCl_3$+TFA, the protons H-1 (δ = 10.57 ppm) and H-2 (δ = 9.69 ppm) are strongly deshielded due to the vicinity of the two nitrogen atoms (they are grafted on double bond C-N). In the dilute solution the signals for H-1 and H-4 appear as two triplets with a coupling constatant of ca. 0.9 Hz. The multiplicity of H-1 could be explained by long range coupling of H-1 with H-4 and H-5, respectively. Similarly, the proton H-4 is coupled with H-1 and H-8, respectively. These long range couplings were put in evidence by HH decoupling experiments.

The phthalazinium *N*-ylide **10**, being unstable, was generated *in situ* by deprotonation of the cycloimmonium bromide **9** with triethylamine. Treatment of phthalazinium bromide **9** with dimethyl acetylenedicarboxilate (DMAD) in the presence of an excess of triethylamine gave the 1,10b-dihydrophthalazine derivative **12a** (Scheme 2). The reaction was performed in methylene chloride at room temperature for ca. 15 min. Similar results were obtained when DMAD was replaced by diethyl and diisopropyl acetylenedicarboxylate, respectively.



7 **8** **9** **10** **11a-c** **12a-c** **13a-c**

Ar = 3-$NO_2C_6H_4$; a: E = CO_2Me; b: E = CO_2Et; c: E = CO_2iPr

Scheme 2

The structure of dihydroderivative **12a** was assigned by ^{1}H- and ^{13}C-NMR spectroscopy. The protons H-1 (δ = 4.53 ppm) and H-10b (δ = 5.35 ppm) appeared in the ^{1}H-NMR spectrum as two doublets with J = 12.9 Hz. The large value of the vicinal coupling constant between H-1 and H-10b indicates a *cis* configuration.

The position of the double bond in the pyrroline moiety resulted from the chemical shifts of the three carbonyl groups. Thus, the large difference between the two carbonyl ester groups (δ = 164.0 ppm and 172.6 ppm) shows that they are grafted on a Csp^2 (δ = 164.0 ppm) and a Csp^3 (δ = 172.6 ppm), respectively. Also, the shielding of the ketone group (δ = 186.2 ppm) is a good evidence for conjugation with a double bond of the pyrroline ring. This value is close with to those of the corresponding of aromatic compounds **13a**, which were found to be 184.4 ppm.

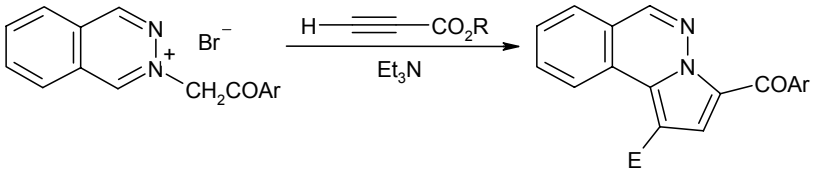
The formation of dihydroderivatives **12** may be explained by the regio- and stereoselective prototropic rearrangement of the primary cycloadducts **11** in the presence of an excess of triethylamine. The isolation

and characterization of such primary cycloadducts in the phthalazine series was recently reported.[6,9] The aromatization of dihydroderivative **12a** to pyrrolophthalazine **13a** was performed, in good yields, by action of tetrakis-pyridinoCo(II)dichromate (TPCD). This reagent was used recently for aromatization of cycloadducts obtained by reaction between heteroaromatics *N*-ylides and activated olefines.[19-21]

Pyrrolo[2,1-a]phthalazine derivatives **12a-c** were also obtained by the "one pot" synthesis from phthalazinium bromide **9**, esters of acetylenedicarboxylic acid, triethylamine and TPCD, in DMF, at 90 °C.

The 1,3-dipolar cycloaddition between *N*-ylide **10** and non-symmetrical acetylenic dipolarophiles such as ethyl or isopropyl propiolate is regiospecific. The reaction of *N*-ylide **10** with ethyl or isopropyl propiolate in methylene chloride at room temperature produced directly the aromatic pyrrolophthalazine **13d,e** (Scheme 3). The regioselectivity in the cycloaddition of **10** with activated non-symmetrical acetylenes can be satisfactorily explained by strong polarization of the dipolarophile triple bond.

From the above results it can be concluded that the intermediates in the 1,3-dipolar cycloaddition of phthalazinium *N*-ylides with activated non-symmetrical alkynes are less stable than in the case of activated symmetrical acetylenes.



9 **13d,e**

Ar = 3-$NO_2C_6H_4$; d: E = CO_2Et; e: E = CO_2iPr

Scheme 3

The new pyrrolophthalazine derivatives were characterized by elemental analysis and NMR spectroscopy.

In the ^{1}H-NMR spectra of compounds **13a-e** the signal of H-10 (δ = 8.84-9.84) is deshielded in comparison with the protons of the phthalazine moiety which appeared in the range 7.68-7.90 ppm. The high chemical shift for proton H-10 can be explained by deshielding and this is a consequence of the presence of carboalkoxy groups in the position 1 of the pyrrole ring. Also, the proton H-6 (δ = 8.46-8.77) is deshielded because is grafted on a double C-N bond. It is interesting to note that the signal for the proton H-6 appeared as a doublet with J ≈ 0.8 Hz when ^{1}H-NMR spectra of the compounds **13** were recorded in dilute solution. The multiplicity of H-6 can be explained by presence of a long range coupling between protons H-6 and H-10. This coupling was confirmed by HH decoupling experiments.

CONCLUSIONS

The 1,3-dipolar cycloaddition between *N*-ylide **10** with diesters of acetylenedicarboxylic acid, performed in dichloromethane, gave dihydroderivatives **12** which were subsequently aromatized with TPCD to **13a-c**. In the same reaction conditions, **10** and esters of acetylenemonocarboxylic acid gave directly pyrrolophthalazine derivatives **13d,e**.

The pyrrolophthalazine **13a-c** were obtained in 'one pot' reactions from pthalazinium bromide **9**, diesters of acetylenedicarboxylic acid, triethylamine and TPCD.

EXPERIMENTAL PART

Melting points were determined on a Boëtius hot plate and are uncorrected. The NMR spectra were recorded on a Varian Gemini 300 BB instrument, operating at 300 MHz for ^{1}H and 75 MHz for ^{13}C.

2-[2-(3-nitrophenyl)-2-oxoethyl]phthalazinium bromide (**9**)

1.3 g (10 mmol) Phthalazine and 2.7 g (11 mmol) 2-bromo-3'-nitroacetophenone in 80 mL of acetone were stirred at room temperature for 5-6 hrs. The precipitate was collected on the filter paper and washed with acetone. The crude product was in 94% yield. The phthalazinium bromide **9** was purified by recrystallization from methanol, giving white crystals with m.p. 254-7°C.

Required for $C_{16}H_{12}N_3O_3$: C, 51.35; H, 3.23; Br, 21.35; N, 11.23. Found: C, 51.72; H, 3.58; Br, 21.76; N, 11.51. ^{1}H-NMR ($CDCl_3$ + TFA; δ, ppm; *J*, Hz): 6.79 (s, 2H, CH_2); 7.85 (t, 1H, 8.0, H-5'); 8.41-8.49 (m, 3H, H-6, H-7, H-4'); 8.53-8.61 (m, 2H, H-5, H-6'); 8.76-8.79 (m, 1H, H-8); 8.92 (t, 1H, 2.0, H-2'); 9.69 (s, 1H, H-4); 10.57 (s, 1H, H-1). ^{13}C-NMR ($CDCl_3$ + TFA; δ, ppm): 69.4 (CH_2); 123.4 (C-2'); 127.9 (C-5); 128.0 (C-4a, C-8a); 129.4 (C-5'); 130.8 (C-4'); 131.6 (C-8); 134.2 (C-6'); 134.3 (C-1'); 137.0 (C-7); 140.6 (C-6); 148.7 (C-3'); 153.5 (C-1); 154.1 (C-4); 187.9 (CO).

Dimethyl cis-3-(3-nitrobenzoyl-1,10b-dihydropyrrolo[2,1-a]phthalazine-1,2-dicarboxylate (**12a**)

1.1 g (3 mmol) Phthalazinium bromide **9** were suspended in dichloromethane (20 mL) and then 3.3 mmol dimethyl acetylenedicarboxylate were added. Under stirring, triethylamine (4 mmol) was added drop wise in 5 mL of methylene chloride. After 10 min. the reaction mixture was washed twice with water and the solvent evaporated. The product was recrystallized from acetonitrile and orange crystals with m.p. 163-5°C were obtained. Yield 92%. Required for $C_{22}H_{17}N_3O_7$: C, 60.69; H, 3.94; N, 9.65. Found: C, 61.02; H, 4.28; N, 9.97. ^{1}H-NMR ($CDCl_3$; δ, ppm; *J*, Hz): 3.54, 3.93 (2s, 6H, 2 Me); 4.53 (d, 1H, 12.9, H-1); 5.35 (d, 1H, 12.9, H-10b); 7.25-7.28 (m, 1H, H-7); 7.39-7.46 (m, 2H, H-9); 7.45 (s, 1H, H-6); 7.52-7.57 (m, 1H, H-10, H-8); 7.74 (t, 1H, 8.1, H-5'); 8.34 (bd, 1H, 8.1, H-6'); 8.46-8.50 (m, 1H, H-4'); 8.85 (bs, 1H, H-2'). ^{13}C-NMR ($CDCl_3$; δ, ppm): 51.5, 52.3 (2 OMe); 53.0 (C-1); 62.0 (C-10b); 102.3 (C-2); 123.4 (C-10); 123.7 (C-2'); 124.6, 130.6 (C-6a, C-10a); 128.4, 130.2 (C-4', C-5'); 128.8 (C-8); 132.0 (C-9); 134.7 (C-6'); 136.2 (C-1'); 142.7 (C-6); 148.7 (C-3'); 151.8 (C-2); 164.0 (2-COO); 172.6 (1-COO); 186.2 (COAr).

General procedure for the synthesis of compounds **13a-c**

Phthalazinium bromide **9** (1.9 g, 5 mmol) was suspended in dimethylformamide (25 mL) and then dimethyl (diethyl or diisopropyl) acetylenedicarboxylate (5.5 mmol) was added. Under stirring, triethylamine (5.5 mmol) and tetrakispyrdine cobalt(II)-dichromate (TPCD) (2 g, 3.2 mmol) were added to the reaction mixture. The reaction mixture was stirred at 80-90°C for 5 hrs. and then cooled at room temperature and poured into a 5 % aqueous HCl solution (100 mL). The solid product was filtered, washed with water and purified either by recrystallization or column chromatography on neutral Al_2O_3, using CH_2Cl_2 as eluent.

Compound **13a** was also obtained from **12a** as it follows: 2.5 mmol of dihydroderivative **12a** and 1 g of TPCD in 15 mL of DMF were stirred at 90°C for 5 hrs. The reaction mixture was treated as above to give the pyrrolophthalazine **13a**, in 74% yield.

Dimethyl 3-(3-nitrobenzoyl)-pyrrolo[2,1-a]phthalazine-1,2-dicarboxylate (**13a**)

The product was recrystallized from acetonitrile and white crystals with m.p. 180-2°C were obtained. Yield 78%. Required for $C_{22}H_{15}N_3O_7$: C, 60.97; H, 3.49; N, 9.70. Found: C, 61.22; H, 3.88; N, 9.97. ^{1}H-NMR ($CDCl_3$; δ, ppm; *J*, Hz): 3.70 (s, 3H, 1-CO_2CH_3); 4.01 (s, 3H, 2-CO_2CH_3); 7.67 (t, 1H, 7.9, H-5'); 7.68-7.73 (m, 1H, H-8); 7.81-7.90 (m, 2H, H-7, H-9); 8.14-8.17 (m, 1H, H-4'); 8.43-8.47 (m, 1H, H-6'); 8.46 (s, 1H, H-6); 8.66 (t, 1H, 1.9, H-2'); 8.91 (ddd, 8.2, 0.8, H-10). ^{13}C-NMR ($CDCl_3$; δ, ppm): 52.4, 52.6 (2 OMe); 107.9 (C-1); 121.3, 121.4, 125.3, 126.5, 126.9 (C-2, C-3, C-6a, C-10a, C-10b); 124.2 (C-2'); 125.1 (C-10); 127.5 (C-4'); 128.3, 129.4 (C-7, C-8); 129.7 (C-5'); 133.6 (C-9); 134.9 (C-6'); 139.1 (C-1'); 147.2 (C-6); 148.3 (C-3'); 163.9, 165.9 (2 COOMe); 184.4 (COAr).

Diethyl 3-(3-nitrobenzoyl)-pyrrolo[2,1-a]phthalazine-1,2-dicarboxylate (**13b**)

The product was recrystallized from ethyl acetate or from a mixture of isopropanol + acetonitrile (2:1) and white crystals with m.p. 148-9°C were obtained. Yield 82%. Required for $C_{24}H_{19}N_3O_7$: C 62.47 H 4.15 N 9.11. Found: C 62.75 H 4.48 N 9.34. ^{1}H-NMR ($CDCl_3$; δ, ppm; *J*, Hz): 1.15 (t, 3H, 7.1, 1-$CO_2CH_2CH_3$); 1.43 (t, 3H, 7.1, 2-$CO_2CH_2CH_3$); 4.15 (q, 2H, 7.1, 1-CO_2CH_2); 4.49 (q, 2H, 7.1, 2-CO_2CH_2); 7.67 (t, 1H, 7.9, H-5'); 7.68-7.72 (m, 1H, H-8); 7.81-7.89 (m, 2H, H-7, H-9); 8.15-8.19 (m, 1H, H-4'); 8.43-8.47 (m, 1H, H-6'); 8.46 (s, 1H, H-6); 8.68 (t, 1H, 1.9, H-2'); 8.91 (bd, 1H, 8.3, H-10). ^{13}C-NMR ($CDCl_3$; δ, ppm): 13.8, 14.0 (2 Me); 61.5, 61.8 (2 CH_2O); 108.2 (C-1); 121.3, 121.5, 125.1, 126.5, 126.8 (C-2, C-3, C-6a, C-10a, C-10b); 124.2 (C-2'); 125.0 (C-10); 127.5 (C-4'); 128.3, 129.4 (C-7, C-8); 129.7 (C-5'); 133.6 (C-9); 134.9 (C-6'); 139.1 (C-1'); 147.1 (C-6); 148.3 (C-3'); 163.4, 164.8 (2 COOEt); 184.5 (COAr).

Diispropyl 3-(3-nitrobenzoyl)-pyrrolo[2,1-a]phthalazine-1,2-dicarboxylate (**13c**)

The product was recrystallized from ethanol and white crystals with m.p. 121-3°C were obtained. Yield 80%. Required for $C_{26}H_{23}N_3O_7$: C, 63.80; H, 4.74; N, 8.58. Found: C, 64.12; H, 5.08; N, 8.97. ^{1}H-NMR ($CDCl_3$; δ, ppm; *J*, Hz): 1.10 (d, 6H, 6.3, 1-CO_2CHMe_2); 1.45 (d, 6H, 6.3, 2-CO_2CHMe_2); 5.04 (sep, 1H, 6.3, 1-CO_2CH); 5.39 (sep, 1H, 6.3, 2-CO_2CH); 7.65-7.71 (m, 1H, H-8); 7.67 (t, 1H, 7.9, H-5'); 7.80-7.88 (m, 2H, H-7, H-9); 8.16-8.19 (m, 1H, H-4'); 8.42-8.47 (m, 1H, H-6'); 8.43 (s, 1H, H-6); 8.71 (t, 1H, 1.9, H-2'); 8.84 (bd, 1H, 8.3, H-10). ^{13}C-NMR ($CDCl_3$; δ, ppm): 21.4, 21.7 (4 Me); 69.4, 69.7 (2 $CHMe_2$); 108.9 (C-1); 121.2, 121.4, 124.6, 126.5, 126.8 (C-2, C-3, C-6a, C-10a, C-10b); 124.2 (C-2'); 124.9 (C-10); 127.5 (C-4'); 128.3, 129.2 (C-7, C-8); 129.7 (C-5'); 133.5 (C-9); 135.1 (C-6'); 139.3 (C-1'); 147.0 (C-6); 148.4 (C-3'); 162.6, 162.5 (2 COOiPr); 184.6 (COAr).

General procedure for the synthesis of compounds **13d,e**

1.9 g (5 mmol) Phthalazinium bromide **9** were suspended in 25 mL of dichloromethane and then 6 mmol of ethyl (or isopropyl) propiolate were added. Under vigorous stirring, 0.7 mL (5 mmol) of triethylamine (dissolved in 5 mL of methylene chloride) were dropped. After 15 min., the reaction mixture was washed with water and the solvent evaporated. The residue was triturated with ethanol and the precipitate was collected on a filter and washed with ethanol.

Ethyl 3-(3-nitrobenzoyl)-pyrrolo[2,1-a]phthalazine-1-carboxylate (**13d**)

The product was recrystallized from acetonitrile and white crystals with m.p. 205-7°C were obtained. Yield 77%. Required for $C_{21}H_{15}N_3O_5$: C, 64.78; H, 3.88; N, 10.79. Found: C, 65.12; H, 4.07; N, 11.07. ^{1}H-NMR ($CDCl_3$; δ, ppm; *J*, Hz): 1.41 (t, 3H, 7.1,

CH_3); 4.43 (q, 2H, 7.1, CH_2); 7.72 (t, 1H, 7.9, H-5'); 7.73 (s, 1H, H-2); 7.76-7.82 (m, 1H, H-8); 7.91-7.97 (m, 2H, H-7, H-9); 8.24-8.27 (m, 1H, H-4'); 8.45-8.49 (m, 1H, H-6'); 8.74 (t, 1H, 1.9, H-2'); 8.77 (s, 1H, H-6); 9.86 (bd, 8.1, 0.8, H-10). ^{13}C-NMR ($CDCl_3$; δ, ppm): 14.4 (Me); 60.9 (CH_2); 109.1 (C-1); 122.2, 126.2, 128.2, 130.9 (C-3, C-6a, C-10a, C-10b); 124.4 (C-2'); 125.2 (C-10); 126.6, 129.7, 130.0 (C-6, C-5', C-7); 127.6, 127.7 (C-2, C-4'); 133.1 (C-9); 135.1 (C-6'); 140.6 (C-1'); 146.7 (C-6); 148.1 (C-3'); 163.8 (COO); 181.9 (COAr).

Isopropyl 3-(3-nitrobenzoyl)-pyrrolo[2,1-a]phthalazine-1-carboxylate (**13e**)

The product was recrystallized from acetonitrile and white crystals with m.p. 210-2°C were obtained. Yield 78%. Required for $C_{22}H_{17}N_3O_5$: C, 65.50; H, 4.25; N, 10.42. Found: C, 65.88; H, 4.57; N, 10.69. ^{1}H-NMR ($CDCl_3$; δ, ppm; *J*, Hz): 1.41 (d, 6H, 6.2, 2 Me); 5.31 (sep, 1H, 6.2, $CHMe_2$); 7.72 (t, 1H, 7.9, H-5'); 7.73 (s, 1H, H-2); 7.75-7.81 (m, 1H, H-8); 7.90-7.97 (m, 2H, H-7, H-9); 8.24-8.27 (m, 1H, H-4'); 8.45-8.49 (m, 1H, H-6'); 8.74 (s, 1H, H-6); 8.74 (t, 1H, 1.9, H-2'); 9.86 (bd, 8.1, 0.9, H-10). ^{13}C-NMR ($CDCl_3$; δ, ppm): 22.0 (2 Me); 68.4 ($CHMe_2$); 109.5 (C-1); 122.2, 126.3, 126.8, 130.7 (C-3, C-6a, C-10a, C-10b); 124.4 (C-2'); 125.1 (C-10); 126.6, 129.5, 130.0 (C-6, C-7, C-5'); 127.6, 127.7 (C-2, C-4'); 133.9 (C-9); 135.1 (C-6'); 140.6 (C-1); 146.6 (C-6); 148.2 (C-3'); 163.4 (COO); 182.0 (COAr).

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