66-2; $PhSCH{=}CH_2$, 1822-73-7; $PhS(CH_2)_2OH$, 699-12-7; $PhS(CH_2)_2Cl$, 5535-49-9; $(n\text{-}C_4H_9)_2S$, 544-40-1; $(s\text{-}C_4H_9)_2S$, 626-26-6; $(t\text{-}C_4H_9)_2S$, 107-47-1; Me-p-$C_6H_4S(O)Me$, 934-72-5; Me-p-$C_6H_4SO_2Me$, 3185-99-7; PhS(O)Me, 1193-82-4; $PhSO_2Me$, 3112-85-4; $PhS(O)CH_2Ph$, 833-82-9; PhS(O)Ph, 945-51-7; $PhSO_2CH{=}CH_2$, 5535-48-8; $PhS(O)CH{=}CH_2$, 20451-53-0; $PhSO_2(CH_2)_2OH$, 20611-21-6; $PhS(O)(CH_2)_2Cl$, 27998-60-3; $(n\text{-}C_4H_9)_2S(O)$, 2168-93-6; $(s\text{-}C_4H_9)_2S(O)$, 13153-06-5; $(t\text{-}C_4H_9)_2S(O)$, 2211-92-9; benzenesulfonamide, 98-10-2; 4-nitrobenzaldehyde, 555-16-8; potassium peroxymonosulfate, 10361-76-9; thietane, 287-27-4; thianthrene, 92-85-3; dibenzothiophene, 132-65-0; thietane, 1-oxide, 13153-11-2; thietane, 1,1-dioxide, 5687-92-3; thianthrene 5,10-dioxide, 951-02-0; dibenzothiophene 5-oxide, 1013-23-6; dibenzothiophene 5,5-dioxide, 1016-05-3; 2-(phenylthio)cyclohexanone, 27920-40-7; 2-(phenylsulfinyl)cyclohexanone, 55705-17-4.

Properties of a Tetracyanopentacenequinone and Its Tetracyanoquinodimethane Derivative

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A soluble tetracyanopentacenequinone was prepared. This quinone was converted to a TCNQ derivative by base-catalyzed reaction with malononitrile at high temperature. Variable-temperature NMR spectroscopy revealed a slow conformation change for the TCNQ derivative. Electrochemical reductions of the quinone and its TCNQ derivative were studied and UV–vis–NIR spectra of the neutrals, anion radicals, and dianions were recorded. The ions from the tetracyanopentacenequinone show very long wavelength NIR bands ascribed to strong perturbation of the quinone$^{\bullet-}$ by the cyano groups.

Tetracyanoquinodimethane (TCNQ) and its analogues have attracted continuing attention. Most fundamentally, this attention has resulted because these compounds are excellent electron acceptors. As such they form anion radicals, e.g., TCNQ$^{\bullet-}$, at very positive potentials and form charge-transfer complexes with many donors. The most famous of these is TTF–TCNQ, which was the first organic solid to show metallic conductivity. A number of derivatives have been synthesized in order to explore the conductivity. Of particular interest in the present context are several anthracene derivatives like **3-CN**,[1-3] which were prepared and studied to provide a TCNQ with an extended π-system.

Recent work from this laboratory has been directed toward the preparation of quinones with extended π-systems and also linear arrays of such polyacenequinones.[4,5] One point of particular interest has been the unusual near-infrared (NIR) absorption bands exhibited by the anion radicals of these compounds.[6] It was of interest to extend the scope of this work by preparing TCNQ derivatives of our unusual quinones. We report here on a five-ringed tetracyano quinone (**5-O**) and its TCNQ derivative (**5-CN**). This structure was selected as a synthetic goal out of considerations of solubility, synthetic accessibility, and redox potential. Thus, the aryl groups provide solubility and the four aromatic cyano groups keep the quinone reduction potential from being too negative. Our findings include unusual ^{1}H NMR results showing a conformational change in **5-CN** and UV–vis–NIR results on **5-O**, **5-O**$^{\bullet-}$, **5-O**$^{2-}$, **5-CN**, and **5-CN**$^{2-}$. It is shown that the strong interaction of the aromatic cyano groups with the quinone on **5-O**$^{\bullet-}$ leads to NIR bands not observed for pentacenequinone, **6**$^{\bullet-}$, which lacks the aromatic cyano groups.

Synthesis. Previous studies have demonstrated the utility of Diels–Alder addition to the bis diene **4**.[4] This precursor is of special utility because the *p-tert*-butylphenyl groups provide solubility even to rigid polyacenequinone derivatives 75 Å long.[5] In the present study, addition of fumaronitrile to **4** followed by aromatization gave **5-O**, which was identified spectrally.

The quinone **5-O** could not be converted to **5-CN** by using the published conditions[2] ($TiCl_4$, pyridine in chloroform), but a modicum of success was achieved by using 1,8-bis(dimethylamino)naphthalene as the base in the higher boiling solvent 1,1,2,2-tetrachloroethane. The product **5-CN** was purified by flash chromatography and gave satisfactory IR, NMR, UV, and high resolution mass spectra.

^{1}H NMR of 5-CN. This NMR spectrum is interesting because it reveals a slow conformational change. Comparison of the spectrum of **5-CN** with that of **5-O** and other derivatives allows a firm assignment of the peaks. Of interest are the aryl hydrogens of the attached *p-tert*-butylphenyl groups. The spectra of **5-O** and other derivatives show a pair of A–B doublets for the ortho and meta hydrogens. In contrast, **5-CN** in $CDCl_3$ shows four pairs of doublets.

This complexity is due to a slow conformational change, as shown by a variable-temperature ^{1}H NMR study. Toluene-d_8 was used as solvent to get a more suitable liquid range. This change in solvent changed the chemical shifts and coupling constants, but the spectrum at 0 °C was recognizably similar to that obtained at room temperature in $CDCl_3$. There were four sets of slightly broadened doublets in the range of 7.5–8.0 ppm. When the temperature was raised to 35 °C these peaks broadened further

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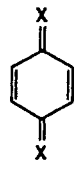

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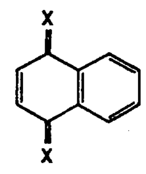

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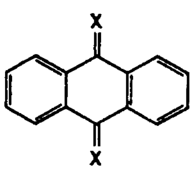

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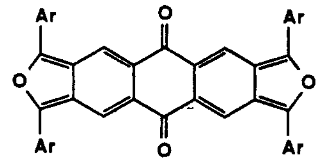

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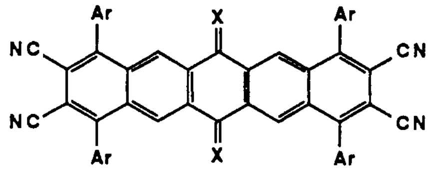

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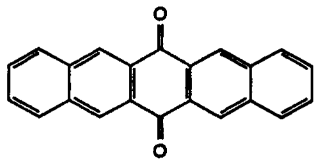

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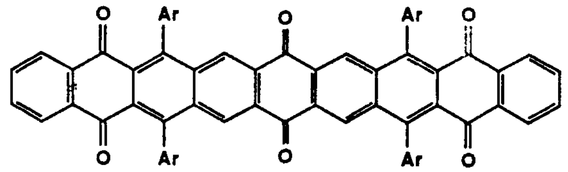

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1,2,3,5-CN: X=C((CN_2))

1,2,3,5-O: X=O

Ar=p-$(CH_3)_3C$-C_6H_4

and started to coalesce (see supplementary material). At 105 °C a pair of doublets—like those from 5-O—was observed. Lowering the temperature to −20 °C gave very narrow lines, and at this temperature, long-range (presumably meta) couplings, which split all the peaks further, can be discerned. These long-range couplings suggest that there are four different hydrogens on each ring in the stable conformation which is locked in at −20°. It refutes an alternative that at room temperature there are two kinds of aryl rings each with equivalent ortho and equivalent meta hydrogens.

Rationalizing these observations is not difficult as the crystal structure of 3-CN has been determined and the central ring has been shown to be distorted into a nonplanar boat conformation.[7] It is proposed that in 5-CN the *tert*-butylphenyl side groups are substantially twisted out of the plane of the pentacene to which they are attached. This arrangement (Figure 1) would place the magnetically anisotropic cyano groups closer to two of the *tert*-butylphenyl protons than the other two, giving four nonequivalent protons on each aryl ring. If *both* the rate of the boat-boat interconversion of the central TCNQ unit *and* the rate of a (full) rotation of the aryl side groups are slow on the NMR timescale, the observed −20 °C spectrum will result.[8]

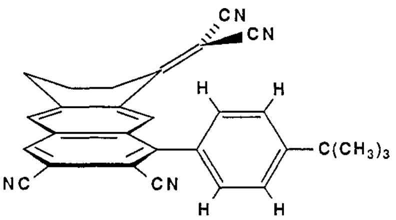

Figure 1. Partial structure of 5-CN.

Table I. $E_{1/2}$ Values from Cyclic Voltammetry[a]

cmpd	$E_{1/2}$ (−V, SCE)	cmpd	$E_{1/2}$ (−V, SCE)
1-O	0.43, 1.34	1-CN[b]	−0.18 0.42
2-O	0.61, 1.41	2-CN[b]	0.00 0.43
3-O	0.85, 1.58	3-CN[b]	0.41 (2 e)
5-O	0.56, 1.09	5-CN	0.27 (2 e)
6	1.08, 1.62		

[a] DMF, 0.1 M $TBABF_4$. [b] Converted to SCE by adding 0.30 V to reported value.[1]

Table II. Vis-NIR Data for Anion Radicals and Dianions in DMF

cmpd	λ_{max} (nm) (log ϵ)
5-CN^{2-}	1060 (3.95), 590 (4.34)
5-O$^{\bullet-}$	1020 (4.38)
5-O^{2-}	915 (4.61)
6$^{\bullet-}$	612 (4.00)
6^{2-}	573 (4.04)
7$^{\bullet-}$	1730 (4.43) 485 (4.11) 435 (4.17)
7^{2-}	1900 (4.87) 490 (4.24)

Electrochemistry

Cyclic voltammetry was employed to evaluate the reduction potentials as well as the reversibility of the reduction process for 5-O and 5-CN. As shown in Table I, 5-O gave two reversible one-electron couples corresponding to formation of the quinone anion radical and dianion. A comparison of the $E_{1/2}$ values of several quinones (Table I) shows that the cyano groups have an expectedly profound effect on the reduction potential. Thus comparison of 1-O, 2-O, 3-O, and 6 shows that benzo or higher annulation of a quinone causes the $E_{1/2}$ to become progressively more negative. The nitrile substitution in 5-O reverses this trend so that 5-O has a first reduction potential similar to that of naphthoquinone. Interestingly, the separation between the two reversible waves is reduced by increased π-conjugation. This phenomenon has been documented for other quinones. It presumably is a result of extensive delocalization, which minimizes electron-electron repulsion in the dianion.

The reduction of 5-CN mimicked the unusual behavior shown by 3-CN.[1] That is, the first reduction process consumed two electrons, generating the dianion. The cyclic voltammogram in DMF/Bu_4NBF_4 shows a reversible couple, $E_{1/2}$ = −0.27 V (SCE) with peak separation of 40

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(7) Schubert, U.; Hunig, S.; Aumuller, A. *Liebigs Ann. Chem.* 1985, 1216.

(8) In addition to the kinetic phenomenon revealed by the peak coalescence, the spectra show temperature-dependent chemical shifts. Thus, the coalesced peaks are not midway between the low-temperature peaks as expected. This may result from population of new conformers at high temperature. Whatever the explanation, it makes it impossible for us to use standard approaches to compute $\Delta H^{\ddagger}$ and we are reluctant to attempt a more detailed mechanistic explanation. The process is complex, and we do not have enough information to unravel it.

mV and peak current about twice that for an equal concentration of **5**-O. There were two additional one-electron couples at −0.94 V and −1.39 V. The observation of a two-electron couple can be rationalized as in the case of **3**-CN[1] by considering the conformational changes that take place upon reduction. Thus, **5**-CN reduction leads to a dianion with a flattened pentacene ring and dicyanomethylene groups that are rotated to become more or less perpendicular to the pentacene ring. This minimizes electron repulsions and steric interactions. If this conformational change takes place for the anion radical as well as the dianion it allows the first and second one-electron peaks to merge into a single two-electron peak as observed.

Electronic Spectra

The UV spectra of quinones have been widely reported and effectively analyzed by Nepras and Titz.[9] A comparison of TCNQ and quinone derivatives is informative. In general, TCNQ derivatives display their lowest energy (π–π^*) absorbance at longer wavelength than the corresponding quinone. Thus, **1**-CN has its lowest energy $\lambda_{\max}$ 395 nm, whereas for **1**-O, the low energy π–π^* band is at $\lambda_{\max}$ 282 nm. This is also seen for **2**-CN (409 nm) and **2**-O (325 nm). In the quinone series, further benzoannulation results in a further red shift of the π–π^* band in a regular fashion: **3**-O (318 nm), **6** (405 nm), **5**-O (425 nm). For TCNQ derivatives, such a trend is not found. Due to their nonplanar geometry, **3**-CN and **5**-CN absorb at higher energy than predicted on the basis of a comparison to **1**-CN and **2**-CN. **3**-CN shows its lowest energy π–π^* band at 343 nm. This is blue-shifted relative to **2**-CN. Also for **5**-CN, the low energy π–π^* is at $\lambda_{\max}$ 432 nm. This is similar to that of **5**-O.

Recent investigations in this laboratory have revealed the unusual near-IR absorption bands of certain quinone anion radicals.[6] These semiquinones have sharp and intense bands in the 1–2 μm region. The reported examples refer to diquinones and a triquinone, although we have found related anion radicals that show intense NIR bands. Simple monoquinone anion radicals do not display such bands. Since it is suspected that the origin of near-IR electronic transitions for anions of di- and triquinones involves interaction between multiple electrophores, it was of obvious interest to observe Vis–NIR spectra of **5**-O$^{\bullet-}$, **5**-O^{2-}, and **5**-CN^{2-}. The additional CN groups in **5**-O$^{\bullet-}$ compared to **6**$^{\bullet-}$ were expected to have a substantial effect and it was thought that the TCNQ derivative **5**-CN^{2-} may exhibit very low energy electronic transitions.

Table II gives these results and comparable data. **5**-O$^{\bullet-}$ has a strong band at 1020 nm, the dianion **5**-O^{2-}, even stronger at 920 nm. These $\lambda_{\max}$ values for **5**-O$^{\bullet-}$ and **5**-O^{2-} (near 1000 nm) are intermediate between those for **6**$^{\bullet-}$, **6**$^{2-}$ (near 600 nm) and **7**$^{\bullet-}$, **7**$^{2-}$ (near 1800 nm). Most notable is the *400-nm shift resulting from substitution of four cyano groups on* **6**$^{\bullet-}$ *to give* **5**-O$^{\bullet-}$. In comparison, for the neutrals, adding four cyano groups to **6** to give **5**-O gives only a 25-nm shift. It is suggested that the 400-nm shift has the same origin as the unusual NIR bands in **7**$^{\bullet-}$ and certain diquinone$^{\bullet-}$. In this model the dicyanobenzo groups (like quininoe groups in **7**$^{\bullet-}$) interact strongly with the quinone$^{\bullet-}$ and the spectrum is therefore strongly perturbed.

Turning finally to consideration of **5**-CN^{2-}, we find that this dianion provides a pair of transitions at 1060 and 590, but not at wavelengths much longer than those for **5**-O^{2-}. The absence of very long wavelength NIR absorption for **5**-CN^{2-} is not surprising considering its conformational situation, which was described above.

Experimental Section

General. ^{1}H NMR (300 MHz) and ^{13}C NMR (75.4 MHz) spectra were measured in deuteriochloroform on an IBM-NR-300-AR spectrometer. Chemical shifts are in ppm downfield of tetramethylsilane and carbon shifts have the number of attached protons in parentheses. Coupling constants are in hertz. Variable-temperature studies were conducted in toluene-d_8, using a Nicolet NT-300-WB spectrometer. Infrared spectra (KBr pellet) were mesured on a Perkin-Elmer 7100 Fourier transform instrument and were calibrated with the 1601.8 cm^{-1} absorption of polystyrene. Fast atom bombardment (FAB) mass spectra were obtained on a VG-7070E-HF instrument. Elemental analyses were performed by M-H-W Laboratories, Phoenix, AZ.

Cyclic voltammetry was performed on a BAS-100 electrochemical analyzer with IR compensation. Anhydrous DMF was used as obtained from Aldrich Chemical Company. Bu_4NBF_4 was obtained from Southwestern Analytical Chemical Company and recrystallized in 3:1 water/methanol. A single-compartment three-electrode cell was used. The working electrode was a glassy carbon disk and the reference was SCE. Redox potentials are measured with respect to an internal standard of ferrocene, whose anodic couple was set at +0.47 V. It was shown that the presence of this internal standard had no effect on the cathodic couples of interest. Bulk electrolyses were performed at constant potential on a PAR-173 potentiostat. A simple two-compartment cell was used with a carbon felt working electrode. Vis–NIR spectra of the electrochemically prepared anion solutions were measured in a 0.1-cm quartz cell adapted for inert atmosphere work. A Cary-14 spectrometer was used, and background absorbances were subtracted.

Preparation of 5-O. Fumaronitrile (123 mg, 1.58 mmol) and **4** (137 mg, 0.168 mmol)[4] in chloroform (30 mL) were stirred under nitrogen in the dark for 30 h to give a brownish solution. The solvent was removed and the resulting solid was agitated with concentrated sulfuric acid (40 mL) for 60 min by using a vortex mixer. The resulting mixture was poured onto ice and extracted with three portions of chloroform. The chloroform extracts were washed with brine and dried over anhydrous magnesium sulfate and the solvent was removed to give a brownish solid. The crude product was purified by flash chromatography (230–400-mesh silica gel, chloroform) to give a bright, golden yellow product (86 mg, 55%). ^{1}H NMR: 8.89 (s, 4 H), 7.65 (d, J = 8.4, 8 H), 7.43 (d, J = 8.4, 8 H), 1.45 (s, 36 H). ^{13}C NMR: 181.1 (0), 153.7 (0), 149.3 (0), 135.6 (0), 132.4 (0), 130.8 (0), 130.2 (1), 126.4 (1), 115.2 (0), 114.0 (0), 35.0 (0), 31.3 (0). IR: 2223, 1689. UV-vis (CH_2Cl_2) $\lambda_{\max}$ (log ϵ): 425 nm, (4.22), 282 (4.97). High-resolution mass spectrum: 937.4551, corresponding to protonated molecular ion; calcd for $C_{66}H_{57}N_4O_2$ 937.4481.

Anal. Calcd for $C_{66}H_{56}N_4O_2$: C, 84.58; H, 6.02; N, 5.98. Found: C, 84.74; H, 6.19; N, 5.77.

Preparation of 5-CN. Titanium tetrachloride (0.120 mL, 1.09 mmol) was added under nitrogen to a stirred solution of **5**-O (88 mg, 0.09 mmol) in dry 1,1,2,2-tetrachloroethane (35 mL). The golden yellow solution at once became dark brown and possibly heterogeneous and was then heated until gently boiling under reflux to give a lighter orange-brown solution. A solution of malononitrile (218 mg, 3.30 mmol) and 1,8-bis(dimethylamino)naphthalene (915 mg, 4.37 mmol) in dry, 1,1,2,2-tetrachloroethane (35 mL) was added dropwise to the boiling reaction mixture over 15 min. The reaction mixture became darker and heterogeneous and was refluxed for 60 min, after which TLC showed that all the starting material had reacted. The reaction mixture was cooled to room temperature and taken in up chloroform to give a brown solution which was then washed twice with 0.5 M sulfuric acid and once with brine and then was dried over anhydrous magnesium sulfate. After the drying agent and any other suspended matter were filtered off, the solution was evaporated to give a dark brown solid. This was washed several times with methanol, using a centrifuge to separate the product from the washings. The brownish yellow product obtained was further purified by flash chromatography (230–400-mesh silica gel, 50/1 chloroform/ethyl acetate) to give **5**-CN as a yellow solid (17 mg, 18%). ^{1}H NMR ($CDCl_3$, 25 °C): 8.59 (s, 4 H), 7.71 (d, J = 7.9, 4 H), 7.62 (d, J = 7.9, 4 H), 7.51 (d, J = 7.9, 4 H), 7.43 (d, J = 7.9, 4 H), 1.43 (s,

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(9) Nepras, M.; Titz, M. *Int. J. Quantum Chem.* **1979**, *16*, 543.

36 H). ^{13}C NMR (25 °C): 158.2 (0), 153.9 (0), 148.3 (0), 133.5 (0), 131.1 (1), 129.9 (0), 129.4 (0), 129.3 (1), 128.9 (1), 126.7 (1), 126.3 (1), 115.0 (0), 114.6 (0), 112.2 (0), 86.4 (0), 35.1 (0), 31.3 (3). IR: 2229. UV-vis (CH_2Cl_2) λ_{max} (log ϵ): 432 nm (4.19), 328 (4.75), 292 (4.94). High resolution mass spectrum: molecular ion 1032.4628; calcd for $C_{72}H_{56}N_8$ 1032.4630.

Acknowledgment. Preliminary electrochemistry was carried out by J. R. Valentine, the variable-temperature ^{1}H NMR study was conducted by R. King, and mass spectra were measured by Dr. E. Larka. This work was supported by a grant from the Office of Naval Research.

Supplementary Material Available: NMR data of **5**-CN (1 page). Ordering information is given on any current masthead page.

Antitumor Xenicane and Norxenicane Lactones from the Brown Alga *Dictyota dichotoma*

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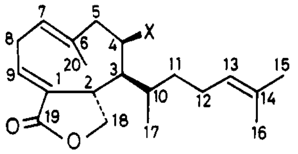
New xenicane diterpenes, 4-acetoxydictyolactone (**1**), dictyotalide A (**6**), dictyotalide B (**3**), and nordictyotalide (**7**), possessing a cyclononane carbon framework, have been isolated from the brown alga *Dictyota dichotoma*, and their structures have been spectroscopically and chemically elucidated. The conformations of these compounds have been determined on the basis of 2D-NOESY spectra. Dictyotalide B (**3**) possesses a highly strained double bond which is located at a bridgehead position of the 9-oxabicyclo[6.2.1]undecane skeleton. The compounds exhibit significant cytotoxic activity against B16 mouse melanoma cells.

Brown algae of the Dictyotaceae family are peculiar in that they produce a series of unique diterpenes consisting of medium-sized rings.[1] Among the diterpenes isolated from seaweeds, xenicanes comprise one of the most interesting classes of compounds because they are composed of a cyclononane skeleton that is seldom found in other natural products. Xenicanes have also been isolated as the ingredients of soft corals and sponges.[2] Herein we describe the structures of four new xenicane lactones from the brown alga *Dictyota dichotoma*, viz., 4-acetoxydictyolactones (**1**), dictyotalide A (**6**) and B (**3**), and nordictyotalide (**7**).

4-Acetoxydictyolactone (**1**), $[\alpha]_D$ −224° (*c* 0.86, $CHCl_3$), $C_{22}H_{32}O_4$ (*m/e* 360.2290), shows IR absorption bands at 1760 and 1740 cm^{-1} assignable to γ-lactone and acetoxy [^{1}H NMR δ 2.03 (3 H, s)] groups, respectively. Analysis of the ^{1}H and ^{13}C NMR (Experimental Section) and mass (*m/e* 109) spectra revealed that a 6-methyl-5-hepten-2-yl group (**8**), the typical side chain of the Dictyotaceae diterpenes, was present. The ^{1}H and ^{13}C NMR spectra were very similar to those of dictyolactone (**2**).[3] The placement of an acetoxy group at C-4 was determined on the basis of the coupling pattern for H-4 [δ 5.27 (dd, *J* = 4.5, 2.3 Hz)]. Consideration of the coupling patterns of the ring protons together with observation of the NOEs (depicted in **1a**) enabled us to propose the relative stereochemistry at C-2, -3, and -4 and the conformation (**1a**) for 4-acetoxydictyolactone. The conformation is essentially the same as that of dictyolactone (**2**) inferred by X-ray analysis.[3]



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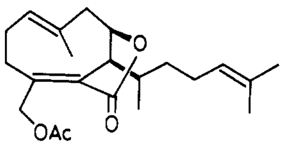
2; X = H



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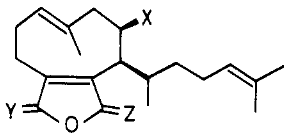
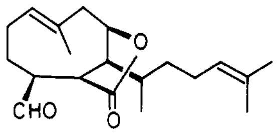


4; X=OAc, Y=O, Z=H$_2$
5; X=OH, Y=H$_2$, Z=O



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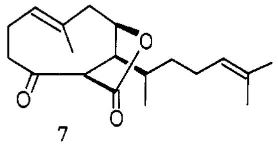

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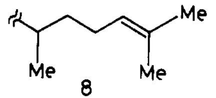

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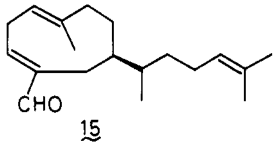

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Dictyotalide B (**3**), $C_{22}H_{32}O_4$ (*m/e* 360.2338), $[\alpha]_D$ +50.3° (*c* 0.59, $CHCl_3$), is an isomer of **1**. The presence of a γ-lactone ring, an acetoxy group, the side chain **8**, and a trisubstituted olefin bearing a methyl group was apparent from the spectral properties (Table I). The ^{1}H NMR spectrum of dictyotalide B lacked the downfield olefin proton signal corresponding to H-9 of **1**, strongly suggesting

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[1] Faulkner, D. J. *Nat. Prod. Rep.* **1984**, *1*, 251.

[2] Faulkner, D. J. *Nat. Prod. Rep.* **1984**, *1*, 551.

[3] Finer, J.; Clardy, J.; Fenical, W.; Minale, L.; Riccio, L.; Battaile, J.; Kirkup, M.; Moore, R. *J. Org. Chem.* **1979**, *44*, 2044.

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