

*The Chemistry of Humulene. Part V.**

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*cyclo*Nona-1 : 3-diene, *cyclodeca*-1 : 3-diene, and *cycloundeca*-1 : 3-diene have been prepared from the corresponding monoenes. The ultra-violet absorption maxima and refractive indices of these substances show a marked correlation with ring size. Infra-red absorption spectra are also recorded.

Comparison of the chemical and physical properties of these compounds with those of humulene confirms the absence of conjugation in the latter. This, together with the original and newer degradative and spectral evidence, leads to the most probable formation of humulene as 2 : 6 : 6-trimethyl-9-methylenecycloundeca-1 : 4-diene (IIIa) or 2 : 5 : 5-trimethyl-9-methylene-cycloundeca-1 : 6-diene (V).

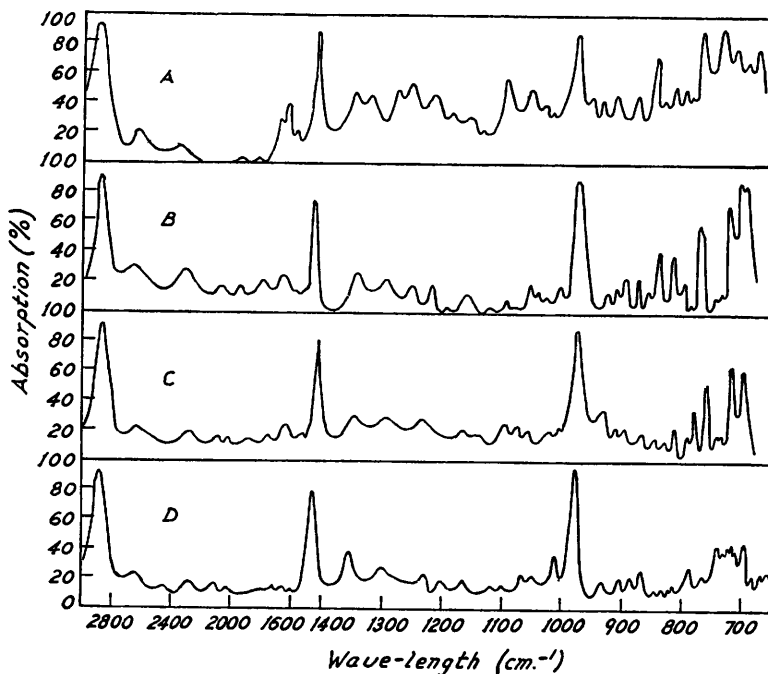
THE structure of the sesquiterpene humulene as a 1 : 4 : 4 : 8-tetramethylcycloundecatriene has been indicated by degradative and synthetic evidence, and from consideration of physical constants (for references see Fawcett and Harris, Part IV *). The exact arrangement of the double bonds has however not been deduced. It was considered from the absence of absorption above 2300 Å, and the failure to obtain a maleic anhydride adduct or to reduce humulene with sodium and alcohol, that the natural hydrocarbon was unconjugated. Pesch and Friess's observation (*J. Amer. Chem. Soc.*, 1950, **72**, 5756) that the ultra-violet absorption maxima of *cyclo*hepta- and *cyclo*octa-1 : 3-dienes occur at shorter wave-lengths than that of *cyclo*hexa-1 : 3-diene, but absence of information regarding the hitherto undescribed nine-, ten-, and eleven-membered ring compounds, rendered the ultra-violet absorption measurements inconclusive. Similarly the inability to form a maleic anhydride adduct from *cyclo*octa-1 : 3-diene (Cope and Estes, *ibid.*, 1950, **72**, 1128; Ziegler and Wilms, *Annalen*, 1950, **567**, 1) and lack of experimental evidence for the immediately higher ring homologues made further information desirable. So *cyclonona*-, *cyclodeca*-, and *cycloundeca*-1 : 3-diene have been prepared, all as colourless liquids by allylic bromination of the corresponding *trans*-monoenes with *N*-bromosuccinimide followed by dehydrohalogenation with quinoline. *cyclo*Deca-1 : 3-diene was also prepared by Hofmann degradation of *cycloundec*-2-enyltrimethylammonium iodide and, after purification, was identical (infra-red spectrum) with the sample obtained by the alternative method. The conjugated dienes were purified by chromatography on alumina. All three showed marked absorption (see Fig. 1) at 975 cm.⁻¹, characteristic of a *trans*-disubstituted olefinic linkage. They therefore possess a *cis-trans*-configuration, since a *trans-trans*-arrangement can only be accommodated, on Stuart models, for *cyclododeca*-1 : 3-diene and higher homologues. However, the infra-red absorption pattern in the 950—1020 cm.⁻¹ region in all cases resembles more closely that recently associated with a *trans-trans*-configuration in aliphatic compounds—for *cis-trans*-compounds bands of similar intensity occur at 950, 985, and 1020

* Part IV, preceding paper.

cm^{-1} (Celmer and Solomons, *J. Amer. Chem. Soc.*, 1953, **75**, 1372), for the presence of which there is some indication also in *cis-trans-cycloocta-1 : 3*-diene (cf. Cope and Estes, *loc. cit.*). A definite decision on the stereoisomeric form of these new dienes is therefore not possible.

cycloUndecene, not previously described, was prepared by dehydration of *cycloundecanol* with phthalic anhydride. This leads to a mixture of *cis*- and *trans-cyclononenes* from *cyclononanol*, in which case Stuart models show the *cis*-form to be less strained, whereas in the case of *cyclodecanol* where models show both *cis*- and *trans-cyclodecene* to be strainless only the *trans*-isomer is formed. Hence it may be assumed that in this preparation of *cycloundecene*, where both geometric isomers are also equally strainless, the *trans*-isomer is also produced. This is confirmed by the marked absorption at 975 cm^{-1} .

FIG. 1.



Ultra-violet absorption spectra for : A, cyclonona-1 : 3-diene ; B, cyclodeca-1 : 3-diene ; C, cycloundeca-1 : 3-diene ; D, *trans*-cycloundecene.

The hitherto undescribed *cycloundecane-1 : 2*-dione was also prepared by the chromic acid oxidation of the corresponding acyloin, and characterised.

The undecane-1 : 11-dioic acid required for this synthesis was prepared by Sauer's method (*ibid.*, 1947, **69**, 2444) from δ -ethoxyvaleryl chloride. Also, undecylenic ester epoxide (Lévy and Willisich, *Bull. Soc. chim.*, 1929, **45**, 930) on catalytic hydrogenation over Raney nickel at $156^\circ/60 \text{ atm}$. gave a mixture of 11- and 10-hydroxyundecanoic acid, in which the former predominated. Oxidation of this mixture gave the required undecane-1 : 11-dioic acid and 10-oxoundecanoic acid, which were readily separated.

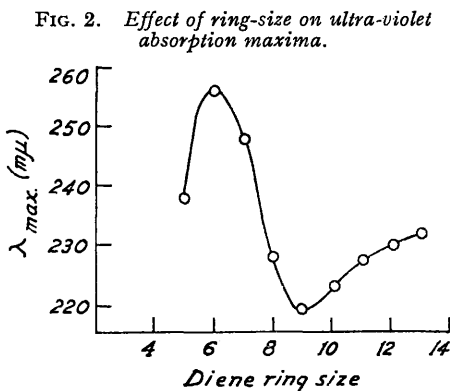
Attempts to obtain an adduct with maleic anhydride and *cyclonona*-, *cyclodeca*-, or *cycloundeca-1 : 3*-diene were abortive. In the possible Stuart models of the *cis-trans*-configurations, whilst all three dienes can assume a "cisoid"-form (cf. Alder and Schumacher, *Annalen*, 1950, **571**, 81), the adducts are all strained, this being least in the C_{11} compound. It may be noted that, although *cyclododeca-1 : 3*-diene forms a liquid adduct with maleic anhydride, this cannot be dehydrogenated but redissociates on pyrolysis (Bartlett, Figdor, and Wiesner, *Canad. J. Chem.*, 1952, **30**, 291).

From these observations coupled with that of Cope and Estes (*loc. cit.*), this reaction

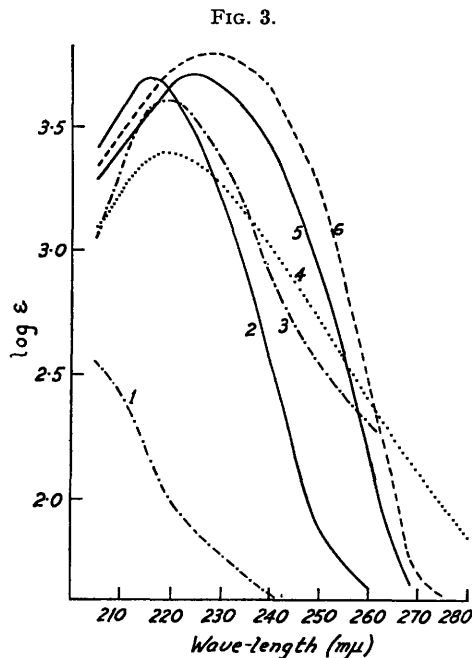
cannot be taken as a criterion for the presence of a conjugated *endo-endo*-system in C_{8-11} cyclic dienes.

The ultra-violet absorption measurements on the dienes showed that the smallest value for the maximum was reached at the C_9 compound (λ_{\max} , 2195 Å) (Figs. 2 and 3). *cyclo*-Undeca-1 : 3-diene has λ_{\max} , 2275 Å, and a pure sample of humulene showed a maximum of 2155 Å. This therefore appears to exclude the presence of an *endo-endo*-conjugated diene grouping in humulene in which the additional alkyl groups would be expected to produce a bathochromic shift.

The refractive indices also show a minimum value at *cyclonona*-1 : 3-diene.



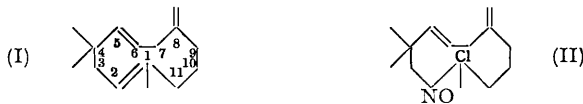
References : 1, Pesch and Friess, *J. Amer. Chem. Soc.*, 1950, **72**, 5756; 2, Bartlett, Figdor, and Wiesner, *Canad. J. Chem.*, 1952, **30**, 291; 3, Ingraham, MacDonald, and Wiesner, *Canad. J. Res.*, 1950, **28**, B, 453.



Ultra-violet absorption spectra : 1, cycloundecene; 2, humulene; 3, dihydrohumulene; 4, *cyclonona*-1 : 3-diene; 5, *cyclodeca*-1 : 3-diene; 6, *cycloundeca*-1 : 3-diene.

The fact that *cyclodeca*- and *cycloundeca*-1 : 3-diene undergo normal reduction to the corresponding monoenes with sodium and alcohol also supports the absence of conjugation in humulene.

Structure (I) is the only formula, based upon a 1 : 4 : 4 : 8-*cycloundecane* skeleton, in which a precise designation of the double bonds of humulene has been given (Šorm, Streibl,

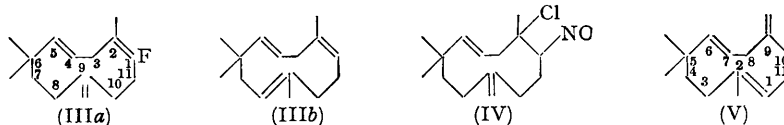


Pliva, and Herout, *Chem. Listy*, 1951, **45**, 308; 1953, **46**, 30). Consideration of all the possible trebly unsaturated structures which may be based on this skeleton leads to 5 : 6 as the best position for the R·CH:CH·R system, the presence of which is clearly shown by the absorption at 975 cm^{-1} (Šorm, *Coll. Czech. Chem. Comm.*, 1949, **14**, 718). No other position leads to a non-conjugated structure which accounts readily for $\alpha\alpha$ -dimethylsuccinic acid and *l*ævulaldehyde as the first products of ozonolysis (Clemo and Harris, *loc. cit.*). The nitrosochloride is probably formed at the endocyclic disubstituted olefinic linkage. This is supported by the strong absorption at 980 cm^{-1} in this derivative indicative of the presence

still of $R\cdot CH:CH\cdot R$; and absorption at 890 cm.^{-1} shows the retention of the $CH_2\cdot CR_2$ group also present in humulene. Presence of the exocyclic methylene group is confirmed in the nitrosochloride by the formation of formaldehyde on ozonolysis. Thus, on these grounds, formula (I) would lead to (II) as the structure of the nitrosochloride, which cannot readily accommodate all the degradative evidence, since reduction to the fully saturated amine followed by methylation, Hofmann degradation, and oxidation (Clemo and Harris, *J.*, 1952, 665) would not lead to a C_{15} acid of the established structure (preceding paper).

However the structure (IIIa) now suggested, with some co-existent endocyclic form (IIIb), whilst accounting satisfactorily for the products of ozonolysis and the spectral data would lead to (IV) as the formula of the nitrosochloride. This would give on the above degradation 2 : 6 : 6 : 9-tetramethylundecane-1 : 11-dioic acid, a permissible structure.

A unique structure which accounts directly for most of the degradative evidence is (V). This leads to the other permissible structure for the C_{15} degradation acid, and the lävul-



aldehyde could arise by decarboxylation of an intermediate β -keto-acid. However, the dimethylsuccinic acid can only arise if allylic oxidation is assumed to occur (cf. Ruzicka and Kaufmann, *Helv. Chim. Acta*, 1940, 23, 1346). The choice between these structures can obviously be arrived at by the exact formulation of the C_{15} degradation acid.

EXPERIMENTAL

For the infra-red spectra see preceding paper. Ultra-violet spectra were measured on a Hilger Unispectrometer.

3-Bromocyclodecene.—*trans-cyclo*Decene (4.5 g.) (Blomquist, *J. Amer. Chem. Soc.*, 1952, 74, 3636) in carbon tetrachloride (22 c.c.) was heated for 45 min. at 100° , under nitrogen, with *N*-bromosuccinimide (5.8 g.) and benzoyl peroxide (50 mg.). The mixture was filtered, the filtrate washed with dilute sodium hydrogen carbonate solution and dried ($MgSO_4$), the solvent removed, and the residue distilled rapidly and then refractionated. *3-Bromocyclodecene* distilled as a colourless liquid which soon became dark red (4.0 g., 56%), b. p. $86-88^\circ/2$ mm. (Found : C, 55.6; H, 8.1. $C_{10}H_{11}Br$ requires C, 55.3; H, 7.8%).

*cyclo*Deca-1 : 3-diene.—(a) *By dehydrohalogenation.* *3-Bromocyclodecene* (4.0 g.) and quinoline (4.8 g.), both freshly distilled, were heated at 230° in a short-necked flask attached to a cooled receiver. The distillate (b. p. $165-185^\circ$) was washed with 5% hydrochloric acid (10 c.c.), extracted with ether, separated, and dried (Na_2SO_4); after removal of the solvent the diene distilled as a colourless, highly mobile liquid with a strong characteristic odour (2.2 g., 45%), b. p. $76-77^\circ/14$ mm. A sample, purified by chromatography on an alumina column, elution being with *n*-hexane, had n_D^{25} 1.4972, λ_{max} (in EtOH) 223 μ ($\log \epsilon$ 3.70) (Found : C, 88.4; H, 11.9. $C_{10}H_{16}$ requires C, 88.2; H, 11.8%). Quantitative hydrogenation over Adams catalyst in glacial acetic acid at room temperature and pressure confirmed the presence of two double bonds. With concentrated silver nitrate solution the diene forms a white addition compound, m. p. $176-178^\circ$ (decomp.).

(b) *By Hofmann degradation.* *3-Bromocyclodecene* (8.5 g.) and dimethylamine (4.4 g.) in benzene (37 c.c.) were kept overnight; dimethylamine hydrobromide was filtered off, the filtrate was extracted with 10% hydrochloric acid (15 c.c.), and the extracts were warmed at $55-60^\circ$ for 3-5 min. to hydrolyse any vinylamines present, and extracted with ether (15 c.c.). The aqueous solution was basified and extracted with ether; the extract was dried (Na_2SO_4) and evaporated, and distillation of the residue yielded colourless *3-dimethylaminocyclodecene* (3.2 g., 45%), b. p. $88-90^\circ/1$ mm. (Found : C, 79.6; H, 13.0. $C_{12}H_{23}N$ requires C, 79.6; H, 12.7%). The *picrate* recrystallised from alcohol in long fine needles, m. p. 158° (Found : C, 52.3; H, 6.5. $C_{18}H_{26}O_2N_4$ requires C, 52.7; H, 6.3%).

The base (3.1 g.) and methyl iodide (5.0 g.) in *cyclohexane* (38 c.c.) were heated under reflux for $2\frac{1}{2}$ hr. The quaternary salt was filtered off (4.8 g., 84%). A sample crystallised from ethanol had m. p. $205-208^\circ$ (decomp.) (Found : C, 48.3; H, 8.5. $C_{13}H_{26}NI$ requires C, 48.3; H, 8.1%).

This salt (7.0 g.) was stirred for 2 hr. at room temperature with an excess of freshly prepared

silver oxide suspended in water (50 c.c.). The silver halide were filtered off, stirred with water (50 c.c.), and filtered off again, and the combined aqueous fractions (100 c.c.) were treated with solid sodium hydroxide (100 g.). The solution was heated under reflux for $1\frac{1}{2}$ hr. The oil which separated was extracted with ether, dried (Na_2SO_4), and distilled. *cyclo*Deca-1 : 3-diene (2.2 g., 70%) was obtained as a colourless liquid, b. p. $89-91^\circ/24$ mm., n_D^{25} 1.4972 (Found : C, 88.1; H, 12.1%). This material, after chromatographic purification as described previously, was identical with the material obtained as above.

*cyclo*Deca-1 : 3-diene (250 mg.) and maleic anhydride (710 mg.) in dry xylene (20 c.c.) were refluxed overnight in an atmosphere of nitrogen. Removal of the solvent yielded only the unchanged starting materials.

Sodium and Alcohol Reduction.—*cyclo*Deca-1 : 3-diene (400 mg.) in absolute alcohol (10 c.c.) was heated under reflux, sodium (2.0 g.) was added in small portions during 30 min., and heating continued for 1 hr. The mixture was cooled, diluted with water, and extracted with ether, the extract was evaporated, and the residue shaken with 50% aqueous silver nitrate (0.5 c.c.). The solid from the unreduced diene was filtered off and washed with ether, and this ether was combined with the ether extract of the filtrate. The ethereal solutions were dried (Na_2SO_4) and concentrated, and the residue was distilled. Quantitative reduction of the product showed the presence of one double bond.

3-Bromocyclononene.—*cyclo*Nonene (2.2 g.), prepared by the phthalic anhydride dehydration of cyclononanal (*idem, ibid.*, p. 3643) was brominated as described for cyclodecene. *3-Bromo-cyclononene* (1.6 g., 45%) distilled as a colourless liquid which gradually darkened (Found : C, 53.0; H, 7.4. $\text{C}_9\text{H}_{15}\text{Br}$ requires C, 53.2; H, 7.4%).

*cyclo*Nona-1 : 3-diene.—The foregoing bromide (1.5 g.) was dehalogenated as described for the cyclodecene derivative. The colourless diene (0.4 g., 42%) had b. p. $105-107^\circ/170$ mm., and a characteristic odour. After adsorption on alumina and elution with *n*-hexane it had n_D^{25} 1.4921, λ_{max} (in EtOH) 219.5 μ . ($\log \epsilon$ 3.40) (Found : C, 88.7; H, 11.8. C_9H_{14} requires C, 88.5; H, 11.5%). Neither a maleic anhydride adduct nor an addition compound with silver nitrate could be prepared.

Undecane-1 : 11-dioic Acid and Ethyl 10-Oxoundecanoate.—Ethyl 10 : 11-epoxyundecanoate (10.0 g.) was catalytically reduced over Raney nickel (2 g.) in absolute alcohol (120 c.c.) at $150^\circ/60$ atm. until uptake of hydrogen ceased (3 hr.). The catalyst and alcohol were removed, the residue was distilled, and the distillate (8.0 g., 76%), b. p. $135-140^\circ/0.8$ mm., collected.

Chromic anhydride (8.7 g.) in concentrated sulphuric acid (4 c.c.) and water 40 c.c.) was added slowly at $<25^\circ$ with stirring to the hydroxy-ester (7.8 g.) in acetone (95 c.c.), and the mixture allowed to reach room temperature overnight. The acetone was removed under reduced pressure and the residue extracted with ether. The acidic products were extracted by sodium carbonate solution, then re-extracted after acidification into ether, the solvent was removed, and the residue was hydrolysed for 4 hr. with potassium hydroxide (5.2 g.) in water (25 c.c.). Acidification gave colourless undecane-1 : 11-dioic acid (5.0 g.), m. p. 101° undepressed on admixture with an authentic sample prepared by Sauer's method (*loc. cit.*) (Found : C, 61.3; H, 9.6. Calc. for $\text{C}_{11}\text{H}_{20}\text{O}_4$: C, 61.1; H, 9.3%).

The above ethereal extract of neutral oxidation products was dried (Na_2SO_4), the ether removed, and the neutral residue distilled, to give colourless ethyl 10-oxoundecanoate (1.3 g., 13%), b. p. $151-154^\circ/5$ mm. (Found : C, 68.7; H, 10.8. Calc. for $\text{C}_{13}\text{H}_{24}\text{O}_2$: C, 68.4; H, 10.5%). This yielded in alcohol the 2 : 4-dinitrophenylhydrazone, in orange needles, m. p. 65° , from alcohol (Found : C, 56.0; H, 7.0. $\text{C}_{13}\text{H}_{28}\text{O}_2\text{N}_4$ requires C, 55.9; H, 6.9%).

2-Hydroxycycloundecanone.—Diethyl undecane-1 : 11-dioate (53.0 g.) in xylene (400 c.c.) with sodium was cyclised by the acyloin procedure (*cf. Blomquist, loc. cit.*). A crude pale yellow acyloin fraction, b. p. $95-100^\circ/0.4$ mm., was obtained. *2-Hydroxycycloundecanone* (12.5 g., 36%) was obtained pure by cooling a light petroleum (b. p. $40-60^\circ$) solution; it had m. p. 38° (Found : C, 71.7; H, 11.1. Calc. for $\text{C}_{11}\text{H}_{20}\text{O}_2$: C, 71.7; H, 10.9%).

*cyclo*Undecanone.—*2-Hydroxycycloundecanone* (10.7 g.), acetic acid (24 c.c.), zinc wool (24 g.), and concentrated hydrochloric acid (24 c.c.) were heated under reflux at $100-110^\circ$. Three similar quantities of hydrochloric acid were added at half-hourly intervals, after which heating was continued for 2 hr. The solution was filtered, diluted, and extracted with ether, the extract was washed with dilute sodium carbonate solution and then water and dried (Na_2SO_4), the ether removed, and the residue distilled. *cyclo*Undecanone (8.6 g., 88%) was obtained as a colourless liquid, b. p. $100-103^\circ/2$ mm. (Found : C, 78.5; H, 12.0. Calc. for $\text{C}_{11}\text{H}_{20}$: C, 78.6; H, 11.9%).

*cyclo*Undecanol.—*cyclo*Undecanone (8.6 g.) in dry ether (10 c.c.) was added with stirring to

a solution of lithium aluminium hydride in ether (45 c.c., 100-vol.)* so that refluxing was just maintained. Excess of reagent was destroyed by water, and the complex decomposed with 10% sulphuric acid (65 c.c.). The ether layer, and ether extracts of the aqueous layer, were dried (Na_2SO_4), the solvent was removed, and the residue distilled. *cycloUndecanol* (7.8 g., 91%) was obtained as a colourless viscous liquid, b. p. 79—82°/0.2 mm. (Found: C, 77.5; H, 13.2. Calc. for $\text{C}_{11}\text{H}_{22}\text{O}$: C, 77.7; H, 12.9%).

trans-cycloUndecene.—*cycloUndecanol* (7.7 g.) was added in portions to phthalic anhydride (16.0 g.), heated to b. p. in a Claisen flask with a refractionating column packed with carborundum lumps; when the addition was nearly complete the temperature was raised to 340°. The mixture of olefin and phthalic anhydride which distilled was warmed with 10% sodium hydroxide solution, cooled, and extracted with ether, the extract was dried (Na_2SO_4), the solvent removed, and the residue distilled. *trans-cycloUndecene* (5.7 g., 80%) distilled as a colourless liquid, b. p. 116—119°/25 mm. Purified by chromatography on an alumina column and elution with hexane this had n_D^{25} 1.4802, λ_{max} . (in EtOH) <200 m μ (Found: C, 87.0; H, 13.4. $\text{C}_{11}\text{H}_{20}$ requires C, 86.9; H, 13.1%).

3-Bromocycloundecene.—The above olefin (5.7 g.) was brominated as described for *cycloUndecene*. *3-Bromocycloundecene* (5.7 g., 66%) distilled as a colourless liquid, b. p. 116—119°/3 mm., which soon darkened (Found: C, 57.3; H, 8.6. $\text{C}_{11}\text{H}_{19}\text{Br}$ requires C, 57.2; H, 8.2%).

cycloUndeca-1:3-diene.—The above bromide (5.7 g.) was dehalogenated as described for the *cyclodeca-1:3-diene* derivative. *cycloUndeca-1:3-diene* (2.2 g., 59%) distilled as a highly mobile liquid with a characteristic odour, b. p. 107—109°/17 mm.; purified by chromatography on alumina and elution with *n*-hexane, it had n_D^{25} 1.5034, λ_{max} . (in EtOH) 225 m μ ($\log \epsilon$ 3.79) (Found: C, 87.8; H, 11.9. $\text{C}_{11}\text{H}_{18}$ requires C, 88.0; H, 12.0%). Quantitative hydrogenation over Adams catalyst in glacial acetic acid confirmed the presence of two double bonds. It gave with silver nitrate solution a white addition compound, m. p. 155° (decomp.), from which the diene was regenerated on shaking with a saturated solution. Attempts to make the maleic anhydride adduct, as described above, yielded only the starting materials. The diene was reduced by sodium and alcohol to *cycloundecene* under conditions similar to those described above.

Ultra-violet Absorption of Humulene.—A sample of humulene (Clemo and Harris, *J.*, 1952, 22) was purified by chromatography on an alumina column and elution with *n*-hexane. A maximum at 215.5 m μ ($\log \epsilon$ 3.69) in EtOH was observed.

Ozonolysis of Humulene Nitrosochloride.—Ozone was passed through a solution of humulene nitrosochloride (4.0 g.) in chloroform (50 c.c.) at 0°, to saturation. The solvent was removed under reduced pressure at <20° and the residue steam-distilled. The distillate, treated with dimedone solution, gave a solid which separated from alcohol as white needles, m. p. 189° alone or mixed with the dimedone formaldehyde compound. In Nujol mull in the 10—12 μ region, the pure nitrosochloride showed bands at 1018, 998, 980, 954, 942, 919, 910, 892, 870, 837, and 826 cm^{-1} .

cycloUndecane-1:2-dione.—2-Hydroxycycloundecanone (1.0 g.) in acetic acid (2 c.c.) was cooled and a cold solution of chromic anhydride (0.6 g.) in water (1 c.c.) and acetic acid (2.5 c.c.) was added slowly with stirring. Next morning saturated sodium chloride solution (5 c.c.) was added and the product extracted with ether. The extract was washed with dilute sodium carbonate solution and then water, dried (Na_2SO_4), and evaporated, and the residue was distilled. *cycloUndecane-1:2-dione* (0.5 g., 50%) distilled as a bright yellow liquid, b. p. 87—90°/2 mm. (Found: C, 72.8; H, 10.3. $\text{C}_{11}\text{H}_{18}\text{O}_2$ requires C, 72.5; H, 10.0%). The red *bis-2:4-dinitrophenylhydrazone* had m. p. 285—286° (Found: C, 50.1; H, 5.0. $\text{C}_{23}\text{H}_{26}\text{O}_8\text{N}_8$ requires C, 50.9; H, 4.8%). The diketone in glacial acetic acid was warmed with *o*-phenylenediamine in alcohol, and the *quinoxaline* which separated recrystallised from alcohol as white needles, m. p. 145° (Found: C, 80.3; H, 9.0. $\text{C}_{17}\text{H}_{22}\text{N}_2$ requires C, 80.3; H, 8.7%).

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* See footnote, p. 2670.