

Studies in Quinoxaline Series. Part 18.¹ Structure of Products of a New Reaction of Tetrazolo[1,5-*a*]quinoxaline 5-Oxide with Carbanions. X-Ray Molecular Structure of 4-Acetyl-4-methyl-3b,4-dihydroazirino[1,2-*a*]tetrazolo[5,1-*c*]quinoxaline

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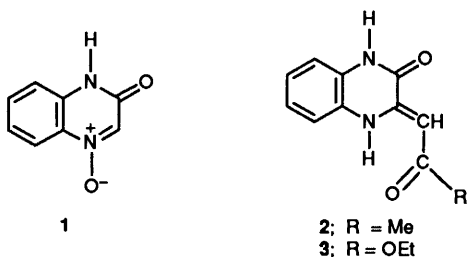
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Tetrazolo[1,5-*a*]quinoxaline-5-oxide **4** reacts with the carbanions of β -diketones and β -keto esters to give enaminketones and enamino esters, respectively. The *N*-oxide **4** reacts with 3-methyl- and 3-ethyl-pentane-2,4-dione under similar conditions to give a racemic mixture of the derivatives of aziridinotetrazoloquinoxaline **10** and **11**. Their structure and the *trans* configuration of hydrogen atom and methyl group on the aziridine ring of compound **10** were estimated by means of ¹H and ¹³C NMR spectroscopy, and were in accord with the results of X-ray diffraction measurements, which also provided bond lengths, and valence, torsion, and dihedral angles.

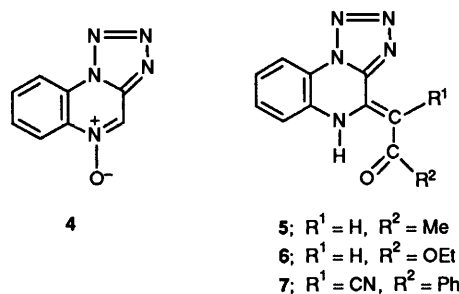
3,4-Dihydro-3-oxoquinoxaline 1-oxide **1** undergoes condensation² with pentane-2,4-dione in boiling ethanol in the presence of piperidine to give 2-acetylmethylene-3-oxo-1,2,3,4-tetrahydroquinoxaline **2**. Both the oxide **1** and its substitution derivatives³ also react with other *C*-acids; e.g., the reaction of the oxide **1** with ethyl acetoacetate under similar conditions produces 2-ethoxycarbonylmethylene-3-oxo-1,2,3,4-tetrahydroquinoxaline **3**. These reactions were shown^{4,5} to proceed *via* nucleophilic attack of the carbanion of the *C*-acid on the C-2 of the oxide **1** with participation of the *N*-oxide oxygen atom *via* an isoxazolidine intermediate which is subsequently deacylated. No such reactions have been observed with quinoxaline 1-oxides which do not contain a 3-oxo and/or a 3-nitro group.⁶



Results and Discussion

We presumed that tetrazolo[1,5-*a*]quinoxaline 5-oxide **4** would react in a similar way with carbanions. The reaction with pentane-2,4-dione gave 4-acetylmethylene-4,5-dihydro-tetrazolo[1,5-*a*]quinoxaline **5**, whose enamine structure was verified by means of IR and NMR spectroscopy. Under similar conditions the *N*-oxide **4** reacts with ethyl acetoacetate to give 4-ethoxycarbonylmethylene-4,5-dihydro-tetrazolo[1,5-*a*]quinoxaline **6**, which is identical with the reaction product⁸ of ethyl (3-chloroquinoxalin-2-yl)acetate with sodium azide. The IR spectrum of ester **6** exhibits intense bands at 1660 and 1635 cm⁻¹ which were assigned to the $\nu(\text{C}=\text{O})$ and $\nu(\text{C}=\text{C})$ modes; the $\nu(\text{N}-\text{H})$ band was found at 3220 cm⁻¹ (a low-intensity band). All these characteristics resemble those of other amino esters.⁹ The

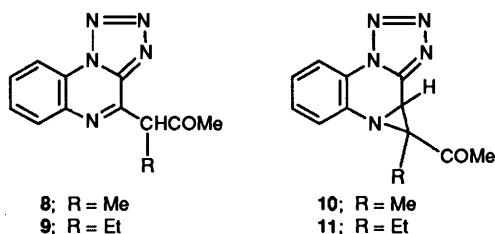
band at 1742 cm⁻¹, which was incorrectly assigned⁸ to an ester carbonyl group, is of medium intensity and should rather be interpreted as being due to combination vibrations of other modes. The *N*-oxide **4** was also treated with the carbanion of benzoylacetonitrile under similar reaction conditions and gave 4-benzoyl(cyano)methylene-4,5-dihydro-tetrazolo[1,5-*a*]quinoxaline **7**.



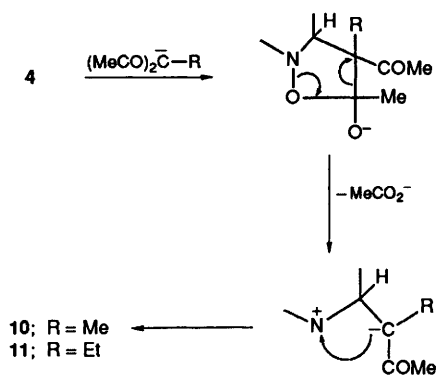
In order to examine further the reactivity of *N*-oxide **4** we carried out experiments involving the reactions of compound **4** with 3-methyl- or 3-ethyl-pentane-2,4-dione in ethanol in the presence of piperidine. We expected the formation of 4-(1-methyl-2-oxopropyl)- **8** and 4-(1-ethyl-2-oxopropyl)-tetrazolo[1,5-*a*]quinoxaline **9**, respectively, with the ketimine tautomeric structure, rather than the formation of the respective enamino ketones, because of the steric hindrance to coplanarity of the carbonyl group due to the presence of the alkyl group at the α -carbon atom. The wavenumber values $\nu(\text{C}=\text{O})$ 1703 and 1701 cm⁻¹, respectively, and the absence of $\nu(\text{N}-\text{H})$ bands are in accord with the proposed ketimine structures.

Further decisive information was obtained from an analysis of the aliphatic sections of the respective ¹H and ¹³C NMR spectra. If the CH(Me)COMe group of the methyl derivative **8** were present, one would have to observe a quartet and a doublet with the coupling constant ³J(H,H) ca. 7 Hz in the ¹H NMR spectrum, and four signals due to CH, Me and COMe groups in the ¹³C NMR spectrum. The molecular mass estimated from the mass spectrum of the compound studied corresponded to the respective molecular formula and did not exclude our

original presumption. However, the ^1H NMR spectrum exhibited three singlets (δ_{H} 4.29, 2.32, 0.97) with relative intensities 1:3:3, and a dilution of the sample did not cause any change in their respective positions. The protons were not exchangeable by deuterium after shaking of the sample with deuterium oxide. The aromatic protons gave two signals, at 8.01 (1 H) and 7.44 (the centre of a multiplet, 3 H). The sample is decomposed on addition of trifluoroacetic acid. The ^1H NMR spectra measured in deuteriochloroform and in hexadeuteriodimethyl sulphoxide were identical. Four signals were observed in the aliphatic part of the ^{13}C NMR spectrum, *viz.* δ_{C} 44.53 (C_q), 38.56 (CH), 23.59 (COMe) and 7.79 (Me), whereas the aromatic section exhibited signals of another eight carbon atoms [δ_{C} 204.18 (COMe), 144.63, 132.40, 126.43 (all C_q), 129.65, 128.13, 127.43, and 116.72 (all CH)]. The above mentioned values and the reactivity observed are in accord with the presence of an aziridine ring; hence, the product of the reaction of *N*-oxide 4 with 3-methylpentane-2,4-dione is 4-acetyl-4-methyl-3b,4-dihydroazirino[1,2-*a*]tetrazolo[5,1-*c*]quinoxaline 10. The same methods were used to verify the structure of the respective 3-ethyl analogue 11.



Aziridinoquinoxalines as products of the reaction of heterocyclic *N*-oxides with carbanions have not previously been observed. They represent a third new isomer among the products of this reaction (beside the above-mentioned tautomers). Stable aziridine derivatives were observed as products of 1,3-dipolar cycloadditions of 3*H*-indole 1-oxide with alkynes¹⁰ proceeding *via* the respective 4-isoxazoline intermediates.¹¹ Our reaction represents a 1,3-cycloaddition of a carbanion to give an unstable isoxazolidine intermediate which undergoes deacylation. The subsequent nucleophilic attack of the carbanion on the nitrogen heteroatom (which is significantly supported by the steric effect of the alkyl group attached at C-4 of the isoxazolidine nucleus) results in ring closure to give the aziridine ring.



We tried to estimate the position of the substituents with regard to the hydrogen atom in the aziridine ring of the derivative 10. The low value found for the nuclear Overhauser effect excludes the *cis* configuration for the methyl group and hydrogen at the 4 and 3b positions, hence the compounds 10 and 11 represent racemic mixtures of the enantiomers **a** and **b**. Out of the three chiral centres present (3b, 4, 5) only the carbon

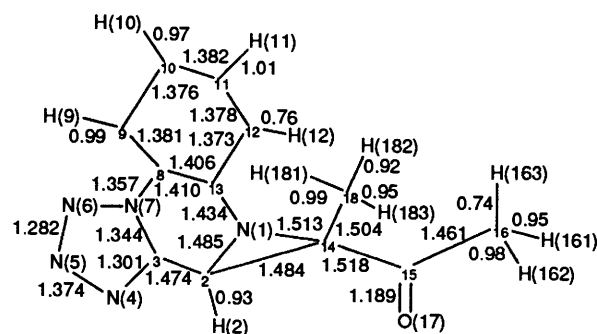


Fig. 1. Interatomic distances (Å) for compound 10. The e.s.d.s are in the range 0.003–0.006 Å for non-H bonds and 0.3–0.08 Å for C–H bonds.

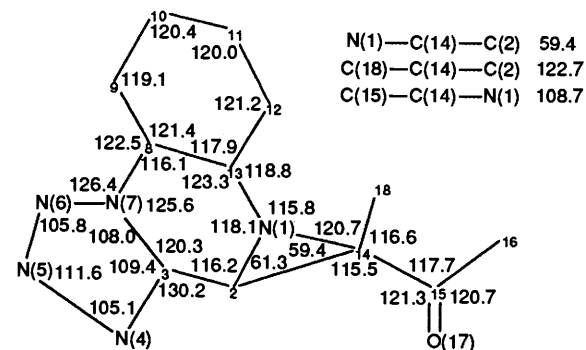
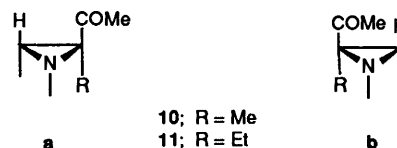


Fig. 2. Interbond angles (°) for compound 10. The e.s.d.s are in the range 0.2–0.3° for non-H-atoms and 2–4° for angles involving H-atoms.

atom C-4 acts as a true chiral centre, whereas both the carbon atoms 3b and nitrogen 5 cannot be chiral because of the rigidity of the heteroaromatic skeleton of the molecules 10 and 11.



The full determination of the molecular structure of compound 10 has been made by X-ray diffraction. The centrosymmetric space group $P\bar{1}$ confirms the presence of the racemic mixtures of enantiomers **a**, **b** in the crystalline state. Fig. 1 shows the numbering scheme of one enantiomer alone with the interatomic distances. The distances are not corrected for thermal motion of the atoms. The corrections calculated according to ref. 12 are lower than 0.01 Å except for the bonds C(15)–O(17) and C(15)–C(16) which, after correction, are 1.21 and 1.54 Å, respectively. The reason for this is because of high anisotropic thermal movement of the methyl C(16) [B_{eq} 12.6(4) Å²] and of the oxygen O(17) [B_{eq} 7.7(1) Å²].

A review of interbond angles is given in Fig. 2. The torsion angles H(2)–C(2)–C(14)–C(18) 150(2)° and H(2)–C(2)–C(14)–C(15) –3(2)° confirm an approximate *trans*-position of the methyl and hydrogen moieties at positions 4 and 3b. H(181) is *trans* with respect to the carbon C(15) [torsion angle C(15)–C(14)–C(18)–H(181) 178(2)°]. Nonetheless, the methyl C(16) is placed so that one hydrogen is in an eclipsed position with respect to oxygen O(17) [torsion angle O(17)–C(15)–C(16)–H(162) 2(4)°]. Another short intramolecular contact is C(18)–H(181)···N(7) [H(181)···N(7) 2.65(2) Å, angle C(18)–H(181)–N(7) 126(3)°].

Torsion angles showing the conformation around the aziridine ring are given in Table 1.

Table 1. Selected torsion angles in molecule **10**

Bond	Angle (°)
C(13)–N(1)–C(14)–C(15)	142.1(3)
C(13)–N(1)–C(14)–C(18)	3.5(4)
C(13)–N(1)–C(2)–C(3)	–7.9(4)
C(13)–N(1)–C(2)–H(2)	–144.3(20)
N(1)–C(14)–C(15)–C(16)	–91.9(5)
N(1)–C(14)–C(15)–O(17)	82.2(4)
C(18)–C(14)–C(15)–C(16)	48.7(6)
C(2)–C(14)–C(15)–O(17)	17.9(5)
C(3)–C(2)–C(14)–C(15)	–157.6(3)
C(3)–C(2)–C(14)–C(18)	–4.2(5)
N(4)–C(3)–C(2)–H(2)	–37.0(20)

Table 2. The angles between the planes in molecule **10**

Plane	Atoms	χ^2
A	N(1),C(2),C(3),N(7),C(8),C(13)	760.0
B	C(8),C(9),C(10),C(11),C(12),C(13)	27.3
C	C(3),N(4),N(5),N(6),N(7)	0.2
D	N(1),C(2),C(14)	
E	C(14),C(15),C(16),C(17)	78.0

Dihedral angles (°)

A,B	3.3(1)	A,C	4.9(1)	B,C	6.2(1)
A,D	71.0(2)	A,E	37.2(2)	D,E	75.3(2)

Table 3. Positional parameters and estimated standard deviations in parentheses for compound **10**. All data $\times 10^4$

Atom	<i>x</i>	<i>y</i>	<i>z</i>
N(1)	6 138(2)	8 147(3)	7 199(2)
C(2)	4 553(3)	7 273(3)	7 482(2)
C(3)	3 655(3)	4 861(4)	7 132(2)
N(4)	2 179(2)	3 635(4)	7 120(2)
N(5)	1 946(3)	1 516(3)	6 633(2)
N(6)	3 211(2)	1 457(3)	6 362(2)
N(7)	4 325(2)	3 581(3)	6 671(2)
C(8)	5 903(3)	4 334(3)	6 519(2)
C(9)	6 574(3)	2 927(4)	6 146(2)
C(10)	8 127(4)	3 791(5)	6 031(3)
C(11)	8 988(3)	6 034(4)	6 259(3)
C(12)	8 303(3)	7 422(4)	6 628(2)
C(13)	6 771(3)	6 623(3)	6 781(2)
C(14)	6 133(3)	8 463(3)	8 640(2)
C(15)	6 583(3)	10 867(4)	9 226(3)
C(16)	8 334(7)	12 317(8)	9 956(10)
O(17)	5 572(2)	11 587(3)	9 009(2)
C(18)	6 851(4)	7 324(4)	9 574(3)

Table 2 shows the planarity of the rings in the molecule **10**. The atoms of both the six- and the five-membered ring lie almost in a single plane. The maximum deviation between normals of planes is 6.2(1)°. The plane of the aziridine ring makes an angle of 71.0(2)° with the adjacent ring. The angles between C(2)–H(2) bond and the adjacent rings A and D (see Table 2) are 55(2)° and 30(2)°, respectively.

The most significant intermolecular contact is C(18)–H(183)···O(17) [H(183)···O(17) 2.59(5) Å, angle C(18)–H(183)–O(17) 125(3)°].

Experimental

General.—M.p.s were determined on a Kofler apparatus and are uncorrected. The course of reactions and the purity of products were monitored by TLC [Silufol plates; cyclohexane–acetone (3:2) (S1) or benzene–ethyl acetate (1:1) (S2)]. IR spectra were recorded in chloroform on an IR 75 spectrophotometer (Zeiss); the wavenumber scale was calibrated with polystyrene. ¹H and ¹³C NMR spectra were measured in the standard way with a JNM-FX 100 (JEOL) apparatus at 300 K at 99.602 and 25.047 MHz, respectively. Both the ¹H and ¹³C chemical shifts are related to internal tetramethylsilane (δ 0.00).

X-Ray Diffraction.—Bright yellow crystal of compound **10** was of dimensions 0.35 \times 0.25 \times 0.20 mm. The crystal quality was checked and the space group was determined by Weissenberg photographs. The crystals are monoclinic, space group *P*1̄, with two molecules in the unit cell. Unit-cell parameters *a* = 9.199(6), *b* = 6.785(5), *c* = 10.593(5) Å, α = 97.99(5), β = 105.01(4), γ = 111.63(5)° and *V* = 573.0(7) Å³ were calculated from 15 reflections measured in the range of 11° < 2 θ < 31° using a SYNTAX P2₁ diffractometer with Cu-K α radiation, *T* = 296 K, *F*(000) = 252. A total of 1950 symmetry-independent reflections were measured by a θ –2 θ scan (2 θ < 120°, scan rate 2.5–29.5° min^{–1}). Three standard reflections: 4,0,0, 0,3,0, 0,0,4, showed no significant fluctuations during the measurement. 1580 Reflections which fulfil the criterion *I* > 1.96 \times σ (*I*) were used for the refinement. The absorption was ignored.

The phase problem was solved by MULTAN80¹³ and the structure was refined by SHELX76.¹⁴ The function minimized was $\Sigma w\Delta^2$, where $w = 1.14 (\sigma^2(F_o) + 0.002|F_o|^2)^{-1}$ and $\Delta = ||F_o| - |F_c||$. All non-H-atoms were refined with anisotropic temperature parameters and all H-atoms, localized on a difference map, were refined isotropically. The refinement of 209 parameters in three blocks was stopped when $\Delta/\sigma < 0.06$. No significant correlation among the refined parameters were observed. The resulting statistical criteria were: *R* = $\Sigma\Delta/\Sigma|F_o| = 0.057$, *R*_g = $\Sigma w\Delta^2/\Sigma w|F_o|^2 = 0.072$, *S* = 1.65. The extinction coefficient was refined to 3.3×10^{-6} . The maximum and minimum peaks on the final difference map were 0.22 and –0.15 e Å^{–3}, respectively. The atomic scattering factors were taken from International Tables for X-ray Crystallography.¹⁵ The geometry was calculated with PARST¹⁶ and IMC¹⁷ programs. The refined atomic parameters are deposited with the Cambridge Structural Data Base.^{18,*} Atomic co-ordinates are given in Table 3.

Tetrazolo[1,5-a]quinoxaline 5-Oxide 4.—A solution of 3-chloroquinoxaline 1-oxide (1.8 g, 10 mmol) and sodium azide (0.7 g, 11 mmol) in ethanol (50 cm³)–HCl (0.2 mol dm^{–3}; 50 cm³) was boiled for 30 h. The formation of the product was followed by TLC (S1). After the addition of sodium azide (0.5 g) and the mixture had been refluxed for 24 h the solution was filtered through charcoal. After the mixture had cooled orange crystals (1.2 g, 64%) was isolated, m.p. 196.5–198.5 °C [lit.,⁷ 52% yield for recrystallized sample; m.p. 189–190 °C (decomp.)] (Found: C, 51.5; H, 2.8; N, 37.6. Calc. for C₈H₅N₅O: C, 51.3; H, 2.7; N, 37.4%).

4-Acetylmethylene-4,5-dihydro-tetrazolo[1,5-a]quinoxaline 5.—A hot solution of *N*-oxide **4** (1.2 g, 6.4 mmol) and 2,4-pentane-2,4-dione (1.2 g, 11.8 mmol) in ethanol (80 cm³) was treated with piperidine (3.5 cm³). A small amount of brown, non-melting precipitate was filtered off, and the filtrate was refluxed for one hour and filtered hot. The solid residue was washed with hot ethanol (2 \times 20 cm³), and the combined filtrates were evaporated under reduced pressure to 25 cm³ and left for 12 h at 0 °C. The red precipitate was collected by suction and recrystallized from ethanol to give chromatographically

* For details see 'Instructions for Authors (1990)', *J. Chem. Soc., Perkin Trans. 1*, 1990, Issue 1.

[TLC (S1)] pure product (0.15 g, 10%). After sublimation (140–150 °C at 0.26 kPa) the product had m.p. was 218–221 °C; ν_{\max} 1626 (C=O) and 1605 cm^{-1} (C=C); δ_{H} 2.32 (COMe), 6.46 (=CHCO), 7.60–7.11 (3 H, ArH), 8.22 (1 H, ArH), and 13.3 (NH) (Found: C, 58.0; H, 4.15; N, 30.8. $\text{C}_{11}\text{H}_9\text{N}_5\text{O}$ requires C, 58.1; H, 4.0; N, 30.8%).

Ethyl-(4,5-Dihydro-1,5-a)quinoxalin-4-ylidene)-acetate 6.—A solution of the *N*-oxide **4** (0.09 g, 5 mmol), ethyl acetoacetate (0.1 g, 8 mmol) and piperidine (0.2 cm^3) in ethanol (5 cm^3) was heated on a steam-bath until the reaction was complete [20 min, TLC (S1)]. After storage for 12 h at 0 °C yellow crystals (0.075 g, 60%) were isolated, m.p. 127–130 °C. The identity with the authentic compound⁸ was verified by means of mixed m.p., TLC and IR spectra.

(α -Benzoyl-4,5-dihydro-1,5-a)quinoxalin-4-yl)-acetonitrile 7.—A boiling solution of the *N*-oxide **4** (0.55, 2.94 mmol) with benzoylacetonitrile (1 mol equiv.) in ethanol (30 cm^3) was treated with piperidine (2 cm^3). After the mixture was boiled for 30 min, and stored for 12 h at 0 °C, followed by separation of a small amount of dark, insoluble solid, the filtrate was evaporated under reduced pressure to dryness. Compound **7** was separated from the residue by means of column chromatography (silica gel; 30 \times 4.5 cm; CHCl_3). From the fraction corresponding to the main spot (R_f 0.62) the solvent was evaporated off and the residue was washed with ethanol (20 cm^3). After crystallization from 1,4-dioxane (charcoal), yellow prisms (0.15 g, 16%) were isolated, m.p. 269–274 °C (decomp.). ν_{\max} 2220 (C \equiv N) and 1635 cm^{-1} (C=O); δ_{H} 7.25–8.02 (7 H, ArH), 8.54 (2 H, *peri*) and 15.97 (N–H) (Found: C, 64.9; H, 3.4; N, 26.6. $\text{C}_{17}\text{H}_{10}\text{N}_6\text{O}$ requires C, 65.0; H, 3.2; N, 26.7%).

4-Acetyl-4-methyl-3b,4-dihydroazirino[1,2-a]tetrazolo[5,1-c]-quinoxaline 10.—A boiling solution of *N*-oxide **4** (0.67 g, 3.6 mmol) and 3-methylpentane-2,4-dione (0.6 g, 5.3 mmol) in ethanol (50 cm^3) was treated with piperidine (1.2 cm^3). After reflux for 1.5 h the reaction was complete [TLC (S2)], and the hot solution was filtered through charcoal and evaporated to ten cm^3 under reduced pressure. After being dried over phosphorus pentoxide and cooled to 0 °C the crystals were collected by suction and washed with methanol (6 cm^3) to remove an oily contaminant. After crystallization (3 \times) from ethanol the tetracycle **10** was obtained as yellow, chromatographically pure crystals (0.1 g, 11.5%), m.p. 149–151 °C (Found: C, 59.6; H, 4.6; N, 29.1. $\text{C}_{12}\text{H}_{11}\text{N}_5\text{O}$ requires C, 59.7; H, 4.6; N, 29.0%).

4-Acetyl-4-ethyl-3b,4-dihydroazirino[1,2-a]tetrazolo[5,1-c]-quinoxaline 11.—A solution of the *N*-oxide **4** (0.55 g, 2.94 mmol), 3-ethylpentane-2,4-dione (0.7 g, 5.5 mmol) and piperidine (1.1 cm^3) in ethanol (100 cm^3) was heated on a boiling water-bath until the reaction was complete [TLC (S2)]. The hot solution was filtered with charcoal, the solvent was evaporated off under

reduced pressure to ten cm^3 , and the solution was submitted to free crystallization at room temperature. After separation of a colourless substance (0.1 g) of m.p. 130–132 °C and washing with methanol the filtrate was submitted to further free crystallization. The resulting precipitate was recrystallized twice from ethanol to give compound **11** as yellow needles (0.2 g, 28%), m.p. 134–135 °C; ν_{\max} 1705 cm^{-1} (C=O); δ_{H} 0.70 (Me), 1.22 (1H) (protons of prochiral CH_2Me groups), 1.42 (1 H), 2.34 (COMe), 4.21 (CH), 7.44 (centre of multiplet, 3 H) and 8.01 (1 H); δ_{C} 203.78 (COMe), 144.51, 132.34, 129.42, 129.70, 127.14, 126.26, 116.49, 48.63 (C_q), 37.74 (CH), 24.41 (COMe), 16.80 (CH_2) and 8.84 (Me) (Found: C, 61.2; H, 5.3; N, 27.5. $\text{C}_{13}\text{H}_{13}\text{N}_5\text{O}$ requires C, 61.2; H, 5.2; N, 27.4%).

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