Electronic Supplementary Information for

Quinoline synthesis: scope and regiochemistry of photocyclisation of substituted benzylidenecyclopentanone *O*-alkyl and *O*-acetyloximes.

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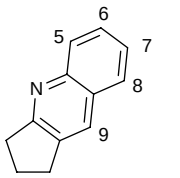
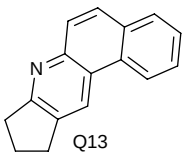
Preparation of the 2-benzylidenecyclopentanone *O*-acetyloximes

Data for the 2,3-dihydro-1*H*-cyclopenta[*b*]quinolines and related photoproducts





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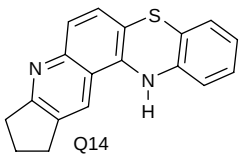

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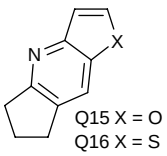

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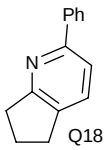
Q16 X = S



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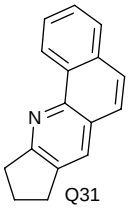

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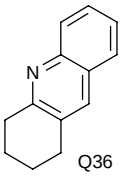

Q36

Introductory remarks

NMR assignments were generally supported by 2D ^{1}H-^{1}H homouclear and ^{1}H-^{13}C heteronuclear correlation spectroscopy. IR spectra were recorded for liquids as thin films on NaCl plates and for solids as dispersions in KBr discs, using either Nicolet 205 FT-IR or Perkin Elmer 983G spectrophotometers. UV spectra were recorded in methanol on a Hewlett Packard 8452A diode array UV-Vis. spectrophotometer. Extinction coefficients (ε) are in dm^3 mol^{-1} cm^{-1}. Oils were purified by short path distillation under reduced pressure. Elemental analyses were carried out by the Microanalytical Laboratory at University College Dublin. TLC was on silica gel plates containing a fluorescent indicator (Riedel-de-Haen, DC-Cards SiF, layer thickness 0.2 mm). Radial centrifugal chromatography (RCC) was with a Harrison Research model 7924T Chromatotron system using rotors coated with silica gel PF_{254} containing 5% calcium sulphate as binder. Light petroleum used for RCC and for column chromatography on silica had b.p. 40-60 °C. HPLC was with a Waters 510 HPLC pump and Waters Microbondapak C_{18} RCM cartridges with 15μm packing (RCM 8x10 cartridge for analytical work, and RCM 25x10 cartridge for preparative work). GC was carried out using a Carlo Erba Strumentazione 4130 gas chromatograph fitted with a Quadrex Corporation fused silica capillary column (25 m, 007 series methyl silicone, 0.32 mm ID, 1.0 μm film thickness).

Aldehydes required for synthesis of the 2-benzylidenecyclopentanones were commercial materials (Aldrich) except for 1-formylphenothiazine, 3-*t*-butylbenzaldehyde, 3-*N*,*N*-dimethylaminobenzaldehyde and 3-methoxy-4-methylbenzaldehyde.

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1-Formylphenothiazine M1 was prepared by a published procedure.[1]

A 2.5M solution of *n*-butyllithium in hexane (37.5 cm^3, 94 mmol) was added to anhydrous diethyl ether (200 cm^3), then phenothiazine (7.42 g, 37.5 mmol) was added slowly and the mixture stirred under nitrogen for 48 hours, then cooled to –70 ^{o}C. Dimethylformamide (2.74 g, 37.5 mmol) was added dropwise at –70 ^{o}C, then the temperature was raised to room temperature and the mixture stirred for 1 hour. Hydrochloric acid (0.5M; 100 cm^3) was added with vigorous stirring and the organic layer was separated. The aqueous layer was extracted

with diethyl ether (3x50 cm^3) and the combined extracts washed with water (100 cm^3) and dried. Rotary evaporation gave a red residue which was chromatographed over silica (90:10 light petroleum/ethyl acetate). Recrystallisation (light petroleum/ethyl acetate) yielded bright red crystals of **1-formylphenothiazine M1** (4.1 g, 48%), mp 81-82 ^oC (lit.,[2] 80-81 ^oC); δ_H 6.62 (1H, dd, *J* 8.5, 1.0, arH), 6.81 (1H, t, *J* 7.6, arH), 6.85 (2H, m, 2 x arH), 7.00 (1H, m, arH), 7.04 (1H, dm, J_d 7.6, arH), 7.25 (1H, dd, *J* 8.0, 1.4, arH), 9.80 (1H, s, CHO) and 10.11 (1H, s, NH); δ_C 116.41, 117.57, 118.61, 119.73, 121.11, 124.18, 126.89, 128.11, 131.92, 133.87, 139.13, 144.87 (12 x arC) and 194.72 (CHO).

3-*t*-Butylbenzaldehyde M2 was prepared by the following sequence of steps.

(i) Conversion of 4-t-butylaniline to 2-bromo-4-t-butylaniline, *using a procedure based on that for conversion of 4-methylaniline to 2-bromo-4-methylaniline.*[3]

4-*t*-Butylaniline (25.0 g, 16.8 mmol) in glacial acetic acid (100 cm^3) was heated under reflux for 3 hours and allowed to cool to about 45 $^\circ C$. Bromine (27.2 g, 17 mmol) was added with rapid stirring, below 55 $^\circ C$. Stirring was continued for 1 hour then the mixture was poured in a thin stream into well stirred ice/water (100 cm^3 each). Sodium metabisulphite was added until the bromine colour no longer persisted. The solid was filtered off, washed with water and then light petroleum (b.pt. 40-60 ^oC). After drying overnight **N-(2-bromo-4-t-butylphenyl)acetamide** was obtained as pink/white crystals (42.2 g, 93%) mp 159-160 ^oC (lit.[4], 159.5 ^oC) and was used in the next step without further purification; δ_H 1.31 (9H, s, CMe_3), 2.25 (3H, s, MeCO) 7.35 (1H, dd, *J* 8.9, 2.2, arH-5), 7.44 (1H, d, *J* 8.9, arH-6) 7.54 (1H, d, *J* 2.2, arH-3) and 8.20 (1H, s, NH); δ_C 25.18 (MeCO), 31.74 (CMe_3), 34.89 (CMe_3), 120.27, 122.21, 125.84, 126.20, 129.47, 133.40 (arC), and 168.53 (C=O).

To dried **N-(2-bromo-4-t-butylphenyl)acetamide** (45.5 g 168.3 mmol) in ethanol (50 cm^3) was added concentrated hydrochloric acid (50 cm^3) and the mixture was heated under reflux for 3 hours. On cooling, water (50 cm^3) was added. Ethanol/water (75 cm^3) was distilled off and the mixture cooled and made alkaline by addition of 10% sodium hydroxide. The heavy organic bottom layer was separated and washed twice with water (50 cm^3). **2-Bromo-4-*t*-butylaniline** was obtained as a deep red oil (31.5 g, 82%) which was used in the next step without further purification; δ_H 1.24 (9H, s, CMe_3), 4.70 (2H, s, NH_2), 6.75 (1H, d, *J* 8.4, H-6), 7.12 (1H, dd, *J* 8.4, 2.1, H-5) and 7.40 (1H, d, *J* 2.1, H-3); δ_C 31.32 (CMe_3), 33.96 (CMe_3), 109.65, 116.01, 125.36, 129.30, 140.70 and 143.30 (arC).[5]

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(ii) Conversion of 2-bromo-4-t-butylaniline to 3-t-butylbromobenzene *was based on a published procedure for reductive deamination of arylamines by alkyl nitrites.*[6]

t-Butyl nitrite (21.4 g, 20.7 mmol) in DMF (40 cm^3) was heated to 65 °C and 2-bromo-4-t-butylaniline (32.1 g, 140.7 mmol) in DMF (20 cm^3) was added dropwise with rapid stirring which was continued for an additional 20 minutes. The mixture was then added to stirred 20% hydrochloric acid (150 cm^3). Extraction with diethyl ether (2x100 cm^3) and washing of the extract with 10% hydrochloric acid (50 cm^3) and water (50 cm^3), followed by drying and rotary evaporatoration, yielded a brown oil which was purified by chromatography on silica (99:1 light petroleum/ethyl acetate). **3-*t*-Butylbromobenzene**[7] was obtained as a yellow oil (13.5 g, 45%) and was used in the next step without further purification; δ_H 1.40 (9H, s, CMe_3), 7.24 (1H, t, *J* 8.0, arH-5), 7.40 (2H, m, arH-4 and arH-6) and 7.62 (1H, t, *J* 1.8, arH-2); δ_C 31.68 (C*Me*$_3$), 35.30 (*C*Me_3), 122.92, 124.48, 129.02, 129.12, 130.15 and 154.02 (arC).

(iii) Conversion of 3-t-butylbromobenzene to 3-t-butylbenzaldehyde *was based on a literature method* [8] *for conversion of 3-t-butyl-6-methylbromobenzene to 3-t-butyl-6-methylbenzaldehyde.*

3-*t*-Butylbromobenzene (3.92 g, 18.4 mmol) in anhydrous diethyl ether (100 cm^3) was cooled to 0 °C. A 2.5M solution of *n*-butyl lithium in hexane (15 cm^3, 37 mmol) was added dropwise and the reaction mixture was stirred for a further 30 minutes. Dimethylformamide (1.68 g, 22.3 mmol) was added dropwise and stirring was continued for 5 minutes. The reaction mixture was warmed to room temperature then poured into 20% aqueous phosphoric acid with vigorous stirring, and extracted with diethyl ether (2x50 cm^3). The extract was washed with 10% sodium bicarbonate (50 cm^3), water (50 cm^3) and dried over magnesium sulphate. Removal of the ether yielded a dark orange oil, which was purified by chromatography on silica (90:10 light petroleum/ethyl acetate), giving **3-*t*-butylbenzaldehyde M2** as a yellow oil (2.43 g, 81%); δ_H[9] 1.25 (9H, s, CMe_3), 7.36 (1H, t, *J* 7.6, arH-5), 7.58 (2H, m, arH-4 and arH-6), 7.81 (1H, t, *J* 1.8, arH-2) and 9.91 (1H, s, CHO); δ_C 31.06 (C*Me*$_3$), 34.69 (*C*Me_3), 126.14, 127.32, 128.67, 131.59, 136.21, 152.06 (arC) and 192.61 (CHO). *Table of Contents*

3-(*N,N*-Dimethylamino)benzaldehyde M3 *was obtained by the following sequence.*

(i) Conversion of 3-bromoaniline to 3-(N,N-dimethylamino)bromobenzene.

Reaction of 3-bromoaniline with dimethyl sulphate[10] yielded 3-(*N,N*-dimethylamino)bromobenzene as a light orange oil (20%) following reduced pressure distillation; δ_H 2.80 (6H, s, NMe_2), 6.49 (1H, m), 6. 70 (2H, m) (3xarH) and 6.95 (1H, t, *J* 8.4, arH-5);.δ_C 40.79 (NMe_2), 111.34, 115.46, 119.46, 123.83, 130.72 and 152.06 (arC).

(ii) Conversion of 3-(N,N-dimethylamino)bromobenzene to 3-(N,N-dimethylamino)benzaldehyde

A 2.5M solution of *n*-butyllithium in hexane (8.0 cm^3, 20 mmol) was added dropwise under nitrogen at 0 ^oC to 3-(*N,N*-dimethylamino)bromobenzene (4.02 g, 20 mmol) in diethyl ether (50 cm^3) and the mixture was stirred for 30 minutes. Dimethylformamide (1.83 g, 25 mmol) was added dropwise and stirring was continued for 5 minutes. The mixture was allowed to reach room temperature and was then poured into 20% aqueous phosphoric acid (50 cm^3) with rapid stirring, then neutralised with 10% sodium carbonate solution and extracted with diethyl ether (2 x 50 cm^3). The extract was washed with 10% sodium carbonate solution (25 cm^3), then water (25 cm^3) and dried. Rotary evaporation yielded a dark orange oil which was purified by silica chromatography (25% ethyl acetate in light petroleum), yielding **3-*N,N*-dimethylaminobenzaldehyde M3** as an orange oil (2.9 g, 97%). IR, ^{1}H-nmr and ^{13}C-nmr data were in agreement with reported[11] literature values: δ_H 2.87 (6H, s, NMe_2), 6.82 (1H, dd, J 8.0, 2.5, arH-4), 7.05-7.10 (2H, m, 2 x arH-2, -6), 7.26 (1H, t, *J* 8.0, arH-5) and 9.83 (1H, s, CHO); δ_C 40.50 (NMe_2), 111.94, 118.48, 118.87, 129.91, 137.58, 151.01 (arC) and 193.39 (CHO).

3-Methoxy-4-methylbenzaldehyde M4 was prepared from 3-methoxybenzaldehyde by a literature procedure.[12]

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A 2.5M solution of *n*-butyllithium in hexane (41.2 cm^3, 103 mmol) was added dropwise to a solution of *n*-methylpiperazine (11.8 cm^3, 107 mmol) in THF (200 cm^3) at –20 ^oC and stirring under nitrogen was continued for a further 15 minutes. 3-Methoxybenzaldehyde (13.6 g, 100 mmol) was added and the solution was stirred for a further 15 minutes. Tetramethylethylenediamine (45.3 cm^3, 30 mmol) was then added, followed by a 1.4M

solution of *sec*-butyllithium in cyclohexane (214 cm^3, 300 mmol). The reaction was stirred for a further 10 minutes, then stored in the freezer overnight. The reaction vessel was cooled to –78 ^oC and methyl iodide (37.3 cm^3, 600 mmol) added slowly with stirring, following which the mixture was allowed to reach room temperature. It was poured into stirring 10% HCl, extracted with diethyl ether (2x100 cm^3), washed with brine (50 cm^3), water (2x50 cm^3) and dried. Purification by silica chromatography (90:10 light petroleum/ethyl acetate) yielded light yellow needles of **3-methoxy-4-methylbenzaldehyde M4** (4.3 g, 29%), mp 45-46 ^oC (lit., 45-46 ^oC); δ_H 2.13 (3H, s, Me), 3.72 (3H, s, OMe), 7.12 (1H, d, *J* 7.5, arH-5), 7.17 (1H, br s, arH-2), 7.19 (1H, dd, *J* 7.5, 1.5, arH-6) and 9.76 (1H, s, CHO); δ_C 17.11 (Me), 55.67 (OMe), 108.18, 124.68, 131.17, 135.05, 136.25, 158.59 (arC) and 192.21 (C=O).

2-Diphenylmethylenecyclopentanone M5 (24%, from reaction[13] of phenylmagnesium bromide with 2,2-ethylenedioxycyclopentanonecarboxylate[14] and subsequent treatment with acidified methanol), mp 115-116 °C (lit., 115-116 °C); δ_H 1.91 (2H, qn, *J* 7.1, $CH_2CH_2CH_2$), 2.36 (2H, t, *J* 7.1, CH_2C=O), 2.81 (2H, t, *J* 7.1, CH_2C=C), 7.12 (2H, m, 2 x arH), 7.18 (2H, m, 2 x arH) and 7.31 (6H, m, 6 x arH); δ_C[15] 20.50, 32.94, 39.78 (3 x CH_2), 127.80, 127.96, 128.36, 129.42, 129.60, 134.31, 140.12, 141.78, 148.27 (arC + C=C) and 206.56 (C=O).

Preparation of *E,E*-2-Benzylidenecyclopentanone *O*-allyloxime *E,E*-5: *N*-Allyloxyphthalimide[16] (4.5 g, 20 mmol) was heated under reflux in 6M hydrochloric acid (50 cm^3) for 2 hours. On cooling, phthalic acid crystallised out and was filtered off. Concentration of the filtrate and drying over sodium hydroxide yielded highly hygroscopic **allyloxyamine hydrochloride** (1.6 g, 80%); δ_H (D_2O) 4.33 (2H, d, *J* 5.8, OCH_2), 4.77 (4H, br s, $-NH_3Cl$ and H_2O), 5.31 (2H, m, CH=CH_2) and 5.72 (1H, m, C*H*=CH_2). The crude hydrochloride was used without further purification. *Table of Contents*

2-Benzylidenecyclopentanone **K1** (3.0 g, 15 mmol), allyloxyamine hydrochloride (1.6 g, 15 mmol), and pyridine (5 cm^3) in ethanol (30 cm^3) were heated under reflux for 30 minutes, then concentrated. Water (30 cm^3) was added, the mixture was cooled in ice and the solid

product dried ($MgSO_4$) and recrystallised (methanol) to yield *E,E*-2-benzylidenecyclopentanone *O*-allyloxime ***E,E*-5** (2.9 g, 73%), mp 45-46 °C (Found: C, 79.5; H, 7.6; N, 6.0. $C_{15}H_{17}NO$ requires: C, 79.3; H, 7.5, N, 6.2%); λ_{max}/nm 222 (13 200) and 302 (27 100); ν_{max}/cm^{-1} (KBr pellet): 3083, 3023, 2953, 2873 , 1646 (C=N), 1590, 1570, 1488, 1446, 1420, 1348, 1319, 1298, 1261, 1221, 1203, 1181, 1157, 1126, 1108, 1079, 1031, 997, 924, 889, 866, 849, 825, 788, 751 and 694cm^{-1}; δ_H 1.86 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.63 (2H, t, *J* 7.6, CH_2C=N), 2.79 (2H, td, J_t 7.3, J_d 2.5, CH_2C=CPh), 4.69 (2H, dt, J_d 5.6, J_t 1.5, OCH_2), 5.25 (1H, J_d 11, J_q 1.5, CH=C*H*H), 5.35 (1H, J_d 17, J_q 1.5, CH=CH*H*), 6.08 (1H, m, C*H*=CH_2), 7.24 (1H, t, *J* 2.5, C=C*H*Ph), 7.27 (1H, t, *J* 7.7, arH-4), 7.36 (2H, t, *J* 7.7, arH-2/6), 7.42 (2H, d, *J* 7.7, arH-3/5); δ_C 22.64 (CH_2CCH_2), 27.65 (CH_2C=N), 31.38 (CH_2C=CPh), 75.16 (OCH_2), 117.36 (C=CH_2), 122.89, 127.25, 128.31, 129.27 (*C*HPh + arC-2/6 + arC-3/5 + arC-4), 134.52 (*C*H=CH_2), 136.85, 137.24 (arC-1 + C=*C*$(C)_2$) and 162.69 (C=N).

Geometrical photoisomerisation of *E,E*-2-benzylidenecyclo-pentanone *O*-allyloxime *E,E*-5 in ethyl acetate

E,E-2-Benzylidenecyclopentanone *O*-allyloxime **A** (1.0 g, 0.004 mol) in ethyl acetate (300 cm^3) was irradiated under the standard conditions and the reaction course followed by GC.[17] After 5 minutes a new product **B** was observed, the concentration of which steadily increased. On further irradiation (30 minutes) the gradual formation of two additional photoproducts **C** and **D** was observed. Irradiation was continued until a photostationary state had been reached (5 hours), comprising **A** (19%), **B** (48%), **C** (23%) and **D** (10%). The solvent was removed by rotary evaporation and the product mixture applied to a 4mm Chromatotron plate (mobile phase: 1% ethyl acetate in light petroleum). *Table of Contents*

The first eluted fraction contained only **C**, *E,Z*-2-benzylidenecyclopentanone *O*-allyloxime ***E,Z*-5** (117 mg, 12%), bp 45-50 °C (0.2mbar)[18] (Found: C, 79.5; H, 7.6; N, 6.0. $C_{15}H_{17}NO$ requires: C, 79.3; H, 7.5; N, 6.2%); λ_{max}/nm 224 (15 500) and 304 (12 200); δ_H 1.83 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.69 (2H, td, J_t 7.4, J_d 2.0, CH_2C=CPh), 2.73 (2H, t, *J* 7.4, CH_2C=N), 4.63 (2H, d, *J* 5.5, OCH_2), 5.27 (1H, dm, J_d 13.0, CH=C*H*H), 5.35 (1H, dm, J_d 19.0, CH=CH*H*), 6.08 (1H, m, C*H*=CH_2), 6.62 (1H, t, *J* 2.0, HC=C), 7.27 (1H, t, *J* 7.6, arH-4), 7.33 (2H, t, *J* 7.6, arH-3/5) and 7.89 (2H, d, *J* 7.6, arH-2/6); δ_C 21.75 (CH_2CCH_2), 29.61 (CH_2C=N), 37.07 (CH_2C=CPh), 75.08 (OCH_2), 117.48 (C=CH_2), 127.33, 127.37, 127.44

(*C*HPh + arC-4 + arC-3/5), 130.06(arC-2/6), 134.91(*C*H=CH_2), 135.47, 136.26 (arC-1 + C=*C*$(C)_2$) and 160.19 (C=N).

The second eluted fraction contained only **A**, recovered *E*,*E*-2-benzylidenecyclopentanone *O*-allyloxime (***E*,*E*-5)** (176 mg, 18%).

The third eluted fraction contained a mixture of **B** and **D.** It was concentrated and then cleanly separated using preparative HPLC (mobile phase 70:30 methanol/water). The methanol was removed from the eluted fractions by rotary evaporation, the resulting emulsions were extracted into diethyl ether, dried ($MgSO_4$) and concentrated.

The first component from the HPLC separation contained only **D**, *Z*,*Z*-2-benzylidenecyclopentanone *O*-allyloxime (***Z*,*Z*-5)** (72 mg, 7%), boiling range 49-53 °C (0.2 mbar) (Found: C, 79.0; H, 7.7; N, 6.0. $C_{15}H_{17}NO$ requires: C, 79.3; H, 7.5; N, 6.2%); λ_{max}/nm 222 (12 300) and 288 (11 000); δ_H 1.89 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.66 (4H, m, CH_2C=N and CH_2C=CPh), 4.25 (2H, dt, J_d 4.8, J_t 1.1, OCH_2), 4.92 (2H, m, CH=CH_2), 5.36 (1H, m, C*H*=CH_2), 6.72 (1H, m, C=C*H*Ph) and 7.27 (5H, m, 5 x arH); δ_C 21.03 (CH_2CCH_2), 31.44 , 34.32 (*C*H_2C=N + *C*H_2C=CPh), 74.69 (OCH_2), 117.29 (C=*C*H_2), 130.66 (*C*HPh), 127.09, 127.11 and 128.62 (arC-2/6 + arC-3/5 + arC-4), 133.26 (*C*H=CH_2), 131.37 and 138.43 (arC-1 + C=*C*$(C)_2$) and 157.53 (C=N).

The second component from the HPLC separation contained only **B**, *Z*,*E*-2-benzylidenecyclopentanone *O*-allyloxime (***Z*,*E*-5)** (124 mg, 12%), boiling range 42-44 °C (0.2mbar) (Found: C, 79.45; H, 7.3; N, 6.1. $C_{15}H_{17}NO$ requires: C, 79.3; H, 7.5; N, 6.2%); λ_{max}/nm 226 (5 900) and 296 (13 200); δ_H 1.73 (2H, qn, *J* 7.3, $CH_2CH_2CH_2$), 2.45 (2H, t, *J* 7.2, CH_2C=N), 2.69 (2H, td, J_t 7.2, J_d 2.4, CH_2C=CPh), 4.60 (2H, dt, J_d 6.5, J_t 1.5, OCH_2), 5.17 (1H, dm, J_d 10.7, CH=C*H*H), 5.30 (1H, dm, J_d 17.3, CH=CH*H*), 6.00 (1H, m, C*H*=CH_2), 7.18 (1H, tt, J 6, 2, arH-4), 7.29 (4H, m, 4 x arH) and 8.11 (1H, t, *J* 2.4, C=CHPh); δ_C 23.62 (CH_2CCH_2), 32.50 (*C*H_2C=N), 32.95 (*C*H_2C=CPh), 75.52 (OCH_2), 117.19 (C=*C*H_2), 127.58 (arC-4), 128.15, 129.47 (arC-2/6 + arC-3/5), 134.44 (*C*H=CH_2), 134.98 (C=*C*HPh), 134.19, 137.75 (arC-1 + C=*C*$(C)_2$) and 156.65 (C=N).

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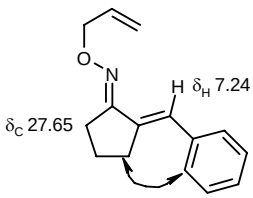
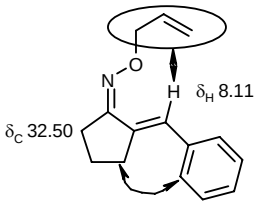
Stereochemical assignments of the geometrical isomers of 5

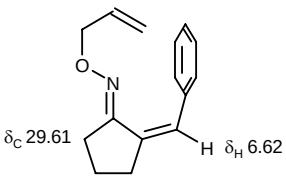
NOE difference spectra for the starting isomer **5A** showed strong enhancement of the H-2/5 aromatic proton absorption when the ring methylene signal at δ2.79 was saturated. Saturation of this aromatic resonance in turn resulted in strong enhancement of the methylene signal. The most rapidly formed photoproduct **5B** also showed similar strong enhancements for the corresponding signals. No such enhancements were observed however between the ring methylene and aromatic signals in the NOE difference spectra of isomers **5C** and **5D**. Taken together, these interactions confirm the stereochemistries at the benzylidene positions for all four isomers, i.e. *E*- for **5A** and **5B,** and *Z*- for **5C** and **5D**, additionally supported by the almost identical chemical shifts for the benzylidene protons in **5C** and **5D (**δ_H 6.62 *vs.* 6.72). For aliphatic oximes and oxime ethers the chemical shift of the oximino α-methylene carbon depends on its orientation relative to the oximino oxygen, with the resonance for the *syn* configuration being found at a lower chemical shift than that for the *anti* arrangement.[19] The observed chemical shifts (δ_C 27.65 and 29.61 for **5A** and **5C** *vs.* δ_C 32.50 and 31.44 for **5B** and **5C**, respectively) confirm the carbon-nitrogen double bond stereochemistries as *E*- for **5A** and **5C**, and *Z*- for **5B** and **5D**. Consistent with these assignments, **5B** and **5D** showed small enhancements in their NOE difference spectra involving the allyloxy group and (for **5B)** the benzylidene proton and (for **5D)** an aromatic proton, whereas **5A** and **5C** showed no such interactions.



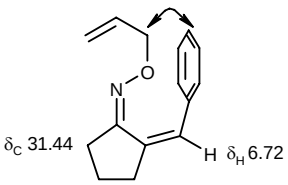
A E,E-5



B Z,E-5



C E,Z-5



D Z,Z-5

Comparison with the chemical shift data for the *O*-methyl- and *O*-acetyloximes confirms that these too have an *E*-configuration at the C=C, as the oximes and ketones must also have.

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Photocyclisation of *E,E*-2-benzylidenecyclopentanone *O*-allyl-oxime (*E,E*-5)

***In methanol*:** *E,E*-2-Benzylidenecyclopentanone *O*-allyloxime (*E,E*-**5**) (1.0 g, 0.004 mol) in methanol (300 cm^3) was irradiated for 5 hours. GC analysis again showed rapid formation of *Z,E*-**5**, with slower formation of the other photoproducts, *E,Z*-**5** and *Z,Z*-**5**, also being seen. In addition, gradual formation of a new photoproduct, absent from the photolysis in ethyl acetate, was also noted. Its concentration increased on continued irradiation, at the expense of the four oxime ether isomers, until it was the major component of the photolysis mixture. Removal of the methanol, and separation using a 4mm Chromatotron plate (mobile phase 90:10 light petroleum / ethyl acetate) gave an oil which was distilled under reduced pressure on to a cold finger to give off-white solid **2,3-dihydro-1*H*-cyclopenta[*b*]quinoline 7** (174 mg, 24%), mp 60-61 °C (lit.,[20] 60-61 °C); ν_{max}/ cm^{-1} 3033, 2955, 2937, 2849, 1614, 1496, 1463, 1405, 1322, 1280, 1265, 1204, 1130, 1093, 1074, 1047, 979, 754, 703; δ_H 2.18 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 3.06 (2H, t, *J* 7.4, CH_2Ar), 3.14 (2H, t, *J* 7.4, CH_2Ar), 7.43 (1H, t, *J* 7.7) and 7.59 (1H, t, *J* 7.9) (arH-7 and arH-6), 7.70 (1H, d, *J* 7.7, arH-8), 7.85 (1H, br s, arH-9) and 8.00 (1H, d, *J* 7.9, arH-5); δ_C 23.62, 30.50, 34.60 (3 x CH_2), 125.49, 127.43, 128.30 and 128.51(benzenoid-*C*H), 130.29 (pyridyl-*C*H), 127.37, 135.77, 147.48 and 167.91 (quaternary *C*s).

In methanol containing 1% w/v potassium carbonate: *E,E*-2-Benzylidenecyclopentanone *O*-allyloxime (*E,E*-**5**) (500 mg, 0.002 mol) in methanol (300 cm^3) containing anhydrous potassium carbonate***** (3.0 g) was similarly irradiated. After 3 hours, irradiation was discontinued, the methanol was removed and water (100 cm^3) added. Extraction with diethyl ether (100 cm^3) and GC analysis showed it to contain a mixture of the four oxime ether isomers of **5** in low concentrations with the quinoline derivative **7** being the major photoproduct present.

> ***** *to eliminate any residual solvent acidity and to confirm that acid conditions are not necessary for photocyclisation in methanol.*

In acetonitrile: Irradiation of *E,E*-2-benzylidenecyclopentanone *O*-allyloxime (*E,E*-**5**) (500 mg, 0.002 mol) in acetonitrile (300 cm^3) showed formation of the geometrical isomers of (**5**), with formation of the *Z,E*-isomer being the fastest process. Continued irradiation led to very slow formation of 2,3-dihydro-1*H*-cyclopenta[*b*]quinoline **7** (approx. 4% of the rate in methanol).

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In methanol containing isoprene: Five solutions of *E,E*-2-benzylidenecyclopentanone *O*-allyloxime (*E,E*-**5**) (0.0025M) in methanol containing different concentrations of freshly distilled isoprene (0.0M, 0.001M, 0.01M, 0.1M and 1.0M) were prepared. A 20 cm^3 aliquot of each solution was placed in a separate quartz tube, the solutions were degassed by bubbling argon through each one for 10 minutes and the tubes were then stoppered and placed on a carousel apparatus circling an immersion well containing a photolysis lamp fitted with a Pyrex filter. The solutions were irradiated for 15 minutes after which samples from each tube were analysed by GC. Each sample contained a mixture of all four isomers of oxime **5** and cyclopenta[b]quinoline **7** in the same ratios irrespective of isoprene concentration present.

General procedure for preparation of 2-benzylidenecyclopentanones

Preparations were conducted on a 25-100 mmol scale using the following typical procedure, based on that of Birkofer *et al*.[21] Cyclopentanone (100 mmol) and morpholine (9.0 cm^3, 100 mmol) in toluene (100 cm^3) were heated under reflux in a Dean and Stark apparatus until no further water collected. The appropriate aldehyde (100 mmol) was then added to the cooled solution and the mixture heated under reflux until no further water collected. A 1:1 mixture of water and concentrated hydrochloric acid (50 cm^3) was added dropwise to the stirring cooled solution which was left for a further hour, then washed with 10% aqueous sodium carbonate (100 cm^3) and dried ($MgSO_4$). Rotary evaporation and recrystallisation from methanol, unless stated otherwise, yielded the corresponding 2-benzylidenecyclopentanone.

2-Benzylidenecyclopentanone K1 (63%, from benzaldehyde), mp 69-70 °C (ethanol) (lit.,[22] 71-72 °C); δ_H 2.03 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.41 (2H, t, *J* 7.9, $CH_2C{=}O$), 2.98 (2H, td, J_t 7.5, J_d 2.5, $CH_2C{=}C$), 7.39 (4H, m, HC=C + 3 x arH) and 7.53 (2H, d, *J* 7.5, 2 x arH); δ_C 20.11, 29.28, 37.74 (3 x CH_2), 128.63, 129.10, 130.46, 132.39, 135.39, 135.98 (4 x arC + C=C) and 208.47 (C=O).

2-(2-Methylbenzylidene)cyclopentanone K2 (60%, from 2-methylbenzaldehyde), mp 68-70 °C (lit. [23] 67-68 °C) (Found: C, 83.5; H, 7.5. $C_{13}H_{14}O$ requires: C, 83.8; H, 7.5%); δ_H 1.81 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.23 (3H, s, Me), 2.54 (2H, t, *J* 7.5, $CH_2C{=}O$), 2.72 (2H, td, J_t 7.5, J_d 2.6, $CH_2C{=}C$), 7.07 (3H, m, 3 x arH), 7.25 (1H, d, *J* 7.3, arH) and 7.43 (1H, t, *J* 2.6, HC=C); δ_C 19.88 (Me), 20.43, 29.32, 38.98 (3 x CH_2), 126.66, 128.53, 129.03, 129.68, 130.41, 134.18, 136.71, 138.77 (6 x arC + C=C) and 207.87 (C=O). Table of Contents

2-(3-Methylbenzylidene)cyclopentanone K3 (75%, from 3-methylbenzaldehyde), mp 30-31°C (dichloromethane) (Found: C, 83.6; H, 7.6. $C_{13}H_{14}O$ requires: C, 83.8; H, 7.6%); δ_H 2.01 (2H, qn, *J* 7.2, $CH_2CH_2CH_2$), 2.38 (2H, t, *J* 7.2, CH_2CO), 2.39 (3H, s, Me), 2.95 (2H, td, J_t 7.2, J_d 2.4, $CH_2C=C$), 7.15 (1H, d, *J* 2.4, arH) and 7.25 (4H, m, 3 x arH + HC=C).

2-(4-Methylbenzylidene)cyclopentanone K4 (59%, from 4-methylbenzaldehyde), mp 64-65 °C (lit.,[24] 62-63 °C); δ_H 1.93 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.30 (5H, m, Me + $CH_2C=O$), 2.87 (2H, td, J_t 7.5, J_d 2.5, $CH_2C=C$), 7.14 (2H, d, *J* 8.0, 2 x arH), 7.29 (1H, t, *J* 2.5, HC=C) and 7.35 (2H, d, *J* 8.0, 2 x arH); δ_C 21.35(Me), 20.09, 29.22, 37.64 (3 x CH_2), 129.33, 130.46, 132.21, 132.69, 134.95, 139.57 (4 x arC + C=C) and 208.03 (C=O).

2-(3,4-Dimethylbenzylidene)cyclopentanone K5 (43%, from 3,4-dimethylbenzaldehyde), mp 85-87 °C (Found: C, 83.9; H, 7.9. $C_{14}H_{16}O$ requires C, 84.0; H, 8.05%); δ_H 1.91 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.19 (6H, s, Me), 2.29 (2H, t, *J* 7.6, CH_2CO), 2.86 (2H, td, J_t 7.6, J_d 2.5, $CH_2C=C$), 7.07 (1H, d, *J* 7.6) and 7.19 (2H, m) (3 x arH) and 7.25 (1H, t, *J* 2.5, HC=C); δ_C 19.66, 19.71, 20.02, 29.24, 37.63 (Me and 3 x CH_2), 127.92, 129.86, 131.79, 132.38, 133.01, 134.80, 136.77, 138.36 (6 x arC + C=C) and 208.04 (C=O).

2-(3-Methyl-4-methoxybenzylidene)cyclopentanone K6 (74% from 4-methoxy-3-methylbenzaldehyde), mp 80-82 °C (Found: C, 77.5; H, 7.5. $C_{14}H_{16}O_2$ requires C, 77.75; H, 7.5%); δ_H 1.95 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.16 (3H, s, Me), 2.32 (2H, t, *J* 7.5, CH_2CO), 2.89 (2H, td, J_t 7.5, J_d 2.4, $CH_2C=C$), 3.79 (3H, s, OMe), 6.79 (1H, d, *J* 8.4) and 7.31 (1H, dd, *J* 8.4, 2.0) (2 x arH), 7.26 (2H, m, arH + HC=C); δ_C 16.33 (Me), 20.13, 29.30, 37.74 (3 x CH_2), 55.38 (OMe), 109.92, 126.99, 127.75, 130.15, 132.52, 132.96, 133.40, 158.85 (6 x arC + C=C) and 208.16 (C=O).

2-(3-*t*-Butylbenzylidene)cyclopentanone K7 (92%, from 3-*t*-butylbenzaldehyde), mp 54-55 °C (Found: C, 83.8; H, 8.9. $C_{16}H_{20}O$ requires C, 84.2; H, 8.8%); δ_H 1.27 (9H, s CMe_3), 1.96 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.34 (2H, t, *J* 7.6, CH_2CO), 2.92 (2H, td, J_t 7.6, J_d 2.8, $CH_2C=C$), 7.28-7.35 (4H, m, 4 x arH) and 7.49 (1H, t, *J* 2.8, HC=C); δ_C 20.21, 29.38, 31.22 (3 x CH_2), 34.65 (CMe_3), 37.80 (CMe_3), 126.53, 127.39, 127.84, 128.39, 132.92, 135.14, 135.62, 151.50 (6 x arC + C=C) and 208.16 (C=O). Table of Contents

2-(2-Methoxybenzylidene)cyclopentanone K8 (72%, from 2-methoxybenzaldehyde), mp 85-86 °C (lit, [25] 77 °C, from hexane) (Found: C, 76.9; H, 6.9. $C_{13}H_{14}O_2$ requires: C, 77.2; H, 7.0%); δ_H 2.00 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.40 (2H, t, *J* 7.5, $CH_2C=O$), 2.92 (2H, td, J_t 7.5,

J_d 2.4, $CH_2C=C$), 3.86 (3H, s, OMe), 6.91 (1H, d, *J* 7.9, arH), 6.98 (1H, t, *J* 7.9, arH), 7.34 (1H, t, *J* 7.9, arH), 7.47 (1H, d, *J* 7.9, arH) and 7.81 (1H, t, *J* 2.4, HC=C); δ_C 20.43, 29.52, 37.96 (3 x CH_2), 55.48 (OMe), 110.71, 120.22, 124.55, 126.91, 129.67, 130.81, 136.05, 158.87 (6 x arC + C=C) and 208.06 (C=O).

2-(3-Methoxybenzylidene)cyclopentanone K9 (74%, from 3-methoxybenzaldehyde), mp 57-58 °C (light petroleum) (lit.,[26] 58-59 °C) (Found: C, 77.0; H, 7.1. $C_{13}H_{14}O_2$ requires C, 77.2; H, 7.0%.); δ_H 2.02 (2H, qn, *J* 7.2, $CH_2CH_2CH_2$), 2.39 (2H, t, *J* 7.3, CH_2CO), 2.96 (2H, td, J_t 7.2, J_d 2.7, $CH_2C=C$), 3.83 (3H, s, OMe), 6.89 (1H, m, arH), 7.03 (1H, br s, HC=C), 7.10 (1H, d, *J* 7.3, arH) and 7.29 (2H, m, 2 x arH); δ_C 19.99, 29.19, 37.63 (3 x CH_2), 55.08 (OMe), 114.79, 115.54, 122.88, 129.49, 131.99, 136.14, 136.65, 159.43 (6 x arC and C=C) and 207.97 (C=O).

2-(4-Methoxybenzylidene)cyclopentanone K10 (71%, from 4-methoxybenzaldehyde), mp 68-69 °C (lit.,[27] 68-69 °C); δ_H 1.83 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.20 (2H, t, *J* 7.5, $CH_2C=O$) 2.75 (2H, td, J_t 7.5, J_d 2.5, $CH_2C=C$), 3.65 (3H, s, OMe), 6.74 (2H, d, *J* 8.8, 2 x arH), 7.16 (1H, t, *J* 2.5, HC=C) and 7.31 (2H, d, *J* 8.8, 2 x arH); δ_C 20.06, 29.18, 37.77 (3 x CH_2), 55.27 (OMe), 114.213, 128.22, 132.21, 132.30, 133.67, 160.50 (4 x arC + C=C) and 208.13 (C=O).

2-(2,5-Dimethoxybenzylidene)cyclopentanone K11 (68%, from 2,5-dimethoxybenzaldehyde), mp 73-75 °C (Found: C, 72.4; H, 7.0. $C_{14}H_{16}O_3$ requires C, 72.4; H, 7.0%); δ_H 1.93 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.33 (2H, t, *J* 7.6, $CH_2C=O$), 2.86 (2H, td, J_t 7.6, J_d 2.8, $CH_2C=C$), 3.72 (3H, s, OMe), 3.75 (3H, s, OMe), 6.77 (1H, d, *J* 8.8, arH), 6.81 (1H, dd, *J* 8.8, 3.0, arH), 6.96 (1H, d, *J* 3.0, arH) and 7.69 (1H, t, *J* 2.8, HC=C); δ_C 20.33, 29.43, 37.88 (3 x CH_2), 55.75, 56.04 (OMe), 111.69, 115.39, 125.31, 126.82, 126.87, 136.29, 153.03, 153.42 (6 x arC + C=C) and 207.87 (C=O).

2-(3,4-Dimethoxybenzylidene)cyclopentanone K12 (82%, from 3,4-dimethoxybenzaldehyde), mp 111-112 °C (lit.,[28] 113-114 °C); δ_H 1.97 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.33 (2H, t, *J* 7.4, CH_2CO), 2.91 (2H, td, J_t 7.4, J_d 2.4, $CH_2C=C$), 3.84 (3H, s, OMe), 3.85 (3H, s, OMe), 6.84 (1H, d, *J* 8.4), 7.00 (1H, d, *J* 1.8) and 7.09 (1H, dd, *J* 8.4, 1.8) (3 x arH), 7.27 (1H, t, *J* 2.4, HC=C); δ_C 20.05, 29.16, 37.63 (3 x CH_2), 55.76, 55.85 (both OMe), 111.00, 113.08, 124.30, 128.42, 132.39, 133.79, 148.76, 150.13 (6 x arC + C=C) and 207.90 (C=O).

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2-(3-Methoxy-4-methylbenzylidene)cyclopentanone K13 (50%, from 3-methoxy-4-methylbenzaldehyde), mp 91-92 ^{o}C (Found: C, 77.3; H, 7.5. $C_{14}H_{16}O_2$ requires C, 77.75; H, 7.5%); δ_H 2.06 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.27 (1H, s, Me), 2.43 (2H, t, *J* 7.5, CH_2CO), 3.01 (2H, td, J_t 7.5, J_d 2.6, CH_2C=C), 3.88 (1H, s, OMe), 7.00 (1H, s), 7.10 (1H, d, *J* 7.6) and 7.20 (1H, d, *J* 7.6) (3 x arH), 7.39 (1H, t, *J* 2.6, HC=C); δ_C 16.29 (Me), 20.15, 29.36, 37.77 (3 x CH_2), 55.21 (OMe), 111.82, 122.74, 128.75, 130.74, 132.73, 134.34, 135.05, 157.70 (6 x arC + C=C) and 208.11 (C=O).

2-(3-Hydroxybenzylidene)cyclopentanone K14 (72%, from 3-hydroxybenzaldehyde), mp 161-162 ^{o}C (Found: C, 76.5; H, 6.5. $C_{12}H_{12}O_2$ requires C, 76.6; H, 6.4%; δ_H 2.02 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.40 (2H, t, *J* 7.6, CH_2C=O), 2.96 (2H, td, J_t 7.6, J_d 2.6, CH_2C=C), 5.73 (1H, s, OH), 6.86 (1H, dd, *J* 7.6, 2.6), 7.03 (1H, t, *J* 1.8), 7.09 (1H, d, 7.6) and 7.27 (1H, t, *J* 8.0) (4 x arH), 7.33 (1H, t, *J* 2.8, HC=C); δ_C CD_3OD 21.15, 30.38, 38.61 (3 x CH_2), 116.77, 117.85, 123.28, 130.79, 133.61, 137.37, 137.77, 158.82 (6 x arC + C=C) and 210.49 (C=O).

2-(4-Hydroxybenzylidene)cyclopentanone K15 (57%, from 4-hydroxybenzaldehyde), mp 187-188 °C (lit.,[29] 188-192 °C); δ_H 1.95 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.35 (2H, t, *J* 7.5, CH_2C=O), 2.88 (2H, td, J_t 7.5, J_d 2.7, CH_2C=C), 6.11 (1H, s, OH), 6.83 (2H, d, *J* 8.8, 2 x arH), 7.30 (1H, t, *J* 2.7, HC=C) and 7.39 (2H, d, *J* 8.8, 2 x arH); δ_C CD_3OD 21.48, 30.48, 38.84 (3 x CH_2), 116.99, 128.33, 134.08, 134.32, 134.40, 160.77 (4 x arC + C=C) and 210.94 (C=O).

2-(3-*N*,*N*-Dimethylaminobenzylidene)cyclopentanone K16 (48%, from 3-*N*,*N*-dimethylaminobenzaldehyde; the reaction was analogous to the general synthesis of the 2-benzylidenecyclopentanones, except that, following acid hydrolysis, the aqueous layer was separated, neutralised with 10% aqueous sodium hydroxide, extracted with diethyl ether and the extract washed with water, dried, concentrated and the product recrystallised), mp 92-93 ^{o}C (Found: C, 77.8; H, 8.0; N, 6.5. $C_{14}H_{17}NO$ requires C, 78.1; H, 8.0; N, 6.5%); δ_H 1.95 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.33 (2H, t, *J* 7.6, CH_2CO), 2.91 (8H, m, NMe_2 and CH_2C=C), 6.68 (1H, dd, *J* 8.4, 2.4), 6.79 (1H, br s), 6.85 (1H, d, *J* 7.6) and 7.21 (1H, t, *J* 7.9) (4 x arH), 7.30 (1H, t, *J* 2.8, HC=C); δ_C 20.17, 29.35, 37.80 (3 x CH_2), 40.47 (NMe_2), 113.64, 114.72, 118.57, 129.22, 133.48, 135.51, 136.09, 150.58 (6 x arC + C=C) and 208.22 (C=O).

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2-(4-*N*,*N*-Dimethylaminobenzylidene)cyclopentanone K17 (72%, from 4-*N*,*N*-dimethylaminobenzaldehyde, using the method reported above for 2-(3-*N*,*N*-

dimethylaminobenzaldehyde), mp 149-151 °C (Found: C, 77.7; H, 8.0; N, 6.5. $C_{14}H_{17}NO$ requires C, 78.1; H, 8.0; N, 6.5%); δ_H 2.01 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.37 (2H, t, *J* 7.5, $CH_2C=O$), 2.95 (2H, td, J_t 7.5, J_d 2.4, $CH_2C=C$), 3.03 (6H, s, NMe_2), 6.71 (2H, d, *J* 8.8, 2 x arH), 7.35 (1H, t, *J* 2.4, HC=C) and 7.47 (2H, d, *J* 8.8, 2 x arH); δ_C 20.21, 29.46, 37.85 (3 x CH_2), 40.16 (NMe_2), 111.82, 123.31, 131.07, 132.54, 133.39, 150.96 (4 x arC + C=C) and 208.26 (C=O).

2-(1-Naphthymethylene)cyclopentanone K20 (72%, from 1-naphthaldehyde), mp 67-70 °C (Found: C, 86.1; H, 6.4%. $C_{16}H_{14}O$ requires: C, 86.45; H, 6.35%); δ_H 1.87 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.35 (2H, t, *J* 7.5, $CH_2C=O$), 2.78 (2H, td, J_t 7.5, J_d 2.6, $CH_2C=C$), 7.42 (4H, m, 4 x arH), 7.75 (2H, m, 2 x arH) and 8.03 (2H, m, HC=C + arH); δ_C 20.54, 29.65, 38.22 (3 x CH_2), 123.93, 125.07, 126.18, 126.64, 127.01, 128.68, 128.80, 129.70, 132.25, 132.27, 133.53, 138.31 (10 x arC + C=C) and 207.71 (C=O).

2-(2-Naphthylmethylene)cyclopentanone K21 (79%, from 2-naphthaldehyde), mp 128-129 °C (Found: C, 86.2; H, 6.4. $C_{16}H_{14}O$ requires: C, 86.5; H, 6.35%); δ_H 2.04 (2H, qn, *J* 7.2, $CH_2CH_2CH_2$), 2.42 (2H, t, *J* 7.2, CH_2CO), 3.05 (2H, td, J_t 7.2, J_d 2.4, $CH_2C=C$), 7.48 (4H, m, 3 x arH + HC=C) and 7.80 (4H, m, 4 x arH); δ_C 19.73, 28.99, 37.34 (3 x CH_2), 126.11, 126.63, 126.71, 127.22, 127.84, 128.07, 130.57, 131.96, 132.60, 132.73, 132.96, 135.80 (10 x arC and 2 x C=C) and 207.64 (C=O).

2-(10*H*-Phenothiazin-1-ylmethylene)cyclopentanone K22 (crude waxy solid washed through a silica column with 80:20 light petroleum/ethyl acetate prior to recrystallisation; 17%, from 1-formylphenothiazine **S1**), mp 171-172 °C (Found: C, 73.5; H, 5.1; N, 4.65. $C_{18}H_{15}NOS$ requires C, 73.7; H, 5.15; N, 4.8%); δ_H 1.98 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.45 (2H, t, *J* 7.5, $CH_2C=O$), 2.83 (2H, td, J_t 7.5, J_d 2.7, $CH_2C=C$), 6.37 (1H, s, NH), 6.69 (1H, dd, *J* 7.8, 1.0, arH), 6.84 (2H, m, 2 x arH), 7.01 (4H, m, 4 x arH) and 7.45 (1H, t, *J* 2.7, HC=C); δ_C 20.37, 29.27, 38.09 (3 x CH_2), 115.28, 118.40, 119.29, 120.51, 121.62, 123.12, 125.29, 126.55, 127.44, 127.53, 127.61, 139.34, 140.89, 140.98 (12 x arC + C=C) and 207.61 (C=O).

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2-(2-Furylmethylene)cyclopentanone K23 (37%, from 2-furaldehyde), mp 58-59 °C (lit.,[30] 60.5 °C); δ_H 2.03 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.40 (2H, t, *J* 7.9, $CH_2C{=}O$), 2.98 (2H, td, J_t 7.4, J_d 2.6, $CH_2C{=}C$), 6.51 (1H, dd, *J* 3.4, 2.0, furyl H-4), 6.66 (1H, d, *J* 3.4, furyl H-3), 7.16 (1H, t, *J* 2.6, HC=C) and 7.56 (1H, d, *J* 2.0, furyl H-5); δ_C 19.60, 28.87, 37.86 (3 x CH_2), 112.35, 115.78, 118.61, 133.48, 144.76, 152.10 (C=C + furyl) and 207.70 (C=O).

2-(2-Thienylmethylene)cyclopentanone K24 (60%, from thiophene-2-carboxaldehyde), mp 77-78 °C (lit.,[31] 79 °C) (Found: C, 67.3; H, 5.45; S, 17.8. $C_{10}H_{10}OS$ requires: C, 67.4; H, 5.65; S, 18.0%); δ_H 2.05 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.40 (2H, t, *J* 7.4, $CH_2C{=}O$), 2.85 (2H, t, *J* 7.4, $CH_2C{=}C$), 7.12 (1H, t, *J* 3.7, thienyl H-4), 7.32 (1H, d, *J* 3.7, thienyl H-3), 7.51 (1H, d, *J* 3.7, thienyl H-5), and 7.56 (1H, br s, C=CH); δ_C 19.63, 29.01, 38.01 (3 x CH_2), 124.93, 127.89, 129.92, 132.60, 133.58, 139.90 (C=C + thienyl) and 207.50 (C=O).

2-Cinnamylidenecyclopentanone K25 (47%, from *E*-cinnamaldehyde), mp 89-90 °C (lit.,[32] 87-89 °C); δ_H 1.95 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.33 (2H, t, *J* 7.5, $CH_2C{=}O$), 2.75 (2H, td, J_t 7.5, J_d 2.8, $CH_2C{=}C$), 6.81 (1H, dd, *J* 15.7, 10.5, HC=C), 6.89 (1H, d, *J* 15.7, HC=C), 7.02 (1H, dt, J_d 10.5, J_t 2.8, HC=C), 7.21-7.31 (3H, m, 3 x arH) and 7.41 (2H, dd, *J* 7.2, 1.6, 2 x arH); δ_C 19.85, 27.36, 38.65 (3 x CH_2), 124.69, 127.16, 128.80, 128.99, 131.53, 136.44, 137.04, 141.48 (4 x arC + 2 x C=C) and 207.57 (C=O).

2-(2-Nitrobenzylidene)cyclopentanone K26 (40%, from 2-nitrobenzaldehyde), mp 83-86 °C (Found: C, 66.05; H, 5.05; N, 6.25. $C_{12}H_{11}NO_3$ requires: C, 66.35; H, 5.1; N, 6.45%); δ_H 1.89 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.30 (2H, t, *J* 7.5, $CH_2C{=}O$), 2.67 (2H, td, J_t 7.5, J_d 2.7, $CH_2C{=}C$), 7.40 (2H, m, 2 x arH), 7.48 (1H, t, *J* 2.7, HC=C), 7.54 (1H, td, J_t 7.6, J_d 1.1, arH) and 7.91 (1H, dd, *J* 8.7, 1.1, arH); δ_C 20.14, 28.64, 37.85 (3 x CH_2), 124.71, 127.01, 129.29, 130.46, 131.00, 132.96, 139.41, 148.70 (6 x arC + C=C), 206.69 (C=O).

2-(3-Nitrobenzylidene)cyclopentanone K27 (67%, from 3-nitrobenzaldehyde), mp 111-112°C (Found: C, 66.1; H, 5.1; N, 6.3. $C_{12}H_{11}NO_3$ requires C, 66.35; H, 5.1; N, 6.45%); δ_H 2.07 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.43 (2H, t, *J* 7.6, CH_2CO), 3.05 (2H, td, J_t 7.6, J_d 2.9, $CH_2C{=}C$), 7.36 (1H, t, *J* 2.9, HC=C), 7.58 (1H, t, *J* 8.0), 7.79 (1H, d, *J* 8.0), 8.18 (1H, d, *J* 8.0) and 8.31 (1H, s) (4 x arH); δ_C 20.04, 29.21, 37.54 (3 x CH_2), 124.45, 124.11, 129.19, 129.67, 136.13, 137.16, 138.78, 148.39 (6 x arC and C=C) and 207.35 (C=O).

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2-(4-Nitrobenzylidene)cyclopentanone K28 (45%, from 4-nitrobenzaldehyde), mp 141-142 °C (lit.,[33] 145-146 °C) (Found: C, 66.05; H, 5.05; N, 6.3. $C_{12}H_{11}NO_3$ requires: C, 66.35; H, 5.1; N, 6.45%); δ_H 2.02 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.39 (2H, t, *J* 7.5, $CH_2C{=}O$), 2.94 (2H, td, J_t 7.5, J_d 2.7, $CH_2C{=}C$), 7.32 (1H, t, *J* 2.7, HC=C), 7.60 (2H, d, *J* 8.8, 2 x arH) and 8.19 (2H, d, *J* 8.8, 2 x arH); δ_C 20.02, 29.36, 37.63 (3 x CH_2), 123.81, 129.23, 130.75, 139.83, 141.86 (4 x arC + C=C) and 207.53 (C=O).

2-(2-Chlorobenzylidene)cyclopentanone K29 (69%, from 2-chlorobenzaldehyde), mp 55-57 °C (lit., bp 128 °C /0.15 mm) (Found: C, 69.6; H, 5.2. $C_{12}H_{11}ClO$ requires: C, 69.7; H, 5.4%); δ_H 1.87 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.28 (2H, t, *J* 7.5, $CH_2C{=}O$), 2.75 (2H, td, J_t 7.5, J_d 2.7, $CH_2C{=}C$), 7.15 (2H, m, 2 x arH), 7.29 (1H, m, arH), 7.36 (1H, m, arH) and 7.56 (1H, t, *J* 2.7, HC=C); δ_C 20.25, 29.16, 37.78 (3 x CH_2), 126.48, 128.00, 129.83, 130.01, 130.14, 133.55, 135.76, 138.20 (6 x arC + C=C), and 207.40 (C=O).

2-(3-Chlorobenzylidene)cyclopentanone K30 (63% from 3-chlorobenzaldehyde), mp 47-48°C[34] (dichloromethane) (Found: C, 69.8; H, 5.4; Cl, 17.5. $C_{12}H_{11}ClO$ requires C, 69.7; H, 5.4; Cl, 17.15%); δ_H 2.03 (2H, qn, *J* 7.8, $CH_2CH_2CH_2$), 2.41 (2H, t, *J* 7.8, CH_2CO), 2.94 (2H, td, J_t 7.8, J_d 2.9, $CH_2C{=}C$) and 7.25-7.48 (5H, m, 4 x arH + HC=C); δ_C 19.92, 29.09, 37.52 (3 x CH_2), 128.48, 128.98, 129.70, 129.80, 130.34, 130.45, 134.39, 137.12 (6 x arC + C=C) and 207.65 (C=O).

2-(4-Chlorobenzylidene)cyclopentanone K31 (61%, from 4-chlorobenzaldehyde), mp 77-79 °C (lit.,[35] 78-80 °C); δ_H 2.05 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.42 (2H, t, *J* 7.5, $CH_2C{=}O$), 2.95 (2H, td, J_t 7.5, J_d 2.6, $CH_2C{=}C$), 7.33 (1H, t, *J* 2.6, HC=C), 7.38 (2H, d, *J* 8.5, 2 x arH) and 7.47 (2H, d, *J* 8.5, 2 x arH); δ_C 20.10, 29.25, 37.70 (3 x CH_2), 128.83, 130.87, 131.57, 133.96, 135.01, 136.47 (4 x arC + C=C) and 207.84 (C=O).

2-(3-Cyanobenzylidene)cyclopentanone K32 (68%, from 3-cyanobenzaldehyde), mp 72-74 °C (Found: C, 79.2; H, 5.7; N, 7.1. $C_{13}H_{11}NO$ requires C, 79.2; H, 5.6; N,7.1%); δ_H 2.05 (2H, qn, *J* 7.8, $CH_2CH_2CH_2$), 2.39 (2H, t, *J* 7.8, CH_2CO), 2.91 (2H, td, J_t 7.8, J_d 2.8, $CH_2C{=}C$), 7.22 (1H, t, *J* 2.8, HC=C), 7.49 (1H, t, *J* 7.8), 7.58 (1H, d, *J* 7.8), 7.68 (1H, d, *J* 7.8) and 7.72 (1H, s) (4 x arH); δ_C 19.85, 29.01, 37.43 (3 x CH_2), 112.76 (C≡N), 118.15, 129.10, 129.40, 131.97, 132.92, 134.23, 136.53, 138.31 (6 x arC + C=C) and 207.29 (C=O).

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2-(4-Cyanobenzylidene)cyclopentanone K33 (72%, from 4-cyanobenzaldehyde), mp 112-113 °C (Found: C, 78.9; H, 5.7; N, 7.3. $C_{13}H_{11}NO$ requires: C, 79.2; H, 5.6; N,7.1%); δ_H 2.03 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.40 (2H, t, *J* 7.5, $CH_2C=O$), 2.95 (2H, td, J_t 7.5, J_d 2.9, $CH_2C=C$), 7.29 (1H, t, *J* 2.9, HC=C), 7.56 (2H, d, *J* 8.9, 2 x arH) and 7.65 (2H, d, *J* 8.9, 2 x arH); δ_C 19.90, 29.19, 37.49 (3 x CH_2), 118.38 (CN), 112.04, 129.57, 130.43, 132.18, 139.16, 139.82 (4 x arC + C=C) and 207.32 (C=O).

2-(2,4-Difluorobenzylidene)cyclopentanone K34 (67%, from 2,4-difluorobenzaldehyde), mp 76-78 °C (Found: C, 69.1; H, 4.8%. $C_{12}H_{10}F_2O$ requires C, 69.2; H, 4.8%); δ_H 1.98 (2H, qn, *J* 7.8, $CH_2CH_2CH_2$), 2.35 (2H, t, *J* 7.8, $CH_2C=O$), 2.83 (2H, td, J_t 7.8, J_d 2.9, $CH_2C=C$), 6.79 (1H, td, J_t 8.9, J_d 2.9, arH), 6.88 (1H, td, J_t 7.8, J_d 2.0, arH), 7.41-7.50 (2H, m, HC=C + arH).

2-(3-Fluorobenzylidene)cyclopentanone K35 (77%, from 3-fluorobenzaldehyde), mp 58-60 ^oC (light petroleum) (Found: C, 75.6; H, 5.9; F, 10.2. $C_{12}H_{11}FO$ requires C, 75.8; H, 5.8; F, 10.0%); δ_H 1.95 (2H, qn, *J* 7.8, $CH_2CH_2CH_2$), 2.34 (2H, t, *J* 7.8, CH_2CO) , 2.87 (2H, td, J_t 7.8, J_d 2.9, $CH_2C=C$), 6.98 (1H, td, J_t 8.9, J_d 2.9) and 7.13 (1H, d, *J* 8.9) (2 x arH) and 7.25 (3H, m, 2 x arH + HC=C).

2-(4-Aminobenzylidene)cyclopentanone K18: 2-(4-Nitrobenzylidene)cyclopentanone **K28** (5.86 g, 27 mmol) and tin(II)chloride dihydrate (30.5 g, 135 mmol) in ethanol (85 cm^3) were heated under reflux for 30 minutes with stirring under nitrogen. The mixture was cooled, brought to pH~8 by addition of 20% aqueous sodium carbonate and extracted with ethyl acetate (2 x 50 cm^3). The extract was washed with brine (50 cm^3), dried ($MgSO_4$), concentrated and recrystallised to yield red/orange crystals of 2-(4-aminobenzylidene)cyclopentanone **K18** (2.6 g, 41%), mp 177-179 °C (decomp.) (Found: C, 76.7; H, 7.0; N, 7.4. $C_{12}H_{13}NO$ requires C, 77.0; H, 7.0; N, 7.5%); δ_H 1.95 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.31 (2H, t, *J* 7.5, $CH_2C=O$), 2.91 (2H, td, J_t 7.5, J_d 2.6, $CH_2C=C$), 3.96 (2H, s, NH_2), 6.67 (2H, d, *J* 8.6, 2 x arH) 7.30(1H, t, *J* 2.6, HC=C) and 7.32 (2H, d, *J* 8.6, 2 x arH); δ_C 20.11, 29.35, 37.74 (3 x CH_2), 114.74, 125.71, 131.96, 132.54, 132.94, 147.87 (4 x arC + C=C) and 208.23 (C=O).

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2-(3-Aminobenzylidene)cyclopentanone K19 (68%, from 2-(3-nitrobenzylidenecyclopentanone **K27**, using the method reported above for 2-(4-aminobenzylidene)cyclopentanone **K18**), mp 119-120 ^{o}C (Found: C, 76.8; H, 7.0; N, 7.4. $C_{12}H_{13}NO$ requires C, 77.0; H, 7.0; N, 7.5%); δ_H 2.00 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.38 (2H, t, *J* 7.5, CH_2CO), 2.95 (2H, td, J_t 7.5, J_d 2.7, $CH_2C{=}C$), 3.72 (2H, s, NH_2), 6.68 (1H, dd, *J* 8.0, 2.0), 6.73 (1H, t, *J* 2.0), 6.94 (1H, d, *J* 8.0) and 7.18 (1H, t, *J* 8.0) (4 x arH), 7.28 (1H, t, *J* 2.7, $HC{=}C$); δ_C 20.11, 29.36, 37.77 (3 x CH_2), 116.22, 116.73, 120.91, 129.47, 132.62, 135.81, 136.41, 146.55 (6 x arC + C=C) and 208.32 (C=O).

2-Benzylidenecyclohexanone K36 (59%, using cyclohexanone in the general method), mp 52-54°C (lit.,[36] 54°C); δ_H 1.74 (2H, m, $CH_2CH_2CH_2$), 1.91 (2H, m, $CH_2CH_2CH_2$), 2.52 (2H, t, *J* 6.7, CH_2CO), 2.82 (2H, td, J_t 6.4, J_d 2.1, $CH_2C{=}C$), 7.34 (5H, m, 5 x arH) and 7.50(1H, t, *J* 2.1, C=CHPh).

General procedure for preparation of 2-benzylidenecyclopentanone oximes[37]

Hydroxylamine hydrochloride (70 mmol; 5.0g), ethanol (50 cm^3), pyridine (5 cm^3) and the 2-benzylidenecylopentanone (50 mmol) were heated under reflux for 1 hour. The ethanol was removed by rotary evaporation and chloroform (75 cm^3) was added. The mixture was washed with 1 M hydrochloric acid (2x100 cm^3) and water (2x100 cm^3) and dried ($MgSO_4$). Rotary evaporation and recrystallisation from methanol yielded the required benzylidenecyclopentanone oxime.

***E*,*E*-2-Benzylidenecyclopentanone oxime O1** (71%, from **K1**), mp 127-129 °C (lit., 129 °C); δ_H 1.87 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.68 (2H, t, *J* 7.6, $CH_2C{=}N$), 2.80 (2H, td, J_t 7.4, J_d 2.5, $CH_2C{=}C$), 7.21 (1H, t, *J* 2.5, $HC{=}C$), 7.26 (1H, t, *J* 7.4, arH), 7.35 (2H, t, *J* 7.4, 2 x arH), 7.41 (2H, d, *J* 7.4, 2 x arH) and 9.55 (1H, br s, OH); δ_C 22.45, 27.09, 31.45 (3 x CH_2), 123.25, 127.35, 128.30, 129.28, 136.59, 137.02 (4 x arC + C=C) and 163.73 (C=N).

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2-Diphenylmethylenecyclopentanone oxime O1a (74%, from **S5**), mp 178-180 °C (Found: C, 82.0; H, 6.6; N, 5.1. $C_{18}H_{17}NO$ requires: C, 82.1; H, 6.5; N, 5.3%); δ_H 1.72 (2H, qn, *J* 7.3, $CH_2CH_2CH_2$), 2.54 (4H, m, $CH_2C{=}N$ and $CH_2C{=}C$), 7.12 (4H, m) and 7.24 (6H, m) (10 x arH) and 7.85 (1H, br s, NOH); δ_C 21.90, 27.75, 34.38 (3 x CH_2), 126.82, 127.15, 127.73, 127.94, 129.53, 133.95, 139.40, 142.13, 143.08 (arC + C=C) and 162.06 (C=N).

2-(2-Methylbenzylidene)cyclopentanone oxime O2 (67%, from **K2**), mp 74-76 °C (Found: C, 77.5; H, 7.5; N, 6.9. $C_{13}H_{15}NO$ requires: C, 77.6; H, 7.5; N, 7.0%); δ_H 1.77 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.30 (3H, s, Me), 2.63 (4H, m, $CH_2C{=}N$ and $CH_2C{=}C$), 7.13 (3H, m) and 7.27 (2H, m) (HC=C + 4 x arH) and 9.41 (1H, br s, OH); δ_C 19.99 (Me), 22.47, 27.33, 30.97 (3 x CH_2), 121.34, 125.46, 127.48, 128.52, 130.03, 135.95, 137.04, 137.30 (6 x arC + C=C) and 163.60 (C=N).

2-(3-Methylbenzylidene)cyclopentanone oxime O3 (82%, from **K3**), mp 147-149 °C (Found: C, 77.4; H, 7.45; N, 6.7. $C_{13}H_{15}NO$ requires: C, 77.6; H, 7.5; N, 7.0%); δ_H 1.87 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.31 (3H, s, Me), 2.65 (2H, t, *J* 7.4, $CH_2C{=}N$), 2.95 (2H, t *J* 7.4, $CH_2C{=}C$), 7.16 (s, 1 H, HC=C), 7.07 (1H, d, *J* 7.4) and 7.25 (3H, m) (4 x arH), 8.95 (1H, broad, OH).

2-(4-Methylbenzylidene)cyclopentanone oxime O4 (67%, from **K4**), mp 132-134 °C (decomp.) (lit., 139-140 °C) (Found: C, 77.9; H, 7.6; N, 6.7. $C_{13}H_{15}NO$ requires: C, 77.6; H, 7.5; N, 7.0%); δ_H 1.87 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.35 (3H, s, Me), 2.67 (2H, t, *J* 7.5, $CH_2C{=}N$), 2.79 (2H, td, J_t 7.5, J_d 2.6, $CH_2C{=}C$), 7.12 (3H, m, HC=C + 2 x arH) 7.31 (2H, d, *J* 8.4, 2 x arH) and 9.14 (1H, br s, OH); δ_C 21.26 (Me), 22.48, 27.07, 31.45 (3 x CH_2), 123.18, 129.07, 129.27, 134.27, 135.64, 137.35 (4 x arC + C=C) and 163.85 (C=N).

2-(3,4-Dimethylbenzylidene)cyclopentanone oxime O5 (87%, from **K5**), mp 157-159 °C (decomp.) (Found: C, 77.8; H, 7.95; N, 6.4. $C_{14}H_{17}NO$ requires C, 78.1; H, 8.0; N, 6.5%); δ_H[38a] 1.80 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.19 (6H, s, Me), 2.60 (2H, t, *J* 7.6, $CH_2C{=}N$), 2.73 (2H, td, J_t 7.6, J_d 2.4, $CH_2C{=}C$) and 7.04-7.11 (4H, m, HC=C + 3 x arH); δ_C 19.59, 19.84, 22.49, 27.06, 31.48 (2 x Me and 3 x CH_2), 123.29, 127.72, 129.63, 130.71, 134.69, 135.46, 136.14, 136.46 (6 x arC + C=C) and 163.87 (C=N).

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2-(3-Methyl-4-methoxylbenzylidene)cyclopentanone oxime O6 (92%, from **K6**), mp 136-138 ^{o}C (Found: C, 72.6; H, 7.1; N, 6.0. $C_{14}H_{17}NO_2$ requires C, 72.7; H, 7.4; N, 6.1%); δ_H 1.81 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.15 (3H, s, Me), 2.59 (2H, t, *J* 7.5, CH_2C=N), 2.72 (2H, td, J_t 7.5, J_d 2.6, CH_2C=C), 3.78 (3H, s, OMe), 6.75 (1H, d, *J* 8.4), 7.15 (1H, br s) and 7.17 (dd, *J* 8.4, 2.0) (3 x arH), 7.04 (1H, t, *J* 2.6, HC=C); δ_C 16.33 (Me), 22.52, 26.86, 31.36 (3 x CH_2), 55.33 (OMe) 109.70, 122.94, 126.48, 128.18, 129.38, 131.75, 134.07, 157.22 (6 x arC + C=C) and 164.11 (C=N).

2-(3-*t*-Butylbenzylidene)cyclopentanone oxime O7 (62%, from **K7**), mp 109-111 ^{o}C (Found: C, 78.9; H, 8.8; N, 5.5. $C_{16}H_{21}NO$ requires C, 79.0; H, 8.7; N, 5.8%); δ_H 1.16 (9H, s, CMe_3), 1.71 (2H, qn, *J* 7.3, $CH_2CH_2CH_2$), 2.51 (2H, t, *J* 7.3, CH_2C=N), 2.65 (2H, td, J_t 7.3, J_d 2.3, CH_2C=C), 7.05-7.14 (4H, m, 3 x arH + HC=C) and 7.26 (1H, s, arH); δ_C 22.54, 27.11, 31.28, 31.55, 34.62 (CH_2, *C*Me_3 and C*Me*$_3$), 123.78, 124.54, 126.28, 126.61, 128.04, 136.23, 136.68, 151.06 (6 x arC + C=C) and 163.78 (C=N).

2-(2-Methoxybenzylidene)cyclopentanone oxime O8 (70%, from **K8**), mp 130-132 °C (decomp.) (Found: C, 72.1; H, 7.1; N, 6.3. $C_{13}H_{15}NO$ requires: C, 71.9; H, 7.0; N, 6.45%); δ_H 1.83 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.66 (2H, t, *J* 7.5, CH_2C=N), 2.72 (2H, td, J_t 7.5, J_d 2.4, CH_2C=C), 3.83 (3H, s, OMe), 6.87 (1H, d, *J* 8.3), 6.94 (1H, t, *J* 8.3), 7.25 (1H, t, *J* 7.9) and 7.35 (1H, d, *J* 7.9) (4 x arH), 7.44 (1H, t, *J* 2.4, HC=C) and 9.15 (1H, br s, OH); δ_C 22.44, 27.11, 31.48 (3 x CH_2), 55.39 (OMe), 110.28, 117.96, 119.99, 126.09, 128.81, 129.44, 136.68, 157.55 (6 x arC + C=C) and 163.47 (C=N).

2-(3-Methoxybenzylidene)cyclopentanone oxime O9 (81%, from **K9**), mp 98-100 ^{o}C (Found: C, 71.50; H, 7.0; N, 6.6. $C_{13}H_{15}NO_2$ requires C, 71.9; H, 7.00; N, 6.45%); δ_H 1.87 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.68 (2H, t, *J* 7.4, CH_2C=N), 2.81 (2H, td, J_t 7.4, J_d 2.9, CH_2C=C), 3.82 (3H, s, OMe), 6.81 (1H, m), 6.95 (1H, s), 7.01 (1H, d, *J* 7.3) and 7.26 (1H, t, *J* 7.3) (4 x arH), 7.18 (1H, t, *J* 2.9, HC=C) and 9.5 (1H, broad, OH); δ_C 22.43, 27.09, 31.51 (3 x CH_2), 55.15 (OMe), 113.02, 114.63, 121.91, 123.13, 129.23, 136.92, 138.38, 159.38 (6 x arC + C=C) and 163.67 (C=N). *Table of Contents*

2-(4-Methoxybenzylidene)cyclopentanone oxime O10 (71%, from**K10**), mp 154-156 °C (Found: C, 72.0; H, 7.0; N, 6.5. $C_{13}H_{15}NO$ requires: C, 71.9; H, 7.0; N, 6.45%); δ_H 1.88 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.67 (2H, t, *J* 7.5, CH_2C=N), 2.78 (2H, td, J_t 7.5, J_d 2.6, CH_2C=C), 3.82 (3H, s, OMe), 6.89 (2H, d, *J* 7.9) and 7.36 (2H, d, *J* 7.9) (4 x arH), 7.14 (1H, t, *J* 2.6,

HC=C), and 8.45 (1H, br s, OH); δ_C 22.48, 27.03, 31.35 (3 x CH_2), 55.26 (OMe), 113.82, 122.79, 129.88, 130.69, 134.36, 158.91 (4 x arC + C=C) and 163.95 (C=N).

2-(2,5-Dimethoxybenzylidene)cyclopentanone oxime O11 (84%, from **K11**), mp 119-121 °C (Found: C, 67.8; H, 7.0; N, 5.6. $C_{14}H_{17}NO_3$ requires C, 68.0; H, 6.9; N, 5.7%); δ_H 1.85 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.66 (2H, t, 7.5, CH_2C=N), 2.76 (2H, td, J_t 7.5, J_d 2.5, CH_2C=C), 3.79 (3H, s, OMe), 3.81 (3H, s, OMe), 6.80 (2H, m) and 6.94 (1H, d, *J* 2.0) (3 x arH), 7.42 (1H, t, *J* 2.5, HC=C) and 8.03 (1H, s, NOH); δ_C 22.49, 27.01, 31.48 (3 x CH_2), 55.74, 56.04 (OMe), 111.30, 113.19, 115.41, 117.74, 126.97, 137.10, 152.12, 152.99 (6 x arC + C=C) and 163.61 (C=N).

2-(3,4-Dimethoxybenzylidene)cyclopentanone oxime O12 (79%, from **K12**), mp 151-153 °C; δ_H[38a] 1.82 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.60 (2H, t, *J* 7.5, CH_2C=N), 2.74 (2H, td, J_t =7.5, J_d 2.4, CH_2C=C), 3.82 (3H, s, OMe), 3.83 (3H, s, OMe), 6.80 (1H, d, *J* 8.4), 6.88 (1H, d, *J* 1.8), and 6.94 (1H, dd, *J* 8.4, 1.8) (3 x arH), 7.07 (1H, t, *J* 2.4, HC=C); δ_C 22.51, 27.30, 31.38 (3 x CH_2), 55.82, 55.88 (OMe), 110.99, 112.40, 122.33, 123.07, 130.17, 134.64, 148.59, 148.63 (6 x arC + C=C) and 163.91 (C=N).

2-(3-Methoxy-4-methylbenzylidene)cyclopentanone oxime O13 (95%, from **K13**), mp 147-149 °C (Found: C, 72.6; H, 7.5; N, 6.0. $C_{14}H_{17}NO_2$ requires C, 72.7; H, 7.4; N, 6.1%); δ_H[38a] 1.91 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.25 (3H, s, Me), 2.71 (2H, t, *J* 7.5, CH_2C=N), 2.85 (2H, td, J_t 7.5, J_d 2.6, CH_2C=C), 3.86 (3H, s, OMe), 6.89 (1H, br s), 6.97 (1H, dd, *J* 7.6, 0.8) and 7.14 (1H, d, *J* 7.6) (3 x arH), 7.20 (1H, t, *J* 2.6, HC=C); δ_C 16.11 (Me), 22.50, 27.07, 31.53 (3 x CH_2), 55.20 (OMe), 110.91, 121.38, 123.48, 126.34, 130.42, 135.72, 135.90, 157.48 (6 x arC + C=C) and 163.81 (C=N).

2-(3-Hydroxybenzylidene)cyclopentanone oxime O14 (71%, from **K14**), mp 130-131 °C (Found: C, 70.9; H, 6.6; N, 6.7. $C_{12}H_{13}NO_2$ requires C, 70.9; H, 6.45; N, 6.9%); δ_H 1.83 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.63 (2H, t, *J* 7.4, CH_2C=N), 2.76 (2H, td, J_t 7.4, J_d 2.6, CH_2C=C), 6.73 (1H, dd, *J* 8.0, 2.2), 6.85 (1H, t, *J* 2.0), 6.94 (1H, d, *J* 8.0) and 7.19 (1H, t, *J* 8.0) (4 x arH), 7.09 (1H, t, *J* 2.6, HC=C), 9.21[38] (1H, s, OH) and 10.74 (1H, s, OH); δ_C 22.44, 27.03, 31.47 (3 x CH_2), 114.68, 116.00, 121.94, 123.06, 129.52, 137.00, 138.55, 155.52 (6 x arC + C=C) and 164.14 (C=N).

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2-(4-Hydroxybenzylidene)cyclopentanone oxime O15 (66%, from **K15**), mp 179-181 °C (decomp.) (Found: C, 70.90; H, 6.64; N, 6.53. $C_{12}H_{13}NO_2$ requires C, 70.92; H, 6.45; N, 6.89%); δ_H $(CD_3)_2SO$ 1.76 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.44 (2H, t, *J* 7.5, $CH_2C=N$), 2.67 (2H, td, J_t 7.5, J_d 2.4, $CH_2C=C$), 6.77 (2H, d, *J* 8.6, 2 x arH) and 7.26 (2H, d, *J* 8.6) (4 x arH), 6.96 (1H, t, *J* 2.4, HC=C), 9.62 and 10.73 (NOH and OH); δ_C CD_3OD 23.55, 27.88, 32.36 (3 x CH_2), 116.29, 123.53, 130.23, 131.82, 135.04, 158.18 (4 x arC + C=C) and 164.57 (C=N).

2-(3-*N*,*N*-Dimethylaminobenzylidene)cyclopentanone oxime O16 (82%, from **K16**), mp 157-158 °C (Found: C, 72.9; H, 7.95; N, 11.9. $C_{14}H_{18}N_2O$ requires C, 73.0; H, 7.9; N, 12.2%); δ_H 1.79 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.60 (2H, t, *J* 7.6, $CH_2C=N$), 2.75 (2H, td, *J* 7.6, 2.6, $CH_2C=C$), 2.88 (6H, s, NMe_2), 6.61 (1H, dd, *J* 7.8, 2.4), 6.70 (1H, br s), 6.75 (1H, d, *J* 7.6) and 7.16 (1H, t, *J* 7.8) (4 x arH), 7.10 (1H, t, *J* 2.6, HC=C), 8.65 (1H, br s, OH); δ_C 22.53, 27.06, 31.55 (3 x CH_2), 40.69 (NMe_2), 112.13, 113.86, 117.91, 124.11, 128.93, 136.20, 137.72, 150.50 (6 x arC + C=C) and 163.83 (C=N).

2-(4-*N*,*N*-Dimethylaminobenzylidene)cyclopentanone oxime O17 (85%, from **K17**), mp 196-198 °C (decomp.) (Found: C, 72.81; H, 7.92; N, 12.16. $C_{14}H_{18}N_2O$ requires C, 73.01; H, 7.88; N, 12.16%); δ_H 1.80 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.58 (2H, t, 7.6, $CH_2C=N$), 2.72 (2H, td, J_t 7.6, J_d 2.4, $CH_2C=C$), 2.91 (6H, s, NMe_2), 6.63 (2H, d, *J* 9.0) and 7.26 (2H, d, *J* 9.0) (4 x arH), 7.04 (1H, t, *J* 2.4, HC=C) and 8.38 (1H, s, NOH); δ_C 22.33, 27.16, 31.50 (3 x CH_2), 40.44 (NMe_2), 112.12, 123.55, 125.57, 130.75, 132.11, 149.76 (4 x arC + C=C) and 164.44 (C=N).

2-(3-Aminobenzylidene)cyclopentanone oxime O18 (92%, from **K18**), mp 150-151 °C (decomp.) (Found: C, 71.0; H, 6.95; N, 13.6. $C_{12}H_{14}N_2O$ requires C, 71.3; H, 7.0; N, 13.85%); δ_H 2.06 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.64 (2H, t, *J* 7.5, $CH_2C=N$), 2.73 (2H, td, J_t 7.5, J_d 2.8, $CH_2C=C$), 3.60 (2H, broad, NH_2), 6.65 (1H, dd, *J* 8.0, 1.6), 6.72 (1H, t, 1.6), 6.83 (1H, d, *J* 8.0) and 7.14 (1H, t, *J* 8.0) (4 x arH), 7.09 (1H, t, *J* 2.8, HC=C) 8.01 (1H, broad, OH); δ_C 22.51, 26.95, 31.52 (3 x CH_2), 114.49, 115.87, 119.97, 123.29, 129.22, 136.57, 138.12, 146.25 (6 x arC + C=C) and 163.90 (C=N). *Table of Contents*

2-(4-Aminobenzylidene)cyclopentanone oxime O19 (80%, from **K19**), mp 206-207 °C (decomp.) (Found: C, 71.0; H, 6.95; N, 13.6. $C_{12}H_{14}N_2O$ requires C, 71.3; H, 7.0; N, 13.85%); λ_{max}/nm 236 (10 800) and 330 (21 900); δ_H 1.80 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.57 (2H, t, *J* 7.5, $CH_2C=N$), 2.70 (2H, td, J_t 7.5, J_d 2.6, $CH_2C=C$), 3.70 (2H, br s, NH_2), 6.60 (2H, d, *J* 8.8)

and 7.18 (2H, d, *J* 8.8) (4 x arH), 7.02 (1H, t, *J* 2.6, HC=C); δ_C $(CD_3)_2SO$ 22.12, 26.70, 30.96 (3 x CH_2), 113.68, 121.40, 124.69, 129.25, 131.50, 148.25 (4 x arC + C=C) and 161.11 (C=N).

2-(1-Naphthylmethylene)cyclopentanone oxime O20 (80%, from **K20**), mp 134-136 °C (decomp.) (Found: C, 80.9; H, 6.4; N, 5.8. $C_{16}H_{15}NO$ requires: C, 81.0; H, 6.4; N, 5.9%); δ_H 1.81 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.70 (2H, td, J_t 7.4, J_d 2.4, CH_2C=C), 2.74 (2H, t, *J* 7.4, CH_2C=N), 7.47 (4H, m), 7.77 (1H, dd, *J* 7.4, 1.5), 7.85 (2H, m), 8.13 (1H, m) (HC=C + 7 x arH) and 9.70 (1H, br s, OH); δ_C 22.46, 27.58, 31.69 (3 x CH_2), 120.52, 124.52, 125.13, 125.85, 126.11, 126.49, 128.01, 128.43, 131.97, 133.48, 134.03, 138.19 (10 x arC + C=C) and 163.44 (C=N).

2-(2-Naphthylmethylene)cyclopentanone oxime O21 (75%, from **K21**), mp 196-198 °C (Found: C, 80.8; H, 6.3; N, 5.65. $C_{16}H_{15}NO$ requires: C, 81.0; H, 6.4; N, 5.9%); δ_H $(CD_3)_2SO$ 1.88 (2H, qn, *J* 7.3, $CH_2CH_2CH_2$), 2.60 (2H, t, *J* 7.3, CH_2C=N), 2.93 (2H, td, J_t 7.3, J_d 2.3, CH_2C=C), 7.33 (1H, s, arH), 7.58 (2H, m, HC=C + arH), 7.67 (1H, d) and 8.01 (4H, m) (5 x arH), 11.20 (1H, broad, OH); δ_C $(CD_3)_2SO$ 22.19, 26.78, 31.27 (3 x CH_2), 120.57, 126.18, 126.39, 127.07, 127.50, 127.84, 127.88, 128.06, 131.98, 133.11, 134.68, 138.32 (2 x C=C + 10 x arC) and 160.82 (C=N).

2-(10*H*-Phenothiazin-1-ylmethylene)cyclopentanone oxime O22 (93%, from **K22**), mp 129-130 °C (decomp.) (Found: C, 70.3; H, 5.6; N, 9.4. $C_{18}H_{16}N_2OS$ requires C, 70.1; H, 5.2; N, 9.18 %); δ_H 1.80 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.61 (2H, dd, *J* 7.4, 2.4, CH_2C=C), 2.69 (2H, t, *J* 7.4, CH_2C=N), 6.21 (1H, s, NH), 6.62 (1H, dd, *J* 7.8, 1.0), 6.82 (2H, m) and 6.96 (4H, m) (7 x arH), 7.09 (1H, t, *J* 2.5, HC=C) and 8.31 (1H, s, NOH); δ_C 22.76, 28.02, 31.62 (3 x CH_2), 115.62, 117.41, 118.91, 119.05, 122.02, 122.75, 123.21, 126.66, 126.99, 127.72, 128.03, 140.36, 140.73, 141.87 (12 x arC + C=C) and 163.73 (C=N).

2-(2-Furymethylene)cyclopentanone oxime O23 (75%, from **K23**), mp 116-117 °C (lit., 116-117 °C); δ_H 1.90 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.68 (2H, t, *J* 7.5, CH_2C=N), 2.84 (2H, td, J_t 7.5, J_d 2.5, CH_2C=C), 6.38 (1H, d, *J* 3.4), 6.44 (1H, dd, *J* 3.4, 1.5) and 7.44 (1H, d, *J* 1.5) (3 x furyl-H), 7.02 (2H, t, *J* 2.5, HC=C), , 9.75 (1H, br s, OH); δ_C 21.95, 27.47, 31.32 (3 x CH_2), 110.93, 111.23, 111.76, 134.16, 142.64, 153.16 (4 x furyl-C + C=C) and 163.39 (C=N).

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2-(2-Thienylmethylene)cyclopentanone oxime O24 (85%, from **K24**), mp 122-123 °C (Found: C, 62.2; H, 5.7; N, 7.1; S, 16.5. $C_{10}H_{11}NOS$ requires C, 62.15; H, 5.7; N, 7.25; S, 16.6%); δ_H 1.95 (2H, qn, *J* 7.9, $CH_2CH_2CH_2$), 2.70 (2H, t, *J* 7.9, $CH_2C{=}N$), 2.79 (2H, td, J_t 7.9, J_d 1.7, $CH_2C{=}C$), 7.05 (1H, t, *J* 5.0), 7.13 (1H, d, *J* 5.0), 7.35 (1H, d, *J* 5.0), 7.40 (1H, br s) (3 x thienyl-H and HC=C) and 8.78 (1H, br s, NOH); δ_C 22.13, 27.48, 31.32 (3 x CH_2), 116.35, 126.68, 127.36, 128.84, 134.21, 141.28 (C=C + thienyl) and 163.46 (C=N).

2-Cinnamylidenecyclopentanone oxime O25 (75%, from **K25**), mp 167-168 °C (Found: C, 78.6; H, 7.1; N, 6.6. $C_{14}H_{15}NO$ requires C, 78.8; H, 7.1; N, 6.6%); δ_H 1.82 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.59-2.65 (4H, m, $CH_2C{=}N$ and $CH_2C{=}C$), 6.63 (1H, d, *J* 12.8, HC=C), 6.84-6.90 (2H, m, HC=C), 7.13-7.17 (1H, t, *J* 7.6), 7.35 (2H, t, *J* 7.6) and 7.36 (2H, d, *J* 7.6) (2 x arH), 8.29 (1H, s, NOH); δ_C 21.87, 27.94, 29.55 (3 x CH_2), 123.04, 125.73, 126.54, 127.82, 128.62, 135.05, 137.22, 137.33 (4 x arC + 2 x C=C) and 162.77 (C=N).

2-(2-Nitrobenzylidene)cyclopentanone oxime O26 (78%, from **K26**), mp 146-148 °C (decomp.) (Found: C, 61.8; H, 5.3; N, 11.8. $C_{12}H_{12}N_2O_3$ requires: C, 62.1; H, 5.2; N, 12.1%); δ_H 1.85 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.63 (2H, td, J_t 7.5, J_d 2.5, $CH_2C{=}C$), 2.68 (2H, t, *J* 7.5, $CH_2C{=}N$), 7.42 (2H, m, HC=C + arH), 7.49 (1H, dd, *J* 7.6, 1.5), 7.59 (1H, td, J_t 7.6, J_d 1.5), 8.00 (1H, dd, *J* 8.4, 1.5) (3 x arH) and 8.66 (1H, br s, OH); δ_C 22.35, 27.16, 31.17 (3 x CH_2), 118.24, 124.68, 128.00, 130.99, 132.39, 132.65, 140.24, 146.67 (6 x arC + C=C) and 162.51 (C=N).

2-(3-Nitrobenzylidene)cyclopentanone oxime O27 (87%, from **K27**), mp.175-176°C (Found: C, 61.65; H, 5.2; N, 12.0. $C_{12}H_{12}N_2O_3$ requires C, 62.1; H, 5.2; N, 12.1%).

2-(4-Nitrobenzylidene)cyclopentanone oxime O28 (74%, from **K28**), mp 146-148 °C (decomp.) (Found: C, 62.1; H, 5.4; N, 11.9. $C_{12}H_{12}N_2O_3$ requires: C, 62.1; H, 5.2; N, 12.1%); δ_H 1.95 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.71 (2H, t, *J* 7.5, $CH_2C{=}N$), 2.84 (2H, td, J_t 7.5, J_d 2.4, $CH_2C{=}C$), 7.22 (1H, t, *J* 2.4, HC=C), 7.53 (2H, d, *J* 8.4) and 8.22 (2H, d, *J* 8.4) (4 x arH) and 8.86 (1H, br s, OH); δ_C 22.42, 26.95, 31.71 (3 x CH_2), 120.94, 123.72, 129.61, 141.31, 143.51, 146.30 (4 x arC + C=C) and 163.26 (C=N). *Table of Contents*

2-(2-Chlorobenzylidene)cyclopentanone oxime O29 (70%, from **K29**), mp 176-178 °C (decomp.) (Found: C, 65.3; H, 5.4; N, 6.05. $C_{12}H_{12}NOCl$ requires: C, 65.0; H, 5.5; N, 6.3%); δ_H 1.86 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.70 (4H, m, $CH_2C{=}N$ and $CH_2C{=}C$), 7.23 (2H, m) and 7.41 (3H, m) (HC=C + 4 x arH) and 8.35 (1H, br s, OH); δ_C 22.49, 27.09, 31.36 (3 x CH_2),

119.54, 126.33, 128.55, 129.58, 129.87, 134.15, 135.16, 138.92 (6 x arC + C=C) and 163.21 (C=N).

2-(3-Chlorobenzylidene)cyclopentanone oxime O30 (84%, from **K30**), mp 122-123°C (Found: C, 65.0; H, 5.5; N, 6.2; Cl, 15.9. $C_{12}H_{12}NOCl$ requires C, 65.0; H, 5.5; N, 6.3; Cl, 16.0%); δ_H 1.90 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.72 (2H, t, *J* 7.4, CH_2C=N), 2.80 (2H, td, J_t 7.4, J_d 2.5, CH_2C=C), 7.16 (1H, t, *J* 2.5, HC=C), 7.25 (3H, m), 7.39 (1H, s) (4 x arH-2) and 10.05 (1H, broad, OH); δ_C 22.33, 27.08, 31.40 (3 x CH_2), 121.83, 127.22, 127.33, 128.89, 129.45, 134.12, 138.00, 138.80 (6 x arC + C=C) and 163.43 (C=N).

2-(4-Chlorobenzylidene)cyclopentanone oxime O31 (63%, from **K31**), mp 117-120 °C (decomp.) (Found: C, 64.9; H, 5.4; N, 6.1. $C_{12}H_{12}NOCl$ requires: C, 65.0; H, 5.5; N, 6.3%); δ_H 1.89 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.68 (2H, t, *J* 7.5, CH_2C=N), 2.77 (2H, td, J_t 7.5, J_d 2.7, CH_2C=C), 7.13 (1H, t, *J* 2.7, HC=C), 7.32 (4H, br s, 4 x arH) and 9.45 (1H, br s, OH); δ_C 22.45, 27.10, 31.45 (3 x CH_2), 122.05, 128.55, 130.46, 133.11, 135.49, 137.22 (4 x arC + C=C) and 163.62 (C=N).

2-(3-Cyanobenzylidene)cyclopentanone oxime O32 (73%, from **K32**), mp 134-136°C (decomp.) (Found: C, 73.45; H, 5.8; N, 13.1. $C_{13}H_{12}N_2O$ requires C, 73.6; H, 5.7, N, 13.2%); δ_H 1.90 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.66 (2H, t, *J* 7.4, CH_2C=N), 2.78 (2H, td, J_t 7.4, J_d 2.5, CH_2C=C), 7.12 (1H, t, *J* 2.5, HC=C), 7.5 (4H, m, 4 x arH) and 9.8 (1H, broad, OH); δ_C 22.30, 27.00, 31.37 (3 x CH_2), 118.60 (C≡N), 112.42, 120.73, 129.11, 130.42, 132.22, 133.21, 138.11, 139.33 (6 x arC + C=C) and 163.00 (C=N).

2-(4-Cyanobenzylidene)cyclopentanone oxime O33 (80%, from **K33**), mp 130-133 °C (decomp.) (Found: C, 73.1; H, 5.7; N, 13.0. $C_{13}H_{12}N_2O$ requires C, 73.6; H, 5.7; N,13.2%); δ_H: 1.88 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.66 (2H, t, *J* 7.4, CH_2C=N), 2.78 (2H, td, J_t 7.4, J_d 2.5, CH_2C=C), 7.14 (1H, t, *J* 2.5, HC=C), 7.42 (2H, d, *J* 8.4) and 7.60 (2H, d, *J* 8.4) (4 x arH) and 9.9 (1H, br s, NOH); δ_C 22.28, 26.99, 31.55 (3 x CH_2), 118.77 (cyano), 110.16, 121.30, 129.43, 132.01, 140.38, 141.44 (C=C + 4 x arC) and 163.22 (C=N). *Table of Contents*

2-(2,4-Difluorobenzylidene)cyclopentanone oxime O34 (76%, from **K34**), mp 155-158 °C (decomp.) (Found: C, 64.4; H, 5.0; N, 6.1; F, 16.6. $C_{12}H_{11}F_2NO$ requires C, 64.6; H, 5.0; N, 6.3; F, 17.0%); δ_H 1.88 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.70 (4H, m, CH_2C=N and CH_2C=C), 6.82 (2H, m), 7.22 (1H, s), 7.38 (1H, m) and 8.70 (1H, br s, NOH); δ_C 22.26, 26.93, 31.31 (3 x CH_2), 103.80 (t, *J* 26), 110.90 (dd, *J* 18, 3.0), 121.25 (t, *J* 4.6), 130.35 ("q", *J* 4.6) (4 x arC),

114.04 (d, *J* 4.6, $CH_2C=CH$), 138.44 ($CH=CCH_2$), 159.31 (d, *J* 252) and 159.43 (d, *J* 252) (2 x arCF) and 163.02 C=N).

2-(3-Fluorobenzylidene)cyclopentanone oxime O35 (80%, from **K35**), mp 110-112°C (Found: C, 70.2; H, 5.9; N, 6.8; F, 9.55. $C_{12}H_{12}NOF$ requires C, 70.2; H, 5.9; N, 6.8; F, 9.3%); δ_H 1.91 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.70 (2H, t, *J* 7.4, $CH_2C=N$), 2.81 (2H, td, J_t 7.4, J_d 2.5, $CH_2C=C$), 7.20 (m, 5H, 4 x arH + HC=C) and 9.8 (1H, broad, OH); δ_C 22.31, 27.03, 31.37 (3 x CH_2), 114.10 (d, *J* 21), 115.50 (d, *J* 23), 125.10 (d, *J* 3.0), 129.60 (d, *J* 9.1) and 139.20 (5 x arC), 122.10 (d, *J* 3.0, *C*H=C), 137.83 ($CH_2C=C$), 161.36 (d, *J* 244, arC-F) and 163.51 (C=N).

General procedure for preparation of benzylidenecyclopentanone *O*-methyl oximes

Solutions of dimethyl sulphate (5 cm^3) in acetone (20 cm^3) and of aqueous sodium hydroxide (40% w/v, 12 cm^3) were added simultaneously and dropwise with stirring to the benzylidenecyclopentanone oxime (2.0g.) in acetone (50 cm^3) and the resulting solution was heated under reflux until TLC (light petroleum / ethyl acetate 90:10) showed that all the oxime had reacted (~1 hour). Further sodium hydroxide solution (10 cm^3) was added and the acetone removed. Water (50 cm^3) was then added and the mixture extracted with diethyl ether (50 cm^3). The extract was washed with dilute hydrochloric acid (2x50 cm^3) and water (2x50 cm^3), dried ($MgSO_4$) and the ether removed. Light petroleum/ethyl acetate (90:10, 100 cm^3) was added and the solution passed through a short plug of silica gel, concentrated and recrystallised from methanol, unless otherwise stated, to yield the required 2-benzylidene cyclopentanone *O*-methyloxime.

2-Benzylidenecyclopentanone *O*-methyloxime MO1 (67%, from **O1**), mp 78-79 °C (light petroleum) (Found: C, 77.3; H, 7.6; N, 6.7. $C_{13}H_{15}NO$ requires: C, 77.6; H, 7.5; N, 7.0%); λ_{max}/nm 226 (11 600) and 302 (24 200); δ_H 1.85 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.58 (2H, t, *J* 7.5, $CH_2C=N$), 2.78 (2H, td, J_t 7.5, J_d 2.6, $CH_2C=C$), 3.97 (3H, s, OMe), 7.24 (2H, m, HC=C + arH), 7.35 (2H, t, *J* 7.9) and 7.41 (2H, d, *J* 7.9) (4 x arH); δ_C 22.61, 27.43, 31.35 (3 x CH_2), 62.03 (OMe), 122.80, 127.26, 128.31, 129.23, 136.79, 137.18 (4 x arC + C=C) and 162.50 (C=N).

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2-Diphenylmethylenecyclopentanone *O*-methyloxime MO1a (38%, from **O1a**; bp 55-60 °C (1.0mbar) (Found: C, 82.1; H, 7.0; N, 5.2. $C_{19}H_{19}NO$ requires: C, 82.3; H, 6.9; N, 5.05%); λ_{max}/nm 235 (8 500) and 298 (7 200); δ_H 1.74 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.54 (2H, t, *J* 7.4), 2.59 (2H, t, *J* 7.4) (CH_2C=N and CH_2C=C), 3.50 (3H, s, OMe), and 7.17-7.31 (10H, m, 10 x arH); δ_C 22.24, 27.91, 34.05 (3 x CH_2), 61.59 (OMe), 126.58, 127.03, 127.69, 127.74, 129.54, 129.64, 134.18, 139.44, 142.51, 142.84 (8 x arC + C=C) and 160.79 (C=N).

2-(2-Methylbenzylidene)cyclopentanone *O*-methyloxime MO2 (67%, from **O2**), mp 45-47 °C (Found: C, 77.75; H, 7.9; N, 6.2. $C_{14}H_{17}NO$ requires: C, 78.1; H, 8.0; N, 6.5%); λ_{max}/nm 226 (8 500) and 299 (18 300); δ_H 1.81 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.41 (3H, s, Me), 2.61 (2H, t, *J* 7.6, CH_2C=N), 2.69 (2H, td, J_t 7.6, J_d 2.3, CH_2C=C), 4.02 (3H, s, OMe), 7.21 (3H, m, 3 x arH) and 7.35 (2H, m, HC=C + arH); δ_C 20.02 (Me), 22.56, 27.55, 31.25 (3 x CH_2), 61.93 (OMe), 120.97, 125.39, 127.34, 128.29, 129.94, 136.04, 137.24, 137.26 (6 x arC + C=C) and 162.10 (C=N).

2-(4-Methylbenzylidene)cyclopentanone *O*-methyloxime MO4 (63%, from **O4**), mp 63-65 °C (Found: C, 77.9; H, 8.0; N, 6.3. $C_{14}H_{17}NO$ requires: C, 78.1; H, 7.9; N, 6.2%); λ_{max}/nm 230 (5 100) and 306 (24 900); δ_H 1.86 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.37 (3H, s, Me), 2.59 (2H, t, *J* 7.5, CH_2C=N), 2.79 (2H, td, J_t 7.5, J_d 2.6, CH_2C=C), 4.00 (3H, s, OMe), 7.19 (2H, d, *J* 7.8) and 7.33 (2H, d, *J* 7.8) (4 x arH), 7.23 (1H, t, *J* 2.6, HC=C); δ_C 21.23 (Me), 22.58, 27.41, 31.31 (3 x CH_2), 61.94 (OMe), 122.73, 129.03, 129.19, 134.33, 135.73, 137.18 (4 x arC + C=C) and 162.59 (C=N).

2-(2-Methoxybenzylidene)cyclopentanone *O*-methyloxime MO8 (65%, from **O8**), mp 62-64 °C (Found: C, 72.4; H, 7.4; N, 5.8. $C_{14}H_{17}NO$ requires: C, 72.7; H, 7.4; N, 6.1); λ_{max}/nm 230 (12 300) and 318 (17 000); δ_H 1.80 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.57 (2H, t, *J* 7.5, CH_2C=N), 2.71 (2H, td, J_t 7.5, J_d 2.4, CH_2C=C), 4.15 (3H, s, OMe), 4.27 (3H, s, OMe), 6.88 (1H, d, *J* 8.1), 6.94 (1H, t, *J* 7.4), 7.25 (1H, td, J_t 8.1, J_d 1.5) and 7.35 (1H, dd, *J* 7.4, 1.5) (4 x arH), 7.48 (1H, t, *J* 2.4, HC=C); δ_C 22.54, 27.46, 31.38 (3 x CH_2), 55.35 and 61.95 (OMe), 110.23, 117.59, 119.97, 126.18, 128.68, 129.41, 136.85, 157.53 (6 x arC + C=C) and 162.29 (C=N).

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2-(3-Methoxybenzylidene)cyclopentanone *O*-methyloxime MO9 (64%, from **O9;** an oil, purified by reduced pressure condensation onto a cold finger) (Found: C, 73.1; H, 7.5; N, 6.1. $C_{14}H_{17}NO$ requires C, 72.7; H, 7.4; N, 6.1%); λ_{max}/nm 224 (10 100) and 302 (18 000); δ_H 1.81

(2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.56 (2H, t, *J* 7.4, $CH_2C{=}N$), 2.76 (2H, t, *J* 7.4, $CH_2C{=}C$), 3.75 (3H, s, OMe), 3.95 (3H, s, OMe), 6.79 (1H, m), 6.93 (1H, s), 7.01 (1H, d, *J* 2.0) and 7.25 (1H, t, *J* 2.0) (4 x arH), 7.19 (1H, s, HC=C); δ_C 22.67, 27.50, 31.46 (3 x CH_2), 55.21 ($COCH_3$), 62.07 ($NOCH_3$), 112.98, 114.71, 121.93, 122.78, 129.31, 137.15, 138.62, 159.51 (6 x arC + C=C) and 162.45 (C=N).

2-(4-Methoxybenzylidene)cyclopentanone *O*-methyloxime MO10 (59%, from **O10**), mp 93-94 °C (Found: C, 72.4; H, 7.4; N, 5.9. $C_{14}H_{17}NO$ requires: C, 72.7;H, 7.4; N, 6.1%); λ_{max}/nm 224 (11 000) and 314 (27 000); δ_H 1.85 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.57 (2H, t, *J* 7.5, $CH_2C{=}N$), 2.75 (2H, td, J_t 7.5, J_d 2.0, $CH_2C{=}C$), 3.84 (3H, s, OMe), 3.99 (3H, s, OMe), 6.90 (2H, d, *J* 8.5) and 7.38 (2H, d, *J* 8.5) (4 x arH), 7.19 (1H, t, *J* 2.0, HC=C); δ_C 22.68, 27.50, 31.30 (3 x CH_2), 55.31 and 62.00 (OMe), 113.86, 122.49, 130.06, 130.71, 134.52, 158.89 (4 x arC + C=C) and 162.82 (C=N).

2-(1-Naphthylmethylene)cyclopentanone *O*-methyloxime MO20 (69%, from **O20**), mp 109-111 °C (Found: C, 81.5; H, 6.9; N, 5.1. $C_{17}H_{17}NO$ requires: C, 81.2; H, 6.8; N, 5.6%); λ_{max}/nm 326 (16 600) and 230 (26 400); δ_H 1.77 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.64 (4H, m, $CH_2C{=}N$, and $CH_2C{=}C$), 4.02 (3H, s, OMe), 7.48 (4H, m, HC=C + 3 x arH), 7.77 (1H, m), 7.84 (2H, m), and 8.12 (1H, m) (4 x arH); δ_C 22.57, 27.75, 31.54 (3 x CH_2), 62.06 (OMe), 119.98, 124.52, 125.14, 125.81, 126.00, 126.49, 127.88, 128.44, 131.93, 133.47, 134.16, 138.89 (10 x arC + C=C) and 161.91 (C=N).

2-(2-Naphthylmethylene)cyclopentanone *O*-methyloxime MO21 (58%, from **O21**), mp 113-114°C (Found: C, 81.2; H, 6.7; N, 5.5. $C_{17}H_{17}NO$ requires C, 81.2; H, 6.8; N, 5.6%); λ_{max}/nm 224 (21 900), 282 (22 800) and 322 (27 300); δ_H 1.90 (2H, qn, *J* 7.9, $CH_2CH_2CH_2$), 2.63 (2H, t, *J* 7.9, $CH_2C{=}N$), 2.92 (td, J_t 7.9, J_d 2.9, $CH_2C{=}C$), 4.03 (3H, s, OMe), 7.42 (1H, t, *J* 2.9, HC=C), 7.49 (2H, m), 7.59 (1H, m) and 7.85 (4H, m) (7 x arH); δ_C 22.62, 27.42, 31.46 (3 x CH_2), 62.03 (OMe), 122.85, 126.07, 126.16, 127.12, 127.52, 127.77, 128.09, 128.45, 132.42, 133.30, 134.74, 137.15 (10 x arC + C=C) and 162.47 (C=N). *Table of Contents*

2-(2-Furylmethylene)cyclopentanone *O*-methyloxime MO23 (42%, from **O23**), bp 62-66 °C (0.5 mm Hg) (Found: C, 69.2; H, 6.8; N, 7.3. $C_{11}H_{13}NO_2$ requires: C, 69.1; H, 6.85; N, 7.3%); λ_{max}/nm 224 (5 500) and 320 (30 600); δ_H 1.84 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.55 (2H, t, *J* 7.5, $CH_2C{=}N$), 2.79 (2H, td, J_t 7.5, J_d 2.5, $CH_2C{=}C$), 3.88 (3H, s, OMe), 6.36 (1H, m), 6.41 (1H, m) and 7.42 (1H, m) (3 x furyl-H), 7.02 (1H, t, *J* 2.5, HC=C); δ_C 22.07, 27.62, 31.02 (3 x

CH_2), 61.86 (OMe), 110.28, 110.86, 111.63, 134.29, 142.46, 153.24 (4 x furyl + C=C) and 161.82 (C=N).

2-(2-Nitrobenzylidene)cyclopentanone *O*-methyloxime MO26 (62%, from **O26**), mp 82-84 °C (Found: C, 63.1; H, 5.7; N, 11.1. $C_{13}H_{14}N_2O_3$ requires: C, 63.4; H, 5.7; N, 11.4%); λ_{max}/nm 220 (5 900) and 272 (13 700); δ_H 1.82 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.59 (4H, m, $CH_2C{=}N$ and $CH_2C{=}C$), 3.98 (3H, s, OMe), 7.39, (1H, td, J_t 8.4, J_d 1.5), 7.46 (1H, dd, *J* 7.4, 1.5), 7.58 (1H, td, J_t 8.4, J_d 1.5) and 7.97 (1H, dd, J_t 8.4, J_d 1.5) (4 x arH), 7.43 (1H, t, *J* 2.6, HC=C); δ_C 22.46, 27.50, 31.12 (3 x CH_2), 62.19 (OMe), 117.83, 124.59, 127.88, 130.95, 132.40, 132.67, 140.41, 148.41 (6 x arC + C=C) and 160.96 (C=N).

2-(4-Nitrobenzylidene)cyclopentanone *O*-methyloxime MO28 (58%, from **O28)**, mp 122-124 °C (Found: C, 63.2; H, 5.7; N, 11.1. $C_{13}H_{14}N_2O_3$ requires: C, 63.4; H, 5.7; N, 11.4%); λ_{max}/nm 268 (8 800) and 346 (20 200); δ_H 1.90 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.59 (2H, t, *J* 7.5, $CH_2C{=}N$), 2.80 (2H, td, J_t 7.5, J_d 2.5, $CH_2C{=}C$), 3.99 (3H, s, OMe), 7.26 (1H, t, *J* 2.5, HC=C), 7.53 (2H, d, *J* 8.4) and 8.19 (2H, d, *J* 8.4) (4 x arH); δ_C 22.51, 27.27, 31.57 (3 x CH_2), 62.26 (OMe), 120.45, 123.65, 129.49, 141.48, 143.72, 146.17 (4 x arC + C=C) and 161.61 (C=N).

2-(2-Chlorobenzylidene)cyclopentanone *O*-methyloxime MO29 (36%, from **O29**), bp 75-79 °C (1mmHg) (Found: C, 66.1; H, 5.95; N, 5.8. $C_{13}H_{14}ClNO$ requires: C, 66.2; H, 6.0; N, 5.9%); λ_{max}/nm 230 (9 700) and 298 (16 100); δ_H 1.82 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.58 (2H, t, *J* 7.6, $CH_2C{=}N$), 2.67 (2H, td, J_t 7.6, J_d 2.5, $CH_2C{=}C$), 3.95 (3H, s, OMe), 7.21 (2H, m) and 7.40 (3H, m) (HC=C + 4 x arH); δ_C 22.58, 27.47, 31.29 (3 x CH_2), 62.17 (OMe), 119.34, 126.29, 128.44, 129.54, 129.94, 134.38, 135.32, 139.12 (6 x arC + C=C) and 161.73 (C=N).

2-(4-Chlorobenzylidene)cyclopentanone *O*-methyloxime MO31 (55%, from **O31**), mp 50-51 °C (Found: C, 66.2; H, 6.0; N, 5.8. $C_{13}H_{14}ClNO$ requires: C, 66.2; H, 6.00; N, 5.9%); λ_{max}/nm 228 (9 600) and 306 (28 100); δ_H 1.85 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.57 (2H, t, *J* 7.5, $CH_2C{=}N$), 2.73 (2H, td, J_t 7.5, J_d 2.4, $CH_2C{=}C$), 3.97 (3H, s, OMe), 7.16 (1H, t, *J* 2.4, HC=C) and 7.31 (4H, m, 4 x arH); δ_C 22.64, 27.44, 31.36 (3 x CH_2), 62.14 (OMe), 121.56, 128.57, 130.44, 132.98, 135.70, 137.47 (4 x arC + C=C) and 162.27 (C=N).

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2-Benzylidenecyclohexanone *O*-methyloxime MO36 (46%, from **K36**; oil solidified following short path distillation under reduced pressure), mp 23.5-25.5 °C (Found: C, 78.3; H,

7.9; N, 6.75. $C_{14}H_{17}NO$ requires: C, 78.1; H, 8.0; N, 6.5%); λ_{max}/nm 276 (15 900); δ_H 1.63 (2H, m, $CH_2CH_2CH_2$), 1.69 (2H, m, $CH_2CH_2CH_2$), 2.59 (2H, t, *J* 6.6) and 2.66 (2H, t, *J* 6.2) ($CH_2C{=}C$ and $CH_2C{=}N$), 3.94 (3H, s, OMe), 6.92 (1H, s), 7.24 (1H, m) and 7.32 (4H, m) (5 x arH + HC=C); δ_C 23.39, 24.91, 25.62, 28.96 (4 x CH_2), 61.63 (OMe), 126.99, 127.43, 127.99, 129.69, 134.80, 136.86 (4 x arC + C=C) and 159.77 (C=N).

General procedure for preparation of the benzylidenecyclopentanone *O*-acetyloximes

Acetyl chloride (2.1 g, 20 mmol) was added dropwise to a solution of the benzylidenecyclopentanone oxime (15 mmol) in pyridine (25 cm^3) at 5 °C with vigorous stirring. After a further hour at room temperature crushed ice (50 g) was added, the benzylidenecyclopentanone *O*-acetyloxime suspension was filtered off, dissolved in chloroform (50 cm^3), washed with dilute hydrochloric acid and dried ($MgSO_4$). Rotary evaporation and recrystallisation from methanol, unless otherwise stated, yielded the corresponding *O*-acetyloxime.

2-Benzylidenecyclopentanone *O*-acetyloxime AO1 (90%, from **O1**), mp 114-115 °C (light petroleum) (Found: C, 73.4; H, 6.65; N, 6.1. $C_{14}H_{15}NO_2$ requires: C, 73.3; H, 6.6; N, 6.1%); λ_{max}/nm 302 (22 700); δ_H 1.82 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.15 (s, 3H, MeCO), 2.66 (2H, t, *J* 7.4, $CH_2C{=}N$), 2.77 (2H, td, J_t 7.4, J_d 2.5, $CH_2C{=}C$), 7.22 (1H, m), 7.30 (2H, m) and 7.37 (2H, m) (5 x arH), 7.47 (1H, t, *J* 2.5, HC=C); δ_C 19.60 (Me), 22.33, 28.92, 31.18 (3 x CH_2), 127.22, 128.13, 128.46, 129.66, 134.80, 136.42 (C=C + 4 x arC), 168.99 and 170.20 (C=N and C=O)

2-(3-Methylbenzylidene)cyclopentanone *O*-acetyloxime AO3 (75%, from **O1**), mp 78-79°C (Found: C, 74.0; H, 7.0; N, 5.7; $C_{15}H_{17}NO_2$ requires C, 74.05; H, 7.0; N, 5.8%); λ_{max}/nm 282 (22 200); δ_H 1.88 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.20 (3H, s, MeAr), 2.37 (3H, s, MeCO), 2.72 (2H, t, *J* 7.4, $CH_2C{=}N$), 2.83 (2H, t, *J* 7.4, $CH_2C{=}C$), 7.11 (1H, d, *J* 5.6, arH), 7.25 (3H, m, 2 x arH + HC=C) and 7.50 (1H, d, *J* 3.7, arH); δ_C 19.61 and 21.34 (2 x Me), 22.32, 28.92, 31.21 (3 x CH_2), 126.65, 127.20, 128.22, 128.85, 130.26, 134.45, 136.24, 137.87 (6 x arC + C=C), 168.84 and 170.07 (C=N and C=O).

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2-(3,4-Dimethylbenzylidene)cyclopentanone *O*-acetyloxime AO5 (89%, from **O5**), mp 99-101 °C (Found: C, 74.65; H, 7.5; N, 5.4. $C_{16}H_{19}NO_2$ requires C, 74.7; H, 7.4; N, 5.4%); λ_{max}/nm 234 (6 350) and 312 (23 400); δ_H 1.91 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.24 (3H, s) and 2.30 (6H, s) (3 x Me), 2.74 (2H, t, *J* 7.6, CH_2C=N), 2.86 (2H, td, J_t 7.6, J_d 2.4, CH_2C=C), 7.15-7.25 (3H, m, 3 x arH) and 7.51 (1H, t, *J* 2.4, HC=C); δ_C 19.63, 19.72, 19.85 (MeC=O and Me), 22.40, 29.02, 31.28 (3 x CH_2), 127.16, 127.31, 129.72, 130.99, 133.49, 134.02, 136.59, 137.02 (6 x arC + C=C), 168.98 and 170.31 (C=N and C=O).

2-(3-Methyl-4-methoxy-benzylidene)cyclopentanone *O*-acetyloxime AO6 (77%, from **O6**), mp 77-79 °C (decomp.) (Found: C, 70.3; H, 7.0; N, 5.15. $C_{16}H_{19}NO_3$ requires C, 70.3; H, 7.0; N, 5.1%); λ_{max}/nm 238 (9 600) and 326 (26 400); δ_H 1.82 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.14 (3H, s), 2.15 (3H, s) (MeCO and ArMe), 2.64 (2H, t, *J* 7.6, CH_2C=O), 2.75 (2H, td, J_t 7.6, J_d 2.6, CH_2C=C), 3.78 (3H, s, OMe), 6.76 (1H, d, *J* 8.4), 7.18 (1H, s) and 7.21 (1H, dd, *J* 8.4, 1.8) (3 x arH), 7.39 (1H, t, *J* 2.6, HC=C); δ_C 16.32 (Me), 19.72 (MeC=O), 22.40, 29.03, 31.20 (3 x CH_2), 55.31 (OMe) 109.74, 126.61, 127.17, 128.70, 128.91, 131.87, 132.05, 157.80 (6 x arC + C=C), 169.06 and 170.51 (C=N and C=O).

2-(3-*t*-Butylbenzylidene)cyclopentanone *O*-acetyloxime AO7 (68%, from **O7**), yellow oil (Found: C, 75.7; H; 8.35; N, 4.8. $C_{18}H_{23}NO_2$ requires C, 75.8; H, 8.1; N, 4.9%); λ_{max}/nm 224 (32 200) and 304 (47 800); δ_H 1.24 (9H, s, CMe_3), 1.81 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.13 (3H, s, MeCO), 2.64 (2H, t, *J* 7.4, CH_2C=N), 2.76 (2H, td, J_t 7.4, J_d 2.4, CH_2C=C), 7.23 (3H, m) and 7.39 (1H, s) (4 x arH), 7.48 (1H, t, *J* 2.4, HC=C); δ_C 19.77 (*Me*CO), 22.37, 28.95, 31.15, 31.21, 34.52 (3 x CH_2, *C*Me$_3$ and C*Me*$_3$), 125.16, 126.33, 127.13, 127.64, 128.06, 134.15, 135.92, 151.12 (6 x arC + C=C), 168.79 and 170.05 (C=N and C=O).

2-(3-Methoxybenzylidene)cyclopentanone *O*-acetyloxime AO9 (80%, from **O9**), mp 69-70°C (Found: C, 69.7; H, 6.6; N, 5.6. $C_{15}H_{17}NO_3$ requires C, 69.5; H, 6.6, N, 5.4%); λ_{max}/nm 298 (23 150); δ_H 1.91 (2H, qn, *J* 7.8, $CH_2CH_2CH_2$), 2.23 (3H, s, MeCO), 2.72 (2H, t, *J* 7.8, CH_2C=N), 2.85 (2H, td, J_t 7.8, J_d 1.9, CH_2C=C), 3.82 (3H, s, OMe), 6.86 (1H, dd, *J* 8.8, 1.5), 6.98 (1H, br s), 7.05 (1H, d, *J* 7.8) and 7.30 (1H, t, *J* 7.8) (4 x arH) and 7.52 (1H, t, *J* 1.9, HC=C); δ_C 19.75 (MeCO), 22.48, 29.12, 31.42 (3 x CH_2), 55.47 (OMe), 113.78, 114.95, 122.12, 127.04, 129.34, 135.02, 137.67, 159.44 (6 x arC + C=C), 168.86 and 170.02 (C=N and C=O).

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2-(4-Methoxybenzylidene)cyclopentanone *O*-acetyloxime AO10 (83%, from **O10**), mp 88-89 °C (light petroleum) (Found: C, 69.6; H, 6.65; N, 5.4. $C_{15}H_{17}NO_3$ requires C, 69.5; H, 6.6; N, 5.4%); λ_{max}/nm 226 (11 300) and 322 (27 000); δ_H 1.89 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.23 (s, 3H, MeCO), 2.71 (2H, t, *J* 7.4, $CH_2C=N$), 2.82 (2H, td, J_t 7.4, J_d 2, $CH_2C=C$), 3.85 (3H, s, OMe), 6.90 (2H, d, *J* 6.4) and 7.41 (2H, d, *J* 6.4) (4 x arH), 7.50 (1H, br s, HC=C); δ_C 19.63 (Me), 22.33, 28.96, 31.11 (3 x CH_2), 55.23 (OMe), 113.98, 126.93, 129.24, 131.26, 132.29, 159.60 (C=C + 4 x arC), 169.52 and 170.52 (C=N and C=O).

2-(2,5-Dimethoxybenzylidene)cyclopentanone *O*-acetyloxime AO11 (84%, from **O11**), mp 112-113 °C (Found: C, 66.4; H, 6.7; N, 4.7. $C_{16}H_{19}NO_4$ requires C, 66.4 H, 6.6; N, 4.8%); λ_{max}/nm 292 (15 100) and 348 (9 200); δ_H 1.79 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.16 (s, 3H, MeCO), 2.65 (2H, t, *J* 7.4, $CH_2C=N$), 2.70 (2H, td, J_t 7.4, J_d 2.7, $CH_2C=C$), 3.71 (s, 3H, OMe), 3.73 (s, 3H, OMe), 6.76 (2H, m), and 6.85 (1H, d, *J* 1.6) (3 x arH), 7.66 (1H, t, *J* 2.7, HC=C); δ_C 19.76 (Me), 22.36, 29.02, 31.37 (3 x CH_2), 55.74, 55.94 (OMe), 111.43, 114.00, 115.74, 122.27, 126.29, 135.23, 152.44, 152.93 (C=C + 6 x arC), 169.19 and 170.00 (C=N and C=O).

2-(3,4-Dimethoxybenzylidene)cyclopentanone *O*-acetyloxime AO12 (80%, from **O12**), mp 122-123 °C (Found: C, 66.6; H, 6.5; N, 4.6. $C_{16}H_{19}NO_4$ requires C, 66.4; H, 6.6; N, 4.8%); λ_{max}/nm 246 (8 100) and 330 (22 000); δ_H 1.84 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.14 (3H, s MeCO), 2.64 (2H, t, *J* 7.6, $CH_2C=N$), 2.76 (2H, td, J_t 7.6, J_d 2.5, $CH_2C=C$), 3.81 (3H, s, OMe), 3.83 (3H, s, OMe), 6.81 (1H, d, *J* 8.4), 6.92 (1H, d, *J* 1.6) and 6.99 (1H, dd, *J* 8.4, 1.6) (3 x arH), 7.41 (1H, t, *J* 2.5, HC=C); δ_C 19.64 (MeC=O), 22.37, 28.95, 31.11 (3 x CH_2), 55.77, 55.83 (OMe), 111.00, 112.81, 122.81, 127.04, 129.44, 132.45, 148.66, 149.15 (6 x arC + C=C), 168.84 and 170.19 (C=N and C=O).

2-(3-Methoxy-4-methylbenzylidene)cyclopentanone *O*-acetyloxime AO13 (85%, from **O13**), mp 108-109 °C (Found: C, 70.05; H, 7.0; N, 5.1. $C_{16}H_{19}NO_3$ requires C, 70.3; H, 7.0; N, 5.1%); λ_{max}/nm 242 (8 500) and 310 (18 600); δ_H 1.93 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.24 (3H, s) and 2.25 (3H, s) (Me and MeCO), 2.75 (2H, t, *J* 7.5, $CH_2C=N$), 2.87 (2H, td, J_t 7.5, J_d 2.6, $CH_2C=C$), 3.86 (3H, s, OMe), 6.92 (1H, br s), 7.01 (1H, dd, *J* 7.6, 0.8) and 7.15 (1H, d, *J* 7.6) (3 x arH), 7.54 (1H, t, *J* 2.6, HC=C); δ_C 16.14 (Me), 19.73 (MeCO), 22.44, 29.04, 31.29 (3 x CH_2), 55.20 (OMe), 111.43, 121.61, 127.23, 127.51, 130.53, 133.69, 135.24, 157.54 (6 x arC + C=C), 168.91 and 170.20 (C=N and C=O).

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2-(3-Hydroxybenzylidene)cyclopentanone *O*-acetyloxime AO14 (80%, from **O14**), mp 189-190 °C (Found: C, 68.1; H, 6.2; N, 5.6. $C_{14}H_{15}NO_3$ requires C, 68.6; H, 6.2; N, 5.7%); λ_{max}/nm 224 (8 300) and 300 (20 000); δ_H $(CD_3)_2SO$ 1.83 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.18 (s. 3H, Me), 2.67 (2H, t, *J* 7.5, $CH_2C=N$), 2.78 (2H, td, J_t 7.5, J_d 2.7, $CH_2C=C$), 6.76 (1H, dd, *J* 7.8, 2.0), 6.92 (1H, t, *J* 2.0) and 6.94 (1H, d, *J* 8.0) (3 x arH), 7.23 (2H, m, arH + HC=C) and 9.56 (1H, s, OH); δ_C $(CD_3)_2SO$ 19.58 (*Me*CO), 22.04, 28.63, 31.00 (3 x CH_2), 115.52, 115.89, 120.71, 125.77, 129.65, 135.41, 137.14, 157.43 (6 x arC + C=C), 168.30 and 169.70 (C=N and C=O).

2-(4-Hydroxybenzylidene)cyclopentanone *O*-acetyloxime AO15 (80%, from **O15**), mp 153-154 °C (Found: C, 68.3; H, 6.4; N, 5.6. $C_{14}H_{15}NO_3$ requires C, 68.6; H, 6.2; N, 5.7%); λ_{max}/nm 230 (7 600) and 324 (22 800); δ_H 1.89 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.23 (s, 3H, MeCO), 2.71 (2H, t, *J* 7.5, $CH_2C=N$), 2.78 (2H, td, J_t 7.5, J_d 2.5, $CH_2C=C$), 6.27 (1H, br s, OH), 6.88 (2H, d, *J* 8.4) and 7.27 (2H, d, *J* 8.4) (4 x arH), 7.41 (1H, t, *J* 2.5, HC=C); δ_C 19.75 (Me), 22.41, 29.09, 31.15 (3 x CH_2), 115.59, 127.33, 128.84, 131.45, 131.70, 156.27 (C=C + 4 x arC), 169.45 and 170.82 (C=N and C=O).

2-(3-*N*,*N*-Dimethylaminobenzylidene)cyclopentanone *O*-acetyloxime AO16 (82%, from **O16**), mp 96-97 °C (Found: C, 70.8; H, 7.4; N, 10.2. $C_{16}H_{20}N_2O_2$ requires C, 70.6; H, 7.4; N, 10.3%); λ_{max}/nm 280 (19 600) and 302 (21 200); δ_H 1.82 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.15 (3H, s, MeCO), 2.65 (2H, t, *J* 7.6, $CH_2C=N$), 2.79 (2H, td, *J* 7.6, 2.6, $CH_2C=C$), 2.89 (6H, s, NMe_2), 6.63 (1H, dd, *J* 8.0, 2.4), 6.73 (1H, d, *J* 2.4), 6.78 (1H, d, *J* 8.0) and 7.18 (1H, t, *J* 8.0) (4 x arH), 7.45 (1H, t, *J* 2.6, HC=C); δ_C 19.75 (MeCO), 22.47, 29.09, 31.34 (3 x CH_2), 40.55 (NMe_2), 112.59, 114.29, 117.78, 128.28, 129.02, 134.13, 137.02, 150.52 (6 x arC + C=C), 168.97 and 170.28 (C=N and C=O).

2-(4-*N*,*N*-Dimethylaminobenzylidene)cyclopentanone *O*-acetyloxime AO17: (86%, from **O17**), mp 91-92 °C (Found: C, 70.6; H, 7.50; N, 10.2. $C_{16}H_{20}N_2O_2$ requires C, 70.6; H, 7.4; N, 10.3%); λ_{max}/nm 250 (8 800) and 372 (24 600); δ_H 1.88 (2H, m, *J* 7.5, $CH_2CH_2CH_2$), 2.21 (s, 3H, MeCO), 2.70 (2H, t, *J* 7.5, $CH_2C=N$), 2.75 (2H, td, J_t 7.5, J_d 2.4, $CH_2C=C$), 2.99 (s, 6H, NMe_2), 6.71 (d, 2H, *J* 8.8) and 7.37 (d, 2H, *J* 8.8) (4 x arH), 7.46 (1H, t, *J* 2.4, HC=C); δ_C 19.85 (Me), 22.51, 29.20, 31.34 (3 x CH_2), 40.25 (NMe_2), 111.89, 124.61, 127.76, 129.53, 131.31, 150.13 (C=C + 4 x arC), 169.22 and 170.96 (C=N and C=O). *Table of Contents*

2-(2-Naphthylmethylene)cyclopentanone *O*-acetyloxime AO21 (73%, from **O21**), mp 95-96°C (Found: C, 77.35; H, 6.2; N, 5.0%. $C_{18}H_{17}NO_2$ requires C, 77.4; H, 6.1; N, 5.0%); λ_{max}/nm 222 (48 800), 282 (35 700) and 322 (33 500); δ_H 1.92 (2H, qn, *J* 7.9, $CH_2CH_2CH_2$), 2.23 (3H, s, MeCO), 2.73 (2H, t, *J* 7.9, $CH_2C{=}N$), 2.92 (2H, td, J_t 7.9, J_d 2.9, $CH_2C{=}C$), 7.51 (3H, m) and 7.85 (4H, m) (7 x arH), 7.70 (1H, d, *J* 2.9, HC=C); δ_C 19.64 (Me), 22.36, 28.93, 31.31 (3 x CH_2), 126.26, 126.46, 126.96, 127.16, 127.38, 127.90, 128.22, 129.25 (7 x arCH + HC=C), 132.74, 133.12, 133.84, 135.01 (3 x arC + C=C(C_2), 168.86 and 170.05 (C=N and C=O).

2-(10*H*-Phenothiazin-1-ylmethylene)cyclopentanone *O*-acetyloxime AO22 (75%, from **O22**), mp 159-160 °C (Found: C, 68.5; H, 5.3; N, 7.9. $C_{20}H_{18}N_2O_2S$ requires C, 68.55; H, 5.2; N, 8.0%); λ_{max}/nm 252 (18 300), 312 (8 400) and 400 (1 800); δ_H 1.84 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.24 (s, 3H, MeCO), 2.63 (2H, td, J_t 7.4, J_d 2.3, $CH_2C{=}C$), 2.76 (2H, t, *J* 7.4, $CH_2C{=}N$), 6.16 (1H, s, NH), 6.64 (1H, dd, *J* 8.0, 1.2), 6.81 (2H, m) and 6.96 (4H, m) (7 x arH), 7.40 (1H, t, *J* 2.3, HC=C); δ_C 19.63 (Me), 22.18, 29.34, 31.22 (3 x CH_2), 115.15, 118.30, 118.62, 121.02, 121.56, 121.64, 122.89, 126.54, 126.71, 127.33, 127.81, 138.88, 139.91, 141.03 (C=C + 12 x arC), 168.73 and 169.17 (C=N and C=O).

2-(2-Thienylmethylene)cyclopentanone *O*-acetyloxime AO24 (82%, from **O24**), mp 120-121 °C (Found: C, 61.2; H, 5.6; N, 5.9; S, 13.65. $C_{12}H_{13}NOS$ requires C, 61.25; H, 5.6; N, 6.0; S, 13.6%); λ_{max}/nm 324 (25 400); δ_H 1.96 (2H, qn, *J* 7.9, $CH_2CH_2CH_2$), 2.22 (3H, s, MeCO), 2.75 (2H, t, *J* 7.9, $CH_2C{=}N$), 2.80 (2H, td, J_t 7.9, J_d 2.9, $CH_2C{=}C$), 7.08 (1H, t, *J* 4.9), 7.20 (1H, d, *J* 4.9) and 7.41 (1H, d, *J* 4.9) (3 x thienyl-H), 7.75 (1H, t, *J* 2.9, HC=C); δ_C 19.69 (Me), 22.05, 29.47, 31.05 (3 x CH_2), 120.22, 127.56, 127.93, 130.22, 131.99, 140.56 (vinyl + thienyl), 168.84 and 169.72 (C=N and C=O).

2-Cinnamylidenecyclopentanone *O*-acetyloxime AO25 (85%, from **O25**), mp 97-99 °C (Found: C, 75.2; H, 6.9; N, 5.5. $C_{16}H_{17}NO_2$ requires C, 75.3; H, 6.7; N, 5.5%); λ_{max}/nm 234 (9 700) and 334 (37 100); δ_H 1.82 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.14 (s, 3H, MeCO), 2.65 (4H, m, $CH_2C{=}N$ and $CH_2C{=}C$), 6.70 (1H, d, *J* 15.5, HC=C), 6.82 (1H, dd, *J* 15.6, 11.4, HC=C), 7.19 (2H, m, HC=C + arH), 7.26 (2H, t, *J* 7.6) and 7.38 (2H, d, *J* 7.6) (4 x arH); δ_C 19.70 (Me), 21.81, 29.28, 29.84 (3 x CH_2), 125.14, 126.79, 126.97, 128.29, 128.66, 135.28, 136.82, 137.20 (2 x C=C + 4 x arC), 168.93 and 169.09 (C=N and C=O). *Table of Contents*

2-(3-Nitrobenzylidene)cyclopentanone *O*-acetyloxime AO27 (78%, from **O27**), mp119-121°C (Found: C, 61.1; H, 5.2; N, 10.2. $C_{14}H_{14}N_2O_4$ requires C, 61.3; H, 5.1; N, 10.2%); λ_{max}/nm 288 (27 280); δ_H 1.92 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.20 (3H, s, Me), 2.73 (2H, t, *J* 7.4, $CH_2C{=}N$), 2.86 (2H, td, J_t 7.4, J_d 2.4, $CH_2C{=}C$), 7.52 (2H, m), 7.68 (1H, d, *J* 7.9) and 8.10 (1H, m) (4 x arH), 8.25 (1H, t, *J* 2.4, HC=C); δ_C 19.53 (*Me*CO), 22.27, 28.79, 31.10 (3 x CH_2), 122.41, 123.37, 124.24, 129.37, 135.32, 137.83, 138.02, 148.19 (6 x arC + C=C), 168.56 and 169.24 (C=N and C=O).

2-(3-Chlorobenzylidene)cyclopentanone *O*-acetyloxime AO30 (69%, from **O30**), mp 77-78°C (Found: C, 63.8; H, 5.4; N, 5.3; Cl, 13.6. $C_{14}H_{14}NO_2Cl$ requires C, 63.8; H, 5.35; N, 5.3; Cl, 13.4%); λ_{max}/nm 222 (9 700), and 298 (22 600); δ_H 1.87 (2H, qn, *J* 7.9, $CH_2CH_2CH_2$), 2.18 (3H, s, Me), 2.68 (2H, t, *J* 7.9, $CH_2C{=}N$), 2.77 (2H, td, J_t 7.9, J_d 2.9, $CH_2C{=}C$), 7.23 (3H, m) and 7.36 (1H, s) (4 x arH), 7.41 (1H, t, *J* 2.9, HC=C); δ_C 19.56 (Me), 22.26, 28.84, 31.11 (3 x CH_2), 125.45, 127.73, 127.90, 128.95, 129.56, 134.18, 136.26, 138.03 (6 x arC + C=C), 168.67 and 169.61 (C=N and C=O).

2-(3-Cyanobenzylidene)cyclopentanone *O*-acetyloxime AO32 (76%, from **O32**), mp 111-112°C (Found: C, 70.9; H, 5.6; N, 11.0. $C_{15}H_{14}N_2O_2$ requires C, 70.85; H, 5.55; N, 11.0%); λ_{max}/nm 226 (14 400) and 294 (25 600); δ_H 1.91 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.19 (3H, s, Me), 2.72 (2H, t, *J* 7.4, $CH_2C{=}N$), 2.79 (2H, td, J_t 7.4, J_d 2.9, $CH_2C{=}C$), 7.44 (1H, t, *J* 2.9, HC=C), 7.54 (2H, m), 7.60 (1H, d, *J* 7.9) and 7.66 (1H, s) (4 x arH); δ_C 19.53 (Me), 22.26, 28.79, 31.10 (3 x CH_2), 112.58, 118.45, 124.40, 129.28, 131.05, 132.25, 133.63, 137.42, 137.61 (cyano, 6 x arC + C=C), 168.60 and 169.33 (C=N and C=O).

2-(4-Cyanobenzylidene)cyclopentanone *O*-acetyloxime AO33 (81%, from **O33**), mp 120-121 °C (Found: C, 70.9; H, 5.6; N, 11.0. $C_{15}H_{14}N_2O_2$ requires C, 70.85; H, 5.55; N, 11.0%); λ_{max}/nm 228 (8 700) and 308 (30 000); δ_H 1.95 (2H, qn, *J* 7.9, $CH_2CH_2CH_2$), 2.24 (3H, s, MeCO), 2.75 (2H, t, *J* 7.9, $CH_2C{=}N$), 2.82 (2H, td, J_t 7.9, J_d 2.9, $CH_2C{=}C$), 7.51 (3H, m, HC=C + 2 x arH) and 7.66 (2H, d, *J* 8.9, 2 x arH); δ_C 19.69 (Me), 22.44, 28.96, 31.43 (3 x CH_2), 118.79 (cyano), 111.17, 125.14, 129.92, 132.24, 138.70, 140.85 (arC + C=C), 168.70 and 169.49 (C=N and C=O). *Table of Contents*

2-(2,4-Difluorobenzylidene)cyclopentanone *O*-acetyloxime AO34 (70%, from **O34**), mp 117-118 °C (Found: C, 63.3; H, 4.9; N, 5.2; F, 14.5. $C_{14}H_{13}NO_2F_2$ requires C, 63.4; H, 4.9; N, 5.3; F, 14.3%); λ_{max}/nm 218 (7 800) and 292 (20 000); δ_H 1.85 (2H, qn, *J* 7.0, $CH_2CH_2CH_2$),

2.20 (3H, s, MeCO), 2.70 (4H, m, CH_2C=C and CH_2C=N), 6.82 (2H, m) and 7.35 (1H, m) (3 x arH), 7.51 (1H, br s, HC=C); δ_C 19.60 (Me), 22.22, 28.92, 31.19 (3 x CH_2), 104.60 (t, *J* 26), 111.70 (dd, *J* 21, 3), 120.75 (d, *J* 17), 131.50 (dd, *J* 6, 3) (4 x arC), 118.30 (d, *J* 3, *C*H=CCH_2), 136.92 (CH=*C*CH_2), 162.51 (d, *J* 252) and 162.53 (d, *J* 252) (2 x arC-F), 168.90 and 169.45 (C=O and C=N).

2-(3-Fluorobenzylidene)cyclopentanone *O*-acetyloxime AO35 (74%, from **O35**), mp 84-85°C (Found: C, 68.0; H, 5.7; N, 5.7; F, 7.9. $C_{14}H_{14}NO_2F$ requires C, 68.0; H, 5.7; N, 5.7; F, 7.7%); λ_{max}/nm 220 (9 700) and 298 (25 800); δ_H 1.88 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.20 (3H, s, Me), 2.71 (2H, t, *J* 7.4, CH_2C=N), 2.80 (2H, td, J_t 7.4, J_d 2.5, CH_2C=C), 6.75 (1H, td, J_t 8.4, J_d 2.5), 7.11 (1H, dt, J_d 10.3, J_t 2.0), 7.18 (1H, d, *J* 7.9), 7.30 (1H, m) (4 x arH) and 7.46 (1H, t, *J* 2.5, HC=C).

2-(3-Acetoxybenzylidene)cyclopentanone *O*-acetyloxime AO37

2-(3-Hydroxybenzylidene)cyclopentanone *O*-acetyloxime **AO14** (0.71 g, 2.9 mmol) was dissolved in 5M sodium hydroxide solution (10 cm^3) and ice (10 g) was added. Acetic anhydride (1.25 g, 12.2 mmol) was then added quickly with vigorous stirring. The reaction was stirred for 10 minutes, then filtered and recrystallised to yield white crystals of 2-(3-acetoxybenzylidene)cyclopentanone *O*-acetyloxime **AO37** (0.78 g, 94%), mp 110-111 °C (Found: C, 66.75; H, 6.0; N, 4.8. $C_{16}H_{17}NO_4$ requires C, 66.9; H, 6.0; N, 4.9%); λ_{max}/nm 224 (15 800) and 298 (23 800); δ_H 1.92 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.25 (3H, s, MeCO), 2.34 (3H, s, MeCO), 2.75 (2H, t, *J* 7.5, CH_2C=N), 2.85 (2H, dd, *J* 7.5, 2.5, CH_2C=C), 7.05 (1H, dd, *J* 8.0, 1.6), 7.18 (1H, s), 7.31 (1H, d, *J* 8.0) and 7.40 (1H, t, *J* 8.0) (4 x arH), 7.52 (1H, t, *J* 2.5, HC=C); δ_C 19.69, 21.16, 22.40, 28.99, 31.24 (2 x *Me*CO and 3 x CH_2), 121.22, 122.23, 126.16, 127.28, 129.37, 135.87, 137.88, 150.59 (6 x arC + C=C), 168.93, 169.41 and 169.88 (C=N + 2 x C=O).

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2-(4-Acetoxybenzylidene)cyclopentanone *O*-acetyloxime AO38

Ice (10 g) was added to a solution of 2-(4-hydroxybenzylidene)cyclopentanone *O*-acetyloxime **AO15** (0.76 g, 3.1 mmol) in 5M sodium hydroxide (10 cm^3). Acetic anhydride (1.25 g, 12.2 mmol) was then added rapidly with vigorous stirring. After 10 minutes the product was filtered off and recrystallised to yield yellow crystals of 2-(4-acetoxybenzylidene)cyclopentanone *O*-acetyloxime **AO38** (0.85 g, 95%), mp 107-108 °C

(Found: C, 66.8; H, 6.0; N, 4.85. $C_{16}H_{17}NO_4$ requires C, 66.9; H, 6.0; N, 4.9%); λ_{max}/nm 224 (10 700) and 306 (22 500); δ_H 1.89 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.22 (s, 3H) and 2.30 (s, 3H) (MeCO), 2.72 (2H, t, *J* 7.5, $CH_2C{=}N$), 2.80 (2H, td, J_t 7.5, J_d 2.5, $CH_2C{=}C$), 7.09 (d, 2H, *J* 8.6) and 7.43 (d, 2H, *J* 8.6) (4 x arH), 7.50 (1H, t, *J* 2.5, HC=C); δ_C 19.69, 21.13 (Me), 22.40, 29.02, 31.17 (3 x CH_2), 121.62, 126.23, 130.68, 134.14, 134.82, 150.22 (C=C + 4 x arC), 168.98, 169.39 and 170.09 (C=N + 2x C=O).

Data for 2,3-Dihydro-1*H*-cyclopenta[*b*]quinoline and related compounds

2,3-Dihydro-1*H*-cyclopenta[*b*]quinoline: See page 12.

9-Phenyl-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q10 (from **MO1a**) mp 132-134 °C (methanol) (lit.,[39] 134-135 °C); ν_{max}/ cm^{-1} 3058, 2965, 2917, 1608, 1589, 1571, 1486, 1436, 1425, 1385, 1342, 1311, 1274, 1210, 1179, 1142, 1078, 1026, 766, 724, 704, 668 and 612; δ_H 2.16 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.90 (2H, t, *J* 7.5, CH_2Ar), 3.24 (2H, t, *J* 7.5, CH_2Ar), 7.36 (3H, m), 7.49 (3H, m) and 7.62 (2H, m) (8 x arH), 8.08 (1H, d, *J* 8.4, arH-5); δ_C 23.42, 30.22, 35.08 (3 x CH_2), 125.39, 125.54, 126.09, 127.88, 128.13, 128.39, 128.68, 129.18, 133.55, 136.62, 142.59, 147.81 and 167.31 (13 x arC).

8-Methyl-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q12a (from **MO2**), mp 64-65 °C (light petroleum) (Found: C, 85.35; H, 7.1; N, 7.4. $C_{13}H_{13}N$ requires: C, 85.2; H, 7.15; N, 7.6%); λ_{max}/nm 240 (29 800) and 322 (4 800); ν_{max}/ cm^{-1} 3058, 2957, 2895, 2861, 2837, 1609, 1572, 1494, 1455, 1424, 1399, 1374, 1364, 1311, 1229, 1203, 1152, 1114, 1039, 970, 909, 892, 873, 810, 756, 695, 649 and 627; δ_H 2.21 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.65 (3H, s, Me), 3.11 (2H, t, *J* 7.4, CH_2Ar), 3.16 (2H, t, *J* 7.4, CH_2Ar), 7.28 (1H, d, *J* 8.4, arH-7), 7.50 (1H, t, *J* 8.4, arH-6), 7.87 (1H, d, *J* 8.4, arH-5) and 8.07 (1H, s, arH-9); δ_C 18.83 (Me), 23.65, 30.72 and 34.50 (3 x CH_2), 126.12, 126.53, 126.75, 126.89, 127.94, 133.98, 135.22, 147.68 and 167.23 (9 x arC).

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6-Methyl-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q12b (from **MO4**), mp 86-88 °C (light petroleum) (Found: C, 85.2; H, 7.1; N, 7.8. $C_{13}H_{13}N$ requires: C, 85.2; H, 7.15; N, 7.6%); λ_{max}/nm 236 (11 900) and 326 (3 600); ν_{max}/ cm^{-1} 3032, 2948, 2924, 1626, 1566, 1498, 1455, 1438, 1427, 1411, 1360, 1305, 1277, 1223, 1152, 1090, 1035, 1009, 923, 881 and 808; δ_H 2.20 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.54 (3H, s, Me), 3.07 (2H, t, *J* 7.5, CH_2Ar), 3.15 (2H, t, *J*

7.5, CH_2Ar), 7.30 (1H, d, *J* 8.2, arH-7), 7.63 (1H, d, *J* 8.2, arH-8), 7.79 (1H, s, arH-5) and 7.85 (1H, s, arH-9); δ_C 21.82 (Me), 23.66, 30.48 and 34.63 (3 x CH_2), 125.37, 127.07, 127.70, 128.98, 130.15, 131.93, 134.73, 138.49 and 167.80 (9 x arC).

8-Methoxy-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q12c (48%, from 2-(2-methoxybenzylidene)cyclopentanone *O*-methyloxime **MO8**), mp 76-77 °C (light petroleum) (Found: C, 78.3; H, 6.6; N, 6.85. $C_{13}H_{13}NO$ requires: C, 78.4; H, 6.6; N, 7.0%); λ_{max}/nm 248 (21 900) and 308 (2 600); ν_{max}/ cm^{-1} 3070, 2952, 2927, 2836, 1613, 1573, 1473, 1433, 1399, 1367, 1311, 1259, 1219, 1193, 1119, 1065, 958, 911, 878, 811 and 759; δ_H 2.21 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 3.09 (2H, t, *J* 7.6, CH_2Ar), 3.15 (2H, t, *J* 7.6, CH_2Ar), 3.99 (3H, s, MeO), 6.81 (1H, d, *J* 8.1, arH-7), 7.51 (1H, t, *J* 8.1, arH-6), 7.61 (1H, d, *J* 8.1, arH-5) and 8.33 (1H, s, arH-9); δ_C 23.61, 30.64, 34.62 (3 x CH_2), 55.68 (OMe), 103.62, 119.56, 120.89, 125.06, 128.17, 134.76, 148.34, 155.07 and 168.05 (9 x arC).

6-Methoxy-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q12d (53%, from **MO10**), mp 58-60 °C (light petroleum) (Found: C, 78.3; H, 6.5; N, 7.1. $C_{13}H_{13}NO$ requires: C, 78.4; H, 6.6; N, 7.0%); λ_{max}/nm 236 (8 400) and 336 (1 200); ν_{max}/ cm^{-1} 3014, 2952, 2831, 1620, 1569, 1499, 1467, 1450, 1411, 1379, 1369, 1301, 1278, 1262, 1227, 1201, 1168, 1150, 1132, 1087, 1028, 961, 914, 876, 845, 814, 767 and 659; δ_H 2.20 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 3.06 (2H, t, *J* 7.5, CH_2Ar), 3.15 (2H, t, *J* 7.5, CH_2Ar), 3.93 (3H, s, OMe), 7.12 (1H, dd, *J* 8.8, 2.2, arH-7), 7.37 (1H, d, *J* 2.2, arH-5), 7.62 (1H, d, *J* 8.8, arH-8) and 7.82 (1H, s, arH-9); δ_C 23.58, 30.32, 34.59 (3 x CH_2), 55.35 (OMe), 106.95, 118.20, 122.34, 128.30, 130.23, 133.31, 148.98, 159.86 and 167.98 (9 x arC).

6-Hydroxy-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q12e (from **AO15**), mp 168-169 °C (Found: C, 77.9; H, 6.1; N, 7.5. $C_{12}H_{11}NO$ requires C, 77.8; H, 6.0; N, 7.6%); λ_{max}/nm 214 (24 859) and 338 (4 346); ν_{max}/ cm^{-1} (KBr pellet): 3132, 2966, 2927, 2854, 1620, 1466, 1401, 1243, 1135, 920, 860 and 816 cm^{-1}; δ_H $(CD_3)_2SO$ 2.09 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.93-3.01 (4H, m, CH_2Ar), 7.00 (1H, dd, *J* 8.8, 2.2, arH-7), 7.24 (1H, d, *J* 2.2, arH-5), 7.50 (1H, d, *J* 8.8, arH-8), 7.73 (1H, s, arH-9) and 9.65 (1H, br s, OH); δ_C $(CD_3)_2SO$ 22.64, 29.27, 33.59 (3 x CH_2), 109.30, 117.16, 120.73, 127.36, 129.43, 131.50, 148.06, 157.06 and 166.67 (9 x arC).

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6-Acetoxy-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q12f (from **AO38**), mp 96-97 °C (Found: C, 74.0; H, 5.85; N, 6.2. $C_{14}H_{13}NO_2$ requires C, 74.0; H, 5.8; N, 6.2%); λ_{max}/nm 236 (19 500) and 322 (5 500); ν_{max}/ cm^{-1} (KBr pellet): 2957, 2925, 1758, 1621, 1573, 1486, 1416, 1372, 1206, 1132, 1015, 972 and 926 cm^{-1}; δ_H 2.21 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.36 (s, 3H, MeCO), 3.08 (2H, td, J_t 7.6, J_d 1.0, CH_2Ar), 3.08 (2H, t, *J* 7.6, CH_2Ar), 7.24 (1H, dd, *J* 8.8, 2.4, arH-7), 7.71-7.74 (2H, m, arH-5/8) and 7.88 (1H, br s, arH-9); δ_C 21.19 (Me), 23.58, 30.42, 34.57 (3 x CH_2), 119.64, 120.80, 125.40, 128.34, 130.15, 135.62, 147.91, 150.46, 168.57 and 169.41(9 x arC + C=O).

6-*N*,*N*-Dimethylamino-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q12g (from **AO17**), mp 104-106 °C (Found: C, 79.4; H, 7.6; N, 13.3. $C_{14}H_{16}N_2$ requires C, 79.2; H, 7.6; N, 13.2%.); λ_{max}/nm 258 (18 878), 292 (4 733) and 378 (4 581); ν_{max}/ cm^{-1} (KBr pellet): 2962, 2929, 1624, 1515, 1450, 1417, 1383, 1368, 1306, 1262, 1163, 1141, 974, 915, 928 and 801 cm^{-1}; δ_H 2.16 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 3.01 (2H, td, J_t 7.5, J_d 1.0, CH_2Ar), 3.06 (6H, s, NMe_2), 3.09 (2H, t, *J* 7.5, CH_2Ar), 7.13 (1H, dd, *J* 8.8, 2.6, arH-7), 7.14 (1H, d, *J* 2.6, arH-5), 7.55 (1H, d, *J* 8.8, arH-8) and 7.72 (1H, s, arH-9); δ_C 23.60, 30.33, 34.70 (3 x CH_2), 40.60 (NMe_2), 107.11, 115.15, 119.93, 127.86, 130.08, 131.41, 149.20, 150.67 and 167.77 (9 x arC).

6-Amino-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q12h (from **O19**), mp 94-96 °C (Found: C, 78.1; H, 6.7; N, 15.2. $C_{12}H_{12}N_2$ requires C, 78.2; H, 6.6; N, 15.2%); λ_{max}/nm 226 (12 100), 258 (18 100), 300 (4 800) and 390 (4 250); ν_{max}/ cm^{-1} (KBr pellet): 3438, 3157, 2964, 2927, 2857, 1636, 1563, 1515, 1460, 1401, 1322, 1198, 1113 and 1046 cm^{-1}; δ_H 2.15 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 3.02 (2H, td, J_t 7.6, J_d 1.0, CH_2Ar), 3.26 (2H, t, *J* 7.6, CH_2Ar), 4.22 (2H, br, NH_2), 6.89 (1H, dd, *J* 8.5, 2.3, arH-7), 7.36 (1H, s, arH-9), 7.50 (1H, d, *J* 8.5, arH-8) and 7.80 (1H, d, *J* 2.3, arH-5); δ_C 22.73, 30.31, 31.17 (3 x CH_2), 99.83, 118.84, 121.96, 123.55, 129.47, 133.34, 141.82, 148.14 and 152.25 (9 x arC). *Table of Contents*

9,10-Dihydro-*8H*-benzo[*f*]cyclopenta[*b*]quinoline Q13: (from **MO20)**, mp 126-128 °C (light petroleum) (Found: C, 87.8; H, 6.1; N, 6.1. $C_{16}H_{13}N$ requires: C, 87.6; H, 6.0; N, 6.4%); λ_{max}/nm 236 (35 800), 280 (17 200), 334 (5 800) and 350 (6 900); ν_{max}/ cm^{-1} 3058, 3038, 2955, 2929, 1612, 1567, 1485, 1444, 1426, 1406, 1380, 1354, 1279, 1231, 1191, 1163, 1122, 1029, 1006, 983, 946, 902, 877, 867, 838, 754, 746 and 699; δ_H 2.27 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 3.19 (2H, t, *J* 7.4, $ArCH_2$), 3.22 (2H, *J* 7.4, $ArCH_2$), 7.61 (1H, t, *J* 7.4) and 7.66 (1H, t, *J* 7.4) (arH-2/3), 7.92 (3H, m; arH-1/4/5), 8.60 (1H, d, *J* 7.8, arH-6) and 8.74 (1H,

s, arH-11); δ_C 23.66, 30.92, 34.47 (3 x CH_2), 122.36, 123.99, 125.75, 126.60, 126.69, 127.91, 128.61, 129.63, 129.82, 131.48, 135.82, 147.08 and 166.86 (13 x arC).

1,2,3,12-Tetrahydrocyclopenta[5,6]pyrido[3,2-*a*]phenothiazine Q14: (from **AO22**), mp 67-68 °C (Found: C, 74.4; H, 4.6; N, 9.7. $C_{18}H_{14}N_2S$ requires C, 74.45; H, 4.9; N, 9.65%.); λ_{max} /nm 206 (12 000), 232 (10 200), 252 (9 300) and 324 (2 400); ν_{max}/ cm^{-1} (KBr pellet): 3453, 2962, 2926, 1650, 1475, 1426, 1384, 1262, 1095, 1024, 868, 802 and 738 cm^{-1}.; δ_H 2.14 (2H, qn, *J* 7.2, $CH_2CH_2CH_2$), 2.44 (2H, t, *J* 7.2) and 3.29 (2H, t, *J* 7.2) (2 x CH_2Ar), 6.38 (1H, s, NH), 6.68 (1H, dd, *J* 7.2, 0.8), 6.95 (2H, m), 7.10 (3H, m) and 7.32 (1H, dd, *J* 8.8, 1.0) (7 x arH); δ_C 16.69, 24.56, 29.78 (3 x CH_2), 107.64, 116.34, 116.46, 116.74, 117.19, 119.18, 123.02, 123.77, 124.74, 126.52, 127.56, 128.25, 136.08, 136.18 and 137.56 (15 x arC).

6,7-Dihydro-5*H*-cyclopenta[*b*]furo[2,3-*e*]pyridine Q15 (45%, from 2-furfurylidenecyclopentanone *O*-methyloxime **MO23**), mp 64-65 °C (light petroleum) (Found: C, 75.6; H, 5.65; N, 8.6. $C_{10}H_9NO$ requires: C, 75.45; H, 5.7; N, 8.8%); λ_{max}/nm 238 (1 700) and 298 (4 800); ν_{max}/ cm^{-1} 3118, 2898, 2847, 1575, 1531, 1434, 1395, 1334, 1266, 1209, 1154, 1121, 1066, 1019, 913, 887, 872, 784 and 743; δ_H 2.15 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.97 (2H, t, *J* 7.4, CH_2Ar), 3.02 (2H, t, *J* 7.4, CH_2Ar), 6.84 (1H, d *J* 2.5, furoH-3), 7.51 (1H, s, pyridylH-8) and 7.70 (1H, d, *J* 2.5, furoH-2); δ_C 24.14, 30.70, 33.65 (3 x CH_2), 107.68, 114.64, 133.36, 145.67, 147.31, 147.74 and 162.15 (furopyridine).

6,7-Dihydro-5*H*-cyclopenta[*b*]thieno[2,3-*e*]pyridine Q16: (from **AO24**), mp 84-85 °C (Found: C, 68.4; H, 5.2; N, 7.9; S, 18.7. $C_{10}H_9NS$ requires C, 68.5; H, 5.2; N, 8.0; S, 18.3%); λ_{max}/nm 228 (29 800) and 302 (9 900); ν_{max}/ cm^{-1} (KBr pellet): 3089, 3039, 2968, 2918, 2854, 1545, 1538, 1431, 1395, 1381, 1310, 1232, 1075, 1033, 919, 890, 833, 776, 691 and 684 cm^{-1}; δ_H 2.21 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 3.03 (2H, t, *J* 7.4, CH_2Ar), 3.11 (2H, t, *J* 7.4, CH_2Ar), 7.47 (1H, d, *J* 5.9) and 7.52 (1H, d, *J* 5.9) (thienoH-2/3), 7.95 (1H, s, pyridylH-8); δ_C 23.83, 30.45, 33.87 (3 x CH_2), 124.43, 125.70, 128.66, 131.24, 133.10, 154.64 and 164.39 (7 x arC).

2-Phenyl-6,7-dihydro-5*H*-cyclopenta[*b*]pyridine Q18: (from **AO25**), mp 79-80 °C (lit.,[40] 81-82 °C); λ_{max}/nm 248 (12 000) and 292 (11 500); ν_{max}/ cm^{-1} (KBr pellet): 2960, 2960, 2927, 2857, 1656, 1544, 1441, 1401, 1263, 842, 772, 734 and 695 cm^{-1}; δ_H 2.04 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.84 (2H, t, *J* 7.6, CH_2Ar), 2.96 (2H, t, *J* 7.6, CH_2Ar), 7.24 (1H, tt, *J* 7.6, 1.2), 7.32 (3H, m), 7.42 (1H, d, *J* 7.6) and 7.81 (2H, m) (7 x arH); δ_C 23.23, 30.48, 34.41 (3 x CH_2), 118.25, 126.89, 128.32, 128.64, 132.56, 135.41, 140.00, 155.87 and 165.83 (9 x arC).

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5-Methyl-2,3-dihydro-[1*H*]-cyclopenta[*b*]quinoline Q26a: (from **AO3**), mp 92-94°C (light petroleum) (Found: C, 85.2; H, 7.5; N, 7.00. $C_{13}H_{13}N$ requires C, 85.2; H, 7.15; N, 7.6%); λ_{max}/nm (methanol) 240 (28 400), 305 (3 880) and 322 (4 350); ν_{max}/cm^{-1} (KBr pellet): 3025, 2954, 2925, 2854, 1623, 1495, 1473, 1409, 1352, 918, 776 and 769; δ_H 2.19 (2H, qn, *J* 7.4, $CH_2CH_2CH_2$), 2.80 (3H, s, Me), 3.05 (2H, t, *J* 7.4, CH_2), 3.16 (2H, t, *J* 7.4, CH_2), 7.32 (t, 1H, J 8.3, arH-7), 7.44 (1H, d, *J* 8.3, arH-8 or -6), 7.55 (1H, d, *J* 8.3, arH-6 or -8) and 7.81 (1H, s, arH-9); δ_C 18.32 (Me), 23.72, 30.43, 34.87 (3 x CH_2), 125.04, 125.52, 127.22, 128.54, 130.46, 135.08, 136.20, 146.63 and 166.88 (arC).

5,6-Dimethyl-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q26b: (from **AO5**), mp 90-91 °C (Found: C, 85.5; H, 7.95; N, 6.8. $C_{14}H_{15}N$ requires C, 85.2; H, 7.7; N, 7.1%); λ_{max}/nm 242 (24 000), 314 (3 440) and 326 (3 700); ν_{max}/cm^{-1} (KBr pellet): 2954, 2843, 1625, 1509 (C=C and (C=N), 1474, 1434, 1358, 1247, 1212, 1160, 1063, 1016 and 850; δ_H 2.10 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.40 (3H, s, Me), 2.67 (3H, s, Me), 2.98 (2H, t, *J* 7.5, CH_2), 3.09 (2H, t, *J* 7.5, CH_2), 7.19 (1H, d, *J* 8.2) and 7.40 (1H, d, *J* 8.2) (arH-7 and arH-8), 7.71 (1H, s, arH-9); δ_C 13.48, 20.64 (2 x Me), 23.75, 30.33, 34.88 (3 x CH_2), 124.45, 125.55, 128.17, 130.46, 133.49, 133.97, 135.91, 146.53 and 166.75 (9 x arC).

5-Methyl-6-Methoxy-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q26c: (from **AO6**), mp 89-91 °C (Found: C, 78.7; H, 7.0; N, 6.6. $C_{14}H_{15}NO$ requires C, 78.8; H, 7.1; N, 6.6%); λ_{max}/nm 244 (23 000) and 334 (5 500); ν_{max}/cm^{-1} (KBr pellet): 2961, 2926, 2854, 1738, 1613, 1503, 1461, 1407, 1344, 1255, 1171, 1105, 1030, 803 and 783; δ_H 2.12 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.62 (3H, s, Me), 2.98 (2H, t, *J* 7.5, CH_2), 3.10 (2H, t, *J* 7.5, CH_2), 3.90 (3H, s, OMe), 7.15 (1H, d, *J* 9.0, arH-7), 7.50 (1H, d, *J* 9.0, arH-8) and 7.73 (1H, s, arH-9); δ_C 9.94 (Me), 23.78, 30.30, 35.00 (3 x CH_2), 56.41 (OMe), 112.25, 121.54, 122.47, 125.56, 130.47, 132.81, 147.15, 156.68 and 167.65 (9 x arC). *Table of Contents*

5-*t*-Butyl-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline[41] **Q26d:** (from **AO7**) mp 62-64 °C (light petroleum) (Found: C, 85.15; H, 8.6; N, 6.5. $C_{16}H_{19}N$ requires C, 85.3; H, 8.5; N, 6.2%; λ_{max}/nm 238 (20 600) and 320 (3 200); ν_{max}/cm^{-1} (KBr pellet): 3048, 2957, 2925, 2855, 1630, 1607, 1488, 1466, 1411, 1353, 1265, 1148, 1097, 1082, 1026, 891, 800 and 763; δ_H 1.70 (9H, s, C*Me*$_3$), 2.20 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 3.07 (2H, t, *J* 7.6, $ArCH_2$), 3.14 (2H, t, J 7.6, Ar'CH_2), 7.36 (1H, t, *J* 7.6, arH-7), 7.58 (2H, coincident doublets, *J* 7.6, arH-6 and arH-8) and 7.83 (1H, s, arH-9); δ_C 22.61, 28.68, 29.39, 30.03, 33.82 (3 x CH_2, *C*Me$_3$ and C*Me*$_3$), 123.73, 123.78, 125.35, 127.04, 129.50, 132.94, 145.59, 146.31 and 163.70 (9 x arC).

7-Methoxy-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q27e: (from **AO9** or **MO9**), mp 96-97°C (light petroleum) (lit.[42] 99-100 °C) (Found: C, 78.45; H, 6.7; N, 7.0; $C_{13}H_{13}NO$ requires C, 78.4; H, 6.6; N, 7.0%); λ_{max}/nm (methanol) 236 (8 420) and 334 (1 250); ν_{max}/cm^{-1} (KBr pellet): 3040, 2960, 2882, 2840, 1619, 1500, 1388, 1366, 1299, 1217, 1158, 1118, 1090, 1032, 902, 874, 828 and 616; δ_H 2.72 (2H, qn, *J* 7.9, $CH_2CH_2CH_2$), 3.58 (2H, t, *J* 7.9, $ArCH_2$), 3.65 (2H, t, *J* 7.9, $ArCH_2$), 4.42 (3H, s, OMe), 7.53 (1H, d, *J* 2.5, arH-8), 7.81 (1H, dd, *J* 8.9, 2.5, arH-6), 8.32 (s, 1H, arH-9), and 8.45 (1H, d, *J* 8.9, arH-5); δ_C 23.28, 30.18, 33.93 (3 x CH_2), 55.06 (OMe), 105.16, 120.07, 127.87, 128.93, 129.46, 135.52, 143.05, 156.70 and 164.98 (arC).

6,7-Dimethoxy-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q27g: (from **AO12**), mp 99-100 °C (lit.,[43] 112-113 °C; lit. 120-121 °C) (Found: C, 73.4; H, 6.5; N, 5.8. $C_{14}H_{15}NO_2$ requires C, 73.3; H, 6.6; N, 6.1%); (λ_{max}/nm 222 (36 400), 324 (10 450) and 336 (14 280); ν_{max}/cm^{-1}.(KBr pellet): 3000, 2958, 2834, 1619, 1505, 1456, 1421, 1384, 1284, 1243, 1150, 1011, 910, 884 and 748; δ_H 2.12 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.98 (2H, t, *J* 7.6, CH_2), 3.04 (2H, t, *J* 7.6, CH_2), 3.92 (3H, s, OMe), 3.94 (3H, s, OMe), 6.92 (1H, s, arH-8), 7.31 (1H, s, arH-5) and 7.68 (1H, s, arH-9); δ_C 23.59, 30.46, 34.34 (3 x CH_2), 55.90, 55.94 (both OMe), 105.21, 107.53, 122.44, 129.10, 133.69, 143.99, 148.82, 151.34 and 165.24 (9 x arC).

6-Methyl-7-methoxy-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q27h: (from **AO13**), mp 129-130 °C (Found: C, 78.6; H, 7.3; N, 6.5. $C_{14}H_{15}NO$ requires C, 78.8; H, 7.1; N, 6.6%); λ_{max}/nm 224 (34 000), 246 (15 500), 322 (6 700) and 336 (8 280); ν_{max}/cm^{-1}.(KBr pellet): 2954, 2927, 2855 (OMe), 1628, 1498 (C=C and (C=N), 1461, 1426, 1383, 1370, 1232, 1136, 1100, 1026, 923, 884 and 832; δ_H 2.10 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.31 (3H, s, Me), 2.96 (2H, t, *J* 7.4, CH_2), 3.03 (2H, t, *J* 7.6, CH_2), 3.84 (3H, s, Me), 6.85 (1H, s, arH-8) and 7.68 (2H, coincident singlets, arH-5/9); δ_H $(CD_3)_2SO$ 2.01 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.21 (3H, s, Me), 2.88 (4H, m, $CH_2C{=}C$ and $CH_2C{=}N$), 3.80 (3H, s, OMe), 7.03 (1H, s, arH-8), 7.55 (1H, s) and 7.74 (1H, s) (arH-5/9); δ_C 17.01 (Me), 23.62, 29.49, 34.24 (3 x CH_2), 55.34 (OMe), 103.72, 126.75, 129.05, 129.20, 130.93, 134.54, 143.07, 156.21 and 164.83 (9 x arC).

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7-Hydroxy-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q27i: (from **AO14**), mp 142-143 °C (Found: C, 77.75; H, 5.8; N, 7.7. $C_{12}H_{11}NO$ requires C, 77.8; H, 6.0; N, 7.6%); λ_{max}/nm 284 (2 900) and 336 (4 500); ν_{max}/cm^{-1} (KBr pellet) 3450, 3154, 2955, 2956, 2855, 1655, 1619, 1461, 1399, 1276, 1235, 1127, 907 and 827; δ_H $(CD_3)_2SO$ 2.08 (2H, qn, *J* 7.6 $CH_2CH_2CH_2$),

2.96 (4H, m, $ArCH_2$ and $Ar'CH_2$), 6.96 (1H, d, *J* 2.4, arH-8), 7.13 (1H, dd, *J* 8.9, 2.4, arH-6), 7.63 (1H, s, arH-9), 7.70 (1H, d, *J* 8.9, arH-5) and 9.35 (1H, s, OH); δ_C $(CD_3)_2SO$ 23.19, 30.00, 33.68 (3 x CH_2), 108.45, 120.09, 128.14, 128.48, 128.91, 135.22, 141.99, 154.44 and 164.02 (9 x arC).

7-*N*,*N*-Dimethylamino-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q27j: (from **AO16**), mp 122-123 °C (B) (Found: C, 79.3; H, 7.6; N, 13.4. $C_{14}H_{16}N_2$ requires C, 79.2; H, 7.6; N, 13.2%); λ_{max}/nm 260 (24 700) and 374 (3 600); ν_{max}/cm^{-1} (KBr pellet): 3164, 2955, 2927, 2858, 1619, 1513, 1400, 1363, 1142, 968, 906 and 815; δ_H 2.10 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.96 (2H, t, *J* 7.5, $ArCH_2$), 2.98 (6H, s, NMe_2), 3.03 (2H, t, *J* 7.5, $ArCH_2$), 6.71 (1H, d, *J* 2.6, arH-8), 7.21 (1H, dd, *J* 9.2, 2.6, arH-6), 7.65 (1H, s, arH-9) and 7.80 (1H, d, *J* 9.2, arH-5); δ_C 23.48, 30.57, 34.14 (3 x CH_2), 40.88 (NMe_2), 105.75, 118.27, 128.70, 128.83, 128.87, 135.75, 141.29, 148.18 and 163.56 (9 x arC).

7-Amino-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q27k: (from **O18**), mp 121-122 °C (Found: C, 78.0; H, 6.6; N, 15.1. $C_{12}H_{12}N_2$ requires C, 78.2; H, 6.6; N, 15.2%); λ_{max}/nm 354 (4969), 248 (29679) and 224 (27141); ν_{max}/cm^{-1} 3423, 3312, 3138, 2953, 2936, 1638, 1626, 1510, 1401, 1368, 1322, 1245, 1220, 1136, 1032, 913, 821 and 747; δ_H 2.00 (2H, qn, *J* 7.6, $CH_2CH_2CH_2$), 2.85 (2H, t, *J* 7.6, $ArCH_2$), 2.94 (2H, t, *J* 7.6, $ArCH_2$), 3.80 (2H, s, NH_2), 6.67 (1H, d, *J* 2.4, arH-8), 6.89 (1H, dd, *J* 8.8, 2.4, arH-6), 7.47 (1H, s, arH-9) and 7.68 (1H, d, *J* 8.8, arH-5); δ_C 23.51, 30.37, 33.93 (3 x CH_2), 107.91, 120.13, 128.39, 128.59, 128.46, 135.75, 142.14, 143.81 and 163.90 (9 x arC).

7-Acetoxy-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q27l: (from **AO37**), mp 118-119 °C (Found: C, 74.25; H, 5.8; N, 5.8. $C_{14}H_{13}NO_2$ requires C, 74.0; H, 5.8; N, 6.2%); λ_{max}/nm 234 (23 400), 282 (5 700) and 332 (6 100); ν_{max}/cm^{-1} (KBr pellet): 2957, 2925, 2853, 1765, 1620, 1498, 1462, 1431, 1371, 1192, 1145, 1014, 966, 922, 878, 836 and 744; δ_H 2.13 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 2.27 (3H, s, MeCO), 2.99 (2H, td, J_t 7.5, J_d 1.2, $ArCH_2$), 3.07 (2H, t, *J* 7.5, $Ar'CH_2$), 7.27 (1H, dd, *J* 8.9, 2.5, arH-6), 7.39 (1H, d, *J* 2.5, arH-8), 7.76 (1H, s, arH-9) and 7.95 (1H, d, *J* 8.9, arH-5); δ_C 20.17 (Me), 22.59, 29.49, 33.45 (3 x CH_2), 117.21, 122.27, 126.62, 128.84, 129.05, 135.35, 144.38, 146.81, 166.94 (9 x arC) and 168.53 (C=O).

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5-Hydroxy-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q26l': (from **AO37**), mp 74-75 °C (Found: C, 77.6; H, 5.8; N, 7.6. $C_{12}H_{11}NO$ requires C, 77.8; H, 6.0; N, 7.6 %); λ_{max}/nm 248 (23 000) and 310 (2 200); ν_{max}/cm^{-1} (KBr pellet): 3450, 2958, 2925, 2854, 1498, 1462, 1379,

1331, 1237, 1210, 1129, 964, 916, 870 and 755; δ_H $(CD_3)_2SO$ 2.14 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 3.05 (4H, m, $CH_2C{=}C$ and $CH_2C{=}N$), 6.98 (1H, dd, *J* 8.0, 1.6, arH-6) and 7.28 (1H, dd, *J* 8.0, 1.6, arH-8), 7.32 (1H, t, *J* 8.0, arH-7), 8.03 (1H, s, arH-9) and 8.29 (1H, s, OH); δ_C 23.65, 30.47, 34.11 (3 x CH_2), 109.15, 117.65, 126.39, 127.53, 130.35, 136.60, 137.35, 151.49 and 165.70 (9 x arC).

5,8-Dimethoxy-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline Q29: (from **AO11**), mp 104-105 °C (lit.,[44] 98-100 °C) (Found: C, 72.8; H, 6.5; N, 6.0. $C_{14}H_{15}NO_2$ requires C, 73.3; H, 6.6; N, 6.1%); λ_{max}/nm 260 (31 000) and 336 (2 300); ν_{max}/ cm^{-1} (KBr pellet): 3370, 2994, 2952, 2937, 2839, 1614, 1486, 1460, 1378, 1327, 1279, 1264, 1211, 1143, 1094, 1068, 977, 911, 814, 793 and 724; δ_H 2.11 (2H, qn, *J* 7.5, $CH_2CH_2CH_2$), 3.00 (2H, td, J_t 7.5, J_d 1.0, CH_2Ar), 3.13 (2H, t, *J* 7.5, CH_2Ar), 3.86 (3H, s, OMe), 3.94 (3H, s, OMe), 6.61 (1H, d, *J* 8.8) and 6.77 (1H, d, *J* 8.8) (arH-6/7), 8.22 (1H, s, arH-9); δ_C 22.62, 29.64, 33.86 (3 x CH_2), 54.76, 54.89 (OMe), 101.89, 104.76, 119.58, 124.20, 134.44, 138.64, 147.73, 148.24 and 166.25 (9 x arC).

9,10-Dihydro-8*H*-benzo[*h*]cyclopenta[*b*]quinoline Q31: (from **AO21** or **MO21**), mp 115-116 °C; δ_H 2.23 (2H, qn, *J* 7.9, $CH_2CH_2CH_2$), 3.07 (2H, t, *J* 7.9, $ArCH_2$), 3.26 (2H, t, *J* 7.9, $ArCH_2$), 7.59 (1H, d, *J* 8.9), 7.70 (m, 3H), 7.84 (s, 1H, arH-7), 7.89 (1H, dd, *J* 7.9, 0.9Hz) and 9.35 (1H, d, *J* 8.4); δ_C 23.57, 30.49, 34.70 (3 x CH_2), 124.12, 124.90, 125.52, 126.30, 126.48, 127.34, 127.55, 130.60, 131.42, 133.20, 135.85, 145.32 and 166.14 (13 x arC).

1,2,3,4-Tetrahydroacridine Q36: (from **MO36**), mp 52-53 °C (light petroleum) (lit.,[45] 54.5 °C); ν_{max}/ cm^{-1} 3055, 2940, 2875, 1623, 1600, 1562, 1492, 1438, 1414, 1242, 1151, 1010, 966, 913, 846, 805, 778, 748 and 614; δ_H 1.89 (2H, m, CH_2), 1.99 (2H, m, CH_2), 2.98 (2H, t, *J* 6.1, CH_2Ar), 3.13 (2H, t, *J* 6.4, CH_2Ar), 7.43 (1H, t, *J* 7.8, arH-6) and 7.60 (1H, t, *J* 8.3, arH-7), 7.69 (1H, d, *J* 7.8, arH-8), 7.80 (1H, s, arH-9) and 7.97 (1H, d, *J* 8.3, arH-5); δ_C 22.90, 23.23, 29.26, 33.56 (4 x CH_2), 125.54, 126.89, 127.19, 128.23, 128.51, 130.98, 135.02, 146.56 and 159.32 (9 x arC).

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Nitro-, chloro-, cyano-, fluoro-substitued precursors: Similar irradiations of 2-(2-nitrobenzylidene)cyclopentanone *O*-methyloxime **MO26**, 2-(4-nitrobenzylidene)-cyclopentanone *O*-methyloxime **MO28**, 2-(3-nitrobenzylidene)cyclopentanone *O*-acetyloxime **AO27**, 2-(2-chlorobenzylidene)cyclopentanone *O*-methyloxime **MO29**, 2-(4-chloro-

benzylidene)cyclopentanone *O*-methyloxime **MO31**, 2-(4-cyanobenzylidene)cyclopentanone *O*-acetyloxime **AO33**, 2-(3-cyanobenzylidene)cyclopentanone *O*-acetyloxime **AO32**, 2-(2,4-difluorobenzylidene)cyclopentanone *O*-acetyloxime **AO34**, 2-(3-chlorobenzylidene)-cyclopentanone *O*-acetyloxime **AO30**, and 2-(3-fluorobenzylidene)cyclopentanone *O*-acetyloxime **AO35** resulted only in the rapid formation of complex mixtures lacking any major product, and were not investigated further. Nmr spectroscopic investigation of the mixtures indicated the presence of only trace amounts at most of the corresponding quinoline derivatives and tlc examination showed the presence of a plethora of very difficult-to-separate components.

Thermolysis of 2-diphenylmethylenecyclopentanone *O*-methyloxime MO1a

2-Diphenylmethylenecyclopentanone *O*-methyloxime **MO1a** (50 mg, 0.18 mmol) in ethylene glycol (50 cm^3) was heated under reflux for 10 hours. TLC did not show any product formation. Removal of the solvent by vacuum distillation yielded only solid **MO1a** (IR, TLC).

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