Supplementary Information

Preparation of coumpounds

All chemicals and solvents were obtained from commercial suppliers and used without further purification. Reactions were monitored by thin layer chromatography. Melting points were determined on a Büchi 530 apparatus and are uncorrected. Infrared (IR) spectra were recorded on a Perkin-Elmer 833 spectrophotomere. ^{1}H-NMR spectra were taken in Deuteriochloroform or Hexadeuterodimethyl sulfoxide and recorded on a Bruker DRX 400 (400 MHz) spectrometer, and the spectra are reported in δ. ^{13}C-NMR spectra were taken at 200 MHz Bruker DRX 400 spectrometer, respectively. Microanalyses were carried out by the NCSR Demokritos, Greece, and the results obtained had a maximum deviation of ± 0.4% from the theoretical value.

Compound **8:** 3-Cyclopentyl-1-tricyclo[3.3.1.1^{3,7}]decylmethyl 4-toluene sulfonate

To a stirred solution of 3-cyclopentyl-1-adamantanemethanol[1] (4.22 g, 18 mmol) in dry pyridine (18 ml), tosyl chloride (4.3 g, 22.5 mmol) was added in small portions, at 0 ^{o}C, under argon. The resultant mixture was stirred at room temperature overnight and then acidified, under cooling, with HCl (10%). The mixture was extracted with ether, the combined ether phases were washed with water, dried over Na_2SO_4 and evaporated under vacuum to give 6.78 g of a viscous product, which was crystallized in the fridge and was used without further purification in the next step. Yield 97%. Mp 55-57 ^{o}C; ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.16 (s, 2H, 6-H), 1.21 (m, 2H, 3,4-$H_{C,ax}$), 1.34-1.46 (complex m, 17H, 2,4,8,9,10-H, 1,2,5-HC, 3,4-$H_{C,eq}$), 2.02 (br. s, 2H, 5,7-H), 2.46 (s, 3H, CH_3), 3.58 (s, 2H, CH_2O), 7.34-7.36 (d, 2H, AA'BB', J_{AB}=$J_{A'B'}$≈8Hz, $J_{AA'}$=$J_{BB'}$≈0Hz, 3,5-H_{ar}), 7.78-7.80 (d, 2H, AA'BB', J_{AB}=$J_{A'B'}$≈8Hz, $J_{AA'}$=$J_{BB'}$≈0Hz, 2,6-H_{ar}).

Compound **9:** 3-Cyclopentyl-1-tricyclo[3.3.1.1^{3,7}]decaneacetonitrile

Sodium cyanide (0.94 g, 19 mmol) was added to a solution of tosylate **8** (2.63 g, 6.75 mmol) in anhydrous DMSO (15 ml). The mixture was stirred under argon at 120 ^{o}C for 20h and after cooling was poured into water, and extracted with ether. The combined ethereal extracts were washed with water, dried over Na_2SO_4 and evaporated. The residue was purified by flash column chromatography by using a mixture of *n*-hexane:ether 8:2 as eluent. Acetonitrile **9** was obtained as an oil in 73% yield (1.2 g). ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.18 (m, 2H, 3,4-$H_{C,ax}$), 1.27 (s, 2H, 6-H), 1.31-1.50 (very complex m, 15 H, 4,8,9,10-H, 1,2,5-H_C, 3,4-$H_{C,eq}$), 1.53 (br. s, 2H, 2-H), 2.02 (m, 2H, 5,7-H), 2.05 (s, 2H, α-H); ^{13}C-NMR ($CDCl_3$), δ(ppm): 25.52 (3,4-C_C), 25.70 (2,5-C_C), 28.76 (5,7-C, α-C), 32.26 (3-C), 32.89 (1-C), 36.19 (2-C), 39.27 (4,10-C), 41.71 (8,9-C), 44.96 (6-C), 51.05 (1-C_C), 117.90 (C≡N)

Compound **10:** 3-Cyclopentyl-1-tricyclo[3.3.1.1^{3,7}]decaneacetic acid

A mixture of 3-cyclopentyl-1-adamantaneacetonitrile (**9**) (2.39 g, 9.8 mmol), KOH (4 g) and ethylene glycol (15 ml) was stirred at 140-150 ^{o}C for 24 h. After cooling, the reaction mixture was poured into boiling water (80-100 ml) and charcoal was added into the mixture. Charcoal was removed by filtration and the filtrate was acidified under cooling by the dropwise addition of concentrated hydrochloric acid. The liberated carboxylic acid was extracted with ether, the combined ether phases were washed with water, dried over Na_2SO_4 and evaporated to yield 1.8 g of acetic acid **10** as a sticky solid, which was used in the next step without further purification. Yield 70%.

Compound **6h:** Ethyl 3-cyclopentyl-1-tricyclo[3.5.1.1^{3,7}]decaneacetate

A mixture of thionyl chloride, freshly distilled over quinoline (6.2 g, 4 ml), and crude acetic acid **10** (1.3 g, 5 mmol) was gently refluxed for 60 min. The excess of thionyl chloride was evaporated in vacuo and the last traces were removed azeotropically with dry benzene. Ethanol was added to the residue and the mixture was gently refluxed for 90 min. The solvent was evaporated under vacuum and water was added. The mixture was extracted with ether, the combined ethereal extracts were washed with water, dried over Na_2SO_4 and evaporated. The residue was purified by flash column chromatography, using as eluent with a mixture of *n*-hexane:ether, 9:1, ethylester **6h** was obtained as an oil. Yield 1 g (67%). IR (film), ν(C=O): 1732 cm^{-1}; ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.23-1.27 (t+m, 5H, A_3X_2, J_{AX}≈7.12 Hz, CH_3, 3,4-$H_{C,ax}$), 1.32 (s, 2H, 6-H), 1.35-1.55 (complex m, 15H, 4,8,9,10-H, 1,2,5-H_C, 3,4-$H_{C,eq}$), 1.57 (br. s, 2H, 2-H), 2.01 (m, 2H, 5,7-H), 2.03 (s, 2H, α-H), 4.08-4.13 (q, 2H, A_3X_2, J_{AX}≈7.12Hz, CH_2O); ^{13}C-NMR ($CDCl_3$), δ(ppm): 14.56 (CH_3), 25.53 (3,4-C_C), 25.78 (2,5-C_C), 29.08 (5,7-C), 33.48 (3-C), 34.93 (1-C), 36.66 (2-C), 39.66 (4,10-C), 42.30 (8,9-C), 45.61 (6-C), 49.01 (α-C), 51.31 (1-C_C), 59.94 (CH_2O), 172.02 (C=O).

Compound **13:** 3-Phenyl-1-tricyclo[3.3.1.1^{3,7}]decylmethyl 4-toluene sulfonate

Tosylate **13** was prepared in a similar manner as tosylate **8** by using 3-phenyl-1-adamantane methanol **12**[1] as starting material. Yield 90 %. Mp 105-107 ^{o}C; ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.44 (s, 2H, 4,10-H), 1.53 (s, 2H, 6-H), 1.56-1.65 (q, 2H, 2-H), 1.70-1.82 (q, 4H, 8,9-H), 2.10 (br. s, 2H, 5,7-H), 2.37 (s, 3H, CH3), 7.11-7.13 (m, 1H, , 4-H_{ar}), 7.22-7.28 (m, 6H, 2,3,5,6-H_{ar}, 3',5'-H_{ar}), 7.70-7.72 (d, 2H, AA'BB', J_{AB}=$J_{A'B'}$≈8.2Hz, $J_{AA'}$=$J_{BB'}$≈0Hz, 2',6'-H_{ar}).

Compound **14:** 3-Phenyl-1-tricyclo[3.3.1.1^{3,7}]decaneacetonitrile

Acetonitrile **14** was prepared from tosylate **13** in a similar manner as nitrile **9**. After purification by flash column chromatography, using a mixture of *n*-hexane:ether, 9:1, as eluent, nitrile **14** was obtained in 92% yield as a solid. Mp 64-60 ^{o}C; ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.55-1.66 (complex m, 6H, 4,6,10-H), 1.69 (s, 2H, 2-H), 1.76-1.85 (q, 4H, 8,9-H), 2.11 (s, 2H, α-H), 2.18 (br. s, 2H, 5,7-H), 7.13 (m, 1H, 4-H_{ar}), 7.23-7.28 (m, 4H, 2,3,5,6-H_{ar}); ^{13}C-NMR ($CDCl_3$), δ(ppm): 29.04 (5,7-C), 32.19 (α-C), 33.31 (3-C), 35.52 (2-C), 37.07 (1-C), 41.08 (4,10-C), 42.01 (8,9-C), 47.46 (6-C), 117.80 (C≡N), 124.88 (3,5-C_{ar}), 126.08 (4-C_{ar}), 128.93 (2,6-C_{ar}), 149.68 (1-C_{ar}).

Compound **15:** 3-Phenyl-1-tricyclo[3.3.1.1^{3,7}]decaneacetic acid

Carboxylic acid **15** was prepared by the alkaline hydrolysis of nitrile **14** under the same conditions used for the preparation of

acid **10**. The product was extracted with benzene. Yield 70% of a crude solid, which was used as such in the next step.

Compound **6k:** Ethyl 3-phenyl-1-tricyclo[3.3.1.1^{3,7}]decaneacetate

Ethylester **6k** was prepared from the crude acetic acid **15** via the carbonyl chloride **16** in a similar manner used for the preparation of ester **6h**. Purification of the obtained residue, by flash chromatography, was performed by gradient elution with mixtures of *n*-hexane:ether from 95:5 to 90:10 to give **6k**, as an oil in 65% yield. IR (film), ν(C=O): 1728.8 cm^{-1}; ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.67-1.20 (t, 3H, A_3X_2, J_{AX}≈7.12Hz, CH_3), 1.55-1.62 (m, 6H, 4,6,10-H), 1.69 (s, 2H, 2-H), 2.08 (s, 2H, α-H), 2.11-2.12 (m, 2H, 5,7-H), 4.02-4.07 (q, 2H, A_3X_2, J_{AX}≈7.12Hz, CH_2O), 7.09-7.13 (m, 1H, 4-H_{ar}), 7.22-7.28 (m, 4H, 2,3,5,6-H_{ar}); ^{13}C-NMR ($CDCl_3$), δ(ppm): 14.54 (CH_3), 29.34 (5,7-C), 33.84 (3-C), 35.98 (2-C), 37.12 (1-C), 41.62 (4,10-C), 42.43 (8,9-C), 48.10 (6-C), 48.82 (α-C), 60.06 (CH_2O), 124.99 (3,5-C_{ar}), 125.82 (4-C_{ar}), 128.28 (2,6-C_{ar}), 150.55 (1-C_{ar}), 171.80 (C=O).

Compound **18:** Ethyl (*E*)-2-tricyclo[3.3.1.1^{3,7}]decanepropenoate

To a stirred suspension of sodium hydride (600 mg, 25 mmol, 1 g dispersion 60% in paraffin oil, prewashed with *n*-pentane) in anhydrous THF (30 ml) triethyl phosphonoacetate (5.6 g, 25 mmol) in anhydrous THF (10 ml) was added dropwise, at 0 ^{o}C, under an argon atmosphere. Stirring was continued at 0 ^{o}C, under argon for 30 min and then a solution of 2-adamantanecarboxaldehyde (**17**) (2.75 g, 18.3 mmol) in dry benzene (30 ml), prepared from 2-adamantanone (4.3 g, 28.7 mmol) according to the Färcaşiu method[2], was added dropwise. After stirring for 60 min the reaction mixture was hydrolyzed under cooling by quenching with an ammonium chloride saturated solution. The organic solvents were removed in vacuo, water was added to the residue and the mixture was extracted with ether. The combined ethereal extracts were washed with water, dried over Na_2SO_4 and evaporated. The residue was purified by flash column chromatography, using a mixture of *n*-hexane:ether 4:1, as eluent, to give unsaturated ester **18** as an oil. Yield 2.5 g (38.5% from 2-adamantanone). IR (film), ν(C=O): 1717 cm^{-1}, ν(C=C): 1647 cm^{-1}; ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.22-1.25 (t, 3H, A_3X_2, J_{AX} ≈ 7.2 Hz, CH_3), 1.48-1.51 (br. d, 2H, 4,9- H_{eq}), 1.62-1.66 (br. d, 2H, 4,9-H_{ax}), 1.67 (s, 2H, 6-H), 1.72-1.85 (complex m, 8H, 1,3,5,7,8,10-H), 2.47-2.48 (~dd, 1H, $J_{2,\beta}$≈5Hz, $J_{2,\alpha}$≈2Hz, 2-H), 4.10-4.15 (q, 2H, A_3X_2, J_{AX} ≈7.2Hz, CH_2O), 5.74-5.79 (dd, 1H, $J_{\alpha,\beta}$ ≈16Hz, $J_{2,\alpha}$≈ 2Hz, α-H), 7.07-7.13 (dd, 1H, $J_{\alpha,\beta}$≈16 Hz, $J_{2,\beta}$≈5Hz, β-H).

Compound **20:** Ethyl (*E*)-3-cyclopentyl-1-tricyclo[3.3.1.1^{3,7}]decanepropenoate

To a stirred suspension of sodium hydride (480 mg, 20 mmol, 800 mg dispersion 60% in paraffin oil, prewashed with *n*-pentane) in anhydrous THF (30 ml) triethyl phosphonoacetate (4.2 g, 18.5 mmol) in THF anhydrous (10 ml) was added dropwise, at 0 ^{o}C, under an argon atmosphere. Stirring was continued at 0 ^{o}C and under argon for 30 min and then a solution of 3-cyclopentyl-1-adamatanecarboxaldehyde (**19**)[1] (4.25 g, 18.3 mmol) in anhydrous THF (30 ml) was added dropwise. After stirring for 60 min the reaction mixture was hydrolyzed under cooling by quenching with an ammonium chloride saturated solution. THF was evaporated in vacuo, water was added to the residue and the mixture was extracted with ether. The combined ethereal extracts were washed with water, dried over Na_2SO_4 and evaporated. Flash column chromatography of the obtained residue, by gradient elution with mixtures of *n*-hexane:ether from 98:2 to 95:5 gave 3.15 g of the desired ester **20**, as an oil, in 57% yield. IR ($CHCl_3$), ν(C=O): 1709-1682 cm^{-1}, ν(C=C): 1647.5 cm^{-1}; ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.26-1.30 (t, 3H, A_3X_2, J_{AX}≈7.1Hz, CH_3), 1.33 (s, 2H, 6-H), 1.42-1.56 (complex m, 17H, 4,8,9,10-H, 1,2,3,4,5-H_C), 1.61 (br. s, 2H, 2-H), 2.07 (br. s, 2H, 5,7-H), 4.15-4.20 (q, 2H, A_3X_2, J_{AX}≈7.1Hz, CH_2O), 5.64-5.68 (d, 1H, AX, J_{trans}≈16Hz, α-H), 6.80-6.84 (d, 1H, J_{trans}≈16Hz, β-H); ^{13}C-NMR ($CDCl_3$), δ(ppm): 14.43 (CH_3), 25.52 (3,4-C_C), 25.80 (2,5-C_C), 28.63 (5,7-C), 34.51 (3-C), 35.13 (1-C), 36.55 (2-C), 39.62 (4,10-C), 41.05 (8,9-C), 44.22 (6-C), 51.27 (1-C_C), 60.27 (CH_2O), 116.28 (β-C), 158.2 (α-C), 167.63 (C=O).

Compound **22:** Ethyl (*E*)-3-phenyl-1-tricyclo[3.3.1.1^{3,7}]decanepropenoate

Ester **22** was obtained by application of the Emmons-Horner reaction on 3-phenyl-1-adamantanecarboxaldehyde (**21**)[1], under the same conditions used for the preparations of ester **20**. The obtained residue was submitted to flash column chromatography, by using a mixture of n-hexane:ether, 4:1, as eluent, to afford ester **22** as a solid. Yield 90%. Mp 63-65 ^{o}C; IR ($CHCl_3$), ν(C=O): 1713 cm^{-1}, ν(C=C): 1648.5 cm^{-1}; ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.19-1.23 (t, 3H, A_3X_2, J_{AX}≈7.14Hz, CH_3), 1.55-1.70 (m, 4H, 4,10-H), 1.59 (s, 2H, 6-H), 1.70 (s, 2H, 2-H), 1.76-1.86 (q, 4H, 8,9-H), 2.15 (br. s, 2H, 5,7-H), 4.08-4.16(q, 2H, A_3X_2, J_{AX}≈7.14Hz, CH_2O), 5.63-5.67 (d, 1H, AX, J_{trans}≈16Hz, α-H), 6.79-6.83 (d, 1H, J_{trans} ≈ 16 Hz, β-H), 7.10-7.14 (m, 1H, 4-H_{ar}), 7.22-7.29 (m, 4H, 2,3,5,6-H_{ar}); ^{13}C-NMR ($CDCl_3$), δ(ppm): 14.39 (CH_3), 28.89 (5,7-C), 35.83 (2-C), 36.64 (3-C), 36.81 (1-C), 40.44 (4,10-C), 42.30 (8,9-C), 46.63 (6-C), 60.32 (CH_2O), 117.08 (α-C), 124.89 (3,5-C_{ar}), 125.95 (4-C_{ar}), 128.33 (2,6-C_{ar}), 150.14 (1-C_{ar}), 158.14 (β-C), 167.47 (C=O).

Compound **24:** Diethyl (*E,E*)-1,3-tricyclo[3.3.1.1^{3,7}]decanedipropenoate

Diester **24** was prepared by application of the Emmons-Horner reaction on the 1,3-adamatanedicarboxaldehyde (**23**)[3], by using 2 equiv. of sodium hydride and triethyl phosphonoacetate. Flash column chromatography of the obtained residue, by eluting with a mixture of *n*-hexane:ether 4:1, gave diester **24**, as an oil, in 55% yield. IR (CH_2Cl_2), ν(C=O): 1712 cm^{-1}, ν(C=C): 1648 cm^{-1}; ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.26-1.30 (t, 6H, A_3X_2, J_{AX}≈7Hz, 2xCH_3), 1.49 (br. s, 2H, 6-H), 1.54-1.65 (br. q, 8H, 4,8,9,10-H), 1.66 (br. s, 2H, 5,7-H), 2.14-2.15 (~d, 2H, J≈2.5Hz, 2-H), 4.14-4.20 (q, 4H, A_3X_2, J_{AX}≈7Hz, 2xCH_2O), 5.66-5.70 (d, 2H, AX, J_{trans}≈16Hz, 2xα-H), 6.79-6.83 (d, 2H, J_{trans}≈16Hz, 2xβ-H); ^{13}C-NMR ($CDCl_3$), δ(ppm): 14.40 (CH_3), 28.12 (2-C), 35.67 (5,7-C), 36.11 (1,3-C), 40.38 (4,8,9,10-C), 45.07 (6-C), 60.39 (CH_2O), 117.35 (α-C), 157.51 (β-C), 167.32 (C=O).

Compound **6f:** Ethyl 2-tricyclo[3.3.1.1^{3,7}]decanepropanoate

Ester **6f** was prepared by hydrogenation of the unsaturated ester **18**, under the same conditions used in the preparation of the ester **6e**. Colorless oily product. Yield almost quantitative. IR

(film), *ν*(C=O): 1724 cm^{-1}; ^{1}H-NMR ($CDCl_3$), *δ*(ppm): 1.23-1.27 (t, 3H, A_3X_2, J_{AX}≈7 Hz, CH_3), 1.48-1.51 (br. d, 2H, 4,9-H_{eq}), 1.55-1.60 (m, 1H, 5-H), 1.62-1.88 (very complex m, 14H, 1,2,3,6,7,8,10-H, 4,9-H_{ax}, *β*-H), 2.25-2.29 (t, 2H, A_2X_2, J_{AX}≈8Hz, *α*-H), 4.10-4.14 (q, 2H, A_3X_2, J_{AX}≈7Hz, CH_2O);^{13}C-NMR ($CDCl_3$), *δ*(ppm): 14.40 (CH_3), 28.01 (*α*-C), 28.15 (5-C), 28.37 (7-C), 31.67 (4,9-C), 31.78 (1,3-C), 32.84 (*β*-C), 38.47 (6-C), 39.28 (8,10-C), 44.20 (2-C), 174.64 (C=O).

The following saturated esters were prepared in a similar way from the corresponding unsaturated esters.

Compound **6i:** Ethyl 3-cyclopentyl-1-tricyclo[3.3.1.1^{3,7}]decanepropanoate

Prepared by hydrogenation of ester **20** in almost quantitative yield. Colorless oil. IR ($CHCl_3$), *ν*(C=O): 1722.3 cm^{-1}; ^{1}H-NMR ($CDCl_3$), *δ*(ppm): 1.16 (s, 2H, 6-H), 1.23-1.26 (t, 3H, A_3X_2, J_{AX}≈7.1Hz, CH_3), 1.35-1.47 (complex m, 19H, 4,8,9,10-H, 1,2,3,4,5-H_C, *β*-H), 1.56 (br. s, 2H, 2-H), 1.99 (s, 2H, 5,7-H), 2.22-2.27 (~t, 2H, *α*-H), 4.08-4.13 (q, 2H, A_3X_2, J_{AX}≈7.1Hz, CH_2O); ^{13}C-NMR ($CDCl_3$), *δ*(ppm): 14.22 (CH_3), 25.39 (3,4-C_C), 25.65 (2,5-C_C), 28.33 (*α*-C), 28.95 (5,7-C), 32.49 (3-C), 34.66 (1-C), 36.85 (2-C), 38.86 (*β*-C), 39.84 (4,10-C), 41.83 (8,9-C), 45.07 (6-C), 51.25 (1-C_C), 60.18 (CH_2O), 174.69 (C=O).

Compound **6l:** Ethyl 3-phenyl-1-tricyclo[3.3.1.1^{3,7}]decanepropanoate

Prepared by hydrogenation of ester **22** in almost quantitative yield. Colorless oil. IR (film), *ν*(C=O): 1734 cm^{-1}; ^{1}H-NMR ($CDCl_3$), *δ*(ppm): 1.16-1.19 (t, 3H, A_3X_2, J_{AX}≈7.12Hz, CH_3), 1.42-1.47 (m, 6H, 4,10-H, *β*-H), 1.53 (s, 2H, 6-H), 1.57-1.65 (br. q, 2H, 2-H), 1.73-1.82 (br. q, 4H, 8,9-H), 2.09-2.10 (~s, 2H, 5,7-H), 2.20-2.24 (m, 2H, *α*-H), 4.02-4.07 (q, 2H, A_3X_2, J_{AX}≈7.14Hz, CH_2O), 7.09-7.13 (m, 1H, 4-H_{ar}), 7.24-7.29 (m, 4H, 2,3,5,6-H_{ar}); ^{13}C-NMR ($CDCl_3$), *δ*(ppm): 14.37 (CH_3), 28.40 (*α*-C), 29.34 (5,7-C), 33.12 (3-C), 36.29 (2-C), 37.03 (1-C), 38.86 (*β*-C), 41.30 (4,10-C), 42.53 (8,9-C), 42.67 (6-C), 60.42 (CH_2O), 124.97 (3,5-C_{ar}), 125.79 (4-C_{ar}), 128.27 (2,6-C_{ar}), 150.79 (1-C_{ar}), 174.70 (C=O).

Compound **6o:** Diethyl 1,3-tricyclo[3.3.1.1^{3,7}]decanedipropanoate

Prepared by hydrogenation of ester **24** in almost quantitative yield. Colorless oil. IR (film), *ν*(C=O): 1732 cm^{-1}; ^{1}H-NMR ($CDCl_3$), *δ*(ppm): 1.14 (s, 2H, 6-H), 1.22-1.25 (t, 6H, A_3X_2, J_{AX}≈7.16Hz, 2xCH_3), 1.31-1.41 (complex m, 12H, 4,8,9,10-H, *β*-H), 1.55 (s, 2H, 2-H), 2.01(br. s, 2H, 5,7-H), 2.21-2.25 (~t, 4H, *α*-H), 4.07-4.13 (q, 4H, A_3X_2, J_{AX}≈7.16Hz, 2xCH_2O); ^{13}C-NMR ($CDCl_3$), *δ*(ppm): 14.35 (CH_3), 28.38 (*α*-C), 28.98 (5,7-C), 32.76 (1,3-C), 36.54 (2-C), 38.72 (*β*-C), 41.56 (4,8,9,10-C), 46.74 (6-C), 60.73 (CH_2O), 174.68 (C=O).

Compound **5a**: 1-Tricyclo[3.3.1.1^{3,7}]decanecarbohydrazide

Hydrazine hydrate (2 ml) was added to a solution of ethyl 1-adamantanecarboxylate (**6a**) (2 g, 9.6 mmol) in diethyleneglycol (8 ml) and the mixture was irradiated in a microwave apparatus (initially 900 W) for 2 min and then (500 W) at 210 °C for 90 min. After cooling the reaction mixture was poured into ice-water, the solid precipitate filterted, washed with water and dried over phosphorus pentoxide under vacuum. Yield 1.2 g (62%) of crystalline solid. Mp 156-157 °C (MeOH-H_2O); ^{1}H-NMR ($CDCl_3$), *δ*(ppm): 1.68-1.76 (br. q, 6H, 4,6,10-H), 1.85 (s, 6H, 2,8,9-H), 2.04 (br. s, 3H, 3,5,7,-H), 3.52 (br. s, 2H, NH_2), 6.98 (br. s, 1H, NH); ^{13}C-NMR ($CDCl_3$), *δ*(ppm):28.10 (3,5,7-C), 36.60 (4,6,10-C), 39.10 (2,8,9-C), 40.18 (1-C), 178.67 (C=O).

Compound **5b**: 1-Tricyclo[3.3.1.1^{3,7}]decaneacetohydrazide

Hydrazide **5b** was prepared from ester **6b** in a similar manner as the hydrazide **5a**. Yield 80%. Mp 108-110 °C (Et_2O); ^{1}H-NMR ($CDCl_3$), *δ*(ppm): 1.54 (s, 6H, 2,8,9-H), 1.54-1.65 (br. q, 6H, 4,6,10-H), 1.85 (s, 2H, *α*-H), 1.90 (s, 3H, 3,5,7,-H), 3.65 (br. s, 2H, NH_2), 6.92 (br. s, 1H, NH); ^{13}C-NMR ($CDCl_3$), *δ*(ppm): 28.70 (3,5,7-C), 32.87 (1-C), 36.80 (4,6,10-C), 42.70 (2,8,9-C), 49.45 (*α*-C), 171.99 (C=O); Anal. Calcd. for $C_{12}H_{20}N_2O$ (%):C, 69.19; H, 9.68; found (%): C, 69.33; H, 9.72.

Compound **5c**: 1-Tricyclo[3.3.1.1^{3,7}]decanepropanohydrazide

Method A. Hydrazide **5c** was prepared under the same conditions used in the preparation of hydrazide **5a**. Yield 70%.

Method B. Hydrazine hydrate (2 ml) was added to a solution of ethyl 1-adamantanepropionate **6c** (2.3 g, 9.6 mmol) in ethanol (10 ml) and the mixture was irradiated in a microwave apparatus (500 W) at 150 °C for 150 min. After cooling ethanol was removed in vacuo and water was added to the residue. The solid precipitate filtered, washed with water and dried over phosphorus pentoxide to yield 1.4 g (67%) of crystalline product. Mp 91-93 °C (MeOH-H_2O); ^{1}H-NMR ($CDCl_3$), *δ*(ppm): 1.37-1.40 (~t, 2H, A_2X_2, J_{AX}≈8Hz, *β*-H), 1.42 (s, 6H, 2,8,9-H), 1.56-1.68 (br. q, 6H, 4,6,10-H), 1.92 (s, 3H, 3,5,7,-H), 2.07-2.11 (t, 2H, A_2X_2, J_{AX}≈8Hz, *α*-H), 3.78 (br. s, 2H, NH_2), 7.26 (br. s, 1H, NH); ^{13}C-NMR ($CDCl_3$), *δ*(ppm): 28.25 (*α*-C), 28.63 (3,5,7-C), 32.06 (1-$C_{,rot}$), 32.27 (1-$C_{,rot}$) 37.12 (4,6,10-C), 36.69 (*β*-C), 41.27 (*β*-$C_{,rot}$), 42.13 (2,8,9-C), 174.97 (C=O); Anal. Calcd. for $C_{13}H_{22}N_2O$ (%):C, 70.23; H, 9.97; found (%): C, 70.44; H, 9.89.

Compound **5d**: 2-Tricyclo[3.3.1.1^{3,7}]decanecarbohydrazide

Compound **5d** was prepared from the ester **6d** by the same method used for the hydrazide **5a**. Yield 65%. Mp 132-134 °C (MeOH-H_2O); ^{1}H-NMR ($CDCl_3$), *δ*(ppm): 1.57-1.64 (~d, 2H, 4,9-H_{eq}), 1.72-1.96 (complex m, 10H, 4,9-H_{ax}, 5,6,7,8,10-H), 2.22 (s, 2H, 1,3-H), 2.47 (s, 1H, 2-H), 3.73 (br. s, 2H, NH_2), 7.20 (br. s, 1H, NH); ^{13}C-NMR ($CDCl_3$), *δ*(ppm): 27.40 (5-C), 27.59 (7-C), 29.91 (1,3-C), 33.36 (8,9-C), 37.43 (6-C), 38.41 (4,10-C), 49.00 (2-C), 175.34 (C=O); Anal. Calcd. for $C_{11}H_{18}N_2O$ (%):C, 68.01; H, 9.34; found (%): C, 68.04; H, 9.23.

Compound **5e**: 2-Tricyclo[3.3.1.1^{3,7}]decaneacetohydrazide

Hydrazide **5e**[40] was prepared from the ester **6e**[39] by the same method used for the hydrazide **5a**. Yield 89%. Mp 162-164 °C (MeOH-H_2O); Σ.τ.: 162-164 °C (MeOH-H_2O); ^{1}H-NMR ($CDCl_3$), *δ*(ppm): 1.54-1.57 (~d, 2H, 4,9-H_{eq}), 1.70-1.79 (complex m, 10H, 1,3,5,6,7,8,10-H), 1.82-1.85 (~q, 2H, 4,9-H_{ax}), 1.97-2.00 (m, 1H, 2-H), 2.88-2.90 (d, 2H, J≈7.6Hz, *α*-H), 4.63 (s, 2H, NH_2), 7.30 (s, 1H, NH); ^{13}C-NMR ($CDCl_3$), *δ*(ppm): 27.66 (2-C), 28.03 (5-C), 28.07 (7-C), 31.63 (α-$C_{,rot}$), 31.70 (α-$C_{,rot}$), 31.76 (1,3-$C_{,rot}$), 31.91 (1,3-$C_{,rot}$), 38.28 (6-C), 38.96 (4,8,9,10-C), 174.50 (C=O); Anal.

Calcd. for $C_{12}H_{20}N_2O$ (%):C, 69.19; H, 9.68; found (%): C, 68.85; H, 9.38.

Compound **5f**: 2-Tricyclo[3.3.1.1^{3,7}]decanepropanohydrazide

Hydrazide **5f** was prepared from the ester **6f** by the same method used for the hydrazide **5c**. By application of **Method A** (Yield 63%) or by application of **Method B** (Yield 58%). Mp 116-118 ^{o}C (MeOH-H_2O); ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.67-1.70 (~d, 2H, 4,9-H_{eq}), 1.77-1.85 (very complex m, 14H, 4.9-H_{ax}, 1,3,5,6,7,8,10-H, β-H), 2.11-2.15 (t, 2H, J≈7.6Hz, α-H), 2.69-2.73 (t, 1H, J≈8Hz, 2-H), 3.60-3.75 (br. s, 2H, NH_2), 7.10 (s, 1H, NH); ^{13}C-NMR ($CDCl_3$), δ(ppm): 28.12 (5-C), 28.34 (7-C), 28.53 (α-C), 31.66 (4,9-C), 31.75 (β-C), 38.43 (6-C), 39.26 (8,10-C), 44.27 (2-C), 174.49 (C=O); Anal. Calcd. for $C_{13}H_{22}N_2O$ (%):C, 70.23; H, 9.97; found (%): C, 70.58; H, 10.19.

Compound **5g**: 3-Cyclopentyl-1-tricyclo[3.3.1.1^{3,7}]decanecarbohydrazide

Hydrazide **5g** was prepared from the ester **6g** by the same method used for the hydrazide **5a**. Yield 75%. Mp 98-100 ^{o}C(MeOH-H_2O); ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.26-1.72 (very complex m, 21H, 2,4,6,8,9,10-H, 1,2,3,4,5-H_C), 2.03 (br. s, 2H, 5,7-H), 3.77 (very br. s, 2H, NH_2), 7.14 (br. s, 1H, NH); ^{13}C-NMR ($CDCl_3$), δ(ppm): 25.49 (3,4-C_C), 25.74 (2,5-C_C), 28.50 (5,7-C), 34.54 (3-C), 36.39 (2-C), 38.88 (4,10-C), 39.42 (8,9-C), 40.91 (1-C), 42.16 (6-C), 51.23 (1-C_C), 178.82 (C=O); Anal. Calcd. for $C_{16}H_{26}N_2O$ (%):C, 73.24; H, 9.99; found (%): C, 73.04; H, 10.16.

Compound **5h**: 3-Cyclopentyl-1-tricyclo[3.3.1.1^{3,7}]decaneacetohydrazide

Prepared from the ester **6h** by the same procedure used for the hydrazide **5a**. After cooling, the reaction mixture was diluted with cold water and the hydrazide **5h**, which was separated as an oil, taken up by extraction with chloroform. The combined chloroform extracts were washed with water, dried over Na_2SO_4 and evaporated to give hydrazide **5h** as an oily product. Yield 72%. ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.17-1.18 (m, 2H, 3,4-$H_{C,eq}$), 1.25 (s, 2H, 6-H), 1.29-1.50 (very complex m, 17H, 1,2,5-H_C, 3,4-$H_{C,ax}$, 2,4,8,9,10-H), 1.86 (s, 2H, α-H), 1.96 (br. s, 2H, 5,7-H), 3.30-4.25 (very br. s, 2H, NH_2), 6.82 (s, 1H, NH); ^{13}C-NMR ($CDCl_3$), δ(ppm): 25.52 (3,4-C_C), 25.70 (2,5-C_C), 29.01 (5,7-C), 33.50 (3-C), 34.94 (1-C), 36.57 (2-C), 39.55 (4,10-C), 42.44 (8,9-C), 45.93 (6-C), 49.36 (α-C), 51.29 (1-C_C), 171.99 (C=O).

Compound **5i**: 3-Cyclopentyl-1-tricyclo[3.3.1.1^{3,7}]decanepropanohydrazide

Prepared and isolated as an oily product in a similar manner as hydrazide **5h**. Yield 97%. ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.15 (σ, 2H, 3,4-$H_{C,eq}$), 1.23 (s, 2H, 6-H), 1.35-1.46 (very complex m, 17H, 1,2,5-H_C, 3,4-$H_{C,ax}$, ,4,8,9,10-H, β-H), 1.55 (s, 2H, 2-H), 1.99 (br. s, 2H, 5,7-H), 2.09-2.13 (t, 2H, α-H), 3.54 (br. s, 2H, NH_2), 7.08 (s, 1H, NH); ^{13}C-NMR ($CDCl_3$), δ(ppm): 25.54 (3,4-C_C), 25.78 (2,5-C_C), 28.72 (α-C), 29.06 (5,7-C), 32.71 (3-C), 34.81 (1-C), 36.95 (2-C), 39.63 (β-C), 39.93 (4,10-C), 42.00 (8,9-C), 45.32 (6-C), 51.37 (1-C_C), 174.98 (C=O).

Compound **5j**: 3-Phenyl-1-tricyclo[3.3.1.1^{3,7}]decanecarbohydrazide

Prepared from the ester **6j** by the same procedure used for the hydrazide **5a**. Yield 92%. Mp 91-93 ^{o}C (MeOH-H_2O); ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.70 (s, 2H, 6-H), 1.74-1.93 (m, 8H, 4,8,9,10-H), 2.00 (s, 2H, 2-H), 2.25 (br. s, 2H, 5,7-H), 3.59 (very br. s, 2H, NH2), 7.17-7.18 (m, 1H, 4-H_{ar}) 7.19-7.36 (m, 4H, 2,3,5,6-H_{ar}), 7.85 (very br. s, 1H, NH); ^{13}C-NMR ($CDCl_3$), δ(ppm): 28.81 (5,7-C), 35.71 (2-C), 36.63 (3-C), 38.32 (4,10-C), 41.23 (1-C), 42.16 (8,9-C), 44.47 (6-C), 124.86 (3,5-C_{ar}), 126.05 (4-C_{ar}), 128.35 (2,6-C_{ar}), 149.76 (1-C_{ar}), 178.26 (C=O); Anal. Calcd. for $C_{17}H_{22}N_2O$ (%):C, 75.52; H, 8.20; found (%): C, 75.84; H, 8.50.

Compound **5k**: 3-Phenyl-1-tricyclo[3.3.1.1^{3,7}]decaneacetohydrazide

Prepared from the ester **6k** by the same procedure used for the hydrazide **5a**. By diluting the reaction mixture with cold water compound **5k** was separated as an oil, which was taken up by extraction with chloroform and crystallized in the refrigerator. Yield 75%. Mp 113-115 ^{o}C (MeOH-H_2O); ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.64-1.68 (m, 6H, 4,6,10-H), 1.73 (s, 2H, 2-H), 1.85 (br. s, 4H, 8,9-H), 1.99 (s, 2H, α-H), 2.18 (br. s, 2H, 5,7-H), 3.91 (br. s, 2H, NH_2), 6.97 (s, 1H, NH), 7.16-7.20 (m, 1H, 4-H_{ar}), 7.29-7.36 (m, 4H, 2,3,5,6-H_{ar}); ^{13}C-NMR ($CDCl_3$), δ(ppm): 29.27 (5,7-C), 33.88 (3-C), 35.89 (2-C), 37.10 (1-C), 41.82 (4,10-C), 42.86 (8,9-C), 48.39 (6-C), 49.13 (α-C), 124.96 (3,5-C_{ar}), 125.85 (4-C_{ar}), 128.24 (2,6-C_{ar}), 150.34 (1-C_{ar}), 171.69 (C=O); Anal. Calcd. for $C_{18}H_{24}N_2O$ (%):C, 76.02; H, 8.51; found (%): C, 75.85; H, 8.46.

Compound **5l**: 3-Phenyl-1-tricyclo[3.3.1.1^{3,7}]decanepropanohydrazide

Prepared from the ester **6l** and taken up by chloroform extraction, as viscous oil. Yield 90%. ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.38-1.46 (m, 6H, 4,10-H, β-H), 1.52 (s, 2H, 6-H), 1.59-1.62 (m, 2H, 2-H), 1.76-1.79 (m, 4H, 8,9-H), 2.10 (m, 4H, 5,7-H, α-H), 3.52 (br. s, 2H, NH_2), 6.99 (br. s, 1H, NH), 7.11-7.13 (m, 1H, 4-H_{ar}), 7.25-7.26 (m, 4H, 2,3,5,6-H_{ar}); ^{13}C-NMR ($CDCl_3$), δ(ppm): 28.33 (α-C), 29.28 (5,7-C), 33.16 (3-C), 36.23 (2-C), 37.01 (1-C), 39.45 (β-C), 124.92 (3,5-C_{ar}), 125.81 (4-C_{ar}), 128.28 (2,6-C_{ar}), 150.68 (1-C_{ar}), 174.77 (C=O).

Compound **5m**: 1,3-Tricyclo[3.3.1.1^{3,7}]decanedicarbohydrazide

Dihydrazide **5m** was prepared from the diester **6m**, by the same procedure used for hydrazide **5a**. After dilution of the reaction mixture with water, the solution was evaporated in vacuo and the excess of hydrazine was removed azeotropically. Upon addition of ethanol to the residue dihydrazide **5m** was crystallized and collected by filtration. Yield 89%. Mp 206-208 ^{o}C (EtOH); ^{1}H-NMR (DMSO-d_6), δ(ppm): 1.57 (br. s, 2H, 6-H), 1.64-1.73 (br. q, 4,8,9,10-H), 1.78 (s, 2H, 5.7-H), 2.02 (s, 2H, 2-H), 4.13 (s, 4H, 2xNH_2), 8.72 (s, 2H, 2xNH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 27.71 (5,7-C), 35.19 (2-C), 37.82 (4,8,9,10-C), 39.52 (1,3-C), 40.13 (6-C), 175.83 (C=O); Anal. Calcd. for $C_{12}H_{20}N_4O_2$ (%):C, 57.12; H, 7.99; found (%): C, 56.84; H, 8.18.

Compound **5n**: 1,3-Tricyclo[3.3.1.1^{3,7}]decanediacetohydrazide

Hydrazine hydrate (2 ml) was added to a solution of the diester **6n** (1.34 g, 4.3 mmol) in ethanol (8 ml) and the mixture was stirred in an autoclave, as 150-160 °C, for 170 h. After cooling ethanol was removed in vacuo and water (40 ml) was added to the residue. Water was evaporated by heating under reduced pressure and the excess of hydrazine was removed azeotropically. The last traces of water were removed azeotropically with the aid of dry benzene. The viscous residue was treated with ether and crystallized on standing in the fridge to afford 1.1 g (91%) of a solid, Mp 121 °C, which was used as such for the next step. ^{1}H-NMR (DMSO-d_6), δ(ppm): 1.35-1.47 (complex m, 12H, 2,4,6,8,9,10-H), 1.77 (s, 4H, α-H), 1.94 (s, 2H, 5,7-H), 4.20 (br. s, 4H, 2xNH_2), 8.86 (br. s, 2H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.36 (5,7-C), 32.90 (1,3-C), 35.72 (2-C), 40.90 (6-C), 41.32 (4,8,9,10-C), 47.32 (α-C), 169.40 (C=O).

Compound **5o**: 1,3-Tricyclo[3.3.1.1^{3,7}]decanedipropanohydrazide

Dihydrazide **5o** was prepared from diester **6o** according to **Method B**, which was used for the preparation of hydrazide **5c**. After cooling, ethanol was removed under vacuum and water was added to the residue. The mixture was extracted with chloroform, the combined chloroform extracts washed with water, dried over Na_2SO_4 and evaporated to give dihydrazide **5o** as a sticky solid, which was used in the next step without further purification. Yield 57%. ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.11 (br. s, 2H, 6-H), 1.29-1.41 (complex m, 12H, 2,4,8,9,10-H, β-H), 2.00 (s, 2H, 5,7-H), 2.06-2.10 (t, 4H, A_2X_2, J_{AX}≈8Hz, α-H), 3.96 (br. s, 4H, 2xNH_2), 7.66 (m, 2H, 2xNH); ^{13}C-NMR ($CDCl_3$), δ(ppm): 28.32 (α-C), 28.90 (5,7-C), 32.84 (1,3-C), 36.48 (2-C), 39.35 (β-C), 41.62 (4,8,9,10-C), 46.54 (6-C), 174.87 (C=O).

Compound **5p**: 3-Ethoxycarbonylmethyl-1-tricyclo[3.3.1.1^{3,7}]decaneacetohydrazide

Hydrazine hydrate (2 ml) was added to a solution of the diester **6n** (1 g, 3.2 mmol) in ethanol (8 ml) and the mixture was irradiated under stirring in a microwave apparatus at 150 °C for 150 min. After cooling, ethanol was removed in vacuo and water was added to the residue. The mixture was extracted with DCM, the combined DCM layers were washed with water and dried over Na_2SO_4. The aqueous phase was re-extracted with ethyl acetate, the combined ethyl acetate layers washed with water and dried over Na_2SO_4. The dried organic phases were combined and evaporated. The residue was submitted to flash column chromatography eluting with a mixture of DCM:MeOH, 9:1, to give hydrazidoester **5p** (396 mg, 42%), as a viscous liquid, which was used as such in the next step. IR ($CDCl_3$), ν(C=O): 1719-1717 cm^{-1} (ester), ν(C=O): 1699-1685 cm^{-1} (hydrazide); ^{1}H-NMR ($CDCl_3$), δ(ppm): 1.22-1.26 (t, 3H, A_3X_2, J_{AX}≈7.1Hz, CH_3), 1.45 (br. s, 2H, 6-H), 1.51 (br. s, 2H, 2-H), 1.45-1.56 (br. m, 8H, 4,8,9,10-H), 1.93 (s, 2H, α-H), 2.04 (s, 2H, 5,7-H), 2.07 (s, 2H, α'-H), 3.71 (br. s, 2H, NH_2), 4.06-4.12 (q, 2H, A_3X_2, J_{AX}≈7.1Hz, CH_2O), 7.08 (very br. s, 1H, NH); ^{13}C-NMR ($CDCl_3$), δ(ppm): 14.49 (CH_3), 28.94 (5,7-C), 33.60 (1,3-C), 35.84 (2-C), 41.50 (4,10-C), 41.75 (8,9-C), 47.50 (6-C), 48.50 (α-C), 48.92 (α'-C), 60.07 (CH_2O), 171.80 (C=O).

Compound **4a**: N^2-(5-Nitro-2-furanemethylene)-1-tricyclo[3.3.1.1^{3,7}]decanocarbohydrazide

To a solution of 5-nitro-2-furaldehyde (0.4 g, 2.84 mmol) in ethanol (4 ml) a solution of 1-adamantanecarbohydrazide (**5a**) (0.5 g, 2.58 mmol) in ethanol (4 ml) was added. The mixture was stirred under argon in darkness for 12 h. The precipitated solid was obtained by filtration, washed with a small amount of ethanol and dried to give **4a** (625 mg, 77%), as a yellow solid. Mp 268 °C (dec) (EtOH); ^{1}H-NMR (DMSO-d_6), δ(ppm): 1.68 (br. s, 6H, 4,6,10-H), 1.85 (br. s, 6H, 2,8,9-H), 2.00 (br. s, 3H, 3,5,7-H), 7.17 (s, 1H, 3-H_f), 7.77 (s, 1H, 4-H_f), 8.36 (s, 1H, CH=N), 11.25 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 27.47 (3,5,7-C), 38.09 (4,6,10-C), 38.25 (2,9-C), 40.62 (1-C), 114.68 (3,4-C_f), 134.42 (C=N), 152.12 (2,5-C_f), 173.71 (C=O); Anal. Calcd. for $C_{16}H_{19}N_3O_4$ (%): C, 60.55; H, 6.04; N, 13.24; found (%) C, 60.21; H, 6.08; N, 13.50.

The following carbohydrazones **4b-p** were prepared in a similar manner.

Compound **4b**: N^2-(5-Nitro-2-furanmethylene)-1-tricyclo[3.3.1.1^{3,7}]decanoacetohydrazide

Yield 78%. Mp 218-220 °C ($CHCl_3$)

Mixture of two conformers **A** and **B**. **A**/**B**≈1.63

Conformer A

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.55-1.63 (very br. s, 12H, 2,4,6,8,9,10-H), 1.88 (s, 3H, 3,5,7-H), 1.95 (s, 2H, α-H), 7.15 (s, 1H, 3-H_f), 7.74 (s, 1H, 4-H_f), 8.10 (s, 1H, CH=N), 11.63 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.00 (3,5,7-C), 32.80 (1-C), 36.32 (4,6,10-C), 42.03 (2,8,9-C), 48.02 (α-C), 113.76 (3-C_f), 114.89 (4-C_f), 133.62 (C=N), 151.98 (2,5-C_f), 166.96 (C=O).

Conformer B

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.55-1.63 (very br. s, 12H, 2,4,6,8,9,10-H), 1.88 (s, 3H, 3,5,7-H), 2.38 (s, 2H, α-H), 7.13 (s, 1H, 3-H_f), 7.73 (s, 1H, 4-H_f), 7.86 (s, 1H, CH=N), 11.50 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.06 (3,5,7-C), 32.88 (1-C), 36.32 (4,6,10-C), 42.20 (2,8,9-C), 44.47 (α-C), 113.76 (3-C_f), 114.63 (4-C_f), 130.04 (C=N), 151.98 (2,5-C_f), 172.70 (C=O).

Anal. Calcd. for $C_{17}H_{21}N_3O_4$ (%): C, 61.62; H, 6.39; N, 12.68; found (%) C, 61.41; H, 6.52; N, 12.48.

Compound **4c**: N^2-(5-Nitro-2-furanmethylene)-1-tricyclo[3.3.1.1^{3,7}]decanopropanohydrazide

Yield 60%. Mp 237 °C (EtOH)

Mixture of two conformers **A** and **B**. **A**/**B**≈1.25

Conformer A

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.35 (br. m, 2H, β-H), 1.47 (br. s, 6H, 2,8,9-H), 1.57-1.68 (br. q, 4,6,10-H), 1.92 (br. s, 3H, 3,5,7-H), 2.50-2.54 (~t, 2H, A_2X_2, J_{AX}≈7.8Hz, α-H), 7.15-7.16 (~d, 1H, J≈3.2Hz, 3-H_f), 7.76 (s, 1H, 4-H_f), 7.90 (CH=N), 11.60 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 25.80 (α-C), 28.01 (3,5,7-C), 31.72 (1-C), 36.59 (4,6,10-C), 38.53 (β-C), 41.58 (2,8,9-C), 114.18 (3-C_f), 114.69 (4-C_f), 130.38 (C=N), 151.85 (2,5-C_f), 175.60 (C=O).

Conformer B

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.35 (br. m, 2H, β-H), 1.44 (br. s, 6H, 2,8,9-H), 1.57-1.68 (br. q, 4,6,10-H), 1.92 (br. s, 3H, 3,5,7-H), 2.16-2.20 (~t, 2H, A_2X_2, J_{AX}≈7.8Hz, α-H), 7.19-7.20 (~d, 1H, J≈3.2Hz, 3-H_f), 7.76 (s, 1H, 4-H_f), 8.12 (CH=N), 11.74 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.01 (3,5,7-C), 28.13 (α-C), 31.67 (1-C), 36.59 (4,6,10-C), 38.97 (β-C), 41.58 (2,8,9-C), 114.18 (3-C_f), 114.69 (4-C_f), 133.58 (C=N), 151.85 (2,5-C_f), 169.95 (C=O).

Anal. Calcd. for $C_{18}H_{23}N_3O_4$ (%): C, 62.59; H, 6.71; N, 12.17; found (%) C, 62.88; H, 6.44; N, 12.02.

Compound **4d**: N^2-(5-Nitro-2-furanmethylene)-2-tricyclo[3.3.1.1^{3,7}]decanocarbohydrazide

Yield 71%. Mp 281-282 ^{o}C (dec) ($CHCl_3$)

Mixture of two conformers **A** and **B**. **A**/**B**≈2.4-2.5

Conformer A

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.52-1.55 (~d, 2H, 4,9-H_{eq}), 1.68 (br. s, 2H, 6-H), 1.73-1.87 (br. m, 6H, 5,7,8,10-H), 2.02-2.05 (d, 2H, 4,9-H_{ax}), 2.18 (br. s, 2H, 1,3-H), 2.51 (s, 1H, 2-H), 7.16 (~s, 1H, 3-H_f), 7.75 (~s, 1H, 4-H_f), 8.16 (s, 1H, CH=N), 11.59 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 26.79 (5-C), 27.07 (7-C), 29.36 (1,3-C), 32.50 (4,9-C), 36.96 (6-C), 37.88 (8,10-C), 48.01 (2-C), 114.69 (3,4-C_f), 133.27 (C=N), 152.14 (2,5-C_f), 171.03 (C=O).

Conformer B

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.52-1.55 (~d, 2H, 4,9-H_{eq}), 1.68 (br. s, 2H, 6-H), 1.73-1.87 (br. m, 6H, 5,7,8,10-H), 2.11-2.14 (d, 2H, 4,9-H_{ax}), 2.18 (br. s, 2H, 1,3-H), 3.24 (s, 1H, 2-H), 7.10 (~s, 1H, 3-H_f), 7.86 (~s, 1H, 4-H_f), 8.32 (s, 1H, CH=N), 11.44 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 26.79 (5-C), 27.36 (7-C), 29.36 (1,3-C), 32.50 (4,9-C), 36.96 (6-C), 38.19 (8,10-C), 45.37 (2-C), 114.48 (3,4-C_f), 129.76 (C=N), 151.66 (2,5-C_f), 171.03 (C=O).

Anal. Calcd. for $C_{16}H_{19}N_3O_4$ (%): C, 60.55; H, 6.04; N, 13.24; found (%) C, 60.88; H, 5.95; N, 13.42.

Compound **4e**: N^2-(5-Nitro-2-furanmethylene)-2-tricyclo[3.3.1.1^{3,7}]decanoacetohydrazide

Yield 85%. Mp 231-233 ^{o}C ($CHCl_3$)

Mixture of two conformers **A** and **B**. **A**/**B**≈1.05-1.09

Conformer A

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.49-1.52 (br. d, 2H, 4,9-H_{eq}), 1.66-1.85 (very complex m, 10H, 1,3,5,6,7,8,10-H), 1.85-1.95 (m, 2H, 4,9-H_{ax}), 2.17-2.21 (t, 1H, A_2X, J_{AX}≈7.5 Hz, 2-H), 2.38-2.40 (d, 2H, A_3X_2, J_{AX}≈7.5 Hz, α-H), 7.19-7.20 (d, 1H, J≈4 Hz, 3-H_f), 7.77-7.78 (d, 1H, J≈4Hz, 4-H_f), 8.14 (s, 1H, CH=N), 11.77 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 27.31 (5,7-C), 31.10 (4,9-C), 31.25 (6-C), 31.42 (1,3-C), 37.85 (α-C), 38.39 (8-C), 38.49 (10-C), 40.66 (2-C), 114.02 (4-C_f), 114.93 (3-C_f), 133.79 (C=N), 151.89 (2,5-C_f), 174.59 (C=O).

Conformer B

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.49-1.52 (br. d, 2H, 4,9-H_{eq}), 1.66-1.85 (very complex m, 10H, 1,3,5,6,7,8,10-H), 1.89-1.95 (m, 2H, 4,9-H_{ax}), 2.17-2.21 (t, 1H, A_2X, J_{AX}≈7.5Hz, 2-H), 2.75-2.77 (d, 2H, A_3X_2, J_{AX}≈7.5Hz, α-H), 7.19-7.19 (d, 1H, J≈4Hz, 3-H_f), 7.76-7.77 (d, 1H, J≈4Hz, 4-H_f), 7.91 (s, 1H, CH=N), 11.67 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 27.31 (5,7-C), 31.10 (4,9-C), 31.14 (1,3-C), 31.25 (6-C), 34.91 (α-C), 37.71 (10-C), 38.39 (8-C), 40.91 (2-C), 113.92 (4-C_f), 114.63 (3-C_f), 130.46 (C=N), 151.89 (2,5-C_f), 168.78 (C=O).

Anal. Calcd. for $C_{17}H_{21}N_3O_4$ (%): C, 61.62; H, 6.39; N, 12.68; found (%) C, 61.49; H, 6.75; N, 12.48.

Compound **4f**: N^2-(5-Nitro-2-furanmethylene)-2-tricyclo[3.3.1.1^{3,7}]decanopropanohydrazide

Yield 58%. Mp 205-206 ^{o}C (dec) ($CHCl_3$)

Mixture of two conformers **A** and **B**. **A**/**B**≈1.3-1.4

Conformer A

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.43-1.49 (br. d, 2H, 4,9-H_{eq}), 1.68-1.82 (very complex m, 12H, 1,3,5,6,7,10-H, β-H), 2.22 (t, 1H, J≈7.5Hz, 2-H), 2.52 (m, 2H, α-H), 7.15-7.17 (~d, 1H, J≈4Hz, 3-H_f), 7.75-7.77 (~d, 1H, J≈4Hz, 4-H_f), 7.91 (s, 1H, CH=N), 11.62 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 27.19 (α-C), 27.38 (5,7-C), 30.08 (β-C), 31.05 (4,9-C), 31.07 (1,3-C), 37.83 (6-C), 38.60 (8,10-C), 43.40 (2-C), 114.88 (4-C_f), 119.74 (3-C_f), 130.55 (C=N), 150.10 (2,5-C_f), 175.03 (C=O).

Conformer B

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.43-1.49 (br. d, 2H, 4,9-H_{eq}), 1.68-1.82 (very complex m, 12H, 1,3,5,6,7,10-H, β-H), 2.22 (t, 1H, J≈7.5Hz, 2-H), 2.52 (m, 2H, α-H), 7.45-7.47 (~d, 1H, J≈4Hz, 3-H_f), 7.81-7.83 (~d, 1H, J≈4Hz, 4-H_f), 8.13 (s, 1H, CH=N), 11.73 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 27.38 (5,7-C), 27.70 (α-C), 31.05 (4,9-C), 31.07 (1,3-C), 32.56 (β-C), 37.83 (6-C), 38.60 (8,10-C), 43.40 (2-C), 114.02 (3-C_f), 114.28 (4-C_f), 133.60 (C=N), 150.10 (2,5-C_f), 169.48 (C=O).

Anal. Calcd. for $C_{18}H_{23}N_3O_4$ (%): C, 62.59; H, 6.71; N, 12.17; found (%) C, 62.25; H, 6.99; N, 12.01.

Compound **4g**: 3-Cyclopentyl-N^2-(5-nitro-2-furanmethylene)-1-tricyclo[3.3.1.1^{3,7}]decanocarbohydrazide

Yield 65%. Mp >240 ^{o}C ($CHCl_3$); ^{1}H-NMR (DMSO-d_6), δ(ppm): 1.27 (br. s, 2H, 3,4-$H_{c,ax}$), 1.39-1.58 (complex m, 11H, 1,2,5-Hc, 3,4-$H_{c,eq}$, 8,9-H), 1.57-1.64 (m, 6H, 2,6-H), 1.78 (br. q, 4H, 4,10-H), 2.06 (br. s, 2H, 5,7-H), 7.18 (s, 1H, 3-H_f), 7.77 (s, 1H, 4-H_f), 8.37 (s, 1H, CH=N), 11.24 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 24.97 (3,4-C_C), 25.20 (2,5-C_C), 27.88 (5,7-C), 34.02 (3-C), 35.73 (2-C), 37.93 (4,10-C), 38.68 (8,9-C), 40.72 (1-C), 41.07 (6-C), 50.71 (1-C_C), 114.56 (3-C_f), 114.67 (4-C_f), 134.46 (C=N), 152.13 (2,5-C_f), 173.79 (C=O); Anal. Calcd. for $C_{21}H_{27}N_3O_4$ (%): C, 65.43; H, 7.06; N, 10.90; found (%) C, 65.28; H, 6.91; N, 11.02.

Compound **4h**: 3-Cyclopentyl-N^2-(5-nitro-2-furanmethylene)-1-tricyclo[3.5.1.1^{3,7}]decanoacetohydrazide

Yield 62%. Mp 181-183 ^{o}C ($CHCl_3$)

Mixture of two conformers **A** and **B**. **A**/**B**≈1.5

Conformer A

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.20 (s, 2H, 6-H), 1.30-1.51 (complex m, 19H, 1,2,3,4,5-H_C, 2,4,8,9,10-H), 1.96 (s, 2H, 5,7-H), 1.98 (s, 2H, α-H), 7.18 (s, 1H, 3-H_f), 7.76 (s, 1H, 4-H_f), 8.12 (s, 1H, CH=N), 11.65 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 24.98 (3,4-C_C), 25.20 (2,5-C_C), 28.32 (5,7-C), 33.42 (3-C), 34.33 (1-C), 36.12 (2-C), 39.13 (4,10-C), 41.82 (8,9-C), 45.15 (6-C), 48.47 (α-C), 50.73 (1-C_C), 113.93 (3-C_f), 114.58 (4-C_f), 133.62 (C=N), 151.96 (2-C_f), 152.02 (5-C_f), 167.11 (C=O).

Conformer B

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.20 (s, 2H, 6-H), 1.30-1.51 (complex m, 19H, 1,2,3,4,5-H_C, 2,4,8,9,10-H), 1.96 (s, 2H, 5,7-H), 2.42 (s, 2H, α-H), 7.18 (s, 1H, 3-H_f), 7.76 (s, 1H, 4-H_f), 7.87 (s, 1H, CH=N), 11.62 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 24.98 (3,4-C_C), 25.20 (2,5-C_C), 28.32 (5,7-C), 33.53 (3-C), 34.33 (1-C), 36.12 (2-C), 39.23 (4,10-C), 41.49 (8,9-C), 44.58 (α-C), 45.32 (6-C), 50.67 (1-C_C), 114.98 (3,4-C_f), 129.96 (C=N), 151.96 (2-C_f), 152.02 (5-C_f), 172.96 (C=O).

Anal. Calcd. for $C_{22}H_{29}N_3O_4$ (%): C, 66.14; H, 7.32; N, 10.53; found (%) C, 65.92; H, 7.45; N, 10.41.

Compound **4i**: 3-Cyclopentyl-*N*^{2}-(5-nitro-2-furanmethylene)-1-tricyclo[3.3.1.1^{3,7}]decanepropanohydrazide

Yield 62%. Mp 181-183 ^{o}C ($CHCl_3$)

Mixture of two conformers **A** and **B**. **A**/**B**≈1.26

Conformer A

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.16-1.77 (~d, 2H, 6-H), 1.23 (br. m, 2H, 3,4-$H_{C,eq}$), 1.33-1.77 (complex m, 17H, 3,4-$H_{C,ax}$, 1,2,5-H_C, 4,8,9,10-H, β-H), 1.54 (br. s, 2H, 2-H), 1.98 (br. s, 2H, 5,7-H), 2.17-2.21 (~t, 2H, α-H), 7.76-7.77 (~t, 2H, 3,4-H_f), 8.12 (s, 1H, CH=N), 11.73 (s, 1H. NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 24.97 (3,4-C_C), 25.33 (2,5-C_C), 25.80 (α-C), 28.34 (5,7-C), 32.02 (3-C), 34.20 (1-C), 36.34 (2-C), 38.37 (β-C), 39.39 (4,10-C), 41.37 (8,9-C), 44.53 (6-C), 50.73 (1-C_C), 114.18 (3-C_f), 114.52 (4-C_f), 133.54 (C=N), 151.76 (2,5-C_f), 169.94 (C=O).

Conformer B

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.16-1.77 (~d, 2H, 6-H), 1.23 (br. m, 2H, 3,4-$H_{C,eq}$), 1.33-1.45 (complex m, 17H, 3,4-$H_{C,ax}$, 1,2,5-H_C, 4,8,9,10-H, β-H), 1.54 (br. s, 2H, 2-H), 1.98 (br. s, 2H, 5,7-H), 2.52-2.55 (~t, 2H, α-H), 7.15-7.15 (d, 1H, 3-H_f), 7.19-7.21 (d, 1H, 4-H_f), 7.91 (s, 1H, CH=N), 11.60 (s, 1H. NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 24.97 (3,4-C_C), 25.33 (2,5-C_C), 28.27 (α-C), 28.34 (5,7-C), 32.29 (3-C), 34.20 (1-C), 36.34 (2-C), 38.37 (β-C), 39.37 (4,10-C), 41.37 (8,9-C), 44.63 (6-C), 50.73 (1-C_C), 114.43 (3,4-C_f), 130.37 (C=N), 151.76 (2,5-C_f), 175.60 (C=O).

Anal. Calcd. for $C_{23}H_{31}N_3O_4$ (%): C, 66.80; H, 7.56; N, 10.16; found (%) C, 66.65; H, 7.79; N, 10.22.

Compound **4j**: 3-Phenyl-*N*^{2}-(5-nitro-2-furanmethylene)-1-tricyclo[3.3.1.1^{3,7}]decanocarbohydrazide

Yield 75%. Mp 228 ^{o}C (dec) ($CHCl_3$); ^{1}H-NMR (DMSO-d_6), δ(ppm): 1.71 (s, 2H, 6-H), 1.86-1.89 (m, 8H, 4,8,9,10-H), 1.95 (s, 2H, 2-H), 2.20 (br. s, 2H, 5,7-H), 7.18 (s, 2H, 4-H_{ar}, 3-H_f), 7.32-7.40 (dm, 4H, 2,3,5,6-H_{ar}), 7.77 (s, 1H, 4-H_f), 8.37 (s, 1H, CH=N), 11.32 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.25 (5,7-C), 35.01 (2-C), 36.40 (3-C), 37.39 (4,10-C), 41.13 (1-C), 41.47 (8,9-C), 43.39 (6-C), 114.60 (3,4-C_f), 124.68 (3,5-C_{ar}), 125.71 (4-C_{ar}), 128.11 (2,6-C_{ar}), 134.47 (C=N), 149.40 (2-C_f), 151.73 (1-C_{ar}), 152.11 (5-C_f), 173.45 (C=O); Anal. Calcd. for $C_{22}H_{23}N_3O_4$ (%): C, 67.16; H, 5.89; N, 10.68; found (%) C, 69.94; H, 6.02; N, 10.77.

Compound **4k**: 3-Phenyl-*N*^{2}-(5-nitro-2-furanmethylene)-1-tricyclo[3.3.1.1^{3,7}]decanoacetohydrazide

Yield 97%. Mp 191 ^{o}C ($CHCl_3$)

Mixture of two conformers **A** and **B**. **A**/**B**≈1.5-1.6

Conformer A

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.61 (br. s, 6H, 4,6,10-H), 1.69 (s, 2H, 2-H), 1.76 (br. s, 4H, 8,9-H), 2.07 (s, 2H, α-H), 2.10 (br. s, 2H, 5,7-H), 7.16-7.31 (very complex m, 6H, 2,3,4,5,6,-H_{ar}, 3-H_f), 7.75 (s, 1H, 4-H_f), 8.12 (s, 1H, CH=N), 11.70 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.70 (5,7-C), 33.81 (3-C), 35.41 (2-C), 37.08 (1-C), 41.48 (4,10-C), 41.78 (8,9-C), 47.78 (6-C), 48.35 (α-C), 114.65 (3-C_f), 115.02 (4-C_f), 124.62 (3,5-C_{ar}), 125.54 (4-Car), 128.11 (2,6-C_{ar}), 133.74 (C=N), 150.21 (2,5-C_f), 151.91 (1-C_{ar}), 166.90 (C=O).

Conformer B

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.61 (br. s, 6H, 4,6,10-H), 1.69 (s, 2H, 2-H), 1.76 (br. s, 4H, 8,9-H), 2.10 (br. s, 2H, 5,7-H), 2.48 (s, 2H, α-H), 7.16-7.31 (very complex m, 6H, 2,3,4,5,6,-H_{ar}, 3-H_f), 7.77 (s, 1H, 4-H_f), 7.88 (s, 1H, CH=N), 11.66 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.70 (5,7-C), 33.81 (3-C), 35.41 (2-C), 36.56 (1-C), 41.30 (4,10-C), 41.78 (8,9-C), 44.37 (α-C), 47.94 (6-C), 113.89 (3-C_f), 114.82 (4-C_f), 124.62 (3,5-C_{ar}), 125.54 (4-Car), 128.04 (2,6-C_{ar}), 130.11 (C=N), 150.21 (2,5-C_f), 151.91 (1-C_{ar}), 172.68 (C=O).

Anal. Calcd. for $C_{23}H_{25}N_3O_4$ (%): C, 67.79; H, 6.18; N, 10.31; found (%) C, 67.56; H, 6.42; N, 10.33.

Compound **4l**: 3-Phenyl-*N*^{2}-(5-nitro-2-furanmethylene)-1-tricyclo[3.3.1.1^{3,7}]decanepropanohydrazide

Yield 72%. Mp 188-190 ^{o}C ($CHCl_3$)

Mixture of two conformers **A** and **B**. **A**/**B**≈1.15

Conformer A

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.41-1.49 (m, 6H, 4,10-H, β-H), 1.54 (br. s, 2H, 6-H), 1.58-1.64 (~q, 2H, 2-H), 1.72-1.80 (~q, 4H, 8,9-H), 2.10 (br. s, 2H, 5,7-H), 2.53-2.57 (t, 2H, A_2X_2, J_{AX}≈8Hz, α-H), 7.11-7.18 (complex m, 2H, 4-H_{ar}, 3-H_f), 7.24-7.33 (complex m, 4H, 2,3,5,6-H_{ar}), 7.72-7.73 (d, 1H, J≈3.5Hz, 4-H_f), 7.88 (s, 1H, CH=N), 11.61 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.27 (α-C), 28.71 (5,7-C), 32.80 (3-C), 35.66 (2-C), 36.51 (1-C), 40.35 (β-

C), 40.70 (4,10-C), 42.10 (8,9-C), 47.40 (6-C), 114.70 (3,4-C_f), 124.70 (3,5-C_{ar}), 125.53 (4-C_{ar}), 128.08 (2,6-C_{ar}), 130.46 (C=N), 150.44 (1-C_{ar}), 151.84 (2,5-C_f), 175.58 (C=O).

Conformer B

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.41-1.49 (m, 6H, 4,10-H, β-H), 1.54 (br. s, 2H, 6-H), 1.58-1.67 (~q, 2H, 2-H), 1.72-1.80 ~q, 4H, 8,9-H), 2.10 (br. s, 2H, 5,7-H), 2.19-2.23 (t, 2H, A_2X_2, J_{AX}≈8Hz, α-H), 7.11-7.18 (complex m, 2H, 4-H_{ar}, 3-H_f), 7.24-7.33 (complex m, 4H, 2,3,5,6-H_{ar}), 7.74-7.75 (d, 1H, J≈3.5Hz, 4-H_f), 8.09 (s, 1H, CH=N), 11.73 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 25.85 (α-C), 28.71 (5,7-C), 32.80 (3-C), 35.66 (2-C), 36.51 (1-C), 40.35 (β-C), 40.70 (4,10-C), 42.10 (8,9-C), 47.40 (6-C), 114.21 (3-C_f), 114.94 (4-C_f), 124.70 (3,5-C_{ar}), 125.53 (4-C_{ar}), 128.08 (2,6-C_{ar}), 133.61 (C=N), 150.44 (1-C_{ar}), 151.84 (2,5-C_f), 169.97 (C=O).

Anal. Calcd. for $C_{24}H_{27}N_3O_4$ (%): C, 68.39; H, 6.46; N, 9.97; found (%) C, 68.57; H, 6.38; N, 9.82.

Compound **4m**: *N*^{2},*N*$^{2\prime}$-Bis(5-nitro-2-furanmethylene)-1,3-tricyclo[3.3.1.1^{3,7}]decanedicarbohydrazide

Yield 75%. Mp 230 ^{o}C (dec) ($CHCl_3$); ^{1}H-NMR (DMSO-d_6), δ(ppm): 1.69 (br. s, 2H, 6-H), 1.83-1.90 (br. q, 8H, 4,8,9,10-H), 2.02 (s, 2H, 2-H), 2.18 (s, 2H, 5,7-H), 7.20-7.21 (~d, 2H, J≈3.9 Hz, 3-H_f), 7.78-7.79 (~d, 2H, J≈3.9 Hz, 4-H_f), 8.35 (s, 2H, CH=N), 11.38 (s, 2H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 27.56 (5,7-C), 34.81 (2-C), 37.16 (4,8,9,10-C), 39.52 (1,3-C), 40.46 (6-C), 114.67 (3-C_f), 114.83 (4-C_f), 134.66 (C=N), 151.81 (2-C_f), 151.99(5-C_f), 173.11 (C=O); Anal. Calcd. for $C_{22}H_{22}N_6O_8$ (%): C, 53.22; H, 4.47; N, 16.93; found (%) C, 52.88; H, 4.62; N, 16.73.

Compound **4n**: *N*^{2},*N*$^{2\prime}$-Bis(5-nitro-2-furanmethylene)-1,3-tricyclo[3.3.1.1^{3,7}]decanediacetohydrazide

Yield 92%. Mp 158-160 ^{o}C ($CHCl_3$)

It seems to be a mixture of three conformers **A** and **B** and **C**. **A**/**B**/**C**≈10/9/3

Conformer A

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.47 (br. s, 2H, 6-H), 1.49-1.54 (m, 8H, 4,6,8,9,10-H), 1.56 (br. s, 2H, 2-H), 1.99 (~s, 4H, α-H), 2.02 (s, 2H, 5,7-H), 7.46-7.47 (~d, 2H, J≈4Hz, 3-H_f), 7.82-7.83 (~d, 2H, J≈4Hz, 4-H_f), 8.73 (s, 2H, CH=N), 11.62 (s, 2H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.37 (5,7-C), 33.55 (1,3-C), 35.57 (2-C), 41.35 (8,9,10-C), 46.90 (6-C), 48.17 (α-C), 119.74 (3-C_f), 130.08 (4-C_f), 150.10 (C=N), 151.38 (2-Cf), 151.92 (5-C_f), 166.90 (C=O).

Conformer B

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.47 (br. s, 2H, 6-H), 1.49-1.54 (m, 8H, 4,6,8,9,10-H), 1.56 (br. s, 2H, 2-H), 2.02 (s, 2H, 5,7-H), 2.45 (s, 4H, α-H), 7.16-7.14 (~d, 2H, J≈4Hz, 3-H_f), 7.75-7.76 (~d, 2H, J≈4 Hz, 4-H_f), 8.11 (s, 2H, CH=N), 11.65 (s, 2H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.37 (5,7-C), 33.55 (1,3-C), 35.58 (2-C), 41.46 (8,9,10-C), 44.05 (α-C), 46.90 (6-C), 114.03 (3-C_f), 114.61 (4-C_f), 133.58 (C=N), 151.38 (2-Cf), 151.92 (5-C_f), 172.61 (C=O).

Conformer C

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.47 (br. s, 2H, 6-H), 1.49-1.54 (m, 8H, 4,6,8,9,10-H), 1.56 (br. s, 2H, 2-H), 2.02 (s, 2H, 5,7-H), 2.43 (s, 4H, α-H), 7.14-7.15 (~d, 2H, J≈4Hz, 3-H_f), 7.72-7.73 (~, 2H, J≈4Hz, 4-H_f), 8.09 (s, 2H, CH=N), 11.57 (s, 2H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.37 (5,7-C), 33.55 (1,3-C), 35.58 (2-C), 41.46 (8,9,10-C), 46.58 (α-C), 46.90 (6-C), 113.75 (3-C_f), 114.78 (4-C_f), 133.58 (C=N), 152.77 (2,5-Cf), 172.61 (C=O).

Anal. Calcd. for $C_{24}H_{26}N_6O_8$ (%): C, 54.96; H, 5.00; N, 16.02; found (%) C, 55.12; H, 5.23; N, 15.82.

Compound **4o**: *N*^{2},*N*$^{2\prime}$-Bis(5-nitro-2-furanmethylene)-1,3-tricyclo[3.3.1.1^{3,7}]decanedipropanohydrazide

Yield 58 %. Mp 218 ^{o}C (dec) ($CHCl_3$)

It seems to be probably a mixture of four conformers **A** and **B**, **C** and **D**. **A**+**B**/**C**+**D**≈1.2

Conformers A+B

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.17 (s, 2H, 6-H), 1.37 (m, 8H, 4,8,9,10-H), 1.41 (~s, 4H, β-H), 1.53 (br. s, 2H, 2-H), 2.00 (br. s, 2H, 5,7-H), 2.50 (br. t, 2H, α-H), 7.17 (br. s, 2H, 3-H_f), 7.74 (br. s, 2H, 4-H_f), 7.86,7.88 (d, 2H, CH=N), 11.59 (s, 2H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 25.95 (α-C), 28.42 (5,7-C), 32.51 (1,3-C), 36.06 (2-C), 39.76 (β-C), 41.11 (4,8,9,10-C), 46.33 (6-C), 114.12 (3-C_f), 114.74, 114.91 (4-C_f), 130.40,130.49 (C=N), 151.83 (2,5-C_f), 175.57,175.63 (C=O).

Conformers C+D

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.20 (s, 2H, 6-H), 1.37 (m, 8H, 4,8,9,10-H), 1.41 (~s, 4H, β-H), 1.53 (br. s, 2H, 2-H), 2.00 (br. s, 2H, 5,7-H), 2.17 (br. t, 2H, α-H), 7.10, 7.28 (s, 2H, 3-H_f), 7.69-7.70, 7.80-7.81 (2xd, 2H, 4-H_f), 8.08 (~d, 2H, CH=N), 11.73 (d, 2H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.27 (α-C), 28.42 (5,7-C), 32.44 (1,3-C), 36.06 (2-C), 40.21 (β-C), 40.78 (4,8,9,10-C), 46.33 (6-C), 114.69 (3-C_f), 114.74, 114.95 (4-C_f), 133.58 (C=N), 151.91 (2,5-C_f), 169.92 (C=O).

Anal. Calcd. for $C_{26}H_{30}N_6O_8$ (%): C, 56.51; H, 5.47; N, 15.21; found (%) C, 56.38; H, 5.69; N, 14.99.

Compound **4p**: 3-Ethoxycarbonylmethyl-*N*^{2}-(5-nitro-2-furanmethylene)-1-tricyclo[3.3.1.1^{3,7}]decaneacetohydrazide

Yield 48%. Mp 124 ^{o}C ($CHCl_3$)

Mixture of two conformers **A** and **B**. **A**/**B**≈1.5

Conformer A

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.11-1.15 (t, 3H, A_3X_2, J_{AX}≈7Hz, CH_3), 1.41-1.54 (very complex m, 12H, 2,4,6,8,9,10-H), 1.95 (br. s, 2H, 5,7-H), 1.98 (s, 2H, CH_2CON), 2.02 (s, 2H, CH_2CO_2), 3.97-4.02 (q, 2H, A_3X_2, J_{AX}≈7Hz, CH_2O), 7.16 (3-H_f), 7.75 (4-H_f), 8.11 (CH=N), 11.66 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 14.65 (CH_3), 28.72 (5,7-C), 33.39 (3-C), 33.95 (1-C), 35.90 (2-C), 41.47 (4,10-C), 41.65 (8,9-C), 44.49 (6-C), 46.99 ($\underline{C}H_2CO_2$), 48.15 ($\underline{C}H_2CON$), 59.80 (CH_2O), 114.36 (3-C_f), 115.25 (4-Cf), 134.10 (CH=N), 152.38 (2,5-C_f), 171.10 (C=O, hydrazide), 173.08 (C=O, ester).

Conformer B

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.10-1.13 (t, 3H, A_3X_2, J_{AX}≈7Hz, CH_3), 1.41-1.54 (very complex m, 12H, 2,4,6,8,9,10-H), 1.95 (br. s, 2H, 5,7-H), 2.00 (s, 2H, CH_2CON), 2.42 (s, 2H, CH_2CO_2), 3.94-4.00 (q, 2H, A_3X_2, J_{AX}≈7Hz, CH_2O), 7.17 (3-H_f), 7.76 (4-H_f), 7.87 (CH=N), 11.63 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 14.65 (CH_3), 28.72 (5,7-C), 33.39 (3-C), 33.95 (1-C), 35.90 (2-C), 41.19 (4,10-C), 41.81 (8,9-C), 44.49 (6-C), 47.19 ($\underline{C}H_2CO_2$), 48.59 ($\underline{C}H_2CON$), 59.80 (CH_2O), 113.47 (3-C_f), 115.43 (4-Cf), 130.52 (CH=N), 152.06 (2,5-C_f), 167.37 (C=O, hydrazide), 180.67 (C=O, ester).

Anal. Calcd. for $C_{21}H_{27}N_3O_6$ (%): C, 60.42; H, 6.52; N, 10.07; found (%) C, 60.22; H, 6.68; N, 9.78

Compound **4q**: N^2-(2-furanmethylene)-1-tricyclo[3.3.1.1^{3,7}]decanecarbohydrazide

Carbohydrazone **4q** was prepared by reacting 2-furaldehyde with 1-adamantane carbohydrazide **5a**, in ethanol under the same conditions used for the preparation of hydrazine **4a**. Ethanol was removed under vacuum and the last traces of water, formed during the reaction, were removed azeotropically with the aid of dry benzene. The residue was recrystallized from a mixture of ether-*n*-pentane. Yield 53%. Mp 237 ^{o}C (dec); ^{1}H-NMR (DMSO-d_6), δ(ppm): 1.65-1.72 (m, 6H, 4,6,10-H), 1.85 (∼s, 6H, 2,8,9-H), 1.99 (s, 3H, 3,5,7-H), 6.59-6.60 (q, 1H, $J_{3,4}$≈3.2Hz, $J_{4,5}$≈3.12Hz, 4-H_f) 6.83-6.84 (d, 1H, $J_{3,4}$≈3.2Hz, $J_{4,5}$≈3.12Hz, 3-H_f), 7.80 (s, 1H, 5-H_f), 8.30 (s, 1H, CH=N), 10.75 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 27.55 (3,5,7-C), 36.02 (4,6,10-C), 38.29 (2,8,9-C), 39.70 (1-C), 112.03 (4-C_f), 112.57 (3-C_f), 136.03 (C=N), 144.82 (5-C_f), 149.70 (2-C_f), 173.19 (C=O); Anal. Calcd. for $C_{16}H_{20}N_2O_2$ (%): C, 70.56; H, 7.40; N, 10.29; found (%) C, 70.49; H, 7.45; N, 10.49.

Compound **4r**: N^2-(2-furanmethylene)-1-tricyclo[3.3.1.1^{3,7}]decaneacetohydrazide

Hydrazone **4r** was prepared in a similar procedure used in the preparation of hydrazine **4q**. Yield 66%. Mp 175-177 ^{o}C (Et_2O-*n*-pentane)

Mixture of two conformers **A** and **B**. **A**/**B**≈1.48-1.5

Conformer A

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.53-1.63 (m, 12H, 2,4,6,8,9,10-H), 1.89 (∼s, 5H, 3,5,7-H, α-H), 6.56 (∼s, 1H, 4-H_f), 6.81 (∼d, 1H, 3-H_f), 7.76 (s, 1H, 5-H_f), 8.02 (s, 1H, CH=N), 11.16 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.04 (3,5,7-C), 32.69 (1-C), 36.39 (4,6,10-C), 42.09 (2,8,9-C), 48.69 (α-C), 112.08 (3-C_f), 112.90 (4-C_f), 135.64 (C=N), 144.66 (2-C_f), 144.89 (5-C_f), 166.36 (C=O).

Conformer B

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.53-1.63 (m, 12H, 2,4,6,8,9,10-H), 1.89 (s, 3H, 3,5,7-H), 2.34 (s,2H, α-H), 6.56 (∼s, 1H, 4-H_f), 6.76 (∼d, 1H, 3-H_f), 7.76 (s, 1H, 5-H_f), 7.80 (s, 1H, CH=N), 11.11 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.04 (3,5,7-C), 32.87 (1-C), 36.39 (4,6,10-C), 42.27 (2,8,9-C), 44.57 (α-C), 112.00 (3-C_f), 112.49 (4-C_f), 132.15 (C=N), 144.89 (5-C_f), 149.53 (2-C_f), 172.10 (C=O).

Anal. Calcd. for $C_{17}H_{22}N_2O_2$ (%): C, 71.30; H, 7.74; N, 9.78; found (%) C, 71.46; H, 7.52; N, 9.80

Compound **4s**: N^2-(2-furanmethylene)-1-tricyclo[3.3.1.1^{3,7}]decanepropanohydrazide

Yield 60%. Mp 190 ^{o}C (Et_2O-*n*-pentane)

Mixture of two conformers **A** and **B**. **A**/**B**≈1.3

Conformer A

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.53-1.63 (m, 12H, 2,4,6,8,9,10-H), 1.89 (∼s, 5H, 3,5,7-H, α-H), 6.56 (∼s, 1H, 4-H_f), 6.81 (∼d, 1H, 3-H_f), 7.76 (s, 1H, 5-H_f), 8.02 (s, 1H, CH=N), 11.16 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.04 (3,5,7-C), 32.69 (1-C), 36.39 (4,6,10-C), 42.09 (2,8,9-C), 48.69 (α-C), 112.08 (3-C_f), 112.90 (4-C_f), 135.64 (C=N), 144.66 (2-C_f), 144.89 (5-C_f), 166.36 (C=O).

Conformer B

^{1}H-NMR (DMSO-d_6), δ(ppm): 1.53-1.63 (m, 12H, 2,4,6,8,9,10-H), 1.89 (s, 3H, 3,5,7-H), 2.34 (s,2H, α-H), 6.56 (∼s, 1H, 4-H_f), 6.76 (∼d, 1H, 3-H_f), 7.76 (s, 1H, 5-H_f), 7.80 (s, 1H, CH=N), 11.11 (s, 1H, NH); ^{13}C-NMR (DMSO-d_6), δ(ppm): 28.04 (3,5,7-C), 32.87 (1-C), 36.39 (4,6,10-C), 42.27 (2,8,9-C), 44.57 (α-C), 112.00 (3-C_f), 112.49 (4-C_f), 132.15 (C=N), 144.89 (5-C_f), 149.53 (2-C_f), 172.10 (C=O).

Anal. Calcd. for $C_{18}H_{24}N_2O_2$ (%): C, 71.97; H, 8.05; N, 9.33; found (%) C, 71.79; H, 8.16; N, 9.35

Molecular Mechanics Methods

Coordinate scan using Molecular Mechanics

All the structures were built in Maestro 9.3 (Schrodinger Suite 2012.1). The minimization was done using MacroModel 9.9, the OPLS_2005 force field and the Polak-Ribiere conjugate gradient to a convergence threshold of 0.001 kJ mol^{-1} Å^{-1}. The coordinate scan of the three dihedrals was also done in MacroModel 9.9 using two different force fields (OPLS_2005 and MMFFs) for the calculations. A dielectric constant of 47.24 was used to simulate DMSO as a solvent. The dihedral was incremented by 1 degree in each step.

Relaxed coordinate scan using quantum mechanical methods

The Jaguar 8.0 module of Schrödinger was used to perform a relaxed coordinate scan for all compounds to confirm the results given using molecular mechanics. The dihedral was incremented by 5 degrees in each step. The DFT level of theory and the B3LYP/6-31G** hybrid functional were used for the optimization of the structures during the coordinate scan for all the intermediate steps. The calculations were performed *in vacuo* and in solvent using the Poisson−Boltzmann finite element solvent model, with DMSO as the solvent.

Calculation of the octanol/water partition coefficient.

Starting geometries for all the structures were generated using the LigPrep 3.4 [4] protocol implemented in the Maestro 10.2 modeling package. The structures were minimized in Macromodel 10.8 using the OPLS3 force field with a constant dielectric of 1.0. The octanol/water partition coefficient ($QPlogP_0/w$) predicted by Qikprop 4.4 [5] was considered as the LogP value.

Cytotoxic activity against rat skeletal myoblast L6 cells

Cytotoxicity against mammalian cells was assessed using microtitre plates. Briefly, L6 cells (a rat skeletal muscle line) were seeded at 1 x 104 ml-1 in 200 µl of growth medium containing 7 different compound concentrations in a range previously established to encompass both the IC_{50} and IC_{90} values. The plates were incubated for 6 days at 37 °C and 20 µl Alamar Blue (Biosource UK Ltd) was then added to each well. After an additional 8 hours incubation, the fluorescence was determined using a Gemini fluorescent plate reader (Molecular Devices). Inhibition of growth was calculated by comparison with control values and IC_{50} and IC_{90} values were determined in triplicate using linear regression analysis.

***Trypanosoma brucei* culturing and drug testing**

Bloodstream form *T. brucei* (strain 427) were cultured at 37 °C in modified Iscove's medium. Trypanocidal activity was assessed by growing parasites in microtitre plates in the presence of various drug concentrations. Parasites were seeded at 0.25 x 10-5 ml-1 in 200 µl of growth medium containing 7 different compound concentrations in a range previously established to encompass both the IC_{50} and IC_{90} values. The plates were incubated for 48 hours at 37 °C and 20 µl Alamar Blue (Biosource UK Ltd) was then added to each well. After an additional overnight incubation, the fluorescence was determined using a Gemini fluorescent plate reader (Molecular Devices). Inhibition of growth was calculated by comparison with control values and IC_{50} and IC_{90} values were determined in triplicate using linear regression analysis.

***Trypanosoma cruzi* culturing and drug testing**

T. cruzi epimastigotes (strain CL Brener) were cultured at 28 °C in supplemented RPMI-1640 medium. Trypanocidal activity was assessed by growing parasites in microtitre plates in the presence of various drug concentrations. Parasites were seeded at 2.5 x 10-5 ml-1 in 200 µl of growth medium containing 7 different compound concentrations in a range previously established to encompass both the IC_{50} and IC_{90} values. The plates were incubated for 4 days at 28 °C and 20 µl Alamar Blue (Biosource UK Ltd) was then added to each well. After an additional 3 days incubation, the fluorescence was determined using a Gemini fluorescent plate reader (Molecular Devices). Inhibition of growth was calculated by comparison with control values and IC_{50} and IC_{90} values were determined in triplicate using linear regression analysis.

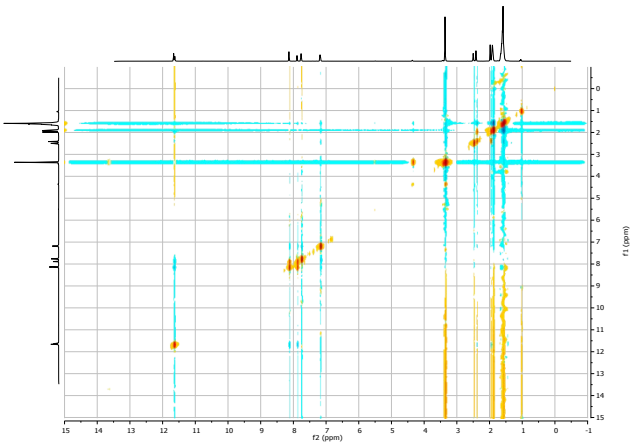

Fig. 2: 1D NOEDY spectrum of the major and minor conformers of compound **4b.**

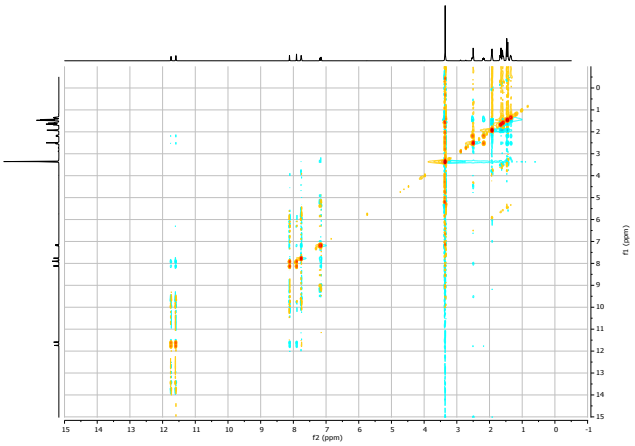

Fig. 3: 1D NOEDY spectrum of the major and minor conformers of compound **4c.**

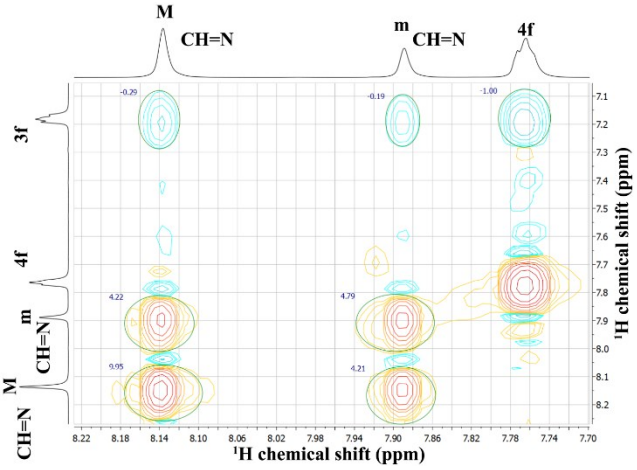

Fig. 4: 2D NOESY spectrum of the major and minor conformers of compound **4b**. Integrations of the circled peaks were obtained using TOPSPIN 3.1.

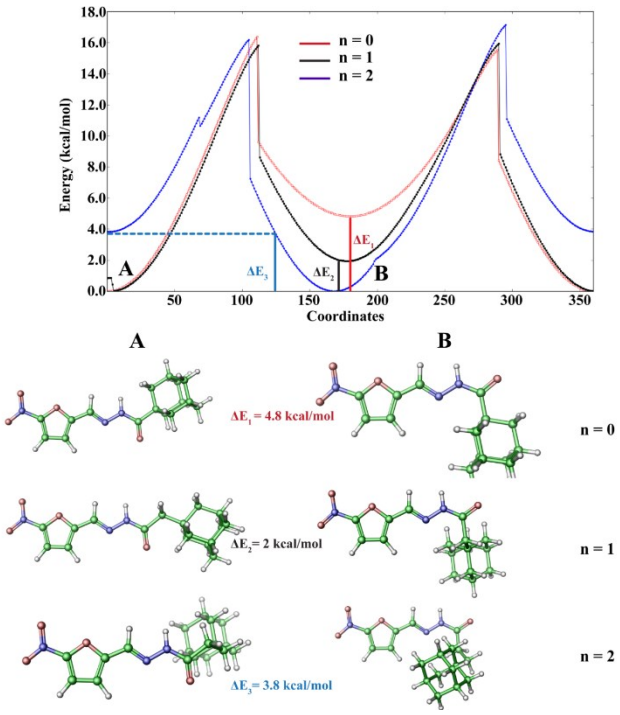

Fig. 5: Grid scans around the amide bond of adamantane derivatives **4a**, **4b**, **4c**.

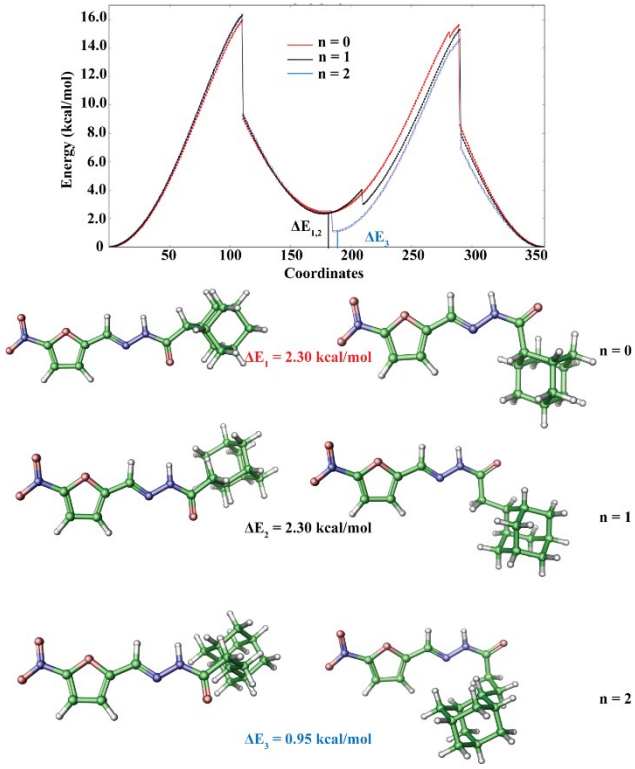

Fig. 6: Grid scans around the amide bond of the adamantane derivatives **4d**, **4f** and **4e**.

Table VII Lipophilicity of the most important compounds of the study

Cmpd	QPlogP0/w
4a	2.840
4b	3.176
4c	3.448
4d	2.778
4e	3.146
4f	3.418
4g	4.194
4h	4.514
4i	4.810
4j	4.423
4k	4.755
4l	5.037

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