Fused Triazines Via a Tandem Wittig/Ring Closure Strategy: Synthesis of Pyrazolo[5,1-*c*]-1,2,4-triazines and 1,2,4-triazolo[5,1-*c*]-1,2,4-triazines

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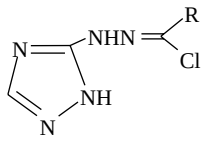
A new method for the synthesis of azolo[5,1-*c*]-1,2,4-triazine ring systems is reported by the sequential formation of triazine ring based on the regioselective formation of the azophosphoranes from hydrazonyl chlorides, followed by the intermolecular Wittig reaction with carboxylic acid chlorides, phenylisocyanate, and carbon disulfide.

Keywords: Hydrazonylchloride; Azophosphorane; Azolotriazines.

The considerable biological and medicinal activities of pyrazolotriazines and triazolotriazines, as adenine analogues, antagonists, antischistosomal and antitumor agents[1-4] have stimulated recent interest in the synthesis of these ring systems. In our previous work,[5] we reported the synthesis of azolo- and triazolo[5,1-*c*][1,2,4]triazepines starting from azophosphoranes **3** derived from hydrazonyl chlorides **1** and **2**. It has been our interest to examine this method and its application to the synthesis of new azolo[5,1-*c*]-1,2,4-triazine derivatives. **3** reacted smoothly with acyl chlorides in anhydrous toluene in the presence of triethylamine (2.0 equiv.) at room temperature to provide the expected azolotriazine derivatives **5**. Here, the tertiary amine seems to be essential for the reaction as demonstrated. In fact, without Et_3N only 9% product was obtained.



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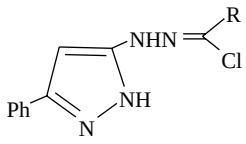

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The azophosphoranes **3** apparently act as Wittig reagents and azolyl chlorides **4** are assumed to be the key intermediate for the consecutive reaction. The driving force in this reaction is the formation of the stable phosphine oxide, which is an inevitable product in the Wittig reactions. Subsequent cyclization of intermediate **4** by intramolecular nucleophilic attack of the ring nitrogen to the newly generated electrophilic carbon center would give the corresponding azolo-[5,1-*c*]-1,2,4-triazine derivatives **5a-h** (Scheme I).

Azophosphoranes **3** reacted also with phenylisocyanate in dry toluene at reflux temperature to give azolotriazines **7a-d**. The yield of the isolated products was higher than 85%. We believe that the mechanism of this conversion involves an initial Wittig reaction between the azophosphoranes **3** and the phenylisocyanate to furnish a carboimide **6** as highly reactive intermediate, which easily undergoes a heterocyclic ring closure to give the fused triazines **7**.

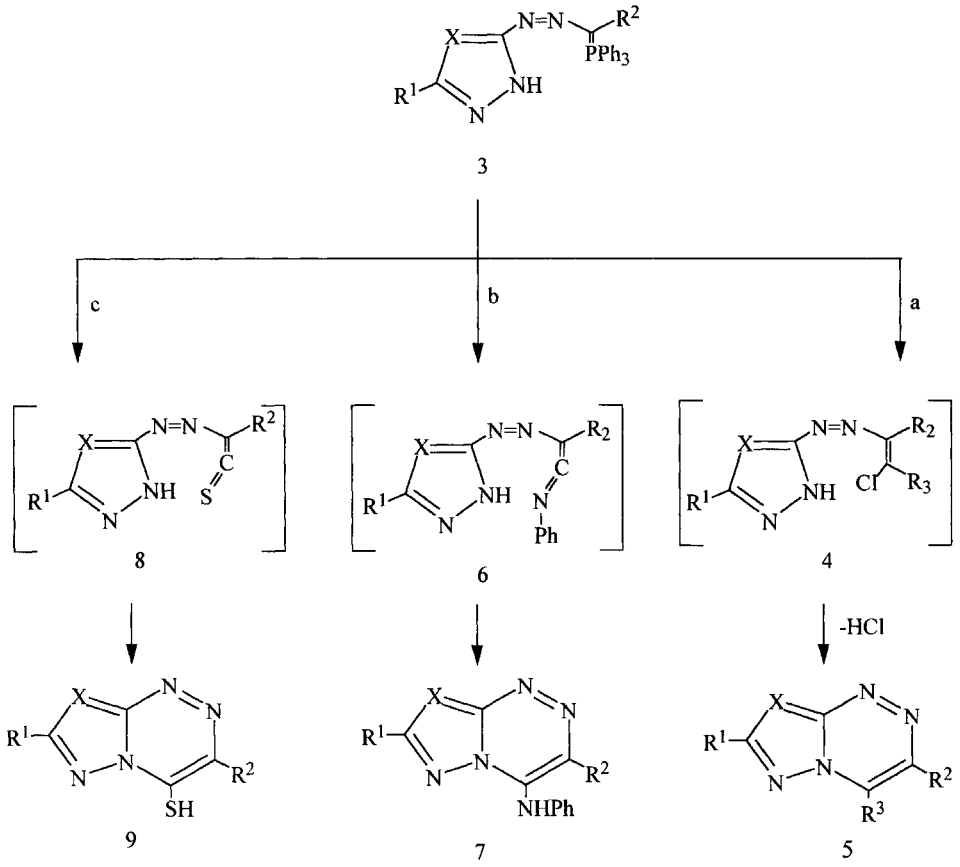
The treatment of azophosphoranes **3** with excess of carbon disulfide in dry methylene chloride at room temperature, under nitrogen atmosphere, leads to the formation of azolotriazines **9a-d**. The possible intermediate **8** in the conversion **8** $\rightarrow$ **9** is shown in Scheme I. Isolation of this intermediate was not successful.

We were interested to see if this procedure might be extended to include other heterocycles, thus hydrazonyl chlorides **11a**, **b**[6] were treated with triphenylphosphine and triethylamine under the same reaction conditions. No expected azophosphoranes **12a**, **b** were found and pyrazolo[3,4-*c*]pyrazole derivatives **13a**, **b** were isolated instead (Scheme II). This result showed that azophosphorane **12** is a highly reactive and not isolable compound which, upon formation, undergoes immediate intramolecular cyclization through the nucleophillic attack of the -N=N- group on the carbonyl group of compound **12**, and subsequent ethanol elimination would give **13**.

The -N=N- group in these compounds, in fact, has shown a great propensity to react as a nucleophile,[7] and particularly is able to proceed intramolecular nucleophilic attack on electron-deficient carbon atoms located at the ortho position on the aromatic ring.[8,9]

In conclusion, the synthetic method presented herein provides a new and general route to fused triazines with variable substituents from readily available precursors with high yield under mild reaction conditions. Further studies along

Scheme I Reagents and conditions: a) R^3COCl/Et_3N/toluene, 6-12 h; b) PhNCO/toluene, reflux; c) CS_2/CH_3CN/nitrogen gas, 6 h. r.t.



7,9 a, R^1=Ph,R^2=COOEt, X=CH
b, R^1=Ph, R^2=$COCH_3$, X=CH
c, R^1=H, R^2=COOEt, X=N
d, R^1 =H, R^2=$COCH_3$, X=N

5 a, R^1=Ph, R^2 =COOEt, R^3 =Ph, X=CH
b, R^1=Ph, R^2 =COOEt, R^3 =CH_3, X=CH
c, R^1=Ph, R^2 =COCH3,R^3 =Ph, X=CH
d, R^1=Ph, R^2 =$COCH_3$, R^3 =CH_3, X=CH
e, R^1=H, R^2 =COOEt,R^3 =Ph, X=N
f, R^1=H, R^2 =COOEt,R^3 =CH_3, X=N
g, R^1=H, R^2 =$COCH_3$,R^3 =Ph, X=N
h, R^1= H, R^2 = $COCH_3$, R^3=CH_3, X=N

these lines are in progress at our laboratory.

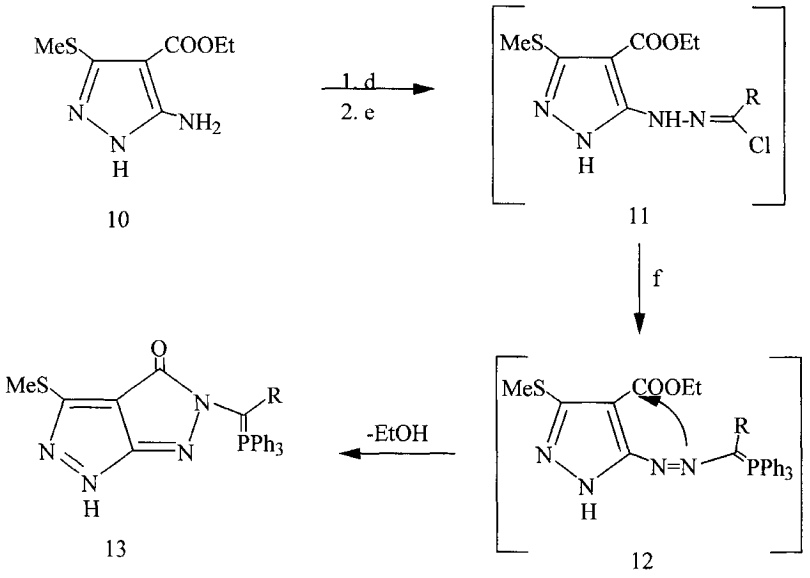
EXPERIMENTAL

Melting points were determined on a Gallenkamp electrothermal melting point apparatus and are uncorrected. IR spectra were recorded as potassium bromide pellets on a Pye Unicame SP 3-300 and FT IR 8101 PC Shimadzu infrared spectrophotometer. The ^{1}H NMR spectra were obtained in deuterated dimethyl sulfoxide on a Varian Gemini 200 NMR spectrometer using tetramethylsilane (TMS) as an internal reference. Mass spectra were recorded on a GC MS-QP 1000 EX Shimadzu mass spectrometer at 70 eV. Elemental analysis was carried out at the Microanalytical Center of Cairo University, Giza, Egypt.

General procedure for the preparation of 4-[aryl (or alkyl)-7-phenyl-3-substituted pyrazolo[5,1-*c*][1,2,4]triazines 5a-d and 4-[aryl (or alkyl)-3-substituted-1,2,4-triazolo[5,1-*c*][1,2,4]triazines 5e-h

To a solution of azophosphorane **3a-d** (0.60 mmol) in dry toluene (10 mL) was added dropwise appropriate acyl chloride (benzoyl chloride or acetyl chloride) (1.20 mmol,

Scheme II Reagents and conditions: d) $NaNO_2$/HCl, 0 °C; e) $CH_3CO(Cl)R/CH_3COONa$, EtOH, 0 °C; f) PPh_3/Et_3N/CH_3CN, 10 min. r.t.



2.0 equiv.) and Et_3N (0.17 mL, 1.0 mmol, 2.0 equiv.), and the reaction mixture was stirred at room temperature for 12 h. After completion of the reaction (TLC control), the precipitate was filtered off, and the filtrate was washed with water, dried, and crystallized from acetonitrile to give **5a-h**.

Ethyl 4,7-diphenylpyrazolo[5,1-*c*][1,2,4]triazine-3-carboxylate 5a

Yield 86%; yellow crystals; mp 290 °C; IR (KBr) 1725, 1650, 1610, 1585 cm^{-1}; ^1H NMR (DMSO-d_6) δ 1.43 (s, 3H, CH_3), 3.91 (q, 2H, CH_2), 6.30 (s, 1H, pyrazole), 6.92-7.31 (m, 10H, Ar-H); ^{13}C NMR (DMSO-d_6, 50 MHz) δ 168.1 (CO_2Et), 131.5 (C-3), 129.1 (C-4), 134.9 (C-7), 128.4 (C-8), 134.0 (C-9), 113.8, 125.6, 128.3, 130.0, 130.2, 128.2, 134.1, 131.6 (aromatic carbons), 62.3 (OCH_2), 14.7 (CH_3); MS (EI) *m/z* (rel. intensity) 344 (3, M^+), 288 (7), 261 (3), 221 (3), 183 (10), 128 (20), 101 (27), 77 (10). 51 (69). Anal. Calcd. for $C_{20}H_{16}N_4O_2$ (344.36) C, 69.75; H, 4.68; N, 16.27. Found C, 69.59; H, 4.49; N, 16.27%.

Ethyl 4-methyl-7-phenylpyrazolo[5,1-*c*][1,2,4]triazine-3-carboxylate 5b*

Yield 75%; pale yellow solid; mp 185 °C; IR (KBr) 2957, 1715, 1630, 1615 cm^{-1}; ^1H NMR (DMSO-d_6) δ 1.4 (s, 3H, CH_3), 2.7 (s, 3H, CH_3), 4.22 (q, 2H, CH_2), 6.21 (s, 1H, pyrazole), 6.81-7.21 (m, 5H Ar-H); MS (EI) *m/z* (rel. intensity) 282 (5, M^+), 267 (15), 238 (10), 190 (20), 77 (90), 51 (50). Anal. Calcd. for $C_{15}H_{14}N_4O_2$ (282.29) C, 63.81; H, 4.99; N, 19.84. Found C, 63.73; H, 4.81; N, 19.65%.

3-Acetyl-4,7-diphenylpyrazolo[5,1-*c*][1,2,4]triazine 5c

Yield 88%; pale yellow solid; mp 235 °C; IR (KBr) 3160, 1680, 1620, 1610, cm^{-1}; ^1H NMR (DMSO-d_6) δ 3.1 (s, 3H, $COCH_3$), 6.3 (s, 1H, pyrazole), 7.0-7.40 (m, 10H, Ar-H); ^{13}C NMR (DMSO-d_6, 50 MHz) δ 169.1 (CO), 134.1 (C-3), 130.2 (C-4), 134.9 (C-7), 128.4 (C-8), 134.2 (C-9), 127.7, 119.8, 134.6, 130.1, 128.8, 129.9, 127.8, 131.0 (aromatic carbons), 14.2 (CH_3); MS (EI) *m/z* (rel. intensity) 314 (38, M^+), 237 (62), 194 (12), 117 (15), 77 (20), 52 (30). Anal. Calcd. for $C_{19}H_{14}N_4O$ (314.33) C, 72.59; H, 4.48; N, 17.82. Found C, 72.48; H, 4.41; N, 17.60%.

3-Acetyl-4-methyl-7-phenylpyrazolo[5,1-*c*][1,2,4]triazine 5d

Yield 85%; white solid; mp 225 °C; IR (KBr) 2920, 1680, 1635, 1610 cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.3 (s, 3H, CH_3), 3.1 (s, 3H, $COCH_3$), 6.22 (s, 1H, pyrazole), 6.9-7.20 (m, 5H, Ar-H); MS (EI) *m/z* (rel. intensity) 252 (15, M^+), 237 (17), 194 (20), 117 (30), 77 (20), 51 (40). Anal. Calcd. for $C_{14}H_{12}N_4O$ (252.27) C, 66.65; H, 4.79; N, 22.21. Found C,

* Compound **5b** have been previously prepared through otherwise different proceedure1 0,11

66.60; H, 4.65; N, 22.00%.

Ethyl 4-phenyl-1,2,4-triazolo[5,1-*c*][1,2,4]triazine-3-carboxylate 5e

Yield 75%; yellow crystals; mp 230 °C; IR (KBr) 1725, 1630, 1615, cm^{-1}; ^1H NMR (DMSO-d_6) δ 1.29 (t, 3H, CH_3), 4.31 (q, 2H, CH_2), 7.41-8.62 (m, 6H, Ar-H and H-3 triazole protons), ^{13}C NMR (DMSO-d_6, 50 MHz) δ 168.3 (CO_2Et), 142.3 (C-3), 135.6 (C-4), 134.9 (C-7), 134.0 (C-9), 128.6, 129.1, 130.2, 141.7 (aromatic carbons), 59.9 (OCH_2), 14.4 (CH_3); MS (EI) *m/z* (rel. intensity) 296 (84, M^+), 212 (60), 135 (20), 111 (15), 77 (30), 52 (20). Anal. Calcd. for $C_{13}H_{11}N_5O_2$ (296.26) C, 57.98; H, 4.11; N, 26.01. Found C, 57.89; 4.15; N, 25.90%.

Ethyl 4-methyl-1,2,4-triazolo[5,1-*c*][1,2,4]triazine-3-carboxylate 5f

Yield 67%; pale yellow solid; mp 185 °C; IR (KBr) 2990, 1715, 1625, 1610 cm^{-1}; ^1H NMR (DMSO-d_6) δ 1.3 (t, 3H, CH_3), 2.4 (s, 3H, CH_3), 4.40 (q, 2H, CH_2), 8.18 (s, 1H, triazole H-3), ^{13}C NMR (DMSO-d_6, 50 MHz) δ 169.0 (CO), 142.1 (C-3), 137.5 (C-4), 133.6 (C-7), 133.0 (C-9), 14.2 (CO*CH*$_3$), 20.2 (CH_3); MS (EI) *m/z* (rel. intensity) 207 (15, M^+), 192 (20), 119 (35), 95 (40), 57 (100). Anal. Calcd. for $C_8H_9N_5O_2$ (207.19) C, 46.37; H, 4.37; N, 33.80. Found C, 46.35; H, 4.31: N, 33.66%.

3-Acetyl-4-phenyl-1,2,4-triazolo[5,1-*c*][1,2,4]triazine 5g

Yield 80%; pale yellow solid; mp 198 °C; IR (KBr) 1710, 1640, 1625 cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.8 (s, 3H, $COCH_3$), 7.90-8.51 (m, 6H, Ar-H and triazole H-3 protons), MS (EI) *m/z* (rel. intensity) 239 (2, M^+), 185 (2), 171 (2), 161 (3), 130 (2), 97 (5), 71 (6), 57 (100), 45 (20). Anal. Calcd. for $C_{12}H_9N_5O$ (239.23) C, 60.24; H, 3.79; N, 29.27. Found C, 60.20; H, 3.73; N, 29.17%

3-Acetyl-4-methyl-1,2,4-triazolo[5,1-*c*][1,2,4]triazine 5h

Yield 62%; pale yellow solid; mp 215 °C; IR (KBr) 2999, 1715, 1635, 1610 cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.17 (s, 3H, CH_3), 2.80 (s, 3H, $COCH_3$), 8.11 (s, 1H, triazole H-3); MS (EI) *m/z* (rel. intensity) 177 (5, M^+), 123 (6), 97 (8), 57 (70), 53 (30). Anal. Calcd. for $C_7H_7N_5O$ (177.16) C, 47.45; H, 3.98; N, 39.53. Found C, 47.39; H, 3.97; N, 39.42%.

General procedure for the preparation of 4-(*N*-phenylamino)-7-ph-3-substituted pyrazolo[5,1-*c*][1,2,4]triazines 7a,b and, 4-(*N*-phenylamino)-3-substituted-1,2,4-triazolo-[5,1-*c*][1,2,4]triazines 7c,d

A solution of phenylisocyanate (0.95 g, 8.0 mmol) in 10 mL of dry toluene was added dropwise to a stirred solution of azophosphoranes **3a-d** (8.0 mmol) in 20 mL of the same solvent; the resulting solution was stirred at reflux temperature for 1-5 h. (in some cases the solid separated on heat). After cooling, the solid product formed, was collected by filtration and recrystallized from dioxane/H_2O.

Ethyl 4-(*N*-phenylamino)-7-phenylpyrazolo[5,1-*c*][1,2,4]-triazine-3-carboxylate 7a

Yield 90%; white solid; mp 237 °C; IR (KBr) 1718, 1690, 1620, 1580 cm^{-1}; ^1H NMR (DMSO-d_6) δ 1.2 (t, 3H, CH_3), 3.2 (q, 2H, CH_2), 6.11 (s, 1H, pyrazole H-3), 7.3-8.1 (m, 11H, Ar-H and NH protons); MS (EI) *m/z* (rel. intensity) 360 (84, M^++1), 248 (40), 171 (20), 98 (10), 57 (50), 51 (70). Anal. Calcd. for $C_{20}H_{17}N_5O_2$ (359.38) C, 66.83; H, 4.76; N, 19.48. Found C, 66.80; H, 4.75; N, 19.39%.

3-Acetyl-4-(N-phenylamino)-7-phenylpyrazolo[5,1-*c*]-[1,2,4]triazine 7b

Yield 87%; pale yellow solid; mp 245 °C; IR (KBr) 3224, 1690, 1620, 1519, cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.3 (s, 3H, $COCH_3$), 6.12 (s, 1H, pyrazole H-3), 7.2-8.1 (m, 11H, Ar-H and NH protons); MS (EI) *m/z* (rel. intensity) 330 (93, M^++1), 218 (50), 169 (100), 125 (56), 139 (75), 117 (81), 108 (62), 102 (56). Anal. Calcd. for $C_{19}H_{15}N_5O$ (329.35) C, 69.28; H, 4.59; N, 21.26. Found C, 69.20; H, 4.51; N, 21.16%.

Ethyl 4-(N-phenylamino)-1,2,4-triazolo[5,1-*c*][1,2,4]triazine-3-carboxylate 7c

Yield 90%; white solid; mp 280 °C; IR (KBr) 3059, 1715, 1640, 1615 cm^{-1}; ^1H NMR (DMSO-d_6) δ 1.3 (t, 3H, CH_3), 4.21 (q, 2H, CH_2), 7.20-7.82 (m, 7H, Ar-H and NH protons), 8.17 (s, 1H, triazole H-3); MS (EI) *m/z* (rel. intensity) 285 (16, M^++1), 264 (15), 239 (32), 185 (18), 147 (30), 108 (49), 99 (66), 63 (100), 51 (62). Anal. Calcd. for $C_{13}H_{12}N_6O_2$ (284.28) C, 54.92; H, 4.25; N, 29.56. Found C, 54.81;H, 4.20; N, 29.44%.

3-Acetyl-4-(N-phenylamino)-1,2,4-triazolo[5,1-*c*][1,2,4]-triazine 7d

Yield 88%; pale yellow solid; mp 228 °C; IR (KBr) 3021, 2952, 1751, 1640, 1625, cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.8 (s, 3H, $COCH_3$), 7.21-7.90 (m, 7H, Ar-H and NH protons), 8.20 (s, 1H, triazole H-3); MS (EI) *m/z* (rel. intensity) 255 (93, M^++1), 218 (50), 169 (100), 125 (56), 139 (75), 117 (81), 108 (62), 102 (56). Anal. Calcd. for $C_{12}H_{10}N_6O$ (254.25) C, 56.68; H, 3.69; N, 33.15. Found C, 56.61; H, 3.91; N, 33.15%.

General procedure for the preparation of 4-mercapto-7-phenyl-3-substituted pyrazolo[5,1-*c*][1,2,4]triazines 9a,b and, 4-mercapto-3-substituted-1,2,4-triazolo[5,1-*c*][1,2,4]-triazines 9c,d

To a solution of azophosphoranes **3a-d** (5 mmol) in 25 mL of dry methylene chloride was added excess (7 mL) of carbon disulfide. The reaction mixture was stirred under nitrogen at room temperature for 6 h., the solution was evaporated to dryness, and the residue material was purified by crystallization from EtOH or EtOH/H_2O mixture.

Ethyl 4-mercapto-7-phenylpyrazolo[5,1-*c*][1,2,4]triazine-3-carboxylate 9a

Yield 65%; green solid; mp 185 °C; IR (KBr) 1725, 1655, 1622, 1610 cm^{-1}; ^1H NMR (DMSO-d_6) δ 1.3 (t, 3H, CH_3), 3.1 (s, 1H, SH), 4.2 (q, 2H, CH_2), 6.03 (s, 1H, pyrazole H-4), 7.21-7.90 (m, 5H, Ar-H); MS (EI) *m/z* (rel. intensity) 300 (16, M^+), 268 (20), 222 (25), 145 (70), 77 (100), 51 (50). Anal. Calcd. for $C_{14}H_{12}N_4O_2S$ (300.33) C, 55.98; H, 4.02; N, 18.65; S, 10.67. Found C, 55.93; H, 4.08; N, 18.53; S, 10.61%.

3-Acetyl-4-Mercapto-7-phenylpyrazolo[5,1-*c*][1,2,4]triazine 9b

Yield 63%; white solid; mp 210 °C; IR (KBr) 3116, 1715, 1650, 1612 cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.8 (s, 3H, $COCH_3$), 2.99 (s, 1H, SH), 6.08 (s, 1H, pyrazole), 7.65-8.22 (m, 5H, Ar-H), MS (EI) *m/z* (rel. intensity) 270 (3, M^+), 222 (5), 194 (10), 117 (20), 77 (90), 53 (40). Anal. Calcd. for $C_{13}H_{10}N_4OS$ (270.30) C, 57.76; H, 3.72; N, 20.72; S, 11.86. Found C, 57.71; H, 3.70; N, 20.61; S, 11.80%.

Ethyl 4-mercapto-1,2,4-triazolo[5,1-*c*][1,2,4]triazine-3-carboxylate 9c

Yield 70%; pale green solid; mp 230 °C; IR (KBr) 1725, 1630, 1615 cm^{-1}; ^1H NMR (DMSO-d_6) δ 1.2 (t, 3H, CH_3), 3.3 (s, 1H, SH), 4.2 (q, 2H, CH_2), 8.15 (s, 1H, triazole H-3); MS (EI) *m/z* (rel. intensity) 225 (16, M^+), 192 (15), 147 (42), 92 (70), 62 (30). Anal. Calcd. for $C_7H_7N_5O_2S$ (225.23) C, 37.32; H, 3.13; N, 31.09; S, 14.23. Found C, 37.30; H, 3.14; N, 31.11; S, 14.02%.

3-Acetyl-4-Mercapto-1,2,4-triazolo[5,1-*c*][1,2,4]triazine 9d

Yield 65%; pale green solid; mp 205 °C; IR (KBr) 1720, 1635, 1620, 1615 cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.6 (s, 3H, $COCH_3$), 3.12 (s, 1H, SH), 8.17 (s, 1H, pyrazole); MS (EI) *m/z* (rel. intensity) 195 (5, M^+), 162 (15), 147 (30), 119 (50), 61 (25). Anal. Calcd. for $C_6H_5N_5OS$ (195.20) C, 36.91; H, 2.58; N, 35.88; S, 16.42. Found C, 36.93; H, 2.57; N, 35.79; S, 16.39%.

General procedure for the preparation of pyrazolo[4,5-*c*]pyrazoles 13a and 13b

Et_3N (9.5 mmol) was added to a stirred mixture of the appropriate hydrazonyl chlorides **11a** and/or **11b** (6.0 mmol) and PPh_3 (6.0 mmol), in CH_3CN (10 mL) at room temperature; white crystals formed within a few minutes. The mixture was stirred for a further 1/2 h to complete precipitation; the yielded product was collected by filtration, washed with a least amount of CH_3CN and crystallized from DMF.

4-Oxo-3-methylthiopyrazolo[4,5-*c*]pyrazol-5-yl(ethoxycarbonyl)methylenetriphenylphosphorane 13a

Yield 95%; white crystals; mp 310 °C; IR (KBr) 1715, 1650, 1635, 1569 cm^{-1}; ^1H NMR (DMSO-d_6) δ 1.32-1.40 (t, 3H, CH_3), 2.5 (s, 3H, CH_3), 4.24-4.40 (q, 2H,CH_2), 6.43-7.95 (m, 15H, Ar-H); MS (EI) *m/z* (rel. intensity) 514 (100, M^+), 468 (60), 391 (25), 269 (60), 262 (80), 237 (40), 77 (20), 51 (50). Anal. Calcd. for $C_{27}H_{23}N_4O_3SP$ (514.51) C, 63.02; H, 4.50; N, 10.88, S, 6.23. Found C, 63.14; H, 4.51; N, 10.61; S, 6.20%.

4-Oxo-3-methylthiopyrazolo[4,5-*c*]pyrazol-5-yl(methoxycarbonyl)methylenetriphenylphosphorane 13b

Yield 93%; white crystals; mp 315 °C; IR (KBr) 2290, 1710, 1680, 1620 cm^{-1}; ^1H NMR (DMSO-d_6) δ 2.24 (s, 3H, CH_3), 2.81 (s, 3H, $COCH_3$), 6.43-7.81 (m, 15H, Ar-H); MS (EI) *m/z* (rel. intensity) 484 (100, M^+), 438 (41), 361 (15), 274 (20), 263 (60), 197 (15), 183 (80), 77 (40), 51 (50). Anal. Calcd. for $C_{26}H_{21}N_4O_2SP$ (484.49) C, 64.45; H, 4.36; N, 11.56, S, 6.60. Found C, 64.38; H, 4.32; N, 11.39; S, 6.52%.

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