Fig 2.2 3D overlapping of compound II a and series IV a
All the melting points were determined in open capillaries and are uncorrected. The IR spectra were recorded in potassium bromide discs on Perkin-Elmer model-841 Spectrophotometer. Ultraviolet spectra were recorded on Schematzu model 25-spectrophotometer. $^1$HNMR spectra were taken on Varion A-60 spectrometer at 60 MHz using Tetramethyl silane (TMS) as an internal standard. The Mass spectra were obtained on Varion Atlas CH-7 spectrometer at 70eV ionizing beam using direct insertion probe. Thin layer chromatography was performed on microscopic slides (2 x 7.5 cms) coated with silica gel G and the spots visualized by exposure to iodine vapour and UV radiation. Satisfactory microanalysis (± 0.4 % of the calculated values) was obtained for the compounds.
3.1 Synthesis of Di (methylmercapto) methylene ethylcyanoacetate\textsuperscript{96,97}

To an ice-cold solution of 13.2 g (0.2 mole) of potassium hydroxide (85%) in 10 ml of water and 30 ml of dimethylformamide was added with cooling and stirring, 11.3 ml (0.1 mole) of Ethylcyanoacetate followed by 7.6 g (0.1 mole) of carbon disulphide. The mixture was stirred for one hour at room temperature, cooled and treated drop wise with 25.2 g (0.2 mole) of Dimethylsulfate, maintaining the temperature at 20°C. The reaction mixture was allowed to stand at room temperature for 12 hours and poured into ice water. The solid obtained was filtered, washed with water and dried and recrystallised from methanol to get yellow crystalline compound.

Analysis:

<table>
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<td>M.P.</td>
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<tr>
<td>Yield</td>
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<tr>
<td>TLC</td>
<td>Solvent: Benzene: Methanol [9:1]</td>
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<tr>
<td>Rf</td>
<td>0.73</td>
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</table>


3.2 Synthesis of 1-Methylmercapto-1-amino methyleneethylcyanoacetate$^{100,101}$

To a solution of 3.1, [Di (methylmercapto)methylene ethylcyanoacetate] (10 g, 0.04 mole) in rectified spirit, was added 50 ml of conc. Ammonium hydroxide solution. The mixture was then refluxed for about 6 hours. The solution was cooled, the crystals were filtered, washed with alcohol and dried and recrystallised from ethanol-chloroform mixture.

Analysis:

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<td>Yield</td>
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<td>TLC</td>
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<td>Rf</td>
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</tr>
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3.3 Synthesis of N,N-diarylisothioureas\textsuperscript{107-113}

**GENERAL PROCEDURE**

To a solution of potassium hydroxide (5.6 g; 0.1 mole) in ethanol (30 ml, 100%), was added the appropriate monoaryl amine (0.2 mole) and carbondisulphide (20 ml). The mixture was refluxed on a steam bath until the crystals of N,N-diarylisothioureas started separating out. Excess of organic solvent was removed by distillation under reduced pressure. The residue was washed with HCl (10% V/V) and water, dried and recrystallised with ethanol.

3.3 a) (Symm.) Di-(4-methyl phenyl) isothiourea

This compound was obtained through the reaction of p-toludine and carbondisulphide as colourless needles (Ethanol)

Yield : 73%
Melting point : 175-178\textdegree C.

3.3 b) (Symm.) Di-(2-methyl phenyl) isothiourea

This compound was obtained through the reaction of o-toludine and carbondisulphide as colourless needles (Ethanol)

Yield : 78%
Melting point : 127-128\textdegree C.
3.3 c) (Symm.) Di- (4-methoxy phenyl) isothioureia

This compound was obtained through the reaction of p-anisidine and carbonbisulphide as colourless needles (Ethanol)

Yield : 73 %
Melting point : 174-176°C.

3.3 d) (Symm.) Di- (4-bromo phenyl) isothioureia

This compound was obtained through the reaction of p-bromo aniline and carbonbisulphide as colourless needles (Ethanol)

Yield : 80 %
Melting point : 144-146°C.

3.3 e) (Symm.) Di- (4-iodo phenyl) isothioureia:

This compound was obtained through the reaction of p-iodo aniline and carbonbisulphide as violet coloured product (Ethanol).

Yield : 67 %
Melting point : 188-190°C.
3.3 f) (Symm.) Di-(4-flouro phenyl) isothiourea

This compound was obtained through the reaction of p-flouro aniline and carbondisulphide as grayish scaly product (Ethanol)

Yield : 75 %
Melting point : 185-187°C.
3.4 Synthesis of S-methyl diarylisothioureas$^{107-113}$

**GENERAL PROCEDURE**

To the ice cold suspension of the appropriate monoaryltiourea (7.5 g, 0.1 mole) in minimum quantity of acetone (30 ml), Dimethylsulphate (0.1 mole) was added dropwise with continuous stirring over a period of half an hour. The mixture was thereafter stirred at room temperature for an additional hour then refluxed for 3 hours. The acetone removed by distillation. The heavy oily residue was taken up in ice cold water, filtered. The filtrate basified (10 % aq. Na$_2$CO$_3$). The solid separated out was filtered, washed with cold water dried and recrystallised.

3.4 a) S-methyl di (4-methyl phenyl) isothiourea

This compound was obtained through the reaction of (Symm.) di (4-methyl phenyl) isothiourea and dimethylsulphate as white crystalline solid (Benzene : Petroleum ether).

3.4 b) S-methyl di (2-methyl phenyl) isothiourea

This compound was obtained through the reaction of (Symm.) di (2-methyl phenyl) isothiourea and dimethylsulphate as white crystalline solid (Benzene : Petroleum ether).
3.4 c) S-methyl di (4-methoxy phenyl ) isothiourea

This compound was obtained through the reaction of ( Symm.) di (4-methoxy phenyl ) isothiourea and dimethylsulphate as white crystalline solid ( Benzene : Petroleum ether ).

3.4 d) S-methyl di (4-bromo phenyl ) isothiourea

This compound was obtained through the reaction of ( Symm.) di (4-bromo phenyl ) isothiourea and dimethylsulphate as white crystalline solid ( Benzene : Petroleum ether ).

3.4 e) S-methyl di (4-iodo phenyl ) isothiourea

This compound was obtained through the reaction of ( Symm.) di (4-iodo phenyl ) isothiourea and dimethylsulphate as violet crystalline solid ( Benzene : Petroleum ether ).

3.4 f) S-methyl di (4-fluro phenyl ) isothiourea

This compound was obtained through the reaction of ( Symm.) di (4-fluro phenyl ) isothiourea and dimethylsulphate as white crystalline solid ( Benzene : Petroleum ether ).
3.5 Synthesis of 5-cyano-6-methylmercapto-3-(substituted)-aryl-2-(substituted) arylaminopyrimidin-4 (3H)-ones (SERIES - I)

GENERAL PROCEDURE

A solution of 1-methylmercapto-1-amino methylene ethyl cyanoacetate (0.1 mole) and S-methyl N,N-diarylisothioureas (0.1 mole) in dimethyl formamide was refluxed for 6-7 hours with intermittent addition of pinch of K₂CO₃. The reaction mixture was cooled and poured in 400 ml of ice-cold water. The solid separated out was filtered, washed with water, dried and recrystallized from ethanol: chloroform mixture

3.5 a) Synthesis of 5-cyano-6-methylmercapto-3- (4-methyl phenyl)-2-(4-methylphenyl)aminopyrimidin-4 (3H)-ones

A solution of 1-methylmercapto-1-amino methylene ethylcyanoacetate (0.1 mole) and S-methyl di (4-methyl phenyl ) isothiourea (0.1 mole) in dimethyl formamide was refluxed for 6-7 hours with intermittent addition of pinch of K₂CO₃. The reaction mixture was cooled and poured in 400 ml of ice-cold water. The solid separated out was filtered, washed with water, dried and recrystallized from ethanol: chloroform mixture.
3.5 b) Synthesis of 5-cyano-6-methylmercapto-3-(2-methylphenyl)-2-(2-methylphenyl)aminopyrimidin-4 (3H)-ones:

A solution of 1-methylmercapto-1-amino methylene ethylcyanoacetate (0.1 mole) and S-methyl di (2-methyl phenyl) isothioure (0.1 mole) in dimethyl formamide was refluxed for 6-7 hours with intermittent addition of pinch of K₂CO₃. The reaction mixture was allowed to cool and poured in 400 ml of ice-cold water. The solid separated out was filtered, washed with water, dried and recrystallized from ethanol: chloroform mixture.

3.5 c) Synthesis of 5-cyano-6-methylmercapto-3-(4-methoxy phenyl)-2-(4-methoxyphenyl)aminopyrimidin-4(3H)-ones:

A solution of 1-methylmercapto-1-amino methylene ethylcyanoacetate (0.1 mole) and S-methyl di (4-methoxy phenyl) isothioure (0.1 mole) in dimethyl formamide was refluxed for 6-7 hours with intermittent addition of pinch of K₂CO₃. The reaction mixture was cooled and poured in 400 ml of ice-cold water. The solid separated out was filtered, washed with water, dried and recrystallized from ethanol: chloroform mixture.

3.5 d) Synthesis of 5-cyano-6-methylmercapto-3-(4-bromo phenyl)-2-(4-bromophenyl)aminopyrimidin-4(3H)-ones:

A solution of 1-methylmercapto-1-amino methylene ethylcyanoacetate (0.1 mole) and S-methyl di (4-bromo phenyl) isothioure (0.1 mole) in dimethyl formamide was refluxed for 7-8 hours with intermittent addition of pinch of K₂CO₃. The reaction mixture cooled and poured in 400 ml of ice-cold water.
The solid separated out was filtered, washed with water, dried and recrystallized from ethanol : chloroform mixture

3.5 e) Synthesis of 5 – cyano – 6 – methylmercapto – 3 - ( 4 - iodo phenyl ) -2-(4-iodophenyl)aminopyrimidin-4(3H)-ones :

A solution of 1-methylmercapto-1-amino methylene ethylcyanoacetate (0.1 mole) and S-methyl di (4-iodo phenyl ) isothiourea ( 0.1 mole) in dimethyl formamide was refluxed for 7-8 hours with intermittent addition of pinch of K₂CO₃. The reaction mixture cooled and poured in 400 ml of ice-cold water. The solid separated out was filtered, washed with water, dried and recrystallized from ethanol : chloroform mixture.

3.5 f) Synthesis of 5 – cyano – 6 – methylmercapto – 3 - ( 4 - fluro phenyl ) – 2 - ( 4-fluorophenyl)aminopyrimidin-4(3H)-ones :

A solution of 1-methylmercapto-1-amino methylene ethylcyanoacetate (0.1 mole) and S-methyl di (4-fluoro phenyl ) isothiourea ( 0.1 mole) in dimethyl formamide was refluxed for 7-8 hours with intermittent addition of pinch of K₂CO₃. The reaction mixture cooled and poured in 400 ml of ice-cold water. The solid separated out was filtered, washed with water, dried and recrystallized from ethanol : chloroform mixture.

3.6 Synthesis of 2, 4 – diamino – 6 - ( substituted ) aryl -7-(substituted)arylamino-pyrimido [4,5-d] pyrimidin-5 (6H)ones (SERIES-II).
The target series was synthesized by the condensation of the pyrimidines, 5 - cyano - 6 - methyl mercapto - 3 - (substituted) aryl - 2 - (substituted) arylamino pyrimidin-4(3H)ones with guanidine nitrate in dimethylformamide in the presence of catalytic amounts of anhydrous potassium carbonate till the evolution of methylmercaptan ceased.89

3.6 a) Synthesis of 2, 4 - diamino - 6 - (4-methyl phenyl) - 7 -(4-methyl phenyl) amino-pyrimido [4,5-d] pyrimidin-5 (6H)one :

A mixture of 6-methylmercapto-5-cyano-3-(4-methylphenyl)-2-(4-methylphenyl) aminopyrimidin-4(3H) one (0.1 mole ) and Guanidine nitrate (0.8 moles ) and Dimethyl formamide (50ml) was refluxed until the methylmercaptan gas was ceased with intermittent addition of anhydrous potassium carbonate. The reaction mixture was cooled to room temperature, poured into ice water. The solid obtained was filtered, dried and recrystallised.

3.6 b) Synthesis of 2,4-diamino-6- (2-methyl phenyl)-7- (2-methyl phenyl) amino-pyrimido [4,5-d] pyrimidin-5 (6H)one :

A mixture of 6 - methylmercapto - 5 - cyano - 3 - (2-methylphenyl)-2-(2-methylphenyl) aminopyrimidin-4(3H) one (0.1 mole ) and Guanidine nitrate (0.8 moles ) and Dimethyl formamide (50ml) was refluxed until the methylmercaptan gas was ceased with intermittent addition of anhydrous potassium carbonate. The reaction mixture was cooled to room temperature, poured into ice water. The solid obtained was filtered, dried and recrystallised.
3.6 c) Synthesis of 2,4-diamino-6- (4-methoxy phenyl)-7- (4-methoxyphenyl) amino-pyrimidido [4,5-d]pyrimidin- 5(6H)one

A mixture of 6 - methylmercaptto - 5 - cyano - 3 - ( 4 - methoxyphenyl)-2-(4-methoxyphenyl) aminopyrimidin-4(3H) one (0.1 mole ) and Guanidine nitrate (0.8 moles ) and Dimethyl formamide (50ml) was refluxed until the methylmercaptan gas was ceased with intermittent addition of anhydrous potassium carbonate. The reaction mixture was cooled to room temperature, poured into ice water. The solid obtained was filtered, dried and recrystallised.

3.6 d) Synthesis of 2,4-diamino-6- (4-bromo phenyl)-7- (4-bromo phenyl) amino-pyrimidido [4,5-d] pyrimidin-5 (6H)one

A mixture of 6 - methylmercaptto - 5 - cyano - 3 - ( 4 - bromophenyl)-2-(4-bromophenyl) aminopyrimidin-4(3H) one (0.1 mole ) and Guanidine nitrate (0.8 moles ) and Dimethyl formamide (50ml) was refluxed until the methylmercaptan gas was ceased with intermittent addition of anhydrous potassium carbonate. The reaction mixture was cooled to room temperature, poured into ice water. The solid obtained was filtered, dried and recrystallised.

3.6 e) Synthesis of 2,4-diamino-6- (4-iodo phenyl)-7- (4-iodo phenyl) amino-pyrimidido [4,5-d] pyrimidin-5 (6H)one

A mixture of 6-methylmercaptto-5-cyano-3-(4-iodophenyl)- 2-(4-iodophenyl) aminopyrimidin-4(3H) one (0.1 mole ) and Guanidine nitrate (0.8 moles ) and Dimethyl formamide (50ml) was refluxed until the methylmercaptan gas was ceased with intermittent addition of anhydrous potassium carbonate. The
reaction mixture was cooled to room temperature, poured into ice water. The solid obtained was filtered, dried and recrystallised.

3.6 f) Synthesis of 2,4-diamino-6-(4-fluro phenyl)-7-(4-fluro phenyl) amino-pyrimido [4,5-d] pyrimidin-5 (6H)one

A mixture of 6 - methylmercaptan - 5 - cyano - 3 - ( 4 - methylphenyl)-2-(4-methylphenyl) aminopyrimidin-4(3H) one (0.1 mole ) and Guanidine nitrate (0.8 moles ) and Dimethyl formamide (50ml) was refluxed until the methylmercaptan gas was ceased with intermittent addition of anhydrous potassium carbonate. The reaction mixture was cooled to room temperature, poured into ice water. The solid obtained was filtered, dried and recrystallised.
3.7 Synthesis of 5 - cyano - 6 - methyl sulphonyl - 3 - (substituted)aryl-2- (substituted)-arylaminoypyrimidin-4 (3H)-ones (SERIES III)

5 - cyano - 6 - methylsulphonyl - 3 - (substituted) aryl - 2 - (substituted)-arylaminoypyrimidin-4 (3H)-ones were synthesized from the corresponding 5-cyano-6-methylmercapto-3-(substituted ) aryl-2- (substituted)-arylaminoypyrimidin-4 (3H)-ones by oxidation using 30% H₂O₂ (15-20 ml ) in glacial acetic acid (20-30 ml) at 60-65°C for variable time. The reaction mixture was further stirred for an additional hour. The product was isolated by pouring the reaction mixture in ice-water, product obtained was filtered, dried and recrystallised from suitable solvent.¹¹⁸

3.7 a ) Synthesis of 5-cyano-6-methylsulphonyl-3- (4-methyl phenyl) -2-(4-methylphenyl)aminopyrimidin-4(3H)ones

To a mixture of 1.8g (0.02 moles) of 5-cyano-6-methylmercapto - 3 - (4 - methylphenyl ) - 2 - (4-methylphenyl)-aminopyrimidin-4(3H)-ones in 20 ml glacial acetic acid was added 8 ml, 30%, H₂O₂ dropwise. The reaction mixture was heated on steam bath for 1.5 hr, cooled and poured into a mixture of ice-water. The precipitated 5-cyano-6-methylsulphonyl- 3 - (4 - methylphenyl ) - 2 - (4-methylphenyl)-aminopyrimidin-4(3H)-ones was filtered, washed with cold water and air dried. The compound was recrystallised from ethanol-chloroform mixture.
3.7 b) Synthesis of 5-cyano-6-methyl sulphonyl-3-(2-methylphenyl)-2-(2-methylphenyl)aminopyrimidin-4(3H)ones

To a mixture of 1.8g (0.02 moles) of 5-cyano-6-methylmercapto-3-(2-methylphenyl)-2-(2-methylphenyl)aminopyrimidin-4(3H)-ones in 20 ml glacial acetic acid was added 8 ml 30%, H₂O₂ dropwise. The reaction mixture was heated on steam bath for 1.5 hr, cooled and poured into a mixture of ice-water. The precipitated 5-cyano-6-methyl sulphonyl-3-(2-methylphenyl)-2-(2-methylphenyl)aminopyrimidin-4(3H)-ones was filtered, washed with cold water and air dried. The compound was recrystallised from ethanol-chloroform mixture.

3.7 c) Synthesis of 5-cyano-6-methyl sulphonyl-3-(4-methoxy phenyl)-2-(4-methoxy phenyl)aminopyrimidin-4(3H)ones

To a mixture of 1.97 g (0.02 moles) of 5-cyano-6-methylmercapto-3-(4-methoxyphenyl)-2-(4-methoxyphenyl)aminopyrimidin-4(3H)-ones in 20 ml glacial acetic acid was added 8 ml, 30%, H₂O₂ drop wise. The reaction mixture was heated on steam bath for 1.5 hr, cooled and poured into a mixture of ice-water. The precipitated 5-cyano-6-methyl sulphonyl-3-(4-methoxy phenyl)-2-(4-methoxyphenyl)aminopyrimidin-4(3H)-ones was filtered, washed with cold water and air dried. The compound was recrystallised from ethanol-chloroform mixture.
3.7 d) **Synthesis of 5 - cyano - 6 - methylsulphonyl - 3 - (4-bromophenyl)-2-(4-bromophenyl)aminopyrimidin-4(3H)ones**

To a mixture of 1.97 g (0.02 moles) of 5-cyano-6-methylmercapto-3-(4-bromophenyl)-2-(4-bromophenyl)aminopyrimidin-4(3H)-ones in 20 ml glacial acetic acid was added 8 ml, 30%, H₂O₂ dropwise. The reaction mixture was heated on steam bath for 1.5 hr, cooled and poured into a mixture of ice-water. The precipitated 5 - cyano - 6 - methylsulphonyl-3-(4-bromophenyl)-2-(4-bromophenyl)-aminopyrimidin-4(3H)-ones was filtered, washed with cold water and air dried. The compound was recrystallised from ethanol-chloroform mixture.

3.7 e) **Synthesis of 5 - cyano - 6 - methyl sulphonyl - 3 - (4-iodophenyl) -2-(4-iodophenyl)aminopyrimidin-4(3H)ones**

To a mixture of 1.97 g (0.02 moles) of 5-cyano-6-methylmercapto-3-(4-iodophenyl)-2-(4-iodophenyl)aminopyrimidin-4(3H)-ones in 20 ml glacial acetic acid was added 8 ml, 30%, H₂O₂ dropwise. The reaction mixture was heated on steam bath for 1.5 hr, cooled and poured into a mixture of ice-water. The precipitated 5 - cyano - 6 -methyl sulphonyl-3-(4-iodophenyl)-2-(4-iodophenyl)-aminopyrimidin - 4 (3H)-ones was filtered, washed with cold water and air dried. The compound was recrystallised from ethanol-chloroform mixture.
3.7 Synthesis of 5 - cyano - 6 - methylsulphonyl - 3 -(4 - fluorophenyl) -2-(4-fluorophenyl)aminopyrimidin-4(3H)ones

To a mixture of 1.97 g (0.02 moles) of 5-cyano-6- methylmercapto - 3 -(4 - fluorophenyl) - 2 -(4 - fluorophenyl) - aminopyrimidin-4(3H)-ones in 20 ml glacial acetic acid was added 8 ml, 30%, H₂O₂ dropwise. The reaction mixture was heated on steam bath for 1.5 hr, cooled and poured into a mixture of ice-water. The precipitated 5 - cyano - 6 -methyl sulphonyl- 3 -(4-fluorophenyl)-2-(4-fluorophenyl)-amonopyrimidin-4(3H)-ones was filtered, washed with cold water and air dried. The compound was recrystallised from ethanol-chloroform mixture.

3.8 Synthesis of 5-cyano-6-guanidino-3- (substituted)aryl-2- (substituted) -arylaminopyrimidin-4 (3H)-ones (series IV)

The target series was synthesized by the reaction of the 5-cyano-6-methylsulphonyl-3- (substituted)aryl-2- (substituted)-arylaminopyrimidin-4 (3H)-ones with guanidine nitrate in dimethylformamide in the presence of catalytic amounts of anhydrous potassium carbonate. ( Other than halogen substituted compounds the reaction was carried out at lower temperature i.e. at 80-90°C for 6-7 hours ).
3.8 a) Synthesis of 5-cyano-6-guanidino-3- (4-methylphenyl)-
2- (4-methylphenyl)amino pyrimidin-4 (3H)-one

A mixture of 5 – cyano – 6 – methylsulphonyl – 3 -(4-methyl
phenyl) -2-(4-methylphenyl)aminopyrimidin-4(3H)-one (0.1 mole
) and Guanidine nitrate (0.8 mole) in 50 ml Dimethyl formamide
was heated with stirring ( condensor attached with calcium
chloride guard tube) till the reaction completed. Intermittently
anhydrous potassium carbonate was added. The reaction
mixture was cooled to room temperature, poured into ice-water.
The solid obtained was filtered dried and recrystallised.

3.8 b) Synthesis of 5-cyano-6-guanidino-3- (2-methylphenyl)-
2- (2-methylphenyl)amino pyrimidin-4 (3H)-one

A mixture of 5-cyano-6-methylsulphonyl-3-(2-methyl
phenyl) -2-(2-methylphenyl)aminopyrimidin-4(3H)-one (0.1 mole
) and Guanidine nitrate (0.8 mole) in 50 ml Dimethyl formamide
was heated with stirring ( condensor attached with calcium
chloride guard tube) till the reaction completed. Intermittently
anhydrous potassium carbonate was added. The reaction
mixture was cooled to room temperature, poured into ice-water.
The solid obtained was filtered dried and recrystallised.
3.8 c) Synthesis of 5-cyano-6-guanidine-3-(4-methoxyphenyl)-2-(4-methoxyphenyl) amino pyrimidin-4(3H)-one

A mixture of 5-cyano-6-methylsulphonyl-3-(4-methoxy phenyl)-2-(4-methoxyphenyl)aminopyrimidin-4(3H)-one (0.1 mole) and Guanidine nitrate (0.8 mole) in 50 ml Dimethyl formamide was heated with stirring (condenser attached with calcium chloride guard tube) till the reaction completed. Intermittently anhydrous potassium carbonate was added. The reaction mixture was cooled to room temperature, poured into ice-water. The solid obtained was filtered dried and recrystallised.

3.8 d) Synthesis of 5-cyano-6-guanidino-3-(4-bromophenyl)-2-(4-bromophenyl) amino pyrimidin-4(3H)-one

A mixture of 5-cyano-6-methylsulphonyl-3-(4-bromo phenyl)-2-(4-bromophenyl) aminopyrimidin-4(3H)-one (0.1 mole) and Guanidine nitrate (0.8 mole) in 50 ml Dimethyl formamide was heated with stirring (condenser attached with calcium chloride guard tube) till the reaction completed. Intermittently anhydrous potassium carbonate was added. The reaction mixture was cooled to room temperature, poured into ice-water. The solid obtained was filtered dried and recrystallised.
3.8 e) Synthesis of 5-cyano-6-guanidino-3- (4-iodophenyl)-2-(4-iodophenyl)amino pyrimidin-4 (3H)-one

A mixture of 5-cyano-6-methylsulphonyl-3-(4-iodo phenyl) -2-(4-iodophenyl)aminopyrimidin-4(3H)-one (0.1 mole ) and Guanidine nitrate (0.8 mole) in 50 ml Dimethyl formamide was heated with stirring ( condensor attached with calcium chloride guard tube) till the reaction completed. Intermittently anhydrous potassium carbonate was added. The reaction mixture was cooled to room temperature, poured into ice-water. The solid obtained was filtered dried and recrystallised.

3.8 f) Synthesis of 5-cyano-6-guanidino-3- (4-fluorophenyl)-2-(4-fluorophenyl)amino pyrimidin-4 (3H)-one

A mixture of 5-cyano-6-methylsulphonyl-3-(4-fluro phenyl) -2-(4-fluoro phenyl)aminopyrimidin-4(3H)-one (0.1 mole ) and Guanidine nitrate (0.8 mole) in 50 ml Dimethyl formamide was heated with stirring ( condensor attached with calcium chloride guard tube) till the reaction completed. Intermittently anhydrous potassium carbonate was added. The reaction mixture was cooled to room temperature, poured into ice-water. The solid obtained was filtered dried and recrystallised.
3.9 Synthesis of 5-cyano-6-methylmercapto-1,3-di(substitutedaryl)-2-thio-pyrimidin-4(3H)-ones. (SCHEME V)

The 1-(dimethylmercapto)methylene ethylcyanoacetate was refluxed with the appropriate symmetrical disubstituted diaryl isothioureas in Dimethylformamide in the presence of catalytic amount of anhydrous potassium carbonate till the evolution of methyl mercaptan ceased.

The workup of the reaction mixture, involves pouring the reaction mixture in ice water, filtering, drying the separated residue to yield a solid, which was recrystallised from ethanol-chloroform to give the corresponding 5-cyano-6-methylmercapto-1,3-di(substitutedaryl)-2-thio pyrimidin-4(3H)ones.

3.9 a) Synthesis of 5-cyano-6-methylmercapto-1,3-di(4-methylphenyl)-2-thio-pyrimidin-4(3H)-ones

A solution of 1- (dimethyl mercapto ) methylene ethylcyanoacetate (0.1 mole) and (Symm.) Di- (4-methyl phenyl) isothiourea_ (0.1 mole) in dimethyl formamide (30 ml) was refluxed for 6-7 hours with intermittent addition of pinch of K₂CO₃. The reaction mixture was cooled and poured in 400 ml of ice-cold water. The solid separated out was filtered, washed with water, dried and recrystallized from ethanol: chloroform mixture.
3.9 b) Synthesis of 5 - cyano - 6 - methylmercaptop - 1, 3 - di(2-methylphenyl) -2-thio-pyrimidin-4(3H) -ones

A solution of 1 - (dimethyl mercapto) methylene ethylcyanoacetate (0.1 mole) and (Symm.) Di- (2-methyl phenyl) isothiourea (0.1 mole) in dimethyl formamide (30 ml) was refluxed for 6-7 hours with intermittent addition of pinch of $\text{K}_2\text{CO}_3$. The reaction mixture was cooled and poured in 400 ml of ice-cold water. The solid separated out was filtered, washed with water, dried and recrystallized from ethanol: chloroform mixture.

3.9 c) Synthesis of 5 - cyano - 6 - methyl mercapto -1, 3-di(4-methoxyphenyl) -2-thio-pyrimidin-4(3H) -ones

A solution of 1 - (dimethyl mercapto) methylene ethylcyanoacetate (0.1 mole) and (Symm.) Di- (4-methoxy phenyl) isothiourea (0.1 mole) in dimethyl formamide (30 ml) was refluxed for 6-7 hours with intermittent addition of pinch of $\text{K}_2\text{CO}_3$. The reaction mixture was cooled and poured in 400 ml of ice-cold water. The solid separated out was filtered, washed with water, dried and recrystallized from ethanol: chloroform mixture.
3.9 d) Synthesis of 5 – cyano – 6 – methylmercapto – 1, 3-di(4-bromophenyl) -2-thio-pyrimidin-4(3H) –ones

A solution of 1 - (dimethyl mercapto) methylene ethylcyanoacetate (0.1 mole) and (Symm.) Di- (4-bromophenyl) isothioureia (0.1 mole) in dimethyl formamide (30 ml) was refluxed for 8-9 hours with intermittent addition of pinch of K₂CO₃. The reaction mixture was cooled and poured in 400 ml of ice-cold water. The solid separated out was filtered, washed with water, dried and recrystallized from ethanol: chloroform mixture.

3.9 e) Synthesis of 5 – cyano – 6 – methylmercapto – 1, 3-di (4-iodophenyl) -2-thio-pyrimidin-4(3H) –ones

A solution of 1 - (dimethyl mercapto) methylene ethylcyanoacetate (0.1 mole) and (Symm.) Di- (4-iodo phenyl) isothioureia (0.1 mole) in dimethyl formamide (30 ml) was refluxed for 8-9 hours with intermittent addition of pinch of K₂CO₃. The reaction mixture was cooled and poured in 400 ml of ice-cold water. The solid separated out was filtered, washed with water, dried and recrystallized from ethanol: chloroform mixture.

3.9 f) Synthesis of 5 – cyano – 6 – methylmercapto – 1, 3-di (4-fluorophenyl) -2-thio-pyrimidin-4(3H) –ones

A solution of 1 - (dimethyl mercapto) methylene ethylcyanoacetate (0.1 mole) and (Symm.) Di- (4-fluro phenyl) isothioureia (0.1 mole) in dimethyl formamide (30 ml) was refluxed for 8-9 hours with intermittent addition of pinch of K₂CO₃. The reaction mixture was cooled and poured in 400 ml of ice-cold water. The solid separated out was filtered, washed with water, dried and recrystallized from ethanol: chloroform mixture.
3.10 2, 4 - Diamino - 6, 8 - di ( substitutedaryl ) - 7 - thio-
pyrimido pyrimidin-5- ones ( SERIES VI )

The target series was synthesized by the condensation of the 5 - cyano - 6 - methylmercapto -1, 3- di ( substitutedaryl ) -
2 - thio - pyrimidin-4(3H) -ones with guanidine nitrate in dimethylformamide in the presence of catalytic amounts of anhydrous potassium carbonate till the evolution of methylmercaptan ceased.

3.10 a) Synthesis of 2, 4 - diamino - 6 , 8 - di ( 4-methyl
phenyl) -7-thio pyrimido pyrimidin-5 one

A mixture of 5 - cyano - 6 - methylmercapto -1, 3 - di ( 4-
methylphenyl) -2-thio-pyrimidin-4(3H) -one (0.1 mole ) and Guanidine nitrate (0.8 moles ) in Dimethyl formamide (50ml) was refluxed until the methylmercaptan gas ceased with intermittent addition of anhydrous potassium carbonate. The reaction mixture was cooled to room temperature, poured into ice water. The solid obtained was filtered, dried and recrystallised.

3.10 b) Synthesis of 2,4-diamino-6,8- di (2-methyl phenyl) –
7-thio pyrimido pyrimidin-5 one

A mixture of 5 - cyano - 6 - methyl mercapto -1, 3-di ( 2-
methylphenyl) -2-thio-pyrimidin-4(3H) -one (0.1 mole ) and Guanidine nitrate (0.8 moles ) in Dimethyl formamide (50ml) was refluxed until the methylmercaptan gas ceased with intermittent addition of anhydrous potassium carbonate. The reaction mixture was cooled to room temperature, poured into ice water. The solid obtained was filtered, dried and recrystallised.
3.10 c) Synthesis of 2,4-diamino-6,8- di (4-methoxy phenyl) -7-thio pyrimido pyrimidin-5 one

A mixture of 5 - cyano - 6 - methylmercapto -1, 3 - di ( 4 - methoxyphenyl) -2-thio-pyrimidin-4(3H) –one (0.1 mole ) and Guanidine nitrate (0.8 moles ) in Dimethyl formamide (50ml) was refluxed until the methylmercaptan gas ceased with intermittent addition of anhydrous potassium carbonate. The reaction mixture was cooled to room temperature, poured into ice water. The solid obtained was filtered, dried and recrystallised.

3.10 d) Synthesis of 2,4-diamino-6,8- di (4-bromo phenyl) -7-thio pyrimido pyrimidin-5 one

A mixture of 5-cyano-6-methylmercapto-1, 3-di(4-bromophenyl) -2-thio-pyrimidin-4(3H) –one (0.1 mole ) and Guanidine nitrate (0.8 moles ) in Dimethyl formamide (50ml) was refluxed until the methylmercaptan gas ceased with intermittent addition of anhydrous potassium carbonate. The reaction mixture was cooled to room temperature, poured into ice water. The solid obtained was filtered, dried and recrystallised.
3.10 e) Synthesis of 2,4-diamino-6,8-di (4-iodo phenyl) -7-thio pyrimido pyrimidin-5 one

A mixture of 5-cyano-6-methylmercapto-1, 3-di (4-iodo phenyl)-2-thio-pyrimidin-4(3H) -one (0.1 mole) and Guanidine nitrate (0.8 moles) in Dimethyl formamide (50ml) was refluxed until the methylmercaptan gas ceased with intermittent addition of anhydrous potassium carbonate. The reaction mixture was cooled to room temperature, poured into ice water. The solid obtained was filtered, dried and recrystallised.

3.10 f) Synthesis of 2,4-diamino-6,8-di (4-fluro phenyl) -7-thio pyrimido pyrimidin-5 one

A mixture of 5-cyano-6-methylmercapto-1, 3-di(4-fluro phenyl)-2-thio-pyrimidin-4(3H) -one (0.1 mole) and Guanidine nitrate (0.8 moles) in Dimethyl formamide (50ml) was refluxed until the methylmercaptan gas ceased with intermittent addition of anhydrous potassium carbonate. The reaction mixture was cooled to room temperature, poured into ice water. The solid obtained was filtered, dried and recrystallised.