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Contamination of Groundwater by Usage of Pesticide in Cotton Growing Area

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Abstract
Cotton is cultivated throughout the world under subtropical climatic conditions. The high water requirements and indiscriminate use of pesticides in cotton cultivation in India have resulted in contamination of associated surface water, such as streams, ditches, rivers, lakes and also the groundwater aquifers through percolation of pesticides from the contaminated soil. Through contaminated soil, pesticide also leaches down to groundwater. Cotton is a widely cultivated important cash crop in India and grows in largest area of 90 million hectares. In India 5% of total cultivable land is used in cotton cultivation and consumes 50% of total pesticides produced in India. Therefore a study was conducted to analyze the pesticide contamination of groundwater in and nearby areas of cotton growing fields. Pesticides used in the cotton field were surveyed from the farmers. Ground water samples were taken from eight different tubewells of cotton growing farm fields. Area selected for study was Sirsa belt in Haryana, India. A Cotton variety grown in the fields was Desi Cotton (RG-8 and HD-123). Pesticides spray in the cotton fields according to the survey with farmers were Chlorpyriphos, Carbofuran, Phorate, Imidacloprid, Endosulfan, Lindane, DDT, Carbandazim, Quinalphos, Monocrotophos, Carbaryl, and Methyl Parathion at different time interval. Among the various pesticides analyzed, 65.2% samples were found contaminated with organochlorine pesticides and 91.6% samples were contaminated with organophosphate pesticides above MRL values while synthetic pyrethroids were not detected in groundwater samples. Concentration of organochlorine pesticide ranged from 0.005- 1.055µg/l in groundwater samples. While concentration of organophosphate pesticides ranged from 0.004 to 4.364µg/l. Almost 93% samples were found contaminated with organochlorine pesticides, organophosphate pesticides respectively. According to the analysis, residues of the banned organo chlorine pesticide residues (DDT and BHC) were found above MRL value. It seems that they are present either due to the high persistence of these pesticides or due to their continuous use in the form of adulterated pesticides by farmer. Study reveals that rigorous extension services are required to make farmers aware about harmful residues in water and switching to safer molecules like synthetic pyrethroids and Integrated Pest Management techniques for judicious use of pesticides.

Keywords: Groundwater; Contamination; Leaching; Residues; Pesticides.

1. Introduction
The use of cotton, world over, has been on the upswing despite competition from synthetic and animal origin fibers [1]. About 130 different species of insects and mites found to devour cotton at different stages of crop growth in India among which bollworms *viz., Helicoverpa armigera* (Hubner), the American bollworm, *Earias vittella* (Fab.), spotted bollworm *Earias insulana* (Biosdual), spiny bollworm and *Pectinophora gossypiella* (Saunders),the pink bollworm occupy major pest status contributing to lower yields [2]. A recent estimation indicated that the loss caused by *H. armigera* and leaf hopper (*Amrasca biguttula biguttala* Ishida) was 31.0 and 18.0 per cent respectively [2]. A loss of US $ 1.0 billion worth cotton has been accounted for dreaded pest, *H. armigera* every year [4]. Cotton occupies only 5% percent of the cropped area, on an average it used to receive 48.0 per cent of the total pesticide used for agriculture purpose in India [5]. The indiscriminate use of pesticides, however, has adverse effects on the natural predators and parasites of bollworms. Use of pesticides also leads to environmental pollution (of soil and water), increase in the cost of cultivation, and sometimes, development of resistance in insects against insecticides [6].
Agriculture accounts for 83% of total water usage (79.7% in surface water and 89.6% in ground water). Globally, cotton production is associated with large amount of water withdrawal. About 53% of the global cotton area is under irrigation [7]. In India however, most of the cotton growing regions come under rain-fed (69%) and few areas (31%) are irrigated. According to estimates, the share of cotton water use from rainfall to irrigation is 405:133.20 [8]. However, it's the quality of the water that is affected most in cotton cultivation due to leaching and run off

chemicals in ground and surface water. Contamination in groundwater has been reported on a global scale. A study observed that less than 0.1% of pesticides applied for pest control reach their target pest. Thus more than 99.9% of pesticides used move into the environment [9]. In Bahawalnagar, Muzafargarh, D.G. Khan and Rajan Pur districts of Punjab, Pakistan, groundwater in four intensive cotton growing district were found contaminated with six pesticides. The percentage of detection of bifenthrin, λ-cyhalothrin, carbofuran, endosulfan, methyl parathion and monocrotophos was, respectively 13.5%, 5.4%, 59.4%, 8%, 5.4% and 35.1% in July; 16.2%, 13.55%, 43.2%, 8%, N.D. (not detected) and 24.3% in October [10]. Water samples collected from Syrian costal area reported that the residue level DDE was the highest among the rest of the detected organochlorine residues and it ranged from 8.54 to 22.50µgl^{-1}. However the range of residues concentration of the other detected organochlorine pesticides were as follow; $_{4,4}$DDD from 3.81 to 14.37 µgl^{-1}, $_{4,4}$DDT from 2.22 to 12.38 µgl^{-1}, $_{2,4}$DDT from 2.94 to 5.28 µgl^{-1}, HCH from 3.18 to 571 µgl^{-1}, $_{2,4}$DDD from 0.87 to 4.28 µgl^{-1} and the combination of Endosulfan and Endosulfan sulfate ranged from 0.05 to 9.18 µg/l [11]. Various studies have the presence of pesticide residue in water samples collected from cotton fields, rice growing fields and municipal areas. DDT and its metabolites were found in all areas but were not present in all samples. Concentration of pesticide residues varies from sample to sample and was in the range of 0.017-1.06 ng ml^{-1}. Overall, recoveries ranged from 84%-91% for all target pesticides [12].

To evaluate the level of contamination of ground water, a sampling campaign of tubewell water was carried out from cotton growing areas of Sirsa. The objective of this work is based on the identification of the residues of pesticides and their impacts on the quality of water.

2Materials and methods

2.1Study area

In Haryana, Sirsa (Panniwala) was selected as the study area for groundwater samples of cotton growing area. Within the city, sampling sites were selected using EICHER Map shown in Figure 1.

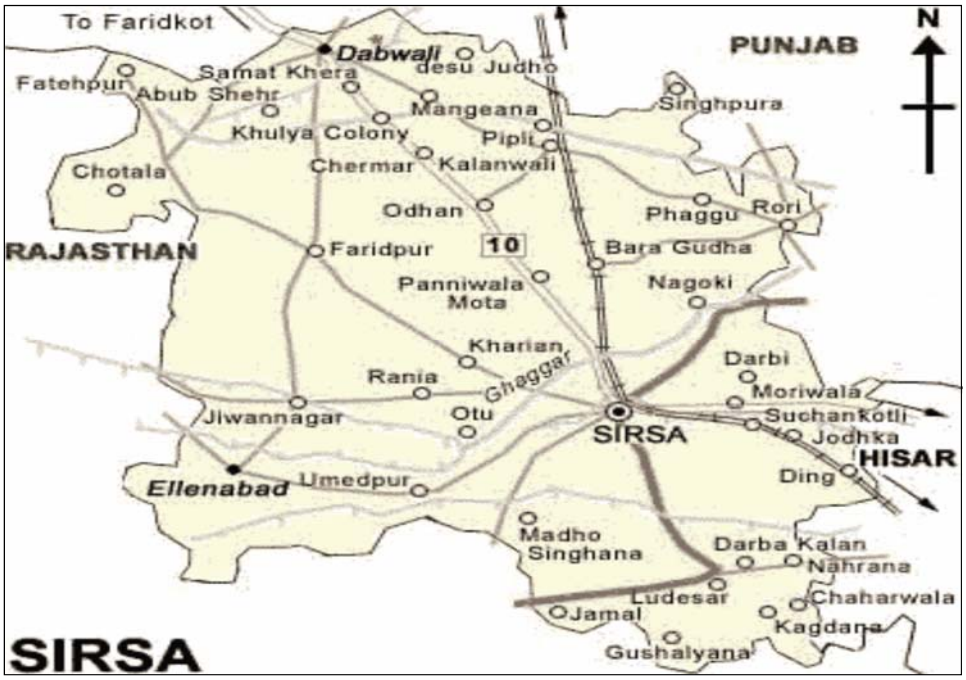

Fig 6: Haryana map depicting the areas (Punjuwan, Sirsa) from where Ghaggar River flows

2.2 Survey of crops grown and pesticide usage

Surveys with the farmers were carried out to get the information about the pesticides used in the crops of the study area as shown in Table 1.

Table 1: Survey of cotton grown and pesticide usage

Cotton Crop Varity	Pesticide
Desi Bt cotton(RG-8 and HD-123)	Monocrotophos, Chlorpyriphos
Desi Bt cotton(RG-8 and HD-123)	Endosulfan, Acetamopid
Desi Bt cotton(RG-8 and HD-123)	Monocrotophos, Pendimethalin
Desi Bt cotton(RG-8 and HD-123)	Imidacloprid, Acetamopid
Desi Bt cotton(RG-8 and HD-123)	Quinalaphos, Phorate
Desi Bt cotton(RG-8 and HD-123)	Endosulfan, Chlorpyriphos
Desi Bt cotton(RG-8 and HD-123)	Chlorpyriphos
Desi Bt cotton(RG-8 and HD-123)	Monocrotophos, Methyl parathion

2.3 Sampling

Groundwater samples were collected from eight tube wells of different cotton farm fields, periodically. The collected samples were kept in refrigerator till further analysis.

2.4 Chemical and solvents used

HPLC grade dichloromethane, HPLC grade n- hexane, HPLC grade ethyl acetate, sodium chloride (analytical grade) and sodium sulphate (analytical grade)

2.5 Extraction and clean-up: Pesticide residue extraction and clean-up was performed according to the standard protocols.

Water: 500 ml of water was taken in one litre separatory funnel. 10-15g sodium chloride was dissolved and extracted thrice (50, 25, 25ml) with 15% dichloromethane in hexane by vigorous shaking. Organic phase was collected, concentrated upto 10 ml and divided into two equal parts. One part was

taken for estimation of organochlorine pesticides and synthetic pyrethroids whereas the second part was processed for estimation of organophosphate pesticides. In both the cases, the solvent was evaporated to near dryness in the rotator evaporator and the residues were redissolved in n-hexane for organochlorine pesticides and synthetic pyrethroid and in ethyl acetate for organophosphate pesticides. Meticulous care was taken to remove traces of dichloromethane from the extract used for estimation of organochlorine pesticides and synthetic pyrethroid.

2.6 Estimation: Groundwater samples were analysed for 19 pesticides including 4 organochlorines (HCH and its four isomers, p,p' DDE, Endosulfan and its three isomers and Dicofol), six synthetic pyrethroids (Beta Cyfluthrin, Fenpropathrin, Lemda Cyhalothrin, AlphaCypermethrin, Deltamethrin, Fenvelerate) and nine organophosphate (Phorate, Dimethoate, Phosphamedion**,** Methyl Parathion**,** Malathion**,** Chlorpyriphos**,** Quinalaphos, Profenophos, Monocrotophos). The final extracts were analyzed on Shimadzu (GC model no. 2010) gas chromatograph equipped with capillary columns (280m x 280m x 175 mm) using Electron Capture Detector (ECD) and Flame Photometric Detector (FPD). Operating conditions were as per details: (1) for organochlorine pesticides and synthetic pyrethroids: detector ECD (^{63}Ni), capillary column (280m x 280m x 175 mm) with spiltless system. Temperature: initial temperature 170 ^{o} C and detector temperature 350^{o} C, carrier gas: Nitrogen gas, Flow rate: 1.0 ml/min. (2) Operating conditions for organophosphate pesticides: Detector: FPD, capillary column (280m x 280m x 175 mm), initial temperature 150 ^{o} C, and detector temperature 290^{o} C, carrier gas: Nitrogen gas, Flow rate: 1.0 ml/min.

For quality assurance, blank and laboratory control samples were run with each set of samples. Both were subjected to the same analytical procedure as those used on the study samples. Recoveries < 70% or >130% for laboratory control samples were promptly investigated and if necessary reanalyzed. Mixed standards were injected before and after the sample run. Appropriate quality assurance and quality control was performed including analysis of procedural blank (analyte concentrations were <MDL, method detection limit), random duplicate samples (standard deviation <5), and calibration curves with r^2 value of 0.999.

The instrument detection limits were established by using 3:1 signal to noise ratio to determine a peak as a valid quantifiable peak. Each sample was analyzed in duplicate and the average was used in analytical calculations. Calculated concentrations were reported as less than the limit of detection if the peak area did not exceed the specified threshold (three times the noise). Concentrations below the limit of detection were assigned zero values for the statistical analysis. Method detection limits were established by processing eight aliquots of the sample spiked with a quantity sufficient to produce a detectable response (s/n > 3) and multiplying the standard deviation by 3 (the $t_{student}$ value for eight replicates). MDL for organochlorine pesticides were 0.004µg/L, organophosphorous pesticides were 0.02µg/L but for phosphamedion and dimethoate MDL were 0.05µg/L, while for synthetic pyrethroids the MDL were 0.04µg/L respectively.

3 Result

Sampling area and location of groundwater on cotton growing areas in district Sirsa were illustrated in table 2.

Table 2: Sampling area and location of ground water in cotton growing area

Area		Water bodies			
Field size biga	**Location**	**Well**	**Tube wells**	**Pond**	**River**
2biga	Panniwala	-	1	-	Ghaggar
12biga	Panniwala	-	1	-	Ghaggar
10biga	Panniwala	-	1	-	Ghaggar
10biga	Panniwala	-	1	-	Ghaggar
7biga	Panniwala	-	1	-	Ghaggar
10biga	Panniwala	-	1	-	Ghaggar
1biga	Panniwala	-	1	-	Ghaggar
5biga	Panniwala	-	1	-	Ghaggar

Concentration of organochlorine pesticide residues in groundwater samples for cotton growing area are showed in table 3 (contents and cumulated contents of organochlorine pesticides in groundwater samples)

Table 3: Concentration of organochlorine pesticide in groundwater samples of cotton growing area

S/p µg/l	C_{TB-1}	C_{TB-2}	C_{TB-3}	C_{Tb-4}	C_{TB-5}	C_{TB-6}	C_{TB-7}	C_{TB-8}
α-HCH	0.131± 0.004	0.718± 0.004	0.023± 0.008	BDL	BDL	0.010± 0.006	0.032± 0.002	BDL
β-HCH	BDL	0.632± 0.007	0.961± 0.005	0.416± 0.003	0.324± 0.005	0.779± 0.008	0.058± 0.004	0.063± 0.006
γ-HCH	0.173± .006	0.190± 0.005	0.088± 0.004	BDL	0.012± 0.006	0.080± 0.004	0.059± 0.003	0.020± 0.003
δ-HCH	0.209± 0.004	0.040± 0.005	0.037± 0.009	BDL	0.059± 0.002	0.010± 0.006	0.030± 0.004	0.026± 0.009
Endosulfan-I	0.028± 0.006	0.056± 0.005	0.079± 0.004	0.034± 0.006	0.018± 0.009	0.017± 0.009	BDL	0.021± 0.005
Endosulfan-I I	0.547± 0.005	0.007± 0.004	0.011± 0.006	0.827± 0.006	0.946± 0.004	0.010± 0.006	0.979± 0.003	1.055± 0.006
Endosulfansulfate	0.516± 0.003	0.011± 0.006	0.714± 0.003	0.009± 0.004	0.008± 0.004	0.118± 0.004	0.251± 0.003	0.015± 0.007
Dicofol	0.413± 0.002	0.117± 0.06	0.514± 0.003	0.010± 0.005	0.216± 0.001	0.007± 0.003	0.124± 0.008	0.210± 0.002
p, p'-DDE	0.310± 0.004	0.069± 0.004	0.222± 0.007	0.005± 0.002	0.010± 0.005	0.211± 0.005	0.280± 0.003	0.468± 0.005

S/P= sample code/parameters; $C_{TB(1\ TO\ 8)}$ = groundwater samples from tubewell; BDL= below detectable limit; Minimum Detection limit of all organochlorine pesticide is: 0.004 µg/l

Similarly the concentration of organophosphate, synthetic pyrethroid residues in groundwater samples has been illustrated in table 4, 5.

Table 4: Concentration of organophosphate pesticide in groundwater samples of cotton growing area

S/P µg/l	C_{TB-1}	C_{TB2}	C_{TB-3}	C_{Tb-4}	C_{TB-5}	C_{TB-6}	C_{TB-7}	C_{TB-8}
Phorate	0.414± 0.001	0.058± 0.005	0.321± 0.006	0.009± 0.003	0.018± 0.003	0.915± 0.002	0.022± 0.006	1.010± 0.003
Dimethoate	0.614± 0.006	0.783± 0.005	0.536± 0.006	0.882± 0.006	0.916± 0.003	0.718± 0.003	0.618± 0.004	0.942± 0.008
Malathion	0.143± 0.008	0.009± 0.005	0.004± 0.001	0.044± 0.007	0.011± 0.006	0.020± 0.004	0.010± 0.006	0.009± 0.004
Chlorpyriphos	0.882± 0.006	1.969± 0.003	1.618± 0.002	0.746± 0.009	0.626± 0.003	0.910± 0.004	0.913± 0.002	1.109± 0.005
Quinalaphos	0.187± 0.005	0.217± 0.003	0.419± 0.006	0.182± 0.005	0.251± 0.005	0.364± 0.007	0.281± 0.006	0.452± 0.006
Profenophos	0.075± 0.003	1.09± 0.003	1.039± 0.004	2.332± 0.005	0.018± 0.003	2.915± 0.003	0.615± 0.002	0.004± 0.003
Monocrotophos	BDL	0.615± 0.003	2.732± 0.005	BDL	0.623± 0.002	BDL	0.007± 0.003	4.364± 0.005
Phosphamedion	0.228± 0.004	0.213± 0.003	0.287± 0.003	0.181± 0.005	0.312± 0.008	0.277± 0.006	0.358± 0.004	0.382± 0.005
Methyl Parathion	0.230± 0.004	0.146± 0.007	0.284± 0.004	0.178± 0.005	0.314± 0.002	0.278± 0.005	0.358± 0.003	0.382± 0.005

S/P= sample code/parameters; $C_{TB(1\ TO\ 8)}$ = groundwater samples from tubewell; BDL= below detectable limit; Minimum Detection limit of organophosphorous pesticide is: 0.002 µg/l except Phosphamedion and Quinalaphos: 0.005 µg/l

Table 5: Concentration of synthetic pyrethroids in groundwater samples of cotton growing area

S/P µg/l	C_{TB-1}	C_{TB2}	C_{TB-3}	C_{Tb-4}	C_{TB-5}	C_{TB-6}	C_{TB-7}	C_{TB-8}
Beta Cyfluthrin	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Fenpropathrin	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Lemda Cyhalothrin	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Alpha Cypermethrin	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Deltamethrin	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Fenvelerate	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL

S/P= sample code/parameters; $C_{TB(1\ TO\ 8)}$ = groundwater samples from tubewell; BDL= below detectable limit; Minimum Detection limit of synthetic pyrethroids is: β Cyfluthrin: 0.06 µg/l, Fenpropathrin: 0.05 µg/l, Lemda Cyhalothrin: 0.06 µg/l, α Cypermethrin: 0.07 µg/l, Deltamethrin: 0. 05µg/l, Fenvelerate: 0.03 µg/l

The analysis of the groundwater samples collected from the eight tubewells of cotton growing areas of Sirsa showed that residues of eighteen pesticides were detected in approximately the totality of the tubewells sampled close to the cotton plantations which indicate that almost, all the eight tubewells were contaminated with pesticides. The detected residues of eighteen pesticides including: organochlorine, organophosphate, with cumulated contents of organochlorine pesticides reaching a mean concentration range of 1.055µg/L, similarly for organophosphate the range was 4.364µg/L. On the contrary, synthetic pyrethroids, owing to their biodegradable property, were found below degradable limit and hence can be considered as safe pesticides.

4. Discussion

All the molecules of pesticides analyzed were detected and the lowest concentration recorded was 0.005µg/L for organochlorine pesticides, 0.004µg/L for organophosphate. For some pesticides molecules like endosulfan II, the concentration largely exceeds the permissive limit which is for endosulfan-II (1.055±0.006µg/L), monocrotophos (4.364±0.005µg/L).

Additionally, other insecticides such as DDT and its metabolites were also found in the analyzed samples. The concentrations recorded for organochlorine were relatively in the range (1.055µg/L); they were largely above the maximum permissive limit (0.004µg/L), similarly organophosphate range 4.364µg/L and the maximum permissive limit (0.002µg/L). Average contents of HCH, one of the isomers of lindane, which was detected in all groundwater sample, were higher than the standard limit 0.004µg/L. In this study endosulfan I and II and their metabolite endosulfan sulfate were detected in all the groundwater samples. The maximum concentrations of endosulfan I and endosulfan II measured in all the samples were respectively 0.079µg/L and 1.055µg/L. With regard to endosulfan sulfate, the concentration was 0.714µg/L in the C_{fb-3} sample of groundwater. Their presence in all groundwater samples indicates the use of these products despite the restrictions of their use. In addition, the persistence and reminisce of certain pesticides cannot alone explain the values recorded even if several concentrations do not exceed the standard for drinking water. This concentration is 92% higher than the permissive value which is 0.004µg/L. the other found residues of pesticides were 82% higher than the permissive value 0.004µg/L. The organophosphate pesticides present the average concentrations highest in monocrotophos sample, all field together taken was (0.615±4.364µg/L). The levels of these concentrations, could find their explanations in the abusive frequent use of these active molecules for the treatment of the cultures [13]. Their detection in the medium, despite their capacity of being degraded quickly in the environment [14] represented not only their strong use but also the proximity of the plantations. Organochlorine pesticides were also present in all contaminated groundwater. The mean concentration in the sample were 0.012 to 0.961µg/L for lindane, 0.008 to 1.055µg/L for endosulfan, 0.007 to

0.514µg/L for dicofol and 0.005 to 0.468µg/L for pp' DDE in eight different tubewells of cotton growing fields. The detection of these chemicals in the groundwater samples, in spite of various bans on their use [15, 16] could be due to their persistence in the environment [17] or to a deceptive use. The residues of pesticides are omni present in the majority of the samples. Approximately, some of the detected pesticides are prohibited since years [18] which indicate that the stability of these chemicals in the environment and persist ability in the soil and that they infiltrate slowly in groundwater. In fact in the case of organochlorine (endosulfan, lindane, DDT) pre-prevails in water of the cotton fields. Elsewhere, the most remarkable pesticides are organophosphate. This shows that in the fields, the cultures are very diverse [19] and occupy of large surfaces and testifies too many plant health problems. Also they receive very intensive chemical treatments, in particular with the Nematicides and fungicides, the weed killers. The results of our analyses show that the subsoil water was of good quality contrary to the traditional belief which supposes that the ground removes from water all its contaminatants at the time of its infiltration towards the underground table.

5. Conclusion

The analyses carried out on tubewell water showed a contamination of groundwater by the pesticides with low disparities of contamination according to the zone of culture considered, even if some tubewells were contaminated than others. Concentrations sometimes are higher than the maximum residue limits relative to the drinking water. This report constitutes one of the main public health problems in the study area, because of the cumulative effects of these products on health. It is a growing concern, knowing that water is consumed in great quantity. There is thus a real risk to health. And this risk is higher as the molecules present were used more and more in significant quantity in the zone of studies and also stable in water. Added to this, the risk related to the vulgarizing made of the use of pesticides. It is advisable to recall that the most effective means to reduce the health risk remains the decline of the use of chemicals.

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