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USE, CONTAMINATION AND EXPOSURE OF PESTICIDES IN PAKISTAN: A REVIEW

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Pesticides contamination is a global environmental and public health concern due to their carcinogenic and highly toxic nature for all kind of living organisms including plants, animals and humans. Pakistan as an agriculture-based country, annually use a huge amount of organochlorine pesticides (OC) to control insect pests and different diseases of crops, resulting in environmental contaminations and their subsequent exposure to humans. Unfortunately, more than half million people are endured annually from OC pesticides and other agro-chemicals poisoning in Pakistan. However, recent data on the uses, contamination and exposure of pesticides in Pakistan are largely unavailable. This review summarizes the recent information and provides data on consumption and contamination of pesticides with a particular focus on their exposure to humans in Pakistan. Studies must be conducted to precisely determine the current use, contamination level and exposure of pesticides in Pakistan.

Keywords: Pesticides, pollution, vulnerability, environment, Pakistan.

INTRODUCTION

Pesticides consist of various compounds for wide range applications such as herbicides, nematicides, insecticides, molluscicides, fungicides, rodenticides and plant growth regulators. Due to widespread uses and toxic nature of pesticides (Bashir *et al.*, 2018), these are pollutants of a highly global concern (Damalas and Eleftherohorinos, 2011). It has the highest potential in spreading cancer risk in humans. Approximately 98.0% cases of cancer and 94.0% patients with other health issues are stated to contain enough concentration of organochlorine pesticides (OC) in their serum (Attaullah *et al*., 2016). Moreover, the risk extrapolation data of both animals and humans showed that in developing countries, 37,000 cases per year of cancer have direct or indirect relationship with pesticides applications (WHO, 1990). According to the reports of WHO (2014), from 2003 to 2014-worldwide poisoning cases have been increased from 10.0 to 14.0 million people and majority of the cases belonged to the developing countries (McGuire, 2016).

Pesticides are classified according to their hazardous levels by WHO (2009) (Table 1), unfortunately, the most of applied pesticides fall in the category of highly poisonous (34.2%) and moderate poisonous (35.0%) (Damalas and Khan, 2017). The high intoxications of these pesticides appear because of inappropriate application technology, knowledge, and attitudes towards their applications (Koizumi *et al*., 2017). Farmers' poor knowledge about pesticides applications or absence of protective measures before or during application has increased their risk and exposure in environment (Khan *et al*., 2015). About 58.0% farmers have moderate levels of knowledge about the pesticide application, only 16.0% farmers are trained to use pesticide and merely 13.0% farmers have access to the related information (Damalas and Khan, 2017).

The worldwide utilization of pesticides is approximately 5.6 billion pounds per year and their consumption is increasing unexpectedly (Alavanja, 2009), which is about 45.0%, 25.0% and 25.0% in Europe, USA and rest of the world, respectively (Bourguet and Guillemaud, 2016). China and USA are among the top pesticides producers in the world. From the South Asian countries, Pakistan ranks second in the overall consumption of pesticides and major use of these pesticides is in agriculture sector (Figure 1) (Yadav *et al.*, 2015; Waheed *et al.*, 2017).

Unfortunately, a huge amount of pesticides was applied during 1980s to 1990s throughout agriculture area of Punjab, Pakistan. The sale and distribution of pesticides were transferred from public to private sector which increased consumption by five-time in one year with no significant effect on crop yield. Currently, more than 30 types of fungicides, 5 types of acaricides, 39 types of weedicides, 6 different types of rodenticides, and 108 types of insecticides are being used in Pakistan (PPSGDP, 2002; Mehmood *et al*., 2017). The previous data show that more than 69.0% pesticides were used for cotton crop only (Figure 2) and the

Table 1. Classification of pesticides according to their hazardous levels and concentrations (WHO, 2009)

Routes of exposure	Extremely hazardous Ia	Highly hazardous Ib	Moderately hazardous II	Slightly hazardous III	Unlikely to present acute hazard U	Obsolete as pesticide, not classified O
Oral LD_{50}	< 5 mg/kg	Up to and including 50 mg/kg	50-500 mg/kg	500-500 mg/kg	> 5000 mg/kg	NA
Inhalation LD_{50}	< 0.2 mg/kg	Up to and including 0.2 mg/L	0.2-2 mg/L	2-20 mg/L	> 20 mg/L	NA
Dermal LD_{50}	< 200 mg/kg	Up to and including 200 mg/kg	200-2000 mg/kg	2000-20000 mg/kg	> 20000 mg/kg	NA
Eye effects	Corrosive corneal opacity not reversible	Corrosive corneal opacity not reversible within 7 days	Corneal opacity reversible within 7 days; irritation persisting for 7 days	No corneal opacity; irritation reversible within 7 days	No irritation	NA
Skin effects	Corrosive	Corrosive	Severe irritation	Moderate irritation	Mild or slight irritation	NA
Signal word	Danger poison	Danger poison	Warning	Caution	Caution	NA

LD_{50}: Lethal dose per body weight (150 lb.) to infect 50% population, NA: Use for non-agriculture purpose but their concentration influences normal mechanism.

remaining 31.0% is used for other crops such as sugarcane, rice, tobacco, paddy, maize, fruits and vegetables (Hakeem *et al*., 2016; Randhawa *et al*., 2016).

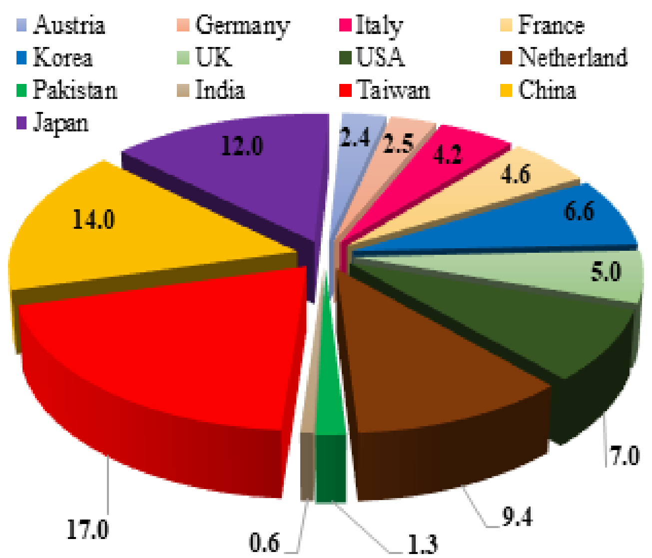


Figure 1. Pesticide consumption in Pakistan and other countries (Modified from Yadav *et al.*, 2015)

The use of pesticides was increased by 1169% in last 20 years with more than 10 sprays per crop season which become the alarming situation for human beings (Ejaz et al., 2004; Khwaja *et al*., 2013). Although, import of pesticides has been reduced significantly by 2016-17 (Table 2), their demands and uses remained high which affected the yield of the major crops to some extent. The high application of pesticides caused accumulation of toxic active ingredients in the food chain (Damalas and Khan, 2017).

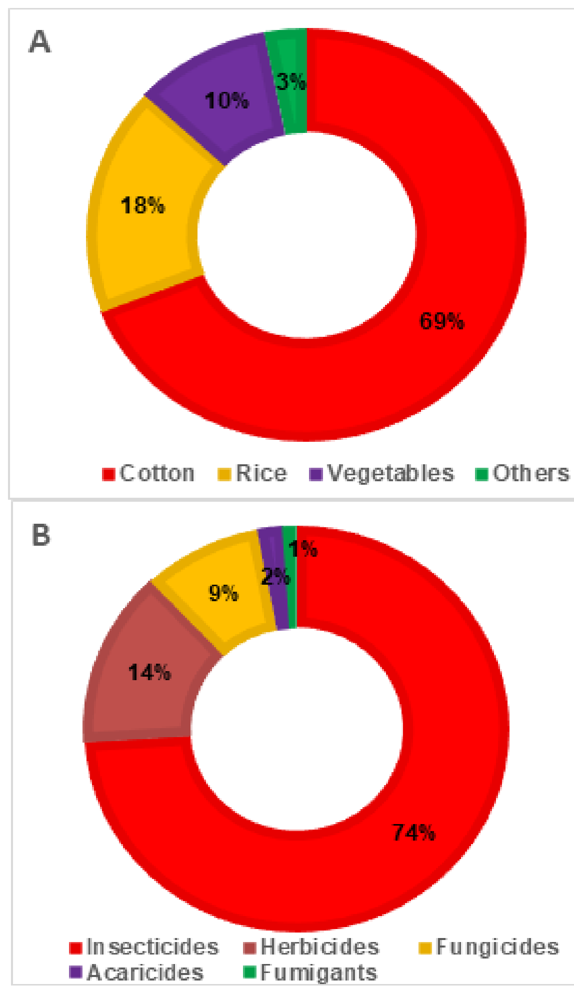


Figure 2. Use (%) and share (%) of different pesticides in Pakistan (modified from Hakeem *et al.*, 2016)

Table 2. Cotton, area, production, yield and pesticides imported or consumed.

Year	Area (thousand hectares)	Production (thousand bales)	Yield (kg/ha)	Total pesticides imported (tons)
2000-01	2927	10732	570	21255
2001-02	3116	10613	579	31783
2002-03	2794	10211	622	22242
2003-04	2989	10048	572	41406
2004-05	3193	14265	760	41561
2005-06	3096	12417	682	33954
2006-07	3075	12856	711	29089
2007-08	3054	11655	649	27814
2008-09	2820	11819	713	28839
2009-10	3106	12913	695	38227
2010-11	2689	11460	725	36183
2011-12	2835	13595	815	32152
2012-13	2879	13031	769	17882
2013-14	2806	12769	774	23546
2014-15	2961	13960	802	23157
2015-16	2902	9917	582	17386
2016-17	2489	10671	730	12806

Source: Economic Survey of Pakistan, 2009-2017

The prevailing situations reveal that the developing countries such as Pakistan may have the higher pesticides poisoning cases than reported due to misdiagnosis and absence of information (Tariq, 2005) and there are significant occupational exposures of pesticides despite their strict regulations. Here, the main objective of this review is to summarize the all possible uses, recent trend in consumption of pesticides, contamination levels and exposure of pesticides in Pakistan and to plan the future research on the basis of current knowledge in this area.

***Pesticides use*:** Historically, among different pesticides, dichloro-diphenyl-trichloroethane (DDT) is the first organo-pesticide which was prepared in the laboratory as insecticide in 1874. DDT is a colorless and crystalline solid which has tendency to dissolve only in organic solvents. It accumulates in fats as bio-accumulative toxin and thus enters and remain in the food chain (Brevik *et al*., 1996). Due to its hazardous effects, it was banned in the European countries (i.e. Norway and Sweden) in 1970 and in the United Sates in 1972. A treaty was also signed among 170 countries known as Stockholm Convention on persistent organic pollutants (POPs) to resist the use of DDT (Lallanilla, 2017). Hydrocyanic acid, carbon bisulfide, arsenicals were first reported chemicals used as weedicide and fungicide after 1886 (Aktar *et al*., 2009) and their restrictions to use in technologically advanced countries were also reported in 1960 (NRC, 2000). Some hazardous pesticides were also restricted in the developing countries due to their poisonous impact but at the same time these toxic pesticides are being used in these countries (Sankararamakrishnan *et al*., 2005). In Pakistan, pesticides were standardized by the Federal Government through Department of Plant Protection (DPP) before 1971. However, the agricultural pesticide rules and agricultural pesticides ordinance (APO) were promulgated in 1971 and 1973 to regulate registration and standardization of pesticides as well as formulation, production, imports, sale, distribution, and their uses in the country (Mazari, 2005).

Although, in Pakistan, the pesticide usage is not very high (Figure 1) as compared to total cultivated area, insecticides are extensively used in the cotton areas (Iqbal *et al*., 2001). Several reports show that pests attack have been reducing annul cotton yield by 30-40% but less reduction rate has observed in the genetically modified BT (*Bacillus thuringiensis*) cotton (Tariq, 2005; Malik and Ahsan, 2016). To avoid further loss in upcoming years, Government of Pakistan in collaboration with pesticides industries introduced an act of high use of pesticide in monoculture crop which eventually increased the cotton yield during year 2000-2001 (Mazari, 2005; Tariq, 2005; Economic Survey of Pakistan, 2005-6). According to the economic survey of Pakistan, significant relationships exist between the total cotton cultivated area vs its yield (R^2 = 0.58) and total area under pesticide consumption (R^2 = 0.52). However, a weak relationship exists between the total pesticide usage and total yield (R^2 = 0.22). This weak relationship shows that the current usage of pesticide by the farmers was continued even after the resolution of the low yield issue. Farmers used higher doses of pesticide (8 to 13 sprays per season) than the recommendation (Tariq *et al*., 2004a). The use of different pesticides (i.e. cypermethrin, methamidophos, monocrotophos, and dimethorate) was increased many folds (19-725 fold) to control ballworm (*Helicoverpa armigera*) and whitefly (*Bemisia tabaci*) (Tariq *et al*., 2007).

In addition, higher usage of pesticides or insecticides resulted in development of resistance in insect-pests against pesticides making the situation worst and challenging regarding human health and environmental protection. Results of several studies clearly revealed development of resistance in two major cotton pests, American bollworm and whitefly against the common pesticides at Central Cotton Research Institute (CCRI), Multan (Ahmad *et al*., 2002a, 2003, 2007). These heavy usages of pesticides were an indication of treadmill of pesticides according to the Ministry of Food and Agriculture, Pakistan. In 1983-1997, 5.0 to 10.0% cotton area of Punjab was applied with these pesticides (Tariq, 2005), but this consumption has been raised up to 80.0 to 90.0% in 2008 (Khooharo *et al*., 2008).

Pesticides contamination

***Surface water and ground water contamination*:** Green revolution had brought the fundamental application of pesticides to control the insect pests of plants (Palikhe, 2003). The harmful impacts of higher pesticides use have converted the advantages of chemistry to disadvantages due to its interference and inclusion in the food chain particularly in sources of drinking water (Tariq *et al*., 2003). Water, the fundamental requirement of living beings, is being severely polluted due to several anthropogenic activities and natural processes (Hussain and Asi, 2008). The pesticides concentration in the water reaches up to dozens of mg/L and interlinked with the heavy watering and fertilization in the field. The vigorous plant growth attracting the competitors increase the pesticide requirements (Gurr, 2016).

Pesticides contents in different types of water were found to be in following increasing order: cropland water > field ditch water > runoff > pond water > groundwater > river water > deep groundwater > sea water (Zhang *et al*., 2011). Pesticides contamination in surface water and ground water is controlled by point (municipal and industrial wastes) and nonpoint (erosion, seepage and runoff) sources (Tankiewicz *et al*., 2010). The level or degree of contamination can be determined with three factors; (1) presence and levels of the pesticide, (2) mobility of pesticides in soil, and (3) the carrying agents of pesticides especially water in the field (Mensah *et al*., 2014; Anderson *et al*., 2018).

Natural agents or nonpoint sources that support the pesticide contamination also harm the aquatic life by polluting the surface water including rivers, lakes, and sea. Toxicity, exposure time, amount, and their persistence in the surface waters have disturbed the aquatic food chains (Helfrich *et al*., 2009). The accumulation of contamination in water and fish's tissues causes the adverse effects on the human health due to consumption of aquatic products, particularly by people inhabiting and traveling to watershed lands (Lee *et al*., 2014).

In Pakistan, some studies reported on pesticides contamination and/or presence of different pesticides (Table 3) in surface water (Parveen and Masud, 1988b; Ahad *et al*., 2006), shallow water (Ali and Jabbar, 1992; Tariq *et al*., 2004a; Muhammad *et al*., 2015; Ali *et al*., 2016) and in fresh water sources (Mahmood *et al*., 2014b; Mahboob *et al*., 2014; Ali *et al*., 2016). Cotton areas of Sindh plains and southeastern Punjab were found to be the most affected ones (Ali and Jabbar, 1992; Tariq *et al*., 2004a; Memon *et al*., 2016).

Rawal Lake, which was became press news due to its fish killing disasters, is situated in the capital city of Pakistan (Islamabad) and its water is supplied and used in Rawalpindi city after some treatments. The water samples from this lake were analyzed by different research groups and found the presence of up to 4-time higher levels of residues of pyrethroids pesticides than the Environment- European Commission (EEC) standards for drinking water. The pesticide in the lake was intentionally thrown by some miscreant for catching the fish (Ahad *et al*., 2006).

Table 4 shows the summary of water pollution data which is sorted out to show historical change in water resources. These findings show the pollutant capabilities and the presence of contaminations due to many factors including soil characteristics, shallow water tables, and intensive sprayings (Jabbar *et al*., 1993; Tariq *et al*., 2004a, b, 2006) and their infiltration through heavy and intensive water irrigation (Flury *et al*., 1994). Careless disposal of pesticides containers, point and nonpoint sources are the direct factors through which the aquatic environment is being contaminated (Ahad *et al*., 2000, 2001; Tariq *et al*., 2004a, 2006).

***Soil contamination*:** Soils provides a long-term storage compartment to pesticides with organic carbon due to its buffer and filter capacity, and degradation potential (Burauel and Bassmann, 2005). Pesticides in the soil expose through direct application in the field or through indirect means such as accidental leakage or spillage and run-off through plant surface, undergo the fates such as chemical, photochemical and microbial degradation, volatilization, adsorption, movement and uptakes by organisms (Bailey and White, 1970; Rashid *et al*., 2010) (Figure 3).

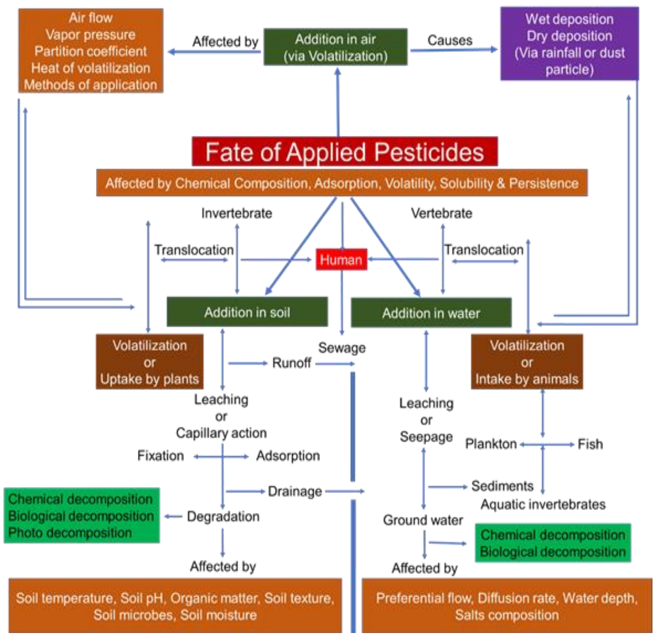
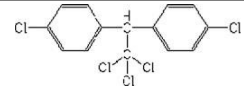
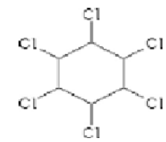
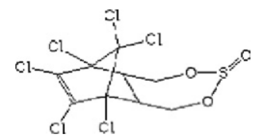
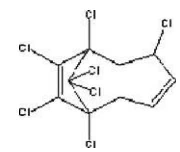
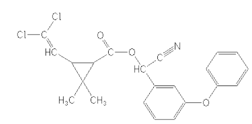
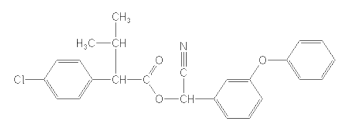
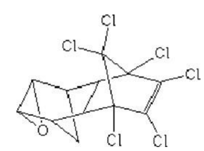
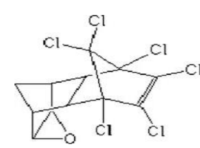
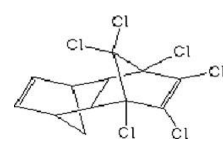
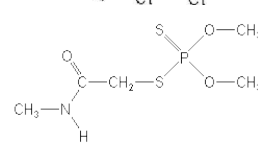
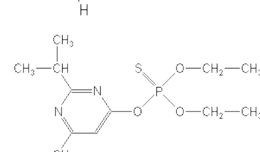


Figure 3. Fate of applied pesticides in soil, air and water

Soil is known to have potential pathway of pesticides transport to pollute the food, air, plants and particularly water which ultimately affects the living organisms including human. These contaminations supported by soil properties including runoff, interflow, leaching, soil erosion, subsurface drainage and transportation of the pesticides with mineral nutrients move into plant and animal's body to become constituents of food chain (Abrahams, 2002; Nakata *et al*., 2002) (Figure 3).

Pesticides in the soil have persistent behavior and degraded slowly by soil biota and thus become source of pollution (Stephenson and Solomon, 1993). The contamination sources are mainly linked with human activities such as insecticides use and other chlorinated chemicals applications, deforestation, industrial and domestic discharge that increased the soil erosion (Bhattacharya *et al*., 2003).

Table 3. Pesticides information, detected from different samples of surface and ground water and soil (modified from Tariq *et al.*, 2007)

Sr.#	Common name	IUPAC name	Structural formula	Case No.
1	DDT ($C_{14}H_9Cl_5$)	1,1,1-Trichloro-2,2-bis(4-chlorophenyl) ethane		50-29-3
2	HCH, ($C_6H_6Cl_6$)	Benzene hexachloride		608-73-1
3	Endosulfan ($C_9H_6Cl_6O_3S$)	6,7,8,9,10,10-Hexachloro-1,5,5a,6,9,9a-hexahydro-6,9-methano-2,4, 3-benzodioxathiepin 3-oxide		115-29-7
4	Heptachlor ($C_{10}H_5Cl_7$)	1,4,5,6,7,8,8-Heptachloro-3a,4,7,7a-tetrahydro-4,7-methanoindene		76-44-8
5	Cypermethrin ($C_{22}H_{19}Cl_2NO_3$)	(RS)-α-cyano-3-phenoxybenzyl (1RS,3RS;1RS,3SR)-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate		52315-07-8
6	Fenvalerate ($C_{25}H_{22}ClNO_3$)	(αRS)-α-cyano-3-phenoxybenzyl (2RS)-2-(4-chlorophenyl)-3-methylbutyrate		51630-58-1
7	Dieldrin ($C_{12}H_8Cl_6O$)	1,2,3,4,10,10-Hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-endo-1, 4-exo-5,8-dimethanonaphthalene		60-57-1
8	Endrin ($C_{12}H_8Cl_6O$)	1,2,3,4,10,10-Hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-exo-1, 4-exo-5,8-dimethanonaphthalene		72-20-8
9	Aldrin ($C_{12}H_8Cl_6$)	(1R,4S,4aS,5S,8R,8aR)-1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1, 4:5,8-dimethanonaphthalene		309-00-2
10	Dimethoate ($C_5H_{12}NO_3PS_2$)	O, O-dimethyl S-methylcarbamoylmethyl phosphorodithioate		60-51-5
11	Diazinon $C_{12}H_{21}N_2O_3PS$)	O, O-diethyl O-2-isopropyl-6-methylpyrimidin-4-yl phosphorothioate		333-41-5

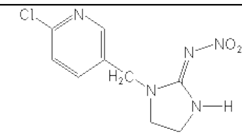
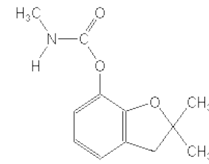
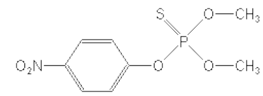
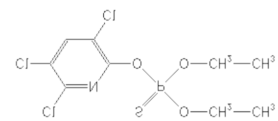
Sr.#	Common name	IUPAC name	Structural formula	Case No.
12	Imidacloprid ($C_9H_{10}ClN_5O_2$)	(E)-1-(6-chloro-3-pyridylmethyl)-N-nitroimidazolidin-2-ylideneamine		138261-41-3
13	Carbofuran ($C_{12}H_{15}NO_3$)	2,3-dihydro-2,2-dimethylbenzofuran-7-yl methylcarbamate		1563-66-2
14	Parathion-methyle ($C_8H_{10}NO_5PS$)	O, O-dimethyl O-4-nitrophenyl phosphorothioate		298-00-0
15	Chlorpyrifos ($C_9H_{11}Cl_3NO_3PS$)	O, O-diethyl O-3,5,6-trichloro-2-pyridyl phosphorothioate		5598-13-0

Table 4. Pesticides detected in surface waters and ground waters of different areas of Pakistan with their hazardous classes

Location	Year	Detected	Concentration (µg/L)	CASRN	Detection Technique	LD_{50} mg/kg	Classes	Reference
Coastal areas,	1990	DDT	2.7-9.2	50-29-3	GC (ECD)	113	II	Bano and Siddique (1991)
		α-BHC	0.06-0.2	58-89-9		88	II	
		Heptachlor	1.1-3.5	76-44-8		NA	O	
		Dieldrin	2.7-9.2	60-57-1		NA	O	
Cropland (Faisalabad, Kala Shah Kaku, Samundri)	1990	Aldrin	0.001-0.002	309-00-2	GC (ECD)	NA	O	Ali and Jabbar (1992), Jabbar *et al.* (1993)
		Cyhalothrin	0.003-0.01	68085-85-8		144	II	
		Cypermethrin	0.4	52315-07-8		250	II	
		Dieldrin	Traces-0.0002	60-57-1		NA	O	
		Dimethoate	Traces-0.002	60-51-5		150	II	
		DDT	0.0002-0.0006	50-29-3		113	II	
		Endrin	Traces	72-20-8		NA	O	
		Fenvalerate	0.3-0.6	51630-58-1		NA	II	
		Monocrotophos	Traces	6923-22-4		87	Ib	
Deg Nullah, Lahore	1991	o,p′DDT	0.2-1.1	50-29-3	GC (ECD)	113	II	Tehseen *et al.* (1994)
		p,p′-DDT	0.006-2.4	NA		NA	NA	
		o,p′-DDD	0.2-2.0	NA		NA	NA	
		p,p′-DDD	0.6-2.3	NA		NA	NA	
		o,p′-DDE	0.002-0.3	NA		NA	NA	
		p,p′-DDE	0.0008-0.8	NA		NA	NA	
		HCB	0.0001-0.09	118-74-1		NA	NA	
		BHC	Traces	58-89-9		88	II	
Swabi, Chota Lahore, Takhbai, Mardan (Mardan division)	2000	Chlorpyrifos	0.0-0.03	5598-13-0	GC (ECD)	>300	II	Ahad *et al.* (2000)
		Dichlorvos	0.03-0.5	62-73-7		56	Ib	
		Dimethoate	0.0-0.2	60-51-5		150	II	
Cotton growing area of Multan (Punjab)	2001	Carbofuran	0.0-0.3	1563-66-2	GC (ECD)	8	Ib	Ahad *et al.* (2001)
		Diazinon	0.0-0.03	333-41-5		300	II	
		Dichlorvos	0.0-0.2	62-73-7		56	Ib	
		Esfenvalerate	0.01-0.2	66230-04-4		87	II	
		Lindane	0.0-0.1	58-89-9		88	II	
Haleji Lake, Thatta (Sindh)	1999	HCB	0.4-1.7	118-74-1	GC (ECD)	NA	NA	Sanpera *et al.* (2002)
		HCH	Traces	608-73-1		100	II	
		DDT	0.0-6.5	50-29-3		113	II	
		PCB	Traces	-		NA	NA	

Location	Year	Detected	Concentration (µg/L)	CASRN	Detection Technique	LD_{50} mg/kg	Classes	Reference
Bahawalnagar, D.G. Khan, Muzafargarh, Rajanpur	2004	Bifenthrin	4.3	82657-04-3	GC (ECD)	55	II	Tariq *et al.* (2004a)
		λ- Cyhalothrin	2.9	68085-85-8		144	II	
		Carbofuran	23.1	1563-66-2		8	Ib	
		Endosulfan	2.8	115-29-7		80	II	
		Monocrotophos	8.3	6923-22-4		14	Ib	
Rawal Lake, Islamabad (Capital of Pakistan)	2006	Azinophos-methyle	0.06-13.3	-	GC (ECD)	NA	NA	Ahad *et al.* (2006)
		Fenitrothion	0.08-8.3	122-14-5		503	II	
		Parathion-methyle	0.02-2.7	298-00-0		13	Ia	
		α-Cypermethrin	0.2-18.8	68085-85-8		144	II	
Samples collected from various water sources in different districts of Punjab	2008	o,p′-DDD	0-0.8	NA	HPLC	NA	NA	Asi *et al.* (2008)
		o,p′-DDE	0-0.7	NA		NA	NA	
		o,p′-DDT	0-0.9	50-29-3		113	II	
		p,p′-DDE	0-0.2	NA		NA	NA	
		p,p′-DDT	0-1.1	NA		NA	NA	
Water samples from nearby area of demolished DDT factory, Amangarh, Nowshera (KP)	2009	p,p′-DDT	70-400	NA	GC-14A	NA	NA	Jan *et al.* (2009)
Samples collected at different spots from Rawal Lake and Simly Lake, Islamabad	2009	2,4-DDT	0.9-2.9	50-29-3	GC (ECD)	113	II	Iram *et al.* (2009)
		4,4-DDD	0.8-2.4	NA		NA	NA	
		Alpha-HCH	0.8	608-73-1		100	II	
		Azinphos-methyl	1.5-1.8	NA		NA	NA	
		Diazinon	1.8-3.6	333-41-5		300	II	
		Endosulfan	0.7-0.7	115-29-7		80	II	
		Fenitrothion	2.1-2.3	122-14-5		503	II	
Southern Punjab	2012	Imidacloprid	0.6	138261-41-3	HPLC	450	II	Baig *et al.* (2012)

GC (ECD): Gas Chromatography-Electron Capture Detector, HPLC: High-Performance Liquid Chromatography, GC-14A: Gas Chromatography series 2014, LD50: Lethal dose per body weight (150 lb.) to infect 50% population, CASRN: Chemical Abstracts Service Registry Number, NA: Not available

Several research studies have been conducted in Pakistan for detecting the pesticide in soils (Bano and Siddique, 1991; Jabbar *et al*., 1993; Tehseen *et al*., 1994; Sanpera *et al*., 2002; Tariq *et al*., 2006; Syed *et al*., 2013; Syed *et al*., 2014; Bajwa *et al*., 2016; Aamir *et al*., 2017). These studies have quite similar results as the findings of studies conducted by the researchers from the developed world (Masiá *et al*., 2015; Chakraborty *et al*., 2015; Sánchez-Osorio *et al*., 2017). Table 5 summarizes data regarding the different types of chlorinated pesticides (organochlorine) detected from the soil or sediments of different areas of Pakistan.

In various studies of water and soil contamination, the buffering and degradation potential of different soil series were also calculated on the basis of half-lives of pesticides and soil organic carbon sorption coefficient (K_{oc}) (Tariq *et al*., 2004b; Marín-Benito *et al*., 2017). The K_{oc} values of these studies are inconsistent with those of Wauchope *et al*. (1992) and Tomlin (1997). These variability and inconsistencies in the results could be due to several possible reasons such as changing and variable nature of soil organic matter (SOM) (Ahmad *et al*., 2001b; Spark and Swift, 2002), clay contents (Li *et al*., 2003; Sanchez-Martin *et al*., 2006), soil pH (Sheng *et al*., 2005). Solid-state CP/MAS ^{13}C NMR (Cross-Polarization Magic Angle Spinning Carbon-13 Nuclear Magnetic Resonance) was used to analyze the different soil samples (Ahmad *et al*., 2001a) to observe the relationship of K_{oc} values of different pesticides with SOM, supposing its structural composition.

The results showed a linear positive relationship between aromatic SOM and K_{oc} values and further revealed that aromatic SOM has potential to bind the non-ionic pesticides (Spark and Swift, 2002). The binding ability of pesticides in soil also varies due to the clay contents in soil and soil pH (Sanchez-Martin *et al*., 2006; Sheng *et al*., 2005). Furthermore, persistence and hydrophobicity, the two basic properties of pesticides, have also been studied and found to control the accumulation of pesticides in various soil series of Pakistan depending on the charge density of organic cations and clay minerals (Tariq *et al*., 2004b, 2006; Sanchez-Martin *et al*., 2006). Results showed that if the solubility of pesticides is less than 1mg/L in water, octanol–water partition coefficient (K_{ow}) is more than 1000 and half-life in soil is longer than 30 days then they have more accumulation potential in the aquatic biota and sediments (Tariq *et al*., 2004b, 2006; Tariq, 2005).

Table 5. Pesticides detected in Pakistani soils/sediments with hazardous classes

Location	Year	Detected	Concentration (µg/L)	CASRN	Detection Technique	LD50	Classes	Reference
Coastal areas,	1990	DDT	2.7-9.2	50-29-3	GC (ECD)	113	II	Bano and Siddique (1991)
		DDE	0.06-0.2	NA		NA	II	
		α-BHC	1.1-3.5	58-89-9		88	II	
		Heptachlor	2.7-9.2	76-44-8		NA	O	
		Dieldrin	0.5-12.5	60-57-1		NA	O	
Cropland soils (village 452, 547, 550, 499, 498 GB, Faisalabad (Punjab)	1990	Aldrin	0.002-0.01	309-00-2	GC (ECD)	NA	O	Ali and Jabbar (1992), Jabbar *et al.* (1993)
		Cyhalothrin	Traces-0.6	68085-85-8		144	II	
		Cypermethrin	Traces	52315-07-8		250	II	
		Dielelrin	0.003-0.01	60-57-1		NA 150	O	
		Dimethoate	0.4	60-51-5		113	II	
		p,pV-DDT	Traces-0.0002	50-29-3		NA	NA	
		o,p′-DDD	Traces-0.002	NA		NA	NA	
		p,p′-DDE	Traces-0.002	NA		NA	II	
		Endrin	0.0006-0.01	72-20-8		87	O	
		Fenvalerate	Traces	51630-58-1		14	II	
		Monocrotophos	0.3-0.6	6923-22-4		NA	Ib	
		Prorenofos	Traces	NA		NA	NA	
Deg Nullah, Lahore (Punjab)	1991	o,p′-DDT	0.2-1.1	50-29-3	GC (ECD)	113	II	Tehseen *et al.* (1994)
		p,p′-DDT	0.006-2.4	NA		NA	NA	
		o,p′-DDD	0.2-1.9	NA		NA	NA	
		p,p′-DDD	0.6-2.3	NA		NA	NA	
		o,p′-DDE	0.002-0.3	NA		NA	NA	
		p,p′-DDE	0.001-0.8	NA		NA	NA	
		HCB	0.0001-0.09	NA		NA	NA	
		BHC	Traces	58-89-9		88	II	
Haleji Lake, Thatta	1999	HCB	0.4-1.7	NA	GC (ECD)	NA	NA	Sanpera *et al.* (2002)
		HCH	Traces	608-73-1		100	II	
		DDT	6.5	50-29-3		113	II	
		PCB	Traces	NA		NA	NA	
Swat (KPK)	2009	Endosulfan	3.5-29.6	115-29-7	GC (ECD)	80	II	Nafees *et al.* (2009)
		Cypermt	4.6-47.6	NA		NA	NA	
Samples collected from nearby areas of obsolete pesticides stores in KPK, Sindh and Punjab	2009	2,4-DDD	84	NA	GC (ECD)	NA	NA	Ahad *et al.* (2009)
		2,4-DDT	299	50-29-3		NA	II	
		4,4-DDE	62	NA		113	NA	
		4,4-DDT	333	NA		NA	NA	
		Dieldrin	3	60-57-1		NA	O	
		Endrin	5	72-20-8		NA	O	
		Heptachlor	43	76-44-8		NA	O	
River Chenab (Punjab)	2011	HCHs	0-9.2	50-29-3	GC (ECD)	100	II	Malik *et al.* (2011)
		DDT	0.2	608-73-1		113	II	
		Heptachlor	0-41.6	76-44-8		NA	O	
River Chenab (Punjab)	2011	HCHs	2.1-18.2	50-29-3	GC (ECD)	100	II	Eqani *et al.* (2011)
		DDT	7.6-59.9	608-73-1		113	II	
Nawabshah (Sindh)	2011	Dichlorvos	0.04	62-73-7	GC (ECD)	56	Ib	Anwar *et al.* (2011)
		Mevinphos	0.4	26718-65-0		4	Ia	
		Dimethoate	0.6	60-51-5		150	II	
		Fenitrothion	0.1	122-14-5		503	II	
		Chlorpyrifos	0.5	5598-13-0		>300	II	
		Endosulfan	0.5	115-29-7		80	II	
Industrial and cotton growing areas (Punjab)	2013	ΣDDTs	40	50-29-3	GC (ECD)	113	II	Syed *et al.* (2013)
		ΣHCHs	7.8	608-73-1		100	II	
		α-Endosulfan	0.3	115-29-7		80	II	
		Chlordanes	3.8	57-74-9		460	II	
Indus River catchment area	2016	p,p'-DDE	0.7	NA	GC (MC)	NA	NA	Bajwa *et al.* (2016)
		o,p'-DDE	0.5	NA		NA	NA	
		o,p'-DDT	0.4	50-29-3		113	II	
		b-HCH	0.3	608-73-1		100	II	
Sediments collected from Kabul River (KPK & Punjab)	2017	ΣHCHs	5.9	50-29-3	GC (ECD)	100	II	Aamir *et al.* (2017)
		ΣDDTs	3.9	608-73-1		113	II	

GC (ECD): Gas Chromatography-Electron Capture Detector, GC (MC): Gas Chromatography-Mass Spectrometry, LD50: Lethal Dose per body weight (150 lb.) to infect 50% population, CASRN: Chemical Abstracts Service Registry Number, NA: Not available

Table 6. Pesticides detected in Pakistani air with hazardous classes

Location	Year	Detected	Concentration (pg/m^3)	CASRN No	Sample Collector	LD_{50} mg/kg	Classes	Reference
Faisalabad, Lahore, Sheikhupura, Cheechawatni, Mianchanu, Shahdra (Punjab)	2013	ΣPCBs	34 - 390	NA	PUF-PAS	NA	NA	Syed *et al.* (2013)
Nullah Aik and Palkhu tributaries of River Chenab (Punjab)	2014	ΣPCBs	41 - 299	NA	PUF-PAS	NA	NA	Mahmood *et al.* (2014a)
Chashma, Taunsa, Sukkar Barrage, Rahimyar Khan (Sindh and Punjab)	2014	ΣDDT	108 - 744	50-29-3	PUF-PAS	113	II	Sultana *et al.* (2014)
		Σchlordanes	2.0 - 43	57-74-9		460	II	
		ΣEndosulfan	0.4 - 10	115-29-7		80	II	
		HCB	2.0 - 22	118-74-1		NA	NA	
		Heptachlor	1.0 - 20	76-44-8		NA	O	
Four sites of Hyderabad city (Sindh)	2014	ΣDDT	1850 - 90700	50-29-3	PUF-PAS	113	II	Alamdar *et al.* (2014)
		ΣHCH	1500 - 97400	608-73-1		100	II	
		ΣChlordane	91 - 188	57-74-9		460	II	
		Heptachlor	54 - 620	76-44-8		NA	O	
Taunsa, Kot Mithan, Guddu and Sukkur (Sindh and Punjab)	2016	HCH	34 - 119	608-73-1	PUF-PAS	100	O	Bajwa *et al.* (2016)
		ΣEndosulfan	87	115-29-7		80	II	
		p'-p'DDE	65 - 515	NA		NA	NA	
		Heptachlor	20	76-44-8		113	O	
		p'-p' DDT	14 - 205	NA		NA	NA	

PUF-PAS: Passive Air Samplers (PAS) using Polyurethane Foam (PUF), LD_{50}: Lethal dose per body weight (150 lb.) to infect 50% population, CASRN: Chemical Abstracts Service Registry Number, NA: Not available

On the basis of findings of above studies, an equation (1) could be formulated consisting of K_{ow} and K_{oc} values of different soil series from the developing countries such as Pakistan to observe the leaching potential of pesticides, in particular where experimental values are not accomplishable due to unavailability of analytical instruments and lack of funds (Tariq *et al*., 2004b).

$$\log K_{oc} = 0.586 + 0.713 \log K_{ow} \qquad (1)$$

After accumulation, persistence of pollutants is another important factor in toxicological point of view. Persistence of pesticides in soil is mainly controlled by the soil conditions, humidity, temperature and microbial activities in the soil (Tariq *et al*., 2006). Microbes play major role for bioremediation of pesticides and their intermediate products from soil (Bollag and Liu, 1990; Vidali, 2001). In cotton agro-system, soil fertility, structure, pH, moisture, microbial density, temperate, redox potential and nature of pollutants control the soil microbial activity which plays an important role in pesticides degradation and providing important inorganic materials and organic metabolites to the plants (Asha and Sandeep, 2013). Furthermore, the indigenous or externally added microbes may further mineralize pesticides through their diverse enzymatic activities (Rodrigo *et al*., 2014). Organic matter in the soil not only support the microbial community with nutrients but also provide the sorbed and porous sites as a niche to multiply their community and improve degradation rate (Xiong *et al*., 2017).

***Air contamination*:** According to the U.S. Environmental Protection Agency (USEPA, 2018), indoor and outdoor air quality directly affects the living organisms which can be measured by presence of pollutants, their concentrations and persistence in air. These pollutants may be settled down to Oceans and grounds, degraded by the presence of air humidity and radiations or dispersed into air. Pesticides pollution in the air vary due to their volatilization behavior and persistence. Most of the pesticides release into the air from tropical and subtropical regions (Wania and Mackay, 1995), and distribute on global scale due to their long-range transportation in atmosphere (Alegria *et al*., 2008; Park *et al*., 2011). Approximately 2.0% of the sprayed pesticide is volatilized to the atmosphere during application. This application loss which can be defined as the concentration of applied doses in non-target area is due to the spray drift in the field. The nozzles pore size can control droplets distribution within the target area to limit evaporation losses (Van-den-Berg et al., 1999). Evaporation of pesticides is less affected by its physio-chemical properties and more dependent on the formulation and application method, carrier (diluent) and the other environmental conditions including wind speed, temperature, relative humidity and radiation (FOCUS, 2008).

Pesticides in the atmosphere have major two types of fates i.e. degradation and deposition. The volatilized pesticides go through the photochemical reactions and free radical reactions. The sun produces radiations having wavelength of 290-400nm which supports pesticides to go for photochemical reactions. These radiations contain enough energy to breakdown the chemical bond of pesticides or excite the other molecules to react with these pesticides. In the free radical reactions, sun light produces unpaired and unstable electron through excitation of H_2, O_3, O_2, or N_2O. Unstable products react quickly with molecules in the air to form stable and less toxic compounds (Linde, 1994). The changing environmental conditions support the deposition of pesticides all over global environment far away from their target areas. Through atmospheric transportation, tons of compounds have been observed in the ecosystems of cold polar region (McConnell *et al*., 1998).

Several studies have been conducted to find the level of pesticides in the marine ecosystem which revealed that the marine water is the major recipients of these contamination (Goldberg *et al*., 1975; Strand and Hov, 1996). The high level of pesticides in oceans is due to wet and dry depositions, gaseous exchange between air and water and riverine inputs. The marine water becomes the source and sink of pesticides due to air exchanges. The transported pesticides sink in the deep oceans by absorbing on particles (Dachs *et al*., 2002; Lohmann *et al*., 2006) or oceans becomes the secondary source of contamination in the atmosphere because of gaseous exchange of seas to atmosphere (Zhong *et al*., 2014).

A recent study reported the presence of organo-thiophosphate insecticides especially chlorpyrifos in air of Arctic region, which is due to long-range transportation of these chemicals from point and non-point sources (Anjum *et al*., 2017). Bajwa *et al*. (2016) observed the presence of organochlorine pesticides (OCPs) in air. The authors selected the four important ecological regions of River Indus such as Taunsa, Kot Mithan, Guddu and Sukkur for sampling (Table 6). It was found that the ∑DDTs concentration (147.4 pgm^{-3}) was higher in the air than the concentration found from the most developed countries like UK and Spain ranged from 6 to 83 pgm^{-3} however, this concentration had consistent nature with values observed by Sultana *et al*. (2014). The DDTs values were surveyed by ∑HCHs which ranged from 176 to 325 pgm^{-3}. The overall trend they observed was in following order ∑DDTs > ∑HCHs > ∑endosulfan > ∑HCB > ∑Chlordane > ∑Heptachlor. Sultana *et al*. (2014) conducted the same experiment at six selected locations near center of Indus Basin but observed different trend of OCPs as ΣDDTs> ΣHCHs> ΣChlordanes> ΣEndosulfans> HCB> Heptachlor. The total DDTs that they found from the ambient air samples were attributed 80.0% out of total OCPs and ranged from 108 to 744 pgm^{-3}. The total OCPs were ranged from 123 to 856 pgm^{-3}. The temperature differences between upstream and lower stream also affected the trend of OCPs from Head Panjnad > Sukkar > Dera Ismail Khan > Taunsa > Chashma > Rahimyar Khan (Table 5). In other studies, conducted by Alamdar *et al*. (2014) and Mahmood *et al*. (2014a) the concentration of DDTs and HCHs out of total OCPs were detected from four sites of Hyderabad city and Nullah Aik and Palkhu streams of Sialkot and Gujranwala districts, respectively. It was observed that DDTs and HCHs concentrations were about 70.0% and 80.0% in samples in Hyderabad cities and 66.0% and 16.0% in samples in Nullah Aik and Palkhu streams. All previous observations show the extensive use of illegal pesticides in both up and down stream areas. These illegal pesticides contain persistent toxic substances which have the potential to cause cancer in near residential areas and harmful for natural biota and habitats.

Pesticides enter into body after long- and short-term exposure from air during or after application. Delayed effect of pesticides occurs through inhalation of residual concentration or through the dose present on the surface of body or clothes, pesticides containers and spraying equipment. Increased exposure of pesticides through poor protective equipment and their usages, more frequency and diameter of spill through nozzles, and deposition on body and cloth cause the severe respiratory problems (Damalas and Eleftherohorinos, 2011; Cattani *et al*., 2001).

***Food contamination*:** Advancement in technology, for determining trace amounts of pesticides in the food, increases anxiety among the people about how much concentration of a pesticides they are consuming (Tucker, 2008). Strict government regulations, with reliable and precise analysis, have been implemented in the developed world about the toxic pesticides residues in food (Fong *et al*., 1999). A group of researchers from western Canada introduced a new production system named as pesticide free production (PFP) to produce crops on sustainable basis. In this system, the crops are produced following the organic and integrated pest management (IPM) approaches, without application of chemicals for pest control (Magnusson and Cranfield, 2005). From sowing to harvesting, crop plants or soil may not be treated with any type of pesticides, with the exception of recommended fertilizers, because the active ingredients of pesticides may accumulate as residues in the soil and enter in the food chain and cause severe health issues (Magnusson and Cranfield, 2005).

According to FAO, approximately 80.0% of food produced in the developing countries comes from the previous cultivated lands which increases the chances of the pesticides exposure in the agriculture land particularly for higher crop production (WHO, 2017). Residual contents of pesticides in plant body mainly comes from soil and water routes. Plants take up nutrients from the soil and transport them only through water within the plant body. Several researchers have found the soil residual pesticides in the plant body. The residues that enter in the plant body expectedly present in three forms as (i) freely extractable form, (ii) bounded extractable form, and (iii) unextractable bounded form while the later ultimately becomes the part of plant's constituents and transfers to the end consumers of the plant constituents (Bhat and Gómez-López, 2014).

The presence of pesticide residues in milk, feed, cottonseed, different vegetables, fruits, and fresh meal has been reported throughout the Pakistan (Cheema and Shah, 1987; Masud and Farhat, 1985; Masud and Hasan, 1992; Parveen and Masud, 1988a, b; Munshi *et al*., 2004; Saqib *et al*., 2005; Hassan *et al*., 2014; Yaqub *et al*., 2017). Table 7 shows the presence of OCPs in majority of the samples with higher concentrations than the residual safe limits for the end consumers.

Table 7. Pesticide poisoning in Pakistani people (modified from Tariq *et al.* 2007)

Affectees	Affected (%)	Exposure	Pesticides	Effects	Reference
Hospital patients, Karachi (Sindh)	100	Accidental, suicidal	Organophosphates	15 deaths out of total 53 registered cases	Jamil *et al.* (1977)
Farm-workers	60 -70	Occupational	Methyl-parathion	Limbs, feeling dizziness, chest pressure, change in defecation and eye tearing	Shihab (1976), Baker et al. (1978)
Spray-men and persons associated with job during malaria control program	40	Occupational	Parathion	5 died and 2800 affected. Severe depressions of red blood and cell cholinesterase activity appeared	Shihab (1976), Baker et al. (1978)
Villagers (adults and children) Talagang, Chakwal (Punjab)	80	Accidental	Endrin	Among 194 affected persons, 70 % was children and 10 % affected persons was died	Jabbar (1992)
Cotton picker		Non-occupational	General	Nausea, abdominal pain, headache, cough, throat irritation, eye irritation, diarrhea, unusual tiredness, wheezing and dizziness	Baloch (1985), Khwaja (2001)
25 hospital patients (9 females and 16 males) (Baluchistan)	100	Non-occupational	DDT DDE HCH	Blood and fat concentration of pesticides in the patients were significantly higher amongst 21 blood samples and 20 fat tissues of the patients	Krawinkel *et al.* (1989)
Male and female cotton pickers, Multan (Punjab)	100	Non-occupational	General	Amongst 88 females, 1 had a sufficiently low AchE level, 74% had moderate pesticide poisoning while 1/4 of the sample had dangerous level AchE levels (50-87.5%). Whereas, the comparative figures for 33 males were 12, 51, and 36% respectively	Masud and Baig (1992)
Hospital patients, Karachi (Sindh)	100	Non-occupational	General	Low AChE level	Naqvi and Jahan (1999), Azmi et al. (2005)
Fruit and vegetable farm-station workers, Gadap, Karachi (Sindh)	100	Occupational	Cypermethrin, Deltamethrin Polytrin-C Diazinon Monocrotophos DDT DDE	Significant increase in enzyme level of GOT, GPT and ALP. Suffering hepatitis, dyspnea and burning sensation in urine	Azmi *et al*. (2006)
Population of Khawara valley in Nowshehra (KPK)	Not specified		Spilled of obsolete pesticides in 1995	Intestinal, eye and lung diseases; especially amongst the children	Environmental News Service (1998)
Female cotton pickers, Multan (Punjab)	18 (22% for married whereas 9% for unmarried)	Occupational	General	Amongst 38 females, 42.86% had AChE level at alarming situation while 52.63% participants were in dangerous condition. Whereas the effect on reproductive hormones (LH and FSH) was recorded major in married women as compared to unmarried	Ahmad *et al.* (2004)
Female cotton pickers, Khairpur (Sindh)	18 (22% for married whereas 9% for unmarried)	Occupational/ non-occupational	Methamatophos Baythroids Cypermethrin Endone, Aldrin	Amongst 34 female workers, a significant increase was recorded in reproductive hormones (LH and FSH, PG and estradiol) both in cotton pickers and non-pickers	Rizwan *et al.* (2005)
Male industrial workers of pesticide manufacturing industry, Multan (Punjab).	NA	Occupational	NA	A significant genotoxic basis on significant (Pb0.001) decrease in the level of SChE in 35 industrial workers	Bhalli *et al.* (2006)
Factory areas in Lahore, Multan (Punjab), Karachi (Sindh)	29% small and 8% medium industrial workers	Occupational	Endosulfan Imidacloprid Thiodicarb Carbofuran Methamidophos	Serum AST, ALT, Creatinine GGT, Malondialdehyde, Total antioxidant, and CRP significantly higher	Khan *et al.* (2010)
Village (Chak No. 105) (Punjab)	NA	Non-occupational	Chlorfenapyr	70 lives infected to food poisoning and 11 members of same family died	Mohiuddin et al. (2016)
Hospital patients, Karachi (Sindh)	8% mortality rate	Non-occupational	Organophosphate	Acute respiratory distress syndrome to aspiration pneumonia.	Mohiuddin *et al.* (2016)
Male, female and children from three different farm locations, Vehari (Punjab)	NA	Occupational	Endosulfan Aldrin Dieldrin pp-DDT	Farm workers contained 1.13 ngmL^{-1} pp-DDT, 0.92 ngmL^{-1} aldrin, 0.68 ngmL^{-1} dieldrin and 1.96 ngmL^{-1} endosulfan in their blood samples.	Saeed *et al.* (2017)

AChE: Acetyl cholinesterase level, KPK: Khyber Pakhtunkhwa (former North-West Frontier Province (NWFP) of Pakistan), SChE: Serum cholinesterase, GPT: Glutamate pyruvate transaminase, GOT: Glutamate oxaloacetate transaminase, ALP: Alkaline phosphatase, LH: Luteinizing hormone, FSH: Folliclestimulating hormone, PG: Progesterone, AST: Aspartate aminotransferase, ALT: Alanine aminotransferase, GGT: Gamma glutamyltransferase, CRP: C-reactive protein, NA: Not available

Although vegetables and fruits have various residual level of pesticides which may be due to different climatic conditions of the zone (hot, humid, and cold) or due to the variation in plant species, MRL (maximum residue limit) can be defined as tolerant level to store residues in the food commodities, as already set by the USEPA. The MRL of the food impacts the acceptable daily intake (ADI) of residues per kilogram body weight (kBW) in a day. United States and European Union have set the ADI as 1.75 and 0.3 mg/kg/BW/day, respectively to reduce its impact on the human health (Gillam, 2017).

Various fruits samples were collected to see year-wise distribution of contamination with the MRL. Substantial reduction has been noted in fruits varieties particularly in citrus and apple which was supposed to be due to awareness in the growers about regularity in use of pesticides. The higher values of bifenthrin, cypermethrin, cyhalothrin and endosulfan were also observed in citrus fruits; however, their presence was not justified scientifically (Parveen *et al*., 2004). Irrigation is supposed as a common practice and a faster way to contaminate food through soil and water particularly in Pakistan. More than 50.0% of the collected fruits and vegetables were found to have higher level of contamination than the permissible level and MRL due to the presence of higher residues of dieldrin, DDT, methamidophos, and diazinon. The fruits samples collected from Nawab Shah, Karachi and Hyderabad regions were found to be 8.0%, 36.0% and 12.5% contaminated, respectively. The meat samples that were collected from Faisalabad and Karachi regions were 100.0% contaminated and all these samples were found to contain residual level 100 times higher than MRL (Faheem *et al*., 2015).

The contamination of recent stockpiled pesticides is very high with their persistent nature in different environmental matrices and crucially needs a comprehensive study about the surveillance, risk assessments, residual limits, and monitoring of these pesticides. However, the polices of Pakistan related to the food items are around 40 years old and lacking regularity in measurement, this reflects the high level of pesticide residues in 105 food items (Syed *et al*., 2014).

***Pesticides exposure to human*:** Some of the pesticides such as triazines, tetrachloro-p-dioxin (TCDD), 1,1,1-trichloro-2,2-bis-(p-chlorophenyl)-ethane (DDT), butylated hydroxyanisole (BHA), Di-(2-ethylhexyl)- phthalate (DEHP), poly- chlorinated biphenyls (PCBs), tributyltin (TBT) were found to be still in use even after these were banned by the regulating authorities in Pakistan (Ejaz *et al*., 2004; Khwaja *et al*., 2013). Additionally, misuse of pesticides in Pakistan is very high due to the less literacy rate and awareness to their safe uses (Tariq *et al*., 2004a). According to the report of Sustainable Development Policy Institute (SDPI), the pesticide exposure is more dangerous for the women in the cotton field (Jabbar and Mallick, 1994). The main reason behind this fact is that traditionally more than 90.0% of cotton picking in the field is done by women workers in Pakistan (Khwaja, 2001; Ahmad *et al*., 2004; Rizwan *et al*., 2005). Furthermore, health issues and different symptoms observed in cotton-picking women include (but not limited to) nausea (12.0%), skin irritation (27.0%), gastroenteritis (10.0%) and headache (26.0%) (Jabbar and Mallick, 1994).

In October, 2006, dengue virus caused 44 deaths and 732 severe fevers out of 4659 cases in Pakistan (Xinhua, 2006). The Government announced to spray deltamethrin to control this vector born disease in populated areas without preventive measures. This dengue control activity significantly increased the cases of respiratory disorders (i.e. more than 20.0% than previous hospital records) (Special Report, 2006; Orme and Kegley, 2006). Moderate persistence of deltamethrin in atmosphere was observed especially in Lahore and Faisalabad due to the hazy environmental conditions of these regions at the time (having 400–600 $\mu g/m^3$ suspended particulate matter at 13-20˚C) (Shahid, 2007).

Krawinkel *et al*. (1989) collected 20 and 21 samples of fat tissues and blood respectively from the hospital of Quetta city to observe the pesticides level especially organochlorine (i.e. HCH, DDT and DDE). It was reported that the concentrations of DDT had association with the level of DDE and both samples of fat tissues and blood had high concentration of DDE/DDT. Seed *et al*. (2017) collected 56 blood samples from different cultivated areas of Vehari and found significant concentration of pp-DDT, aldrin, dieldrin and endosulfan from blood samples. Parveen and Masud (2001) collected 75 samples of blood from cotton cultivated areas of Multan and found 85% of the blood samples containing organophosphate (OP), synthetic pyrethroids (SP) and organochlorines (OC). Pesticides such as DDT and its metabolites, profenofos, fenpropathrin, monocrotophos and cyhalothrin were found more frequently in different blood samples. Ilyas *et al*. (2003) collected 116 blood samples of spray men, pesticide dealers and common men and found the same trend. Higher concentration of pesticides also causes the neurological disturbance to impair the nerve functions and affect equally at the central nerves and peripheral nerves system (Hong *et al*., 2007).

Higher concentration of acetyl-cholinesterase enzyme (AChE) 50.0 to 88.0% per quarter of sample) was found in 88 blood samples of cotton picker females which was probably due to the higher use of organophosphate and/or OCPs (Masud and Baig, 1992). The same AChE was also observed in the blood samples of urban male and female groups from Karachi, Pakistan. In this study, correlation between organophosphate pesticides and AChE was not significant (Naqvi and Jahan, 1999; Azmi *et al*., 2005).

Results of several previous studies showed residues of endocrine disrupting organochlorine pesticide in the blood samples of the residents of Punjab, Baluchistan and Sindh provinces of Pakistan (Table 7), where higher rates of cancer were observed during 1994-2002 (Krawinkel *et al*., 1989; Naqvi and Jahan, 1999; Parveen and Masud, 2001). Ahmad *et al*., (2002b) reported that pesticide poisoning center, Nishtar Hospital, Multan, Pakistan registered 578 patients and 370 patients were found the victim of poisoning (i.e. OP, OC) exposure (27.0% females and 73.0% males). The toxic effects of these pesticides on the reproductive hormones [i.e. follicle stimulation hormone (FSH), luteinizing hormone (LH), progesterone (PG)] and estradiol with AChE's level in the blood plasma were evaluated in cotton picker women. Considerable differences were observed in FSH and LH of married workers than the unmarried; however, overall effect on these hormones was negligible. Results of this study were found to be consistent with that of Larsen *et al*. (1998) who observed that short exposure of these pesticides did not likely affect the reproductive hormones and serum quality. Rizwan *et al*. (2005) also conducted the similar research in Khairpur District, Sindh, Pakistan and correlated endocrine disruption with pesticides (i.e. cypermethrin, methamatophos, endone and baythroids).

Exposure and effects of polytrin-C, monocrotophos, cypermethrin, diazinon, deltamethrin, DDE and DDT were reported in workers blood from 14 different fruit and vegetable farms in Gadap, Karachi, Pakistan. The residual effect was also observed on the enzyme secretions i.e. glutamate pyruvate transaminase (GPT), alkaline phosphatase (ALP) and glutamate oxaloacetate transaminase (GOT). High level of these enzymes was detected in 78 blood samples out of 247 samples which may be due to mutation in genetic expression to detoxify the pesticides (Azmi *et al*., 2006). To assess the effect of pesticides on genetic materials, Bhalli *et al*., (2006) monitored the micronucleus assay, serum cholinesterase (SChE), liver enzymes and some hematological parameters as biomarkers in group of 35 male labor in the pesticide industry in Multan, Pakistan and found significant reduction in the level of SChE in the labor group suggesting genotoxic effect of pesticides.

Conclusions and future recommendations: Extensive use of pesticides for agricultural production is a major environmental and health issue in the farming community of Pakistan. Most of the farmers use a mixture of chemical pesticides for the better control of different insect pests and diseases. Major portion of the used pesticides is highly or moderately toxic (70.0%) to the biological system. Careless use and handling of these toxic pesticides result in contamination of all three kinds of environmental matrices including air, water and land. Through all these matrices, directly or indirectly, these pesticides enter into food chain and human body and cause a number of health issues in human beings. The sole reliance of farmers on chemical pesticides, poor knowledge and limited access of farmers to training facilities, and improper handlings are the major factors leading to the presence of higher pesticides residues in agricultural commodities and thus higher risk of their exposure to the end users (i.e. human). Strict government regulations on pesticides production, distribution, and easy access of farming community to training facilities and effective extension services are required for improving farmers' knowledge on use and proper handlings of pesticides. This will help to reduce the risk of exposure and subsequently mitigate the adverse effects of pesticides on environmental health. Alternate pests controlling substances such as biopesticides (i.e. natural or microbes-based products) or allelopathic plant extracts (i.e. allelochemicals) must be explored and developed for possible use in crop production.

Although some data on use, contamination and exposure of pesticides in Pakistan are available, research studies are too limited to know the real picture of the subject matter. On the basis of previous studies, we suggest that considerable efforts must be made to examine the current status of pesticides contamination and related health issues in Pakistan. Studies must be conducted to precisely determine the current use, contamination level and exposure of pesticides in Pakistan. In addition, substantial research work must be done to develop new, eco-friendly and cost-effective methodologies and explore suitable ways to remove these pollutants from the contaminated environments.

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