Freely dissolved Organochlorine Pesticides and Polychlorinated Biphenyls along the Indus River Pakistan: Spatial pattern and risk assessment

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Figure S7: Percentage contribution of various sources of POPs at four studied zones in surface water along Indus River

Text S1. More details on Study Area and Field Strategies:

The present study categorized the Indus River into four major zones - the first is Frozen Mountainous Zone (FMZ), the second is Wet Mountainous Zone (WMZ), the third is Alluvial Riverine Zone (ARZ) and fourth is Low Lying Zone (LLZ) **(Fig. S1)**, based on the altitudes and geographical characteristics. The FMZ includes Hunza, Skardu, Gilgit and Swat and is characterized by having world's greatest mountain ranges (Hindukush, Karakorum and Himalaya) which are covered with the snow throughout the year. These valleys' elevation ranges from 750 to 3000 m from sea level (CIESIN, 2014) with minimal anthropogenic activities in the area. It has been reported that extensive contribution of LRAT of dust in these areas originates from India and China (Carmichael et al., 2009). In these areas, sources are related to LRAT of regional POPs/metals (China, India), as atmospheric dust particles deposition, which are derived from the anthropogenic activities contaminated with POPs/metals, resulted into the retaining and buildup of POPs into the forest soils. Moreover, these areas also possess the fruit orchards and agricultural fields, where pesticides application is common for the insect control. The WMZ comprises of Khyber Pakhtunkhwa (KPK) districts, including Swabi, and Nowshera. These regions are characterized by more rainfall with stretches of the plains near to hills, where population settlement are situated. These region are also covered with the temperate forests, with an elevation range 750 to 1500m from the sea level (CIESIN, 2014). Currently, various anthropogenic activities are being observed in these regions including mining, agriculture and mining activities etc., and contributing to short range atmospheric dust transport. The alluvial riverine zone (ARZ) includes the Punjab Province districts such as Mianwali, Layyah, Bhakkar and Dera Ghazi Khan which are the parts of upper Indus plains. These areas lie on the Indus River bank, which is the major water source for the domestic and agriculture purposes. Indus River brings the minerals and sediments from the mineral rich mountains that are then deposited into the plains and becomes part of soil composition (Eqani et al., 2012a). Agriculture is the main livelihood of the region, with massive usage of pesticides and other chemicals for several agricultural and urban purposes. In addition, industrial activities along with the huge population and the poor drainage might contribute to elevated burden of POPs along Indus River. The fourth zone [i.e. Low-Lying Zone (LLZ)] comprises the low-Lying region of Sindh such as Khairpur, Sukkar and Hyderabad. These regions located on left/right bank of the southern irrigated and lower Indus Plains downward towards the Arabian Sea. These regions are characterized by alluvial materials because it is composed of silt loam and permeable sand close to river with the increasing clay when going away from river. Agricultural and industrial activities are main factors, which may be responsible for the contamination of POPs in this zone (Alamdar et al., 2014). In addition, the climatic condition (semi-arid characteristic with higher temperature, sandstorms and the low vegetation cover) make the associated human population and other wild life of LLZ and ARZ more vulnerable to the POPs/metals exposure. The detailed studied site location description is given as **Table. S1.**

Text S2. Calculating sampling rate (Rs) of water

Final dissolved fraction of OCPs and PCBs were calculated by using the equilibrium/disequilibrium of performance reference compounds (PRCs) added to the LPDE-samplers. Assuming that uptake and elimination rates are equivalent, the freely dissolved (or gas-phase) concentrations of individual OCPs and PCBs (C_{water}) were then calculated by the following equation:

$$C_{water} = C_{LDPE}/K_{LDPE\text{-}water} \times [1\text{-}\exp{-Rs} \times t/K_{LDPE\text{-}water} \times m_{LDPE}] \quad (3)$$

Whereas C_{LDPE} is the OCP or PCBs concentration in the LDPE (L/kg), Rs is the sampling rate [L /day], t is the deployment period (days), m_{LDPE} is the mass of the LDPE sheet (kg) and $K_{LDPE\text{-}water}$ is the sampler-water partitioning coefficients. Sampling rates were calculated using the PRCs as follows:

$$f = \exp(-Rs \times t/K_{LDPE\text{-}water} \times m_{LDPE}) \quad (4)$$

Where f is the fraction of PRC retained in the passive sampler after deployment. More details on the PRC method/sampling rate calculation of LPDE- have been described elsewhere (Khairy and Lohmann, 2013). The OCPs and PCBs concentrations in the air were estimated by assuming sampling rate and the partitioning for a sample unit throughout the sampling period. A standard of 3.5 m3 air/day per sampler of the sampling rate was calculated in the previous calibration studies (Shoeib and Harner, 2002).

Text S3. Human Health Risk Assessment of POPs contaminated water

The health risk assessment was performed to evaluate the toxic impacts of POPs present in Indus River water on the human health in study sites (USEPA, 1998). The health risk assessment for human was done to find the POPs poisoning from the drinking water and dermal exposure. POPs chronic daily intake (CDI) through oral and dermal exposure from contaminated water was evaluated in the present study.

Oral Exposure

For calculating Oral Exposure, $CDI_{(oral)}$ of POPs due to ingestion of POPs-contaminated water was presented by the Eq. 1: (USEPA, 1998).

$$CDI_{(oral)} = \frac{C \times IR \times ED \times EF}{BW \times AT} \quad \ldots\ldots (1)$$

Where C is OCPs and PCBs concentration in water (mg/L), IR is ingestion rate of water (L/day), ED is the exposure duration (70 years), EF is the exposure frequency (365 days/ year), BW is the body weight (72 kg) (Sultana et al., 2014) and AT is the person average life time in days (24,455 days).

Dermal Exposure

For dermal exposure the $CDI_{(dermal)}$ of OCPs and PCBs through Indus River was measured by Eq. 2: (Li and Zhang, 2010).

$$CDI_{(dermal)} = \frac{L \times SA \times Kp \times ET \times EF \times ED \times Fcl \times Fc2}{BW \times day} \quad \ldots\ldots (2)$$

Where CDI (dermal) is chronic daily exposure of OCPs and PCBs via dermal water contact (mg/kg/day), L is OCPS and PCBs levels in Indus Riverr (µg/L), SA is exposed skin area(cm2) to water, Kp is dermal permeability coeffeicient (cm/h), EF is the exposure frequency (days/year), ET is the exposure time (min/day), ED is the exposure duration (year), BW is the body weight (kg), Fc1 is conversion factor from μg to mg (0.001), and Fc2 is the conversion factor of unit (L 1000 cm-3 (0.001). According to USEPA database the value of Kp is 1.10^{-3} cm/h (USEPA, 2004).

Hazard Quotient (HQ)

Hazard quotient (oral) were presented by the Eq. 3 (USEPA, 1998).

$$HQ_{(oral)} = \frac{CDI_{(oral)}}{RfD_{(oral)}} \quad (3)$$

Where RfD (oral) is the oral reference dose.

Carcinogenic Risk (CR)

Carcinogenic risks (CR) for the oral and dermal OCPs and PCBs exposure were determined by using the following Eq. (4, 5).

$$CR_{(oral)} = CDI_{(oral)} \times SF \quad (4)$$

Where $CSF_{(oral)}$ is called the cancer slope factor of oral exposure

$$CR_{(dermal)} = CDI_{(dermal)} \times SF \quad (5)$$

Where $CSF_{(dermal)}$ denote cancer slope factor of dermal exposure to OCPs and PCBs

Table S1

Sites	Zones	Latitudes	Longitudes	Site characteristics
S-1. Hunza	FMZ	36°18'35.25"N	74°38'22.70"E	Low or cold/freezing temperature Pristine sites, less anthropogenic activities were observed in regards of pesticides for crop protection, less human settlement were found, routine consumer product (electrical materials, electronics, plastic, furniture) were also observed.
S-2. Gilgit	FMZ	35°54'28.74"N	74°22'46.85"E	Low temperature, less anthropogenic activities were observed in regards of pesticides for crop protection, orchards protection and malarial control, routine consumer product (electrical materials, electronics, plastic, furniture, foam) were observed.
S-3. Skardu	FMZ	35°18'47.13"N	75°38'24.34"E	Pristine environment, less anthropogenic activities were observed in regards of pesticides for crop, orchards and malarial control protection, human settlement are less, electrical materials, electronics, textiles, furniture, plastic, and foam were observed.
S-4. Swat	FMZ	34°42'24.54"N	72°48'10.64"E	Anthropogenic activities were observed in regards of pesticides for crop protection and malarial control, human settlement are less, routine consumer product such as electrical materials, electronics, textiles, plastic, furniture and foam were observed.
S-7. Swabi	WMZ	34° 0'39.14"N	72°36'47.35"E	Moderate temperature having anthropogenic activities were observed in regards of pesticides for crop protection and malarial control, more human settlement were found, routine consumer product (electrical materials, electronics, textiles, plastic, foam, furniture) were also observed

S-8. Nowshera	WMZ	34° 1'2.99"N	72° 1'14.64"E	Pesticides burial ground in Amangarh Nowshera present here where still dumping is present; Anthropogenic activities were observed in regards of pesticides for crop protection and malarial control, more human settlement was found, routine consumer product (electrical materials, electronics, textiles, plastic, foam, furniture) were also observed
S-9. Mianwali	ARZ	32°26'58.47"N	71°22'15.10"E	Agriculture zone containing cereal crops, sugarcane, cotton, fodder, and vegetable were grown, huge human activities, pesticides application. Electronics, and electric materials, textile, foam, furniture were also common.
S-10. Bhakkar	ARZ	31°36'57.07"N	70°52'44.76"E	Agriculture zone containing cereal crops, sugarcane, cotton, fodder, and vegetable were grown, huge human activities, pesticides application. Electronics, and electric materials, textile, foam, furniture were also common.
S-11. Layyah	ARZ	30°57'47.72"N	70°50'27.01"E	Agriculture zone containing cereal crops, sugarcane, cotton, fodder, and vegetable were grown, huge human activities, pesticides application. Electronics, and electric materials, textile, foam, furniture were also common.
S-12. D.G Khan	ARZ	30° 2'41.07"N	70°48'27.57"E	Agriculture zone containing cereal crops, sugarcane, cotton, fodder, and vegetable were grown, rapid population growth and unplanned urbanization, pesticides application. Electronics, and electric materials, textile, foam, furniture were also common.
S-13. Sukkur	LLZ	27°41'10.24"N	68°51'35.48"E	Pesticides usage for crop protection and malarial control were observed, more human settlement were found, routine consumer

				product (electrical materials, electronics, textiles, plastic, foam, furniture) were also observed
S-14. Khairpur	LLZ	27°31'32.03"N	68°45'18.26"E	Anthropogenic activities were observed in regards of pesticides for crop, and also for date form protection and malarial control, more human settlement was found, routine consumer product (electrical materials, electronics, textiles, plastic, foam, paint, furniture) were also observed.
S-15. Hyderabad	LLZ	25°22'25.39"N	68°18'26.74"E	Agriculture and urban zone. Large human settlement and heavy anthropogenic activities were observed in regards of pesticides for crop protection and malarial control. Routine consumer product (electrical materials, electronics, textiles, plastic, foam, furniture) were also observed.

Table S2

Code	Sites description	Zones	Water (L/day)
S -1	Hunza	FMZ	27.20
S -2	Gilgit	FMZ	9.00
S -3	Skardu	FMZ	17.16
S -4	Swat	FMZ	23.13
S -7	Swabi	WMZ	10.32
S -8	Nowshera	WMZ	53.33
S -9	Mianwali	ARZ	10.90
S -10	Bhakkar	ARZ	15.94
S -11	Layyah	ARZ	19.60
S -12	D.G Khan	ARZ	46.15
S -13	Sukkar	LLZ	55.30
S -15	Hyderabad	LLZ	91.00

Table S3.

Sampling Location	HCBs (pg/L)	DDTs (pg/L)	HCHs (pg/L)	PCBs (pg/L)	References
Indus River Pakistan	0.23–6.41	0.48–224	0–2.64	3–226	Current Study
Water, River Chenab		0.55–580 (ng/L)	3.0–330 (ng/L)	7.7–110 (ng/L)	(Eqani et al., 2012b)
Baiertang waters, China		30–2700	400–5900		(Luo et al., 2004)
River Xanaes argentina		70-340			(Schreiber et al. 2013)
Yangtze River, China		N.D–16700	550–2800	290–2000	(Tang et al., 2008)
S-SE Brazil		0.2–0.3			(Meire et al. 2016)
Kolleru Lake, India		1980	5440		(Sreenivasa Rao and Ramamohana Rao, 2000)
Rocky Mountains, Canada	4.0–7.2				(Blais et al., 2001)
Grossenkölle, Austria	2.2–5.8	7.7–20			(Vilanova et al., 2001)
Pyrenees, Spain	7.3–9.5	13–18			(Vilanova et al., 2001)
Øvre Neådalsvatn, Norway	5.2–7.2	0.2–1.0			(Vilanova et al., 2001)
Pyrenees, Spain	0.5–35	16–29			(Catalan et al., 2004)
Lake Erie USA				1.50−83.8 (29)	(Liu et al., 2015)
Lake Ontario USA				6.00−105 (29)	(Liu et al., 2015)
Great lake USA				2.0−55 (29)	(Khairy et al., 2015)
Daya Bay China				91.1–1355.3	(Zhou et al., 2001)
Tonghui River China				31.58–344.9 (ng/L)	(Zhang et al., 2004)
Minjiang Estuary China				203.9–2473	(Zhang et al., 2003)

Table S4

Compound	FMZ	WMZ	ARZ	LLZ	CMC	CCC	MAC	AA
o,p′-DDE	1.3	0.3	4.7	1.1				
p,p′-DDE	29.3	3.3	50.1	17.7	1100000	1000		
o,p′-DDD	1.7	0.4	3.5	0.6				
p,p′-DDD/ o,p′-DDT	4.2	0.7	6.5	1.6	1100000	1000		
p,p′-DDT	11.5	1.8	12.3	5.3	1100000	1000		10000
ΣDDTs	48.	6.5	77	26				2500
γ-HCH/lindane			0.6		950000		40000	
Chlordane	0.77	0.51	1.6	0.61	2400000	4300		
α-Endosulfan	0.91	6.4	2.31	1.27	220000	56000	10000	
PCBs	13.8	12.2	23.4	5.9		14000		

Table S5

Dioxin Like PCBs pgTEQs/L	TEF	Studied Zones: FMZ			WMZ			ARZ			LLZ		
		Min	**Max**	**Mean**	**Min**	**Max**	**Mean**	**Min**	**Max**	**Mean**	**Min**	**Max**	**Mean**
PCB-118	0.00003	0.0000045	0.000013	0.000006	0	0	0	0.000007	0.000021	0.000013	0.000001	0.000024	0.000012
PCB-114	0.00003	0	0	0	0	0	0	0	0	0	0	0	0
PCB-105	0.00003	0.000003	0.00003	0.0000025	0	0.000005	0.0000025	0	0.000004	0.0000025	0	0.0000009	0.0000004
PCB-156	0.00003	0	0	0	0	0	0	0	0.000009	0.0000022	0	0	0
∑Mono-ortho DL-PCBs		0.0000075	0.000043	0.0000085	0	0.000005	0.0000025	0.000007	0.000034	0.0000177	0.000001	0.0000249	0.0000124
PCB-77	0.0001	0	0	0	0	0	0	0	0	0	0	0	0
PCB-126	0.1	0	0	0	0	0	0	0	0	0	0	0	0
PCB-169	0.03	0	0	0	0	0	0	0	0.0003	0.000075	0	0	0
Non-ortho DL-PCBs		0	0	0	0	0	0	0	0.0003	0.000075	0	0	0

Table S6

OCPs	FMZ	WMZ	ARZ	LLZ
p,p′-DDD	2.83E-11	4.33E-12	4.34E-11	1.05E-11
p,p′-DDE	2.40E-09	3.16E-11	4.74E-10	1.67E-10
p,p′-DDT	1.09E-10	1.74E-11	1.16E-10	5.02E-11
γ-HCH/lindane			2.38E-11	
ΣChlordane	6.51E-12	4.96E-12	1.58E-11	5.93E-12

Table S7

OCPs	FMZ	WMZ	ARZ	LLZ
p,p′-DDD	6.89E-14	1.06E-14	1.06E-13	2.57E-14
p,p′-DDE	6.74E-13	7.71E-14	1.15E-12	4.07E-13
p,p′-DDT	2.65E-13	4.23E-14	2.83E-13	1.22E-13
γ-HCH/lindane			5.81E-14	
ΣChlordane	1.82E-14	1.21E-14	3.84E-14	2.27E-14

Table S8

Compounds	**PC-1**	**PC-2**	**PC-3**	**PC-4**
OPDDE	**.838**	-.191	.427	.256
PPDDE	**.804**	-.311	.422	.263
OPDDD	**.904**	.037	.426	-.009
PPDDDOPDDT	**.887**	.085	.423	-.149
PPDDT	**.599**	.459	.254	**-.558**
HCBs	.384	**.857**	-.140	.309
3CBs	.466	**-.848**	-.204	.047
4CBs	.428	**-.553**	-.414	.229
5CBs	**.640**	.296	**-.605**	.369
6CBs	.308	**.920**	-.193	.131
7CBs	**-.710**	.121	**.544**	.419
8CBs	**-.696**	.084	**.648**	.241
Eigenvalue	5.3529463	3.0670997	2.13569691	1.005977
Variance %	33.330085	26.187061	23.809193	13.02132
cumulative %	33.330085	59.517147	83.326340	96.34766
Identified Sources	Mixed	Mixed	Electronics	Pesticides
Percentage Contribution (100%)	50.2824858	0.8474576	44.915254	3.954802

Table S9

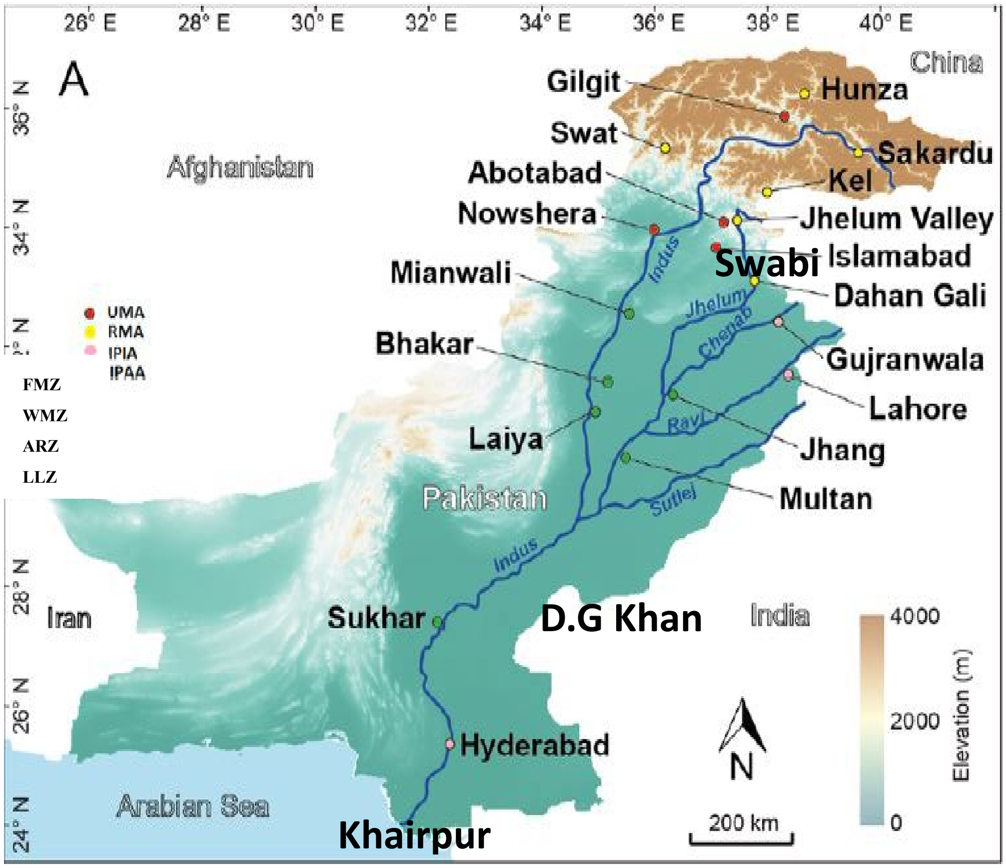
Compounds	**PC-1**
OPDDE	**1.000**
PPDDE	**1.000**
OPDDD	**1.000**
PPDDDOPDDT	**1.000**
PPDDT	**1.000**
HCBs	**-1.000**
3CBs	**1.000**
4CBs	**1.000**
5CBs	**1.000**
6CBs	**1.000**
7CBs	**1.000**
8CBs	**1.000**
Eigenvalue	12.000
Variance %	100.000
cumulative %	100.000
Identified Sources	Mixed
Percentage Contribution (100%)	100

Table S10

Compounds	**PC-1**	**PC-2**
OP-DDE	**.990**	.102
PP-DDE	**.996**	.045
OP-DDD	**.959**	.275
PP-DDD/OP-DDT	**.965**	.235
PP-DDT	**.974**	.172
HCBs	-.440	**.897**
3CBs	-.423	**.893**
4CBs	-.137	**.986**
5CBs	.362	**.905**
6CBs	**.592**	-.123
7CBs	**-.907**	.409
8CBs	.225	**.973**
Eigenvalue	6.519626	4.696721
Variance %	54.18559	39.28396
cumulative %	54.18559	93.46955
Identified Sources	Mixed	Industrial
Percentage Contribution (100%)	50	50

Table S11

Compounds	**PC-1**
OP-DDE	**1.000**
PP-DDE	**1.000**
OP-DDD	**1.000**
PP-DDD/OP-DDT	**1.000**
PP-DDT	**1.000**
HCBs	**1.000**
3CBs	**1.000**
4CBs	**1.000**
5CBs	**1.000**
6CBs	**1.000**
7CBs	**1.000**
8CBs	**-1.000**
Eigenvalue	12.000
Variance %	100.000
cumulative %	100.000
Identified Sources	Mixed
Percentage Contribution (100%)	100.000



26° E
28° E
30° E
32° E
34° E
36° E
38° E
40° E
A
36° N
34° N
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26° N
24° N
China
Gilgit
Hunza
Swat
Sakardu
Afghanistan
Abotabad
Kel
Nowshera
Jhelum Valley
Islamabad
Swabi
Mianwali
Dahan Gali
UMA
RMA
IPIA
IPAA
FMZ
WMZ
ARZ
LLZ
Indus
Jhelum
Chenab
Bhakar
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Ravi
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Sutlej
Pakistan
Indus
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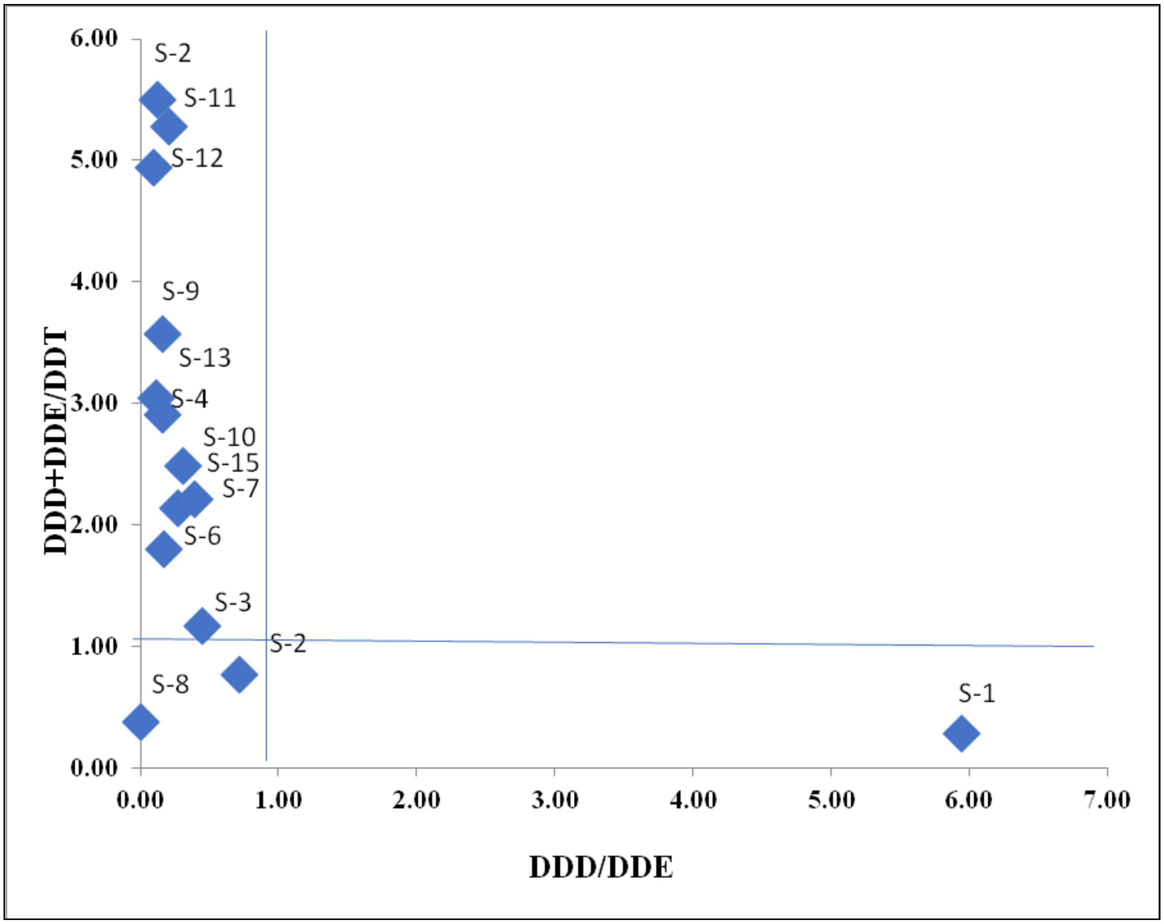
N
Arabian Sea
200 km
Khairpur

Figure S1



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3.00
2.00
1.00
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DDD+DDE/DDT
S-2
S-11
S-12
S-9
S-13
S-4
S-10
S-15
S-7
S-6
S-3
S-2
S-8
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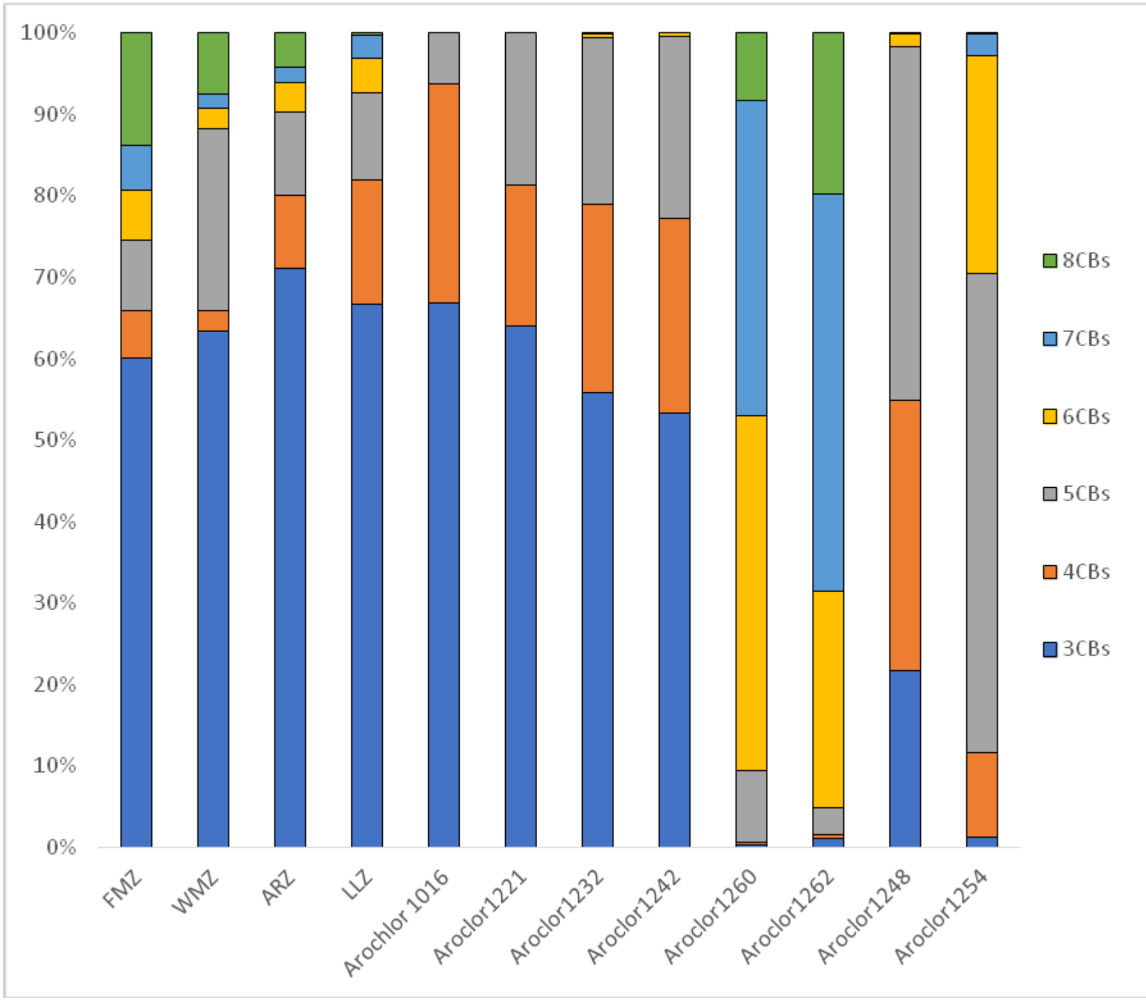
3.00
4.00
5.00
6.00
7.00
DDD/DDE

Figure S2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60%
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30%
20%
10%
0%
FMZ
WMZ
ARZ
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Aroclor1221
Aroclor1232
Aroclor1242
Aroclor1260
Aroclor1262
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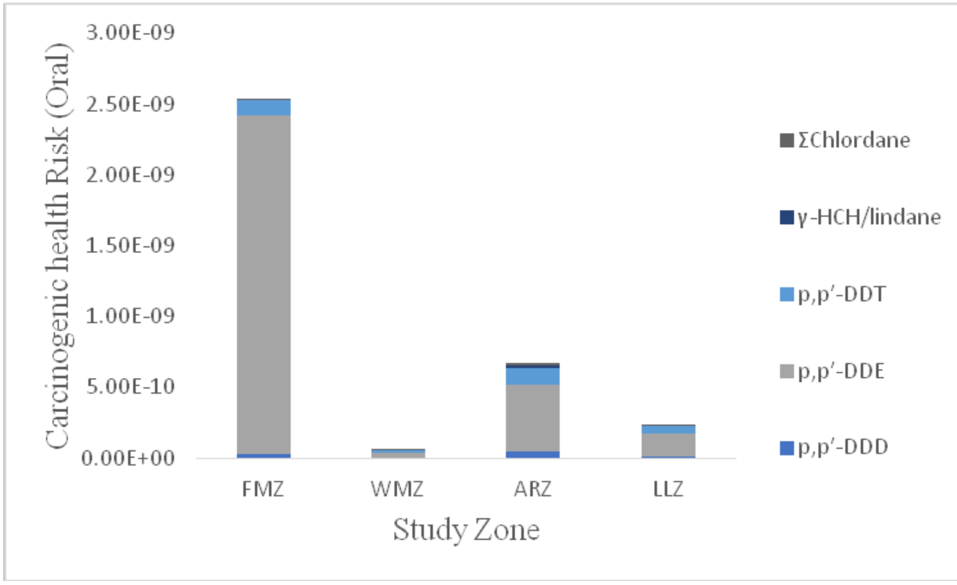
3CBs

Figure S3

Figure S4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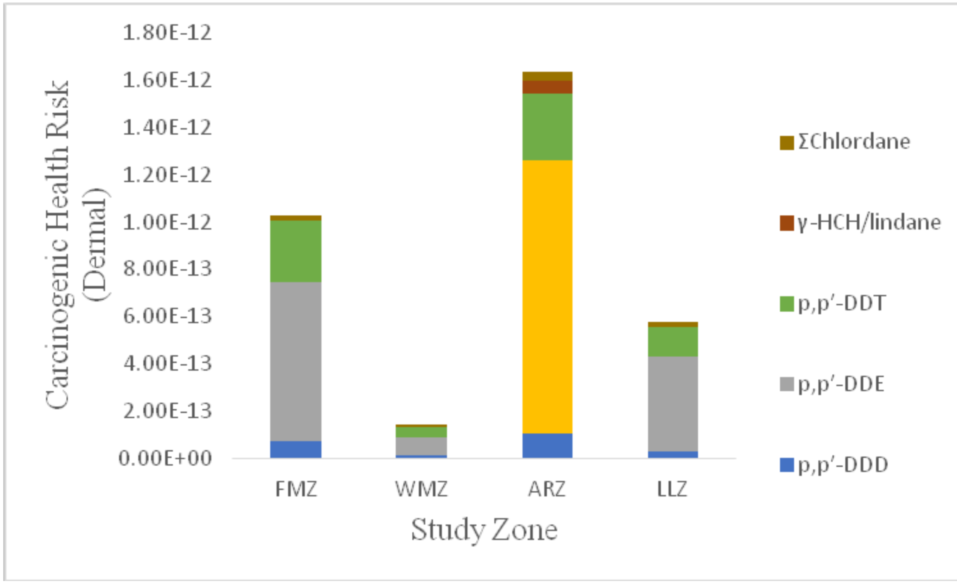
2.50E-09
2.00E-09
1.50E-09
1.00E-09
5.00E-10
0.00E+00
FMZ
WMZ
ARZ
LLZ
Study Zone
ΣChlordane
γ-HCH/lindane
p,p′-DDT
p,p′-DDE
p,p′-DDD

Figure S5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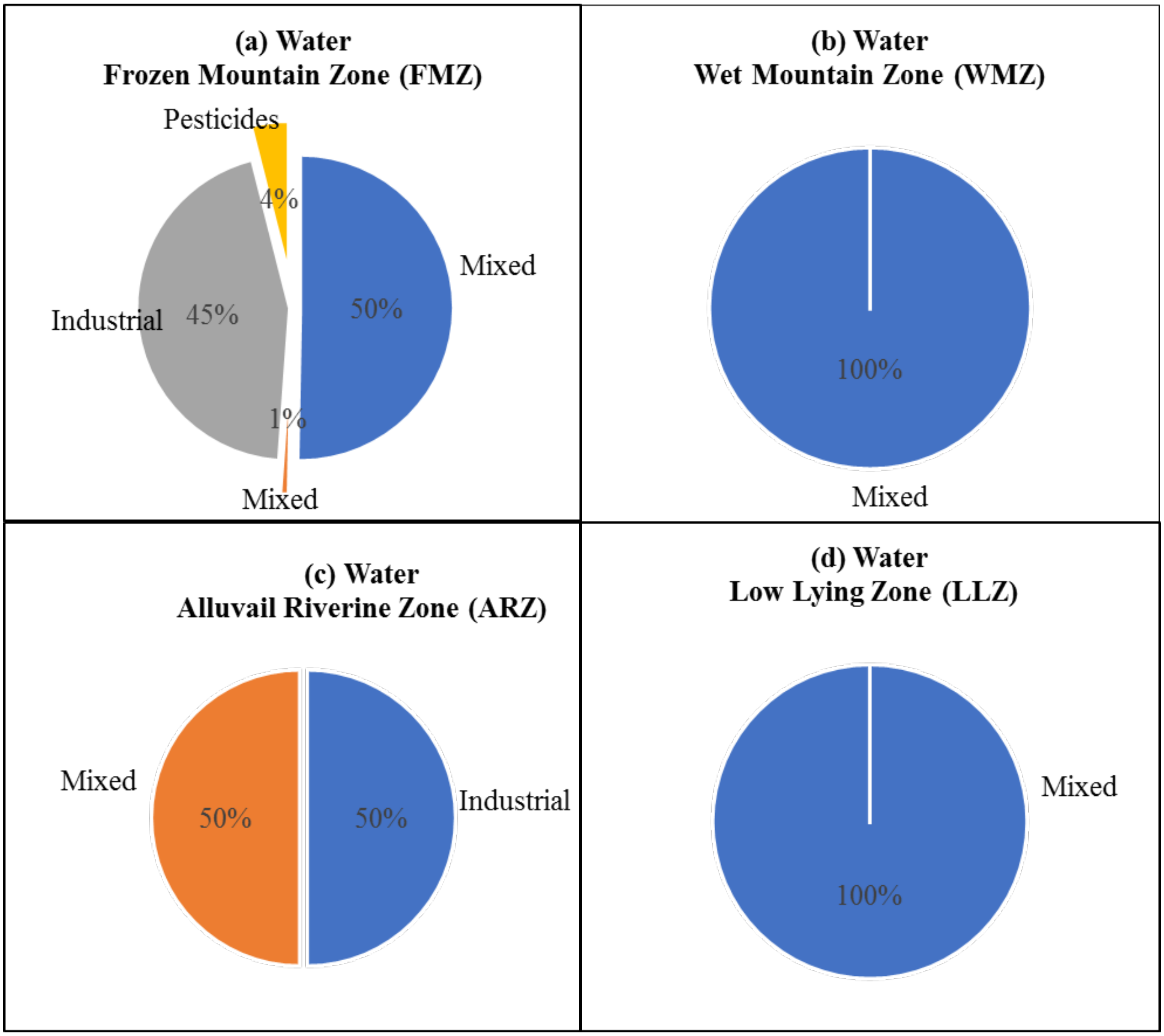
8.00E-13
6.00E-13
4.00E-13
2.00E-13
0.00E+00
FMZ
WMZ
ARZ
LLZ
Study Zone
ΣChlordane
γ-HCH/lindane
p,p′-DDT
p,p′-DDE
p,p′-DDD

Figure S6



Mixed: both pesticides and electronic usage

Figure S7

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