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ORIGINAL ARTICLE

Distribution and health risk assessment of organochlorine pesticides in agricultural soils of the Aghili plain, Southwest Iran

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Abstract

Organochlorine pesticides (OCPs) are a well-known group of persistent man-made chemicals. In the present study, 55 agricultural soil samples were collected from the Aghili plain and analyzed for 20 OCPs with the aim of determining contamination profiles, spatial distribution, influencing factors and carcinogenic risks. Among different OCPs, only aldrin, dieldrin, 4,4-dichlorodiphenyldichloroethylene, and 4,4-dichlorodiphenyldichloroethane were detected. The other OCPs dosages were lower than the detection limit in all samples. The results of cancer risk assessment demonstrated no carcinogenic risks to an exposed population.

Keywords Organochlorine pesticides · Agricultural soils · Health risk assessment · Pollution · Cancer risk assessment

Introduction

Organochlorine pesticides (OCPs) are a well-known group of persistent man-made chemicals that are extremely harmful to humans and wildlife (Pirard et al. 2018; Li et al. 2018; Behfar et al. 2013). Their accumulation in the environment, especially in soils, can cause significant stress on ecosystems (Artikov et al. 2018). The world consumption of OCPs in 1950–2000 was approximately two million tons per year (Fang et al. 2017). Although the application of OCPs was banned in most countries, they are still detected in different media such as soil, due to their previous and/or illegal use or disposal (Pirard et al. 2018; Li et al. 2018; Gao et al. 2013). OCPs are strongly retained by soil due to their insolubility in water (Artikov et al. 2018; Kafilzadeh 2015). Furthermore, they can be decomposed by environmental factors into other more complex and toxic compounds (Artikov et al. 2018). OCPs have the potential to enter into the food chain due to their lipophilicity and persistency in living organisms (Shahmoradi et al. 2019; Behrooz et al. 2009a). Upon entry, these compounds may cause neuro developmental disruption, reproductive disorders, thyroid-related diseases, diabetes and cancers, in organisms (Pirard et al. 2018; Barron et al. 2017).

Soil media have tremendous retention capabilities for OCPs (Qu et al. 2015; Alamdar et al. 2014) and, as a consequence, would be considered an important source of OCPs for other media as well as organisms (Yang et al. 2012). However, the distribution and fate of OCPs in soil are mainly influenced by soil characteristics, the geochemical behavior of chemicals, exposure period, and magnitude of application (Gopalan and Chenicherry 2018; Artikov et al. 2018; Zhou et al. 2013).

The Aghili plain is one of the most important agricultural centers in Khuzestan province, southwest of Iran. Many industries such as paper and pulp manufacturing, metallurgical industries, and cane factories are located on the plain (Ahmadi et al. 2019). It should be noted that OCPs are still used in Iran (Shahmoradi et al. 2019) and Behfar et al. (2013) reported different OCP levels along the Karun river in Khuzestan province (22.25–89.34 μg/L). In addition,

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previous investigations have demonstrated OCPs in agricultural products and food (Amrollahi et al. 2018; Amirahmadi et al. 2013), dairy products (Ashnagar et al. 2009; Koohi et al. 2011; Bayat et al. 2011), fish (Ebadi et al. 2006; Kafilzadeh 2015; Seifzadeh et al. 2018), sturgeon (Kajiwara et al. 2003), and human breast milk (Behrooz et al. 2009a; b; Shahmoradi et al. 2019; Cok et al. 1999). However, to the best of our knowledge, the study of OCP concentrations in agricultural soils in Iran is scarce. Since OCPs have been identified as endocrine-disrupting chemicals (Qu et al. 2015; Chen et al. 2015), the objectives of the present research were to investigate the spatial distribution, source identification, and risk assessment of OCPs in the collected agricultural soil samples of the Aghili plain, Khuzestan, Iran.

Materials and methods

Study area and sampling stations

The Aghili plain covers an area of 11,000 ha and is located between 32°1′ to 32°7′N and 48°52′ to 48°56′E. The plain is situated in a semi-arid zone of central Asia and has hot and dry climate. According to meteorological data, the average annual temperature and rainfall of plain are 33 °C and 326 mm, respectively. From the geological point, the Aghili plain can be considered as quaternary unconsolidated alluvial sediments (Fig. 1). In Spring 2017, 55 topsoil samples (0–30 cm) were collected using a stainless steel hand auger. At each sampling point, 1 ± 0.5 kg soil was taken from the mixed samples using a quartile method and then kept in polyethylene bags in the laboratory. Prior to further analysis, the sample was air dried and then sieved using a 2-mm steel sieve, and frozen at − 18 °C.

Extraction and analysis

Soil extraction was carried out using the QuEChERS method. First, 5 g of air-dried sample was weighed and transferred to the Falcon tube. A 1-mL mixture of surrogate standards of decachlorobiphenyl (PCB-209) and 2,4,5,6-tetrachloro-*m*-xylene (TCmX) was then added to each sample.

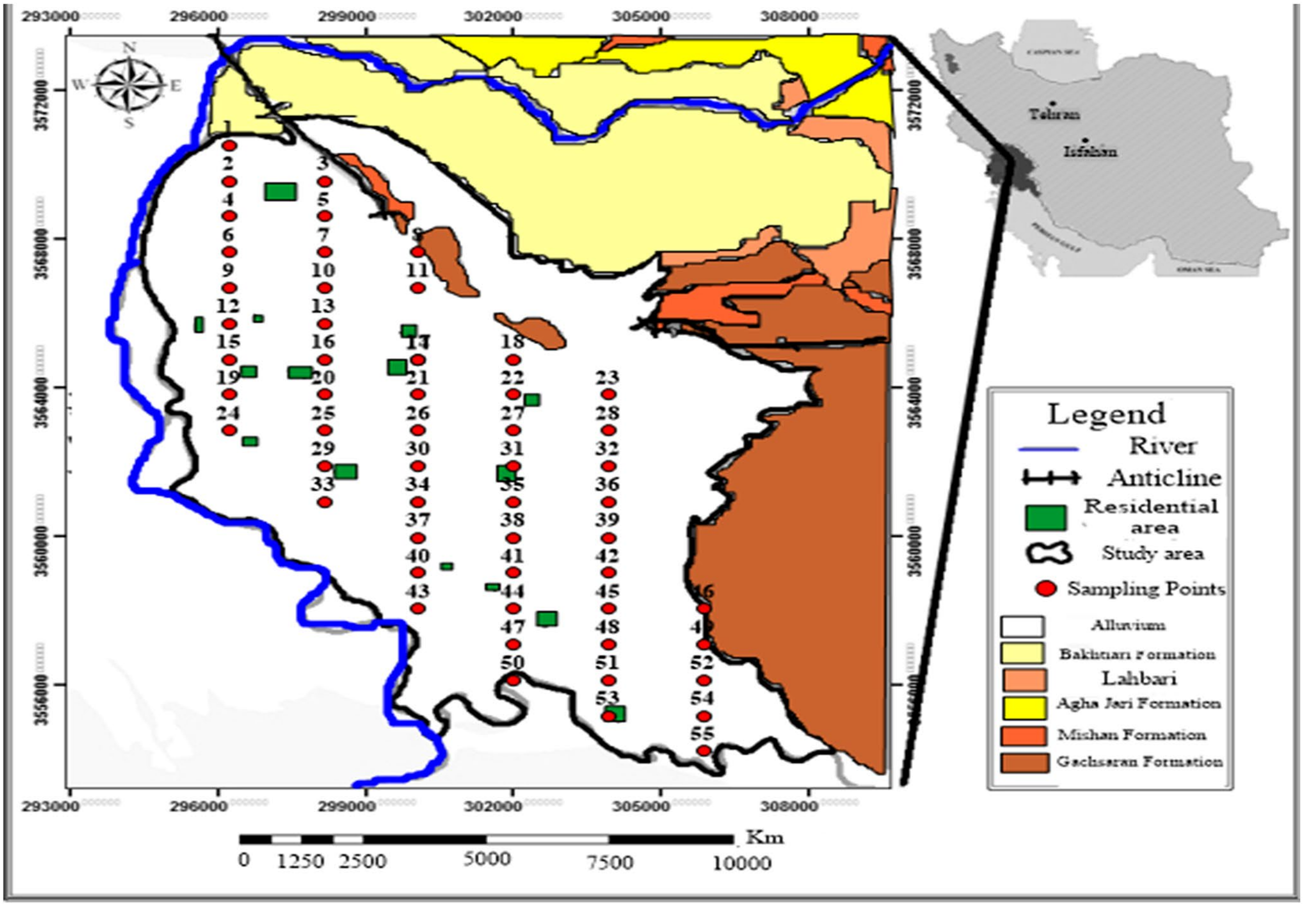

Fig. 1 Geological map of Aghili plain and sampling points in the study area

All materials were of analytical grade (Merck Company) and used without any further purification.

Analysis of OCPs was carriedout using an Agilent GC-ECD HP-6890 (Agilent Technologies) series system with a DB-5 capillary column (30 m, 0.32 mm, 0.25 µm). Nitrogen was used as a carrier gas at a flow rate of 1 mL/min under constant flow mode. The oven temperature began at 110 °C and increased to 280 °C at a rate of 5 °C/min. The injector temperature was 280 °C. A 2-µL sample was injected into the GC-ECD for analysis.

Physiochemical characteristics of soil samples

The hydrometer method was used to find the size distribution of soil particles (Gee et al. 1986). In addition, electrical conductivity (EC) and acidity (pH) of soil samples were measured using a mixture of soil and distilled water with a 1:2.5 (w/v) ratio; these were shaken continuously for 15 min prior to pH measurement (Ryan et al. 2007).

Quality control and quality assurance (QC/QA)

All chemicals were of analytical grade and supplied (Merck and Sigma-Aldrich Co.). All laboratory glassware was washed with 30% nitric acid, followed by rinsing using deionized water and drying in an oven, prior to analysis. The instruments were calibrated daily with calibration standards. The target substances were identified via the retention times and quantified using an internal standard. Duplicates, standard OCP solution, and blank reagents were analyzed to confirm the concentrations. Solvent blanks, procedural blanks and real samples were analyzed using the same procedure.

Statistical analysis

Analysis of experimental data was performed via Surfer 14, Excel, XLSTAT 2016 and Excel 2016 software for windows. The original concentration data normality was evaluated using Kolmogorov–Smirnov test (significance level was considered as P value ≤ 0.05). Spearman correlation analysis was applied for assessment of correlation among OPCs. In the statistical analysis, those original concentrations lower than the detection limit (DL) were presumed to be equal to 0.75 of DL.

Risk assessments

The incremental lifetime cancer risk (ILCR) shows the probability that an individual will develop cancer during his/her lifetime as a result of exposure to carcinogenic compounds (Qu et al. 2015). Three pathways of human exposure to carcinogenic chemicals through soil contamination are (1) direct ingestion of substrate particles, (2) dermal absorption. and (3) inhalation of re-suspended particles emitted from soil (Qu et al. 2015; Moore et al. 2015):

$$\text{ILCR}_{\text{ingest}} = \frac{C_{\text{Soil}} \times \text{IngR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \times \text{CSF}_{\text{oral}} \times \text{CF},$$

$$\text{ILCR}_{\text{dermal}} = \frac{C_{\text{Soil}} \times \text{SA} \times \text{FE} \times AF_{\text{Soil}} \times \text{ABS} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \times \text{CF} \times \text{CSF}_{\text{oral}} \times \text{ABS}_{\text{GI}},$$

$$\text{ILCR}_{\text{inhale}} = \frac{C_{\text{Soil}} \times \text{InhR} \times \text{AF}_{\text{inh}} \times \text{EF} \times \text{ED}}{\text{PEF} \times \text{AT}} \times \text{CSF}_{\text{inh}},$$

$$\text{Total CR} = \sum \text{ILCR}_{\text{ingest}} + \text{ILCR}_{\text{dermal}} + \text{ILCR}_{\text{inhale}},$$

where, C_{soil} is the contaminant concentration in soil (mg/kg), CSF is the carcinogenic slope factor $(\text{mg/kg/day})^{-1}$, IngR is the soil ingestion rate (mg/day), EF is the frequency of exposure (day/year), ED is the exposure duration (year), BW is the average body weight (kg), AT is the average lifespan (day), CF is the convention factor (10^{-6} kg/mg), SA is the surface area of the skin (cm^2), FE is the fraction of dermal exposure ratio to soil, AF_{soil} is the skin adherence factor for soil (mg cm^2), ABS is the dermal absorption factor (chemical-specific), ABSGI is the fraction of contaminant absorbed in the gastrointestinal tract, InhR is the inhalation rate (m^3/day) and PEF is the particle emission factor (Ge et al. 2013). Table 1 shows the values used in the applied risk assessments. Furthermore, total cancer risk is the estimation of cancer risk through different pathways (ingestion, dermal contact, and inhalation). The total cancer risk is categorized as—value $\le 10^{-6}$: very low; $10^{-6} < \text{value} \le 10^{-4}$: low; $10^{-4} < \text{value} \le 10^{-3}$: moderate; $10^{-3} < \text{value} \le 10^{-1}$: high; and value $\ge 10^{-1}$: very high (Ge et al. 2013).

Results and discussion

Grain size and soil characteristics in the Aghili plain

Hydrometer analysis revealed the agricultural soils of the Aghili plain to have a silty clay loam texture. The mean standard deviation values of pH and EC of soil samples from the Aghili plain were 7.85 ± 0.31 and 2.20 ± 1.51 ms/cm, respectively.

Concentration of organochlorine pesticides

The statistical range, mean, standard deviation, and detection frequencies for each OCP in the agricultural soil of the Aghili plain are summarized in Table 2. Most of the analyzed compounds (16 of the 20) were below the detection

Table 1 Parameters used in human health risk assessments (Ge et al. 2013; Qu et al. 2015)

Variable (symbol)	Unit	Values	
		Child	Adult
Ingestion rate (IngR)	mg/day	200	100
Exposure frequency (EF)	days/year	350	350
Exposure duration (ED)	year	6	30
Surface area (SA)	cm^2	2800	5700
Body weight (BW)	kg	15	70
Dermal exposure ration (FE)	unitless	0.61	0.61
Dermal surface factor (AF)	mg/cm	0.2	0.07
Inhalation rate (InhR)	m^3/day	10.9	15.8
Particle emission factor (PEF)	m^3/kg	$1.36* 10^9$	$1.36* 10^9$
Averaging time (AT)	year	For carcinogenic effects AT = 70	For carcinogenic effects AT = 70
Dermal absorption fraction (ABS)	–	0.13	0.13

Table 2 Concentrations of organochlorine pesticides in agricultural soils of Aghili plain, Iran (ng/g)

OPCs	*n*	Minimum	Maximum	Mean ± STD	Detection frequencies (%)
Alfa-HCH	55	ND	ND	–	0
Beta-HCH	55	ND	ND	–	0
Gamma-HCH	55	ND	ND	–	0
Delta-HCH	55	ND	ND	–	0
Heptachlor	55	ND	ND	–	0
Aldrin	55	ND	1.52	0.54 ± 0.41	60
Heptachlor epoxide	55	ND	ND	–	0
Gamma chlordane	55	ND	ND	–	0
Alpha-endosulfane	55	ND	ND	–	0
Alpha-chlordane	55	ND	ND	–	0
Dieldrin	55	ND	0.27	0.15 ± 0.016	1.8
4,4-DDE	55	ND	2.08	0.31 ± 0.31	52.7
Endrin	55	ND	ND	–	0
Beta endosulfane	55	ND	ND	–	0
4,4-DDD	55	ND	1.29	0.54 ± 0.11	3.6
Endrin aldhyde	55	ND	ND	–	0
4,4-DDT	55	ND	ND	–	0
Endosulfan sulfate	55	ND	ND	–	0
Methoxychlor	55	ND	ND	–	0
Endrin keton	55	ND	ND	–	0

limits in all samples and their detection frequencies were also 0%. However, the concentration ranges were from not detected (ND) to 1.52 ng/g with a mean value of 0.54 ng/g for aldrin, ND to 0.27 ng/g (mean 0.15 ng/g) for dieldrin, ND to 2.08 ng/g (average 0.31 ng/g) for 4,4-dichlorodiphenyldichloroethylene (DDE), and ND to 1.29 ng/g (mean 0.54 ng/g) for 4,4-dichlorodiphenyldichloroethane (DDD).

A low concentration of OCPs in agricultural soils indicated their complete degradation via microorganisms, surface run-off, leaching, wind erosion, and volatilization (Artikov et al. 2018; Gopalan and Chenicherry 2018). Moreover, almost complete degradation of non-detectable OCPs confirms no recent application (Artikov et al. 2018). On the other hand, 60, 52.7, 3.6, and 1.8% of the soil samples contained aldrin, dieldrin, 4,4-DDD, and 4,4-DDE, respectively. The presence of these detectable OCPs confirmed the possible persistence against decay, adsorption onto soil particles, and recent use (Artikov et al. 2018). Aldrin quickly breaks into dieldrin, which is more resistant to environmental degradation (Gopalan and Chenicherry 2018; Ssebugere et al. 2010). Hence, the high concentration of aldrin, in comparison with dieldrin confirms its fresh input in the study area. Under aerobic and anaerobic soil conditions, dichlorodiphenyltrichloroethane (DDT) can be converted into DDD and DDE (Gopalan and Chenicherry 2018). Generally, the ratio of DDE + DDD/DDT < 1 might suggest the presence of fresh DDT sources (Barron et al. 2017; Fang et al. 2017; Gao et al. 2013). The concentration of DDT in the Aghili plain agricultural soils was found to be below detection at all stations; however, the presence of its toxic metabolites at some stations confirmed its persistent nature in the environment (Gopalan and Chenicherry 2018; Li et al. 2018). In addition, the higher levels of DDE compared to DDD at most stations, indicates that parental DDT was subjected to aerobic degradation (in the presence of air and sunlight) (Alamdar et al. 2014; Tarcau et al. 2013). On the other hand, the higher ratio of DDD/DDE at stations 7 and 35 displayed the anaerobic degradation of parental DDT (Alamdar et al. 2014; Tarcau et al. 2013).

In comparison with previous reports about OCPs in agricultural soils from different regions (Table 3), the levels of aldrin in this study are much lower than in central China (Zhou et al. 2013) and India (Gopalan and Chenicherry 2018), while their concentration is slightly higher than in Uzbekistan (Artikov et al. 2018) and Shanghai (Jiang et al. 2009). The mean concentration of dieldrin in Aghili soils is lower than in Central China (Zhou et al. 2013), Mexico (Wong et al. 2010), India (Gopalan and Chenicherry 2018), and Uzbekistan (Artikov et al. 2018). However, the mean concentration of dieldrin in Shanghai soils is slightly lower than the Aghili plain (Jiang et al. 2009). In addition, the levels of DDE, and DDD in this study are much lower than in China (Gao et al. 2013; Zhou et al. 2013; Jiang et al. 2009), Mexico (Wong et al. 2010), Uzbekistan (Artikov et al. 2018), and India (Gopalan and Chenicherry 2018).

Distribution of organochlorine pesticides

The spatial distribution of detected OCPs is illustrated in Fig. 2. It shows that the trends for detected OCPs varied at different stations. Generally, vegetables are more vulnerable to damage by insects than grain crops, since they are subject to higher pesticide application per unit area in farmlands (Qu et al. 2015). Based on the results, distribution of all detected OCPs in stations 7 and 35 are

Table 3 Concentrations of OCPs in agricultural soils from different regions mean (range) (ng/g)

Study area	Hongze Lake, China	Central China	Mexico	Uzbekistan	Palakkad, India	Shanghai, China
Sampling point	33	44	16	8	6	36
Alfa-HCH	3.79 (ND–17.53)	264 (ND–44.43)	–	0.45 (ND–3.16)	ND–0.9	0.48 (ND–1.46)
Beta-HCH	4.80 (ND–19.65)	6.41 (ND–53.12)	–	ND	ND–6.8	1.19 (ND–9.52)
Gamma-HCH	1.67 (ND–7.12)	1.20 (ND–8.23)	–	ND	ND–0.7	0.36 (ND–4.15)
Delta-HCH	0.95 (ND–3.36)	5.14 (ND–32.32)	–	ND	–	0.38 (ND–1.76)
Heptachlor	–	5.34 (ND–28.71)	ND	–	ND–7	1.05 (ND–6.07)
Aldrin	–	8.27 (ND–21.56)	ND	0.06 (ND–0.50)	ND–13.2	0.32 (ND–6.62)
Heptachlor epoxide	–	0.20 (ND–3.65)	0.05	–	ND–7.6	0.92 (ND–4.78)
Gamma chlordane	–	0.20 (ND–6.45)	–	–	–	0.20 (ND–1.41)
Alpha-endosulfane	–	0.34 (ND–5.88)	–	ND	–	0.13 (ND–1.77)
Alpha-chlordane	–	0.28 (ND–4.70)	–	–	–	0.10 (ND–1.21)
Dieldrin	–	0.42 (ND–16.99)	0.22	0.26 (ND–1.12)	ND–7.51	0.09 (ND–1.38)
4,4-DDE	12.82 (1.86–73.22)	73.38 (ND–807.82)	–	0.50 (0.07–1.49)	ND–5.5	16.14 (0.17–77.76)
Endrin	–	0.16 (ND–6.12)	–	10.71 (2.41–20.87)	ND–65.1	0.24 (ND–4.32)
Beta endosulfane	–	0.94 (ND–23.04)	57	10.47 (ND–74.56)	–	–
4,4-DDD	6.27 (ND–35.60)	9.26 (ND–62.78)	–	0.67 (ND–1.56)	ND	4.56 (0.15–124.67)
4,4-DDT	7.55 (ND–50.13)	52.74 (ND–436.91)	10	ND	ND	3.26 (ND–24.52)
Endosulfan sulfate	–	1.28 (ND–23.04)	–	–	–	–
Methoxychlor	–	10.47 (ND–169.03)	23	–	–	–
References	Gao et al. (2013)	Zhou et al. (2013)	Wong et al. (2010)	Artikov et al. (2018)	Gopalan and Chenicherry (2018)	Jiang et al. (2009)

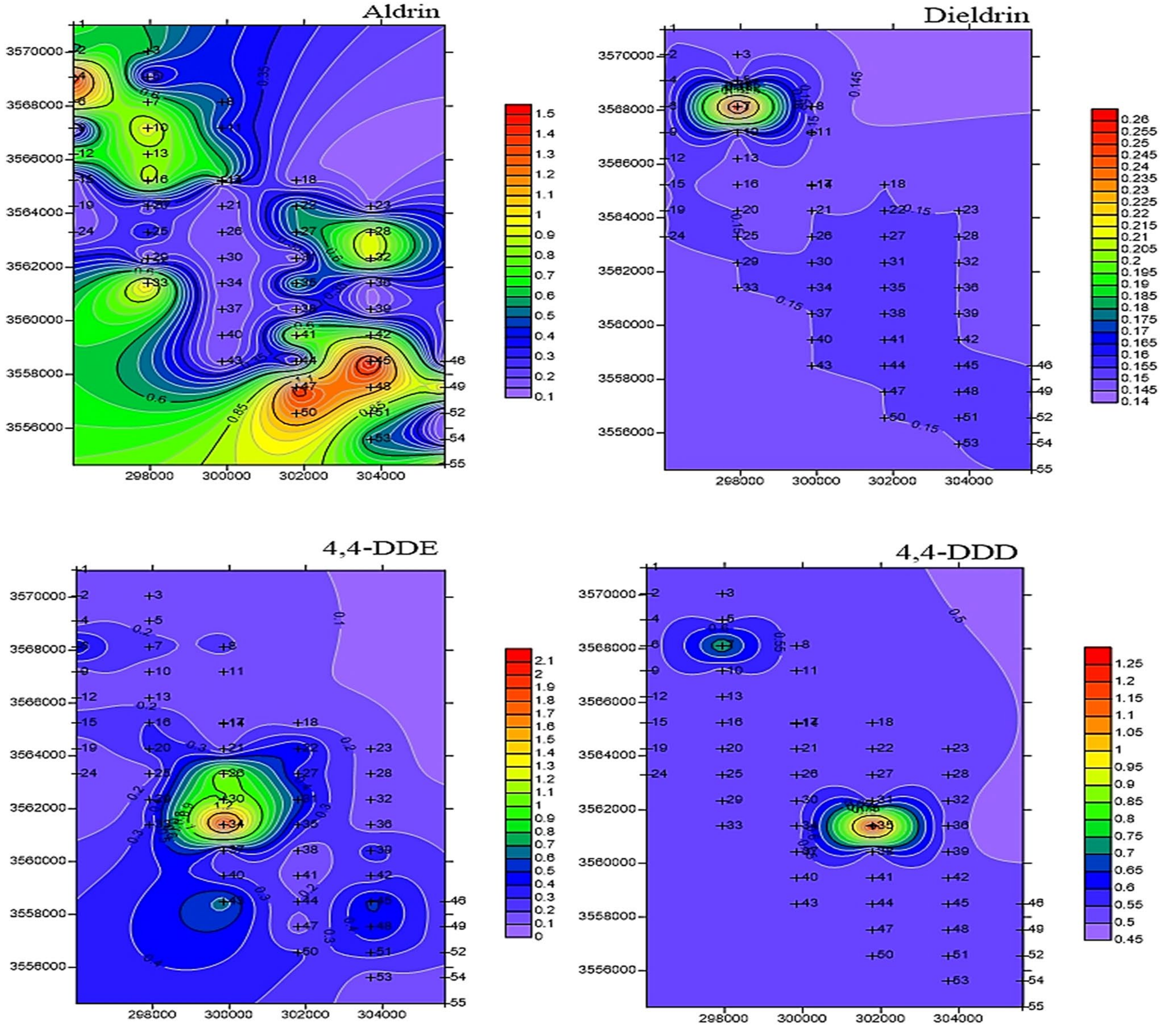

Fig. 2 Spatial distribution of detected OCPs in the agricultural soils of the Aghili plain

noticeable. The variation tendencies of aldrin confirmed that aldrin is the most popular OCP used in the study area.

Correlation and principal component analysis of organochlorine pesticides

The possible correlation between the physiochemical parameters of soil samples and detected OCPs were evaluated by non-parametric Spearman correlation analysis as the results of the normality test showed the lack of normality of data. As shown in Table 4, a significant positive correlation between 4,4-DDD and dieldrin ($P < 0.01$, $r > 0.6$) indicated their persistence in agricultural soils. The significant positive relationship between dieldrin and 4,4-DDD was also reported by Gopalan and Chenicherry (2018). Moreover, a moderate linear relationship between EC and aldrin ($P < 0.05$, $r > 0.3$) suggested that the distribution and sorption of aldrin may be under the control of soil salinity. A weak negative correlation was observed between pH and EC ($P < 0.05$, $r = -0.3$). Furthermore, no significant correlation was found among the other studied parameters. Ge et al. (2013) also reported insignificant relationships between pH and OCPs. Since soil pH can affect the structure and composition of humic acids (Gao et al. 2013), these results are not surprising. Overall, these results indicated that OCP concentration in the agricultural soils of the study area is not controlled by soil characteristics; however, the physiochemical properties of the OCPs do regulate their fate in soils.

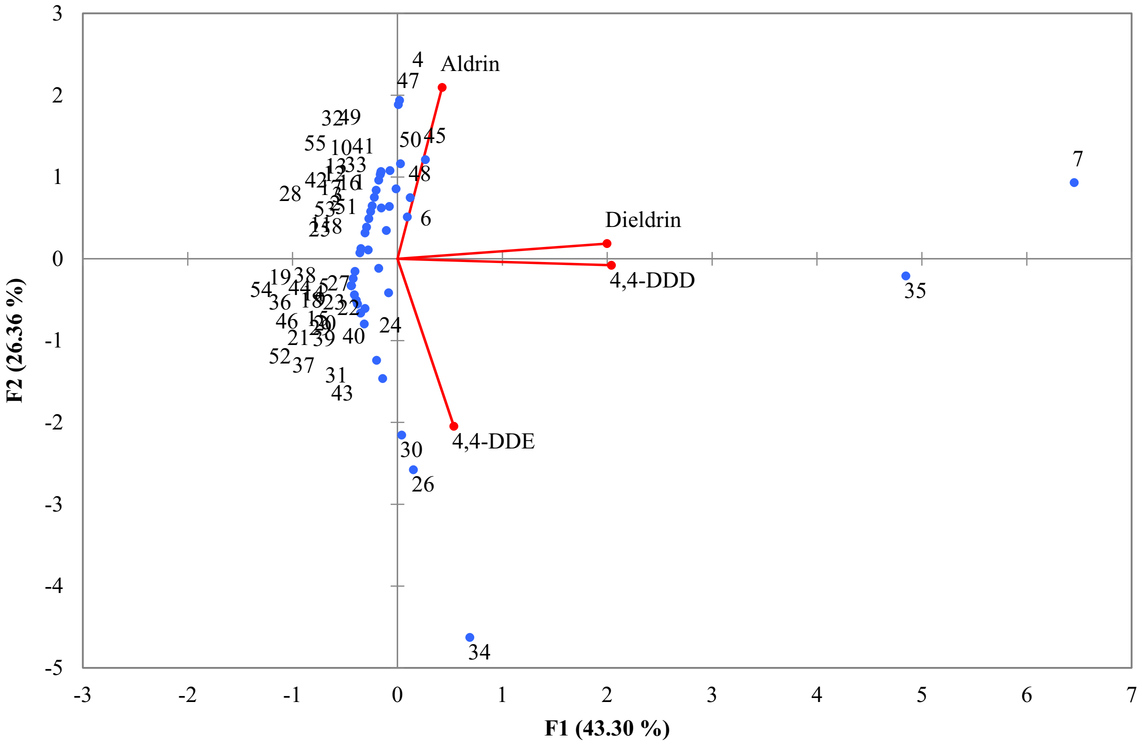

Fig. 3 Principal component (PC) biplot of the sampling sites and OCPs in the agricultural soils of the Aghili plain

Figure 3 illustrates the principal component (PC) biplot of the sampling sites and OCPs in the agricultural soils of the Aghili plain. Results of the applied PC biplot were in agreement with those of spatial distribution and correlation analysis. The first three PCs represented 92.39% of the variance within the data set. The first PC (PC1), second PC (PC2) and third PC (PC3) displayed 43.30, 26.36, and 22.72% of the total variance, respectively. PC1 had a high positive loading of dieldrin and 4, 4-DDD, indicating similar physiochemical properties of these compounds. PC2 was highly associated with aldrin, the most dominant OCP in the study area. PC3 correlated well with 4,4-DDE. The PC biplot (Fig. 3) also shows the spatial distribution of the soil samples based on the OCP concentration. Generally, the stations in the first and fourth quadrants displayed a relatively high concentration of contaminants, while the plotted stations in the second and third quadrants exhibited a low concentration of contaminants (Akhbarizadeh et al. 2017). As shown in Fig. 3, in the Aghili plain, stations 4, 6, 7, 26, 30, 34, 35, 47, 48, and 50 displayed relatively a high concentration of OPCs. Among them, stations 7 and 35 showed a higher level of all detected OCPs, whereas station 4 and 26 had the highest concentration of aldrin and 4,4-DDE, respectively.

Risk assessment of organochlorine pesticides

The lifetime carcinogenic risk of detected OCPs in the agricultural soils of the Aghili plain was calculated through ingestion, dermal contact, and inhalation and the results are presented in Fig. 4. All calculated risk values for both age groups at all sampling sites are well below the target risk level (10^{-6}); hence, the cancer risk of OCPs is negligible in the study area. For different exposure pathways, the decreasing trend in cancer risks for both age groups is ingestion > dermal contact > inhalation. Moreover, the total cancer risk ranks very low (values $< 10^{-6}$).

Conclusion

Concentration, spatial distribution, and risk assessment of OCPs in the agricultural soils of the Aghili plain, Khuzestan, Iran were investigated. The results revealed that the agricultural soils were contaminated with OCPs and recent application of OCPs was highly unlikely. Among different OCPs, only aldrin was still used in agricultural activities in the study area. However, due to the long-term application and persistent nature of OCPs, some residual compounds (i.e., dieldrin, 4,4-DDD, and 4,4-DDE) were detected in the samples. The findings of lifetime carcinogenic risk

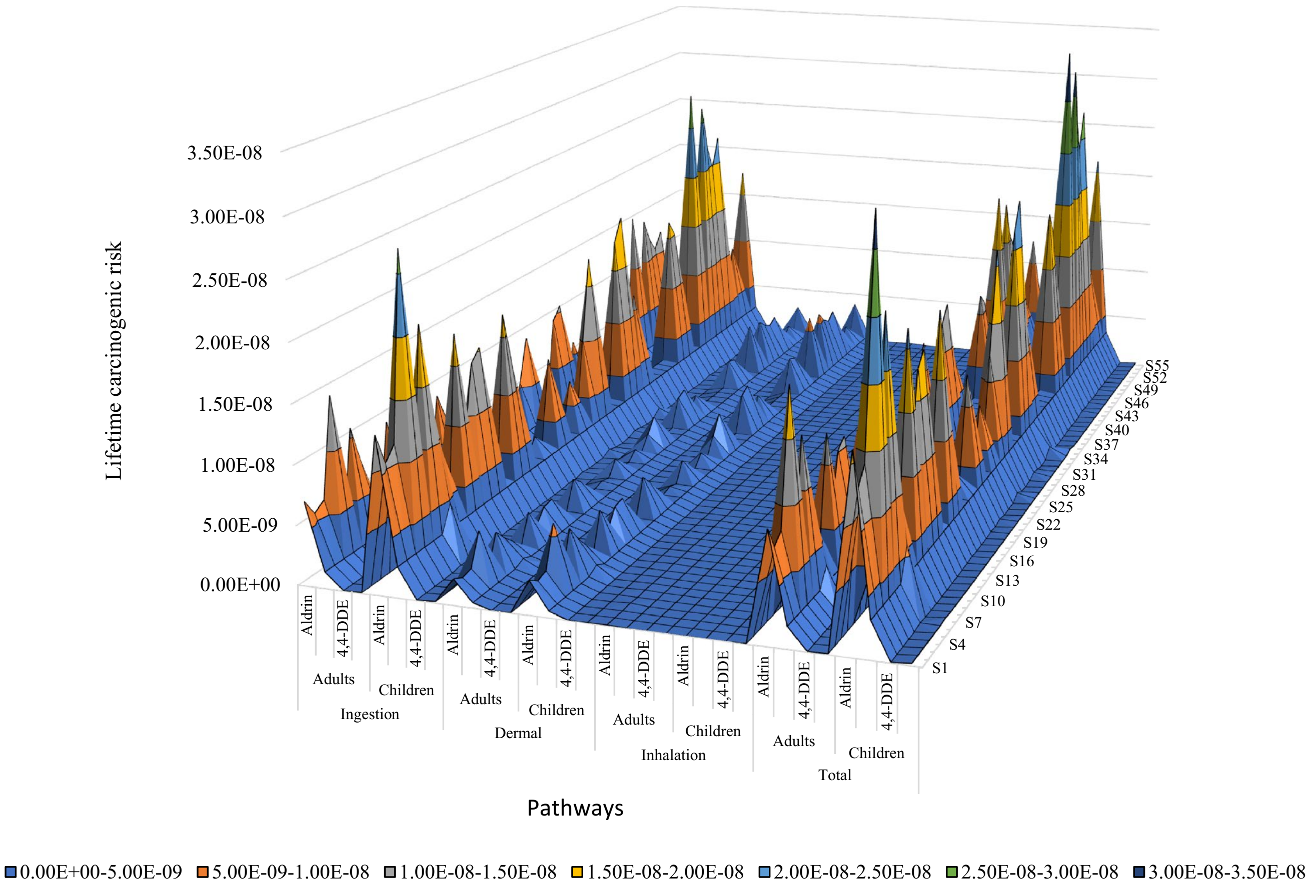

Fig. 4 Lifetime carcinogenic risk (LCR) of the detected OCPs in the agricultural soils of the Aghili plain through different pathways

assessments via ingestion, dermal and inhalation of agricultural soils indicated that the risks for residents are at a very low level. However, the cultivated vegetables may be polluted and should be analyzed in order to guarantee the health of the residents who live in the study area.

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