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ARTICLE

# Organochlorine pesticides (OCPs) in soils near and around Lake Son-Kul in the western Tian Shan Mountains, Central Asia

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## Abstract

**Purpose** Summer pasture development and tourism near and around Lake Son-Kul in the western Tian Shan Mountains has resulted in a local source of organochlorine pesticides (OCPs) that lack monitoring and evaluation.

**Materials and methods** Twenty-eight soil samples were collected from the Lake Son-Kul Basin (SKB) and Naryn River Valley (NRV) to measure OCP concentrations and evaluate their composition, transport processes, and ecological risks.

**Results and discussion** Except for the high OCP concentration measured at one location, site 7 (145.32 ng/g dw), total OCP concentrations in the SKB ranged from 2.36 to 24.13 ng/g dw, slightly higher than concentrations in the NRV (1.69–7.45 ng/g dw) and similar to values in polar regions. Among the OCPs, HCHs and methoxychlor were the predominant compounds that accumulated in soils, followed by DDTs and Aldrins, whereas Chlors and Endos displayed lowest concentrations, indicating that animal husbandry is the main agricultural activity in the study area. Most of the less volatile pesticides, including β-HCH, p,p-DDT, and Chlors, represent past use from local sources, for instance the pesticides warehouse at site 7. More volatile compounds such as α-HCH, γ-HCH, and p'p-DDE represent exogenous atmospheric transport to the study area, as demonstrated by their isomeric ratios. Ecological risk assessment showed that the surface environment of the study area remains in good condition.

**Conclusions** Findings from this study provided a snapshot of the current status of OCP contamination in soils of the Lake Son-Kul region. Although historical OCP contamination has impacted the lake's ecology, catchment soils display low levels of OCP contamination.

**Keywords** Ecological risks · OCPs · Soil · Sources · Son-Kul · Tian Shan Mountains

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## 1 Introduction

Man-made organochlorine pesticides (OCPs), some of the best-known organic pollutants, are of great concern because they are toxic, persistent, and bioaccumulate in nature. As semi-volatile compounds, OCPs can undergo long-range atmospheric transport (LRAT) and be distributed widely, even to remote mountain regions where they are generally considered to be derived from distant sources (Khairy et al. 2016; Yang et al. 2018). Remote mountain regions are considered to reflect near-background levels of OCPs on Earth because of their low human population densities and agro-engineering activities (Daly and Wania 2005; Yuan et al. 2012). Recently, however, rapid development of agriculture, animal husbandry, and tourism in mountain regions has led to greater

pollutant accumulation, often more than the local region can handle (Yu et al. 2012; Brahney et al. 2015). Such development in remote mountain regions has resulted in local sources of OCP release, leading to higher-than-expected OCP concentrations. In view of the risks that OCPs pose to water resources, sensitive species, and local residents, OCP pollution has been identified as a potential environmental problem in remote mountain regions (Tao et al. 2014; Li et al. 2018).

The Tian Shan Mountains (40°~45°N, 67°~95°E) extend west-east for approximately 2500 km from northwest China to Kazakhstan, Kyrgyzstan, Uzbekistan, and Tajikistan, and represent one of the largest mountain systems in the world. Because the Tian Shan Mountains are located in the Asian hinterland, at an average altitude of 4000 m a.s.l., the region is thought to be a pristine environment. It also supports numerous species of rare flora and fauna (Baumer 2012; Zhang et al. 2015). In the past few years, the environment of the Tian Shan Mountains has been threatened by several natural and anthropogenic processes, i.e., glacier recession that causes debris flows and floods (Erokhin et al. 2018) and contamination by uncontrolled agricultural and industrial activities (Zhang et al. 2015; Bekmamat et al. 2016; Shen et al. 2017). Human activity has been an important driver of environmental degradation, especially in the areas surrounding mountain lakes, where human populations concentrate. The Lake Bosten region (~ 1000 m a.s.l.), in the eastern Tian Shan Mountains, has been subject to pollution by polycyclic aromatic hydrocarbons (PAHs) (25.2–491.0 ng/g dw) and OCPs (6.9–16.7 ng/g dw), a consequence of human activities such as farming and tourism (Shen et al. 2016a, 2017). The Lake Issyk-Kul region (~ 1600 m a.s.l.) in the western Tian Shan region is threatened by radioactive material and other toxic products derived from the mining industry (Bekmamat et al. 2016; Li et al. 2018). Another mountain lake in the western Tian Shan Mountains, Lake Son-Kul (~ 3000 m a.s.l.), which is surrounded by extensive pastures in summer, was the site of an environmental accident in which a pesticide warehouse filled with OCPs was destroyed by a flood in 1976, causing widespread fish deaths the following year (Hadjamberdiev and Begaliev 2004). Follow-up surveys of OCP contamination in the Lake Son-Kul region, however, have been limited or nonexistent. Hence, it would be prudent to evaluate the legacy and current ecological risks of OCPs in the Lake Son-Kul region. Lake Son-Kul lies at high altitude and has a cold climate. The area can be regarded as a major condensation point that is subject to the "distillation effect," and studies there can help us gain a better understanding of long-range atmospheric transport and deposition of OCPs in such environments.

Soils are the major terrestrial repository of OCPs and are a suitable matrix for studies that evaluate long-term accumulation of these contaminants, as well as transfers among different reservoirs that accumulate OCPs in the mountains, e.g., soils, lake sediments, plants, and animals (Vighi 2006; Zhao

et al. 2013). Soils play an important role in the global distribution and transport of OCPs because they not only have a large retention capacity but also emit old organic pollutants into the air as a secondary source, providing a complete picture of the main distribution phenomena that occur in an area (Meijer et al. 2003; Zhan et al. 2017). In this study, we analyzed OCPs in soil samples near and around Lake Son-Kul in the western Tian Shan Mountains. Our general objectives were to (1) determine residual concentrations of OCPs in soils, (2) ascertain the transport pathways and possible sources of such contaminants, and (3) evaluate the potential ecological risks from OCPs in local alpine ecosystems. This study is the first to assess the fate and transport of OCPs in the Lake Son-Kul region.

## 2 Materials and methods

### 2.1 Study area

The study area is located near and around Lake Son-Kul, in the Naryn River Basin, central Kyrgyzstan (Fig. 1a). The Naryn River originates in the western part of the Tian Shan Mountains and flows west through the Ferghana Valley, where it merges with the Kara Darya River to form the Syr Darya River. The Naryn River is one of the main tributaries of the Syr Darya, and it ultimately flows toward the northern Aral Sea (Fig. 1b) (Hagg et al. 2013). The study area is located in the upper reaches of the Naryn River Basin, which flows through the Naryn River Valley (1500–2500 m a.s.l.) (Fig. 1c). The climate of the Naryn River Valley is dominated by the prevailing westerlies that supply the largest share of annual precipitation in the spring and winter. Annual precipitation ranges from 280 to 450 mm, depending on altitude, and the average temperature is approximately 3 °C (Kriegel et al. 2013). Through the connection of the Son-Kul and Naryn Rivers, Lake Son-Kul fills the central depression of a plateau between two generally east-west-trending Tian Shan mountain ranges, the Son-Kul-Too Range in the north and the Boor-Albas Range in the south (Fig. 1c). Mean annual temperature in the lake drainage basin is − 3.5 °C, with mean temperatures of − 20 °C in January and 11 °C in July. Mean seasonal precipitation is 300–400 mm from April to October and 100–150 mm from November to March (Mathis et al. 2014; Pacton et al. 2015). The Lake Son-Kul Basin is famous for summer (June–September) pasturing and tourism.

### 2.2 Sample collection and preparation

Twenty-eight soil samples were collected near and around Lake Son-Kul (41.3744° N~42.6263° N, 75.0186°
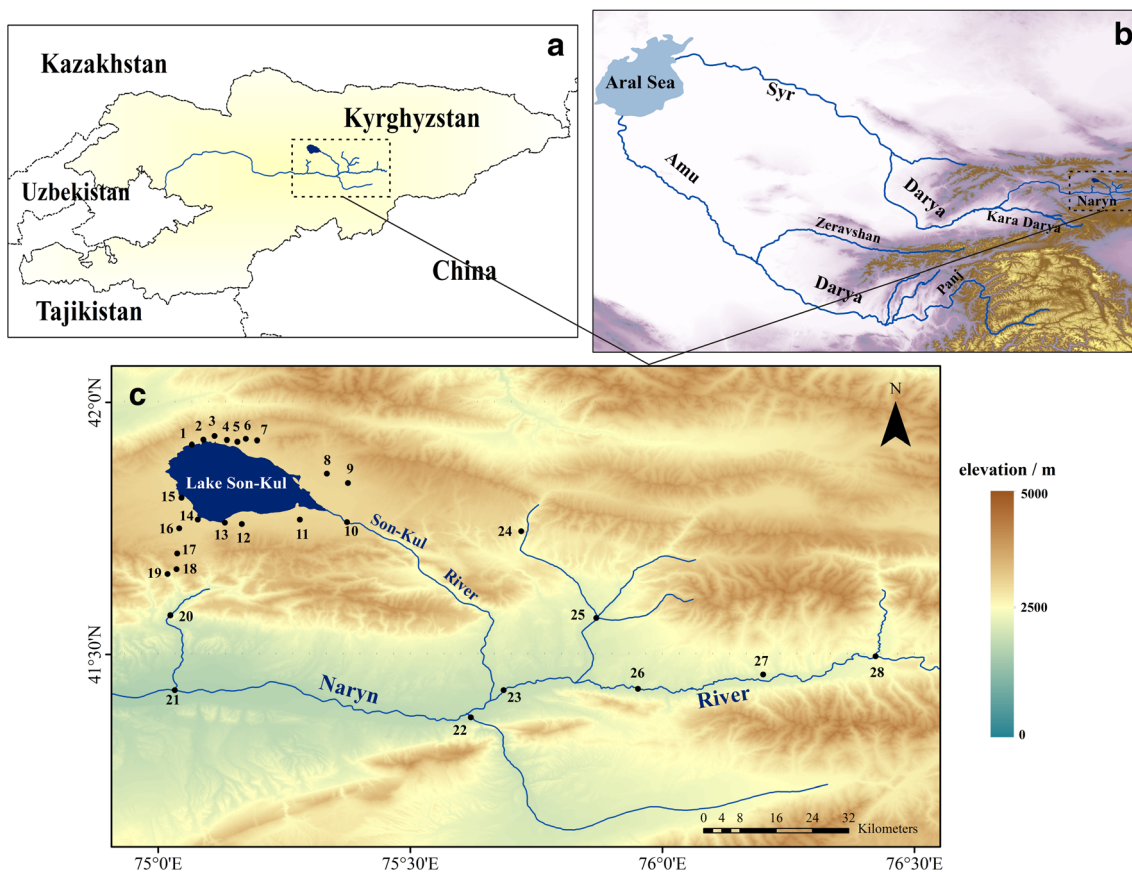
**Fig. 1** Map of the study area with locations of sampling sites. **a** The location of the study area. **b** The geomorphic map of the Aral Sea watershed. **c** The geomorphic map of sampling sites from the SKB (sites 1–19) and NRV (sites 20–28)

E~76.4229° E) in August 2013 (Fig. 1c), of which 19 were from the Son-Kul Basin (SKB) and 9 were from the Naryn River Valley (NRV). Soil samples were taken at intervals of 2 m within a plot, and five or more samples were collected from each plot and mixed together to represent a single, composite sample. A clean, stainless steel, 30-cm-diameter cylinder was used for sampling, and a Garmin 12 XL global positioning system (GPS) was employed to record the location of each sampling plot. Samples were wrapped in aluminum foil and sealed in clean polyethylene bags. As soon as possible, samples were stored at − 20 °C prior to analysis.

All soil samples were freeze-dried and ground in an agate mortar to obtain 100-mesh (149 μm) powders for extraction. Five grams of homogenized soil sample was mixed with 3 g of anhydrous sodium sulfate, baked at 550 °C for 6 h, and extracted using dichloromethane using an accelerated solvent extraction (ASE) system (Dionex ASE-350, Sunnyvale, California) at a temperature of 100 °C and pressure of 1500 psi. Freshly prepared copper granules were added to each sample to avoid potential interference of sulfur. The extract was further purified with a silica gel-alumina (2:1) column, and the target n-hexane/dichloromethane elution was then concentrated and treated according to Zhao et al. (2016).

## 2.3 Chemicals and reagents

A mixture of standard solution including α, β, γ, δ-HCH, heptachlor (HEPT), heptachlor epoxide (HEPX), α-chlordane (α-chlor), γ-chlordane (γ-chlor), aldrin, endrin, α-endosulfan (α-endo), γ-endosulfan (γ-endo), endrin aldehyde, endosulfan sulfate, endrin ketone, p,p'-DDE, p,p'-DDD, p,p'-DDT, and methoxychlor was purchased from Supelco (Bellefonte, PA, USA). Stock standard solutions were dissolved in n-hexane and stored in a dark glass container at 4 °C before use. All solvents (dichloromethane, n-hexane, and methyl alcohol) used for sample processing and analysis were chromatographic grade (Sinopharm Chemical Reagent Co. Ltd., Shanghai, China). The silica gel (100–200 mesh) and alumina (120–200 mesh) were extracted for 48 h in a Soxhlet apparatus, activated for 12 h at 180 and 250 °C, respectively, and then deactivated with deionized water at a ratio of 3% (m/m). Deionized water (Millipore Milli-Q system, MA, USA) was used throughout the analysis.

## 2.4 Instrument analysis

Purified sample extracts for OCP analysis were analyzed quantitatively using a gas chromatograph (Agilent 7890A,

Palo Alto, CA, USA) equipped with a [63]Ni μ-electron capture detector (GC-μECD) and a HP-5MS fused silica capillary column (30 m × 0.25 mm × 0.25 μm, J&W Scientific, Inc., USA). Detailed instrumental conditions were described in Zhao et al. (2016).

## 2.5 Quality control and assurance

Identification of OCPs was based on the retention time of the standard components analyzed under the same instrumental conditions, and quantification was achieved using the external standard method. The calibration curves of individual compounds were established using the peak area by analyzing a series of increasing concentrations of standard solutions. Additionally, a standard solution of intermediate concentration was analyzed after every eight samples to recalibrate the retention time of the target compounds. Precision of the measurements was determined through replicate analysis of certified reference materials (GSB 07-2973-2013) purchased from the Institution for Environmental Reference Materials Ministry of Environmental Protection. All data were subject to strict quality assurance and quality control procedures. During the entire test, method detection limits (MDLs) for individual compounds were determined as the concentration of analytes in a sample that yielded a peak signal-to-noise (S/N) ratio of 3 in the range 0.01–0.08 ng/g dw. For each set of samples, a procedural blank consisting of all reagents was run to check for cross contamination. Results indicated that no interferents were detected during the entire study. Furthermore, spiked recovery of the sample matrix ranged from 76 to 103% for individual compounds. Relative standard deviations (RSDs) of the instrument, determined using seven different standard solutions, were all < 10%; each standard solution was injected six times under the same instrumental conditions.

## 2.6 Statistical analysis

OCP concentrations were expressed on a dry weight basis (ng/g dry weight, ng/g dw). For samples with the concentrations below the MDLs, or that could not be detected, zero was assigned for statistical purposes. The statistical analysis was performed using SPSS Microsoft version 22.0 and R studio 3.5.1 for windows. A $p$ value < 0.05 (two-tailed test) was used as a cut-off for statistical significance.

# 3 Results and discussion

## 3.1 OCP concentrations and relative abundances

Concentrations of OCPs and relative abundances of individual compounds in soils from 19 sites in the SKB (sites 1–19) and

9 sites in the NRV are shown in Fig. 2. Total OCP concentrations in the SKB varied ~ 60-fold, from 2.36 to 145.32 ng/g dw, with the highest concentration found at site 7 (Fig. 2a). As mentioned, we concluded that site 7 was where a pesticide warehouse experienced an accidental release of OCPs in the SKB 40 years ago. Other than this site, total residual OCP concentrations in the remaining sites ranged from 2.36 to 24.13 ng/g dw (7.23 ± 5.21 ng/g dw), slightly higher than values observed in the NRV (range 1.69–7.45 ng/g dw, mean 4.11 ± 1.67 ng/g dw). To elucidate the distributions of OCPs and possible spatial associations in the study area, principal component analysis (PCA) was performed on concentrations of OCPs across different sampling sites (Table 1). In this study, four factors (eigenvalue > 1.0) that account for 96.8% of the total variance were extracted and rotated by varimax rotation. The first (PC1), second (PC2), third (PC3), and forth components (PC4) represent 32.7, 30.0, 20.8, and 13.3%, respectively. The principal components were classified as "strong," "moderate," and "weak," corresponding to absolute loading values of > 0.70, 0.70–0.50, and 0.50–0.30, respectively (Juahir et al. 2011; Wu et al. 2017). Therefore, PC1 with strong positive loadings on sampling sites 11–19 and with moderate positive loadings on sites 3–4 and sites 9–10 were generally located in the southern part of the SKB. Sites 24–28 were also moderately loaded on PC1. They were located in the upper reaches of the NRV, in the southeast of the SKB. Because they are downstream in a western wind direction, OCPs accumulated in sites 24–28 may be influenced by OCPs accumulated in the SKB. In addition, PC3 was dominated by sites 20–28, all of which are located in the NRV, suggesting that OCPs in sites 24–28 were also influenced by the OCPs in the lower reaches of the NRV (sites 20–23), but no spatial associations were found for OCPs between the low reaches of NRV and the SKB. PC2 had strong positive loadings on sites 1–2 and moderate positive loadings on sites 5 and 8–9, whereas PC4 was dominated by sites 5–7, especially high loading on site 7, demonstrating that the high concentrations of OCPs in site 7 exerted a strong effect on ambient sites 5–6 and a weak effect on sites 1–4, but no obvious effect on other sites in the SKB.

For individual OCP compounds, HCHs (including α, β, γ, δ-HCH) were detected at the highest concentrations in soils from both the SKB and NRV (2.20 ± 1.31 ng/g dw for SKB and 1.88 ± 1.38 ng/g dw for NRV), followed by methoxychlor (2.03 ± 2.16 ng/g dw for SKB and 0.95 ± 0.43 for NRV) (Fig. 2b). DDTs (including p,p'-DDD, p,p'-DDE, and p,p'-DDT) and Aldrins (aldrin, endrin, endrin aldehyde, and endrin ketone included) were the main pesticides, with average detection levels of 1.37 ± 1.11 ng/g dw (DDTs) and 1.24 ± 1.34 ng/g dw (Aldrins) for SKB and 0.50 ± 0.16 ng/g dw (DDTs) and 0.72 ± 0.44 ng/g dw (Aldrins) for NRV. In contrast, Endos (including α-,β-endosulfan and endosulfan sulfate) and Chlors (including α-,γ-chlordane, heptachlor, and heptachlor
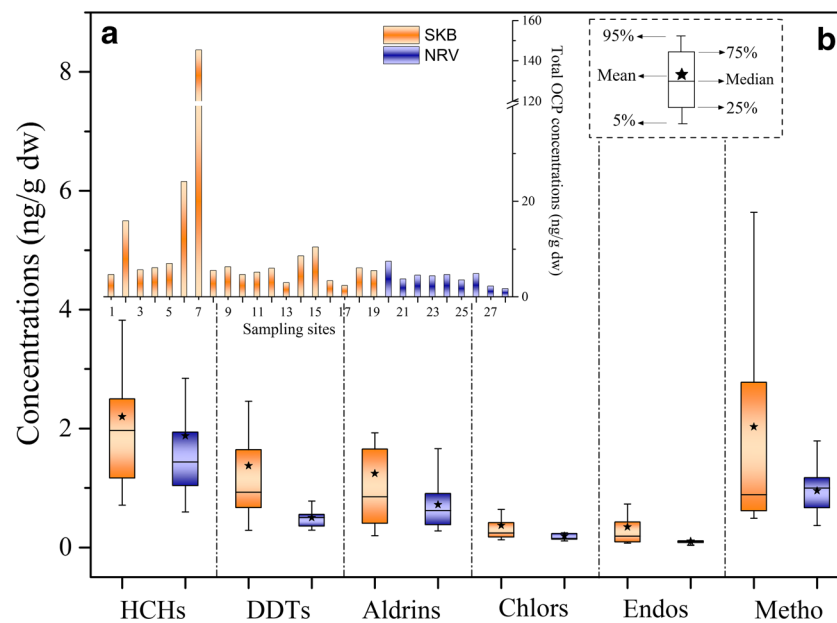
**Fig. 2** Concentrations of OCPs in soils from the SKB and NRV. **a** The total OCP concentrations at different sampling sites in the SKB and NRV. **b** Box plot of the individual OCP concentrations in soils from the SKB and NRV, except at site 7 (HCHs: the sum of α-, β-, γ-, δ-HCH; DDTs: the sum of p,p′-DDE, p,p′-DDD, and p,p′-DDT; Aldrins: the sum of aldrin, endrin, endrin aldehyde, and endrin ketone; Chlors: the sum of heptachlor, heptachlor epoxide, α-chlordane, γ-chlordane; Endos: the sum of endosulfan I, endosulfan II, and endosulfan sulfate; Metho: methoxychlor; 25%: 25th percentile; 75%: 75th percentile; the range of whiskers: 5–95%)

epoxide) were observed at low concentrations (Fig. 2b). Using the MANOVA test, it was found that there were significant differences of heptachlor (sig ≈ 0.000), α-HCH (sig = 0.007), α-endosulfan (sig = 0.023), and p,p′-DDT (sig = 0.037) concentrations between SKB and NRV but other compounds had no significant difference. This suggests that the sources of heptachlor, α-HCH, α-endosulfan, and p,p′-DDT in the SKB may be different from those in the NRV, which provide a clue to OCP source apportionment and be further analyzed below. HCHs, methoxychlor, DDTs, and Aldrins were used primarily as insecticides for a wide range of pests including biting flies, houseflies, mosquito larvae, and termites, around stored grains, livestock, wood, and wooden structures. Chlors and Endos are broad-spectrum insecticides that have been used mainly on agricultural crops, including vegetables, small grains, maize, fruits, and cotton (EPA). Therefore, the fact that residual levels of HCHs, methoxychlor, DDTs, and Aldrins are higher than those for Chlors and Endos in soils of the study area indicates that animal husbandry has been the dominant agricultural activity, whereas farming has remained relatively undeveloped. This finding is consistent with the agricultural structure of the SKB (Borchardt et al. 2011).

## 3.2 Regional comparison of OCP concentrations

Because HCHs and DDTs are ubiquitous in the environment and have been reported commonly by researchers, levels of HCHs and DDTs in the study area (except site 7) were compared to values from other remote mountain and polar regions

around the world. For mountain regions in China, concentrations of HCHs and DDTs in soils from the SKB were similar to those of the Ruoergai Highlands (~3552 m a.s.l) (Gai et al. 2014), where levels of HCHs and DDTs in soils were 0.43–6.72 ng/g dw (1.86 ± 1.49 ng/g dw) and 0.29–4.34 ng/g dw (1.63 ± 1.20 ng/g dw), respectively. Concentrations in the study area were lower than those in the Changbai Mountain area (253–1547 m a.s.l; HCH ranges 3.10–25.6 ng/g dw, mean 13.87 ± 11.28 ng/g dw; DDT ranges 4.15–9.06 ng/g dw, mean 6.48 ± 2.46 ng/g dw) (Wang et al. 2017), but were slightly higher than those in cultivated soils on the Tibetan Plateau (3000–5000 m a.s.l; mean 0.39 ng/g dw and 1.05 ng/g dw for HCHs and DDTs, respectively) (Chen et al. 2017). HCHs in the Kaidu River Basin from the eastern Tianshan Mountains of China were found at higher concentrations (range 3.7–4.5 ng/g dw, mean 3.9 ng/g dw) than the HCHs in the SKB, whereas DDTs were observed at much lower levels (ranges 0.14–1.1 ng/g dw, mean 0.2 ng/g dw) (Shen et al. 2016b). Elevated concentrations of DDTs were detected in soils from the Indian Himalayan Region (range 0.28–1448 ng/g dw, mean 148 ± 355 ng/g dw) (Devi et al. 2015), mountain regions in Azerbaijan (range 1.43–1115 ng/g dw, mean 236 ± 364 ng/g dw) (Aliyeva et al. 2012), and mountain regions in the Czech Republic (275–1050 m a.s.l; range 8.8–1908 ng/g dw, mean 315 ng/g dw) (Holoubek et al. 2009), indicating that those countries were contaminated by substantial application of DDTs and/or recent use of DDTs there. HCHs were found at similar or slightly higher levels elsewhere compared with the SKB (range nd-2.79 ng/g dw, mean

**Table 1** Results of principal component analysis (PCA) performed using concentrations of OCPs in different sampling sites. Eigenvalues, % contribution to the total variation, and eigenvectors for each of four principal components (PCs)

| | PC1 | PC2 | PC3 | PC4 |
|---|---|---|---|---|
| Eigenvalues | 16.9 | 3.41 | 1.78 | 1.05 |
| % variation | 32.7% | 30.0% | 20.8% | 13.3% |
| Sampling sites | | | | |
| 1 | 0.367 | *0.762* | 0.127 | 0.321 |
| 2 | 0.226 | *0.777* | 0.049 | 0.430 |
| 3 | *0.622*[a] | 0.082 | 0.488 | 0.379 |
| 4 | *0.683* | 0.380 | 0.143 | 0.415 |
| 5 | 0.144 | 0.156 | 0.179 | *0.724* |
| 6 | 0.271 | *0.506* | 0.100 | *0.849* |
| 7 | 0.071 | 0.326 | 0.218 | *0.947* |
| 8 | 0.229 | *0.554* | 0.124 | 0.112 |
| 9 | *0.537* | *0.564* | 0.038 | 0.188 |
| 10 | *0.690* | 0.199 | 0.078 | 0.113 |
| 11 | *0.741* | 0.026 | 0.392 | 0.296 |
| 12 | *0.965* | 0.051 | 0.088 | 0.143 |
| 13 | *0.815* | 0.346 | 0.180 | 0.375 |
| 14 | *0.713* | 0.284 | 0.103 | 0.320 |
| 15 | *0.910* | 0.314 | 0.081 | 0.173 |
| 16 | *0.865* | 0.329 | 0.033 | 0.043 |
| 17 | *0.754* | 0.380 | 0.069 | 0.230 |
| 18 | *0.780* | 0.379 | 0.294 | 0.042 |
| 19 | *0.848* | 0.378 | 0.220 | 0.066 |
| 20 | 0.409 | 0.208 | *0.886* | 0.017 |
| 21 | 0.342 | 0.112 | *0.554* | 0.335 |
| 22 | 0.365 | 0.360 | *0.503* | − 0.022 |
| 23 | 0.305 | 0.033 | *0.935* | −0.017 |
| 24 | *0.553* | 0.390 | *0.797* | − 0.132 |
| 25 | *0.638* | 0.199 | *0.643* | 0.054 |
| 26 | *0.595* | 0.056 | *0.719* | 0.101 |
| 27 | *0.570* | 0.368 | *0.862* | − 0.008 |
| 28 | *0.599* | 0.243 | *0.744* | 0.055 |

[a] Italic type: strong and moderate positive loadings on PCs

1.12 ± 0.71 ng/g dw for the Indian Himalayan Region; range 0.26–1.66 ng/g dw for the Czech Republic; and range 0.90–24.46, mean 10.78 ± 6.38 ng/g dw for Azerbaijan). Concentrations of HCHs and DDTs in European and North American mountain regions, such as those of Mt. Legnone, Italy (~ 245–2600 m a.s.l; HCH range 0.01–1.88 ng/g dw, mean 0.51 ± 0.26 ng/g dw; DDT range 0.18–11.0 ng/g dw, mean 2.2 ± 3.1 ng/g dw) (Tremolada et al. 2008), the Pyrenees (~ 1700–2057 m a.s.l.; HCH range 0.08–0.19 ng/g dw; DDT range 1.70–3.40 ng/g dw) (Grimalt et al. 2004), and Pico de Teide, Spain (~ 10–3400 m a.s.l.; HCH range nd-1.00 ng/g dw; DDT range 0.01–40.0 ng/g dw) (Ribes et al. 2002) displayed values of a similar order of magnitude as the

SKB. Overall, concentrations of HCHs and DDTs in soils from the SKB were comparable to or lower than those in soils from most other mountain regions. Furthermore, compared with polar regions, levels of HCHs and DDTs in the SKB were lower than those in Svalbard, the Norwegian Arctic (HCH range 0.8–6.5 ng/g dw, mean 4.1 ng/g dw; DDT range 0.2–5.4 ng/g dw, mean 2.2 ng/g dw) (Jiao et al. 2009), but higher than those on James Ross Island, Antarctica (HCH range 0.49–1.34 ng/g dw; DDT range 0.51–3.68 ng/g dw) (Klánová et al. 2008) and Prydz Bay and Peninsula, Antarctica (HCH range 0.28–1.23 ng/g dw, mean 0.57 ng/g dw; DDT range 0.45–6.5 ng/g dw, mean 1.7 ng/g dw) (Xue 2014), where residual levels of two such compounds were comparable to those in the NRV. This suggests that concentrations of OCPs in the NRV soils can be characterized as background values and that soils of the region have not been subject to substantial contamination from human activities.

### 3.3 Sources and transport pathway of OCPs

Remote mountain regions have many similarities to polar regions, with respect to climate conditions (low temperature and ice cover), remoteness, and the ecological structure of the communities. There are, however, great differences, such as the relative proximity to contaminant emission sources, the greater importance of local meteorological conditions, and the short distances over which climate and ecological gradients change. Sampling sites in the SKB were located at an average altitude of 3028 m, whereas the Naryn River flows mainly west through the valley (average altitude 2059 m), making the relative height difference between these two areas approximately 1000 m and resulting in a temperature difference of 6 °C. Temperature plays a role in shaping the biotic community and intensity of microbial activity, both of which affect the degradation of OCP compounds (Manz et al. 2001; Oliveira et al. 2016).

Long-range transport of OCPs is also driven by changes in the subcooled liquid vapor pressure of each compound with temperature. Semi-volatile OCP compounds have subcooled liquid vapor pressures of 0.1 to 0.001 Pa at 25 °C (Hinckley et al. 1990). Although these vapor pressures are sufficient for all such compounds to be transported to some degree, the most volatile compounds are more readily distilled. Thus, more volatile OCP compounds, such as HCHs, would be expected to occur at relatively higher concentrations at higher altitudes. Less volatile compounds such as Chlors and Endos should not be transported to high altitudes as effectively, and their environmental concentrations should thus reflect local use (Simonich and Hites 1995). Therefore, OCP concentrations from both long-range transport and local application should be considered when attempting to determine the source and transport mechanisms of OCPs in mountain regions. Several factors can help discern sources and transport processes,

including specific distribution patterns, in combination with wind direction and local temperature. Additionally, differences in concentration of individual compounds, along with their isomer ratios, can help identify the source and age of contaminant deposition.

### 3.3.1 Chlors

HEPT is a chlorinated cyclodiene insecticide. Not only is HEPT included in technical chlordane, but it can also be used independently to control soil insects. HEPX is a breakdown product of HEPT that can remain in the soil for a long time and is produced by photolysis or epoxidation in plants and animals (Nasir et al. 2014). A higher ratio of HEPT/HEPX (if ratio > 1) indicates the fresh application of HEPT, whereas a low ratio indicates there has been sufficient time for breakdown of HEPT. In this study, the HEPT/HEPX ratio was zero at all sites except site 7 because HEPT could only be detected at site 7 (3.85 ng/g dw). There, the HEPT/HEPX ratio was 1.12, indicating slow degradation and/or fresh input of HEPT. Presence of HEPT at site 7 indicated that the site was unique; probably it was the location of the pesticide warehouse in the SKB. Furthermore, HEPT has a shorter half-life and is less volatile than other OCPs (Baek et al. 2011), and its more stable degradation product (HEPX) displayed a trend of increasing concentration approaching the pesticide warehouse and low levels in those far from the pesticide warehouse, thereby indicating the point source of HEPT in the study area.

Technical chlordane is a mixture of γ-Chlor (13%), α-Chlor (11%), HEPT (5%), and trans-nonachlor (Zhang et al. 2012), with a γ-/α-Chlor ratio of 1.2 (Chakraborty et al. 2010). γ-Chlor is more easily degraded than α-Chlor in the environment. Hence, the γ-/α-Chlor ratio can be utilized to determine whether technical chlordane in the environment is recent or "aged." Ratios of γ-/α-Chlor were all < 1.2 in soils from both the SKB ($0.35 \pm 0.49$) and the NRV ($0.12 \pm 0.17$), implying that technical chlordane is mainly derived from historical, rather than recent inputs, with greater degradation in the NRV (Fig. 3a). Differences in the degree of degradation between these two areas were likely related to differences in temperature (~ 6 °C) related to altitude, and different distances from the pollution source. Higher altitude in the SKB enhanced retention capacity of soils, contributing to elevated concentrations of technical chlordane. At lower altitude in the NRV, higher temperatures promoted microbial activity that resulted in faster pollutant degradation. In addition, relatively higher γ-/α-Chlor ratios were observed in and around the pesticide warehouse (sites 5–8). This probably indicates that the local point source influenced values in the neighboring environments, owing to the lower volatilization of technical chlordane and its accumulation near the original site of use.

### 3.3.2 HCHs

HCH has two formulations, i.e., technical grade and lindane. Technical-grade HCH contains α-HCH, β-HCH, γ-HCH, and δ-HCH isomers with proportions of 60–70, 5–12, 10–12, and 6–10%, respectively, while lindane contains > 99% γ-HCH. Therefore, the ratio of α-/γ-HCH is relatively stable, with values of 4–7 for technical HCH mixtures and values near zero for lindane (> 99% γ-HCH), which may increase during degradation in the environment (Syed et al. 2014). In addition, β-HCH has the lowest water solubility and vapor pressure, is the most stable, and is relatively resistant to microbial degradation in soil (Bai et al. 2015). Both α- and γ-HCH can be transformed into β-HCH in the environment. Thus, the higher the percentage of β-HCH, the longer the period over which HCHs had been used. In this study, the ratios of α-/γ-HCH ranged from 0.29 to 2.40 ($0.87 \pm 0.50$) in the SKB, and they ranged from 0.08 to 1.05 ($0.55 \pm 0.42$) in the NRV (Fig. 3b), both indicating the dominance of lindane contamination in the study area. The slightly higher ratios of α-/γ-HCH in the SKB, compared to the NRV, however, imply faster degradation and relatively smaller fresh inputs of lindane in the SKB, even though the SKB is characterized by colder temperatures and reflects local point source pollution from the pesticide warehouse there. The high contribution of β-HCH in the SKB ($26 \pm 19\%$ of total HCHs) indicates that there are no recent local HCH sources. Nevertheless, there may have been exogenous volatile HCHs from other regions via long-range atmospheric transport input into the study area, especially α-HCH and γ-HCH. Ambient air monitoring a in Central Asia, carried out by the Reach Center for Environmental Chemistry and Ecotoxicology (RECETOX) from 2004 to 2008, showed the continuous persistent air contamination with HCHs, not only in Kyrgyzstan, but in Kazakhstan and even more remote countries such as Romania, where the highest median HCH levels were measured (~ 2 µg/filter) (Klánová et al. 2009). Lindane was found to be the principal compound of HCHs in the global atmosphere (Shunthirasingham et al. 2010). Thus, under prevailing westerly winds and cold condensation, an exogenous source of HCHs from long-range transport could be deposited in the study area. Furthermore, α-HCH is shown to be enriched with greater latitude and altitude, more so than γ-HCH (Chernyak et al. 1995; Pereira et al. 2006). This is in agreement with our results, in that the average concentration of α-HCH in the SKB was three times higher than that in the NRV. Statistical analysis suggested a positive correlation between α-HCH concentration and altitude ($r = 0.63$, $p < 0.05$, $n = 28$). The γ-HCH concentrations between the two areas, however, were similar and displayed no significant correlation with altitude ($r = 0.23$, $p > 0.05$, $n = 28$), contributing to the higher ratio of α-/γ-HCH in the SKB. In general, local HCHs generally reflect historic usage, but there is also an exogenous, fresh input of HCHs, especially lindane, via
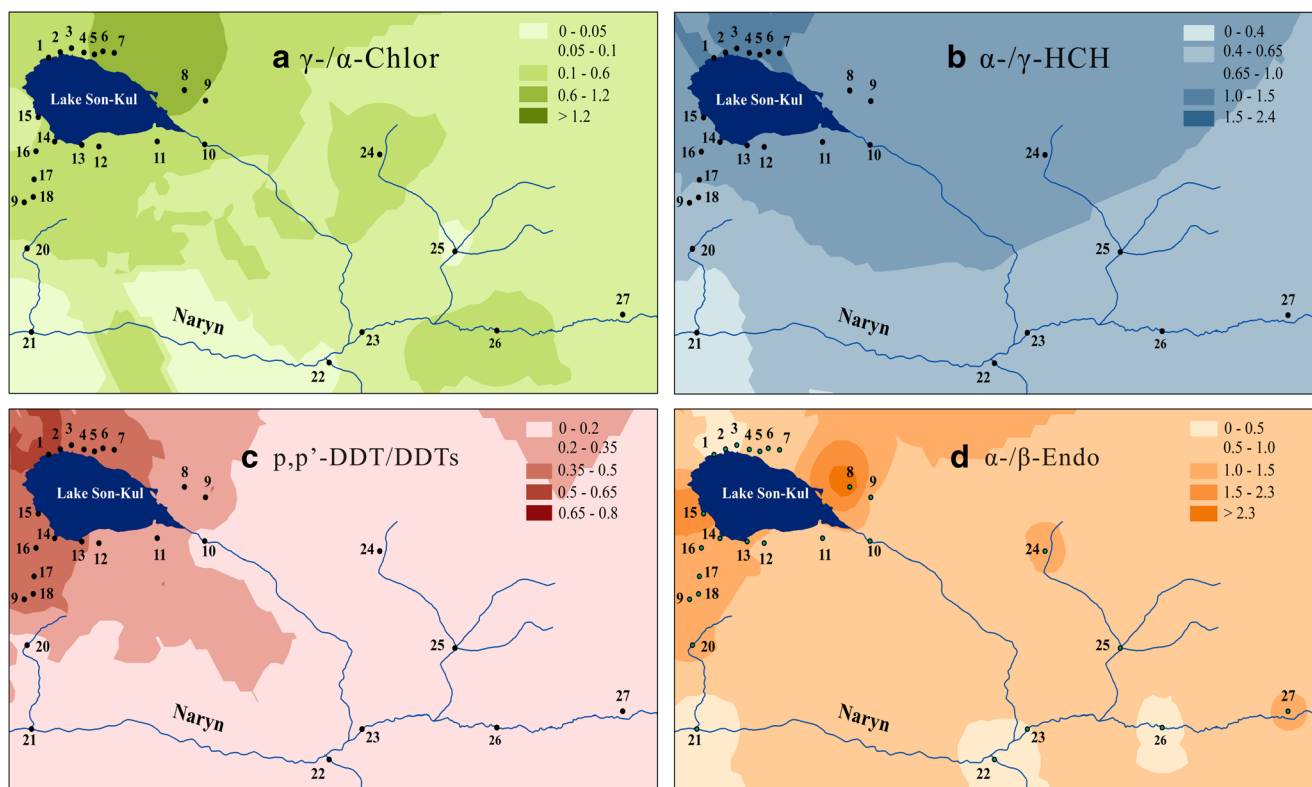
**Fig. 3** The Kriging interpolation plots of isomeric and parent substance/metabolite ratios of OCPs in soils around Lake Son-Kul and Naryn River. (**a**: γ-/α-chlor > 1.2, indicating that chlordanes originated mainly from historical use. **b**: α-/γ-HCH was in the range of 4.0–7.0 for technical

HCH mixtures and nearly zero for pure lindane. **c**: p,p'-DDT/DDTs > 0.5, indicating the fresh inputs of DDTs; p,p'-DDT/DDTs < 0.5, indicating no fresh inputs of DDTs. **d**: α-/β-Endo < 2.3 means no fresh endosulfan inputs)

long-distance atmosphere transport to the study area. Higher ratios of α-/γ-HCH in the SKB are a consequence of relatively higher α-HCH at higher, colder altitudes.

### 3.3.3 DDTs

p,p'-DDT is the most active isomer and typically accounts for approximately 80% of the technical formulation (Zhao et al. 2016). p,p'-DDT can degrade slowly to p,p'-DDE under aerobic conditions and to p,p'-DDD under anaerobic conditions (Pandit et al. 2002; Wang et al. 2012). Therefore, the relative abundances of the parent compound and the metabolites (p,p'-DDT/DDTs) can be used to indicate recent application (ratio > 0.5) or historical use (ratio < 0.5), and the ratio of p,p'-DDE/p,p'-DDD has been used to estimate the biodegradation conditions of the environment. The dominance of p,p'-DDE over p,p'-DDD in soils from the study areas may reveal the dechlorination of p,p'-DDT to p,p'-DDE under aerobic conditions. Ratios of p,p'-DDT/DDTs in the SKB ranged from 0.00 to 0.79 (0.42 ± 0.25) (Fig. 3c), implying relatively slow degradation of p,p'-DDT, and minimal new DDT input in this area. Ratios of p,p'-DDT/DDTs in the NRV, however, were near zero, suggesting faster degradation of p,p'-DDT to p,p'-DDE and little or no new DDT input, except at site 24, which is located on the mountaintop (p,p'-DDT/DDTs = 0.24).

Compared to HCHs, DDTs are less volatile, are not as effectively distilled, and hence do not undergo long-range atmospheric transport. DDTs tend to remain near their original site of use, and only short-range transport via air is possible. Highest p,p'-DDT/DDT ratios (> 0.5) occur at sites 1–2 (Fig. 3c), where grazing herds are abundant in summer, and higher p,p'-DDT/DDT ratios (0.35–0.5) occur around the SKB, where summer tourism is popular. This accounts for local emissions of DDT and secondary pollution from original DDT-contaminated soils, contributing to a high percentage of DDT in DDTs. In addition, thousands of tons of DDTs were imported into Kyrgyzstan in 2002 because of poor customs control. Some local independent farmers used these obsolete pesticides at levels exceeding the recommended application rates, resulting in the high DDT values in the environment (Shakirov and Bekkoesnov 2002). This phenomenon was widespread in western Kyrgyzstan (Toichuev et al. 2017). According to air monitoring data collected by RECETOX, median concentration of DDTs in the air over Kyrgyzstan from 2004 to 2008 were approximately 50 ng/filter. Some monitoring sites were > 100 ng/filter (Klánová et al. 2009). Prevailing western winds enhance the chance that mountains in Kyrgyzstan are affected and short-range transport of DDTs from the application sites in Kyrgyzstan might have led to an exogenous influence of atmospheric DDT burden on the high-

altitude study areas. Given that the environment in the NRV is basically pristine, i.e., has little anthropogenic interference, the predominance of p,p'-DDE in the NRV revealed that p,p'-DDE was the chief compound of exogenous DDTs deposited in the study area.

### 3.3.4 Endos

Endosulfan has two isomers, α-endo and β-endo, with a α-/β-endo ratio of approximately 2.3 in the technical mixture (Chakraborty et al. 2010; Zhao et al. 2016). Since α-endo degrades more rapidly in soils than γ-endo (WHO 1984), having a shorter soil half-life (60 days) than γ-endo (900 days) (Stewart and Cairns 1974), a lower ratio of α-/β-endo (< 2.3) represents past usage of technical endosulfan, whereas a ratio > 2.3 suggests recent usage (Qu et al. 2015). Figure 3d shows that the ratios of α-/β-endos were almost all < 2.3 (site 8 excluded) in soils from the SKB and the NRV, implying that technical endosulfan was mainly derived from historical use in the study areas. Unlike other isomeric ratios, α-/β-endo values near the pesticide warehouse were comparable to those in the NRV. The reason for this may be that animal husbandry is the dominant agriculture in the SKB, especially on the northwestern shore of Lake Son-Kul, where farming is largely absent. As technical endosulfan is used to protect agricultural crops, it is rarely applied by herdsmen, which accounts for the low residual levels and low α-/β-endo ratios. Site 8, however, had the highest α-/β-endo ratio (> 2.3), which may indicate that some farming activity occurred in the area. There is a permanent settlement nearby where people live to manage the nature reserve (Terghazaryan and Heinen 2006). They may have removed some natural vegetation to plant crops, and thus may have been responsible for the technical endosulfan inputs.

In summary, the only residues and fresh input of HEPT, observed at site 7, demonstrate that the pesticide warehouse was indeed at that location and had an influence on the local environment. Several volatile pesticides, including HEPX and technical chlordane, were primarily historically input from the pesticide warehouse, and likely remained near their original site of use. Less volatile technical endosulfan showed a similar pattern, that is, the nearer that sites were to the pollution source (site 8), the higher the residual concentrations and lower the degree of degradation. In contrast, more volatile compounds such as HCHs, through long-distance atmospheric transport, influenced by prevailing westerly winds, contributed to major fresh inputs into the study area. γ-HCH was dominant and α-HCH was more effectively deposited at higher altitudes compared to γ-HCH. p,p'-DDE is the main exogenous input into the study area, and is carried via short-range atmospheric transport. Additionally, local emissions of DDT and secondary pollution from original DDT-contaminated soils around the SKB also generated new endogenous DDTs in the SKB.

### 3.4 Assessment of OCP ecological risks

OCP concentrations in the SKB are relatively low, and many sites have values that are comparable to environmental background values in the NRV. According to a Screening Level Ecological Risk Assessment (USEPA 1997, 1998), most sites in the study areas had concentrations below their threshold effect levels (TELs), indicating that they exhibit low ecological risk (Fig. 4). A large quantity of the OCPs released from the pesticide warehouse did not have a long-lasting toxic effect on the environment, probably attributable to the natural capacity for recovery (e.g., rapid degradation) and protection measures. The pesticide warehouse was destroyed by a flood and most of toxic pesticides washed into the lake rapidly,



**Fig. 4** OCP concentrations in the SKB and NVR in comparison to guideline values. *TEL* threshold effect level for potential ecological effects, *PEL* probable effect levels for adverse ecological effects
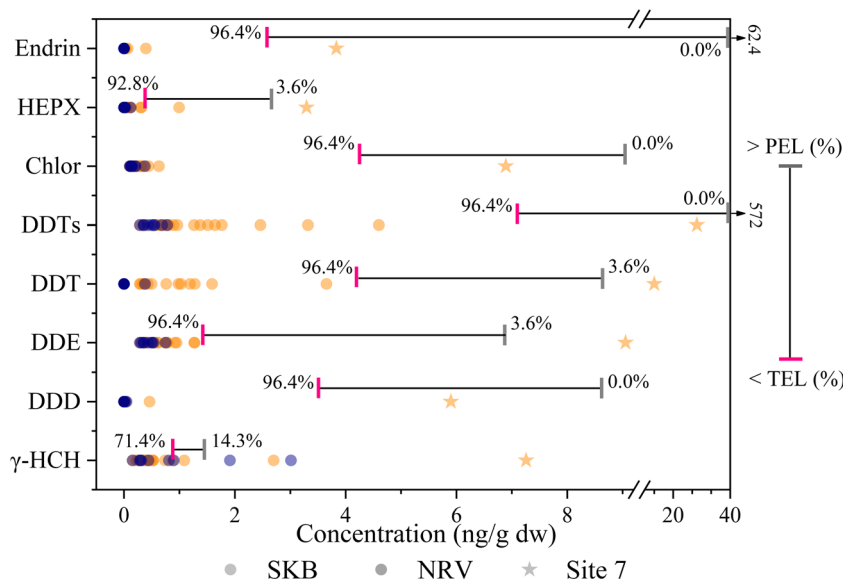
leaving little residual contaminant in the surface soil. Furthermore, after the environmental accident, the public and scientists launched campaigns to eliminate the use of OCPs (Pechenyuk 2006). Local authorities have taken steps to enact laws that ban some pesticides, control the import of others, and establish a nature reserve in the SKB to manage recovery of the environment in the SKB (Kustareva and Naseka 2015; Zhou et al. 2017). Nevertheless, the ecological risks of OCPs at the pesticide warehouse (site 7) should not be ignored. Concentrations of γ-HCH, p,p-DDE, and HEPX at site 7 were above probable effect levels (PELs) at which adverse ecological risks are frequently found (Fig. 4), suggesting the need for disposal of remaining pesticide stocks, as well as local remediation and clean-up efforts. Otherwise, the pesticide warehouse may continue to have some detrimental effects on the surrounding environment (sites 5–6). Moreover, seasonal herding and tourism bring OCP contamination into the SKB and this requires monitoring. In terms of individual compounds, γ-HCH was the main OCP species of ecological concern. Given the elevation-related temperature dependence of HCH degradation, which decreases at higher altitude and lower temperature, long-term studies are needed to investigate temporal trends of HCH concentrations and address the importance of remote alpine areas as sinks for more volatile organic pollutants. With the exception of the pesticide warehouse site (site 7) and seasonal human pressures, the environment of the SKB, with respect to pesticide contamination, is in generally good condition.

## 4 Conclusions

This study explored the residual levels, possible sources, and ecological risks of OCPs in mountain soils of the SKB and NRV in the western Tian Shan Mountains, Kyrgyzstan. High OCP concentrations were measured in the SKB, and represent residual pesticides released from an accident (flooding) at a pesticide warehouse 40 years ago. Aside from this unique site, OCP concentrations in the SKB were relatively low compared to those of most mountain regions, and only slightly higher than those in the NRV, which can be characterized as background values. HCHs, methoxychlor, DDTs, and Aldrins were the primary OCP compounds, indicating that animal husbandry is the dominant agricultural activity in the study area, whereas crop farming remains relatively undeveloped. Most of the pesticides originated from historical usage, and the degree of degradation of such chemicals in the SKB was lower than that in the NRV, which was ascribed to the proximity to the local pollution source and the relatively colder temperatures associated with higher altitude. Comparatively, HCHs and DDTs showed new inputs partly because of local anthropogenic emissions and partly because of input via exogenous atmospheric transport, controlled by the vapor pressure of

chemicals. This mechanism accounts for the new input of volatile compounds such as α-HCH, γ-HCH, and p,p'-DDE. Also, α-HCH residues displayed greater accumulation at higher altitudes in the SKB. An ecological risk assessment showed that, with the exception of the adverse ecological effect from high OCP concentrations near the site of the pesticide warehouse, which requires improved management and remediation, the ecological risk of OCPs in the broader study area is low.

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