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Distribution and occurrence of persistent organic pollutants in the waters of the Sevan Lake basin (Armenia): Evidence of agricultural runoff

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ABSTRACT

The ecological status of Sevan Lake, the largest freshwater reservoir in Armenia, is increasingly threatened by legacy agricultural pollution. This study assessed the transport of organochlorine pesticides from surrounding agricultural soils into Sevan Lake and its inflowing rivers via snowmelt and precipitation-driven runoff. Water sampling was conducted during November-December 2024 and January-March 2025. A total of 65 water samples were collected from seven nearshore zones of the lake and the mouths of six major inflowing rivers. Qualitative and quantitative analyses were performed using gas chromatography with an electron capture detector. This study presents the seasonally resolved, basin-scale quantitative assessment of organochlorine pesticides in Sevan Lake and its major tributaries. DDT, DDD, DDE, heptachlor, α-HCH, and lindane (γ-HCH) were detected in surface waters. Concentrations increased after the snowmelt period, particularly in March 2025, and frequently exceeded threshold values. Concentration ratios of DDT/DDD and γ-HCH/α-HCH indicate historical pesticide use and ongoing remobilization from contaminated soils. The results show that surrounding agricultural lands act as active secondary sources of organochlorine pesticide contamination, posing risks to aquatic ecosystems and fish populations.

Keywords: freshwater lake, surface water, organic pollutants, organochlorine pesticides.

INTRODUCTION

Lakes are complex aquatic ecosystems that support biodiversity, including aquatic organisms and migratory birds, while also providing important economic services such as fisheries, irrigation, and tourism. Although lakes represent only a small fraction of global freshwater resources, they play a crucial role in the biosphere and are increasingly important under conditions of global freshwater scarcity (Benateau et al., 2019). However, lake ecosystems worldwide are subject to growing anthropogenic pressures, including climate change, nutrient enrichment, and chemical pollution, which significantly affect their ecological status and water quality (Zalasiewicz et al., 2017; Shikhani et al., 2021; Zebardast et al., 2023). Lake Sevan is the largest freshwater lake on the Transcaucasian region and one of the largest high-altitude freshwater lakes globally, located at an elevation of approximately 1916 m above sea level. The lake has major economic and ecological importance for Armenia, serving as a source of drinking water, irrigation, hydropower generation, fisheries, and tourism. It supplies a substantial proportion of the country's fish and crayfish production and represents a natural and cultural heritage site with high biodiversity value. Consequently, maintaining the ecological integrity and water quality of Sevan Lake is of national importance. Until the early twentieth century, Sevan Lake maintained a relatively stable ecological condition. However, large-scale anthropogenic interventions during the Soviet period, including the artificial lowering of the lake level and intensified exploitation of water resources, triggered significant ecological degradation (Hovanesian and Bronozian, 1994; Hayrapetyan, 2025). By the beginning of the 2000s, the lake experienced substantial reductions in water level, surface area, and volume, accompanied by increased pollution from agricultural, industrial, and domestic sources (Varotsos, 2020). Population growth, tourism development, and insufficient wastewater management further exacerbated eutrophication and water quality deterioration (Varotsos, 2020; Khosrovyan, 2023). In addition, mining activities in the catchment area have contributed to the input of heavy metals and other contaminants into the lake via tributary rivers (Gevorgyan, 2023).

Agriculture represents one of the major sources of diffuse pollution in the Sevan Lake basin. During the Soviet period, agricultural lands surrounding the lake were intensively treated with agrochemicals, particularly organochlorine pesticides. According to available estimates, significant quantities of pesticides were applied annually within the basin from the mid-1960s onward (Gabrielyan, 2022). Although the use of many organochlorine pesticides has been prohibited for decades, their high persistence, hydrophobicity, and tendency to bioaccumulation allow them to remain in soils and sediments for long periods and to be continuously remobilized into aquatic environments. Organochlorine pesticides belong to the group of persistent organic pollutants (POPs), which undergo long-range environmental transport and cycling between air, water, and soil compartments. These compounds pose serious ecological and human health risks due to their toxicity and bioaccumulation potential (Aravind Kumar et al., 2022; Bashkin, 2024; Ruiz-Ciau et al., 2025; Matei et al., 2023; Vaccher et al., 2020; Rostami and Juhasz, 2011; Guo et al., 2019; Melake, 2024).

In the post-Soviet period, additional risks have arisen from the presence of obsolete and banned pesticide stockpiles stored in deteriorating facilities or informal disposal sites within the Sevan Lake basin (Milieukontakt. Annex 06. 2006). The absence of a centralized pesticide registration and monitoring system in Armenia further complicates the assessment of current pesticide use and environmental exposure pathways. Despite the recognized vulnerability of Sevan Lake to chemical pollution, systematic quantitative data on organochlorine pesticide contamination in the surface waters of the lake and its inflowing rivers remain limited. Available information is fragmented and largely derived from non-academic sources (Khachatryan et al., 2004). In particular, the contribution of contaminated agricultural soils to the transport of organochlorine pesticides into the aquatic system during snowmelt and precipitation events has not been sufficiently quantified.

The aim of this study is to assess the qualitative and quantitative occurrence of organochlorine pesticides in the surface waters of Sevan Lake and its major inflowing rivers. By analyzing seasonal variations and diagnostic concentration ratios, the study seeks to elucidate the pathways through which historical pesticide residues are remobilized from surrounding agricultural soils and transported into the lake.

MATERIAL AND METHODS

Lake Sevan is situated in the eastern part of Armenia at 40°19′ N latitude and 45°21′ E longitude, at a high altitude. It has an elongated shape and is divided into two unequal parts, known as Major Sevan and Minor Sevan (Figure 1) (Wilkinson, 2020).

The lake is fed by 28 rivers and two major springs and drains through the Hrazdan River, which subsequently flows into the Araks River. The primary sources of river inflow are snowmelt, rainwater, and groundwater. The 28 tributaries pass through urban, agricultural, and industrial areas, carrying industrial, agricultural, and domestic wastes into the lake, thereby inducing changes in the lake ecosystem (Gabrielyan, 2022; Wilkinson, 2020).

The water volume of the lake is approximately 37.9×10^{12} l, the surface area is 1,276.4 km², and the maximum and mean depths are 81 m and 29.8 m, respectively (Hovanesian and Bronozian, 1994; Gabrielyan, 2022; Wilkinson, 2020).

The study area comprised Major Sevan and Minor Sevan, together with the largest rivers discharging into Sevan Lake: Argichi (Ar; 51 km), Gavaraget (G; 50 km), Masrik (Ms; 45 km), Martuni (M; 28 km), Karchaghbyur (K; 26 km), and Dzknaget (Dz; 22 km) (Figure 1) (ArmMonitoring.am, Sevan basin environmental data). Water sampling sites in the lake were selected along the littoral zones heavily influenced by agricultural activities. From the rivers, major rivers primarily affected by agricultural practices were chosen.

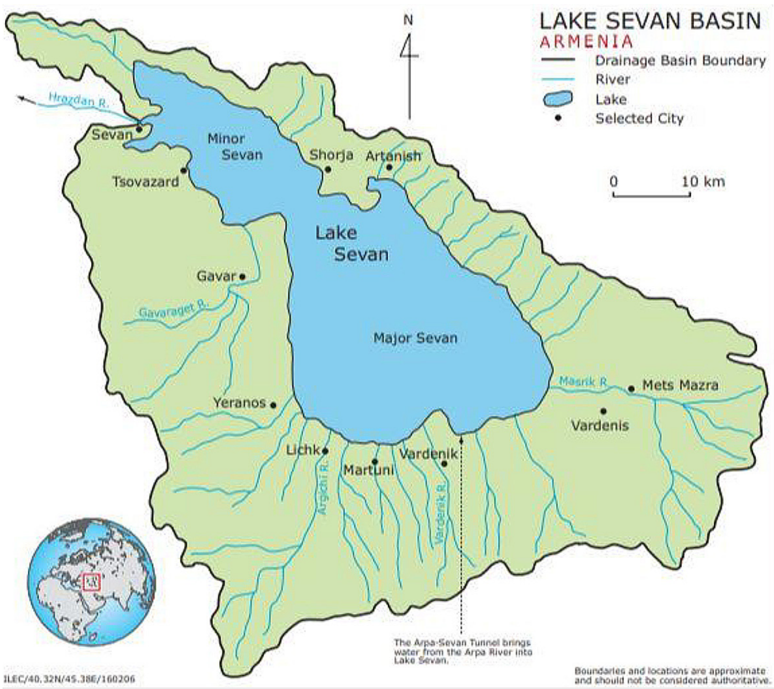

Figure 1. Map of Sevan Lake and its basin rivers

According to Table 1, in March 2025, the rivers discharging into the lake contributed 1.85 times more water to Sevan Lake than in February, even though precipitation volumes over the lake surface in March and February were nearly similar. The rivers entering the lake in March 2025 carried snowmelt and precipitation waters, which washed soils from the adjacent areas into the lake.

Sampling was conducted in November and December 2024 and in January, February, and March 2025. A total of 35 water samples were collected from the coastal areas adjacent to Major Sevan and Minor Sevan, and 30 water samples were collected from the mouths of six rivers flowing into Sevan Lake (Figure 1). Sampling, preservation, extraction, and concentration of organochlorine pesticides were carried out in accordance with national and international standard methods (Armenian Standard, 2005; Armenian Standard, 2016). The samples were stored in a cold room at approximately 4 °C until extraction and analysis. Qualitative and quantitative analyses of organochlorine pesticides were performed in the water samples. The following organochlorine pesticides were identified: DDT, DDD, DDE, heptachlor, α-BHC, and lindane (γ-BHC), and their concentrations were determined in the samples. Water quality assessment was conducted in accordance with Decision of the Government of the Republic of Armenia No. N75-N of January 27, 2011 on water quality standards (Government of the Republic of Armenia 2011, January 27).

For the extraction of organochlorine pesticides from water samples, 1 l of each water sample was transferred into a separatory funnel, and 100 ml of the organic solvent hexane was added. The funnel was sealed and shaken to allow the pesticides to partition from the aqueous phase into the organic solvent, after which it was left to stand to allow phase separation. The aqueous phase remained at the bottom, while the organic phase formed the upper layer. The organic layer was collected and dried over anhydrous sodium sulfate to remove residual moisture.

In the subsequent step, the pesticide extract was concentrated using a rotary evaporator to evaporate the solvent from the organic solution. After evaporation, hexane was added, and the final volume of the extract was adjusted to 2 ml.

The final concentrated extract was subjected to the identification and quantification of residual pesticides by gas chromatography. Analyses were performed using a Varian CP-3800 gas chromatograph equipped with a VF-5 ms capillary column (30 m × 0.25 mm i.d., 0.25 µm film thickness) and an electron capture detector (ECD), operated in split less injection mode. Helium was used as the carrier gas at a constant flow rate of 1 ml min^{-1}.

The gas chromatography oven temperature program was as follows: the initial temperature

Table 1. Water balance of Sevan Lake for January-March 2025 by input sources; data for November and December 2024 were not available on the website (ArmMonitoring.am, water quality data and reports)

Sampling months	January 2025	February 2025	March 2025
Precipitation on the lake surface (l)	2.00×10^9	28.5×10^9	30.0×10^9
Inflow from rivers (l)	33.99×10^9	31.0×10^9	57.39×10^9
Other sources (l)	9.82×10^9	9.44×10^9	9.74×10^9
Total (l)	45.81×10^9	68.94×10^9	97.13×10^9
Average monthly air temperature (˚C)	-3.5	-5.9	1.8

was set at 100 °C and held for 1 min, then increased to 280 °C at a rate of 10 °C min^{-1} and held for 5 min. Calibration was performed using a standard mixture of organochlorine pesticides based on AccuStandard ISO 6468-PEST standards (New Haven USA).

Quality assurance and quality control (QA/QC) procedures included calibration with certified reference standards, analysis of procedural blanks, and replicate sample analysis to ensure analytical accuracy and precision. The limits of detection (LOD) ranged from 0.01 to 0.2 ng/l, and the limits of quantification (LOQ) ranged from 0.03 to 0.6 ng/l for the analyzed compounds, as determined using GC-ECD analysis.

RESULTS AND DISCUSSION

The detected concentrations of organochlorine pesticides were compared with the maximum permissible concentrations (MPCs) for pesticides in surface waters, established at 10 ng/l (Avagyan, 2019). According to the World Health Organization (WHO), the guideline value for DDT (including its metabolites) in drinking water is 0.001 mg/l (1 μg/l), a limit designed primarily for the protection of human health rather than aquatic ecosystems (World Health Organization, 2011). Due to the high persistence, strong bioaccumulation potential, and pronounced ecotoxicity of DDT, international environmental frameworks recommend substantially lower threshold concentrations for surface waters. The European Union Environmental Quality Standards (EQS) define an annual average concentration of approximately 25 ng/l for DDT in surface waters, while the Canadian Council of Ministers of the Environment (CCME) proposes values close to 1 ng/l for the protection of aquatic life (European Commission 2013; Canadian Council of Ministers of the Environment, 1999). Similarly, the United States Environmental Protection Agency (US EPA) establishes a chronic exposure criterion of 0.23 ng/l for aquatic organisms, reflecting their high sensitivity to DDT contamination (United States Environmental Protection Agency, 2016).

In this context, the reference concentration of 10 ng/l adopted in the present study represents a conservative and ecologically justified threshold for Sevan Lake, balancing international precautionary approaches with regional regulatory applicability. This approach is consistent with Decision No. 75-N of January 27, 2011 of the Government of the Republic of Armenia, which permits the application of international guideline values for water quality assessment in the absence of specific national standards (Government of the Republic of Armenia, 2011).

All detected concentrations of organochlorine pesticides were above the method LOQs (0.03-0.6 ng/l), indicating that the reported values reliably reflect environmental contamination levels in Lake Sevan and its tributaries.

Figures 2 and 3 illustrate the occurrence of DDT, DDD, and DDE in the waters of Sevan Lake and its inflowing rivers during November and December 2024 and January, February, and March 2025.

Figures 4 and 5 present the concentrations of α-HCH, lindane (γ-HCH), and heptachlor in lake and river waters over the same period.

The results demonstrate that all six organochlorine pesticides were detected in both lake and river waters, with concentrations frequently exceeding the MPC during the snowmelt and precipitation period, particularly in March 2025. During the remaining months, pesticide residues were still detectable, although their occurrence was spatially heterogeneous and not observed at all sampling sites.

This pronounced seasonal pattern indicates that hydrological processes play a key role in controlling pesticide transport into the aquatic system.

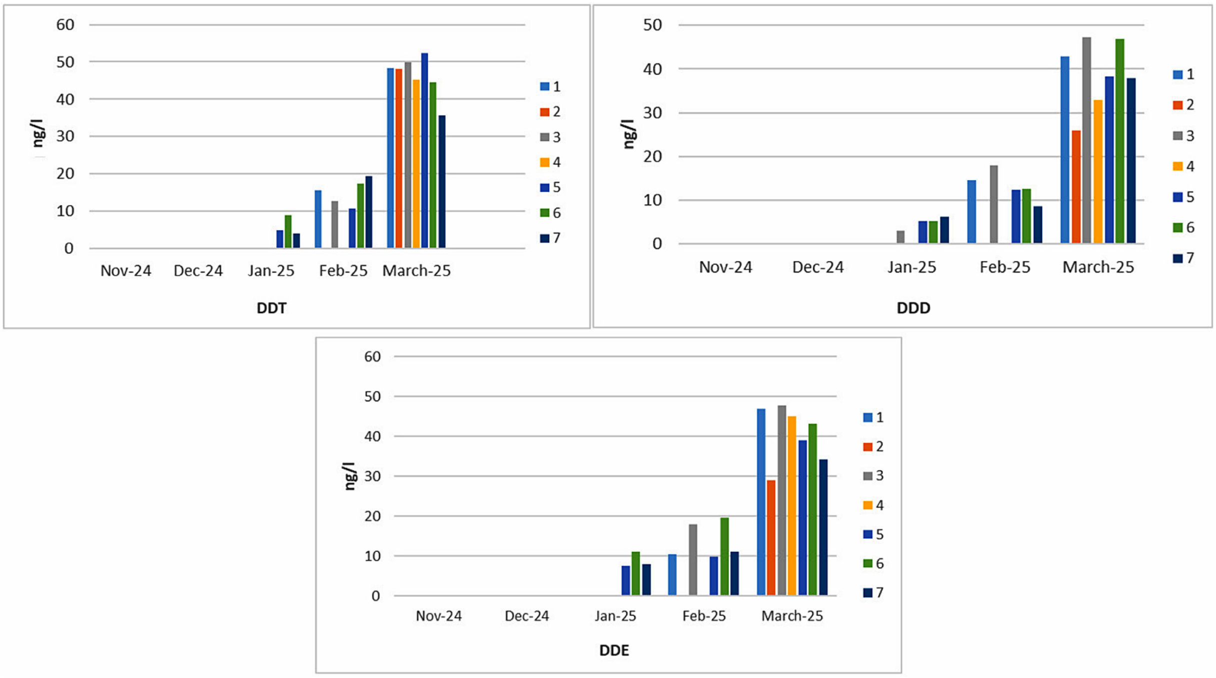

Figure 2. Organochlorine pesticides (DDT, DDD, DDE) in Sevan Lake. MPC for surface waters: 10 ng/l. **Note:** Sampling sites 1–7 are located within the lake

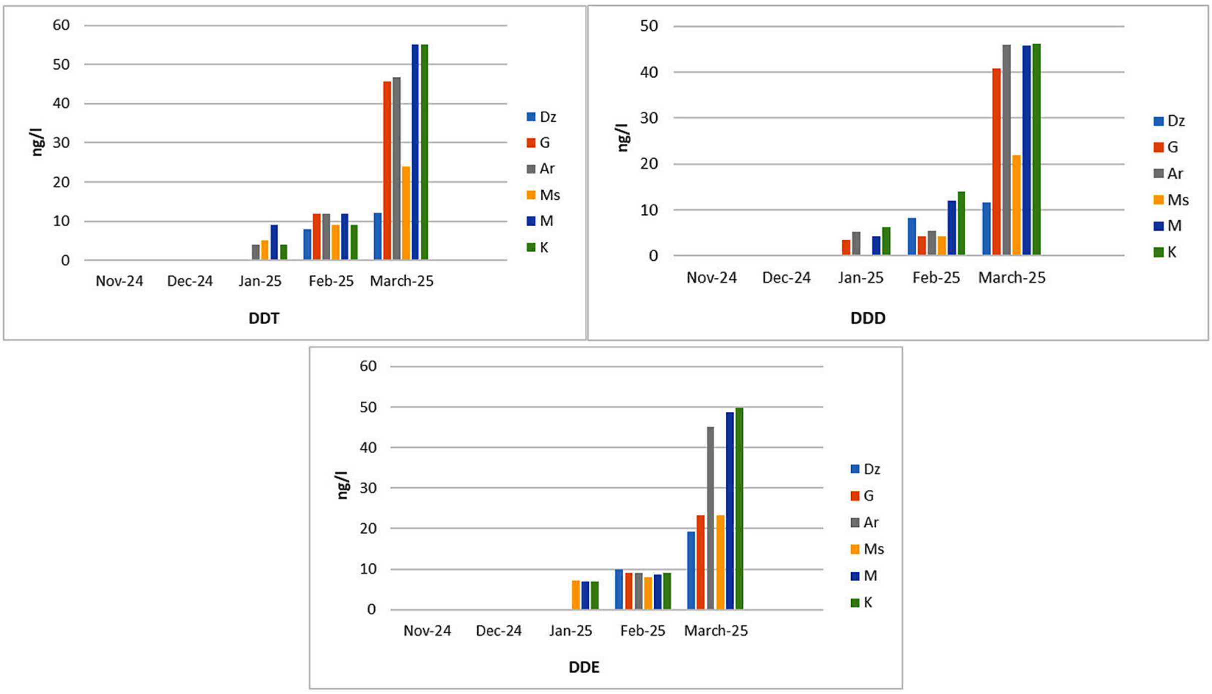

Figure 3. Organochlorine pesticides (DDT, DDD, DDE) in rivers flowing into Sevan Lake. MPC for surface waters: 10 ng/l. Note: Dz – Dzknaget, G – Gavaraget, Ar – Argichi, Ms – Masrik, M – Martuni, K – Karchaghbyur

With increasing air temperatures (Table 1), snowmelt and precipitation runoff likely mobilize pesticide residues from contaminated agricultural soils and from soil-covered burial sites containing obsolete and banned pesticide stockpiles. These residues are subsequently transported into tributary rivers and ultimately into Sevan Lake. In addition, atmospheric deposition, including the transport of pesticides bound to dust particles, may represent a secondary input pathway, particularly in spring when surface soils are exposed and wind activity increases (Kurek 2019).

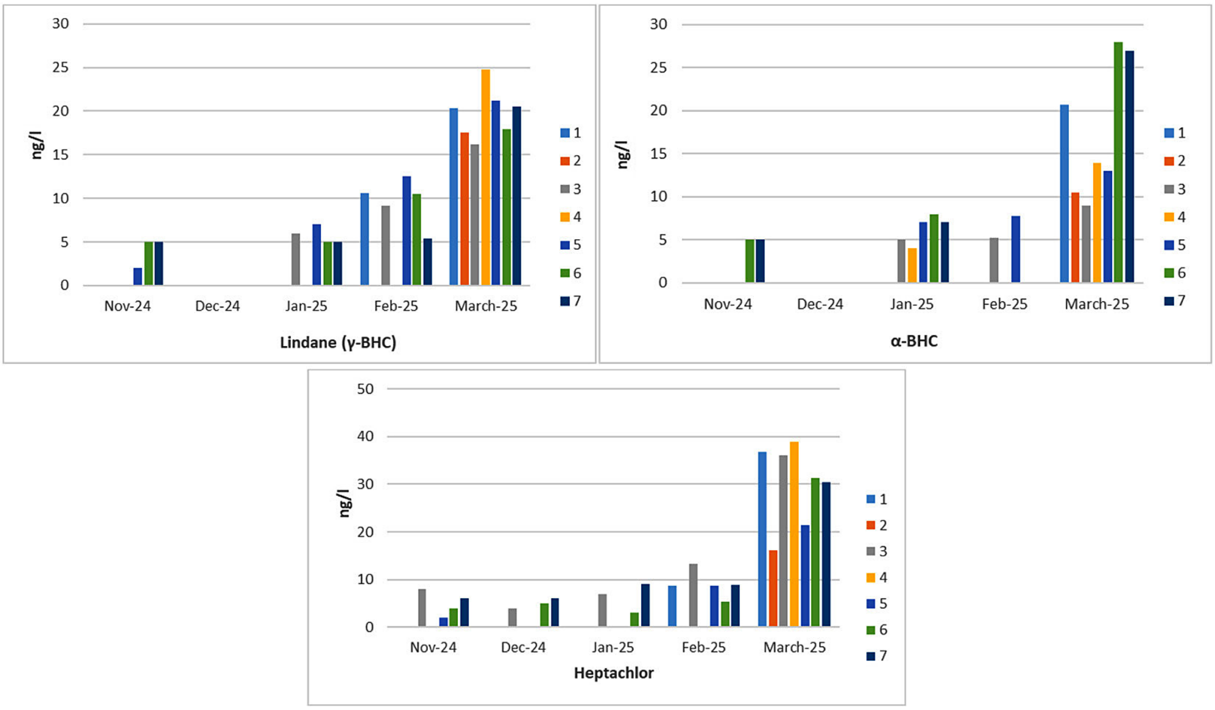

Figure 4. Organochlorine pesticides (Lindane (γ-BHC), α-BHC, Heptachlor) in Sevan Lake. MPC for surface waters: 10 ng/l.
Note: Sampling sites 1–7 are located within the lake

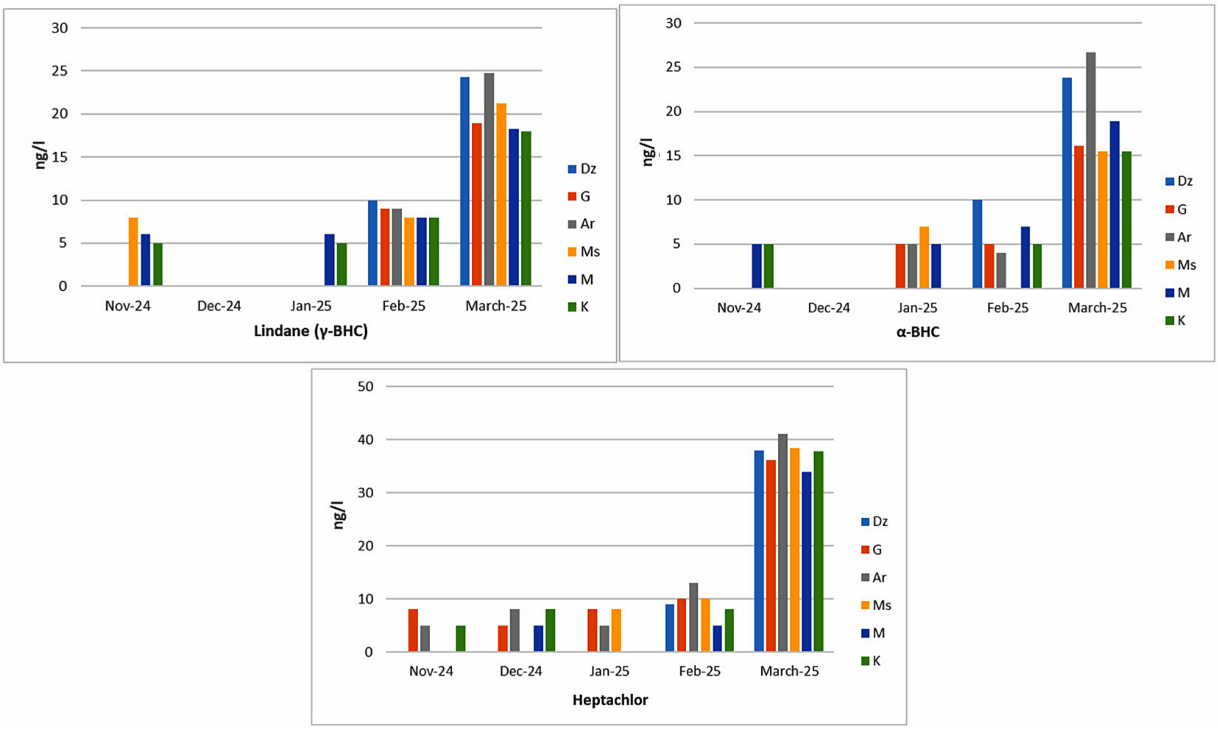

Figure 5. Organochlorine pesticides (Lindane (γ-BHC), α-BHC, Heptachlor) in rivers flowing into Sevan Lake. MPC for surface waters: 10 ng/l. Note: Dz – Dzknaget, G – Gavaraget, Ar – Argichi, Ms – Masrik, M – Martuni, K – Karchaghbyur

The ratios of DDT to DDD concentrations in rivers (Table 2) are close to unity. Such ratios are widely interpreted as indicators of historical rather than recent DDT inputs, reflecting the progressive degradation of parent DDT into its metabolites DDD and DDE in soils and sediments (Aravind

Table 2. Ratios of DDT to DDD concentrations in river waters as indicators of historical DDT use

March 2025 (snowmelt period)	Dz	G	Ar	Ms	M	K
DDT (ng/L)	12.15	45.60	46.84	24.03	55.33	55.17
DDD (ng/L)	11.54	40.84	46.06	21.98	45.67	46.16
DDT/DDD	1.05	1.12	1.02	1.09	1.21	1.20

Note: Dz - Dzknaget, G - Gavaraget, Ar - Argichi, Ms - Masrik, M - Martuni, K – Karchaghbyur.

Table 3. Ratios of Lindane (γ-BHC) to α-BHC concentrations in river waters as indicators of historical Lindane use

March 2025 (snowmelt period)	Dz	G	Ar	Ms	M	K
Lindane(γ--BHC) (ng/L)	24.23	18.97	24.76	21.22	18.22	17.96
A--BHC (ng/L)	23.82	16.1	29.71	18.87	18.84	15.46
Lindane(γ--BHC)/α--BHC	1.0	1.18	0.83	1.12	0.97	1.16

Note: Dz – Dzknaget, G – Gavaraget, Ar – Argichi, Ms – Masrik, M – Martuni, K – Karchaghbyur.

Kumar 2022). The observed values therefore suggest that current contamination is primarily driven by the remobilization of legacy pesticide residues rather than by ongoing application. Similarly, the concentration ratios of lindane (γ-HCH) to α-HCH (Table 3) fluctuate around 1.

These ratios indicate that the detected HCH isomers likely originate from aged pesticide formulations or contaminated soils rather than from fresh lindane use. Snowmelt and rainfall appear to facilitate the mobilization of both γ-HCH and α-HCH from soil-covered stockpiles of obsolete or banned pesticides into river waters.

Although γ-HCH (lindane) exhibits the highest insecticidal activity among HCH isomers, the other isomers (α-, β-, and δ-HCH) are also environmentally persistent and toxic, despite their lower biological efficacy. The simultaneous presence of multiple HCH isomers therefore reflects both the historical composition of applied formulations and the long-term environmental persistence characteristic of POPs.

The contamination patterns observed in Lake Sevan are consistent with those reported for other freshwater and coastal ecosystems worldwide, where legacy POPs continue to affect aquatic environments decades after their ban through soil erosion, runoff, and atmospheric transport (Aravind Kumar et al., 2022; Ruiz-Ciau et al., 2025; Melake et al., 2024; Kurek et al., 2019; Gong 2024; Li 2023; Wu 2025; McGill 2024). This similarity underscores the global relevance of the findings and highlights the role of contaminated catchment soils as long-term secondary sources of organochlorine pesticides to surface waters.

CONCLUSIONS

The obtained data indicate that organochlorine pesticides are widespread in Sevan Lake and its inflowing rivers, with concentrations exceeding the MPCs.

The lindane (γ-BHC)/α-BHC and DDT/DDD ratios are approximately equal to 1, suggesting that no recent inputs of lindane (γ-BHC) or DDT have occurred, and the existing residues have partially degraded into α-BHC and DDD, respectively.

The organochlorine pesticides detected in Sevan Lake waters, along with their concentrations, confirm that soils in the lake's vicinity constitute a significant source of organochlorine contamination in the lake's waters.

Organochlorine pesticide contamination of the waters of Sevan Lake constitutes a significant ecological burden, posing a threat to the lake's ecosystem and its fish fauna. Furthermore, because these waters are widely used for irrigation of agricultural lands, organochlorine residues may be transferred to surrounding soils, thereby expanding the spatial extent of environmental contamination.

Based on the findings of this study, the following conclusions were drawn:

- Applied environmentally safe and effective bioremediation approaches, during the restoration of contaminated soils to facilitate detoxification of persistent pollutants.
- Developed long-term monitoring programs for soils, water, and adjacent environmental components to assess organochlorine pesticide levels.

- Adopted, as a precautionary study-based recommendation, a threshold concentration of 10 ng/l for organochlorine pesticides in freshwater bodies in order to minimize ecological risks.

These results are essential for maintaining the ecological security and sustainable use of the lake's water resources for precise and effective remediation and risk-management strategies.

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