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ORGANOCHLORINE PESTICIDE LEVELS IN TASAUL LAKE

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ABSTRACT

The paper presents data regarding the level of organochlorine pesticides collected in 2005 - 2007 from different sites round Tasaul Lake and its tributaries in monitoring system, in a network of stations.

The level of organochlorine pesticides in Tasaul Lake is influenced not necessarily by hot spot sites like Tasaul's tributaries, but more probably by diffused pollution.

KEY-WORDS: organochlorine pesticides, water, sediments, Tasaul Lake

AIMS AND BACKGROUND

Significant sources of pollution with pesticides are atmospheric deposition, other water bodies, influences and terrestrial coasts of lakes. Pollutants come from rivers, domestic and industrial sources, from the air, by rain and snow, falling particles dissolved into the water, gas absorption into the water. The chemicals can be deposited to the ground or water bodies; when or if it is warm enough they can revolatilize back into the air. This cycle can be produced numerous times. This is why high levels of pollutants can be found in the areas which are far away from the main sources, cities and factory activities.

The evaluation of pesticides is very important because these persistent bioaccumulative toxic substances can be transferred into wildlife, causing reproductive problems and other harmful effects, including carcinogens; they also produce problems at people level, inducing reproduction, developmental, neurological and immunity disorders, accumulated by skin contact, inhalation and ingestion.

Monitoring for pesticides is generally poor in the world and can be mentioned especially in developing countries. Key pesticides are present in some countries monitoring of waters, but the cost of analysis is very expensive and sampling is at critical times of the year, needing to extend the data set. Many problems are related with inadequate facilities, impure reagents, and financial constraints. New techniques are necessary and may reduce costs and increase reliability.

The use of pesticides is extremely variable, in present also in large parts of Africa to Central America, with extremely heavy dosage in intensive agricultural areas of Brazil (world's third largest user of "agrototoxic" substances). France and the United States are on the first place. The concentrations found in the world, with few exceptions, are lower, in particular in developed countries which have a longer history of high pesticide consumption and intense use. In coastal waters, sediments and biota are less contaminated than inland waters, with few hot spots exception.

The Danish Government initiated an Action Plan in 1986, for sustainable agriculture to prevent the use of pesticides: research has been enlarged to all developed countries taking into consideration two main reasons (WWF, 1992): 1) Safeguard human health - from risks, adverse effects, preventing intake with pesticides by food and drinking water; 2) Protect the environment - wild flora and fauna, cultivated land and aquatic environments.

The International Code of Conduct on the Distribution and Use of Pesticides, formulated and being implemented by FAO (1985) is very relevant, in general, to pesticide pollution control and environmental protection. In Romania it is prepared a Manual for the improvement and development of the Integrated Monitoring for Romanian Waters (IMRW) which will be authorised by an official order about the quality of surface waters.

The catchment area of the Casimcea River, as main tributary, passes also through many villages and agricultural areas. Surrounded by agriculture fields, local communities and important industrial objectives, Tasaul Lake needed to be evaluated for the pesticides concentration, due to the impact of discharges from different diffusion sources. The network of the sampling stations is the same as of the other variables.

MATERIAL AND METHODS

The assay was done on water and sediment samples collected between May 2005 and March 2007 from 8 different stations along Tasaul Lake and 3 sediment depth cores (0-15 cm, 15-30 cm, > 30 cm) collected from sites 3 and 6. In 2007, there were collected 4 water samples from Tasaul's inflluents: Sibioara, Casimcea, Delafac, Piatra (Fig. 1).

The detection of organochlorine pesticides was done with a Hewlett - Packard gas chromatograph with electron capture detector.

The extraction of water samples was done in a separation funnel using a mixture of 7/3 hexane/dichloromethane. The extraction of sediment samples collected from the same locations was done in Soxhlet apparatus for eight hours with hexane.

After extraction, the samples were prepared as follows: they were concentrated to about 20 ml in a rotary evaporator; the sulphur compounds were removed with copper powder in an ultrasonic bath; the samples were fractionated on florisil columns and the final concentration was done in Kuderna-Denish concentrators and in nitrogen stream up to 1 ml.

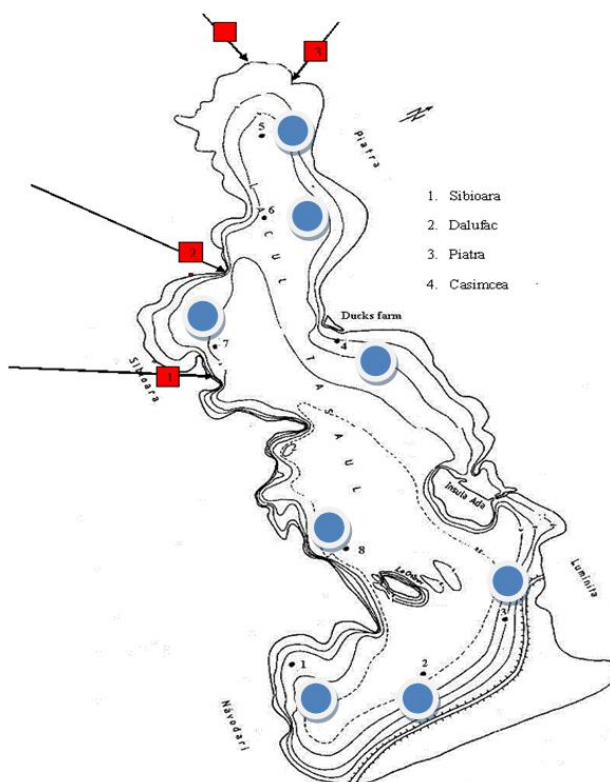


Fig. 1. The main influents of Tasaul Lake and their coordinates

RESULTS AND DISCUSSION

The results obtained for water samples from Tasaul Lake collected in May, August and November 2005 and March 2006 are listed in Table 1, Table 2, Table 3 and Table 4.

Table 1. Data obtained for organochlorine pesticides analyses in water samples collected from Tasaul Lake in May 2005

| ng/l | Tasaul 1 | Tasaul 2 | Tasaul 3 | Tasaul 4 | Tasaul 5 | Tasaul 6 | Tasaul 7 | Tasaul 8 |
|-------------------|----------|----------|----------|----------|----------|----------|----------|----------|
| HCB | 320.95 | 288.84 | < 4.26 | 419.35 | <4.26 | 94 | 297.4 | 400.62 |
| Lindane | < 3.78 | < 3.78 | < 3.78 | < 3.78 | < 3.78 | < 3.78 | < 3.78 | < 3.78 |
| Heptachlor | 156.38 | 129.184 | 121.63 | 182.16 | 37.22 | 24.75 | 61.52 | 178.65 |
| Aldrin | 101.48 | 129.28 | 177.73 | 273.8 | 178.91 | < 3.16 | < 3.16 | 186.79 |
| Dieldrin | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 |
| Endrin | 18.9 | 52.65 | 52.56 | 61.41 | 43.37 | 14.81 | 20.36 | 52.36 |
| p,p' DDE | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 |
| p,p' DDD | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 |
| p,p' DDT | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 |
| Total OCPs | 597.71 | 599.95 | 351.92 | 678.85 | 259.95 | 133.56 | 379.28 | 818.42 |

Table 2. Data obtained for organochlorine pesticides analyses in water samples collected from Tasaul Lake in August 2005

| ng/l | Tasaul 1 | Tasaul 2 | Tasaul 3 | Tasaul 4 | Tasaul 5 | Tasaul 6 | Tasaul 7 | Tasaul 8 |
|-------------------|----------|----------|----------|----------|----------|----------|----------|----------|
| HCB | 118,21 | 422,08 | 108,01 | 182,01 | 46,3 | 59,98 | 6,58 | <4,26 |
| Lindane | < 3.78 | 131.97 | 64.78 | 49.79 | < 3.78 | 138.52 | < 3.78 | 218.72 |
| Heptachlor | 82.01 | 161.79 | 150.95 | 200.17 | 14.74 | 51.64 | 190.19 | 300.29 |
| Aldrin | 143.4 | 342.17 | 381.81 | 514.37 | 93.49 | 290.72 | 443.12 | 85.92 |
| Dieldrin | < 2.41 | < 2.41 | < 2.41 | 20.49 | < 2.41 | < 2.41 | < 2.41 | 38.25 |
| Endrin | 29.84 | 165.84 | 89.72 | 145.73 | 66.16 | 36.13 | 160.67 | 127.58 |
| p,p' DDE | < 2.35 | < 2.35 | 90.82 | 142.46 | < 2.35 | < 2.35 | 15,89 | < 2.35 |
| p,p' DDD | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2,35 |
| p,p' DDT | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 |
| Total OCPs | 1793.13 | 1799.86 | 1055.76 | 2810.16 | 778.5 | 400.68 | 1137.84 | 2455.26 |

Table 3. Data obtained for organochlorine pesticides analyses in water samples collected from Tasaul Lake in November 2005

| ng/l | Tasaul 1 | Tasaul 2 | Tasaul 3 | Tasaul 4 | Tasaul 5 | Tasaul 6 | Tasaul 7 | Tasaul 8 |
|-------------------|----------|----------|----------|----------|----------|----------|----------|----------|
| HCB | 83.51 | 157,63 | 195,04 | <4,26 | 163,22 | 165,77 | 603 | 24.91 |
| Lindane | 346.46 | < 3.78 | 10.2 | 71.89 | < 3.78 | < 3.78 | 234.5 | < 3.78 |
| Heptachlor | 69.38 | 129.37 | 209.03 | 67.8 | 150.9 | 95.11 | 245 | 168.66 |
| Aldrin | 324.24 | 327.74 | 145.63 | 255.42 | 266.27 | 224 | 120.35 | 609.08 |
| Dieldrin | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 |
| Endrin | 57.65 | 69.71 | 221.55 | 75.78 | 99.21 | 94.3 | 174.5 | 122.24 |
| p,p' DDE | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 |
| p,p' DDD | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 |
| p,p' DDT | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 |
| Total OCPs | 888.04 | 689.55 | 788.25 | 477.69 | 686.4 | 585.97 | 1384.15 | 724.14 |

Table 4. Data obtained for organochlorine pesticides analyses in water samples collected from Tasaul Lake in March 2006

| ng/l | Tasaul 1 | Tasaul 2 | Tasaul 3 | Tasaul 4 | Tasaul 5 | Tasaul 6 | Tasaul 7 | Tasaul 8 |
|-------------------|----------|----------|----------|----------|----------|----------|----------|----------|
| HCB | 52 | 118.57 | 137.84 | 89.68 | 90.77 | 143.84 | 120.22 | 117.02 |
| Lindane | 122.02 | 85.67 | 96.72 | 71.85 | 48.43 | 243.42 | 83.08 | 104.46 |
| Heptachlor | 45.44 | 26.34 | 42.5 | 20.82 | 24.51 | 20.117 | 10.49 | 26.71 |
| Aldrin | 237.92 | 177.32 | 332.64 | 228.24 | 193.78 | 168.22 | 275.97 | 236.68 |
| Dieldrin | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 |
| Endrin | 183.7 | 149.98 | 254.62 | 146.98 | 76.55 | 97.92 | 64.96 | 75.72 |
| p,p' DDE | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 |
| p,p' DDD | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 | < 2.35 |
| p,p' DDT | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 | <2.78 |
| Total OCPs | 641.08 | 557.88 | 864.32 | 557.57 | 434.04 | 673.517 | 554.72 | 560.59 |

In May, 4 (HCB, heptachlor, aldrin, endrin) from the 9 investigated compounds had concentrations between 14.81 ng/l and 400.62 ng/l (Fig. 2). The other compounds had concentrations below the detection limit (Table 1).

In August, 7 (HCB, linden, heptachlor, aldrin, endrin, p,p' DDE) from the 9 investigated compounds had concentrations between 6.58 ng/l and 514.37 ng/l (Fig. 3). The other compounds had concentrations below the detection limit (Table 2).

In November, 6 (HCB, lindane, heptachlor, aldrin, endrin) from the 9 investigated compounds had concentrations between 10.2 ng/l and 514.37 ng/l (Figure 4). The other compounds had concentrations below the detection limit (Table 3).

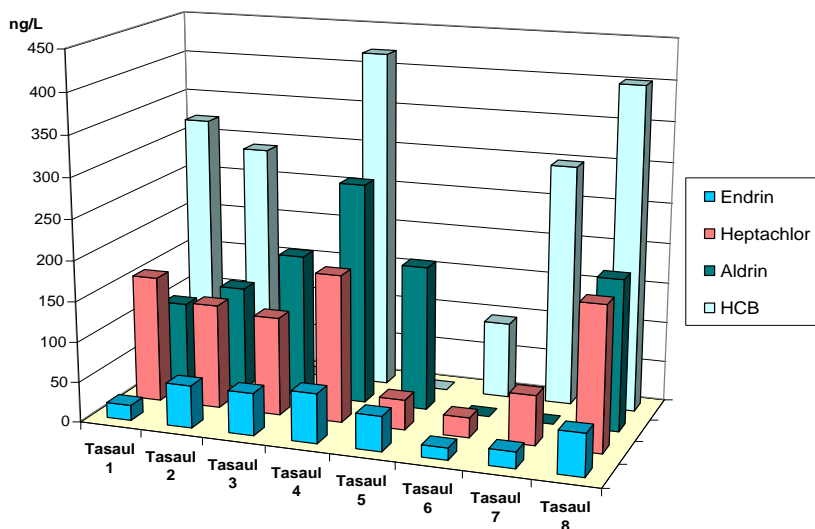


Fig. 2. Organochlorine pesticide levels in water samples collected from Tasaul Lake in May 2005

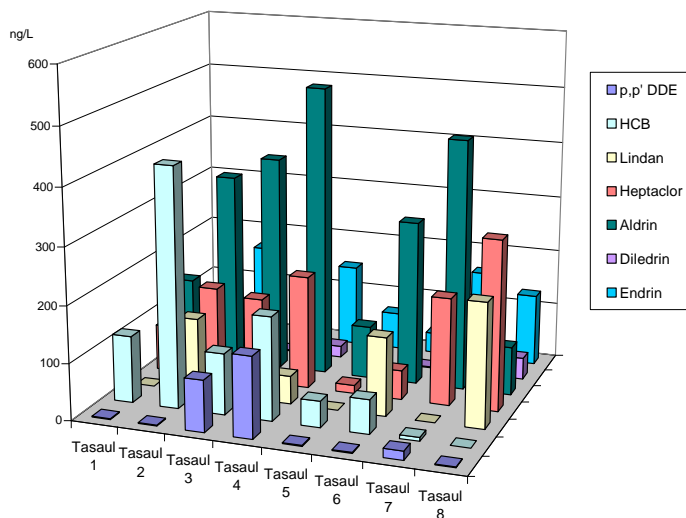


Fig. 3. Organochlorine pesticide levels in water samples collected from Tasaul Lake in August 2005

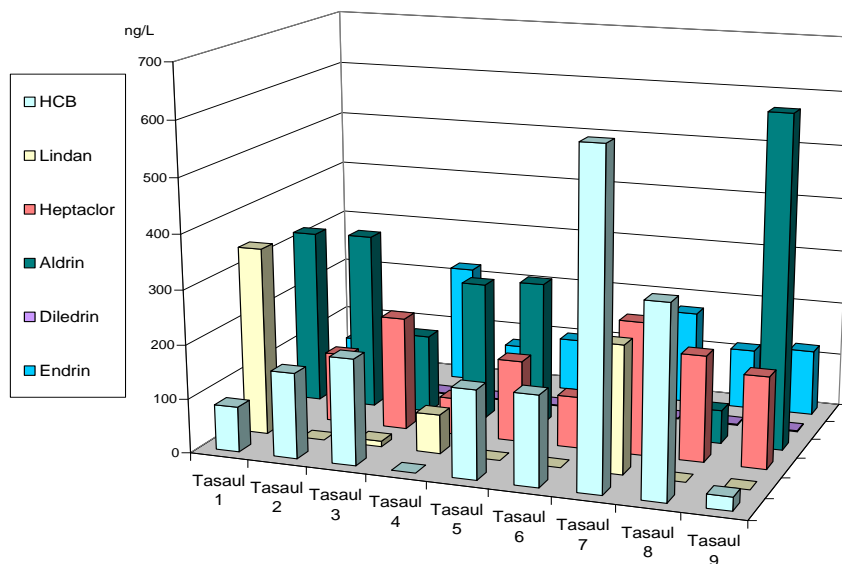


Fig. 4. Organochlorine pesticide levels in water samples collected from Tasaul Lake in November 2005

In March, 5 (HCB, lindane, heptachlor, aldrin, endrin) from the 9 investigated compounds had concentrations between 10.49 ng/l and 332.42 ng/l (Fig. 5). The other compounds had concentrations below the detection limit (Table 4).

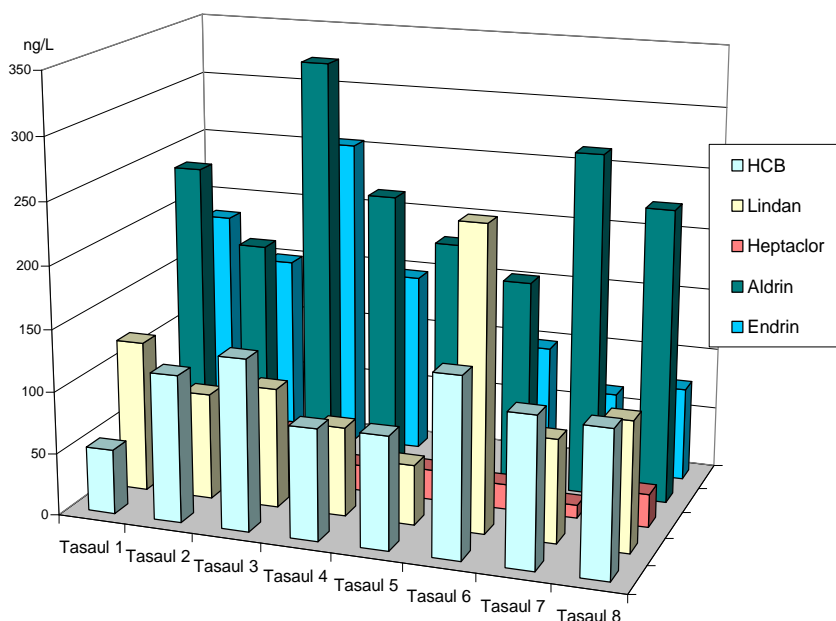


Fig. 5. Organochlorine pesticide levels in water samples collected from Tasaul Lake in March 2006

It is notable that DDT and its metabolites are below the detection limit in almost all samples. From the point of view of total organochlorine pesticide concentration, the most polluted waters are in Tasaul 1, 2, 4, 7 and 8, especially in August, when waters in sites 4 and 8 had total organochlorine pesticides concentration higher than 2 000 ng/l (Fig. 6).

The results obtained for water samples collected from influents of Tasaul Lake in 2007 are listed in Table 5.

Table 5. Data obtained for organochlorine pesticide analyses in water samples collected in 2007 from influents of Tasaul Lake

| ng/L | Sibioara | Casimcea | Delafac | Piatra |
|-------------------|----------|----------|---------|--------|
| HCB | 0 | 0 | 0 | 0 |
| Lindan | 0 | 0 | 0 | 0 |
| Heptaclor | 207.90 | 153.77 | 160.57 | 136.26 |
| Aldrin | 0 | 11.67 | 54.37 | 65.84 |
| Dieldrin | 55.63 | 0 | 25.18 | 42.71 |
| Endrin | 0 | 252.83 | 0 | 0 |
| p,p' DDE | 0 | 0 | 13.14 | 0 |
| p,p' DDD | 70.18 | 56.17 | 0 | 0 |
| p,p' DDT | 0 | 0 | 0 | 0 |
| Total OCPs | 333.71 | 474.44 | 253.26 | 244.81 |

Some of the compounds found in the lake's waters (HCB, lindane) are not present in influents' waters (Fig. 7). This means the influents do not represent the only source of pollution for organochlorine pesticides, but there is also a diffused pollution that has a significant importance.

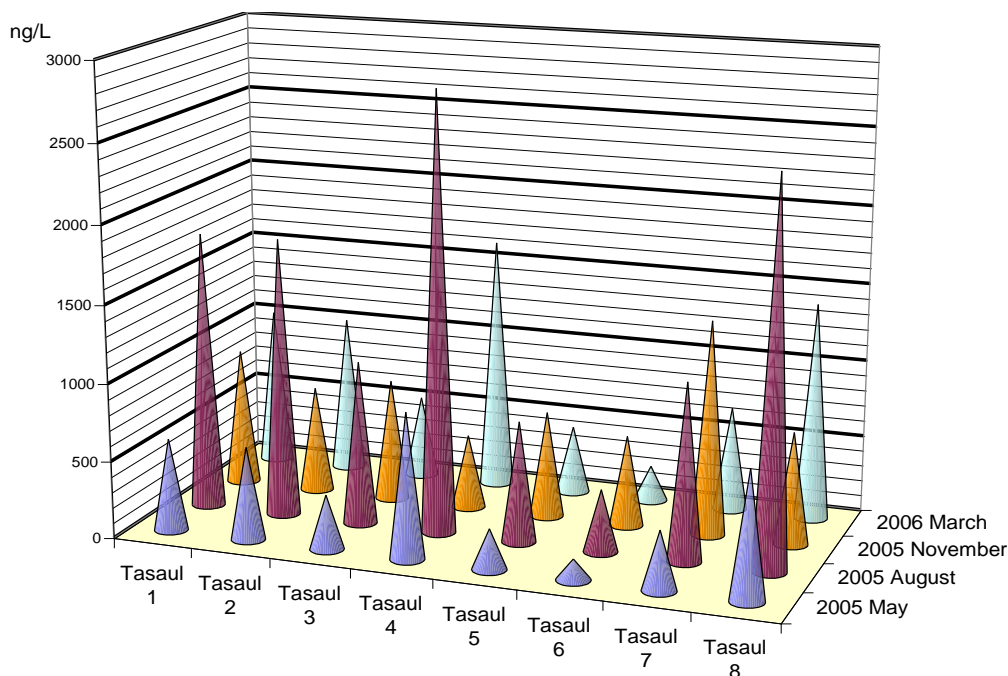


Fig. 6. Total organochlorine pesticide levels in water samples collected from Tasaul Lake in 2005 - 2006

The results obtained for sediment samples are listed in Table 6 and Table 7.

Table 6. Organochlorine pesticide levels in surface sediment samples collected from Tasaul Lake in August 2005

| ng/g dry sediment | Tasaul 1 | Tasaul 2 | Tasaul 3 | Tasaul 4 | Tasaul 5 | Tasaul 6 | Tasaul 7 | Tasaul 8 |
|-------------------|----------|----------|----------|----------|----------|----------|----------|----------|
| HCB | 8.184 | 8.69 | < 0.3 | < 0.3 | < 0.3 | < 0.3 | < 0.3 | < 0.3 |
| Lindane | 43.25 | 189.77 | 649.45 | 234.63 | 230.15 | 43.68 | 131.63 | 2.88 |
| Heptachlor | 6.68 | < 3.12 | < 3.12 | 12.69 | < 3.12 | 1.56 | < 3.12 | < 3.12 |
| Aldrin | < 3.16 | < 3.16 | < 3.16 | < 3.16 | < 3.16 | < 3.16 | < 3.16 | < 3.16 |
| Dieldrin | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 | < 2.41 |
| Endrin | 13.29 | 167.61 | 92.44 | < 3.72 | < 3.72 | < 3.72 | < 3.72 | < 3.72 |
| p,p' DDE | < 0.18 | < 0.18 | < 0.18 | < 0.18 | < 0.18 | < 0.18 | < 0.18 | < 0.18 |
| p,p' DDD | < 0.18 | < 0.18 | < 0.18 | < 0.18 | < 0.18 | < 0.18 | < 0.18 | < 0.18 |
| p,p' DDT | < 0.21 | < 0.21 | < 0.21 | < 0.21 | < 0.21 | 124.21 | < 0.21 | 159.11 |
| Total OCPs | 78.204 | 371.17 | 148.69 | 254.12 | 233.55 | 172.85 | 136.73 | 163.69 |

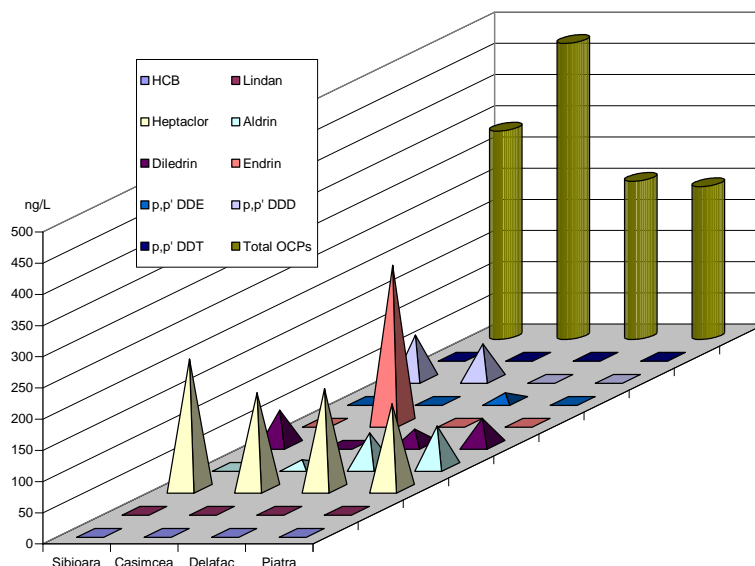


Fig. 7. Organochlorine pesticide levels in water samples collected in 2007 from influents of Tasaul Lake

Table 7. Organochlorine pesticides levels in depth core samples collected from Tasaul Lake in August 2005

| ng/g dry sediment | Tasaul 3 (0-15 cm) | Tasaul 3 (15-30 cm) | Tasaul 3 (>30 cm) | Tasaul 6 (0-15 cm) | Tasaul 6 (15-30 cm) | Tasaul 6 (>30 cm) |
|-------------------|--------------------|---------------------|-------------------|--------------------|---------------------|-------------------|
| HCB | < 0.3 | < 0.3 | < 0.3 | 44.0 | < 0.30 | < 0.3 |
| Lindane | 49.58 | 16.61 | 26.64 | < 0.30 | < 0.30 | 19.99 |
| Heptachlor | 71.07 | < 3.12 | < 312 | 23.37 | < 3.12 | < 3.12 |
| Aldrin | 140.06 | < 3.16 | < 316 | 47.7 | < 3.16 | < 3.16 |
| Dieldrin | < 2.41 | < 2.41 | 11.33 | < 2.41 | < 2.41 | < 2.41 |
| Endrin | 204.25 | 112.63 | 116.74 | < 3.72 | 150.36 | 14.15 |
| p,p' DDE | < 0.18 | < 0.18 | < 0,18 | 73.53 | < 0.18 | 1.38 |
| p,p' DDD | 10.83 | 14.36 | 13.5 | < 018 | < 0.18 | < 0.18 |
| p,p' DDT | < 0.21 | < 0.21 | < 0.21 | < 021 | < 0.21 | < 0.21 |
| Total OCPs | 480.89 | 151.025 | 175.01 | 197.10 | 162.26 | 44.02 |

In sediment samples were identified all 9 investigated compounds: HCB, lindane, heptachlor, aldrin, endrin, p,p DDE, p,p' DDD, p,p' DDT; their concentrations ranged between below detection limit and 234.63 ng/g dry sediment (Fig. 8 and 9).

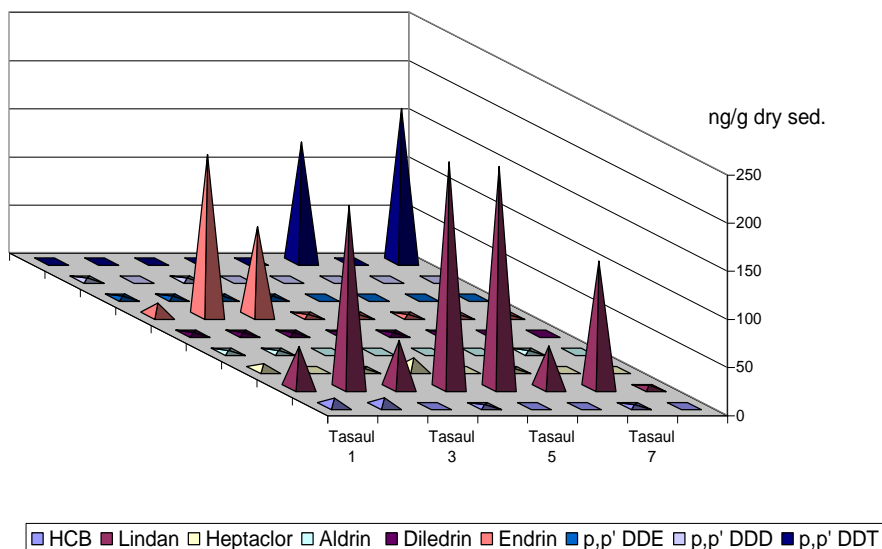


Fig. 8. Organochlorine pesticide levels in surface sediment samples collected from Tasaul Lake in 2005

In surface sediment samples lindane was predominant. Its concentrations were between 2.88 and 234.63 ng/g dry sediment. There was also some DDT in stations 7 and 8 (124.2 ng/g dry sediment, respectively 159.11 ng/g dry sediment) and endrin in stations 1, 2, and 3 (13.29 ng/g dry sediment; 167.61 ng/g dry sediment, respectively 92.44 ng/g dry sediment). Some other compounds (HCB, heptachlor) had concentrations below 15 ng/g dry sediment.

In depth core sediment samples from site 3 endrin was predominant, its concentrations ranged between 112.63 ng/g dry sediment and 204.25 ng/g dry sediment. Aldrin had also a high concentration (140.06 ng/g dry sediment) in station 3 in 0-15 m core. Some other compounds had levels below 75 ng/g dry sediment. The concentration of total organochlorine pesticides was highest in the 0-15 cm and decreased in the other two cores. In 15-30 cm core and > 15 cm core the levels were about the same (Fig. 9 a).

The situation is similar in station 6, where total organochlorine pesticides levels decreased from top core (0-15 cm) to bottom core (> 30 cm), but the concentrations were lower than station 3 (Fig. 9 b). The highest concentration was detected for endrin in the middle core (15-30 cm), the other compounds did not exceed 73.5 ng/g dry sediment.

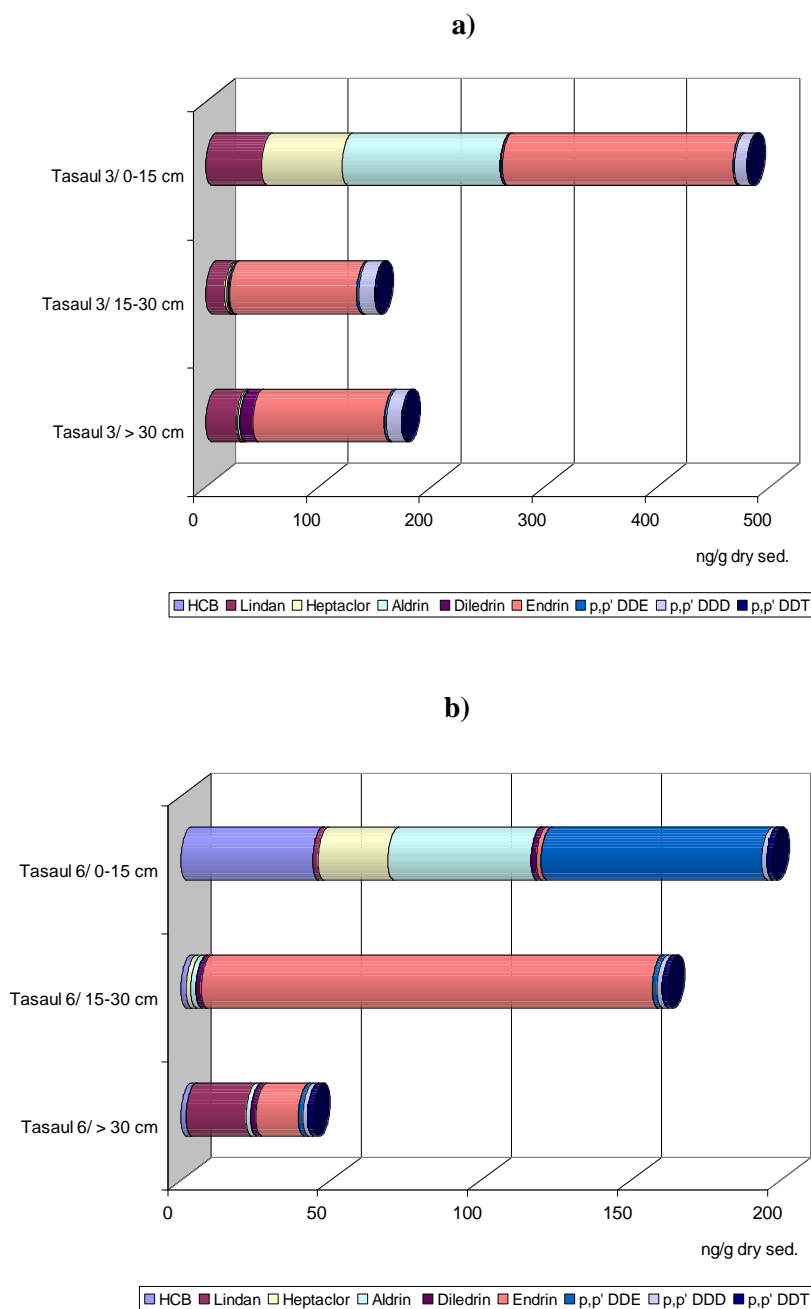


Fig. 9. Organochlorine pesticide levels in depth core sediment samples collected from sites 3 (a) and 6 (b), from Tasaul Lake in 2005

CONCLUSIONS

1. The level of total organochlorine pesticides in lake water is influenced not necessarily by hot spot sites like Tasaul's influents, but more probably by diffused pollution. The highest concentrations were detected in August after a period with a lot of rain that washed out the land around the lake.
2. Generally, the concentrations of total organochlorine pesticides in surface sediments collected from different sites varied in small limits and we can not incriminate a point source of pollution with this kind of compounds. In core sediments, there is an evident concentration of pesticides in the top core (0-15 cm) in both analysed sites.

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