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Monitoring of oil products and hazardous substances in Estonian surface water bodies

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Abstract. Compared to the "old" member states of the European Union little attention has been paid to the monitoring of hazardous substances in Estonia, but it is still possible to draw preliminary conclusions about priority substances. The concentrations of oil products and hazardous substances were studied in Estonian surface waters. In Estonia oil products are not included in the lists of hazardous substances. While Estonia has long-term experience in environmental monitoring of surface water, especially for eutrophication, as well as determination of some heavy metals in Estonian rivers subject to monitoring, in the field of toxic persistent organic compounds and oil products the results are not numerous. Although a large amount of environmental monitoring information is available, a comprehensive overview of priority hazardous substances and oil products covering all major problem areas in Estonia is not available due to lack of risk assessment, risk and data management, cross-national synthesis, and integrated framework projects in this field in Estonia. The aim of this article is to give an overview of hazardous substances and oil products in Estonian surface waters at the beginning of the 21st century.

Key words: hazardous substances, oil, monitoring, Estonia, surface water.

INTRODUCTION

The amount of chemicals on the market is enormous and it is extremely difficult to get exact information on their actual quantity. At the same time it is evident that we cannot manage without using chemicals in the modern society. Rapid economic development has brought about alongside with numerous benefits also distinct pollution problems concerning the air, ground, water bodies, and organisms surrounding us. A substance hazardous to the aquatic environment is any element or compound that due to toxicity, persistence, or bioaccumulation will cause or may cause hazard to human health upon occurrence in the aquatic environment and will

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or may damage other living organisms or ecosystems. In Estonia hazardous substances are divided into two groups on the basis of hazardousness and enumerated in the relevant lists. List 1 includes substances the release or disposal of which into water shall be prevented and List 2 substances the release or disposal of which into water shall be limited. In Estonia oil products are not included in the two hazardous substances lists. So far, hazardous substances and oil products have been found mainly as a result of environmental monitoring carried out after accidents or appearance of problems related to human health [1, 2].

The Estonian National Environmental Monitoring Programme was initiated in 1994 [3]. Presently there are altogether around 1800 monitoring stations in the monitoring set of 68 sub-programmes of 11 monitoring themes, the parameters reaching 250 [4–7].

The main objective of monitoring priority hazardous substances in Estonian surface water bodies is to observe long-term changes of hazardous substances in them and to assess their contamination. The results will serve as the basis for planning further measures for achieving a good condition of surface water bodies. Another main objective directly related to the monitoring of priority hazardous substances, hazardous substances, and other chemical compounds in surface water bodies is development of environmentally safe technologies.

All European Union (EU) member states, among them Estonia, shall specify priority hazardous substances for surface water bodies on national level [8]. Compared to the "old" member states of the EU little attention has been paid to the monitoring of hazardous substances in Estonia, but it is still possible to draw preliminary conclusions about priority substances.

It is planned to launch the first monitoring project with a monitoring network in 2007. It would be the basis for further monitoring of priority hazardous substances, hazardous substances, and other chemicals in surface water [9].

The aim of this article is to give an overview of hazardous substances and oil products in Estonian surface waters at the beginning of the 21st century.

MATERIALS AND METHODS

Methods used in the analysis of oil products and hazardous substances in water bodies

The Estonian Environmental Research Centre (EERC) is specialized in chemical analyses in the field of environmental protection. EERC facilities are well equipped, enabling precise determination of environmentally dangerous substances and oil products in different sample types. EERC provides a comprehensive range of analyses, all made in compliance with international standards. EERC is accredited by the German accreditation bureau Deutsches Akkreditierungssystem Prüfwesen GmbH (DAP) (reg. No. DAP-PL-3131.00) and the Estonian Standard Board (reg. No. L008).

Analyses of oil products from water bodies were made in accordance with the method specified by ISO 9377-2:2000 [10] and the Estonian standard EVS-EN

ISO 9377-2:2001 [11]. The method is suitable for surface water, waste water, and water from sewage treatment plants and allows the determination of a hydrocarbon oil index in concentrations above 0.1 mg/L. The oil products analysis was performed with a gas chromatograph Agilent GC 6890 (Agilent Technologies, Palo Alto, CA, USA); injection technique: programmed temperature vaporization (PTV). Chromatographic separation of 1 μL was performed on a DB 5 MS column with a length of 30 m, ID 0.25 mm, film thickness 0.25 μm. The GC oven was programmed as follows: 40 °C initial hold for 5 min, increased at a rate of 10 °C/min to 300 °C, then 300 °C for 20 min.

Description of the sampling techniques and analytical procedures for persistent organic pollutants can be found in [6, 12]. Chlorinated compounds such as polychlorinated biphenyl congeners and chlororganic pesticides (aldrin, dieldrin, endrin, DDT, lindane, HCB) were analysed on a 90 m capillary column (DB 5) using gas chromatography (Varian 3380) with an electron capture detector.

The analysis of heavy metals followed ISO 8288-1986 (E) [13]. An AAS Varian SpectrAA-250 Plus atom absorption spectrophotometer with graphite and flame furnaces [6] was used for the analysis.

RESULTS AND DISCUSSION

At the moment, the authors can base only on some national programmes of environmental monitoring (see Fig. 1 for the location of monitoring points in

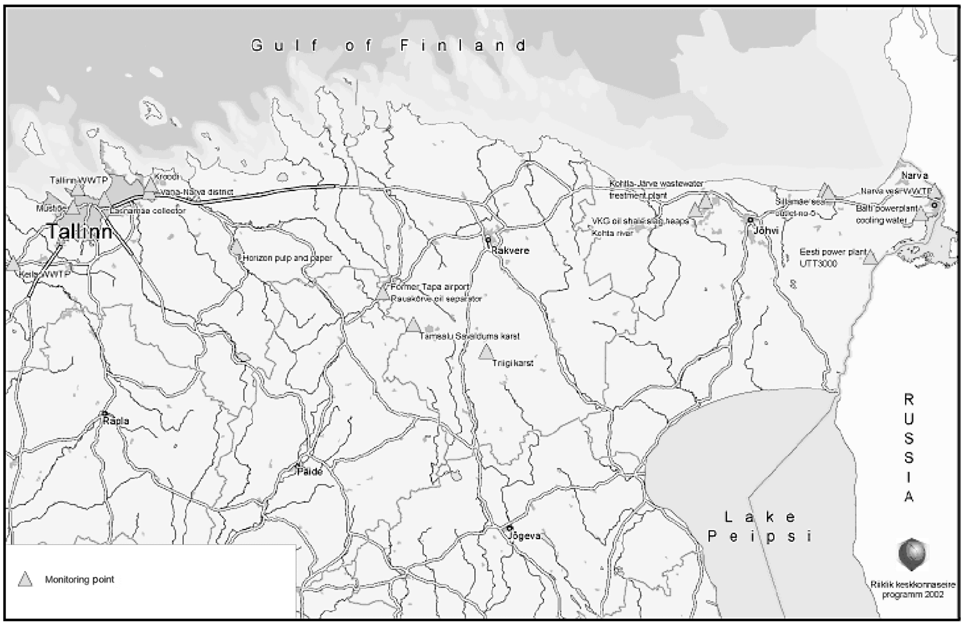

Fig. 1. Monitoring points of hazardous substances in Estonia in 2002–2003.

2002–2003) and inventory results of hazardous substances from the period 1999–2001. The last national programme covered 90% of substances polluting water in Estonia; however, single sampling and analysis of a few random samples do not suffice for drawing adequate conclusions about the state of surface water.

Inventory of discharges of chemical compounds in 1999–2001

The objective of the inventory of hazardous substances was to get a systematic overview of emissions of hazardous substances into the environment. Mainly emissions of hazardous substances from industry in general and from industry to the public sewerage system were studied. Emissions caused from residual pollution and hazardous wastes were of secondary importance. Studying the effect of emissions on the recipient is the objective of further projects.

This overview is based on the results of three nation-wide inventories of hazardous substances performed in 1999–2001:

– inventory of emissions of hazardous substances and programme for the reduction of emissions in Hiiu, Jõgeva, Järva, Lääne, Tartu, Põlva, Pärnu, Rapla, Saare, Valga, Viljandi, and Võru counties [14]
– inventory of emissions of hazardous substances in Lääne-Viru and Ida-Viru counties [15]
– research of emissions of hazardous substances in Tallinn and Harju County [16].

The inventories covered the following:

– identification of generators of potential emissions of hazardous substances
– assessment of the pollution load – indirect emissions into groundwater, emissions into water bodies, and direct emissions into the public sewerage system
– pinpointing the companies that should be obliged to monitor hazardous substances.

The amounts of substances hazardous to the aquatic environment included in List 1 and List 2 discharged into the aquatic environment in Estonia calculated on the basis of the inventories are presented in Tables 1 and 2. The total calculated emissions of oil products into water via the sewerage system in Estonia amount to 15 771 kg/year [14–16].

Table 1. Total calculated emissions of substances of List 1 of hazardous substances of Estonia into water via the sewerage system [14–16]

Substance	Total emission, kg/year
Carbon tetrachloride	304.0
Perchloroethene	21.9
Trichloroethene	10.4
1,2-Dichloroethane	3.9
Cd	3.1
Chloroform	2.1
Pentachlorophenol	1.7
Hg	0.3
Cyanide	0.2
Lindane	0.02

Table 2. Total calculated emissions of substances of List 2 of hazardous substances of Estonia into water via the sewerage system [14–16]

Substance	Total emission, kg/year
Ba	7531
Ni	4980
Zn	2566
1-Basic phenols of List 2	866
Cr	451
Cu	398
As	104
Co	90.7
Pb	83.2
Mo	76.7
Benzene	19.7
Sn	2.1
V	1.7
Se	0.6
Polycyclic aromatic hydrocarbons of List 2	0.09

Monitoring of oil products and hazardous substances for Estonian surface water bodies

National monitoring programme "Monitoring of hazardous substances in Estonian rivers"

Oil products (hydrocarbons). The concentration of oil hydrocarbons is determined according to the monitoring programme in rivers falling to the sea and in the Emajõgi River 6 times a year and in other rivers 1 or 2 times a year. In 2003 the concentration of oil hydrocarbons was studied in 17 rivers (in 2002 in 22 rivers). The concentrations of oil hydrocarbons in Estonian rivers are low, remaining in most rivers below 50 μg/L or below the detecion limit of this method [17, 18]. Like in previous years, in 2003 the highest levels were measured in the Purtse River, where the concentration of oil products remained within the range 37–271 μg/L. Single higher concentrations were measured also in the Pärnu River in August (320 μg/L), in the Selja River in July (73 μg/L), and in the Püha River in March (78 μg/L). According to the limit values used in Europe (excellent – 0 μg/L, good – 20 μg/L, satisfactory – 50 μg/L), the water of most Estonian rivers is of good and satisfactory quality for the concentration of oil hydrocarbons, because the concentrations remain within the range 1–50 μg/L (there are only single higher concentrations, except the Purtse River, where 65% of the measurement results of the year exceeded the permitted limit value). Compared to the previous years the situation had not changed [17, 18]. The requirements of the EU Directive on Habitats of Freshwater Fish (78/659/EEC) have been taken into account in national monitoring programme of rivers since 2002. Table 3 shows that the studied rivers met the requirements specified in the directives of the European Union.

Table 3. Concentrations of hazardous substances in Estonian fishing rivers in 2002 [18]

Sampling location	Target fish	Hg, μg/L	Cd, μg/L	Cu, μg/L	Pb, μg/L	Zn, μg/L
Kasari R.	Cyprinids	0.1	0.03	1.5	0.2	4
Mouth of Keila R.	Salmonids	<0.1	0.05–0.54	8–19	<0.2–0.8	3–22
Pirita R.	Salmonids	<0.1	0.29	2.6	0.2	19
Pärnu R.	Salmonids	<0.05	<0.1	3	<1	<10
Mouth of Selja R.	Salmonids	0.15	0.06	2	1	10
Mouth of Kunda R.	Salmonids	0.1–0.65	0.06–0.08	10–33	1–4	<10–21
Narva R.	Salmonids	0.13	0.09	36	<1	<10
Emajõgi R.	Cyprinids	<0.1	<0.02–0.04	<1–2.5	0.4–1.0	3–9

Regulation No. 58 of the Estonian Minister of the Environment from 9 October 2002 “Requirements for the quality and monitoring of waterbodies protected as habitats for salmonids and cyprinids and stations of national environmental monitoring of salmonids and cyprinids” and the EU Directive on Habitats of Freshwater Fish (78/659/EEC) specify also limit values for the concentrations of heavy metals (Cu, Zn), oil products, and phenols. These indicators have been determined in 15 monitoring points of rivers according to sampling frequency specified in the monitoring programme. As there are no direct sources of heavy metals in Estonia, concentrations of Cu, Cd, Pb, Zn, and Hg have been determined once a year, and only in four rivers (mouth of the Keila River, mouth of the Kunda River, Oore monitoring point on the Pärnu River, and Kavastu monitoring point on the Emajõgi River) studies of heavy metals have been carried out 6 times a year. According to the monitoring programme, oil products have been determined 6 times a year and the concentration of phenols in the rivers of North-East Estonia 6–12 times a year. According to regulation No. 58 of the Estonian Minister of the Environment, in salmonid rivers the limit value for phenols is 5 μg/L, for oil products 20 μg/L, for Cu 40 μg/L, and for Zn 300 μg/L [18]. (Limit values are given for water hardness of 100–300 mg/L $CaCO_3$, which corresponds to the hardness of the water of Estonian rivers.)

The rivers studied within the frames of the monitoring programme meet the above-mentioned standards. Higher levels were found only in concentrations of oil products in the rivers of North-East Estonia, which exceed the limit values established in Estonia (in the Püha River <10–78 μg/L and in the Selja River <10–73 μg/L). Levels specified for oil products and phenols in Regulation No. 58 of the Estonian Minister of the Environment are very strict, limit values of these indicators are either close to the detection limit or even below (determination of oil products with gas chromatography). The results show that Estonian rivers face no problems with hazardous substances. Concentrations of heavy metals in most rivers of Estonia are low. The majority of Estonian rivers belong into the class of clean waters according to the classification used in Europe and into quality class I according to the standards established in Estonia [5, 18]. The Purtse and Pühajõgi rivers are polluted with oil hydrocarbons and phenols. Other rivers belong into the water class of good quality according to the standards used in Europe.

National monitoring programme "Hazardous substances in surface waters"

Oil products in water. Rotation monitoring of hazardous substances in water bodies with the length of three years started from North-East Estonia, which is the most polluted region in Estonia. Monitoring points of recipients and compounds subject to analysis were selected by experts from the Ministry of the Environment, University of Tartu, and Estonian Environmental Research Centre.

For example, in the course of monitoring in 2002 a pollution source from the Russian side was detected in the Plyussa River downstream from Slantsy within the frames of a Swedish–Russian–Estonian joint expedition. As a result, also the mouth of the Plyussa River was included into monitoring.

The total concentration of oil products in the water of the Narva River downstream from the effluent of the town of Narva exceeded the limit value (10 µg/L) established for surface and sea waters (Table 4). As to groundwater

Table 4. Concentration of hazardous substances in water samples from North-East Estonia [19]

Substance	Viru Keemia				Narva River, downstream from effluent of Narva WWTP	Mouth of Plyussa River (Russia)
	Kohtla River, down-stream from the outlet	Kohtla River at Lüga-nuse	Purtse River down-stream from Kohtla	Mouth of Purtse River		
Aldrin, ng/L	<10	<10	<10	<5	<10	<10
Dieldrin, ng/L	<10	<10	<10	<5	<10	<10
Endrin, ng/L	<10	<10	<10	<5	<10	<10
DDT, ng/L	<10	<10	<10	<5	<10	<10
Lindane, ng/L	1	<10	<10	<5	<10	<10
HCB, ng/L	<10	<10	<10	<5	<10	<10
1,2-Dichloroethane, µg/L	<1	<1	<1	<1	<1	<1
Trichloromethane, µg/L	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Trichloroethylene, µg/L	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Tetrachloroethylene, µg/L	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Tetrachloromethane or carbon tetrachloride, µg/L	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Hg, µg/L	0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Cd, µg/L	0.1	<0.1	<0.1	<0.1	0.1	<0.02
PAHs, µg/L	–	–	–	–	–	0.011
Oil products, µg/L	–	–	–	–	87.6	–
Sulphides, mg/L	–	–	–	–	<0.02	–
Sn, µg/L	–	–	–	–	<0.005	–
Ni, µg/L	–	–	–	–	<1	–
Cu, µg/L	–	–	–	–	18	–
Pb, µg/L	–	–	–	–	2	–
Zn, µg/L	–	–	–	–	<0.01	–
Cr, µg/L	–	–	–	–	<1	–

– not determined.

standards, the effluent contained over four times more oil products than their target value (i.e. the concentration in the case of whose equal or lower value the condition of groundwater is good, that is not hazardous to humans or the environment) but still below the relevant limit value.

Cadmium and mercury concentrations in water samples remained considerably lower than the limit values for concentrations of hazardous substances in the effluents discharged into water bodies and the limit values of hazardous substances in groundwater.

Concentrations of lead, copper, nickel, chromium, tin, and zinc in water samples were determined in the Narva River downstream from the effluent of Narva. The results did not exceed target values established to groundwater or limit values of concentrations of hazardous substances in effluents. Concentrations of organochlorine pesticides and hexachlorobenzene in the water samples also remained below the established target values in groundwater and limit values set to effluents. The concentrations of polycyclic aromatic hydrocarbons (PAHs) in the water of the mouth of the Plyussa River did not exceed the limit values valid in Estonia.

Concentrations of trichloroethylene, chloroform, tetrachloromethane, and tetrachloroethylene in surface water were analysed in the Kohtla River downstream from the outlet of Viru Keemia Grupp, in the Narva River downstream from the effluent of Narva, and in the mouth of the Plyussa River. The results did not exceed the respective limit values. Also the concentrations of 1,2-dichloroethane in surface water samples were below the limit values.

The monitoring reports of hazardous substances from the years 2002, 2003, and 2004 as well as inventory reports of discharges of hazardous substances from earlier years were studied as well (Table 5).

Oil products in bottom sediments. The total concentration of oil products in the bottom sediments of the Narva River downstream from the effluent of Narva did not exceed the target value established for soil (100 mg/kg).

In order to get a better overview of the pollution of water bodies with hazardous chemicals, we compared the results of analyses of bottom sediments and water samples from water bodies with the target values valid in Estonia.

Cadmium concentrations in bottom sediments remained below the established target value in all samples of bottom sediments. The highest cadmium concentrations, 0.48 mg/kg, were measured in bottom sediments of the Purtse River downstream from its confluence with the Kohtla River. The cadmium concentration in the bottom sediments in the mouth of the Plyussa River (Russia) and in the Kohtla River downstream from the outlet of Viru Keemia Grupp amounted to one third of the target value, or 0.33 mg/kg (Table 6).

Table 5. Average annual concentrations and minimum and maximum levels of some priority hazardous substances included into List 1 on the basis of directive 92/446/EEC in 2002–2004 [9, 19]. The number of samples analysed during the year is given in parentheses

Substance	2002[a]	2003[b]	2004[c]
Mercury	<0.05–0.05 μg/L (5)	<0.05 μg/L <0.05–<0.05 μg/L (10)	<0.05 μg/L <0.05–<0.05 μg/L (10)
Cadmium	0.28 μg/L <0.1–1.0 μg/L (5)	0.15 μg/L 0.02–0.46 μg/L (10)	0.27 μg/L <0.1–0.40 μg/L (10)
Hexachlorocyclo-hexane	<10 ng/L (5)	<10 ng/L (2)	–
Tetrachlorocarbon (tetrachloromethane)	<0.1 μg/L (5)	<0.1 μg/L (4)	–
DDT	<10 ng/L (5)	<10 ng/L (2)	–
Drins			
Aldrin	<5 ng/L	<10 ng/L	–
Dieldrin	<5 ng/L	<10 ng/L	–
Endrin	<5 ng/L (5)	<10 ng/L (2)	–
Hexachlorobenzene, HCB	<10 ng/L (5)	<10 ng/L (2)	–
Chloroform (trichloromethane)	<0.1 μg/L (5)	<0.1 μg/L (4)	–
1,2-Dichloroethane	<1 μg/L (5)	<1 μg/L (2)	–
Trichloroethylene	<1 μg/L (5)	<1 μg/L (2)	–

[a] Sampling region: Ida-Viru County; sampling point: Viru Keemia Grupp (Kohtla and Purtse rivers) and Narva Vesi (Narva River downstream from Narva).
[b] Sampling region: Harju County; sampling points: Kroodi Creek, Lasnamäe sewer, Vana-Narva Road, Mustaoja Creek (Paldiski Road), and outlet of Keila WWTP.
[c] Sampling regions: Harju and Põlva counties; sampling points: area of Vana-Narva Road and AS Räpina Paber.
– not determined.

Mercury concentrations in bottom sediments remained below the established target value in all samples. The highest mercury concentrations, 0.43 mg/kg, were measured in bottom sediments of the Purtse River after its confluence with the Kohtla River. The mercury concentration in the bottom sediments of the mouth of the Plyussa River was 0.13 mg/kg.

The concentrations of aldrin, dieldrin, endrin, DDT, hexachlorocyclohexane, and hexachlorobenzene remained below the established target values in all bottom sediments.

Table 6. Concentration of hazardous substances in bottom sediments (sample range) [19, 20]

Substance	Viru Keemia, Kohtla River	Viru Keemia, Purtse River	Estonian TPS, Mustajõe downstream from cooling water dis-charge canal	Narva Vesi, Narva River downstream from effluent of Narva	Baltic TPS, cooling water discharge canal	Mouth of Plyussa River
Aldrin, µg/kg	<5	<5	<5	<5	<1	<1
Dieldrin, µg/kg	<5	<5	<5	<5	<1	<1
Endrin, µg/kg	<5	<5	<5	<5	<1	<1
DDT, µg/kg	<5	<5	<5	<5	<1	<1
Lindane, µg/kg	<5	<5	<5	<5	<1	<1
HCB, µg/kg	<5	<5	<5	<5	<1	<1
Hg, mg/kg	0.03–0.04	0.05–0.43	0.02–0.04	<0.02	0.045–0.047	0.131–0.132
Cd, mg/kg	0.196–0.331	<0.25–0.484	<0.25	<0.25	0.119–0.123	0.329–0.331
PAHs, mg/kg	–	–	–	–	–	0.15
Oil products, mg/kg	–	–	–	36.9	–	–
Sn, mg/kg	0.283–0.724	0.255–1.026	–	<0.25	–	–
Ni, mg/kg	3.54–3.84	5.14–15.5	–	1.44	–	–
Cu, mg/kg	6.11–11.7	5.1–17.5	–	2.8	–	–
Pb, mg/kg	<2.5–4.01	10.0–15.8	–	<2.5	–	–
Zn, mg/kg	14.1–25.2	19.0–61.4	–	9.06	–	–
Cr, mg/kg	10.3–11.6	5.56–6.96	–	<1.25	–	–
1-Basic phenols, mg/kg	–	–	–	–	0.14	–
2-Basic phenols, mg/kg	–	–	–	–	2.0	–
PCB, µg/kg	–	–	–	–	–	<5

– not determined.

The concentrations of chromium, copper, nickel, lead, and tin in the samples of bottom sediments taken from the mouth of the Purtse River and from the Purtse River after its confluence with the Kohtla River remained below the established target value. The concentrations of all the mentioned heavy metals in the samples of bottom sediments taken from the mouth of the Purtse River were lower than the relevant parameters in the Purtse River after its confluence with the Kohtla River.

1-Basic phenols in the bottom sediments of the mouth area of the cooling water discharge canal of the Baltic TPS did not exceed the established target value. However, 2-basic phenols in the same sampling area exceeded twice the target value, although were noticeably below the established target value for residential zones.

The total concentration of PAHs in the bottom sediments taken from the mouth of the Plyussa River remained below the established target value. From isomers of PAH, naphthalene formed the main part (65%), followed by phenantrene and benzo(*b*,*k*)fluoranthene, both 5%. The concentrations of polychlorinated biphenyls in the bottom sediments of the mouth of the Plyussa River remained below the established target value.

CONCLUSIONS

Compared to the "old" member states of the EU little attention has been paid to the monitoring of hazardous substances in Estonia. However, it is possible to draw some conclusions about priority substances. The list of priority hazardous substances of Estonia should include heavy metals (Cr, Cu, Ni, Zn, Ba) and phenols (1-basic and 2-basic phenols) as data of inventories and a report [9] suggest. As phenols are of special interest from the standpoint of Estonian environmental protection, we shall discuss them more thoroughly in the future. Problems are directly related to the mining and processing of oil shale in North-East Estonia.

The priority substances list is not final. Upon reception of new information hazardous substances should be added into the list or, if no hazard exists, removed from the list.

The concentrations of heavy metals [18], persistent organic pollutants [6, 19], and oil products in most Estonian water bodies are low. In most cases Estonian rivers belong into the class of clean waters according to the classification used in Europe (waters with good and satisfactory quality) and into quality class I according to the standards established in Estonia. Presently Estonian rivers have no problems with hazardous substances [5, 17, 18].

Therefore, in monitoring hazardous substances in water bodies, we shall observe in the future also their emissions into the surrounding environment and shall pay attention to the improvement of conditions of production, transport, incineration, and disposal of (hazardous) waste. Monitoring shall cover also hazardous substances used earlier that are now prohibited in Estonia, but the waste of which can still be found here.

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Naftaproduktide ja ohtlike kemikaalide seire Eesti pinnaveekogudes

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Eestis pole naftaproduktide ja ohtlike kemikaalide seirele nn vanade Euroopa Liidu liikmesriikidega võrreldes siiani piisavalt tähelepanu pööratud. Artiklis on püütud anda ülevaade ohtlike kemikaalide ja naftaproduktide senitehtud analüüsidest Eesti pinnaveekogudes. Käesoleval ajal ei kuulu naftaproduktid Eesti ohtlike ainete nimekirjadesse number 1 ja 2. Neis on loetelu ainetest, mille tootmine ja kasutamine tuleb lõpetada või mille kasutamist tuleks piirata. Artikkel põhineb Eesti Riikliku Keskkonnaseire programmi raames tehtud uurimustel ja aastatel 1999–2001 kogu Eestis läbi viidud ohtlike ainete inventuuri andmetel.