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Baltic Marine Environment Protection Commission

The sixth pollution load compilation (PLC-6)

PRESSURE





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Preface

Various land-based pollution remains the major source of environmental pressure on the Baltic Sea ecosystem. This pressure includes eutrophication, caused by excess supply of the marine environment by nutrients (nitrogen and phosphorus) and pollution of the marine environment by heavy metals, persistent organic pollutants and other synthetic hazardous substances. One of the largest HELCOM projects, Pollution Load Compilation (PLC), is missioned to supply the most up-to-date information on landbased input of nutrients and selected hazardous substances to the marine environment, their sources and pathways. Compilations of pollution load data have been an integral part of the HELCOM assessment system since 1987, including annual and periodic assessments. The current assessment is the sixth one, which found reflection in the abbreviation of the project title - PLC-6. The assessment creates scientific background for the Contracting Parties to undertake measures to prevent and eliminate pollution of the Baltic marine environment in accordance with Article 16 of the Convention on the Protection of the Marine Environment of the Baltic Sea Area, 1992 (Helsinki Convention).

To fulfill all the tasks PLC-6 project delivers six major products:

- 1. Updated HELCOM Core Pressure Indicator on nutrient inputs (MAI fulfilment follow-up) covering data from 1995 to 2015
- Follow-up progress towards national reduction targets for nutrients, CART follow-up assessment, covering data from 1995 to 2014 (policy message and scientific report)
- Assessment of sources and pathways of nutrients into the Baltic Sea environment in 2014
- 4. Input of nutrients by the seven biggest rivers
- 5. Evaluation of effectiveness of measures to reduce nutrient inputs to the Baltic Sea
- 6. Assessment of input of selected hazardous substances

Figure 1. Baltic Sea catchment and PLC subcatchment areas with monitoring status of areas.

PLC data

Scientifically sound and compatible data on pollution loads across the region is the key factor for reliable assessment results. The HELCOM Pollution Load Compilation is supplied by national data on waterborne inputs from all Contracting Parties obtained mainly through national monitoring programmes (map illustrating monitoring data coverage, Fig. 1) based on HELCOM monitoring and reporting guidelines. The waterborne data for the period from 1995 to 2014 which is used for PLC-6 assessment products are stored in the HELCOM PLC-water database. The database also provides public access to the raw data. All reported data is also spatially referenced and can be visualized as maps using the HELCOM map and data service. Data on atmospheric inputs is annually provided by EMEP based on national reports on nitrogen emissions. Atmospheric deposition of phosphorus is accounted as a fixed rate of 5 kg phosphorus per km².

In the compilation of data for the HELCOM PLC assessment, special attention is paid to assure the quality of reported data. HELCOM PLC data is to pass four quality control stages. The WEB-based Reporting Tool, specifically designed for reporting data on pollution loads, automatically verifies the technical parameters of the reported data and performs the statistical evaluation of their quality. The data which does not satisfy statistical verification criteria is automatically marked as suspicious and further manually validated by national experts responsible for PLC data reporting. The final data quality verification is performed by experts as part of the assessment procedures.



Progress towards Maximum Allowable Inputs (MAI)

A significant reduction of nutrient inputs has been achieved for the whole Baltic Sea by 2015. The last assessment shows that the normalized input of nitrogen was reduced by 12% and phosphorus by 25% since the reference period (1997-2003) (Results for N&P Fig. 2, 3). The Maximum Allowable Inputs (MAI) of nitrogen in this period were fulfilled in the Kattegat, Danish Straits, Bothnian Bay and Bothnian Sea (Table 1a). The nitrogen input into the Gulf of Riga is below MAI but cannot be considered as fulfilled due to statistical uncertainty. MAI for phosphorus input is fulfilled in the Kattegat, Danish Straits and Bothnian Sea (Table 1b). The inputs to the Gulf of Finland and Bothnian Bay are below MAI but cannot be considered as fulfilled due to statistical uncertainty.

> Baltic Sea (BAS) entire waterbody 🔵 – Nitrogen (N) Phosphorous (P)

> > Ν

-12%

p

-25%

Ν



Figure 2.

Reduction of nitrogen (N) and phosphorous (P) inputs achieved in 2015 since the reference period (1997-2003), with classification of achieving MAI.

Danish Straights (DS)

– Nitrogen (N) Phosphorous (P)

-17%

Ρ

-2%

Analyses of nutrient input trends for the whole observation period starting from 1995 show statistically significant steady reduction of total inputs of nitrogen and phosphorus to the Baltic Sea amounting correspondingly to 20% and 27% by 2015 (Fig. 4, 5). A steady reduction of nitrogen input throughout the whole period was observed in Danish Straits, Kattegat, Gulf of Riga and Baltic Proper. A significant decrease of nitrogen input to the Bothnian Sea was observed after 2003, to the Gulf of Finland after 2004 and to the Bothnian Bay only after 2006. The input of phosphorus decreased significantly throughout the whole period in the Kattegat. Four basins demonstrate significant decreases in the first section of the time series: Baltic Proper (1995 to 2011), Bothnian Bay (1995 to 2002), Bothnian Sea (1995 to 2002) and Danish Straits (1995 to 1999). The reduction of Pinput to the Gulf of Finland has been achieved in the last years starting from 2009. Unfortunately, P input to the Gulf of Riga was increasing until 2007 and no statistically significant reduction has been observed in the last period.

MAI (Maximum Allowable Inputs)

Maximum Allowable Inputs (MAI) of nutrients are a part of the HELCOM nutrient reduction scheme, indicating the maximum level of total (waterand airborne) input of nitrogen and phosphorus to the Baltic Sea sub-basins (map of MAI fulfilment at the HELCOM map service) that is allowed to fulfil the targets for a sea unaffected by eutrophication. The targets were first included into the Baltic Sea Action Plan in 2007 and then updated in the Copenhagen Ministerial Declaration in 2013. Input of nutrients is one of HELCOM core pressure indicators.

Table 1a (N) and 1b (P):

The average normalized annual inputs of nitrogen and phosphorus during 2015. The table also includs statistical uncertainty, inputs in 2015 including statistical uncertainty and the remaining reduction needed to reach MAI. Classification of achieving MAI: () green=MAI fulfilled, () yellow= fulfilment is not determined due to statistical uncertainty, and () red=MAI not fulfilled. (() to tenth, hundreds or thousands) has been performed in the indicator.

Nitrogen (N) Baltic Sea sub-basin	MAI*	N input 2015	Statistical uncertainty 2015	N input in- cluding stat. uncertainty 2015	Exceedance of MAI	Input 2015 including stat. uncertainty in % of MAI	Classification of achieved reduction
Bothnian Bay (BOB)	57,622	54,092	1,852	55,944		97	۲
Bothnian Sea (BOS)	79,372	71,305	2,479	73,784		93	٠
Baltic Proper (BAP)	325,000	387,711	12,358	400,069	75,069	123	٠
Gulf of Finland (GUF)	101,800	107,746	7,535	115,280	13,480	113	٠
Gulf of Riga (GUR)	88,417	83,398	5,533	88,931	514**	101	
Danish Straits (DS)	65,998	55,033	2,009	57,042		86	٠
Kattegat (KAT)	74,000	65,422	1,394	66,816		90	٠
Baltic Sea (BAS)***	792,209	827,773	17,752	845,667	121,835	107	•

Phosphorus (P)		Dinnut	Statistical	P input in- cluding stat.	Evcondanco	Input 2015 including stat.	Classification
Baltic Sea sub-basin	MAI*	2015	2015	2015	of MAI	in % of MAI	reduction
Bothnian Bay (BOB)	2,675	2,574	149	2,724	49**	102	•
Bothnian Sea (BOS)	2,773	2,569	133	2,702		97	٠
Baltic Proper (BAP)	7,360	15,327	1,554	16,881	9,521	229	•
Gulf of Finland (GUF)	3,600	2,853	1,610	4,463	863**	124	•
Gulf of Riga (GUR)	2,020	2,372	227	2,599	579	129	•
Danish Straits (DS)	1,601	1,530	49	1,579		99	٠
Kattegat (KAT)	1,687	1,477	59	1,563		91	٠
Baltic Sea (BAS)***	21,716	30,026	1,229	31,255	9,539	144	۲

*) As adopted by the 2013 HELCOM Copenhagen Ministerial Meeting (HELCOM 2013a). **) Exceedance of MAI is caused by statistical data uncertainty. ***) Input to the Baltic Sea (BAS) assessed by separate statistical procedure and might deviate from the sum of inputs to sub-basins.



Figure 3. Total inputs of nitrogen to the Baltic Sea.



Figure 4.

Total inputs of phosphorous to the Baltic Sea.





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Progress toward national targets for nutrients input reduction

The assessment of progress is based on the evaluation of nutrient net input reduction towards the net nutrient input ceilings. Net inputs of nitrogen and phosphorus from each country to each sub-basin in 2014 were computed using the method of statistical estimation based on a time series of normalized net inputs for the period 1995–2014. The method also takes into account uncertainty of the estimation. A comparison of the estimated net inputs and corresponding nutrient input ceilings illustrates progress towards national input targets by 2014 (evaluation of fulfilment N&P Fig. 6, 7). The annual water flow variation is given in Figure 8.

Meeting of national nitrogen input ceilings

Denmark is the only country that has fulfilled nitrogen ceilings to all Baltic Sea sub-basins. Finland and Sweden met their nitrogen ceilings to all sub-basins except the Baltic Proper and the Gulf of Finland, where the missing reduction is less than 10% of the input ceilings for these countries. Russia exceeded national ceilings to all sub-basins. Atmospheric nitrogen inputs from Baltic Sea shipping and non-HELCOM countries exceeded their target values to all sub-basins. It is important to note that the total nitrogen inputs to the Bothnian Sea, Bothnian Bay, Danish Straits and Kattegat were below the MAIs for these sub-basins and the countries which exceeded national input ceilings for these sub-basins have only minor airborne contribution. Almost all countries demonstrated progress towards reduction targets, though the progress in some cases cannot be justified being within statistical uncertainty of the assessment data.

Country/basin	BOB	BOS	BAP	GUF	GUR	DS	КАТ
Denmark	\checkmark	\checkmark	\downarrow	4	\downarrow	\downarrow	\downarrow
Estonia	\checkmark	Ŷ	\downarrow			¥	
Finland			\downarrow		Ŷ	.↓	\downarrow
Germany	↓ ↓	¥	\downarrow	\checkmark	Ŷ	\downarrow	\downarrow
Latvia	¥	\checkmark				\downarrow	\checkmark
Lithuania		¥		\checkmark			
Poland	↓ ↓	¥	\downarrow	↓	V	¥	↓
Russia	1	Ŷ	\uparrow			4	
Sweden	\downarrow	\downarrow	\downarrow	4	\downarrow		\downarrow
Belarus							
Czech Republic							
Ukraine							
Baltic Sea shipping							
Other countries	V	¥	V	¥	\checkmark	V	V
MAI	\downarrow	\downarrow	\downarrow			\downarrow	\downarrow

Figure 6.

Total Nitrogen. Evaluation of input ceilings fulfillment. Based on statistically estimated inputs (see full legend below).

Country/basin	BOB	BOS	BAP	GUF	GUR	DS	КАТ
Denmark			\rightarrow				\downarrow
Estonia				\rightarrow			
Finland	\downarrow						
Germany							
Latvia							
Lithuania			\downarrow		\downarrow		
Poland							
Russia			\uparrow	\downarrow			
Sweden		\downarrow	\downarrow			\downarrow	
Belarus							
Czech Republic							
Ukraine			一 个				
Baltic Sea shipping							
Other countries							
MAI				\downarrow			\downarrow

Figure 7.

Total Phosphorus. Evaluation of input ceilings fulfillment. Based on statistically estimated inputs.



Arrows: statistically significant changes of nutrient inputs since the reference period, taking into account 95% confidence interval for both latest inputs and reference values.

- ψ = significant decrease
- Λ = significant increase

* Yellow, orange and red shades: input ceiling is exceeded with 95 % statistical certainty. The legend illustrates the percentage which reduction left to the target constitutes in the corresponding input ceiling value. "Other countries" includes sources for atmospheric nitrogen deposition as the 20 EU countries not being HELCOM Contracting Parties, countries outside EU including Belarus, Ukraine, North Sea shipping etc.



Daugava river in Latvia © Ivars

Meeting of national phosphorus input ceilings

There was not a single country reaching phosphorus input ceilings for all Baltic Sea sub-basins. All HELCOM countries and non-HELCOM countries with waterborne inputs exceeded input ceilings for the Baltic Proper. Two of three countries, which contribute to the input to the Gulf of Finland, also exceeded their ceilings. The fulfilment of the input ceiling by Russia cannot be justified at the moment due to high uncertainty caused by variability of the assessment data. This also holds true for the Russian phosphorus inputs to the Gulf of Riga. Latvia, Poland, the Czech Republic and Ukraine exceeded their ceilings to the sub-basins to which they have inputs. All countries met national ceilings for total phosphorus inputs to the Danish Straits and Kattegat.

National net input ceilings

Net input of nutrients is an estimate of the amount of nutrients ending up in the Baltic Sea sub-basins coming from a particular country. The calculations integrate waterborne (direct coastal point sources and discharges from rivers), airborne (atmospheric deposition from a particular country or a group of countries) and transboundary (input via rivers through another country).

National net nutrient input ceiling is the maximum allowable amount of nutrients released from a country to a sub-basin, assuring the good environmental status of the Baltic Sea in terms of eutrophication. The sum of input ceilings set for all countries to a specific sub-basin is equal to the Maximum Allowable Input for that sub-basin.

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Internal biogeochemical cycles of nutrients

In addition to the supply of nutrients from land and via atmospheric deposition, there are nutrient sources driven by the internal biogeochemical cycles within the Baltic Sea. A large volume of the nitrogen load (400,000-600,000 t/year) originates from N fixation by cyanobacteria transforming dinitrogen (N2) of the air to bioavailable ammonia. The magnitude of this N fixation grows with the magnitude of cyanobacteria blooms. Denitrification is a microbially facilitated process where nitrate (NO3–) is reduced and ultimately produces molecular nitrogen (N2) completing the nitrogen cycle. Denitrification in water column and sediments plays important role in the Baltic Sea nitrogen budget.

Huge resources of phosphorus have been accumulated in the bottom sediments of the Baltic Sea during the past decades. Internal load, caused by release of phosphorus from sediments in anoxic conditions, is a factor contributing to its elevated concentration in the Baltic Sea.

Algal bloom in the Baltic Sea captured by the Sentinel-2A satellite. © Copernicus Sentinel/ESA, CC BY-SA 3.0

Terms used in PLC

ΤN

TN – total nitrogen

TP

TP - total phosphorus

Sub-basin division units of the Baltic Sea used for the HELCOM assessment of input of nutrients:

- Kattegat (KAT)
- Danish Straights (DS)
- Baltic Proper (BAP)
- Gulf of Riga (GUR)
 Gulf of Finland (GUF)
- Bothnian Sea (BOS)
- Bothnian Bay (BOB)
- Entire Baltic Sea (BAS)

Baltic Sea Shipping

Maritime traffic taking place in the Baltic Sea.

Other countries

Other countries (as referred in Fig. 6 and 7) – include sources for atmospheric nitrogen deposition as the 20 EU countries not being HELCOM countries, countries outside EU including Belarus, Ukraine, etc.

Riverine input

The amount of a substance carried to the maritime area by a watercourse (natural or man-made) per unit of time.

Direct input

Point sources (municipal waste water treatment plants (MWWTP), industrial plants and aquaculture plants) discharging (defined by location of the outlet) directly to the sea.

Pathways of nutrients into the Baltic Sea

Nitrogen reaches the Baltic Sea via three major pathways. It can be transported by rivers (riverine input), deposited from air (airborne input) and discharged directly to the Baltic Sea from various industrial or municipal waste water treatment facilities located on the sea coast. Phosphorus can be released through the same pathways. The difference is that deposition of phosphorus from air is caused mainly by natural factors and for the HELCOM assessment considered as constant.

The riverine input is a dominating pathway for both nutrients in 2014 constituting of almost 70 % of total nitrogen and 89 % of total phosphorus input to the Baltic Sea (Fig. 8, 9). However, the share of pathways differs between sub-basins. The contribution of airborne deposition for nitrogen varies from almost 12% in the Gulf of Riga up to more than 40% in the Danish Straights, and for phosphorus from less than 4% to the Gulf of Finland to more than 15% to the Bothnian Sea. The proportion of direct input of nitrogen deviates from around 1% to the Gulf of Riga and Baltic Proper to more than 8% to the Gulf of Finland. The role of the direct point source of phosphorus also varies between sub-basins from almost 3 % in the Gulf of Riga and Baltic Proper to nearly 20% in the Danish Straights. Also national net inputs show a large variation of proportions between the pathways.

Changes in the main pathways of nitrogen and phosphorus input

The total average inputs for both phosphorus and nitrogen for the entire Baltic Sea have reduced over time. However, the reduction achieved in 2014 since 1995 was not equal for different pathways. The highest reduction was achieved for direct sources. The proportion of the direct source was reduced from 5.5 % to 3.8 % for nitrogen and from almost 11 % to almost 6 % for the phosphorus input. The proportion of airborne input of nitrogen also slightly decreased while the proportion of riverine input of both nutrients increased remarkably.

The pattern illustrating the proportion of different pathways in the total nutrient load differs for the different sub-basins and countries. However, the general tendency of the total nitrogen and phosphorus inputs is a decrease of the share of direct point sources experiencing greatest reduction since 1995.

This pattern is common for the nitrogen input to all sub-basins in the southern part of the Baltic Sea (the Baltic Proper, Danish Straits, and Kattegat) as well as to the Gulf of Finland. The reductions correspond to falling of nitrogen input from the countries responsible for the main nitrogen inputs to these basins. There is also a similar tendency for the total nitrogen inputs to the Gulf of Riga, although there is a large variability in total inputs over time.

The main tendencies of the phosphorus input change reveal that there is a decrease in the total phosphorus inputs since 1995. Especially, it concerns the reduction of proportion of direct point sources. The highest drop down of the share of direct sources was observed in the Gulf of Riga, although this is the only basin without any obvious reduction of total P input. Most countries show a decrease in their share of direct point sources over time except Sweden, which is the only country without any clear trend in changed pathways. This is due to the fact that Sweden together with Denmark, Finland, and Germany started to reduce the emissions from waste water treatment plants before HELCOM measurements began, which imply that these reductions cannot be seen in the HELCOM data. In these countries, measures are now more oriented to diffuse sources such as agricultural losses and scattered dwellings.

Figure 8. Total Nitrogen input to the Baltic Sea.

Figure 9. Total Phosphorus input to the Baltic Sea.

Sources of riverine nutrient loads in the Baltic Sea area

Natural background loads of nitrogen and phosphorus are one third of the total load of nutrients to the Baltic Sea (riverine loads, Fig. 10, 11). However, there are large differences for the different basins. The largest proportion is for the Gulf of Finland (68% for nitrogen, and 59% for phosphorus), and Bothnian Bay (65% for both nitrogen and phosphorus), whereas the lowest proportions are in the Gulf of Riga (12% and 11%, respectively). The anthropogenic diffuse sources (mainly from agricultural activities) make up 46% of the total riverine nitrogen load and 36% of the total riverine phosphorus load to the Baltic Sea. Large differences in the amount of agriculture land are reflected by varying contributions over the Sea area. High impact is found in the Gulf of Riga (57%) for nitrogen, and 42% for phosphorus), and for nitrogen also in the Danish Straits (68%), and Kattegat (59%). Point sources constitute 12% of the total nitrogen load and 24 % of the total phosphorus load. The variability in the importance of point sources is even greater than of the other nutrient sources. A high impact from point sources is found in the Baltic Proper (18% for nitrogen, 33% for phosphorus), and in the Danish Straits (43% for phosphorus). However, it has not been possible to allocate any specific sources to a large proportion of the nutrient load classified as transboundary as they originate in upstream countries, mainly Belarus. For example, the transboundary load to the Gulf of Riga is estimated to be 30 % for nitrogen, and 42% for phosphorus.

Input of nutrients by 7 biggest rivers

The seven biggest rivers of the Baltic Sea (Göta, Kemi, Daugava, Nemunas, Oder, Vistula and Neva) cover 50% of the Baltic Sea catchment area and are inhabited by over 50 million people. There-fore, the load carried by these rivers constitutes an essential part of the waterborne inputs of nu-trients to the Baltic Sea and intensive monitoring of water quality in these rivers is one of the key factors of a reliable evaluation of possible changes in loading. In 2014 the seven rivers exported 227,000 t TN and 15,300 t TP into the Baltic Sea, which was 29 % of the TN input of the Baltic Sea and 46 % of the respective TP input. The anthropogenic pressure is highest in the southern catchments with the most dense population and intensive agricultural activities. Accordingly, the nutrient loads are high in the south: the area specific TN load of the Vistula River in 2014 was 341 kg/km², whereas the respective load of the Kemijoki River was 111 kg/km². The variation in the area specific TP export was even larger: the River Kemijoki 5.1 kg/km² and the River Vistula 42 kg/km². The Neva River contributed with over 40 % of the total flow of the seven biggest rivers, but the Vistula had the highest TN and TP loads: it contributed with 26 % to the TN load and 52 % to the TP load. Both TN and TP loads showed a statistically significant decrease from 1995 to 2014. The TN load was reduced by nearly 81,000 t (21 %) and the TP load by 4,600 t (23 %), but the trends of individual rivers varied greatly.

Phosphorous riverine loads.

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Figure 12. Area specific loads kg/km-2 (load/ area) of total nitrogen and total phosphorus from the catchments or sub-catchment of the seven biggest rivers (2014).

Figure 13.

Proportions of the total flow, normalized total nitrogen load and normalized total phosphorus load for the seven biggest rivers of the Baltic Sea catchment area in 2014.

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The evaluation of the effectiveness of measures to reduce the input of nutrients to the Baltic Sea is one of the additional PLC-6 project deliverables. The evaluation is based on the countries' responses to the questionnaire prepared by the PLC-6 project team.

The Contracting Parties were asked to evaluate the load reductions that took place between the years 1995 and 2014 and also to estimate sector-wise potential reduction of nitrogen and phosphorus from 2015 onwards. National estimates of the reduction effects were mainly based on measures included in the River Basin Management Plans (RBMPs) under WFD, national water protection programmes as well as the effects of the implementation of the relevant EU directives, e.g. Programme of Measures (PoMs) for the development and implementation of the EU Marine Strategy Framework Directive (MSFD) and Urban Waste Water Directive (UWWB).

The questionnaire revealed a very high variability of the reported estimates between countries caused by various non-harmonized assessment methods and a remarkable difference in both temporal, geographical and sectoral data coverage. In general, these data deficiency and inconsistency impeded the quantification of the effects of measures across the region providing information only for a narrative description of the undertaken and planned measures. However, the work resulted in a comprehensive overview of the measures to reduce nutrient loads in the region and laid a basis for further analysis and data specification, paving the way for the assessment within the PLC-7 project.

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Inputs of hazardous substances to the Baltic Sea

Inputs of Cadmium, Mercury and Lead to the Baltic Sea

There are quite large differences in the estimated total amounts of metals that enter the Baltic Sea every year. The main route of entry is also quite variable between metals. In total, it is estimated that the inputs of cadmium, mercury, and lead to the Baltic Sea 2012-2014 have been in the range of 23-42, 4.7-5.3, and 441-550 tonnes per year respectively. Mercury is mainly entering the Baltic Sea via atmospheric deposition (about 70% of the total inputs). For lead and cadmium, the riverine inputs are most important (64%, and 79% respectively). In all cases, the direct point sources make the smallest contribution to the total inputs (4% of the mercury inputs, <1% for cadmium and lead).

Atmospheric deposition of some selected organic pollutants.

The modelled atmospheric deposition of PBDEs, PCBs, PCDD/Fs shows a continuously decreasing trend over time. On the other hand, the Benzo(a)pyrene deposition seems to have reached a steady state where no visible decrease seems to occur any longer.

Pharmaceutical residues

In a first attempt to get an overview of the inputs of pharmaceuticals to the freshwater and marine environment in the Baltic Sea region, as well as to estimate the contamination of the marine environment, a rough estimate showed that the annual release from the main source MWWTPs to the environment is about 1.8 thousand tonnes of pharmaceutical residues. Only nine out of the 118 assessed pharmaceuticals were removed with an efficiency over 95 % in the MWWTPs and almost half of the compounds were removed with an efficiency less than 50 %.

Upcoming work on the input of hazardous substances to the Baltic Sea.

The work on hazardous substances within the Pollution Load Compilations will be intensified in future data reporting and assessments. Based on the outcome of the questionnaire, the first data collection and assessment of the inputs of organic pollutants is suggested to include nonylphenols, octylphenols, and PFAS. These substances were identified as generally having a high concern, and the stated data availability appears to be comparatively good for all three groups of pollutants.

Figure 14. Top 20 pharmaceuticals detected in WWTPs' effluents.