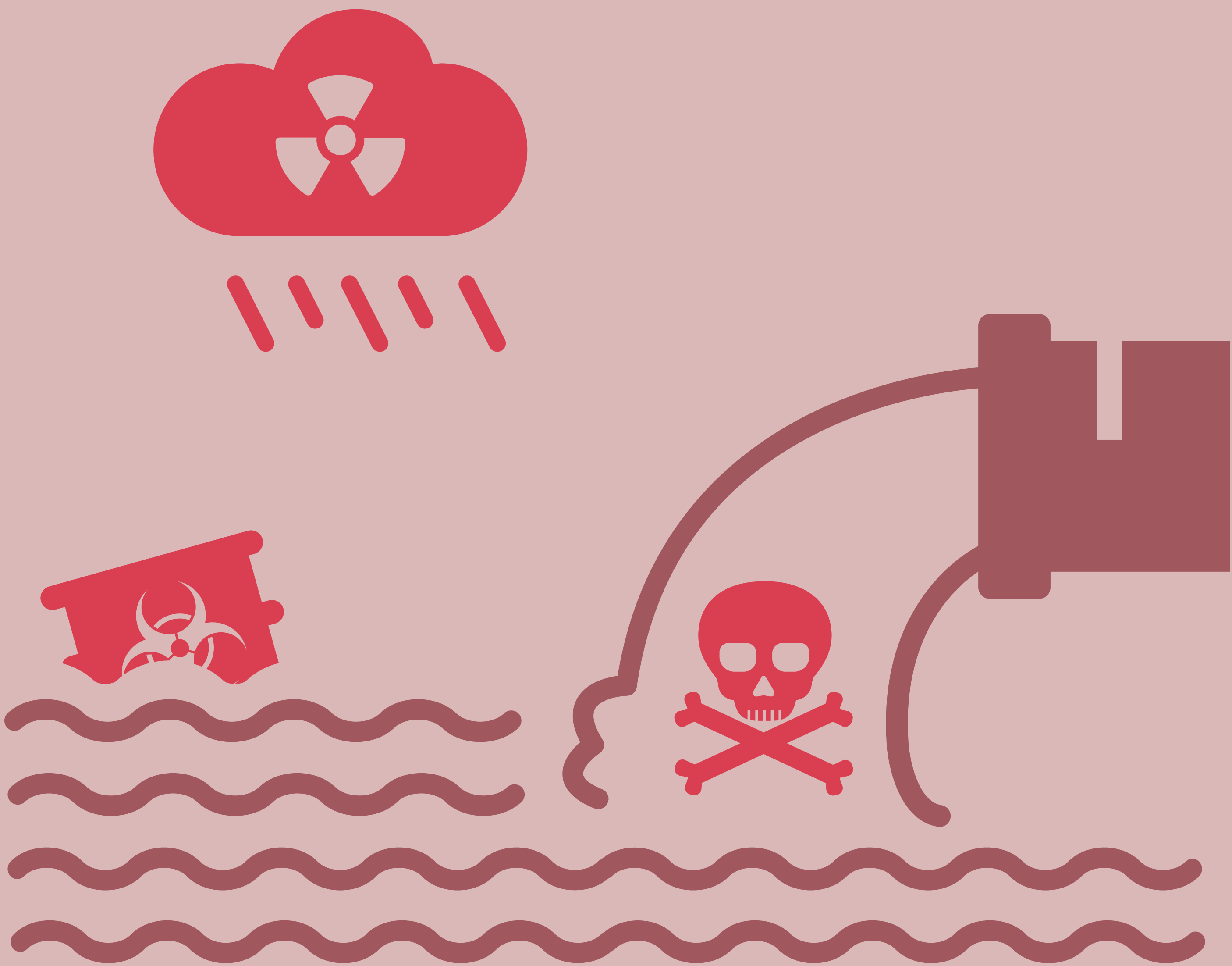
Inputs of hazardous substances to the Baltic Sea

Baltic Marine Environment Protection Commission

Hazardous substances

BSEP n°179



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1. Introduction

Hazardous substances like metals and organic substances may originate from natural or anthropogenic sources, although organic contaminants tend to be more commonly of anthropogenic origin. Accumulation of contaminants in the environment may lead to risk for biota including a risk for human health. The inputs are considered to be mainly waterborne via rivers and direct point sources, and via atmospheric deposition, depending on the substance and data availability.

The monitoring and reporting guidelines for waterborne inputs of hazardous substances to the Baltic Sea (PLC-Water guidelines) are to a large degree focused on metal inputs, whereas in the programme for monitoring air pollution and precipitation, the airborne inputs include both metals and some organic contaminants. Due to this inconsistency between the monitoring and reporting of the different sources, fair estimates for the total inputs are at the moment only possible from some countries for some metals that are included in both programmes. The inputs of these metals are given below. In addition, an assessment of a specific HELCOM data call on nonyl- and octylphenols, as well as per- and polyfluoroalkyl substances (PFASs) in rivers and coastal waters, as well as summarised information on the atmospheric deposition of some selected organic contaminants based on EMEP work for HELCOM are included.

2. Inputs of heavy metals to the Baltic Sea

Mercury, cadmium and lead are the present HELCOM indicators on metal pollution in the Baltic Sea, and high levels of these metals have been detected in sediments and in fish tissue (HELCOM 2018a). Soil properties, industrial activity, high population density, the exploitation of minerals and other natural resources, the application of fertilizers in agricultural areas as well as atmospheric deposition from local and distant emission sources are the main factors that contribute to heavy metal inputs.

Limitations in national monitoring programmes and/or lack of proper laboratory resources have in some cases prevented the reporting of heavy metal input data. As a result, only an indication on the inputs of mercury, cadmium and lead entering the Baltic Sea could be established in PLC-6, and still issues exist regarding reporting completeness for some CPs, as well as data quality issues and the possibilities to quantify the metals at ambient level. The results from the PLC-7 reporting ought to be seen as mainly indicative (cf. “Data handling and quality control”). In addition to the data assessed in PLC-6, the present assessment also includes chromium, copper, nickel, and zinc. Unfortunately, no data is available on the atmospheric deposition for these metals, as they have, at least so far, not been included in the commission to EMEP regarding modelling of metal deposition on the Baltic Sea. Also, it should be noted that in case where there are upstream countries, the transboundary metal loads are included in the metal inputs to the Baltic Sea from the HELCOM CPs that encompasses the river mouth as it has not been possible to correct for these upstream inputs.

According to the PLC-Water guidelines, mercury, cadmium, and lead are mandatory parameters that should be reported wherever concentrations in rivers are not below the recommended quantification limit, whereas copper, zinc, nickel, and chromium may be reported on a voluntary basis. The request is on the total load of the named metals, although most CPs are analysing on filtered samples (cf. “Data handling and quality control”). The PLC-Water guidelines indicate methods for making estimates from measurements below the quantification limits (HELCOM 2019a). The reporting obligations for MWWTPs and industrial point sources are on the other hand regulated by the size of the MWWTPs and if the monitoring is a part of the permissions for a specific industrial plant. Due to the size regulation for the WWTPs, the inputs from smaller facilities are most probably underestimated as they often are not obliged to report metals.

2.1. Data handling and quality control

Metal data have been reported by the Contracting Parties within the framework of the annual Pollution Load Compilations. The reported data has been compiled and assessed as far as possible, and the HELCOM Contracting Parties have been asked to verify their data, especially suspicious outliers, and to fill in potential data gaps. Anyhow, there still remain plentiful issues regarding the temporal and spatial coverage for several CPs. Also, it has been challenging to assure the data quality, as some observations appear to be suspiciously high or low in comparison to observations in time-series from a single CP or compared to inputs from other CPs.

2.2. Data coverage

The assessment of heavy metal inputs to the Baltic Sea has focused on the period 2015-2017 as these are the recent years with the most complete data coverage in the HELCOM PLC database. It is mandatory to report cadmium, mercury and lead inputs, whereas chromium, copper, nickel and zinc are voluntary to report. However, the spatial coverage is far from complete and varies between the metals (Figure 1). The very limited data

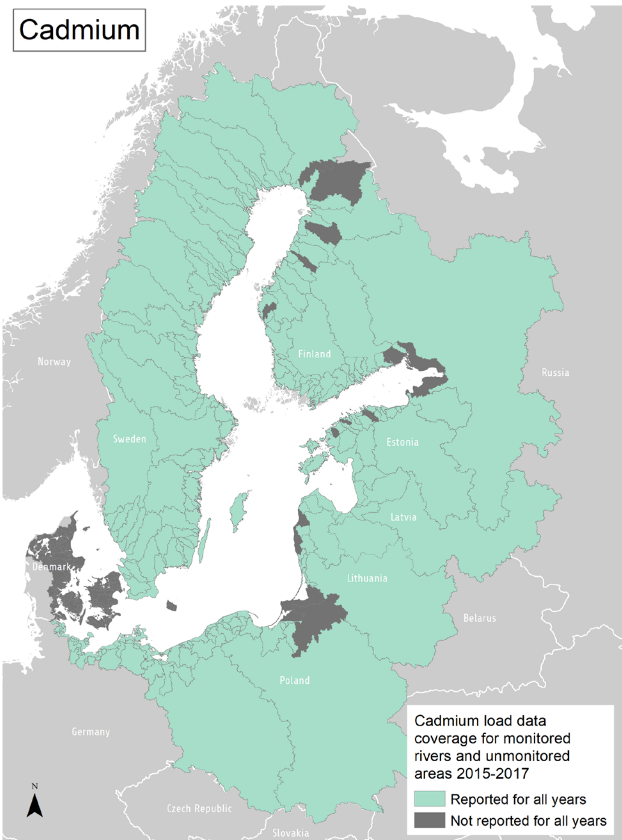


Figure 1. The spatial data coverage 2015–2017 of reported riverine inputs of mandatory metals Cd, Hg and Pb, and voluntary metals Cr, Cu, Ni, Zn

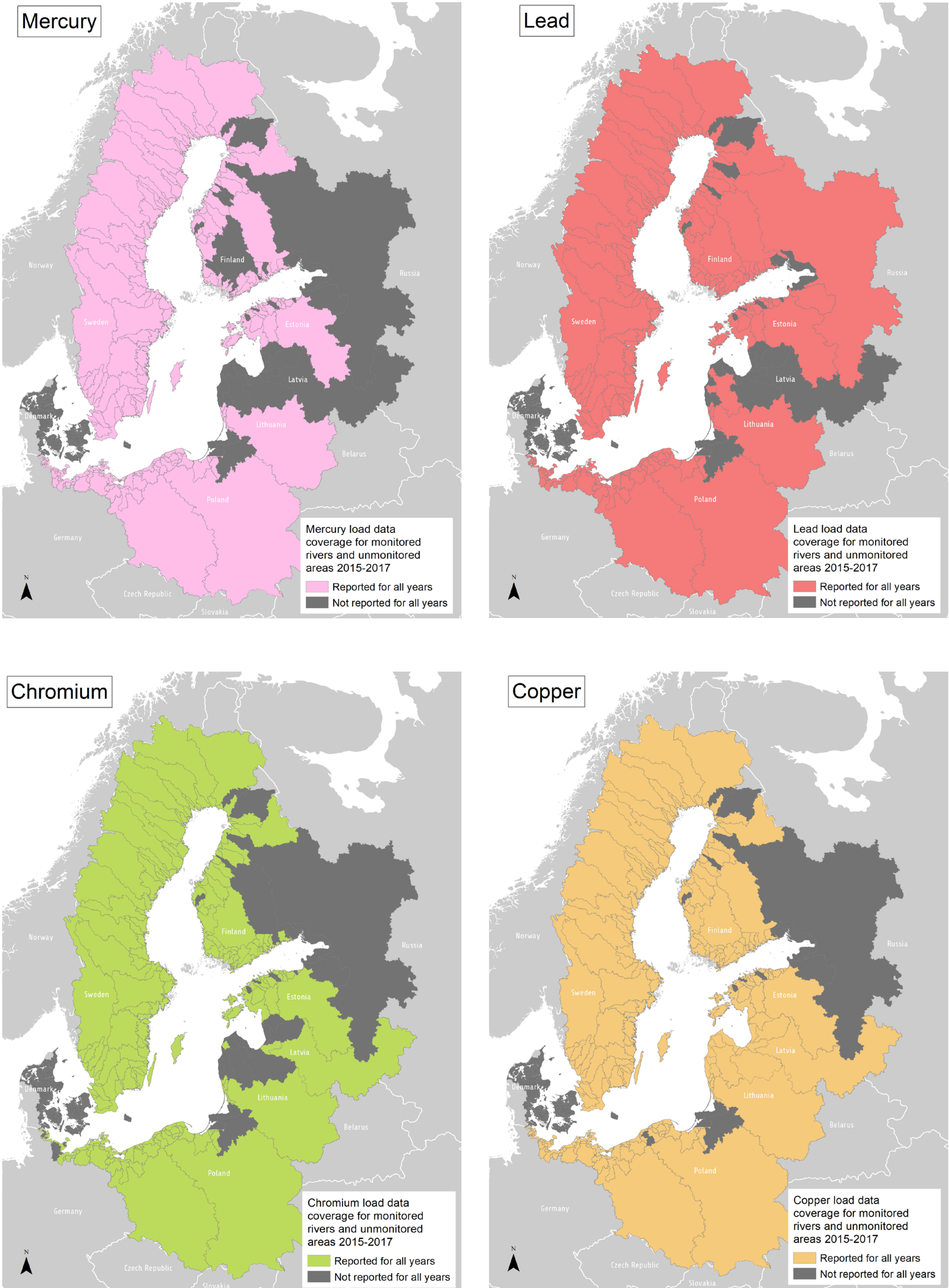


Figure 1. (continued) The spatial data coverage 2015–2017 of reported riverine inputs of mandatory metals Cd, Hg and Pb, and voluntary metals Cr, Cu, Ni, Zn

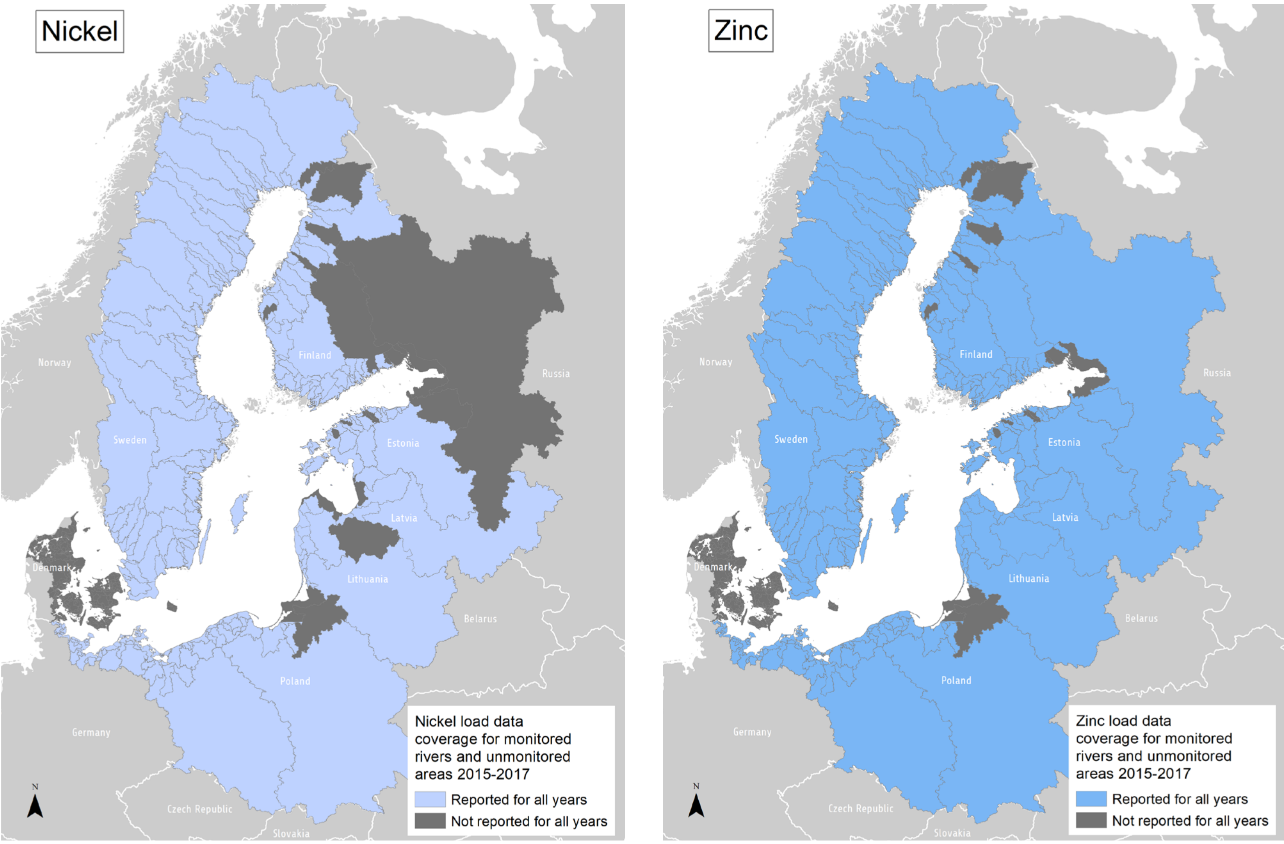


Figure 1. (continued) The spatial data coverage 2015-2017 of reported riverine inputs of mandatory metals Cd, Hg and Pb, and voluntary metals Cr, Cu, Ni, Zn

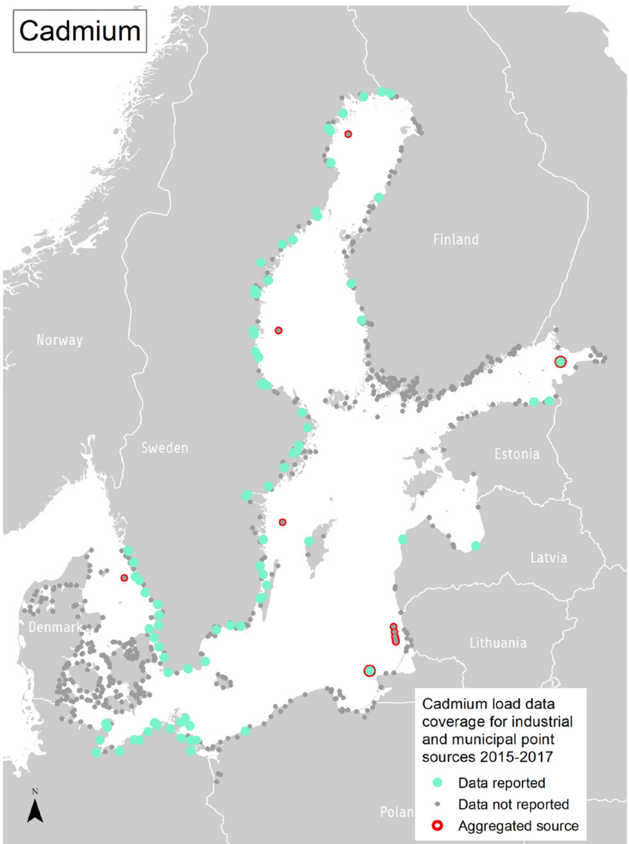


Figure 2. The spatial data coverage 2015-2017 of reported inputs from direct point sources of the mandatory metals Cd, Hg and Pb, and the voluntary metals, Cr, Cu, Ni and Zn. Aggregated point sources are indicated by a red border around the points. The aggregated sources are only positioned on the maps within respective sea basin as the number and location of individual sources are not known.

on riverine inputs from Denmark has not been included due to very poor coverage. The coverage of heavy metal inputs from direct point sources is certainly not fully covered. For instance, Sweden only can report metal inputs from the larger MWWTs, as the smaller plants seldom have reporting obligations on metals in their permits. Maps with the positions of MWWTs and industrial point sources with reported metal loads are given in Figure 2.

In addition, most CPs are analysing the metals on filtered riverine samples although the request according to the PLC-Water Guidelines are on the total loads. This is mainly an adaptation to the Water Framework Directive that requests data on biologically available metals. Among the HELCOM countries, only Finland is actually measuring the total metal concentration (EE analyses both total and filtered metals, but report dissolved metals), whereas Sweden is analysing acid soluble metals that include dissolved metals and metals adsorbed to particulate matter. Except for Lithuania, all other CPs are analysing filtered samples. In Lithuania Cd, Hg, Pb, and Ni are analysed on filtered samples, whereas total concentrations are analysed for Cr, Cu, and Zn. Consequently, the data reported and assessed are in regard to the total metal inputs in most cases an underestimate as the metals associated to particulate matter are not included. The underestimation is higher the higher fraction of a transported metal that is normally particle bound. Hence, due to the very high particle affinity for especially lead, the inputs of these metals are most probably prone to be seriously underestimated.
In the quality control of the reported metal data comparisons have mainly been performed in time series for the total metal inputs for a Contracting Party regarding annual riverine data, as well as on data from direct point-sources such as municipal waste water

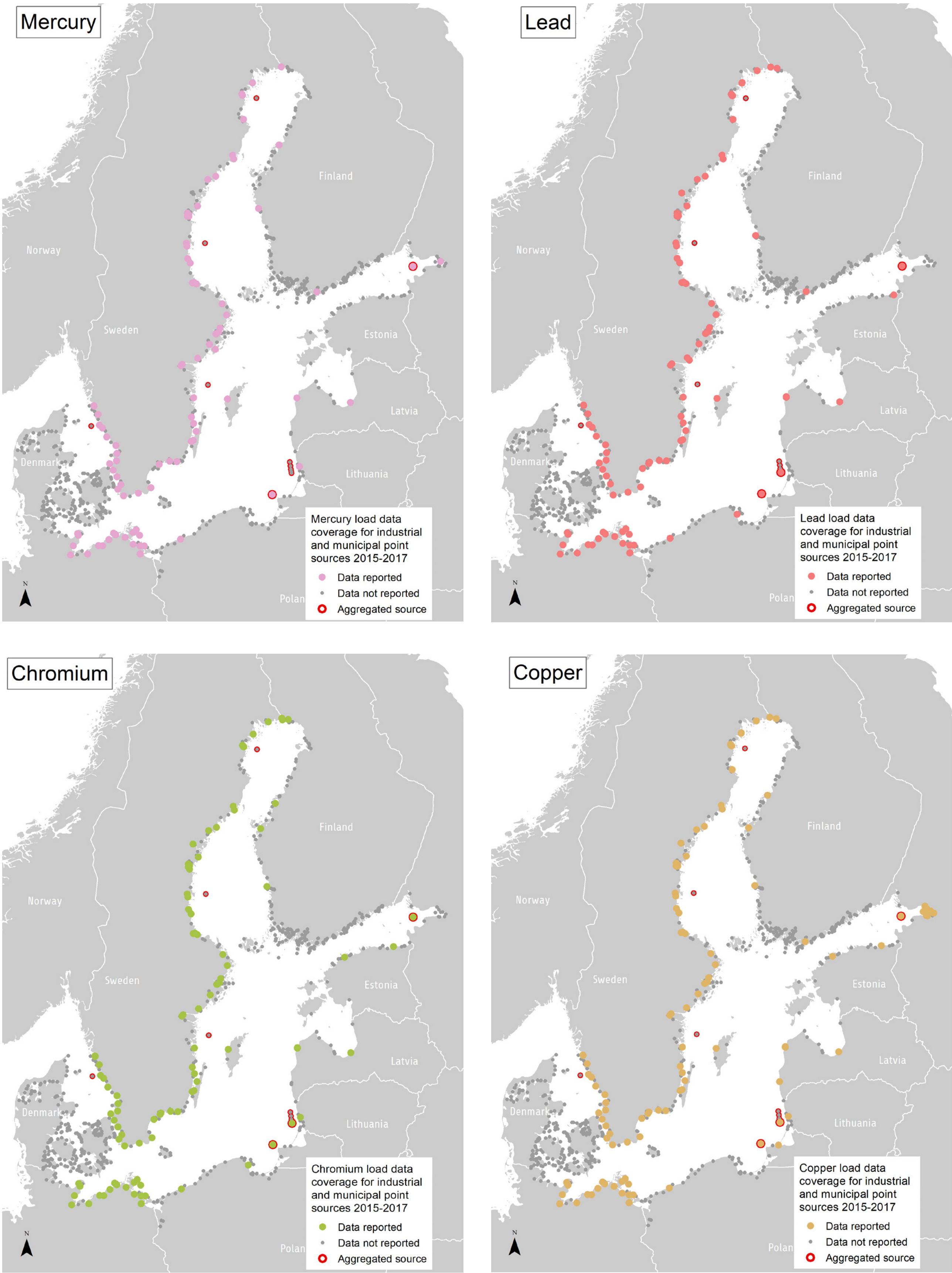


Figure 2. (continued) The spatial data coverage 2015–2017 of reported inputs from direct point sources of the mandatory metals Cd, Hg and Pb, and the voluntary metals, Cr, Cu, Ni and Zn. Aggregated point sources are indicated by a red border around the points. The aggregated sources are only positioned on the maps within respective sea basin as the number and location of individual sources are not known.

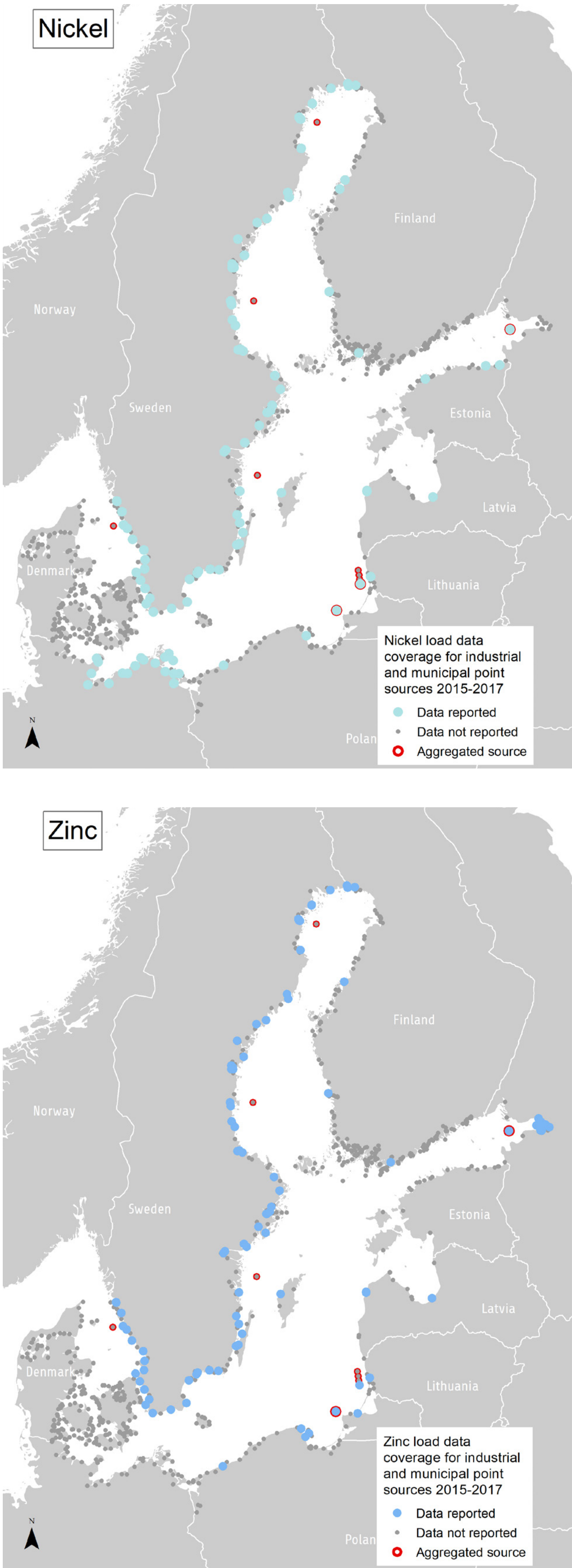


Figure 2. (continued) The spatial data coverage 2015–2017 of reported inputs from direct point sources of the mandatory metals Cd, Hg and Pb, and the voluntary metals, Cr, Cu, Ni and Zn. Aggregated point sources are indicated by a red border around the points. The aggregated sources are only positioned on the maps within respective sea basin as the number and location of individual sources are not known.

treatment plants and industrial facilities. Potential high or low outliers have subjectively been noted, and the CPs have been notified and asked to verify the outliers. Atypical values may be caused by natural causes, like large variation in water flow due to drought or flooding events, or by anomalies in the data handling or in the laboratory analysis. Loads that are considerably higher than expected might result from estimates based on contaminated samples. In the metal input assessment within PLC-6 the flow-normalised riverine metal inputs were used as a tool to reveal inconsistencies (cf. HELCOM 2018b). Unfortunately, due to problems to link the metal inputs to corresponding annual mean water discharges, this quality assurance procedure could not be used in the present assessment.

A special issue in making reliable input estimates is using too high limits of quantification in the laboratory analysis of samples. In these cases, the estimates will be extra sensitive if the quite common procedure to use LOQ/2 to estimate levels below the limit is applied. That is especially problematic if the estimate is based on a large proportion of observations with levels below the limit. A more realistic weighted approach is recommended in the PLC- Water Guidelines (HELCOM 2019a), but this procedure is not always used by CPs. However, this procedure may also create unreliable input estimates when a large part of the observations is below the LOQ. For instance, the Estonian riverine inputs are most probably affected by this computational challenge.

In addition to estimates based on various LOQs and differences in estimating using data below the LOQs, there is also some differences in which metal fraction the CPs are analysing their riverine samples. Most CPs are only analysing metals on filtered river samples. These samples are of course not completely comparable with the total concentrations reported (as required in the PLC-Water Guidelines) by other CPs. Also, Denmark has only been monitoring the riverine metal loads in a total of twelve of their numerous small rivers in the period 2012-2014 (of which only eight for Hg), and no river was monitored for more than a single year. Hence, the Danish riverine inputs are not included in this assessment, since that would result in serious underestimations of the total metal inputs to the Baltic Sea.

Calculations of the area-specific metal inputs to the Baltic Sea reveal that in general there is a quite good agreement between the inputs from the different countries, although there are some suspiciously low inputs that most probably are not correct (Table 2). The Russian inputs of cadmium and lead are most certainly over-estimated due to many observations below the comparably high LOQs. Since these input estimates are based on half the LOQs, the reported inputs are therefore consequently deemed to be over-estimated. Also, as the input data include possible transboundary inputs from upstream countries. Hence, data for individual countries ought to be handled by great care.

Recent metal input data are generally believed to give better estimates of the inputs, as the data coverage is in general better compared to earlier years, but also the data quality appears to be better. In spite of this, there are still some concerns about specific estimates, but the data have been quality assured by the CPs and verified as correct. However, due to these questionable input estimates, some data has been overlooked in the assessment (esp. zero riverine inputs), mainly due to inconsistent reporting coverage. To exclude zero inputs is justified by problems to distinguish between "real" zero inputs and lack of data/information. In addition, as actual metal inputs of these widespread naturally occurring metals never can be zero in reality, but rather reflect insufficient analytical capabilities, and the fact that including zeros in averaging would cause severe underestimates of the inputs, these data have been overlooked in the assessment. Also, in the compilation of metal inputs to the different Baltic Sea basins, it has not been possible to get a full coverage for some of the southernmost basins, mainly due to the limited riverine load data from Denmark. In this assessment, the annual average metal inputs for the period 2015-2017 is judged to be most complete and appears to have the least questionable annual data, as it seems that in general there still need to be some verifications for the annual inputs in 2018. However, for comparison both the annual average inputs for 2015-2017, and 2016-2018 are given in the compiled data tables.

Table 1. Limits of quantification (LOQ) for metals in river water (µg/l). Data for Contracting Parties from PLC 5.5 (HELCOM 2015) or later, and the recommended LOQs from PLC-Water Guideline (HELCOM 2019a).

Metal	Guideline	DE	DK	EE	FI	LT	LV	PL	RU	SE
Cd	0.01	0.02-0.06	0.012	0.02-0.05	0.01	0.05	0.024	0.1	0.1	0.004
Cr	0.05	0.1-0.2	0.03	0.5-1	0.2	0.5	0.8	1.0	1	0.03
Cu	0.1	0.08-0.5	0.09-0.12	1	0.1	0.5	0.9	1.0	1	0.01
Hg	0.005	0.001-0.005	n.a.	0.015-0.1	0.002	0.03	0.01	0.013	0.01	0.0001
Ni	0.05	0.07-0.5	0.09	0.1-1	0.2	1.0	2	1.0	5	0.02
Pb	0.05	0.04-0.2	0.075	0.1-1	0.01	1.0	1.0	1.0	2	0.01
Zn	0.5	0.2-0.5	0.09	1-2	1	5.0	3	1.0	2	0.4

Table 2. Area-specific riverine inputs of cadmium, chromium, cupper, mercury, nickel, lead, and zinc from HELCOM Contracting Parties to the Baltic Sea, as well as the area covered by the estimated inputs, and the coverage of the total area of the specific country. The inputs include possible transboundary inputs from upstream countries, and consequently data for individual countries ought to be handled by care.

CP	Cd (kg/km2)							Area (km2)	Coverage (%)
	2012	2013	2014	2015	2016	2017	2018		
DE	0.0048	0.0043	0.0014	0.0062	0.0025	0.0040	0.0034	23276	81
DK									
EE	0.0197	0.0010	0.0008	0.0008	0.0002	0.0079	0.0005	46329	100
FI	0.0086	0.0056	0.0061	0.0086	0.0055	0.0052	0.0040	316941	100
LT			0.0003					47349	73
LV	0.0001	0.0002	0.0038	0.0061	0.0102	0.0030	0.0066	65874	100
PL	0.0009	0.0009	0.0022	0.0023	0.0015	0.0022	0.0020	304801	98
RU	0.0820	0.0490	0.0290	0.0570	0.0390	0.0410	0.0370	351023-381507	93
SE	0.0054	0.0033	0.0037	0.0048	0.0033	0.0039	0.0039	454259	100

Table 2. (continued) Area-specific riverine inputs of cadmium, chromium, cupper, mercury, nickel, lead, and zinc from HELCOM Contracting Parties to the Baltic Sea, as well as the area covered by the estimated inputs, and the coverage of the total area of the specific country. The inputs include possible transboundary inputs from upstream countries, and consequently data for individual countries ought to be handled by care.

CP				**Cr (kg/km2)**				**Area (km2)**	**Coverage (%)**
	2012	2013	2014	2015	2016	2017	2018		
DE	0.074	0.049	0.007	0.089	0.043	0.071	0.046	23276	81
DK									
EE	0.116	0.01	0.007	0.018	0.001	0.023	0.009	46329	100
FI	0.401	0.293	0.247	0.396	0.229	0.215	0.185	316941	100
LT	0.163	0.170		0.234	0.074	0.271	0.194	47349	73
LV	0.002	0.003	0.489	0.079	0.155	0.090	0.018	65874	100
PL	0.630			0.067	0.088	0.045	0.014	304801	98
RU									
SE	0.178	0.083	0.096	0.102	0.072	0.084	0.096	454259	100

CP				**Cu (kg/km2)**				**Area (km2)**	**Coverage (%)**
	2012	2013	2014	2015	2016	2017	2018		
DE	0.403	0.305	0.134	0.580	0.223	0.328	0.371	23276	81
DK									
EE	1.826	0.721	0.197	0.501	0.716	0.631	0.200	46329	100
FI	0.820	0.547	0.463	0.689	0.494	0.451	0.377	316941	100
LT	0.446	0.658	0.001	0.486	0.402	0.803	0.972	47349	73
LV	0.011	0.009	0.456	0.643	0.871	0.993	0.659	65874	100
PL	0.486	0.369	0.257	0.132	0.177	0.253	0.183	304801	98
RU	0.598	0.623		1.145	0.901			351023	
SE	0.677	0.451	0.506	0.487	0.383	0.378	0.315	454259	100

CP				**Hg (kg/km2)**				**Area (km2)**	**Coverage (%)**
	2012	2013	2014	2015	2016	2017	2018		
DE	0.0008	0.0014	0.0001	0.0023	0.0004	0.0005	0.0005	23276	81
DK									
EE	0.0037	0.0009	0.0008	0.0136	0.0019	0.0020	0.0006	46329	100
FI	0.0015	0.0010	0.0011	0.0009	0.0006	0.0006	0.0005	316941	100
LT		0.0010		0.0008	0.0006	0.0023		47349	73
LV	0.0001	0.0002	0.0001	0.0001	0.0001	0.0097	0.0141	65874	100
PL	0.0010	0.0023	0.0004	0.0013	0.0001	0.0001	0.0003	304801	98
RU			0.0007		0.0720	0.0001		9584 (2014) 15500 (2015) 30484 (2016) 5400 (2017)	
SE	0.0012	0.0007	0.0007	0.0010	0.0007	0.0007	0.0007	454259	100

Table 2. (continued) Area-specific riverine inputs of cadmium, chromium, cupper, mercury, nickel, lead, and zinc from HELCOM Contracting Parties to the Baltic Sea, as well as the area covered by the estimated inputs, and the coverage of the total area of the specific country. The inputs include possible transboundary inputs from upstream countries, and consequently data for individual countries ought to be handled by care.

CP	Ni (kg/km2)							Area (km2)	Coverage (%)
	2012	2013	2014	2015	2016	2017	2018		
DE	0.183	0.132	0.050	0.615	0.182	0.276	0.360	23276	81
DK									
EE	0.504	0.158	0.113	0.156	0.284	0.242	0.134	46329	100
FI	1.024	0.653	0.734	1.099	0.783	0.734	0.533	316941	100
LT	0.212	0.338		0.296	0.338	0.444	0.275	47349	73
LV	0.004	0.007	0.142	0.266	0.008	0.009	0.006	65874	100
PL	0.112	0.273	0.178	0.171	0.184	0.257	0.171	304801	98
RU	0.435	0.675	-	0.526	0.157			350400	
SE	0.387	0.228	0.250	0.279	0.222	0.216	0.230	454259	100

CP	Pb (kg/km2)							Area (km2)	Coverage (%)
	2012	2013	2014	2015	2016	2017	2018		
DE	0.080	0.042	0.015	0.019	0.018	0.035	0.033	23276	81
DK									
EE	0.183	0.041	0.017	0.045	0.019	0.04	0.008	46329	100
FI	0.132	0.087	0.077	0.125	0.080	0.073	0.064	316941	100
LT		0.002	0.007	0.486	0.190	0.486	0.006	47349	73
LV	0.003	0.002	0.123	0.077	0.489	0.619	0.337	65874	100
PL	0.033	0.015	0.004	0.030	0.005	0.008	0.008	304801	98
RU	0.633	0.679	-	0.329	0.326	0.148	-	351023-381507	
SE	0.137	0.064	0.077	0.082	0.073	0.071	0.069	454259	100

CP	Zn (kg/km2)							Area (km2)	Coverage (%)
	2012	2013	2014	2015	2016	2017	2018		
DE	1.24	0.92	0.27	3.79	0.52	0.91	0.73	23276	81
DK									
EE	2.77	0.41	0.25	2.61	4.40	1.40	0.32	46329	100
FI	2.60	1.55	1.81	2.65	1.83	1.61	1.35	316941	100
LT	2.22	2.63		4.58	0.76	1.61	9.19	47349	73
LV	0.04	0.03	0.53	1.05	0.76	0.96	0.92	65874	100
PL	0.41	0.39	0.08	0.47	0.2	0.28	0.17	304801	98
RU	3.21	2.91	2.20	2.77	2.69	8.27	10.7	337823	
SE	2.10	1.16	1.25	1.53	1.09	1.03	1.03	454259	100

2.3. Total inputs of assessed metals to the Baltic Sea 2012–2018

There are quite large differences in the total amounts of the different metals that enter the Baltic Sea every year, as well as the main route of entry is quite variable between the metals. In this context, it ought to be noted that the full picture is not known for chromium, copper, nickel, and zinc due to lack of information on the atmospheric deposition. For cadmium, mercury and lead, where deposition estimates are available, it is estimated that the total average annual inputs to the Baltic Sea 2015-2017 have been 27, 5.3, and 356 tonnes per year, respectively (Table 3). Mercury and lead are characterised as the metals for which the atmospheric deposition is an especially important route of entry to the Baltic Sea which constitute about 47% and 40 %, respectively of the total inputs to the sea (Figure 3). On the other hand, for cadmium the riverine inputs are the predominant route of entry with about 81% of the total inputs. For all assessed metals, the direct point sources make the smallest contribution to the total inputs (about 0.5-2%), although the point sources might be underestimated somewhat as e.g. Sweden only can report metal inputs from larger MWWTs, as smaller plants seldom have reporting obligations on metals in their permits. Anyhow, the importance of the direct point sources may be regarded to be considerably less than the other two routes of entry.

As previously stated, there is no atmospheric deposition data available for chromium, copper, nickel and zinc, and consequently the complete picture of the routes of entry for these metals is not known. However, as with the previously mentioned metals, also for these metals the direct point-sources comprise only a minor part of the total waterborne inputs with about 2% of the totals for all metals except zinc, for which the point-sources constitute almost 5% of the total waterborne inputs (Table 3, and Figure 4).

Table 3. Inputs of cadmium, chromium, copper, mercury, nickel, lead, and zinc to the Baltic Sea from direct point sources, via rivers, total waterborne, and atmospheric deposition 2012–2018. Atmospheric deposition only available for Cd, Hg and Pb. Annual average inputs 2015–2017, and 2016–2018 are also given.

Source	Cd (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
Direct point sources	0.14	0.20	0.15	0.53	0.47	0.48	0.55	0.49	0.50
Riverine	35	21	5	26	19	19	17	21	18
Waterborne	35	21	5	27	19	19	18	21	18
Depositiona	4.9	4.5	4.8	4.3	3.9	4.2	4.2	4.2	4.1
Total	40	25	10	31	23	23	22	25	22

Source	Cr (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
Direct point sources	2.9	4.0	3.8	5.5	4.0	4.0	4.4	4.5	4.1
Riverine	412	137	151	207	143	138	114	163	132
Waterborne	415	141	155	212	147	142	118	167	135
Depositiona									
Total									

Source	Cu (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
Direct point sources	15	16	12	23	16	17	17	19	17
Riverine	1031	770	486	964	802	518	411	883	788
Waterborne	1046	786	498	987	818	535	428	780	593
Depositiona									
Total									

Table 3. (continued) Inputs of cadmium, chromium, copper, mercury, nickel, lead, and zinc to the Baltic Sea from direct point sources, via rivers, total waterborne, and atmospheric deposition 2012–2018. Atmospheric deposition only available for Cd, Hg and Pb. Annual average inputs 2015–2017, and 2016–2018 are also given.

Source				**Hg (tonnes/year)**				**Annual average**	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
Direct point sources	0.094	0.497	0.097	0.177	0.075	0.079	0.085	0.110	0.080
Riverine	1.5	1	0.8	1.7	2.8	1.4	1.5	2.4	2.5
Waterborne	1.6	1.5	0.9	1.9	2.9	1.5	1.6	2.5	2.6
Deposition[a]	3.4	2.9	3.2	2.7	2.6	3.1	2.8	2.8	2.8
Total	5.0	4.4	4.1	4.6	5.5	4.6	4.4	4.9	4.8

Source				**Ni (tonnes/year)**				**Annual average**	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
Direct point sources	14.7	12.7	9.7	23.3	11.7	10.1	9.9	15.0	10.6
Riverine	717	650	408	746	485	440	345	609	460
Waterborne	732	663	418	769	497	450	355	572	434
Deposition[a]									
Total									

Source				**Pb (tonnes/year)**				**Annual average**	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
Direct point sources	2.24	1.87	1.48	2.74	2.30	2.85	2.42	2.63	2.52
Riverine	345	301	252	230	225	177	76	211	160
Waterborne	347	303	254	233	227	180	78	214	163
Deposition[a]	157	167	216	163	127	139		143	
Total	504	472	473	396	354	319		357	

Source				**Zn (tonnes/year)**				**Annual average**	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
Direct point sources	132	106	59	205	125	139	141	156	135
Riverine	3201	2257	1167	2961	2301	4065	5116	3109	3827
Waterborne	3333	2363	1226	3166	2426	4204	5257	3265	3962
Deposition[a]									
Total									

[a] Deposition data from EMEP (HELCOM 2020 for Cd and Hg, HELCOM 2019b for Pb)

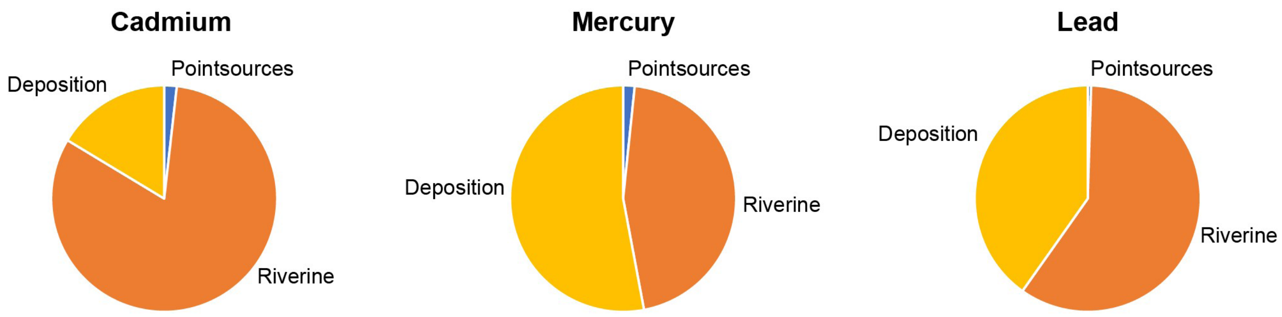


Figure 3. The division of inputs of cadmium, mercury and lead from point sources, via rivers, and atmospheric deposition to the Baltic Sea based on average inputs 2015–2017.

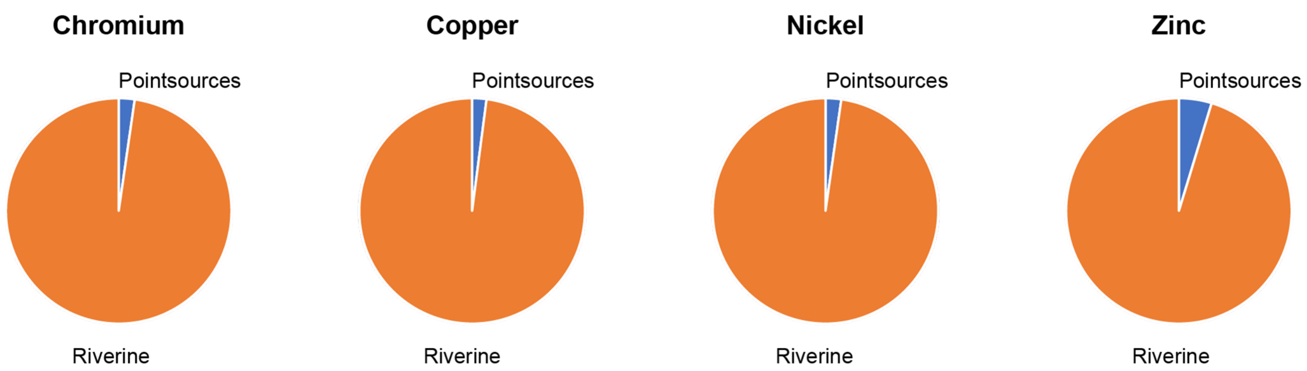


Figure 4. The division of inputs of chromium, copper, nickel, and zinc, from point sources and via rivers to the Baltic Sea based on average inputs 2015–2017.

2.4. Inputs of metals via rivers and direct point-sources 2012–2018

As previously concluded, the metal inputs from direct point-sources to the Baltic Sea are quite low compared to the riverine inputs and the inputs via atmospheric deposition for the metals that have deposition data available. This is also evident when the load data is presented per contracting party (Tables 4 and 5). In general, CPs with large flow to the Baltic Sea, due to either large rivers and/or large surface area, naturally tend to have larger riverine metal loads. For point sources it is more difficult to draw any general conclusions, as it is more complicated than just e.g. the number of inhabitants, but also include industrial release directly to the Baltic Sea. The proportion of point sources inland compared to direct point sources to the Sea is very important, as the former will burden the riverine inputs rather than direct point sources. Also, the composition of waste water, including its origin, is of importance as this will influence the amount of metals in the incoming water to the waste water treatment plants, although the majority of the metals will end-up in the sewage sludge due to their predominantly high particle affinity.

Table 4. Riverine inputs of cadmium, chromium, cupper, mercury, nickel, lead, zinc to the Baltic Sea 2012–2018. Annual average inputs 2015–2017, and 2016–2018 are also given.

CP	Cd (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	0.101	0.099	0.033	0.093	0.057	0.088	0.079	0.079	0.075
DKa									
EEb	0.913	0.046	0.034	0.034	0.007	0.35	0.008	0.130	0.122
FI	2.65	1.66	1.84	2.64	1.68	1.56	1.16	2.0	1.5
LTb	0*	0*	0.014d	0*	0*	0*	0*		
LVb			0.243	0.391	0.644	0.170	0.362	0.402	0.392
PL	0.280	0.210	0.678	0.701	0.461	0.656	0.583	0.6	0.6
RUc	28.8	17.4	10.3d	20.1	14.8	15.2	12.9	16.7	14.3
SE	2.45	1.49	1.68	1.82	1.15	1.46	1.41	1.5	1.3
Total	*35*	*21*	*5*	*26*	*19*	*19*	*17*	*21*	*18*

CP	Cr (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	1.61	1.12	0.13	0.43	0.98	1.48	0.91	0.96	1.1
DKa									
EEb	5.2	0.37	0.24	0.83	0.025	1.00	0.35	0.62	0.46
FI	126	90	76	124	70	66	57	87	64
LTb	7.7	8.0		11.1	3.49	12.8	9.2	9	9
LVb			32.0	5.1	10.1	5.7	1.0	7.0	5.6
PL	192			20.4	26.7	13.6	4.2	20	15
RUc									
SE	80	37	43	45	32	37	42	38	37
Total	*412*	*137*	*151*	*207*	*143*	*138*	*114*	*163*	*132*

Table 4. (continued) Riverine inputs of cadmium, chromium, cupper, mercury, nickel, lead, zinc to the Baltic Sea 2012–2018. Annual average inputs 2015–2017, and 2016–2018 are also given.

CP	Cu (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	9	7	3	6	5	7	8	6	7
DKa									
EEb	84	33	9	23	33	29	9	29	24
FI	257	169	143	215	152	139	116	169	136
LTb	21	31		23	19	38	46	27	34
LVb			29	42	57	65	43	55	55
PL	148	112	78	40	54	77	55	57	62
RUc	210	219		402	316			359	316
SE	302	199	224	213	166	163	134	181	154
Total	*1031*	*770*	*486*	*964*	*802*	*518*	*411*	*883*	*788*

CP	Hg (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	0.018	0.033	0.003	0.015	0.009	0.011	0.012	0.01	0.01
DKa									
EEb	0.172	0.043	0.039	0.63	0.088	0.091	0.026	0.27	0.07
FI	0.428	0.272	0.301	0.235	0.167	0.175	0.135	0.19	0.16
LTb	0	0.049		0.039	0.029	0.109		0.06	0.07
LVb						0.63	0.904	0.63	0.77
PL	0.315	0.306	0.103	0.352	0.018	0.035	0.09	0.14	0.05
RUc			0.007*	0	2.188	0.004*		0.73	1.10
SE	0.523	0.292	0.315	0.418	0.267	0.298	0.298	0.33	0.29
Total	*1.5*	*1*	*0.8*	*1.7*	*2.8*	*1.4*	*1.5*	*2.4*	*2.5*

CP	Ni (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	4	3	1	4	4	6	8	5	6
DKa									
EEb	23	7	5	7	13	11	6	10	10
FI	320	203	228	344	244	230	166	273	213
LTb	10	16		14	16	21	13	17	17
LVb			9	17	0*	0*	0*	17	0*
PL	34	83	54	52	56	78	52	62	62
RUc	153	237		185	55			120	55
SE	173	101	111	123	97	94	100	105	97
Total	*717*	*650*	*408*	*746*	*485*	*440*	*345*	*609*	*460*

Table 4. (continued) Riverine inputs of cadmium, chromium, cupper, mercury, nickel, lead, zinc to the Baltic Sea 2012-2018. Annual average inputs 2015-2017, and 2016-2018 are also given.

CP	Pb (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	1.86	0.97	0.34	0.43	0.42	0.80	0.76	0.6	0.7
DKa									
EEb	8.5	1.9	0.77	2.1	0.87	1.8	0.35	1.6	1.1
FI	41	27	24	39	25	23	20	29	23
LTb	0*	0*	0.34*	23	9	23	0.3	18	11
LVb			8	5	32	40	22	26	31
PL	10	4.6	1.3	9	1.6	2.3	2.5	4.3	2.1
RUc	222	238	182*	115	124	55	0c	98	60
SE	62	29	35	36	32	31	30	33	31
Total	*345*	*301*	*252*	*230*	*225*	*177*	*76*	*211*	*160*

CP	Zn (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	28	21	6	13	12	18	14	14	15
DKa									
EEb	127	18	11	121	204	63	13	129	93
FI	798	461	546	819	559	484	402	621	482
LTb	105	124		217	36	76	435	110	182
LVb			32	67	49	62	58	59	56
PL	97	115	20	140	58	83	47	94	63
RUc	1106	1002		948	948	2869	3735	1588	2517
SE	940	516	552	636	435	410	412	494	419
Total	*3201*	*2257*	*1167*	*2961*	*2301*	*4065*	*5116*	*3109*	*3827*

Note! aDenmark only have a very limited amount of data that is not possible to extrapolate to the whole country, and consequently no data is given here. bThe spatial and/or temporal coverage of load data from EE, LT and LV is not complete. cInputs from Russia may be overestimated due to the used estimation method based on high LOQ's. dData according to BSEP162. eNote that the annual average of the total metal input is not the same as the mean of the three different years, as the sum of the different annual averages for the CPs are not influenced by missing observations. *Estimates based on data mainly <LOQ

Table 5. Inputs of cadmium, chromium, cupper, mercury, nickel, lead, zinc, and annual mean water discharge from point sources to the Baltic Sea 2012–2018. Annual average inputs 2015–2017, and 2016–2018 are also given.

CP	Cd (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	0.011	3E-04	3E-04	0.052	4E-04	0.004	0.001	0.019	0.002
DK	0.013	0.013	0.014	0.014	0.013	0.014	0.012	0.014	0.013
EE			0.002	0.002	0.002	0.016	0.015	0.007	0.011
FI	0.08	0.104	0.109	0.098	0.08	0.091	0.093	0.071	0.069
LT			0*						
LV	0.006	0,015	0,008	0,010	0,030	0,030	0,075	0.023	0.045
PL	0.006	0.055	0.002	0.01	2E-04	0.003	0.022	0.004	0.008
RU	0.002		2E-04a	0.001	0*	0*	4E-05	4E-04	1E-05
SE	0.022	0.018	0.016	0.343	0.352	0.323	0.338	0.339	0.338
Total	*0.14*	*0.20*	*0.15*	*0.53*	*0.47*	*0.48*	*0.55*	*0.49*	*0.50*

CP	Cr (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	0.101	0.019	0.026	1.653	0.018	0.169	0.161	0.613	0.116
DK	0.321	0.307	0.332	0.352	0.328	0.342	0.304	0.341	0.325
EE	0.145	0.085	0.091	0.015	0.004	0.058	0.054	0.026	0.039
FI	1.652	2.723	2.734	1.996	2.3	2.0	1.988	1.604	1.602
LT	0.033	0.04	0.005	0.001	0.002	0.001		0.001	0.002
LV	0.112	0.189	0.19	0.121	0.119	0.216	0.176	0.152	0.17
PL	0.03	0.15	0.11	0.123	0.133	0.055	0.011	0.104	0.066
RU	0.074	0.06		0.086	0*	0.0005	0*	0.029	2E-04
SE	0.47	0.47	0.391	1.146	1.113	1.207	1.721	1.155	1.347
Total	*2.9*	*4.0*	*3.8*	*5.5*	*4.0*	*4.0*	*4.4*	*4.5*	*4.1*

CP	Cu (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	0.371	0.105	0.123	7.51	0.179	0.644	0.645	2.778	0.489
DK	0.810	0.842	0.839	0.888	0.827	0.856	0.764	0.857	0.816
EE	0.6	0.42	0.13	0.19	0.18	0.25	0.26	0.207	0.23
FI	3.0	4.36	3.78	3.34	4.54	3.9	3.41	2.583	2.607
LT	0.133	0.143	0.033	0.013	0.011	0.013	0.008	0.012	0.011
LV	0.7	0.57	1.01	0.37	0.39	0.42	0.42	0.393	0.41
PL	0.12	0.54	0.24	0.21	0.03	0.25	0.69	0.163	0.323
RU	3.43	2.48		1.96	1.84	1.68	1.88	1.827	1.8
SE	5.46	5.96	5.68	8.17	8.07	8.78	8.95	8.34	8.6
Total	*15*	*16*	*12*	*23*	*16*	*17*	*17*	*18*	*17*

Table 5. (continued) Inputs of cadmium, chromium, cupper, mercury, nickel, lead, zinc, and annual mean water discharge from point sources to the Baltic Sea 2012–2018. Annual average inputs 2015–2017, and 2016–2018 are also given.

CP	Hg (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	0.0002	0.0002	0.0002	0.038	0.0002	0.0004	0.0007	0.013	0.0004
DK	0.021	0.020	0.022	0.023	0.021	0.022	0.020	0.022	0.021
EE	2E-04		5E-05		3E-04	3E-05	1E-04	2E-04	1E-04
FI	0.040	0.036	0.041	0.035	0.012	0.012	0.008	0.018	0.009
LT	0.002	4E-04	0.001	9E-04	7E-04	2E-04	6E-04	6E-04	5E-04
LV	0.007	0.012	0.005	0.005	0.006	0.006	0.023	0.006	0.012
PL	6E-04	0.406	0.005	0.034	5E-08	0.003	2E-05	0.012	0.001
RU				7E-04	5E-06	1E-04	1E-04	3E-04	7E-05
SE	0.023	0.022	0.023	0.04	0.035	0.035	0.032	0.037	0.034
Total	*0.094*	*0.497*	*0.097*	*0.177*	*0.075*	*0.079*	*0.085*	*0.110*	*0.080*

CP	Ni (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	0.25	0.08	0.17	10.32	0.24	0.43	0.38	3.67	0.35
DK	1.31	1.24	1.36	1.44	1.34	1.40	1.24	1.39	1.33
EE	0.33	0.31	0.24	0.22	0.16	0.22	0.23	0.2	0.203
FI	4.6	4.11	4.51	4.21	4.1	2.76	3.07	2.943	2.563
LT	0.023	0.021	0	0.007	0.008	0.01	0.009	0.008	0.009
LV	0.27	0.48	0.35	0.54	0.51	0.56	0.41	0.537	0.493
PL	0.13	0.24	0.28	0.26	0.21	0.3	0.06	0.257	0.19
RU	4.86	3.65		2.68	1.46	0.26	0.18	1.467	0.633
SE	2.89	2.57	2.7	3.67	3.75	4.31	4.41	3.91	4.157
Total	*14.7*	*12.7*	*9.7*	*23.3*	*11.7*	*10.1*	*9.9*	*15.0*	*10.6*

CP	Pb (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	0.011	0.002	0.004	0.014	0.002	0.014	0.015	0.01	0.01
DK	0.776	0.736	0.805	0.853	0.794	0.828	0.737	0.825	0.787
EE			0.004	0.006	0.002	0.032	0.02	0.013	0.018
FI	0.96	0.63	0.36	0.55	0.25	0.16	0.2	0.267	0.15
LT	0.004	0.08	0.002	0.001	0.001	0.001	0.001	0.001	0.001
LV	0.22	0.14	0.1	0.09	0.18	0.75	0.22	0.34	0.383
PL	0.06	0.08	0.03	0.07	0.01	0.02	6E-04	0.033	0.01
RU	0.01	0.004	3E-04a	0.01	0*	0.003	0.01	0.004	0.004
SE	0.19	0.19	0.17	1.15	1.06	1.04	1.21	1.083	1.103
Total	*2.24*	*1.87*	*1.48*	*2.74*	*2.30*	*2.85*	*2.42*	*2.63*	*2.52*

Table 5. (continued) Inputs of cadmium, chromium, cupper, mercury, nickel, lead, zinc, and annual mean water discharge from point sources to the Baltic Sea 2012–2018. Annual average inputs 2015–2017, and 2016–2018 are also given.

CP	Zn (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	0.8	0.4	0.4	75.2		3.2	3.1	39.2	3.2
DK	10.7	10.2	11.1	11.7	10.9	11.4	10.1	11.4	10.8
EE	1.3	0.9	0.7			2	2	2	2
FI	26.6	31.6	26.4	21.7	19.9	26.6	25.2	15.8	24.5
LT	0.338	0.418	0.108	0.065	0.065	0.112	0.089	0.081	0.089
LV	2.9	2.1	2.9	2.1	0.8	1.4	2.6	1.433	1.6
PL	27.3	3.6	2.9	2.8	3.1	3.3	3.5	3.067	3.3
RU	48.6	43.7		33.3	31.1	32.4	40.1	32.27	34.53
SE	13.6	12.8	14.8	58	59.3	59	54.3	58.77	57.53
Total	*132*	*106*	*59*	*205*	*125*	*139*	*141*	*156*	*135*

CP	Water flow (m3/s)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
DE	3.2	2.0	2.7	3.3	3.2	3.4	3.3	3.3	3.3
DK	11.2	10.6	11.5	12.2	11.2	11.8	10.1	11.8	11.0
EE	2.5	2.4	1.8	1.9	2.2	2.3	2.0	2.1	2.1
FI	22.6	21.9	21.8	14.1	21.4	34.5		23.3	28.0
LT	0.8	0.6	0.7	0.7	0.9	0.7		0.8	0.8
LV	2.1	2.1	1.9	1.9	2.0	1.8		1.9	1.9
PL		2.5	3.1	2.0	2.1	4.0	2.4	2.7	2.8
RU	26.3	27.3	27.7	27.3	27.8	29.4	26.4	28.2	27.8
SE	21.8	20.4	21.7	35.3	32.2	33.6	32.6	33.7	32.8
Total	894	925	940	933	950	1020	881	968	984

[a] Data according to BSEP162. *Estimates based on data mainly <LOQ

2.5. Total inputs of metals per basin 2012–2018

As was the case in the PLC-6 metal assessment, a basin-wise assessment of the waterborne (riverine + direct point sources) metal inputs is only possible for some of the Baltic Sea basins (Table 6). No data is presented for the southernmost basins mainly due to the lack of total load estimates for Denmark that makes it impossible to make comparisons with the other basins. The waterborne inputs to the other basins are characterised by the large amounts entering the Gulf of Finland due to the very large riverine inputs via Russia (Tables 4 and 5). There is a concern about the Russian estimates based on a considerable number of observations less than comparatively high LOQs, which are replaced by LOQ/2 in the estimations, but the large inputs are also a consequence of the very large amount of riverine water, mainly via River Neva, that enters the Gulf from Russia. These presumed over-estimated inputs caused by the problem with high LOQs could be avoided in future assessments, if Russia would apply more sensitive analytical methods, as the present ones give too high LOQs especially compared to the recommendations in the PLC-Water Guidelines.

For the other basins the metal inputs are quite comparable, except for the Archipelago Sea, which only receive about 1/10 of the total amounts to the other basins. However, taken into consideration that this basin is quite small and sparsely populated, the metal inputs are not insignificant.

Table 6. Total waterborne inputs of cadmium, chromium, cupper, mercury, nickel, lead and zinc, to the Baltic Sea basins 2012–2018. Annual average inputs 2015–2017, and 2016–2018 are also given.

CP				Cd (tonnes/year)				Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
BOB	2.27	1.31	0.38	2.31	1.39	1.24	0.95	1.65	1.19
BOS	1.63	1.07	0.59	1.57	1.08	1.29	1.22	1.31	1.2
ARC	0.13	0.1		0.16	0.06	0.1	0.05	0.11	0.07
BAP	2.19	2.11	1.53	1.15	3.26	4.24	1.08	2.88	2.86
GUF	30.8	17.6	10.6	20.3	12.6	12.9	13.1	15.3	12.9
GUR	0.38	0.03*	0.05*	0.36	0.61	0.22	0.33	0.40	0.39
WEBa									
SOUa									
KATa									

CP				Cr (tonnes/year)				Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
BOB	101.8	53	13.3	74.1	63.4	51	47.6	62.8	54.0
BOS	45.2	25.9	12.5	39.3	19.4	23.2	27.6	27.3	23.4
ARC	12.5	18.8		28.3	6.7	8.2	5.1	14.4	6.7
BAP	30.9	50.4	33.9	38.9	33.4	30.9	19.3	34.4	27.9
GUF	29.3	19.1	9.7*	16.4	6.0	17.7	11.2	17.1	14.5
GUR	6.2	0.41*	0.25*	5.1	9.7	9.7	0.9*	8.2	9.7
WEBa									
SOUa									
KATa									

CP				Cu (tonnes/year)				Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
BOB	246	157	67	175	144	131	96	150	124
BOS	133	93	65	131	89	83	78	101	83
ARC	20	17		28	10	15	7,0	18	11
BAP	280	202	130	120	111	153	138	128	134
GUF	321	280	214	443	375	88	43	302	169
GUR	98	66	12.6*	59	67	65	43	64	58
WEBa									
SOUa									
KATa									

Table 6. (continued) Total waterborne inputs of cadmium, chromium, cupper, mercury, nickel, lead and zinc, to the Baltic Sea basins 2012–2018. Annual average inputs 2015–2017, and 2016–2018 are also given.

CP	Hg (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
BOB	0.522	0.294	0.086	0.34	0.242	0.214	0.18	0.265	0.212
BOS	0.282	0.149	0.12	0.183	0.123	0.151	0.161	0.152	0.145
ARC	0.004	0.027		0.029	0.012	0.013	0.005	0.018	0.010
BAP	0.619	1.056	0.602	0.521	2.262	0.329	0.231	1.037	0.941
GUF	0.053	0.054	0.023	1.783	0.04	0.178	0.064	0.667	0.094
GUR	0.263	0.058	0.038	0.061	0.075	2.148	0.839	0.761	1.021
WEBa									
SOUa									
KATa									

CP	Ni (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
BOB	260	147	23	277	221	184	135	227	180
BOS	124	82	32	100	78	85	82	88	82
ARC	12	12		25	6.0	8.4	4.7	13	6
BAP	112	144	109	112	92	120	93	108	102
GUF	221	273	285	216	83	32	23	110	46
GUR	13.6*	3.6*	2.5*	27.1	3.9*	5.8*	2.7*	12	4
WEBa									
SOUa									
KATa									

CP	Pb (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
BOB	40	18	6	24	27	20	16	24	21
BOS	25	14	10	21	12	15	19	16	15
ARC	5.6	5.4		11.0	3.1	4.4	1.9	6	3
BAP	52	49	19	38	38	49	11	42	33
GUF	237	246	187	122	111	53	4*	95	56
GUR	9.8	2.4	0.97	1.1	30.5	33.3	19.5	22	28
WEBa									
SOUa									
KATa									

Table 6. (continued) Total waterborne inputs of cadmium, chromium, cupper, mercury, nickel, lead and zinc, to the Baltic Sea basins 2012–2018. Annual average inputs 2015–2017, and 2016–2018 are also given.

CP	Zn (tonnes/year)							Annual average	
	2012	2013	2014	2015	2016	2017	2018	2015-2017	2016-2018
BOB	851	444	139	716	557	424	348	566	443
BOS	529	297	195	461	276	288	317	342	294
ARC	57	53		91	32	41	25	55	33
BAP	361	471	252	553	184	228	563	322	325
GUF	1306	1133	830	1053	1066	3048	3876	1722	2663
GUR	145	269	11.6*	219	301	95	67	205	154
WEBa									
SOUa									
KATa									

Note! aThe data is considered too incomplete to be assessed (DK data only as estimates for point sources, and for in total twelve rivers for three basins). *Severe amount of data is missing

2.6. Waterborne inputs of cadmium, mercury, and lead to the Baltic Sea 1995–2018

Overall, most CPs show substantial inter-annual variability in metal inputs to the Baltic Sea during the 24-year period 1995-2018 (Figures 5-7). Complete data series for the whole period is only available for a few countries, although for some countries only a few observations may be lacking. More severe is that in some cases there are considerable problems with the spatial and/or temporal data coverage. Especially for mercury there is quite a lot of missing data in many time series. The mercury data for Latvia and Russia is even too scattered to be shown at all (Figure 6). Denmark reported to the corresponding assessment for PLC-6 data for point sources and in total twelve rivers for the period 2012-2014. This limited data has been excluded in the prevailing assessment as it is not possible to extrapolate to estimate the total inputs for Denmark, and consequently they are not comparable to the data reported by the other countries. Due to these data issues, the assessment of the overall waterborne inputs over time can only be done with great caution, and especially for the oldest data in the time series. Even in complete time series, there might be changes over time in analytical methods and/or LOQs that call for great caution when assessing this kind of data.

The tendencies for the three CPs with the most complete and consistent time series, i.e. Germany, Finland, and Sweden are in general reduced waterborne inputs or at least stable inputs levels over time for all three metals (Figures 5-7). The cadmium and lead inputs for the other CPs with more or less complete time series show quite large inter-annual variability that makes it hard to reveal any tendencies (Figure 5 and 7). Regarding the mercury inputs, all CPs except the already stated for Germany, Finland and Sweden, the variability and/or scarceness of the data is too large to reveal any tendencies (Figures 6).

Figure 5. The annual waterborne inputs of cadmium from the Contracting Parties to the Baltic Sea (tonnes per year) 1995–2018. The bars show the sum of inputs from rivers and direct point sources. Note! Denmark is excluded due to very limited amount of data. Large inter-annual variability may be due to differences in the number of sources between years, but also on estimate methods used when observations are less than LOQ.

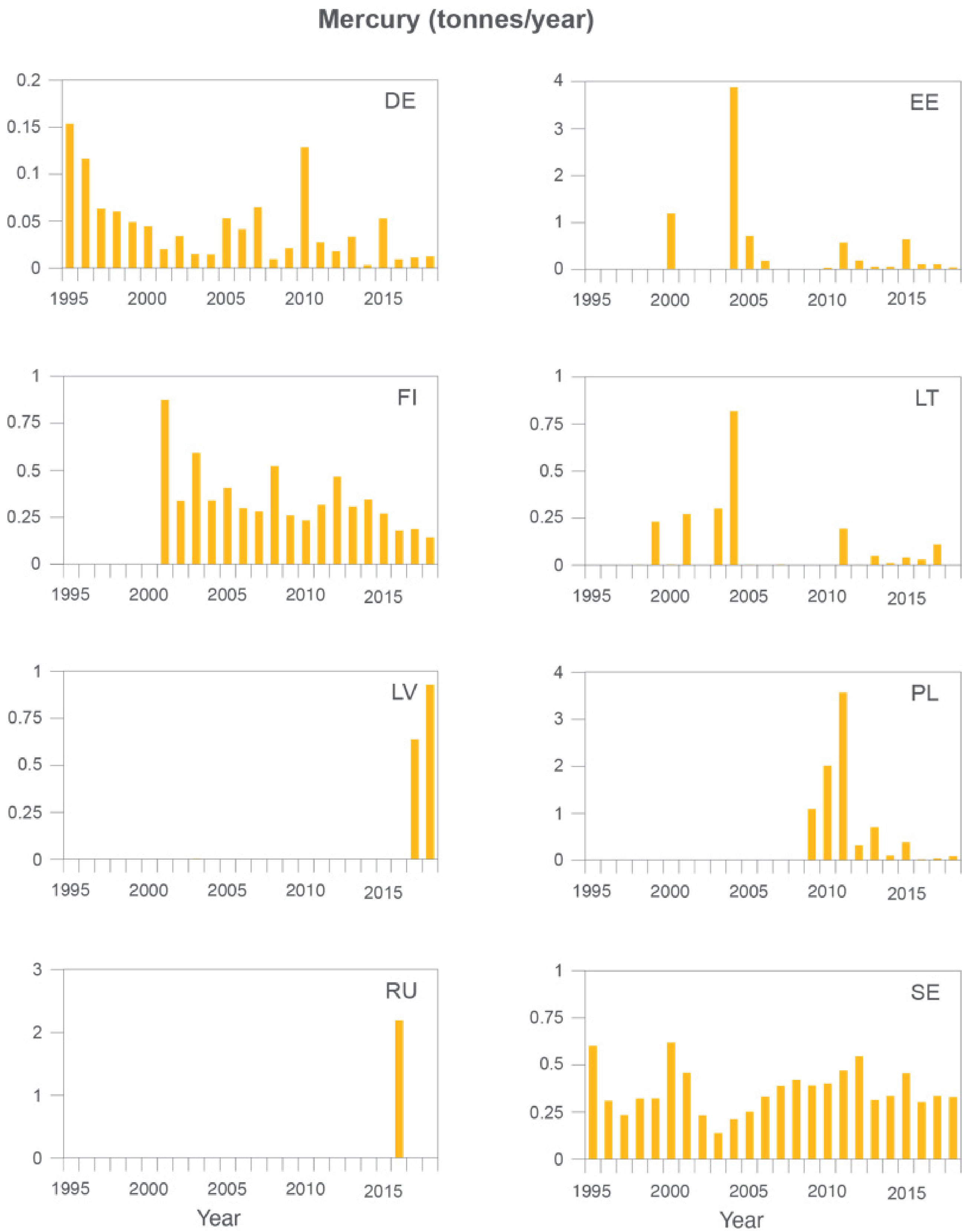


Figure 6. The annual waterborne inputs of mercury from the Contracting Parties to the Baltic Sea (tonnes per year) 1995–2018. The bars show the sum of inputs from rivers and direct point sources. Note! Denmark is excluded due to very limited amount of data. Latvian data only for 2017 and 2018, and Russian data only for 2016. Large inter-annual variability may be due to differences in the number of sources between years, but also on estimate methods used when observations are less than LOQ.

Figure 7. The annual waterborne inputs of lead from the Contracting Parties to the Baltic Sea (tonnes per year) 1995–2018. The bars show the sum of inputs from rivers and direct point sources. Note! Denmark is excluded due to very limited amount of data. Large inter-annual variability may be due to differences in the number of sources between years, but also on estimate methods used when observations are less than LOQ.

2.7. Atmospheric deposition of cadmium, mercury, and lead

All three metals with modelled atmospheric deposition show reducing deposition over time from the start of the time series in 1990 up to present (2018 for cadmium and mercury, 2017 for lead that is not assessed by EMEP for HELCOM every year), and this is valid for both the annual depositions as well as the weather-normalised annual depositions (Figures 8-10). The cadmium and lead depositions are reducing markedly more (-73%, and -81% respectively) than the mercury deposition (-35%). According to the assessment of the cadmium and mercury deposition by EMEP in the Baltic Sea Environmental Fact Sheets (Bartnicki et al. 2016), the reduction of atmospheric inputs is a result of abatement measures as well as of economic contraction and industrial restructuring in Poland, Estonia, Latvia, Lithuania, and Russia in early 1990s. The other CPs had their major emission reductions already before the start of the time series. However, the considerably lower reduction rate in mercury deposition (Figure 9) is probably due to the influence of a much larger long-range transport that makes it considered as a global contaminant (cf. Ilyin et al. 2016).

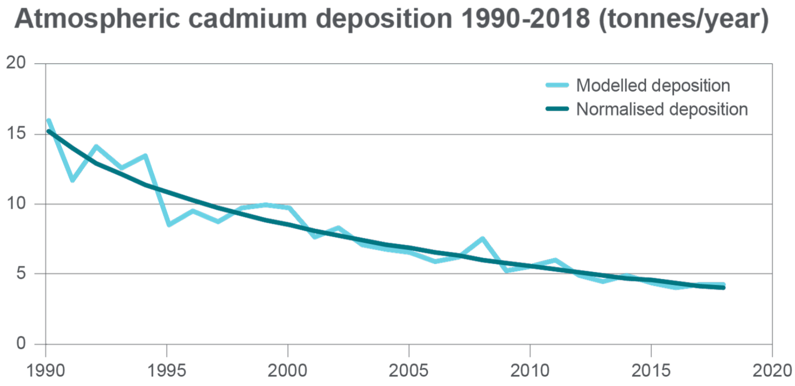


Figure 8. Modelled and normalised atmospheric cadmium deposition (tonnes/year) on the Baltic Sea 1990–2018. Data from EMEP (HELCOM 2020).

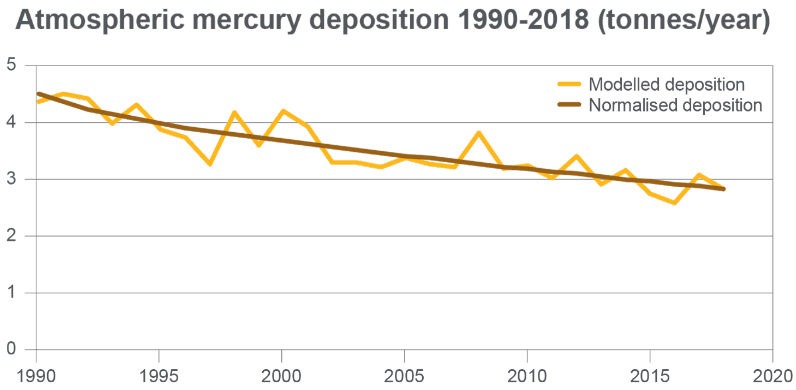


Figure 9. Modelled and normalised atmospheric mercury deposition (tonnes/year) on the Baltic Sea 1990–2018. Data from EMEP (HELCOM 2020).

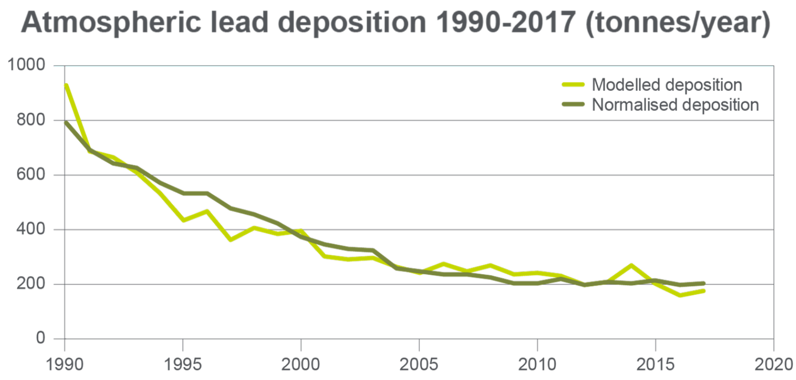
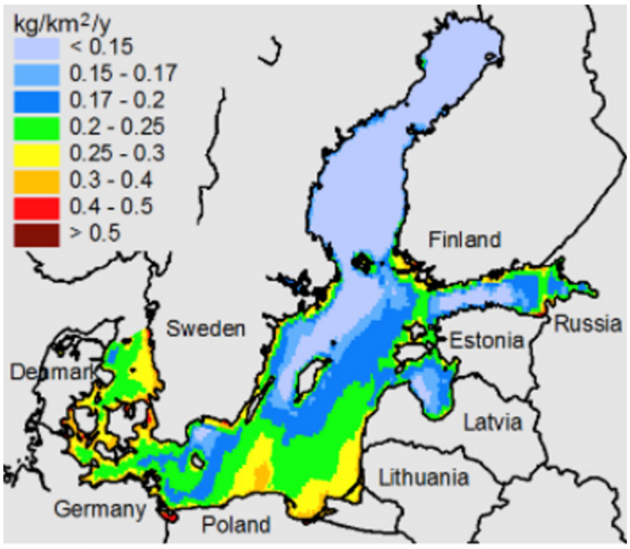
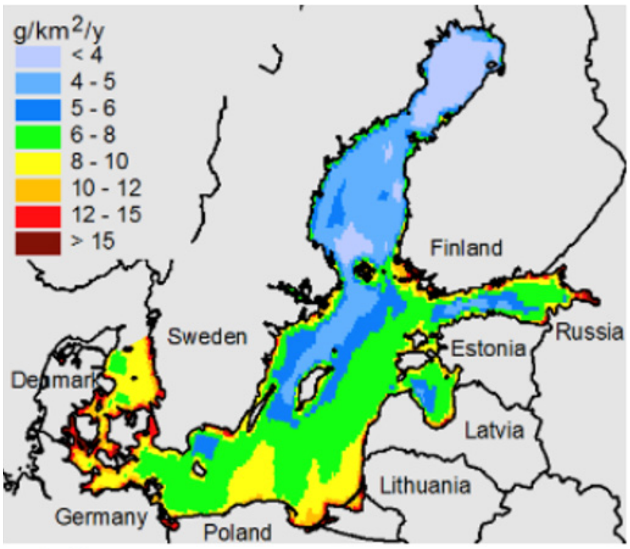


Figure 10. Modelled and normalised atmospheric lead deposition (tonnes/year) on the Baltic Sea 1990–2017. Data from EMEP (HELCOM 2019b).

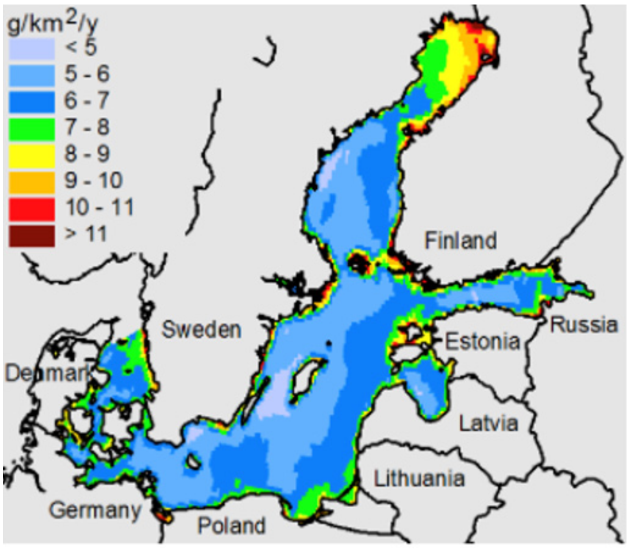
The spatial resolution of the modelled metal deposition and emissions in the Baltic Sea region reveal in general a strong south to north gradient, with both higher emissions and depositions in the southern part of the catchment area compared to the northern part (Figure 11). In addition to this gradient there are also markedly higher emissions as well as depositions of all three metals in Poland, although smaller "hot-spots" also occur in other CPs.



Lead deposition to the Baltic Sea in 2018, kg/km²/y



Cadmium deposition to the Baltic Sea in 2018, g/km²/y



Mercury deposition to the Baltic Sea in 2018, g/km²/y

Figure 11. Total annual deposition of lead, cadmium, and mercury (g/km2/y) in 2018. From EMEP (Ilyin et al. 2020)

3. Concentrations of nonyl– and octylphenols, and PFASs in rivers and coastal waters

In a HELCOM enquiry within the PLC-6 project asking for the HELCOM contracting parties data availability and concern regarding several groups of hazardous substances in rivers and WWTPs, great concerns about nonyl- and octylphenols, PFASs, and heavy metals were raised, which resulted in a dedicated data call on concentrations found in WWTPs, rivers and coastal zones in the Baltic Sea catchment. The results in influents and effluents to WWTPs are reported elsewhere, whereas the information on concentrations of nonyl- and octylphenols, and PFASs found in rivers and coastal waters are presented here.

3.1. Nonyl– and octylphenols in rivers and coastal water

Alkylphenol ethoxylates are chemicals used as surfactants in a broad range of applications, both industrial and consumer products, eg. paints, adhesives, inks, formulation of pesticides, paper and pulp production, industrial and household cleaners[1]. The most commercially important alkylphenols are nonyl- and octylphenol ethoxylates. These compounds are easily degraded by microorganisms into the more persistent and toxic nonylphenols and octylphenols, which are also the substances used to synthesize the ethoxylates. The substances have previously been included in the HELCOM list of priority substances (HELCOM 2010), but are currently not included in the set of Core Indicators. They are, however, Priority Substances listed under the WFD.

The concentrations of nonylphenol in natural waters were, as expected, in general considerably lower than concentrations measured in effluents (Figures 12 and 13). However, many riverine concentrations in Latvia were of the same order of magnitude as the highest measured concentrations in effluents in other countries. Latvia did not report measured concentrations in effluents. Note that Latvian and Swedish river samples were analysed on nonfiltered water, whereas it was not specified for the other countries.

In recent years lower detection limits of analytical methods have been used which has widen the concentration span observed over time, however the higher concentrations observed are in the range of a few µg/l in Finland, Sweden and Latvia.

Large variations over time were observed for the same sampling points in rivers. For example, in Finnish rivers, the limit of the analytical method applied was 200 ng/l for analyses made before 2013 and lower thereafter. The measured concentrations at the same sampling points varied by several hundreds of ng/l between different years. In Swedish rivers, which were sampled 3-4 times during a period of about 1.5 years, the concentrations at the same sampling point varied by a factor of 10 or more between different sampling occasions. Several factors influence concentrations in river water, e.g. emitted amount of the substance and the water flow, as well as the amount of suspended particles. The number of analyses is too small to give a clear picture of how concentrations change over time, but indicate typical levels in these rivers.

Only a few surface water samples in Germany had levels of nonylphenol above the detection limit. The limit was however in the range of 100 – 150 ng/l, which is substantially higher than detection limits reported for Swedish and Latvian samples (often below 10 ng/l).

The concentrations of octylphenol in rivers and coastal areas were lower than the corresponding concentrations of nonylphenol and the detection frequency was also lower (Figure 14 and Figure 15, data for CAS 104-66-9 only).

1 See e.g. Acir, I.-H. & Guenther, K. Endocrine-disrupting metabolites of alkylphenol ethoxylates–a critical review of analytical methods, environmental occurrences, toxicity, and regulation. Sci. Total Environ. 635, 1530–1546 (2018).

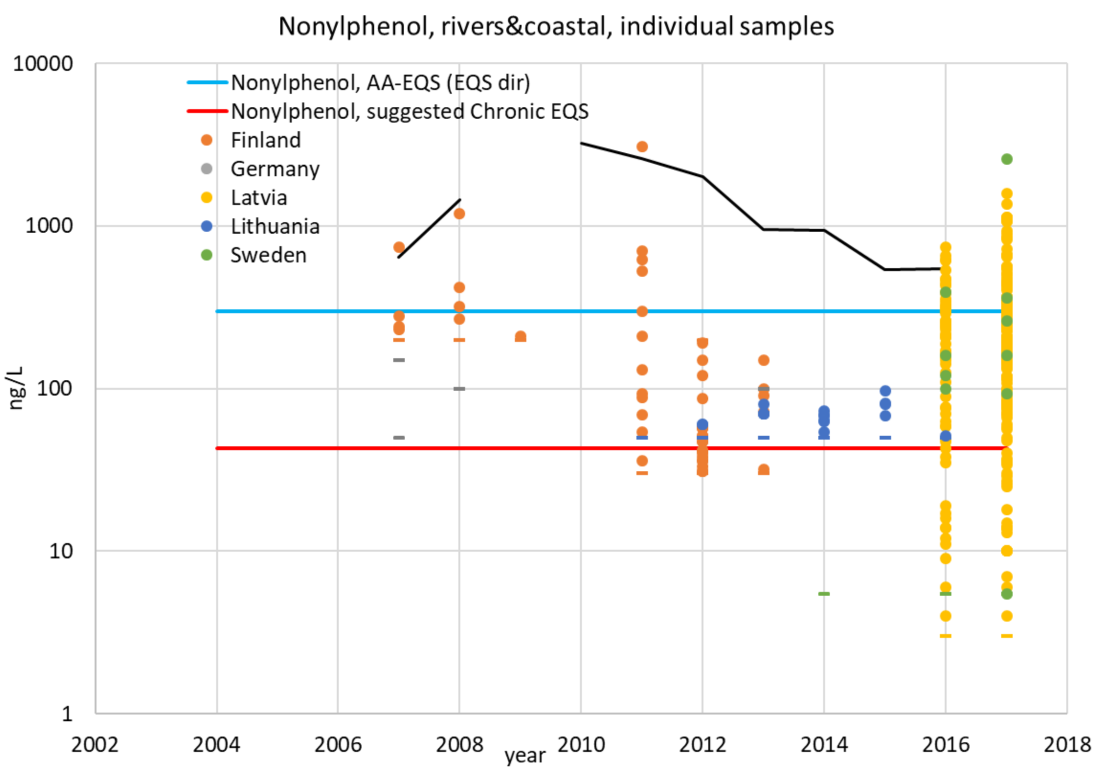


Figure 12. Measured concentrations at all riverine and costal sampling points in the data set in ng/l. Concentration above the detection limit are indicated by circles, concentrations at or below the detection limit are indicated by minus signs. The concentrations are compared to the current AA-EQS[1] of the EQS directive (300 ng/l) and the suggested chronic EQS by the Ecotox Centre Eawag-EPFL[2] (43 ng/l) (blue and red solid lines, respectively). The average measured concentrations in all effluent samples are also included for comparison (black solid line). Note the logarithmic scale.

1 Annual Average Environmental Quality Standard

2 https://www.ecotoxcentre.ch/expert-service/quality-standards/proposals-for-acute-and-chronic-quality-standards/

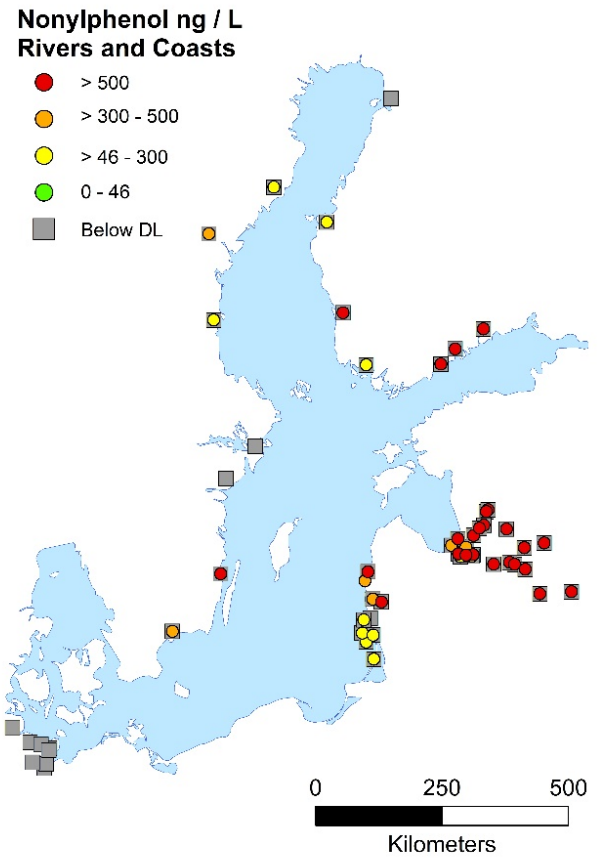


Figure 13. Observed concentrations (ng/l) of nonylphenol (CAS 25154-52-3) in rivers and coastal waters, all stations and years are included.

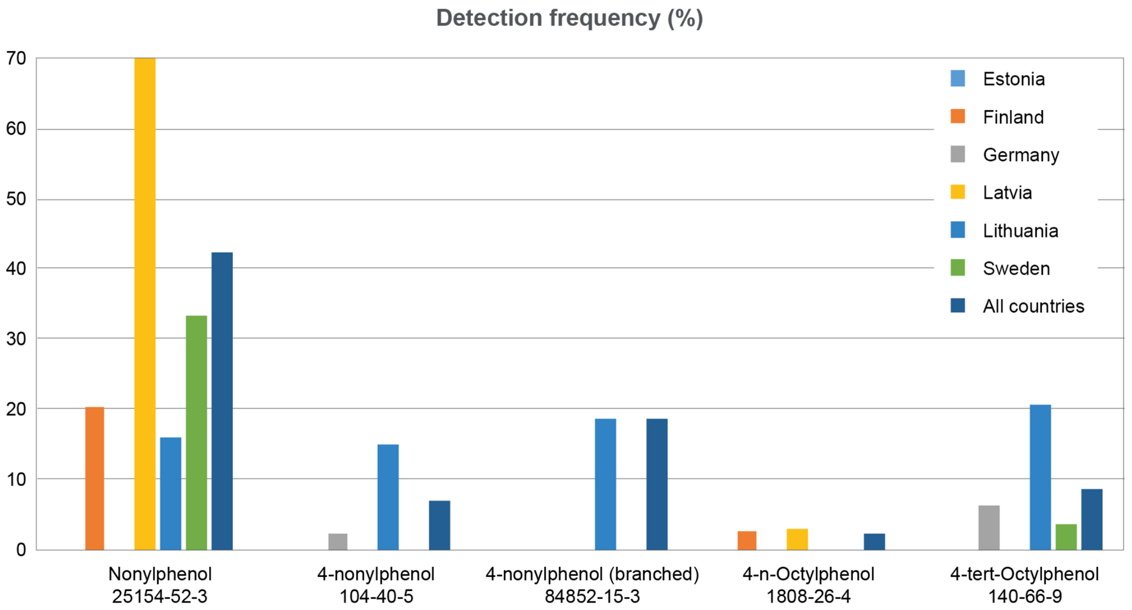


Figure 14. Detection frequencies of nonylphenols and octylphenols in rivers and coastal waters, all stations and years.

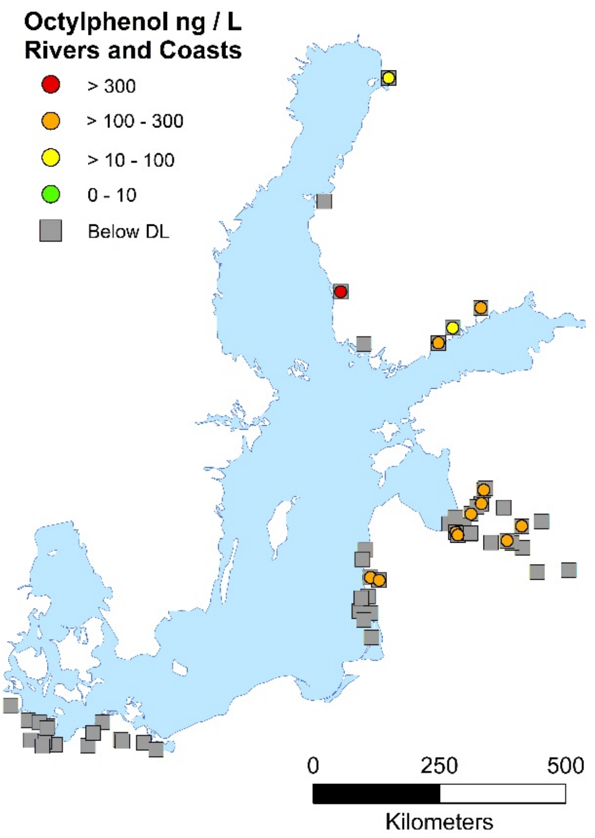


Figure 15. Observed concentrations (ng/l) of octylphenol (CAS 104-66-9) in riverine and coastal water, all stations and years.

3.2. Per- and polyfluoroalkyl substances (PFAS) in rivers and coastal waters

The per- and polyfluoroalkyl substances (PFAS) constitute a large group of synthetic organic chemicals consisting of more than 3000 substances. These chemicals have a wide range of applications such as firefighting foams, textile coatings, cookware, pesticide formulations, metal production, cosmetics, pharmaceuticals, food contact materials, inks etc. Perfluoroalkyl substances have a fully fluorinated carbon chain in the molecular structure, whereas the less stable polyfluoroalkyl substances have only partly fluorinated carbon chains. Data on environmental levels are available only for a small part of all PFASs on the market. PFOS is one of the most well-known PFAS and is a HELCOM Core Indicator, as well as a priority substance under the WFD and also listed for restrictions under the Stockholm Convention.

In total 1827 measurements were reported for a number of PFAS in rivers and coastal waters, 712 (39%) of these were above the detection limit or limit of quantification. The majority of the samples were taken in Swedish waters. Low detection frequencies are partly due to high detection limits of analytical methods used in some countries. Note that Swedish river samples analysed for PFAS were on filtered water, whereas the Latvian samples were not filtered, and consequently include both particulate and dissolved fractions.

The compounds most frequently measured were PFOS and PFOA, and these substances also exhibited the highest detection frequency in rivers and coastal waters (Figure 16).

The reported PFOS concentrations in rivers and coastal waters originate to a large degree from two sampling campaigns in Sweden 2013 and Latvia 2017, respectively (Figure 17). Several analyses from Estonia and Germany were reported with comparatively high detection limits.

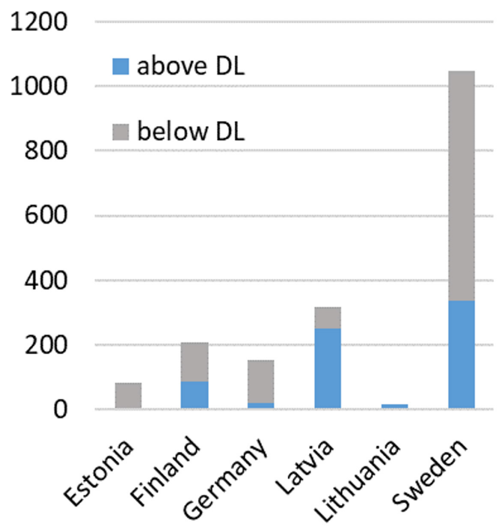
The reported PFOS concentrations show considerable variation both between different sampling sites and over time (Figure 18). The majority of the samples held concentrations below the AA-EQS (0.65 ng/l) and chronic EQS (2 ng/l) suggested by Ecotox Centre Eawag-EPFL[2]. Note that the detection limit of the method used for the Estonian samples (and in some of the German samples) was considerably higher than the AA-EQS.

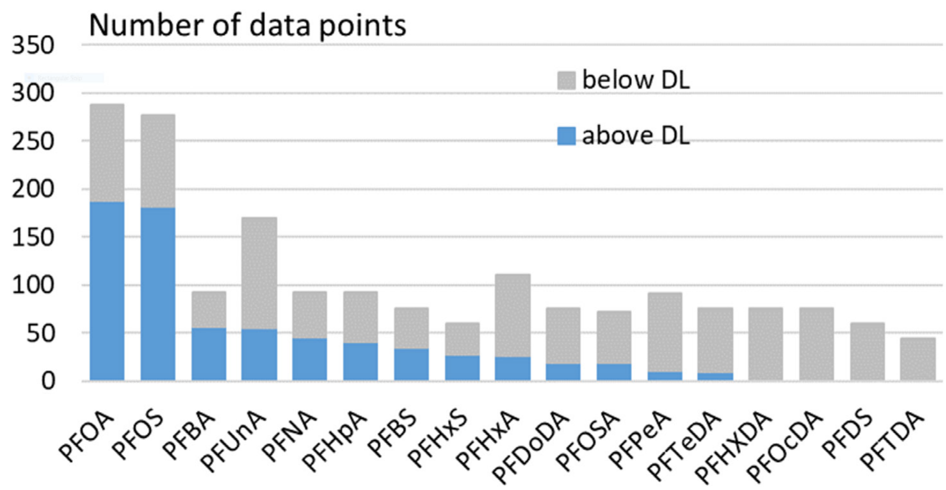
PFOA was detected at similar concentration levels as PFOS (Figures 19 and 20).

The majority of measured concentrations for PFAS other than PFOS and PFOA were reported by Sweden. The arithmetic (normal) and geometric mean concentrations were commonly of similar magnitude as those of PFOS and PFOA, all in the range of below one to a few ng per litre. A comparison between countries is difficult due to few comparable observations outside Sweden, and uncertainty regarding filtration or not of samples before analysis of concentrations. An overview of PFAS concentrations (both arithmetic and geometric means) in Swedish waters and a comparison between average levels in Sweden, Latvia and Lithuania is presented in Figure 21. Note however that Latvian river samples were not filtered, whereas Swedish samples were filtered. The concentrations of PFOA in Germany were reported to be below a comparatively high detection limit of 10 ng/l. PFBA concentrations in German surface water samples were high (on average ca 30 ng/l) compared to Swedish observations, whereas the PFOS concentration reported was only 1.7 ng/l and comparable to Swedish, Lithuanian and Latvian data.

2 Proposals for Acute and Chronic Quality Standards | Oekotoxzentrum (ecotoxcentre.ch)

Perfluorooctane sulfonate	PFOS
Perfluorooctanoic acid	PFOA
Perfluorobutanoic acid	PFBA
Perfluorononanoic acid	PFNA
Perfluorobutane sulfonate	PFBS
Perfluorohexane sulfonate	PFHxS
Perfluoroheptanoic acid	PFHpA
Perfluoroundecanoic acid	PFUnA
Perfluorooctane sulfonamide	PFOSA
Perfluorododecanoic acid	PFDoDA
Perfluorohexanoic acid	PFHxA
Perfluoropentanoic acid	PFPeA
Perfluorotetradecanoic acid	PFTeDA
Perfluorohexadecanoic acid	PFHXDA
Perfluorooctadecanoic acid	PFOcDA
Perfluorodecane sulfonate	PFDS
Perfluorotridecanoic acid	PFTDA





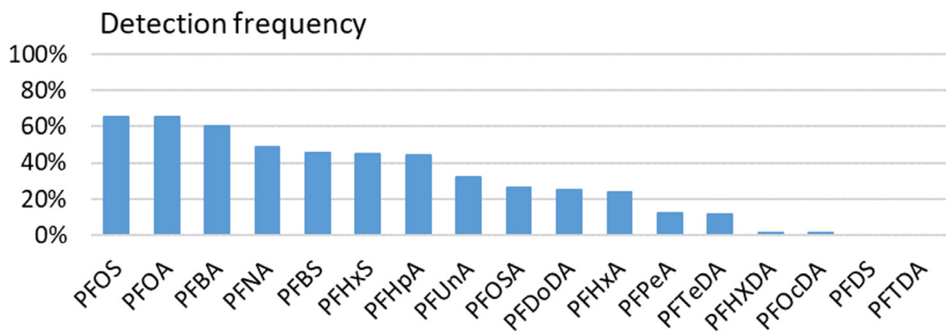


Figure 16. Names and abbreviations for analysed PFAS, number of data points for individual PFAS in rivers and coastal waters in total for all HELCOM CPs, for all PFAS per CP and detection frequencies.

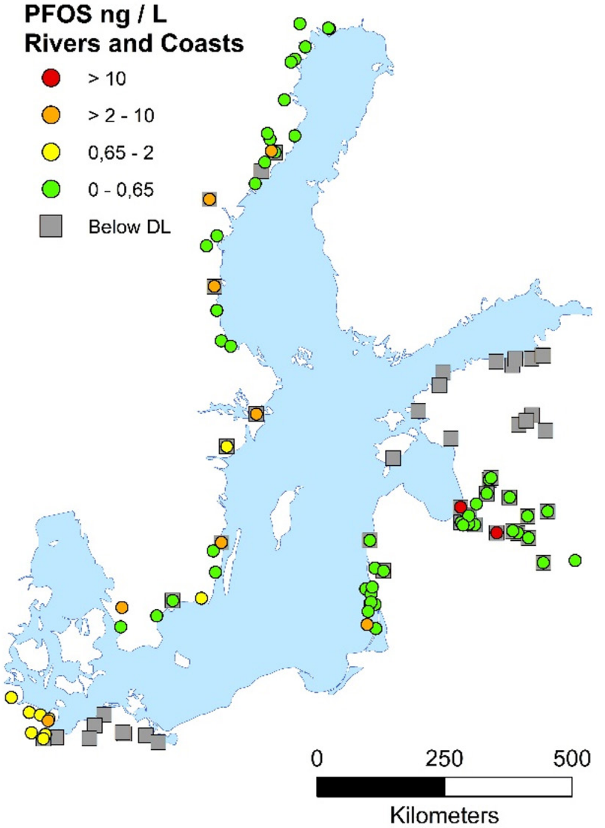


Figure 17. Sampling locations and detected levels of PFOS in rivers and coastal waters in ng/l.

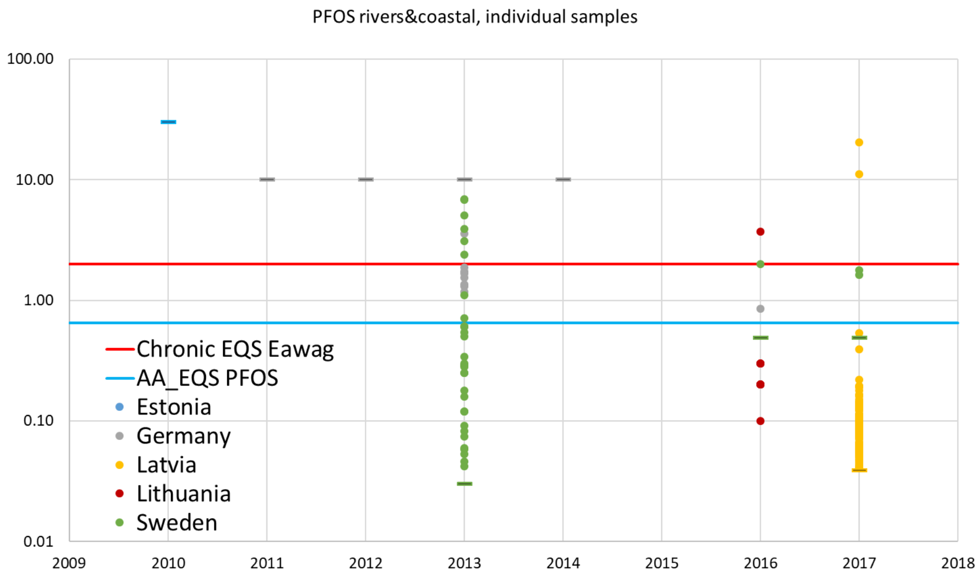


Figure 18. Reported concentrations of PFOS in rivers and coastal waters. Individual data points are plotted. Filled circles indicate observed concentrations above the detection limit. Minus signs indicate the detection limits of analytical methods applied.

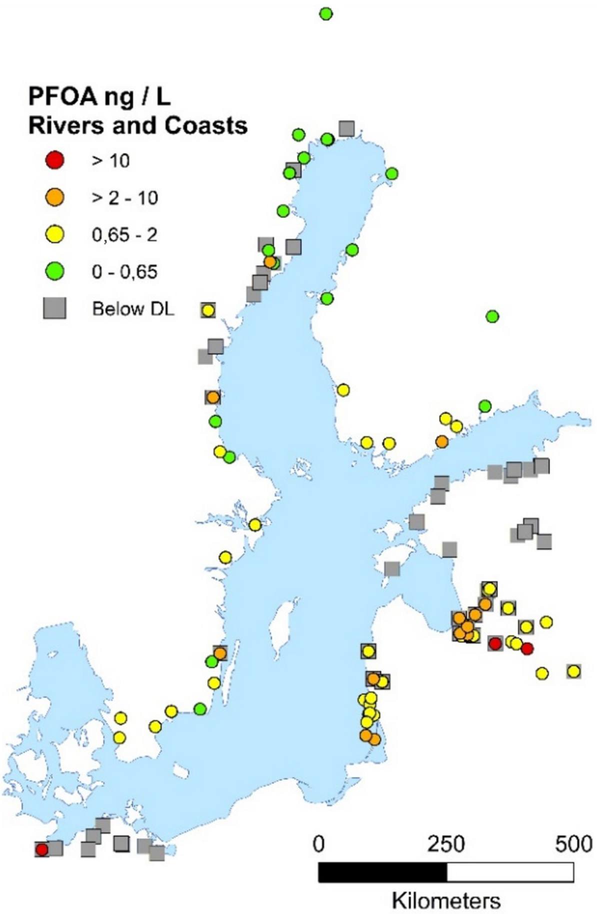


Figure 19. Sampling locations and detected concentrations of PFOA in rivers and coastal waters.

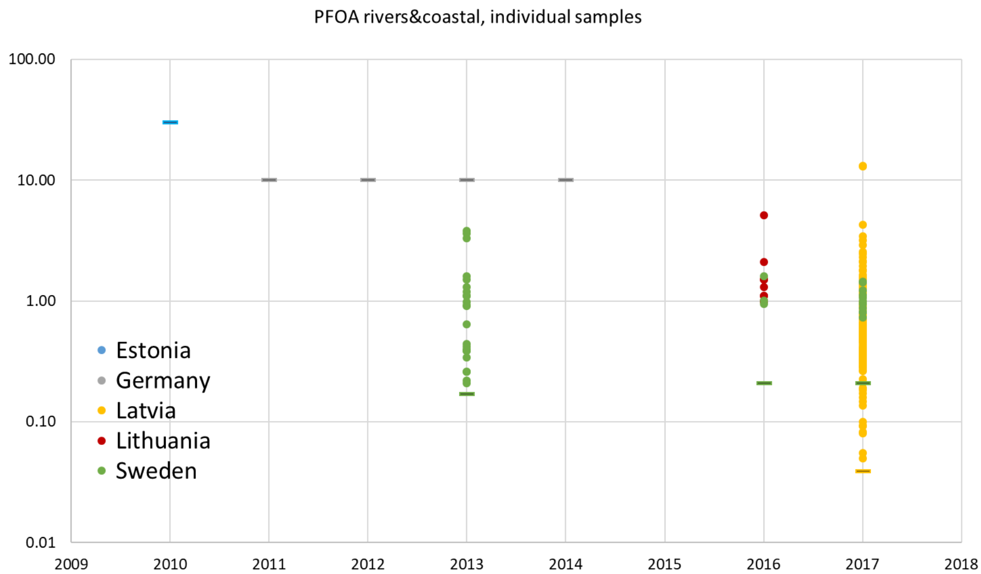
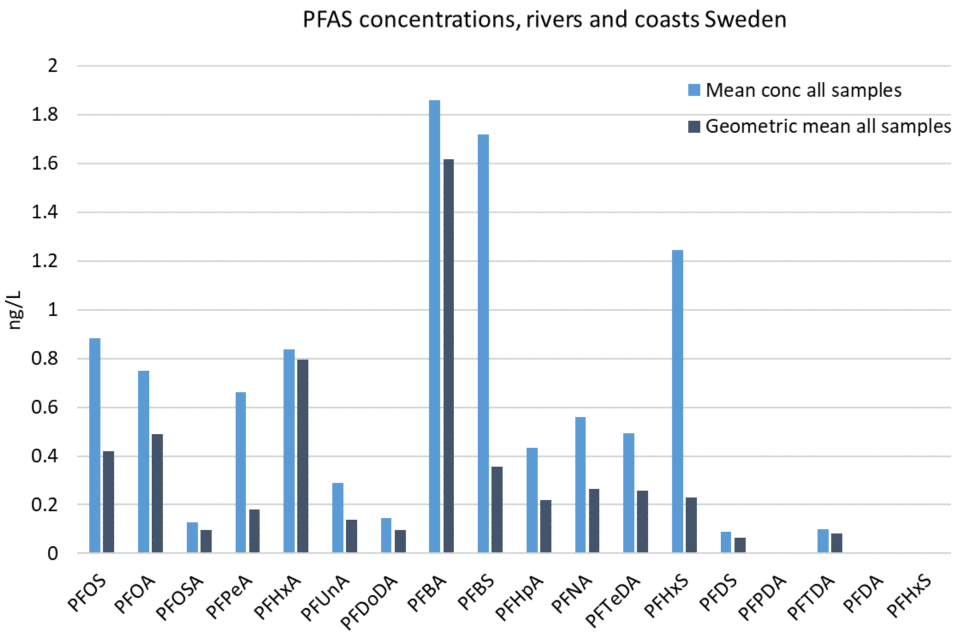


Figure 20. Concentrations of PFOA in rivers and coastal waters in ng/l. Note the logarithmic scale on the y-axis. Filled circles indicate measured concentrations above the detection limit, minus signs indicate the reported detection limits.



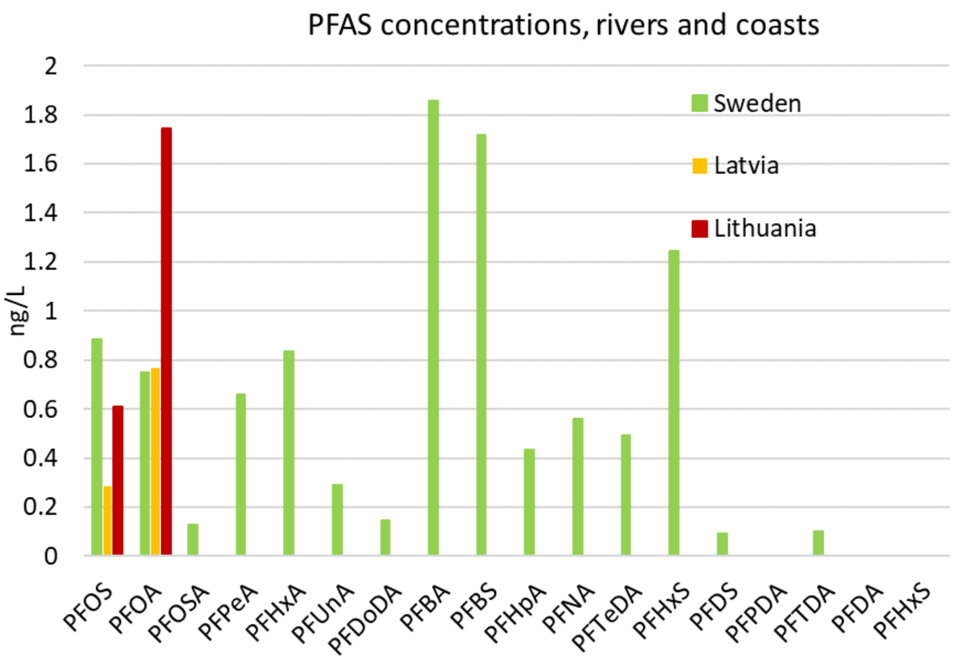


Figure 21. Arithmetic (normal) and geometric mean concentrations calculated for all PFAS in Swedish rivers and coastal water (upper panel) and arithmetic mean concentrations (ng/l) in Sweden, Latvia and Lithuania (lower panel), all years. Swedish samples were filtered. Latvian samples were not filtered, whereas information regarding Lithuanian samples was not provided.

4. Atmospheric deposition of some selected organic contaminants

4.1. Atmospheric deposition of Benzo(a) pyrene to the Baltic Sea

The atmospheric deposition of Benzo(a)pyrene to the Baltic Sea has steadily decreased since 1990 (Figure 22). The spatial pattern of both the deposition and the anthropogenic emissions are rather similar to the patterns for the metals, with a strong south-to-north gradient, and the highest levels in the south to be found in the southern part of Poland (Figure 23). The emissions are heavily dominated by the so-called Sector C "Other Stationary Combustion" (Gauss et al. 2020).

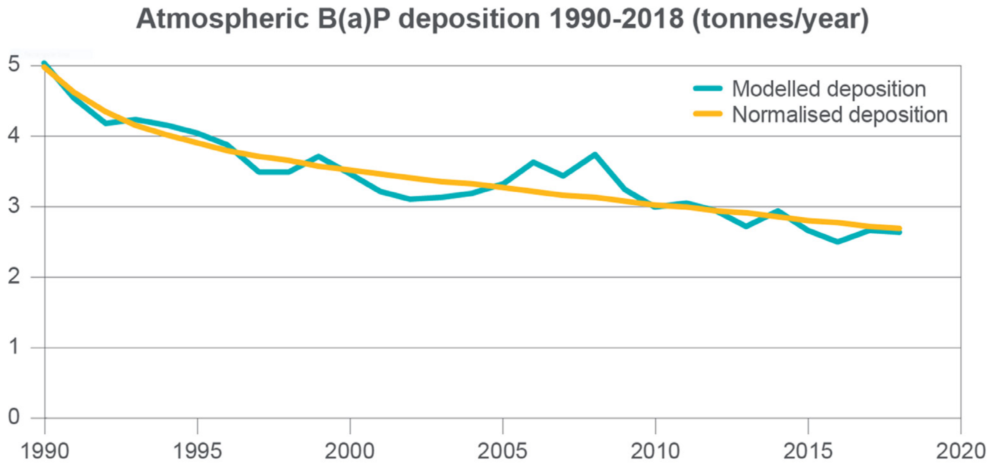
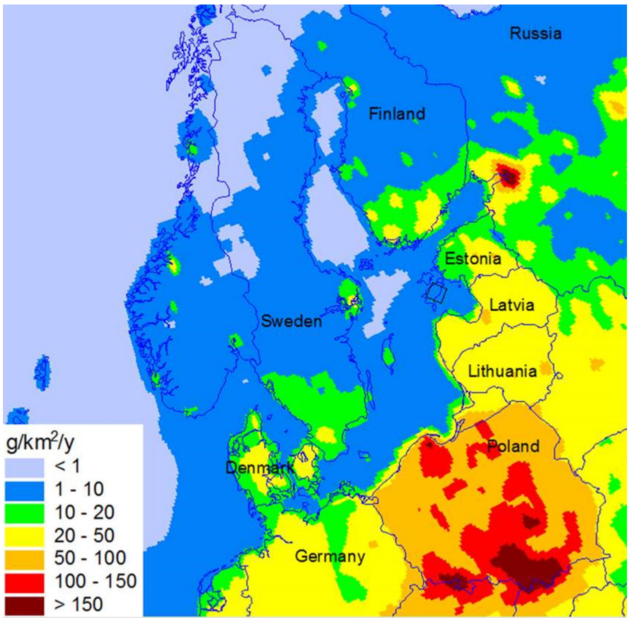


Figure 22. Modelled and normalised atmospheric B(a)P deposition (tonnes per year) on the Baltic Sea 1990–2018. Data from EMEP (HELCOM BSEFS 2020).



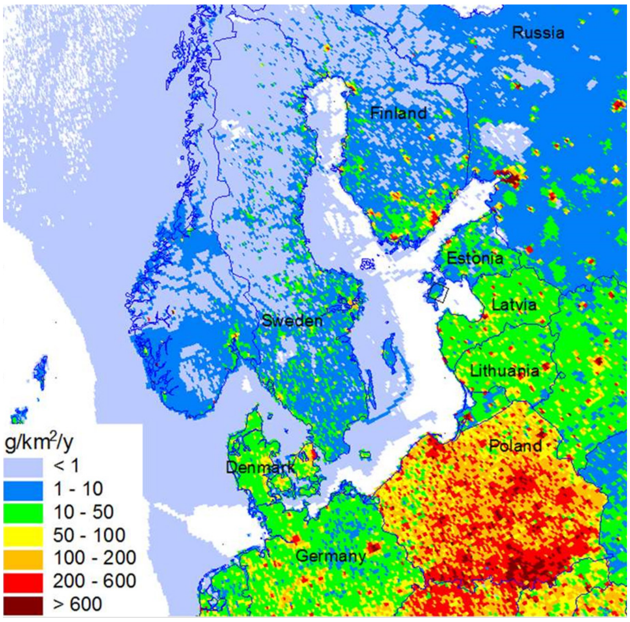


Figure 23. Total annual Benzo(a)pyrene deposition (left) and anthropogenic emissions (right) in the Baltic Sea region 2018 in g/km2/ year. From Gauss et al. 2020.

4.2. Atmospheric deposition of dioxins and furans to the Baltic Sea

The deposition of PCDD/Fs to the Baltic Sea has decreased over the period from 1990 (figure 24). The spatial pattern of deposition and anthropogenic emissions show the common south-to-north gradient, and the highest levels in the south to be found in the southern part of Poland (Figure 25). The emissions are dominated by the so-called Sectors B and C, i.e. "Industry" and "Other Stationary Combustion", and to some extent also Sector A "Public Power" (Gauss et al. 2020).

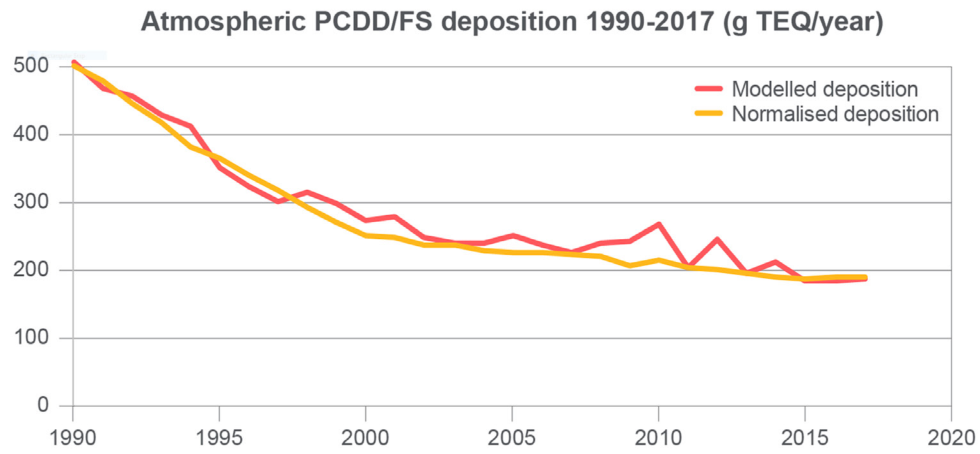


Figure 24. Modelled and normalised atmospheric PCDD/Fs deposition (g TEQ1/year) on the Baltic Sea 1990– 2017. Data from EMEP (BSEFS 2019).

1 Toxic Equivalents

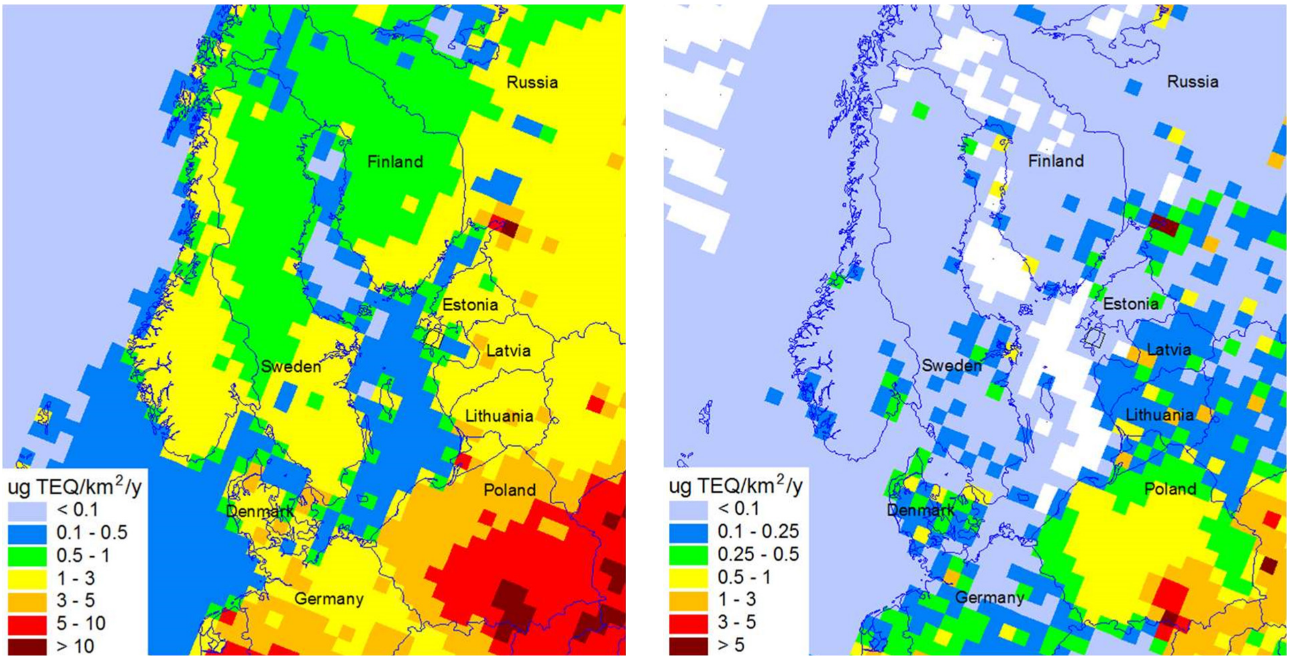


Figure 25. Annual deposition (left) and anthropogenic emissions (right) of dioxins and furans (PCDD/Fs) in the Baltic Sea region 2017 in µg TEQ/km2/year. From Gauss et al. 2020.

4.3. Atmospheric deposition of PCBs to the Baltic Sea

The deposition of PCB-153 to the Baltic Sea has been steadily decreasing since the early 1990's (Figure 26). The spatial pattern for the deposition as well as the anthropogenic emissions show the common strong south-to-north gradient, with highest levels in the western part of Europe (Figure 27). The emissions are dominated by the so-called Sectors A-C, i.e. "Public Power", "Industry", and "Other Stationary Combustion", respectively (Gauss et al. 2020).

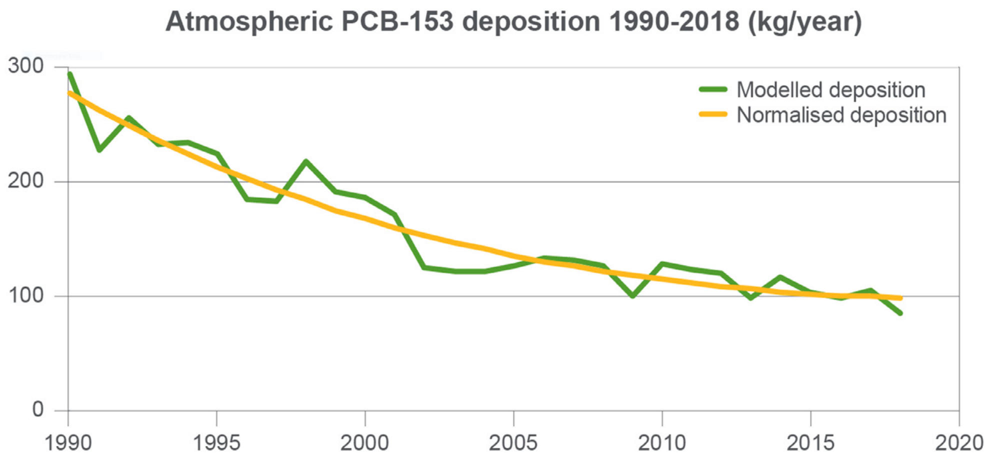


Figure 26. Modelled and normalised atmospheric PCB-153 deposition (kg per year) on the Baltic Sea 1990–2018. Data from EMEP (BSEFS 2020).

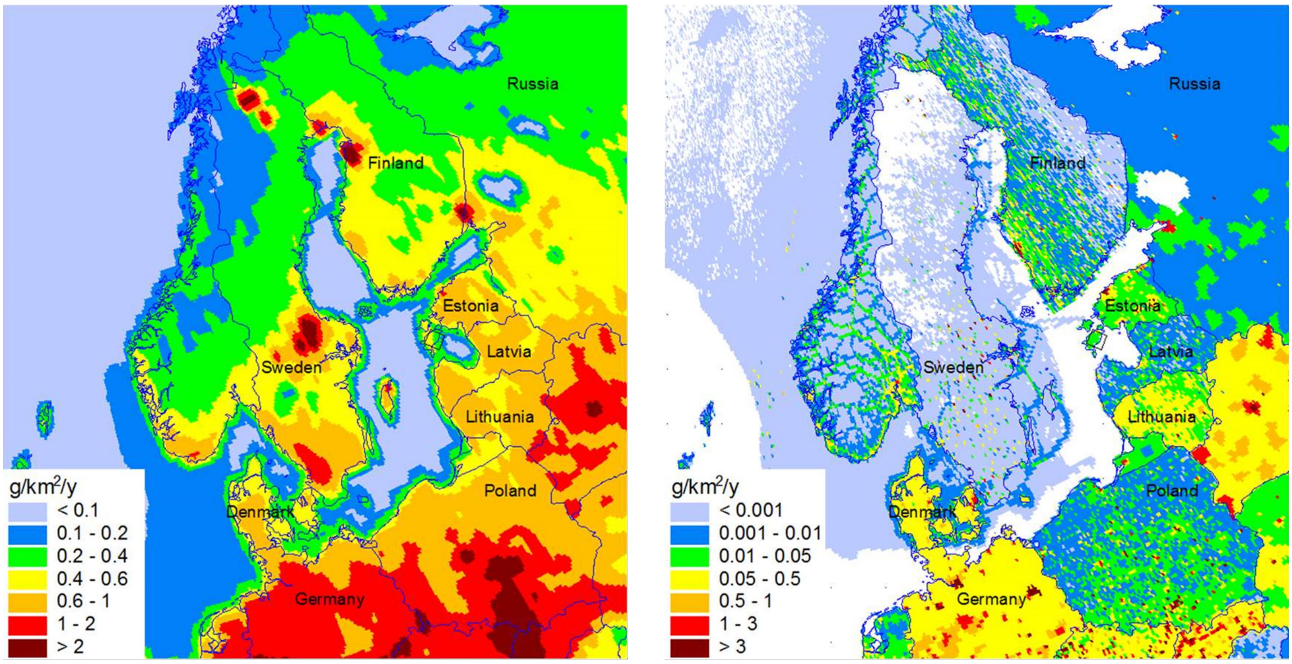


Figure 27. Annual deposition (left) and anthropogenic emissions (right) of Polychlorinated Biphenyls (PCBs) exemplified with the congener PCB-153 in the Baltic Sea region 2018 in g/km2/year. From Gauss et al. 2020.

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