

Inputs of hazardous substances to the Baltic Sea

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Hazardous substances



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Inputs of hazardous substances to the Baltic Sea



Hazardous substances, such as metals and various organic environmental pollutants, may originate from natural or anthropogenic sources, although organic pollutants tend to be most commonly of anthropogenic origin. Excessive levels of pollutants 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.

The monitoring and reporting guidelines for waterborne inputs of hazardous substances to the Baltic Sea (Water Borne Pollution Load Compilation (PLC-Water) guidelines) are to a large degree focused on metal inputs, whereas in the programme for monitoring of the pollution of air and precipitation, the airborne inputs include both metals and some organic pollutants. 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, summarised information on the atmospheric deposition of selected organic pollutants based on the data supplied to HELCOM by European Monitoring and Evaluation Programme (EMEP) are included. Also, to prepare for future monitoring and reporting of at least selected organic pollutants, a questionnaire was sent out to the Contracting Parties (CPs) in 2015, asking for identification of substances of concern as well as indications on data availability. For practical reasons, the questionnaire also included micropollutants in municipal waste water treatment plant (MWWTP) effluents, another closely related mission to cover. The results from this questionnaire are summarised below. Collection of data on the prioritised substances obtained by Contracting Parties will start in early 2018.

Input of heavy metals to the Baltic Sea

Heavy metals in rivers may originate from natural or anthropogenic sources, and excessive metal levels in surface waters may pose a health risk to humans and to the biota in the environment. In the Baltic Sea, high cadmium and mercury concentrations have been detected in sediments and fish tissue (HELCOM 2010). The main factors contributing to heavy metal inputs include soil properties, industrial activity, high population density, exploitation of minerals and other natural resources, application of fertilizers in agricultural areas, and atmospheric deposition from local and distant emission sources.

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, a clear picture of the heavy metal inputs entering the Baltic Sea could not be established in the fifth Pollution Load Compilation (PLC-5). As issues still remain regarding completeness and quality of data reported by some countries, as well as quantification of inputs of the metals with concentrations at the level of natural background **these results from the PLC-6 reporting ought to be seen as indicative only** (cf. “Data coverage” and “Data handling and quality control”). It should also be noted that the transboundary metal loads (from upstream countries) are included in the metal inputs to the Baltic Sea from the HELCOM CPs, 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”). On the other hand, the reporting obligations for municipal waste water treatment plants (MWWTPs) and industrial point sources are regulated by the size of the MWWTPs, and if the monitoring is a part of the permissions for a specific industrial plant. The PLC-Water guidelines indicate methods for making estimates from measurements below the quantification limits (HELCOM 2016).





Data coverage

The assessment of heavy metal inputs to the Baltic Sea has been focused on the period 2012–2014 as these are the years with the most complete data coverage in the HELCOM PLC database. Despite this the spatial coverage is far from complete, ranging from only 11% of total riverine catchment areas (Denmark, although only 2% for Hg) to fully covered CPs (Figure 1). Also, the coverage of heavy metal inputs from point sources are most 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 position of MWWTs and industrial point sources with reported metal loads are given in Figure 2.

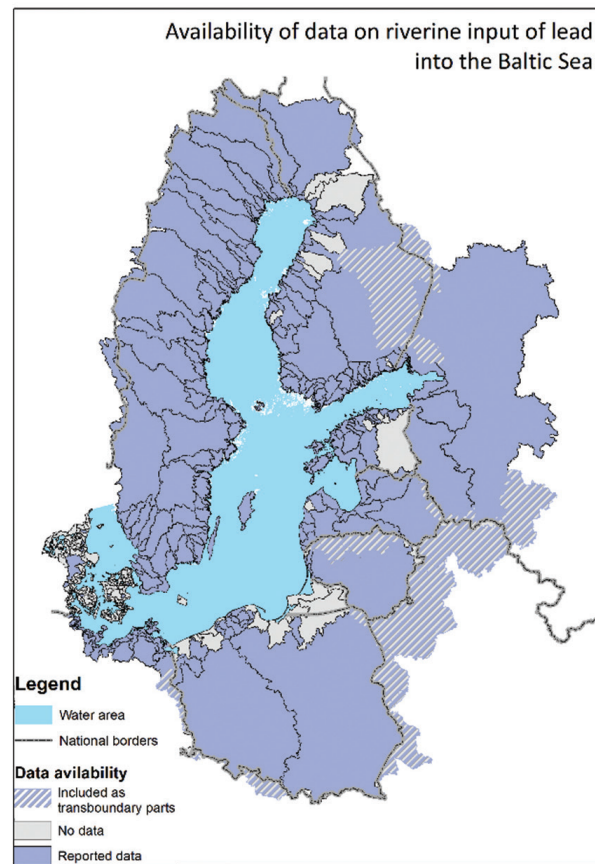
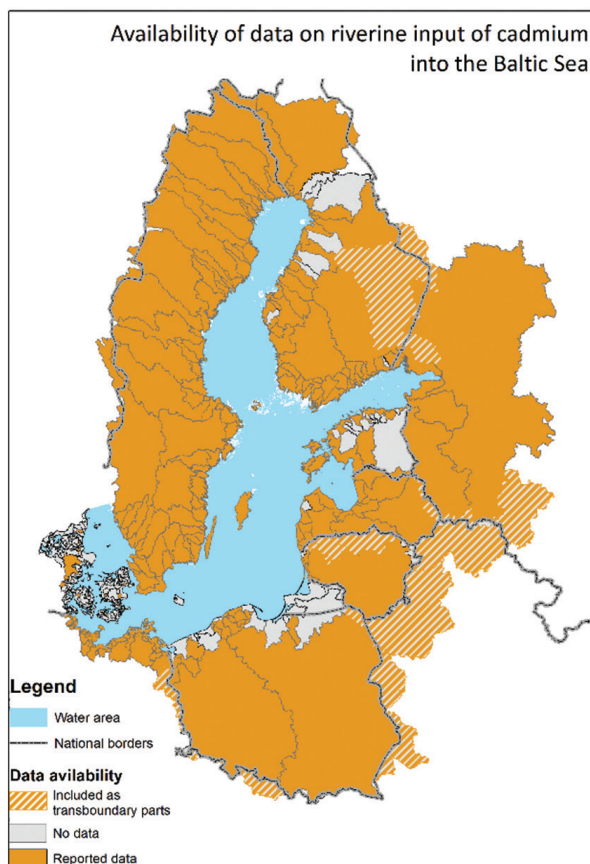
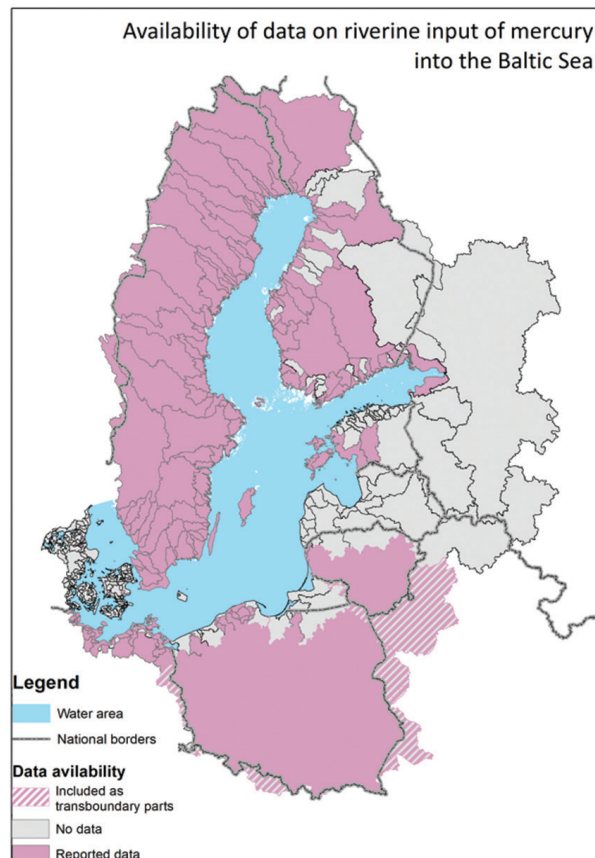


Figure 1.
The spatial data coverage of reported riverine inputs of Cd, Hg, and Pb to the Baltic Sea 2012–2014.

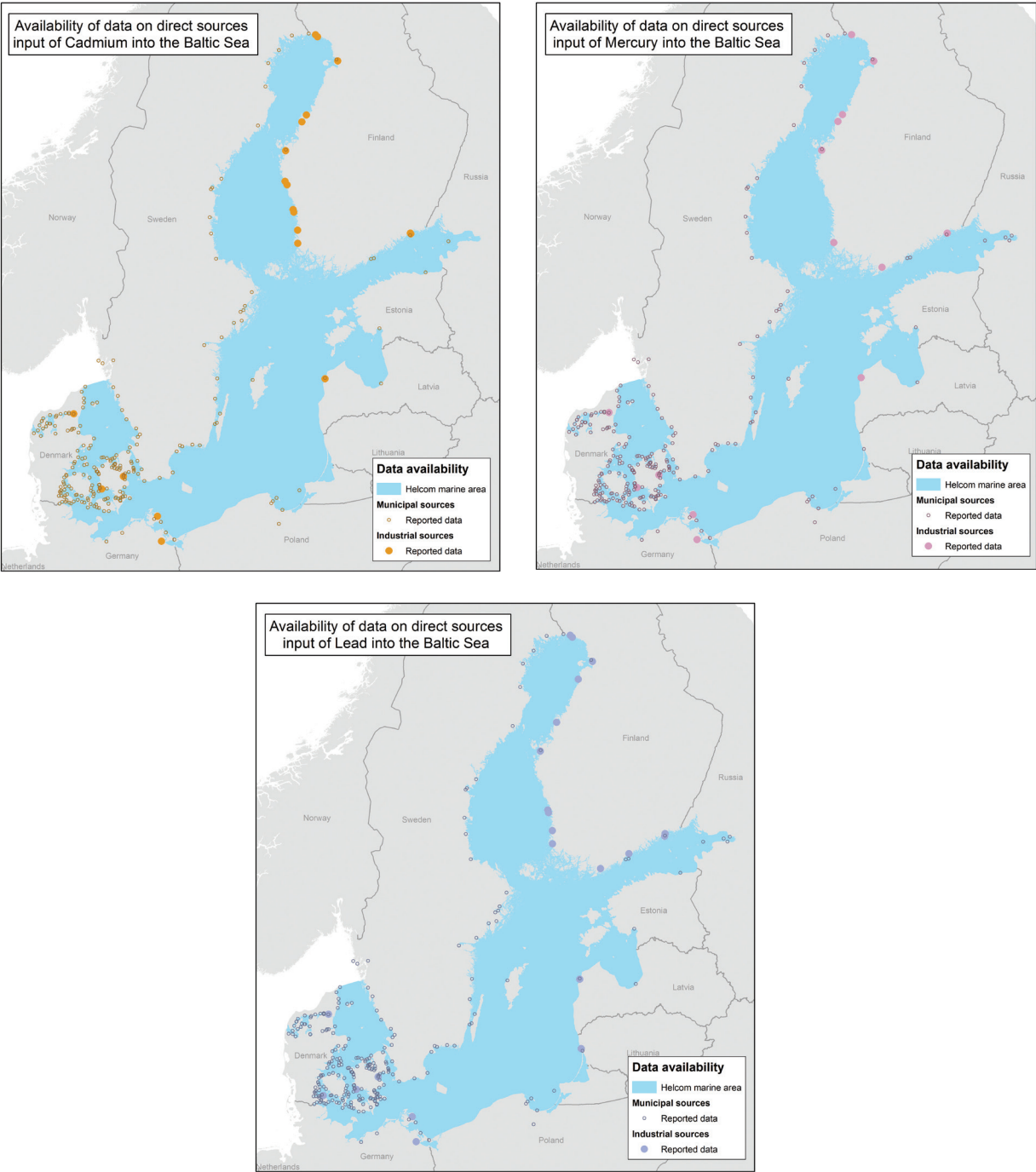


Figure 2.
Reported point sources with direct discharge of Cd, Hg, and Pb to the Baltic
Sea 2012–2014.



Data handling and quality control

Within the frame of the annual 'Pollution Load Compilations', metal data have been reported by the Contracting Parties. The reported data has been compiled and assessed as far as possible, although there are numerous issues regarding the temporal and spatial coverage for several CPs. Also, it has been hard to confirm the data quality, as there are some observations that appear to be suspiciously high or low in comparison to the inputs from other CPs. In addition, most CPs are analysing the metals based on filtered samples although the request according to the PLC-Water Guidelines are for the total loads. This is mainly an adaptation to the EU Water Framework Directive WFD that request data on biologically available metals. Among the HELCOM countries, only Estonia and Finland are actually measuring the total metal concentration (Estonia analyses both total and filtered metals), whereas Sweden is analysing acid soluble metals that include dissolved metals and metals adsorbed to particulate matter. Except from Lithuania, all other CPs are analysing filtered samples. For Lithuania there is a lack of information on the metal fractions analysed and reported. Consequently, the data reported and assessed in regard to the total metal inputs are in most cases an underestimate, since the metals associated with particulate matter are not included.

Total annual flow weighted average riverine concentrations have been calculated based on the annual loads and the average water flow as a part of the data quality control procedures in addition to comparing the input levels between different CPs. In cases

when the computed flow normalised concentrations are very low, close to or even below level of quantifications (cf. Table 1), the reported annual loads are considered as unrealistically low.

Loads that are considerably higher than might be expected are much harder to assess, as they may be result from estimates based on contaminated samples or they can be result from using too high levels of quantification in the laboratory analysis of samples. The estimates will in these cases be extra sensitive if the common procedure to use LOQ/2 is used to estimate levels below the limit of quantification (LOQ), especially if the estimate is based on a large proportion of observations with levels below the limit. A more realistic approach is recommended in the PLC- Water Guidelines (HELCOM 2015), but this procedure is not always used by CPs. Russian input estimates in particular are believed to over-estimate the actual input due to this reason (Oblomkova, Pers. Comm.). 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 in their riverine samples. For instance, Denmark only analyses metals based on filtered river samples, which of course are not completely comparable with the total concentrations from other CPs (as required in the PLC-Water Guidelines). Also, Denmark has only been monitoring the riverine metal loads in twelve of their numerous small rivers during the period 2012-2014 (only eight for Hg), and no river has been monitored for more than a single year. Hence, to be able to include the Danish riverine loads all twelve rivers have been set for 2014 although some were sampled in 2012 or 2013.

Box 1.

Table 1.
Limits of quantification (LOQ) for metals in river water (µg/l). From PLC 5.5 (HELCOM 2015).

Metal	Guideline	DE	DK	EE	FI	LT	LV	PL	RU	SE
Cd	0.01	0.02-0.06	0.015	0.02-0.05	0.01	0.05	0.2-0.3	0.1	0.1	0.005
Hg	0.005	0.001-0.005	0.015	0.015-0.1	0.002	0.03	0.21	0.013	0.01	0.0001
Pb	0.05	0.04-0.2	0.1	0.1-1	0.01	1.0	1.3-1.4	1.0	2	0.02



Box 2.

Table 2.

Area-specific riverine inputs of Cd, Hg, and Pb from HELCOM CPs 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. Questionably low area-specific inputs are marked with red text.

CP	Cd (kg/km ²)			Hg (kg/km ²)			Pb (kg/km ²)			Area	Coverage (%)
	2012	2013	2014	2012	2013	2014	2012	2013	2014		
DE	0.00434	0.00424	0.00286		0.00143	0.00029		0.04157	0.02925	23276	81
DK			0.00320			0.00013			0.03283	3575 ^a	11
EE	0.05739	0.00103	0.00112	0.00583	0.00127	0.00132	0.31809	0.05625	0.02412	46329	100
FI	0.00864	0.00524	0.00567	0.00139	0.00095	0.00080	0.13411	0.08616	0.07387	316941	100
LT			0.00030		0.00103	0.00098			0.00722	47349	73
LV			0.00443						0.14257	65874	100
PL	0.00619	0.00570	0.00399	0.00181	0.00181	0.00179	0.13858	0.14078	0.03926	304801	98
RU	0.09806	0.05902	0.03515			0.00069	0.75608	0.81098	0.61837	294015	93
SE	0.00539	0.00329	0.00371	0.00115	0.00064	0.00069	0.1362	0.06349	0.07648	454259	100

^a Only 711 km² for mercury equivalent to 2% of the total area.

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, though there are some suspiciously low inputs, which when taking the earlier reported LOQs into consideration (see above), most probably are not correct (Table 2). As stated earlier, the Russian inputs of cadmium and lead are most certainly over-estimated due to many observations below the comparably high LOQs, and as the input estimates are based on half the LOQs the estimates are deemed to be over-estimated.

The metal input data from the last three years are in any case believed to be the best estimates

of the inputs as the data coverage is generally better compared with earlier years, but also the data quality appear to be superior. 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 have been censored in the assessment, mainly due to inconsistent reporting coverage. Also, in the compilation of metal inputs to the different Baltic Sea basins, it has not been possible to get full coverage for some of the southernmost basins, mainly due to the limited riverine load data from Denmark.



Total inputs of cadmium, mercury, and lead to the Baltic Sea 2012–2014

There are quite large differences in the total amounts of the different metals that enter the Baltic Sea every year. Furthermore, the main route of entry is quite variable between the 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 23–45, 4.8–5.6, and 443–565 tonnes per year, respectively (Table 3). Of these assessed metals, Mercury is characterised to be entering the Baltic Sea via atmospheric deposition to a major degree, constituting around

70% of the total inputs (Figure 3). For lead and cadmium it is riverine inputs that make up about the same proportion of the total input (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), although the point sources might be underestimated somewhat (for example Sweden only can report metal inputs from the larger MWWTs, as the smaller plants seldom have reporting obligations on metals in their permits). In any case, the importance of the direct point sources may be regarded to be considerably less than the other two routes of entry.

Box 3.

Table 3.
Inputs of cadmium, mercury and lead to the Baltic Sea from direct point sources, via rivers, and atmospheric deposition 2012–2014.

Source	Cd (tonnes/year)			Hg (tonnes/year)			Pb (tonnes/year)		
	2012	2013	2014	2012	2013	2014	2012	2013	2014
Direct point sources	0.09	0.19	0.05	0.04	0.48	0.10	0.96	1.13	1.27
Riverine	39	22	15	1.8	1.3	1.2	386	341	264
Total waterborne	39	23	16	1.8	1.8	1.3	386	342	265
Deposition ^a	6.2	5.7	7.5	3.7	3.1	3.4	179	177	177 ^b
Total	45	28	23	5.6	4.9	4.8	565	520	443

^a Deposition data from EMEP (BSEFS 2015, and BSEFS 2016).

^b Lead deposition for 2014 has not been estimated by EMEP, and the deposition in 2013 has been used as an estimate.

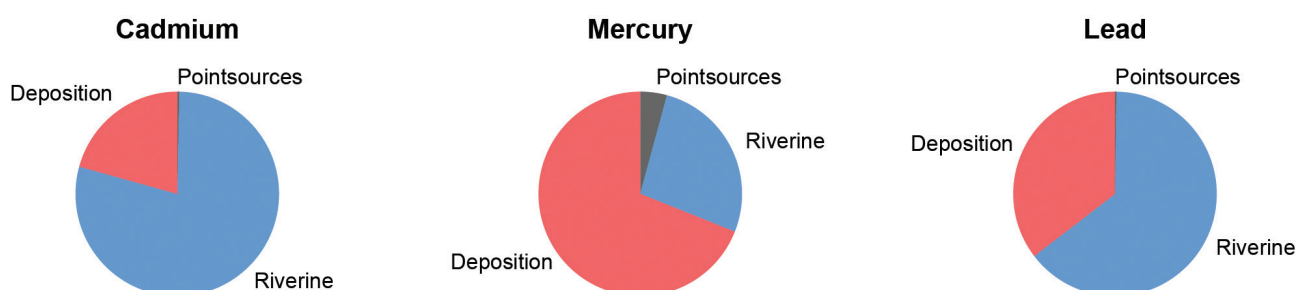


Figure 3.
The importance of inputs of cadmium, mercury and lead from point sources, via rivers, and atmospheric deposition to the Baltic Sea based on average inputs 2012–2014.



Inputs of cadmium, mercury, and lead via rivers and point sources 2012–2014

As stated earlier, the importance of point source inputs for the Baltic Sea is quite low compared to riverine inputs and inputs via deposition from the atmosphere. 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 the number of inhabitants for example. The allocation of point sources inland compared to direct point sources entering 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 the generally high particle affinity of metals.

Box 4.

Table 4.
Riverine inputs of cadmium, mercury and lead to the Baltic Sea 2012–2014.

CP	Cd (tonnes/year)			Hg (tonnes/year)			Pb (tonnes/year)			Flow (m³/s)		
	2012	2013	2014	2012	2013	2014	2012	2013	2014	2012	2013	2014
DE	0.1009	0.0987	0.0666	0.0178	0.0332	0.0068	1.86	0.97	0.68	97	101	71
DK			0.0019			0.0001			0.04			7.4
EE	2.6587	0.0477	0.0518	0.2700	0.0588	0.0613	14.74	2.61	1.12	312	202	164
FI	2.7369	1.6594	1.7965	0.4390	0.3010	0.2528	42.51	27.31	23.41	3519	2612	2519
LT			0.0144		0.0488	0.0463			0.34	515	570	407
LV			0.2919						9.39	1258	1007	661
PL	1.8856	1.7387	1.2149	0.5514	0.5502	0.5447	42.24	42.91	11.97	1542	1884	1608
RU	28.8320	17.3527	10.3350			0.0071	222.3	238.4	181.8	3594	4030	3184
SE	2.4473	1.4945	1.6851	0.5234	0.2922	0.3145	61.87	28.84	34.74	7309	5327	5675
Total	39	22	15	1.8	1.3	1.2	386	341	264	18147	15732	14298

Note!

^a Estimates from Denmark are based on filtered samples from in total twelve rivers 2012–2014 covering only few percent of the Danish catchment area to the Baltic Sea.

^b The interannual variation in Estonian data is notable, mainly due to many observations below the LOQ used.

^c The spatial and/or temporal coverage of load data from EE, LT and LV are not complete. ^d Inputs from Russia are probably overestimated due to the estimation methods based on high LOQ's



Box 5.

Table 5.
Inputs of cadmium, mercury and lead to the Baltic Sea from point sources 2012–2014.

CP	Cd (tonnes/year)			Hg (tonnes/year)			Pb (tonnes/year)			Flow (m³/s)		
	2012	2013	2014	2012	2013	2014	2012	2013	2014	2012	2013	2014
DE	0.0003	0.0003	0.0003	0.0001	0.0002	0.0002	0.0091	0.0025	0.0043	2.7	2.0	2.7
DK			0.0234			0.0253			0.6143			11.4
EE	0.0002		0.0017	0.0002		<0.0001			0.0042	2.5	2.0	1.8
FI	0.0550	0.1036		0.0097	0.0359	0.0413	0.4768	0.6341	0.3583	22.8	21.9	21.4
LT				0.0015	0.0004	0.0013	0.0040	0.0800	0.0018	0.8	0.6	0.6
LV	0.0063	0.0148	0.0079	0.0068	0.0119	0.0050	0.2168	0.1412	0.1009	2.1	2.1	1.9
PL	0.0057	0.0552	0.0020	0.0006	0.4064	0.0045	0.0557	0.0788	0.0254		2.4	5.2
RU	0.0016		0.0002				0.0057	0.0037	0.0003	26.3	27.3	27.7
SE	0.0217	0.0155	0.0146	0.0234	0.0212	0.0213	0.1930	0.1879	0.1601	21.5	18.9	19.9
Total	0.09	0.19	0.05	0.04	0.48	0.10	0.96	1.13	1.27	79	77	93

Total inputs of mercury, cadmium and lead per basin 2012–2014

A basin-wide assessment of the waterborne (riverine plus direct point sources) metal inputs was only possible for some of the Baltic Sea basins (Table 6). For the southernmost basins, the lack of total load estimates for Denmark made it impossible to make comparisons with the other basins, and consequently no data is presented. 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). As stated earlier, there is concern about the reliability of Russian estimates, due to the considerable number of observations

less than the LOQs, but the large inputs are also a consequence of the very large flow of riverine water, mainly from the River Neva, entering the Gulf via Russia. The over-estimated inputs caused by the problem with high LOQs could be avoided in future assessments, if Russia applies more sensitive analytical methods. 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 reasonably comparable, except for the Archipelago Sea, which only receives about one tenth of the total amount, compared to the other basins. However, considering that this basin is quite small and sparsely populated, the metal inputs are not insignificant.



Box 6.

Table 6.

Total waterborne inputs of cadmium, mercury and lead to the Baltic Sea basins 2012–2014.
The total catchment area for the basins is also given (rounded to the closest 1000 km², data from PLC-5).

Basin	Cd (tonnes/year)			Hg (tonnes/year)			Pb (tonnes/year)			Area
	2012	2013	2014	2012	2013	2014	2012	2013	2014	
BOB	2.2696	1.3074	1.3393	0.5215	0.2944	0.2754	40.19	18.02	14.66	261000
BOS	1.6345	1.073	1.1651	0.2824	0.1493	0.1559	25.39	13.95	14.15	221000
ARC	0.1265	0.1011	0.1089	0.0044	0.0266	0.0249	5.61	5.43	6.32	9000
BAP	2.4445	2.1023	1.6210	0.7385	1.0059	0.6500	52.70	48.89	20.98	575000
GUF	31.4701	17.6009	10.4862	0.0638	0.0823	0.0511	239.4	245.8	186.5	413000
GUR	0.3758	0.0304	0.2828	0.2628	0.0580	0.0605	9.75	2.35	9.33	135000
WEB										23000
SOU										5000
KAT										87000

Note!

^a Estimate based on EE data from 2012–2014, and LV data from 2014.

^b The 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).

Waterborne inputs of cadmium, mercury, and lead to the Baltic Sea 1995–2014

Overall, most CPs show substantial inter-annual variability in metal inputs to the Baltic Sea during the 20-year period 1995–2014 (Figures 4–6). Complete data series for all three metals are only available for Germany and Sweden. For cadmium and lead Poland and Russia have complete datasets. For the other CPs either data is totally lacking for some years, or in some cases there are considerable problems with the spatial and/or temporal data coverage. Denmark has only been able to report data for point sources and for a total of twelve rivers for the period 2012–2014. This data is shown as 2014 data to reduce the scarcity as much as possible, although some samples were taken in 2012–2013. For mercury in particular there is quite a lot of missing data in many time series. The mercury data for Latvia and Russia is too scattered to be shown at all (Figure 3). Due to these data issues,

the assessment of the overall waterborne inputs over time can only be carried out with great caution, particularly 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 (Germany, Finland, and Sweden) is a general reduction in waterborne inputs or at least stabilising of inputs levels over time for all three metals (Figures 2–4). 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 4 and 6). One exception might be the Polish lead inputs that appear to be reducing over this period (Figure 4). Regarding the mercury inputs, all CPs (except the already stated inputs for Germany, Finland and Sweden), have data that is too variable and/or scarce to reveal any tendencies (Figure 5).





Cadmium (tonnes/year)

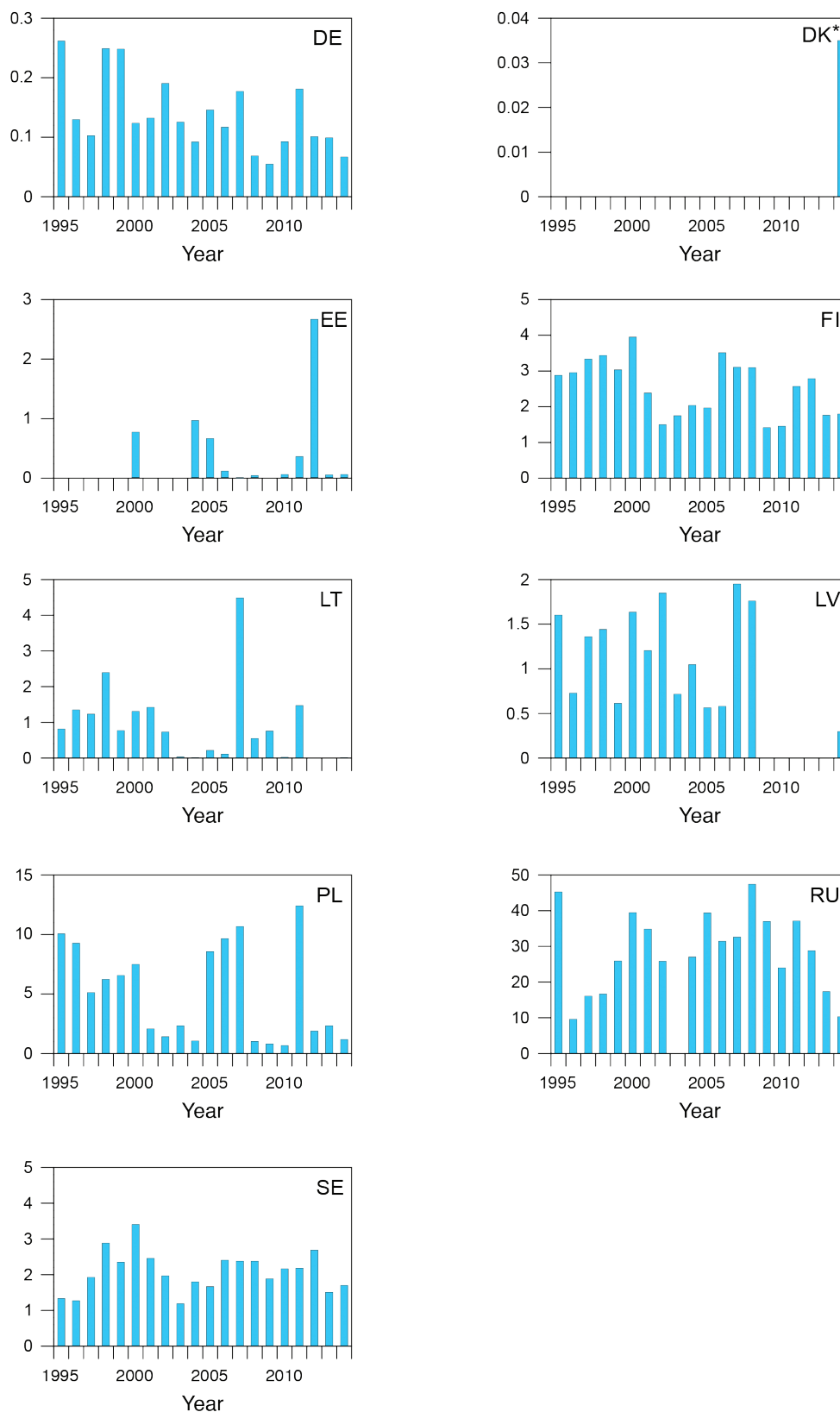


Figure 4. The annual waterborne inputs of cadmium from the Contracting Parties to the Baltic Sea (tonnes per year) 1995-2014. The bars show the sum of inputs from rivers and direct point sources. Note! The load from Denmark is based on only twelve rivers 2012-2014. Large inter-annual variability may be due to differences in the number of sources between years, but also on estimate methods used when observations are less than LOQ.



Mercury (tonnes/year)

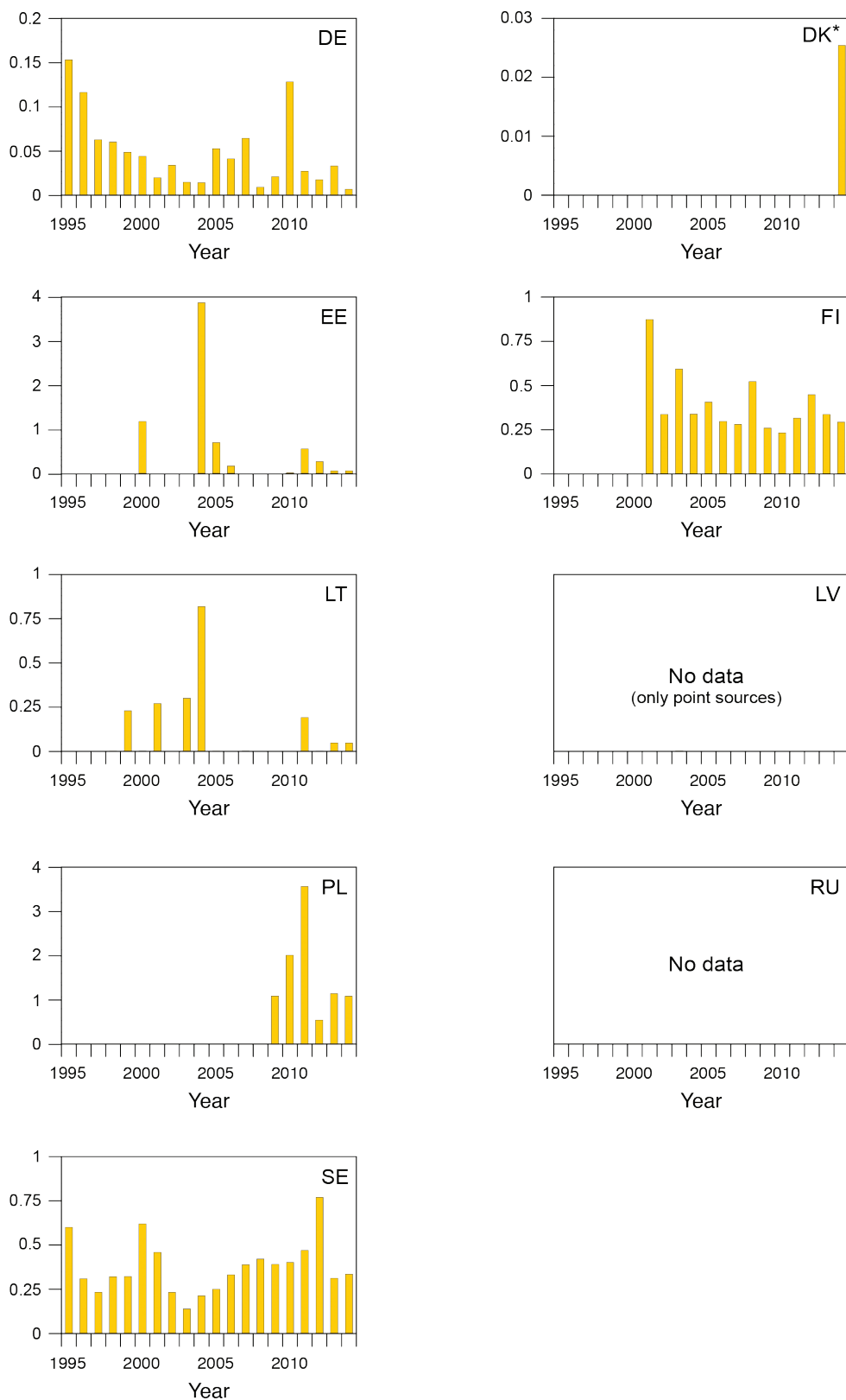


Figure 5.

The annual waterborne inputs of mercury from the Contracting Parties to the Baltic Sea (tonnes per year) 1995–2014. The bars show the sum of inputs from rivers and direct point sources. Note! Inputs from Denmark are based on only eight rivers 2013–2014. The very limited data for Latvia and Russia is not shown in the figure. 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.



Lead (tonnes/year)

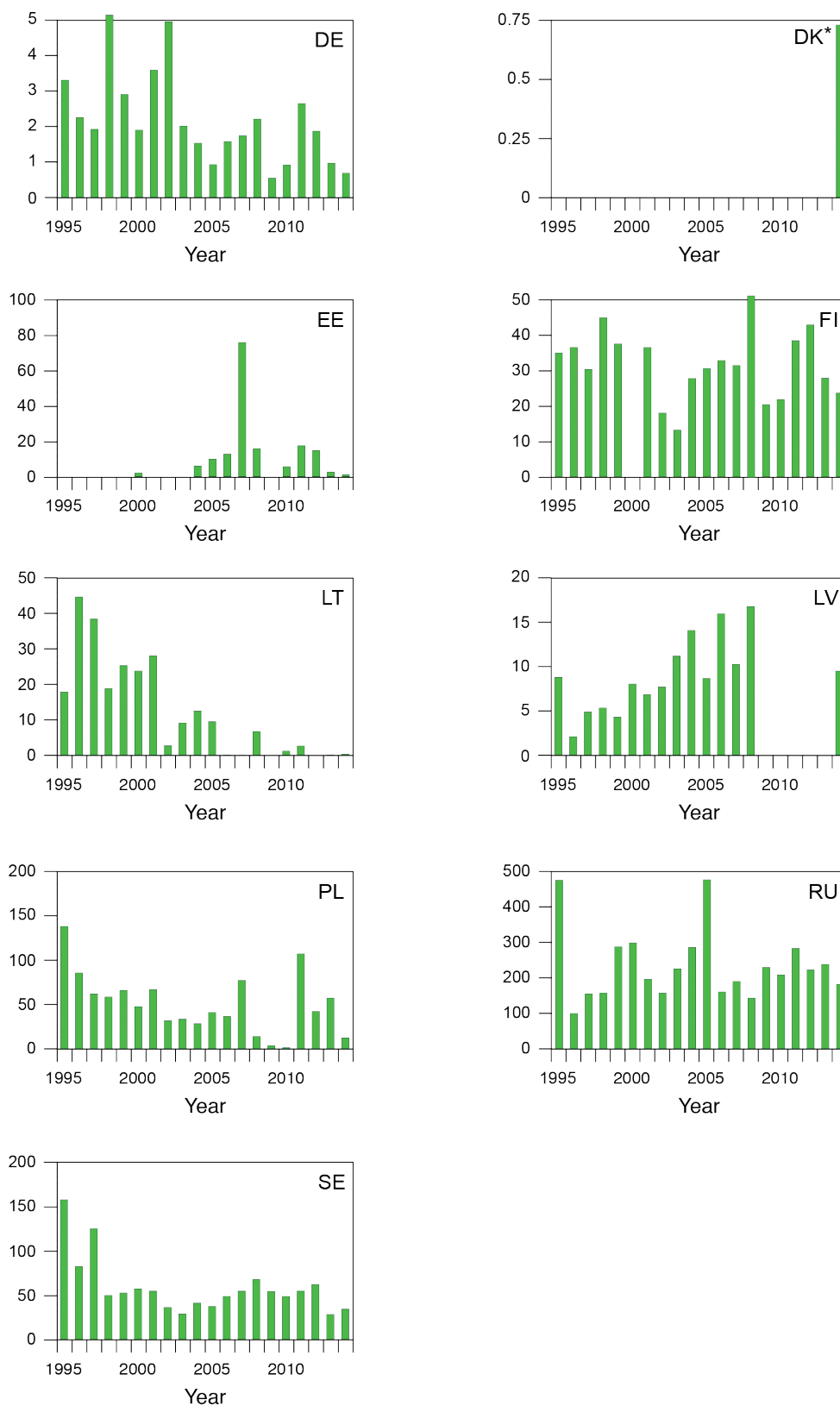


Figure 6.

The annual waterborne inputs of lead from the Contracting Parties to the Baltic Sea (tonnes per year) 1995–2014. The bars show the sum of inputs from rivers and direct point sources. Note! The load from Denmark is based on only twelve rivers 2012–2014. 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.



Atmospheric deposition of cadmium, mercury and lead

The modelled atmospheric deposition of all three metals are reducing over time from the start of the time series in 1990 up to the present (2014 for cadmium and mercury, 2013 for lead that is not assessed by EMEP for HELCOM every year). This is valid for both the annual deposition as well as the weather-normalised annual deposition (Figures 7-9). The cadmium and lead deposition are reducing notably more (60%, and 80% respectively) than the mercury deposition (15%). According to as-

essment of 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 economic contraction and industrial restructuring in Poland, Estonia, Latvia, Lithuania, and Russia in early 1990s. The other CPs had their major emission reductions previously, before the start of the time series. However, the considerably lower reduction rate in mercury deposition (Figure 6) is probably due to the influence of the much larger long-range transport, which makes mercury be considered as a global pollutant (cf. Ilyin *et al.* 2016).

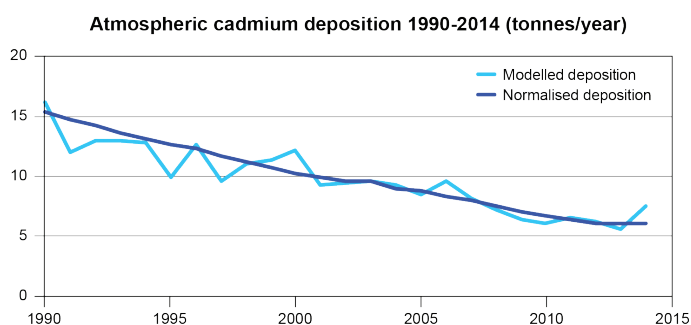


Figure 7.
Modelled and normalised atmospheric cadmium deposition (tonnes/year) on the Baltic Sea 1990-2014. Data from EMEP (Bartnicki *et al.* 2016).

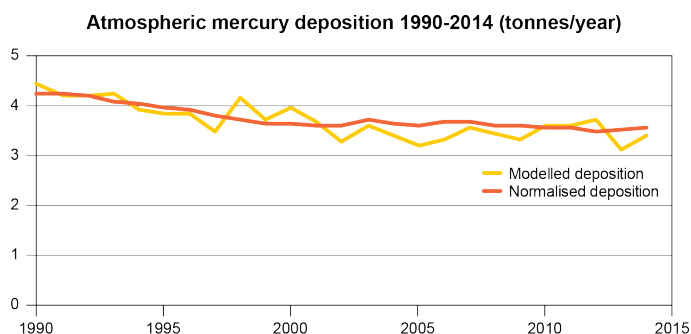


Figure 8.
Modelled and normalised atmospheric mercury deposition (tonnes/year) on the Baltic Sea 1990-2014. Data from EMEP (Bartnicki *et al.* 2016).

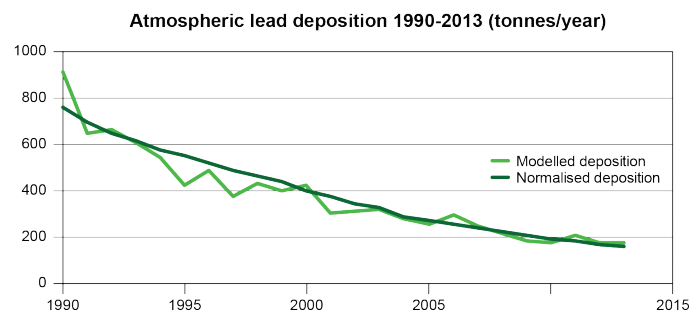


Figure 9.
Modelled and normalised atmospheric lead deposition (tonnes/year) on the Baltic Sea 1990-2013. Data from EMEP (Shamsudheen *et al.* 2015). Note! Lead deposition is not modelled by EMEP every year. Hence, the data only covers up to 2013.



The spatial resolution of the modelled metal deposition and emissions in the Baltic Sea region reveal a strong south to north gradient in general, with both higher emissions and depositions in the southern part of the catchment area compared to the northern

part (Figures 10-12). In addition to this gradient there are also markedly elevated emissions in Poland, although relatively smaller “hot-spots” also occur in other CPs.

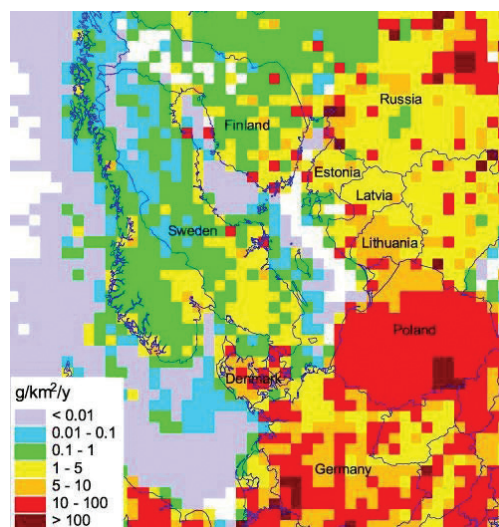
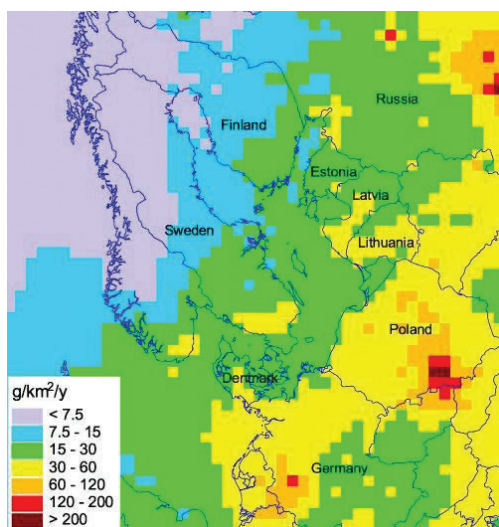


Figure 10.
Total annual cadmium deposition (left) and anthropogenic emissions (right) in the Baltic Sea region 2014 in g/km²/year. From Bartnicki et al. (2016).

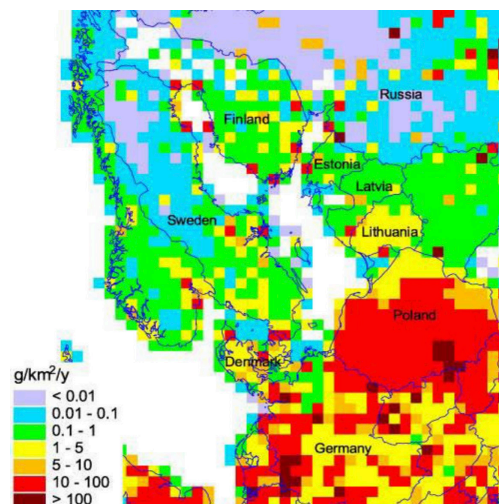
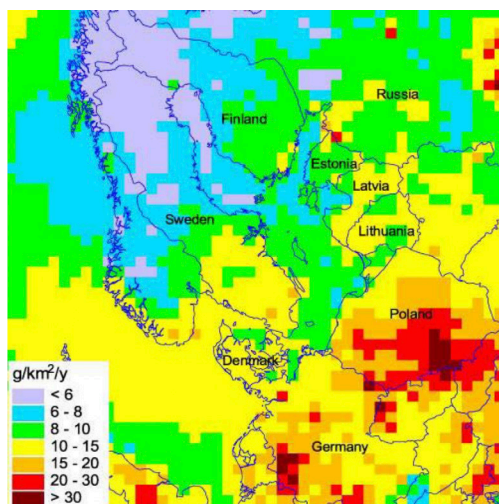


Figure 11.
Total annual mercury deposition (left) and anthropogenic emissions (right) in the Baltic Sea region 2014 in g/km²/year. From Bartnicki et al. (2016).

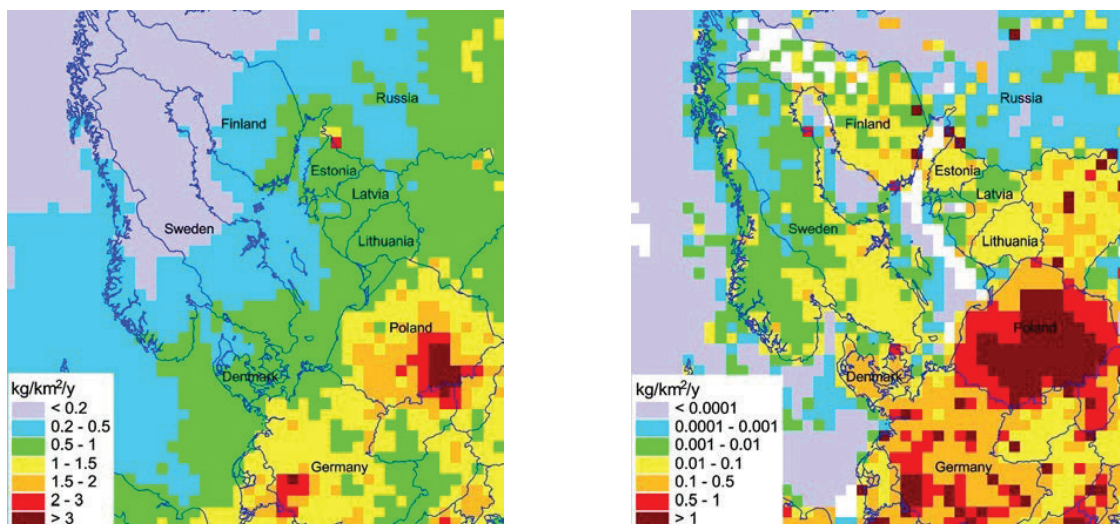


Figure 12.

Total annual lead deposition (left) and anthropogenic emissions (right) in the Baltic Sea region 2013 in kg/km²/year. Note! Data coverage is 2013 as EMEP do not assess lead emissions and deposition every year for HELCOM. From Shamsudheen *et al.* (2015).



Atmospheric deposition of selected organic pollutants

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

After an initial decrease in atmospheric deposition of Benzo(a)pyrene to the Baltic Sea in the early 1990's, the level has been quite stable on an annual basis (Figure 13). The spatial pattern of both the deposition and the anthropogenic emissions are rather similar to the patterns observed for 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 14). The emissions are heavily dominated by the so-called Sector C "Other Stationary Combustion" (EMEP 2016).

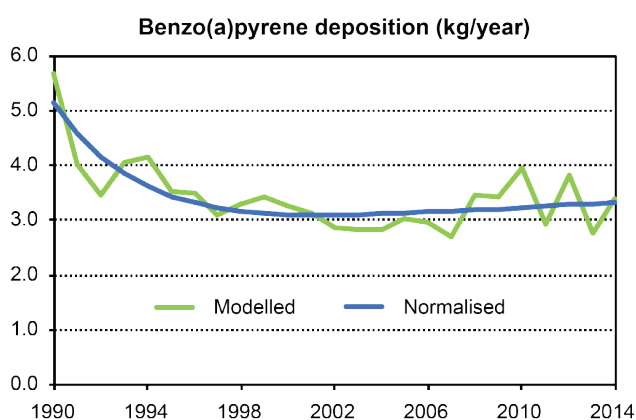


Figure 12.
Total annual lead deposition (left) and anthropogenic emissions (right) in the Baltic Sea region 2013 in kg/km²/year. Note! Data coverage is 2013 as EMEP do not assess lead emissions and deposition every year for HELCOM. From Shamsudheen et al. (2015).

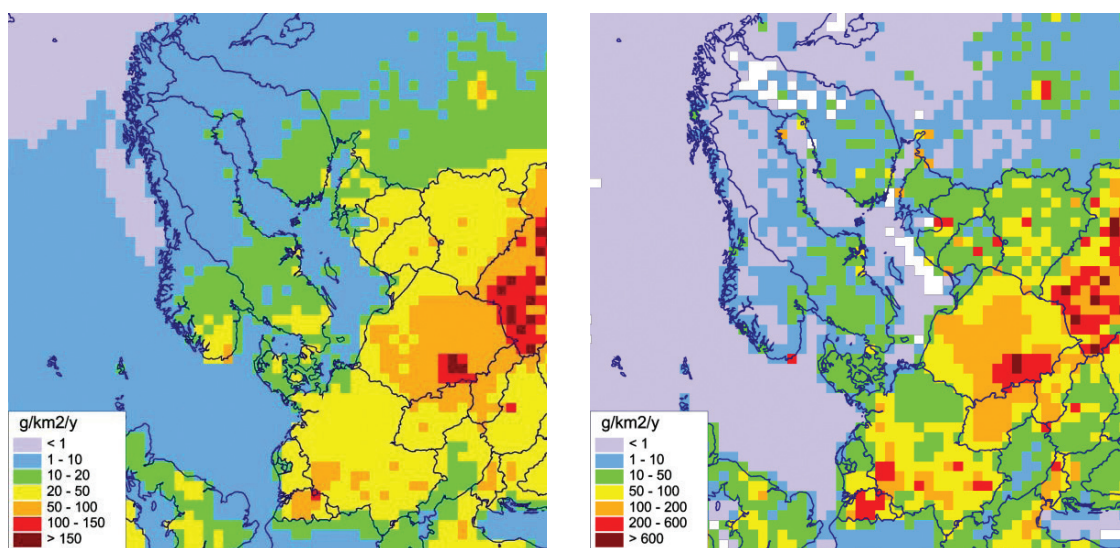


Figure 14.
Total annual Benzo(a)pyrene deposition (left) and anthropogenic emissions (right) in the Baltic Sea region 2014 in g/km²/year. From Bartnicki et al. (2016).



Atmospheric deposition of polybrominated diphenyl ethers (PBDEs) to the Baltic Sea

In test modelling by EMEP the spatial pattern of BDE-99 deposition and anthropogenic emissions was found to slightly differ from the benzo[a]pyrene B(a)P and the metals as, except from the common south-to-north gradient, the highest levels in the south are to be found in the western part of Europe (Figure 15).

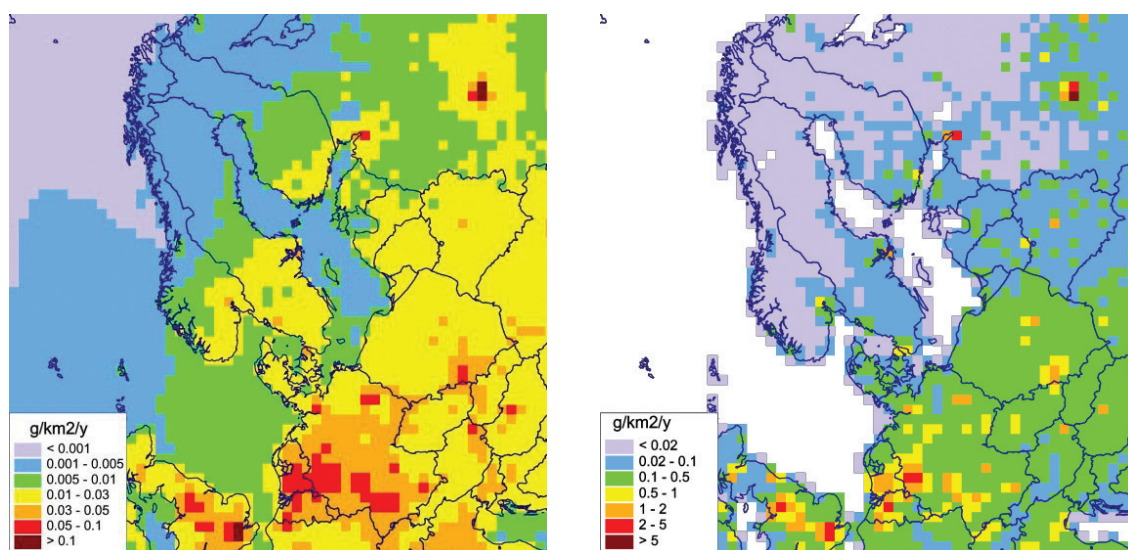


Figure 15. Annual deposition (left) and anthropogenic emissions (right) of Polybrominated diphenyl ethers (PBDEs) exemplified with the indicator congener BDE-99 in the Baltic Sea region 2000 in g/km²/year. From Bartnicki et al. (2016).

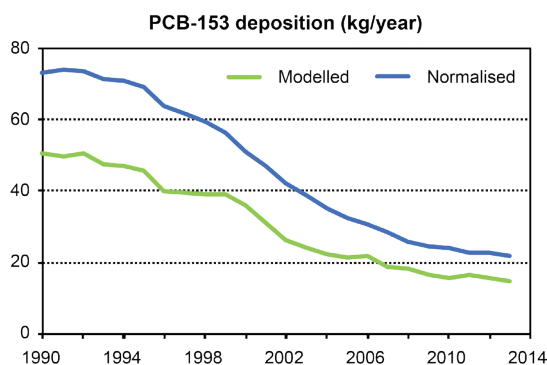


Figure 16.
Modelled and normalised atmospheric PCB-153 deposition (kg/year) on the Baltic Sea 1990–2013. Data from EMEP (Shamsuddeen et al. 2015).

Atmospheric deposition of polychlorinated biphenyls (PCBs) to the Baltic Sea

The deposition of PCB-153 to the Baltic Sea has been steadily decreasing since the early 1990's (Figure 16). The spatial pattern for the deposition as well as the anthropogenic emissions of PCB-153 resemble the pattern for BDE-99 (PBDEs), with highest levels in the western part of Europe, as well as the common strong south-to-north gradient (Figure 17).

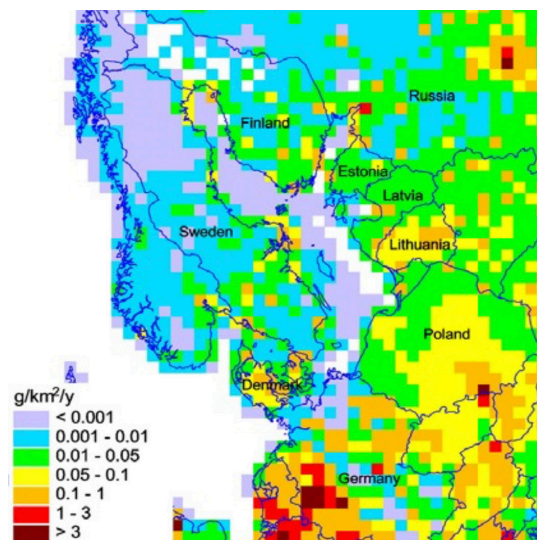
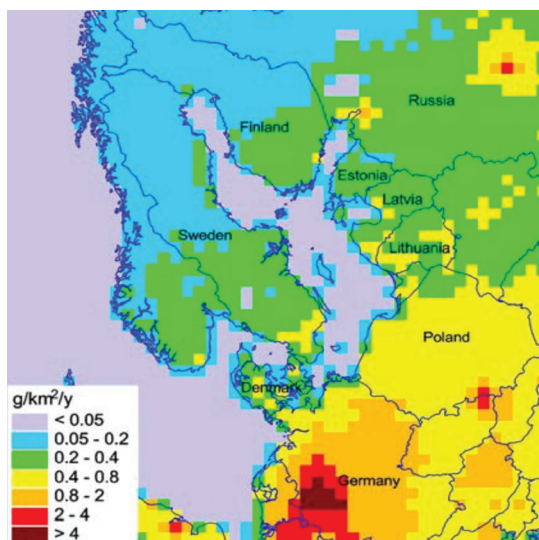


Figure 17.
Annual deposition (left) and anthropogenic emissions (right) of Polychlorinated Biphenyls (PCBs) exemplified with the congener PCB-153 in the Baltic Sea region 2013 in g/km²/year. From Shamsuddeen et al. (2015).



Atmospheric deposition of PCDD/Fs to the Baltic Sea

The deposition of PCDD/Fs to the Baltic Sea has decreased by 67% since the early 1990's (Figure 18). The spatial pattern for the deposition as well as the anthropogenic emissions of PCDD/Fs resemble the pattern for the metals (Figures 8-10), with highest levels in the southern part of the catchment area, as well as the common strong south-to-north gradient (Figure 19).

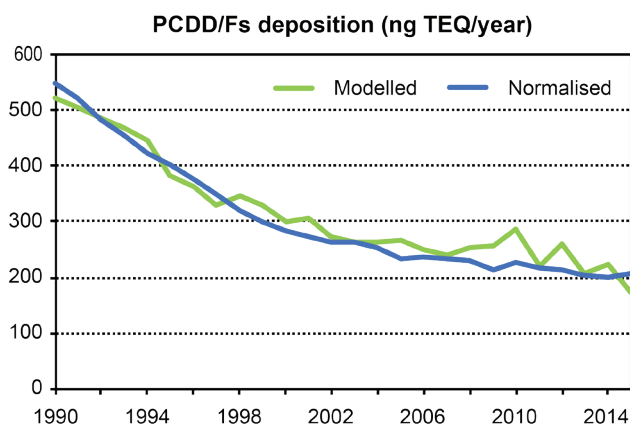


Figure 18. Annual deposition of Polychlorinated dibenzodioxins and dibenzofurans (PCDD/Fs) in the Baltic Sea region 1990–2015. Deposition in Toxic equivalent mass (ng TEQ/year). Data from Bartnicki et al. (2017).

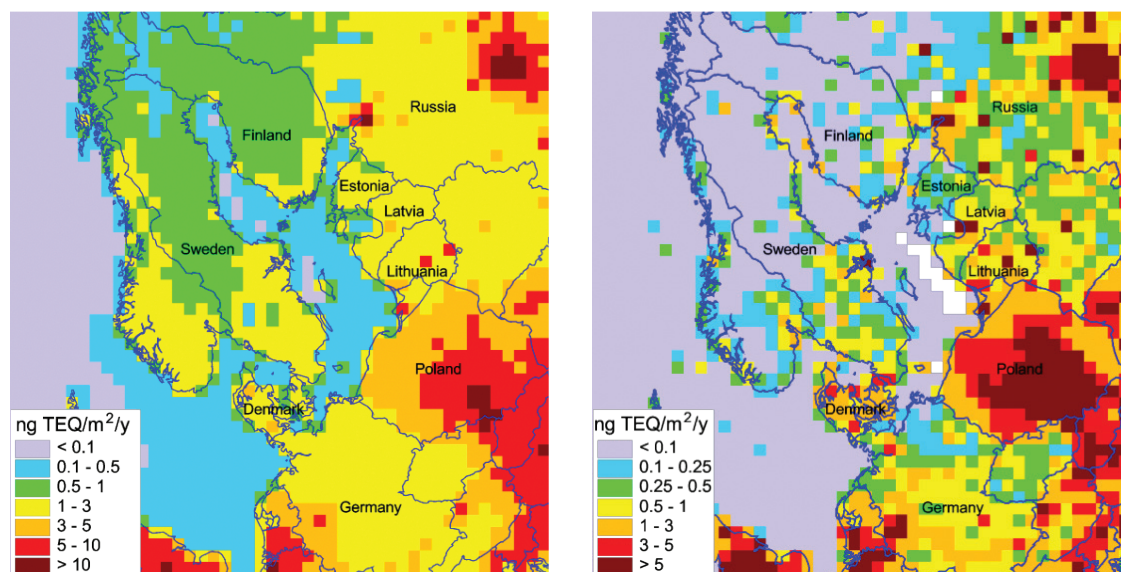


Figure 19. Annual deposition 2015 (left) and anthropogenic emissions 2012 (right) of Polychlorinated dibenzodioxins and dibenzofurans (PCDD/Fs) in the Baltic Sea region in Toxic equivalent mass (ng TEQ/year). From Bartnicki et al. (2017).



Pharmaceutical residues

In 2015 a joint project between HELCOM and the Policy Area Hazards within the EU Strategy for the Baltic Sea Region was initiated as a first attempt to provide a comprehensive regional overview of the extent of inputs of pharmaceuticals to the freshwater and marine environment in the Baltic Sea region, as well as to estimate contamination of the marine environment. The project has made a first assessment based on information on pharmaceutical residues in the Baltic Sea as well as in rivers entering to the Sea, and data on the use of pharmaceutical compounds in the Baltic Sea region. At a later stage UNESCO also took part in the assessment, and the final report was published in 2017 as a report within UNESCO Emerging Pollutants in Water Series (No 1, Pharmaceuticals in the aquatic environment of the Baltic Sea region – A status report) as well as a Baltic Sea Environmental Proceedings (HELCOM 2015).

The assessment is based on data covering the period 2003-2014. In total 47,600 observations on sources and pathways of pharmaceuticals (for example information from wastewater influents and effluents, sludge and river water) and 4,600 observations of pharmaceuticals in the Baltic Sea. The report includes data on 167 different phar-

maceutical substances analysed in the marine environment and 156 pharmaceutical substances and 2 metabolites analysed in surface freshwaters and in influents, effluents and sludge of municipal wastewater treatment plants (MWWTPs). The most frequently observed substances in the Baltic Sea marine environment belong to the therapeutic groups of anti-inflammatory and analgesics, cardiovascular and central nervous system agents.

The main sources of pharmaceuticals in the freshwater and marine environment in the Baltic Sea region was found to be the excretion of active substances consumed by humans and animals via their faeces and urine. Hence, the main pathway of pharmaceutical residues into the freshwater and marine environment, according to the data collected within the project, is via the MWWTPs effluents. A rough estimate gives that the annual release from MWWTPs to the environment is about 1.8 thousand tons 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%.

The work on pharmaceutical residues in the Baltic Sea region that started with this project will be continued, as this is a prioritized area within the HELCOM work on hazardous substances, as well within the Policy Area Hazards of the European Union Strategy for the Baltic Sea Region (EUSBSR). In early 2018 the Baltic Sea Pharma Platform (Baltic Sea Region cooperation platform to reduce pharmaceuticals in the Baltic Sea environment) was launched, and the joint efforts on the matter within HELCOM and Policy Area Hazards of the EU Strategy for the Baltic Sea Region will be performed within this new cooperative platform.



Persistent organic pollutants and other substances of concern in the Baltic Sea area – results from a questionnaire to the HELCOM Contracting Parties

This assessment is based on an information request that was sent to the Contracting Parties in 2015 regarding concern about inputs of various persistent organic pollutants (POPs) and some other substances to the Baltic Sea. The request also included other micropollutants of concern in effluents from waste water treatment plants. By "concern" in this context the interest is in substances that the Contracting Parties consider or believe to a significant degree are transported to the Baltic Sea via atmospheric deposition or via riverine transport.

The information gathered from the questionnaire is intended to be used for a better understanding on POPs and other substances that are included in air and riverine monitoring, and to ascertain which substances may need to be focused on in future HELCOM activities. By autumn 2017 the work reached the next stage when requests on data availability were sent to the CPs. The long-term goal, is to gather a sufficient knowledge base to revise the HELCOM list of Priority Substances, as well as to continue the work to reduce the amount of hazardous substances in the Baltic Sea region.

Results from the questionnaire

The information was based on information provided by the eight Contracting Parties that answered the questionnaire (DE, DK, EE, FI, LT, LV, SE and RF). Unfortunately, some of the answers did not cover all subjects, probably due to lack of (time for) national coordination. This gives somewhat unbalanced final results, but in any case some general tendencies may be seen in the replies.

To summarise the concern from the CPs on the various kinds of substances and groups of substances, a weighted approach was performed using a weight of 3 for major concern, 2 for intermediate, and 1 for little concern, whereas if the

CP indicated lack of knowledge a value of 0 was applied, and for substances that were indicated as not relevant a weight of -1 was used.

The weighed scores for the different substances indicate that for air monitoring and atmospheric deposition the substances of most concern are dioxins including dibenzo-furans and dioxin-like PCBs (Table 7). The second largest concern are substances belonging to the groups non-dioxin-like PCBs, PBDEs, PFAS, and HCHs, whereas the other substances were indicated to be of less concern or on some cases not relevant for air monitoring or atmospheric deposition.

In water (in this case both rivers and micropollutant MWWTP effluents) the patterns of concern are fairly similar with nonylphenols, and octylphenols, as well as PFAS, having high or medium concern on both cases. A medium concern is also the case for most other substances on the common list, whereas less concern emerges for medium-chained chlorinated paraffins, endosulfans, and heptachlor. Also, there is tendency that dioxins, PBDEs, HBCDD, DDTs and HCHs are of higher concern in rivers than for MWWTP effluents. For the MWWTP effluents also other kinds of substances where included in the questionnaire and of these a major concern was expressed for heavy metals and pharmaceutical residues in the effluents. In addition, medium concern was recorded for different groups of pesticides, disinfectants, and endocrine disrupting substances in the MWWTP effluents, whereas veterinary drug residues seem to be of low concern, although these substances are very closely related to the pharmaceutical residues (actually, in many cases the same kind of substances are used both for humans and for animals). It is not possible to ascertain if the lower concern for the veterinary drug residues originate from less knowledge, or from information that these substances are found to a lesser extent in MWWTP effluents, from the replies to these high-level questions.

The monitoring activities in air, atmospheric deposition, and in riverine waters are to a large degree reflecting the concern expressed by the Contracting Parties, but the monitoring is also heavily dependent on what it is feasible to monitor or detect by present analytical methods, and also on demands from different kind of legislative directives. No major changes in the monitoring of air and atmospheric deposition seems to be planned by the CPs, whereas the riverine monitoring seems mainly to shift more in the direction of fulfilling of the demands within the Water Framework Directive.



Box 7.

Table 7.

Weighted summary of the concern by CPS on the various kinds of substances and groups of substances in air and atmospheric deposition, and in rivers and MWWTP effluents. The weighing process is described in the main text.

Substance (group)	Air	Rivers	MWWTP
Dioxins (PCDD, PCDF, dioxin-like PCBs)	13	5	3
Other PCBs (other than dioxin-like)	6	6	5
Organotin compounds (TBT, TPhT, etc)	-4	7	6
PBDEs (pentaBDE, octaBDE, decaBDE)	6	9	4
PFAS (PFOS, PFOA)	6	10	8
HBCDD	2	6	4
Nonylphenols (NP, NPE)	-4	10	12
Octylphenols (OP, OPE)	-4	8	12
Short-chain chlorinated paraffins (C10-13)	1	4	5
Medium-chain chlorin. paraffins (C14-17)	-2	2	3
Endosulfan	2	3	2
DDTs (sum-DDT, DDE, etc)	4	6	2
PAHs (incl. metabolites)	15	9	8
BFRs (PBDEs etc)	3	6	5
HCHs (alpha, beta, gamma)	5	6	4
Heptachlor	0	1	4
Heavy metals			14
Pharmaceutical residues			12
Herbicides (except listed above)			6
Fungicides (except listed above)			5
Insecticides (except listed above)			5
Endocrine disrupting substances (EDS, except listed above)			9
Animal/veterinary drug residues (except listed above)			2
Disinfectants (except listed above)			5



Upcoming work on the input of POPs and micropollutants

Based on the outcome of the questionnaire, the first data collection and assessment of inputs of organic pollutants is suggested to include nonylphenols, octylphenols, and PFAS. These substances were identified as generally of high concern, and the stated data availability appears to be comparatively good for all three groups of pollutants. This also holds true for the MWWTP effluents, which will be included here for practical reasons as already stated. A proposition for future work would be to include pharmaceutical residues as the next group of pollutants to consider for PLC-reporting. The concern and the data availability for this group is very similar to the suggested groups to be prioritised, but will probably involve considerably more labour to compile and to assess, and hence is not proposed to be included in the first step.

Steps forward

The work on hazardous substances within the Pollution Load Compilations will be intensified in future data reporting's and assessments.

This is true for both the substances that already covered in the PLC-Water Guidelines, and consequently already are supposed to be monitored and reported to HELCOM, but also for new substances that will be incorporated according to the priorities found in the questionnaires to the Contracting parties. The new substances will firstly be included in test reporting to be assessed if they are possible to incorporate in future regular work. Especially for the already included substances, the work on increasing the data coverage and data quality need to be intensified, to ensure that future assessments will give reliable results.

A test data reporting of riverine and MWWTP effluents of selected substances of concern will be performed in early 2018. As it will be a test reporting, it will not at this stage be incorporated in the ordinary PLC data reporting. Airborne PFAS/PFOS will be provided by EMEP from a test modelling, whereas atmospheric deposition is not considered to be a significant source for inputs of nonyl- and octylphenols. The results will, together with the outcome from a similar process on micropollutants in MWWT effluents and the continuation of the initial work on pharmaceutical residues, be assessed in the PLC-7 reporting with tentative termination in early 2020.





References

Bartnicki J., Gusev A., Aas W. and Benedictow A. 2016. Atmospheric Supply of Nitrogen, Cadmium, Mercury, Benzo(a)pyrene, and PBDEs to the Baltic Sea in 2014. EMEP/MSC-W TECHNICAL REPORT 1/2016.

Bartnicki J., Gusev A., Aas W., Gauss M. and Jonson J.F. 2017. Atmospheric Supply of Nitrogen, Cadmium, Mercury, Lead, and PCDD/Fs to the Baltic Sea in 2015. EMEP/MSC-W TECHNICAL REPORT 2/2017.

HELCOM, 2015. Updated Fifth Baltic Sea pollution load compilation (PLC-5.5). Baltic Sea Environment Proceedings No. 145.

HELCOM 2016. HELCOM Guidelines for the annual and periodical compilation and reporting of waterborne pollution inputs to the Baltic Sea (PLC-Water).

Ilyin I., Rozovskaya O., Travnikov O., Varygina M., Aas W., and Pfaffhuber K.A. 2016. Assessment of heavy metal transboundary pollution, progress in model development and mercury research. EMEP Status Report 2/2016.

Shamsudheen S.V., Bartnicki J., Gusev A. and Aas W. 2015. Atmospheric Supply of Nitrogen, Lead, Cadmium, Mercury and PCBs to the Baltic Sea in 2013. EMEP/MSC-W TECHNICAL REPORT 2/2015.