An analysis of catchment factors associated with heavy metal export into the Baltic Sea and Nature-Based Solutions aimed at its limitation

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Notification: The preprint has been submitted to The Journal of Hazardous Materials

Abstract

The aim of the article was to determine the shares of individual Baltic countries participating in the inflow of metal loads to the Baltic Sea and identify patterns of similarity between these countries regarding the causes of heavy metal load generation.

The analyses used HELCOM and EUROSTAT data. The findings indicate that Finland and Sweden generate the highest total loads of heavy metals flowing in through rivers. However, Lithuania and Finland are distinguished by high metal loads calculated per km² of catchment area. Clustering countries in terms of their similarity in the heavy metal loads provided to the Baltic resulted in three groups. Finland and Lithuania generates the highest mean loads of cadmium, chromium, nickel and zinc per unit area [kg/km²/year]. Estonia and Latvia generates the highest mean annual loads of lead, mercury and copper. Poland, Germany and Sweden generates the lowest heavy metal loads.

Multidimensional data analysis showed a strong correlation between aquaculture production in the Baltic Sea catchment area, the number of cattle, beef, mutton, pigs, poultry, and meat produced from them, the amount of waste, trucks, cereal production, the use of nitrogen fertilizers, and the loads of heavy metals reaching the Baltic Sea with river waters.

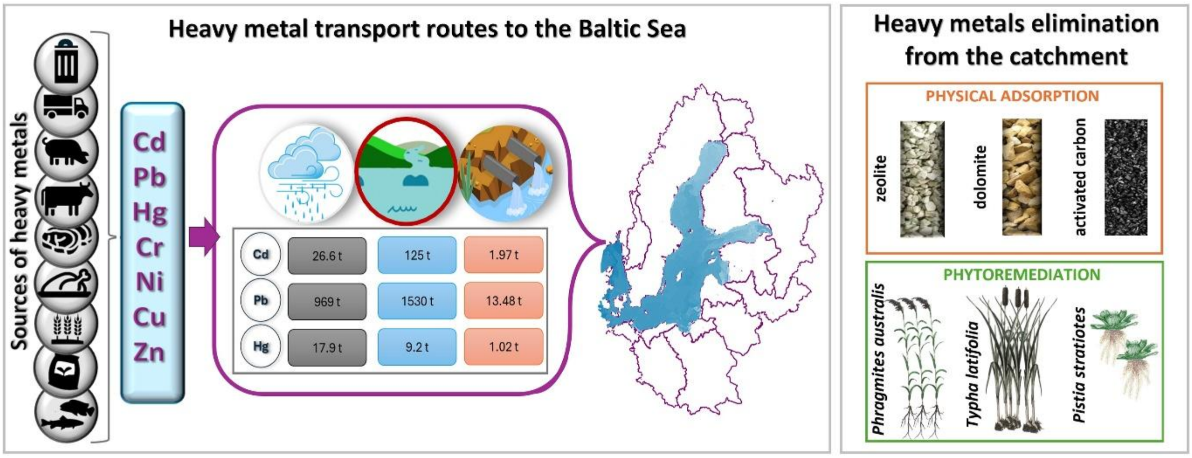
Therefore, there is a need for continuous monitoring of the loads and transfer of heavy metals to the Baltic Sea, and for activities aimed at eliminating them from the environment. For this purpose, Nature-Based Solutions can be used, as they represent inexpensive, nature-friendly methods for removing pollutants from surface waters.

Keywords: Baltic Sea catchment, Baltic Sea surrounding countries, Ecohydrology, heavy metals, Nature-Based Solutions, water contamination

Environmental Implication

Heavy metals, after entering rivers, are not only transported along the river continuum, but also deposited in sediments and living organisms. Even small increase in the concentration of metals in ecosystem can lead to a toxic effect on the trophic chain and humans. These effects can manifest themselves i.a. in impaired reproductive capacity, organs damage, increased mortality, behavioral changes and disturbances in the population structure. All this indicates a constant need for identifying the level of pollution of the aquatic environment with heavy metals and determining the sources of its emissions to develop modern, nature-friendly solutions for their elimination.

Graphical Abstract



Contents

1. Introduction

Water, as a basic element of life, plays a key role in maintaining the balance of ecosystems and human well-being. However, the dynamic development of urbanization and industry significantly increases the risk of pollution of the aquatic environment with hazardous substances, such as heavy metals. Heavy metals are defined as metallic elements characterized by high density, i.e. above 5g/cm^3. A few, such as zinc, copper, iron, nickel or molybdenum, are necessary for the proper functioning of living organisms, while most of them such as cadmium, lead and mercury, are highly toxic even at low concentrations (Piwowarska et al., 2024), the latter being known as the "toxic trio" (Wilk et al., 2021).

Due to the danger associated with their emission and accumulation in the environment, heavy metals are included in many international monitoring programs (UNEP, 2023; HELCOM, 2021a). Cadmium, lead and mercury are listed among 45 priority substances posing a serious threat to surface waters, which are included in Annex X of Directive 2013/39/EU of the European Parliament and of the Council of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy (European Union, 2013). Despite the fact that these substances have been covered by priority actions and constant monitoring for many years, they are still detected in the natural environment around the world (Piwowarska et al., 2024).

Although heavy metals enter aquatic ecosystems from both natural and anthropogenic sources, it is the latter that play a significant role in their emission to the environment (Dixit et al., 2015; Paul, 2017). Among these anthropogenic sources, the largest amounts of heavy metals are emitted to the environment by mining, metallurgy, land and water transport, metallurgical industry, paint and varnish production , agriculture, combustion of fossil fuels, amber mining in the coastal zone and discharge of sewage from treatment plants (Piwowarska et al., 2024; Rashmi & Pratima, 2013).

After entering rivers, heavy metals are transported along the river continuum. During their transport, they are deposited in sediments and in the tissues of *inter alia* small zooplankton, mussels, fish and amphibians, posing a direct threat to the biodiversity of aquatic ecosystems (Wang et al., 2018). Even a small increase in the concentration of metals in aquatic ecosystems can lead to a toxic effect on organisms in the trophic chain, including humans (HELCOM, 2018b). These toxic effects can manifest themselves in various ways, such as impaired reproductive capacity, organs damages, increased oxidative stress (Chen et al., 2017), paralysis of the nervous system (Berg et al., 2010), metabolic disorders and increased mortality (Sierra-Marquez et al., 2019; Guo et al., 2018). Heavy metal pollution can also result in behavioral changes and disturbances in the population structure, which can lead to disruptions in entire aquatic ecosystems (Mukherjee et al., 2022; Singh & Saxena, 2020; Huang et al., 2011; Berg et al., 2010).

As such, there is a constant need for monitoring aimed at identifying the level of heavy metal pollution in the aquatic environment, and for determining the sources of these emissions in order to develop modern, nature-friendly solutions for their deactivation and elimination. Such a holistic approach to environmental research fits perfectly into the One Health Approach concept, which states that maintaining the well-being of people, animals and entire ecosystems is closely linked, and solving any potential or existing threats must be based on a coordinated, multidisciplinary approach (Mackenzie & Jeggo, 2019).

Heavy metal loads entering the Baltic Sea result from a complex interaction between natural processes and human activity in the agricultural, industrial and service sectors. All these factors are interconnected, and their mutual connections create a complex network of ecological and social dependencies, whose analysis is needed to design strategies for reducing heavy metal emissions to the aquatic environment. With the above in mind, this article sets itself the following goals:

(I) Quantify the heavy metal pollution of the Baltic Sea, including a precise determination of the share of individual Baltic countries in the inflow of metal loads to the sea;

(II) Determine the hierarchy of factors generating the runoff of heavy metals from the Baltic Sea catchment area, taking into account both natural processes and anthropogenic sources of pollution;

(III) Perform cluster analysis of the Baltic countries to identify common patterns regarding the sources of heavy metal emission and determine relationships between socio-economic factors and metal loads reaching the Baltic Sea.

The novelty of the article lies in its use of a holistic, multi-dimensional approach combining spatial analysis and a modeling of metal loads flowing out of the catchment areas of the Baltic countries, combined with an assessment of the impact of anthropogenic factors. It also performs an interdisciplinary synthesis of methods from the field of Nature-Based Solutions (NBS), which can effectively reduce the runoff of heavy metals from the urban and agricultural landscape and their transfer to aquatic ecosystems. The innovative nature of the research it that it not only makes a precise assessment of the scale of the problem, but also provides tools for effective management of the risk of pollution of the Baltic Sea, taking into account both environmental and socio-economic aspects.

2. Materials and methods

2.1. Characteristic of the Baltic Sea basin

The Baltic Sea is the only almost completely closed inland sea in Europe (ESaTDOR, 2013). Its surface area is 420,000 km², with the catchment area being almost four times larger. The Baltic Sea can hold up to about 20,000 km³ of water, with a renewal time of 25–35 years for complete exchange with the North Sea (HELCOM, 2018a; Lodenius, 2016). It is not a deep sea, with a mean depth of only 60 m (Ojaveer et al., 2010). Although its salinity is below 30

PSU, which is associated with the inflow of significant amounts of fresh water, the sea is classified as one of 66 *large marine ecosystems* (Snoeijs-Leijonmalm & Andrén, 2017). The total catchment area of the Baltic Sea is 1,729,500 km² (HELCOM, 2019).

The Baltic Sea is fed with freshwater from about 200 rivers (Kiedrzyńska et al., 2014). Among them, the main role is played by the Neva, Vistula, Neman, Daugava, Odra, Göta älv and Kemijoki, whose total catchment area is 869,891 km² (HELCOM, 2018c). The sum of annual river and precipitation runoffs feeding the Baltic Sea is about 400-500 km³ of freshwater. However, as the Baltic is supplied by waters flowing from nine highly industrialized countries, it is constantly exposed to the inflow and accumulation of pollutants, including heavy metals and biogenic compounds (Kiedrzyńska et al., 2014; Zalewski et al., 2020).

2.2. Characteristic of the surrounding countries

The catchment area of the Baltic Sea is 93% owned by nine countries with direct access: Poland-PL, Sweden-SE, Germany-DE, Finland-FI, Denmark-DK, Lithuania-LT, Latvia-LV, Estonia-ES and Russia-RU. The remaining 7% of the catchment area belongs to territories not adjacent to the sea and located in the intermediate catchment area (Czech Republic, Slovakia, Norway, Belarus, Ukraine) (HELCOM, 2019). The largest part of the Baltic Sea catchment area belongs to Sweden, with 440,050 km² (25%) followed by Russia (19.7%), Poland (19.4%) and Finland (18.8%). Of all the countries with direct access to the Baltic Sea, Germany has the smallest share (1.8%). The total catchment area outside the boundaries of the Contracting Parties is 125,030 km², covering mainly the territory of Belarus (HELCOM, 2019).

The Baltic Sea catchment area is characterized by a high levels of both industrial and agricultural development (Kiedrzyńska et al., 2014). In 2014 alone, the Baltic countries were home to 487 treatment plants discharging treated sewage directly into the sea, 4143 treatment plants discharging sewage into surface waters constituting indirect sources of pollution, and 1814 industrial plants located in the direct and indirect catchment area (HELCOM, 2019).

Moreover, in the countries belonging to the European Union included in the Baltic Sea catchment area (Germany, Denmark, Lithuania, Latvia, Estonia, Finland, Poland, Sweden), in 2013 alone there were 2,147,000 farms, representing almost 20% of the total number in the entire EU (EUROSTAT, 2016b). It is estimated that in the same year the area of agricultural land in these countries was about 447,115 km² (EUROSTAT, 2015b).

The leader in average annual grain production is Denmark, with a supply of 222.15 t/km² (mean value from 2012-2015), compared to 135.87 t/km² for Germany, and 93.44 t/km² for Poland. Lithuania also recorded a high average grain production per km², with a value of 77.74 t/km² (SI2). Denmark, Germany and Poland also use the largest amounts of nitrogen and phosphorus fertilizers per km² on average (data for 2012-2015) (SI2). In the period 2012-2015, these countries were also leaders in the average annual production from aquaculture, average annual production of poultry, beef and pork meat per km² (SI2).

Taken together, these data indicate the intensification of anthropogenic processes, leading to an intensification of contamination pressure on the environment. The demographic and socio-economic characteristics of individual countries located in the direct catchment area of the Baltic Sea are presented in the Supplementary Materials (SI2).

2.3. Data set

The chemical parameters of water quality (SI1a, SI1b) were based on HELCOM data for the years 2012-2021. The demographic and economic data for eight countries belonging to the Baltic Sea catchment area, *viz.* Sweden, Finland, Estonia, Latvia, Lithuania, Poland, Germany and Denmark, were taken from the EUROSTAT database for the years 2012-2015 (SI2). Russia was not included in the analyses due to lack of data for key parameters. Additionally, Denmark was also excluded from some analyses due to insufficient data.

EUROSTAT data were unified and converted to the area (km²) of each country to standardise analyses, and then proportionally adjusted to the areas of the individual countries within the

Baltic Sea catchment area. This conversion of EUROSTAT data based on the catchment area of individual countries enabled their unification and allowed them to be compared with the HELCOM data, thus ensuring consistency of analysis. The resulting database was used for statistical analyses. However, in the process of its development, difficulties were encountered related to the incompleteness of data and the varying time intervals for which data on heavy metal loads were available. For example, data on atmospheric deposition were available for cadmium (Cd) for the period 2012-2020, for lead (Pb) for 2012-2017, and for mercury (Hg) for 2012-2018. Data for riverine inflow and direct point sources covered the period 2012-2021 (based on HELCOM 2021a, 2024).

2.4 Statistical data analysis

The spatial variability and drivers of heavy metal loads among countries in the Baltic Sea catchment were analysed using R software (R Core Team, 2024). Data preparation included combining annual heavy metal concentration data with catchment area sizes to compute area-specific loads (kg/km²/year) and total loads (kg and tonnes/year). Transformations, aggregations, and data wrangling were performed using tidyverse tools (Wickham et al., 2019). The chart illustrating the characteristics of heavy metal loads entering into the Baltic Sea from various sources was created using Statistica ver. 13.3 (StatSoft Poland).

Maps were created to illustrate the spatial distribution of average annual heavy metal loads (tonnes/year) across Baltic Sea catchment countries. Country boundaries were retrieved using the “rnaturalearth” and “sf” packages. For each metal, spatial data were combined with average load values, and centroids of country geometries were calculated to position load-specific markers. The size of each marker represented the mean annual load for the given metal, while text annotations indicated numerical values and country names.

Visualizations were generated using “ggplot2” (Wickham, 2016) and “ggrepel”, ensuring clear representation of spatial patterns. Individual maps for each metal were produced, highlighting the variability in loads between countries. These maps provided insights into the geographical distribution of metal emissions within the Baltic Sea catchment.

Correlations between heavy metal loads (Cd, Cr, Cu, Hg, Ni, Pb, Zn) and environmental factors were assessed using Spearman’s correlation methods. The Shapiro-Wilk test, implemented via “rstatix” (Kassambara, 2021), was used to evaluate the normality of factor-metal distributions. Correlation coefficients and p-values were calculated for each pair, structured into matrices, and visualized using “ggcorrplot” (Kassambara, 2019) and “corrplot” (Wei & Simko, 2021). Heatmaps and annotated matrices were used to highlight significant relationships ($p=0.05$) across spatial and temporal patterns.

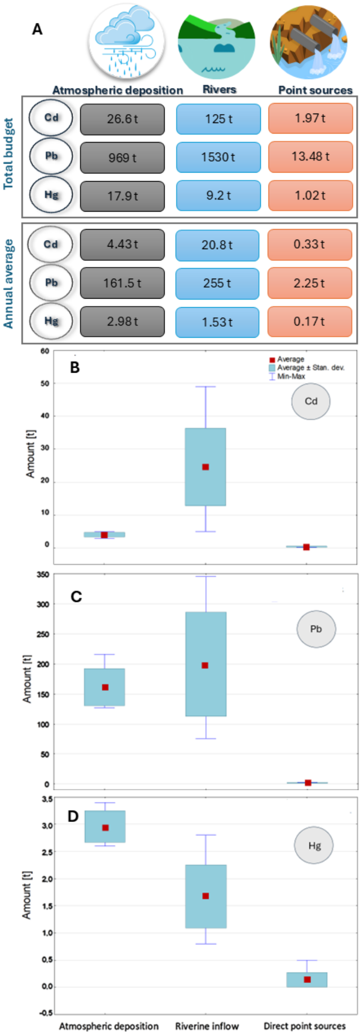
Principal Component Analysis (PCA) was used to investigate relationships between countries and heavy metal loads. PCA and biplot visualizations were conducted using “FactoMineR” (Lê et al., 2008) and “factoextra” (Kassambara & Mundt, 2020).

Hierarchical clustering was applied to classify countries based on mean annual area-specific heavy metal loads, using Ward’s Method with a Euclidean distance matrix. Clusters were visualized using “factoextra” and “dendextend” (Galili, 2015). Linear Discriminant Analysis (LDA), implemented with the MASS package (Venables & Ripley, 2002), was used to assess the contribution of specific metals to group separation. Figures illustrating clustering results, group-specific mean values, and LDA plots were created using “ggplot2” (Wickham, 2016) and “ggpubr” (Kassambara, 2022).These analyses provided insights into the spatial and temporal variability of heavy metal loads, emphasizing the role of environmental factors and highlighting differences among countries in the Baltic Sea catchment.

3. Results

3.1. Total budget of heavy metals in the sea

The main sources of heavy metal pollution in the Baltic Sea include atmospheric deposition, riverine inflow from the catchment area and point sources located in the immediate catchment area (Fig.1A).



		Atmospheric deposition	Rivers	Point sources
Total budget	Cd	26.6 t	125 t	1.97 t
	Pb	969 t	1530 t	13.48 t
	Hg	17.9 t	9.2 t	1.02 t
Annual average	Cd	4.43 t	20.8 t	0.33 t
	Pb	161.5 t	255 t	2.25 t
	Hg	2.98 t	1.53 t	0.17 t

Fig.1 Balance and mean annual loads of cadmium (Cd), lead (Pb) and mercury (Hg) entering the Baltic Sea in 2012-1017 (this period was selected due to the completeness of data on loads-based on database HELCOM 2021a) (A) also presents the characteristics of the loads of cadmium (B), lead (C) and mercury (D) reaching the Baltic Sea from various source groups. Data on atmospheric deposition are available for cadmium (Cd) for the period 2012-2020, for lead (Pb) for 2012-2017, and for mercury (Hg) for 2012-2018. Data for riverine inflow and direct point sources cover the period 2012-2021 (database HELCOM 2021a, 2024).

Maximum, minimum and mean values of cadmium (Fig.1B), lead (Fig.1C) and mercury (Fig.1D) loads reaching the Baltic Sea via three main routes: atmospheric deposition, river runoff and direct point sources. In the case of atmospheric deposition, the maximum mean annual load of cadmium reaching the Baltic Sea was 4.9 tonnes (t), and the lowest was 3 t (mean load 4.43 t) (Fig. 1A,B). In contrast, the highest mean annual values of lead and mercury loads were 216 t (Pb) and 3.4 t (Hg), respectively, and the lowest 127 t Pb and 2.6 t Hg (Fig.1C,D), giving mean values of 161.5 t Pb and 2.98 t Hg (Fig.1A). In the case of heavy metal loads entering the Baltic Sea via river runoff, the highest values over the multi-year period (2012-2021) were 49 t Cd, 345 t Pb and 2.8 t Hg, and the lowest were 5 t Cd, 76 t Pb and 0.8 t Hg (Fig.1B-D), giving mean values of 20.8 t Cd, 255 t Pb, 1.53 t Hg (Fig.1A). Direct point sources constitute the smallest source of heavy metal loads entering the Baltic Sea among the three main emission categories. In the years 2012-2021 the highest recorded annual mean loads were 0.57 t Cd, 2.85 t Pb, and 0.5 t Hg, while the lowest values were 0.14 t Cd, 1.48 t Pb, and 0.06 t Hg, respectively (Fig.1B,C,D) (HELCOM, 2021a; 2024). The Baltic Sea was polluted from direct point sources with a mean annual load of 0.33 t Cd, 2.25 t Pb, 0.17 t Hg (Fig.1A).

3.2 Heavy metal loads by country

3.2.1 Riverine outflows

Detailed data on the share of individual countries in the inflow of cadmium, lead and mercury, together with other heavy metals (i.e. chromium, nickel, copper and zinc), from rivers to the Baltic Sea are presented in Fig. 2. The analysis of the mean annual loads of heavy metals transported by rivers in the years 2012-2021 indicates that the highest values came from Finland and Sweden, while the other Baltic countries delivered significantly lower, diversified loads of metals (Fig.2A-G).

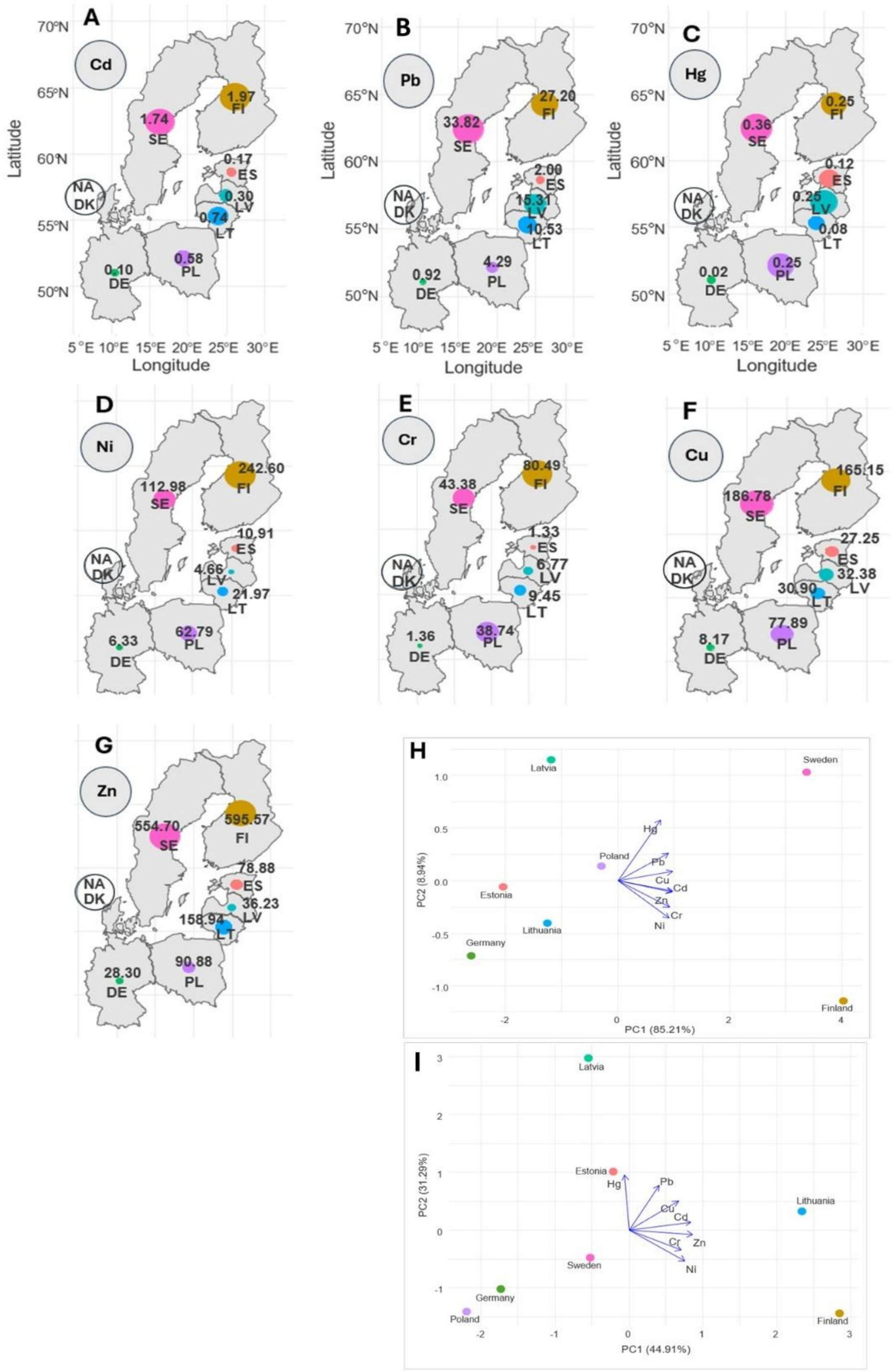


Fig.2 (A-G) Mean annual load of heavy metals [t] transported by rivers to the Baltic Sea from individual countries in 2012-2021 (data: HELCOM, 2021a, 2021d, 2024);

(H) PCA ordination of countries based on annual total heavy metal loads (t/year) transported by rivers during the period 2012-2021 (based on the data from HELCOM 2021a, 2021d, 2024);

(I) PCA ordination of countries based on annual area-normalized heavy metal loads (t/km^2/year) transported by rivers during the period 2012-2021 (based on data from HELCOM, 2021a, 2024)

The PCA ordination based on the annual load of heavy metals from riverine sources arranged countries along a gradient of increasing loads; this gradient was primarily arranged along the first PCA axis, which explains 85.21% of the variation (Fig.2H). Sweden and Finland are positioned on the side associated with high annual loads of metals (Fig.2A-G). Sweden's position can be attributed to its particularly high values of Hg, Pb, and Cu, while Finland is influenced by elevated levels of Cd, Zn, Cr, and Ni. Latvia's position in the PCA ordination space is driven by high loads of Hg and Pb, with a stronger correlation to the second PCA axis. Poland is positioned closest to the mean values, while Estonia, Lithuania, and Germany scored low in relation to PCA1 (Fig.2H).

A different distribution of variance can be seen for the PCA ordination based on data normalized per km^2 (t/km2/year) of catchment area of each country. PC1 explains 44.91% of the variance, while PC2 accounts for 31.29% (Fig.2I). The arrowheads of Pb, Cu, and Cd, indicating increasing loads, are directed towards Lithuania. Similarly, the arrowheads of Ni, Zn, and Cr, as in the analysis of annual average loads per country, point towards Finland. Notably, Latvia is positioned significantly away from the center of the plot, with the vector for Hg pointing in its direction. Estonia is located slightly closer to this vector, suggesting that its data provide less distinct information compared to Latvia.

Sweden, Germany, and Poland are located on the opposite side of the plot relative to the vectors representing heavy metals, indicating that the loads are negligible compared to other countries, when considering normalized data per km^2 of catchment area (Fig.2I). Poland and Germany are notably distant from the center of the plot and are positioned at the negative extremes of both PC1 and PC2, indicating that the loads they generate are the smallest.

In addition, an analysis of the correlation between the unit loads (kg/km^2/year) of different types of heavy metal entering the Baltic Sea with river runoff between individual countries was performed for the years 2012-2021 (Fig.3). The research showed that Finland and Sweden exhibited moderate to strong correlations (in most cases above 0.7) between heavy metal loads (Fig.3). In Germany, correlations above 0.5 were found for all metal pairs except those involving lead (<0.5), with the Pb-Ni pair even showing a slight negative correlation. In Estonia, strong positive correlations were identified for pairs including Cd-Cr, Cd-Cu, Cd-Ni, Cd-Pb, Cr-Cu, Cu-Ni, Cu-Pb, and Ni-Pb. For Zn, a strong positive correlation was observed only with Ni, while other correlations were weaker (<0.5), and the Ni-Hg pair showed a negative correlation. In Poland, strong correlations were observed only for Cd-Ni, Cr-Cu, Cr-Pb, Cu-Pb, Hg-Pb, Hg-Zn, and Pb-Zn, while other pairs showed either weak positive or negative correlations. Similarly, in Latvia, strong positive correlations were mainly found for pairs with Cd (Cd-Cu, Cd-Pb, Cd-Zn), as well as Cu-Pb, Cu-Zn, Hg-Pb, and Pb-Zn, with other correlations being either negative or weakly positive (Fig. 3).

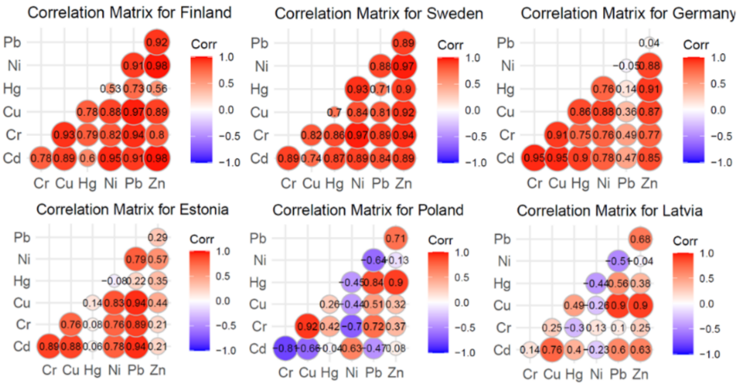


Fig. 3 Correlations between specific loads (kg/km²/year) of individual heavy metals entering the Baltic Sea via river runoff from the Baltic countries in 2012-2021 (based on data from HELCOM, 2021a, 2024)

3.2.2 Direct point sources

An analysis of direct point sources, such as industrial plants and wastewater treatment plants, discharging pollutants directly into the Baltic Sea showed that the largest loads of heavy metals come from Finland and Sweden. The mean heavy metal loads released from direct point sources in the years 2012-2021 are given in Figure 4. The highest cadmium loads came from Finland (0.086 t) and Sweden (0.249 t).

The highest mean annual lead loads were recorded in Sweden (0.875 t) and Denmark (0.790 t) (Fig.4B). The highest mean mercury loads were recorded for Poland (0.045 t), Sweden (0.04 t), Finland (0.023 t) and Denmark (0.021 t) (Fig.4C). For nickel, the highest mean loads came from Finland (3.927 t) and Sweden (4.159 t), similarly to copper (FI-3.665 t; SE-8.417 t), zinc (FI-21.78 t; SE-45.87 t) and chromium (FI-2.055 t; SE-1.128 t). Detailed source values are presented in the SI1b.

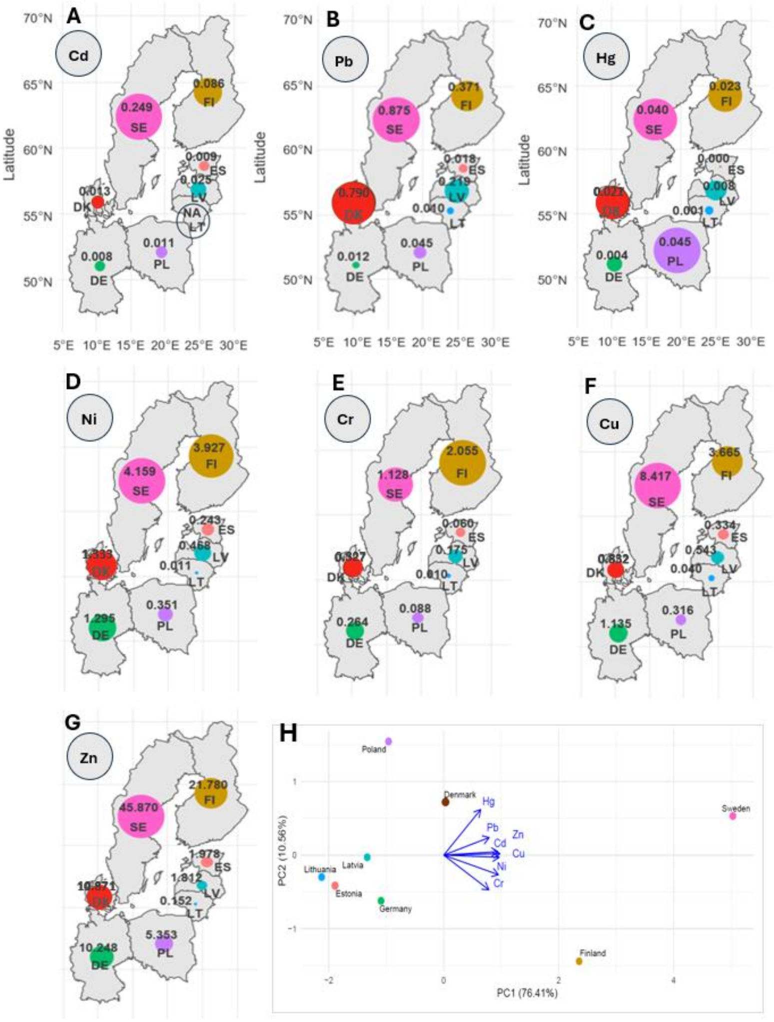


Fig. 4 (A-G) Mean annual load of heavy metals [t] from direct point sources, transported from individual Baltic countries to the Baltic Sea in 2012-2021 (based on data from HELCOM, 2021a, 2024). (H) PCA ordination of Baltic countries based on the annual heavy metals load from the point sources for the period 2012-2021 (based on data from HELCOM, 2021a, 2024)

The PCA ordination performed for the annual mean loads of heavy metals generated in individual countries from point sources (Fig.4H) shows that the first PCA axis explains more than 76% of the variance, while the second axis, accounts for approximately 10%. This findings indicate that the vectors representing elevated loads of Hg, Pb, Cd, Zn, and Cu are directed towards the most distant point on the plot, representing Sweden, suggesting that it is responsible for the highest generated loads of these metals. The vectors for Cr, Ni, and Cu, on the other hand, point towards Finland. Poland and Denmark are located in the same region of the plot, with evidence of elevated Hg loads in these countries. Lithuania, Latvia, Estonia, and Germany form a single group, with their points positioned close to each other, indicating that the variance between them is minimal (Fig.4H).

For Sweden and Denmark, a strong positive correlation was observed between all pairs of heavy metals from point sources (Fig.5). In Germany, strong positive correlations (>0.5) were found for all pairs of heavy metals except those involving lead, as well as the Pb-Zn pair, which showed weak positive correlations. For the remaining countries, correlations between heavy metals were more variable, ranging from strongly positive to strongly negative depending on the metal pair and the country (Fig.5).

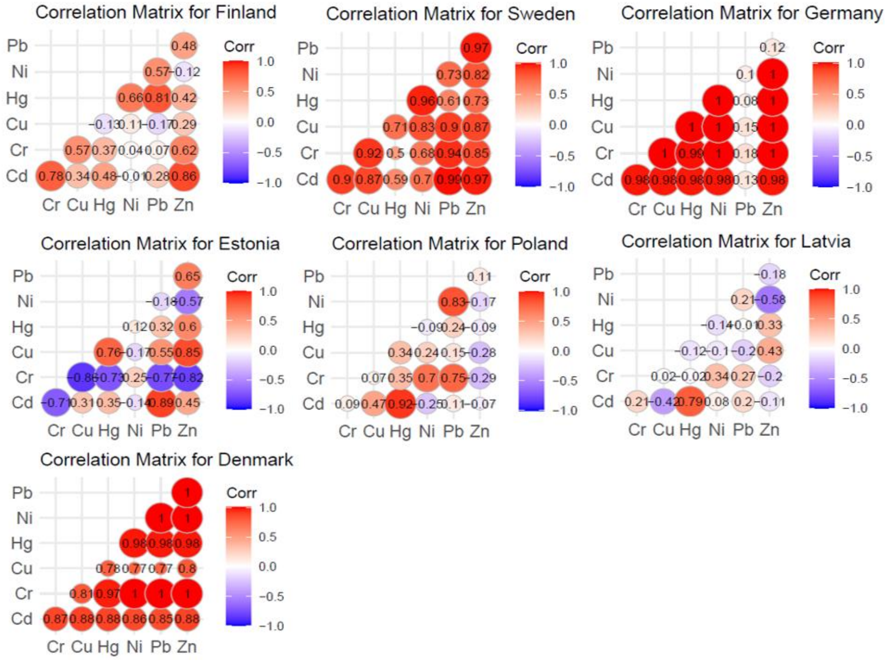


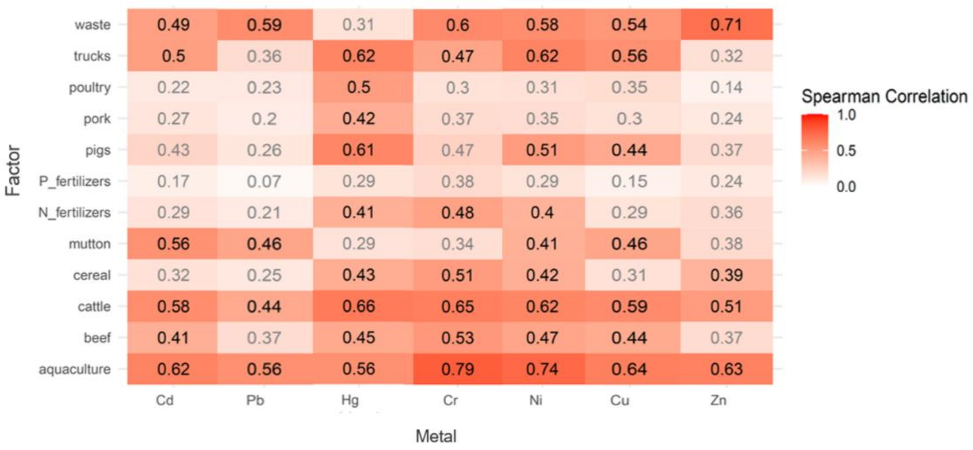
Fig. 5 Correlations between loads (kg/km²/year) of heavy metals entering the Baltic Sea from direct point sources from individual Baltic countries in 2012-2021 (based on HELCOM, 2021a, 2024).

3.3 Hierarchy of factors affecting the contamination of the Baltic Sea with heavy metals

The economic factors influencing the release of heavy metals and their transport with river waters to the Baltic Sea were determined using Spearman's correlation analysis based on data from 2012-2015 (Fig.6). It was found that aquaculture production and the number of cattle had significant influences on the loads of all analyzed heavy metals entering with river waters. Additionally, beef production demonstrated a strong positive correlation with all metals except lead and zinc (Fig.6). The number of pigs correlated with mercury, nickel and copper loads. Interestingly, pork production only exhibited a correlation with mercury loads. Poultry production was also correlated with mercury. For mutton, a positive correlation was demonstrated for Cd, Pb, Ni and Cu loads.

Analyses also showed that the amount of waste generated by Baltic Sea basin residents influences the loads of all heavy metals except mercury. Strong correlations were also shown between the number of trucks registered in the Baltic countries and the presence of cadmium, mercury, chromium, nickel and copper, although no correlation was shown with lead and zinc.

A significant positive relationship was found between crop production in the Baltic countries and the load of mercury, chromium, nickel and zinc discharged. The use of nitrogen fertilisers in agriculture also has an impact on increased loads of Hg, Cr and Ni, while no significant correlation was shown with metal and phosphate fertilisers. The numerical values for individual economic factors included in this analysis were included in the SI.



Factor	Cd	Pb	Hg	Cr	Ni	Cu	Zn
waste	0.49	0.59	0.31	0.6	0.58	0.54	0.71
trucks	0.5	0.36	0.62	0.47	0.62	0.56	0.32
poultry	0.22	0.23	0.5	0.3	0.31	0.35	0.14
pork	0.27	0.2	0.42	0.37	0.35	0.3	0.24
pigs	0.43	0.26	0.61	0.47	0.51	0.44	0.37
P_fertilizers	0.17	0.07	0.29	0.38	0.29	0.15	0.24
N_fertilizers	0.29	0.21	0.41	0.48	0.4	0.29	0.36
mutton	0.56	0.46	0.29	0.34	0.41	0.46	0.38
cereal	0.32	0.25	0.43	0.51	0.42	0.31	0.39
cattle	0.58	0.44	0.66	0.65	0.62	0.59	0.51
beef	0.41	0.37	0.45	0.53	0.47	0.44	0.37
aquaculture	0.62	0.56	0.56	0.79	0.74	0.64	0.63

Metal

Fig. 6 Spearman’s correlation matrix indicating significant relationships between heavy metal loads discharged in 2012-2015 via riverine outflows from individual Baltic Sea basin countries and their specific economic factors. Economic factors were converted proportionally to the Baltic Sea basin area and calculated per km^2 of a given country. Statistically insignificant values ($p>0.05$) are grayed out. (Based on data from HELCOM, 2019, 2021a; EUROSTAT, 2013; 2014; 2015a 2015b; 2016a, 2016b, 2017a, 2017b, 2018a)

3.4 Clustering of countries by area-specific loads of heavy metals riverine input to the Baltic Sea

The cluster analysis of annual area-specific heavy metal loads (kg/km^2/year) revealed three groups of countries (Fig.7A). Group 1, characterized by high loads of Cd (0.009), Cr (0.206), Ni (0.571), and Zn (2.207), contained Finland and Lithuania. Group 2, with the lowest heavy metal loads, included Poland, Germany and Sweden. Group 3 , with the highest annual average loads of Pb (0.141), Hg (0.003), and Cu (0.552), included Estonia and Latvia. Due to insufficient data, Denmark was excluded from the analysis (Fig.7B).

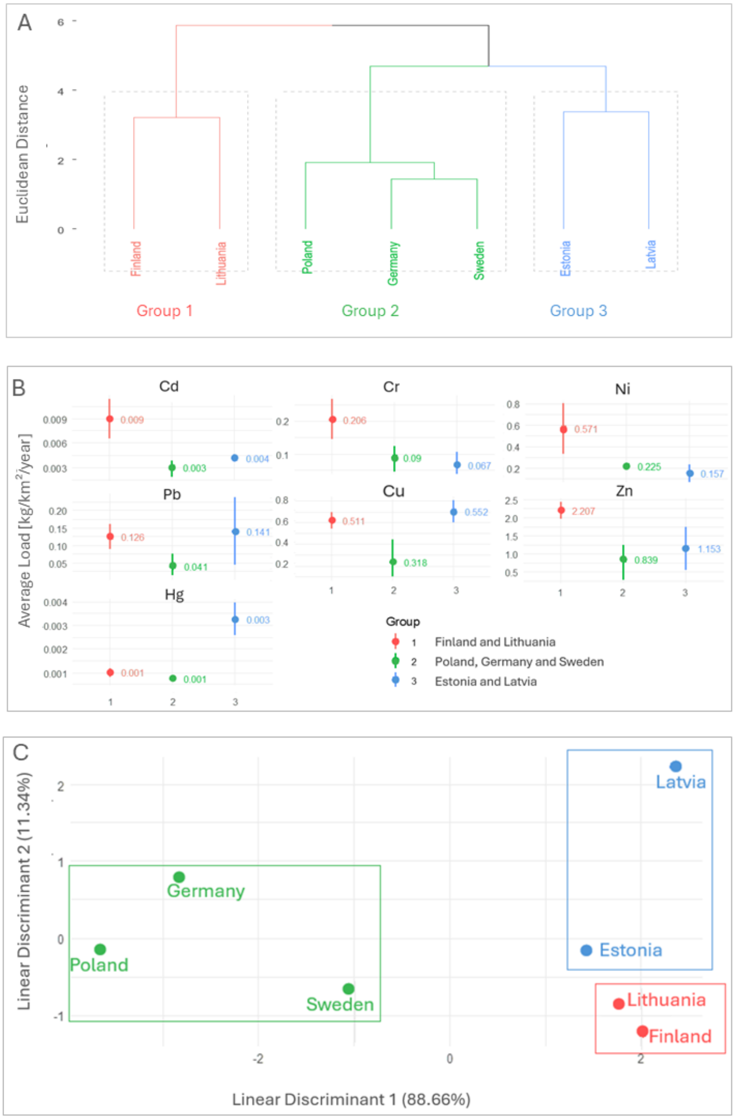


Fig.7A Hierarchical clustering of countries based on annual area-specific loads of heavy metals (kg/km²/year). Distance metric: Euclidean. Clustering method: Ward's Method. 7B Annual area-specific load of heavy metals for groups of countries according to cluster analysis from Fig.7A. Min-max values are marked with lines, and mean values are illustrated with points (based on data from HELCOM 2021a, 2024). 7C Linear Discriminant Analysis (LDA) illustrating the discrimination of countries based on area-specific heavy metal riverine loads to the Baltic Sea. Groups correspond to the hierarchical clustering shown in Fig.7A.

The mean loads in the groups are presented in Fig.7B. With regard to unit area, the first group (Finland and Lithuania), generates the highest mean loads of cadmium (0.009 kg/km²/year), chromium (0.206 kg/km²/year), nickel (0.571 kg/km²/year) and zinc (2.207 kg/km²/year). The third group (Estonia and Latvia) generates the highest loads of lead (0.141 kg/km²/year), mercury (0.003 kg/km²/year) and copper (0.552 kg/km²/year). The second group (Poland, Germany, Sweden) generates the lowest loads.

Discriminant analysis revealed that LD1 (Linear Discriminant 1) accounted for 88.66% of the variability, while LD2 explained an additional 11.34% (Fig.7C). Along LD1, the most significant contributor was Hg (713.1), indicating that its variability played a dominant role in differentiating the groups. A substantial contribution was also noted for Cd (83.6). In contrast, Cu (7.1) and Pb (8.7) contributed moderately, while Cr (-0.19), Ni (2.7), and Zn (0.52) had minimal influence on LD1. Along LD2, Hg again had the largest contribution (1032.5), emphasizing its key role across both dimensions. Cd (-114.5) was the second most important contributor to LD2, but its negative coefficient suggested an inverse relationship compared to LD1. Other metals, including Cr (-17.0), Cu (-11.0), Pb (12.5), and Ni (6.9), had moderate contributions to LD2, while Zn (0.005) showed an almost negligible impact. These results highlight Hg as the dominant factor influencing group separation, followed by Cd, with moderate contributions from Cu, Pb, and Cr, and minimal influence from Ni and Zn (SI3).

4. Discussion

4.1 Baltic Sea - Total Balance of Heavy Metals

Human activity has been exerting strong pressure on the ecosystem of the Baltic Sea and its catchment area for many years. Its specific hydrological and geographical conditions make the Baltic Sea highly sensitive to anthropogenic pollution and the accumulation of harmful substances. It has been classified by the Marine Environment Protection Committee of the International Maritime Organization as a Particularly Sensitive Sea Area (Popek et al., 2021). The main migration routes of heavy metals from highly industrialized areas to the Baltic Sea are atmospheric deposition and river runoff (Kuprijanov et al., 2021).

In 2012–2017, atmospheric deposition delivered a total of 26.6 t Cd, 969 t Pb and 17.9 t Hg to the Baltic Sea (HELCOM, 2021a). In general, atmospheric heavy metal emissions are decreasing in the European Union, with Pb falling 89%, and Cd 66% from 1990 to 2012. Nevertheless, these compounds are still detected (Kuprijanov et al., 2021). It is estimated that the heavy metal content of the Baltic Sea may be up to 20 times higher than the North Atlantic, and high levels of metals are still identified *inter alia* in marine organisms (Garnaga, 2012).

Water emissions include runoff from diffuse sources and from urban and industrial areas. Large rivers such as the Vistula and Neva play a significant role in the transport of metal loads such as cadmium and lead (Lodenius, 2016). In addition, up to 99% of heavy metals flowing into rivers can be stored in their sediments; however, changes in the physicochemical conditions in the aquatic and benthic environment can drive their re-emission into the water column, and thus to surface waters (Shen et al., 2020).

Another significant source of heavy metals entering the Baltic Sea is represented by point sources such as industrial plants and sewage treatment plants. In 2012-2021, these sources were responsible for the emission of 26.3 t of Cd, Pb and Hg into the Baltic, of which 15% were cadmium, 80% lead and 5% mercury (HELCOM, 2021a).

4.1.1 Cadmium

In the case of cadmium, up to 90% of deposition is dry, occurring in areas downwind of point sources, and typically 30–70% occurs on land, depending on the anthropogenic source and land use type. This land-deposited cadmium can be mobilized, resulting in higher concentrations in aquatic ecosystems such as lakes and rivers. Importantly, as an aerosol, cadmium demonstrates 80–100% solubility, and can completely dissolve within six hours of exposure to water. Therefore, wet deposition is also important, when precipitation leads to the washing out of small particles and aerosols (Cullen & Maldonado, 2012). This may explain the increased values of cadmium loads reaching the Baltic Sea with river runoff.

In large cities most airborne cadmium comes from tire abrasion and plastic, paint and adhesive emissions. It can also enter the environment as a by-product of metallurgical processes and from household and industrial waste. In surface waters, significant amounts of cadmium are leached from agricultural land, which it enters with phosphate fertilizers (Moiseenko & Gashkina, 2018). Cadmium is also easily released from the mineral environment into aquatic ecosystems under the influence of acid precipitation (Moiseenko & Gashkina, 2018).

4.1.2 Lead

Lead transport in the aquatic environment is strongly influenced by the adsorption and desorption processes occurring in bottom sediments. The adsorption and retention capacities of Pb^{2+} ions on sediments are strongly related to the presence of phosphorus in the sediment and surface waters. It has been found that 50 times less phosphorus is released during desorption of sediments loaded with Pb or Pb with Cd compared to sediments not loaded with metals (Shen et al., 2020). Strong correlations have also been noted between Pb concentrations and TOC (Total Organic Carbon) values (Ning-jing et al., 2015).

4.1.2 Mercury

Global atmospheric deposition of mercury increased three- to fivefold in the Anthropocene compared to the Holocene (Sommar et al., 2020). It is estimated that 60% of atmospheric mercury deposition is of anthropogenic origin, and its re-emission comes from surface reservoirs. Mercury deposited in bottom sediments can be re-released to the atmosphere by changes in temperature, pH or biological activity. In addition, 13% of current deposition is attributed to natural emissions and 27% to primary anthropogenic emissions (direct emissions from deep mineral deposits to the atmosphere) (Amos et al., 2013).

In the period 2012-2017, a total of 17.9 t Hg entered the Baltic Sea from the atmosphere compared to 9.2 t Hg via river waters (Fig.1). The deposition of mercury in rivers is strongly influenced by atmospheric precipitation, resulting in either dilution of mercury in the river or increased leaching from land. Greater leaching is observed in cold periods, as a result of greater mercury emission due to increased fuel combustion, as well as less runoff into rivers with melting snow; conversely, warmer periods are characterized by a lower demand for heating and hence reduced fossil fuel use (Bełdowska et al., 2014). This suggests that the level of mercury pollution in the sea is influenced by the topography of the Baltic Sea catchment area and the prevailing climate. The southwestern part of the catchment area has a temperate Atlantic climate, the eastern part a temperate continental climate, and the northern parts, a boreal and arctic climate characterized by long winters, which influences the prevailing temperatures and the length of the heating period (HELCOM, 2019). According to Wängberg (2001), the mean concentration of total mercury in the Baltic surface water column was 0.7±0.4 $\mu g/m^3$ in summer, and 1.2±0.3 $\mu g/m^3$ in winter.

4.2 Heavy metal loads by country

The analysed HELCOM data indicates that the northern countries of the Baltic have a significant share in its pollution. However, the differences noted between individual countries may be partly due to variation in the methodology of measuring heavy metal concentrations (HELCOM, 2021c).

Finland appears to make a significant contribution to the pollution of the Baltic Sea, both from river inflows and point sources. These sources are both natural and anthropogenic (Vallius, 1999). Sediments in the northern part of the Gulf of Finland contain two to four times higher concentrations of cadmium, lead and mercury than those in its southern part (Vallius & Leivuori, 2003). Finnish soils, due to their acidic pH and high content of organic matter, are prone to cadmium accumulation, and arable soils in southern Finland contain almost twice as much cadmium as soils in the north. This is due to intensive agriculture, industry and cadmium transport from Central Europe (Louekari et al., 2000). In the Gulf of Finland, more than 96% of mercury, 74% of cadmium, 40% of copper, lead and zinc, 31% of cobalt and 26% of chromium come from human activity (Vallius, 1999). Significant sources of pollution are chemical, metallurgical and mining plants in the coastal region, and the pulp and paper industry, especially along the Kymijoki River (Salonen & Korkka-Niemi, 2007). The industry generates about 100,000 t of dry mass of by-products per year, containing Al, As, Ba, Cd, Cr, Cu, Fe, Ni, Pb and Zn (Golmaei et al., 2018). In addition, more than 100 mines have operated in Finland, of which about 11 metal mines and 3 ore mills are currently active. In Finland, 90% of the population uses centralized water supply systems and 84% are connected to municipal sewage systems, which requires more than 200 wastewater treatment plants with a person equivalents of more than 1,000 (Ekholm et al., 2020).

Sweden also contributes high concentrations of heavy metals. These include natural sources of lead and copper, deriving from metal ores in the Bothnian Sea and the eastern coastline, which have been a source of metals for the aquatic ecosystem since the 10th century: they are transported by rivers to Lake Mäleren, which, like Lake Freden, is a point reservoir of heavy metals in the region (Manzetti, 2020). The natural geochemical background may also be responsible for the cadmium, zinc and arsenic in sediments around Gotland and the Gulf of Bothnia (Apler & Josefsson, 2016).

In southern Sweden, 60–85% of lead deposition comes from sources outside the area, with the share of local emissions increasing to the north (Rühling & Tyler, 2001). Significant sources of pollution include industrial plants, metal smelters, the pulp and paper industry, sulphide factories, sawmills and waste incinerators. It is estimated that in the second half of the 20th century, 2.8 t Cd, 14 t Pb, 0.4 t Hg, 129 t Zn, 28.2 t Cu, 7.7 t Cr, 64 t Ni, 51 t Co, 1000 t Fe and 272 t As were discharged annually into the Gulf of Bothnia, resulting in their accumulation in sediments and possible re-emission to surface waters (Manzetti, 2020). Sweden also has had a problem with mercury pollution since the 1950s, when alkyl- and phenyl-mercury compounds were used in agriculture and the pulp industry. Other significant sources of emissions have been the chlor-alkali industry (since 1860), pyrite burning, coal, peat and wood combustion, and processing of sulphide ores (Lindqvist et al., 1991).

Regarding Poland, over 96% of its area is covered by the basins of two large rivers, the Vistula and the Odra, which flow into the Baltic Sea; they also carry pollution from both point and area sources from almost the entire country (Jaskuła & Sojka, 2022). It is estimated that about 71% of the total cadmium emissions to the Odra river system are from municipal sewage treatment plants, 29% comes from diffuse sources, more than half of which are from urban areas (Buszewski & Kowalkowski, 2003). Interestingly, while the heavy metal content of the sediments increases along the course of the river in the Odra, they decrease along the Vistula (Jaskuła & Sojka, 2022). This reduction may be due to deposition of sediments in reservoirs located in river basins, modernization of sewage treatment plants in large cities and effective self-purification processes (Jaskuła & Sojka, 2022). The higher concentrations of metals in the upper reaches of the Vistula may be attributed to its tributaries running through the highly-industrialized and anthropogenically-changed areas of Upper Silesia (Strzebońska et al., 2017), which produce high levels of Pb, Zn and As. Another significant source of Cd emissions is agriculture (Kałmykow-Piwińska & Falkowska, 2024).

In Germany, heavy metal emissions to river basins are dominated by diffuse pathways such as paved urban areas and erosion processes. Regarding point sources, reductions in pollution from have been reported (Scherer et al., 2003). Also, in a sewage treatment plant in Kaunas, Lithuania, significant decreases in Cr, Pb and Hg levels in sludge were reported during 2000-2022, indicating the effectiveness of regulatory measures and improved treatment. Seasonal variability in metal concentrations in Lithuania is related to industrial activity and runoff (Jachimowicz et al., 2025). In Estonia, heavy metals are derived from point sources such as oil shale power plants, oil and cement production, and the chemical industry (Napa et al., 2015).

The territory of Latvia is characterized by a dense network of rivers. The five largest, *viz.* the Venta, Lielupe, Daugava, Gauja, and Salaca, account for over 90% of total runoff. River contamination with cadmium, lead, copper, and nickel is significantly influenced by large cities such as Riga, Daugavpils, and Novopolotsk. In turn, the presence of zinc and manganese may be partially related to the natural geochemical background (Klavinš et al., 2000).

4.2.3 Metal-metal correlations

Different heavy metals engage in complex interactions with each other. Our findings indicate that the occurrence patterns of individual metals vary between countries. In the case of Finland and Sweden, strong correlations were found for all metals carried with river runoff. Other studies performed in the Archipelago of Southwestern Finland identified strong correlations between iron, aluminium, vanadium, titanium and chromium concentrations (Müller, 1999); this was attributed to high proportions of detrital bonds. Nickel shows strong bonds with chromium, aluminium, titanium and vanadium, and this has also been attributed to detrital bonds. In addition, copper is widespread in the Gulf of Bothnia, and is carried in sediments by humic material and manganese/iron-oxides/hydroxides. Cobalt occurs in a partially bound form with Mn/Fe sediments. Moreover, the ratio of cobalt to nickel in organic matter has similar properties as Cd to Ni. Zinc, like nickel and copper, also shows partial detrital binding (Müller, 1999), which would explain the existence of numerous connections between environmental metals. This is also confirmed by the present findings (Fig.3).

As indicated by Chen et al. (2022), significant positive correlations between two different metals generally suggest a closely-related source, while weak positive correlations indicate different origins. In analyses conducted on dust samples, strong positive correlations were found between Cr-Ni (r=0.737), Cr-Zn (r=0.813), Cr-Sn (r=0.802), Cu-Mn (r=0.793), Mn-Fe (r=0.721), Ni-Sn (r=0.772), and Zn-Sn (r=0.783), suggesting that the heavy metals may be homologous. The weak correlations found for As-Cd (r=0.0199), As-Mn (r=0.242), As-Pb (r=0.156), As-Fe (r=0.271), Cd-Cr (r=0.159), Cd-Mn (r=0.0259), Cd-Sn (r=0.278), Co-Pb (r=0.0262), Co-Zn (r=0.252), Cu-Pb (r=0.205), Mn-Ni (r=0.266), Mn-Pb (r=0.155) and Pb-Fe (r=0.237) (Chen et al., 2022).

Other analyses conducted on the Yanghe Reservoir showed Cu correlated strongly with Zn and Cr in the bottom sediments. Also, in the river bottom sediments, Zu showed a significant negative correlation with Pb and Cr. It was also found that Pb in river bottom sediments showed a positive correlation with Zn and Cd, which means that they may come from a common source (Kuang et al., 2016). It should be remembered that correlation analysis can only supplement analyses of the sources of individual metals in the environment, and a comprehensive approach is needed to obtain a full picture of environmental pollution.

4.3 Factors - generating / determining the inflow of heavy metals

4.3.1 Industry

Our findings indicate that heavy metal pollution of the Baltic Sea deriving from river water is influenced *inter alia* by industry and waste. The most metal-emitting industries include the chemical industry, ceramics, electronics, metallurgical, printing, battery and accumulator production and catalyst paint production (Ishchenko, 2018). No significant correlations were found between heavy metal inflow and combustion processes occurring in industrial processes, waste incineration of the operation of power plants; however, according to literature they have been found to make a significant contribution to environmental pollution. For example, in the United States, of 1,100 steam-electric facilities, more than half are coal-fired power plants, which generate millions of tons of cadmium, lead, mercury, arsenic, selenium and boron annually (Sonone et al., 2020). Importantly, metals such as Pb, As, Se and Cr very easily volatilize at high temperatures, and as the temperature of the exhaust gases decreases, some heavy metals can condense, either enriching fine dust particles or being released into the atmosphere (Luo et al., 2020). Once in the atmosphere, these metals can be transported over long distances and deposited on water surfaces through dry and wet atmospheric deposition, thus accumulating in aquatic ecosystems.

4.3.2 Agriculture and meat production

Agriculture emits heavy metals, both from point sources such as factory farms and animal fattening farms, and from diffuse sources such as fertilizer and pesticide runoff (Zahoor & Mushtaq, 2023). Studies have found nitrate fertilizers and grain production to drive heavy metal pollution of the Baltic Sea. Indeed, the use of fertilizers has been associated with the occurrence of Cd, Cu and Zn in greenhouse soils (Wei, 2020), and nitrogen-based fertilizers in crops with increased levels of Cd or Pb in agricultural products (Zhou, 2003). Both Cr and Ni show strong correlations with nitrate fertilizers: their levels in fertilizers range from 3.2 to 19 µg/g (chromium) and from 7 to 34 µg/g (nickel). It is worth noting, however, that they can be present in phosphate fertilizers at concentrations of 66-245 µg/g (chromium) and 7-38 µg/g (nickel) (Sandeep et al., 2019).

Our findings, and literature data, indicate that the loads of heavy metals reaching the Baltic Sea with river runoff are influenced by poultry (Hg), pig (Hg, Ni, Cu) and cattle farming (Cd, Pb, Hg, Cr, Ni, Cu, Zn). Agriculture and animal husbandry have been found to contribute 20% to surface water pollution in the Huangshui River basin in China. Besides presence of heavy metals in fertilizers, these compounds are also commonly detected in animal feed (Zhang et al., 2022). Therefore, both breeding and meat production have a large impact on the generation of heavy metal pollution. This is also confirmed by our present data, which found correlations between poultry and pork production volume and mercury load, as well as between lamb and cadmium, lead, copper and nickel, beef and cadmium, mercury and chromium, copper and nickel.

Indeed, Makridis (2012) report that while heavy metal transfer from feed to animal products was within acceptable risk levels, these metals were detected in animal organs. For example, Cu was detected in elevated concentrations in cow livers and sheep liver and kidneys. Cd was also detected in animal organs. Relatively high concentrations of Cu were noted in pig excrement and Zn in sheep excrement, which can be explained by the presence of metals in feed. Elevated concentrations of Ni, Cr and Pb were also found in sheep feces (Makridis et al., 2012). In Germany, the Pb content was 0.031-0.101 mg/kg feed in cereal feeds and 0.160-0.204 mg/kg feed in legumes. In addition, the Cd content was 0.012-0.530 mg/kg cereal feed and 0.024-0.115 mg/kg legume feed (Wolf & Cappai, 2021). In compound feeds for laying hens, these values ranged from 0.137-0.223 mg Pb/kg feed and 0.019-0.035 mg Cd/kg feed; for broilers 0.081-0.112 mg Pb/kg feed and 0.028-0.029 mg Cd/kg feed; and for turkeys 0.067-0.358 mg Pb/kg feed and 0.013-0.054 mg Cd/kg feed. Despite the elevated values of heavy metals in the analyzed samples, all values were within the limits of the standards established by the European Union (Wolf & Cappai, 2021).

4.3.3 Road transport

Multiparameter analysis revealed strong correlations between the number of trucks and tractors in individual Baltic countries and the load of cadmium, chromium, copper, mercury and nickel (Fig.6). A similar relationship was observed by Ferreira (2016) in the Ribeira dos Covões catchment area in Portugal, where a direct relationship was found between the number of vehicles and the amount of heavy metals present in the flushed water stream. Significant amounts of heavy metals are flushed from roads by rainfall and transported to green urban areas and surface waters. The maximum concentrations of metals detected in the water were 0.01 mg/L cadmium, 0.1 mg/L lead, 0.6 mg/L copper and 5 mg/L zinc, respectively (Ferreira et al., 2016). In 2007, in Denmark, road transport was estimated to have emitted 48 kg of Cd, 6989 kg of Pb, 28 kg of Hg, 8 kg of As, 197 kg of Cr, 51779 kg of Cu, 158 kg of Ni, 33 kg of Se and 28556 kg of Zn. The largest emitters of heavy metals from transport in this country were found to be passenger cars (60.4% of the total analysed Cd emission; 61.5% Pb; 60.7% Hg), followed by vans (20.9% Cd; 35.2% Pb; 17.6% Hg), trucks (14.1% Cd; 1.47% Pb; 16.2% Hg), buses and coaches (4% Cd; 0.65% Pb; 4.4% Hg) and two-wheelers (0.6% Cd; 1.1% Pb; 1% Hg) (Winther & Slentø, 2010).

4.3.4 Aquaculture

Aquaculture plays a significant role in the heavy metal pollution of the Baltic Sea (Fig.6). In 2012-2015, aquaculture production within the Baltic Sea catchment amounted to 472317 tons of fish and seafood (EUROSTAT 2015b, 2016b, 2017b). Copper can enters aquaculture through the use of copper sulphate-based algaecides and herbicides, used to control phytoplankton and aquatic weeds, and copper-based agents used to counter unwanted organisms from nets and cages immersed in the sea (Reckermann et al., 2022). Heavy metals can enter the water column with grey water from fishing vessels and passenger ships. It is estimated that 5.5 million^3 of grey water is emitted into the Baltic Sea annually, which was found to contain an annual load of 0.0009 t Cd, 0.141 t Pb, 0.0009 t Hg, 0.04 t Cr, 0.033 t As, 2.8 t Zn and 1.5 t Cu and 0.14 t Ni in 2012 (Ytreberg et al., 2020).

4.4 Clustering countries by factors correlated to heavy metals input to the Baltic Sea

The following three clusters of countries were formed based on the characteristics and mean annual loads of heavy metals: Finland and Lithuania (Group 1); Poland, Germany and Sweden (Group 2), and Estonia and Latvia (Group 3). Group 1 generates the highest average annual loads of almost all analyzed heavy metals (Cd, Cr, Ni, Zn). The high loads of heavy metals noted in Lithuania derive from the transboundary Neman River, the fourth largest river in the Baltic Sea catchment area, whose catchment area is 98,220 km^2, and the average discharge is 674 m^3/s. The Neman has its source in Belarus, flows through Lithuania for about 359 kilometers, and runs along the border of Lithuania with the Kaliningrad Oblast for 116 km. Since 71.5% of Lithuania's territory belongs to the Neman catchment area, both industrial and agricultural pollution from almost the entire country reaches the river directly or through its tributaries (Kruopiene, 2007). A further analysis based on standardized data per unit area clearly divided the Baltic countries into eastern (Groups 1 and 3) and western (Group 2) directions. This division reflects the complexity and problems of heavy metal transport processes from the Baltic Sea catchment area to its water body.

A clustering analysis of the Baltic countries based on nutrient loads (total nitrogen and total phosphorus) identified a north-south division (Kiedrzyńska et al., 2014). Sweden, Finland, Russia and Estonia were assigned to Group 1, Lithuania, Latvia and Poland to Group 2, and Germany and Denmark to Group 3; this arrangement reflected the hierarchy of factors influencing the nutrient load to the Baltic Sea (Kiedrzyńska et al., 2014).

5. Heavy metal norms

Heavy metals are among the priority substances, i.e. whose content in water and sewage is determined by legal standards. A key documents governing the European strategy for the protection of surface waters, against *inter alia* pollution by heavy metals is The Water Framework Directive (WFD) 2000/60/EC (European Union, 2000). Additionally, in 2008 the European Parliament together with the Council of the European Union established a framework for Community action in the field of marine environmental policy, the so-called Marine Strategy Framework Directive of the European Union. Its aim is to improve the effectiveness of protection of the marine environment throughout Europe, including in the Baltic Sea (European Union, 2008). Furthermore, a list of priority substances is given in Council Directive 2013/39/EU of the European Parliament and of the Council of 12 August, 2013; amending Directives 2000/60/EC and 2008/105/EC; in this directive, the maximum permissible concentrations of cadmium for *inter alia* inland surface waters range from ≤ 0.45 to max 1.5 μg /L depending on the hardness class. Analogous values are 14 μg /L for lead, 0.07 μg /L for mercury, and 34 μg /L for nickel. Regarding mean annual concentrations, the standards range from ≤ 0.08 to 0.25 μg/L (inland surface waters) and max. 0.2 μg/L (other surface waters) for cadmium, 1.2 μg/L or 1.3 μg/L for Pb, and 4 μg/L or 8.6 μg/L for Ni (European Union, 2013). In order to prevent pollution of the marine environment of the Baltic Sea area, the Helsinki Convention was signed in 1974, the executive body of which is HELCOM (Palmowski, 2021). In 2007, the HELCOM member states adopted the Baltic Sea Action Plan (BSAP), the main goal of which was to achieve a good ecological status of the Baltic Sea by 2021. To achieve this, the focus was on four main strategic goals:

1) Significant reduction of eutrophication processes in the Baltic Sea,
2) Reduction of the amount of hazardous substances entering the Baltic Sea,
3) Conducting maritime activities in an environmentally-friendly manner,
4) Protecting biodiversity in the Baltic Sea (HELCOM, 2021b).

Currently, the BSAP update from 2021 assumes the implementation of specific measures and actions to achieve the set environmental goals by 2030. Its actions include regional and national programs aimed at controlling heavy metals and other hazardous substances. It also strengthens HELCOM recommendations on industrial emissions by implementing, among others, chemical-smart purchasing strategies to reduce emissions of hazardous substances. The plans also include monitoring of mercury and other heavy metal concentrations in mining spoil, and preventing its release during the exploitation and transport of aggregates. In addition, the plans also include establishing and implementing rules for handling waste containing mercury, and encouraging society to use less toxic alternatives to lead, e.g. in fishing and angling equipment (HELCOM, 2021b).

6. Nature-Based Solutions for limitating heavy metal levels in the environment

The removal of heavy metals is a costly process and requires advanced techniques, and new, inexpensive, environmental-friendly solutions are constantly sought. One growing area involves ecohydrological biotechnologies and nature-based solutions (NBS). These methods are currently the preferred approach to eliminating pollutants from the environment and are included in various programs of the European Commission (e.g. Horizon Europe), and the 9th phase of the UNESCO Hydrological Program (Piwowarska et al., 2024). Nature-Based Solutions should be understood as a group of biotechnological and ecohydrological tools that contribute to increasing the potential for sustainable development. NBS are based on processes that naturally occur in ecosystems. These solutions can be used both on a micro-(e.g. within the area of individual households) and on a macro-scale (e.g. the entire landscape) (Piwowarska & Kiedrzyńska, 2022). However, these solutions need to be implemented in both the agricultural landscape and urban areas to effectively eliminate heavy metals from the environment; to effectively reduce the outflow of heavy metals into the Baltic, they need to experience widespread implementation throughout the Baltic Sea catchment area.

6.1 Nature-Based Solutions in urban landscape

One element of the urban landscape used to improve water quality is the green roof. These consist of a growth substrate, a filtration layer, and a drainage and insulation layer, and vegetation selected based on drought tolerance, ground cover ability, and aesthetics (Yan et al., 2024). When eliminating heavy metals from water, a key role is played by the substrate used in the green roof. A mixture of 20% vermiculite, 30% perlite, 20% crushed brick, 10% sand, and 20% coconut peat has been shown to be over 97% effective in removing *inter alia* Cr (99.2% elimination of initial concentration) Cd (99.9%), Pb (99.9%), Cu (98.9%) Ni (97.1%) and Zn (97.4%) (Vijayaraghavan, & Raja, 2014). Another solutions are the rain gardens, i.e. specially-designed gardens consisting of specific porous substrates and drought and flood resistant plants. The shallow depressions of the gardens retain water, allowing it to re-enter the soil, and any pollutants are subject to phytoremediation (Sharma & Malaviya, 2021). Analyses of laboratory rain gardens showed that the top-soil system was able to reduce Cu contamination by 69% and

Zn by 71.4%. Sand and sand-topsoil mix removed up to 83.3% of Cu, 94.5% of Zn and 97.3% of Pb (Good et al., 2012).

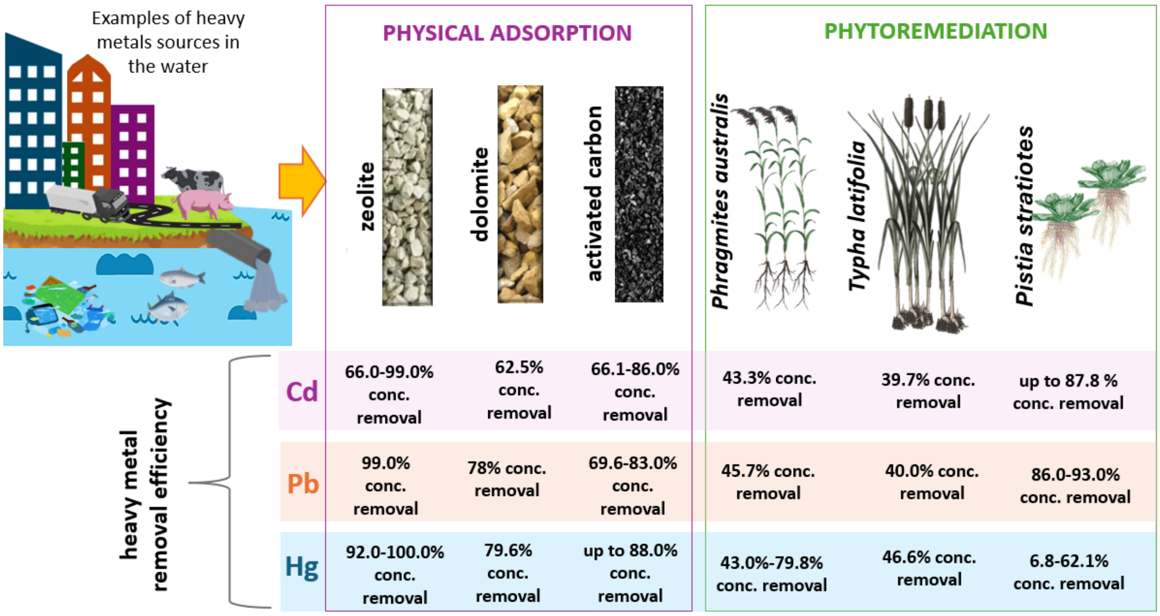
Both green roofs and rain gardens play important roles in the concept of the *sponge city*. This concept assumes the construction of rainwater systems with a low environmental impact; these allow faster adaptation to changes and the free migration of rainwater in the city area. They hence increase water absorption, infiltration, retention and, above all, purification, and release water into the environment during periods of drought (Guan et al., 2021). A particularly promising approach in the bioremediation of limited urban spaces is the use of sequential sedimentation-biofiltration systems (SSBS). This technology combines processes such as sedimentation, filtration and adsorption, as well as phytoaccumulation and rhizofiltration. Such systems are used, for example, to eliminate biogenic compounds and PCBs from rainwater or treated sewage (Jarosiewicz et al., 2024; Kiedrzyńska et al., 2017).

6.2 Nature-Based Solutions in agricultural landscape

NBS can be successfully used to protect aquatic ecosystems from area pollution flowing from catchment areas (Kiedrzyńska et al., 2021). One such example are buffer zones, which act as transition zones between aquatic and terrestrial ecosystems. They serve to protect soil, store water, stop the spread of pollutants and enrich the landscape. One such use is to eliminate heavy metals from mine leachates (Feng et al., 2021; Izydorczyk et al., 2015). Studies conducted on an 8km buffer zone surrounding a non-ferrous metal mine in Suxian, China, found the concentrations of As, Pb, Cu and Zn to decrease with increasing distance from the mine. It was also observed that Cu concentration fell more quickly than the others in the buffer zones: with reductions of 66% within a 1 km buffer zone and 64% in 2–4 km; in comparison, As concentration fell by 64% in 1–2 km, and Pb by 85% in 4–6 km and 87% in 6–8 km. Further out, Cu fell by 82% and Zn by 78% within 4–6 km and by 87% and 74% in the 6–8 km buffer.

In the buffer zones, the remediation processes appear to be most affected by the slope of the terrain and the occurring winds (Ding et al., 2017).

In addition to buffer zones, area pollution can also be eliminated using constructed wetlands (CW), which are low-cost, effective, environmentally-friendly technologies based on the physical and biological processes of *inter alia* macrophyte plants (Piwowarska et al., 2024). It is a hybrid technology combining geofiltration and phytoremediation; the latter is based on the absorption of pollutants by plants and their ability to grow root systems enabling rhizofiltration and rhizodegradation, as well as their symbiosis with microorganisms (Bianchi et al., 2021). CWs can be successfully used to purify heavy metals from, among others, domestic and industrial sewage and pollutants flowing from agricultural areas (Fig. 8).



	PHYSICAL ADSORPTION			PHYTOREMEDIATION		
	zeolite	dolomite	activated carbon	*Phragmites australis*	*Typha latifolia*	*Pistia stratiotes*
Cd	66.0-99.0% conc. removal	62.5% conc. removal	66.1-86.0% conc. removal	43.3% conc. removal	39.7% conc. removal	up to 87.8 % conc. removal
Pb	99.0% conc. removal	78% conc. removal	69.6-83.0% conc. removal	45.7% conc. removal	40.0% conc. removal	86.0-93.0% conc. removal
Hg	92.0-100.0% conc. removal	79.6% conc. removal	up to 88.0% conc. removal	43.0%-79.8% conc. removal	46.6% conc. removal	6.8-62.1% conc. removal

Fig.8 Examples of effective heavy metal elimination using selected components of biofiltration systems (based on Karnib et al., 2014; Merrikhpour & Jalali, 2013; Walker et al., 2005; Melamed & Da Luz, 2006; Egirani et al., 2021; Kumari & Tripathi, 2015; Soto-Ríos et al., 2018; Anning et al., 2013; Kumar et al., 2019; Zhou et al., 2013; Maine et al., 2001).

In wetlands, heavy metals can be removed by adsorption to fine-grained sediments and organic matter, precipitation as insoluble salts such as sulfides, absorption and by deposition of suspended solids at low flow levels (Marchand et al., 2010). Importantly, research on 20-year-old CW systems found them to be efficient (Knox et al. 2021). They demonstrated constant removal of Cu over time, with an approximate 80% reduction being achieved despite large changes in influent concentrations. The mean reduction of Pb from industrial wastewater was 67% in 2004 and 74% in 2020; for Zn, these values were 52% and 65% (Knox et al., 2021). A laboratory study in Canada using *Typha latifolia* and mine leachates showed that constructed wetland was effective in reducing lead concentrations from 0.19 mg/L on entry to 0.10 mg/L at the wetland outlet: a lead elimination efficiency of almost 53% (Etteieb et al. 2021) High reduction rates were also obtained in a pilot CW consisting of *Canna indica* and *Typha latifolia* plants in eastern Sicily. The system achieved 95.6% reduction of Cd, 99.2% reduction of Pb, 93.6% reduction of Cr, 87.8% reduction of Fe, 91.9% reduction of Cu and 97.6% reduction of Zn (Ventura et al., 2021).

7. Conclusions

Growing urbanization, population growth and industrial development in the Baltic Sea region has led to intensive emission of heavy metals to the marine ecosystem. Their accumulation in the aquatic environment, toxicity to organisms and transfer in trophic networks emphasize the need for systematic monitoring and effective management of their emissions. Our findings have cast a clearer light on the contribution of individual Baltic countries to the inflow of heavy metals to the Baltic Sea. It has also allowed these countries to be grouped based on similarities in emission sources and metal transport mechanisms. Results indicate that:

- The largest mean annual loads of cadmium and lead enter the Baltic Sea with river waters, while the highest loads of mercury come from atmospheric deposition.
- The highest total annual loads of heavy metals reaching the Baltic Sea with river waters come from Sweden (Pb, Hg, Cu) and Finland (Cd, Zn, Cr, Ni). However, the highest river

loads of metals per 1 km² of the catchment area are generated by Lithuania (Cd, Pb, Cu) and Finland (Ni, Zn, Cr). Regarding direct point sources, higher loads of Cd, Pb, Hg, Zn and Cu come from Sweden, and Cr, Ni, Cu from Finland.

- Significant factors influencing the emission of heavy metals to river waters, and then to the Baltic, are aquaculture production, animal breeding (cows, pigs, sheep, poultry) and related meat production, as well as agriculture (nitrogen fertilizers), waste generation and transport.
- Clustering analysis, based on *inter alia* annual loads of heavy metals, showed a clear division of the Baltic countries into the eastern and western blocs. This division reflects the complexity of the processes of metal transport to the Baltic Sea from its catchment and indicates common environmental challenges requiring coordinated actions at the regional level.

Eliminating heavy metals from the ecosystem requires a reduction in their emissions, especially from point sources such as sewage treatment plants and urban areas. An effective solution may be Nature-Based Solutions: these are effective and ecological methods of removing pollutants, implemented in both urban and agricultural environments.

8. Acknowledgments

The authors wish to acknowledge Michal Matuszewski MSc. for his support in the analysis of reports and data collection. The research was conducted as part of the Farmikro Project, funded entirely by the National Science Centre, Poland, Opus 22 (Project No. 2021/43/B/ST10/01076). For the purpose of Open Access, the authors have applied a CC-BY public copyright licence to any Author Accepted Manuscript (AAM) version arising from this submission.

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