

## Changes in heavy metal concentrations in the sediments of the Gulf of Finland over two decades

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**Abstract.** The Gulf of Finland was studied in two international campaigns, in 1995 and 2014, with the purpose of mapping heavy metals in surface sediments. The data from the campaigns reveal that the situation with heavy metal concentrations has partly improved, but not at the same pace for all metals. Mercury concentrations have, on average, halved, and cadmium concentrations have decreased considerably across the Gulf of Finland. Copper and zinc show some moderate local increase, but the maximum increase of 61.5% for copper has occurred at the westernmost station. Similarly to the other metals, copper and zinc demonstrate a decrease at the easternmost station. Lead and chromium show a moderate overall decrease across the gulf, while arsenic shows a slight increase in the western part, but a general decrease in the rest of the gulf.

Most of the heavy metal data from 2014 exceed the threshold levels of the sediment quality guidelines, but not in a particularly high degree. We can thus report a very satisfactory general trend for all the studied elements except for zinc, which has not decreased to a degree that would be satisfactory and still exceeds the probable effects level at one site in sediments deeper than 5 cm.

**Keywords:** heavy metals, sediment, toxicity, sediment quality, environment, improvement.

### INTRODUCTION

The Gulf of Finland is known to have been loaded with anthropogenically derived heavy metals for decades. The load was acknowledged, and several authors studied these matters in the 1990s. Annual deposition of heavy metals and arsenic in the sediments of the Gulf of Finland was estimated by Borg and Jonsson (1996) and Vallius and Leivuori (1999). According to these estimates, the annual metal and arsenic deposition in the Gulf of Finland in the early 1990s varied between 1.2 ta<sup>-1</sup> and 1.6 ta<sup>-1</sup> for mercury (Hg), 7.4 ta<sup>-1</sup> and 10.7 ta<sup>-1</sup> for cadmium (Cd), 277 ta<sup>-1</sup> and 452 ta<sup>-1</sup> for lead (Pb), 246 ta<sup>-1</sup> and 374 ta<sup>-1</sup> for copper (Cu), 1400 ta<sup>-1</sup> and 1818 ta<sup>-1</sup> for zinc (Zn) and between 59 ta<sup>-1</sup> and 122 ta<sup>-1</sup> for arsenic (As), depending on the author (summary table in Vallius and Leivuori 1999, table 3, p. 28). In the course of the Gulf of Finland Year 1996 studies, a multitude of sites in the Gulf of Finland were visited and investigated during the previous field season in 1995. The results sometimes showed high contamination of heavy metals in the near-surface

sediments, but so that in most cases a decrease in metal concentrations was detected in the topmost part of the cores (Leivuori 1998; Vallius 1999a; 1999b; Vallius and Leivuori 1999; Leivuori 2000; Vallius and Leivuori 2003). When the Gulf of Finland Year 2014 was launched, one of the ideas was to check whether the situation had improved in some of these old sites at exactly the same coordinates where the studies were carried out about 20 years earlier. This was planned together with Finnish, Russian, and Estonian scientists, and cores were taken by these parties on different cruises during the field season of 2014.

### STUDY AREA

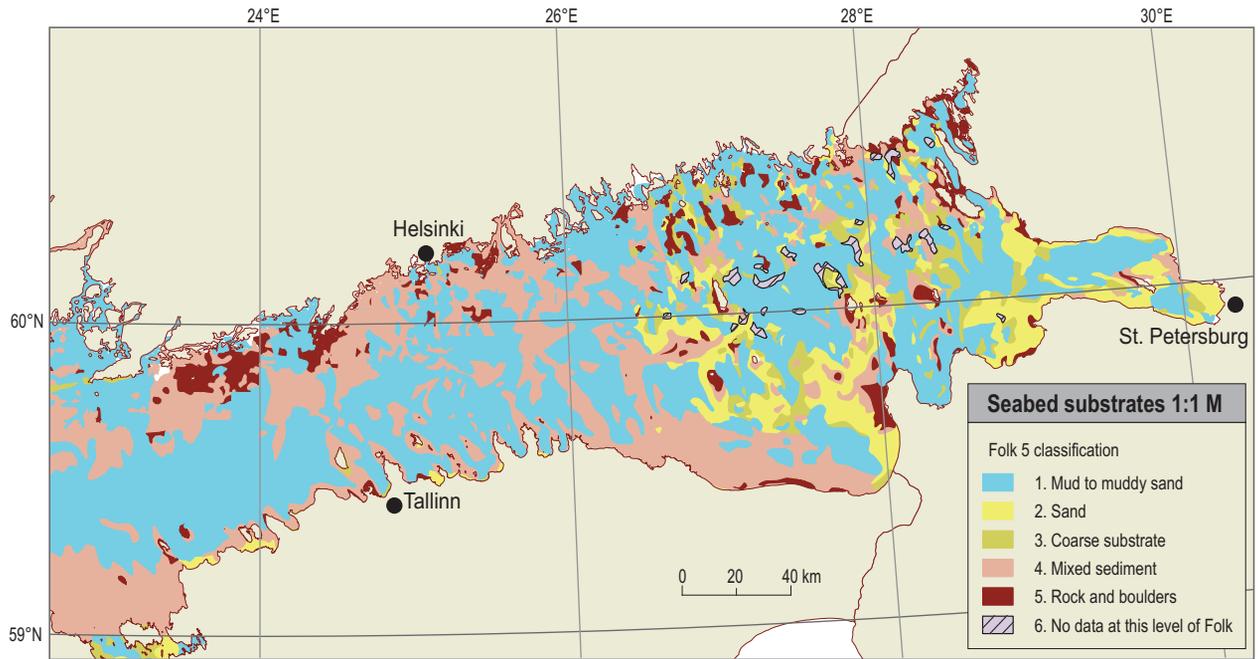
The study area comprises the whole Gulf of Finland, a rather shallow eastward-trending gulf of the main Baltic Sea. The geology of the Gulf of Finland basin is diverse. First, the bedrock in the Gulf of Finland area is divided into two very different parts (Fig. 1).



Fig. 1. Bedrock geology of the Gulf of Finland area (modified from Koistinen 1996 and Tuuling 2019).

The bedrock on the northern side of the gulf consists of Precambrian crystalline rocks, while the bedrock on the southern side is composed of sedimentary rocks ranging in age from the Ediacaran (i.e., ca 600 My) to Middle-Ordovician (450 My), lying on top of the Precambrian basement. The Precambrian basement is considerably harder and more durable than the sedimentary rocks on top of it. Due to the differences in erosion tolerance between the Finnish and Estonian bedrocks, there are clear differences in the bedrock topography between the two sides of the Gulf of Finland, which is also reflected in the nature of the seafloor on the southern and northern sides of the gulf. Seabed depressions formed by bedrock, glacial till, and old sediments act as sedimentation basins where younger sediments have been deposited over the past millennia to form almost horizontal clay and silt-clay deposits (Winterhalter 1992; VSEGEI Atlas 2010; Fig. 2).

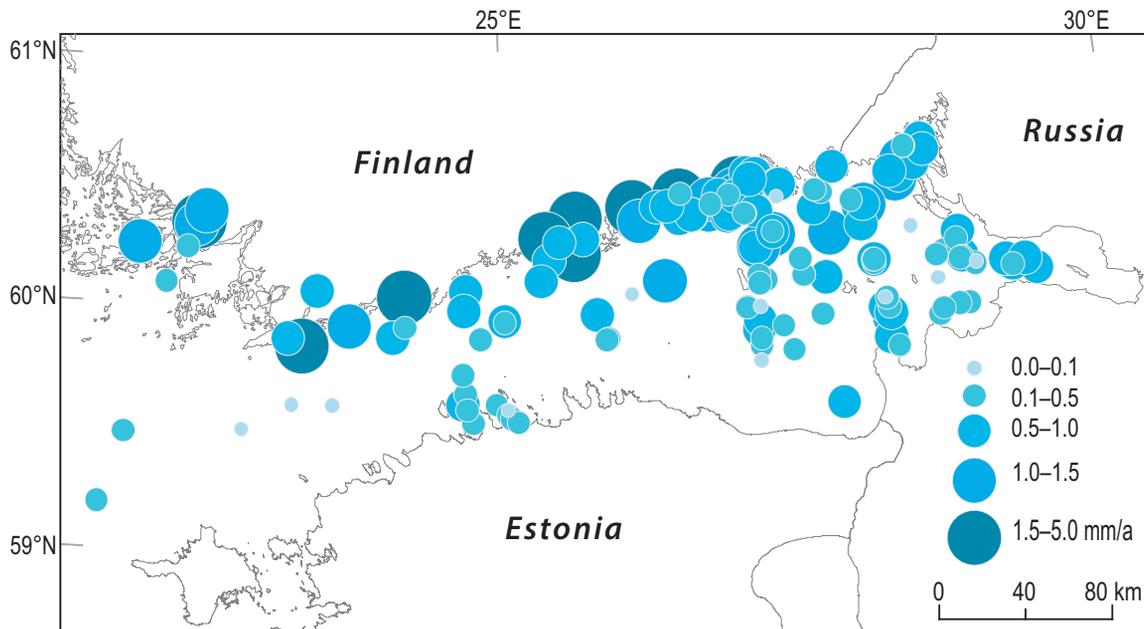
The youngest units, often found in the top layers of these sedimentation basins, are recent clays and muds with higher concentrations of organic matter and heavy metals that are known to adsorb onto fine-grained and organic matter. Due to the different bedrock topography of the northern and southern parts of the gulf, the accumulation basins of fine particulate matter are different. On average, the accumulation basins in the northern part are smaller and more irregular in shape than the basins in the central and southern parts of the gulf. In spite of the heterogeneity of the gulf's bottom, many good representative basins can be found in almost all areas of the Gulf of Finland. The natural accumulation rate in the sedimentation basins of the eastern (Russian) part of the Gulf of Finland reaches 0.6–0.9 cm/year (Ryabchuk et al. 2015), in central and western parts 0.4–0.8 cm/year, with considerably higher rates in sheltered near-coastal basins, up to 2.75 cm/year



**Fig. 2.** Seabed substrates of the Gulf of Finland in Folk 5 classification. Source: EMODnet Geology (<https://www.emodnet-geology.eu>).

(Vallius 2015c). The cores at stations JML, GF1, E5, and XVI, dated for the present study, were observed to have net sedimentation rates of  $0.36 \text{ cm a}^{-1}$ ,  $0.54 \text{ cm a}^{-1}$ ,  $0.39 \text{ cm a}^{-1}$ , and  $0.36 \text{ cm a}^{-1}$ , respectively. Thus, sedimentation rates

vary rather significantly in different parts of the gulf, which was also observed by Ilus et al. (2007). This is also evidenced by the open access EMODnet Geology data (Fig. 3).



**Fig. 3.** Sedimentation rates in the Gulf of Finland (mm/a). Source: EMODnet Geology (<https://www.emodnet-geology.eu>).

## MATERIALS AND METHODS

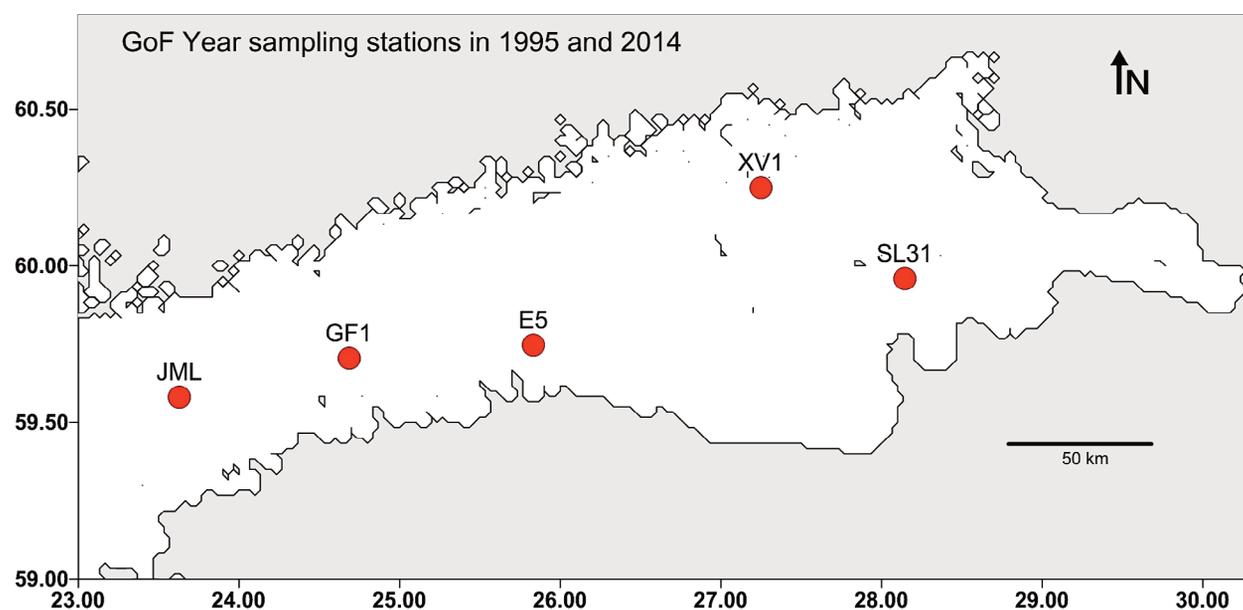
This study involves five sites in different parts of the Gulf of Finland, which were sampled in 1995 and 2014, in the two consecutive Gulf of Finland Year programs (Fig. 4 and Table 1). The samples in 1995 were taken by Finnish scientists on three different spring and summer cruises of R/V Aranda. The 2014 sampling was performed by scientists from Russia, Estonia, and Finland on three research cruises of R/V Aranda, R/V Salme, and R/V Barracuda. The samples from Finnish waters at stations JML and XV1 were taken on 14 and 16 May, respectively, under calm conditions. The Russian sample from station SL31 was taken on 15 July and the Estonian samples from stations GF1 and E5 on 15 May and 22 July, respectively.

In order to have comparable results for this study, it was important to be able to repeat all the procedures that were taken into account in 1995. In practice, the most important thing was to reach the same location during the 2014 year coring as during the previous study. This was achieved by coring accuracy within 20 meters of the target, usually much better. Also, the sampling was performed exactly according to the old routines, including gravity coring, slicing the topmost part of the core into 10 mm subsamples, storing the slices in plastic bags refrigerated at +4 °C until laboratory procedures, and everything was performed systematically, avoiding contamination. The only difference in the methods was the corer, as the corer in 1995 was a slightly smaller version of the GEMAX corer, the GEMINI, with an inner diameter of 80 mm instead of 90 mm for the GEMAX corer used in

**Table 1.** Sampling stations, coordinates in decimal degrees, water depth in meters

Station	Latitude	Longitude	Water depth
JML	59.5817	23.6300	80
GF1	59.7050	24.6850	83
E5	59.7472	25.8340	73
XV1	60.2500	27.2505	63
SL31	59.9592	28.1453	40

2014. This was the only drawback in the 1995 study, as in many cases the amount of dry matter in the topmost centimeter was too low for complete chemical analyses, typically only a couple of grams of dry matter. Thus, the 1995 and 2014 surface concentrations were compared per the second centimeter of the surface (1–2 cm sediment depth). After the fieldwork, the cores were transported to Finland, where they were sent to the chemical laboratory Labtium (today Eurofins Labtium), the same laboratory that made the analyses already in 1995, then called the Chemistry Laboratory of the Geological Survey of Finland. By using the same laboratory, we could avoid any systematic errors between the methods of different laboratories. In the laboratory, the samples were weighed for wet weight, freeze dried, and weighed for dry weight. After that, all subsamples were homogenized and sieved into <2 mm fraction to remove unnecessary objects such as plant or animal remains. In practice, 95% of the sample material itself was <65 µm fraction; thus, no further sieving was necessary.



**Fig. 4.** Gulf of Finland (GoF) with the sampling stations indicated.

After the described pre-treatment, the samples were digested with hydrofluoric-perchloric acids, followed by elemental determination using inductively coupled plasma mass spectrometry (ICP-MS, As, and Cd) or inductively coupled plasma atomic emission spectrometry (ICP-AES, cobalt (Co), chromium (Cr), Cu, Pb, and Zn), depending on the element. Mercury was measured with a Hg analyzer through pyrolytic determination.

The commercial sediment reference materials QCGBMS304-6, QCMES-4, QCNIST8704 were used for assessing measurement accuracy. For all reference materials, element concentrations measured for each sample batch were well within  $\pm 10\%$  of the certified values, except for As, which for QCMES gave an average recovery of 119%, and Pb, which showed an average recovery of 91%, but with the extremes of 61.4% and 117.2%. However, with the two other SRMs (standard reference materials), Pb fell perfectly within  $\pm 3\%$  of the certified values (101.00% and 102.67%, respectively). Thus, it seems that there might have been a problem with the sample matrix of QCMES-4 for the laboratory's method.

In order to assess the quality of the sediments of the Gulf of Finland, the obtained sediment concentrations of As, Cd, Cr, Co, Cu, Hg, Pb and Zn were compared with the sediment quality guidelines (SQGs) established by the Canadian Council of Ministers of the Environment (CCME 2002). This classification uses two reference values, levels referred to as the "interim sediment quality guideline" (ISQG, lower reference value) and the "probable effects level" (PEL, upper reference value), where the ISQG is considered the "threshold effects level" (also known as TEL).

## RESULTS AND DISCUSSION

### Change in arsenic and metal concentrations in the surface sediments between 1995 and 2014

When comparing the results of 2014 with the old data from the 1995 sampling campaign, we had to use the second-to-topmost samples for comparison, as many of the topmost sediment data were missing from the old dataset because of too small samples obtained for chemical analyses by the old GEMINI corer. Thus, to compare these two datasets, slices from the 1–2 cm sediment depth of both sampling campaigns were used (Table 2).

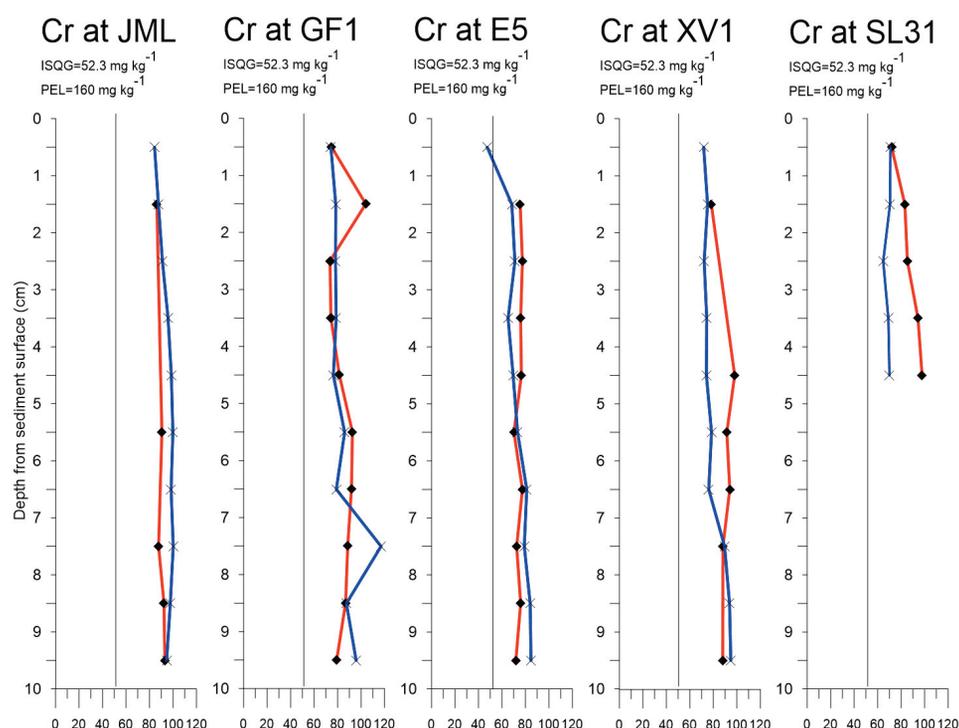
A comparison of these data revealed that Cd, Cr, Hg, and Pb have decreased at all coring sites during the two decades considered. However, the decrease in Cr is almost negligible when looking at the vertical profiles of the Cr concentrations in the cores from both the 1995 and 2014 campaigns (Fig. 5). The strongest decrease is noticed for Hg, with a decrease from 30% at station XV1 up to ca 78% at station E5. The decrease in Cd was almost as strong, with a decrease from ca 16 up to 71%. The patterns of decrease for these metals are rather similar, with the exception of the westernmost station JML, where the decrease in Cd is weaker than elsewhere, only 16%, while the decrease in Hg is quite high there, around 65%. The decrease in Pb varies between 7 and 42%, depending on the site, while Cr has decreased between 4 and 24%, except at the westernmost station JML, where no decrease was observed. It is common for all these metals with a general decrease through the two decades that the lowest decrease is generally observed at the northeastern station

**Table 2.** Arsenic and heavy metal concentrations ( $\text{mg kg}^{-1}$  dry weight) in the surface sediments (1–2 cm sediment depth) of stations JML, GF1, E5, XV1, and SL31 in 1995 and 2014, as well as the degree of change (decrease/increase in %) at each station

Site	As in 1995	As in 2014	Decrease/Increase (%)	Cd in 1995	Cd in 2014	Decrease/Increase (%)	Co in 1995	Co in 2014	Decrease/Increase (%)	Cr in 1995	Cr in 2014	Decrease/Increase (%)
JML	7.81	8.91	<b>14.1</b>	0.66	0.55	<b>-16.2</b>	14.1	16.4	<b>16.3</b>	86.2	87.4	<b>1.4</b>
GF1	10.0	6.58	<b>-34.2</b>	0.69	0.37	<b>-46.1</b>	14.6	16.3	<b>11.6</b>	104	78.4	<b>-24.6</b>
E5	14.1	6.77	<b>-52.0</b>	1.01	0.37	<b>-63.0</b>	16.6	13.5	<b>-18.7</b>	75.5	68.4	<b>-9.4</b>
XV1	12.2	12.6	<b>3.3</b>	2.76	1.65	<b>-40.2</b>	16.9	19.6	<b>16.0</b>	78.3	75.1	<b>-4.1</b>
SL31	8.96	6.90	<b>-23.0</b>	1.55	0.45	<b>-71.0</b>	16.8	12.2	<b>-27.4</b>	83.1	70.5	<b>-15.2</b>

Site	Cu in 1995	Cu in 2014	Decrease/Increase (%)	Hg in 1995	Hg in 2014	Decrease/Increase (%)	Pb in 1995	Pb in 2014	Decrease/Increase (%)	Zn in 1995	Zn in 2014	Decrease/Increase (%)
JML	32.7	52.8	<b>61.5</b>	0.125	0.044	<b>-65.2</b>	44.5	34.6	<b>-22.2</b>	154	185	<b>20.1</b>
GF1	33.9	40.5	<b>19.5</b>	0.133	0.053	<b>-60.2</b>	45.4	32.7	<b>-28.0</b>	133	155	<b>16.3</b>
E5	35.6	34.0	<b>-4.5</b>	0.167	0.037	<b>-77.7</b>	42.0	24.1	<b>-42.6</b>	158	136	<b>-13.9</b>
XV1	54.6	67.2	<b>23.1</b>	0.212	0.148	<b>-30.2</b>	60.8	56.6	<b>-6.9</b>	229	255	<b>11.4</b>
SL31	41.8	35.0	<b>-16.3</b>	0.207	0.076	<b>-63.1</b>	51.8	36.3	<b>-29.9</b>	209	139	<b>-33.5</b>



**Fig. 5.** Chromium concentrations ( $\text{mg kg}^{-1}$ ) in the 1995 core (red lines) and the 2014 core (blue lines) at five stations in the Gulf of Finland lined from west to east (for locations, see Fig. 4). Threshold value (ISQG)  $52.3 \text{ mg kg}^{-1}$  is indicated by the vertical line.

XV1, for which no single reason is clarified in this study. The same applies to the westernmost station JML, except that mercury has also substantially decreased there. All the other metals (Zn, Co, Cu and As) have shown some decrease in some parts of the Gulf of Finland, such as at stations E5 and SL31, but there has also been a notable increase at other sites, up to ca 20%. Cu, however, has clearly increased more at the westernmost station JML, with an increase of 60%, which can be considered a major rise. Except for Hg, the overall degree of decrease or increase in the metals is irrelevant, but it can be observed that the decrease has been smallest at the westernmost station JML, which might be attributed to the fact that most metal concentrations were comparatively low already in 1995, and thus a strong decrease was not foreseen. Similarly, there has been a general trend of a slighter decrease in the metals in the sediment surface at station XV1 in the northeast and a generally observed stronger decrease in concentrations at station SL31 in the east.

#### Sediment quality in the Gulf of Finland as revealed by the core samples

In the Baltic Sea, sediment quality has generally not been assessed according to the SQGs, except for a study in the Gulf of Gdansk (Bettinetti et al. 2009) and two earlier

studies in the Gulf of Finland (Vallius 2015a; 2015b). This study reports a measure of sediment quality as it has been evaluated using the SQGs established by the Canadian Council of Ministers of the Environment (CCME 2002), which was used by Vallius (2015a; 2015b) as a measure of sediment toxicity in the classification of the sediments in the Gulf of Finland. Since Co is not reported in the SQG used, its quality is not discussed here. In practice, the SQG should be considered a screening tool for the probability of toxicity rather than the actual levels of toxicity. As indicated above, the guidelines are based on two fixed levels for each reported element – the ISQG (also called the TEL) and the PEL. The ISQG is the threshold below which toxic effects on biota are observed in less than 10% of the cases. The PEL is the level above which toxicity is probable for most species, as toxicity is observed in more than 50% of the cases for most species. Between the ISQG and the PEL, toxicity is occasional, ranging from 10 to 50% of the cases.

Figure 5 shows the Cr data from the 1995 campaign plotted together with the 2014 data as concentration curves for each station. As seen in the 2014 curve, the Cr concentrations have shown a general decreasing trend during the recent decades, except in the east where a weak increasing trend is observed in the uppermost part of the short core. However, even there, the concentrations are at a relatively

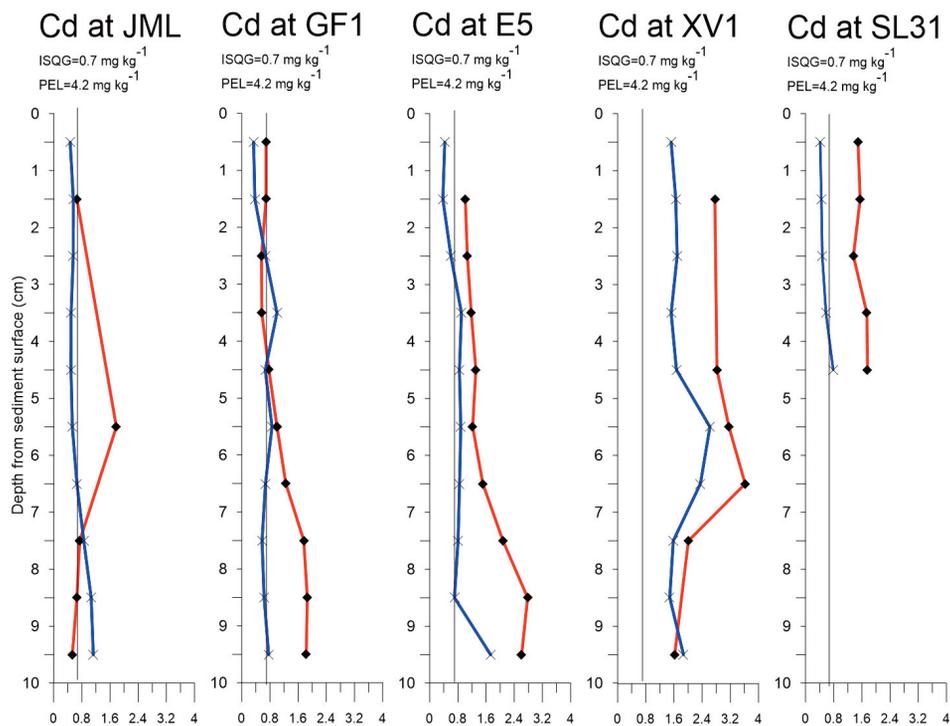
low level, only less than 20 mg kg<sup>-1</sup> above the threshold (ISOQ) value of 52.3 mg kg<sup>-1</sup>, but considerably below the PEL of 160 mg kg<sup>-1</sup>. The same applies to the rest of the cores, where only the westernmost station JML has Cr values slightly higher above the threshold (about 35 mg kg<sup>-1</sup>), but the uppermost sample at station E5 has reached the threshold value of 52.3 mg kg<sup>-1</sup>. Only one sample at a depth of 7–8 cm at station GF1 clearly exceeds 100 mg kg<sup>-1</sup>, but is still comfortably below the PEL level. Chromium should not pose a risk to seafloor biota, according to this study.

Figure 6 shows the Cd (mg kg<sup>-1</sup>) data from the 1995 campaign plotted together with the 2014 data as concentration curves for each station. The concentrations in the 2014 data show mainly lower levels than in the 1995 study and a clearly decreasing trend towards the top of the cores. In all cases, except station XV1 in the northeastern Gulf of Finland, these levels are very close to the threshold value of 0.7 mg kg<sup>-1</sup>. At station XV1, the concentrations are at a clearly lower level than in 1995, but still about twice the threshold value, yet well below the PEL of 4.2 mg kg<sup>-1</sup>. Thus, at most of the studied sites, sediment cadmium concentrations seem to be below the risk level to seafloor biota.

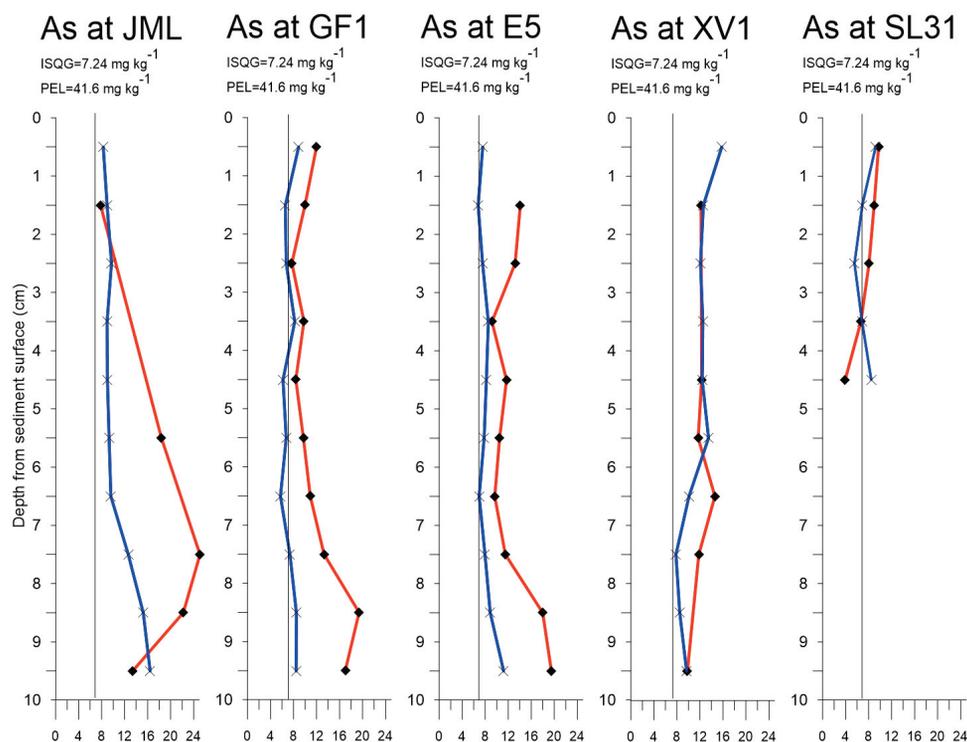
Figure 7 shows the As (mg kg<sup>-1</sup>) data from the 1995 campaign plotted together with the 2014 data as concentration curves for each station. On average, the tem-

poral trend of As seems to increase over time in the east and decrease or remain at previous levels in the central and western parts of the Gulf of Finland, where the concentrations are close to the applied SQG threshold level of 7.24 mg kg<sup>-1</sup>. However, all the obtained concentrations are well below the PEL of As – 41.6 mg kg<sup>-1</sup>. Previous higher As values (from 1995) have decreased, and thus As cannot be seen as posing a threat to seafloor biota at the studied locations.

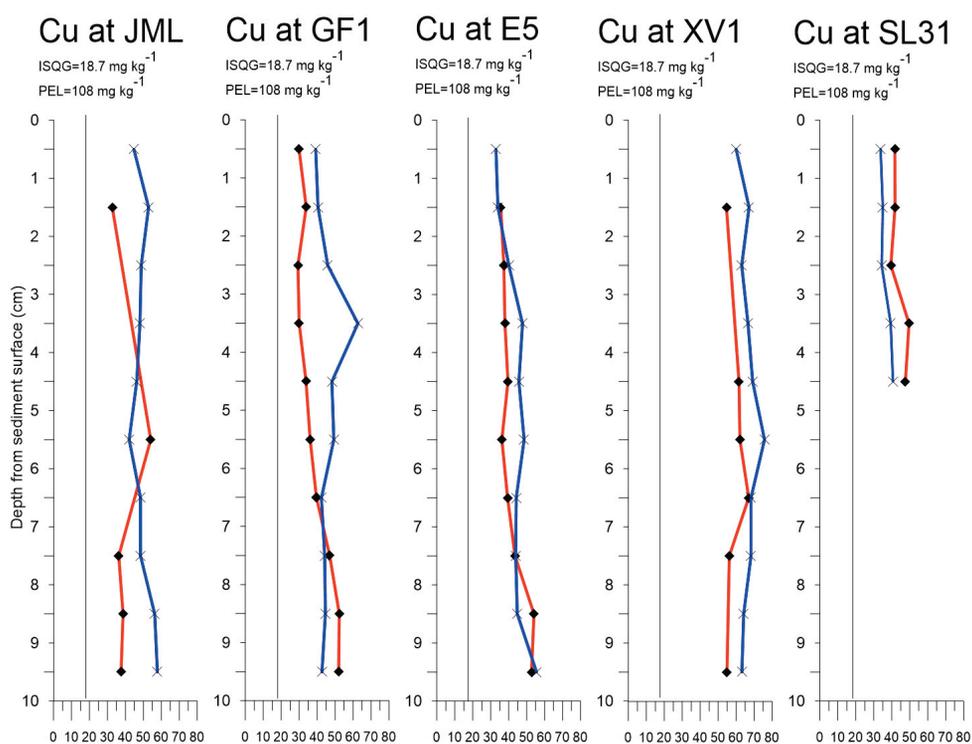
Figure 8 combines the Cu data from 1995 and 2014. Only at station SL31 in the eastern Gulf of Finland were the 2014 concentrations plotted completely below the Cu concentrations curve of the 1995 core. It is disappointing to see that the favorable decreasing trends that prevailed until 1995 seem to have become an increasing trend at some sites in the Gulf of Finland, as the 2014 data on concentration in the western and central parts of the gulf were partly higher than the 1995 data. We have no explanation for this change in the trend for Cu, but still a slight decreasing trend in Cu concentrations can be observed at the top of the 2014 curves during the most recent years. All the Cu concentrations clearly exceed the threshold value (ISOQ) of 18.7 mg kg<sup>-1</sup>, about two times. In the worst case, at station XV1, in the northeastern area of the gulf, the Cu values reach 70 mg kg<sup>-1</sup>, whereas the PEL is 108 mg kg<sup>-1</sup>. Thus, it can be concluded that Cu concen-



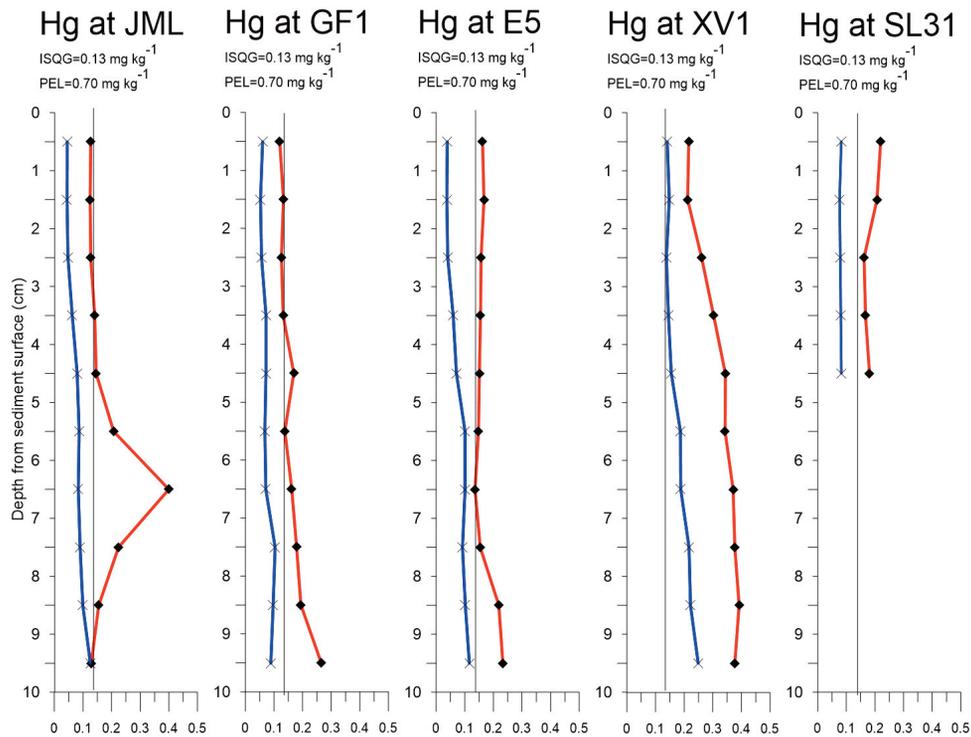
**Fig. 6.** Cadmium concentrations (mg kg<sup>-1</sup>) in the 1995 core (red lines) and the 2014 core (blue lines) at five stations in the Gulf of Finland lined from west to east (for locations, see Fig. 4). Threshold value (ISOQ) 0.7 mg kg<sup>-1</sup> is indicated by the vertical line.



**Fig. 7.** Arsenic concentrations ( $\text{mg kg}^{-1}$ ) in the 1995 core (red lines) and the 2014 core (blue lines) at five stations in the Gulf of Finland lined from west to east (for locations, see Fig. 4). Threshold value (ISQG)  $7.24 \text{ mg kg}^{-1}$  is indicated by the vertical line.



**Fig. 8.** Copper concentrations ( $\text{mg kg}^{-1}$ ) in the 1995 core (red lines) and the 2014 core (blue lines) at five stations in the Gulf of Finland lined from west to east (for locations, see Fig. 4). Threshold value (ISQG)  $18.7 \text{ mg kg}^{-1}$  is indicated by the vertical line.



**Fig. 9.** Mercury concentrations ( $\text{mg kg}^{-1}$ ) in the 1995 core (red lines) and the 2014 core (blue lines) at five stations in the Gulf of Finland lined from west to east (for locations, see Fig. 4). Threshold value (ISQG)  $0.13 \text{ mg kg}^{-1}$  is indicated by the vertical line.

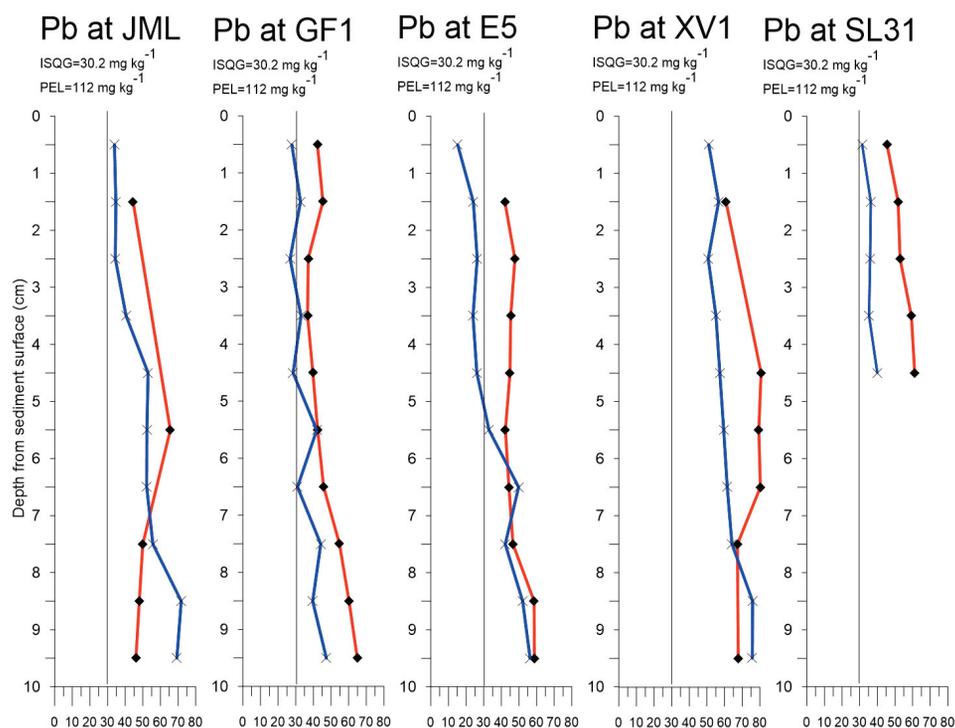
trations in the surface sediments do not probably pose a significant threat to seafloor biota in the surveyed areas.

Figure 9 combines old and new Hg data curves for the five studied stations. The temporal trend for Hg is decreasing over time, and all the observations in 2014 are at substantially lower levels than in 1995, which can perhaps be considered the most satisfactory result of this study. In fact, all the 2014 curves, except the one in the northeastern Gulf of Finland (station XV1), are plotted below the threshold (ISQG) level of  $0.13 \text{ mg kg}^{-1}$ . Even there, the topmost part of the curve lies almost exactly at that value, indicating that Hg load has decreased to a minimum even in the vicinity of the River Kymijoki, which is known to be the main source of Hg pollution in the Gulf of Finland (Verta et al. 2009; HELCOM 2010). Nevertheless, it is typical for Hg, especially in this area, that concentrations are considerably higher in deeper sediment layers, typically below sediment depths of 20 cm (Vallius 2015a; 2015b). Thus, Hg should not, in general, be of major concern for most benthic biota, as only a few species dwell deeper than 20 cm, provided that no human activity or strong bottom currents erode the seafloor in such a way that the older sediments are exposed to benthic animals or biota at the seafloor or in the water column. However, there is one species that has been prominently abundant during recent years and dwells

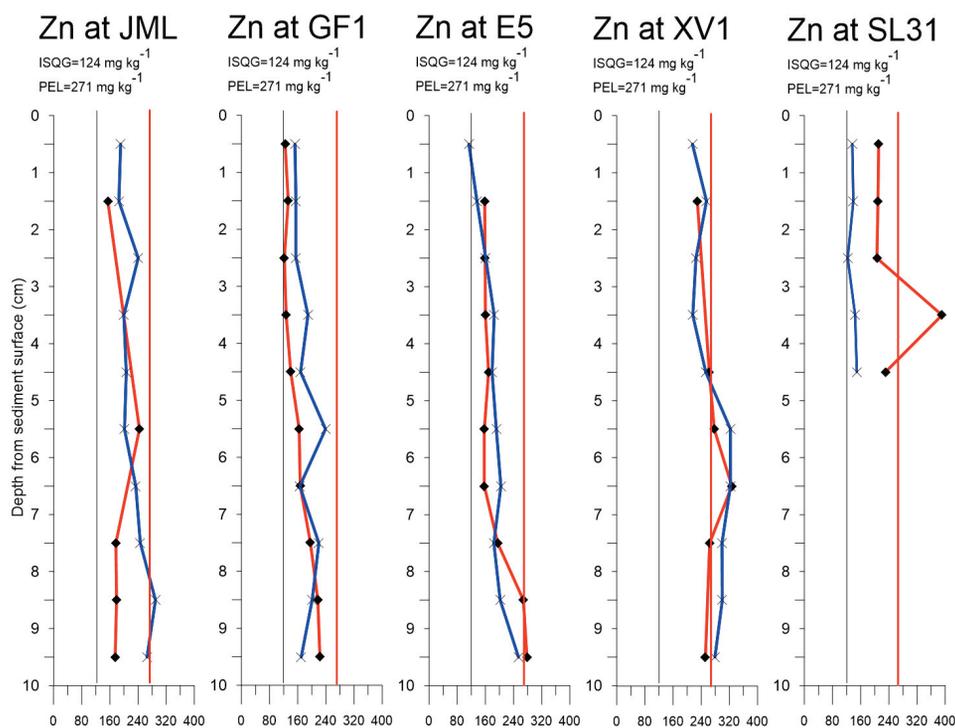
easily deeper than 20 cm, *Marenzelleria* spp. It can cope with poor oxygen and high sulfide conditions, practically thriving in areas of hypoxia (Granberg et al. 2008; Maximov 2011). Thus, it is possible that Hg is transported vertically upwards to the surface of the sediment column during the lifetime of this species. Although Granberg et al. (2008) reported a bioturbation driven release of organic contaminants mediated by *Marenzelleria neglecta*, this is unlikely to occur for Hg to a greater extent since its concentrations are low in the surface sediments of station XV1.

Figure 10 combines the Pb data from 1995 and 2014. The concentrations of the 2014 data show mainly lower levels than in the 1995 study and an interesting feature in this figure is the strong decrease in Pb concentrations over time at all sites, especially at station E5, where the decrease is very strong and concentrations in the topmost sediment are at levels below the threshold level of  $30.2 \text{ mg kg}^{-1}$ . During the sampling in 2014, Pb concentrations were elevated only at station XV1, but even there they were well below the PEL of  $112 \text{ mg kg}^{-1}$ .

Figure 11 combines the Zn data from 1995 and 2014. An interesting feature in Fig. 11 is the fact that, except for SL31 in the east, most of the 2014 curves plot on average similar or even slightly higher concentrations than in the old data, which is not satisfactory considering the state of



**Fig. 10.** Lead concentrations ( $\text{mg kg}^{-1}$ ) in the 1995 core (red lines) and the 2014 core (blue lines) at five stations in the Gulf of Finland lined from west to east (for locations, see Fig. 4). Threshold value (ISQG)  $30.2 \text{ mg kg}^{-1}$  is indicated by the vertical line.



**Fig. 11.** Zinc concentrations ( $\text{mg kg}^{-1}$ ) in the 1995 core (red lines) and the 2014 core (blue lines) at five stations in the Gulf of Finland lined from west to east (for locations, see Fig. 4). Threshold value (ISQG)  $124 \text{ mg kg}^{-1}$  is indicated by the grey vertical line. The red vertical line indicates the PEL of  $271 \text{ mg kg}^{-1}$ .

the surface sediments in the Gulf of Finland. At station XV1, the concentrations in the surface sediments are in fact very close to the PEL of  $271 \text{ mg kg}^{-1}$ , which can be considered a rather alarming trend in the Gulf of Finland. On average, however, the temporal trends of Zn are slightly decreasing but are still partly at too high levels, except at station E5, where in the 2014 core the Zn concentration in the surface sediment layer has decreased below the threshold level of  $124 \text{ mg kg}^{-1}$ . At station SL31, the concentrations have decreased substantially and are now very close to the threshold level. Also, at station GF1, the concentrations are rather close to the threshold level.

## CONCLUSIONS

It is obvious from the comparison of the Gulf of Finland sediment data from the 2014 and the 1995 core analyses that mercury and cadmium concentrations have decreased markedly during the two decades, with mercury concentrations having halved on average across the Gulf of Finland, and cadmium having decreased between 16 and 71%, depending on the station. This is very encouraging, especially as the previous study seemed to indicate that cadmium had remained at the same level, and no improving trend could be envisaged (Vallius and Leivuori 1999). Additionally, the trends of lead and chromium are promising. Although arsenic has not decreased to such a degree in some areas, its concentrations in the sediments of the Gulf of Finland were never alarmingly high. On the other hand, the observed average As recovery of 119% can largely explain the slightly higher levels in 2014. Thus, we report an overall very satisfactory trend for all the studied elements, except for zinc, which has not decreased to the degree that would be sufficiently satisfactory, taking into account the toxicity levels that it presented already in the 1990s and still presents today, at least in the northeast, but partly also in other parts of the Gulf of Finland.

Looking at the decreasing trend of the metal concentrations, the most promising decrease is observed in the east and south and the least promising in the west and north. For mercury, the decrease is very good in all areas, which is also the case for cadmium, perhaps with slight inertia in the northeast, but still a positive trend for both metals. The differences in the decreasing trend for all the studied metals over the two observed decades are most probably attributed to changes in metal loads in the different areas of the Gulf of Finland.

In practice, when comparing the data with the SQGs, much of the heavy metals data from 2014 still exceed the threshold levels. Interestingly, mercury concentrations in the topmost sediments have decreased to such a degree that they are now below or, in the worst case, very close

to its threshold level. None of the samples reaches the PEL in the surface sediments, but zinc is close to that and exceeds the PEL considerably in the sediment at a depth of 5–10 cm at site XV 1 in the northeastern Gulf of Finland, being the metal of most concern according to this study.

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## Muutused Soome lahe setete raskmetallide sisaldustes viimasel kahel aastakümnel

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Aastakümnete jooksul on inimtegevuse tõttu akumulunud Soome lahe setetes raskmetalle. Kahel Soome lahega seonduvatele keskkonnaprobleemidele pühendatud aastal, nn Soome lahe aastal 1995 ja 2014, viisid kolme Soome lahe ümbritseva riigi teadlased läbi ulatuslikud rahvusvahelised uuringud selgitamaks, milline on lahe keskkonnaseisund. Sealhulgas uuriti raskmetallide kontsentratsioone Soome lahe pindmistes setetes.

Käesolevas artiklis võrreldakse aastail 1995 ja 2014 viiest erinevast Soome lahe punktist kogutud pindmiste setteprofiilide raskmetallide (Zn, Cu, Co, Cr, Pb, Hg, Cd, As) sisaldusi. Et tulemused oleksid võrreldavad, koguti setteproovid eri aastatel võimalikult täpselt samadest kohtadest ja raskmetallide analüüsid tehti ühes ja samas laboratooriumis sama meetodikat kasutades.

Tulemused näitavad, et peaaegu 20 aasta möödudes on raskmetallide kontsentratsioonid lahe setetes üldiselt vähenenud, kuid mitte ühetaoliselt kõigi metallide osas. Hg-sisaldused on umbes poole väiksemad ning samuti on Cd-kontsentratsioonid oluliselt vähenenud kogu Soome lahes. Pb- ja Cr-sisaldused on kõigis analüüsitud punktides mõõdukalt vähenenud. Sama kehtib As osas, kuid lahe läänepoolsel alal on selle kontsentratsioonid setetes veidi tõusnud. Cu ja Zn aga näitavad lokaalselt kõrge taset, kusjuures kõige läänepoolsemas punktis on Cu-kontsentratsioon tõusnud lausa üle 60%. Ometi on kõige idapoolsemas punktis Cu- ja Zn-sisaldused sarnaselt teiste raskmetallidega vähenenud.

Raskmetallide kontsentratsioone Soome lahe setetes võrreldi Kanada Keskkonnaministeeriumi kehtestatud setete kvaliteedijuhistega. Enamuse raskmetallide sisaldused 2014. aastal kogutud proovides ületavad sealseid lävitasemeid, kuid mitte oluliselt. Seetõttu võib pidada Soome lahe pindmiste setete seisundit raskmetallide sisalduse osas küllaltki heaks, väljaarvatud Zn-kontsentratsioonid, mis pole vähenenud piisavalt ja ületavad endiselt ühes proovivõtupunktis nn tõenäolise mõju taset.