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Pollution history of Neva Bay bottom sediments (eastern Gulf of Finland, Baltic Sea)

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Abstract Neva Bay is the shallowest and easternmost part of the Gulf of Finland (Baltic Sea). St. Petersburg, Russia's second largest city, occupies the coastal area where the Neva River debouches into Neva Bay. St. Petersburg has a protracted history of industrial, transportation and urban related activity that have affected Neva Bay. By the sealing off the bay from the eastern Gulf of Finland, the St. Petersburg Flood Protective Facility, which was constructed from the 1970‘s to 2011, transformed Neva Bay into a "technogenic" lagoon. Neva Bay sediments record a unique history of pollution near the metropolis. Heavy metal concentrations of most elements studied varied consistently throughout sediment cores. Temporal trends indicate that metals started to accumulate abruptly in the first half of the 20th century. Zinc, lead and copper were the first metals to reach contaminant thresholds implicating the regional base metal industry as a source. Significant increase in cadmium levels a decade or two later suggests pollution from the regional chemical industry. Comparison of geochemical data collected from sediment cores and recent annual sediment surveys highlighted the temporal history and potential sources of pollution in Neva Bay. Intensive dredging in 2007–2008 resuspended and redistributed contaminated sediment around Neva Bay causing a dramatic increase in benthic sediment heavy metal concentrations. Concentrations of all measured metals subsequently declined from 2009–2014 relative to the elevated values observed for 2007–2008. Pollution history of Neva Bay bottom sediments is closely linked with changing of sedimentation conditions. Analyses of sedimentological data collected by 20th and 21st century scientific surveys reveal dramatic shifts in Neva Bay sedimentation processes over the last three centuries. The western part of Neva Bay has transitioned from a sand-dominated system to one of mud accumulation with the aerial extent of mud deposition expanding significantly during the 20th century. This inventory coupled with an understanding of primary natural and anthropogenic processes can help inform decision makers to support the overall ecological health of the bay.

Keywords *• bottom sediments • geochemistry • pollution • Neva Bay*

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INTRODUCTION

The Gulf of Finland plays an important role in ecosystem health of the Baltic Sea. Located between three countries with developed industry, transport and urban activities – Finland, Russia and Estonia – it is highly impacted by anthropogenic processes, e.g. changes in seabed substrate composition and morphology, extraction of sediments and minerals, dredging, dumping, construction of infrastructure, input of hazardous substances, nutrients and litter etc. (Raateoja, Setälä 2016).

Neva Bay is the easternmost and shallowest part of the Gulf of Finland (Fig. 1A). Anthropogenic modification of Neva Bay and its coastal areas began with the foundation of St. Petersburg by Peter the Great in 1703. In the 20th century, St. Petersburg developed into a metropolis of 5 million people with significant industrial and transportation related activities, including several major ports, as well as intense dredging and dumping. These developments have caused high anthropogenic impact on the Gulf of Finland and its ecosystem.

The Gulf of Finland has received a considerable load of anthropogenic harmful substances during the past decades (Raateoja, Setälä 2016). Heavy metal input into the Gulf of Finland began to increase in the 1950's due to the postwar industrialization. The input peaked from the 1960's to the 1970's, and started to decline in the mid-1980's (Vallius 2009, 2012, 2014). Despite this decreasing trend, there are still areas where heavy metal concentrations in the seabed sediments are relatively high (Vallius 2014, 2016). According to a study from the 1990's (Vallius 1999) the surface sediments of easternmost part of the Gulf of Finland was characterized by highest heavy metal concentrations. Sediments of easternmost Gulf of Finland were one of the most important sources of secondary pollution for the westerly parts of the gulf.

Geochemistry of the Gulf of Finland bottom sediments has been studied since early 1980s by Finnish (Leivuori 1998; Vallius 1999) and Russian (Emelyanov 1995) researches and in frame of several pan-Baltic international projects (Borg, Jonsson 1996; Brügmann, Lange 1990; Pertilä 2003 etc.). It is important to mention, however, that Neva Bay as very shallow area has never been sampled during scientifical cruises from large research vessels (e.g. Russian Institute of Oceanology). It has resulted in a gap of published data about Neva Bay sediment geochemistry. For example, maps and description of trace elements in surface layer of sediments, based on results of the project implemented at the beginning of the 1990s under the auspices of ICES and the HELCOM, in a book "Geochemistry of the Baltic Sea" (Uścinowicz 2011, 217–220) did not include any information about Neva Bay.

Results of recent biological research (Golubkov 2014; Maximov 2014; Ryabchuk *et al.* 2017 etc.) revealed significant change in benthic communities linked with transformation of sediment environment caused by anthropogenic processes. Geochemistry of the bottom, including concentration of harmful elements in the bottom sediments and main trends of its change is an important indicator of ecosystem health.

The main goal of this article is to assess the recent status of Neva Bay sediment environment. The tasks of presented research are to establish the level of potentially harmful elements (Co, Ni, Cu, Pb, Zn, Cr, V, As and Cd) concentration and to analyze centennial to annual trends of heavy metal accumulation and redistribution in Neva Bay bottom sediments.

STUDY AREA

Neva Bay spans 21 km in length and reaches a maximum width of 15 km to cover an area of 329 km^2. Its average depth is 3.5 m and it contains a water volume of about 1.2 km^3. Neva Bay generally deepens toward the Kotlin Island. Local shallows appear along southern and northern coasts of the bay. Maximum depths occur in the central - western part of the bay (5–6 m), within former underwater sand-mining careers (10–12 m) and ship-channels (up to 14 m).

The hydrological regime of the bay varies due to its shallow-water depths, frequent changes in hydrometeorological conditions and the strong influence of the Neva River. With a water discharge of 77.6 km^3/year (Bergström, Carlsson 1994) or 75.69 km^3/year (Alenius *et al.* 1998), the Neva River is the largest river draining into the Gulf of Finland. Water level fluctuations, wind waves and currents also represent major hydrodynamic factors affecting Neva Bay, which salinity is fairly low (0–0.3‰). Ice cover forms annually but warmer winters have limited the duration of solid ice cover in recent years (Ryabchuk *et al.* 2011). Neva Bay also experiences flood events with water levels rising more than 1.6 m above mean sea level. The most significant floods in the eastern Gulf of Finland occur due to storm run-up, which in turn results from the combined effects of drift currents and long waves. Wave disturbance reaches depths of 3–3.5 m (Leontiev 2008) exposing virtually the entire Neva Bay benthic surface to periodic wave influence.

For protection from catastrophic floods the St. Petersburg Flood Protection Facility (FPF) – the largest hydrotechnical construction in the Gulf of Finland – was built in 1979–2011 (Fig. 1). At present, Neva Bay connects to the Gulf of Finland through six channels (gates) including the Main Marine Channel. These openings span a total width of about 1 km. When the FPF first separated Neva Bay from the eastern Gulf of Finland in the 1980s, the former became a technogenic lagoon (Ryabchuk *et al.* 2017).

A new phase of anthropogenic modification began in 2006 with the hydraulic filling of 476.7 hectares of the eastern part of the bay near Vasilievsky

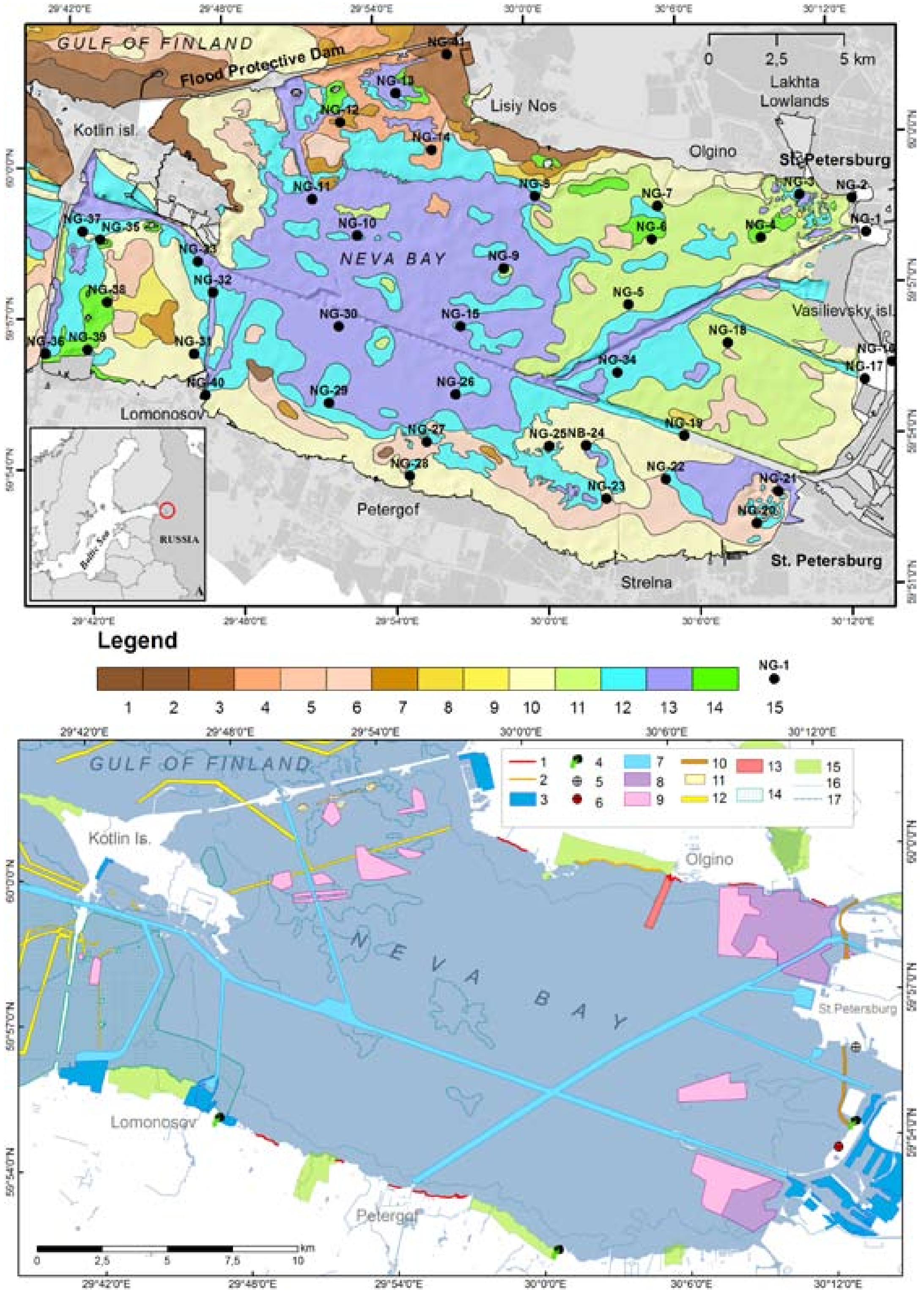

Fig.1 **A**. Sea floor substrate map for Neva Bay and location of monitoring sampling sites. 1 – boulders, pebbles; 2 – pebbles, gravel; 3 – boulders, pebbles, sand; unsorted sand: 4 – mainly coarse-grained sand, 5 – unsorted, mainly medium-grained sand; 6 – mainly fine-grained sand;7 – sand with gravel; 8 – coarse-grained sand; 9 – medium-grained sand;10 – fine-grained sand; 11 – silty sand; 12 – silt; 13 – silty clay-rich mud, 14 – mixed sediments; 15 – sediment sampling sites from the 2011–2015 survey. Compiled by D. Ryabchuk and A. Sergeev, 2016. **B.** Map of technogenic load on Neva Bay bottom. 1 – areas of coastal erosion; 2 – areas of active Aeolian processes; technogenic objects onshore: 3 – area of cargo port; 4 – unauthorized wastewater discharge; 5 – dumping place of industrial and urban waste (with coast protection structures); 6 – dumping place of industrial and urban waste (without coast protection structures); technogenic objects onshore: 7 – navigation channels; 8 – area of former underwater sand excavation; 9 – dumping sites of dredged sediments; 10 – highway on pillar bridge (constructed in 2015–2016); 11 – stone crib-bars; 12 – wooden crib-bars; 13 – underwater pipeline; 14 – area of dredging; 15 – nature protection areas; 16 – isobaths; 17 – rivers. Compiled by V. Zhamoida and A. Sergeev, 2014

Island for the new Passenger Port of St. Petersburg. These operations dredged large amounts of seafloor sediment (e.g. clay-rich material) to deepen channels for vessels/cruisers with draughts of up to 14 m. The sediments were removed and dumped in former sand extraction quarries found in the northeastern corner of the bay near Lakhta. Construction of the Bronka harbor along the western part of Neva Bay began in 2008 (Fig. 1B).

MATERIALS AND METHODS

Given its critical role in St. Petersburg development, detailed records of Neva Bay bathymetry and surface sediment types date back to the first half of the 18th century (Fig.2). Scientific investigations of Neva Bay started in 1920–1924 surveys by Professor Konstantin Deryugin, who collected geochemical, biological and hydrological data (Deryugin 1923). Results from earlier investigations thus allow comparison with more recent geological and geochemical surveys of the bay.

Systematic marine geological surveys of Neva Bay benthic sediments began in the 1980's. Since 1987, scientists from the A.P.Karpinsky Russian Research Geological Institute (VSEGEI) have conducted detailed geological surveys and systematic analysis of Neva Bay benthic sediments (Spiridonov *et al.* 2007; Atlas … 2010). A sedimentological and geochemical dataset has been constructed from more than 1000 sediment samples collected from 1987–2002. This data has contributed to Quaternary maps detailing benthic cover and litho-facies distributions in Neva Bay at different scales (from 1:200,000 to 1:25,000).

From 2005–2007, the joint Russian-Finnish SAMAGOL project (Sediment Geochemistry), which included experts from both VSEGEI and the Geological Survey of Finland (GTK), executed cruises that collected 15 sediment cores around the bay using a Niemistö gravity corer. After on-board subsampling (every cm) and preparation, core material was analyzed by gamma spectrometry, ICP-AES and ICP-MS (Table 1).

From 2004 to 2016, side-scan sonar and echosounding profile surveys mapped more than 50% of the

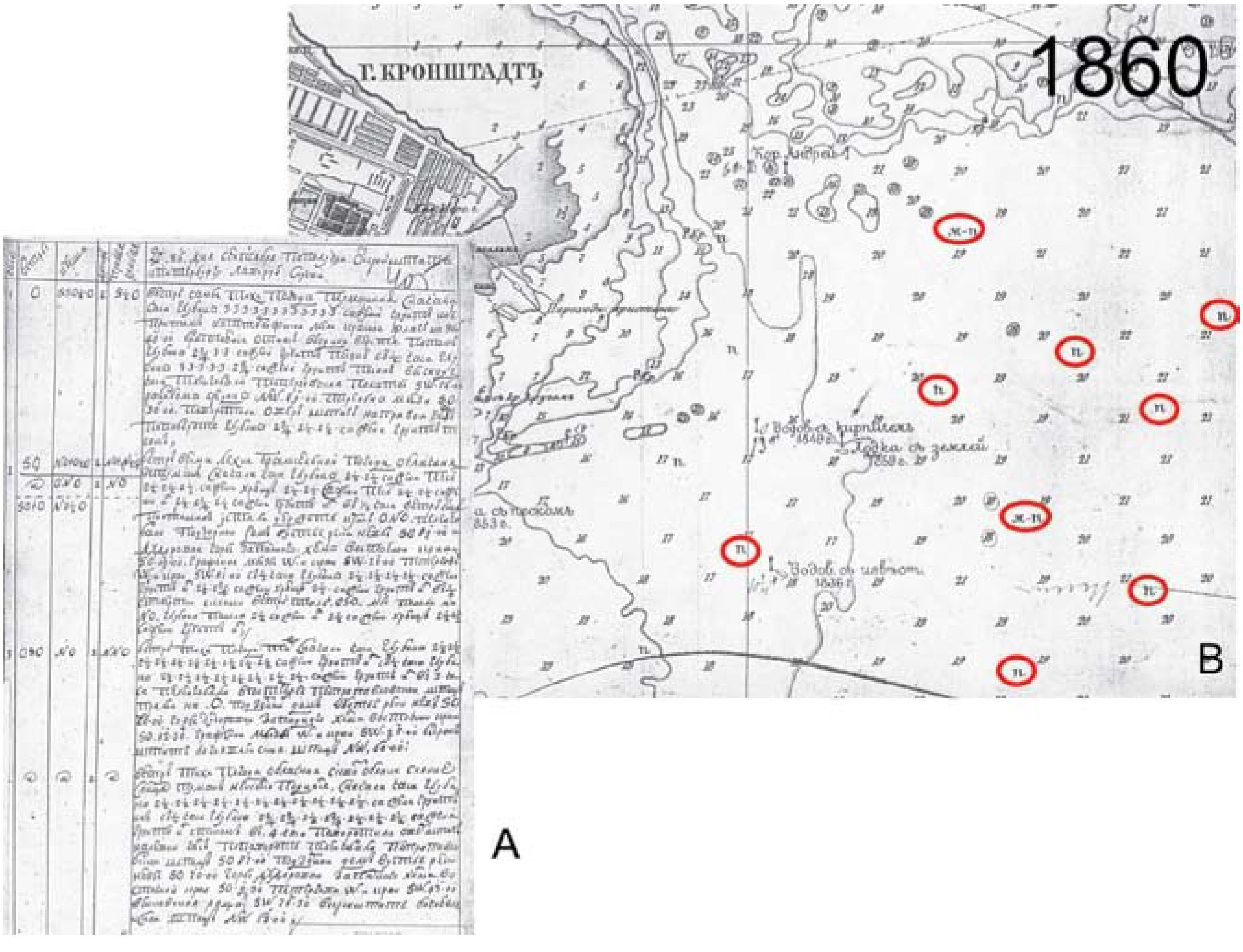

Fig. 2 Historical materials of Neva Bay studies. **A**. Page of hydrographical journal by skippers Vasily Karpov and Matew Verkhovtsev with description on Neva Bay depths and bottom sediments, 1751; **B.** fragment of nautical map of Neva Bay showing sandy sediments within area of recent mud accumulation (marked by circles). Compiled by D. Ryabchuk, 2017

Table 1 Characretistic of method of analysis of heavy metals and detection limits

Year	Milling procedure	Digestion procedure	Equipment	Certified/ standard reference materials	Detection limits	
					Element	Limit, ppm
2005	Samples were freeze dried, homogeni-zed and sieved into <2mm fraction	Complete digestion with hydrofluoric-perchloric acids Sample is digested twice with mixture of hydrofluoric and per chloric acid and evapo-rated until dry on a hot plate. Finally the residue of samples are dissolved to nitric acid and diluted with water	ICP - AES ICP-MS	Commercial standard reference materials MESS-2 and NIST8704	As	0.5
					Cd	0.1
					Co	0.2
					Cr	4
					Cu	2
					Ni	4
					Pb	1
					Zn	5
					V	0.5
1993 2005–2015	Milling (up to 3 mm) using Geological Cutting Machine SCD-6; up to 0.074 mm using litium disk eraser LDI-65	Digestion in muffler LOIP LF-5/11 G1 (T 520^0C)	OESA (optical emission spectral analysis) STE-1	Standard Materials SDPS2	Co	1
					Cr	0.5
					Cu	0.5
					Ni	1
					Pb	2
					Zn	10
					V	2
		HNO_3+HCl (As)	ICP-MS Agilent-7700	State Standard Sample 3484-86	As	1
		HNO_3+HF+$HClO_4$ (Cd)			Cd	0,1

nearshore areas around Neva Bay (Spiridonov *et al.* 2008). Benthic sediments were sampled at more than 150 sites. From 2005 to 2009 the "Sevmorgeo" Institute carried out annual monitoring at 33–40 sampling stations. Since 2011, VSEGEI has conducted annual monitoring in Neva Bay that focuses on contaminants in coastal zone sediments. Monitoring includes geochemical and sedimentological analysis of benthic sediments (upper 2 cm) sampled from the same 35–40 sites every year (Fig.1), supplemented with additional samples from varying localities. The samples are analyzed for grain-size and semiquantitatively for compositional properties using optical emission spectral analysis (27 elements, including Co, Ni, Cu, Pb, Zn, Cr and V) and ICP-MS for Cd and As concentrations in 2014–2015. On an annual basis, 150 to 190 samples from backshore environments (subaerially exposed areas of the coastal zone) have been analyzed by these methods, resulting in a sedimentological and geochemical dataset by 2015, which is partially interpreted below (Information Bulletin 2007, 2008, 2009).

Ascertaining heavy metal contamination in benthic sediments requires comparison with reference values. This study compared Neva Bay sample data with the Swedish Environmental Quality Criteria for sediments (Swedish EPA 2000) and the sediment quality guidelines (SQG's) issued by the Canadian Council of Ministers of the Environment (CCME 2002).

The Swedish EPA (2000) criteria (WGMS 2003) compare total concentrations for a range of elements with reference or background estimates for five gradual levels of contamination. The five levels are numbered as classes 1–5 and assigned qualitative descriptors of "little or none" to "very large". This approach does not help constrain potential ecological impacts or toxicity of contaminated sediment but does provide a categorical framework for comparing different metal concentrations (Table 2). Using the Swedish EPA (2000) criteria also allows for comparison of results with previous studies conducted in the Gulf of Finland (Vallius, Leivuori 2003; Vallius *et al.* 2009). Canadian sediment quality guidelines (CCME 2002) are based on toxicity tests. Vallius (2014) recently used these SQG's for evaluation of Gulf of Finland sediments. The CCME (2002) classification uses two reference value (ISQG, lower reference value) and "probable effect level" (PEL, upper reference value) (Table 2).

Grain-size analyses of all benthic sediments were carried out in VSEGEI laboratories using a "Microsizer 201A" laser diffractometer (VA Instal, Russia) and an analytical sieve shaker (AS 200 Retsch). Heavy metal concentrations from benthic and backshore (coastal/beach) sediments were then interpreted for temporal, spatial, and size-fraction related patterns.

RESULTS

Sediment geochemistry

Fine- to very fine sands and silty-sands and silts dominate among the surficial sediments of Neva Bay bottom. Silty clay-rich mud accumulates in the center

Table 2 Contamination quality criteria for heavy metals used by the Swedish Environmental Protection Agency (2000; ppm relative to dry weight, total analysis; WGMS 2003; Vanadium levels not listed) (Vallius *et al.* 2007) and the Canadian Council of Ministers of the Environment (CCME, 2002). Criteria for Ni, Co and V not listed. Compiled by H. Vallius and D. Ryabchuk

Metal (ppm, dry weight)	WGMS 2003					CCME, 2002		Regional background for Neva Bay coast (VSEGEI, 2006)	Regional background for soils (Gorky et al. 2006)
	Class 1 Little or none	Class 2 Slight	Class 3 Significant	Class 4 Large	Class 5 Very large	ISQG, lower reference value	PEL, upper reference value		
As	<10	10-16	16-26	26-40	>40	7.24	41.6	-	2.6
Cd	<0.2	0.2-0.5	0.5-1.2	1.2-3	>3	0.7	4.2	-	0.17
Co	<14	14-20	20-28	28-40	>40			3.9	4.1
Cr	<80	80-110	110-160	160-220	>220	52.3	160	27	12.5
Cu	<15	15-30	30-60	60-120	>120	18.7	108	18	18
Ni	<33	33-43	43-56	56-80	>80			8.7	15.3
Pb	<31	31-46	46-68	68-100	>100	30.2	112	20	19.1
Zn	<85	85-125	125-195	195-300	>300	124	271	40	43.1

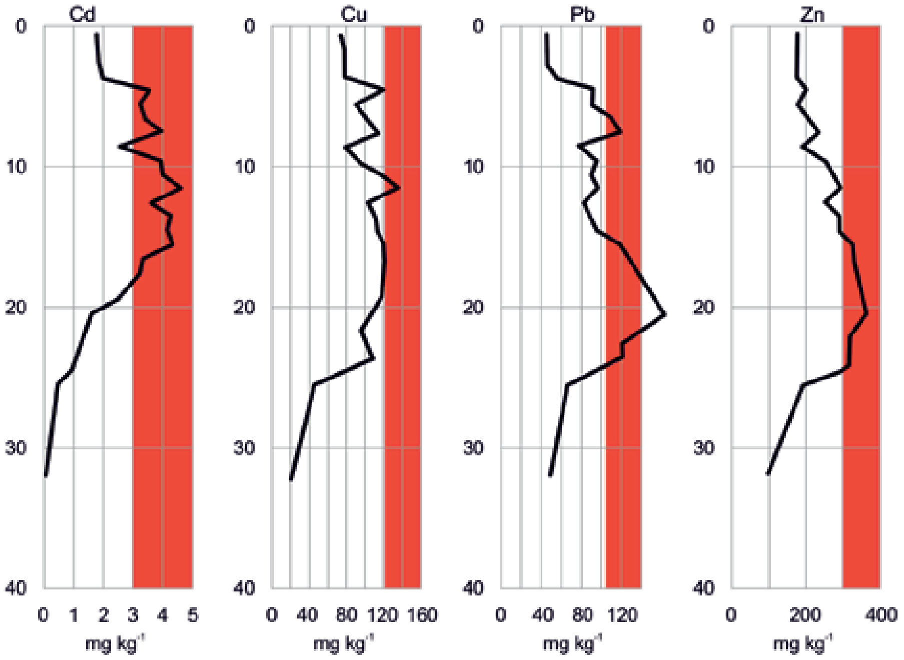

Fig. 3 Concentration curves for cadmium (Cd), copper (Cu), lead (Pb), and zinc (Zn) in Neva Bay sediment cores (NG-2005-9, collected 9 June 2005; sampled every centimeter from 1–26 cm depth and once from the 26–36 cm interval). Vertical axis shows core depth while red zone marks the "very high" contamination levels for respective metals in sediment according to Swedish EPA (2000) guidelines. Compiled by H. Vallius, 2006

of the western part of the bay, where water depths exceed 4 m and within anthropogenic depressions (Fig.1). Coarse-grained sediments cover the bottom surface of the near-shore of the western part of the bay.

The vertical concentration curves of most heavy metals (e.g. Cd, Zn, Pb, Cu) analyzed from cores sampled in 2005, show similar variation patterns throughout sediment profiles – from low values at 30–40 cm of core depth to drastic increase of concentrations, reaching maximal concentrations at core depth from 25 to 5 cm depending on site, and with a final decrease of concentrations in the uppermost 10-5 centimetres of the cores (Fig. 3).

Sevmorgeo monitoring from 2005–2010 documented an increase in sedimentary concentrations of most heavy metals from 2007–2008. Concentrations subsequently decreased from 2009–2010 (Information Bulletin 2007, 2008, 2009). VSEGEI geochemical monitoring from 2011–2014 documented a continued decline in average of Co, Ni, Pb, Cr and V concentrations, whereas average Cu and Zn concentrations showed a slight increase over the last three years (Fig. 4, Tables 3, 4).

Fig. 5 shows the distribution of Cu, Pb, Zn and Cr concentrations (Neva Bay benthic sediments sampled from 2011–2015) relative to CCME (2002)

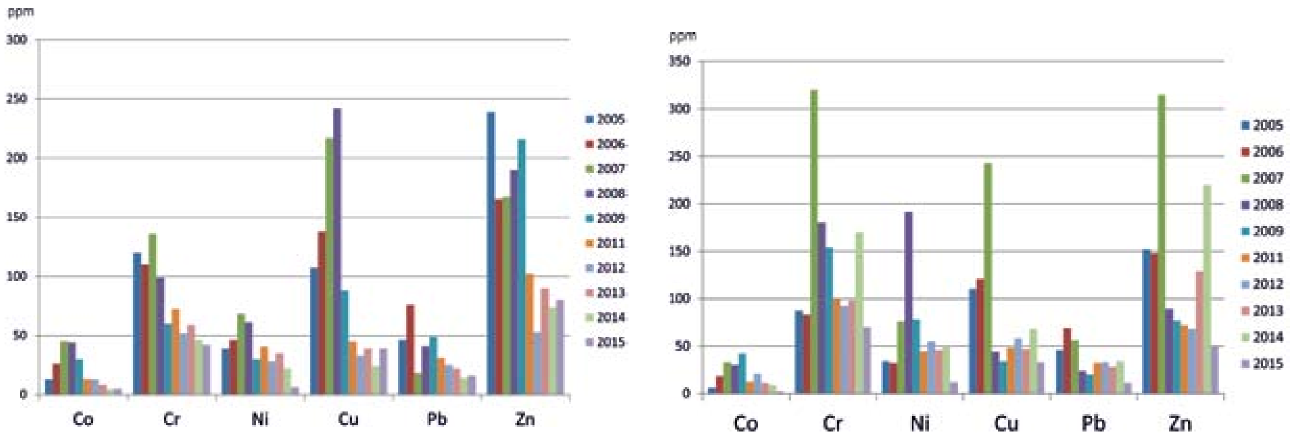

Fig. 4 Upper charts show heavy metal concentrations for **A**) Site NB-3 (from artificial depressions near Lakhta) and **B**) Site NB-10 (from the western sedimentation basin). Data from 2005–2009 collected by Sevmorgeo (Information Bulletin…, 2007; 2008; 2009) and 2011–2015 data collected by VSEGEI. Compiled by D. Ryabchuk, 2016

Table 3 Concentrations (ppm) and descriptive statistics measured from Neva Bay benthic sediments (0–1 cm) in 2011–2015. Data refer to all grain-size fractions. The 'Class' column indicates level of contamination according to Swedish EPA (2000) contamination levels. Compiled by D. Ryabchuk and N. Malysheva

	Co	Class	Ni	Class	Cu	Class	Pb	Class	Zn	Class	Cr	Class	V
2011													
Mean	3.8	1	30.7	1	68.7	4	36.4	2	66.9	1	61.3	1	30.1
SD	1.8		16.1		80.9		26.1		48.9		32.3		16.6
Minimum	0.3	1	2.5	1	11.9	1	11.6	1	15.8	1	21.3	1	2.5
Maximum	7.7	1	71.7	4	477	5	161	5	212.7	4	155	3	66.1
Count	32		32		32		32		32		32		32
2012													
Mean	9.7	1	27.2	1	45.3	3	23.1	1	52.1	1	71.7	1	13.1
SD	5.5		16.0		21.1		8.8		22.9		29.0		5.2
Minimum	0.8	1	3.8	1	10.3	1	13.9	1	22	1	28.8	1	4.9
Maximum	21	3	63	4	90.9	5	57.4	3	113	2	141.3	3	23.8
Count	35		35		35		35		35		35		35
2013													
Mean	4.2	1	23.2	1	59.4	3	27.7	1	100.4	1	61.1	1	36.7
SD	6.2		10.8		54.1		10.5		54.3		30.6		20.9
Minimum	0.9	1	4.1	1	8.7	1	12.0	1	25	1	8.0	1	3.5
Maximum	25.6	3	46	3	266.4	5	64.8	3	222.8	4	137.6	3	87.3
Count	41		41		41		41		41		41		41
2014													
Mean	9.1	1	29.6	1	35.5	4	17.7	1	75.5	1	78.5	1	47.1
SD	4.9		15.8		43.4		7.5		48.5		45.2		25.5
Minimum	2.9	1	9.1	1	6.2	1	8.5	1	20	1	21.8	1	12.4
Maximum	24	3	73	4	210	5	45	2	220	4	231.2	5	119.3
Count	39		39		39		39		39		39		39
2015													
Mean	5.6	1	12.7	1	25.8	2	14.5	1	42.9	1	44.9	1	38.6
SD	4.5		8.2		15.7		3.1		26.7		24.1		19.8
Minimum	1	1	3.4	1	6.4	1	8	1	14	1	6.6	1	9.9
Maximum	19	2	41	2	80	4	22	1	110	2	140	3	87
Count	39		39		39		39		39		39		39

Table 4 Concentrations (ppm) and descriptive statistics measured in 2011–2015 from the sitly clay-rich fraction of benthic sediments (0–1 cm) occupying a sedimentation basin. The 'Class' column indicates level of contamination according to Swedish EPA (2000) contamination levels. Compiled by D. Ryabchuk and N. Malysheva

	Co	Class	Ni	Class	Cu	Class	Pb	Class	Zn	Class	Cr	Class	V
2011													
Mean	5.0	2	44.8	3	123.1	5	53.5	3	88.9	2	99.6	2	41.5
SD	1.4		12.5		126.2		38.9		34.7		32.8		10.6
Minimum	2.4	1	25.7	1	62.4	3	31.2	2	48.2	1	48.3	1	21.6
Maximum	7.7	1	71.7	4	477	5	161	5	165	3	155	3	58
Count	10		10		10		10		10		10		10
2012													
Mean	13.1	1	37.2	2	63.1	4	27.7	1	61.3	1	98.1	1	16.1
SD	4.3		15.3		16.0		11.5		25.7		27.6		4.1
Minimum	6.3	1	14.5	1	40.0	3	16.9	1	27.4	1	58.8	1	9.8
Maximum	21	3	59	3	90.9	4	57.4	3	113	2	141.3	3	22.1
Count	10		10		10		10		10		10		10
2013													
Mean	3.8	1	32.6	1	99.9	4	34.9	2	129.4	3	91.2	1	55.9
SD	5.5		9.9		66.5		13.9		53.1		32.7		19.8
Minimum	0.9	1	16	1	30.6	2	17	1	36	1	40.0	1	24.3
Maximum	25.6	3	46	3	266.4	5	64.8	3	190.4	3	137.4	3	87.3
Count	10		10		10		10		10		10		10
2014													
Mean	9.95	1	36.6	2	55.1	3	23.4	1	106	2	111.1	2	60.7
SD	5.7		16.3		52.4		9.6		58.2		64.2		25.9
Minimum	2.9	1	19	1	16	1	11	1	39	1	44.9	1	28.3
Maximum	24	3	73	4	180	5	45	2	220	4	231.2	4	119
Count	10		10		10		10		10		10		10
2015													
Mean	7.3	1	13.0	1	33.9	3	15.1	1	47.5	1	57.5	1	43.1
SD	4.8		5.3		12.9		2.9		28.5		18.2		16.4
Minimum	1	1	5.6	1	12	1	10	1	23	1	27	1	20
Maximum	19	2	41	4	80	4	22	1	110	2	140	3	80
Count	10		10		10		10		10		10		10

sediment quality guidelines (ISQG and PEL levels for respective elements). Average concentrations of heavy metals currently fall far below their respective PELs. While maximum Zn, Cd and As concentrations did not reach PELs, maximum concentrations of Cu, Pb and Cr exceeded them (Figs 5, 6). While CCME (2002) reference values are not available for Co and Ni, maximum Co and Ni concentrations in Neva Bay sediments did not reach "very high"(Class 5) levels designated by the Swedish EPA (2000). The highest values for Cu and Pb concentrations occurred in areas around the main shipping channel (sites 2011-NB-15 and 2011-NB-26).

Except for Cu, Cd and Pb, average sedimentary concentrations of heavy metals in 2011 did not exceed the "little or none" (Class 1) contaminant level of the Swedish EPA (2000) for most types of sediments. Silty clay-rich muds from the sedimentation basin gave concentrations that classified as slightly contaminated (Class 2) (Fig.6, Tables 3 and 4). Out of 34 samples, four gave Cr concentrations that classified as slightly contaminated (Class 2). Three samples gave Cr concentrations that classified as having "significant" levels of contamination (Class 3). Ni concentrations reached "slight" levels of contamination in eight samples, "significant" levels in seven samples and "large" levels in sample 11-NB-26 (western sedimentation basin). Zn concentrations reached "slight" levels of contamination in six samples, "significant" levels in one sample and "large" levels in two samples (11-NB-2 and 11-NB-28). Most samples analyzed exhibited Class 2 or 3 contamination for Pb and Cu. In terms of Pb concentrations, 13 samples met "slight" contamination criteria, four samples met "significant" contamination criteria, and one sample (11-NB-26) met "very large" contamination criteria. In terms of Cu concentrations, eight samples met "slight" contamination criteria, 7 samples met "significant" contamination criteria, 13 samples met "large" contamination criteria and three samples (11-NB-2, 11-NB-15 and 11-NB-30) met "very large" contamination criteria.

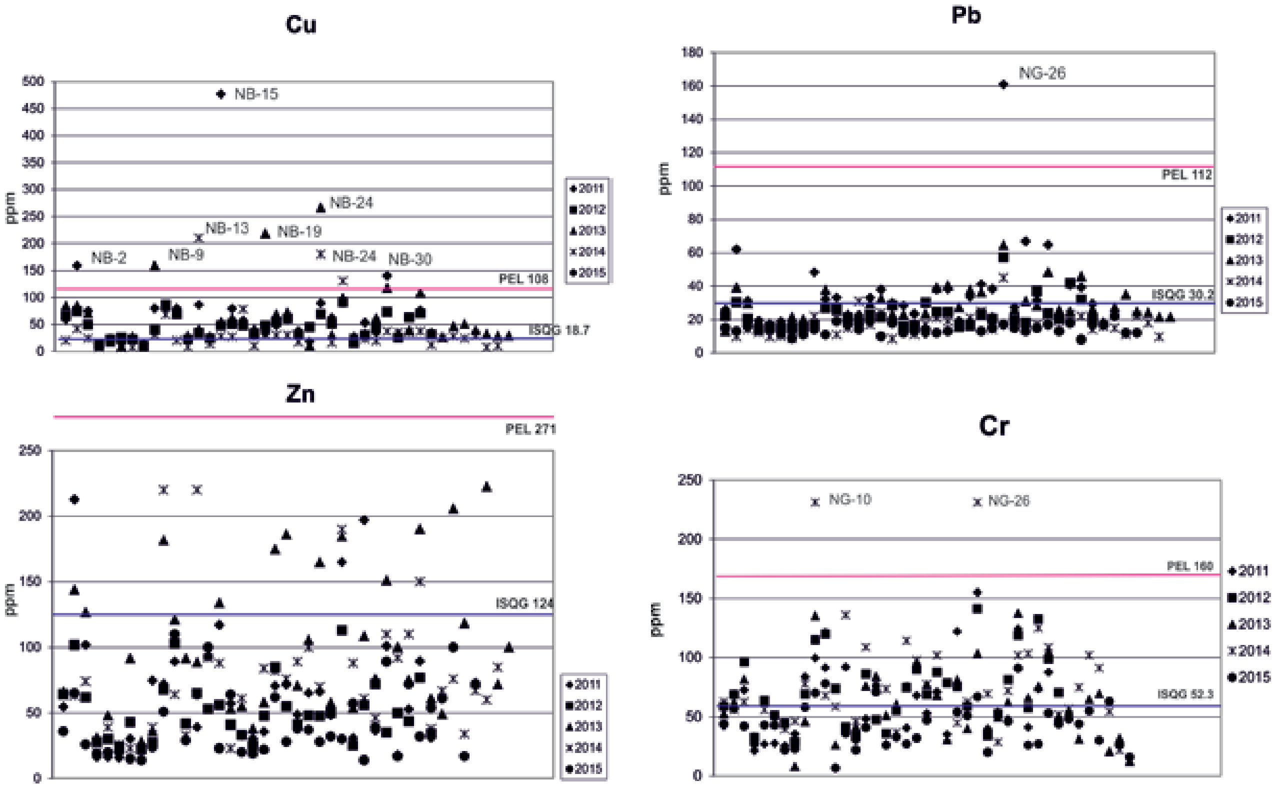

Fig. 5 Concentration of copper, lead, zinc and chromium (ppm) in Neva Bay benthic sediments shown with sediment quality guidelines (horizontal lines; ISQG = interim sediment quality guideline and PEL = probable effect level; CCME, 2002). Fig. 1 gives locations of coring sites. Compiled by D. Ryabchuk, 2017

In 2012, average concentrations of Co, Ni, Pb, Zn and Cr fell below most Swedish EPA (2000) contamination thresholds (Class 1 for all sediment types, Classes 1 or 2 for silty clay-rich mud from the local sedimentation basin in the western part of Neva Bay) (Fig.6, Tables 3 and 4). Concentrations of Co met the "slight" level in four samples (out of 35 total) and the "significant" level in three samples (site 12-NB-10, western sedimentation basin). Ni concentrations reached "slight" contamination levels in three samples, "significant" levels in four samples and "large" levels in two sample (12-NB-20 and 12-NB-11, southeastern part of Neva Bay). Concentrations of Pb reached "slight" levels in 3 samples and "significant" levels in one sample (12-NB-26 from the western sedimentation basin). For Zn concentrations, four samples reached "slight" contamination levels. For Cr concentrations, five samples met "slight" contamination levels and five samples met the "significant" levels. Benthic sediments exhibited relatively high Cu and Cd concentrations in 2012. Average Cu concentrations in all type of sediments (30.9 ppm) categorized as "significant" contamination (Class 3) while just two of 35 samples showed little or no Cu contamination (Class 1). Meanwhile 8 samples exhibited "slight" levels of contamination, 15 showed "significant" levels and 10 showed "large" levels. Of the two samples analyzed by ICP-MS for Cd, one met "large" contamination levels. Thus, 11 out of 35 samples gave Class 4 levels of contamination for at least one heavy metal.

Benthic sediment samples analyzed in 2013 and 2014 gave similar results wherein average concentrations for all heavy metals except Cu fell below Class 1 contamination levels. In 2013, even maximum Cr and Co concentrations (in three and one samples respectively) did not exceed "significant" levels (Fig. 6, Tables 3 and 4). Concentrations of Ni reached "slight" levels in eight samples (out of 41 total) and "significant" levels in one sample. Twenty-nine samples showed little or no Pb contamination while ten samples met "slight" and two met "significant" contamination levels. Concentrations of Zn reached "slight" levels in ten samples, "significant" levels in ten samples and "large" levels in two samples. Average Cu concentrations reached "significant" levels for 8 samples, exceeded "large" levels for 12 samples and "very large" levels for three samples (13-NB-9, 13-NB-19 and 13-NB-24). 14 samples of 41 analyzed in 2013 gave heavy metal concentrations that reached "large" levels and three samples met the "very large" contamination level.

Neva Bay benthic sediments analyzed in 2014 exhibited relatively low average concentrations for all heavy metals except Cu and Cd. A total of 39 samples exhibited little or no contamination (Class 1) (Fig. 6, Tables 3 and 4). Average Ni, Zn and Cr concentrations

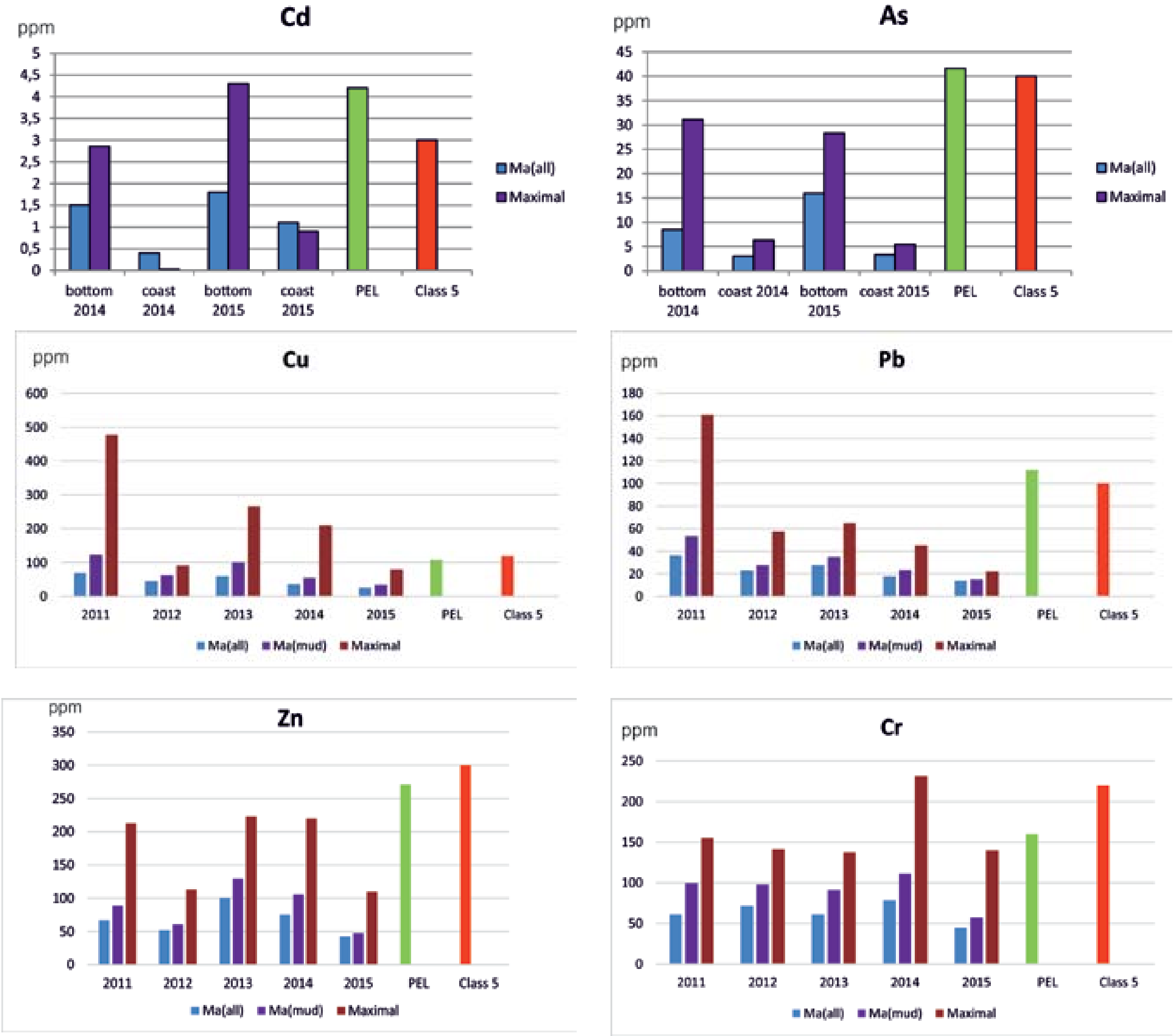

Fig. 6 **A.** Mean (Ma) and maximum Cd and As concentrations (ppm) in Neva Bay benthic and backshore (coastal/beach) sediments shown with bars representing sediment quality contamination levels (PEL= probable effect level, CCME, 2002; Class 5 = "very large" contamination, Swedish EPA, 2000); **B.** Progression of mean and maximum heavy metal concentrations measured in Neva Bay benthic sediments from 2011–2014 shown with contamination levels (PEL= probable effect level, CCME, 2002; Class 5 = "very large" contamination, Swedish EPA, 2000). Ma(all) – arithmetic mean of concentrations across all grain-size fractions; Ma (mud) – arithmetic mean of concentrations in soft muddy sediments from areas of active accumulation. Compiled by D. Ryabchuk, 2017

in silty clay-rich mud from the western sedimentation basin reached "slight" levels of contamination (Class 2). Concentrations of Co and Pb reached "slight" levels of contamination in six and three samples, respectively. In terms of Cr concentrations, 27 samples exhibited little or no contamination, 8 samples exhibited "slight" contamination, three samples met "significant" level, and one sample from the western sedimentation basin (14-NB-10) reached "very large" levels (231 ppm) of contamination. In terms of Zn concentrations, two samples (14-NB-10 and 14-NB-13) met the 220 ppm concentration for "large" levels of contamination, two samples reached "significant" levels of contamination and nine samples categorized as slightly contaminated (Class 2). Similar to previous years, benthic sediments from 2014 generally exhibited relatively high concentrations of Cu. Only 11 out of 39 samples gave Cu concentrations that categorized as having little or no contamination (Class 1). Fifteen samples exhibited "slight" levels of contamination, nine samples exhibited "significant" levels, two samples exhibited "large" levels and three samples (14-NB-13, 14-NB-24 and 14-NB-26) met the 210 ppm "very large" level of contamination. Out of 39 samples analyzed from 2014, three showed "large" levels of contamination for at least one heavy metal and three samples met "very large" contamination criteria.

In 2015, average concentrations of Co, Ni, Pb, Zn and Cr still were below most Swedish EPA (2000) contamination thresholds (Class 1 both for all sediment types and silty clay-rich mud) (Fig. 6, Tables 3 and 4). Co and Pb concentrations did not exceed "little or none" contamination level in all samples. Out of 39 samples, one gave Cr concentrations that classified as slightly contaminated (Class 2). Ni concentrations reached "slight" levels of contamination in two samples. Zn concentrations reached "slight" levels of contamination in four samples. In terms of Cu concentrations, 12 samples met "slight" contamination criteria, 15 samples met "significant" contamination criteria, and one sample exceeds "large" contamination criteria.

Annual monitoring of heavy metal concentrations in Neva Bay benthic sediments showed that in 2015 the average concentrations for all metals except Cu and Cd were slightly higher than corresponding values from 2000–2004, but lower than corresponding values measured in sediment from cores spanning the time period from the 1950's to 1990's. Contamination levels have also fallen over the last three years following a 2006–2008 contamination event. Measurements carried out for benthic sediments sampled in 2011–2015 demonstrate the trend of decrease of all heavy metal concentrations (both average and maximal) with exception of Cu and Cd (Fig. 6, Tables 3 and 4).

Geochemical data from backshore (coastal/beach) sediment around Neva Bay can record information concerning sources of heavy metal contamination (Figs 7, 8, Table 5). Backshore (coastal/beach) sediment samples typically exhibited low average Co, Ni, Zn and Cr concentrations (little or no contamination) and "significant" Cu and Pb contamination for the time period from 2011 to 2014. Maximum concentrations however reached the "very large" contamination level (Class 5) and in several instances exceeded PELs for all heavy metals except Co in every year of the study. This pattern indicates coastal contaminant sources have an intense but limited spatial reach. Some samples collected near industrial and waste disposal sites in the easternmost part of Neva Bay showed contamination levels of up to 10 times the "very large" levels (Vasylievsky Island, Kanonersky Island, Krasnenkaya River mouth) (Fig.8).

DISCUSSION

In the eastern part of Neva Bay, the Neva River discharge exerts primary control on deposition. The total annual bedload volume transported by the Neva River reaches 65,000 tons, while suspended load volume reaches about 510,000 tons. Most of this load settles out in Neva Bay (Raukas, Hyvärinen 1992).

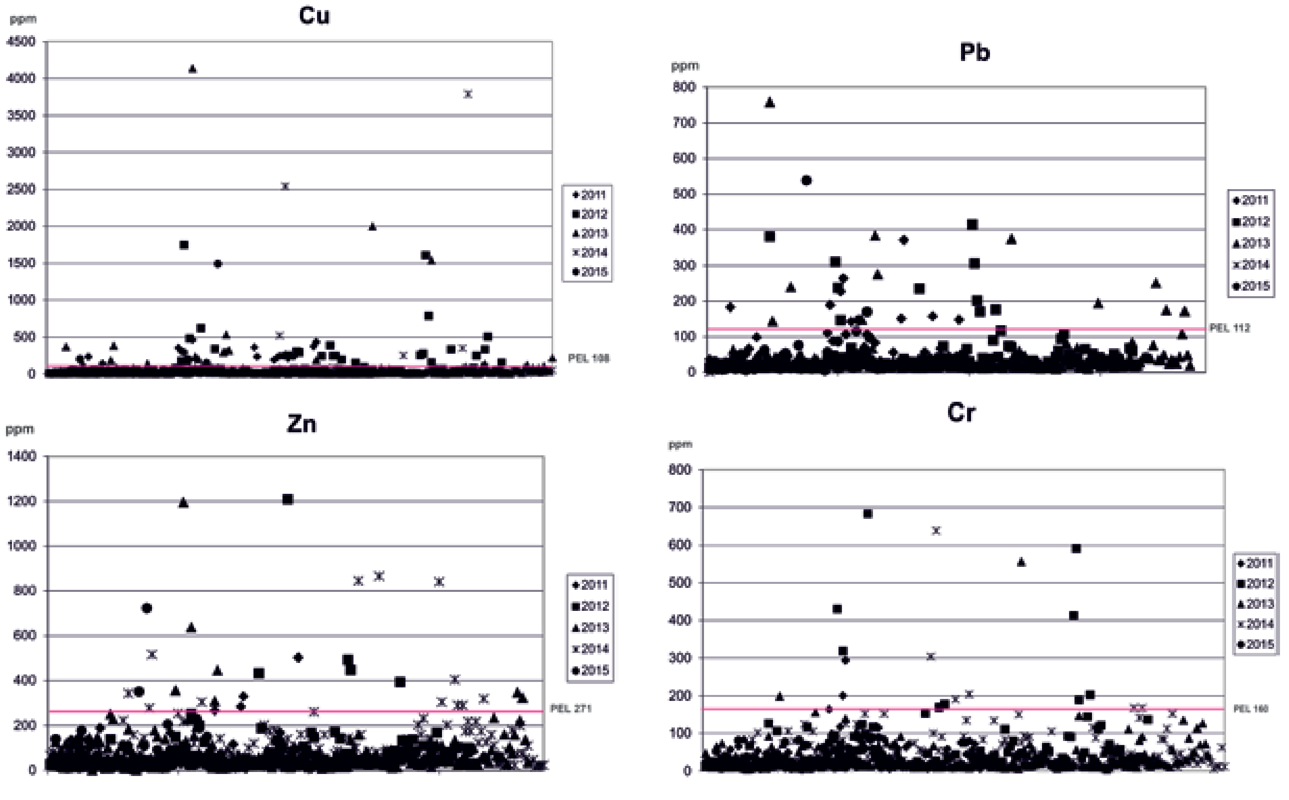

Fig. 7 Concentrations of copper, lead, zinc and chromium (ppm) in backshore (coastal/beach) sediments around Neva Bay with probable sediment contamination level indicated (horizontal line; PEL= probable effect level, CCME, 2002). Compiled by D. Ryabchuk, 2017

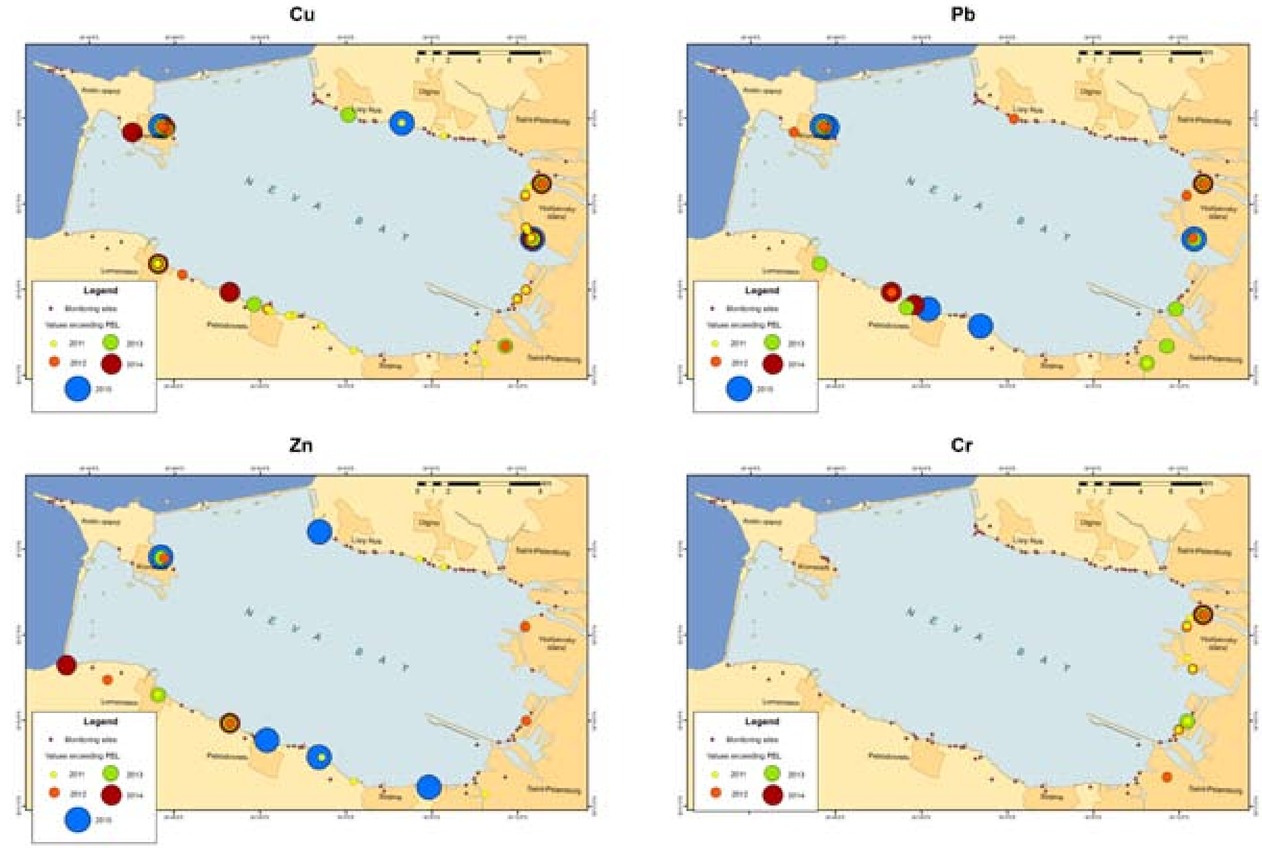

Fig. 8 Location of monitoring sites with heavy metal concentrations, exceeding PEL (PEL= probable effect level). Compiled by N. Deryugina, 2017

Fine- to very fine sands, silty-sands and silts are deposited from east to west according to decreasing current velocities. Silty clay-rich mud accumulates in the center of the western part of the bay within depressions where water depths exceed 4 m. The natural sedimentation rate is about 0.5 mm/year (Spiridonov *et al.* 2004). Accordingly, most of the Neva Bay benthic surface consists of sands and silty sands (Fig. 1). Due to the predominance of silt (0.05-0.005 mm) in the Neva River sediment load, Neva Bay includes more silty benthic sediments relative to other areas in the eastern Gulf of Finland. The high rates of modern-day mud accumulation in Neva Bay represent a significant environmental impact.

Analytical data and archival materials thus consistently record changes in mud accumulation patterns in Neva Bay over the last two centuries. Data from 18th century hydrographic surveys (found in the Russian Navy State Archive) indicate that areas of modern mud accumulation in the western part of the bay previously consisted of uniform sand deposits. All maps from 1830 to 1911 show sandy deposits in western areas of the bay. The 1920-1924 scientific expedition by Professor Deryugin detected silty clay-rich mud in several sampling sites in the central part of Neva Bay. Professor Deryugin's report assumed that previous maps and sediment descriptions had misreported, but comparison of his data with surveys conducted in the 1990's indicates significant expansion of areas of accumulating mud (Fig.9) (Spiridonov *et al.* 2008). Recent monitoring has demonstrated that these areas continue to expand.

Together with the present Neva River discharge, erosion of the late-glacial and lake sediments represents the primary natural source of fine-grained sediment in the eastern Gulf of Finland (Atlas … 2010). Since the end of 19th century, dredging became the other source of silty-clayey particles. By the late 1980's and early 1990's, modification of Neva Bay (e.g. hydraulic infilling of areas) increased suspended load concentrations. Suspended loads in Neva Bay surface waters during active dredging phases reached 200 mg/l, exceeding natural/background levels by an order of magnitude. The FPF, whose construction began in the 1970's, also activated silty-clay accumulation processes. This massive hydro-engineering project was halted in 1993 at which point suspended sediment levels declined (becoming 3–4 times less than values measured in 1998). Technogenic modification of Neva Bay bathymetry includes submarine sand excavation in the northeastern part of the bay (near Lakhta), which formed a series of relatively deep depressions. These depressions currently serve as sediment repositories for dredging activities and

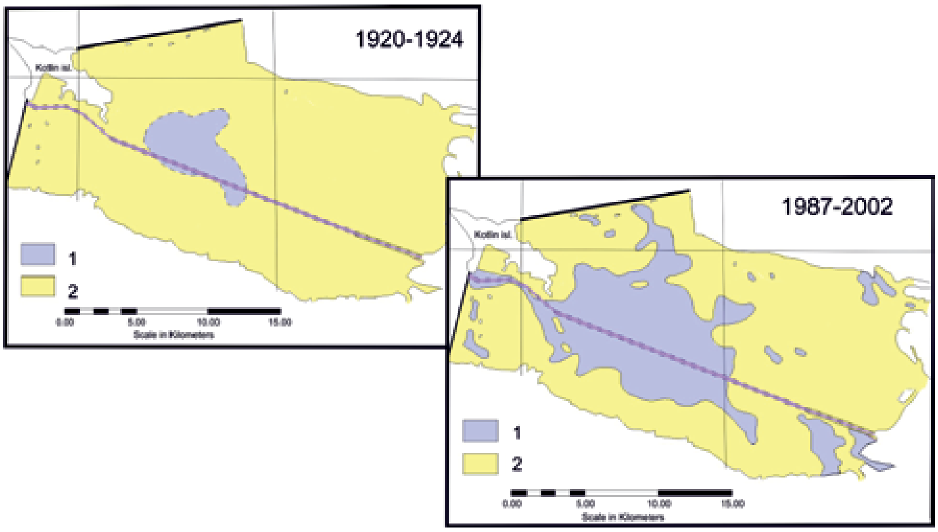

Fig. 9 Expansion of silty-clay mud accumulation area during 20th century based on comparison of K. Derugin's expedition (1923) and results of geological survey (1989–2003). 1 – silty-clayey mud; 2 – other sediment types. Compiled by D. Ryabchuk and E. Nesterova, 2004

thus assume abnormally high artificial sedimentation rates of up to 1–3 cm per year (Ryabchuk *et al.* 2017). Annual sedimentological studies of Neva Bay benthic sediments carried out over the last decade have demonstrated the role of anthropogenic influence on sedimentation and resultant overall modification of sedimentary cover. Sediment sampling of near-shore benthic sediments around the northern coast from 2002 to 2008 revealed a pronounced increase in time of clayey silty mud sediments. There a clay-rich layer of up to 3 cm thickness develops atop the formerly sandy surfaces at water depths of 2–3 m (Ryabchuk *et al.* 2017). Grain-size analyses of the benthic sediment sampling sites from area of silty-clay mud accumulation (sites NG-9, 10, 25, 26 and 30) similarly show up to 15-20% increases in proportions of fine grained particles (<0.01 mm). Monitoring studies from 2011–2013 indicate that sedimentation is gradually reverting back to previous dynamics, however (Fig. 10).

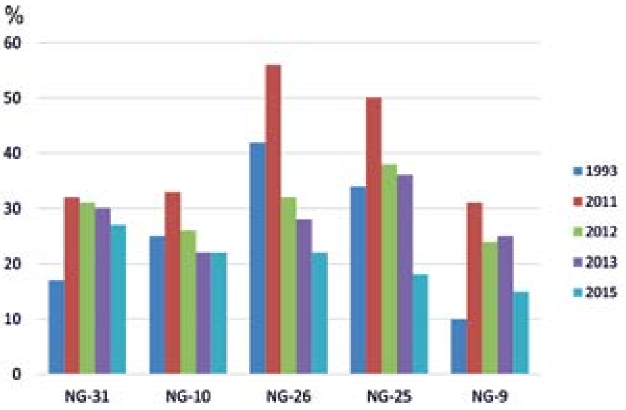

Fig. 10 Relative abundance of <0.01 mm grain-size fraction in the upper 1 cm of Neva Bay benthic sediments (1993–2015). Compiled by D. Ryabchuk, 2015

The sedimentation rate and sediment contamination studies have shown that the entire 40–50 cm deposit of silty-clay formed over the last 100–150 years. The timing of these changes along with the history of regional development indicates anthropogenic causes. According to average sedimentation rates, temporal trends show that metal accumulation began rather abruptly in the first half of the last century. The composition and timing of this contamination implicates the local base metal industry as a source. Zinc, lead and copper were the first metals to reach concentrations qualifying them as major contaminants. The pronounced increase in sedimentary Cd concentrations a decade or two later indicates intensification of activities related to the chemical industry (Fig. 3). The highest concentrations occur in the upper halves of the cores and probably span a time frame from the 1950's to the late 20th century. Sediment core material representing the last 15 years record significant decreases in heavy metal concentrations. Concentrations of all metals decreased significantly from 1995 to 2005.

Decreasing anthropogenic loads in the 1990's partially account for limited declines in heavy metal concentrations. Major efforts by the VODOKANAL State Enterprise in improving St. Petersburg water treatment along with pronounced reductions in phosphorus and nitrogen input to Neva Bay (http://www.vodokanal.spb.ru/en) also account for continuing declines in benthic sediment load.

Major marine infrastructure projects constructed from 2006–2008 also influenced Neva Bay's sedimentological regime. Dredging and dumping processes significantly increased the volume of water column suspended matter by 2007 and traces of the sediment reached Vyborg Bay (Fig. 11). The sus-

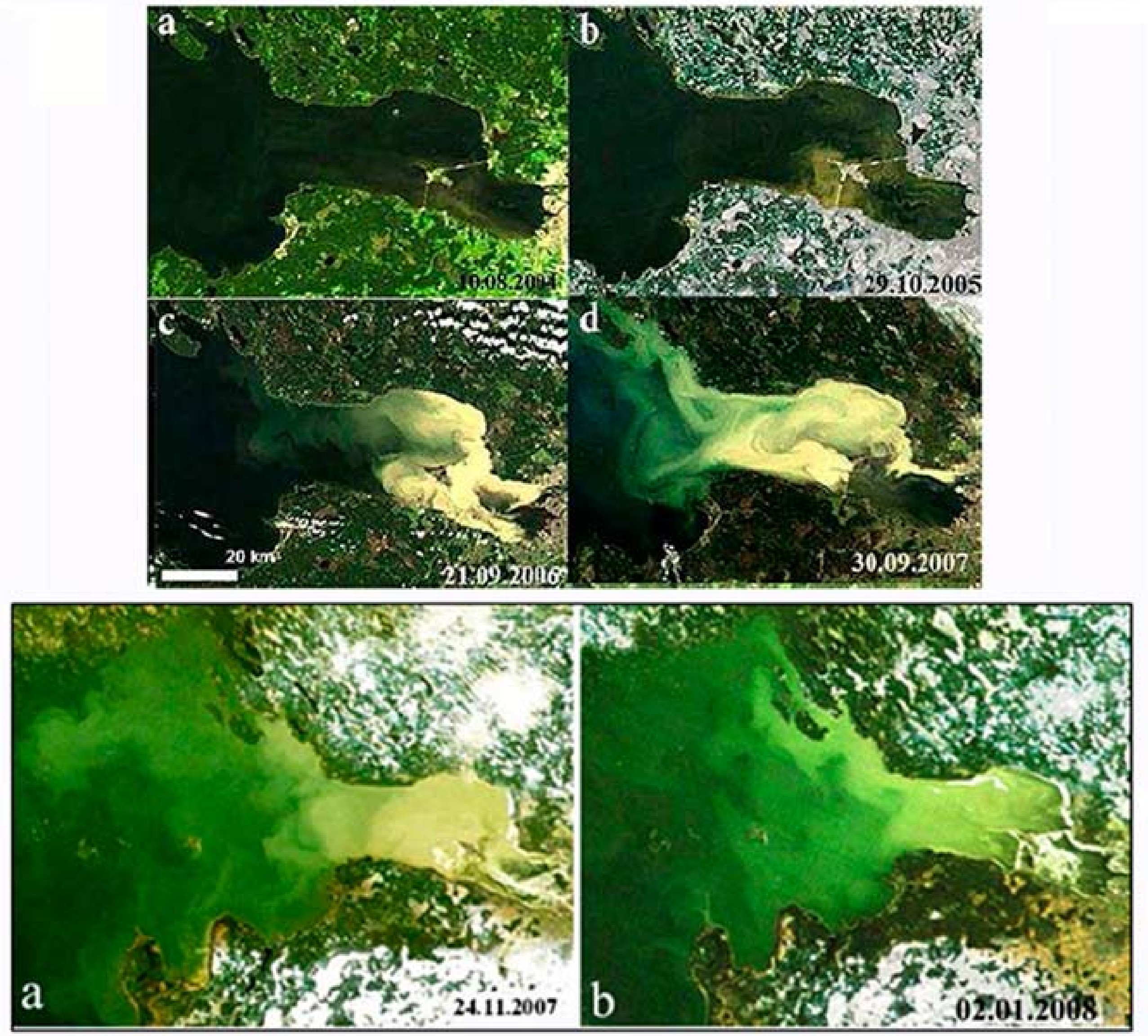

Fig. 11 A. MODIS images of hydro-optical heterogeneities in the area study during implementation of the Sea Façade Project, chosen examples differ by intensity of hydro-technical activity, by cites of underwater ground disposals in the Neva Bay and by hydro-meteorological conditions: a – from 10.08.2004, before dredging has started; b – from 29.10.2005, initial period of hydro-technical activity; c, d from 21.09.2006 and from 30.07.2007, active period of dredging and dumping operations, characterized by very high levels of water contamination by fine suspended sediments. B. MODIS images, winter aspects: a – from 24.11.2007 distribution of suspended sediments after dredging was interrupted; b – from 02.01.2008, in spite of 6 weeks passed, a sufficient amount of SS can be seen. Compiled by L.Sukchacheva, 2017.

pended load then deposited as a silty-clay layer over the natural sand surface at water depths of 2–3 m. Annual geochemical monitoring from 2005–2015 revealed a contamination pulse of 2007–2008 in areas of silty clay-rich mud.

Coupled with changing hydro-optical and chemical conditions of the lower water column, suspended sediment loads have likely contributed to diminished eukaryotic plankton abundance in favor of cyanobacterial biomass, and pronounced shifts in benthic communities (e.g. decline of large *Unionidae* molluscs) (Maksimov 2014). The reworking and redistribution of contaminated sediments thus imposed a second contamination event on the Neva Bay ecosystem. As suggested by Golubkov (2014), contaminants may curtail zooplankton cycles by histopathological mechanisms and other abnormalities affecting the dominant *Cladoceran* species in areas adjacent to St. Petersburg.

Results of the 2011–2015 annual geochemical monitoring revealed slight recent decreases in benthic sediment heavy metal concentrations. The average concentrations of heavy metals currently fall significantly below their respective probably effect levels (PELs). Maximum observed Cu, Pb and Cr concentrations however still exceeded PELs whereas maximum Zn, Cd and As concentrations do not. Average Cu concentrations in benthic sediments reach “significant” contamination and average Cd concentrations meet “large” contamination levels according to Swedish EPA (2000) guidelines.

Received results confirm the conclusion of recent improvment of the GOF environment due to the weakened impact of municipal and industrial discharges (Raateoja, Setälä 2016). The 2011–2015 analysis of backshore (coastal/beach) Neva Bay sediments meanwhile detected numerous sources of significant heavy metal contamination from industrial and waste disposal activities around the easternmost part of Neva Bay (Vasylievsky Island, Kanonersky Island, Krasnenkaya River mouth).

CONCLUSIONS

Coastal sedimentation basins around Neva Bay record a unique history of pollution. Most heavy metals analyzed (e.g. Cd, Zn, Pb, Cu) show similar patterns of variation throughout sediment profiles. Metals began to accumulate rapidly in the first half of the 20th century. Zinc, lead and copper were the first metals to reach contaminant thresholds implicating the regional base metal industry as a source. Pronounced increase of cadmium contamination a decade or two later suggests pollution from the local chemical industry. The highest concentrations occur in upper sections of sediment cores, which likely represent a time frame spanning from the 1950’s almost to the end of the century. Heavy metal concentrations subsequently decreased significantly from 1995 to 2005.

Intensive dredging in 2007–2008 resuspended and redistributed contaminated sediment around Neva Bay causing a dramatic increase in benthic sediment heavy metal concentrations. Concentrations of all measured metals subsequently declined from 2009–2014 relative to the elevated values observed for 2007–2008.

Pollution history of Neva Bay bottom sediments is closely linked with changing of sedimentation conditions. Integrated analysis of sedimentology, geochemistry and archival material reveal dramatic shifts in Neva Bay sedimentation processes over the last three centuries. The western part of Neva Bay has transitioned from a sand-dominated system to one of mud accumulation with the aerial extent of mud deposition expanding significantly during the 20th century. Extensive dredging activity in 2006–2008 caused a dramatic increase in water column suspended load and deposited a clay layer up to 5 mm thick atop the natural sandy sediment surface of Neva Bay at water depths of 2–3 m. Investigations in 2007–2008 showed that anthropogenic processes (ship channels, submarine sand extraction, sediment dumps etc.) have completely transformed the benthic environment in the eastern reaches of Neva Bay. Previously quarried depressions have been repurposed as sedimentation traps with very high accumulation rates.

Extremely high concentrations of heavy metals however persist in backshore (coastal/beach) sediments around Neva Bay and continue to represent an anthropogenically imposed stress on the environment. Identification of off-shore pollution sources should be subject of future investigations.

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