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Essential and non-essential trace elements in fish consumed by indigenous peoples of the European Russian Arctic☆

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ABSTRACT

In present study, the analyses of essential [copper (Cu), cobalt (Co), selenium (Se) and zinc (Zn)] and non-essential elements [mercury (Hg), lead (Pb), cadmium (Cd) and arsenic (As)] in 7 fish species consumed by the indigenous people of the European Russia Arctic were conducted. The Nenets Autonomous Region, which is located in the north-eastern part of European Russia, was chosen as a Region of interest. Within it, the Nenets indigenous group ($n = 6000$) constitutes approximately 10% of the total population. Nearly all of the Nenets live a traditional life with fish caught in the local waters as a subsistence resource.

We found that northern pike contained twice the amount of Hg compared with roach, and 3–4 times more than other fish species commonly consumed in the Russian Arctic (namely, Arctic char, pink salmon, navaga, humpback whitefish and inconnu). Fish Hg concentrations were relatively low, but comparable to those reported in other investigations that illustrate a decreasing south-to-north trend in fish Hg concentrations. In the current study, northern pike is the only species for which Hg bio-accumulated significantly. In all fish species, both Cd and Pb were present in considerably lower concentrations than Hg. The total As concentrations observed are similar to those previously published, and it is assumed to be present primarily in non-toxic organic forms. All fish tissues were rich in the essential elements Se, Cu and Zn and, dependent on the amount fish consumed, may contribute significantly to the nutritional intake by indigenous Arctic peoples. We observed large significant differences in the molar Se/Hg ratios, which ranged from 2.3 for northern pike to 71.1 for pink salmon. Values of the latter <1 may increase the toxic potential of Hg, while those >1 appear to enhance the protection against Hg toxicity.

1. Introduction

A traditional food system comprises food items that are available to communities from local natural resources. The harvested wild-life foods consumed by indigenous populations living in Arctic regions are an example. Unfortunately such foods may be contaminated to an extent above levels acceptable for human consumption (Kuhnlein and Chan, 2000). This is well documented in several studies in which persistent organic pollutants (POPs) and mercury (Hg) have been considered in human health risk assessments (Dudarev et al., 2013; Kuhnlein and Chan, 2000; Odland et al., 2016). Among persistent inorganic pollutants (PIPs), Hg is of special concern as it constitutes a mobile toxic element that bio-magnifies and bio-accumulates in nature.

Atmospheric Hg is transported to the Arctic as a result of worldwide human activities, as well as the occurrence of natural processes. The Hg depositional processes involved are complex and poorly understood, and are considered to be particularly sensitive to climate change. Current scientific information, however, is not

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capable of clearly answering whether this involves increase or decrease of Hg in wild-life food items in the Arctic (Sundseth et al., 2015).

Even though attention has been given to Hg dynamics in the western Arctic (Europe and North-America), limited information is available from the Russian Arctic. A recent study has shown low and declining Hg concentrations in Arctic Russian rivers and fish, although the study suggests that there is a significant amount of heterogeneity in geographical Hg concentrations across the Russian Arctic (Castello et al., 2014). Though an estimation of the Hg intake from the consumption of fish and seafood in Russia indicates that it cannot be a source of high Hg intake by the general Russian population (Lyapunov et al., 2016), there is little information about this for foods consumed by Russian indigenous populations (AMAP, 2011; Dudarev et al., 2013; Odland et al., 2016). Other issues related to food security in the Russian Arctic have been identified and include: a lack of almost all types of vitamins and mineral nutrients [e.g. calcium, magnesium, zinc (Zn) and selenium (Se)] and knowledge about the presence of PIPs other than Hg (Dudarev, 2012; Dudarev et al., 2013).

Predatory and long-lived species of fish are considered ideal for the biomonitoring of contaminants in the environment because environmental pollutants accumulate in both water bodies and prey. However, the accessibility of fish species that are part of the diet varies across the Russian continent. Higher Hg concentrations are typically found in predatory species that feed at higher trophic levels, such as lake trout (*Salvelinus namaycush*), northern pike (*Exos lucius*), and Arctic char (*Salvelinus alpinus*). Fish populations in the Arctic are often older (due to low fishing pressure) and have slower growth rates compared with populations at lower latitudes, and both factors contribute to enhanced Hg bioaccumulation (Lockhart et al., 2005). Mercury concentrations in fish, as methyl mercury (MeHg), tend to be enhanced in smaller lakes because of higher summer epilimnion temperatures that favour higher Hg methylation rates in lakes, as well as higher concentrations of both inorganic Hg and MeHg in the waters entering the lakes from the watershed (Evans et al., 2005). This emphasizes the need of a fish species sampling strategy of the typical available fish species consumed by the different indigenous communities located across Russian Arctic. To address this objective, a long-term study was initiated to monitor changes in the amounts of POPs and PIPs in the diets of pan-Russian Arctic indigenous populations. As part of this project, an analytical biomonitoring laboratory was established at the Northern (Arctic) Federal University (NArFU) in Arkhangelsk, Russia. NArFU complies with The Russian System for Accreditation of Analytical Laboratories (Karpov et al., 2008) and fulfils the requirements of the ISO/IEC 17025 Accreditation. A Russian governmental mega grant was received to conduct this research.

The findings of the current study of trace elements in fish consumed by indigenous peoples in the Nenets autonomous region constitute a first report. The Nenets region is located in the north-eastern part of European Russia and is bounded by the Kara, the Barents and the White seas. The indigenous group ($n = 6000$) makes up approximately 10% of the total population of the Nenets Autonomous Region. In fact, most of those are Nenets (Federal State Statistics Service. Retrieved June 29, 2012). Nearly all of the Nenets live a traditional life with fish caught in the local waters (sea shore, lakes and rivers) as a subsistence resource. The consumption rate and the preferred fish species consumed vary dependent on the location and individual preferences. Being one of the main food items fish is an important source of essential elements [e.g. copper (Cu), cobalt (Co), Se and Zn], as well as of other PIPs such as Hg, lead (Pb), cadmium (Cd) and arsenic (As) (Dudarev, 2012; Rylander et al., 2011).

In the current study, the most common fish species consumed by Nenets living in three villages located away from the regional capital Naryan-Mar were identified employing a nutritional questionnaire and collected for analyses. The main objective of the research described was to identify fish species suitable for inclusion in the mentioned pan-Russian Arctic program for monitoring the nutritional intake of POPs and PIPs among the different Arctic indigenous populations. In the current study, we report the concentrations of selected toxic and essential elements measured in 7 fish species that are part of the Nenets nutritional intake.

2. Materials and methods

2.1. Sampling strategy

The field sampling of fish species was initiated in the territory of the Nenets autonomous region by an agreement between NArFU and the regional government. A food frequency questionnaire was employed to identify the most commonly consumed fish species by the local population. Based on the responses from 150 individuals, Arctic char, pink salmon, navaga, humpback whitefish, northern pike, roach and inconnu fish species were selected for chemical analysis. These species constituted approximately 70% of total fish consumption by the respondents.

Three Nenets villages were chosen for our research and included Krasnoe, Nelmin-Nos and Indiga. The former two were selected because they were included in the 2004 AMAP report on Persistent Toxic Substances, Food Security and Indigenous Peoples of the Russian North (Arctic Monitoring and Assessment Programme, 2004). Since then, no significant research has been carried out in this area. The village of Indiga was chosen because its location is more remote from Naryan-Mar and is closer to the Barents Sea. The main economic activity of the population is reindeer husbandry. The village of Krasnoe is one of four settlements that are part of the municipality of “Primorsko-Kuiskiy Selsovet”. It is located 40 km north of Naryan-Mar downstream on the Pechora river and in 2017 had a population of 1875 in 2017 of whom 869 identified themselves as Nenets and 175 as Komi (Passport of the Municipality Primorsko-Kuiskiy Selsovet, 2017).

Nelmin-Nos is located 60 km downstream from Naryan-Mar. Its population was 1002 in 2017 of whom 681 identified themselves as Nenets (Passport of the Municipality Malozemelskiy Selsovet, 2017). The village of Indiga is situated in the Northwestern region of the Nenets Autonomous Okrug, 280 km west of Naryan-Mar. Its population was 632 in 2018, and 451 identify themselves as Nenets and 44 as Komi (Passport of the Municipality Timan Selsovet, 2018). The locations of the sampling sites are indicated in Fig. 1.

2.2. Sample collection and preparation

Out of fifteen different fish species collected, seven of the most consumed in the study villages were chosen for analyses (see Table 1). The fish were bought from local fishermen from May 2017 to July 2018. The whole fish were wrapped in a food polyethylene film and were then placed in a Zip-lock sealed polyethylene bag. They were then frozen immediately at a temperature of −20 °C before transportation to the Arctic Biomonitoring Laboratory at the Northern (Arctic) Federal University in Arkhangelsk in medical thermo-containers. Upon arrival at the laboratory, the fish were thawed, weighed and their age determined. The muscle tissue was separated from the carcass and homogenized with an ordinary household blender RHB-CB2932 (Redmond Industrial Group LLC, Albany, New York, USA) and then weighed on an analytical balance AUW220 (Shimadzu Corporation, Kyoto, Japan). Subsequently, the homogenized fillet was transferred to plastic containers and frozen at a temperature of −80 °C. Prior to analysis, the muscle was

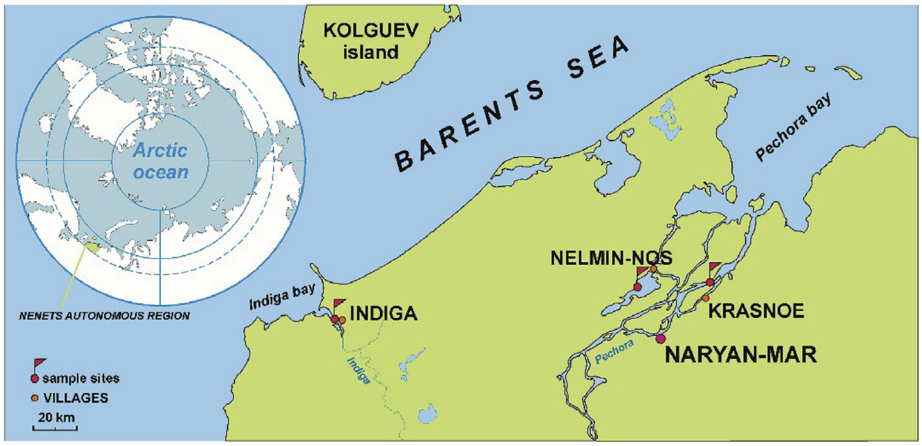

Fig. 1. Map of villages in the Nenets Autonomous Okrug where research was conducted.

Table 1
The fish species selected for chemical analyses and water habitats caught.

Settlement	Water habitat	Fish species
Krasnoe	Pechora River	**Inconnu** *(Stenodus nelma)* **Northern pike** (*Esox lucius)*
Nelmin-Nos	“Golodnaya Guba” of Pechora River	**Roach** *(Rutilus rutilus)*
Indiga	Indiga River	**Arctic char** *(Salvelinus alpinus)* **Navaga** *(Eleginus nawaga)* **Pink salmon** *(Oncorhynchus gorbuscha)* **Roach** *(Rutilus rutilus)* **Humpback whitefish** *(Coregonus pidschian)*

thawed and lyophilized using a FreeZone 2.5 L Benchtop Freeze Dry System (Labconco Corporation, Kansas City, MO, USA).

2.3. Age determination

Fish age was determined by measuring the growth patterns in the otoliths (ear stones) located behind the brain in bony fish and involved counting the number of opaque zones (annuli) from the primordium to the otolith edge. Otoliths from each fish were removed, cleaned and dried before sectioning. The latter involved a modified high-speed gem-cutting saw with a 250 µm thick diamond impregnated blade. Sections from each otolith were mounted on clear glass microscope slides under glass coverslips using resin. Annual increments were then counted on the ventral side of the section from the primordium to the otolith edge adjacent to the sulcus (Howland et al., 2004).

2.4. Elemental measurements

The concentrations of 9 elements, namely Hg, Cd, Pb, As, Co, nickel (Ni), Cu, Se and Zn, were measured in the muscle tissue as this is the only part of the fish which is consumed. Portions of 0.25 g of lyophilized muscle were weighed with an accuracy of ± 0.001 g into 50 ml 115 × 28mm polypropylene tubes (PP) (Sarstedt AG & Co. KG, Nümbrecht, Germany) to which was added 5 ml of in-house double sub-distilled nitric acid (HNO_3) (puriss. p.a., Merck, Darmstadt, Germany) using two DST-1000 (Savillex, Eden Prairie, MN, USA) acid purification systems. Samples were left for one hour at room temperature and then gently heated in an ED36s digestion block (LabTech S.R.L, Sorisole, Italy) to 105 °C using a temperature gradient of 0.5 °C/min. The digestion tubes were held at 105 °C for 2 h. After cooling, 100 µl of an internal standard solution containing 1.25 µg/ml of scandium (Sc), yttrium (Y), indium (In), terbium (Tb) and bismuth (Bi) was added (Bruker Daltonics, Fremont, CA, USA). In addition, 100 µl of a 2.5 µg/ml ^{74}Se enriched stable isotope solution (99.9% of ^{74}Se from STB Isotope Germany GmbH, Hamburg, Germany) was added before dilution to 25 ml with ultrapure water. The quantitation of the elements was performed with an Aurora Elite (Bruker Daltonik GmbH, Bremen, Germany) inductively coupled plasma mass spectrometer (ICP-MS), equipped with a collision reaction interface (CRI) for reducing isobaric mass interferences, a concentric glass nebulizer and a Peltier cooled Scott double-pass spray chamber. The following isotopes with internal standards in brackets; ^{59}Co (^{45}Sc), ^{60}Ni (^{89}Y), ^{65}Cu (^{89}Y), ^{66}Zn (^{89}Y), ^{114}Cd (^{115}In), ^{202}Hg (^{209}Bi), ^{206,207,208}Pb (^{209}Bi) were measured in the no gas mode. To reduce polyatomic interferences for the ^{75}As (^{74}Se) and ^{78}Se (^{74}Se) isotopes the CRI was used with a hydrogen gas flow rate of 115 ml/min in the skimmer cone. Gas flow was optimized to achieve maximum signal for the isotope ratios ^{74}Se/^{75}As and ^{74}Se/^{78}Se when a solution containing 10 µg/l of ^{74}Se in HCl (2% v/v) was measured. The instrument was set up to determine Cd by use of the $^{114}Cd^{+}$ ion with automatic mass correction for $^{114}Sn^{+}$ ionic interference. Although the molybdenum (Mo) concentration in fish tissue is low, any interference from the $^{98}Mo^{16}O^{+}$ ion was considered not to contribute significantly to the overall signal at mass 114. Nitric acid matrix matched calibration solutions were prepared daily in a PP volumetric flask from certified primary stock solutions of the individual elements (Spectrapure Standards AS, Oslo, Norway). The moisture content of each sample was measured during the freeze-drying step for recalculation of individual moisture content in order to present elemental concentrations as mg/kg or µg/kg wet weight (ww).

2.5. Quality assurance

Routine acid leaching of vessels used in the preparations of all solutions, use of clean sample PP tubes and use of ultrapure water and sub-distilled HNO_3 assured that the concentrations of the elements of interest in blank samples were as low as possible in order to obtain adequate limits of quantifications [LOQ was taken as 10 times the standard deviation of 5 laboratory blank solutions]. The LOQs were established daily using reagent blanks; typical LOQ values in μg/kg for each element were Hg: 1, Cd: 0.03, Pb: 0.3, As: 35, Co: 1.0, Ni: 1.1, Cu: 4.0, Se: 18 and Zn: 20. The accuracy of the measurements was established using relevant certified reference materials: ERM-BB422 fish muscle (European Commission-Joint Research Centre, Institute for Research Materials and Measurements, Geel, Belgium), IAEA-436 tuna fish flesh homogenate (International Atomic Energy Agency, Vienna, Austria) and DOLT-5 dogfish liver (National Research Council Canada, Ottawa). For method development and the daily quality assurance of the measurements commercially available, lyophilized fish powder produced from white fish caught in the North-Atlantic Ocean was used (Seagarden, Nestun, Norway). The day-to-day coefficients of variation for this in-house quality control material (analyzed with each batch of samples) were between 3.4 and 11.9% for the measured elements. The recoveries for the different elements in the reference materials ranged from 86 to 110% of the certified values (see Table 2).

2.6. Statistical analysis

The distribution of the variables was visually assessed and the skewness was calculated. Variables were log-transformed when skewness exceeded 2.0. The geometric means (GM) are presented for these variables, while arithmetic means (AM) are otherwise reported. Analysis of variance (ANOVA) was used to assess differences between several groups simultaneously, and least square differences were calculated to assess which groups that differed from each other. Differences between two groups were assessed using Students T-test. Univariate associations were assessed using least square regression analysis, yielding the Pearson's correlation coefficient (r) as the measure of association. Two-tailed p-values <0.05 were considered to be of statistical significance. The statistical package SPSS®, version 25.0 (IBM Corp., Armunk, NY, USA), was used for the statistical calculations.

3. Results and discussion

To our knowledge this is the first study addressing the quantity of toxic and essential elements in fish species most commonly consumed by the Nenets population in the European Russian Arctic. The raw data, as well as details about concentrations of selected elements measured in the muscle tissues of several other fish species and their average consumption rates, are provided in a supplementary article (Sobolev et al., 2019). Anadromous salmonid fish are represented by Arctic char, pink salmon, humpback whitefish, and inconnu and freshwater fish by roach and northern pike. One marine amphidromous cod species, namely navaga is also included in this study. Concentrations of essential and toxic elements are expressed as μg/kg or mg/kg wet weight basis. Fish ages and weights are summarized in Table 3. The average ages of the freshwater fish were quite similar, ranging from 10.5 years for roach caught around the village of Indiga to 7.0 for northern pike and inconnu. The migrating salmonids, Arctic char and pink salmon were significantly younger (4.5 and 1 years) but their weights reflect a faster growth. The lowest fish weights were evident among the marine navaga. The intra-age variability among the different species was larger than for the inter-species age, while the weights were species dependent. The assessed ages and weights of the fish are typical for those consumed in the villages of Indiga, Krasnoe and Nelmin-Nos.

3.1. Non-essential elements

Fish have slower growth rates and longer lifetime cycles in the Arctic, which may cause a higher bioaccumulation of contaminants compared to more productive southern regions. Mercury biomagnifies and bioaccumulates mostly (>80%) as MeHg especially in predatory fish muscles, with its levels increasing from lower to upper trophic levels (Moiseenko and Gashkina, 2016). In fish it is mostly enriched in muscle as it associates with proteins (Mason et al., 1995; Polak-Juszczak, 2018). Total Hg mean concentration in fish muscle in the current study ranged from 18 to 19 in Arctic char and pink salmon to 188 μg/kg in northern pike (see Table 3). Among the collected northern pikes, Hg bioaccumulated significantly with a rate of 40 $ng^{-1}g$ per 1 kg whole fish (p-value = 0.008 and Pearson r = 0.89; see Fig. 2A), while an apparent decrease in Hg with weight among Arctic char was observed (p-value = 0.060, Pearson r = 0.58). As described by Mittelbach and Persson (2011) and Selden et al. (2018), for fish the relationship between predator and prey is complicated. Presumably, this accounts for the observed differences in the sign and magnitude of slopes for the relationship between fish Hg content and weight.

In general (see Table 3), the concentration of Hg was significantly higher in northern pike and roach compared with the salmoniform species (i.e., Arctic char, pink salmon, humpback whitefish and inconnu). The observed Hg concentrations are relatively low but comparable to those reported in other investigations that have shown a decreasing south-to-north Hg concentration

Table 2
Measured arithmetic mean concentrations (±SD) in the certified reference materials compared with certified values (dry mass basis).

Element	Certified value ERM-BB422 Fish muscle $\mu g^{-1}g$ n = 5	Found, $\mu g^{-1}g$	Recovery in %	Certified value IAEA-436 n = 5	Found, $\mu g^{-1}g$	Recovery in %	Certified value DOLT-5 Dogfish liver $\mu g^{-1}g$ n = 2	Found, $\mu g^{-1}g$	Recovery in %	In-house quality control material Whole fish powder $\mu g^{-1}g$ n = 5	Day-to-day variability of the in-house quality control material (RSD), %
As	12.7 ± 0.7	13.0 ± 0.7	102	1.98 ± 0.17	1.78 ± 0.026	90	34.6 ± 2.4	31.2	90	24.1 ± 0.6	6.6
Cd	0.0075 ± 0.0016	0.0080 ± 0.0006	107	0.052 ± 0.007	0.055 ± 0.001	105	14.5 ± 0.6	15.0	103	0.061 ± 0.003	4.6
Cu	1.67 ± 0.16	1.71 ± 0.0034	102	1.73 ± 0.19	1.55 ± 0.03	90	35.0 ± 2.4	32.3	92	1.42 ± 0.04	7.2
Hg	0.601 ± 0.030	0.658 ± 0.003	109	4.19 ± 0.36	4.24 ± 0.01	101	0.44 ± 0.18	0.43	98	0.044 ± 0.001	8.2
Se	1.33 ± 0.13	1.46 ± 0.019	110	4.63 ± 0.48	4.63 ± 0.06	100	8.3 ± 1.8	7.8	94	1.92 ± 0.03	3.4
Zn	16.0 ± 1.1	16.9 ± 0.20	106	19.0 ± 1.3	17.7 ± 0.6	93	105.3 ± 5.4	99.2	94	51.4 ± 2.2	8.3
Co				0.042 ± 0.006	0.045 ± 0.002	107	0.267 ± 0.026	0.238	89	0.083 ± 0.004	7.7
Ni				0.069 ± 0.041	0.064 ± 0.008	93				1.46 ± 0.07	10.3
Pb							0.162 ± 0.032	0.140	86	0.022 ± 0.001	11.9

Table 3
Arithmetic (AM) and geometric means and range of wet weight concentrations of elements in muscle of the different fish species.

	Arctic char (N = 11)		Pink salmon (N = 12)		Navaga (N = 10)		Humpback whitefish (N = 12)		Northern pike (N = 7)		Roach Pechora (N = 10)		Roach Indiga (N = 10)		Inconnu (N = 6)	
	AM	Min-Max	AM	Min-Max	AM	Min-Max	AM	Min-Max	AM	Min-Max	AM	Min-Max	AM	Min-Max	AM	Min-Max
Age (year)	4.5	2.5–6.5	1+	1–2	4.5	3.0–6.5	7.0	5.0–10.0	7.0	3.5–10.5	8.0	6.5–10.0	10.5	8.5–13.0	8.0	5.0–12.5
Weight (kg)	0.713	0.255 –1.045	1.069	0.805 –1.640	0.220	0.125–0.380	0.440	0.380 –0.565	2.811	0.810 –5.620	0.309	0.245 –0.350	0.312	0.260 –0.380	1.113	0.420 –2.300
Hg (μg/kg)	18	10–28	19	15–22	63	29–117	55	29–111	188	119–311	94	74–116	98	65–128	48	<LOQ-120
As[a] (μg/kg)	2590	1940–3760	792	513–1080	30000	10300-65500	753	333–2360	625	68–5380	69	50–113	86	44–131	90	<LOQ-1360
Se (μg/kg)	462	404–551	532	444–653	565	412–682	340	281–396	167	113–215	295	269–315	338	192–420	208	76–357
Cd (μg/kg)	0.5	0.2–1.8	6.6	2.8–9.8	2.1	0.9–4.1	1.6	0.4–5.1	0.6	0.1–1.2	3.3	2.3–4.2	3.5	2.3–6.3	0.1	<LOQ-0.5
Pb (μg/kg)	5.3	1.0–9.8	8.6	0.5–19	9.3	5.4–24	2.3	0.7–6.3	9.4	5.8–14	3.9	2.2–6.0	6.6	0.5–18	1.0	<LOQ-1.6
Co (μg/kg)	3.0	2.3–4.0	2.6	2.0–3.1	9.7	5.5–14	20	7.8–33	1.9	1.3–2.8	3.3	2.7–4.2	3.8	2.4–7.7	11	1.9–31
Ni (μg/kg)	21	9.4–46	17	9.9–23	25	15–35	21	14–29	20	10–40	19	15–22	23	15–32	17	9.8–23
Cu (μg/kg)	443	344–517	540	438–647	722	473–1100	198	149–254	182	130–248	296	193–455	350	224–623	337	179–431
Zn (mg/kg)	5.2	3.8–6.8	7.3	5.8–8.4	13	8.5–18	5.9	4.9–6.8	4.0	3.1–5.4	6.8	5.2–8.4	7.2	4.8–9.5	5.4	4.8–6.7

Number of fish (N), Limit of quantification (LOQ) in μg/kg: Cd: 0.03; Pb: 0.3; Hg: 1; As: 35.
[a] Geometric mean.

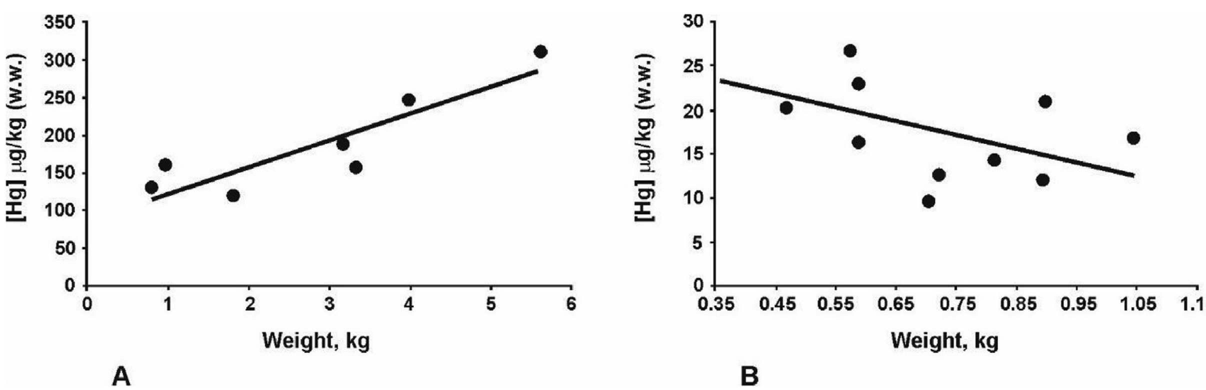

Fig. 2. Dependence of the bioaccumulation of Hg on fish weight: A northern pike, B Arctic char.

trend in fish muscle (Munthe et al., 2007). In the current study northern pike is the only species for which Hg bioaccumulated significantly. This is in accordance with an earlier study which reported that northern pike has generally higher concentrations of Hg compared with fish feeding on lower trophic levels, such as whitefish and Arctic char that feed primarily on zooplankton, salmon eggs, insects and benthos (Lockhart et al., 2005). Only for humpback whitefish was there a significant bioaccumulation of Pb with increasing age (Fig. 3, with Pearson $r = 0.62$ and p-value = 0.001). It seems pertinent to mention that inhalation of Pb fumes released during gun use when hunting with Pb shot and the subsequent consumption of meats contaminated with Pb fragments constitute major Pb exposure sources in Canadian indigenous communities (Liberda et al., 2018).

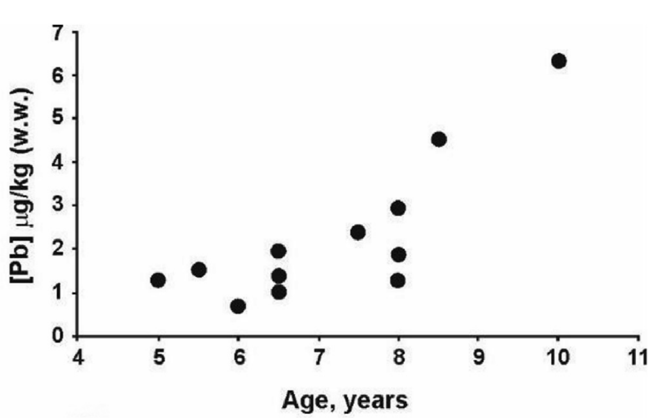

Fig. 3. Pb bioaccumulation rate in humpback whitefish.

If we consider Arctic char, pink salmon and navaga to feed predominantly in a marine habitat, the average concentrations in μg/kg for all elements in our samples (except for Ni) from these three fish species (n = 33) are statistically significantly different when compared with the less anadromous fish (n = 45). The observed mean amounts in μg/kg are: Hg (32 vs 93); As (3540 vs 200); Se (518 vs 285); Cd (3.2 vs 2.0); Pb (7.7 vs 4.5); Co (4.9 vs 8.7); Ni (21 vs 20); Cu (562 vs 270) and Zn (8.3 vs 6.0). All elements except Hg, Co and Ni were present at higher concentrations in fish feeding in marine habitats.

There was a significant species difference for Cd. The highest average amount, 6.6 μg/kg, was measured in pink salmon which feeds only in marine waters where the diet includes a diverse prey of copepods, pteropods and amphipods (Kaeriyama et al., 2000). Studies of zooplankton from the Barents Sea have revealed high Cd concentrations in the copepods which may explain the significantly increased Cd concentration in the one-year old pink salmon (Zauke and Schmalenbach, 2006).

There were also significant differences in the average total arsenic (totAs) concentrations among the fish species, which ranged from 69 to 30000 μg/kg in roach and navaga respectively. Marine species in general contain significantly higher amount of totAs than fresh-water fish, with cod featuring especially high amounts of total As; concentrations as high as 15–17 mg/kg have been reported (Sloth et al., 2005). In the present study, navaga had almost twice the totAs in muscle compared with other cod species. These observations for marine species was shown by Francesconi and Edmonds (1996) to be due to the transformation of inorganic As (iAs) to organic As-compounds. The latter have higher accumulation and retention in marine organisms. The predominant As compound in fish muscle is arsenobetaine (AsB), which is non-toxic and non-carcinogenic to mammals. Minor amounts of arseno-lipids and As-containing fatty acids (in oily fish) have been identified which, in addition to methylated arsenicals, are metabolites of iAs and arsenolipids (Taylor et al., 2017). Although considered to be cancerogenic, the concentration of iAs in marine species is negligible and typically ratios of totAs/iAs are featured in the range of 100 in marine mammals to about 50000 in cod and anglerfish (Sloth et al., 2005). No information on As speciation analysis of navaga was found in the literature.

As for Arctic char, pink salmon and navaga there was a comparable large variation in totAs concentration among whitefish, northern pike, roach and inconnu, even though the totAs concentrations were lower. Arsenic species distribution in freshwater fish has been less studied than in marine habitats, but similar As compounds have been found in muscle for fresh water fish species (Arroyo-Abad et al., 2016; de Rosemond et al., 2008; Komorowicz et al., 2019; Šlejkovec et al., 2004). The totAs concentrations we observed are similar to the results obtained by (de Rosemond et al., 2008) in northern pike and lake white fish from Black Bay in Northwestern Ontario, Canada. Their findings for muscle of lake whitefish and pike predominantly featured non-identified organic As species, 87.8 and 54.0% of the totAs respectively, as well as considerable amounts of AsB and dimethylarsenic acid. It should be noted that there are strong associations between As, Cu and Zn in marine fish which could suggest a common source and uptake of these elements (Table 5). The most significant association in the current study was between As and Hg in Arctic char, pink salmon and navaga ($r = 0.84$) which was primarily due to navaga with a Pearson r-value of 0.60.

The observed concentrations of Hg, Cd and Pb in all the fish species were lower than those specified in the Russian regulations pertaining to inorganic toxicants per wet-weight of fish muscles for human consumption, namely: Hg: 0.3 mg/kg for freshwater non-predator, 0.6 mg/kg for freshwater predators and 0.5 mg/kg for seawater fish; 0.2 mg/kg for Cd in all fish species; and 1.0 mg/kg in all fish species for Pb (SanPiN 2.3.2.1078-01, 2002). The concentrations were also below the EU maximum concentrations for Hg, namely 1.0 mg/kg for pike and 0.5 mg/kg for other fish species; 0.050 mg/kg for Cd and 0.3 mg/kg for Pb (Commission Regulation (EC) No 1881/2006). The amount of As in Arctic char exceeds the Russian regulation for totAs in freshwater fish of 1.0 mg/kg (wet-weight) by 2–3.8 times; in navaga As exceeds the Russian limit of 5.0 mg/kg (wet-weight) in seawater fish 2–13 fold (SanPiN 2.3.2.1078-01, 2002). The Russian regulation for As applies only to total As, although most of it occurs in fish predominantly as the non-toxic organic form arsenobetane (AsB). Thus, there is an urgent need to implement As-species based limits in the Russian regulations. As it is, several fish species in this study are considered not to be recommended for human consumption.

3.2. Essential elements

Selenium is essential for fish as it is incorporated in proteins, e.g. as selenocysteine. Fish may contain the largest quantity of selenoproteins among biota including several not found in other vertebrates (Janz, 2011). Of these, the best characterized are enzymes involved in antioxidant defense and thyroid hormone metabolism. The Se fish muscle concentrations in the present study were highest in the marine fish group (Arctic char, pink salmon and navaga), which also contained some of the lowest Hg concentrations. Since a protective role of Se on MeHg bioaccumulation has been observed and fish consumption is the primary wildlife and human exposure route for Hg, Se concentrations and the Hg/Se ratio are considered to be bioindicators of human risks associated with the consumption of Hg in fish (Burger and Gochfeld, 2011; Kasuya, 1976). Se in the form of selenide has a considerable higher affinity constant with Hg^{2+} than sulfide (10^{49} *versus* 10^{39}), and this favors the metabolic formation of HgSe. Consequently, it is understood that it is not the concentration of either Hg^{2+} nor MeHg that is critical for their toxic effect, but the molar ratio of Se to Hg. A molar Se/Hg ratio <1 appears to increase the toxic potential of Hg, while those which approach or exceed >1 increasingly protect against Hg toxicity (Belzile et al., 2006; Peterson et al., 2009). It is also important to note that an increase in the bioaccumulation of Hg has been observed with decreasing Se concentration in lake waters and in the various trophic compartments of an aquatic food web including amphipodes, zooplankton and perch (Belzile et al., 2006; Ralston et al., 2008). In the present study, there are large significant differences in the molar Se/Hg ratios that range from 2.3 for northern pike and up to 71.1 for pink salmon (see Table 4). This may be explained by the well-known bioaccumulation of Hg in northern pike, the presence of much lower amounts of Hg in salmonid species living predominantly in marine habitats, and the significantly lower amount of Se in northern pike muscle compared with the other species.

Surprisingly, comparable molar ratios exist for Cu/Hg in comparison to Se/Hg. The basis for this may be the highly statistically significant association between the Cu and Se in all fish analyzed (see Table 5 and Fig. 4). Humpback whitefish has a significantly higher Se/Cu ratio. Its exclusion changes the Pearson r-value from 0.16 to 0.40 ($p < 0.01$) for northern pike, roach and inconnu. There are also highly statistically significantly associations between the essential elements Cu, Se and Zn among all fish and the two subgroups. For Co these associations are only significant for the “marine” species. Fish may obtain these essential elements both directly from the ambient water through gills, their entire body surface or natural feed to ensure normal growth survival and essential cellular metabolism (Chanda et al., 2015). To our knowledge, these associations in fish muscle have not been described earlier in the literature, although Julshamn et al. (1990) have reported a strong positive correlation between Se and Cu in liver from farmed and wild Atlantic salmon.

4. Conclusions

Essential and toxic elements have been measured in fish species that are part of the Nenets nutritional intake. Of the fish selected for biomonitoring of environmental and essential inorganic elements, northern pike contained twice the amount of Hg compared with

Table 4
Molar ratios between Se, Hg and Cu in muscle of different species ($n = 78$).

Fish species	Se/Hg	Cu/Hg	Se/Cu
Arctic char	65.2	77.7	0.84
Pink salmon	71.1	89.7	0.79
Navaga	22.8	36.2	0.63
Humpback whitefish	15.7	11.4	1.38
Northern pike	2.3	3.1	0.74
Roach Pechora	8.0	9.9	0.80
Roach Indiga	8.8	11.3	0.78
Inconnu	11.0	22.2	0.50

Table 5
Univariate associations (Pearson's r and p-values) between elements in all fish (n = 78).

		LgAs	Se	Cd	Pb	Co	Ni	Cu	Zn
Hg	a	0.84^{***}	0.24^{ns}	-0.14^{ns}	0.13^{ns}	0.81^{***}	0.39^{*}	0.63^{***}	0.78^{***}
	b	0.20^{ns}	-0.38^{**}	0.07^{ns}	0.51^{***}	-0.55^{***}	0.08^{ns}	-0.21^{ns}	-0.34^{*}
	c	-0.18^{ns}	-0.55^{***}	-0.14^{ns}	0.12^{ns}	-0.19^{ns}	0.06^{ns}	-0.37^{**}	-0.13^{ns}
LgAs	a		0.28^{ns}	-0.46^{**}	0.07^{ns}	0.84^{**}	0.41^{*}	0.55^{**}	0.69^{***}
	b		-0.04^{ns}	-0.32^{*}	0.03^{ns}	0.27^{ns}	0.13^{ns}	-0.50^{**}	-0.32^{*}
	c		0.60^{***}	-0.10^{ns}	0.27^{**}	0.09^{ns}	0.22^{*}	0.55^{***}	0.50^{***}
Se	a			0.28^{ns}	0.28^{ns}	0.41^{*}	0.09^{ns}	0.42^{*}	0.40^{*}
	b			0.50^{**}	-0.02^{ns}	0.40^{**}	0.29^{ns}	0.16^{ns}	0.55^{***}
	c			0.40^{***}	0.34^{**}	0.01^{ns}	0.15^{ns}	0.71^{***}	0.52^{***}
Cd	a				0.04^{ns}	-0.25^{ns}	-0.28^{ns}	0.14^{ns}	$<0.01^{ns}$
	b				0.26^{ns}	-0.31^{*}	0.25^{ns}	0.23^{ns}	0.49^{**}
	c				0.19^{ns}	-0.27^{*}	-0.06^{ns}	0.29^{*}	0.19^{ns}
Pb	a					0.33^{ns}	0.11^{ns}	0.26^{ns}	0.26^{ns}
	b					-0.36^{*}	0.49^{**}	0.19^{ns}	-0.03^{ns}
	c					-0.24^{*}	0.30^{**}	0.39^{***}	0.27^{*}
Co	a						0.44^{**}	0.50^{**}	0.77^{***}
	b						0.09^{ns}	-0.14^{ns}	0.02^{ns}
	c						0.14^{ns}	-0.15^{ns}	0.10^{ns}
Ni	a							0.26^{ns}	0.40^{*}
	b							0.28^{ns}	0.31^{*}
	c							0.21^{ns}	0.33^{***}
Cu	a								0.70^{***}
	b								0.56^{***}
	c								0.70^{***}

a: Arctic char, pink salmon, navaga (n = 33); b: whitefish, northern pike, roach, inconnu (n = 45); c = all fish (n = 78).
***Significant at the <0.001 level.
**Significant at the <0.01 level.
*Significant at the <0.05 level.

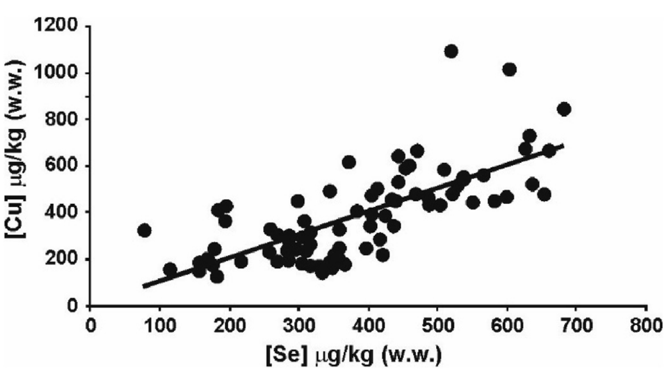

Fig. 4. Association between Se and Cu in fish muscle (n = 78, Pearson r-value = 0.71, p = < 0.001).

roach and 3–4 times more than the other fish species examined and that are commonly consumed in the Russian Arctic. In all fish species tested, both Cd and Pb were present in considerably lower concentrations than Hg. The totAs concentrations observed are similar to those previously published, and this element is assumed to be present in its non-toxic organic forms. Despite considerably information about the amount of totAs and the distribution of As species in marine fish reported in the literature, this is lacking for navaga. Clearly future speciation research pertaining to the characterizing of the different forms of As in the edible part of this Arctic species would be helpful. All fish tissues are rich in the essential elements Se, Cu and Zn and, depending on the amount fish consumed, may contribute significantly to the nutritional intake by indigenous Arctic people.

It is advisable that when fish species are included in a program for monitoring of the nutritional intake of PIPs, the molar Se/Hg ratio must be taken into consideration as values < 1 may increase the toxic potential of Hg and when >1 increase the protection against Hg toxicity. In conclusion, since Arctic char, pink salmon, navaga, northern pike, humpback, whitefish, roach and inconnu are all frequently consumed by the Nenets indigenous population, they should be included in a pan-Russian Arctic Monitoring Program.

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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