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Bioaccumulation of chemical elements in grey seals (*Halichoerus grypus*) from the Baltic Sea

Even Buvarp Helsingen

Environmental Toxicology and Chemistry
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Abstract

Concentrations of Ag, As, B, Ba, Bi, Ca, Cd, Ce, Co, Cs, Cu, Dy, Er, Fe, Ga, Ge, Hg, Ho, K, La, Li, Lu, Mg, Mn, Mo, Na, Nd, Ni, P, Pb, Pr, Rb, S, Sb, Se, Si, Sm, Sn, Sr, Tb, Th, Tl, V, W, Y, Yb and Zn in the livers of grey seals obtained from stranded, hunted and by-caught animals from the Swedish and Polish Baltic coasts were determined by ICP-MS. Stable isotope analysis of $\delta^{13}C$ and $\delta^{15}N$ was applied to link chemical element concentrations to feeding preferences (pelagic vs. benthic) and trophic position of the seals, respectively. The liver tissue samples of the seals from Polish coast were significantly enriched in ^{15}N indicating diet preferences towards species from higher trophic level such as e.g. cod (*Gadus morhua*). This finding is further supported by significantly lower hepatic values for $\delta^{13}C$ in seal from northern basins (Gulf of Bothnia) compared to more southern locations. Depletion of ^{13}C isotope indicates more pelagic feeding preferences (e.g. more herring and common white fish in the diet) in the seals from northern basin then benthic (e.g. cod) in the seals from the south.

Analysis of covariance (ANCOVA) revealed differences in chemical element concentrations between areas for As, B, Cu, Ni, Rb, Sr, Tl with most of these elements exhibiting higher concentrations in seals from the southern Baltic Sea. Additionally age related differences in concentrations of several trace elements were confirmed by analysis of variance (ANOVA). Silver, B, Bi, Cd, Co, Hg, Mo, Sb, Se, Sn, V and Zn and the rare earth elements (REE) were found to increase with age.

The role of Se in detoxification of Hg was confirmed by strong significant correlation between these two elements ($r = 0.99$, $p < 0.0001$). Higher Se:Hg molar ratio were observed in yearlings compared to juvenile and adult seals. This suggests a prominent role of maternal transfer of Se during lactation and the possible existence of a protective mechanism against Hg toxicity, especially in the early stage of development. Several elements revealed statistically significant interrelationships, especially pronounced for the REEs.

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1. Introduction

The anthropogenic pressure on the ecosystem of the Baltic Sea is tremendous. Nine countries border to the sea and more than 85 million people live in its catchment area. Large cities and industrial areas contribute with pollution and marine resources of the sea are heavily overexploited. Many species inhabiting the Baltic Sea are of marine origin, hence constantly on the edge of their physiological tolerance in this brackish water body with the salinity being only 7 ‰, or one fifth of the ocean concentration (Leppäranta and Myrberg 2009). The additional stress deriving from the anthropogenic load and a changing climate will likely have large impacts on these species (Schiedek *et al.* 2007).

During the last century the grey seal (*Halichoerus grypus*) population in the Baltic Sea has undergone some extreme events. In the beginning of the century, the population probably exceeded 90 000 seals, but bounty hunting reduced population numbers to about 20 000 towards the 1940s (Harding and Harkonen 1999; Harding *et al.* 2007). Hunting ceased during the Second World War, but the population continued to decrease until the middle of the 1970s when less than 4000 grey seals inhabited the Baltic Sea (Harding and Harkonen 1999).

Jensen et al. (1969) reported high levels of dichlorodiphenyltrichloroethane (DDT) and polychlorinated biphenyls (PCB) in Baltic seals. Thus, pollution was suspected as one of the main factors responsible for the overall decline of seal populations in the Baltic Sea (Hook and Johnels 1972; Olsson *et al.* 1975). During the same period Helle et al. (1976a; 1976b) found severe pathological changes manifesting in the uterus(occlusions and stenosis) as signs of interrupted pregnancies. Other effects that have been related to organochlorine pollution are hormonal disturbances (adrenocortical hyperplasia), arteriosclerosis and lesions of kidneys, skull, intestines and integument (Bergman and Olsson 1986; Zakharov and Yablokov 1990; Bergman *et al.* 1992; Bergman 1999; Bergman *et al.* 2001; Bäcklin *et al.* 2003; Bredhult *et al.* 2008).

The decreased concentrations of PCBs and DDT in seals (Nyman *et al.* 2002; Bäcklin *et al.* 2010a) and other biota during the last decades (Falandysz 1981; Moilanen *et al.* 1982; Olsson and Reutergardh 1986; Haahti and Perttila 1988; Kannan *et al.* 1992; Bignert *et al.* 1998), the ban of hunting in 1974 (Bäcklin *et al.* 2010b) and a positive trend regarding the gynecological health (Bergman 1999), have been suspected as major factors for the increasing grey seal population in the Baltic Sea. The growth rate of the Swedish population of grey seals was estimated to 7.5% between 1990-2004, with the total population in the

Baltic Sea outnumbering 22000 individuals in 2005 (Karlsson and Helander 2005). However, in recent years the population incline has leveled off. Furthermore a negative population trend was observed between 2008 and 2009, together with a significant decrease in blubber thickness of juvenile and adult seals (Bäcklin *et al.* 2010a).

1.1 Chemical elements and pollution in the Baltic Sea

During the 20th century the Baltic Sea has changed from a clean oligotrophic sea to one of the most polluted areas in the world (Melvasalo 1980). Sediment studies in the Baltic Sea showed a constant increase in concentration of several chemical elements from the 1950s, reaching a peak around 1980 (Jonsson 1992). High levels of cadmium (Cd), zinc (Zn) and lead (Pb) were found in the Baltic proper and arsenic (As) dominated in the Bothnian Bay. Mercury (Hg) have revealed elevated concentrations in the Bothnian Bay, coastal areas of the Bothnian Sea and in areas around Stockholm (Borg and Jonsson 1996). Even though the heavy metals and legacy POPs contaminant levels are decreasing, they are still higher than before the Second World War (Jonsson 1992). At the same time new emerging organic pollutatnts such as brominated flame retardants (BFRs), and perfluoroalkyl and polyfluoroalkyl substances (PFASs) were found to increase in biota of the Baltic Sea ecosystem (Holmström and Urs 2008; HELCOM 2010).

The atmospheric and waterborne inputs of Cd, Pb and Hg have decreased in the period from 1990-2008 (Gusev 2010; PLC-Group 2010). However, only Hg was found to decrease in water concentration in the Baltic Sea since 2000 (Pohl and Hennings 2010). Downward trends for these heavy metals have been reported in fish and other biota (Polak-Juszczak 2009; Bignert *et al.* 2010), but there are differences between areas, metals and species (Bignert *et al.* 2010; HELCOM 2010).

Another problem in the Baltic Sea is the increasing level of eutrophication in large areas of the Baltic Sea. Since the beginning of the 1900, nitrogen (N) and phosphorous (P) input to the Baltic Sea has increased 4- and 8 fold, respectively (Larsson *et al.* 1985). The organic matter settles on the sea floor and with its decomposition exhausting the available oxygen, leaving wide areas of the Baltic seafloor anoxic. Major inflow events of oxygen rich Atlantic water can rapidly change the situation and alter the present redox conditions, which again, can influence the suspension and re-suspension of different chemical elements (Delaune and Smith 1985; Calmano *et al.* 1994; Petersen *et al.* 1997).

Regardless of its reputation as one of the most polluted areas of the world, studies on chemical elements and metals in the Baltic Sea are relatively sparse in comparison to organochlorines. The same holds true for studies addressing concentrations and effects in marine mammals living in the Baltic Sea. Frank et al. (1992) investigated the metal concentrations in seal species from the Swedish coast and found that the concentration of several elements in Baltic grey and ringed seals were higher compared to coastal living Baltic harbor seals. It was also found that Hg, Se and Cd levels were increased with age of the seals. Fant *et al.* (2001) compared the concentration of four selected metals in ringed seals (*Phoca hispida*) from the Baltic Sea and Svalbard and found significantly higher concentrations of Hg and Se, and lower of Cd in Baltic ringed seals. In the study by Szefer et al. (2002a) the concentrations of selected metals were compared in harbor porpoise (*Phocoena phocoena*) from the Baltic Sea, Danish and Greenland coastal waters. Elevated levels of Cd were related to Cd rich diet of animals inhabiting the coast of Greenland. Ciesielski et al. (2006b), investigated the chemical element concentration in different mammals collected from the Polish coast of the Baltic Sea. Cadmium and Hg concentrations were found to be correlated to age and Se, respectively. Furthermore, the seals health condition was regarded as good.

1.2 Effects of chemical element pollution

The effect of metals on marine mammals have been reviewed by several authors (O`Shea 1999; Das *et al.* 2003b; Kakuschke and Prange 2007), but little information is available except for tissue concentrations. However, some relationships between metal contamination and health status are worth mentioning. Hyvärinen and Sapilä (1984) studied a ringed seal (*Phoca hispida saimensis*) population and found significantly higher concentrations of nickel (Ni) in still-born pups compared to live-born pups and adults. Harp seals (*Phagophilus groenlandicus*) experimentally intoxicated with methyl-Hg (MeHg) (25mg/kg/day), experienced lethargy and loss of weight, finally resulting in death (Ronald *et al.* 1977). MeHg have also been reported to alter the *in vitro* biosynthesis of steroids in in adrenal and testicular cell cultures from both grey and harp seals (Freeman *et al.* 1975; Freeman and Sangalang 1977).

Metals have also been suspected to lower the resistance to diseases. In harbor porpoises from German waters, a relationship between high concentrations of Hg and the prevalence of parasitic infections and pneumonia was reported (Siebert *et al.* 2001). Bennet *et al.* (2001) also suspected a reduced resistance in harbor porpoises from the coast of England and Wales and found significantly higher metal burdens in porpoises that died of infectious diseases.

Furthermore, negative relationships between metals and the immune function of marine mammals were found in other studies (De Guise *et al.* 1996; Pillet *et al.* 2000; Nakata *et al.* 2002; Lalancette *et al.* 2003; Kakuschke *et al.* 2008).

1.3 Baltic grey seals

The grey seal is one of four native marine mammals in the Baltic Sea, the others being the cetacean harbor porpoise and two pinniped species – common seal (*Phoca vitulina*) and the ringed seal. Other marine mammals can also appear sporadically (Skóra 1991). The grey seal is the only member of the genus *Halichoerus,* in the family *Phocidae* or true seals. It has its natural distribution in cold temperate and sub-arctic areas in the North Atlantic Ocean and the Baltic Sea (Hall and Thompson 2009). Two different sub-species have been identified - *Halichoerus grypus grypus,* inhabiting both sides of the North Atlantic Ocean and *Halichoerus grypus macroynchus,* in the Baltic Sea (Rice 1998).

In Baltic Sea food web, the grey seal is one of the marine top predators. Their long lifespan and the long biological half-time for elimination of pollutants make them susceptible for accumulating certain pollutants (e.g. Hg). Thus, grey seals have been proposed to be used as a bioindicator pollution monitoring studies (Das *et al.* 2003b). Uptake routes for chemical elements in marine mammals include atmospheric uptake, absorption through the skin and transfer from mother to pup (milk and placenta). However uptake through ingested food is the most important source (Das *et al.* 2003b).

Several studies have examined the diet and feeding habits of marine mammals (see Pauly et al. 1998 for an overview). A recent study by Lundström *et al.* (2007) examinated the digestive tract content of 145 grey seals from the Baltic Sea. They identified 24 different prey species, but only a few of these contributed significantly to the diet. Herring (*Clupea harrengus*) was the most important, followed by common whitefish (*Coregonus lavarentus*), sprat (*Sprattus sprattus*), flounder (*Platichtys flesus*) and salmon (*Salmo salar*). A similar study from the 70s concluded that cod, herring and salmon were the most important prey species (Söderberg 1971) These differences indicate a shift in the diet composition of the grey seals in the last 3-4 decades. Moreover, Lundström *et al.* (2007) found differences in diets between seals of different age and between seals from the northern and southern Baltic Sea. Sprat seems to be more important in the south, while common whitefish in the north (Lundström *et al.* 2010). These distinct differences in diets between north and south are likely resulting from the salinity gradient within the Baltic Sea and the associated species distribution. Such differences in diet of seals from distinct areas in the Baltic have also been confirmed by the use carbon and nitrogen stable isotopes (Svensson *et al.* 2007, unpublished).

To assess the impact of anthropogenic activities on the health of marine mammals and the population viability, information about source, pathways and degree of biomagnification of contaminants is crucial. One of the methods to assess community structures in marine ecosystems are by using C and N stable isotopes. Naturally occurring isotope ratios of carbon ($^{13}C/^{12}C$) and nitrogen ($^{15}N/^{14}N$) can be used to trace dietary patterns and trophic relationships in ecosystems (DeNiro and Epstein 1977; Peterson and Fry 1987). The principle behind the method relies on the fact that C and N isotopes undergo isotopic separation (fractionation) between the trophic levels. This phenomenon results in enrichment of heavier isotope of N and thus $^{15}N/^{14}N$ ratio can reflect organisms position in the food web. On the other hand, the $^{13}C/^{12}C$ isotopic ratio remains almost unchanged and give an indication regarding the isotopic signature of the sources of primary production (Peterson and Fry 1987; Kelly 2000). Compared to studies on stomach contents which give only snap-shot information about what the animals were eating before sampling, stable isotope analysis provide time-integrated dietary information on assimilated food. Additionally, different tissues have different metabolic rates and thus different turnover rates for stable isotopes. This implies that different tissues from an animal can be used to assess dietary information over different time scales (Tieszen *et al.* 1983).

1.4 Aim of the study

According to our knowledge there are no studies on the bioaccumulation of chemical elements in relation to trophic position of marine mammals in the Baltic Sea. Recent studies (Lundström *et al.* 2007; Svensson *et al.* 2007; Lundström *et al.* 2010) suggests that the diet of the grey seals differ in different regions (north-south) of the Baltic Sea, and that the seals therefore occupy different trophic levels in the two regions.

The aim of the present study was to examine the relationships between the trophic position and bioaccumulation of chemical elements in grey seals from different locations from the Baltic Sea It is hypothesized that grey seals from the southern region occupy a higher trophic level than seals from the northern. Furthermore, the differences in trophic position will be reflected in degree of bioaccumulation of chemical elements in hepatic tissue of seals inhabiting distinct geographical areas.

2. Material and method

2.1 Samples

Liver (n = 76) samples used in this study were obtained from grey seals (*Halichoerus grypus*) incidentally caught in fish nets, stranded or shot from the coast of Sweden (n = 69) and the Polish coast (n = 7) as can be seen in **Figure 1**. Samples were collected from December 2007 to January 2010. Additional samples including Atlantic herring (*Clupea harengus*) (n = 20), stickleback (*Gasterosteus aculeatus*) (n = 2) and *Saduria entomon (*n = 32, pooled into 11 samples), were obtained from the Baltic Sea. All samples were freeze dried before analysis

2.2 Chemical element analysis

Liver samples were mechanically crushed and analyzed without further homogenization, while the whole individuals of herring, stickleback and *S. entomon* were homogenized in a mixer mill (MM 400, Reutsch, Haan Germany). Approximately 0.4 - 0.5 g of the sample material was transferred (accurate to 4 significant digits) inside PTFE-Teflon vials (18 mL). Further, 3 mL ultrapure water (Q-option, Elga Labwater, Veolia Water Systems LTD, United Kingdom) and 3 mL concentrated nitric acid (Scan Pure, equal to ultra pure grade, Chemscan, Elverum, Norway) was added to the vials. Digestion of the samples were performed in a high-pressure microwave system (Milestone UltraClave, EMLS, Leutkirch, Germany), that gradually increase temperature to a maximum of 240 ^{o}C within one hour. In addition there was a cooling step that returned the temperature to the initial value within one hour. After cooling of the samples, they were diluted with ultrapure water to a final acid concentration of 0.6 M.

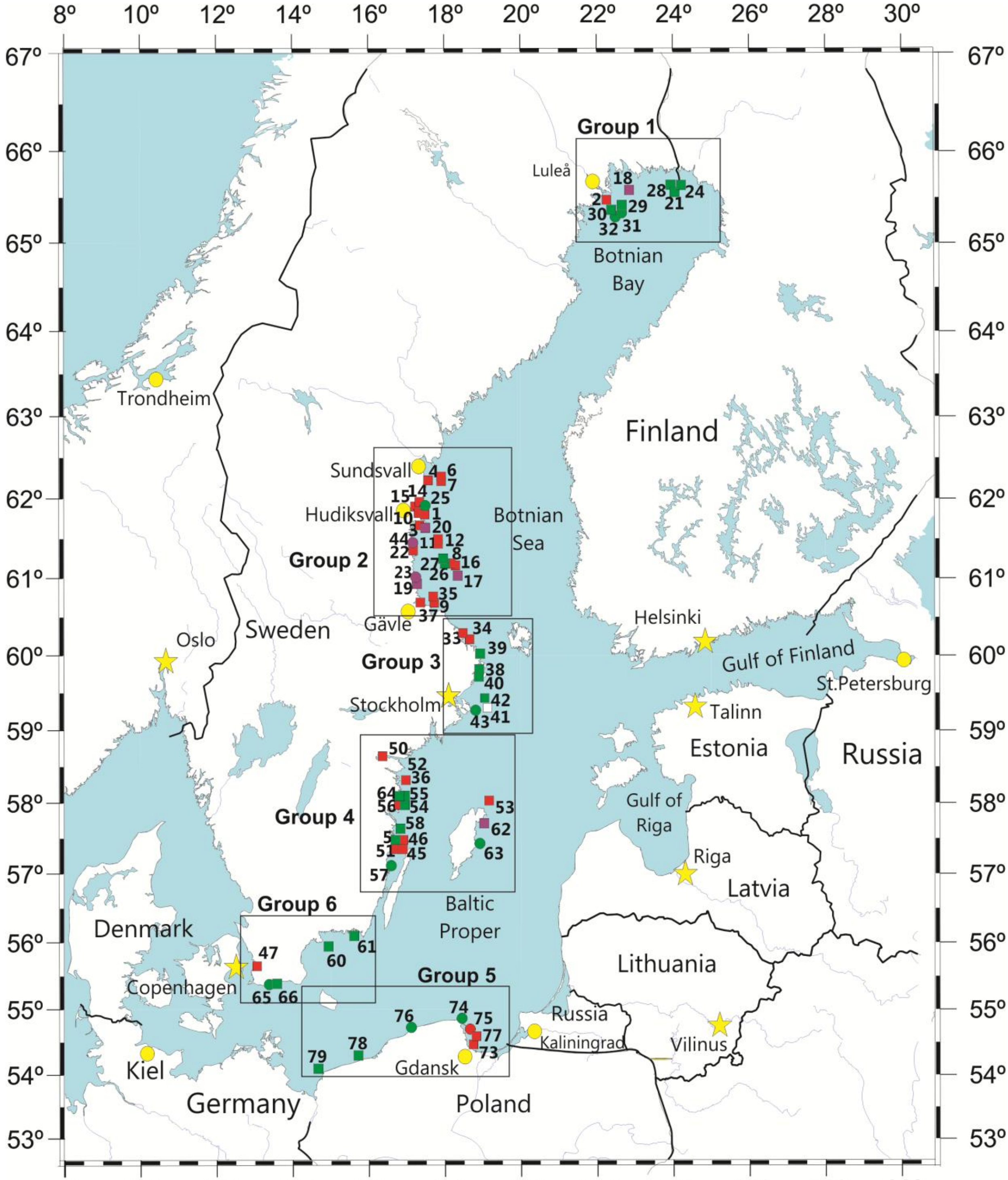

Figure 1: Sample locations for grey seals in the Baltic Sea. Squares and circles refer to males and females respectively, and red, green and purple refers to adults, juveniles and yearlings respectively. See text for group descriptions. No coordinates was obtained for seal no. 13, 48, 49, 59, 68, 69, 70, 71 and 72, but grouping can be found in Appendix IV.

High resolution Inductively Coupled Mass Spectrometry (HR-ICP-MS) analyses were performed using a Thermo Finnigan model Element 2 instrument (Bremen, Germany). The radio frequency power was set to 1400 W. Samples were introduced using a SC-FAST flow injection analysis system (ESI, Elemental Scientific, Inc. Omaha, USA) with a peristaltic pump (1 mL/min). The instrument was equipped with a PFA-ST nebulizer, spray chamber (PFA Barrel 35 mm), demountable torch, quarts standard injector and Al sample skimmer and X skimmer cones. The nebulizer argon gas flow rate was adjusted to give a stable signal with maximum intensity for the nuclides ^{7}Li, ^{115}In and ^{238}U. Methane gas was used in the analysis to minimize interference from carbon and to provide enhanced sensitivity, especially for Se and As. The instrument was calibrated using 0.6 HNO_3 solutions of matrix-matched mulitielement standards. A calibration curve consisting of 5 different concentrations was made from these standards. To check for instrument drift one of these multi-element standards were analyzed for every ten samples. The accuracy of the method was verified by analyzing the certified reference material Bovine Liver NIST 1577b (National Institute of Standards and Technology, Gaithersburd, MD). Results were in good agreement with the certified values, and recovery of the elements ranged from 85 - 113 %.

Method detection limits (MDL) (**Appendix V**) were calculated depending on which method that gave the highest value of 3 times standard deviation of the blanks, or on the instrument detection limit (IDL). Estimation of IDLs was done by subsequent analysis of solutions containing decreasing solutions of the element. Finally the concentration resulting in a relative standard deviation of approximately 25 % (n = 3 scans) were chosen as IDL with baseline corrections applied for these values. Results from chemical analysis that were below the limit of detection (LOD), were replaced with ½*LOD for the measured element. Chemical elements where more than 50 % of the samples where below LOD were omitted from further statistical analysis, including Aluminum (Al), Beryllium (Be), Chromium (Cr), Hafnium (Hf), Niobium (Nb), Scandium (Sc), Tantalum (Ta), Titanium (Ti), Thulium (Tm), Uranium (U) and Zirconium (Zr).

2.3 Stable isotope analysis of C and N

All sample preparations for stable isotope analysis were performed at NTNU. Samples of grey seal liver (n = 73), herring, stickleback and *S. entomon* were ground into a homogenous powder by a mixer mill (MM 400, Reutsch, Haan, Germany). Lipids have bend found to have a low $^{13}C/^{12}C$ ratio, relatively to the diet (DeNiro and Epstein 1977; Tieszen *et al.* 1983), and also varying in concentration between different organisms and tissues. Thus lipids were removed prior to analysis using a modified method of Folch et al. (1957). About 100 - 300 mg of ground tissue was soaked in 3 mL of a 2:1 chloroform:methanol (by volume) solution, and vortexed for 30 seconds. The mixture was then left alone for greater than 30 minutes and centrifuged at 1300 rpm. Then the supernatant was discarded and the pellet was re-suspended in solvent mixture. These steps were repeated until the supernatant was colorless. The sample was then dried at 60 ^{o}C for 24 hours.

Carbon incorporation into tissue and carbonate are of different origin and differs in $\delta^{13}C$. Removal of carbonate is thus recommended (Fry 1988; Rau *et al.* 1992; Cloern *et al.* 2002), therefore samples of *S. entomon*, herring and stickleback were subjected to acid wash. 1 N HCl was added drop by drop until no more CO_2 formed. The samples were then dried at 60 ^{o}C for at least 24 hours and then reground.

Stable isotope analyses were performed at the Institute of Energy Technology (IFE), Kjeller, Norway. Approximately 1.0 mg of sample was weighed into 8 x 5 mm tin capsules and combusted in the presence of O_2 and Cr_2O_3 at 1700 ^{o}C in a Eurovector element analyser. Reduction of NO_x to N_2 was done in a Cu oven at 650 ^{o}C. Water was removed in a chemical trap of $Mg(ClO_4)_2$ before separation of N_2 and CO_2 on a 2 m Poraplot Q gas-chromatography (GC) column. N_2 and CO_2 were directly injected on-line to a Nu Instruments Horizon, Isotope Ratio Mass Spectrometer (IRMS) (Wrexham, North Wales, UK) for determination of $\delta^{13}C$ and $\delta^{15}N$. Isotope values are reported in delta notation according to the equation:

$$\delta X = [(R_{sample}/R_{standard}) - 1] \times 1000 \qquad (1)$$

where X is ^{13}C or ^{15}N, R_{sample} is the ratio of heavy to light isotope ($^{13}C/^{12}C$ or $^{15}N/^{14}N$) in the sample and $R_{standard}$ is the heavy to light istope ratio in the standards. Accuracy and precision of analysis were measured by internal standard (IFE trout) and international standards - USGS-24 (carbon; from the International Atomic Agency [IAEA]) calibrated against PeeDee Belemnite, Vienna (VPDB) for ^{13}C and IAEA-N-1 and IAEA-N-2 ($[(NH_4)_2 SO_4]$ - ammonium sulphate) calibrated against atmospheric N_2 for ^{15}N. Repeated analysis of the internal standard indicated analytical errors of 0.06 ‰ and 0.12 ‰ for carbon and nitrogen, respectively.

2.4 Statistics

All variables were $\log_{10}$-transformed prior to the statistical analyses to approach normal distribution of the data.

A one way analysis of variance (ANOVA) was performed to check for differences between mean concentrations of chemical elements between age classes of seals. Furthermore, the analysis of covariance (ANCOVA) approach, was used to investigate differences between areas, using length to correct for the size of the seals. Length was selected as covariate to approximate for seals age, because of lacking age data. ANCOVA is a merge of the linear regression technique and ANOVA, thus capable of removing the obscuring effects of pre-existing individual differences among the seals (e.g. age or length). To test for differences between groups within the ANOVA and ANCOVA models, Tukey Honestly Significant Difference (HSD) test was performed. Tukey HSD tests for significant differences between the means of the different groups.

Seals were grouped into age and geographical location. For age, the seals were grouped into yearlings (< 1 year), juveniles (1 - 3 years) and adults (> 4 years). For investigation of spatial differences in element concentration, seals were divided into six groups corresponding to adjacent areas of the Baltic Sea (see **Figure 1**) Seals from Group 1 (n = 9) were collected in the Bothnian Bay, Group 2 (n = 26) - Bothnian Sea, Group 3 (n = 8) - the Sea of Åland, Group 4 (n = 23) - the Baltic Proper, Group 5 (n = 7) - the Polish coast and Group 6 (n = 5) - the Bornholm and Arcona basins.

The multivariate technique, principal component analysis (PCA) and the Pearson's product-moment coefficient (Pearson correlation) were used to review the relationships between the chemical elements, Se:Hg molar ratio, isotopic values ($\delta^{13}C$ and $\delta^{15}N$ and biometric data (age, length and weight). In PCA the systematic variation in a dataset can be taken out and described by only a few parameters that more easily can be interpreted. In theory, PCA corresponds to a mathematical decomposition of the data matrix X into two different parts, the score matrix and loading matrix for relationships between observations (samples) and variables (data) respectively. Observations with similar values will group together in a score plot, and samples with different values will be located further away. In loading plots the variables grouping together are likely correlated with each other.

The ANOVA and ANCOVA were performed using STATISTICA (Version 10 for Windows, StatSoft Inc., Tulsa, OK, USA). For PCA analyses SIMCA P+ (Version 12, Umertrics, Umeå, Sweden) were applied.

For all analyses the significance level was set to $p < 0.05$. For comparison of element concentration reported in other studies, fresh weight concentrations were converted to dry weight using the a factor of 3.3 as suggested by Yang and Miyazaki (2003).

3. Results

3.1 Stable isotopes

Stable isotopes of $\delta^{15}N$ in grey seals hepatic tissues ranged from 12.75 ‰ in an individual from the Bothnian Sea to 19.10 ‰ in another from the Baltic Proper, averaging 14.53 ‰. For $\delta^{13}C$, seals from the Baltic Proper exhibited both the lowest (-22.50 ‰) and the highest (-15.61 ‰) values, with an average of about -19.06 ‰. Isotopic means and ranges for all seal groups and food items can be found in **Appendix II** and **III**, respectively.

A positive correlation ($r = 0.37$, $p = 0.001$) between $\delta^{15}N$ and $\delta^{13}C$ (**Figure 2**) was observed. The correlation was further improved ($r = 0.58$, $p < 0.001$) when removing four male seals, no. 21, 46, 50 and 51 that could be considered outliers from the rest of the seals. Significant differences was observed between age groups of both $\delta^{15}N$ ($F_{2,70} = 4.23$, $p < 0.02$) and $\delta^{13}C$ ($F_{2,70} = 3.18$, $p < 0.05$). Adult had higher ratios of $\delta^{15}N$ compared to yearlings (Tukey HSD; $p < 0.02$), but not to juveniles (Tukey HSD; $p > 0.05$). For $\delta^{13}C$, no significant differences could be obtained between the age groups (Tukey HSD; $p > 0.05$), but a general decrease was observed from adults towards yearlings. Between the sexes, no differences could be obtained due to the low number of adult females ($n = 2$).

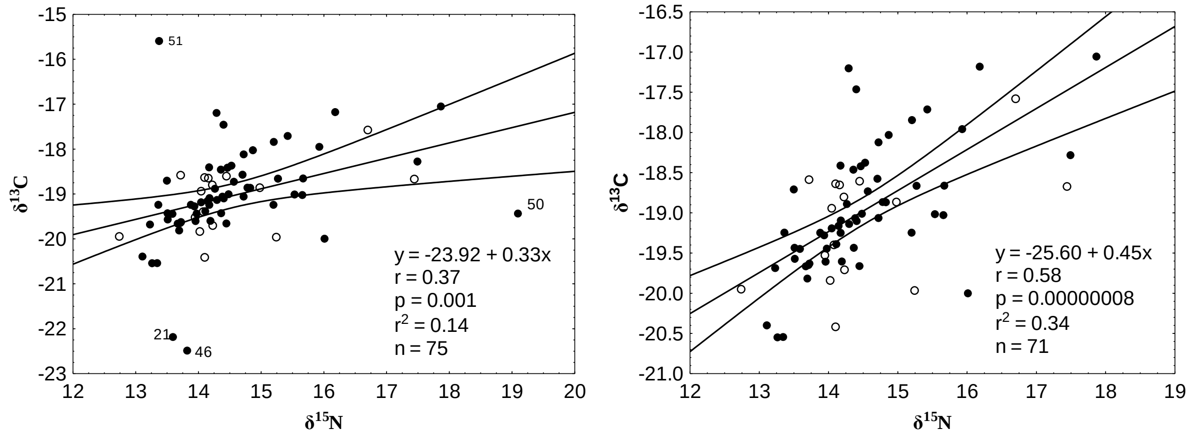

Figure 2: Relationship between of $\delta^{15}N$ and $\delta^{13}C$ in males (●) and females (○) of grey seals (*Halichoerus grypus*) of all age groups from the Baltic Sea. The solid line represents the correlation between $\delta^{15}N$ and $\delta^{13}C$ for all seals and curved lines denotes the 95% confidence interval. In the left figure, all seals with isotopic values are included, while in the right, seal number 21, 46, 50 and 51 are removed.

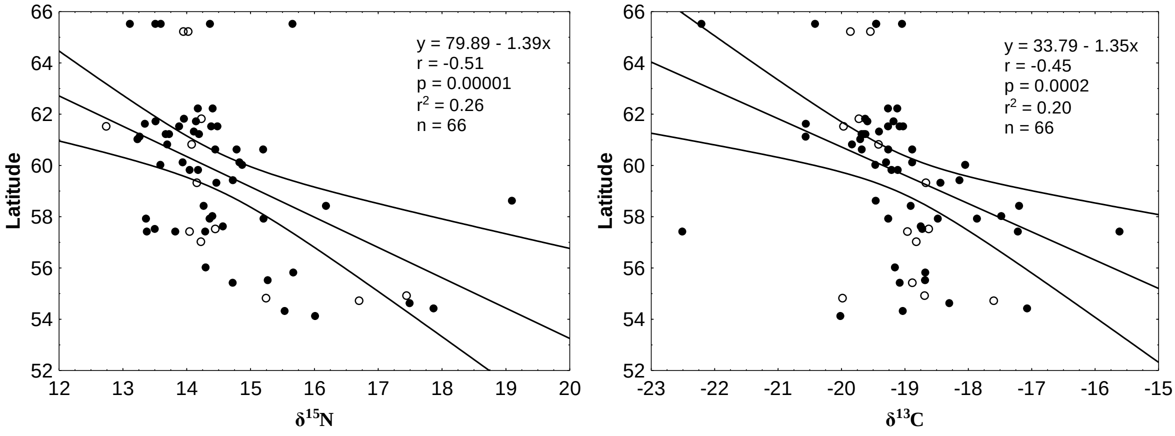

Figure 3: Relationship between of δ^{15}N (left) and δ^{13}C (right) in males (●) and females (○) of grey seals (*Halichoerus grypus*) of all age groups along the latitude gradient of the Baltic Sea. The straight line represents the correlation between the isotope and the latitude for all seals and curved lines denotes the 95% confidence interval

A negative correlation was observed for both δ^{15}N ($r = -0.51$, $p < 0.001$) and δ^{13}C ($r = -0.44$, $p < 0.001$) in liver versus the gradient from north to south of the Baltic Sea (**Figure 3**). The correlation was further improved for δ^{15}N when removing seal no. 50 ($r = -0.57$, $p < 0.001$) and seal no. 46 for δ^{13}C ($r = -0.53$, $p < 0.001$).

Means of δ^{15}N and δ^{13}C for different geographical groups in the Baltic (**Figure 4**), adjusted for length, indicated significant differences ($F_{(5,68)} = 14.75$, $p < 0.0001$ and $F_{(5,68)} = 4.16$, $p = 0.002$ for δ^{15}N and δ^{13}C, respectively). Seals from the Polish coast had significantly higher δ^{15}N values relative to the northern groups from the Sea of Åland and the Gulf of Bothnia (Tukey HSD; $p < 0.0002$), and the group from the Bornholm and Arcona basins (Tukey HSD; $p < 0.02$). The seals from the Sea of Åland, Baltic Proper and the Polish coast had significantly higher δ^{13}C values compared to seals from the Bothnian Bay (Tukey HSD; $p < 0.05$).

The stable isotope means of the suspected food items, herring, stickleback and *S. entomon*, were lower in δ^{15}N (10.09, 8.90 and 8.27 ‰, respectively) relative to the seals. For δ^{13}C the means were more analogous to the seals (-21.20, -20.85 and -18.76 ‰ for herring, stickleback and *S. entomon*, respectively).

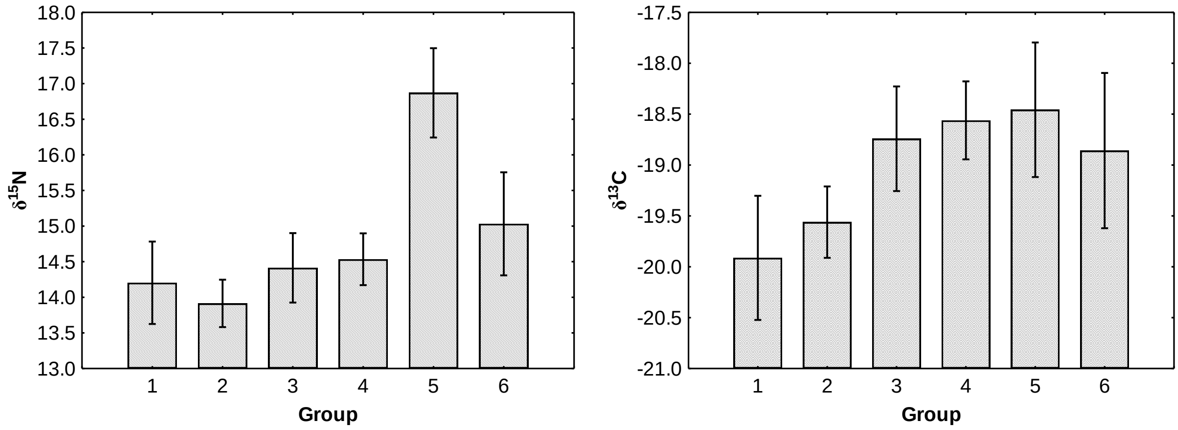

Figure 4: Comparison of length adjusted means ± standard deviation (SD), for $\delta^{15}N$ (left) and $\delta^{13}C$ (right) between different groups of grey seals (*Halichoerus grypus*) in the Baltic Sea. Non transformed values are graphed for illustrative purposes.

3.2 Chemical elements

The concentration of 58 elements was measured in the liver of 76 grey seals, divided into six different groups in the Baltic Sea. Of those, 47 chemical elements were reported and statistically treated. The mean concentrations for macro elements, rare earth elements (REE) and trace elements in hepatic tissue for all geographical and age groups can be found in **Appendix VII - IX**. Chemical element concentration in food items herring, stickleback and *S. entmon* are found in **Appendix VI**. All concentrations are reported in $\mu g \cdot g^{-1}$ dry weight. Biometric data of the seals can be found in **Appendix IV**.

Mercury was the most abundant trace element in the hepatic tissue of the grey seals, with a total concentration of 80.4 ± 151.1 (range 1.5 - 917) $\mu g \cdot g^{-1}$. Seal no. 75, a stranded adult female and no. 51, a shot adult male (the oldest seal) had extremely high values of Hg with 917.3 and 823.4 $\mu g \cdot g^{-1}$, respectively. Four other seals exhibited Hg concentrations above 200 $\mu g \cdot g^{-1}$ (seal no. 53, 50, 70 and 71). The mean concentration differences were significant among adults (139 ± 209.4 $\mu g \cdot g^{-1}$), juveniles (43.2 ± 55.5 $\mu g \cdot g^{-1}$) and yearlings (2.6 ± 0.9 $\mu g \cdot g^{-1}$) (Tukey HSD; $p < 0.001$).

Following Hg, other trace elements found in high total concentrations were Cu (62.9 ± 40.2 $\mu g \cdot g^{-1}$), Se (30.5 ± 54.3 $\mu g \cdot g^{-1}$), Mn (11.3 ± 4.5 $\mu g \cdot g^{-1}$) and Rb (7.1 ± 2.5 $\mu g \cdot g^{-1}$). For Se, concentrations were highest in adults (51.4 ± 75.5 $\mu g \cdot g^{-1}$) followed by juveniles (17.4 ± 19.2 $\mu g \cdot g^{-1}$) and yearlings (2.3 ± 0.2 $\mu g \cdot g^{-1}$). The two seals (no. 75 and 51) that contained high Hg levels, also exhibited higher Se concentrations than the other seals, with 340.0 and 284.5 $\mu g \cdot g^{-1}$ respectively. Total concentrations of Cd, Pb and Sn were 0.8 ± 0.7, 0.05 ± 0.04 and 0.8 ± 1.8 $\mu g \cdot g^{-1}$, respectively. For all these three elements, concentrations increased with age. Ce, Dy and Er were the REEs with the highest concentrations 68.0 ± 82.2, 40.2 ± 49.5 and 26.9 ± 32.8 $ng \cdot g^{-1}$, respectively.

Due to high signal from previous sample during the element analysis, the concentration of Hg in seal no. 74 was overestimated. Therefore, the Hg concentration for this seal was calculated from Se based on exceptionally high correlation between Hg and Se ($y = 1.60 + 0.36x$, $r = 0.99$, $p < 0.001$) to be 7.74 $\mu g \cdot g^{-1}$. Two seals (no. 65 and 69) were suspected of sample contamination, having extreme values of Al, Si, Cd and several of the REEs, and these were thus omitted from all statistical analyses.

No sexual differences in element concentrations could be obtained due to lack of adult female seals ($n = 2$).

3.2.1 Spatial variation

When correcting for length, most elements did not show any clear differences between seals from different areas (**Table 1**). However, a few elements revealed spatial differences ranging from Yb ($F_{5,67} = 2.45$, $p = 0.04$) exhibiting the least significant difference to Ni with the largest ($F_{5,67} = 13.1$, $p < 0.001$). Significantly higher concentrations in seals from the Polish coast compared to the other geographical groups were found for B, Ni and Sr (Tukey HSD; $p < 0.01$), while weaker for Ba, Fe, Ga, Li, Na and Th (Tukey HSD; $p < 0.05$). For Cu, P, Rb, S and Tl, the concentrations were higher in the three northern groups. For Cd, the ANCOVA model did not produce any significant differences, but post hoc test revealed significantly lower concentrations in seals from the Polish coast, compared to seals from the Bothnian Sea and the Sea of Åland (Tukey HSD; $p < 0.007$). In the case of As, significantly higher concentrations were found in the Bothnian Sea (Tukey HSD; $p < 0.002$), Sea of Åland (Tukey HSD; $p < 0.05$) and the Bornholm and Arcona basins (Tukey HSD; $p < 0.01$), compared to the Bothnian Bay.

3.2.2 Effects of age

Between age groups, ANOVA revealed several significant differences (**Table 2**). In general adults contained higher concentrations of most investigated elements compared to juveniles and yearlings, indicating bioaccumulation. Only Cu revealed another pattern, with juveniles having higher concentrations (Tukey HSD; $p < 0.0005$) compared to adults and yearlings.

A few compounds revealed large differences ($F_{2,71} > 20$) between the groups, including Bi, Cd, Hg, Sb, Se, V and most of the REEs ($p < 0.0001$). Weaker differences ($F_{2,71} > 10$) were found for Cu, Ga, Mo, Sn and Lu ($p < 0.001$). Of the macro elements, only S displayed significant age differences. The REEs were found to increase with age, and the lighter REEs (Ce, La and Pr) generally vary more between the groups than the heavier REEs (Ho, Lu and Yb).

Table 1: Influence of age group on the metal concentration in grey seal *(Halichoerus grypus)* from the Baltic Sea (ANOVA). All data reported on log transformed data. Bold F-values indicate significant models.

	$F_{2,71}$-value	**p-value**		**$F_{2,71}$-value**	**p-value**
Trace elements					
Ag	**9.52**	0.0002	V	**56.23**	< 0.0001
As	0.85	0.43	W	1.23	0.29
B	**5.70**	0.005	Zn	**5.03**	0.009
Ba	0.43	0.65			
Bi	**24.92**	< 0.0001	**Macro elements**		
Cd	**22.01**	< 0.0001	Ca	0.14	0.87
Co	**8.27**	0.0006	Fe	1.16	0.32
Cs	0.84	0.43	K	0.50	0.59
Cu	**10.59**	0.0002	Mg	0.50	0.62
Ga	**11.00**	< 0.0001	Na	0.81	0.45
Ge	0.35	0.70	P	0.90	0.39
Hg	**31.90**	< 0.0001	S	**4.00**	0.02
Li	**4.05**	0.02			
Mn	0.53	0.59	**Rare Earth Elements**		
Mo	**14.38**	< 0.0001	Ce	**32.23**	< 0.0001
Ni	0.27	0.76	Dy	**30.55**	< 0.0001
Pb	1.29	0.28	Er	**29.86**	< 0.0001
Rb	0.07	0.93	Ho	**25.56**	< 0.0001
Sb	**22.73**	< 0.0001	La	**33.70**	< 0.0001
Se	**22.84**	< 0.0001	Lu	**15.23**	< 0.0001
Se:Hg	**48.26**	< 0.0001	Nd	**30.85**	< 0.0001
Si	0.70	0.50	Pr	**32.21**	< 0.0001
Sn	**13.75**	< 0.0001	Sm	**31.18**	< 0.0001
Sr	2.69	0.07	Tb	**30.66**	< 0.0001
Th	0.73	0.48	Y	**29.37**	< 0.0001
Tl	1.10	0.34	Yb	**27.73**	< 0.0001

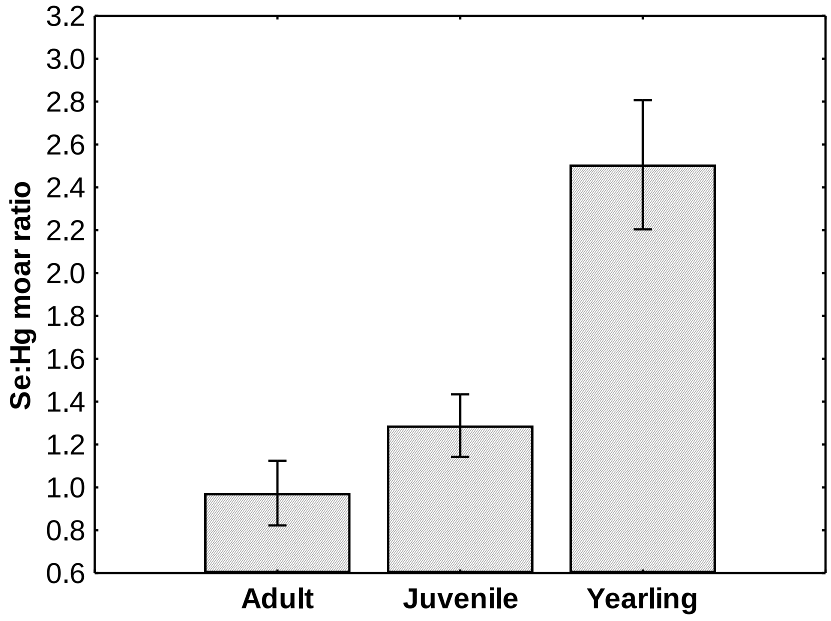

Figure 5: Mean ± standard deviation (SD) of Se:Hg molar ratios between different age classes of grey seals (*Halichoerus grypus*) from the Baltic Sea.

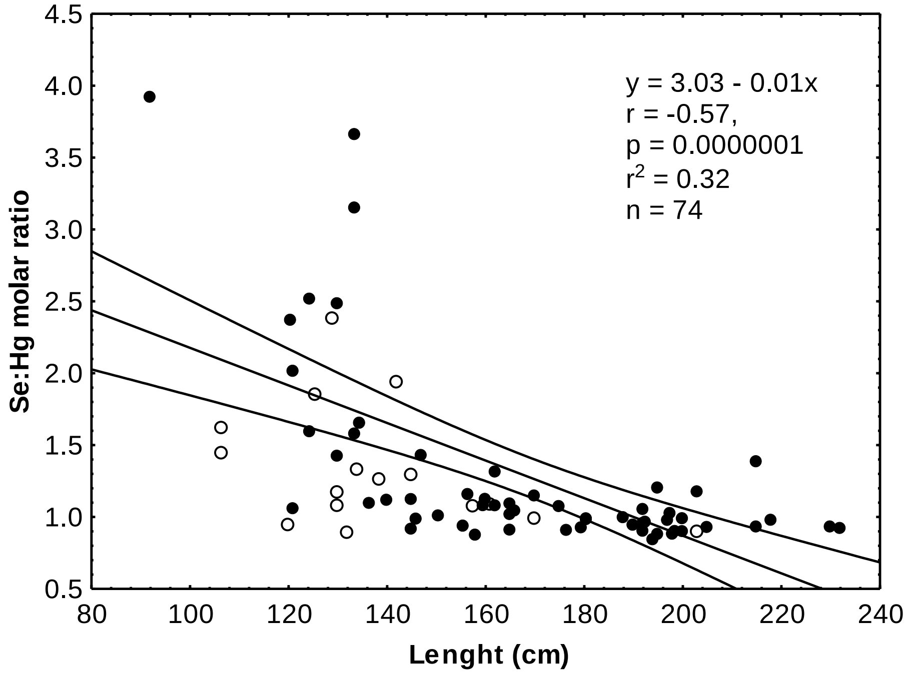

Figure 6: Se:Hg molar ratio versus the length (cm) in male (●) and female (○) grey seals (*Halichoerus grypus*) from the Baltic Sea. The straight line represents the correlation between the Se:Hg molar ratio and the length for all seals and curved lines denotes the 95% confidence interval.

3.2.3 Se:Hg molar ratio

The Hg:Se molar ratio ranged from 0.84 - 3.92. No differences was observed between the molar ratio of Se:Hg and geographical grouping of the seals ($F_{2,68} = 0.23$, $p = 0.95$). However, between age groups (**Figure 5),** yearlings (2.50 ± 0.86) were found to have significantly higher Se:Hg molar ratios than juveniles (1.32 ± 0.51) and adults (0.97 ± 0.11) (Tukey HSD; $p < 0.001$). Differences in Se:Hg between adults and juveniles were also significant (Tukey HSD; $p < 0.0004$). The close to 1:1 Se to Hg molar ratio in seals are presented in **Figure 6**. Se:Hg were negatively correlated with most of the REEs, Ag, Mo and V ($p < 0.01$) (**Appendix XI**).

3.2.4 Relationship between chemical elements, stable isotopes (C and N) and biometric data

Statistical significant relationships (Pearson`s linear correlation coefficient) between the concentrations of the chemical elements in hepatic tissue is presented in **Appendix X**. Several significant differences were observed and the strongest included Ag-Bi, B-Sr, Ba-Mn-Sb-Th-W, Bi-Sn, Ca-Sr, Ga-V, Hg-Se, Mg-P, Mn-Th-W, Mo-Zn, P-Mg and Sb-W ($p < 0.0001$). Between stable isotopes and chemical elements, strong correlations were found for δ^{15}N-Ag, δ^{15}N-Ni, δ^{15}N-Sn, δ^{13}C-Hg and δ^{13}C-Se ($p < 0.001$), and weaker for δ^{15}N against B, Cs, Ga, Sb, Sr, V and Zn ($p < 0.01$). All of the REEs correlated extremely well with each other ($r > 0.84$, $p < 0.001$) and length ($p < 0.005$).

3.3 Principial Component Analysis (PCA)

Because of numerous inter-relationships between the elements in the data set (see section 3.2.4), principal component analysis (PCA) was applied.

Principal component analysis of the hepatic concentrations of the elements resulted in a model ($R^2X = 0.656$, $Q^2 = 0.43$) (**Figure 7**) with 5 significant principal components (PCs) (eigenvalues > 1), explaining 36.2, 11.1 and 8.27, 6.38 and 3.67 % of the total variability, respectively. Thus, PC 1 described most of the variability of the data set.

In the loading plot, biometric variables grouped to the right together with several of the chemical elements including Ag, Bi, Cd, Ga, Hg, Mo, Se, Sn, V and the REEs along PC 1. Correlations between these elements and the biometric variables were confirmed by bivariate analysis ($p < 0.01$) (see **Appendix XI**). In the score plot (**Figure 8**), adults grouped to the right along PC 1, with juveniles and yearlings grouping towards the left. This indicates a gradually decreasing concentration of several elements as a function of age. Se:Hg was the only variable that was negatively correlated with age and most of the chemical element ($p < 0.05$) and were situated in the far left of the loading plot. Removing Se:Hg from the analysis did not affect the general patterns of the model. Stable isotopes and several of the essential macro- and micro-elements were situated in the central part of the loading plot, having average values for PC 1.

Table 2: Influence of geographical location on the metal concentration in grey seals from the Baltic Sea, adjusted for differences in size using length as covariate (ANCOVA). All data reported on log transformed data. Bold F-values indicate significant models.

	$F_{5,67}$-value	**p-value**		**$F_{5,67}$-value**	**p-value**
Trace Element					
Ag	1.11	0.37	V	2.11	0.08
As	**4.66**	0.001	W	1.97	0.09
B	**10.99**	< 0.0001	Zn	**2.53**	0.04
Ba	**3.39**	0.009			
Bi	0.52	0.76	**Macro Elements**		
Cd	2.17	0.07	Ca	**3.26**	0.01
Co	1.34	0.26	Fe	**2.83**	0.02
Cs	1.45	0.22	K	0.65	0.67
Cu	**5.12**	0.0005	Mg	1.30	0.27
Ga	**4.12**	0.003	Na	**4.43**	0.002
Ge	1.54	0.19	P	**4.41**	0.002
Hg	1.29	0.28	S	**3.02**	0.02
Li	**3.23**	0.01			
Mn	1.70	0.15	**Rare Earth Elements**		
Mo	1.79	0.13	Ce	1.16	0.34
Ni	**13.1**	< 0.0001	Dy	2.19	0.07
Pb	1.71	0.14	Er	1.65	0.16
Rb	**3.44**	0.008	Ho	0.94	0.46
Sb	**2.92**	0.02	La	0.97	0.44
Se	1.65	0.16	Lu	**2.35**	0.05
Se:Hg	0.23	0.95	Nd	1.09	0.37
Si	2.30	0.06	Pr	1.04	0.40
Sn	0.74	0.60	Sm	1.21	0.31
Sr	**10.00**	< 0.0001	Tb	1.59	0.18
Th	**3.83**	0.004	Y	1.68	0.15
Tl	**4.68**	0.001	Yb	**2.45**	0.04

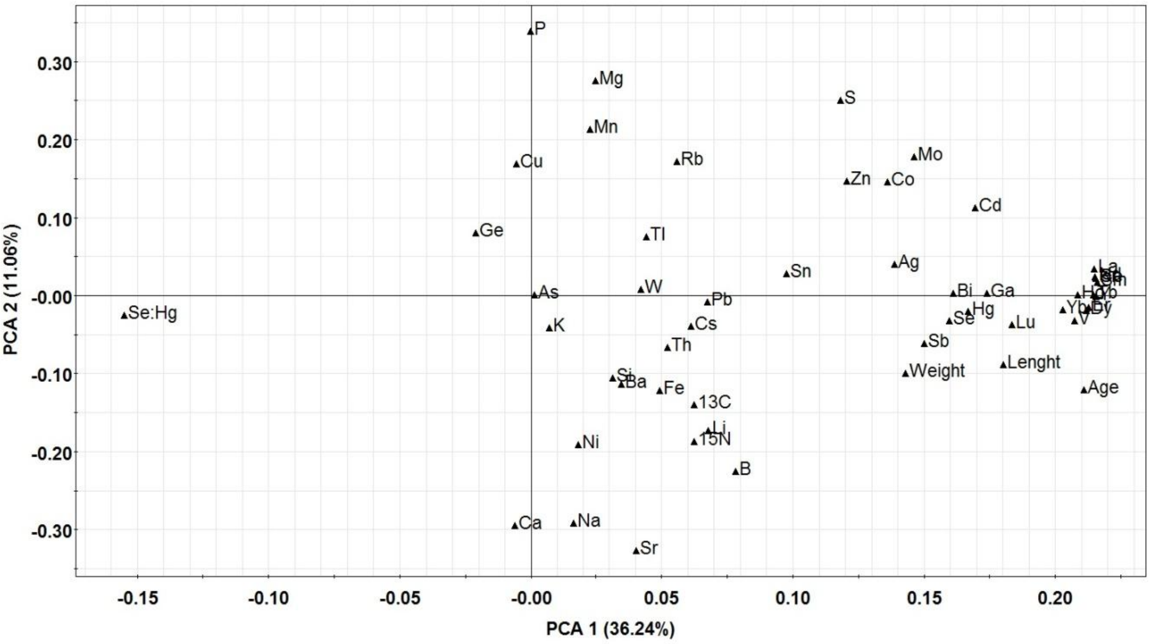

Figure 7: PCA loading plot including all chemical elements, stable isotope, biometric variables and Se:Hg molar ratio in liver samples for grey seals (*Halichoerus grypus*) from the Baltic Sea.

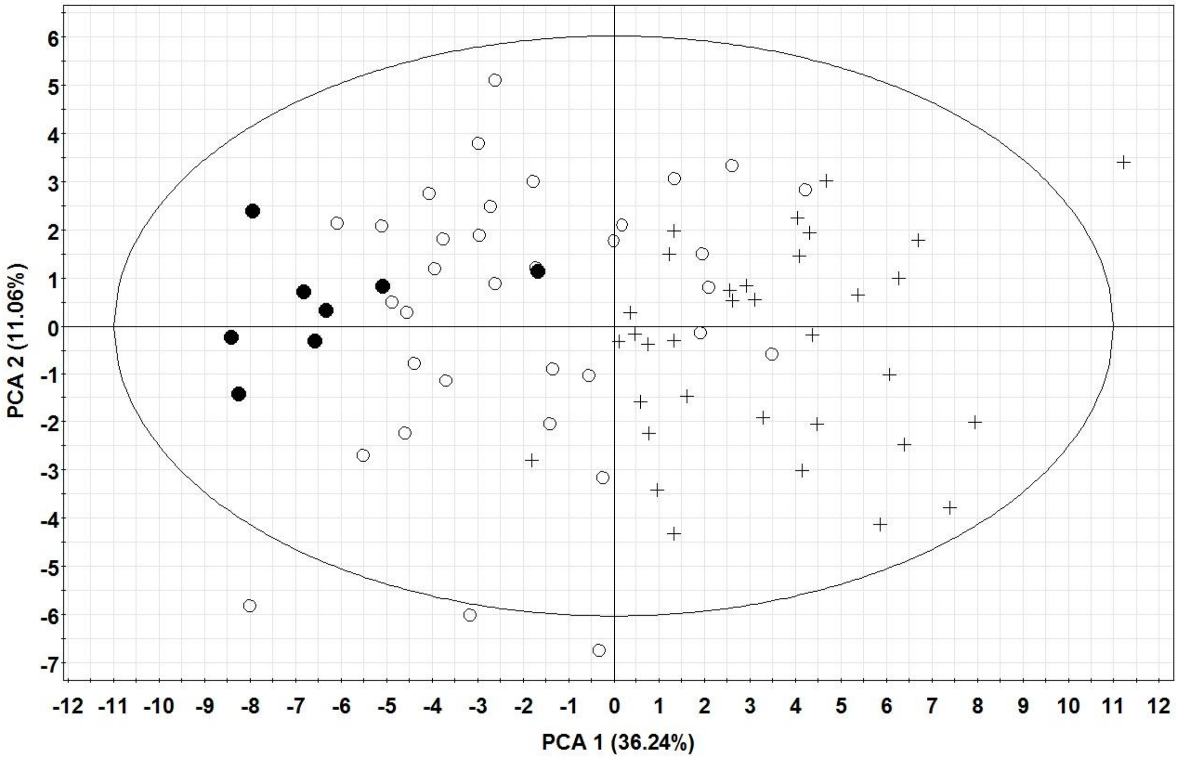

Figure 8: PCA Scoreplot of grey seals (*Halichoerus grypus*) from the Baltic Sea. The age groups are marked as adults (+), juveniles (○) and yearlings (●).

4. Discussion

Only a few studies have reported concentrations of chemical elements in marine mammals in relationship with stable isotopes. The present study is to our knowledge, the first to relate concentrations of a wide range of chemical elements in the Baltic grey seals to their trophic position investigated by C and N stable isotope analysis.

4.1 Stable isotopes

The stable isotope ratios ranged from -17.1 to -20.6 ‰ for $\delta^{13}C$ and 12.7 to 17.8 ‰ for $\delta^{15}N$. Four seal samples were treated as outliers (see explanation below). The values found in this study are in agreement with Svensson *et al.* (2007), reporting isotopic ratios of C and N in grey seals to range from -17.6 to -21.3 ‰ and 11.5 to 18.8 ‰, respectively. Nitrogen and C ratios of seals inhabiting other seas were found to be higher, which indicates that seals from other seas feed higher in the trophic pyramid compared to the Baltic population (Lesage and Hammill 2001; Van De Vijver *et al.* 2003).

Higher $\delta^{15}N$ hepatic values for seals from the southern areas of the Baltic, especially seals from the Polish coast indicate that these seals feed higher up in the food chain in comparison to other areas. Similarly, the heavier isotope of C was generally enriched in the southern areas relative to seals from the Bothnian Bay and Bothnian Sea, indicating that seals in the south feed more in the benthic food web (France 1995). In addition to herring and common whitefish, cod and flounder are important prey species for seals in the southern areas (Lundström *et al.* 2007). Both cod and flounder belong to the benthic food web. In the Gulf of Bothnia, herring and whitefish also dominates the seals diet, but also other pelagic fishes like salmon and trout are important (Lundström *et al.* 2007). Enrichment of ^{15}N in the seals from Polish coast suggests that they forage more on fish from higher trophic levels, e.g. cod. At the same time low hepatic values for $\delta^{13}C$ in seal from northern basins (Gulf of Bothnia) indicates more pelagic feeding preferences e.g. more herring and common white fish in their diet.

A clear difference in the carbon signature was observed between seals in the Gulf of Bothnia and the rest of the seals, indicating a “border” between different feeding habits. The sample size in the group from the Sea of Åland was relatively small and this group is situated between two of the most populated seal areas in the Baltic Sea. This could be because seals collected from this area are more associated with southern areas, thus not reflecting the actual patterns in the seals from the Sea of Åland. Furthermore, seals are known to travel from the

Sea of Åland to the Gulf of Bothnia (Sjöberg *et al.* 1995). Thus it is possible that seals from the sea of Åland occupy an intermediate position.

An interesting observation was a large difference in isotope ratios between seals from the closely located Polish coast and the Bornholm and Arcona basins (**Figure 1**). Seals from the latter seem to feed more on pelagic species, but also on species lower in the trophic pyramid. The water surrounding Bornholm and Arcona basins are generally more species rich than the Polish coast (Ojaveer *et al.* 2010), due to the higher salinity. Furthermore, limited grey seal reproduction is occurring below 58^o in the Baltic Sea. This implies that seals inhabiting the southern waters of Sweden and the Polish coast likely have migrated from nearby breeding colonies in the Baltic proper and Estonia, respectively. Thus, feeding patterns and experience may differ between closely located areas.

Differences were observed in nitrogen and carbon signature between adult, juvenile and yearling seals. Adult seals had higher isotopic ratios of both N and C, indicating that they feed higher in the trophic pyramid and are more associated with the benthic species compared to juveniles and especially yearlings. Younger seals have not developed fully and their hunting may be limited by reduced diving time, diving depth and experience. Thus, yearlings feed on smaller pelagic species while the juveniles and adults gradually shift towards larger prey and more benthic species.

Stable isotopes of C and N were observed to correlate with each other in the seals. In the marine environment, relationships between $\delta^{13}C$ and $\delta^{15}N$ have also been observed for harbor porpoise in the Baltic Sea and Kattegat (Angerbjörn *et al.* 2006) and also other species. However, little quantitative understanding exists on dietary patterns that create this covariation (Kelly 2000) and further investigations are required.

Four grey seals, no. 21, 46, 50 and 51, contained stable isotope ratios that considerably deviated from the rest of the seals. Seal no. 50, were collected from a bay (Bråviken) and thus could be feeding on different species compared to seals from more open waters. For seal no. 21 and 46 low ratio of $\delta^{13}C$ was found, indicating that these seals feed more on the pelagic species. For seal no. 46 (from the Baltic proper) this result seems to be an outlier when comparing to other seals from this area, feeding more in the benthic food web. A possible explanation could be the fact that individual seals can specialize on specific food items.

4.2 Geographical Differences

Bioaccumulation of chemical elements in tissues and organs of marine mammals depends both on concentrations in the surrounding environment (especially content in the food) and the homeostatic processes that govern the uptake, distribution and elimination of the elements. Arsenic, Cu, Zn, Rb, Tl, B, Ni and Sr were found to vary significantly between grey seals from different geographical location in the Baltic Sea (**Table 2**). When comparing results for spatial distribution of chemical elements in marine mammals it is important to take into consideration that grey seals have great mobility. Furthermore, they exhibit a wide seasonal migration range and have been reported to travel large distances in the Baltic Sea (Sjöberg *et al.* 1995). However, most often they are observed close to the haul-out site. Their home ranges have been reported to vary from 1088 - 6400 km^2 in the Baltic Sea (Sjöberg and Ball 2000). For seals inhabiting the closely located Bothnian Sea, Sea of Åland and Baltic proper, placing seals into distinct groups is difficult, given that seals may migrate between areas.

Low values of As were observed in seals from the Bothnian Bay, while highest in the Bothnian Sea and the Bornholm and Arcona basins. No previous reports have reported values for As in hepatic tissues of marine mammals in the Baltic Sea. Frank *et al.* (1992), reported mean values in blubber to range from 10.0 to 12 $\mu g \cdot g^{-1}$ dry weight in juvenile and adult grey seals, respectively. Recent estimates of As input to the Baltic Sea averages about 250 tons per year (Szefer 2002a). Historical sources of As input in the Baltic Sea can also be important, which include dumping of As containing chemicals (about 3400 tons) after the Second World War. Most of the dumping occurred in the southern parts of the Baltic, around Denmark and the Swedish west coast, but also in other areas and still is of concern (HELCOM 2010). Relatively high As concentrations in the sediments in the southern Baltic have also been reported (Szefer 2002b). This could be a possible explanation of the higher levels of As in seal liver from these areas. Furthermore, the more benthic feeding in the south, implies more interaction with the sediments, thus possibly increasing the assimilation of As from the sediments. Thus, the reason for the higher As concentrations in the southern Baltic may be linked to feeding more on benthic organisms. The lower concentrations of As of seals from the Bothnian Bay are in disagreement with sediment studies from the area, reporting relatively high concentrations for the entire Gulf of Bothnia (Leivuori and Niemistö 1995). Seals from the Bothnian Bay have lower $\delta^{13}C$ compared to other areas, indicating that the seals from this area are interacting more with the pelagic species. Thus, the lower As concentrations in these species could result from limited interactions with sediments.

The concentrations of both Cu and Zn are somewhat higher than previously reported from marine mammals of the Baltic Sea. We also observed spatial differences in the concentrations of Cu and Zn, but only significant differences between groups for Cu. The reason of lower Cu concentrations in seals from both the Bothnian Sea and Polish coast can be the higher age and thus, different feeding of the seals in these two areas.

Copper and Zn are essential elements in mammals and their concentrations are regulated mainly by low molecular light weight proteins called metallothioneins (MTs). High Cu and Zn concentrations have been observed in young animals and newborns (Wagemann *et al.* 1988; Caurant *et al.* 1994). These elements are also known to increase in concentration in rapidly developing tissues (Baer and Thomas 1991) and the differences have been proposed to either reflect some special requirement by young animals or low excretion rate of the fetus (Wagemann *et al.* 1988). For Zn, the concentrations increased with age in the seals. This is in contrast to Das *et al*. (2003b), reported that no apparent accumulation of Zn have been observed in tissues of marine mammals.

In this study concentrations of Cu in juvenile male seals were higher in comparison to adults and yearlings. This supports the hypothesis of rapidly developing organisms (juveniles) requiring a substantially higher supply of microelement, such as Cu. Another possible explanation of the higher Cu concentrations observed in juveniles males can be related to hormone homeostasis. In mammals methalotioneins (MT) plays an important role in the homeostatic regulation of metals, including Cu. Metallothioneins are a multi-regulated protein, mainly regulated by the presence of Zn, but also others factors are important, e.g hormon levels such as testosterone (Tohyama *et al.* 1996) and progesterone (Slater *et al.* 1988) and oxidative stress (Hernández *et al.* 2000). Hormonal imbalance in developing juvenile seals cannot be neglected, inducing or inhibiting MTs, resulting in different binding or excretion of Cu between the sexes

Two elements, Rb and Tl, were found to increase in concentration in seals from the south towards the north. Rb belongs to Group I of the periodic table and exhibit similar chemical properties as K and Cs. In mammals Rb is assumed to use the same transporters as K for entering cells (Gallacher *et al.* 1984). A study on goats, have raised speculations on whether or not Rb is an ultra trace essential element, as goats fed low Rb (< 0.28 $mg \cdot kg^{-1} \cdot day$) diet, revealed poor health relative to those fed high Rb diets (> 1 $mg \cdot kg^{-1} \cdot day$) diets (Nielsen 1998). The Rb content of the seals found here, were a little higher compared to long-finned pilot whales *(Globicephala melas)*, from the east-coast of USA (Mackey *et al.* 1995). The higher concentration of Rb in seals inhabiting the northern areas could be a result of higher concentrations in their food items. A survey of Finnish food in the 1980s reported higher values of Rb in fresh water fishes such as rainbow trout *(Salmo gairdnerii)* and whitefish (*Coregonus* sp.) compared to salt water fish (e.g. herring and cod) (Nuurtamo *et al.* 1980). Furthermore, no differences between the age groups were observed, indicating that the element could be actively taken up and homeostatically regulated in the seals. Samples of herring and stickleback analyzed here were depleted in Rb relative to the seals.

Even being regarded as a highly toxic element, studies on Tl is limited in contrast to mercury cadmium and lead. This is primarily because most analytical techniques have poor sensitivity for the element (John Peter and Viraraghavan 2005). No studies have reported concentrations for Tl for marine mammals in the Baltic Sea. The values found here are well within the range of what was found in striped dolphins (*Stenella coeruleoalba*) from Japan (Agusa *et al.* 2008) and Caspian Seals (*Phoca caspica*) from Russia (Anan *et al.* 2002).

Analysis of covariance revealed elevated concentrations of B and Ni in the seals from the southern Baltic (**Table 2**). For B and Ni, a gradual increase in concentrations from close to or below the detection limit in the north, to relatively high concentrations in the Polish coast was observed. The only exception from this latitudinal trend were seals from the Bornholm and Arcona basins for which both elements were found in relatively low concentrations. This trend reflects the pattern observed for the stable isotopes, where high $\delta^{15}N$ and $\delta^{13}C$ values were found in the seals from Polish coast but not from the Bornholm and Arcona basins. This finding confirms that dietary preferences could affect the element concentrations.

Studies on element concentrations in fish and sediments from the Baltic Sea reveals relatively high concentrations of Ni in the southern areas (Szefer 2002b). For B, very limited information is available in Baltic sediments, but a few studies indicate that sediments from southern areas exhibit relatively high concentrations of this element (Szefer 2002b). When comparing the concentrations of Ni and B in the sediments and biota between locations within the southern Baltic area (the Swedish and Polish coasts of the southern Baltic Sea) the literature data do not reflect the large differences observed between the seals from those two regions. However, the differences in C and N stable isotopes signature reflecting the variation in dietary patterns between seals from these closely located areas seem to better explain the differences in hepatic Ni and B concentrations. This was confirmed by the high levels of Ni, B, and δ^{15}N in the seals from the Polish coast.

In general the levels of Ni in marine mammals are low. Hepatic concentrations of Ni in grey and ringed seals from the Swedish and Polish coast were close to the detection limit (Frank *et al.* 1992; Ciesielski *et al.* 2006b). Similarly, Ni in the livers of 8 different species of marine mammals from waters around the British isles, were found to be below the method detection limit (Law *et al.* 1991). No physiological requirement for B in mammals have been found, although evidence of its accumulation suggests that B may be an essential nutrient (Eisler 2000). No studies on B have been performed on marine mammals from the Baltic Sea, but the concentrations in the liver reported herein are lower in comparison to harbour seals (*Phoca vitulina*) from the Arctic (Jenkins 1980). In the present study B was found to weakly, positively correlate with age. For both B and Ni the concentrations in the seal prey were relatively higher than the seals, especially for the *S. entomon*. This observation and the higher δ^{13}C in seals from the Polish coast indicate that feeding on the more benthic species could increase concentrations of these elements.

After adjusting for age of the seals, Sr revealed increasing concentrations from the northern groups towards the south, with the highest values found in the Bornholm and Arcona basins, followed by the Polish coast. Levels of Sr in the seal prey reveal concentrations more than 39, 220 and 1400 times higher than for the seals from both southern groups, for herring, stickleback and *S. entomon* respectively. However, these concentrations are based on analysis of the whole organisms including bones (fish) and exo-skeleton (*S. entomon*), thus it rather reflects the organ and tissue-specific bioaccumulation of the Sr in these organisms.

The increased concentrations of Sr found in the two southernmost groups of seals are likely due to higher levels of Sr in their diet then in the seals from northern Baltic. This observation does not reflect the differences found for stable isotopes of C and especially N between the areas. Strontium concentrations were slightly higher in seal liver samples from Bornholm and Arcona basins then in the Polish coast in contrast to stable isotopes ratios, which were higher in the Polish coast. The likely explanation for this is the fact that Sr is closely correlated with salinity of sea water (Kabata-Pendias and Pendias 1999). Thus, species inhabiting the more saline areas will have elevated concentrations of this element. The hepatic concentration of Sr in the liver match very well growing gradient of salinity in the Baltic sea, with the lowest salinity in Bothnian Bay (2 ‰) and the highest in the Bornholm and Arcona basins (8-10 ‰).

Information about Sr in biota is limited, in particular for marine mammals, given that the element is non-essential for all studied organisms. Furthermore, the toxicity of Sr in humans is low (Pors Nielsen 2004). From animal studies it appears that Sr can replace Ca in certain physiological processes and that Sr can be transported actively over biological membranes by the same transporters as Ca (Pors Nielsen 2004). In mammals most of the Sr present, is accumulated in bone (Dahl *et al.* 2001). Previously observed correlations between Sr-Ca-Ba (Ciesielski *et al.* 2006a; 2006b) were also found in this study confirming similar biogeochemical properties of these elements.

Other trace elements found to vary between geographical areas were Ba, Li, Sb and Th (Table 2). However, for these elements, differences between the areas were small or some concentrations of the element were below or close to the detection limits, thus any clear trends or relationships were not detectable.

4.3 Age Differences

Important factors for evaluation of spatial differences in chemical elements bioaccumulation in marine mammals is the developmental stage (adult, juveniles, yearlings) and sex of the seals. Differences in concentrations of several chemical elements between pups, adult females and males in Baikal seals (*Phoca sibirica*) were attributable to the reproductive cycle of this species (Ciesielski *et al.* 2006a; 2010). Thus both sex and age of the seals are important biological factors when evaluating bioaccumulation of chemical elements in marine mammals. However, sex differences were not observed in harbor porpoise inhabiting Baltic Sea (Ciesielski *et al.* 2006b). In this study sexual differences in the chemical elements bioaccumulation were not tested due to the low number of adult female grey seals (n = 2). It is also worth mentioning that for the interpretation of the results, age was approximated based on individuals length. Adjusting for the length of the seals using analysis of covariance, can bias the results because grey seals are characterized by large sexual dimorphism.

Concentrations of Hg reported here averaged 67 ± 124 (range 1.5 - 917) $\mu g \cdot g^{-1}$ dry weight. The observed differences between areas were not significant. Mercury concentrations reported by Frank *et al*. (1992) in the liver of adult animals from the southern parts of the Swedish coast were 85.8 (range 4,3 - 217), 145 (range 15.7 - 287) and 85.8 (range 75.9 - 303) $\mu g \cdot g^{-1}$ dry weight, for harbor porpoise, ringed seals and grey seals respectively. In comparison mean concentrations observed in this study are almost two times higher, for the same areas. In the before mentioned study (Frank *et al.* 1992), two diseased female grey seals had very high concentrations of Hg (1178 and 2409 $\mu g \cdot g^{-1}$ dry weight). In the study by Ciesielski *et al.* (2006b) hepatic concentration of Hg were 21.9 ± 56.4 (range 1.5 - 217), 2.6 ± 1.0 (range 1.84 - 3.3) and 113 ± 249 (range 0.6 - 557) $\mu g \cdot g^{-1}$ dry weight in harbor porpoise, ringed seals and grey seals, respectively. For grey seals, concentrations found herein are somewhat higher in the group from the Polish coast. Harbor porpoise from the coast of England and Wales that died from infectious diseases and physical trauma contained 66 and 41 $\mu g \cdot g^{-1}$ dry weight, respectively (Bennett *et al.* 2001). Other studies have also reported lower Hg concentrations, from Danish waters (Strand *et al.* 2005), the Norwegian coast (Teigen *et al.* 1993) and from the Black Sea (Joiris *et al.* 2001; Das *et al.* 2004). Grey seals sampled from the Bothnian Bay contained higher average concentrations of 257 ± 277 (range 50 - 1148) $\mu g \cdot g^{-1}$ dry weight (Nyman *et al.* 2002), than reported for that area in the present study. However, data reported by Nyman were for adult seals (mean of 10 < years) while seals from the Bothnian Bay investigated here were mainly juveniles. High concentrations of Hg were also found in grey seals from the Gulf of Finland (mean hepatic value of 329 (range 7 - 1626) $\mu g \cdot g^{-1}$ dry weight (mean of 10 < years) (Perttila *et al.* 1986).

An increase in Hg concentration with age was observed in seals from the Baltic Sea, in agreement with other studies on marine mammals (Frank *et al.* 1992; Das *et al.* 2003a; 2003b; Bustamante *et al.* 2004). Marine mammals assimilate Hg mainly from fish in the organic form of methyl-Hg (MeHg). Herring and stickleback analyzed here contained low values of Hg, with a mean 0.12 and 0.10 $\mu g \cdot g^{-1}$ dry weight respectively. In the benthic isopod *S. entomon* Hg values were not detectable. Mercury concentrations in fish and other biota are largely regulated by the water chemistry that controls the MeHg speciation and uptake at the beginning of the food chain (Mason *et al.* 1995). In its organic state, Hg is significantly more toxic than the inorganic. Thus, measurement of total Hg may be an improper indicator of toxic effects. In the liver of arctic marine mammals, MeHg have been reported to represent 3-12% of the total Hg (Wagemann *et al.* 1998), but it have been observed to decrease with age (Dehn *et al.* 2005). This high ratio of inorganic Hg in the livers of marine mammals is a result of MeHg demethylation, but it is also speculated that Se is involved in the demethylation step. It was found that Hg binds to selenides forming less toxic SeHg complexes (Khan and Wang 2009), and such granules were found in hepatic tissues in marine mammals (Nigro and Leonzio 1996; Arai *et al.* 2004). Moreover, we observed an excellent correlation between Se and Hg ($r = 0.99$, $p < 0.0001$), to our knowledge one of the highest ever observed for marine mammals. This is likely a detoxification mechanism in marine mammals that feed high in the trophic web, possibly explaining the near 1:1 molar ratio of Se:Hg found in these animals. This is also a possible explanation why individuals with very high Hg concentrations, does not exhibit severe toxic effects.

We observed the Se:Hg molar ratio of the seals to decrease strongly with age, from yearlings to adults. Similar findings have also been reported for other marine mammals. The molar ratio of Se:Hg in the liver of ringed, spotted (*Phoca largha*) and bearded (*Erignathus barbatus*) seals, collected from Alaska, were also found to have a strong negative correlation with age (Dehn *et al.* 2005). The fact that younger seals have higher concentrations of Se is likely to be either due to differences in feeding, or transfer of significantly higher concentrations of Se from mother to pups relative to Hg. A recent study by Habran *et al.* (2011), investigated the transfer of mercury and selenium from mother to pup during the lactation period for northern elephant seals (*Mirounga angustirostris*). They observed that maternal transfer of Se is most prominent during lactation, especially in the early stage, while Hg is mainly transferred during gestation. The transfer of Se, greatly increased the molar ratio of Se:Hg in the blood of the pups from 8 to 45, between day 5 and 22 of the lactation period respectively. No studies have been performed on chemical element content in the milk of grey seals, but the milk content (fat, protein and water) have shown similar trends in grey seals as in the northern elephant seals during lactation (Baker 1990).

Concentrations of Hg and Se were found to correlate positively with $\delta^{13}C$ for all seals, indicating that differences in feeding influence the assimilation of these elements. Moreover, seals from the southern areas were found to have higher concentrations of Hg and Se, indicating that feeding in the more benthic food web increase concentrations. However, a similar study by Das *et al*. (2003a) on marine mammals from the Northeast Atlantic identified no such relationship between the hepatic concentrations of Hg and $d^{13}C$.

The present study did not reveal any large spatial differences in Cd concentrations. However, seals from the Polish coast were observed to have somewhat lower concentrations. Moreover, Cd was observed to increase with increasing age. Age dependent accumulation of Cd has been observed in several studies, especially in renal tissue.

Concentrations reported here and in other studies on marine mammals from the Baltic Sea, have revealed low concentrations of Cd compared to Arctic and Atlantic animals. In ringed seals from Northwest Greenland, mean values for hepatic tissue of adult seals were 191.7 $\mu g \cdot g^{-1}$ dry weight, with the highest value exceeding 330 $\mu g \cdot g^{-1}$ dry weight (Dietz *et al.* 1998).

Even higher concentrations were found in renal tissues of the animals in the study, but no morphological changes were observed in relation to higher Cd levels. In these areas Cd levels are naturally high, forcing species inhabiting this environment to adapt to such conditions. In the Baltic Sea, concentrations of Cd are low, thus animals living here may not yet have adapted to elevated concentrations. Therefore, even small fluctuations in the Cd concentration in animals of the Baltic Sea could have negative effects.

Cadmium previously analyzed in grey seals from the Bothnian Bay are in the same range as reported here (Fant *et al.* 2001; Nyman *et al.* 2002). Significant differences in the concentration between males and females have also been observed, with females having higher concentrations (Nyman *et al.* 2002). For the grey seals along the Polish coast, concentrations herein are higher than in previous reports (Szefer 2002b; Ciesielski *et al.* 2006b). The main reason for the higher concentrations is likely the age. Seals in this study were older.

Tin concentrations in hepatic tissue of grey seals ranged from 0.006 to 13.9 $\mu g \cdot g^{-1}$ dry weight and an increase in concentration with age was confirmed. The use of Sn has mainly been in the organic form, as antifouling agent in agricultural biocides, antifouling paints for ship hulls, fish nets and also for other applications. Most studies on Sn have investigated the organic forms such as tributyltin (TBT) and phenyltin (PhT) compounds that have been found to induce e.g. embryotoxic, myotoxic and genotoxic effects in mammals (Boyer 1989; Kang *et al.* 1997; Chao *et al.* 1999). In marine mammals, the organo-Sn have been found to accumulate in the liver (Iwata *et al.* 1995; Kannan *et al.* 1996). However, the relatively high concentrations of inorganic Sn detected in hepatic tissue here, is in contrast to its low bioavailability (Rudel 2003). Thus it can be assumed that Sn was mainly taken up in organic form, through food and then metabolized into and stored in liver as inorganic Sn. *In vitro* dealkylation of TBT by the microsomal monooxygenase system eventually resulting in inorganic Sn has been observed in rats (Kimmel *et al.* 1977). Study of composition of butyltin compounds in samples of marine mammals form the southern Baltic Sea suggested possible metabolism of TBT in liver (Ciesielski *et al*. 2004). However, high amounts of Sn have also been observed in the fur of pinnipeds, thus shedding could be a possible route of removal (Tanabe 1999). Furthermore, in southern sea otters (*Enhydra lutris nereis*) from the coast of California, healthy animals were found to have higher levels of inorganic Sn compared to emaciated and infectious/diseased otters (Kannan *et al.* 2006). Thesefindings could indicate a possible biological function of Sn in marine mammals.

We observed all REEs to strongly accumulate with age in Baltic grey seals and small spatial differences for Lu and Yb. During the last decades the usage of REE has rapidly increased, in particular for agricultural (especially in China) and industrial purposes (Volokh *et al.* 1990; Maestro and Huguenin 1995). However, very limited information is available about their environmental impact, especially in the marine environment. There have been indications of REEs accumulating in experimentally exposed carp (*Cyprinus carpio* L.) (Qiang *et al.* 1994) and REEs have also been found to accumulate with age in human bone tissue (Zaichick *et al.* 2011). To our knowledge this is the first study to report a strong accumulation of REEs with age in marine mammals. On the other hand, numerous papers have investigated the use of REEs as possible growth enhancers in terrestrial mammals, but the findings are ambiguous (He *et al.* 2001; Kraatz *et al.* 2006). Further studies are needed for relating such data to effects in marine mammals. Our observations of lighter REEs accumulating stronger with age than the heavier ones, are also supported by studies on cultured HeLa S-3 cells (Tanaka *et al.* 1999).

The range of V (0.02 - 1.5 $\mu g \cdot g^{-1}$ dry weight) and the strong positive correlation with age observed here, is in concordance with previous reports (Frank *et al.* 1992; Mackey *et al.* 1996; Saeki *et al.* 1999). Currently the information on V in marine mammals is scarce. At the present there is limited information about whether or not V is an essential element in these animals. Several suggestions of biochemical and physiological functions have been made, including inhibition of (Na,K)-ATPase (Cantley *et al.* 1977), stimulation of cyclic AMP synthesis (Schwabe *et al.* 1979) and anti-carcinogenic actions (Thompson *et al.* 1984).

Silver was found to increase with age, but no distinction in concentrations could be made between areas. The element has seldom been investigated in the aquatic environment, especially for marine mammals. The only study on Ag in the Baltic Sea were conducted on juvenile harbor porpoises from the Polish coast, containing an average hepatic concentration more than 29 and 52 times higher than reported for adults and juveniles grey seals here, respectively (Szefer *et al.* 1995). Other studies on seals have confirmed low concentration in these animals (Saeki *et al.* 2001). This indicates that seals from the Baltic Sea and other areas, are able to effectively excrete Ag from their systems, compared to other marine mammals, e.g. cetaceans (Szefer *et al.* 1995; Mackey *et al.* 1996). We observed Ag to strongly correlate with both Hg and Se, in concordance with earlier studies (Saeki *et al.* 2001; Ikemoto *et al.* 2004).

To our knowledge no studies have reported concentrations of Bi in any biota from the Baltic Sea. Concentrations found here ranged from 0.008 - 4.9 $\mu g \cdot g^{-1}$ dry weight in the seals and an age dependent accumulation was observed. Fish and isopods had generally lower values of Bi compared to the seals. The concentrations found here are higher compared to marine mammals from other areas. Polar bears *(Ursus maritimus)* from two sub-populations in Alaska contained an average hepatic concentration of 0.002 and 0.006 (range 0.001 - 0.013) $\mu g \cdot g^{-1}$ dry weight (Kannan *et al.* 2007), while in serum and hair of two Antarctic seal species, Antarctic leopard seals *(Hydrurga leptonyx)* and Weddell seals *(Leptonychotes weddellii)* concentrations were found to be below the detection limit (0.001 $\mu g \cdot g^{-1}$ dry weight) (Gray *et al.* 2008). In livers of southern sea otters found dead along coast of California, concentrations ranged from less than 0.001 to 0.075 $\mu g \cdot g^{-1}$ dry weight (Kannan *et al.* 2006).

Water soluble Mo compounds are readily absorbed via the gastrointestinal tract from assimilated food and concentrated in the liver. We found relatively high concentrations of the element, ranging from 1.1 - 3.7 $\mu g \cdot g^{-1}$ dry weight, and its increasing concentrations with age. Concentrations are comparable to the liver of both Dall’s porpoises (*Phocoenoides dalli*) from Japan waters and stranded California sea lion (*Zalophus californianus*) that contained 3.6 and 1.3 $\mu g \cdot g^{-1}$ dry weight respectively. In the Baltic Sea, Mo have been found to increase in concentrations with increasing depth (Szefer 2002a). Thus, older animals more associated with the benthic environment will be interacting more with the element. High concentrations of Mo have also been found in the sediments surrounding the Bornholm basin (Szefer 2002a), but no significant differences in seal concentrations between the areas could be obtained here. Mo is an essential element in almost all investigated species, being a key element in several enzymes. Moreover, the element is not homeostatically regulated, thus low excretion of the element could result in Mo toxicity (Anke 2004).

Macro elements are frequently participating in biochemical and physiological processes. The uptake and excretion of these elements are closely regulated in most organisms by the process of homeostasis. Values reported for these elements reported here are in the range of previous studies of marine mammals from the Baltic Sea (Frank *et al.* 1992; Szefer 2002b; Ciesielski *et al.* 2006b). For the macro elements Ca, Fe, K, Mg, Na, P and S, no clear differences between areas or age groups of the grey seals inhabiting the Baltic Sea were observed. This is in agreement with the previously mentioned homeostatic regulation. This is in agreement with the previously mentioned homeostatic process.

Other elements found to increase with age in the seals were Co, Ga, Li and Sb (**Table 1**). For Co, Ga and Sb some of the concentrations were low and in a few seals not detectable. For Li seals from the Polish coast had somewhat higher concentrations but differences were small. The current information about these elements in marine mammals and the Baltic environment is limited.

5. Conclusion

The present study reports concentrations of a wide spectrum of chemical elements in grey seals form the Baltic Sea, including elements which were very rarely investigated in marine mammals previously. Analysis of C and N stable isotopes ratios were used to reveal dietary differences between seals from different areas of the Baltic Sea and furthermore relate to the chemical element bioaccumulation. Higher hepatic values of ^{15}N in samples of the seals from Polish coast indicates shift in diet preferences towards species occupying higher trophic position in the Baltic such as for instance cod. At the same time significantly lower values for δ^{13}C in seal from northern basins compared to more southern ones indicates more pelagic foraging, for example herring and common white fish prevalence in the diet. Spatial differences in metal bioaccumulation were found for As, B, Cu, Ni, Rb, Sr, Tl with higher concentrations for most of these elements in seals from the southern Baltic Sea. These differences were fairly well explained by δ^{15}N and δ^{13}C signatures for Ni and B, and by differences in water salinity for Sr. Geographical variation in concentration of As can be attributable to the differences in content of this element in surrounding environment (sediments and biota). Accumulation with age of the seals was found for Ag, B, Bi, Cd, Co, Hg, Mo, Sb, Se, Sn, V and Zn and the rare earth elements (REE). The role of Se in detoxification of Hg was confirmed by very strong significant correlation between these two elements ($r = 0.99$, $p < 0.0001$). Higher Se:Hg molar ratio were observed in yearlings compared to juvenile and adult seals. This suggests the prominent role of maternal transfer of Se during lactation and existence of possible protective mechanisms against Hg toxicity especially in the early stage of development. Several other elements also revealed statistically significant interrelationships, especially pronounced for the REEs.

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Appendix

Appendix I - Accuracy of the method

Table A1: Measured concentration in reference material (Bovine Liver) with standard deviation (SD), standard reference values (Bovine Liver 1577b) and recovery.

Element	Mass number	Concentration (µg/g)	SD	Certified value (µg/g)	Estimated uncertainty	Recovery (%)
Aluminium	27	0.862	0.308	1[a]		86
Antimony	121	0.00274	0.000307	0.003[a]		108
Arsenic	75	0.0435	0.00187	0.05[a]		87
Cadmium	111	0.483	0.00539	0.5	0.03	97
Cadmium	114	0.471	0.00328	0.5	0.03	94
Calcium	43	109	1.78	116	4	94
Cobalt	59	0.228	0.00716	0.25[a]		91
Cupper	63	149	11.3	160	8	93
Iron	57	176	3.77	184	15	96
Lead	208	0.112	0.00105	0.129	0.004	87
Magnesium	25	605	17.5	601	28	101
Manganese	55	10	0.11	10.5	1.7	96
Mercury	202	0.00316	0.00199	0.003[a]		105
Molybdenum	98	3.51	0.0623	3.5[a]		100
Phosphor	31	9810	90.4	11000	300	89
Potassium	39	11 200	480	9940	20	113
Rubidium	85	12,7	0.0636	13.7	1.1	93
Selenium	82	0.728	0.0194	0.73	0.06	100
Selenium	78	0.689	0.0445	0.73	0.06	94
Silver	109	0.0423	0.00313	0.039	0.007	108
Sodium	23	2340	67.7	2420	60	97
Strontium	88	0.132	0.00214	0.136	0.001	97
Sulphur	34	7700	117	7850	60	98
Vanadium	51	0.104	0.00517	0.123[a]		85
Zink	67	123	0.483	127	16	97

[a] Noncertified values are provided for information

Appendix II - Stable isotope ratios of carbon and nitrogen for grey seals

Table A2: Means ± standard deviation and range of $\delta^{15}N$ and $\delta^{13}C$ values from different groups and age classes of grey seals from the Baltic Sea

Region	Age class	n	$\delta^{15}N$	Range	$\delta^{13}C$	Range
Bothnian Bay	All	8	14.05 ± 0.76	13.12 – 15.67	-20.04 ± 1.00	-22.20 – -19.04
	Yearling	1	12.13		-20.41	
	Juvenile	6	13.93 ± 0.32	13.52 – 14.37	-20.15 ± 1.07	-22.20 – -19.04
	Adult	1	15.67		-19.04	
Bothnian Sea	All	24	13.99 ± 0.54	12.75 – 15.21	-19.50 ± 0.47	-20.56 – -18.42
	Yearling	5	13.43 ± 0.50	12.75 – 14.09	-19.89 ± 0.42	-20.55 – -19.41
	Juvenile	4	14.09 ± 0.24	13.73 – 14.24	-19.35 ± 0.62	-19.72 – -18.42
	Adult	15	14.15 ± 0.50	13.27 – 15.21	-19.41 ± 0.41	-20.56 – -18.87
Sea of Åland	All	8	14.25 ± 0.41	13.60 - 14.84	-18.89 ± 0.46	-19.46 – -18.13
	Juvenile	6	14.20 ± 0.39	13.60 – 14.73	-18.83 ± 0.51	-19.46 – -18.13
	Adult	2	14.39 ± 0.63	13.95 – 14.84	-19.08 ± 0.29	-19.29 – -18.88
Baltic proper	All	23	14.63 ± 1.22	13.37 – 19.10	-18.48 ± 1.23	-22.50 – -15.61
	Yearling	3	14.20 ± 0.32	13.98 – 14.58	-19.05 ± 0.37	-19.45 – -18.74
	Juvenile	10	14.45 ± 0.80	13.51 – 16.19	-18.74 ± 1.41	-22.50 – -17.19
	Adult	10	14.95 ± 1.66	13.37 – 19.10	-18.055 ± 1.14	-19.45 – -15.61
Polish coast	All	7	16.62 ± 1.04	15.25 – 17.88	-18.66 ± 1.12	-20.01 – -17.06
	Juvenile	4	16.07 ± 0.98	15.25 – 17.45	-19.42 ± 0.67	-20.01 – -18.68
	Adult	3	17.36 ± 0,60	16.71 – 17.88	-17.65 ± 0.62	-18.29 – -17.06
Bornholm and Arcona basins	All	5	15.00 ± 0.52	14.31 – 15.68	-18.89 ± 0.22	-19.15 – -18.67
	Juvenile	4	14.93 ± 0.58	14.31 –15.68	-18.94 ± 0.21	-19.15 – -18.67
	Adult	1	15.28		-18.67	

Appendix III - Stable isotope ratios of carbon and nitrogen for *S.entomon*, herring and stickleback

Table A3: Means ± standard deviation and range of $\delta^{15}N$ and $\delta^{13}C$ values for different species from the Gulf of Gdansk.

Species	n	$\delta^{15}N$	Range	$\delta^{13}C$	Min
Saduria entomon	4	8.27 ± 0.16	8.07 – 8.41	-18.76 ± 0.58	-19.49 – -18.13
Herring	5	10.09 ± 0.63	9.08 – 10.73	-21.20 ± 1.81	-24.41 – -20.16
Stickleback	2	8.90 ± 0.16	8.79 – 9.02	-20.85 ± 0.77	-21.40 – -20.31

Appendix IV - Biometric data

Table A4. Biometric data of sampled grey seals from the Baltic Sea

ID	Location	Found	Sex[a]	Age [years]	BW[b] [kg]	BL[c] [cm]	BT[d] (CR/AR/CB/AB/HB) [cm]
Group 1							
2	Luleå	02.08.2009	M	6	125	200	5/-/5/-/-
18	Luleå	03.07.2008	M	0	40	92	3/-/3/-/-
21	Haparanda	13.07.2008	M	1	52	146	3/-/3/-/-
24	Haparanda	11.07.2008	M	2	68	175	2.8/-/2.8/-/-
28	Haparanda	13.07.2008	M	2	54	165	2.5/-/2.5/-/-
29	Luleå	14.05.2009	F	2	70	130	5/-/5/-/-
30	Luleå	10.09.2009	M	2	70	170	3/-/3/-/-
31	Luleå	22.05.2009	F	nm[e]	70	145	2.5/-/2.5/-/-
32	Luleå	22.05.2009	F	nm	60	130	1.5/-/1.5/-/-
Group 2							
1	Hudiksvall	09.11.2009	M	6	134.9	179.5	3.8/3.5/3.8/3.5/2.8
3	Hudiksvall	25.05.2009	M	nm	180	200	5/-/5/-/-
4	Björkön	11.07.2009	M	12	123	197.5	3.2/3/3.2/3/1.2
6	Brämön	23.04.2009	M	17	117	185.5	1.5/2/1.5/2/1.8
7	Brämön	22.04.2009	M	nm	128.6	194	2.5/3/2.5/3/2.8
8	Bottenhavet	19.04.2009	M	16	114.5	188	1.4/2.2/1.4/2.2/2
9	Gävle	28.01.2009	M	12	173.7	192.5	4.5/4/4.5/4/4.7
10	Stocka	19.12.2008	M	nm	116.8	180.5	3.6/3.7/3.6/3.7/3.8
11	Ljusne	14.01.2009	M	15	137	198.5	2.8/3/2.8/3/2.7
12	Ljusne	14.01.2009	M	13	169.2	194	4/4/4/4/4.4
13	Bottenhavet	06.08.2008	M	20	154	192	6/-/6/-/-
14	Korvgrund	10.05.2008	M	14	141	195	3.5/-/3.5/-/-
15	Stocka	13.11.2008	M	20	138	192	6/-/6/-/-
16	Söderhamn	06.02.2008	M	16	227.6	203	4.8/-/4.8/-/-
17	Funngrunden	11.06.2009	M		39.4	109	2/2.5/2/2.5/2.3
19	Norrsundet	09.06.2009	M		34	116	2.4/2/2.4/2/1.9
20	Hudiksvall	17.06.2008	M		41.6	120.5	2.8/-/2.8/-/-
22	Söderhamn	01.05.2008	M	9	120	190	3/-/3/-/-
23	Norrsundet	09.06.2009	F	nm	25.9	106.5	1.7/1.5/1.7/1.5/1.6
25	Korvgrund	24.04.2008	F	2	55	132	3/-/3/-/-
26	VingÅland	13.02.2008	M	1	63.3	133.5	nm
27	Söderhamn	12.04.2008	M	1	66.7	147	2.7/2.7/2.7/2.7/2.5
35	Eggegrund	16.12.2008	M	11	158.3	192	3.6/5/3.6/5/4.4
37	Furuvik, Gävle	17.06.2009	M	9	158.9	192	3/2.5/3/2.5/3
44	Korsholmen	14.08.2009	F	nm	40	129	3/-/3/-/-
70	–	18.04.2009	M	nm	92	158	
Group 3							
33	Östhammar	07.11.2009	M	8	180	165	4.5/-/4.5/-/-
34	Östhammar	02.05.2009	M	nm	260	232	4.8/-/4.8/-/-
38	Missing	31.08.2009	M	nm	61	145	2.5/2.5/2.5/2.5/2.6
39	Väddö	02.10.2009	M	2	nm	150.5	3.3/3.5/3.3/3.5/3.5
40	–	07.09.2009	M	nm	81.9	159.6	2.8/2.5/2.8/2.5/2.5
41	Horssten	02.05.2009	–	1	50	130	6/-/6/-/-
42	Själkobbarna	16.04.2009	M	1	60	140	4.5/-/4.5/-/-
43	Grönskär	16.04.2009	F	1	50	142	3/-/3/-/-
Group 4							

5	Grönö	11.09.2009	F	3	83.7	161	2.7/3/2.7/3/2.3
36	Flatlogen	28.06.2009	M	6	150	195	2.5/-/2.5/-/-
45	Figeholm	01.08.2009	M	nm	122.9	180.5	2/2.7/2/2.7/1.8
46	Figeholm	09.10.2009	M	1	60.3	145	2.4/2.5/2.4/2.5/2.3
48	–	21.11.2008	M	5	160	215	4/-/4/-/-
49	–	06.10.2008	M	10	200	230	5/-/5/-/-
50	Bråviken	25.01.2009	M	14	217	215	5/4/5/4/4.8
51	Oskarshamn	09.11.2008	M	26	250	198	4.5/-/4.5/-/-
52	Flatlogen	04.05.2008	M	3	110	165	4/-/4/-/-
53	Fårö, Gotland	11.04.2009	M	nm	180	218	nm
54	Stora Askö	05.09.2008	M	3	76.8	162	2.5/-/2.5/-/-
55	Stora Askö	06.09.2008	M	3	87.8	176.5	2.2/-/2.2/-/-
56	Stora Örskär	06.10.2008	M	4	72	155.5	1.8/2.6/1.8/2.6/2.2
57	Mönsterås	16.10.2008	F	3	72	157.5	2.8/2.8/2.8/2.8/2.8
58	Söreskäret	24.09.2008	M	1	61.3	136.5	2.7/2.7/2.7/2.7/2.7
59	Loftahammar	17.08.2008	M	0	40.6	124.4	2/1.8/2/1.8/1.8
62	Furillen	08.06.2009	M	nm	39.6	121	3/3/3/3/3.1
63	Herrvik	10.12.2007	F	nm	47.1	134	2.5/-/2.5/-/-
64	Stora Askö	24.07.2008	M	3	71.7	156.5	2.3/-/2.3/-/-
68	–	16.09.2009	F	nm	51.6	125.5	nm
69	–	16.04.2010	F	nm	50	150	nm
71	–	16.05.2010	F	nm	120	170	nm
72	–	16.04.2010	M	nm	140	205	nm
Group 5							
73	Westerplatte	14.04.2006	M	nm	89	203	nm
74	Wladysławowo	02.02.2007	F	nm	66.8	106.5	nm
75	Hel	27.03.2007	F	nm	106	188	nm
76	Czołpino	08.07.2008	F	nm	46.2	130	nm
77	Hel	27.11.2008	M	nm	88.9	181	nm
78	Kołobrzeg	24.04.2009	M	nm	78.9	162	nm
79	Dziwnów	29.06.2009	M	nm	40.9	133.5	nm
Group 6							
47	Lomma	27.09.2008	M	nm	109.8	197	1.3/1.3/1.3/1.3/1.5
60	–	13.01.2010	M	nm	85.4	166	1.3/1.9/1.3/1.9/2.4
61	Karlskrona	03.12.2009	M	nm	48.6	134.5	2.6/2.4/2.6/2.4/1.8
65	Smygehuk	01.01.2009	F	1	50.6	138.5	2/1.5/2/1.5/1.7
66	Beddingestrand	10.01.2008	M	3	84.4	160	1.3/2.3/1.3/2.3/2

[a] M, Male; F, Female

[b] Body weight

[c] Body length

[d] Blubber thickness (CR, chest right-side; AR, abdomen right-side; CB, chest back; AB; abdomen back; HB, hip back)

[e] Not measured/missing data

Appendix V - Method detection limits

Table A5: Detection limits used for all chemical elements.

Element	Detection Limit (µg/g)	Element	Detection Limit (µg/g)
Ag	0.003	Nb	0.0003
Al	0.8	Nd	0.0012
As	0.0038	Ni	0.007
B	0.012	P	0.06
Ba	0.002	Pb	0.002
Be	0.0008	Pr	0.00005
Bi	0.00015	Rb	0.0018
Ca	1.5	S	3
Cd	0.0003	Sb	0.0003
Ce	0.00003	Sc	0.0006
Co	0.0006	Se[a]	0.023
Cr	0.011	Se[b]	0.008
Cs	0.00008	Si	0.6
Cu	0.003	Sm	0.000075
Dy	0.0003	Sn	0.00015
Er	0.000045	Sr	0.0038
Fe	2.3	Ta	0.00004
Ga	0.0011	Tb	0.00003
Ge	0.003	Th	0.000075
Hf	0.0004	Ti	0.09
Hg	0.048	Tl	0.00004
Ho	0.00002	Tm	0.00008
K	0.75	U	0.00004
La	0.0003	V	0.00045
Li	0.0045	W	0.00015
Lu	0.00003	Y	0.0002
Mg	0.053	Yb	0.00006
Mn	0.0009	Zn	0.006
Mo	0.003	Zr	0.004
Na	1.5		

[a] Se78 used for herring, stickleback and *Saduria entomon*

[b] Se82 used for grey seals

Appendix VI - Concentration of chemical elements in selected species from the Baltic Sea.

Table A6: Concentrations of metals in μg·g^{-1} dry weight, mean ± standard deviation and range in parentheses in herring, S. entomon and stickleback from the Polish coast, Baltic Sea.

Element	Herring (*C. harengus)* [n=20]	*Saduria entomon* [n=11][a]	Stickleback (*G. aculeatus*) [n=2]
Ag	0.00746 ± 0.00418 (0.00150 - 0.0169)	0.729 ± 0.179 (0.555 - 1.20)	0.0861 ± 0.0633 (0.0413 - 0.131)
Al	70.5 ± 140 (8.04 - 630)	365 ± 283 (176 - 1110)	97.9 ± 72.1 (46.8 - 149)
As	3.58 ± 1.09 (1.81 - 5.43)	4.36 ± 1.16 (2.74 - 6.50)	2.06 ± 0.170 (1.94 - 2.18)
B	0.729 ± 0.283 (0.339 - 1.28)	1.92 ± 0.515 (1.40 - 3.04)	1.93 ± 0.66 (1.46 - 2.40)
Ba	2.06 ± 1.79 (0.341 - 8.38)	48.0 ± 10.1 (37.6 - 73.7)	13.8 ± 3.54 (11.3 - 16.3)
Be	0.00269 ± 0.00520 (0.000400 - 0.0238)	0.0143 ± 0.00939 (0.00675 - 0.0383)	0.00287 ± 0.00172 (0.00165 - 0.00409)
Bi	0.00401 ± 0.00204 (0.00147 - 0.00730)	0.0290 ± 0.0124 (0.0123 - 0.0564)	0.00253 ± 0.000646 (0.00207 - 0.00298)
Ca	20300 ± 7770 (6780 - 39600)	97400 ± 18900 (80100 - 145000)	65800 ± 652 (65300 - 66200)
Cd	0.125 ± 0.0400 (0.0636 - 0.231)	0.770 ± 0.216 (0.435 - 1.10)	0.270 ± 0.0246 (0.252 - 0.287)
Ce	0.138 ± 0.324 (0.0115 - 1.47)	0.680 ± 0.458 (0.290 - 1.91)	0.0769 ± 0.0426 (0.0468 - 0.107)
Co	0.0655 ± 0.0893 (0.0237 - 0.433)	2.22 ± 1.06 (1.20 - 4.95)	0.105 ± 0.0393 (0.0771 - 0.133)
Cr	0.179 ± 0.255 (0.0349 - 1.20)	1.39 ± 1.51 (0.424 - 5.64)	0.178 ± 0.0406 (0.149 - 0.207)
Cs	0.025 ± 0.00576 (0.0173 - 0.0424)	0.0330 ± 0.0242 (0.0181 - 0.103)	0.0401 ± 0.0139 (0.0303 - 0.0499)
Cu	2.94 ± 0.942 (1.74 - 5.00)	77.5 ± 23.0 (44.8 - 134)	3.73 ± 0.175 (3.61 - 3.85)
Dy	0.00597 ± 0.0143 (0.000489 - 0.0647)	0.0232 ± 0.0122 (0.0116 - 0.0489)	0.00258 ± 0.0000305 (0.00256 - 0.00260)
Er	0.00602 ± 0.0145 (0.000383 - 0.0653)	0.0221 ± 0.0104 (0.0125 - 0.0452)	0.00227 ± 0.0000260 (0.00225 - 0.00229)
Fe	183 ± 253 (62.2 - 1200)	554 ± 234 (259 - 1080)	124 ± 5.47 (120 - 128)
Ga	0.0201 ± 0.0413 (0.00217 - 0.187)	0.0933 ± 0.0739 (0.0422 - 0.294)	0.0204 ± 0.0158 (0.00926 - 0.0315)
Ge	0.00782 ± 0.00368 (0.00335 - 0.0205)	0.0150 ± 0.00855 (0.00778 - 0.0329)	0.00855 ± 0.000366 (0.00829 - 0.00881)
Hf		0.0562 ± 0.0714 (0.00865 - 0.224)	0.0396 ± 0.0348 (0.0150 - 0.0642)
Hg	0.123 ± 0.0974 (0.0240 - 0.413)		0.0966 ± 0.00845 (0.0907 - 0.103)
Ho	0.00205 ± 0.00491 (0.000161 - 0.0222)	0.00752 ± 0.00359 (0.00415 - 0.0147)	0.000906 ± 0.0000242 (0.000889 - 0.000923)
K	22500 ± 12100 (10400 - 47800)	16300 ± 5380 (6240 - 29200)	22700 ± 2780 (20700 - 24700)
La	0.0731 ± 0.164 (0.00663 - 0.748)	0.385 ± 0.231 (0.177 – 1.00)	0.0417 ± 0.0246 (0.0243 - 0.0591)
Li	0.0667 ± 0.0567 (0.0211 - 0.297)	0.303 ± 0.192 (0.167 - 0.839)	0.100
Lu	0.000767 ± 0.00180 (0.0000651 - 0.00810)	0.00273 ± 0.00126 (0.00166 - 0.00582)	0.000282 ± 0.0000285 (0.000262 - 0.000302)
Mg	1170 ± 153 (892 - 1460)	4610 ± 804 (3790 - 6410)	2300 ± 191 (2170 - 2440)
Mn	20.9 ± 24.0 (3.15 - 113)	259 ± 135 (143 - 584)	45.1 ± 4.22 (42.2 - 48.1)
Mo	0.0415 ± 0.0219 (0.0207 - 0.125)	0.363 ± 0.118 (0.259 - 0.677)	0.0856 ± 0.0297 (0.0646 - 0.107)

Na	3850 ± 1230	10100 ± 3050	9510 ± 2200
	(1940 - 6310)	(7750 - 18900)	(7950 - 11100)
Nb	0.00754 ± 0.00775	0.0527 ± 0.0451	0.00601 ± 0.000979
	(0.00144 - 0.0280)	(0.0256 - 0.182)	(0.00532 - 0.00670)
Nd	0.0664 ± 0.158	0.308 ± 0.207	0.0333 ± 0.0155
	(0.00537 - 0.717)	(0.135 - 0.862)	(0.0223 - 0.0442)
Ni	0.165 ± 0.114	1.35 ± 0.788	0.273 ± 0.0141
	(0.0841 - 0.622)	(0.515 - 3.41)	(0.263 - 0.283)
P	15000 ± 2670	6440 ± 1320	32500 ± 480
	(11500 - 20700)	(5520 - 10300)	(32100 - 32800)
Pb	2.87 ± 5.73	0.496 ± 0.246	0.280 ± 0.136
	(0.115 - 19.8)	(0.218 - 1.06)	(0.184 - 0.376)
Pr	0.0172 ± 0.0405	0.0830 ± 0.0545	0.00882 ± 0.00442
	(0.00137 - 0.184)	(0.0363 - 0.228)	(0.00570 - 0.0119)
Rb	2.52 ± 0.473	2.65 ± 0.953	3.06 ± 0.326
	(1.56 - 3.42)	(1.96 - 5.33)	(2.83 - 3.29)
S	7750 ± 1090	8280 ± 2500	7510 ± 207
	(5810 - 9910)	(5890 - 15200)	(7370 - 7660)
Sb	0.00911 ± 0.0154	0.0149 ± 0.00881	0.00938 ± 0.000244
	(0.00103 - 0.0652)	(0.00777 - 0.0391)	(0.00921 - 0.00956)
Sc	0.0209 ± 0.0502	0.0687 ± 0.0531	0.00942 ± 0.00238
	(0.00180 - 0.226)	(0.0356 - 0.215)	(0.00774 - 0.0111)
Se	1.08 ± 0.167	1.44 ± 0.464	1.71 ± 0.474
	(0.760 - 1.38)	(0.939 - 2.69)	(1.38 - 2.05)
Si	216 ± 382	1210 ± 703	333 ± 220
	(22.3 - 1720)	(668 - 3030)	(177 - 488)
Sm	0.0129 ± 0.0303	0.0588 ± 0.0396	0.00676 ± 0.00208
	(0.00109 - 0.138)	(0.0254 - 0.163)	(0.00528 - 0.00823)
Sn	0.0271 ± 0.0295	0.0725 ± 0.118	0.0193 ± 0.00177
	(0.00176 - 0.135)	(0.0184 - 0.416)	(0.0180 - 0.0205)
Sr	28.7 ± 11.9	1040 ± 208	166 ± 14.3
	(7.22 - 53.1)	(882 - 1550)	(156 - 176)
Ta	0.000172 ± 0.000116	0.00368 ± 0.00275	0.000302 ± 0.0000462
	(0.0000460 - 0.000494)	(0.00183 - 0.0116)	(0.000269 - 0.000334)
Tb	0.00188 ± 0.00447	0.00823 ± 0.00459	0.00100 ± 0.000299
	(0.000129 - 0.0202)	(0.00403 - 0.0193)	(0.000789 - 0.00121)
Th	0.0138 ± 0.0276	0.0841 ± 0.0853	0.00857 ± 0.00398
	(0.000978 - 0.124)	(0.028 - 0.327)	(0.00576 - 0.0114)
Ti	4.88 ± 9.90	16.4 ± 13.3	1.79 ± 0.514
	(0.660 - 44.2)	(7.86 - 54.0)	(1.43 - 2.16)
Tl	0.00607 ± 0.00170	0.0185 ± 0.00759	0.0180 ± 0.00233
	(0.00386 - 0.00985)	(0.0136 - 0.0399)	(0.0163 - 0.0196)
Tm	0.000813 ± 0.00197	0.00279 ± 0.00128	0.000321 ± 0.0000142
	(0.0000400 - 0.00891)	(0.0016 - 0.00585)	(0.000311 - 0.000331)
U	0.0235 ± 0.0219	0.0853 ± 0.0409	0.0289 ± 0.0129
	(0.00456 - 0.106)	(0.0453 - 0.177)	(0.0198 - 0.0380)
V	0.207 ± 0.423	0.638 ± 0.386	0.163 ± 0.101
	(0.0305 - 1.93)	(0.409 - 1.74)	(0.0914 - 0.234)
W	0.00608 ± 0.0102	0.0123 ± 0.00803	0.00237 ± 0.000115
	(0.00150 - 0.0491)	(0.00708 - 0.0352)	(0.00228 - 0.00245)
Y	0.138 ± 0.154	0.344 ± 0.264	0.273 ± 0.172
	(0.0532 - 0.720)	(0.131 - 0.991)	(0.151 - 0.395)
Yb	0.00524 ± 0.0126	0.0184 ± 0.00784	0.00218 ± 0.0000481
	(0.000340 - 0.0571)	(0.0107 - 0.0369)	(0.00214 - 0.00221)
Zn	120 ± 29.3	69.8 ± 17.6	318 ± 36.1
	(57.1 - 155)	(55.3 - 120)	(293 - 344)
Zr	0.0155 ± 0.0222	1.96 ± 2.64	1.98 ± 1.66
	(0.00200 - 0.0876)	(0.251 - 8.14)	(0.804 - 3.15)

[a] Pooled into 11 samples

Appendix VII - Macro element concentration in grey seal livers

Table A7: Concentrations of macro elements in µg·g^{-1} dry weight, mean ± standard deviation and range in parentheses in grey seal (Halichoerus grypus) livers (n=76) from the Baltic Sea

Group	N	Age	Ca	Fe	K	Mg	Na	P	S
1	1	Adult	128	736	9230	580	3230	8750	9940
	7	Juvenile	268 ± 278	1130 ± 284	9800 ± 2430	608 ± 88.0	4040 ± 1480	9060 ± 1290	10100 ± 618
			(124 - 893)	(557 - 1440)	(6880 - 13900)	(423 - 692)	(2340 - 6520)	(6520 - 10800)	(8850 - 10500)
	1	Yearling	212	635	9720	587	4640	8590	9330
2	17	Adult	172 ± 20.6	952 ± 334	8920 ± 1300	583 ± 66.6	4130 ± 367	8460 ± 796	10400 ± 792
			(144 - 208)	(400 - 1710)	(6970 - 12000)	(476 - 697)	(3530 - 5000)	(7260 - 9780)	(8090 - 11600)
	4	Juvenile	197 ± 62.2	472 ± 133	9580 ± 1870	665 ± 36.6	3440 ± 615	9570 ± 799	10200 ± 638
			(145 - 287)	(275 - 568)	(6910 - 11200)	(627 - 712)	(2550 - 3940)	(8720 - 10600)	(9610 - 11100)
	5	Yearling	176 ± 43.7	882 ± 196	9230 ± 1830	651 ± 66.3	3950 ± 573	9280 ± 748	9610 ± 576
			(128 - 246)	(702 - 1190)	(6160 - 10500)	(568 - 753)	(3450 - 4720)	(8370 - 10400)	(9000 - 10300)
3	2	Adult	214 ± 91.4	872 ± 592	9090 ± 2290	596 ± 124	5440 ± 3830	8880 ± 2070	10500 ± 1560
			(149 - 278)	(453 - 1290)	(7470 - 10700)	(508 - 684)	(2730 - 8150)	(7410 - 10300)	(9440 - 11600)
	6	Juvenile	151 ± 18.3	966 ± 321	9420 ± 1770	630 ± 76.0	3840 ± 932	9220 ± 1370	10300 ± 208
			(130 - 178)	(486 - 1340)	(6420 - 11100)	(536 - 746)	(2640 - 5260)	(7660 - 11000)	(10100 - 10700)
4	10	Adult	169 ± 18.7	1090 ± 471	10400 ± 1300	612 ± 89.2	4410 ± 939	8720 ± 1330	10400 ± 1340
			(148 - 216)	(391 - 2000)	(8010 - 12700)	(463 - 724)	(2790 - 5700)	(6770 - 10500)	(8500 - 12600)
	11	Juvenile*	188 ± 76.7	1010 ± 457	8180 ± 1000	597 ± 51.0	4660 ± 1240	8720 ± 624	9960 ± 805
			(135 - 411)	(290 - 1920)	(6990 - 10000)	(539 - 692)	(3810 - 8170)	(7920 - 9840)	(8650 - 11600)
	2	Yearling	166 ± 9.70	611 ± 132	8900 ± 2600	584 ± 26.0	4640 ± 15.6	8360 ± 496	9540 ± 191
			(159 - 173)	(518 - 705)	(7060 - 10700)	(565 - 602)	(4620 - 4650)	(8010 - 8710)	(9410 - 9680)
5	3	Adult	291 ± 16.9	1850 ± 123	10000 ± 1980	660 ± 70.8	6160 ± 1340	8470 ± 903	10200 ± 116
			(278 - 310)	(1720 - 1970)	(7950 - 11900)	(602 - 739)	(4790 - 7450)	(7650 - 9440)	(10100 - 10300)
	4	Juvenile	224 ± 44.4	1080 ± 267	10100 ± 1400	573 ± 23.0	4900 ± 288	8190 ± 673	9420 ± 609
			(193 - 288)	(830 - 1410)	(8130 - 11300)	(545 - 598)	(4570 - 5180)	(7360 - 8990)	(8690 - 9980)
6	1	Adult	207	1340	10400	592	4110	6650	10300
	4	Juvenile*	355 ± 215	1110 ± 205	9400 ± 2000	509 ± 51.3	4640 ± 807	7150 ± 782	8830 ± 393
			(149 - 558)	(898 - 1300)	(7350 - 12100)	(448 - 553)	(3940 - 5560)	(6190 - 7830)	(8390 - 9270)

* Groups containing samples that are likely contaminated

Appendix VIII - Rare earth element concentrations in grey seal livers

Table A8: Concentrations of rare earth elements in µg·g^{-1} dry weight, mean ± standard deviation and range in parentheses in grey seal (Halichoerus grypus) livers (n=76) from the Baltic Sea

Group	**N**	**Age**	**Ce**	**Dy**	**Er**	**Ho**	**La**	**Lu**	**Nd**
1	1	Adult	42.1	0.449	0.400	0.156	24.4	0.0307	17.5
	7	Juvenile	36.2 ± 26.5 (4.90 - 73.6)	0.394 ± 0.310 (0.150 - 0.845)	0.384 ± 0.291 (0.0494 - 0.794)	0.123 ± 0.0854 (0.0100 - 0.241)	20.3 ± 14.1 (4.22 - 41.2)	0.0380 ± 0.0341 (0.0150 - 0.0934)	15.4 ± 11.7 (2.14 - 33.4)
	1	Yearling	13.6	0.150	0.122	0.0775	7.93	0.0150	5.95
2	17	Adult	110 ± 76.2 (32.2 - 276)	0.941 ± 0.550 (0.304 - 2.28)	0.855 ± 0.573 (0.239 - 2.31)	0.280 ± 0.171 (0.0809 - 0.731)	66.2 ± 47.0 (21.7 - 174)	0.0631 ± 0.0384 (0.0150 - 0.178)	41.9 ± 26.7 (11.8 - 93.5)
	4	Juvenile	22.3 ± 5.20 (16.8 - 27.4)	0.189 ± 0.0778 (0.150 - 0.306)	0.170 ± 0.0397 (0.122 - 0.212)	0.0488 ± 0.0122 (0.0309 - 0.0584)	13.6 ± 3.10 (10.5 - 16.5)	0.0150	8.77 ± 2.01 (7.03 - 11.2)
	5	Yearling	14.4 ± 10.1 (5.03 - 30.2)	0.223 ± 0.163 (0.150 - 0.515)	0.164 ± 0.178 (0.0225 - 0.473)	0.0657 ± 0.0591 (0.0294 - 0.169)	7.24 ± 3.78 (3.24 - 12.2)	0.0286 ± 0.0303 (0.0150 - 0.0828)	5.57 ± 3.32 (1.97 - 9.74)
3	2	Adult	90.4 ± 65.3 (44.3 - 137)	0.971 ± 0.409 (0.682 - 1.26)	0.817 ± 0.313 (0.596 - 1.04)	0.265 ± 0.106 (0.190 - 0.340)	55.9 ± 39.2 (28.2 - 83.7)	0.0554 ± 0.0305 (0.0338 - 0.0770)	35.6 ± 22.8 (19.5 - 51.7)
	6	Juvenile	40.3 ± 23.9 (15.6 - 79.9)	0.380 ± 0.301 (0.150 - 0.909)	0.339 ± 0.256 (0.118 - 0.790)	0.117 ± 0.0948 (0.0510 - 0.302)	23.1 ± 13.0 (9.17 - 44.6)	0.0289 ± 0.0181 (0.0150 - 0.0604)	16.8 ± 10.2 (6.44 - 34.2)
4	10	Adult	118 ± 153 (37.9 - 546)	0.980 ± 1.19 (0.338 - 4.29)	0.841 ± 0.905 (0.287 - 3.29)	0.275 ± 0.307 (0.0967 - 1.12)	69.2 ± 91.2 (23.1 - 324)	0.0580 ± 0.0581 (0.0150 - 0.214)	47.2 ± 62.2 (13.0 - 221)
	11	Juvenile*	50.5 ± 40.6 (9.64 - 121)	1.20 ± 2.44 (0.150 - 8.47)	1.31 ± 3.09 (0.0691 - 10.6)	0.427 ± 0.967 (0.0350 - 3.33)	29.6 ± 24.2 (5.88 - 72.7)	0.165 ± 0.440 (0.0150 - 1.49)	20.6 ± 16.0 (3.85 - 48.8)
	2	Yearling	15.2 ± 12.4 (6.43 - 24.0)	0.150	0.117 ± 0.0824 (0.0592 - 0.176)	0.0373 ± 0.0386 (0.0100 - 0.0646)	9.43 ± 8.03 (3.75 - 15.1)	0.0150	6.28 ± 4.90 (2.82 - 9.75)
5	3	Adult	209 ± 64.5 (135 - 251)	1.51 ± 0.602 (0.969 - 2.16)	1.41 ± 0.538 (0.902 - 1.97)	0.460 ± 0.191 (0.309 - 0.675)	125 ± 41.8 (76.7 - 150)	0.0865 ± 0.0399 (0.0614 - 0.132)	78.3 ± 25.5 (49.3 - 97.5)
	4	Juvenile	21.0 ± 17.4 (4.28 - 36.2)	0.272 ± 0.145 (0.150 - 0.434)	0.235 ± 0.167 (0.0737 - 0.393)	0.0687 ± 0.0561 (0.0100 - 0.118)	11.4 ± 9.97 (1.42 - 20.1)	0.0326 ± 0.0143 (0.0150 - 0.0499)	8.57 ± 6.99 (1.24 - 14.8)
6	1	Adult	62.9	0.792	0.554	0.234	35.6	0.0463	27.5
	4	Juvenile*	22.6 ± 7.32 (16.0 - 33.1)	0.188 ± 0.0751 (0.150 - 0.300)	0.183 ± 0.0312 (0.144 - 0.209)	0.0695 ± 0.0221 (0.0469 - 0.0979)	12.9 ± 4.96 (8.72 - 20.1)	0.0150	9.82 ± 2.41 (7.11 - 12.8)

* Groups containing samples that are likely contaminated

Table A10 Continued

Group	N	Age	Pr	Sm	Tb	Y	Yb
1	1	Adult	4.60	2.58	0.218	3.66	0.216
	7	Juvenile	3.98 ± 2.91 (0.559 - 8.26)	2.30 ± 1.73 (0.374 - 5.22)	0.196 ± 0.143 (0.0333 - 0.442)	3.59 ± 2.34 (0.952 - 6.61)	0.248 ± 0.189 (0.0300 - 0.528)
	1	Yearling	1.40	0.971	0.0775	1.26	0.0616
2	17	Adult	11.3 ± 7.47 (3.41 - 27.3)	5.45 ± 3.27 (1.37 - 11.8)	0.441 ± 0.256 (0.129 - 0.970)	7.29 ± 4.34 (2.25 - 18.5)	0.510 ± 0.306 (0.191 - 1.32)
	4	Juvenile	2.31 ± 0.508 (1.75 - 2.86)	1.25 ± 0.290 (1.00 - 1.61)	0.100 ± 0.0182 (0.0804 - 0.123)	1.60 ± 0.399 (1.19 - 2.12)	0.137 ± 0.0339 (0.0929 - 0.176)
	5	Yearling	1.42 ± 0.859 (0.492 - 2.54)	0.768 ± 0.461 (0.331 - 1.41)	0.0689 ± 0.0595 (0.0150 - 0.169)	1.45 ± 1.38 (0.518 - 3.87)	0.121 ± 0.128 (0.0300 - 0.348)
3	2	Adult	9.49 ± 6.44 (4.94 - 14.0)	4.81 ± 2.97 (2.71 - 6.90)	0.419 ± 0.227 (0.259 - 0.580)	6.97 ± 2.64 (5.10 - 8.83)	0.430 ± 0.223 (0.272 - 0.587)
	6	Juvenile	4.31 ± 2.59 (1.63 - 8.77)	2.42 ± 1.52 (0.862 - 5.16)	0.215 ± 0.152 (0.0852 - 0.498)	3.11 ± 2.22 (1.13 - 7.16)	0.169 ± 0.151 (0.0300 - 0.452)
4	10	Adult	12.3 ± 15.9 (3.79 - 56.7)	6.31 ± 8.03 (1.99 - 28.8)	0.476 ± 0.599 (0.145 - 2.14)	7.20 ± 8.08 (2.33 - 29.3)	0.446 ± 0.394 (0.147 - 1.52)
	11	Juvenile*	5.37 ± 4.25 (0.992 - 12.6)	3.14 ± 2.11 (0.497 - 6.54)	0.376 ± 0.514 (0.0379 - 1.85)	10.3 ± 22.9 (0.757 - 78.8)	1.22 ± 3.30 (0.0300 - 11.2)
	2	Yearling	1.58 ± 1.27 (0.682 - 2.48)	0.928 ± 0.621 (0.489 - 1.37)	0.0638 ± 0.0469 (0.0307 - 0.0970)	1.35 ± 1.07 (0.593 - 2.10)	0.0715 ± 0.0586 (0.0300 - 0.113)
5	3	Adult	21.2 ± 6.67 (13.6 - 26.0)	10.3 ± 3.61 (6.31 - 13.4)	0.743 ± 0.292 (0.440 - 1.02)	11.2 ± 4.73 (7.12 - 16.4)	0.721 ± 0.361 (0.506 - 1.14)
	4	Juvenile	2.22 ± 1.87 (0.323 - 4.05)	1.23 ± 0.917 (0.309 - 2.08)	0.122 ± 0.0927 (0.0373 - 0.219)	2.00 ± 1.38 (0.524 - 3.30)	0.192 ± 0.0970 (0.0747 - 0.299)
6	1	Adult	7.03	4.13	0.327	5.33	0.387
	4	Juvenile*	2.50 ± 0.716 (1.84 - 3.51)	1.34 ± 0.305 (1.04 - 1.65)	0.112 ± 0.0166 (0.0937 - 0.130)	1.52 ± 0.287 (1.22 - 1.83)	0.0956 ± 0.0340 (0.0619 - 0.134)

* Groups containing samples that are likely contaminated

Appendix IX - Trace element concentrations in grey seal livers

Table A9: Concentrations of trace elements in µg·g^{-1} dry weight, mean ± standard deviation and range in parentheses in grey seal (Halichoerus grypus) livers (n=76) from the Baltic Sea.

Group	**N**	**Age**	**Ag**	**As**	**B**	**Ba**	**Bi**	**Cd**	**Co**
			µg/g	µg/g	ng/g	ng/g	ng/g	µg/g	µg/g
1	1	Adult	0.486	0.512	6.00	3.68	263	0.944	0.0502
	7	Juvenile	0.443 ± 0.213 (0.161 - 0.770)	0.246 ± 0.0552 (0.169 - 0.313)	6.00	21.2 ± 19.4 (2.88 - 55.4)	131 ± 76.7 (70.7 - 287)	0.637 ± 0.268 (0.411 - 1.04)	0.0533 ± 0.0112 (0.0325 - 0.0653)
	1	Yearling	0.107	0.276	6.00	3.59	8.34	0.184	0.0669
2	17	Adult	0.391 ± 0.209 (0.113 - 0.898)	0.551 ± 0.242 (0.347 - 1.24)	23.5 ± 21.7 (6.00 - 90.2)	18.5 ± 33.8 (2.41 - 139)	406 ± 271 (52.5 - 934)	1.42 ± 0.57 (0.543 - 2.18)	0.0725 ± 0.0240 (0.0317 - 0.130)
	4	Juvenile	0.378 ± 0.313 (0.0989 - 0.742)	0.653 ± 0.310 (0.272 - 0.954)	11.2 ± 6.80 (6.00 - 20.3)	10.2 ± 6.69 (3.94 - 17.3)	405 ± 627 (54.7 - 1340)	0.720 ± 0.217 (0.554 - 1.03)	0.0537 ± 0.0143 (0.0391 - 0.0731)
	5	Yearling	0.158 ± 0.0660 (0.0902 - 0.259)	0.503 ± 0.0851 (0.382 - 0.592)	9.75 ± 5.46 (6.00 - 18.0)	20.0 ± 19.7 (5.18 - 54.2)	29.8 ± 31.6 (8.88 - 84.7)	0.144 ± 0.142 (0.0525 - 0.395)	0.0523 ± 0.0162 (0.0343 - 0.0787)
3	2	Adult	0.263 ± 0.0812 (0.206 - 0.321)	0.438 ± 0.170 (0.318 - 0.558)	24.5 ± 2.08 (23.0 - 26.0)	17.9 ± 21.0 (2.99 - 32.7)	292 ± 222 (135 - 449)	1.39 ± 0.175 (1.26 - 1.51)	0.0869 ± 0.0490 (0.0522 - 0.121)
	6	Juvenile	0.362 ± 0.147 (0.167 - 0.555)	0.450 ± 0.101 (0.328 - 0.565)	9.17 ± 4.92 (6.00 - 15.6)	7.18 ± 9.09 (2.04 - 25.5)	142 ± 82.0 (32.9 - 258)	0.592 ± 0.342 (0.256 - 1.10)	0.0591 ± 0.0273 (0.0391 - 0.111)
4	10	Adult	0.735 ± 0.483 (0.0785 - 1.46)	0.474 ± 0.227 (0.104 - 0.825)	85.9 ± 153 (6.00 - 515)	20.4 ± 20.5 (2.23 - 60.9)	1070 ± 1500 (10.7 - 4900)	1.11 ± 1.14 (0.176 - 4.03)	0.0639 ± 0.0322 (0.0317 - 0.145)
	11	Juvenile*	0.312 ± 0.236 (0.00443 - 0.726)	0.544 ± 0.258 (0.207 - 1.06)	15.0 ± 8.24 (6.00 - 28.2)	17.9 ± 23.8 (2.15 - 83.7)	241 ± 256 (23.9 - 687)	4.67 ± 13.4 (0.145 - 45.2)	0.0614 ± 0.0361 (0.0351 - 0.164)
	2	Yearling	0.0813 ± 0.0130 (0.0721 - 0.0905)	0.556 ± 0.0244 (0.539 - 0.573)	34.0 ± 39.6 (6.00 - 61.9)	17.3 ± 2.55 (15.5 - 19.1)	44.6 ± 50.6 (8.81 - 80.4)	0.115 ± 0.0775 (0.0601 - 0.170)	0.0436 ± 0.0177 (0.0311 - 0.0561)
5	3	Adult	1.68 ± 1.57 (0.149 - 3.29)	0.457 ± 0.227 (0.197 - 0.616)	195 ± 121 (59.1 - 293)	46.2 ± 3.27 (42.6 - 49.0)	1030 ± 1220 (232 - 2430)	1.20 ± 0.561 (0.602 - 1.71)	0.0931 ± 0.0337 (0.0584 - 0.126)
	4	Juvenile	0.195 ± 0.163 (0.037 - 0.423)	0.392 ± 0.188 (0.254 - 0.651)	73.4 ± 25.8 (52.0 - 107)	43.3 ± 22.2 (21.9 - 73.0)	78.2 ± 72.5 (15.3 - 169)	0.0793 ± 0.112 (0.00641 - 0.243)	0.0303 ± 0.00671 (0.0203 - 0.0344)
6	1	Adult	1.44	0.930	131	23.7	676	3.03	0.0673
	4	Juvenile*	0.415 ± 0.345 (0.112 - 0.911)	0.549 ± 0.173 (0.415 - 0.777)	56.7 ± 40.1 (6.00 - 98.9)	11.0 ± 4.23 (5.52 - 15.8)	203 ± 178 (28.8 - 452)	0.258 ± 0.0900 (0.156 - 0.351)	0.0419 ± 0.0137 (0.0339 - 0.0624)

* Groups containing samples that are likely contaminated

Table 12A Continued

Group	**N**	**Age**	**Cs** µg/g	**Cu** µg/g	**Ga** ng/g	**Ge** ng/g	**Hg** µg/g	**Li** ng/g	**Mn** µg/g	**Mo** µg/g
1	1	Adult	0.0971	115	1.19	4.14	102	2.25	10.7	2.42
	7	Juvenile	0.0429 ± 0.0202 (0.0256 - 0.075)	85.7 ± 28.9 (42.6 - 136)	1.17 ± 0.719 (0.550 - 2.45)	14.6 ± 19.3 (4.69 - 58.0)	43.7 ± 32.1 (12.0 - 98.6)	2.75 ± 1.33 (2.25 - 5.78)	11.5 ± 3.43 (4.89 - 14.8)	2.38 ± 0.615 (1.51 - 3.11)
	1	Yearling	0.0299	26.3	1.77	4.10	1.50	18.3	15.4	1.18
2	17	Adult	0.0504 ± 0.0127 (0.0342 - 0.0787)	35.0 ± 25,0 (9.96 - 97.9)	2.14 ± 0.867 (1.16 - 3.95)	10.0 ± 13.3 (3.95 - 57.9)	52.3 ± 33.8 (14.9 - 124)	6.57 ± 4.14 (2.25 - 17.7)	11.7 ± 8.33 (6.02 - 40.0)	2.02 ± 0.415 (1.12 - 2.66)
	4	Juvenile	0.0563 ± 0.0351 (0.0345 - 0.109)	101 ± 54.3 (45.4 - 175)	0.862 ± 0.360 (0.550 - 1.19)	5.83 ± 1.03 (4.56 - 7.08)	83.2 ± 133 (8.59 - 282)	3.38 ± 2.26 (2.25 - 6.77)	11.6 ± 2.14 (9.79 - 14.6)	2.02 ± 0.309 (1.74 - 2.30)
	5	Yearling	0.0479 ± 0.00640 (0.0384 - 0.0549)	30.9 ± 11.1 (15.5 - 41.4)	0.950 ± 0.895 (0.550 - 2.55)	17.4 ± 21.7 (5.29 - 56.0)	2.49 ± 0.889 (1.50 - 3.90)	3.78 ± 3.42 (2.25 - 9.90)	12.0 ± 3.67 (8.01 - 17.5)	1.43 ± 0.283 (1.16 - 1.82)
3	2	Adult	0.0493 ± 0.0410 (0.0203 - 0.0783)	28.2 ± 18.7 (14.9 - 41.4)	1.40 ± 1.21 (0.550 - 2.26)	6.16 ± 2.98 (4.05 - 8.27)	38.1 ± 7.26 (32.9 - 43.2)	5.85 ± 0.0685 (5.80 - 5.89)	11.9 ± 4.62 (8.66 - 15.2)	2.09 ± 0.593 (1.67 - 2.51)
	6	Juvenile	0.0481 ± 0.0101 (0.0356 - 0.0612)	91.0 ± 38.4 (54.1 - 158)	1.45 ± 0.926 (0.550 - 3.01)	15.3 ± 19.6 (5.34 - 55.2)	24.2 ± 19.5 (4.23 - 55.2)	2.68 ± 1.04 (2.25 - 4.80)	11.5 ± 1.71 (9.26 - 14.3)	1.95 ± 0.184 (1.66 - 2.14)
4	10	Adult	0.0592 ± 0.0173 (0.0345 - 0.0936)	83.2 ± 43.4 (34.7 - 167)	2.28 ± 1.44 (0.550 - 5.60)	5.91 ± 1.81 (3.43 - 9.12)	217 ± 241 (6.45 - 823)	6.22 ± 7.00 (2.25 - 23.6)	10.7 ± 3.89 (7.57 - 20.6)	2.41 ± 0.623 (1.62 - 3.74)
	11	Juvenile*	0.0581 ± 0.0116 (0.0409 - 0.0736)	78.3 ± 35.5 (19.4 - 133)	2.03 ± 2.23 (0.550 - 8.37)	5.91 ± 3.16 (1.50 - 12.6)	48.1 ± 51.2 (3.94 - 157)	5.00 ± 4.76 (2.25 - 16.7)	10.1 ± 2.88 (3.88 - 14.3)	1.79 ± 0.524 (0.411 - 2.42)
	2	Yearling	0.0506 ± 0.0150 (0.0400 - 0.0612)	40.4 ± 11.7 (32.1 - 48.6)	0.550	5.07 ± 0.786 (4.52 - 5.63)	3.36 ± 0.827 (2.77 - 3.94)	6.78 ± 2.03 (5.35 - 8.21)	11.5 ± 1.84 (10.2 - 12.8)	1.45 ± 0.150 (1.34 - 1.55)
5	3	Adult	0.063 ± 0.0124 (0.0553 - 0.0774)	26.7 ± 23.0 (10.0 - 52.9)	4.56 ± 0.388 (4.19 - 4.96)	6.44 ± 3.57 (3.58 - 10.4)	380 ± 474 (23.4 - 917)	12.3 ± 6.65 (4.60 - 16.3)	14.9 ± 2.22 (12.8 - 17.3)	2.54 ± 0.127 (2.39 - 2.64)
	4	Juvenile	0.0533 ± 0.0168 (0.0330 - 0.0736)	40.1 ± 17.5 (20.4 - 58.8)	3.28 ± 3.12 (0.550 - 7.63)	8.67 ± 2.56 (4.96 - 10.8)	7.17 ± 9.79 (0.0240 - 21.5)	8.28 ± 2.43 (6.06 - 11.6)	10.9 ± 2.42 (7.46 - 12.8)	1.61 ± 0.258 (1.23 - 1.78)
6	1	Adult	0.0554	36.3	2.02	9.49	193	9.88	8.10	2.07
	4	Juvenile*	0.0697 ± 0.00873 (0.0580 - 0.0788)	62.0 ± 44.3 (31.9 - 127)	9.90 ± 18.2 (0.550 - 37.1)	5.10 ± 1.37 (3.47 - 6.83)	41.6 ± 26.0 (5.51 - 61.1)	5.13 ± 2.27 (2.25 - 7.62)	6.53 ± 2.79 (3.88 - 9.82)	1.66 ± 0.134 (1.51 - 1.83)

* Groups containing samples that are likely contaminated

Table 12A Continued

Group	**N**	**Age**	**Ni** ng/g	**Pb** µg/g	**Rb** µg/g	**Sb** ng/g	**Se** µg/g	**Si** µg/g	**Sn** µg/g
1	1	Adult	16.1	0.0302	12.1	2.25	39.5	1.85	0.555
	7	Juvenile	9.75 ± 7.72	0.0354 ± 0.0100	8.73 ± 4.39	3.16 ± 2.53	18.5 ± 13.1	3.65 ± 4.26	0.245 ± 0.193
			(3.50 - 23.1)	(0.0219 - 0.0545)	(5.39 - 18.0)	(1.07 - 8.12)	(5.51 - 41.7)	(0.300 - 12.6)	(0.0580 - 0.548)
	1	Yearling	52.1	0.0284	5.67	1.24	2.32	4.99	0.0390
2	17	Adult	12.1 ± 17.7	0.0479 ± 0.0253	6.25 ± 1.21	5.06 ± 4.18	19.1 ± 11.7	4.00 ± 3.47	1.09 ± 0.973
			(3.50 - 70.5)	(0.0244 - 0.130)	(3.97 - 8.18)	(1.80 - 18.8)	(6.15 - 43.6)	(0.300 - 12.2)	(0.138 - 4.20)
	4	Juvenile	3.50	0.0447 ± 0.0357	6.89 ± 4.21	1.94 ± 1.76	29.6 ± 44.7	1.76 ± 1.72	0.305 ± 0.0867
				(0.0160 - 0.0963)	(3.59 - 13.0)	(0.935 - 4.57)	(4.97 - 96.5)	(0.300 - 4.14)	(0.258 - 0.435)
	5	Yearling	8.99 ± 5.42	0.0502 ± 0.00990	6.46 ± 1.25	1.38 ± 0.803	2.27 ± 0.181	7.74 ± 12,0	0.0402 ± 0.0114
			(3.50 - 15.6)	(0.0393 - 0.0660)	(5.61 - 8.64)	(0.472 - 2.67)	(2.11 - 2.48)	(0.300 - 29.1)	(0.0253 - 0.0553)
3	2	Adult	18.6 ± 3.46	0.0679 ± 0.0300	7.32 ± 5.36	3.20 ± 0.0786	14.4 ± 1.74	8.45 ± 5.84	1.18 ± 0.185
			(16.2 - 21.1)	(0.0467 - 0.0891)	(3.53 - 11.1)	(3.14 - 3.25)	(13.1 - 15.6)	(4.32 - 12.6)	(1.05 - 1.31)
	6	Juvenile	5.06 ± 2.49	0.0388 ± 0.0266	7.67 ± 0.622	1.68 ± 0.665	10.9 ± 8.15	3.60 ± 3.67	0.187 ± 0.110
			(3.50 - 9.10)	(0.0218 - 0.0913)	(6.70 - 8.44)	(1.19 - 2.94)	(3.22 - 24.3)	(0.668 - 10.6)	(0.00602 - 0.302)
4	10	Adult	5.95 ± 4.54	0.0737 ± 0.0591	8.60 ± 2.99	6.44 ± 4.10	79.6 ± 83.3	3.31 ± 3.28	2.60 ± 4.24
			(3.50 - 17.0)	(0.0274 - 0.205)	(4.53 - 13.2)	(1.69 - 13.1)	(3.51 - 285)	(0.300 - 11.5)	(0.0208 - 13.9)
	11	Juvenile*	8.95 ± 6.02	0.0643 ± 0.0302	7.24 ± 1.95	2.67 ± 1.34	18.9 ± 17.5	5.89 ± 8.04	0.515 ± 0.504
			(3.50 - 22.1)	(0.0221 - 0.109)	(5.03 - 11.1)	(1.20 - 5.92)	(2.86 - 55.7)	(0.683 - 28.8)	(0.0358 - 1.48)
	2	Yearling	3.50	0.0524 ± 0.00772	7.22 ± 0.0311	0.822 ± 0.523	2.33 ± 0.193	5.73 ± 5.18	0.0644 ± 0.0270
				(0.0469 - 0.0578)	(7.20 - 7.24)	(0.452 - 1.19)	(2.20 - 2.47)	(2.06 - 9.39)	(0.0453 - 0.0834)
5	3	Adult	190 ± 128	0.0712 ± 0.0189	5.91 ± 0.985	8.09 ± 5.08	140 ± 176	9.58 ± 4.50	0.0283 ± 0.00446
			(46.4 - 292)	(0.0494 - 0.0833)	(4.93 - 6.90)	(3.21 - 13.3)	(9.71 - 340)	(4.74 - 13.6)	(0.0255 - 0.0335)
	4	Juvenile	54.9 ± 29.9	0.0898 ± 0.0710	5.79 ± 0.0523	3.11 ± 0.748	5.75 ± 3.68	7.86 ± 3.64	0.295 ± 0.0969
			(17.8 - 90.9)	(0.0276 - 0.167)	(5.73 - 5.86)	(2.30 - 3.93)	(2.76 - 11.1)	(4.51 - 11.6)	(0.213 - 0.420)
6	1	Adult	7.09	0.0999	5.90	13.1	74.0	3.01	0.0867
	4	Juvenile*	10.4 ± 6.73	0.0358 ± 0.0117	4.40 ± 0.330	5.45 ± 2.74	18.7 ± 10.5	1.35 ± 0.529	0.366 ± 0.542
			(3.50 - 19.4)	(0.0238 - 0.0486)	(3.91 - 4.61)	(2.70 - 9.19)	(3.58 - 26.9)	(0.796 - 1.85)	(0.0445 - 1.17)

* Groups containing samples that are likely contaminated

Table 12A Continued

Group	**N**	**Age**	Sr µg/g	Th ng/g	Tl ng/g	V µg/g	W ng/g	Zn µg/g
1	1	Adult	0.114	0.108	10.0	0.173	0.772	205
	7	Juvenile	0.324 ± 0.698 (0.0516 - 1.91)	0.386 ± 0.565 (0.0375 - 1.65)	9.60 ± 5.71 (4.56 - 19.5)	0.086 ± 0.055 (0.0373 - 0.164)	1.07 ± 0.331 (0.731 - 1.55)	203 ± 35.6 (144 - 260)
	1	Yearling	0.0897	0.417	9.31	0.0386	1.64	144
2	17	Adult	0.174 ± 0.140 (0.0371 - 0.565)	0.205 ± 0.267 (0.0375 - 1.14)	9.79 ± 4.86 (2.70 - 23.5)	0.423 ± 0.209 (0.164 - 0.929)	4.08 ± 7.81 (0.630 - 30.7)	174 ± 51.2 (97.5 - 305)
	4	Juvenile	0.112 ± 0.0608 (0.0669 - 0.197)	0.140 ± 0.0324 (0.108 - 0.176)	9.25 ± 4.51 (5.19 - 15.5)	0.0585 ± 0.0126 (0.0442 - 0.0749)	2.90 ± 1.99 (1.04 - 4.81)	167 ± 34.0 (137 - 216)
	5	Yearling	0.0823 ± 0.0458 (0.0441 - 0.162)	0.600 ± 0.626 (0.0891 - 1.61)	11.7 ± 8.99 (5.04 - 27.4)	0.0514 ± 0.0176 (0.0343 - 0.077)	2.35 ± 2.03 (0.823 - 5.87)	142 ± 24.0 (117 - 181)
3	2	Adult	0.113 ± 0.00309 (0.111 - 0.115)	0.176 ± 0.0242 (0.159 - 0.193)	5.29 ± 3.76 (2.64 - 7.95)	0.305 ± 0.0447 (0.273 - 0.336)	2.72 ± 1.38 (1.75 - 3.69)	158 ± 34.3 (133 - 182)
	6	Juvenile	0.0624 ± 0.00559 (0.0553 - 0.0704)	0.179 ± 0.145 (0.0375 - 0.453)	5.73 ± 3.59 (2.45 - 10.9)	0.130 ± 0.108 (0.0324 - 0.294)	2.06 ± 0.943 (1.08 - 3.26)	164 ± 19.9 (136 - 195)
4	10	Adult	0.187 ± 0.148 (0.0658 - 0.547)	0.196 ± 0.145 (0.0776 - 0.537)	6.75 ± 3.02 (3.45 - 11.1)	0.335 ± 0.285 (0.0727 - 0.968)	2.47 ± 0.985 (1.07 - 4.00)	210 ± 68.5 (131 - 362)
	11	Juvenile*	0.166 ± 0.132 (0.0418 - 0.450)	0.215 ± 0.227 (0.0375 - 0.717)	6.95 ± 3.92 (2.85 - 15.2)	0.144 ± 0.120 (0.0402 - 0.409)	1.74 ± 1.05 (0.706 - 4.09)	167 ± 25.4 (131 - 212)
	2	Yearling	0.0794 ± 0.0313 (0.0573 - 0.102)	0.0834 ± 0.065 (0.0375 - 0.129)	3.69 ± 1.46 (2.66 - 4.72)	0.0311 ± 0.00276 (0.0292 - 0.0331)	2.41 ± 0.199 (2.27 - 2.55)	129 ± 20.3 (114 - 143)
5	3	Adult	0.423 ± 0.142 (0.329 - 0.587)	0.474 ± 0.191 (0.360 - 0.694)	6.39 ± 6.32 (1.71 - 13.6)	0.816 ± 0.612 (0.437 - 1.52)	3.40 ± 1.96 (1.21 - 4.99)	291 ± 68.2 (213 - 343)
	4	Juvenile	0.354 ± 0.262 (0.143 - 0.715)	0.780 ± 0.434 (0.243 - 1.18)	4.06 ± 1.36 (2.83 - 5.95)	0.0665 ± 0.0388 (0.0203 - 0.107)	2.55 ± 1.32 (1.16 - 4.12)	149 ± 43.7 (111 - 204)
6	1	Adult	0.880	0.140	4.26	0.686	1.77	162
	4	Juvenile*	0.733 ± 0.434 (0.313 - 1.28)	0.114 ± 0.0324 (0.0777 - 0.150)	4.23 ± 1.81 (2.52 - 6.53)	0.0808 ± 0.0190 (0.0620 - 0.101)	1.09 ± 0.472 (0.427 - 1.50)	120 ± 22.4 (104 - 153)

* Groups containing samples that are likely contaminated

Appendix X - Relationships between chemical elements in liver tissue

Table A10: Pearson correlations for all elements in livers (n = 74) (log transformed data). Bold numbers indicate $p < 0.01$, while rest $p < 0.05$.

	Ag	As	B	Ba	Bi	Ca	Cd	Ce	Co	Cs	Cu	Dy	Er	Fe	Ga	Hg	Ho	La	Li	Lu	Mg
Ag					**0.79**		**0.51**	**0.46**	0.29	**0.44**	**0.37**	**0.45**	**0.43**		**0.32**	**0.83**	**0.42**	**0.47**		0.30	
As																					
B				0.28	0.29	**0.33**		0.26			-0.27	**0.34**	**0.30**		**0.38**	0.27	0.23	0.23	**0.63**	**0.34**	
Ba			0.28			**0.30**													0.27	0.26	
Bi	**0.79**		0.29				**0.56**	**0.56**	0.26	**0.39**		**0.56**	**0.54**		**0.39**	**0.87**	**0.48**	**0.57**		**0.36**	
Ca			**0.33**	**0.30**							-0.23								**0.33**		**-0.34**
Cd	**0.51**				**0.56**			**0.72**	**0.66**			**0.65**	**0.66**		**0.42**	**0.60**	**0.69**	**0.75**		**0.50**	
Ce	**0.46**		0.26		**0.56**		**0.72**		**0.62**			**0.92**	**0.96**		**0.76**	**0.59**	**0.95**	**0.99**	0.24	**0.80**	
Co	0.29				0.26		**0.66**	**0.62**				**0.59**	**0.57**		**0.44**		**0.62**	**0.64**		**0.53**	0.27
Cs	**0.44**				**0.39**											**0.38**					
Cu	**0.37**		-0.27			-0.23													**-0.37**		
Dy	**0.45**		**0.34**		**0.56**		**0.65**	**0.92**	**0.59**				**0.94**		**0.77**	**0.55**	**0.92**	**0.91**	**0.34**	**0.91**	
Er	**0.43**		**0.30**		**0.54**		**0.66**	**0.96**	**0.57**			**0.94**			**0.77**	**0.56**	**0.96**	**0.94**	**0.32**	**0.87**	
Fe																					
Ga	**0.32**		**0.38**		**0.39**		**0.42**	**0.76**	**0.44**			**0.77**	**0.77**			**0.40**	**0.75**	**0.75**	**0.39**	**0.72**	
Ge																					
Hg	**0.83**		0.27		**0.87**		**0.60**	**0.59**		**0.38**		**0.55**	**0.56**		**0.40**		**0.52**	**0.61**		**0.37**	
Ho	**0.42**		0.23		**0.48**		**0.69**	**0.95**	**0.62**			**0.92**	**0.96**		**0.75**	**0.52**		**0.94**	0.26	**0.84**	
K																					
La	**0.47**		0.23		**0.57**		**0.75**	**0.99**	**0.64**			**0.91**	**0.94**		**0.75**	**0.61**	**0.94**			**0.78**	
Li			**0.63**	0.27		**0.33**		0.24			**-0.37**	**0.34**	**0.32**		**0.39**		0.26			**0.36**	
Lu	**0.30**		**0.34**	0.26	**0.36**		**0.50**	**0.80**	**0.53**			**0.91**	**0.87**		**0.72**	**0.37**	**0.84**	**0.78**	**0.36**		
Mg						**-0.34**			0.27												

	Mn	**Mo**	**Na**	**Nd**	**Ni**	**P**	**Pb**	**Pr**	**Rb**	**S**	**Sb**	**Se**	**Si**	**Sm**	**Sn**	**Sr**	**Tb**	**Th**	**Tl**	**V**	**W**	**Y**	**Yb**	**Zn**
Ag		**0.62**		**0.46**				**0.46**	**0.36**	**0.36**	**0.55**	**0.84**		**0.46**	**0.32**		**0.46**		0.29	**0.53**		**0.44**	**0.37**	**0.48**
As									**-0.39**															
B			**0.31**	0.25	**0.38**	-0.29	0.27	0.25			**0.34**	0.30	**0.30**	0.27		**0.57**	0.28			**0.35**		0.28	**0.31**	
Ba					**0.46**		**0.62**				**0.37**		**0.41**			0.23		**0.39**			0.29			
Bi		**0.52**		**0.56**				**0.56**	0.27	**0.40**	**0.66**	**0.84**		**0.57**	**0.49**	0.24	**0.55**			**0.68**		**0.53**	**0.49**	**0.35**
Ca	**-0.36**		**0.58**		**0.40**	**-0.45**			**-0.31**	**-0.33**						**0.72**		0.26						
Cd		**0.63**		**0.72**				**0.73**		**0.58**	**0.45**	**0.53**		**0.72**	**0.35**		**0.72**			**0.68**		**0.71**	**0.59**	**0.44**
Ce		**0.55**		**1.00**			0.24	**0.99**		**0.50**	**0.53**	**0.56**		**0.99**	**0.36**		**0.98**	0.27		**0.88**		**0.96**	**0.90**	**0.46**
Co	**0.36**	**0.53**		**0.62**		0.25	0.25	**0.62**		**0.61**	**0.32**			**0.61**		-0.24	**0.61**		0.27	**0.59**		**0.62**	**0.52**	**0.42**
Cs									**0.48**			**0.40**				**0.39**			**0.33**					
Cu		**0.38**	**-0.31**		**-0.30**				**0.34**															**0.31**
Dy		**0.53**		**0.92**			**0.34**	**0.91**		**0.48**	**0.53**	**0.53**		**0.93**	**0.34**		**0.93**	**0.36**		**0.89**		**0.96**	**0.92**	**0.45**
Er		**0.50**		**0.95**			0.24	**0.95**		**0.46**	**0.52**	**0.52**		**0.96**	**0.34**		**0.96**	**0.38**		**0.86**		**0.98**	**0.95**	**0.44**
Fe			**0.41**			-0.28														0.23				0.28
Ga	0.25	**0.48**		**0.76**	0.23		0.29	**0.76**		**0.44**	**0.49**	**0.41**		**0.76**			**0.77**	**0.45**		**0.72**	0.28	**0.77**	**0.76**	**0.49**
Ge																								
Hg		**0.57**		**0.59**				**0.60**	**0.36**	0.29	**0.60**	**0.99**		**0.60**	**0.49**	0.30	**0.58**			**0.63**		**0.56**	**0.48**	**0.37**
Ho		**0.49**		**0.96**			0.25	**0.96**		**0.45**	**0.52**	**0.49**		**0.96**	0.29		**0.96**	**0.38**		**0.85**		**0.96**	**0.91**	**0.44**
K																								
La		**0.57**		**0.99**			0.24	**0.99**		**0.51**	**0.52**	**0.57**		**0.98**	**0.37**		**0.97**			**0.88**		**0.95**	**0.88**	**0.46**
Li			**0.33**		**0.33**	-0.24	0.27				0.28		0.23	0.23		**0.40**	0.25	0.25		0.29		0.30	**0.34**	
Lu		**0.44**		**0.80**	0.26		**0.31**	**0.79**		**0.41**	**0.44**	**0.35**	0.26	**0.81**			**0.84**	**0.47**		**0.75**		**0.88**	**0.87**	**0.40**
Mg	**0.52**	**0.46**	**-0.44**			**0.84**			0.26	**0.55**						**-0.39**								**0.50**

	Ag	As	B	Ba	Bi	Ca	Cd	Ce	Co	Cs	Cu	Dy	Er	Fe	Ga	Hg	Ho	La	Li	Lu	Mg
Mn						**-0.36**			**0.36**						0.25						**0.52**
Mo	**0.62**				**0.52**		**0.63**	**0.55**	**0.53**		**0.38**	**0.53**	**0.50**		**0.48**	**0.57**	**0.49**	**0.57**		**0.44**	**0.46**
Na			**0.31**			**0.58**					**-0.31**			**0.41**					**0.33**		**-0.44**
Nd	**0.46**		0.25		**0.56**		**0.72**	**0.99**	**0.62**			**0.92**	**0.95**		**0.76**	**0.59**	**0.96**	**0.99**		**0.80**	
Ni			**0.38**	**0.46**		**0.40**					-0.30				0.23				**0.33**	0.26	
P			-0.29			**-0.45**			0.25					-0.28					-0.24		**0.84**
Pb			0.27	**0.62**				0.24	0.25			**0.34**	0.24		0.29		0.25	0.24	0.27	**0.31**	
Pr	**0.46**		0.25		**0.56**		**0.73**	**0.99**	**0.62**			**0.91**	**0.95**		**0.76**	**0.60**	**0.96**	**0.99**		**0.79**	
Rb	**0.36**	**-0.39**			0.27	**-0.31**				**0.48**	**0.34**					**0.36**					0.26
S	**0.36**				**0.40**	**-0.33**	**0.58**	**0.50**	**0.61**			**0.48**	**0.46**		**0.44**	0.29	**0.45**	**0.51**		**0.41**	**0.55**
Sb	**0.55**		**0.34**	**0.37**	**0.66**		**0.45**	**0.53**	**0.32**			**0.53**	**0.52**		**0.49**	**0.60**	**0.52**	**0.52**	0.28	**0.44**	
Se	**0.84**		0.30		**0.84**		**0.53**	**0.56**		**0.40**		**0.53**	**0.52**		**0.41**	**0.99**	**0.49**	**0.57**		**0.35**	
Si			**0.30**	**0.41**															0.23	0.26	
Sm	**0.46**		0.27		**0.57**		**0.72**	**0.99**	**0.61**			**0.93**	**0.96**		**0.76**	**0.60**	**0.96**	**0.98**	0.23	**0.81**	
Sn	**0.32**				**0.49**		**0.35**	**0.36**				**0.34**	**0.34**			**0.49**	0.29	**0.37**			
Sr			**0.57**	0.23	0.24	**0.72**			-0.24	**0.39**						0.30			**0.40**		**-0.39**
Tb	**0.46**		0.28		**0.55**		**0.72**	**0.98**	**0.61**			**0.93**	**0.96**		**0.77**	**0.58**	**0.96**	**0.97**	0.25	**0.84**	
Th				**0.39**		0.26		0.27				**0.36**	**0.38**		**0.45**		**0.38**		0.25	**0.47**	
Tl	0.29								0.27	**0.33**											
V	**0.53**		**0.35**		**0.68**		**0.68**	**0.88**	**0.59**			**0.89**	**0.86**	0.23	**0.72**	**0.63**	**0.85**	**0.88**	0.29	**0.75**	
W				0.29											0.28						
Y	**0.44**		0.28		**0.53**		**0.71**	**0.96**	**0.62**			**0.96**	**0.98**		**0.77**	**0.56**	**0.96**	**0.95**	0.30	**0.88**	
Yb	**0.37**		**0.31**		**0.49**		**0.59**	**0.90**	**0.52**			**0.92**	**0.95**		**0.76**	**0.48**	**0.91**	**0.88**	**0.34**	**0.87**	
Zn	**0.48**				**0.35**		**0.44**	**0.46**	**0.42**		**0.31**	**0.45**	**0.44**	0.28	**0.49**	**0.37**	**0.44**	**0.46**		**0.40**	**0.50**

	Mn	**Mo**	**Na**	**Nd**	**Ni**	**P**	**Pb**	**Pr**	**Rb**	**S**	**Sb**	**Se**	**Si**	**Sm**	**Sn**	**Sr**	**Tb**	**Th**	**Tl**	**V**	**W**	**Y**	**Yb**	**Zn**
Mn		**0.35**				**0.53**				**0.38**						**-0.39**		0.26			**0.46**			**0.42**
Mo	**0.35**		-0.27	**0.56**		**0.40**		**0.56**	0.28	**0.63**	**0.49**	**0.55**		**0.56**	**0.34**		**0.57**			**0.50**		**0.55**	**0.46**	**0.75**
Na		-0.27			0.26	**-0.60**			**-0.38**	-0.36						**0.45**								
Nd		**0.56**					0.25	**0.99**		**0.50**	**0.53**	**0.56**		**0.99**	**0.36**		**0.98**	0.27		**0.88**		**0.97**	**0.89**	**0.46**
Ni			0.26						-0.24				**0.48**			**0.31**		**0.36**			0.26			
P	**0.53**	**0.40**	**-0.60**						**0.35**	**0.57**						**-0.60**								**0.37**
Pb				0.25				0.24			**0.33**		0.27	0.25				0.29		**0.30**		0.25	**0.31**	0.30
Pr		**0.56**		**0.99**			0.24			**0.49**	**0.53**	**0.56**		**0.99**	**0.37**		**0.98**	0.26		**0.88**		**0.96**	**0.89**	**0.46**
Rb		0.28	**-0.38**		-0.24	**0.35**				**0.31**		**0.38**							**0.34**					0.27
S	**0.38**	**0.63**	**-0.36**	**0.50**		**0.57**		**0.49**	**0.31**			0.25		**0.49**		**-0.36**	**0.48**			**0.44**		**0.50**	**0.46**	**0.56**
Sb		**0.49**		**0.53**			**0.33**	**0.53**				**0.58**		**0.53**	**0.39**	**0.32**	**0.52**			**0.63**	0.28	**0.52**	**0.54**	**0.40**
Se		**0.55**		**0.56**				**0.56**	**0.38**	0.25	**0.58**			**0.56**	**0.46**	**0.33**	**0.55**		0.24	**0.60**		**0.53**	**0.45**	**0.37**
Si					**0.48**		0.27											0.27						
Sm		**0.56**		**0.99**			0.25	**0.99**		**0.49**	**0.53**	**0.56**			**0.35**		**0.99**	0.28		**0.88**		**0.97**	**0.90**	**0.46**
Sn		**0.34**		**0.36**				**0.37**			**0.39**	**0.46**		**0.35**			**0.36**			**0.39**		**0.34**	**0.31**	
Sr	**-0.39**		**0.45**		**0.31**	**-0.60**				**-0.36**	**0.32**	**0.33**												
Tb		**0.57**		**0.98**				**0.98**		**0.48**	**0.52**	**0.55**		**0.99**	**0.36**			**0.31**		**0.86**		**0.98**	**0.90**	**0.45**
Th	0.26			0.27	**0.36**		0.29	0.26					0.27	0.28			**0.31**				0.28	**0.36**	**0.46**	
Tl									**0.34**			0.24												
V		**0.50**		**0.88**			**0.30**	**0.88**		**0.44**	**0.63**	**0.60**		**0.88**	**0.39**		**0.86**					**0.87**	**0.83**	**0.40**
W	**0.46**				0.26						0.28							0.28						
Y		**0.55**		**0.97**			0.25	**0.96**		**0.50**	**0.52**	**0.53**		**0.97**	**0.34**		**0.98**	**0.36**		**0.87**			**0.94**	**0.47**
Yb		**0.46**		**0.89**			**0.31**	**0.89**		**0.46**	**0.54**	**0.45**		**0.90**	**0.31**		**0.90**	**0.46**		**0.83**		**0.94**		**0.43**
Zn	**0.42**	**0.75**		**0.46**		**0.37**	0.30	**0.46**	0.27	**0.56**	**0.40**	**0.37**		**0.46**			**0.45**			**0.40**		**0.47**	**0.43**	

Apppendix XI - Relationship between chemical elements, biometric variables, stable isotope ratios(C and N) and Se:Hg molar ratios

Table 11: Relationship between biometric data, stable isotopes and Se:Hg versus the chemical elements. Bold numbers indicate $p < 0.01$, while rest $p < 0.05$

Element	**Age** (n = 44)	**Weight** (n = 73)	**Lenght** (n = 74)	**δ^{15}N** (n = 72)	**δ^{13}C** (n = 72)	**Se:Hg** (n = 74)
Ag		**0.42**	**0.43**	**0.32**		**-0.63**
As						
B	**0.40**	0.28		**0.35**	0.29	
Ba						
Bi	**0.46**	**0.60**	**0.52**	**0.39**	0.28	**-0.77**
Ca				**0.32**		
Cd	**0.39**	**0.58**	**0.55**			**-0.72**
Ce	**0.65**	**0.64**	**0.60**			**-0.59**
Co	0.33	**0.32**				-0.28
Cs				**0.39**		-0.24
Cu	**-0.54**					
Dy	**0.63**	**0.62**	**0.58**			**-0.51**
Er	**0.62**	**0.62**	**0.58**			**-0.57**
Fe						
Ga	**0.54**	**0.37**	**0.33**	0.27		-0.27
Ge						
Hg	**0.44**	**0.67**	**0.61**	**0.34**	**0.32**	**-0.83**
Ho	**0.60**	**0.59**	**0.58**			**-0.51**
K						
La	**0.67**	**0.64**	**0.60**			**-0.61**
Li	0.37					
Lu	**0.54**	**0.45**	**0.43**			**-0.33**
Mg		-0.25	-0.27			
Mn			-0.26			
Mo		**0.32**	**0.36**			**-0.52**
Na					0.24	
Nd	**0.63**	**0.63**	**0.60**			**-0.58**
Ni				**0.36**		
P		-0.29	**-0.30**			
Pb						
Pr	**0.64**	**0.64**	**0.61**			**-0.59**
Rb						-0.25
S						**-0.36**
Sb	**0.43**	**0.53**	**0.43**	**0.36**	0.25	**-0.51**
Se	**0.41**	**0.63**	**0.56**	**0.36**	**0.33**	**-0.73**
Si						
Sm	**0.59**	**0.62**	**0.59**			**-0.58**
Sn	**0.50**	**0.61**	**0.55**			**-0.49**
Sr		0.24		**0.50**	**0.34**	
Tb	**0.59**	**0.63**	**0.60**			**-0.57**
Th						0.25
Tl					-0.28	
V	**0.72**	**0.71**	**0.65**	0.26		**-0.60**
W						
Y	**0.61**	**0.62**	**0.60**			**-0.55**
Yb	**0.60**	**0.59**	**0.56**			**-0.50**
Zn				0.25		**-0.32**