Methylmercury biomagnification in an Arctic pelagic food web

Anders Ruus

Norwegian Institute for Water Research (NIVA), Gaustadalléen 21, 0349 Oslo, Norway

Phone: +47 22 18 51 00

Fax: +47 22 18 52 00

anders.ruus@niva.no

Total number of words: 6,949

Methylmercury biomagnification in an Arctic pelagic food web

Anders Ruus, *† Ida B. Øverjordet, ‡, § Hans Fredrik V. Braaten, † Anita Evenset, ‖, # Guttorm Christensen, ‖ Eldbjørg S. Heimstad, †† Geir W. Gabrielsen, ‡‡ Katrine Borgå, †, §§

† *Norwegian Institute for Water Research, Gaustadalléen 21, NO-0349 Oslo, Norway*

‡ *Norwegian University of Science and Technology, Department of Biology, NO-7491 Trondheim, Norway*

§ *SINTEF Materials and Chemistry, Marine Environmental Technology, Brattørkaia 17B, NO-7010 Trondheim*

‖ *Akvaplan-niva, Fram Centre, NO-9296 Tromsø, Norway*

University of Tromsø. The Arctic University of Norway, NO-9037 Tromsø, Norway

†† *Norwegian Institute for Air Research, Fram Centre, NO-9296 Tromsø, Norway*

‡‡ *Norwegian Polar Institute, Fram Centre, NO-9296 Tromsø, Norway*

§§ *University of Oslo, Department of Biosciences, PO Box 1066 Blindern, NO-0316 Oslo, Norway*

* To whom correspondence may be addressed (anders.ruus@niva.no).

Abstract

Mercury (Hg) is a toxic element entering the biosphere from natural and anthropogenic sources, and emitted gaseous Hg enters the Arctic from lower latitudes by long-range transport. In aquatic systems, anoxic conditions favour the bacterial transformation of inorganic mercury to methylmercury (MeHg), which has a greater potential for bioaccumulation than inorganic mercury, and is the most toxic form of Hg. The main objective of this study was to quantify the biomagnification of MeHg in a marine pelagic food web, comprising species of zooplankton, fish and seabirds, from the Kongsfjorden system (Svalbard, Norway), by use of Trophic Magnification Factors (TMFs). As expected, tissue concentrations of MeHg increased with increasing trophic level in the food web, however, at greater rates than observed in several earlier studies, especially at lower latitudes. There was strong correlation between MeHg and total Hg (TotHg) concentrations through the food web as a whole. The concentration of MeHg in kittiwake decreased from May to October, contributing to seasonal differences in TMFs. The ecology and physiology of the species comprising the food web in question may have large influence on the magnitude of the biomagnification. A significant linear relationship was also observed between concentrations of selenium (Se) and TotHg in birds but not in zooplankton, suggesting the importance of Se in Hg detoxification for individuals with high Hg concentrations.

Key Words: Methylmercury, Trophic magnification, Bioaccumulation, Arctic, Food Web

Introduction

Mercury (Hg) is a potentially toxic element entering the biosphere from natural and anthropogenic sources. The awareness of Hg as a threat to human and environmental health has led to international agreements to reduce emissions, such as the Minamata Convention on Mercury of the United Nations Environmental Programme (UNEP), agreed at the fifth session of the Intergovernmental Negotiating Committee in Geneva, Switzerland in 2013. However, discharges prevail and current anthropogenic sources account for approximately 30% of annual Hg-emissions to air, while approximately 60% is from re-emissions of previously released mercury [1]. Gold mining and coal combustion account for the largest proportions of anthropogenic emissions [2].

In aquatic systems, anoxic conditions favour the bacterial transformation of inorganic mercury to methylmercury [3]. Methylmercury (MeHg) is the most toxic form of Hg, and has a greater potential for bioaccumulation than inorganic mercury. In marine ecosystems, organisms at the top of food chains are especially exposed, due to the biomagnifying behaviour of methylmercury [4]. Furthermore, there is some evidence of higher biomagnification of mercury in food webs of Northern environments [5].

MeHg binds to sulfhydryl -groups of amino acids, which are the building stones of proteins [6]. Methylmercury is also readily absorbed from the gastrointestinal tract (90-95%) and crosses the blood brain-barrier [6]. Toothed wales (Odontoceti) appear to be a particularly vulnerable group, accumulating high concentrations of mercury in the central nervous system, leading to neurochemical effects [7]. Other adverse effects of MeHg include cardiovascular and reproductive effects, as well as impaired immune function [6].

Correlating concentrations of mercury and selenium has been observed in for instance mammals and birds, and it has been suggested that selenium plays a protective role against the toxic effects of inorganic and organic mercury [e.g. 8]. The mechanism of Se mediated detoxification of mercury in organisms is not fully understood, but may be related to synthesis of metal binding proteins or binding of Hg as insoluble selenide compounds [8, 9]. Potential Hg-Se compounds that have been suggested responsible for the antagonism include bis[methylmercuric]selenide, methylmercury selenocysteinate, selenoprotein P-bound HgSe clusters and the biominerals $HgSe_xS_{1-x}$ [9].

The Intergovernmental Panel on Climate Change (IPCC) predicts prospective climatic changes and consequences for the ecosystem that will occur fastest and with largest magnitude in Polar Regions [10]. Changes in climatic parameters may affect mercury transport, speciation and cycling in the Arctic [11]. Furthermore, primary productivity and food web energetics may be affected by climate changes [12], which may impact the trophic transfer of mercury. Emitted anthropogenic gaseous elemental Hg enters the Arctic from lower latitudes by long-range transport (in the atmosphere and the oceans; [13]). A net loss of gaseous mercury from the atmosphere to snow surface in the Arctic during spring has been shown, and global atmospheric Hg modelling indicates that the Arctic is a sink for Hg [14]. Concentrations of Hg in some Arctic marine organisms are currently approximately a factor of 12 higher than in pre-industrial times [2].

There are few studies pertaining to trophic transfer of MeHg, specifically, from the Svalbard area (Norwegian Arctic; [15]). The main objective of the present study was to quantify the biomagnification of MeHg in an Arctic pelagic food web, comprising species of

zooplankton, fish and seabirds (specified below) from the Kongsfjorden system (northwest Spitzbergen, Svalbard, Norway). Furthermore, an objective was to elucidate possible seasonal changes in MeHg biomagnification. The biomagnification was quantified by use of Trophic Magnification Factors (TMFs) that give the factor of increase in concentrations of contaminants per trophic position. TMFs have recently been amended to Annex XIII of the Regulation of the European parliament and of the Council on the Registration, Evaluation, Authorization and Restriction of Chemicals (REACH; [16]) for possible use in weight of evidence assessments of the bioaccumulative potential of chemicals as contaminants of concern. A second order objective was to quantify the relationship between total mercury and methylmercury, as well as between total mercury and selenium in the food web, to better understand mercury dynamics and the role of Se in Hg detoxification, respectively.

Material and Methods

Study site and sampling

Seabirds, fish, and zooplankton were collected in the Kongsfjorden system, northwest Spitzbergen, Svalbard, Norway 12th to 18th of May, 26th to 29th of July and 1st to 10th of October, 2007, during three cruises with R/V Lance and R/V Jan Mayen. Kongsfjorden (79°N, 12°E) is an open fjord system and the sill-less entrance facilitates exchange of Atlantic and Arctic water masses across the shelf-fjord boundary, which affects the physical and biological environment of the fjord [17].

Adult black legged kittiwake (*Rissa tridactyla*) and little auk (*Alle alle*), were collected with a shotgun in the inner to middle part of the fjord, by permission from the Governor of Svalbard. Polar cod (*Boregadus saida*), and capelin (*Mallotus villosus*) were caught by

gillnets (mesh size: 10, 12.5, 15, 18.5, 22, 26, 35, and 45mm divided into five sections each 5m and 1.5m high, to a total length of 40 m). Zooplankton (copepods: *Calanus hyperboreus*, *C. glacialis*, *C. finmarchicus*; krill/euphausiids: mostly *Thysanoessa inermis*; amphipods: *Themisto abyssorum* and *T. libellula*) were collected at two stations in Kongsfjorden, one in the middle of the fjord (inner station; 78°96 N, 11°94 E) and one outside on the shelf break (outer station; 78°94 N, 8°54 E; [18]). Zooplankton were collected by use of WP-3 (1000 mm mesh, 1 m^2 opening) and MIK (Method Isaac Kid; mesh size 1000 mm and 500 mm at the end, 3.14 m^2 opening) nets. Samples were taken from the entire water column (depth at inner and outer stations were ~330 m and ~290 m, respectively; hauling speed ~1 m/s). Live zooplankton specimens were quickly sorted by species (species specific samples of several pooled individuals, except for some samples sorted to genus; *Calanus sp.*) and stored at -20 °C until preparation for analyses of mercury (Hg), selenium (Se), methylmercury (MeHg) and stable isotopes of nitrogen (a smaller sub-sample for the latter). Biometric measures of seabirds and fish were taken prior to dissection (Supplemental Data, Table S1). Pectoral muscle of birds was analyzed for (organo-)metals and stable isotopes. Muscle tissue of fish was analyzed for MeHg and stable isotopes (TotHg and Se not analysed in fish, i.e. polar cod and capelin).

Element analysis

The element analyses were conducted at the Norwegian University of Science and Technology (NTNU), Norway. The samples were lyophilized for 24 h prior to digestion [19]. Dry samples (~0.15 g) were transferred to PTFE-vials (18 mL) and added ultrapure water and nitric acid (4.2 g; HNO_3; Scanpure/ultrapure grade), before digestion by use of a high pressure microwave emitter (Milestone Ultra Clave, EMLS). Subsequently, samples were diluted in

ultrapure water to a final volume of 60 mL (0.6 M HNO_3). Total Hg and Se were determined by high resolution inductively coupled plasma mass spectrometer (HR-ICP-MS; Thermo Finnigan model Element 2 instrument), with instrument settings as previously described [20]. No concentrations were below the limit of detection (Hg: 0.24 ng/g dry wt.; Se: 60 ng/g dry wt.). The average relative standard deviations (RSD) of multiple scans were below 3 % for both elements. Blank samples and the standard reference materials Bovine liver (National Institute of Standards and Technology; NIST 1577b), Oyster tissue (NIST 1566b) and Chicken (National Research Center of Certified Reference Materials; GBW 10018) were included (n>6) for quality assurance/quality control (QA/QC). The recovery of Se was 114, 123 and 102% in bovine liver, chicken and oyster, respectively. Mercury recovery was only assessed in oyster, and was 105% [19].

Methylmercury analysis

The MeHg analyses were conducted at the Norwegian Institute for Water Research (NIVA). All samples were extracted/analyzed as previously described [21] by use of an acid extraction method based on Hintelmann and Nguyen [22]. Samples (≥0.03 g) were added 10 mL 30% HNO_3 and heated at 60°C overnight (~15 h). Prior to analysis, the extraction solution was added 10 mL deionized water, and thereafter 0.050 mL of the solution was neutralized with 0.050 mL 15% KOH and ethylated before purge/trap and gas chromatography with cold vapor atomic fluorescence spectrometry (GC-CVAFS) analysis and detection based on USEPA Method 1630 [23]. Automated systems, standardized for MeHg, were used for analysis (Brooks Rand Labs MERX automated systems with Model III AFS Detector). For every run of MeHg analysis (n = 30) QA/QC measures included method blanks (n = 4), sample duplicates (n = 3), matrix spikes (n = 3) and certified standard materials (CRMs; n = 6). The certified MeHg concentrations of the CRMs used were 0.355 ±

0.056 mg/kg , 0.152 ± 0.013 mg/kg and 28.09 ± 0.31 µg/kg for DORM- 3 (fish protein; National Research Council of Canada, CNRC), TORT-2 (lobster hepatopancreas; CNRC) and SRM-2976 (mussel tissue; NIST), respectively. Samples that were analyzed in duplicates were also used for matrix spike samples. Samples chosen for matrix spiking were added 1000 pg (1.0–100 ng/g; 0.1 mL of 10.0 ng/mL MeHg hydroxide; MeHgOH) or 10 000 pg (100–1000 ng/g; 1.0 mL of 10.0 ng/ mL MeHgOH) depending on the concentration in the biological sample. Concentrations of MeHg in blank digestions correspond to a method detection limit (MDL) of 1 ng/g dry wt. or better (3 standard deviations of blank concentrations). The actual MDL will vary depending on the weight of sample available for analysis, but are typically in the range of 0.2 – 1.0 ng/g dry wt. for samples weights (0.03 – 0.1 g) included in this study. MeHg recovery of matrix spikes (75 – 125 %) and CRM (0.299 – 0.411 mg/kg, 139 – 165 mg/kg and 27.78 – 28.40 µg/kg for DORM-3, TORT-2 and SRM-2976, respectively) were within expected ranges. The RPD between duplicate samples was found to be satisfactory (< 20 %). If QA/QC measures were not met, samples were re-analyzed.

Stable isotope analysis

The stable isotope analyses were conducted at the Institute of Energy Technology at Kjeller, Norway, as previously described [24]. Prior to analysis, removal of lipids was performed by Soxhlet extraction. Samples (900 – 1500 µg; Mettler Toledo MT5, precision ±0.001 mg) were loaded into tin cups (9 × 15 mm) and were analyzed on a Micromass Optima Isotope Ratio Mass Spectrometers (IRMS; Waters). Stable isotope ratios were expressed in δ notation as the deviation from standard in ‰, according to:

$$\delta^{15}N‰ = [(R_{sample}/R_{standard}) - 1] \times 1000 \quad \text{(Eqn. 1)}$$

where R is the molar ratio of $^{15}N:^{14}N$ in the sample and in standard, respectively. Atmospheric air was used as standard for isotopic ratios of nitrogen. Replicate measurements of internal laboratory standards (muscle tissue of fish) are done routinely and were performed with the samples. This internal standard has been calibrates against the reference standards IAEA-N-1 and IAEA-N-2 (International Atomic Energy Agency) and the mean value in 2008 was $\delta^{15}N_{AIR}$= 11.63‰ ±0.20 (1σ). The mean value for the present study was $\delta^{15}N_{AIR}$= 11.62‰ ±0.16 (1σ). Blanks run routinely generally showed ~10 µg N.

Trophic position (TP) was calculated for each species relative to the copepod *C. finmarchicus* in the same season (May, July or October). *C. finmarchicus* is a primary consumer and therefore is defined as inhabiting TP = 2. TP was calculated by assuming that isotopic enrichment was constant for each trophic step and of the order 3.8‰ [19, 24-27].

$$TP_{consumer} = 2 + (\delta^{15}N_{consumer} - \delta^{15}N_{C.\ finmarchicus})/3.8 \quad \text{(Eqn. 2)}$$

where $\delta^{15}N_{consumer}$ is the species in question and $\delta^{15}N_{C.\ finmarchicus}$ is the stable isotope ratio found in *C. finmarchicus* (in the same season).

However, studies on piscivorous birds have indicated that the $\delta^{15}N$ isotopic fractionation between bird diet and muscle tissue is less than that derived for the other trophic steps, and according to Mizutani et al. [28], a bird diet-muscle isotopic fractionation factor of 2.4‰ is appropriate. Thus, Equation. 2 is then modified to:

$TP_{bird} = 3 + (\delta^{15}N_{bird} - (\delta^{15}N_{C.\ finmarchicus} + 2.4))/3.8$ (Eqn. 3)

Data treatment and statistical methods

Statistical analysis (linear regressions; general linear models) was performed with the use of Statistica software (Ver 11; Statsoft). A significance level of $\alpha = 0.05$ was chosen.

The trophic magnification factor (TMF) was calculated as the antilogarithm (base 10) of the slope (*b*) of the linear regression between $\log_{10}$ concentration (dry wt.) and the trophic position (TP) of the sample/species in question:

$\text{Log}_{10}\ \text{Concentration} = a + b\text{TP}$ (Eqn. 4)

$\text{TMF} = 10^{b}$ (Eqn. 5)

Results and Discussion

General observations

The highest concentrations of total mercury (TotHg) and methylmercury (MeHg) were found in birds (kittiwake and little auk), while the lowest concentrations were measured in zooplankton (Table 1; Figure 1). General linear models with ($\log_{10}$) concentrations of MeHg and TotHg, and amount of MeHg relative to TotHg (%), respectively, as response variables, and season (May, July and October) and food web compartment (bird, fish [applicable only to MeHg] and zooplankton) as predictors, showed all predictors significant ($p<0.0007$). The concentrations of TotHg varied somewhat between seasons, most noticeable for the birds

(Table 1). In kittiwake, concentrations decreased from May, through July, to October [19]. Similarly, in little auk concentrations were lower in July, than in May (little auk were not available in Kongsfjorden in October). The concentrations of MeHg in the birds also decreased from May to July, and to October for kittiwake. Thus, the relative amount of MeHg (MeHg as % of TotHg) in the birds was relatively stable through seasons (Table 1). The zooplankton showed a higher variation in the relative amount of MeHg (Table 1). The concentrations of TotHg and MeHg in the organisms were mostly within the same order of magnitude as in previous studies from the Arctic [15, 29-31].

A general linear model was used to analyze the effect of trophic position (TP) and season (May, July and October) on (Log_{10}) MeHg concentrations:

$$Log_{10}\,[MeHg] = a + bTP + c_i season_i + d_i TP \times season_i + \varepsilon \qquad \text{(Eqn. 6)}$$

where a to d are constants and ε is the error term (*i* pertains to the three different seasons). In addition to significant TP and seasonal terms, the interaction TP×season was significant, indicating different increase in Hg concentration with trophic position (and thus different TMFs) among seasons ($p<0.015$; $TMF_{May} = 24.4$, $TMF_{July} = 15.0$, $TMF_{October} = 8.8$). Krill was only sampled in May and July, and if krill is omitted from the analysis (see below), the interaction term would not be significant, although still with a fairly low *p* value ($p=0.065$; $TMF_{May} = 15.5$, $TMF_{July} = 13.3$, $TMF_{October} = 8.8$).

As for mercury, the concentrations of Se in the birds were also reduced from May to July (and to October for kittiwake; Table 1).

Lower TotHg and MeHg concentrations in birds in July than May (Table 1; $p<0.000001$ for both TotHg and MeHg in kittiwake; $p<0.0002$ and $p<0.0007$ for TotHg and MeHg, respectively, in Little Auk) may suggest that kittiwakes changed from a diet dominated by fish to a diet predominantly constituted of invertebrates (as discussed by Øverjordet et al. [19]). It may partly also be a result of the trophic position of the birds declining from May to July (Table 1; Figure 1; $p<0.000001$ both for kittiwake and for little auk), which in turn may partly be attributed to a shift (increase) in the $\delta^{15}N$ baseline (*Calanus finmarchicus*, defined as TP 2 at all seasons). On the other hand, the lower concentrations in birds, later in the year may also be a result of increased elimination of mercury, bound to feather keratin, through molting (full molt occurring June to July) [19]. Keratin is a group of fibrous structural proteins abundant in feathers, rendering feather growth as an excretion pathway of Hg [8]. Female birds may also excrete Hg via their eggs (egg-laying occurring in June) [32].

Biomagnification

Concentrations (log_{10}-transformed) of MeHg in organisms of the Kongsfjorden system (all seasons included) showed a significant linear relationship with trophic position ($p<0.0001$; $R^2=0.68$; Figure 1). Krill showed somewhat deviating MeHg concentrations and trophic positions from the other organisms (in May; Figure 1). Omitting krill from the regression would change the intercept of the regression line, but leave the slope nearly unchanged (Figure 1), as well as increase the goodness-of-fit ($R^2=0.84$). The slope of the regression corresponded to a trophic magnification factor (TMF) of 8.7 (8.6 without krill).

The concentrations of MeHg in the food web were highly correlated with the concentrations of TotHg (Figure 2; $p<0.0001$; $R^2=0.96$), indicating an average fraction of 63% MeHg (of TotHg; deduced from the slope of the regression) in the food web. As mentioned (Table 1),

this fraction was generally slightly higher in birds, than in zooplankton (p<0.0007; but note that TotHg was not quantified in fish). Since MeHg has a higher bioaccumulative potential than inorganic Hg, it could be expected that this fraction would increase with higher trophic level [5, 33, 34]. The linear relationship between MeHg and TotHg entails a similar TMF for TotHg and MeHg (TMF = 8.8 for TotHg; 8.7 without krill).

The observed TMFs for MeHg and TotHg in the present study are higher (greater biomagnification) than previously observed in the Arctic, and especially higher than observed at lower latitudes [e.g. 15, 30, 33, 35, 36]. Examples of findings from different geographic/climate zones are as follows:

Jæger et al. [15] showed a TotHg TMF = 4.87 for fish and sea birds (muscle) and a MeHg TMF = 4.26 for fish and sea birds (liver) in Kongsfjorden (Svalbard, Norwegian Arctic). It must be noted that concentrations of Hg (total and methyl) are higher in bird liver, than muscle [15, 19]. In a study from the Northwater Polynya, Baffin Bay, Canada, Campbell et al. [30] quantified TotHg and MeHg biomagnification in a food web including ice algae, zooplankton, fish and pinnipeds. They found a concentration increase per trophic level corresponding to TMFs of 5.6 and 7.0 for TotHg and MeHg, respectively (assuming a $\delta^{15}N$ enrichment per integer trophic step (ΔN) of the order 3.8‰, as in the present study). Furhermore, Atwell et al. [29] studied TotHg accumulation in 27 Arctic species from the Lancaster Sound, northwest Territories, Canada, with samples ranging from particulate organic matter through invertebrates, fish, sea birds, marine mammals (cetaceans and pinnipeds) and polar bear (*Ursus maritimus*). They found a concentration increase per trophic level corresponding to a TMF of 5.8 (assuming ΔN = 3.8), while Lavoie et al. [31] found a concentration increase per trophic level corresponding to TMFs of 4.43 and 7.82 for TotHg and MeHg, respectively (assuming ΔN = 3.8) in a Gulf of St. Lawrence (Canada) food web

(particulate organic matter, invertebrates, fish and seabirds). Riget et al. [27] reported concentration increases per trophic level corresponding to TMFs of 2.00 and 3.63 for TotHg and MeHg, respectively (assuming $\Delta N = 3.8$), in a central West Greenland food web including fish, sea birds and marine mammals. In a temperate estuary (Masan Bay, Korea), Kim et al. [36] studied biomagnification of mercury in a benthic food web comprised of invertebrates and fish. They found a concentration increase per trophic level corresponding to TMFs of 2.8 and 4.3 for TotHg and MeHg, respectively (assuming $\Delta N = 3.8$). In a sub-tropical food web (fish at different trophic levels), Cheng et al. [33] found TMFs = 2.32-2.60 for MeHg and TMFs = 1.94-2.03 for TotHg, also indicating an increased fraction of MeHg with higher trophic level. In another subtropical coastal food web (Southwest Florida, US), comprising 57 species (invertebrates and fish), Thera and Rumbold [37] found a TMF = 5.05 for TotHg. In a study of different fish from a tropical marine ecosystem in the Arabian sea, Al-Reasi et al. [35] found a concentration increase per trophic level corresponding to TMFs of 3.1 and 3.4 for TotHg and MeHg, respectively (assuming $\Delta N = 3.8$), while Kehrig et al. [38] found a TMF for TotHg of 5.4 in a Brazilian coastal food web comprised of invertebrates, fish and ceteceans.

The apparent latitude dependence of the magnitude of Hg accumulation, showing higher biomagnification at higher latitude, is in accordance with findings of Lavoie et al. [5], who conducted a worldwide meta-analysis of mercury biomagnification in aquatic food webs (fresh water and marine), compiling data from 69 studies (analyzing TotHg or MeHg), comprising 205 aquatic food webs. They found a mean TMF for TotHg of 4.7 (± 4.7), and for MeHg a mean TMF = 8.1 (± 7.2). For marine locations, the mean TMFs were 6.2 (± 4.1) and 7.0 (± 4.9) for TotHg and MeHg, respectively. The MeHg biomagnification was, on average, a factor of 1.5 higher than for TotHg, and the biomagnification of both MeHg and TotHg was

significantly and positively correlated with latitude. Hence, their results suggested that the biomagnification of mercury is highest in cold and low productivity systems, though for reasons much still unknown. They argued, however, that several mechanisms pertaining to temperature may be important [5]. Warmer temperatures induce growth, which leads to growth dilution. Additionally, colder temperatures lead to slower excretion rates. Furthermore, these authors hypothesized that less complex food webs in the north could lead to higher bioaccumulation, since a larger choice of prey organisms at lower latitudes may potentially reduce the efficiency of trophic mercury transfer. Al-Reasi et al. [35] also argued that mercury biomagnification was lower in tropical system subject of their study, compared to temperate and Arctic ecosystems, likely due to diverse diet items with different Hg content, rendering large variation in the body burden of fish species with similar trophic position.

The ecology and physiology of the species comprising the food web in question may also have large influence on the biomagnification. For instance, Lavoie et al. [31] found that the biomagnification was greater for pelagic and benthopelagic species, compared to benthic species, and suggested that Hg is more bioavailable to benthic species at the base of the food web, but trophic transfer efficiency is higher in pelagic and benthopelagic species. Kim et al. [36] also found that MeHg concentrations were lower in benthic-feeding species, than in pelagic-feeding species, but attributed this to possible biodilution at the base of the benthic food web, as a consequence of higher carbon turnover rates, suggesting that the mercury dynamics at the base of the food web is likely of high importance. High biomagnification of mercury in Arctic pelagic systems, such as that in the present study also corroborates these observations.

Furthermore, according to a review by Lehnherr [4], in Arctic marine ecosystems, increasing evidence suggest Hg methylation in the water column, rather than in sediments, as the primary source of MeHg. It has also been suggested that dimethylmercury (DMHg; the other naturally occurring organic Hg species, only present in low concentrations in the deep areas of the oceans), might be an important, mobile pre-cursor for MeHg in the Arctic marine environment [39].

Another interesting observation with regard to methylation of mercury was done by Pućko et al. [40], who studied transformation of mercury at the base of the Arctic food web and observed that the copepod *Calanus hyperboreus* shifts Hg from mainly inorganic forms in the pelagic particulate organic matter (POM) and seawater to primarily organic forms in their tissue. Furthermore, they observed that the dietary intake of MeHg could supply only ~30% of the MeHg body burden, suggesting transformation within *C. hyperboreus*, possibly mediated by microbes in the gut, or bioconcentration from ambient seawater being of high importance. They argued that acidic and suboxic/anoxic conditions in the gut of *C. hyperboreus* promote mercury methylation by iron dissolution and anaerobic microbial activities. Thus, they hypothesize that the lowest trophic levels of Arctic marine food webs could present a very important point of in vivo Hg transformation, shifting the MeHg:TotHg ratio towards higher values.

Wang et al. [34] also reported differences in the relative amount of MeHg (MeHg as % of TotHg) suggesting biomagnification of MeHg between different size classes of zooplankton. Atwell et al. [29], on the other hand, found no biomagnification among invertebrates (as a subset of the sampled food web), suggesting different transfer mechanisms for mercury at different trophic levels.

A physiological trait of the organisms in the food web, which may have an impact on biomagnification is the issue of thermoregulation. Since homeotherms (or more specifically endotherms) have higher energy requirement and lower food conversion efficiencies than poikilotherms, their higher Hg intake may theoretically lead to larger biomagnification in food webs where homeotherms are included, compared to food webs where homeotherms are not considered [26, 31]. The inclusion of birds in the food web of the present study may therefore be partly responsible for the high TMFs. Higher biomagnification in food webs where homeotherms are included, compared to food webs where homeotherms are not considered is also observed for persistent organic pollutants [e.g. 24]. Lavoie et al. [5], however, found that neither the species composition nor the percentage of homeotherms in food webs affected the magnitude of the biomagnification of mercury. In the study by Campbell et al. [30] from the Northwater Polynya, TotHg and MeHg biomagnification was also lower than in the present study (a concentration increase per trophic level corresponding to TMFs of 5.6 and 7.0 for TotHg and MeHg, respectively, assuming $\Delta N = 3.8$), despite inclusion of substantially more homoeothermic species/samples.

Besides the homeothermy, another influential property of birds is their migratory behavior, since they experience spatiotemporal variations in contaminant exposure, impeding sampling of a static food web [41]. In the study by Atwell et al. [29], vertebrates also had, in general, wider ranges of mercury concentrations than invertebrates, possibly linked to the fact that they are migratory and have larger foraging ranges. The authors therefore argued that high trophic level organisms thus also may be exposed to different levels of dietary mercury during different seasons. Fort et al. [42] also showed that little auks were more contaminated with Hg when outside the Arctic breeding area/season. As mentioned, the concentrations of TotHg in

423 the birds of the present study changed with season (Table 1; Figure 1). Furthermore,
424 segregating the data on season produced significant differences in TMFs (a trend towards
425 lower TMF in October, than in May and July; see above).
426
427 *Selenium*
428 Mercury is not an essential element and is not maintained at a stable level by homeostasis,
429 while Se, being an essential trace element, must be present at a certain level to maintain
430 physiological functions. As mentioned, it has been suggested that selenium plays a protective
431 role against the toxic effects of mercury, although the mechanism is not fully understood. As
432 such, concentrations of mercury and selenium are often correlated in organisms [e.g. 8]. A
433 significant linear relationship was observed between the (log_{10}-transformed) concentrations of
434 Se and TotHg in birds (all individuals of both species, all seasons pooled; $p<0.00001$,
435 $R^2=0.61$; Figure 3). In contrast, the same relationship was not found within the zooplankton
436 group (Figure 3), in which concentrations of Hg were substantially lower than in birds.
437 Looking at kittiwake, separately, the relationship between Se and TotHg was also significant
438 (all seasons pooled; $p<0.00001$, $R^2=0.61$; [19]).
439
440 Kim et al. [8] found a clear relationship between the concentrations of TotHg and Se in the
441 liver of sea bird individuals with TotHg concentrations above a certain level, while such a
442 relationship was unclear in other individuals with lower Hg levels, suggesting the importance
443 of Se in Hg detoxification for individuals with high Hg concentrations. It is known that Se
444 mitigate Hg-toxicity through formation of Hg-Se complexes at Se:Hg molar ratios above 1
445 [9]. Looking at Kittiwakes from October, separately, when Hg concentrations were lowest, no
446 relationship could be observed between concentrations of Se and TotHg (Figure 3). In fact,

when seasons were addressed separately, such a relationship could only be observed in May, when Hg concentrations were highest ($p<0.05$, $R^2=0.40$).

Bjerregaard et al. [43] found that dietary exposure of selenium to the brown shrimp (*Crangon crangon*) enhanced the elimination of MeHg, and that the effect was dose dependent, suggesting that selenium present at lower trophic levels of marine food webs may play an important role in inhibiting MeHg accumulation. Thus, no observed relationship between concentrations of Se and TotHg in zooplankton may be a consequence of too low concentrations of Hg, and not that Se plays a less important role in zooplankton. It is also known from multi-generational studies of cladocerans that selenium deficiency has a negative effect on fertility and development [44], suggesting the importance of Se for prevention of oxidative damage.

Concluding remarks

As expected, tissue concentrations of MeHg increased with increasing trophic level in the food web (biomagnification) in an exponential manner, however, at greater rates than observed in several earlier studies, especially at lower latitudes. There was strong correlation between the MeHg and the TotHg content through the food web as a whole, thus although MeHg has a much higher bioaccumulative potential than inorganic mercury, measures of MeHg and TotHg depict similar trends. It must be noted, however, that TotHg was not quantified in fish. The concentration of MeHg in kittiwake decreased from May (through July) to October, contributing to seasonal differences in trophic magnification factors. The ecology and physiology of the species (e.g. pelagic versus benthic species, homeotherms versus poikilotherms) comprising the food web in question may also have large influence on the magnitude of the biomagnification. A significant linear relationship was observed between

concentrations of Se and TotHg in birds but not in zooplankton, suggesting the importance of Se in Hg detoxification for birds with high Hg concentrations.

Acknowledgements

This work was mainly funded through the Fram Centre flagship "Hazardous substances – effects on ecosystem and health". The samples were collected through the Project Contaminants in Polar Regions (COPOL; 176073/S30), funded by the Norwegian Research Council via the International Polar Year (IPY) Program. COPOL was a large collaboration between the following institutions and thanks are due to everyone involved: the Norwegian Polar Institute, the Norwegian Institute for Water Research (NIVA), Akvaplan-niva, the Norwegian Institute for Air Research (NILU), the Norwegian Institute for Nature Research (NINA) and the Norwegian University for Science and Technology (NTNU). Ingeborg G. Hallanger, Merete Schøyen and Paul Renaud are thanked for outstanding fieldwork. Additional support by the Norwegian Research Council, through Grant number 234388 (COCO).

Supplemental data

Table S1. Biometric measures for birds

References

[1] Amos HM, Jacob DJ, Streets DG, Sunderland EM. 2013. Legacy impacts of all-time anthropogenic emissions on the global mercury cycle. *Global Biogeochem Cy* 27:410-421.

[2] UNEP. 2013. Global mercury assessment 2013: Sources, emissions, releases and environmental transport. UNEP Chemicals Branch, Geneva, Switzerland.

497 [3] Morel FMM, Kraepiel AML, Amyot M. 1998. The chemical cycle and
498 bioaccumulation of mercury. *Annu Rev Ecol Syst* 29:543-566.
499 [4] Lehnherr I. 2014. Methylmercury biogeochemistry: a review with special reference to
500 Arctic aquatic ecosystems. *Environ Rev* 22:229-243.
501 [5] Lavoie RA, Jardine TD, Chumchal MM, Kidd KA, Campbell LM. 2013.
502 Biomagnification of mercury in aquatic food webs: a worldwide meta-analysis. *Environ Sci*
503 *Technol* 47:13385-13394.
504 [6] Wolfe MF, Schwarzbach S, Sulaiman RA. 1998. Effects of mercury on wildlife: A
505 comprehensive review. *Environ Toxicol Chem* 17:146-160.
506 [7] Dietz R, Sonne C, Basu N, Braune B, O'Hara T, Letcher RJ, Scheuhammer T,
507 Andersen M, Andreasen C, Andriashek D, Asmund G, Aubail A, Baagoe H, Born EW, Chan
508 HM, Derocher AE, Grandjean P, Knott K, Kirkegaard M, Krey A, Lunn N, Messier F, Obbard
509 M, Olsen MT, Ostertag S, Peacock E, Renzoni A, Riget FF, Skaare JU, Stern G, Stirling I,
510 Taylor M, Wiig O, Wilson S, Aars J. 2013. What are the toxicological effects of mercury in
511 Arctic biota? *Sci Tot Environ* 443:775-790.
512 [8] Kim EY, Saeki K, Tanabe S, Tanaka H, Tatsukawa R. 1996. Specific accumulation of
513 mercury and selenium in seabirds. *Environ Pollut* 94:261-265.
514 [9] Khan MAK, Wang F. 2009. Mercury-selenium compounds and their toxicological
515 significance: toward a molecular understanding of the mercury-selenium antagonism. *Environ*
516 *Toxicol Chem* 28:1567-1577.
517 [10] IPCC. 2001. Intergovernmental Panel on Climate Change Third assessment report:
518 Climate change 2001—Impacts, adaptation and vulnerability. Cambridge University Press,
519 Cambridge, UK.

520 [11] Stern GA, Macdonald RW, Outridge PM, Wilson S, Chetelat J, Cole A, Hintelmann
521 H, Loseto LL, Steffen A, Wang F, Zdanowicz C. 2012. How does climate change influence
522 Arctic mercury? *Sci Tot Environ* 414:22-42.
523 [12] Wassmann P, Duarte CM, Agusti S, Sejr MK. 2011. Footprints of climate change in
524 the Arctic marine ecosystem. *Global Change Biol* 17:1235-1249.
525 [13] Dastoor AP, Durnford DA. 2014. Arctic ocean: is it a sink or a source of atmospheric
526 mercury? *Environ Sci Technol* 48:1707-1717.
527 [14] Ariya PA, Dastoor AP, Amyot M, Schroeder WH, Barrie L, Anlauf K, Raofie F,
528 Ryzhkov A, Davignon D, Lalonde J, Steffen A. 2004. The Arctic: a sink for mercury. *Tellus B*
529 56:397-403.
530 [15] Jaeger I, Hop H, Gabrielsen GW. 2009. Biomagnification of mercury in selected
531 species from an Arctic marine food web in Svalbard. *Sci Tot Environ* 407:4744-4751.
532 [16] EC. 2006. *Regulation (EC) No. 1907/2006 of the European Parliament and of the*
533 *Council of 18 December 2006.* European Commission, Brussels, Belgium.
534 [17] Hop H, Pearson T, Hegseth EN, Kovacs KM, Wiencke C, Kwasniewski S, Eiane K,
535 Mehlum F, Gulliksen B, Wlodarska-Kowalczuk M, Lydersen C, Weslawski JM, Cochrane S,
536 Gabrielsen GW, Leakey RJG, Lonne OJ, Zajaczkowski M, Falk-Petersen S, Kendall M,
537 Wangberg SA, Bischof K, Voronkov AY, Kovaltchouk NA, Wiktor J, Poltermann M, di
538 Prisco G, Papucci C, Gerland S. 2002. The marine ecosystem of Kongsfjorden, Svalbard.
539 *Polar Res* 21:167-208.
540 [18] Hallanger IG, Ruus A, Herzke D, Warner NA, Evenset A, Heimstad ES, Gabrielsen
541 GW, Borga K. 2011. Influence of season, location, and feeding strategy on bioaccumulation
542 of halogenated organic contaminants in Arctic marine zooplankton. *Environ Toxicol Chem*
543 30:77-87.

544 [19] Øverjordet IB, Kongsrud MB, Gabrielsen GW, Berg T, Ruus A, Evenset A, Borgå K,
545 Christensen G, Jenssen BM. 2015. Toxic and essential elements changed in black-legged
546 kittiwakes (*Rissa tridactyla*) during their stay in an Arctic breeding area. *Sci Tot Environ*
547 502:548-556.
548 [20] Sormo EG, Ciesielski TM, Overjordet IB, Lierhagen S, Eggen GS, Berg T, Jenssen
549 BM. 2011. Selenium moderates mercury toxicity in free-ranging freshwater fish. *Environ Sci*
550 *Technol* 45:6561-6566.
551 [21] Braaten HFV, Harman C, Øverjordet IB, Larssen T. 2014. Effects of sample
552 preparation on methylmercury concentrations in Arctic organisms. *Int J Environ An Ch*
553 94:863-873.
554 [22] Hintelmann H, Nguyen H. 2005. Extraction of methylmercury from tissue and plant
555 samples by acid leaching. *Anal Bioanal Chem* 381:360-365.
556 [23] USEPA (United States Environmental Protection Agency). 1998. *Method 1630:*
557 *Methylmercury in water by distillation, aqueous ethylation, purge and trap, and cold vapor*
558 *atomic fluorescence spectrometry*. United States Environmental Protection Agency - Office of
559 Water, Washington DC, USA.
560 [24] Hallanger IG, Warner NA, Ruus A, Evenset A, Christensen G, Herzke D, Gabrielsen
561 GW, Borga K. 2011. Seasonality in contaminant accumulation in Arctic marine pelagic food
562 webs using trophic magnification factor as a measure of bioaccumulation. *Environ Toxicol*
563 *Chem* 30:1026-1035.
564 [25] Hobson KA, Welch HE. 1992. Determination of trophic relationships within a high
565 arctic marine food web using $\delta^{13}C$ and $\delta^{15}N$ analysis. *Mar Ecol Prog Ser* 84:9-18.
566 [26] Hop H, Borga K, Gabrielsen GW, Kleivane L, Skaare JU. 2002. Food web
567 magnification of persistent organic pollutants in poikilotherms and homeotherms from the
568 Barents Sea. *Environ Sci Technol* 36:2589-2597.

569 [27] Riget F, Moller P, Dietz R, Nielsen TG, Asmund G, Strand J, Larsen MM, Hobson
570 KA. 2007. Transfer of mercury in the marine food web of West Greenland. *J Environ Monitor*
571 9:877-883.
572 [28] Mizutani H, Kabaya Y, Wada E. 1991. Nitrogen and carbon isotope compositions
573 relate linearly in cormorant tissues and its diet. *Isotopenpraxis* 27:166-168.
574 [29] Atwell L, Hobson KA, Welch HE. 1998. Biomagnification and bioaccumulation of
575 mercury in an arctic marine food web: insights from stable nitrogen isotope analysis. *Can J*
576 *Fish Aquat Sci* 55:1114-1121.
577 [30] Campbell LM, Norstrom RJ, Hobson KA, Muir DCG, Backus S, Fisk AT. 2005.
578 Mercury and other trace elements in a pelagic Arctic marine food web (Northwater Polynya,
579 Baffin Bay). *Sci Tot Environ* 351:247-263.
580 [31] Lavoie RA, Hebert CE, Rail J-F, Braune BM, Yumvihoze E, Hill LG, Lean DRS.
581 2010. Trophic structure and mercury distribution in a Gulf of St. Lawrence (Canada) food
582 web using stable isotope analysis. *Sci Tot Environ* 408:5529-5539.
583 [32] Lewis SA, Becker PH, Furness RW. 1993. Mercury levels in eggs, tissues, and
584 feathers of herring-gulls Larus-argentatus from the german wadden sea coast. *Environ Pollut*
585 80:293-299.
586 [33] Cheng Z, Liang P, Shao D-D, Wu S-C, Nie X-P, Chen K-C, Li K-B, Wong M-H.
587 2011. Mercury biomagnification in the aquaculture pond ecosystem in the Pearl River delta.
588 *Arch Environ Contam Toxicol* 61:491-499.
589 [34] Wang Q, Feng X, Yang Y, Yan H. 2011. Spatial and temporal variations of total and
590 methylmercury concentrations in plankton from a mercury-contaminated and eutrophic
591 reservoir in Guizhou province, China. *Environ Toxicol Chem* 30:2739-2747.

592 [35] Al-Reasi HA, Ababneh FA, Lean DR. 2007. Evaluating mercury biomagnification in
593 fish from a tropical marine environment using stable isotopes ($\delta^{13}C$ and $\delta^{15}N$). *Environ*
594 *Toxicol Chem* 26:1572-1581.
595 [36] Kim E, Kim H, Shin K-h, Kim M-s, Kundu SR, Lee B-g, Han S. 2012.
596 Biomagnification of mercury through the benthic food webs of a temperate estuary: Masan
597 Bay, Korea. *Environ Toxicol Chem* 31:1254-1263.
598 [37] Thera JC, Rumbold DG. 2014. Biomagnification of mercury through a subtropical
599 coastal food web off Southwest Florida. *Environ Toxicol Chem* 33:65-73.
600 [38] Kehrig HA, Seixas TG, Malm O, Di Beneditto APM, Rezende CE. 2013. Mercury and
601 selenium biomagnification in a Brazilian coastal food web using nitrogen stable isotope
602 analysis: A case study in an area under the influence of the Paraiba do Sul River plume. *Mar*
603 *Pollut Bull* 75:283-290.
604 [39] Hintelmann H. 2010. Organomercurials. Their formation and pathways in the
605 environment. *Met Ions Life Sci* 7:365-401.
606 [40] Pucko M, Burt A, Walkusz W, Wang F, Macdonald RW, Rysgaard S, Barber DG,
607 Tremblay JE, Stern GA. 2014. Transformation of mercury at the bottom of the Arctic food
608 web: an overlooked puzzle in the mercury exposure narrative. *Environ Sci Technol* 48:7280-
609 7288.
610 [41] Ofukany AFA, Hobson KA, Wassenaar LI. 2012. Connecting breeding and wintering
611 habitats of migratory piscivorous birds: implications for tracking contaminants (Hg) using
612 multiple stable isotopes. *Environ Sci Technol* 46:3263-3272.
613 [42] Fort J, Robertson GJ, Gremillet D, Traisnel G, Bustamante P. 2014. Spatial
614 ecotoxicology: migratory Arctic seabirds are exposed to mercury contamination while
615 overwintering in the Northwest Atlantic. *Environ Sci Technol* 48:11560-11567.

616 [43] Bjerregaard P, Christensen A. 2012. Selenium reduces the retention of methyl mercury
617 in the brown shrimp *Crangon crangon*. *Environ Sci Technol* 46:6324-6329.
618 [44] Elendt BP. 1990. Selenium deficiency in crustacea – An ultrastructural approach to
619 antennal damage in *Daphnia-magna* Straus. *Protoplasma* 154:25-33.
620

Figure Legends:

Figure 1. Trophic level (TL; estimated from $\delta^{15}N$) vs. Log_{10}-transformed concentrations of methylmercury (ng/g dry wt.) in the organisms from the pelagic food web of Kongsfjorden (Svalbard, Norwegian Arctic), sampled in 2007 (May, July and October). Data clustered by species/food web compartment:

a. Zooplankton *(Calanus finmarchicus*, *C. hyperboreus*, *C. glacialis*, *Themisto libellula*, *T. abyssorum*).

b. Krill (mostly *Thysanoessa inermis*)

c. Capelin (*Mallotus villosus*)

d. Polar cod (*Boreogadus saida*)

e. Little Auk (*Alle alle*)

f. Kittiwake (*Rissa tridactyla*; Data from Øverjordet et al. [19])

Regression lines for the linear regression including (solid line;

$Log_{10}[MeHg] = -1.189 + 0.9411 \times TL$; $p<0.0001$, $R^2=0.68$) and excluding (stippled line;

$Log_{10}[MeHg] = -1.0468 + 0.9363 \times TL$; $p<0.0001$, $R^2=0.84$) krill are depicted.

Figure 2. Total mercury (TotHg; ng/g dry wt.) vs. methylmercury (MeHg; ng/g dry wt.) in the organisms of the pelagic food web of Kongsfjorden (Svalbard, Norwegian Arctic), sampled in 2007 (May, July and October). $[MeHg] = 12.1973 + 0.6314 \times [TotHg]$; $p<0.0001$; $R^2=0.96$.

Figure 3. Concentrations of Selenium (Se; ng/g dry wt.; Log_{10}-transformed) vs. concentrations of total mercury (TotHg; ng/g dry wt.; Log_{10}-transformed) in birds (black

legged kittiwake, *Rissa tridactyla*, and little auk, *Alle alle*) and zooplankton (*Calanus finmarchicus*, *C. hyperboreus*, *C. glacialis*, *Themisto libellula*, *T. abyssorum* and krill/mostly *Thysanoessa inermis*) from the pelagic food web of Kongsfjorden (Svalbard, Norwegian Arctic), sampled in 2007 (May, July and October; season specified/clustered for the birds). (Kittiwake data from Øverjordet et al. [19]; $Log_{10}[TotHg] = -2.1123 + 1.2754\times Log_{10}[Se]$; $p<0.00001$, $R^2=0.61$).

SUPPLEMENTAL DATA:

Methylmercury biomagnification in an Arctic pelagic food web

Anders Ruus, *† Ida B. Øverjordet, ‡, § Hans Fredrik V. Braaten, † Anita Evenset, ‖, # Guttorm Christensen, ‖ Eldbjørg S. Heimstad, †† Geir W. Gabrielsen, ‡‡ Katrine Borgå, †, §§

† *Norwegian Institute for Water Research, Gaustadalléen 21, NO-0349 Oslo, Norway*

‡ *Norwegian University of Science and Technology, Department of Biology, NO-7491 Trondheim, Norway*

§ *SINTEF Materials and Chemistry, Marine Environmental Technology, Brattørkaia 17B, NO-7010 Trondheim*

‖ *Akvaplan-niva, Fram Centre, NO-9296 Tromsø, Norway*

University of Tromsø. The Arctic University of Norway, NO-9037 Tromsø, Norway

†† *Norwegian Institute for Air Research, Fram Centre, NO-9296 Tromsø, Norway*

‡‡ *Norwegian Polar Institute, Fram Centre, NO-9296 Tromsø, Norway*

§§ *University of Oslo, Department of Biosciences, PO Box 1066 Blindern, NO-0316 Oslo, Norway*

*Corresponding author: Anders Ruus

Phone: +47 98227786

Fax: +47 22185200

e-mail: anders.ruus@niva.no

Table S1. Biometric measures for birds (black legged kittiwake, *Rissa tridactyla*, and little auk, *Alle alle*) and fish (Polar cod, *Boregadus saida*, and capelin, *Mallotus villosus*) from Kongsfjorden (Svalbard, Norwegian Arctic), sampled in 2007 (May, July and October). Values are mean (and standard deviation).

Species	Season	n [a.]	Body mass (g)	Wing length (cm)	Gonys depth (mm)	Head-bill (mm)	Tarsus length (mm)
Kittiwake [b.]	May	10	428	31.8	11.0	89.6 [c.]	40.8 [d.]
		(4 M, 6 F)	(59)	(1.7)	(0.43)	(4.4)	(1.5)
	July	10	380	32.1	10.7	89.6	39.8
		(7 M, 3 F)	(35)	(0.8)	(0.37)	(3.3)	(1.7)
	October	10	438	31.2	10.2	92.1	38.9
		(8 M, 2 F)	(54)	(1.3)	(0.7)	(3.7)	(3.5)
Little Auk	May	10	160	12.7	7.8	52.9 [e.]	24.2 [f.]
		(7 M, 3 F)	(11)	(0.4)	(1.8)	(2.5)	(2.1)
	July	10	165	12.9	8.8	53.4	25.5
		(4 M, 6 F)	(10)	(0.4)	(0.4)	(1.5)	(0.7)

Species	Season	n	Body mass (g)	Length (cm)
Polar cod	July	5	11.8	12.8
			(1.7)	(0.8)
Capelin	July	8	9.6	12.4
			(1.5)	(0.6)

[a.] Total number of samples (as well as the number of males, M, and females, F); [b.] Data from Øverjordet et al. [1] (where data are reported by sex);
[c.] n = 8; [d.] n = 9; [e.] n = 8; [f.] n = 9.

References

[1] Øverjordet IB, Kongsrud MB, Gabrielsen GW, Berg T, Ruus A, Evenset A, Borgå K, Christensen G, Jenssen BM. 2015. Toxic and essential elements changed in black-legged kittiwakes (*Rissa tridactyla*) during their stay in an Arctic breeding area. *Sci Tot Environ* 502:548-556.