Temporal trend of mercury in relation to feeding habits and food availability in arctic foxes (*Vulpes lagopus*) from Svalbard, Norway

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HIGHLIGHTS

- We analyzed liver concentrations of THg in 109 arctic foxes from Svalbard.
- THg levels in arctic foxes increased with 7.2% per year from 1997 to 2014.
- The increase in THg per year is lower than in other Arctic regions.
- Terrestrial food consumption reduced THg levels in arctic foxes.

GRAPHICAL ABSTRACT
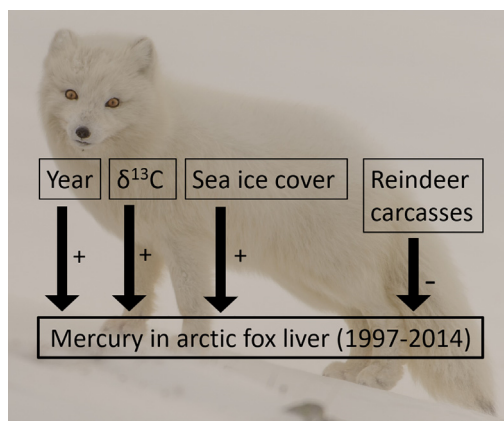
ABSTRACT

We investigated the temporal trend of mercury (Hg) in arctic foxes from Svalbard, Norway sampled in the period 1997–2014 ($n = 109$, from 11 trapping seasons). We used linear models to investigate the effect of trapping season, feeding habits ($\delta^{13}C$), food availability from marine and terrestrial ecosystems (reindeer carcasses and sea ice cover), sex, age and body condition on liver total Hg (THg) levels. Liver THg levels increased in arctic foxes with 7.2% (95% CI: 2.3, 9.6) per year when the concentrations were adjusted for variation of $\delta^{13}C$, sea ice cover, and reindeer carcasses, whereas the raw annual trend was 3.5% (CI: −0.11, 7.2). However, the THg levels in arctic foxes from Svalbard are still lower than other marine mammals. We also demonstrate that arctic fox terrestrial food consumption is important for lowering the overall THg levels in this species.

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1. Introduction

Mercury (Hg) in Arctic environments is of particular concern to wildlife because of its toxicity and ability to bioaccumulate and biomagnify in food webs (Douglas et al., 2011). Presently, over 90% of

Hg found in high trophic level Arctic species is derived from anthropogenic sources (Dietz et al., 2009). Although Hg emissions from Asia and Oceania increased in the period 1990–2010, the global Hg emissions have declined in the same period (Streets et al., 2009; Zhang et al., 2016). Projections for future estimated Hg emissions are highly variable as they depend on uncertainties in estimated emissions, energy production plans in individual countries as well as technology development scenarios (Giang et al., 2015; Pacyna et al., 2016; Streets et al., 2009). However, emission estimates are expected to become more accurate because of requirements posed by the global Minamata Convention of Mercury, the global treaty to reduce anthropogenic Hg emissions, which entered into force in August 2017 (http://www.mercuryconvention.org/). The Convention commits its parties to take action to protect human health and the environment from anthropogenic emissions and releases of mercury (Selin, 2018).

Mercury input to the Arctic is coming from a variety of sources which consist mainly of wet deposition, coastal erosion, riverine input, seawater import, and deposition due to atmospheric Hg depletion events (Outridge et al., 2008). Within the Arctic Ocean riverine input of Hg is an important source, being slightly larger than atmospheric deposition in summer (Zhang et al., 2015). In the past three decades global background concentrations of total gaseous mercury have declined, particularly in the North America, Europe and over the North Atlantic Ocean (−1.5 and −2.2% per year) (Cole et al., 2013; Streets et al., 2009; Zhang et al., 2016). However, in the Arctic air measuring stations the total gaseous mercury has been more stable (0.0 to −0.2% per year) (Berg et al., 2013; Cole et al., 2013). Concentrations of mercury in the Arctic as well as global physical environment may depend on various factors. Accelerating land use, energy use, Hg emissions control strategies, and climate change are all expected to have significant effects on global, regional, and local Hg levels and cycles (Obrist et al., 2018).

Temporal trends of Hg in Arctic biota were summarized by Riget et al. (2011), based on 83 different time series from terrestrial, fresh water and marine species. Average Hg levels changed between −8.6% to +10% per year across all 83 time series depending on species and study site. Terrestrial mammals showed either a decline or no annual changes, whereas variable trends were observed in marine mammals. However, the authors highlighted the lack of studies from higher trophic level species from the European Arctic (Riget et al., 2011). Temporal trends of Hg levels in biota is an integrated result of several biological processes and anthropogenic impacts (e.g. food availability, change in food items abundance, organism growth, climate change, emission rates), which can change from year to year or gradually over time (McKinney et al., 2015; Riget et al., 2007). The need to measure effects of multiple stressors simultaneously, such as pollutant and climate change, was emphasized already several years ago (Letcher et al., 2010). However, only a limited number of studies have investigated climate-related ecological impacts on temporal trends of Hg in Arctic biota (Braune et al., 2016; Fort et al., 2016; Gaden et al., 2009; McKinney et al., 2017; McKinney et al., 2015).

The arctic fox (*Vulpes lagopus*) is a circumpolar predator and scavenger. The population from Svalbard, Norway, is among the highest contaminated apex predators of Arctic ecosystems (Fuglei et al., 2007; Pedersen et al., 2015). Due to the lack of small rodents in most of Svalbard, the arctic fox population feeds extensively on resources from the marine food web in addition to terrestrial resources (Ehrich et al., 2015; Eide et al., 2005; Frafjord, 1993; Pond and Gilmour, 1997). In early spring, arctic foxes may prey on new-born ringed seal pups (*Phoca hispida*) (Lydersen and Gjertz, 1986), while in late spring and summer food items are locally abundant with the arrival of large number of seabirds and geese. Food availability decreases during autumn and winter since most birds leave Svalbard by October (Anker-Nilssen et al., 2000). During winter, arctic foxes rely on carcasses of Svalbard reindeer (*Rangifer tarandus platyrhynchus*) and seals (*Phocidae spp.*), and cached food (Frafjord, 1993). Previous studies reporting Hg levels in arctic fox potential prey species show generally higher levels of Hg in marine prey, such as seals (Fant et al., 2001; Riget et al., 2005) and

seabirds (Braune et al., 2002; Jaeger et al., 2009; Overjordet et al., 2015; Sagerup et al., 2009), and low levels in terrestrial prey, such as the herbivores reindeer (Riget et al., 2004), ptarmigan (Braune and Malone, 2006b) and geese (Braune and Malone, 2006a). The large variation in feeding habits and food availability of arctic foxes in Svalbard has likely consequences on their Hg exposure as already demonstrated for persistent organic pollutants (POPs) and perfluoroalkyl substances (Andersen et al., 2015; Fuglei et al., 2007; Routti et al., 2017). For example, tissue concentrations of Hg in arctic foxes from Iceland, Norwegian Sea, and Bering Island and Mednyi Island, northern Pacific Ocean, were related to their feeding habits (Bocharova et al., 2013). Furthermore, temporal variation in arctic fox feeding habits and food availability are likely to affect temporal trends of Hg in arctic fox tissues.

To increase the knowledge of temporal trends of Hg exposure in relation to feeding habits and food availability in Arctic biota, we conducted a study on total mercury (THg) in arctic foxes from Svalbard, Norway. THg is the sum of all waterborne mercury species (e.g. divalent mercury (Hg(II)), dimethylmercury (DMHg) and monomethylmercury (MMHg)). We expected that total mercury (THg) concentrations in arctic foxes were related to both emissions and feeding habits as well as food availability. We assumed that arctic foxes with a diet dominated by marine prey had higher THg levels than those with a terrestrial dominated diet for any given year. Further, we hypothesized that decreasing sea ice cover and high abundance of reindeer carcasses lowered the THg levels in arctic foxes over time.

2. Materials and methods

2.1. Sample collection

Arctic foxes ($n = 109$) were caught, using baited traps, during the annual harvest (trapping season spanning from 1997 to 1998 to 2013–2014) by local trappers in Spitsbergen, Svalbard, Norway. The trappers report mandatory information about the catch to the Governor of Svalbard and deliver the carcasses for laboratory measurements to the Norwegian Polar Institute. Data from 11 trapping seasons, which starts 1st of November and ends 15th of March, were included in this study (Table 1). Detailed information on sample collection has been reported by Andersen et al. (2015). Briefly, to evaluate arctic fox body condition we used a fat index based on the visible amount of subcutaneous and abdominal fat (Prestrud and Nilssen, 1992). The fat index was evaluated during autopsy and ranged from zero (no visible fat) to four (subcutaneous fat deposits cover most of the body and abdominally). However, since very few foxes had the index value of zero, only foxes of body condition 1–4 were included in this study. Age of the foxes was determined by counting the annuli in the cementum of a sectioned canine tooth (Grue and Jensen, 1976). To avoid confounding effects of maternal transfer of Hg from vixen to pups, we only included foxes between 1 and 2 years old because they had not yet reproduced. All vixens were investigated for dark placental scars in the uteri and none of the sampled foxes had given birth. Samples of skeletal muscle and liver were packed in aluminum foil and stored at −20 °C until further laboratory analysis of stable isotopes and THg, respectively.

2.2. Stable isotope analysis

We used stable isotope of carbon ($\delta^{13}C$) as a proxy for terrestrial vs. marine feeding habits as described by (Andersen et al., 2015; Routti et al., 2017). The stable isotope ratios measured in the arctic foxes reflect the autumn and winter feeding habits since muscle tissue gives an integrated dietary information from approximately the previous 1–2 months before capture (Roth and Hobson, 2000). Briefly, isotopic ratios of ^{14}C and ^{13}C ($\delta^{13}C$) were measured in muscle tissue of arctic foxes. The samples were dried at 60 °C for at least 48 h and ground to a fine powder in a bead-mill homogenizer (TissueLyzerII, Qiagen Gmbh, Hilden, Germany). Combustion analysis was performed at the

Table 1
Average, median (in parenthesis), standard deviations (SD) and range [min, max] values of liver total mercury (THg) levels (mg/kg ww), muscle δ^{13}C (‰) in arctic foxes from Svalbard, Norway, and sea ice cover (km^2) from Isfjorden, Svalbard, Norway. Reindeer carcasses is expressed as number of carcasses the preceding year of trapping (n), Fox (n) is the number of individual arctic foxes, F/M is the number of females and males, 1y/2y is the number of 1 year old and 2 year old individuals per trapping season (1997–2014).

Trap season	Fox (n)	F/M	1y/2y	THg ± SD [range]	δ^{13}C ± SD [range]	Reindeer carcasses (n)	Sea ice cover ± SD [range]
1997–1998	10	7/3	5/5	0.23 (0.19) ± 0.17 [0.02, 0.53]	−22.82 ± 1.33 [−24.82, −20.62]	12	1782 (1875) ± 824 [0, 2820]
1998–1999	10	2/8	5/5	0.17 (0.16) ± 0.1 [0.04, 0.34]	−22.84 ± 1.53 [−24.67, −19.39]	22	2213 (1875) ± 1015 [1, 3189]
1999–2000	10	6/4	3/7	0.07 (0.05) ± 0.06 [0.01, 0.22]	−23.2 ± 1.22 [−24.43, −20.72]	25	1215 (1398) ± 746 [58, 2096]
2001–2002	10	3/7	5/5	0.21 (0.17) ± 0.13 [0.03, 0.48]	−22.25 ± 1.79 [−24.8, −19.49]	29	879 (616) ± 1018 [0, 3369]
2002–2003	10	5/5	6/4	0.08 (0.05) ± 0.06 [0.01, 0.18]	−23.52 ± 1.22 [−24.59, −20.76]	191	860 (1359) ± 657 [0, 1359]
2003–2004	10	5/5	4/6	0.16 (0.16) ± 0.12 [0.02, 0.39]	−23.52 ± 1.17 [−25.32, −21.31]	4	3496 (2602) ± 1155 [2602, 4838]
2007–2008	10	6/4	5/5	0.18 (0.09) ± 0.26 [0.05, 0.94]	−23.34 ± 1.51 [−24.83, −21.17]	26	388 (216) ± 492 [0, 1438]
2010–2011	10	2/8	6/4	0.22 (0.07) ± 0.26 [0.02, 0.75]	−23.22 ± 1.37 [−25.43, −20.62]	92	2315 (2821) ± 1780 [316, 4945]
2011–2012	10	6/4	7/3	0.23 (0.13) ± 0.31 [0.03, 1.11]	−23.07 ± 1.27 [−24.58, −20.12]	8	431 (450) ± 193 [110, 607]
2012–2013	10	3/7	6/4	0.22 (0.21) ± 0.15 [0.04, 0.58]	−22.92 ± 1.12 [−24.77, −21.19]	114	418 (302) ± 287 [106, 738]
2013–2014	9	3/6	9/0	0.37 (0.3) ± 0.28 [0.05, 0.93]	−23.41 ± 0.78 [−24.8, −22.25]	26	455 (433) ± 283 [112, 723]

Stable Isotopes in Nature Laboratory (SINLAB), New Brunswick, Canada, following the protocols of Ehrich et al. (2011). The value of δ^{13}C was lipid normalized using model based normalization for muscular tissue in samples with a C/N ratio between 3.5 and 7 as previously described (Ehrich et al., 2011). Four samples were not corrected as these had a C/N ratio < 3.5 and were considered lean. None of the samples had a C/N ratio > 7. Quality assurance for analyses of δ^{13}C has been presented elsewhere (Andersen et al., 2015).

2.3. Proxies for food availability

We used availability of Svalbard reindeer carcasses and sea ice cover as proxies for food availability as described by Andersen et al. (2015) and (Routti et al., 2017). Briefly, we used total number of reindeer carcasses observed during the summer preceding the arctic fox trapping season as a proxy for terrestrial food availability. Most reindeer die in late winter and spring (Reimers, 2012), and will thus be available as food items for foxes only after the current trapping season.

Extent of sea ice cover (km^2) was used as a proxy for availability of marine food resources, such as seals. Average sea ice cover was calculated for each month (November–March; 1997–2014) using daily sea ice maps of Isfjorden (met.no; http://polarview.met.no). The threshold for sea ice cover, which foxes can use as hunting habitat, was set to 80% cover (i.e. close drift ice) and thus only days with ice cover ≥80% was included in the calculations. For each fox with known trapping date ($n = 99$) we used the sum of the monthly average sea ice from first of November to the month when the fox was trapped. The sum of the monthly average sea ice included the month when the fox was trapped only if trapping occurred after the 15th of the given month. For the foxes without known trapping date ($n = 10$) the average sea ice cover from first of November to 30th of February divided by two was used.

2.4. Mercury analysis

Analyses of total mercury (THg) in arctic fox liver samples was done by the accredited laboratory of ALS Scandinavia in Luleå, Sweden following the Environmental Protection Agency's (EPA) methods 200.7 and 200.8 with modifications. In brief, individual liver samples were homogenized and an aliquot of the homogenate was used for sample processing. The samples were solubilized by gentle refluxing with nitric acid (Sigma Aldrich) and hydrogen peroxide (Sigma Aldrich). After cooling, the samples were made up to volume and centrifuged prior to analysis. Samples were analyzed using inductively coupled plasma mass spectrometry (ICP-MS, Thermo Fischer Scientific). The laboratory regularly participates in interlaboratory comparisons and uses certified reference materials (CRM). The analytical uncertainty was calculated (range 18–35%) and included all significant sources of error such as instrument instability, uncertainty in balances and volumetric equipment, and errors in calibration standards. ALS Scandinavia

uncertainty is based on the international guidelines and adapted the laboratory's analyses and QC routines.

2.5. Data analysis

We used linear models to investigate the effect of age, sex, body condition, trapping season, δ^{13}C, reindeer carcasses and sea ice index on THg levels in arctic foxes. The continuous predictor variables δ^{13}C, reindeer carcasses and sea ice cover were standardized (mean = 0, standard deviation = 1) before analysis to facilitate comparison of effect sizes. The THg concentrations were $\log_{10}$ transformed and diagnostic plots were used to determine if the model residuals met linear models' assumptions, that is constant variance and approximate normality (Zuur et al., 2010). We ranked 12 candidate models (Table S1) according to their Akaike's Information Criterion (Burnham and Anderson, 2002) corrected for small sample size (AIC_c). Each model was given a normalized weight of evidence, that is AIC_c weight ($e^{(0.5(AICc\ best\ model - AICc\ model\ in\ hand)})$), which was further used for model-averaging to make inference from all models using R package AICcmodavg (Mazerolle, 2013). Model-averaged confidence intervals have better coverage properties than post-model selection confidence intervals (Burnham and Anderson, 2002). Yearly change (%) was derived by $100 * (10^{estimate\ for\ year} - 1)$ using the model-averaged estimates, i.e. the trend was adjusted for biological and environmental changes and thus reflect THg concentrations in arctic fox food web. We compared the adjusted trend to non-adjusted trend derived from a model that included only year as predictor variable. These back-transformed estimates reflect changes in median concentrations.

Statistical analyses were carried out using the statistical program R- version 3.3.1 (R Development Core Team, 2016).

3. Results and discussion

3.1. THg levels in arctic foxes

Generally, liver THg concentrations varied largely within trapping seasons; the highest THg concentration measured within one trapping season was 10 to 40 times higher than the lowest THg concentration during the same season (Table 1, Fig. 1). Average THg concentrations varied up to five-fold between trapping seasons. We measured the highest average concentrations in trapping season 2013–2014 (mean ± SD; 0.37 ± 0.28 mg/kg wet weight [ww]) and the lowest in trapping season 1999–2000 (0.07 ± 0.06 mg/kg ww, Table 1). Previously reported THg levels in liver of arctic foxes from Svalbard sampled in 1984–1986 (0.37 mg/kg ww) (Prestrud et al., 1994), were similar to the highest average levels measured in the present study (e.g. trapping season 2013–2014; 0.37 mg/kg ww, Table 1). Liver THg levels in arctic foxes from Svalbard sampled in trapping season 1999–2000 and 2001–2002 (0.07 and 0.21 mg/kg ww, respectively, Table 1) were considerably
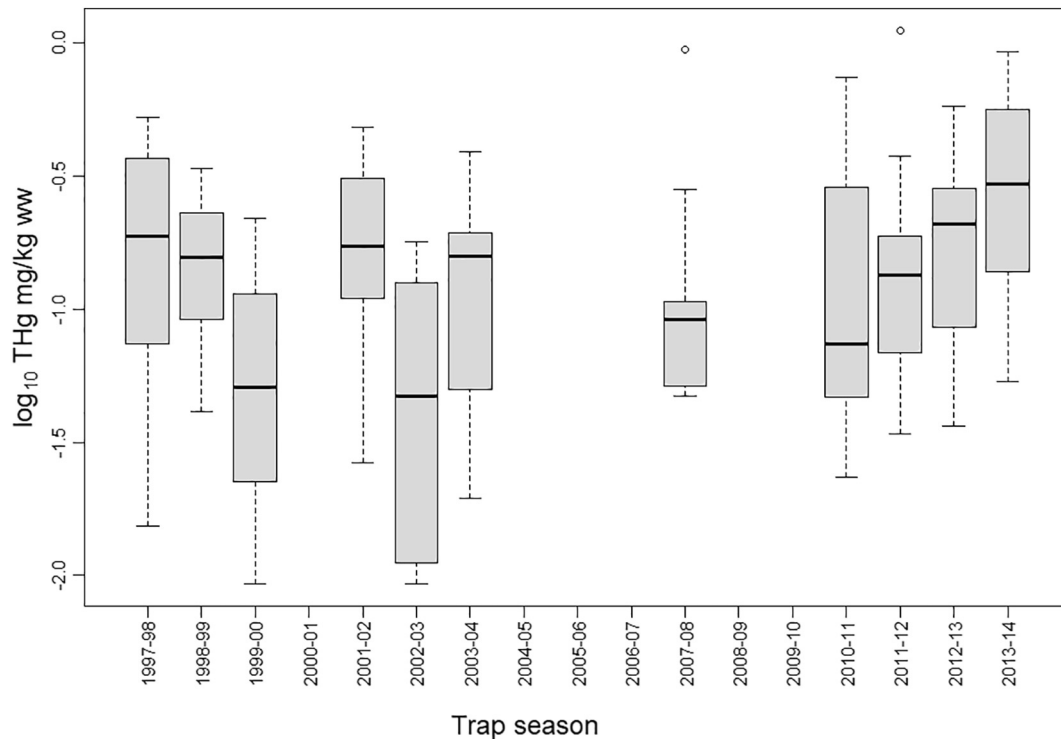
Fig. 1. Boxplot of ($\log_{10}$) total mercury (THg) (mg/kg ww) in the liver of arctic foxes (n = 109) from Svalbard, Norway, for the different trap seasons (1998–2014). The horizontal line is the median, the bottom and top of the box show 25th and 75th percentiles, respectively and whisker show the maximum and minimum values.

lower compared to concentrations measured in arctic foxes from the Canadian Arctic, Ulukhaqtuuq (Holman) and Arviat, in 1998–2001 (~0.8 mg/kg ww) (Hoekstra et al., 2003), and 0.25 and 0.32 mg/kg ww from Holman and Barrow, Alaska, respectively, in 1999 and 2000 (Dehn et al., 2006). Liver THg levels in arctic foxes from Svalbard sampled in 1997–1998 (0.23 mg/kg ww, Table 1) were similar to previously reported liver THg concentrations in arctic foxes from Prudhoe Bay, Alaska from 1994 (0.25 mg/kg ww) (Ballard et al., 2003). The spatial trend in arctic foxes coincide with the circumpolar Hg trend in ringed seals and polar bears (*Ursus maritimus*) with the highest Hg levels in western Canada followed by eastern Canada, Greenland, Alaska, Svalbard and White Sea (Renzoni and Norstrom, 1990; Riget et al., 2005; Routti et al., 2011; Rush et al., 2008). However, arctic fox liver THg concentrations from Iceland, sampled between 2011 and 2012, is much higher than any other regions reported with an average 8.24 mg/kg ww (Treu et al., 2018). This similarity in the general spatial trends can reflect a circumpolar use of marine resources by arctic foxes in wintertime when terrestrial prey, such as rodents, are scarce (Roth, 2002; Roth, 2003).

Liver THg levels in arctic foxes from Svalbard are below levels that have been associated with liver abnormalities or lethal and harmful effects in mammals (Rawson et al., 1993; Thompson, 1996). These were 61 mg/kg ww in liver for Atlantic bottlenose dolphin (*Tursiops truncates*) (Rawson et al., 1993), and 30 mg/kg ww for terrestrial free-ranging wild-life, such as red fox (*Vulpes vulpes*), Eurasian and river otters (*Lutra lutra* and L. *canadensis*) and American mink (*Mustela vison*) (Thompson, 1996).

3.2. Sex, age and body condition

Individual characteristics of arctic foxes (sex, age and body condition) did not influence liver THg concentrations significantly (Table 2). Similar to our results, previous studies on arctic foxes (Bocharova et al., 2013; Prestrud et al., 1994; Treu et al., 2018), polar bears (Routti et al., 2012), and ringed seals (Riget et al., 2012) have not reported differences in Hg exposure between sexes. Bocharova et al. (2013) reported increasing Hg levels with increasing age from juvenile to adult

(≥1 year), while Prestrud et al. (1994) and Treu et al. (2018) did not have an effect of age on the Hg levels between adults (≥1 year) and juveniles. In this study only foxes of 1 and 2 years were included to avoid confounding effects of maternal transfer of Hg from vixen to pups. This short span in age was too short to reveal any age related THg differences in the arctic foxes from Svalbard. Body condition, which reflects the amount of adipose tissue, was not expected to be influential since Hg tends to aggregate in muscular and hepasomatic tissue in mammals (Wagemann et al., 1998; Yamamoto et al., 1987). In the period for the trapping season (November 1 to March 15) the body fat arctic foxes may vary from 10 to 40% depending on the food supply (Prestrud and Nilssen, 1992). Though, in polar bears increasing body condition, defined as BMI (Body mass (kg)/standard length^2 (m^2)) was associated with decreasing THg levels in hair (McKinney et al., 2017). Decreasing THg with higher BMI was partly explained by possible re-circulation of Hg due to catabolism of proteinaceous tissues during fasting.

Table 2
Averaged linear model estimates (β) with 95% confidence intervals (CI) from the selected models (Table S1) predicting the levels of $\log_{10}$ transformed total mercury (THg) in arctic fox livers (n = 109) from Svalbard, Norway (1997–2014). The following 7 predictor variables were included in the models selected for inference. Sex [female vs male; as categorical variable] and trap season [1997/1998–2013/2014], age [1 or 2 years old], body condition [scale 1–4 from lean to fat], δ^{13}C [levels in muscle (Table 1)], reindeer carcasses [number of carcasses the preceding year of trapping] and sea ice cover [average sea ice cover (km^2) of Isfjorden, Svalbard, Norway] as continuous variables. Reindeer carcasses, sea ice cover and δ^{13}C were standardized (mean = 0, standard deviation = 1) prior to analyses.

$\log_{10}$ THg	β	CI
Intercept	−1.16	−1.32, −1.00
Trap season	0.03	0.01, 0.04
δ^{13}C	0.27	0.20, 0.34
Reindeer carcasses	−0.09	−0.16, −0.02
Sea ice cover	0.08	0.00, 0.15
Sex	0.00	−0.14, 0.14
Age	0.07	−0.07, 0.21
Body condition	0.02	−0.05, 0.09
Multiple adjusted R^2	0.42	

3.3. Effect of feeding habits and food availability

Feeding habits (δ^{13}C) and food availability (reindeer carcasses and sea ice cover) influenced liver THg levels in the arctic fox (Table 2). The δ^{13}C value was the most important predictor to explain the THg levels followed by reindeer carcasses and sea ice cover. Our results are consistent with a study by Bocharova et al. (2013) highlighting the importance of feeding habits on Hg exposure in arctic foxes.

Liver levels of THg increased with δ^{13}C values indicating that arctic foxes with a more marine diet were more exposed to Hg than foxes mainly feeding on a terrestrial diet (Table 2, Fig. 2). This is in accordance with our assumption based on previous studies reporting generally higher levels of Hg in marine prey (Braune et al., 2002; Fant et al., 2001; Jaeger et al., 2009; Overjordet et al., 2015; Riget et al., 2005; Sagerup et al., 2009), and lower Hg levels in terrestrial prey (Braune and Malone, 2006a; Braune and Malone, 2006b; Riget et al., 2004). δ^{13}C values did not change over time between 1997 and 2014 (linear model, $\beta = -0.02$, 95% confidence intervals [CI]: -0.07, 0.03), which is in accordance with our previous study based to a large degree on the same individuals (Routti et al., 2017). There was a high individual variation in measured δ^{13}C values within each trap season (Table 1). The range of δ^{13}C was similar to previously reported values (Ehrich et al., 2015; Fuglei et al., 2007), confirming that the foxes used in the present study consist of a mixture of individuals with different feeding strategies. In Svalbard, arctic foxes belong to the coastal ecotype (i.e. living in an ecosystem without rodents as main food) and feed within the terrestrial and marine food web (Eide et al., 2005; Frafjord, 1993; Pond and Gilmour, 1997).

Based on model average estimates increased reindeer carcasses was associated with decreasing liver THg levels in arctic foxes ($\beta = -0.09$, 95% CI: -0.16, -0.02, $R^2 = 42\%$, Table 2, Fig. 2). In years with extremely high availability of reindeer carcasses on the tundra, the THg levels in arctic foxes were low (e.g. trap season 2002–2003, Table 1). Excluding the year with the highest number of reindeer carcasses from the multiple linear model removed the significance of reindeer carcasses on the THg levels ($\beta = -0.08$, 95% CI: -0.22, 0.05, $R^2 = 36\%$) but, the consistent estimates (-0.09 vs. -0.08) suggest reducing effect of reindeer carcasses on the levels of THg in arctic foxes.

In Svalbard, the large valleys (Adventdalen, Sassendalen and Reindalen) that houses a large part of the reindeer population, have a coastal outlet, with short distances from the inner part of the valleys to the coast and to the inner fjords with winter sea ice and access to marine food items (Eide et al., 2005). Therefore, in years with high number of carcasses all foxes will have easy access to reindeer carcasses. The importance of reindeer carcasses on Hg exposure in arctic foxes from Svalbard has also been discussed in a previous study

(Prestrud et al., 1994). The authors suggested that higher Hg levels in arctic foxes caught north compared to south of Isfjorden were related to differences in availability of reindeer as food. The difference was 0.62 ± 0.28 mg/kg ww north of Isfjorden compared to 0.14 ± 0.14 mg/kg ww south of Isfjorden. Arctic foxes trapped on the north side of Isfjorden relied more on marine food items as reindeer were almost absent there when the study was conducted (Prestrud et al., 1994; Tyler and Oritsland, 1989). In our study, THg concentrations were similar in foxes caught on both sides of Isfjorden. This could be because reindeer currently appear to be more commonly distributed also on the north side of the Isfjorden increasing the terrestrial food availability (pers. obs. Å.Ø. Pedersen).

The availability of other terrestrial prey species, which were not included in our study, such as Arctic-nesting geese (*Anser brachyrhynchus* and *Branta leucopsis*) and Svalbard rock ptarmigan (*Lagopus muta hyperborea*), may also affect Hg levels in the arctic fox. For example, glaucous gulls (*Larus hyperboreus*) with a terrestrial dominated diet, including goose eggs and goslings as major components, had lower Hg and POP levels than glaucous gulls with a more marine dominated diet (Braune et al., 2002). THg liver levels in Svalbard reindeer and ptarmigan has been reported from 1993 and is low with ~0.01 mg/kg dry weight for reindeer and ~0.02 mg/kg dry weight for ptarmigan (Severinsen and Skaare, 1997). The goose populations in Svalbard (i.e. pink-footed goose and barnacle goose) have rapidly increased during the study period (Fox et al., 2010; Fox and Madsen, 2017; Madsen and Williams, 2012). These migratory species are available as prey for arctic foxes from mid - late May to September (Anker-Nilssen et al., 2000). As the foxes were trapped between November and March, and the turnover time of muscle δ^{13}C is only 1–2 months, the reported muscle δ^{13}C values unlikely reveal intake of geese.

Levels of THg in the arctic fox increased with increasing spatial extent of sea ice cover (Table 2, Fig. 2). The presence of sea ice facilitates arctic fox movement and the sea ice acts as hunting or scavenging grounds for marine food items such as seal pups or remains from polar bear kills (Roth, 2003), which contain more Hg than terrestrial food items (Braune et al., 2002; Braune and Malone, 2006a; Braune and Malone, 2006b; Riget et al., 2004). Arctic foxes are extremely mobile and can use the sea ice extensively as a hunting platform during winter and spring (Lai et al., 2015; Pamperin et al., 2008; Roth, 2002; Tarroux et al., 2010).

3.4. Temporal trend

Based on the model averaged estimates, levels of THg adjusted for variation in feeding habits and food availability increased with 7.2% (95% CI: 2.3, 9.6, $R^2 = 42\%$) per year in liver of arctic foxes during the
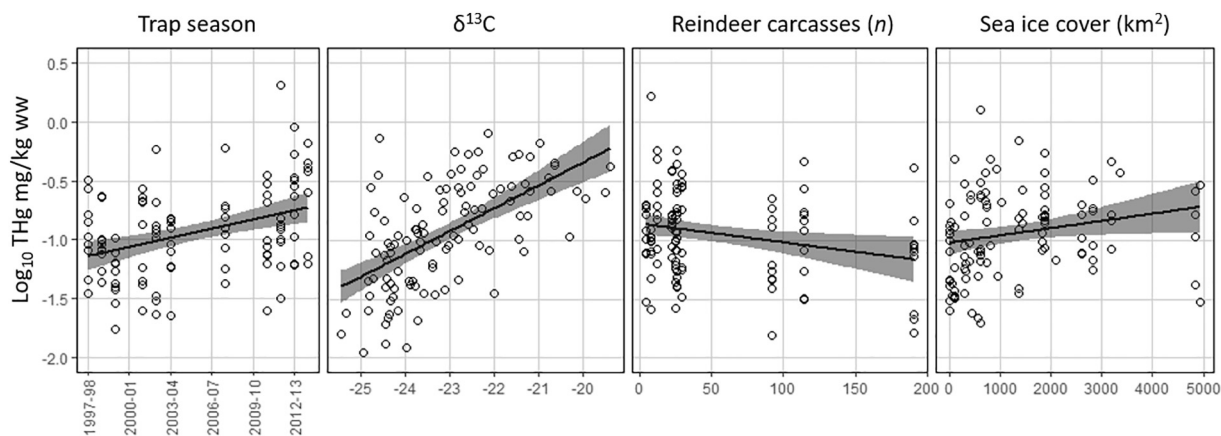

Fig. 2. Relationship between $\log_{10}$ total mercury (THg) (mg/kg ww) in the liver of arctic foxes (n = 109) from Svalbard, Norway, and the scaled predictor variables: trap season (1998–2014), δ^{13}C (‰), reindeer carcasses (n), and sea ice cover (km²). The plots are partial residual plots (i.e. the effect of each predictor variable has been controlled for the other variables in the model). Regression lines present significant relationship between the variables.

study period (1997–2014; Table 2). The annual change in THg concentrations during the period 1997–2014 that only has year as predictor variable was lower than the adjusted trend, as would be expected given the decreasing sea ice cover over time (β:3.5, 95% CI: −0.11, 7.2, R^2 = 2.4%).

Variable temporal trends in THg have been observed in Arctic species with different population trends (increasing, decreasing and stable) and geographical location (Riget et al., 2011). In addition, the explanation for the variable trends differ between studies. For example, increase in Hg levels in little auks (*Alle alle*) during breeding period was related to increasing Hg levels in their zooplankton diet (Fort et al., 2016). Temporal changes, both increasing and decreasing, in ringed seals from Greenland seemed not to be related to dietary changes (i.e. changes in δ^{15}N) (Aubail et al., 2010; Riget et al., 2012), though Riget et al. (2007) suggested that changes in ringed seals Hg levels could be due to changes in feeding behavior that was not reflected in trophic position. The increase in THg in polar bears from East Greenland could be explained by changes in emission, food web or climate-induced changes in Hg cycle (Routti et al., 2011). Changes in foraging ecology and not declining environmental levels of Hg were driving the short term declines in southern Beaufort Sea polar bears (McKinney et al., 2017). Decreasing levels of Hg in eggs of Brünnich's guillemot (*Uria lomvia*), northern fulmars (*Fulmarus glacialis*), black guillemots (*Cepphus grylle*) and black legged kittiwakes (*Rissa tridactyla*) have been observed from 1993 to 2015 in the Canadian Arctic (Braune et al., 2016). The latter trend seemed to reflect atmospheric Hg trends. However, in the same study Hg levels in eggs of glaucous gulls decreased from 1993 to 2003 before increasing again during the period 2008–2013. The changes in Hg in glaucous gulls were not related to changes in δ^{15}N (Braune et al., 2016). Further, no significant temporal trends were observed in eggs of herring gulls (*Larus argentatus*), kittiwakes, common guillemots (*Uria aalge*), and Atlantic puffins (*Fratercula arctica*) from Northern Norway between 1983 and 2003 (Helgason et al., 2008). Nor in short-horn sculpins (*Myoxocephalus scorpius*) from Greenland between 1987 and 2004 (Riget et al., 2007), or Greenland caribou (*Rangifer tarandus groenlandicus*) between 1996 and 2000 (Riget et al., 2004).

3.5. Conclusions

Our study showed that liver THg concentrations in arctic foxes from Svalbard increased by 7.2% per year during the period 1997–2014. As the concentrations were adjusted for variation in feeding habits and food availability, the trend reflects changes in the food web of the artic fox. The arctic fox in Svalbard is a top predator and scavenger both in the terrestrial and marine food web. Their liver THg levels are lower than other marine top predators, such as ringed seals, polar bears, and glaucous gulls. This highlights the importance of the terrestrial diet of the arctic fox with less contaminant burden.

3.6. Future perspectives

To predict the future levels and changes in THg in arctic foxes is challenging. Climate-induced changes within the Arctic can both decrease and increase the exposure of THg in arctic foxes. At present "rain-on-snow" is a common weather phenomena in the Arctic and the frequency of it has increased and is expected to increase (Hansen et al., 2014; Peeters et al., 2019). These events lead to basal ground-ice that encapsulates the vegetation in a solid ice-layer. Which, in turn, has led to high starvation-induced mortality in all monitored populations of the wild reindeer by merely blocking access to the winter food source (Hansen et al., 2013). Increased availability of reindeer carcasses will likely reduce the THg exposure in arctic foxes. However, increased precipitation promotes return of Hg from the air back to soil or sea and possibly increases the exposure of Arctic organisms to Hg. On the other hand, sea ice is retreating rapidly in the European Arctic (Dobricic et al., 2016; Laidre et al., 2015), leaving in particular the western coast of Svalbard

ice free year round (Prop et al., 2015). Reduced sea ice cover decreases the barrier for sea to air exchange and promotes oceanic evaporation, which subsequently increases atmospheric Hg levels (Hirdman et al., 2009). Melting sea ice and snow also releases Hg to ocean and fresh water that is readily reducible and available for evasion or uptake in the food webs (Fisher et al., 2012). In the southern Baltic area lack of sea ice cover increased the mean annual Hg pool in phytoplankton (mass of Hg in phytoplankton per liter of seawater) by 30% (Beldowska and Kobos, 2016). This increases the potential for additional uptake of Hg in zooplankton and bioaccumulation in the food web to top predators such as the arctic fox.

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