# **Supporting Information for:**

**Title:** Contrasting temporal patterns of mercury, niche dynamics, and body condition of polar bears and ringed seals in a melting icescape

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# **Appendix S1**

age class. Adult female polar bears are separated by reproductive stage at time of capture.					
Stage	n	δ <sup>13</sup> C (‰)	$\delta^{15}$ N (‰)	THg (µg/g)	logTHg (µg/g)
Polar bear					
Cub	64	$-17.6\pm0.4$	$19.2\pm1.0$	$2.1\pm0.7$	$0.32\pm0.13$
Yearling	57	$-17.2\pm0.5$	$19.0\pm0.6$	$4.0 \pm 1.2$	$0.59\pm0.12$
Two-year old	54	$-17.2\pm0.5$	$18.9\pm0.5$	$4.3\pm0.9$	$0.63\pm0.10$
Subadult	107	$-17.2\pm0.5$	$18.9\pm0.6$	$4.8 \pm 1.6$	$0.68\pm0.13$
Adult male	353	$-17.1\pm0.5$	$19.8\pm0.5$	$4.5\pm1.1$	$0.65\pm0.10$
Adult female	110	$\textbf{-17.3}\pm0.4$	$19.2\pm0.5$	$4.8\pm1.6$	$0.68\pm0.13$
Adult female w/ cub	59	$-17.4\pm0.5$	$18.7\pm0.6$	$4.9\pm1.2$	$0.69\pm0.10$
Adult female w/ yearling	43	$-17.4 \pm 0.4$	$18.9\pm0.4$	$5.2 \pm 1.3$	$0.72\pm0.11$
Adult female w/ 2 cubs	70	$-17.4 \pm 0.4$	$18.7\pm0.5$	$4.6\pm1.4$	$0.67\pm0.07$
Adult female w/ 2 yearlings	22	$\textbf{-17.1}\pm0.4$	$18.8\pm0.2$	$5.2 \pm 1.0$	$0.71\pm0.08$
Adult female w/ cub and yearling	2	$-17.2 \pm 0.1$	$18.8\pm0.1$	$3.8\pm0.3$	$0.57\pm0.04$
Adult female w/ 2 two-year olds	2	$\textbf{-16.9} \pm 0.4$	$18.9\pm0.2$	$6.4\pm1.0$	$0.81\pm0.05$
Ringed seal					
Adult	144	$-20.5\pm1.4$	$16.2\pm0.7$	$0.4\pm0.3$	$\textbf{-0.40} \pm 0.10$
Subadult	139	$-20.8\pm1.2$	$15.6\pm1.0$	$0.4\pm0.5$	$-0.40 \pm 0.10$

**Table S1.** Sample sizes (*n*) and mean  $\pm$  SD for  $\delta^{13}$ C,  $\delta^{15}$ N and total Hg (THg) of polar bear hair and ringed seal muscle samples collected from 2003-2016 in western Hudson Bay, Canada by age class. Adult female polar bears are separated by reproductive stage at time of capture.

# **Appendix S2**

### **Supplementary Materials and Methods:**

#### **Total mercury analysis**

Polar bear hair samples were washed before THg analysis which consisted of agitation by using stainless steel forceps in warm Milli-Q water containing a small amount of mild Ivory dishwashing detergent, then rinsed three times with Milli-Q water and air-dried overnight in the University of Alberta Biogeochemical Laboratory (Edmonton, AB, Canada). Standard protocols for THg analysis were performed in the University of Alberta Biogeochemical Laboratory using an automated Tekran 2600 THg analyzer (Toronto, ON, Canada) and are detailed elsewhere<sup>1,2,3</sup>. Mean relative standard deviation between duplicate hair samples were  $1.74 \pm 1.45\%$ . The analytical detection limit for both methods was 0.0005 µg/g. All polar bear hair THg data are provided in µg g<sup>-1</sup> dry weight.

Ringed seal muscle was subsampled (2-5 g), taking care to use tissue that was not in contact with polyethylene plastic storage bags, and homogenized with a hand blender. Subsampled muscle tissue (0.2 g wet weight) was analyzed for THg using a Direct Mercury Analyzer (Milestone Inc., Shelton, CT, USA) in Environment and Climate Change Canada laboratories (Burlington, ON, Canada) following USEPA Method 7473 (USEPA, 2007). All ringed seal muscle THg data are presented in  $\mu$ g g<sup>-1</sup> wet weight. Quality assurance steps included the analysis of DORM-3 (fish muscle protein), DOLT-2 and DOLT-4 (dogfish liver) certified reference materials (National Research Council of Canada), and/or National Institute of Standards and Technology (NIST) 2976 muscle tissue with each batch of 20 samples. Average deviation from certified values for Hg ranged from 1.6 to 6.0% (DOLT-2 = 2.91 ± 1.88%, *n* = 7; Dolt 4 = 1.59 ± 1.36%, *n* = 14, Dorm 3 = 5.96 ± 7.28, *n* = 20, 2.77 ± 2.43, *n* = 12).

#### Stable isotope analysis

A cleaned polar bear hair sample weighing between 700-900 µg was transferred into tin capsules and analyzed for  $\delta^{13}$ C and  $\delta^{15}$ N using a EuroVector EuroEA3028-HT elemental analyzer (Eurovector, Milan, Italy) coupled to a GV Instruments IsoPrime continuous-flow isotope ratio mass spectrometer (GV Instruments, Manchester, UK) at the University of Alberta Biogeochemical Laboratory. Due to the presence of lipids<sup>4</sup>, ringed seal muscle tissues were lyophilized for 48 h, homogenized using a mortar and pestle, and lipid-extracted using 2:1 chloroform:methanol solvent following the methods of Bligh and Dyer<sup>5</sup>. Following lipid extraction, 1 mg of muscle was weighed into tin capsules and analyzed for  $\delta^{13}$ C and  $\delta^{15}$ N on a Thermo Finnigan DeltaPlus XL mass spectrometer (Thermo Finnigan, Bremen, Germany) equipped with an elemental analyzer (Carlo Erba, Milan, Italy) at the University of Waterloo (Waterloo, ON). At the University of Waterloo, analytical precision for  $\delta^{15}N$  and  $\delta^{13}C$  of ringed seals were < 0.3% and 0.2‰, respectively, based on repeated measurements of working laboratory standard material cross-calibrated to the international standards USGS-40, IAEA-CH6, -CH3, -N1 and -N2. A mix of these international and lab standards (organic and inorganic) were used with NIST-1577b. A duplicate was run for every 10<sup>th</sup> sample, and analytical precision for  $\delta^{13}$ C and  $\delta^{15}$ N values was < 0.3‰ and 0.2‰, respectively. A small subset of ringed seal muscle samples (n = 20) were analyzed for  $\delta^{34}$ S in the Chemical Tracers Lab at the University of Windsor using a Delta V Plus IRMS (ThermoFinnigan, San Jose, CA USA) coupled with a Costech 4010 EA (Costech Analytical Technologies Inc., Valencia, CA, USA). Precision, assessed by the standard deviation of replicate analyses of five standards: NIST1577c, internal lab standard (tilapia muscle), USGS 42, NIST 8555 and NIST 8529 (n=5 for all), measured  $\leq 0.17\%$  for  $\delta^{34}$ S. The accuracy, based on the certified value of USGS 42 (n=5) analyzed

throughout runs and not used to normalize samples showed a difference of 0.08‰ for  $\delta^{34}$ S from the certified value. At the University of Alberta, NIST-8415 whole egg powder was used as a standard reference material and analytical precision and accuracy was  $\leq 0.1\%$  for  $\delta^{13}$ C and  $\leq$ 0.2‰ for  $\delta^{15}$ N. All stable isotope ratios are expressed in per mil (‰) in standard delta ( $\delta$ ) notation relative to the international standards Pee Dee Belemnite for carbon, atmospheric N<sub>2</sub> for nitrogen, and Vienna Cañon Diablo Triolite for sulphur<sup>6,7</sup> using the following equation:  $\delta X =$ [(Rsample/Rstandard) -1] x 10<sup>-3</sup>, where X is <sup>13</sup>C, <sup>15</sup>N or <sup>34</sup>S and R equals <sup>13</sup>C/<sup>12</sup>C, <sup>15</sup>N/<sup>14</sup>N <sup>34</sup>S/<sup>32</sup>S.

# **Supplementary Results:**

Polar bear THg concentrations were significantly different between classes ( $F_{1011} = 128$ , p < 0.01) with cubs-of-the-year being significantly lower than all other classes (Tukey HSD, p < 0.01), adult males being significantly higher than adult females and yearlings (Tukey HSD, p < 0.04) and yearlings being significantly lower than adult females and subadults (Tukey HSD, p < 0.04) and yearlings being significantly lower than adult females and subadults (Tukey HSD, p < 0.04). Polar bear  $\delta^{13}$ C and  $\delta^{15}$ N values were significantly different between classes (F = 10.46, p < 0.01). Polar bear  $\delta^{13}$ C and  $\delta^{15}$ N values were significantly different between classes (F = 10.46, p < 0.01 for  $\delta^{13}$ C and F = 91.9, p < 0.01 for  $\delta^{15}$ N) with adult males having significantly higher  $\delta^{13}$ C and  $\delta^{15}$ N values than all other classes (all Tukey HSDs, p < 0.01). Adult females also had significantly higher  $\delta^{15}$ N values than cubs-of-the-year (Tukey HSD, p = 0.03). Adult males and cubs-of-the-year were excluded from subsequent niche analyses due to both groups being significantly different from many of the other classes across THg,  $\delta^{13}$ C, and  $\delta^{15}$ N. Although yearlings had significantly lower THg than subadults and adult females, differences in mean THg concentrations were  $\leq 0.8 \ \mu g \ g^{-1}$  and there were no significant differences for  $\delta^{13}$ C and  $\delta^{15}$ N values between age classes, therefore yearling, subadult, and adult female classes were pooled for subsequent niche analyses (Table SI). Differences in THg concentrations,  $\delta^{13}$ C, and  $\delta^{15}$ N

values between western Hudson Bay polar bear classes is consistent with Bechshoft et al.<sup>2</sup> where THg was slightly higher in adult females than younger age classes partly due to differences in food intake and Hg accumulation in liver and kidneys<sup>3,8</sup> despite similar  $\delta^{13}$ C and  $\delta^{15}$ N values between adult females and subadults<sup>9</sup>. Adult females, subadults, two-year-olds and yearlings exhibit a narrower niche than adult males by relying heavily on ringed seals as prey (>50%) with a high dietary contribution from harbour seals (*Phoca vitulina*; 11-20%)<sup>10,11,12</sup>. In turn, this higher reliance on ringed seals as the dominant prey item increases the susceptibility of adult females, subadults and yearlings to consequences of changing sea ice dynamics and inter-annual variation in ringed seal demographics and availability<sup>12,13,14</sup>.

For ringed seals, there was a significant difference in  $\delta^{13}$ C values ( $t_{286} = 2.11$ , p = 0.04) and  $\delta^{15}$ N values ( $t_{286} = 5.42$ , p < 0.01) between adults and subadults, but differences in mean values were minimal (0.3‰ for  $\delta^{13}$ C and 0.6‰ for  $\delta^{15}$ N; Table S1 in Appendix S1, Supporting Information) and slightly higher than analytical error (0.2‰ for  $\delta^{13}$ C and 0.3‰ for  $\delta^{15}$ N). Therefore, given minimal differences in mean  $\delta^{13}$ C and  $\delta^{15}$ N values, no difference in THg between adults and subadults and a documented similar proportional diet of invertebrates and fishes between adults and subadults at lower latitude<sup>15,16,17</sup>, both age classes of ringed seals were grouped for subsequent niche analyses. Furthermore, the diet of subadult and adult ringed seals in western Hudson Bay are similar and mainly piscivorous, dominated by sand lance (*Ammodytes* spp.)<sup>15</sup>, with the contribution of capelin (*Mallotus villosus*) increasing over time<sup>17</sup>. Despite a piscivorous diet,  $\delta^{15}$ N, THg and persistent organic pollutant concentrations of western Hudson Bay ringed seals have generally been lower than conspecifics at higher latitudes since 2000, partially due to natural geological variation, a closer proximity to southern contaminant sources that are more directly impacted by regulations and bans, temporal changes in forage fish species composition, and lower trophic position or food chain length driven by more first-year ice in the Hudson Bay system<sup>16,18,19</sup>.

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