

## Ecological Change Drives a Decline in Mercury Concentrations in Southern Beaufort Sea Polar Bears

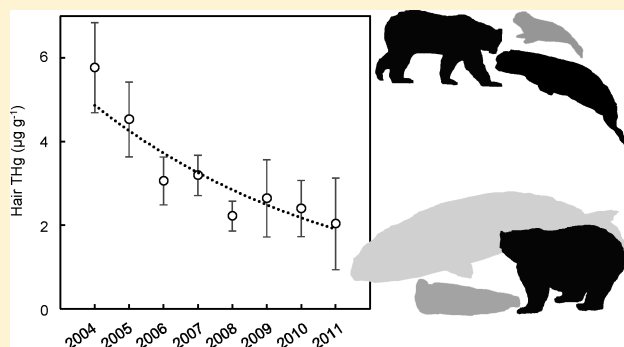
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### S Supporting Information

**ABSTRACT:** We evaluated total mercury (THg) concentrations and trends in polar bears from the southern Beaufort Sea subpopulation from 2004 to 2011. Hair THg concentrations ranged widely among individuals from 0.6 to 13.3  $\mu\text{g g}^{-1}$  dry weight (mean:  $3.5 \pm 0.2 \mu\text{g g}^{-1}$ ). Concentrations differed among sex and age classes: solitary adult females  $\approx$  adult females with cubs  $\approx$  subadults  $>$  adult males  $\approx$  yearlings  $>$  cubs-of-the-year  $\approx$  2 year old dependent cubs. No variation was observed between spring and fall samples. For spring-sampled adults, THg concentrations declined by 13% per year, contrasting recent trends observed for other Western Hemispheric Arctic biota. Concentrations also declined by 15% per year considering adult males only, while a slower, nonsignificant decrease of 4.4% per year was found for adult females. Lower THg concentrations were associated with higher body mass index (BMI) and higher proportions of lower trophic position food resources consumed. Because BMI and diet were related, and the relationship to THg was strongest for BMI, trends were re-evaluated adjusting for BMI as the covariate. The adjusted annual decline was not significant. These findings indicate that changes in foraging ecology, not declining environmental concentrations of mercury, are driving short-term declines in THg concentrations in southern Beaufort Sea polar bears.



### INTRODUCTION

Polar bears (*Ursus maritimus*) face multiple anthropogenic threats.<sup>1</sup> Climate-change-driven declines in sea ice habitat have occurred and have resulted in reduced dietary proportions of traditional prey, namely ice seals (e.g., ringed seals).<sup>2,3</sup> Consequently, some subpopulations of polar bears have been reported to have increased their use of land and accompanying land-based food resources, including subsistence-harvested and beach-cast bowhead whale (*Balaena mysticetus*) carcasses.<sup>4,5</sup> Such changes in habitat and resource use may expose polar bears to health risks, such as disease and environmental contaminants.<sup>6</sup> Mounting evidence suggests that polar bears already face harmful dietary exposures to mercury (Hg) and persistent organic pollutants (POPs), such as polychlorinated biphenyls (PCBs).<sup>7–11</sup>

Mercury, largely in the form of methylmercury (MeHg), bioaccumulates and biomagnifies, leading to tissue concentrations in Arctic marine predators, including polar bears that often exceed general threshold values for biological effects.<sup>7</sup> In polar bears and other Arctic biota, such elevated Hg concentrations have been associated with impaired neurological development and function, as well as hepatotoxicity, nephrotoxicity, and immunotoxicity.<sup>7,12</sup> In the most-recent circumpolar assessment, and similar to previous assessments, total Hg (THg) concentrations were found to be highest in 2007-

sampled polar bear liver tissues in the western North American Arctic, and specifically the Southern (SB) and Northern Beaufort Sea (NB) subpopulations ( $\sim 70 \mu\text{g g}^{-1}$  wet weight (ww)).<sup>13</sup> These SB and NB concentrations were above those associated with liver abnormalities in bottlenose dolphins ( $61 \mu\text{g g}^{-1}$  ww)<sup>14</sup> and above a general effect threshold for terrestrial wildlife ( $30 \mu\text{g g}^{-1}$  ww).<sup>15</sup> Similarly, hair THg concentrations (which are correlated with liver THg concentrations ( $r = 0.69$ ,  $P < 0.05$ ,  $n = 19$ ))<sup>16</sup> in SB polar bears in 2005 ( $7.5 \mu\text{g g}^{-1}$  dry weight (dw))<sup>17</sup> exceeded the mean hair concentration ( $5.4 \mu\text{g g}^{-1}$  dw) in another polar bear subpopulation, in which brain THg and MeHg concentrations were negatively correlated with activity of the neurochemical receptor, N-methyl-D-aspartate (NMDA).<sup>18</sup> As measured in the previous circumpolar assessment,<sup>13</sup> high liver concentrations in SB polar bears in 2007 were similar to those measured in 2002 and higher than those measured in 1982–1984. It is not yet known whether these concentrations have continued to remain elevated and of concern to polar bear health.

Received: February 13, 2017

Revised: May 19, 2017

Accepted: May 23, 2017

Over the last two decades, substantial changes in habitat use and related variation in food resources have been reported for the SB polar bear subpopulation. Historically, SB polar bears have remained on the sea ice nearly or entirely year-round, following the retreating pack ice northward when annual ice melts near the coast in the late spring and early summer.<sup>19</sup> However, both the seasonal duration of sea ice and its southward extent in the Beaufort Sea during the annual reduced ice period (July–October) have been greatly reduced. These changes have led to an increase in individuals of the SB subpopulation making extended use (>21 consecutive days) of onshore habitat during the annual reduced ice season from 2000 and continuing through 2014.<sup>20</sup> While on the sea ice, foraging efforts by polar bears largely focus on ringed seal (*Pusa hispida*) and, to a lesser extent, bearded seal (*Erignathus barbatus*).<sup>21–23</sup> For polar bears on shore, chemical tracer-based feeding estimates suggest that whale remains comprise 50–70% of polar bear diets.<sup>24</sup> During the period of increased land use, annual dietary proportions for SB polar bears of bowhead whale, ringed seal, and beluga whale (*Delphinapterus leucas*) fluctuated with annual climate and sea ice conditions.<sup>5</sup> That is, in poorer ice years, diet estimates showed higher proportions of bowhead whale and lower proportions of ringed seal and beluga whale. Generally speaking, ringed seal, beluga whale, and bearded seal in the Beaufort Sea have varied diets that include both fish and invertebrates.<sup>25,26</sup> In contrast, bowhead whales filter feed on zooplankton.<sup>27</sup> Thus, bowhead carcasses are at a lower trophic position and potentially represent a less-contaminated food source with respect to Hg and POPs relative to other Arctic prey species.<sup>28,29</sup> Differences in seasonal movements among these polar bear prey species could also contribute to variation in their tissue contaminant loads.<sup>30</sup> Tissue contaminant concentrations in polar bears have also been related to differences in body condition.<sup>31,32</sup> Reduced body condition among polar bears resulting from changes in sea ice habitat may increase lipid and protein tissue catabolism and release contaminants into circulation,<sup>33</sup> thus increasing their risk of health effects. In light of these ecological changes, their influence on exposures to contaminants considered to be of major potential health concern in SB polar bears, such as MeHg, should be investigated.

In this study, we assessed hair THg concentrations in SB polar bears from 2004 to 2011, a period during which land use steadily increased and diet substantially changed.<sup>20</sup> Hair serves as a suitable matrix for determining THg because (1) hair accumulates Hg from blood, thus representing circulating concentrations when the hair was grown (blood THg correlation with hair THg determined in SB polar bears:  $r = 0.46$ ,  $P = 0.01$ ,  $n = 30$ );<sup>17</sup> (2) hair can be collected noninvasively; (3) hair Hg concentrations have been reported in SB and other subpopulations previously; and (4) adverse effects guidelines have been reported in the literature based on hair THg concentrations.<sup>34</sup> Hair THg concentrations were evaluated by sex and age class and season and over time. We additionally evaluated whether the temporal trends in hair THg among adult males and females were related to diet, as estimated by chemical tracers (from ref 5), and body condition. We hypothesized that THg concentrations in SB polar bears would be lower in recent years due to increased use of lower trophic position food resources available on shore (e.g., bowhead whales).<sup>28,29</sup>

## MATERIALS AND METHODS

**Sampling.** Hair subsampling used a retrospective approach. Adipose biopsies with attached skin and hair were collected for the purposes of other studies as detailed elsewhere.<sup>5,35</sup> Briefly, biopsies were collected from the rump region of captured individuals by biopsy punch and from remotely sampled individuals by biopsy dart. Prior to taking biopsy punches, hair was trimmed to a short length. Biopsy darts also tended to return only short hairs. A total of 198 samples had sufficient hair for analysis. Most ( $n = 166$ ) were collected from March to mid-May (“spring-sampled”) of 2004–2011, while a smaller number ( $n = 32$ ) were collected in August to October (“fall-sampled”) of 2008, 2009, and 2011 (Table S1).

Biological information was obtained, including sex and age class and genetic identification.<sup>36</sup> Individuals were classified according to sex and age as cubs-of-the-year ( $n = 7$ ), yearlings ( $n = 21$ ), 2 year old dependent cubs ( $n = 13$ ), subadult (independent 2, 3, and 4 year old) females ( $n = 18$ ), and males ( $n = 14$ ), solitary adult (5 years and older) females ( $n = 29$ ), adult females with cubs ( $n = 38$ ), and adult males ( $n = 58$ ). For first-time captures, age was determined by counting growth layer groups in the cementum of a vestigial premolar tooth.

**Hair Total Mercury Analysis.** Concentrations of THg in hair were measured as a proxy for MeHg because MeHg represents >97% of THg in polar bear hair.<sup>34</sup> Hair samples were cut from the biopsy close to the skin and surface contamination was removed by standard protocols.<sup>37</sup> Clean hairs were subject to acid digestion, followed by THg analysis by cold-vapor atomic absorption spectrometry.<sup>38</sup> Concentrations of THg were reported as  $\mu\text{g g}^{-1}$  dw.

Quality-control procedures included analysis of blanks, sample duplicates, matrix spikes, and certified standards from the National Research Council of Canada (DORM-3 and DOLT-4). All blanks were below the detection limit. Based on an average sample size of ~0.001 g, the detection limit was determined to be  $0.3 \mu\text{g g}^{-1}$ . Precision was indicated by relative standard deviation of duplicate samples between 9 and 21%. Recoveries of matrix spikes ranged from 78 to 111%. Accuracy was indicated by THg concentrations measured in DORM-3 and DOLT-4 of  $85 \pm 16\%$  and  $93 \pm 10\%$  of the certified values, respectively.

**Diet Determination.** The diets of these polar bears have already been reported.<sup>5</sup> Here, we use these diet findings to interpret THg trends. In brief, estimates of the proportional contribution of ringed seal, bearded seal, beluga whale, and bowhead whale to the diets of individual polar bears were determined using quantitative fatty acid signature analysis (QFASA).<sup>39</sup> The weighted mixture of prey fatty-acid signatures that best matched each individual polar bear signature after accounting for differences between predator and prey due to predator metabolism was used to generate diet estimates.<sup>39</sup> The prey fatty acid library containing 89 ringed seal, 20 bearded seal, 64 bowhead whale, and 29 beluga whale samples from the SB region was generated in previous studies.<sup>23,40</sup> The model was run using the “QFASApack” package in R (version 3.1.3).<sup>41</sup>

**Body Condition.** Body mass index (BMI: body mass (kg)/standard length<sup>2</sup> (m<sup>2</sup>)) was used as an indicator of body condition. This standard quantitative condition index for polar bears compares well with other indices that have been used previously.<sup>35</sup>

**Statistical Analysis.** Time trend analysis for hair THg was performed using the procedures outlined by the Arctic

Monitoring and Assessment Program using the PIA statistical software.<sup>42</sup> Log-linear THg trends were evaluated using annual median concentrations for all adults and for adult males and females separately. The slope  $b$  of the log-linear analysis was used to calculate annual percent change as  $(1-10^b) \times 100\%$ . Nonlinear THg trend components were also considered using a simple 3 year running-mean smoother. The nonparametric Mann–Kendall test was used to confirm that a significant log-linear trend was not likely due to leverage of end points in the time series. To evaluate the potential influence of ecological variation (proportional diet estimates, BMI), logTHg trends were reevaluated using the most informative and statistically significant ecological variable as a covariate held at its mean. Correlations were used to assess the relationships of diet estimates and BMI to log-THg concentrations and outliers were excluded ( $n = 3$ ) based on residuals analysis and Cook's distances.

All other statistical analyses were performed using Statistica version 13 (Dell Statistica, Tulsa, OK). Summary statistics were reported as arithmetic means and standard errors of the mean of the THg concentration. As the dependent variable for all inferential statistics, THg concentrations were first log-transformed to meet normality. We examined differences in log-THg concentrations among sex and age classes by one-way ANOVA followed by Tukey's HSD for unequal  $n$ . Because of low sample sizes for other sex and age classes (Table S1), concentrations of THg between fall and spring collected samples could only be compared for adult females during 2008, 2009, and 2011. For each of these years, Student  $t$ -tests were used to compare THg concentrations between spring-sampled and fall-sampled adult females. All results were considered to be statistically significant at  $P < 0.05$ .

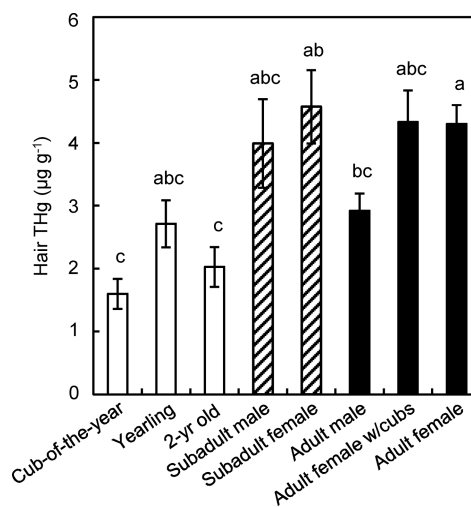
## RESULTS AND DISCUSSION

Considering all bears across all years and seasons, hair THg concentrations in individual SB polar bears ranged widely from 0.6 to 13.3  $\mu\text{g g}^{-1}$  dw, with an arithmetic mean of  $3.5 \pm 0.2 \mu\text{g g}^{-1}$  ( $2.9 \pm 0.3 \mu\text{g g}^{-1}$  for adult males and  $4.3 \pm 0.3 \mu\text{g g}^{-1}$  for adult females). This mean is less than half of that reported for hair THg concentrations in SB bears sampled in spring of 2005 ( $7.5 \pm 0.6$  ( $\pm\text{SD}$ )  $\mu\text{g g}^{-1}$ ), which was the most-recent previous report of hair THg concentrations available for this subpopulation.<sup>17</sup> Unlike in 2005,<sup>17</sup> the 2004–2011 mean was below the revised human hair no-observed-effects-level (NOEL)<sup>43</sup> of  $6.0 \mu\text{g g}^{-1}$  dw and below a subpopulation mean of  $5.4 \mu\text{g g}^{-1}$  dw, for which negative correlations in brain neurochemical receptor activity has been reported in East Greenland polar bears.<sup>18</sup> Our 2004–2011 average was also lower than previously reported in Western Hudson Bay polar bears over a similar time period, 2004–2012 (means of 4.3 and  $4.7 \mu\text{g g}^{-1}$  for males and females, respectively)<sup>44</sup> and also than concentrations reported in polar bears from Baffin Bay in 2001–2008 (median of  $9.4 \mu\text{g g}^{-1}$  versus SB median of  $2.9 \mu\text{g g}^{-1}$ ).<sup>34</sup>

Below, we assess whether these apparently lower hair THg concentrations relative to recent studies in this and other subpopulations in the Western Hemisphere may be a consequence of changes in THg concentrations in SB polar bears from 2004 to 2011 while considering the possible confounding influence sex and age class and season. If THg concentrations in the subpopulation have changed concomitantly with increasing land use, what might be the role of ecological changes? Specifically, we investigate the potential

influence of shifts in feeding habits and body condition on tissue concentrations of mercury, one of the key environmental contaminants of concern with respect to health risks in this and other polar bear subpopulations.

**Sex and Age Class Variation.** Concentrations of hair THg significantly differed among sex and age classes (one-way ANOVA:  $F_{7,190} = 6.51$ ,  $P < 0.001$ ), with this factor explaining 19% ( $\eta^2$ ) of the variation among individuals. Levels in descending order were solitary adult females  $\approx$  adult females with cubs  $\approx$  subadults  $>$  adult males  $\approx$  yearlings  $>$  cubs-of-the-year  $\approx$  2 year old dependent cubs (Figure 1). Post hoc analysis



**Figure 1.** Arithmetic mean total mercury (THg) concentrations ( $\pm\text{SE}$ ) in hair sampled from 198 polar bears of the southern Beaufort Sea subpopulation in 2004 to 2011. Individuals were grouped according to their sex and age class, as cub-of-the-year ( $n = 7$ ), yearling ( $n = 21$ ), 2 year old dependent cub ( $n = 13$ ), subadult (independent 2, 3, and 4 year old) male ( $n = 14$ ), subadult female ( $n = 18$ ), adult (5 years and older) male ( $n = 58$ ), adult female with cubs ( $n = 38$ ), and solitary adult female ( $n = 29$ ). Bars not sharing the same letter denote statistically significant differences.

showed that hair THg levels were higher in solitary adult females than in adult males ( $P = 0.03$ ). Solitary adult females and adult females with cubs showed nearly identical mean THg concentrations ( $4.3 \pm 0.3$  and  $4.3 \pm 0.5 \mu\text{g g}^{-1}$ , respectively). Both solitary adult females and subadult females exhibited higher THg levels than cubs-of-the-year and 2 year old dependent cubs ( $P < 0.05$ ). The mean concentration in females with cubs was higher, though not significantly so ( $0.05 \leq P \leq 0.10$ ), than in adult males, cubs-of-the-year, and 2 year old dependent cubs.

Previous studies in polar bears from the SB and Western Hudson Bay, but not those from East Greenland, also reported higher hair THg concentrations in adult females compared to adult males.<sup>16,17,44</sup> Both former studies suggested that higher concentrations in females in some regions are likely in part driven by sex differences in prey species consumed. Supporting this, we recently showed that during 2004–2012, adult SB males ate nearly twice as much bowhead whale as adult SB females on a biomass basis ( $19.6\% \pm 1.4\%$  versus  $11.0\% \pm 1.1\%$ ), and adult females consumed more ringed seal than adult males ( $48.2\% \pm 1.9\%$  versus  $36.8\% \pm 1.7\%$ ).<sup>5</sup> These findings may partly be related to sexual dimorphism, whereby larger-bodied males may successfully hunt larger prey, such as bearded seals, and thus rely less on ringed seals than females.<sup>3,23</sup> A

higher prevalence of males has also been documented at the onshore bowhead carcasses.<sup>45</sup> Thus, additionally or alternatively, behavioral sexual segregation at the bowhead carcasses may allow dominant adult males to make greater use of this easy-to-access resource, while females and subadults avoid interspecific interactions, instead focusing on the more foraging-intensive resource, ringed seal. The ringed seal represents a higher trophic position food resource (and, thus, a likely greater source of THg) than bearded seal and bowhead whale (trophic positions of  $4.1 \pm 0.2$  versus  $3.8 \pm 0.1$  and  $2.8 \pm 0.1$ , respectively, calculated from  $\delta^{15}\text{N}$ ).<sup>28</sup> Additionally, because ringed seals are smaller than bowheads, belugas, and bearded seals, in addition to lipophilic blubber tissues, other more-proteinophilic (and, thus, Hg-rich tissues) may be consumed more frequently, like meat and internal organs.<sup>17</sup> Sex differences in consumption of prey tissue types has been found in other ursids.<sup>46</sup>

In addition to variation with sex, hair THg concentrations increased with age, with the lowest concentrations in cubs-of-the-year and increasing until the subadult stage when concentrations reached those of adult females (Figure 1). An earlier study did not find differences in blood THg concentrations between SB adult females and young, but age-related differences may not have been distinguishable there because low sample sizes necessitated young polar bears being analyzed as a single group of 1 to 4 year olds.<sup>47</sup> Another study reported no significant variation in liver Hg concentrations among cubs, subadults, and adults from Alaska, although it was likely low-powered as adult concentrations were nonsignificantly higher, and sample sizes were low for cubs and subadults.<sup>48</sup> Conversely, and consistent with our findings, hair THg concentrations were recently reported to increase with age in a detailed analysis of polar bear family groups from Western Hudson Bay, although independent subadults were not evaluated in that study.<sup>49</sup>

Higher concentrations in adult females than in dependent young is likely related to the fact that, unlike for lipophilic organic compounds such as PCBs, transfer to milk does not appear to be major route of elimination for more proteinophilic Hg compounds in adult female polar bears (milk/blood (ww) ratio of 26:1 for PCBs versus 0.6:1 for Hg).<sup>47</sup> Consistent with limited lactational transfer, we did not find differences in THg concentrations between solitary adult females and females with cubs. Similarly, once bears were independent (i.e., subadults), and thus, Hg exposure switched from lactational to dietary, Hg levels increased to those of adult females. We recently showed that the diets of dependent and subadult SB polar bears were not significantly different from those of adult females.<sup>5</sup> Therefore, dietary differences of juveniles and adult females relative to adult males, along with limited lactational transfer of Hg, well-describe the sex- and age-related variation we observed. Interestingly, log-THg concentrations increased linearly with age excluding adult males ( $r = 0.30$ ,  $P < 0.001$ ,  $n = 128$ ). This observation suggests that as young males age, become independent, and increase in size, they are likely exhibiting a transition from food sharing of smaller prey with their mothers to larger, but lower-trophic-level, resources that ultimately decrease their THg exposures. Further investigation of the dietary transitions of cohorts and seasonal habitat choices is warranted to monitor the health and reproductive success of this subpopulation.

Based on sex- and age-related differences, as well as smaller sample sizes for all juvenile classes, we considered only adult

males and females (hereafter combining solitary females and females with cubs), both together and separately, in assessing temporal trends in hair THg concentrations in the SB subpopulation.

**Seasonal Variation.** Evaluation of the potential influence of season on hair THg concentrations was limited. First, samples had to be separated by year to avoid the potential influence of temporal changes because fall sampling only occurred in later years (2008–2011). Second, fall sample sizes were very small, representing just 16% (32/198) of all samples collected. In fact, the only sex and age group and years that had large-enough fall sample sizes ( $n \geq 3$ ) to test for seasonal differences were adult females and in the years of 2008, 2009, and 2011. In each of these three years, hair THg concentrations did not significantly differ between spring and fall sampled adult females ( $P > 0.24$ ; Figure S1). Because this seasonal analysis is based on small sample sizes ( $4 \leq n \leq 7$ ), the findings must be interpreted cautiously.

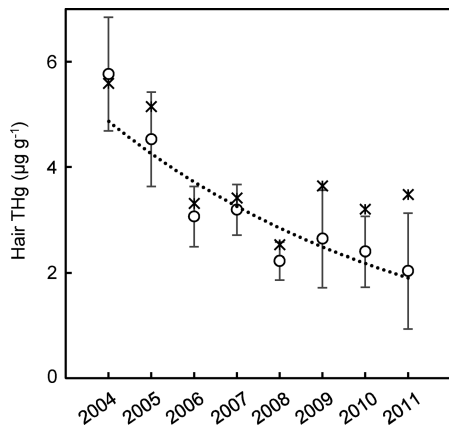
It is often reported that, at least for this polar bear subpopulation, hair growth occurs in the spring and summer. Rogers et al.<sup>24</sup> assumed that, for guard hairs sampled in spring, the hair root stable isotope data could be used to evaluate fall diets, while the hair-tip data could be used to assess summer diets. A study on East Greenland and Svalbard polar bears suggested that the molt starts in May and is complete by September or maybe October.<sup>16</sup> Hair used in the current study was short and cropped close to the skin/hair root. Based on these earlier assumptions, we can speculate then that the close-cropped hairs from spring-sampled bears represent THg exposure from late summer and early fall of the previous year, while the close-cropped hairs from fall-sampled bears represent THg exposure from late summer and early fall of the current year. Nonetheless, hair growth in wild polar bears is not well-studied, and many questions remain in relation to the exact timing and duration of molt, extent of coat replacement, and the possibility of interindividual and interpopulation variation.

Similar, although somewhat higher, hair THg concentrations were found in this subpopulation in 2005 ( $5.7 \mu\text{g g}^{-1}$  in adult males and  $9.0 \mu\text{g g}^{-1}$  in adult females)<sup>17</sup> as we found in the same year ( $4.5 \mu\text{g g}^{-1}$  in adult males and  $5.9 \mu\text{g g}^{-1}$  in adult females). This previous report was based on a larger 2005 sample size and used analysis of the longest trailing guard hairs. Thus, the hairs may have captured Hg concentrations over the entire assumed spring-summer hair growth, while our close-cropped hair samples may have integrated Hg deposited just near the end (perhaps the late summer or early fall) of hair growth when the absence of sea ice over the continental shelf drives some bears to land. These difference in hair sampling, combined with our finding of temporal variation in Hg concentrations (see the **Temporal Trends** section), may explain our finding of slightly lower concentrations relative to the 2005 study.

If the assumption of spring–summer molt is correct, one could reasonably expect some seasonal variation in hair Hg concentrations if, for instance, feeding habits varied interannually as these spring hairs would have been grown one year earlier than the fall hairs. We recently showed significant differences in diets in this subpopulation among the years of 2004 through 2012, related to sea ice conditions.<sup>5</sup> Thus, even though we did not find significant differences in hair THg concentrations between spring and fall, based on the possibility of simply not having enough statistical power to detect seasonal differences and on knowledge of temporal variation in diets

over this time period, we focused the subsequent temporal trends analysis on hair THg concentrations from spring-sampled polar bears only.

**Temporal Trends.** From 2004 to 2011, annual median THg concentrations declined log-linearly in hair from spring-sampled adult polar bears by 13% per year ( $r^2 = 0.84$ ,  $P = 0.002$ ,  $n = 8$  years) (Figure 2). The nonparametric Kendall test was



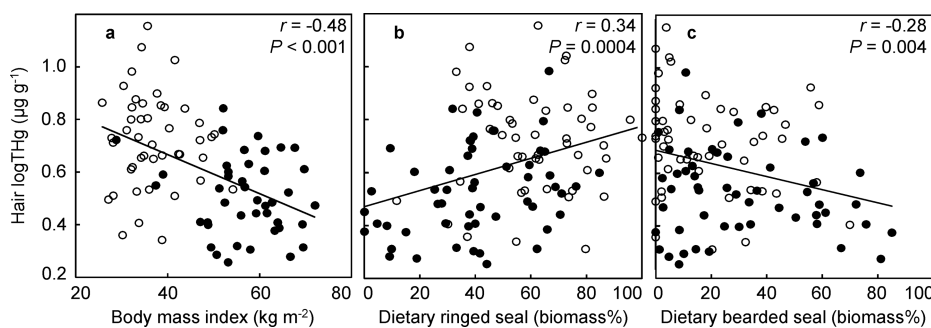
**Figure 2.** Time trends in median hair total mercury (THg) concentrations ( $\pm$ SE) (open circles, dashed line) from adult (males and females combined) southern Beaufort Sea polar bears sampled in the spring from 2004 to 2011. Log-linear declines were significant ( $-13\%$  per year,  $r^2 = 0.84$ ,  $P = 0.002$ ). Annual medians adjusted to the covariate body mass index (BMI) are also shown (asterisks, no line). Log-linear trends were not significant for adjusted median hair THg concentrations ( $-6.6\%$  per year,  $r^2 = 0.41$ ,  $P = 0.08$ ). Raw medians, not log-transformed values, are shown for ease of visualizing the concentration changes.

also significant, suggesting no undue influence of end points ( $P = 0.006$ ), while a nonlinear component based on the 3 year running-mean smoother was not detected ( $P = 0.19$ ). When separated by sex, log-linear declines remained significant for adult males but not for adult females. Concentrations of THg decreased in adult males by 15% per year ( $r^2 = 0.78$ ,  $P = 0.004$ ,  $n = 8$  years; the Kendall test was also significant,  $P = 0.006$ ), from a median in 2004 of  $5.7 \mu\text{g g}^{-1}$  to just  $1.5 \mu\text{g g}^{-1}$  in 2011. Nonlinear trend components were also detected for adult males ( $P = 0.01$ ). While THg concentrations were higher in earlier years (2004:  $6.1 \mu\text{g g}^{-1}$ ) than in recent years (2011:  $5.1 \mu\text{g g}^{-1}$ ) for adult females as well, the slower decline of 4.4% per year was not statistically significant ( $r^2 = 0.13$ ,  $P = 0.38$ ,  $n = 8$  years;

the Kendall test was also not significant,  $P = 0.46$ ). However, nonlinear trend components were also detected for adult females ( $P = 0.03$ ). Log-linear rates of contaminant decrease below 5–10% per year in studies spanning less than 10 years are often not possible to statistically detect due to interannual variation in biological and ecological covariates.<sup>50</sup> The fact that males and females differ in THg declines supports our conclusion that the drivers of THg exposures may differ between sexes.

The declining trend of THg in adult SB polar bears over our 8 year time series is qualitatively consistent with temporal changes reported for liver THg concentrations of Alaskan polar bears from 1995 to 2002.<sup>48</sup> In that study, THg concentrations declined by 34%. A decline in Hg concentrations in blood was also reported for adult SB polar bears between 2005 and 2007.<sup>51</sup> Consistent with these studies, our later time series from 2004 to 2011 suggests a continued and possibly steeper decline by 65% in adult polar bears. It was previously reported that SB polar bear liver THg concentrations were similar in 2007–2008<sup>13</sup> to concentrations reported in an earlier study from 2002,<sup>52</sup> which, in turn, were higher than those reported in 1982.<sup>53</sup> However, as the authors point out, these limited time-point comparisons (especially across multiple data sets and laboratories) should be treated with caution as interannual variation combined with just a few years of data may prevent detection of trends or lead to erroneous assessment of trends.<sup>50</sup> Annual sampling from larger numbers of years, such as was done in the current study and Kannan et al.,<sup>48</sup> revealed significant declines of THg in SB polar bears. Considering these studies together, we can speculate that Hg concentrations peaked in this subpopulation sometime in the 1980–1990s and subsequently declined.

These declining temporal trends appear to differ from those reported for other polar bear subpopulations and marine biota of the Western and Central Arctic (i.e., Alaska, Canadian Arctic, and western Greenland). In a large data set, hair THg concentrations in polar bears in Northwest Greenland increased from 1987 to 2008 by 2% per year.<sup>34</sup> An earlier study similarly showed increases in this subpopulation over 1892–1991.<sup>37</sup> No trend in Hg concentrations was found in muscle of ringed seals from the Beaufort Sea from 1973 to 2007.<sup>54</sup> In a meta-analysis of Hg trends in Arctic biota,<sup>55</sup> only nonsignificant or increasing trends were found in studies on biota from the western North American Arctic. In contrast, in the Eastern Arctic, Hg concentrations slowly declined by 0.8% per year in East Greenland polar bears from 1973 to 2001.<sup>37</sup> Differences between these North Atlantic (Eastern) Arctic



**Figure 3.** Correlation between hair total mercury (THg) concentrations in adult female (open circles) and adult male (closed circles) 2004–2011 southern Beaufort Sea polar bears and (a) body mass index (BMI), (b) dietary proportions of ringed seal, and (c) dietary proportions of bearded seal.

trends versus Northern Canadian and Western Greenland (Western and Central) trends for polar bears have previously been suggested to be related to decreases in North American and European Hg emissions but increased emissions from Asia over the past few decades.<sup>37</sup> As SB polar bears are one of the most westerly located North American Arctic subpopulations, it is thus surprising that we found declines, especially at such a relatively rapid rate of 13% per year. Although it is possible that changes in Hg inputs, deposition, or biogeochemical cycling in the region may have influenced Hg trends in SB polar bears, such explanations do not fit well with the apparent lack of Hg decline in their main prey within the region, albeit with a time series only extending until 2007.<sup>54</sup> Declines in Hg concentrations in SB polar bears, therefore, likely involved ecological changes, which have been documented in this subpopulation,<sup>5,24,31,32</sup> specifically shifts in feeding habits and body condition.

**Influence of Diet and Body Condition.** Concentrations of hair THg in adult male and female SB polar bears from 2004 to 2011 were associated with both body condition and feeding habits. There was a log-linear decrease in THg concentrations with increasing BMI ( $r = -0.48$ ,  $P < 0.001$ ; Figure 3a). Concentrations of THg also increased with greater proportional biomass of ringed seal consumed ( $r = 0.34$ ,  $P = 0.0004$ ; Figure 3b) and decreased with greater proportions of bearded seal consumed ( $r = -0.28$ ,  $P = 0.004$ ; Figure 3c). A lack of significant correlation of THg with bowhead and beluga whale could, in part, be due to the higher frequency of nondetects of these prey as food items, relative to ringed and bearded seal. The BMI was negatively correlated with ringed seal consumption ( $r = -0.35$ ,  $P = 0.0007$ ) and positively correlated with bearded seal consumption ( $r = 0.30$ ,  $P = 0.004$ ). Although not reaching statistical significance, BMI additionally tended to increase with bowhead whale consumption ( $r = 0.20$ ,  $P = 0.06$ ) and decrease with beluga whale consumption ( $r = -0.20$ ,  $P = 0.06$ ). Thus, BMI, overall dietary patterns, and hair THg concentrations were all linked.

The directionality of these relationships is consistent with our expectations of the effects that body condition and diet should have on hair THg concentrations. When prey is unavailable, polar bears rely heavily on built-up fat reserves for their energetic requirements.<sup>56</sup> However, significant protein loss also occurs during fasting and, to a greater extent, for polar bears with lower initial fat stores prior to fasting.<sup>57</sup> Because MeHg is proteinophilic, we suggest that catabolism of proteinaceous tissues can lead to its release into circulation and, thus, greater deposition into hair in these animals. Fasting-induced increases in the blood concentrations of lipophilic PCBs and other POPs have already been documented in polar bears,<sup>33</sup> other ursids,<sup>58</sup> and other Arctic carnivores.<sup>59</sup> In the SB, polar bear fasting rates (based on serum urea-to-creatinine ratios) were shown to increase from 1985–1986 to 2005–2006, corresponding with large-scale changes in sea ice.<sup>60</sup> It has been predicted that such increased fasting rates in polar bears would lead to higher concentrations of POPs in blood and target tissues and thus increases in the toxicological risks posed by these compounds.<sup>1</sup> In support of this, a negative relationship between body condition and blood POPs concentrations was reported in polar bears sampled in Svalbard in 2012–2013 and in SB polar bears in 2007.<sup>61,62</sup> Under such climate change scenarios,<sup>1</sup> our results suggest that the circulating concentrations and thus toxicological risks of MeHg and other proteinophilic contaminants for polar bears would increase,

similar to the POPs. Support for this phenomenon comes from a recent study demonstrating a >10% increase in circulating THg concentrations in zebra finches (*Taeniopygia guttata*) after an 18–21 h fast, during which lean body mass decreased.<sup>63</sup>

Higher THg concentrations in polar bears that consumed proportionally more ringed seal and less bearded seal could be explained by (1) the relative trophic position of these two prey items or (2) differences in the types of tissues consumed for each prey. Ringed seals in the Beaufort–Chukchi Sea sampled in 1999–2000 appeared to feed at a somewhat higher trophic position (3.8 versus 4.1 calculated using a  $\delta^{15}\text{N}$  approach) and showed higher blubber concentrations of PCBs and a number of other POPs than bearded seals.<sup>64</sup> Because ringed seal are a smaller-bodied prey relative to bearded seal, polar bears may consume their blubber, as well as meat and viscera, the latter two being proteinaceous tissues where MeHg is stored; conversely, polar bears may nearly exclusively eat the less Hg-contaminated blubber of larger prey, such as bearded seal.<sup>65</sup>

The significant association between body condition and specific prey items consumed prevented us from evaluating the effects on hair THg concentrations of each ecological factor in isolation. Because the strongest correlation to THg concentrations was for body condition, we used BMI as the covariate for testing how ecological shifts may have contributed to the observed temporal changes in hair THg concentrations from 2004 to 2011.

When the annual median concentrations of hair THg were adjusted to the mean value of the covariate BMI, the annual decline fell to a nonsignificant rate of 6.6% per year ( $r^2 = 0.41$ ,  $P = 0.08$ ; the Kendall test was also not significant,  $P = 0.22$ ). Nonlinear trend components were similarly not significant ( $P = 0.07$ ). Although the 2004–2005 medians remained relatively high after adjustment, the 2006–2011 medians stabilized (adjusted values shown in Figure 2; separate adjusted values for adult males and females in Table S2). That is, after accounting for variation in body condition, hair THg concentrations no longer significantly declined over the 2004–2011 time period, likely due to an increase in BMI over 2004–2011 ( $r^2 = 0.06$ ,  $P = 0.02$ ). This finding, as well as the differences in trends between males and females, is strongly supportive of ecological changes, as opposed to declining environmental concentrations, as the drivers of these short-term temporal declines in hair THg concentrations in SB polar bears.

It is critical to put these results in a broader and longer-term context of potential adverse health effects for SB polar bears. Exposures to Hg represent a top concern regarding health risks posed by contaminants to Arctic biota, and although THg concentrations appear to be in decline in SB polar bears, concentrations appear to be stable or increasing in other subpopulations in the Western North American Arctic.<sup>7,55</sup> Although, as we have previously reported, body condition in this polar bear subpopulation did not decline from 2004 to 2012,<sup>5</sup> earlier studies that included multidecadal time periods have reported declines in the body condition of SB polar bears.<sup>31,32</sup> In addition, if the bears that come ashore in the fall show year-round fidelity to the coast, it is possible that the spring capture samples in recent years could be skewed toward those bears with more ample supply of coastal foods. Therefore, the short-term declines in THg concentrations observed in our study may not persist if considered over a longer time period or over other spatial scales. If improved body condition is related to the consumption of onshore bowhead whale subsidies,<sup>5</sup> we

can speculate that if sea ice continues to decline, more bears will continue to come ashore to take advantage of this resource. At some point, the subsidies may not be sufficient for the number of individuals making use of them. Approximately 20% of the SB population currently makes use of bowhead whale carcasses as a food resource, leaving the remaining 80% on the sea ice during summer and fall months, possibly fasting until freeze-up, when they are able to hunt in more-productive near-shore habitats. Animals remaining onshore and on the sea ice during summer, therefore, can be anticipated to experience further declines in body condition with projected changes in the sea-ice habitat.<sup>6</sup> Extended fasting periods that lead to reductions in body condition are expected to also increase the circulating levels of lipophilic and proteinophilic compounds and, thus, the toxicological risks posed by a wide range of environmental pollutants.

## ■ ASSOCIATED CONTENT

### ● Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b00812.

Tables showing sample sizes and separate adjusted trends for adult males and females. A figure showing seasonal variation. (PDF)

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### Notes

The authors declare no competing financial interest.

## ■ ACKNOWLEDGMENTS

We thank Sniega Stapcinskaite (University of Connecticut) for THg instrumental analysis and Katrina Knott for reviewing an earlier version of the manuscript. Karyn Rode, George Durner, Anthony Pagano, Kristin Simac, Tyrone Donnelly, and Karen Oakley (U.S. Geological Survey) are acknowledged for field logistics and sample collection. Funding came from the USGS's Changing Arctic Ecosystem Initiative and from the Wildlife Program of the USGS Ecosystem Mission Area. Further funds and in-kind support were from the USGS Climate and Land Use Change Mission Area, the Bureau of Ocean Energy Management, the Bureau of Land Management, and the Arctic National Wildlife Refuge. The research was approved under the Marine Mammal Protection Act and Endangered Species Act with USFWS permit no. MA690038. Capture protocols were approved by the USGS Institutional Animal Care and Use Committee. Any use of trade, firm or product names is for descriptive purposes only and does not reflect endorsement by the U.S. government.

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