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Comparative review of the distribution and burden of contaminants in the body of polar bears

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Response to Reviewers:	Reponses to reviews R: The pages are not correctly paged, all pages are paged "no. 8"!

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Comparative review of the distribution and burden of contaminants in the body of polar bears

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1. Abstract

Historical (or legacy) contaminants, such as metals and persistent organic pollutants (POPs; e.g., polychlorinated biphenyls) have been measured in circumpolar subpopulations of polar bears, especially from Hudson Bay, East Greenland and Svalbard, but substantially less is currently known about new and/or emerging contaminants such as polychlorinated naphthalenes, current-use pesticides, organotins, and polycyclic aromatic compounds (PACs). The polar bear (*Ursus maritimus*) is an apex Arctic predator that accumulates high levels of bioaccumulative POPs and mercury (Hg), but there is currently no comprehensive profiling of the present knowledge on contaminants in tissue and body compartment in polar bears. Based on current literature reports and data, and including archived museum samples (as far back as the 1300s) and up to 2018, the aim of this review is to utilize available data to examine the comparative distribution and burden of mainly lipophilic contaminants in kidney, liver, fat, and other body compartments, such as milk, blood, and brain. Highlight outcomes from this review include the following: 1) the kidneys are one of the most important tissue depots of contaminants in polar bears; 2) there is a critical lack of data concerning the presence of metals of concern (other than Hg); and 3) there currently are no data available on the concentrations of many newer and emerging contaminants, such as PACs, which is especially relevant given the increasing oil and gas development in regions, such as the Beaufort Sea (Canada). Additionally, given the vulnerability of polar bear populations worldwide, there is a need to develop non-invasive approaches to monitor contaminant exposure in polar bears.

Keywords: Polar bears, Arctic, tissues, body distribution, contaminants, review.

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2. Introduction

The polar bear (*Ursus maritimus*) is a sentinel species of environmental changes and stressors in the Arctic. The total number of polar bears is estimated to be between 23000 and 26000 bears (Hamilton and Derocher, 2018; Wiig et al., 2015). There are 19 subpopulations worldwide with 13 being fully or partly under Canadian jurisdiction (Obbard et al., 2010; Thiemann et al., 2008a). Polar bears live in both terrestrial and marine environments, and their home range can vary between 140000 km^2 to over 350000 km^2 (Auger-Méthé et al., 2016; McCall et al., 2015). Polar bears are considered vulnerable by the International Union of Conservation of Nature (IUCN) (Wiig et al., 2015) as their population is affected by several environmental factors, including climate change (AMAP, 2018; McKinney et al., 2013; Letcher et al., 2010). Another major stress experienced by polar bears is the exposure to a growing number of anthropogenic and natural chemical contaminants via local Arctic sources, but mostly via long-range atmospheric as well as oceanic transport to the Arctic (AMAP, 2017; AMAP, 2016; Routti et al., 2019).

Consumption of the fat-rich blubber layer of seals or other marine mammal prey is crucial for the survival of polar bears (Peacock et al., 2010). As a top marine predator, polar bears biomagnify many contaminants in their tissues via the diet. They feed mainly on ringed seals (*Pusa hispida*), but also on other marine species, e.g., bearded seals (*Erignathus barbatus*), harp seals (*Pagophilus groenlandicus*), harbor seals (*Phoca vitulina*), hooded seals (*Cystophora cristata*), walrus (*Odobenus rosmarus*), beluga (*Delphinapterus leucas*), narwhal (*Monodon monoceros*), and can also scavenge on other whale species such as bowhead whale (*Balaena mysticetus*) carcasses (Thiemann et al.,

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2008b; AMAP, 1998; Routti et al., 2019). There have been shifts in the diet (prey consumption) of polar bears as well as to the underlying food web structure, and in connection to climate change, which influences pollutant concentrations in polar bear tissues. For example, an increase in the proportion of open water-associated seals species and a decrease of the ice-associated seal species consumed has been shown for Hudson Bay and East Greenland polar bears (McKinney et al., 2009). Using fatty acids as tracers of diet for East Greenland bears, McKinney et al. (2013) estimated that there was 14% reduction/decade of ringed seal consumption compared to an 9.5% increase/decade in feeding on hooded seal (from 1984 to 2011). In the same study, it was reported that this diet shift was associated with a reduction in the concentration of several chlorinated POPs (persistent organic pollutants), but with an increase of the brominated POPs, since the ringed seals were less contaminated than the hooded seals in brominated POPs (McKinney et al., 2013). Harp seals yielded higher levels of contaminants compared to ringed seals (e.g., 6-fold for chlordanes, 4-fold for polychlorinated biphenyls (PCBs), and 3-fold for dichlorodiphenyltrichloroethane (DDT)) (Kleivane et al., 2000).

The levels of POPs and bioaccumulative metals such as cadmium or lead are generally the highest in the polar bear as compared to other Arctic wildlife, and they are an ideal wildlife receptor for the biomonitoring of spatial and temporal trends, distribution, dynamics, fate, biomagnification, and potential effects of anthropogenic contaminants as has been reported for Hg (mercury), and legacy and emerging POPs (AMAP, 2018; AMAP, 2017; Letcher et al., 2010). In Hudson Bay polar bears, levels of some legacy POPs, such as PCBs, have been shown to have decreased prior and up to 2000 (starting from 1980), but have since remained relatively unchanged up to recent times (Letcher et al., 2018; Brown et al., 2018). A number of new and emerging environmental pollutants, such as polybrominated diphenyl ether (PBDE) flame retardants and per-/poly-fluoroalkyl substances (PFAS; especially, the highly bioaccumulative perfluorooctane sulfonic acid (PFOS)) have been reported (Letcher et al., 2018; AMAP, 2017; AMAP, 2016). There are potentially many more POPs in polar bears. Using non-target chemical analysis, Liu et al., (2018) recently reported on the discovery of many additional contaminants in the serum of polar bears from Hudson Bay and the Beaufort Sea, including novel PCB metabolites and many fluorinated or chlorinated substances not previously detected. Using non-target chemical analysis of archived polar bear serum samples (mid-1980s), all fluorinated classes of contaminants showed increasing over time. Altogether, polar bears are exposed to a complex array of known contaminants to multiple routes of exposure, but mainly through diet.

As polar bears are being monitored over time by governmental agencies for contaminant burden, it is critical to understand what is presently known about the contaminant exposome in polar bear tissues and body compartments. A recent review by Routti et al. (2019) focused entirely on trends, fate, and potential health effects of contaminants in polar bears. However, to our knowledge, there has yet to be comprehensive comparative assessment and review of the distribution and burdens of contaminants in the body of polar bears. The main objective of the present review is to compile all available contaminant data in polar bears. Reported data came from papers published between 1990 and 2018 based on samples collected as far back as the 1300s because of the availability of archived museum samples, but data come predominantly from the last three decades and up to 2015. The available data were analyzed to provide insights on the relative importance of contaminant burdens in tissues and blood. We also highlighted current deficiencies in the understanding of the distribution of contaminants in polar bears and provided recommendations that should be considered for continued monitoring assessment of contaminants in circumpolar polar bear populations.

3. Methods

In order to understand the distribution of contaminants in the tissues and body compartments of polar bears, an exhaustive review of the literature was conducted. Briefly, the keywords used included: polar bear + contaminant or toxicity and articles were searched in the Scopus database during September 2017 to September 2018. Most literature studies only assessed one tissue type in polar bears and for a select number of contaminants (AMAP, 2017; AMAP, 2016; Letcher et al., 2010). By narrowing down the papers published (meaning papers that were measuring the levels of contaminants in polar bear tissues) and by compiling data from available studies, we were able to estimate the amount of burden for each tissue and contaminant or contaminant class. Furthermore, data from the published reports produced by the Arctic Monitoring and Assessment Programme of the Arctic Council (AMAP) were also included, in addition to the Scopus database research. In total, 81 published research articles and AMAP reports were found, and the median concentration was used in this review (i.e., for each contaminant and tissue). The median concentrations were a better representation of the data as compared to mean concentrations, as the former are less skewed due to exceptionally high or low concentration data. The concentration intervals were based on quantiles, thus the first quartile (25 %) and the third quartile (75 %) were used to show the variation between each median value. Data were distinguished based on pre- and post-2000 and categorized

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according to the year the samples were collected. Available data over sampling periods (e.g., 1993-2008) were classified according to the longest portion of the sampling period occurring pre- or post-2000. The year 2000 is a logical contaminant delineation point for the Arctic as 1) temporal studies show that prior to 2000 many of the legacy contaminants deceased substantially in biota and in wildlife tissues (mainly in the fat) had more-or-less remained constant post-2000, 2) UNEP's Stockholm Convention on POPs came into existence in 2001 and listed the original 12 "dirty dozen" POPs, 3) in the early 2000s "newer" POPs, such as PBDEs and PFOS, had been voluntarily phased out by industry, and 4) the POPs list was expanded in 2009 to include 9 new compounds including PBDEs and PFOS (and related compounds), and subsequently has been expanded further with newer POPs listed up until 2018 (e.g., polychlorinated naphthalenes (PCNs), short-chain chlorinated paraffins (SCCPs) and hexabromocyclododecane (HBCDD)). A comprehensive summary of contaminant data according to the tissue or body compartment can be found in the Supplementary file for total mercury (THg), pesticides, metals, PCBs, and flame retardants (Tables S1 to S5). In order to be included in Table S2~~1~~, metals studied~~s~~ needed to provide data for at least two different metals in the same tissue in order to calculate a median, excluding THg. Metals reported in previous studies were: Ag (silver), As (arsenic), Ba (barium), Bi (bismuth), Cd (cadmium), Co (cobalt), Cr (chromium), Cu (copper), Fe (iron), Mn (manganese), Mo (molybdenum), Pb (lead), Rb (rubidium), Se (selenium), Sr (strontium), V (valium) and Zn (zinc). Studies have reported contaminant levels on a wet, dry, or lipid weight basis and the published concentrations were compiled as is, as performed in Letcher et al., 2010. As a result, most of the contaminants were reported on a wet weight basis, except

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for the fat, which was mostly measured as lipid weight and hair data were entirely reported as fresh weight. Biological information on polar bears, such as age and sex, were not used in the present assessment as the focus of the review is on organic contaminant concentrations in tissues and body compartments regardless of gender or age. The data for specific studies can be found via several Arctic reviews (AMAP, 2018; AMAP, 2016; Sonne, 2010; Letcher et al., 2010; Braune et al., 2005). Statistical analysis was conducted with JMP Genomics 6.0. Non-parametrical data were analyzed with a Wilcoxon test with chi square approximation.

4. Results and discussion

4.1 Cocktail of contaminants present in polar bears

Figure 1 illustrates the sum of all contaminant concentrations reported in polar bear tissues, blood, and other compartments (brain, brainstem, lymph nods, spleen, and teeth) and encompassing all available data published up to 2018. A maximum of 121 individual contaminants have been reported at quantifiable levels in polar bear tissues and/or body compartments, including metals, pesticides, FRs (flame retardants), such as PBDEs, among others (Tables S1 to S5) pre-2000; whereas up to 336 individual contaminants have been measured post-2000. Therefore, data presented in Figure 1 must be carefully interpreted. A general approach was used to present known data of contaminants in polar bears. The median is formed with wet, dry and lipid weight and also depends of contaminants analyzed. If a compound was or was not analyzed and/or detected in polar bear tissues, it changes the values obtained in Figure 1. The main goal of this figure is to give an idea of the distribution of contaminants and a summary of previous research. Of note, out of the 209 PCB congeners, only 127 of them had levels above the detection limit. Except for blood, there were significant differences in the overall median sum concentrations for each tissue or body compartment (i.e., hair and milk) pre- and post-2000. The median level of the overall sum concentration of contaminants were significantly greater pre-2000 ($p < 0.0001$), except for milk, blood, and other tissues where there was no significant difference. For hair, the opposite was observed, as the level of contaminants was significantly greater post-2000 ($p < 0.0003$). Closer examination of the pre- and post-2000, overall median concentrations of POP and THg contaminants in different tissues and body compartments are shown in Figure 1; whereas, Figure 2 shows the median concentrations for the detailed contaminant categories. Also, in Figure 2 "ND" represent compounds that were analyzed but below the detection limit. As presented in Figure 2, there is a lack of consistency in contaminants analysis depending of the period and the tissue. Therefore, it is crucial to gather more information to allow a better monitoring on polar bears. More data will be helpful to compare the burden and distribution of different compounds in polar bear tissues and change of contaminant levels through time.

4.2 Kidneys

As shown in Figure 1, kidneys have the highest concentrations pre-2000, after liver, pre-2000 with medians of 11640 ng/g (25 %: 5405 ng/g; 75 %: 26550 ng/g). For example, in the liver ΣPCBs reached 7240 ng/g; whereas, in the kidneys they yielded 7020 ng/g post-2000. Of note, lipid normalization was more frequent when reporting kidneys' contaminant levels post-2000, but should not have affected this trend given that kidneys are a lean tissue. These high kidney levels are driven by three groups of contaminants:

metals, ΣPCBs, and ΣPFAS (Figure 2A). Elevated levels of Cd (cadmium) and Se (selenium) were reported in kidney samples as compared to all the other tissues (not shown). Cd is a non-essential element, while Se is an essential element and is believed to reduce the toxicity of some metals, and in particular Hg (AMAP, 1998). Inorganic Hg binds with Se, allowing sequestration and making Hg non-toxic (AMAP, 2011). Other studies have shown that Cd and Se concentrations were also higher in the kidneys than in the liver. For example, Woshner et al. (2001) calculated that Cd is at 20 times higher concentration in the kidneys relative to liver. Dietz et al. (2000) found that levels of Cd, Se, THg (includes inorganic Hg and organic MeHg (methylmercury)), and Zn (zinc) were the most concentrated in bears from North-West Greenland subpopulations. That is, median concentration of THg in kidneys was similar pre-2000 (i.e., 12700 ng/g; quantile 25 %: 10543 ng/g; quantile 75 %: 28533 ng/g) and post-2000 (18000 ng/g; quantile 25 %: 11000 ng/g; quantile 75 %: 24000 ng/g). Similarly, THg medians found in liver pre-2000 (12900 ng/g; quantile 25 %: 7560 ng/g; quantile 75 %: 24800 ng/g) were similar to post-2000 (18000 ng/g; quantile 25 %: 11250 ng/g; quantile 75 %: 31468 ng/g) (kidneys: $p = 0.4892$, liver: $p = 0.1442$; Figure 2B). AMAP (2005) and Dietz et al. (1995) have previously reported THg concentrations to be highest in the kidneys, compared to liver and muscle in polar bears from Central East Greenland (sampling years from 1983 to 1987). This discrepancy could be explained by the large numbers of samples assessed for liver as compared to kidneys, which could have skewed the median calculations.

High burdens of organochlorine pesticides, as well as PCBs, had a high burden in tissues of polar bears. Based on mostly on fat tissue levels, chlorinated biphenyls (CB)-99, CB-138, CB-153, CB-170/190 and CB-180 congeners generally constituted 79-95 % of

ΣPCB concentrations in polar bears (Gebbink et al., 2008b; Kannan et al., 2005). The presence of only a few dominant PCB congeners in polar bears is a reflection of the efficient metabolic biotransformation of most PCB congeners (Kannan et al., 2005; Letcher et al., 2000). According to Weijs et al. (2010b), the concentration of PCB congeners, such as CB-153 can be estimated using the lipid content in the kidneys or the liver. Also, PCBs with a higher degree of chlorination tend to accumulate more in the kidneys and liver as compared to fat, probably because these compounds cannot pass through the biological membranes (Weijs et al., 2010a). Those congeners have a high hydrophobicity and membrane permeation resistance slows the uptake of those PCBs, resulting in a less effective food chain transfer (Kannan et al., 1998). Other contaminants such as PBDEs, DDTs, CHLs, and ClBzs had much lower levels in polar bear liver. Future research or monitoring assessments of polar bear populations should include kidneys as one of the tissues of interest for the important role of kidneys in detoxification and in bioconcentration of pollutants (organism sink for contaminants).

4.3 Hair

After the kidney, hair was the third highest contaminated tissue in polar bears (1635 ng/g pre-2000 (quantile 25 %: 29 ng/g; quantile 75 %: 4998 ng/g) Figure 1). Although hair's contaminant levels were reported as fresh weight, hair yielded very high proportion of THg compared to the other contaminants detected (including ΣCHL (chlordane), ΣDDT, ΣHCH (α-, β- and γ-hexachlorocyclohexane isomers), ΣPBDE, and ΣPCBs; Figure 2C). Hg is known to bind to proteins and hair growth is one of the mechanisms of excretion of Hg-bounded proteins in mammalian species (St Louis et al., 2011; AMAP, 2011). Interestingly, older polar bears have higher THg in hair, partly due to differences in food intake (from milk to solid food) (Bechshoft et al., 2016). Similarly, when POPs are present in the blood, they can be incorporated in different tissues, including in hair (Bechshøft et al., 2012). However, there is a point of saturation at which contaminants can no longer bioconcentrate in hair (Bechshøft et al., 2012). Although it is well documented that polar bear hair can accumulate elevated amounts of Hg (Bechshoft et al., 2016; St Louis et al., 2011; Dietz et al., 2011; AMAP, 2011; Dietz et al., 2006; AMAP, 2005), the tissue comparison in Figure 1, highlights the contrast of Hg loading among polar bear tissues and the Hg proportion compared to the other contaminants.

Jaspers et al. (2010) suggested that polar bear hair may reflect the internal body burden of PCBs since its chemical profile is similar to internal tissues (adipose, liver, brain, and blood). However, our data analysis did not come to the same conclusion. On the contrary, our data suggested that hair would significantly misrepresentative of the polar bear body burden for some contaminants, such as PCBs (Figure 1). Jasper and colleagues (2010) also stated that in general, organohalogen contaminant concentrations in hair are lower than concentrations in adipose or liver tissue, but are similar to brain tissue and higher than concentrations in blood (Jaspers et al., 2010), which is the same pattern observed in this present review. Hair could be a good bioindicator of polar bear loading for some contaminants, especially since long-term dietary exposure dependent on the period of hair growth (Cardona-Marek et al., 2009). However, hair does not appear to be the best tissue to represent the overall polar bear pollutant exposure.

4.4 Liver

As shown in Figure 1, the median pre-2000 contaminant concentration is high in the liver, and where the processes of detoxification take place and one of the most studied organs in polar bear ecotoxicology based on this literature review (AMAP 2018; Bechshøft et al., 2018), with a median of 1395 ng/g (quantile 25 %: 41 ng/g; quantile 75 %: 30675 ng/g) pre-2000 and a median of 115 ng/g (quantile 25 %: 19 ng/g; quantile 75 %: 965 ng/g) post-2000. Similar chemical fingerprints were found for the liver (Figure 2B) and kidneys (Figure 2A) for metals and ΣPCBs, where both had the highest levels of contaminants; however, the median analysis found that the metals (other than Hg) and ΣPCBs were approximatively 4- to 5-fold less concentrated in the liver compared to the kidneys. Chlordanes and dieldrin were the next most concentrated contaminant measured in the polar bear liver (Figure 2B). Chlordanes encompassed a group of six congeners, with oxychlordane representing around 72 % and 79 % of ΣCHL (AMAP 2016; Bentzen et al., 2008b; Bernhoft et al., 1997; Letcher et al., 2010, 2018).

As the top Arctic predator, contaminant levels are higher in polar bears compared to its mainly seal prey, although this was not the case in the present study for PBDEs. Most BFRs (brominated flame retardants) were reported to be at lower concentrations in the polar bear compared to the ringed seal, which suggested a different rate of biotransformation and biodegradation (AMAP 2017; Sørmo et al., 2006). Polar bears showed a significantly lower concentration of the congener brominated diphenyl ethers (BDE)-47 compared to ringed seals (Wolkers et al., 2004), but only a few PBDEs have previously been detected (BDE-47, BDE-99, BDE-100, BDE-153 and BDE-154) (Letcher et al., 2009, 2018). PBDEs such as BDE-209 have been shown to be depleted by polar bears microsomes, and a smaller depletion was also found for BDE-99, BDE-100, and BDE-154 (McKinney et al., 2011). Other BFRs such as HBCDD (hexabromocyclododecane) isomers were not found to biomagnify from ringed seals to polar bears, which indicates that it is metabolized in polar bears (Sørmo et al., 2006). Future studies should include a more detailed analysis on the sexual dimorphism of the hepatic enzymes involved in biotransformation and biodegradation to further assess the contaminant burden risk between sexes.

4.5 Milk

Milk (375 ng/g (quantile 25 %: 326 ng/g; quantile 75 %: 2607 ng/g) pre-2000 and 201 ng/g (quantile 25 %: 28 ng/g; quantile 75 %: 374 ng/g) post-2000) showed a more elevated sum contaminant concentrations than in adipose tissue (Figure 1). This is partly explained by the high levels of ΣCHL found in milk. More specifically, ΣCHLs, ΣClBzs, ΣHCHs, ΣPCBs, and THg were reported in polar bears milk (Figure 2D). Polar bears milk contains from 5-46% lipid (Oftedal, 2011; Hedberg et al., 2011; Derocher et al., 1993), whereas liver contains around 5-15% of fat (Jaspers et al., 2010; Gebbink et al., 2008b; Kannan et al., 2005; Corsolini et al., 2002; Wiberg et al., 2000). Milk is a pathway of elimination for the contaminants in reproductive females (Henriksen et al., 2001) due to lipid mobilisation for lactation (Polischuk et al., 1995). This elimination function is highlighted by data that demonstrated that females with young cubs who had nursed for less than a year showed the highest body burden depuration through nursing (Henriksen et al., 2001). In spring, females with yearlings had lower concentrations of β-HCH than other females, most likely a consequence of POP transfer from mother to young (Tartu et al., 2017). In lactating bears, similar levels of CHLs, DDE (dichlorodiphenyldichloroethylene), HCB (hexachlorobenzene), HCHs, and PCBs were found in lipids of milk and plasma (Bernhoft et al., 1997). HCHs are present in the environment in different forms, and β−HCH is the most persistent and bioaccumulative of the HCH isomers (Kannan et al., 2005). It represents around 70 % and 82 % of ΣHCHs levels found in polar bear (McKinney et al., 2010; Bentzen et al., 2008a; Kannan et al., 2005; Bernhoft et al., 1997). According to Bernhoft et al. (1997), high HCH level in milk was found, which was similar to what was found in subcutaneous tissue and plasma, which differs from our results, as milk has a more elevated burden than plasma. However, no differences of ΣPCB levels was observed between adult female and male polar bears (Sonne et al., 2005; Kannan et al., 2005; Henriksen et al., 2001; Bernhoft et al., 1997), suggesting that there was a low rate of transfer of PCBs into milk (Bernhoft et al., 1997). Also, as there are similar PCB congeners found in milk and blood of polar bears, this suggests that there is little selective chemical transfer from the diet to maternal blood to milk during mid to late lactation (Knott et al., 2012). The lactational transfer of contaminants from dietary sources may be more prominent in female polar bears that have minimal fat reserves after maternal fasting (Knott et al., 2012). However, Bernhoft et al. (1997) found lower levels of PCBs in milk compared to plasma, as the most lipophilic PCBs seems less transferable to milk lipids than to plasma lipids (Bernhoft et al., 1997). Also, for the more metabolizable PCBs, the metabolic capacity may make excretion into milk relatively less important than believed (Olsen et al., 2003; Bernhoft et al., 1997). In addition, Knott et al. (2012) found that when THg concentration data are adjusted to lipid level, milk was three times less concentrated than blood. Therefore, pollutants presence in milk seem to vary depending of the chemical compound in the general condition of the female polar bear. Further research is required to understand the possible accumulation of contaminants in milk and their transfer to offspring.

4.6 Adipose tissue

POPs (e.g., ΣPCBs, ΣDDTs, ΣCHLs, ΣHCHs, and ΣClBzs) were the predominant contaminants measured in adipose tissue (fat). Indeed, it is well known that more than 91 % of total body POP burden are sequestered in adipose tissue (Gebbink et al., 2008a). The presence of contaminants in adipose tissue may change throughout the year since fat levels fluctuate as has been reported for Hudson Bay (Polischuk et al., 1995) and Svalbard polar bears (Tartu et al., 2017). For Svalbard bears, POP concentrations in the blood were shown to be higher in the spring than in the other seasons as during the fasting period polar bears metabolize their lipid stores, which releases contaminants into the blood stream (Tartu et al., 2017). On the contrary, in the fall polar bears increase their fat reserves, and along with increased POP deposition (Tartu et al., 2017). Noteworthy, Hg has not been reported or was below the detection limit in polar bear adipose tissue (Woshner et al., 2001). Fat is one of the most studied tissues and the diversity of contaminants analyzed is greater than in other tissues. For example, 13 contaminant groups were measured in pre-2000 and 17 contaminants groups were measured post-2000, which is less than liver with 14 groups (pre-2000) and 8 groups (post-2000) (Figure 2E).

4.7 Muscle

Adipose and muscle tissue have similar overall median contaminant burdens, but with adipose tissue containing higher concentrations of ΣCHLs (Figure 2E), while muscle has higher levels of ΣPCBs (Figure 2F).Though some authors observed lower levels of contaminants in muscles compared to adipose tissue (Welfinger-Smith et al., 2011), here we show that some contaminants do have similar or higher levels than in adipose tissues, such as metals, ΣDDT, dieldrin, and ΣPFAS (Figure 2F). Due to the different physico-chemical properties of the various POPs and metals, and their varying interaction with proteins, lipids, and other endogenous molecules, it is important to understand the properties of the contaminants of interest in order to sample the proper tissues. For example, it has been found that in polar bears Hg accumulates with age in liver and kidney, but not in the muscle (Rush et al., 2008; Dietz et al., 2000; Dietz et al., 1995; Braune et al., 1991). Some studies have attributed this difference to an efficient catabolic function of the polar bear liver (Dietz et al. 1995). Also, muscle is a very heterogeneous tissue as it has a lot of fat and the lipid percentage depends on the muscle type, which could affect concentrations levels (Verreault et al., 2006). However, chemical composition has an impact on its accumulation in polar bear tissues, as the presence of contaminants such as PFOS in muscle may be related to age, which had significant age effects (Greaves et al., 2012). ΣOH-PBDEs are found in the blood and adipose tissue at very low levels (Gebbink et al., 2008a). In addition, no OH-PBDEs were detectable in polar bear liver, suggesting that the low levels in blood and adipose tissue are due to protein-binding (Gebbink et al., 2008a).

4.8 Blood

In polar bear blood (Figure 2G), THg has the highest concentration of all contaminants (pre-2000, 56 ng/g (quantile 25 %: 51 ng/g; quantile 75 %: 73 ng/g) and post-2000, 70 ng/g (quantile 25 %: 51 ng/g; quantile 75 %: 156 ng/g) even if it is relatively low compared to the levels found in kidneys (Figure 2A) or liver (Figure 2C). Blood samples represent real-time dietary and contaminant uptake (from previous weeks to 2 or 3 months) (Cardona- Marek et al., 2009), compared to other tissues that accumulate in the longer-term. OH-PCB metabolites are less lipophilic than their PCB parent compounds and are mainly present and detectable in the blood due to their non-covalent binding to hormone transport proteins (Erdmann et al., 2013; Letcher et al., 2000). OH-PCB concentrations in polar bears blood is largely a reflection of the time of their prey consumption and accompanying PCB exposure and/or mobilization of lipid (and PCBs) from their tissue lipid stores (Olsen et al., 2003). It has been determined that plasma lipid content affects PCB concentrations and the lipid content changes after feeding (Olsen et al., 2003). When polar bears eat seal blubber, the plasma lipid content increases and so does the concentration of PCBs associated with the seal fat (Olsen et al., 2003).

4.9 Other tissues

The “Others” category (Figure 1) groups the other tissues, such as brain, brainstem, cerebellum, lymph nods, spleen, and teeth (Figure 2H). Metals are the most abundant contaminants found for this group of tissues and body compartments, but other contaminants yielded similar median concentrations. For example, in brain tissue of East Greenland polar bears, OH-PCBs were measurable, indicating that OH-PCBs could penetrate the blood-brain barrier (Gebbink et al., 2008a). For many contaminant classes, the lowest concentrations have been reported in the brain, with a total amount of the POPs accounting for a mere 0.002 % of the tissue burdens, for example in East Greenland polar bears (Gebbink et al., 2008a). Jaspers et al. (2010) noted that concentrations found in brain tissue of bears from central East Greenland were similar to hair for the OHCs. Reports show that polar bears do not accumulate high level of THg in the brain stem (Basu et al., 2009). However, the bioaccumulative form of Hg, MeHg, was reported to account for 100 % of the THg in polar bear brain (Krey et al., 2015; Krey et al., 2012). The Hg concentration depends of the brain region, as it was found to be different in frontal lobe (highest THg level), cerebellum and brain stem (lowest THg level) (Krey et al., 2012). As shown in Figure 2H, studies report a median THg concentration of 230 ng/g in brain tissue. This THg concentration does not exceed the level at which neuropathological effects or neurochemical disruptions can be detected, but it is of concern since it is more elevated than 100 ng/g and could affect polar bears (Krey et al., 2015). There is little experimental evidence that reported polar bears from central East Greenland are experiencing Hg-induced neurological damage (Basu et al., 2009). Similar conclusions were made for PFAS (mainly PFOS), as concentrations were the highest in the lowest part of the brain, like brain stem (Eggers Pedersen et al., 2015). For the teeth, low dental concentrations of THg were reported for East Greenland polar bears (Aubail et al., 2012). Teeth were thought to reflect the Hg exposure through diet at the time of formation since dental Hg matches blood Hg at the time of formation and mineralisation of the tooth (Aubail et al., 2012), but might not indicate the overall load of Hg in the animals.

5. PACs have not been measured in polar bears

The Arctic is subject to PAC (polycyclic aromatic compound) contamination as PACs can be transported to the Arctic as a part of large-scale atmospheric and oceanic circulation patterns (AMAP, 1998). PACs differ from many other POPs and chemicals of emerging concern in that they are unintentional by-products of fossil fuel use and emissions are ongoing (AMAP, 2017). Since PACs are less prone to biomagnification, there is less concern about these compounds (AMAP, 1998), and thus, there are few data available for their presence in marine mammals (AMAP, 2017). PACs biodilute, which means that their concentrations decrease with higher trophic level due to the increase of biotransformation rate (De Laender et al., 2011). Recently, modelling of oil spill's impact on polar bears was calculated and it was found that around 27-38 % of polar bears from the Chukchi sea region would be affected (Wilson et al., 2018). Indeed, it was noted that oil spills could probably reach important habitats for polar bears, such as areas of denning or summer terrestrial habitats (Wilson et al., 2018). Wilson et al. (2018) estimated that it would take approximately 2-3 weeks for an oil spill to reach polar bear environment, which likely provides time for clean-up efforts. In addition, a study has detected up to 31 PACs in the surface sediments of the Chukchi Sea (Harvey and Taylor, 2017). The presence of PACs was also measured in muscle tissues of both the of the Neptune whelk (*Neptunea heros)* and the mussel *Musculus discors* (Harvey and Taylor, 2017). Stimmelmayr et al. (2018) found 6.4 ng/g of PACs in muscles and 4.4 ng/g of PACs in skin of unoiled Alaskan ringed seals sampled in 2012-2014, and higher levels where obtained in tissues of oiled seals. Thus, there is a critical need to monitor PACs in polar bears in order to develop baseline that could be used in the event of an Arctic oil spill.

6. Overall recommendations and conclusions

As polar bears are known to be exposed to a variety of contaminants, this review summarized polar bears burden in various tissues. It also allowed a better understanding of contaminant distribution and patterns of accumulation in polar bears. Although CHLs were banned in most countries in the '80s-'90s and were listed in the Annex I of the

Stockholm Convention as one of the POPs in 2001, this group of contaminants are still of interest as it continues to be among the highest concentration of POPs measured in polar bears. Therefore, CHL monitoring should continue, especially that CHLs are known to induce renal lesions, such as thickening kidneys' globular capillary wall in adult male polar bears (Sonne et al., 2012; Sonne et al., 2006) and to interfere with polar bear's reproduction (Sonne et al., 2009). Similarly, for metals, although they have been heavily studied prior to 2000, their presence in the environment remains high and will persist given the importance of metals in daily consumer products and human activities (e.g., rare-earth elements). Thus, additional efforts should be put forward to evaluate the concentrations of metals in polar bear tissues, including the less studied metals, such as rare-earth elements (REEs), which are increasing been used in nowadays technology (MacMillan et. 2017). Other emerging contaminants, such as PFOS and its replacing compounds should be given priority in the Arctic monitoring programs. Future research and monitoring assessments should also include PACs to generate robust baselines for polar bears given the increased risk for oil spills in the Arctic with the development of the Northern regions of Canada.

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Given that some polar bear populations are starting to decline, there is an increasing need to develop novel non-lethal and non-invasive approaches to evaluate polar bear's contaminant burden.

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8. Declaration of interests

The authors declare no conflict of interest.

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List of figures

Figure 1. The median concentrations (ng/g) are the sum of all available contaminant data for a given polar bear tissue or body compartment and based on 81 different studies conducted between 1990 and 2018 (Supplementary file Tables S1 to S5). “Others” include tissue from brain, brainstem, cerebellum, lymph nods, spleen, and teeth. Samples were categorized from pre-2000 and post-2000. Asterisks (*) represent significant differences over periods for a same tissue. Concentrations of contaminants include: aldrin; chlordane; chlorobenzene; dichlorodiphenyltrichloroethane; dieldrin; endosulfan; hexabromocyclododecane; hexachlorobenzene; hexachlorocyclohexane; mirex; total mercury; octachlorostyrene; other metals; brominated biphenyl; polybrominated diphenyl ether; polychlorinated biphenyls ; polychlorinated naphthalenes; pentachlorophenol; per-/poly-fluoroalkyl substances;~~,~~ short-chain chlorinated paraffin and toxaphene.

Figure 2. Concentrations (ng/g) based on individual or sum classes of contaminants in the kidneys, hair, liver, milk, adipose tissue, muscle, other tissues, and blood of polar bears based on data from 81 different studies from 1990 to 2015. Abbreviations: ΣCHL, chlordane; ΣClB~~b~~z, chlorobenzene; ΣDDT, dichlorodiphenyltrichloroethane ; ΣHBCD, hexabromocyclododecane; HCB, hexachlorobenzene; ΣHCH, hexachlorocyclohexane Hg, mercury; OCS, octachlorostyrene; ΣPBB, brominated biphenyl; ΣPBDE, polybrominated diphenyl ether ; ΣPCBs, polychlorinated biphenyls ; ΣPCNs, polychlorinated naphthalenes;

PCP, pentachlorophenol; ΣPFAS, perfluoroalkyl substances; ΣSCCP, short-chain chlorinated paraffin; THg, total mercury. ND (No Data) indicates that no data was available for the specific group contaminant of a tissue. Concentrations lower than the limit of detection (LOD) as analyzed by (1) Gebbink *et al*., 2008a; (2) Bechshøft *et al*., 2012; (3) Letcher *et al*., 2018; (4) Gabrielsen *et al*., 2015; (5) Bentzen *et al*., 2008b; and (6) Tartu *et al*., 2017.

Figures 1 and 2

Figure 1

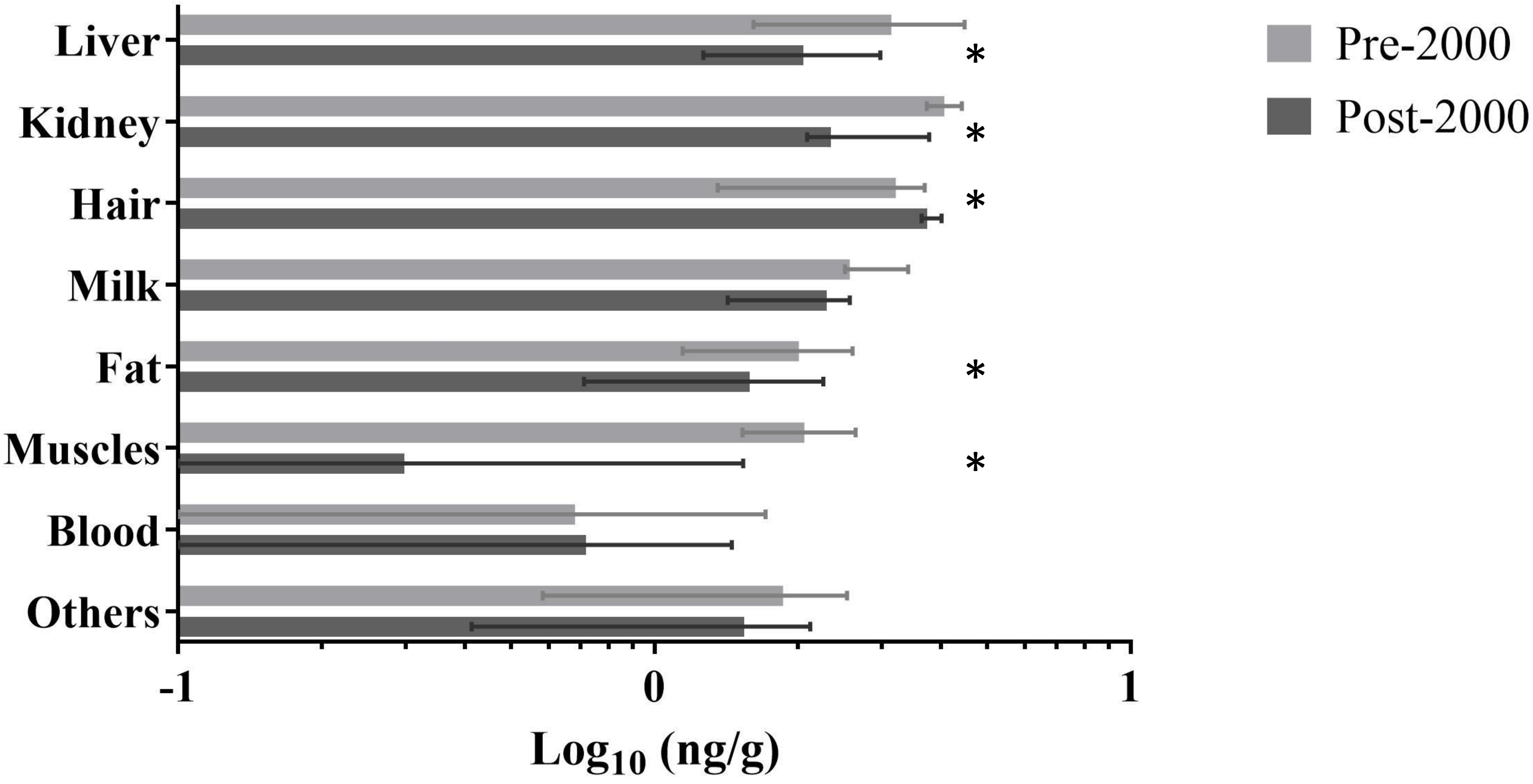


Figure 2

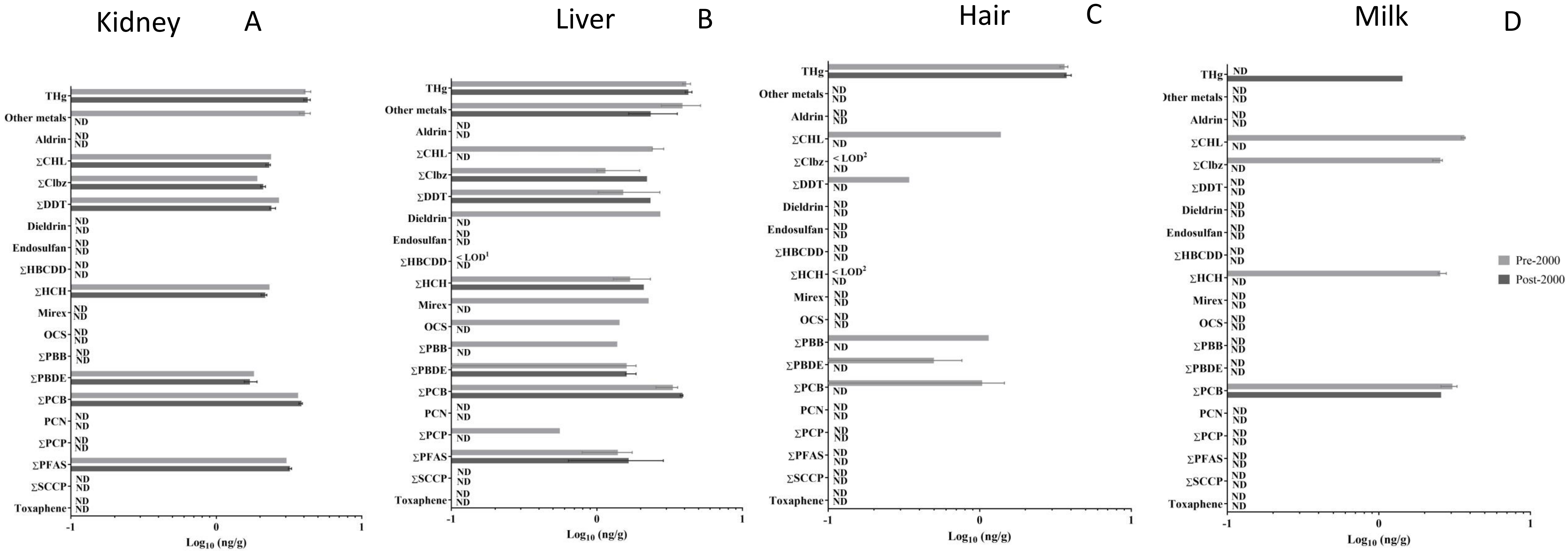
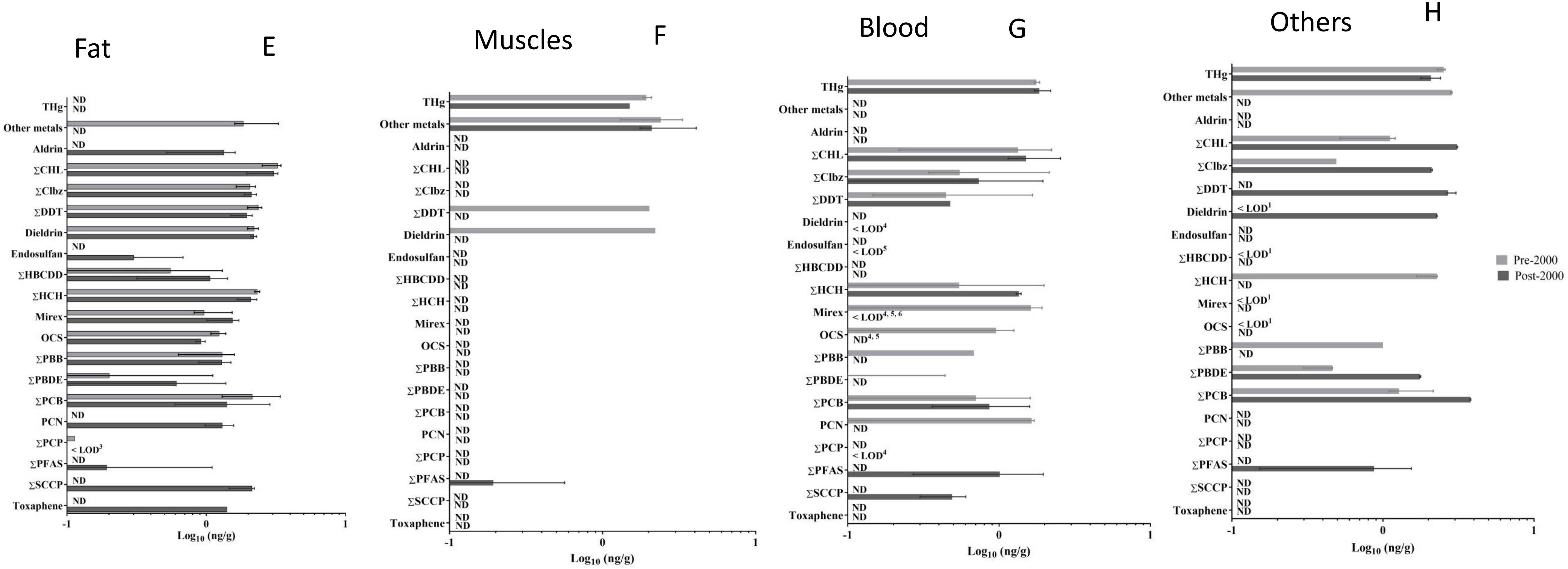

Kidney
A
Liver
B
Hair
C
Milk
D
THg
Other metals
Aldrin
ΣCHL
ΣClbz
ΣDDT
Dieldrin
Endosulfan
ΣHBCDD
ΣHCH
Mirex
OCS
ΣPBB
ΣPBDE
ΣPCB
PCN
ΣPCP
ΣPFAS
ΣSCCP
Toxaphene
ND
< LOD[1]
< LOD[2]
-1
0
1
Log10 (ng/g)
Pre-2000
Post-2000

Figure 2


Fat
E
Muscles
F
Blood
G
Others
H
THg
Other metals
Aldrin
ΣCHL
ΣClbz
ΣDDT
Dieldrin
Endosulfan
ΣHBCDD
ΣHCH
Mirex
OCS
ΣPBB
ΣPBDE
ΣPCB
PCN
ΣPCP
ΣPFAS
ΣSCCP
Toxaphene
ND
< LOD
Log10 (ng/g)
-1
0
1
Pre-2000
Post-2000