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# Quantification de la contamination dans le foie, les muscles et la graisse d'ours polaires de l'Arctique canadien

Par

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# RÉSUMÉ

Depuis l'ère industrielle, les produits chimiques sont omniprésents dans les écosystèmes du monde entier. Cependant, malgré l'arrêt du rejet de plusieurs polluants organiques persistants (POP) hautement toxiques dans l'environnement, certains polluants sont toujours mesurés dans l'Arctique canadien. Ces contaminants sont très préoccupants en raison de leur persistance, de leur toxicité et de leur niveaux de bioaccumulation dans les chaînes alimentaires. Les animaux vivant dans l'Arctique, en particulier les ours polaires qui occupent une position trophique supérieure, sont exposés à ces contaminants principalement par leur alimentation. Notre étude a examiné les niveaux de biphényles polychlorés (BPC), de composés aromatiques polycycliques (CAP), de chlordanes (CHL) et de métaux (y compris le mercure total et le méthylmercure) chez 49 ours polaires de l'Arctique canadien. La charge de contaminants a été mesurée dans le foie, les muscles et la graisse chez des ours polaires de sexe, d'âge et de localisation différents. L'analyse en composantes principales n'était pas assez puissante pour distinguer les différences entre les profils d'âge et de sexe différents. Cependant, les concentrations mesurées et leur répartition dans les tissus confirment les résultats observés dans les études antérieures. Cette étude met en évidence l'importance d'un suivi continu de la santé des ours polaires (par exemple, de nouveaux CAP ont été mesuré par cette étude) et d'une évaluation des impacts de ceux-ci pour les prochaines générations.

**Mots-clés :** Ours polaires, espèces sentinelles, bioamplification, contaminants, métaux, pesticides, retardateurs de flamme, composés aromatiques polycycliques, biphényles polychlorés, Arctique

# ABSTRACT

Since the industrial era, chemicals are ubiquitous in worldwide ecosystems. However, despite the discontinued release of highly toxic Persistent Organic Pollutants (POPs) in the environment, the levels of some persistent pollutants are still being measured in the Canadian Arctic. These contaminants are of great concern due to their persistence, toxicity, and levels of bioaccumulation in food chains. Animals living in the Arctic, in particular the polar bears, which occupy a top trophic position, are exposed to these contaminants mainly through their diet. Our study investigated the levels of (PCBs), polycyclic aromatic compounds (PACs), chlordanes (CHLs), and metals (including total and methyl mercury) in 49 polar bears from the Canadian Arctic. Contaminant burden was measured in liver, muscle and fat in bears of different sex, age and locations.A principal component analysis was not powerful enough to distinguish differences between profiles of different age and sex. However, the concentrations measured and their distribution in the tissues confirm findings observed in past studies. This study highlights the importance of a continual monitoring of polar bears health (e.g., newly PAC were measured within this study) and evaluation of the impacts of those for the next generations.

**Keywords :** polar bears; sentinel species; bioamplification; contaminants; metals; pesticides; flame retardants; polycyclic aromatic compounds; polychlorinated biphenyls; Arctic.

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Al: Aluminium

ASU : Analytical service unit

BB : Baie de Baffin

Be: Béryllium

BPC : Biphényles polychlorés

Ca : Calcium

CAP : Composés aromatiques polycycliques

Cd : Cadmium

CHLs : Chlordanes

Cr : Chrome

CYP1: Cytochrome P450

FB : Bassin de Foxe

Fe: Fer

Ga: Gallium

GB: Golfe de Boothia

GC/MS: Chromatographie gazeuse couplée à la spectrométrie de masse

GC/MS/MS: Chromatographie gazeuse couplée à la spectrométrie de masse en tandem

Hg: Mercure

ICP-AES: Spectroscopie à émission atomique et à plasma à couplage inductif

ICP-OES: Spectroscopie à émission atomique optique et à plasma à couplage inductif

ICP-MS: Spectrométrie de masse à plasma à couplage inductif

INRS: Institut national de la recherche scientifique

K: Potassium

Kow: Coefficient de partition Octanol/Eau

LOD : Limite de détection

MeHg: Méthylmercure

Mg : Magnésium

Na : Sodium

NBS: Nord de la mer de Beaufort

Ni: Nickel

P: Phosphore

PACs : Composés aromatiques polycycliques

Pb: Plomb

PBDE: Polybromodiphényléthers

PCA: Analyse en composante principale

PCBs : Biphényls polychlorés

POPs: Polluants organiques persistants

S: Soufre

Sb: Antimoine

Se: Sélénium

SHB: Sud de la Baie d'Hudson

Si: Silicium

Sr: Strontium

QA/QC: Assurance qualité/ contrôle qualité

THg : Mercure total

V: Vanadium

WHB : Ouest de la Baie d'Hudson

Zn: Zinc

# 1 REVUE DE LITTERATURE

## 1.1 Émission des contaminants dans l'Arctique

Depuis l'ère industrielle, il est connu dans la littérature que de nombreuses substances chimiques qui n'étaient pas présentes naturellement dans l'environnement ont fait leur apparition créant ainsi de nombreux bouleversements dans les écosystèmes. Parmi ces bouleversements, on dénombre les pluies acides, la création d'un trou dans la couche d'ozone et l'augmentation des gaz à effet de serre dans l'atmosphère. Les conséquences de ces bouleversements furent mesurées et présentées par la communauté scientifique et engendrèrent des politiques internationales (Stockholm 2001, Minamata 2013, etc.) afin de remédier à ces problématiques. De cette manière, la production de certains contaminants fut bannie à la suite de conventions internationales telles que les chlorofluorocarbures (CFC) ou l'aldrine. Néanmoins, malgré l'arrêt de l'introduction de ces polluants dans l'environnement, des niveaux de certains de ces polluants les plus persistants sont toujours mesurés dans l'Arctique canadien (Smithwick et al 2005). Beaucoup sont acheminés à cet endroit en raison des courants atmosphérique et marin dominants provenant de l'Asie, de l'Europe et de l'Amérique du Nord (Figure 1.1).

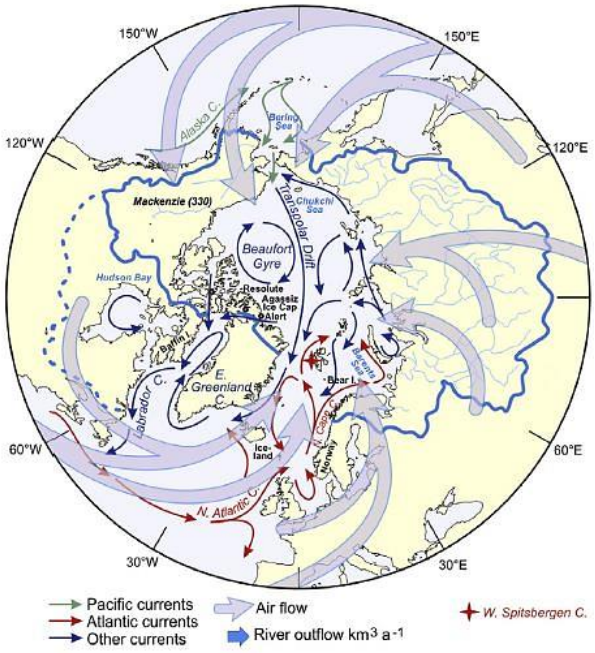


**Figure 1-1 Courants majeurs acheminant les contaminants dans l'Arctique (Brown et al., 2017).**

Les preuves que les écosystèmes arctiques sont exposés à des contaminants se sont accumulées lors des dernières décennies. Ces contaminants sont très préoccupants en raison de leur persistance, de leur toxicité et de leurs niveaux de bioaccumulation. Les animaux vivants en Arctique, en particulier l'ours polaire qui est le prédateur au sommet de la chaîne trophique, sont donc exposés à ces contaminants principalement par leur diète (Tartu et al. 2017). Au Canada, on estime la population d'ours polaires à plus de 15 000, soit environ les 2/3 de la population mondiale dont le Nunavut et les Territoires du Nord-Ouest sont les principaux acteurs de gestion de ces animaux (Aars et al., 2017). Tous les bouleversements que causent les activités humaines sur l'écosystème arctique ont des impacts directs sur les populations d'ours polaires. En effet, ces dernières constituent un bon indicateur de l'état actuel de la contamination de l'Arctique puisqu'elles se déplacent sur une superficie allant jusqu'à 350 000 $km^2$ (Auger-Méthé et al. 2016) et que leur alimentation est variée.

Plusieurs études ont quantifié la charge corporelle en contaminants chez les ours polaires en utilisant des méthodes invasives, notamment en étudiant la matière adipeuse, le foie, les dents ou les reins. On retrouve parmi ces polluants des pesticides, des composés organiques persistants, des retardateurs de flammes ainsi que des métaux. La révision de la littérature semble illustrée que c'est le Nord canadien, l'Alaska, le Svalbard et le Groenland qui furent les plus étudiés pour évaluer la contamination dans les populations d'ours polaires. Par exemple, de cette même revue de littérature, des concentrations moyennes en métaux mesurés dans le foie d'ours polaires de l'Alaska sont répertoriés entre 17 700 – 494 910 ng/g. Le tableau suivant (1-1) indique les concentrations en métaux (ng/g) compilées de la littérature pour le foie, muscle et gras d'ours polaires. La somme des métaux essentiels et non-essentiels est comptabilisée et leurs concentrations est reportées telles quelles (poids sec, humide ou lipidique).

**Tableau 1-1 Niveaux des métaux mesurés chez les ours polaires (ng/g)**

| Tissue | Endroits | Gamme concentrations mesurées (ng/g) | Références |
|---|---|---|---|
| Foie | Alaska | 17 700 – 494 910 | AMAP (2005); Rush *et al.* (2008) ; Woshner *et al.* (2001) |
| | Canada | 119 470- 25 260 760 | Braune *et al.* (1991) Kannan *et al.* (2007) Rush *et al.* (2008) |
| | Groenland | 2630 – 72 900 | AMAP (2005) ; Dietz *et al.* (1995) ; Dietz *et al.* (2000); Rush *et al.* (2008) |
| | Mer des Tchouktches | 353 083 | Kannan *et al.* (2007) |
| Muscle | Alaska | 2800 – 563 200 | AMAP (2005); Rush *et al.* (2008) ; Welfinger-Smith et al. (2011) ; Woshner *et al.* (2001) |
| | Groenland | 95 – 76 215 | AMAP 2005 ; Dietz et al. (1995) ; Dietz et al. (2000) |
| Gras | Alaska | 30 340 | Woshner *et al.* (2001) |

Parmi ces métaux, le mercure reste toujours d'intérêt puisqu'il est, entre autres, neurotoxique sous sa forme méthylée lorsqu'il est absorbé chez les vertébrés (López-Berenguer et al., 2020). De plus, il continue de se déplacer vers l'écosystème arctique en raison d'émissions anthropiques sur toute la planète (Braune et al., 2015). Dans l'Arctique, les niveaux atmosphériques sont généralement à la baisse depuis les deux dernières décennies, alors que les niveaux de mercure dans le biote arctique ont montré des tendances à la fois à la hausse et à la baisse au cours des deux dernières décennies (Landrigan et al., 2020). Les concentrations mesurées de mercure total (THg) chez les ours polaires observées dans la littérature sont affichées dans le tableau 1-2. Les concentrations considérées sont en poids humide, sec ou lipidique.

**Tableau 1-2 : Niveaux de THg mesurés chez les ours polaires (ng/g)**

| Tissue | Endroits | Gamme concentrations mesurées (ng/g) | Références |
|---|---|---|---|
| Foie | Alaska | 10 360 – 52 500 | AMAP (2005); Rush *et al.* (2008) ; Woshner *et al.* (2001) |
| | Canada | 7340 – 200 000 | Braune *et al.* (1991) Kannan *et al.* (2007) ; Rush *et al.* (2008) |
| | Groenland | 1500 – 130 000 | Dietz *et al.* (1990); Dietz *et al.* (1995) ; Dietz *et al.* (2000); Rush *et al.* (2008) ; Sonne *et al.* (2007) ; Sonne *et al.* (2012) |
| | Mer des Tchouktches | 10 100 | Kannan *et al.* (2007) |
| Muscle | Alaska | 30 – 400 | AMAP (2005); Welfinger-Smith (2011) ; Woshner *et al.* (2001) |
| | Groenland | 34 – 570 | AMAP 2005 ; Dietz et al. (1990) ; Dietz et al. (1995) ; Dietz et al. (2000) |
| Gras | Canada | 3000-5300 | Yurkowski et al. (2020) |

Bien que les biphényles polychlorés (BPC) et les chlordanes (CHL), qui étaient utilisés à des fins industrielles et agricoles, soient maintenant réglementés dans certains pays depuis les années 1970 et dans le monde par la Convention de Stockholm signée en 2001, ils sont toujours présents à fortes concentrations dans l'Arctique (Carlsson et al., 2018). Les niveaux de BPC, spécifiquement, ne diminuent pas comme il serait attendu en raison de l'arrêt de leur émission. Les concentrations rapportées (poids sec, humide ou lipidique) dans la littérature pour les CHL et les BPC chez les ours polaires sont présentées dans les tableaux 1-3 et 1-4. Les concentrations affichées sont la somme des composés.

**Tableau 1-2 Niveaux des chlordanes mesurés chez les ours polaires (ng/g)**

| Tissue | Endroits | Gamme concentrations mesurées (ng/g) | Références |
|---|---|---|---|
| Foie | Canada | 250 – 1500 | Kannan *et al.* (2005) ; Wiberg et al. (2000) |
| | Groenland | 121 – 272 | Sonne *et al.* (2012) |
| | Mer des Tchouktches | 458 | Kannan *et al.* (2007) |
| Muscle | Canada | 1287 | Verreault et al. (2006) |
| Gras | Alaska | 313 - 1095 | Bentzen et al. (2008) ; Dietz et al. (2015) ; McKinney et al. (2011) ; Verreault et al. (2005b) |
| | Canada | 979 - 7937 | Dietz et al. (2015) ; Letcher et al. (2018) ; Mckinney et al. (2010) ; Norstrom et al. (1998); Polischuk et al. (2002); Verrault et al. (2005b) |
| | Groenland | 201 - 5044 | Bechshøft et al. (2012b) ; Dietz et al. (2004) ; Dietz et al. (2015) ; Gebbink et al. (2008b) ; Jaspers et al. (2010) ; McKinney et al. (2011) ; Sandala et al. (2004) Sonne et al. (2005) ; Sonne et al. (2013) ; Verreault et al. (2005) ; Verreault et al. (2008) |
| | Mer des Tchouktches | 1016 | Norstrom et al. (1998) |
| | Norvège | 871 - 5616 | Bernhoft et al. (1997) ; Dietz et al. (2015) ; Gabrielsen et al. (2004) ; Kleivane et al. (2000) ; McKinney et al. (2011) ; Norstrom et al. (1998) ; Tartu et al. (2017) ; Verreault et al. (2005b) |

**Tableau 1-3 Niveaux des BPC mesurés chez les ours polaires (ng/g)**

| Tissue | Endroits | Gamme concentrations mesurées (ng/g) | Références |
|---|---|---|---|
| Foie | Alaska | 2110 | Corsolini et al. (2002) |
| | Canada | 857 – 7700 | Kannan *et al.* (2005) ; Wiberg et al. (2000) |
| | Groenland | 2520 – 8185 | Gebbink et al. (2008a/b); Jaspers et al. (2010); Sonne et al. (2012) |
| | Mer des Tchouktches | 466 | Kannan *et al.* (2005) |
| Muscle | Canada | 5628 | Verreault et al. (2006) |
| Gras | Alaska | 445 | Bentzen et al. (2008ab) ; Dietz et al. (2015) ; McKinney et al. (2011) ; Verreault et al. (2005b) ; Welfinger-Smith et al. (2011) |
| | Canada | 1138 – 22 391 | Dietz et al. (2015) ; Letcher et al. (2018) ; Mckinney et al. (2010) ; Mckinney et al. (2011); Norstrom et al. (1998); Polischuk et al. (2002); Verrault et al. (2005b); Wiberg et al. (2000) |
| | Groenland | 5 - 11390 | Bechshøft et al. (2012b) ; Dietz et al. (2004) ; Dietz et al. (2015) ; Erdmann et al. (2013) ; Gebbink et al. (2008b) ; Jaspers et al. (2010) ; McKinney et al. (2011) ; Norstrom et al. (1998) ; Sandala et al. (2004) Sonne et al. (2005) ; Sonne et al. (2006) Sonne et al. (2013) ; Verreault et al. (2005) ; Verreault et al. (2008) ; Vorkamp et al. (2015) |
| | Mer des Tchouktches | 2058 - 5535 | Norstrom et al. (1998) |
| | Norvège | 2259 – 29 409 | Bernhoft et al. (1997) ; Dietz et al. (2015) ; Gabrielsen et al. (2004) ; Henriksen et al. (2001) ; Kleivane et al. (2000) ; McKinney et al. (2011) ; Norstrom et al. (1998) ; Tartu et al. (2017) ; Verreault et al. (2005b) |

Il est difficile d'établir des tendances sur la prévalence des composés aromatiques polycycliques (CAP) dans les régions de l'Arctique en raison de leurs nombreuses sources d'émission, à la fois naturelle (feux de forêt, sédiments) et anthropique (utilisation des combustibles fossiles) (Hodson et al., 2020). Néanmoins, même si des changements au niveau des émissions dues à l'exploitation pétrolière devaient causer une diminution des niveaux des CAP présents dans l'atmosphère de l'Arctique, la pertinence de ce changement dans l'Arctique est incertaine. La bioaccumulation des CAP fut mesurée dans la faune autant chez les poissons, grenouilles, amphibiens et oiseaux (voir revue par Wallace et al. 2020) Par contre, peu de données sont disponibles pour les concentrations tissulaires chez les mammifères en raison des taux élevés de métabolisme et d'excrétion des CAP. Ainsi, aucune concentration de CAP chez les tissues d'ours polaires n'est rapportée dans d'autres études (Wallace et al., 2020).

La lecture de la littérature permet de constater que les études sont majoritairement axées sur la charge des contaminants dans le foie et le gras, et très peu sur les muscles. De plus, les ours polaires du Canada semblent être les plus affectés par les contaminants, présentant fréquemment la gamme de concentration la plus élevée entre les régions du monde. La distribution des contaminants change selon la nature du contaminant : les métaux dont le mercure semble s'accumuler davantage dans le foie alors que les chlordanes et les BPC d'autant plus dans les gras. Aucune étude ne rapporte factuellement la distribution possible des PAC entre les tissues chez les ours polaires. Néanmoins, il fut rapporté que chez les vertébrés terrestres c'est dans le foie que ces composés peuvent se bioaccumuler.

## 1.2 Impacts de l'exposition des contaminants sur les ours polaires en Arctique

Les impacts potentiels sur la santé des ours polaires en raison de l'exposition de ces contaminants sont étudiés dans la littérature. Actuellement, les évaluations des risques pour les ours polaires sont basées sur des extrapolations à partir de recherches toxicologiques sur d'autres espèces de mammifères (Routti et al., 2019). Les paramètres étudiés, tels que la perturbation du métabolisme endocrinien, l'immunotoxicité, la neurotoxicité et les altérations pathologiques, varient selon les sous-populations d'ours polaires (Bechshoft et al., 2017). Dans les dernières années, un nombre croissant de recherches in vitro, d'études sur des espèces de substitution et d'études d'évaluation des risques ont été publiées.

En effet, les contaminants peuvent affecter la santé de l'ours polaire de diverses manières en fonction de leurs concentrations. Il a été démontré que les concentrations de polluants dans les tissus de l'ours polaire sont en corrélation avec les concentrations de certaines hormones et vitamines, affectant ainsi la morphologie et la fonction des tissus (par exemple, le foie, les reins et la glande thyroïde), la densité osseuse et la fonction immunitaire (Daugaard-Petersen et al. 2018; Ciesielski et al. 2018; Sonne 2010). Par exemple, des techniques in vitro ont été utilisées par Desforges et al. (2017) pour évaluer le lien concentration-réponse entre les polluants provenants de la graisse et la fonction immunitaire des ours polaires. Selon cette étude, une quantité présente de PCB dépassant 0,1 g/mL causerait la suppression de prolifération des lymphocytes.

Selon Haave et al. 2003, une augmentation des concentrations plasmatiques de progestérone est corrélée avec des concentrations croissantes de PCB, dans une gamme de concentration pour ces composés allant 1392 à 18 210 ng/g en poids lipidique dans le sang (Haave et al., 2003).

Les contaminants peuvent également affecter le comportement des ours polaires, entre autres au niveau des comportements de survie. En effet, le MeHg affecterait le métabolisme des neurotransmetteurs tels que la sérotonine et la dopamine. Krey et al. (2014) a étudié l'activité de la monoamine impliquée dans le métabolisme de ces neurotransmetteurs dans le cerveau de 24 ours polaires provenant du Nunavik et détermina que son activité était inversement associée aux concentrations de THg dans ces échantillons de cerveau d'ours polaires. Les contaminants organohalogénés tels que les chlordanes ont été liés à des changements morphologiques dans le foie et les reins des ours polaires de l'est du Groenland, y compris l'accumulation de cellules immunitaires, les modifications des voies biliaires et l'accumulation de lipides, qui indiquent toutes des infections chroniques et une exposition toxique (Letcher et al., 2010; Sonne, 2010).

Des études à grande échelle ont également mesuré l'ensemble des concentrations de polluants organiques persistants (POP) dans 11 sous-populations d'ours polaires (de l'Alaska au Svalbard). Selon Dietz et al. (2015), ces populations ont dépassé les seuils pour influencer la santé des ours sur le plan du système reproducteur, immunitaire ainsi qu'au niveau de la cancérogénicité. La population des ours polaires de l'est du Groenland, entre autres, est à risque d'effets de contaminants tels les BPC depuis plusieurs décennies, les quotients de risque ayant culminé au début des années 1980, puis à nouveau en 2013 (Dietz et al., 2018). Dans une autre étude, l'apport quotidien de POP pour les ours polaires a été déterminé à l'aide de données de la littérature, et les résultats ont été comparés à la consommation quotidienne admissible de POP pour les humains (Villa et al., 2017). Les auteurs ont conclu que les mélanges de contaminants présentent un risque important pour les ours polaires adultes et davantage pour les juvéniles.

## 1.3 Impacts sur les communautés inuites en Arctique

Il ne faut pas négliger également la place centrale qu'occupe l'ours polaire dans la culture, la spiritualité et la pratique de la chasse pour les communautés inuites. Certaines communautés autochtones sont d'ailleurs davantage à risque d'être exposées aux mêmes contaminants puisque l'ours polaire fait partie intégrante de leur alimentation traditionnelle (AMAP, 2015). Cette dernière est leur principale source de contamination et bien que la plupart des aliments sauvages de l'Arctique soient connus pour être riches en nutriments et faibles en contaminants, ces communautés restent exposées à une variété de POP et de métaux nocifs lors de la

consommation de mammifères marins comme l'ours polaire. De nombreux polluants mesurés dans les communautés arctiques diminuent, mais les niveaux de POP dans certaines populations arctiques restent plus élevés que chez les personnes dans d'autres parties du monde. Les substances per- et polyfluoroalkylées (PFAS), ainsi que le méthylmercure, restent une source de préoccupation (Gibson et al.2016). Les concentrations en Hg dans le sang sont notamment les plus hautes chez les femmes enceintes provenant de régions arctiques (Figure 1-2).

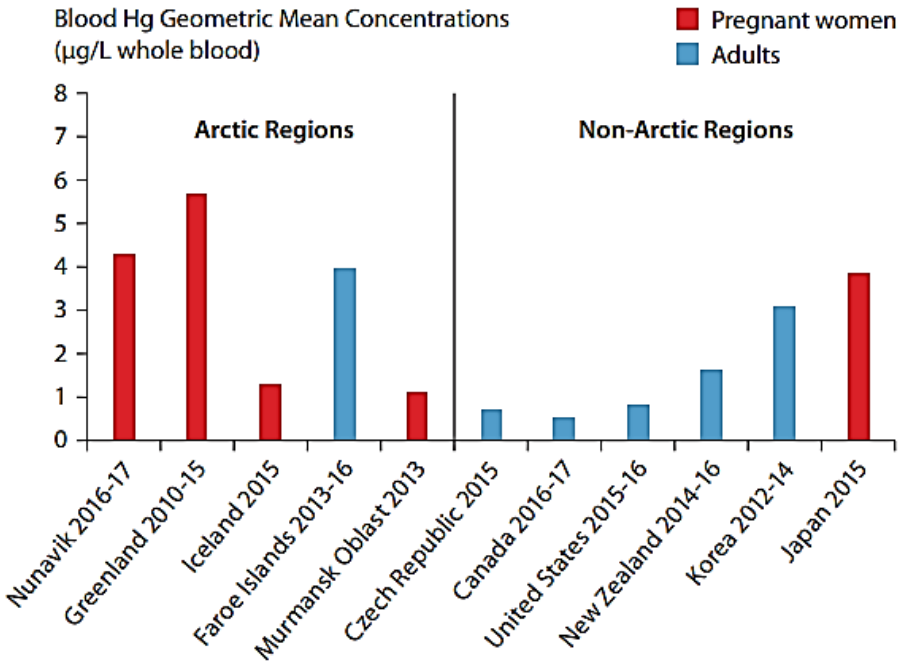


**Figure 1-2 Niveaux de mercure chez les femmes enceintes et les hommes et femmes adultes selon leur lieu de résidence (Gibson et al. 2016)**

Le Groenland et le Nunavik avaient les niveaux moyens de mercure les plus élevés chez les femmes enceintes dans une étude comparant les niveaux de mercure dans sept pays de l'Arctique (Adlard et al., 2021). Selon une étude mondiale, les adultes et les enfants du Nunavik, du Groenland et des îles Féroé présentaient des niveaux de mercure plus élevés que ceux des pays non arctiques. L'exposition alimentaire à certains POP, PFAS et métaux comme le mercure peut occasionner des effets indésirables au cerveau et au système immunitaire, augmenter le

risque d'obésité juvénile, augmenter le risque de diabète de type 2 plus tard dans la vie et nuire à la croissance et au développement prénatals (Donaldson et al. 2010).

Cependant, au cours des dernières décennies, les populations de l'Arctique se sont tournées vers les repas occidentaux achetés en magasin, en raison d'une accessibilité accrue (par exemple, grâce au développement d'infrastructures de transport) et de leur prise de conscience envers les problèmes de santé concernant les composés toxiques présents dans les aliments traditionnels (Mead et al. 2010). Ceci engendra à la fois des conséquences positives sur la teneur en contaminants ingérés tels des niveaux inférieurs de polluants dans le sang des femmes enceintes. Néanmoins, ce virage a causé une augmentation soulevée de l'obésité, des maladies métaboliques et des problèmes dentaires chez ces populations. Des déficiences en vitamine D et de l'iode sont également observés (Mead et al., 2010)

## 1.4 Objectif de recherche et hypothèses

L'objectif de ce projet est de quantifier la distribution des contaminants dans les muscles, la graisse et le foie d'ours polaires d'âge, de sexe et d'endroits différents vivants dans le nord du Canada. De ces mesures, il sera établi si des tendances sont observables et si les faits reportés de cette étude ont déjà été rapportés dans la littérature.

Au total c'est plus de 110 contaminants qui furent analysés. Plus précisément, ce fut 30 métaux essentiels et non essentiels incluant le mercure total et le méthylmercure, six chlordanes incluant l'oxychlordane, l'heptachlor ainsi que les couples d'isomères cis/trans-chlordane et cis/trans-nonachlor., 11 composés parents et leur congénères alkylés pour les CAP ainsi que 20 composés parmi les 209 congénères des BPC furent également évalués. Ces composés seront dans 49 ours polaires chassés entre 2016 et 2019 par des chasseurs locaux pan-canadiens. La majorité des ours étaient des mâles adultes, soit 33 ours. Ces ours furent chassés parmi six régions soit 20 ours du Sud de la baie d'Hudson, 12 du bassin de Foxe, 12 du Golfe de Boothia, trois du nord de la mer de Beaufort, un de l'ouest de la baie d'Hudson et un de la baie de Baffin. L'analyse en composantes principales était l'outil statistique utilisé pour analyser les données présentées dans l'article suivant. L'analyse par composante principale (ACP) servit à observer les différences et similitudes dans les profils des ours et quels contaminants les caractérisent.

Les hypothèses liées à ce projet sont que d'une part les concentrations de composés dont l'utilisation est bannie comme les chlordanes et les BPC devraient être à des niveaux plus bas

qu’à des données publiées antérieurement. Au contraire, les métaux et les CAP devraient être pour leur part être à des niveaux supérieurs. La distribution des contaminants entre les tissus, quant à elle, devrait être similaire à celle qui a été reportée dans la littérature. Finalement, il est difficile d’établir d’avance si des composés seront associés davantage à des caractéristiques comme l’âge, le sexe et la région d’origine des ours.

# 2 SPATIAL CONTAMINANTS ASSESMENTS OF POLAR BEARS OF LIVER, FAT AND MUSCLE IN NORTHERN CANADA

## Distribution spatiale des contaminants dans le foie, gras et muscle d'ours polaires vivant dans le Nord canadien

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**Contribution des auteurs** : **Vincent Boutet** : rédaction première ébauche, analyses chimiques, analyses des données. **Kristin Eccles :** analyses des données, rédaction et révision. **Mélanie Dominique :** rédaction. **Marsha Branigan :** conceptualisation, échantillonnage. **Markus Dyck :** conceptualisation, échantillonnage. **Peter van Coeverden de Groot :** conception, échantillonnage, financement. **Stephen C. Lougheed :** conceptualisation, financement. **Allison Rutter :** conceptualisation**,** analyses chimiques, révision et supervision. **Valerie S. Langlois :** conceptualisation, ressources, rédaction, révision, supervision et financement

## 2.1 Introduction

Arctic biota are subject to numerous natural and anthropogenic stressors, including climate change, hunting pressure, invasive species, emerging pathogens, and changes in food web dynamics (AMAP, 2018) In addition to these factors, a complex mixture of environmental contaminants, including emerging contaminants and persistent organic pollutants (POPs) are being detected into Arctic ecosystems. Even though the use of some biologically hazardous chemicals have been regulated or banned in industrialised countries as a result of several conventions, they still exist in the biosphere (Bentzen., 2008). These contaminants are transported through the atmosphere, oceans, and rivers and deposited at higher latitudes into the Canadian Arctic (AMAP, 2017). Although local sources of Arctic contamination exist in communities and in relation with activities such as petroleum extraction, many contaminants originate from mid-latitude regions in Asia, Europe, and North America (Law et al., 2017). These contaminants include metals, pesticides and flame retardants (Gibson, 2020).

Biomonitoring assesses ongoing changes in an ecosystem using physical and chemical measurements (Bondaruk et al., 2015). Polar bears (*Ursus maritimus*), as a sentinel species, can be used to evaluate the health status of the Arctic biota as they are top predators that are exposed to contaminants that have bioaccumulated and biomagnified along the food chain. Further, population dynamics of polar bears in the Canadian Arctic as have been well-studied and have a large circumpolar distribution covering up to 350 000 $km^2$ (Bromaghin et al., 2015; Auger-Méthé et al., 2016). They are also omnipresent since most of the polar bear population worldwide (16 000 out of 26 000) resides in Canada (Koehler et al., 2019).

The diet of polar bears mainly consists of ringed seals, which is high in fat and a tissue known to preferentially accumulate POPs (Ciesielski et al., 2017). Contaminants have been quantified in polar bear tissues (such as liver, spleen or fat) including total Hg, various pesticides, PCBs (polychlorinated biphenyls), and PBDEs (polybrominated diphenyl ethers) (reviewed in Dominique et al., 2020). Beside to the review of Dominique et al., to our knowledge no studies have reported the presence of PACs (polycyclic aromatic compounds) in polar bears. The concentrations of contaminants in polar bear tissues have shown to disrupt thyroid hormones, lipid metabolism, and changes brain neurochemistry (Routti et al., 2019). Furthermore, risk of exposure to these contaminants is high for Indigenous communities living in the Arctic as polar bear meat is an integral part of their traditional diet (AMAP, 2016). The impact on the health of human, especially on Indigenous communities, have been measured in many ways such as

associations between PCB exposure and carcinogenicity, reproductive impairment, and neurodevelopmental anomalies in adult humans (Quinete et al., 2014).

There remain knowledge gaps on the current contaminant exposure of polar bears and by extension humans, since environmental factors are causing shifts in contaminant exposures. Thus, contaminant profiles in polar bears cannot be generalized due to differences between pollutants, species, and locations (AMAP 2021). The objective of our study was to quantify the distribution of contaminants including of PCBs, PACs, CHLs (chlordanes), and metals (including total and methyl mercury) in the muscle, fat, and liver of polar bears of different age, sex and locations from northern Canada.

## 2.2 Materials and Method

### 2.2.1 Sample collection, information, and preparation

A total of 49 polar bears (33 males,16 females and one with unknown sex) from Northern Canada were used for this study. Of these, 33 were adults, 15 were juveniles. The animals were taken from six locations: Northern Beaufort Sea (NB) (n = 3), Southern Hudson Bay polar bear subpopulations (SH) (n = 20), Western Hudson Bay (n = 1), Baffin Bay (n = 1), Foxe Basin (n = 12), and Gulf of Boothia (n = 12) between 2016 and 2019 by Inuit hunters (Figure 2-1). For each animal, liver, fat, and muscles tissue were collected opportunistically. Tissue samples were taken by Inuit hunters, bagged, labelled, and stored at -20 °C prior to analyses.

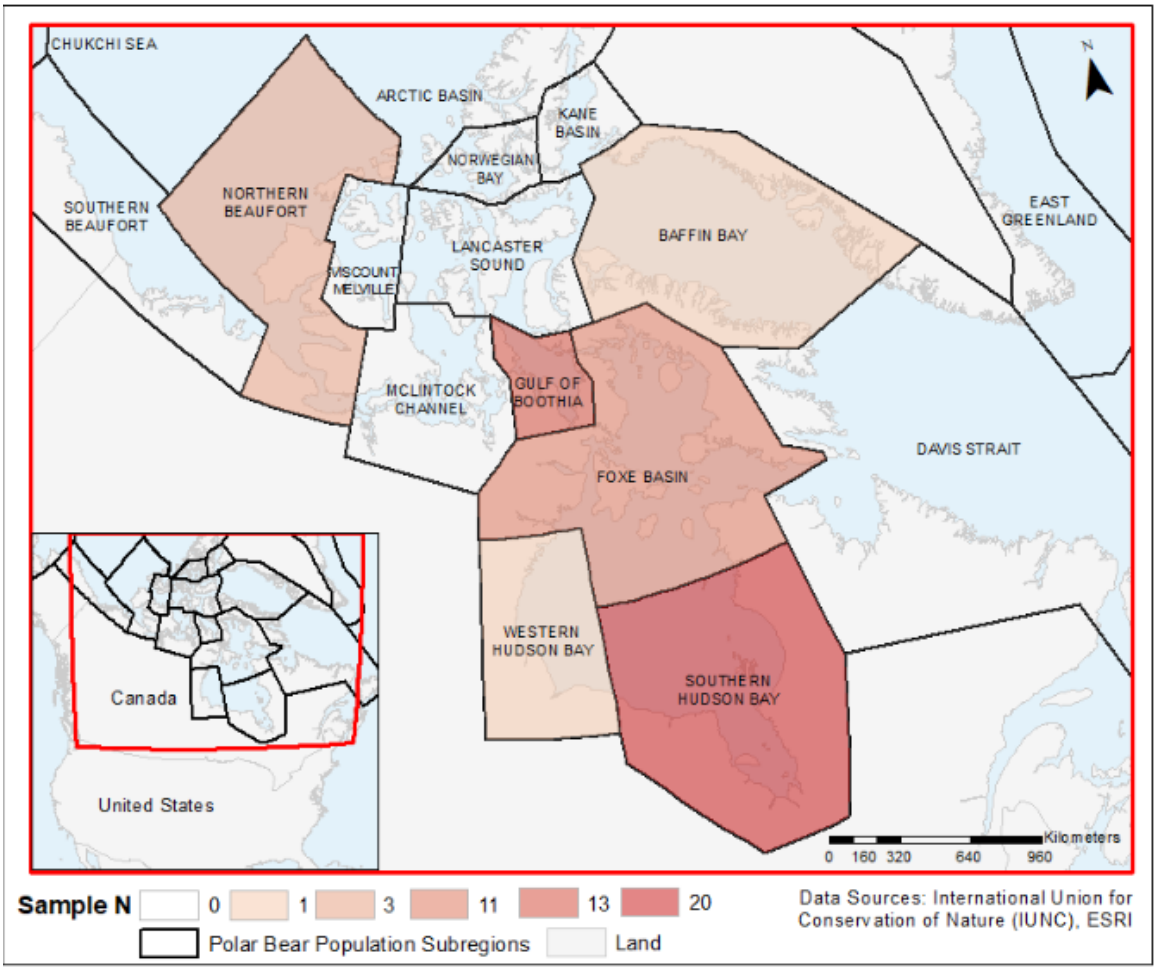


**Figure 2-1 Geographical origin and number of polar bears sampled out of a total 49.**

### 2.2.1 Metal analysis

Metal analyses were done by the Analytical Services Unit (ASU), Queen's University, Kingston, Ontario for all bears. 30 metals were analyzed including essential and non-essential metals. Samples were dried at 70 °C for 24 h and weighed (0.5 g dry weight) for all bears. A combination of commercially available single element and custom multi-element standards were used. Second source standards were used for calibration check solutions, including initial calibration verification and QA/QC solutions. ICP-AES and MS (PlasmaCAL) stock standards were used to make the working stocks and calibration standards. TORT-2 and TORT-3 tissue standards (National Research Council Canada) were used, with one reference material for every five samples tested.The dried samples were put into Digitubes (SCP Science), and 2 mL of $HNO_3$

and 6 mL of HCl (Fisher Chemical) were then added. The samples were capped and then heated at 50 °C for 5 h on a graphite digestion block (DigiPREP LS), then incubated at 90 °C for 2 h, ensuring that the tubes did not go dry. The final volume was approximately 5 mL. The volume was made up to 25 mL with distilled water. Samples were decanted into plastic autosampler tubes, then run on an Agilent 7700X ICP-MS (Santa Clara, California, USA). Samples were also analyzed by ICP-OES (Varian Vista axial ICP-OES) for B, P and S. Samples were filtered with 0.45 µm-syringe filters prior to analysis if particulates were present. Quality assurance quality control (QA/QC) methods included the use of duplicates, blanks and reference materials. QC/QA are presented in Table S9 (Supplementary info).

### 2.2.1 MeHg analysis

Analyses were conducted in Dr. Marc Amyot' lab at the *Département de sciences biologiques,* University of Montreal, Montreal, Quebec for all bears. Methylmercury (MeHg) analysis was performed following the methods of Krey et al., (2012), with tissues dried in an oven at 70 °C. Ultrapure water was used for all experiments (Millipore Milli-Q water) to rinse, clean, and prepare solutions. Reagents used were $CH_3HgCl$ (Alfa Easar Chemicals), methanol, acetic acid, and hydrochloric acid. Samples were weighted to the nearest 0.05 g dried for 12 h and then placed in a glass vial. For digestion of liver and muscle samples, 5 mL of $HNO_3$ 4M was added. For fat samples 5 mL 25% of KOH in methanol was added. All samples were incubated at 60 °C overnight. A 25-µL aliquot was transferred to a vial containing 30 mL Milli-Q water, 250 µL acetate buffer, and 40 µL $NaB(Et)_4$ as an ethylation reagent. The analysis for MeHg was conducted using a Tekran® 2700 Methyl Mercury Auto-Analysis System (Toronto, Ontario, Canada) using a 6-point standard curve ranging from 0.02 to 4.0 µg/L. For QA/QC a lobster hepatopancreas marine certified material (TORT-2) from the National Research Council Canada, duplicate analyses, blanks spikes and matrix spikes were included in each run and were within acceptable limits. QC/QA laboratory controls are presented in Table S10.

### 2.2.2 Total Hg analysis

Total Hg (THg) analyses were conducted by the ASU, Queen's University, Kingston, Ontario for all samples. THg measurement followed the protocol of Sonne et al., 2012, using cold vapour atomic absorption spectrophotometry (Milestone DMA-80 Direct Mercury Analyzer). Samples were air dried 24 h then weighed on an electronic scale (Metler Toledo XS204) on quartz

or nickel weigh boats. An ICP-AES and MS (PlasmaCAL or Sigma) stock standard of 1000 ppm Hg was used to make the working stocks and calibration standards. For tissues, reference materials TORT and TORT-2 (Dogfish liver) from National Research Council Canada were included. An aqueous calibration check sample was included in all runs. Duplicates, blanks, spikes and matrix spikes were within acceptable ranges. QC/QA laboratory controls are presented in Table S10.

### 2.2.1 Polycyclic aromatic compounds (PACs) analysis

Analyses were completed by ASU, Queen's University, Kingston, Ontario for 24 bears. Tissue samples were air-dried overnight in a room temperature (24 ˚C) oven or fume hood and then representative sub-samples were taken. 11 parents compounds and their alkylated congeners were analyzed. Prior to extraction, the surrogate Semivolatile Internal Standard Mix (Supelco), sodium sulphate (40 g), and Ottawa sand (20 g) were added to each sample. Samples were weighed using an electronic scale (Metler Toledo XS204) with one duplicate per run. Extraction was by the Soxhlet method over 6 h with 4-6 cycles per hour, using 300 mL of dichloromethane.

The extract was concentrated by roto-evaporation to an approximate volume of 1 mL and reconstituted to 10 mL in dichloromethane. The extraction was sub-sampled for cleanup and fractionation with a 1 m gel permeation chromatography column (70 g S-X3 BioBead stationary phase, 100% dichloromethane mobile phase). The fractions containing lipids and PACs were determined by pre-calibration. Only the PAC fraction was collected and concentrated, after which the solvent was exchanged with hexane using roto-evaporation to an approximate volume of 1 mL; this fraction was cleaned using a Silica cleanup column and the eluate (>10 mL) was concentrated to a volume of between 0.5-1mL. The samples were further cleaned using a homemade silica column (Silica Gel - Davisil grade 22, pore size 60 Å, 60-200 mesh) in hexane. The samples were flushed with 12 mL of hexane, discarding the first 3 mL; this fraction is set aside. A second fraction, the one of interest, was collected of at least 15 mL of 1:1 hexane:dichloromethane and then concentrated to a final volume of 0.5 mL. A 100-µL aliquot was spiked with internal standard L429-IS and L429-AS (Wellington Laboratories) before analysis.

The samples were analyzed using a GC/MS (gas chromatography with inert mass selective detector) Agilent 6890N GC 5975 Mass Spectrometer, an HP-5MS capillary column (30 m, 0.25 mm i.d. x 0.25 µm film thickness) and Enhanced ChemStation (MSD ChemStation D.02.00.275) software. The conditions were sample volume of 1 µL, pulsed splitless injection,

temperature programmed as ‘ramp’, and constant helium carrier gas pressure. Data were collected for all ions within the mass range of 128-302 m/z. Data selection criteria were based on compound retention time and on the relative intensity of primary and secondary ions for standard reference PACs and extracted samples. Calibration standards containing known concentrations of all reported PACs were used for PAC quantitation. Control samples, duplicates, and blanks were extracted for 10% of the samples. PAC values were reported as ng/g dry weight (ppb). Duplicates, blanks, and controls were made and were within the target range. QC/QA laboratory controls are presented in Table S11.

### 2.2.1 Chlordanes analysis

Analysis of CHLs was done at the *Institut national de recherche scientifique* (INRS), Quebec (Canada) for all bears. The chlodanes analyzed were oxychlordane, cis/trans-chlordane, heptachlor and cis/trans-nonachlor. Tissue samples were dried at room temperature (24 ˚C). For each individual, between 0.2-0.5 g of dried tissue was used for the extraction. Samples were purified after extraction using Thermo Scientific Reacti-Therm which 4 mL of hexane and 1 mL of samples percolate through a filter and the eluant was purged with nitrogene gas. Separation and quantification of analytes were completed using gas chromatography-mass spectrometry (GC-MS Thermo 1310), a capillary column (30 m, 0.25 µm i.d. x 0.25 µm film thickness), and the Chromeleon software. The final volume was 15 mL for all samples. Homogenization of the samples was made via lyophilization over 5 days at 20 °C with 15 mL of hexane and accelerated solvent extraction. Internal standards were spiked with cis-chlordane, trans-chlordane, cis-nanochlor, trans-nonachlor, oxychlordane and heptachlor (Cambridge Isotope Laboratories Inc.). Methods are detailed in Tartu et al., 2017. QC/QA laboratory controls are presented in Table S12.

### 2.2.2 Polychlorinated biphenyl (PCB) congener analysis

Analyses of PCBs were completed by the ASU, Queen’s University, Kingston, Ontario for 24 bears. 20 PCBs were analyzed (see table S4). All samples were dried 24 h at room temperature (24 ˚C). We used the Soxhlet method from Verreault et al., 2005 for extraction over a 6-h period at 4 - 6 cycles per hour, with 250 mL of dichloromethane. The extract was concentrated by roto-evaporation to an approximate volume less than 1 mL and reconstituted to 10 mL in dichloromethane. Each extraction was sub-sampled for cleanup and fractionation in a 1 m gel permeation chromatography column (70 g S-X3 BioBead stationary phase, 100% dichloromethane mobile phase). The lipid fraction was determined gravimetrically. The congener

fraction was collected, concentrated and solvent exchanged with hexane by roto-evaporation to an approximate volume less than 1 mL. This fraction was run through a Florisil cleanup column and the eluate (>10 mL) was concentrated to a volume of 2 mL. The samples are further cleaned using a homemade silica column (Silica Gel - Davisil grade 22, pore size 60 Å, 60-200 mesh) in hexane. Each sample was flushed through a column using at least 25 mL of hexane and then concentrated into a final volume of 0.5 mL. The samples were analyzed using GC/MS/MS (gas chromatography with tandem mass spectrometry using a Varian 4000 GC Mass Spectrometer) with a SGE Forte capillary column (60 m, 0.25 mm i.d. x 0.25 μm film thickness), and using Varian MS Workstation V.6 software. Data were collected for all ions within the mass range of 150-550 m/z. Data selection criteria were based on compound retention time and on the relative intensity of primary and secondary ions for standard reference congeners and extracted samples. Calibration standards containing known concentrations of all 209 PCB congeners (matrix spikes) were used for congener quantification. The congener values were reported as ng/g dry weight (ppb). Lipid values were reported as percent dry weight. Triplicates, blanks, and controls were included and were within the target range. QC/QA laboratory controls are presented in Table S13.

### 2.2.1 Data analysis

To prepare for data analysis the data was compiled and cleaned. Sample replicate values (for QA/QC purposes) were averaged and the mean was used in the analysis. PACs values are reported are the sum of unsubstituted parent compounds and their alkyl-substituted derivatives. Contaminant concentrations that were below the limit of detection (LOD) were replaced with half the LOD. Samples that had missing data (n.a) were imputed by using regularized iterative PCA algorithm from the missMDA package in R (Josse and Husson., 2016). This algorithm is used as a pre-processing step before doing a PCA on a dataset: it consists of imputing missing values with initial values such as the mean of the variable. If the seed argument is supplied to a specified value, a random initialization is conducted, with the initial values selected from a gaussian distribution and the mean and standard deviation derived from the observed values (Josse and Husson., 2016). Tissues that had few missing values were estimated using tissue specific chemical subgroup measurements. For tissues that has a larger number of missing values, all tissues (i.e., liver, fat, muscle) within the chemical subgroup were used for the imputation. With the completed data, a PCA was used to examine the overall variance patterns in each of the tissues across the different contaminant groups. All data analyses were completed in R Version 3.4.4 and the map was made in ArcGIS 10.7.

## 2.3 Results

PCAs show highlight the overall variance in contaminant burden, age, sex, and location. Figures (2-2 to 2-6) are separated into six subplots where A and B are the first four components of liver, C and D are muscle, and E and F are fat. The loadings associated with each biplot and the variance explained by each PCA are in Tables S5-S8.

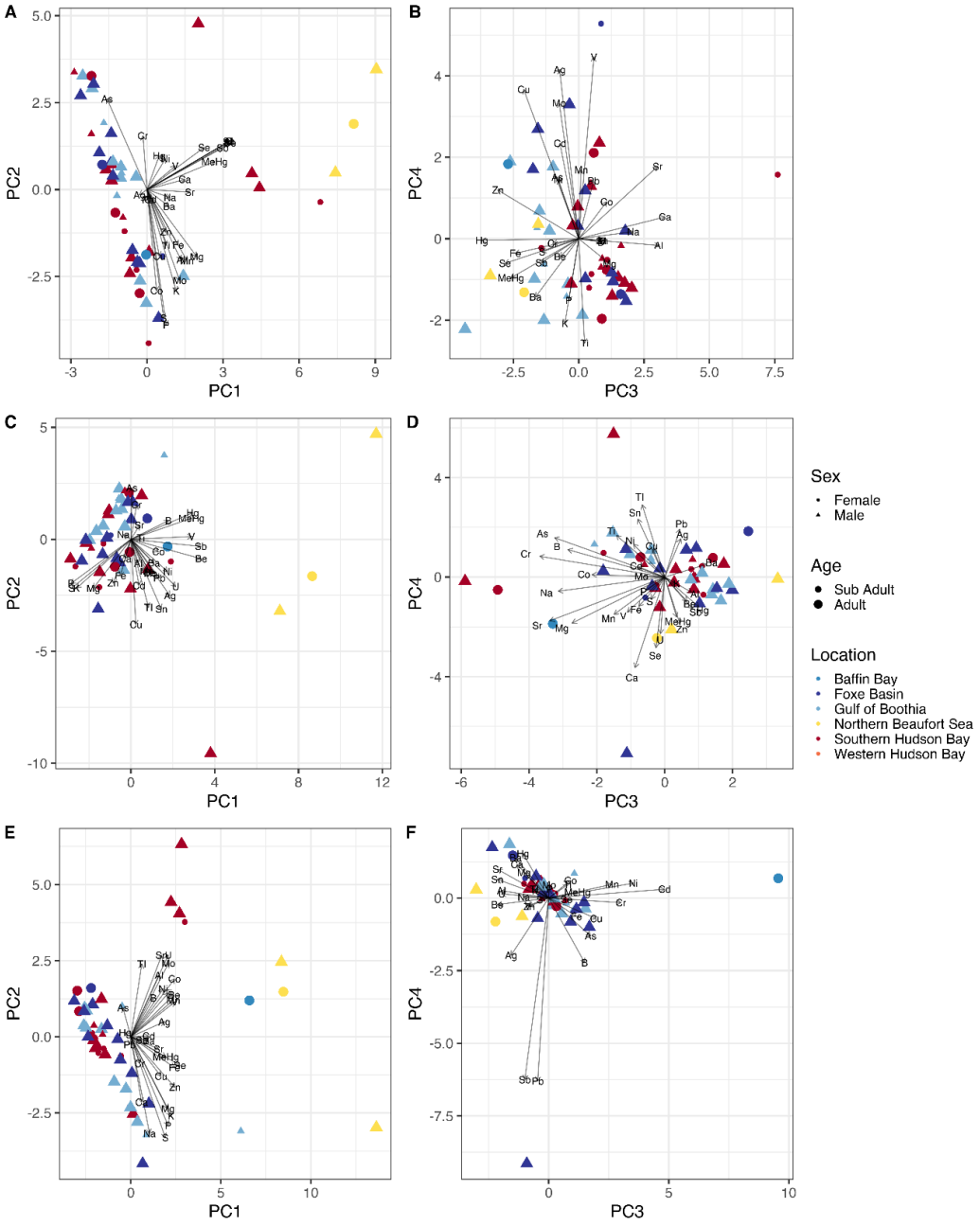


**Figure 2-2 Principals component analyses (PCA) of metals with biplot of the fourth components for Liver (A & B), Muscle (C&D) and Fat (E & F). The locations are shown by different colours using the acronyms BB (Baffin Bay), FB (Foxe Basin), GB (Gulf of B Boothia), NBS (Northern Beaufort Sea), SHB (Southern Hudson Bay) and WHB (Western Hudson Bay)**

### 2.3.1 Metals

Essential elements like phosphorus (P), potassium (K), sulfur (S), and sodium (Na) yielded the highest concentration (1.6-14.5 mg/g). Non-essential metals including MeHg, arsenic (As), cadmium (Cd), lead (Pb) had average concentrations of 2.4 ± 2.5, 1.5 ± 1.4, 2.1 ± 1.2, 0.4 ± 0.5 µg/g in liver and in muscle, at 0.4 ± 0.3, 1.8 ± 1.6, 0.07 ± 0.05 and 0.5 ± 0.4 µg/g, respectively (Table S1).

Figure 2-2 shows the biplot PCAs for metals. Overall, there were no apparent differences in the contaminant profiles from bears of different ages and sexes but location may be an important contributor to variability in contaminant concentrations. In all tissues, the three bears (two adult males and one adult female) from the Northern Beaufort Sea had different contaminant profiles from most of the other bears. This dominated the plots of PC1 an PC2 suggesting the contaminant burdens are markedly different. These bears were typically associated with Hg, MeHg, Se (selenium), Sb (antimony) and Be (beryllium). In PC 3 and 4 of the liver (Figure 2B), bears from Gulf of Boothia clustered together with highly loaded metals Zn (zinc), Hg, Fe (iron), Se, and MeHg, whereas bears from Southern Hudson Bay and Foxe Basin clustered together with highly loaded metals Sr (strontium), Ga (gallium), Na, and Al (Aluminum). In muscle and fat there were no additional location-based patterns observed.

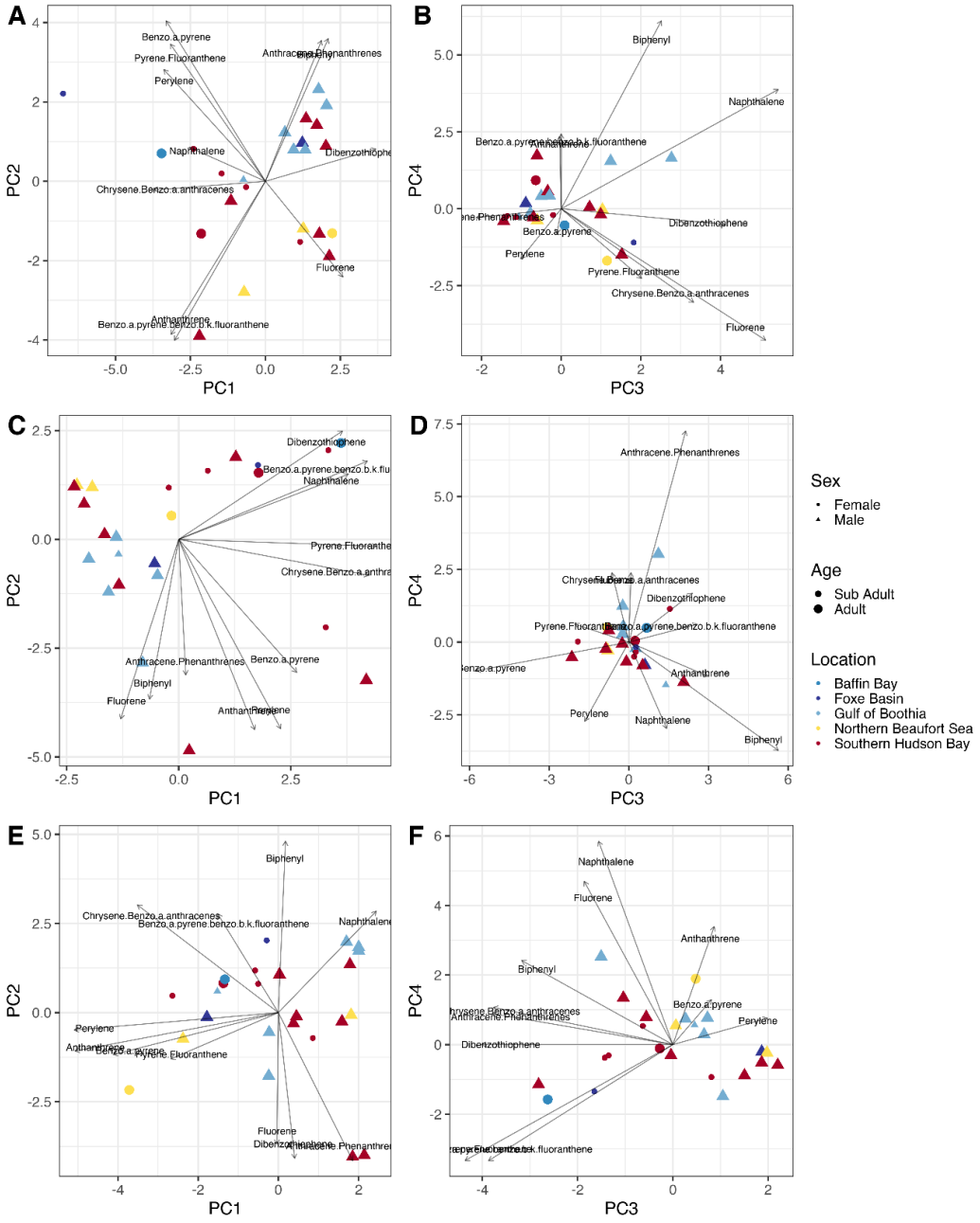


**Figure 2-3 Principal component analyses (PCA) of PACs with biplot of the fourth components for Liver (A & B), Muscle (C & D) and Fat (E & F). The locations are shown by different colours using the acronyms BB (Baffin Bay), FB (Foxe Basin), GB (Gulf of Boothia), NBS (Northern Beaufort Sea), SHB (Southern Hudson Bay) and WHB (Western Hudson Bay).**

### 2.3.1 PACs

The highest concentrations of PAC congeners measured the polar bear tissues were naphthalene, anthracene/phenanthrene, biphenyl, and dibenzothiophene. Liver and fat had the highest PACs measured values, of which naphthalene and anthracene/phenanthrene have the highest concentration (6.1 – 2800 ng/g). In liver PC1 and PC2, Figure 2-3A, bears from Gulf of Boothia profiles were clustered with biphenyl, anthracene/phenanthrene and dibenzothiophene and bears from Northern Beaufort were associated with fluorene. Interestingly, four out of six juveniles were associated to naphthalene and chrysene/benzo(a)anthracene. In muscle PC1 and PC2 (Figure 2-2C), three bears from Southern Hudson Bay dominated the variance. In PC3 and PC4 of muscle (Figure 2-2D), bears from Southern Hudson Bay were associated with benzo(a)pyrene, perylene naphthalene, and biphenyl, whereas bears from Baffin Bay were associated with anthracene/phenanthrene, chrysene/benzo(a)anthracene. This difference may be related to the body burden in benzo(a)pyrene, perylene and naphthalene. In fat PC1 and PC2 (Figure 2-2E), subadults were associated with chrysene/benzo(a)anthracene and benzo(b&k)fluoranthene. There were no patterns observed in PC3 and PC4.

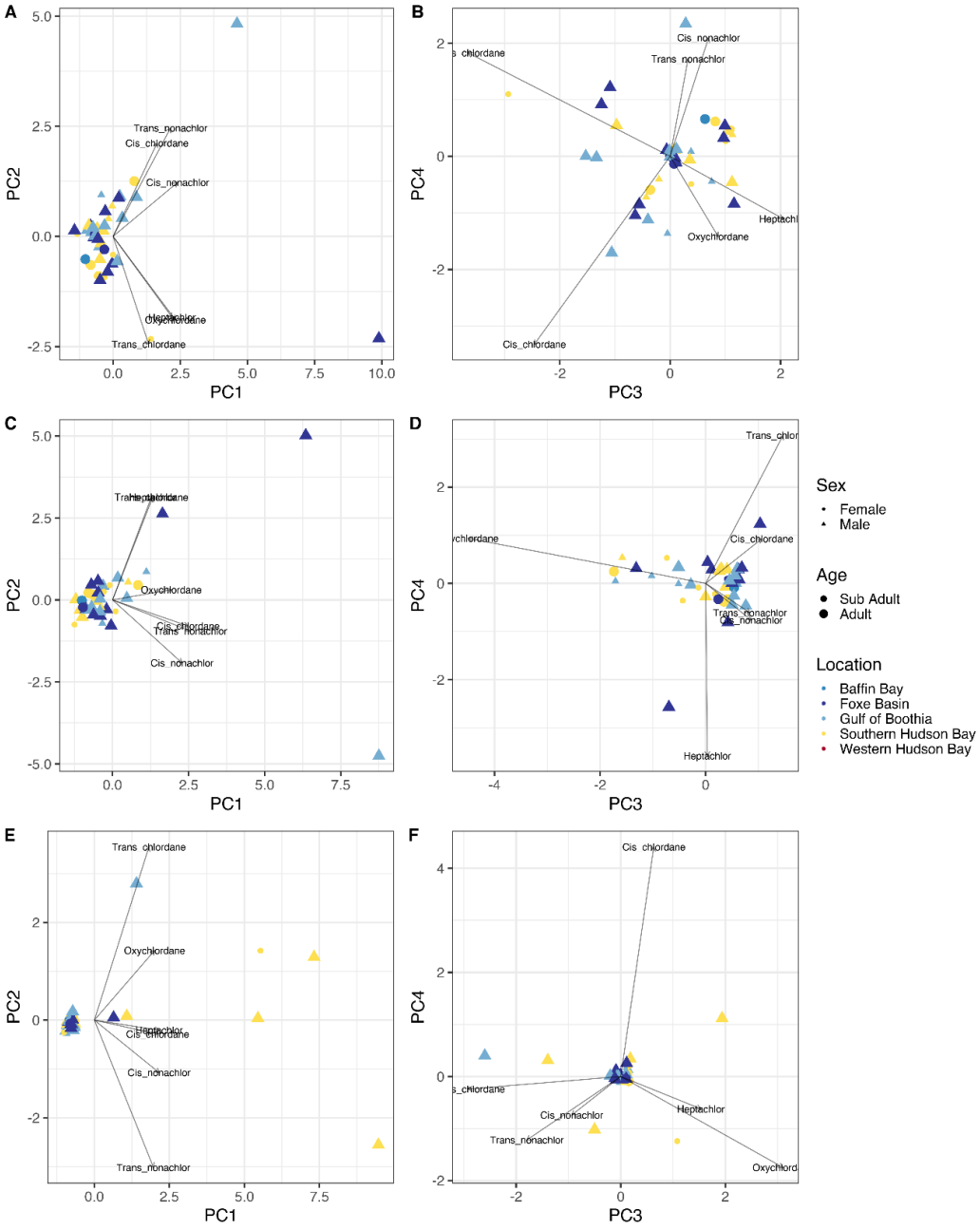


**Figure 2-4 Principal component analyses of chlordanes with biplot of the fourth components for Liver (A & B), Muscle (C & D) and Fat (E & F). The locations are shown by different colours using the acronyms BB (Baffin Bay), FB (Foxe Basin), GB (Gulf of Boothia), NBS (Northern Beaufort Sea), SHB (Southern Hudson Bay) and WHB (Western Hudson Bay).**

### 2.3.1 Chlordanes

Many CHL's values were measured below the LODs for each of the tissues. The liver yielded the highest concentration on average of CHLs; oxychlordane (39 ± 35 ng/g) and heptachlor (8.5 ± 6.1 ng/g) (Table S3). These two compounds also had the highest concentration in fat (50 ± 150 ng/g and 22 ± 64 ng/g, respectively), while muscle tissue had the lowest concentration for each compounds with a greater concentration of 14 ± 20 ng/g for heptachlor. The majority of variance in PC1 and PC2 across all tissue types were driven by a few bears with comparatively higher contaminant burdens. In the liver PC3 and PC4 (Figure 2-4B). In muscle PC3 and PC4 (Figure 2-4D) plots, the variance of contaminant burdens in the juveniles was associated with oxychlordane. In the fat for all PCs (Figures 2-4E and 2-4F), bears from the Southern Hudson Bay had the highest concentrations and as a result were associated with the highly loaded CHLs.

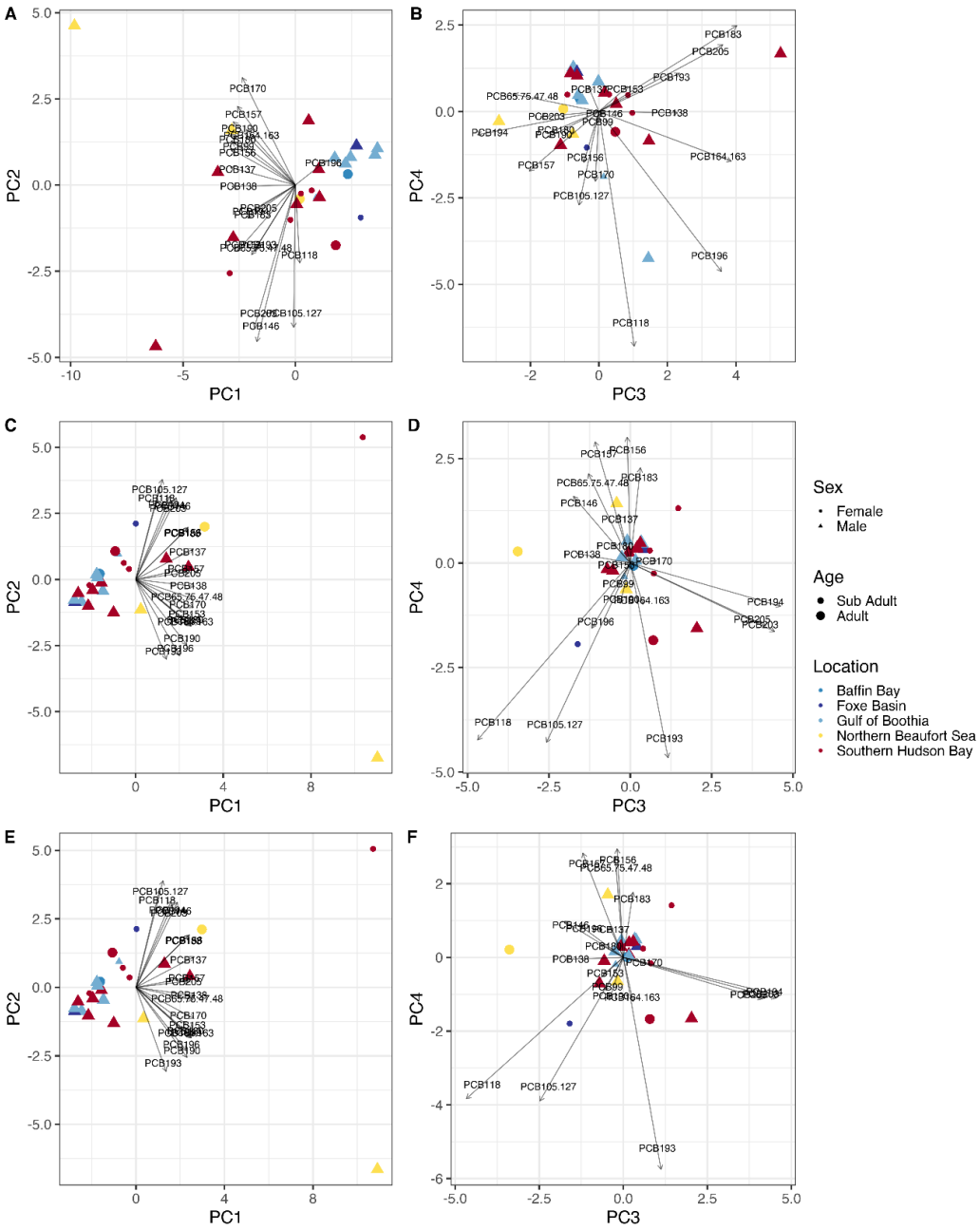


**Figure 2-5 Principal component analyses (PCA) of PCBs with biplot of the fourth components for Liver (A & B), Muscle (C & D) and Fat (E & F). The locations are shown by different colours using the acronyms BB (Baffin Bay), FB (Foxe Basin), GB (Gulf of Boothia), NBS (Northern Beaufort Sea), SHB (Southern Hudson Bay) and WHB (Western Hudson Bay).**

### 2.3.1 PCBs

The PCB congeners with the highest average concentrations were PCB-153, -180, -138, -170 and -99 in liver (1000 ± 580; 550 ± 520; 160 ± 100; 150 ± 120; and 140 ± 120 ng/g d.w respectively; Table S4) and in fat (2300 ± 1600; 990 ± 850; 240 ± 140; 320 ± 260; 200 ± 200 d.w). PCB-153 had the highest concentration all tissues ranging from 27.1 to 1611.6 ng/g. In liver PC1 and PC2 (Figure 2-5A), bears from Baffin Bay, Foxe Basin, and Gulf of Boothia were clustered together by the PCB-196. The three bears from NBS had a large intra-group difference. There were no observable patterns between the age and sex profiles. The PCBs with the highest loadings were PCB-196, PCB-137, PCB-156 and PCB-180. The distribution of PC3 and PC4 (Figure 2-5B) clustered with three profiles of Northern Beaufort Sea highly loaded with PCB-47+48+65+75, PCB-194 and PCB-153 while the profiles in the Southern Hudson were highly loaded with PCB-183, -163 + 164, -205. Profiles from Baffin Bay, Gulf of Boothia, and Foxe Basin are clustered together and have comparable in their body burden of PCB-137. The PCA of muscle (Figure 2-5C and 2-5D) and fat (Figure 2-5E and 2-5F) have similar contaminant and bear distribution. PC1 and PC2 were strongly driven by two profiles from the Northern Beaufort Sea and Southern Hudson Bay. In these PCs bears from the Northern Beaufort Sea also cluster together indicating the PCB profiles are similar. The highest positive loading values for the first component were PCBs-47+48+65+75, PCB-137, PCB-138, PCB-153, PCB-157 and PCB-170.

## 2.4 Discussion

### 2.4.1 Metals

Some metals, such as Cu or Zn, are essential for metabolism and maintenance of homeostasis in polar bears (Braune et al., 1991). Alkalis and alkaline earths, which were measured in this study likely become bioavailable from weathering and erosion of the lithosphere (e.g., Si (silicon), Al, Fe; Cai et al., 2011) and from the sea (e.g., Na, Mg (magnesium), Ca (calcium); Harnung and Johnson., 2012). Depending on the toxicokinetic properties of the metal, some will biomagnify, while others will be biodiluted throughout the food chain (Janssen et al., 1993). In the study, the metal contents measured in liver and muscle were higher than those measured in fat. The metals contents measured in this study were similar to or higher than those of other studies (reviewed in Dominique et al., 2020).

Toxic metals, such as arsenic, come from multiple sources such as mining activities (from tailings and dust), boat traffic, infrastructure development (Campbell et al., 2005), and fossil fuel combustion (Foster et al., 2014). Therefore, airborne particulate metals represent one route of exposure for polar bears even if the main metal exposure comes from their diet. Particulate Pb, As, and Cd were measured in the air in Northern Canada (Becagli et al., 2020). Cr, V, and Ni deposition has also been measured using lichens in Yellowknife which may result from local mine dust (Naeth and Wilkinson., 2008). Toxic metals like As can have both organic and inorganic forms, which are readily absorbed from the intestinal tract (Campbell et al., 2005) and could explain its omnipresence among profiles. Polar bears appear to be exposed to As at high background values owing to content of arsenic found in sediments in estuaries of the Beaufort Sea (Hartwell et al., 2020), values exceeding the effects range low (ERL) of 8.20 µg/g dw. This finding is of interest given that the inorganic form of As is more bioavailable and more toxic than its organic form (Clemente et al., 2019). In the present study, As, Cd, Pb, Cr, Ni, and Zn were detected in polar bear liver. Muscle had similar levels measured and fat yielded less metals. The metal concentrations measured in liver were higher than those reported in other studies. Woshner et al. 2001 measured these elements in liver of 24 bears of Alaska (As = 0.36, Cd= 1.88, Pb = 0.32, Zn = 314.6 ug/g dw). Dietz et al. 2000 measured a range (geometric means) of 78.8 to 304.0 ug/g dw of Zn and levels of Cadmium concentrations in liver of a range 0.48-7.92 ug/g dw in 100 bears in Greenland. It is suspected that exposures to these metals may cause immunotoxicity (López-Berenguer et al. 2020).

Routti et al. (2012) evaluated the trend of Hg in the liver of polar bears between 1980 and 2002 and observed an overall increase in Hg concentrations in the Beaufort Sea polar bears. This prior study measured Hg concentrations at highest levels in the Beaufort Sea and lowest in the Hudson Bay polar bears populations. Even though our sample size is low from this region, this supports results from the PCA which shows bears from the Northern Beaufort Sea had higher concentrations of THg and MeHg. This trend is also similar to other Arctic species, where western Arctic biota generally have higher THg than in the east of Canada (Braune et al., 2014). Further, a 5,200 km transect across Canada (150˚ to 53˚) demonstrated that seawater the highest concentrations of MeHg in the shallow depths of the Northern Beaufort Sea (Wang et al., 2018). The exact mechanism by which Hg enters the food chain are not fully understood (Braune et al., 2014). Lavoie et al., (2013) has reported that Hg to bioamplifies more in the food webs in colder

regions. An emerging exposure of concern is the melting permafrost which promotes the release and bacterial methylation of Hg stored in snow (Lippold et al., 2020).

Our study also found higher concentrations in the liver than in muscle. On average THg concentration in the liver were eight times higher than fat and 76 times higher in muscles. In comparison, in the most common prey, ringed seal, THg was 32 times higher in seal liver than muscle (Dehn et al., 2005). The high concentration of THg in liver is due to the detoxification function, as it is a site where the more toxic MeHg is demethylated into inorganic Hg in preparation for elimination (Krey et al., 2012). MeHg was also found in higher concentrations in the liver than fat and muscle (6-fold in muscle; 80-fold in fat). The order in which MeHg accumulate in organs throughout the bloodstream may explained this last distribution. Isotope tracer analyses have identified that MeHg first accumulates in the liver, kidney, and spleen, then to muscle and the brain; Evan et al.2016. The levels of MeHg measured in the liver in our samples do not suggest causing structural irregularity on the liver. Rawson et al. 1992 estimated that it takes more than 200 µg /g dw of MeHg in the liver of Stranded Atlantic Bottlenose Dauphin to create pigmentation (cell membrane damage).

### 2.4.1 PACs

There are many sources of PACs including incomplete combustion of organic matter, abrasion of materials, fats and oils as well as creosote treatment for railways (Pollock and St. Clair., 2020). Polar bears may be exposed to PACs from a variety of mechanisms, including absorption through the respiratory membranes or skin, food, or maternal transmission (Wallace et al., 2020). Despite the fact that air is the environmental compartment that receives the most current PAC emissions (Berthiaume et al., 2020), there is little known about the importance of inhalation as a route of PAC exposure in the Arctic. It is likely that Canada's polar bears will be exposed via inhalation to these compounds, especially with the increase in wildfires due to climate changes (Flannigan et al. 2009).While PACs are not likely to biomagnify due to the high rate of metabolism in vertebrates (Wallace et al., 2020), the presence of PACs in the polar bear tissue samples suggests that exposure is occurring at a rate that exceeds elimination. Since mammals readily metabolize PACs, few studies evaluate exposure and effect in Arctic mammalian species. The distribution between tissues may reflect the metabolism efficiency to eliminate PACs based on the inducibility of liver CYP1 enzymes.

In this present study, the omnipresence of naphtalene and anthracene/phenanthrene in liver and fat tissues may result from the low molecular weight of the compounds resulting from

the two to three carbon ring structure. Low concentrations of PACs in muscle might suggest that metabolism is more efficient in muscle than in liver or fat. In another way it is possible that liver metabolism and elimination may successful based on the dose so that compounds are eliminated before they are accumulated in other tissues. Polar bears in Canada may be exposed to PACs, particularly low molecular weight compounds, through volatilization in the atmosphere from oil and gas extractions and development related activities in western Canada.

Thomas et al., 2020 measured low molecular weight PACs in river otter (*Lontra canadensis*) livers from sites around the Athabasca Oil Sands Region in northern Alberta and found hepatic concentrations ranging from 0.07 – 23 ng/g lw, with the highest concentrations being PACs of low molecular weight. Furthermore, measured alkylated PACs were consistently detected at higher levels than their parent compounds, as observed in our study suggesting that these chemicals undergo metabolism. According to PCA, PAC content in bear fat was associated by larger, high molecular weight PAC (i.e., anthanthrene, benzo(a)pyrene, perylene) even if their concentration were still low. This could be explained by the higher Kow of those compounds, and therefore, a greater affinity for hydrophobic tissue. The muscle profiles do not allow a precise pattern to be conclusive and seem to indicate that the measurement of PACs in muscles is not an effective tool for establishing trends between these compounds.

### 2.4.1 Chlordanes

While many pollutant concentrations have decreased in the arctic marine biota during the last two decades, PCBs and chlordanes in the arctic wildlife tissues have remained at high levels for the last ten years (AMAP, 2016). Routti et al., (2012) reported that some subpopulation of polar bears in the Arctic have a decreasing trend of chlordane. Samples collected in 2013–2014 relative to those collected in 2007–2008 show that PCB concentrations have declined in the southern Hudson Bay polar bears. Among recent studies that evaluated the biomagnification of these compounds, Dietz et al., (2015) measured a range of 765 – 3480 ng/g lw for ∑CHLs in adipose tissue of 165 polar bears sampled in 11 sub-regions in the Arctic. Our data suggest that polar bears are still being exposed to CHLs, although we detected lower levels of CHLs than Dietz et al., (2015). When exposed to CHL and nonachlor, polar bear livers biotransformed these compounds into oxychlordane and heptachlor epoxide, which may explain why oxychlordane had the highest concentrations in liver. Organochlorine preferenitally accumulate in fatty tissue (Campbell et al., 2005) and therefore 50% of ∑CHLs measured in fat was oxychlordane. Verreault et al., 2006 obtained 66% of oxychlordane while measuring content in fat in 107 adult and sub-

adult polar bears adipose tissue samples from Alaska, Canada, East Greenland, and Svalbard between 1996 and 2002. Low concentrations have been also reported in muscle tissues and might be explained by the heterogeneous structure of fat (Verreault et al., 2006). Past studies evaluate exhaustively chlordane as a whole (∑CHLs) and metabolite (oxychlordane) in fat tissue of polar bears but little is known about the distribution within other tissues and the relationship between chlordane enantiomers and oxychlordane. Oxychlordane and cis/trans-nonachlor were highly loaded in the liver PCA, and thus warrant further study on this relationship.

### 2.4.1 PCBs

Another organochlorine highly present in the Artic are PCBs (Helgason et al., 2013). Even though PCBs were banned since the 1970s, their persistence and large quantity emitted prior to the ban may explain why PCBs were measured in this study. Polar bears are exposed to a range of PCBs, but only a few congeners, especially chlorinated biphenyls 99, 138, 153, 170/190, and 180, account for the majority of PCBs measured in polar bear tissues (79 to 95%) (reviewed in Dominique et al., 2020). Dietz et al., (2004) reported the same PCB congeners and order of magnitudes in fat for polar bears in Greenland (PCB-153 (32%), PCB-180 (21%), PCB-170 (12%), PCB-138 (11%), and PCB-99 (7.3%)), as this study.

The omnipresence of congeneric 190 and (-138, -153, and -180) might be explained with the inability for polar bears to eliminate these compounds which result in significant biomagnification (Kucklick et al., 2002). The presence of PCB-153 and PCB-180 may be related to biomagnification and persistence in the food chain resulting from a high Kow (Knott et al., 2012). The presence of only a few congeners demonstrates the biotransformation efficiency of most PCBs (Kannan et al., 2005). Furthermore, PCBs with a higher chlorination concentration tend to accumulate more in the kidneys and liver than fat, possibly because these compounds cannot pass through the biological membranes (Dominique et al., 2020). Among our sites, polar bears liver profiles from Baffin Bay, Foxe Basin and Gulf of Boothia clustered together on the PCAs while Southern Hudson Bay and Northern Beaufort Sea profiles had the greatest diversity of contaminants profiles.

This observation may be related to differences in prey selection in bears from the Southern Hudson Bay area. Mckinney et al., 2011 assessed concentration of PCBs in fat in the context of fatty acid, $^{15}N$ and $^{13}C$ signature (Diet-index) between bears and ringed seal of Southern Hudson Bay. Higher level of PCBs remained even after adjustment of the Diet-index suggesting that recent sea ice-associated dietary and/or food web changes. Therefore, the chemical exposure for

these bears may be influenced by their ability to adapt to environmental stress (Letcher et al., 2018).

### 2.4.2 Conclusion

Over 110 different contaminants were measured in liver, muscle, and fat from 49 polar bears living in several regions of Canada. PCAs highlighted variation of contaminants by region which may result from different sources and environmental factors. The PCAs were not effective enough to establish trends for profiles of different ages and genders. Overall, concentration distribution measured in the tissues were in agreement with the literature; toxic metals were omnipresent in the liver, while organochlorine PACs and CHLs were mainly detected in fat. This study highlighted the large amount of contaminants present in polar bears and why monitoring their health is still a current issue.

# 3 DISCUSSION GENERALE ET CONCLUSION

## 3.1 Rétrospective

Lors de cette étude, la majorité des contaminants analysés furent détectés dans tous les tissus. En général, ce fut les muscles qui comportèrent la moins grande teneur en contaminants. L'analyse en composante principale (ACP) permit d'établir certaines tendances entre le profil des ours et leur région correspondante, mais aucune tendance ne fut observée entre les profils d'âge ou de sexe différents

Les métaux furent mesurés en plus grandes concentrations dans le foie que dans les muscles ou le gras. Les métaux mesurés en plus grande concentration furent les métaux essentiels soit les alcalins et alcalino-terreux. Les métaux toxiques tels que l'As, le MeHg ou le Cd était à des niveaux plus élevés dans le foie que d'autres études sur des ours polaires provenant de l'Alaska ou le Groenland. L'ACP établit que les profils provenant de la mer de Beaufort étaient ceux qui se distinguaient principalement par leur composition en MeHg et As.

Les CAP furent mesurés en plus grandes concentrations dans le foie que dans les muscles et le gras. Les CAP mesurés en plus grande concentration furent le naphtalène, l'anthracène/phénanthrène, le biphényl et le dibenzothiophène. L'étude de la littérature de Dominique et collègues (2020) mit en lumière le manque de littérature sur l'exposition des mammifères au CAP. Néanmoins l'omniprésence de ces composés répartit entre les trois tissus, et ce même s'ils sont éliminés aisément, suggère une exposition soutenue des ours polaires à ces composés. L'ACP permit d'établir que les profils de foie étaient associés aux CAP à plus petit poids moléculaire alors que les profils de ces contaminants dans le gras étaient plutôt associés à des composés de plus gros poids moléculaires.

Les chlordanes furent mesurés à basse concentration, mais furent détectés dans tous les tissus. Le foie contenu les plus grands niveaux de chlordanes. L'oxychlordane et l'heptachlor furent les composés mesurés en plus grande teneur. La proportion de ces composés par rapport au chlordane total est similaire à ce qui fut déjà observée par le passé dans le foie d'ours polaire. L'exploration des travaux antérieurs relève le manque de littérature sur les relations entre les composés des chlordanes entre autres les paires d'isomères.

Les PCB furent mesurés en plus grande concentration dans le gras. Parmi les 20 congénères, les composés 99, 138, 153, 170/190 et 180 furent ceux mesurés en plus grande concentration. La littérature suggère que l'omniprésence de ces composés serait la cause de

l'incapacité de leur métabolisation. L'ACP mit en relief la distribution étendue des profils de foie d'ours de la baie d'Hudson suggérant une différente exposition de contaminants entre les ours de cette région, et serait probablement causé par une différence au niveau des sélections des proies.

## 3.2 Limites de l'étude

L'ajout de certains éléments du projet aurait été intéressant à étudier. Premièrement, un plus grand nombre d'échantillons provenant de la mer de Beaufort (ou du moins dans les régions environnantes) auraient renforcit statistiquement les faits proposés entre ces deux régions. Ceci aurait pu amener une comparaison d'ordre plus grand i.e., le profil des contaminants d'ours polaires de l'Ouest canadien par rapport de l'Est canadien. Il aurait également été intéressant d'avoir davantage d'ours polaires provenant de l'ouest de la Baie d'Hudson et de faire le comparatif à ceux du sud pour établir si les profils se distinguent parmi cette même région. À cette fin, une analyse telle que le marquage isotopique comme celle du $^{13}C$, $^{15}N$ ou la composition en acides gras aurait été intéressant afin de voir le changement de régime alimentaire des ours face au stress environnemental subit par les répercussions des changements climatiques dans ces régions.

D'autres informations sur les profils auraient été intéressantes telles que le poids des ours chassés ou bien l'identification des paires juvéniles-femelles pour établir des différences de charge de contaminants entre les femelles et leur portée respective. De plus peu d'informations étaient véhiculées sur quel ours était chassé ; si ces derniers étaient les plus faibles parmi la population d'ours (donc les plus faciles à chasser) il se peut qu'ils étaient les plus malades en raison de leur grande consommation de contaminants et donc surévalué la charge de contaminants moyenne pour la population. D'un autre côté, des ours plus faibles pourraient signifier également qu'ils sont sous-alimentés en raison de leur incapacité à trouver des proies et ce faisant ces échantillons pourraient cette fois sous-évaluer la charge en contaminants moyenne. Dans le cas de cette étude, il était connu que la majorité des ours polaires chassés étaient des mâles; sachant que les ours polaires ne sont pas grégaires et que sont les mâles qui se déplacent sur une plus grande superficie de territoire, il est fort possible que les chasseurs se heurtaient à ceux qu'ils avaient la plus grande probabilité de trouver, ce faisant les mâles. Néanmoins, avoir davantage d'information à ce sujet aurait pu prévenir les incertitudes reliées au biais mentionné précédemment..

## 3.3 Nouvelles perspectives

Plusieurs études passées ont quantifié la charge corporelle en contaminants chez les ours polaires, mais elles utilisaient surtout des méthodes invasives, notamment en étudiant la matière adipeuse, le foie, les dents ou les reins. L'utilisation des poils fut également étudiée par certains chercheurs, mais cette méthode peut être biaisée par le dépôt atmosphérique de polluants organiques persistants (Bechshoft et al. 2012; Dietz et al. 2006). Ainsi, dans l'ère des changements climatiques où la population des ours polaires est en déclin, il serait intéressant de pouvoir évaluer la charge en contaminants des ours sans toutefois les tuer ou les perturber dans leur habitat. Une autre facette de ce projet, également financée par Génome Canada, sera d'évaluer le potentiel des fèces comme proxy pour prédire la charge corporelle en contaminants. En effet, puisque la majorité des contaminants présents chez les ours polaires proviennent de leur diète il est ainsi présumé que la quantité de contaminants présente dans les fèces est liée à la quantité présente dans les aliments. En raison d'un manque de temps, ce sera plutôt la co-auteure de l'article, Kristin Eccles, qui complètera cette partie du projet. Affichée sur la figure 3.1 la relation entre la quantité de MeHg dans les fèces et les autres tissus, la relation qui semble avoir le meilleur potentiel dans les résultats préliminaires.

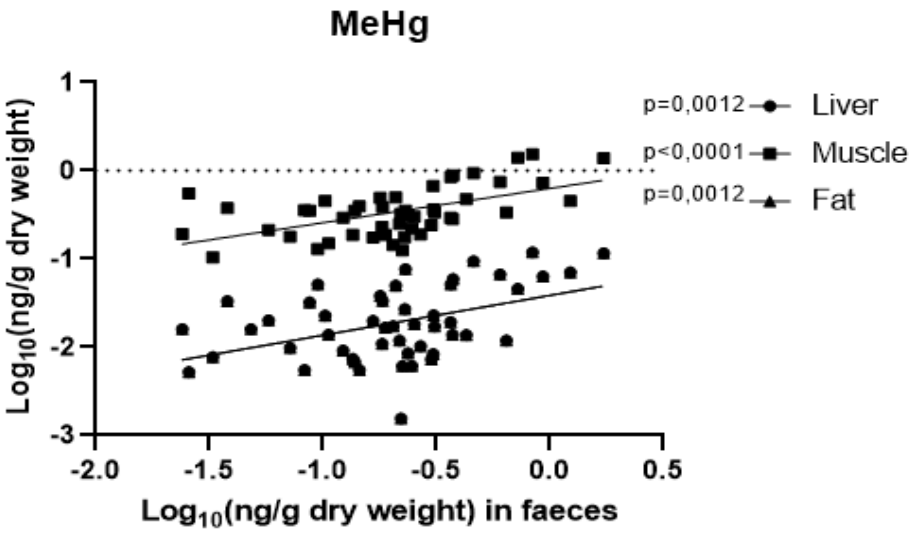


**Figure 3-1 Résultats préliminaires sur la relation entre la charge de contaminants présent dans les fèces selon celle présente dans le foie, les muscle et le gras**

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# ANNEXE I

**Table S1 Laboratory QA/QC Metals**

| Laboratory QA/QC (µg/g) | Blank | Blank | Normal Reporting Limit | Control 1 (ng/ml) | Control 1 (ng/ml) | Control 1 (ng/ml) | Control 1 Target (ng/ml) |
|---|---|---|---|---|---|---|---|
| Aluminium * | <10 | <10 | <10 | 25 | 25 | 25 | 25 |
| Antimony | <0.1 | <0.1 | <0.1 | - | - | - | - |
| Arsenic | <0.5 | <0.5 | <0.5 | 24 | 22 | 25 | 25 |
| Barium | <0.1 | <0.1 | <0.1 | 25 | 24 | 24 | 25 |
| Beryllium | <0.005 | <0.005 | <0.005 | 25 | 24 | 24 | 25 |
| Boron | <10 | <10 | <10 | 1900 | 1900 | - | 2000 |
| Cadmium | <0.005 | <0.005 | <0.005 | 25 | 24 | 24 | 25 |
| Calcium * | <20 | <20 | <20 | - | - | - | - |
| Chromium | <0.1 | <0.1 | <0.1 | 24 | 24 | 23 | 25 |
| Cobalt | <0.01 | <0.01 | <0.01 | 25 | 24 | 24 | 25 |
| Copper | <0.5 | <0.5 | <0.5 | 24 | 24 | 23 | 25 |
| Iron | <10 | <10 | <10 | - | - | - | - |
| Lead | <0.05 | <0.05 | <0.05 | 25 | 24 | 23 | 25 |
| Magnesium | <2.0 | <2.0 | <2.0 | - | - | - | - |
| Manganese | <0.2 | <0.2 | <0.2 | 25 | 24 | 24 | 25 |

**Table S1 Following**

| **Laboratory QA/QC** (µg/g) | **Control 2 (ng/ml)** | **Control 2 (ng/ml)** | **Control 2 (ng/ml)** | **Control 2 Target (ng/ml)** | **TORT-3 CRM (ng/ml)** | **TORT-3 CRM ** (ng/ml)** | **TORT-3 CRM TARGET** |
|---|---|---|---|---|---|---|---|
| Aluminium * | - | - | - | - | - | - | - |
| Antimony | 25 | 23 | 23 | 25 | - | - | - |
| Arsenic | - | - | - | - | 54 | 52 | 59 |
| Barium | - | - | - | - | - | - | - |
| Beryllium | - | - | - | - | - | - | - |
| Boron | - | - | - | - | - | - | - |
| Cadmium | - | - | - | - | 36 | 36 | 36 |
| Calcium * | - | - | - | - | - | - | - |
| Chromium | - | - | - | - | 1.6 | <7.5 | 1.7 |
| Cobalt | - | - | - | - | - | - | - |
| Copper | - | - | - | - | 390 | 370 | 420 |
| Iron | - | - | - | - | 140 | 150 | 120 |
| Lead | - | - | - | - | 0,19 | 0,19 | 0,19 |
| Magnesium | - | - | - | - | - | - | - |
| Manganese | - | - | - | - | 13 | 13 | 13 |

* Reporting limits raised due to interference

**Table S1 Following**

| **Laboratory QA/QC** (µg/g) | **Blank** | **Blank** | **Normal Reporting Limit** | **Control 1 (ng/ml)** | **Control 1 (ng/ml)** | **Control 1 (ng/ml)** | **Control 1 Target (ng/ml)** |
|---|---|---|---|---|---|---|---|
| Molybdenum | - | - | - | - | - | - | - |
| Nickel | 25 | 23 | 23 | 25 | 25 | 23 | 23 |
| Phosphorus | 32000 | 32000 | - | 30000 | 32000 | 32000 | - |
| Potassium | - | - | - | - | - | - | - |
| Selenium | 25 | 24 | 24 | 25 | 25 | 24 | 24 |
| Silver | 25 | 23 | 24 | 25 | 25 | 23 | 24 |
| Sodium | - | - | - | - | - | - | - |
| Strontium | 26 | 24 | 24 | 25 | 26 | 24 | 24 |
| Sulfur | 31000 | 30000 | - | 30000 | 31000 | 30000 | - |
| Thallium | 25 | 24 | 25 | 25 | 25 | 24 | 25 |
| Tin | - | - | - | - | - | - | - |
| Titanium | - | - | - | - | - | - | - |
| Uranium | 25 | 24 | 24 | 25 | 25 | 24 | 24 |
| Vanadium | 25 | 24 | 24 | 25 | 25 | 24 | 24 |
| Zinc | 24 | 23 | 24 | 25 | 24 | 23 | 24 |

**Table S1 Following**

| Laboratory QA/QC (µg/g) | Control 2 (ng/ml) | Control 2 (ng/ml) | Control 2 (ng/ml) | Control 2 Target (ng/ml) | TORT-3 CRM (ng/ml) | TORT-3 CRM ** (ng/ml) | TORT-3 CRM TARGET |
|---|---|---|---|---|---|---|---|
| Molybdenum | - | - | - | - | - | - | - |
| Nickel | 27 | 24 | 26 | 25 | - | - | - |
| Phosphorus | - | - | - | - | 54 | 52 | 59 |
| Potassium | - | - | - | - | - | - | - |
| Selenium | - | - | - | - | - | - | - |
| Silver | - | - | - | - | - | - | - |
| Sodium | - | - | - | - | 36 | 36 | 36 |
| Strontium | - | - | - | - | - | - | - |
| Sulfur | - | - | - | - | 1.6 | <7.5 | 1.7 |
| Thallium | - | - | - | - | - | - | - |
| Tin | - | - | - | - | 390 | 370 | 420 |
| Titanium | 24 | 24 | 24 | 25 | 140 | 150 | 120 |
| Uranium | 28 | 29 | 30 | 25 | 0.19 | 0.19 | 0.19 |
| Vanadium | - | - | - | - | - | - | - |
| Zinc | - | - | - | - | 13 | 13 | 13 |

NOTES: Scandium, Indium and Bismith were used as internal standards. Gas dilution (HMI) used: Y (med).

B, P and S by ICP-OES.

** Cr and Ni results for Tort-2 were outside normal control limits. Reporting limits were raised for samples (where applicable)

**Table S2 Laboratory QA/QC THg/MeHg**

| Laboratory QA/QC | IPRO.5 | IPRO.5 | IPRO.5 | Tort-2 | Tort-2 | Tort-2 |
|---|---|---|---|---|---|---|
| Hg (ppt) | 0.062 | 0.041 | 0.034 | 110 | 120 | 110 |
| MeHg (ng/gt) | 0.64 | 0.49 | 0.47 | 150 | 160 | 160 |
| Recovery (%) | 130 | 97 | 94 | 100 | 110 | 100 |

**Table S3 Laboratory QA/QC PACs**

| Laboratory QA/QC (ng/g) Compound | Blank | Control | Control Target |
|---|---|---|---|
| Naphthalene | <50 | 75.3 | 100 |
| Acenaphthylene | <5.0 | 127 | 100 |
| Acenaphthene* | <5.0 | 148 | 100 |
| Fluorene | <5.0 | 119 | 100 |
| Phenanthrene* | <10 | 65.3 | 100 |
| Anthracene | <10 | 94 | 100 |
| Fluoranthene* | <10 | 143 | 100 |
| Pyrene | <5.0 | 122 | 100 |
| Benzo(a)anthracene | <5.0 | 109 | 100 |
| Chrysene | <10 | 122 | 100 |
| Benzo(bkj)fluoranthene | <10 | 187 | 200 |
| Benzo(a)pyrene | <5.0 | 103 | 100 |
| Dibenzo(ah)anthracene* | <2.0 | 101 | 100 |
| Indeno(123cd)pyrene | <5.0 | 109 | 100 |
| Benzo(ghi)perylene* | <5.0 | 94.3 | 100 |

* Detection limit raised due to interferences

**Table S4 Laboratory QA/QC Chlordanes**

| Laboratory QA/QC (µg/g) | Blank | Blank 2 | Blank 3 | Control (ng/ml) | Control (ng/ml) | Control (ng/ml) | Control Target (ng/ml) |
|---|---|---|---|---|---|---|---|
| Oxychlordane | n.a | 7.427 | 5.762 | 23.522 | 22.516 | 24.779 | 25 |
| Cis-chlordane | 1.735 | n.a | n.a | 22.595 | 20.88 | 20.597 | 25 |
| Trans-chlordane | 1.237 | 0.571 | 0.182 | 22.418 | 25.537 | 24.345 | 25 |
| Heptachlor | 1.067 | 3.447 | 7.321 | 20.519 | 32.04 | 27.78 | 25 |
| Cis-nonachlor | 1.123 | 0.888 | 1.736 | 22.901 | 23.273 | 22.206 | 25 |
| Trans-nonachlor | 0.561 | 0.456 | n.a | 22.417 | 22.605 | 21.988 | 25 |

**Table S5 Laboratory QA/QC PCBs**

| Laboratory QA/QC (ng/g) Compound | Blank | Control | Control Target |
|---|---|---|---|
| PCB-65+75+47+48* | <0.2 | <0.2 | <0.2 |
| PCB-99 | <0.05 | <0.05 | <0.05 |
| PCB-118* | <0.05 | 0.45 | 5.0 |
| PCB-105+127 | <0.1 | <0.1 | <0.1 |
| PCB-146* | <0.2 | <0.2 | <0.2 |
| PCB-153* | <14 | 4.3 | 5.0 |
| PCB-137* | <0.05 | <0.05 | <0.05 |
| PCB-163+164* | <3.0 | <3.0 | <3.0 |
| PCB-138* | <6.0 | 3.7 | 5.0 |
| PCB-156* | <0.05 | <0.05 | <0.05 |
| PCB-157* | <0.05 | <0.05 | <0.05 |
| PCB-183* | <0.05 | <0.05 | <0.05 |
| PCB-180* | <17 | 5.2 | 5.0 |
| PCB-193* | <0.05 | <0.05 | <0.05 |
| PCB-170* | <0.05 | <0.05 | <0.05 |
| PCB-190* | <2.5 | <2.5 | <2.5 |
| PCB-196 | <0.05 | <0.05 | <0.05 |
| PCB-203* | <2.0 | <2.0 | <2.0 |
| PCB-194* | <5.0 | <5.0 | 5.0 |
| PCB-205 | <0.05 | <0.05 | <0.05 |

* Detection limit increased due to interferences

**Table S6 Concentrations of 30 metals (µg/g dry weight) measured in polar bear tissues (n = 49)**

| Metal | Mean ± SD µg/g | | |
|---|---|---|---|
| | Liver | Muscles | Fat |
| Aluminium | 11.4 (± 6.9) | 9.5 (± 8.3) | 6.2 (± 3.1) |
| Antimony | 0.3 (± 0.1) | 0.3 (± 0.1) | 0.5 (± 0.3) |
| Arsenic | 1.5 (± 1.4) | 1.8 (± 1.6) | 1.2 (± 0.8) |
| Barium | 0.2 (± 0.1) | 0.12 (± 0.06) | 0.1 (± 0.1) |
| Beryllium | 0.007 (± 0.003) | 0.007 (± 0.003) | 0.007 (± 0.003) |
| Boron | 5.7 (± 1.8) | 7.6 (± 2.5) | 7.6 (± 2.5) |
| Cadmium | 2.1 (± 1.2) | 0.07 (± 0.05) | 0.04 (± 0.11) |
| Calcium | 111 (± 30) | 200 (± 400) | 70 (± 100) |
| Chromium | 0.01 (± 0.04) | 0.14 (± 0.06) | 0.12 (± 0.05) |
| Cobalt | 0.01 (± 0.01) | 0.012 (± 0.006) | 0.010 (± 0.004) |
| Copper | 110 (± 42) | 5.4 (± 1.8) | 1.0 (± 0.9) |

| Metal | Mean ± SD µg/g | | |
|---|---|---|---|
| | Liver | Muscles | Fat |
| Hg | 46 (± 42) | 0.6 (± 0.4) | 6 (± 24) |
| MeHg | 2.4 (± 2.5) | 0.4 (± 0.3) | 0.03 (± 0.03) |
| Iron | 290 (± 190) | 130 (± 24) | 10.1 (± 7.2) |
| Lead | 0.4 (± 0.5) | 0.5 (± 0.4) | 26* (± 40) |
| Magnesium | 513 (± 52) | 850 (± 120) | 39 (± 29) |
| Manganese | 10.4 (± 2.1) | 0.6 (± 0.3) | 0.2 (± 0.1) |
| Molybdenum | 1.4 (± 0.3) | 0.06 (± 0.03) | 0.05 (± 0.02) |
| Nickel | 0.1 (± 0.1) | 0.12 (± 0.06) | 0.12 (± 0.06) |
| Phosphorus | 9000 (± 1000) | 8090 (± 970) | 420 (± 250) |
| Potassium | 7030 (± 750) | 12200 (± 1600) | 490 (± 340) |
| Selenium | 19 (± 30) | 1.6 (± 0.5) | 0.2 (± 0.1) |

**Table S6 Following**

| Metal | Mean ± SD µg/g | | |
|---|---|---|---|
| | **Liver** | **Muscles** | **Fat** |
| Silver | 0.5 (± 0.4) | 0.02 (± 0.01) | 0.02 (± 0.01) |
| Sodium | 2240 (± 540) | 2160 (± 550) | 800 (± 520) |
| Strontium | 0.4 (± 0.3) | 0.6 (± 0.5) | 0.3 (± 0.2) |
| Sulfur | 6570 (± 660) | 8010 (± 910) | 580 (± 350) |
| Thallium | 0.003 (± 0.001) | 0.02 (± 0.01) | 0.006 (± 0.003) |
| Tin | 0.10 (± 0.04) | 0.2 (± 0.1) | 0.10 (± 0.04) |
| Titanium | 0.3 (± 0.1) | 0.3 (± 0.1) | 0.2 (± 0.1) |
| Uranium | 0.3 (± 0.1) | 0.003 (± 0.001) | 0.003 (± 0.01) |
| Vanadium | 0.2 (± 0.1) | 0.03 (± 0.02) | 0.03 (± 0.01) |
| Zinc | 171 (± 54) | 176 (± 44) | 5.2 (± 4.8) |

* One individual had an abnormally high level of antimony and lead likely due to the presence of a bullet in its body or through a prey that contained a bullet

**Table S7 Concentrations of PACs (µg/g dry weight) measured in polar bear tissues (n = 49)**

| | Mean ± SD ng/g | | |
|---|---|---|---|
| **Compound** | **Liver** | **Muscles** | **Fat** |
| Naphthalene | 1700 (± 1000) | 630 (± 350) | 1260 (± 740) |
| Biphenyl | 340 (± 230) | 102 (± 97) | 370 (± 180) |
| Fluorene | 460 (± 450) | 72 (± 65) | 150 (± 160) |
| Dibenzothiophene | 980 (± 590) | 59 (± 69) | 83 (± 58) |
| Anthracene/Phenanthrenes | 1000 (± 1000) | 93 (± 95) | 410 (± 490) |
| Pyrene/Fluoranthene | 188 (± 140) | 27 (± 18) | 42 (± 13) |
| Chrysene/Benzo[a]anthracenes | 130 (± 120) | 34 (± 16) | 47 (± 25) |
| Benzo[a]pyrene/benzo(b&k)fluoranthene | 72 (± 71) | 18 (± 10) | 49 (± 58) |
| Perylene | 4.2 (± 3.0) | 4.9 (± 3.2) | 6.2 (± 2.7) |
| Anthanthrene | 73 (± 73) | 3.9 (± 2.2) | 5.5 (± 2.8) |
| Benzo[a]pyrene | 13.9 (± 8.8) | 2.3 (± 3.2) | 17.5 (± 6.7) |

**Table S8 Concentrations of CHLs (ng/g dry weight) measured in polar bear tissues (n = 49)**

| | Mean ± SD ng/g | | |
|---|---|---|---|
| **Compound** | **Liver** | **Muscles** | **Fat** |
| Oxychlordane | 39 (± 35) | 8.9 (± 8.5) | 50 (± 150) |
| Cis-chlordane | 6.1 (± 4.9) | 1.0 (± 0.5) | 3.3 (± 8.9) |
| Trans-Chlordane | 2.0 (±1.5) | 1.3 (± 1.6) | 2.9 (± 5.0) |
| Heptachlor | 8.5 (± 6.1) | 14 (± 20) | 22 (± 64) |
| Cis-nonachlor | 1.3 (± 0.2) | 1.3 (± 0.4) | 4.6 (± 18) |
| Trans-nonachlor | 2.1 (± 3.4) | 0.9 (± 1.1) | 8 (± 35) |

**Table S9 Concentrations of PCBs (ng/g dry weight) measured in polar bear tissues (n = 24)**

| | Mean ± SD ng/g | | | | Mean ± SD ng/g | | |
|---|---|---|---|---|---|---|---|
| **Compound** | **Liver** | **Muscle** | **Fat** | **Compound** | **Liver** | **Muscle** | **Fat** |
| PCB 47+48+65+75 | 1.9 (± 1.0) | 0.9 (± 1.2) | 7.5 (± 5.6) | PCB 163+164 | 84 (± 48) | 9 (± 16) | 54 (± 29) |
| PCB 99 | 140 (± 120) | 48 (± 74) | 200 (± 200) | PCB 170 | 150 (± 120) | 38 (± 50) | 320 (± 260) |
| PCB 118 | 9.8 (± 9.2) | 3.5 (± 3.7) | 38 (± 33) | PCB 180 | 550 (± 520) | 120 (± 160) | 990 (± 850) |
| PCB 105+127 | 3.6 (± 3.0) | 1.1 (± 1.2) | 6.0 (± 5.6) | PCB 183 | 13 (± 12) | 1.9 (± 2.5) | 17 (± 13) |
| PCB 137 | 11.7 (± 9.9) | 3.3 (± 4.3) | 23 (± 14) | PCB 190 | 18 (± 21) | 6 (± 12) | 46 (± 40) |
| PCB 138 | 160 (± 100) | 26 (± 30) | 240 (± 140) | PCB 193 | 11.8 (± 6.9) | 1.4 (± 1.7) | 16 (± 12) |
| PCB 146 | 10.4 (± 8.7) | 2.6 (± 3.1) | 25 (± 15) | PCB 194 | 65 (± 73) | 15 (± 26) | 160 (± 190) |
| PCB 153 | 1000 (± 580) | 290 (± 350) | 2300 (± 1600) | PCB 196 | 3.2 (± 4.9) | 17 (± 36) | 170 (± 200) |
| PCB 156 | 14 (± 11) | 4.4 (± 5.6) | 45 (± 31) | PCB 203 | 5.1 (± 7.4) | 0.5 (± 0.9) | 4.0 (± 5.4) |
| PCB 157 | 8.4 (± 10.5) | 2.3 (± 2.7) | 23 (± 19) | PCB 205 | 2.2 (± 3.1) | 1.2 (± 2.0) | 10 (± 16) |

**Table S10 Loading values (eighenvalues) in metals. Highest values in both positive and negative axis have been bolded**

| Metal | Liver Principal Components | | | | Muscle Principal Components | | | |
|---|---|---|---|---|---|---|---|---|
| | **PC1** | **PC2** | **PC3** | **PC4** | **PC1** | **PC2** | **PC3** | **PC4** |
| | **36.6%** | **13.4%** | **8.9%** | **6.6%** | **22.9%** | **14.5%** | **9.1%** | **8.0%** |
| Aluminium | 0.13 | -0.20 | **0.31** | -0.02 | 0.04 | -0.11 | **0.09** | -0.07 |
| Antimony | 0.30 | 0.12 | -0.14 | -0.06 | **0.33** | -0.03 | **0.09** | -0.14 |
| Arsenic | **-0.16** | **0.26** | -0.08 | 0.15 | 0.004 | **0.23** | **-0.36** | 0.18 |
| Barium | 0.09 | -0.05 | -0.17 | **-0.14** | 0.11 | -0.11 | **0.14** | 0.06 |
| Beryllium | **0.33** | 0.13 | -0.07 | -0.04 | **0.34** | -0.09 | 0.07 | -0.11 |
| Boron | 0.32 | 0.14 | 0.08 | -0.01 | 0.18 | 0.08 | -0.32 | 0.12 |
| Cadmium | 0.01 | -0.028 | -0.07 | 0.23 | 0.04 | -0.21 | -0.08 | 0.04 |
| Calcium | 0.15 | 0.03 | **0.33** | 0.05 | -0.03 | -0.09 | -0.10 | **-0.40** |
| Chromium | -0.02 | **0.15** | -0.10 | -0.01 | 0.03 | **0.15** | **-0.41** | 0.09 |
| Cobalt | 0.04 | **-0.29** | 0.11 | 0.09 | 0.13 | -0.06 | -0.24 | 0.01 |
| Copper | 0.05 | -0.19 | -0.21 | **0.37** | 0.02 | **-0.38** | -0.04 | 0,12 |

**Table S10 Following**

| Metal | Liver Principal Components | | | | Muscle Principal Components | | | |
|---|---|---|---|---|---|---|---|---|
| | **PC1** | **PC2** | **PC3** | **PC4** | **PC1** | **PC2** | **PC3** | **PC4** |
| | **36.6%** | **13.4%** | **8.9%** | **6.6%** | **22.9%** | **14.5%** | **9.1%** | **8.0%** |
| Silver | -0.03 | -0.02 | -0.07 | **0.42** | 0.19 | -0.25 | 0.05 | 0.17 |
| Sodium | 0.09 | -0.02 | 0.21 | 0.017 | -0.04 | 0.02 | -0.35 | -0.06 |
| Strontium | 0.17 | -0.01 | **0.30** | 0.18 | 0.04 | 0.06 | **-0.38** | -0.19 |
| Sulfur | 0.07 | **-0.37** | -0.14 | -0.03 | **-0.28** | -0.22 | -0.04 | -0.10 |
| Thallium | **0.32** | **0.14** | 0.09 | -0.01 | 0.08 | **-0.30** | -0.07 | **0.32** |
| Tin | **0.32** | **0.14** | 0.09 | -0.01 | 0.15 | **-0.31** | -0.09 | **0.26** |
| Titanium | 0.08 | -0.16 | 0.02 | **-0.26** | 0.05 | 0.01 | -0.16 | 0.19 |
| Uranium | **0.32** | **0.14** | 0.09 | -0.01 | 0.21 | -0.21 | -0.01 | **-0.25** |
| Vanadium | 0.11 | 0.07 | 0.06 | **0.45** | **0.29** | 0.01 | -0.12 | -0.16 |
| Zinc | 0.07 | -0.12 | -0.31 | 0.12 | -0.09 | -0.20 | 0.05 | -0.21 |

**Table S10 Following**

| Metal | Fat | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | Principal Components | | | | | | | | |
| | **PC1 28.8%** | **PC2 14.9%** | **PC3 10.0%** | **PC4 8.1%** | | **PC1 28.8%** | **PC2 14.9%** | **PC3 10.0%** | **PC4 8.1%** |
| Aluminium | 0.16 | 0.20 | -0.20 | 0.03 | Silver | 0.19 | 0.05 | -0.16 | -0.20 |
| Antimony | 0.06 | -0.01 | -0.10 | **-0.63** | Sodium | 0.10 | **-0.32** | -0.10 | 0.003 |
| Arsenic | -0.04 | 0.09 | 0.17 | -0.13 | Strontium | 0.15 | -0.04 | **-0.21** | **0.10** |
| Barium | 0.10 | -0.01 | -0.14 | 0.14 | Sulfur | 0.19 | **-0.33** | -0.04 | -0.01 |
| Beryllium | **0.24** | 0.14 | **-0.21** | -0.02 | Thallium | 0.06 | **0.24** | -0.06 | 0.03 |
| Boron | 0.13 | 0.13 | 0.15 | **-0.22** | Tin | 0.17 | **0.27** | **-0.21** | 0.06 |
| Cadmium | 0.10 | 0.003 | **0.48** | 0.03 | Titanium | 0.21 | 0.14 | 0.08 | 0.05 |
| Calcium | 0.06 | -0.22 | -0.13 | **0.12** | Uranium | 0.21 | **0.27** | -0.20 | 0.01 |
| Chromium | 0.05 | -0.09 | **0.30** | -0.02 | Vanadium | **0.25** | 0.11 | -0.09 | -0.03 |
| Cobalt | **0.24** | 0.19 | 0.09 | 0.06 | Zinc | **0.24** | -0.17 | -0.08 | -0.03 |

**Table S11 Loading values (eighenvalues) in PACs. Highest values in both positive and negative axis have been bolded.**

| PACs | Liver Principal Components | | | | Muscle Principal Components | | | |
|---|---|---|---|---|---|---|---|---|
| | **PC1** | **PC2** | **PC3** | **PC4** | **PC1** | **PC2** | **PC3** | **PC4** |
| | **45.1%** | **23.7%** | **11.5%** | **6.7%** | **37.8%** | **30.6%** | **9.1%** | **7.8%** |
| Naphthalene | -0.25 | 0.09 | **0.55** | **0.39** | 0.38 | **0.15** | 0.14 | **-0.30** |
| Biphenyl | 0.19 | **0.35** | 0.25 | **0.61** | -0.07 | -0.37 | **0.56** | **-0.37** |
| Fluorene | **0.26** | **-0.24** | **0.51** | **-0.43** | **-0.13** | **-0.41** | -0.06 | **0.24** |
| Dibenzothiophene | **0.37** | 0.08 | **0.41** | -0.05 | 0.37 | **0.25** | 0.24 | 0.17 |
| Anthracene/Phenanthrenes | **0.21** | **0.36** | **-0.21** | -0.03 | 0.02 | -0.31 | 0.21 | **0.73** |
| Pyrene/Fluoranthene | -0.32 | **0.35** | 0.20 | **-0.23** | **0.44** | -0.02 | **-0.20** | 0.06 |
| Chrysene/Benzo[a]anthracenes | **-0.37** | -0.02 | 0.33 | **-0.30** | **0.42** | -0.08 | 0.01 | **0.24** |
| Benzo[a]pyrene/benzo(b&k)fluoranthene | -0.30 | **-0.40** | 0.00 | **0.24** | **0.42** | **0.18** | **0.26** | 0.06 |
| Perylene | **-0.34** | 0.28 | -0.10 | -0.16 | 0.23 | **-0.44** | **-0.17** | **-0.27** |
| Anthanthrene | -0.31 | **-0.39** | 0.00 | 0.23 | 0.17 | **-0.44** | **0.30** | -0.12 |
| Benzo[a]pyrene | **-0.33** | **0.40** | -0.01 | -0.08 | 0.26 | -0.31 | **-0.57** | -0.10 |

**Table S11 Following**

| PACs | Fat Principal Components | | | |
|---|---|---|---|---|
| | **PC1 25.8%** | **PC2 25.0%** | **PC3 18.5%** | **PC4 9.9%** |
| Naphthalene | **0.24** | **0.28** | -0.16 | **0.58** |
| Biphenyl | 0.02 | **0.48** | -0.32 | 0.24 |
| Fluorene | -0.004 | **-0.37** | -0.19 | **0.47** |
| Dibenzothiophene | 0.04 | **-0.41** | **-0.40** | 0.00 |
| Anthracene/Phenanthrenes | **0.19** | **-0.42** | -0.38 | 0.09 |
| Pyrene/Fluoranthene | -0.27 | -0.13 | -0.44 | **-0.33** |
| Chrysene/Benzo[a]anthracenes | -0.35 | **0.30** | **-0.38** | 0.11 |
| Benzo[a]pyrene/benzo(b&k)fluoranthene | -0.15 | **0.28** | **-0.39** | **-0.33** |
| Perylene | **-0.51** | -0.05 | **0.20** | 0.08 |
| Anthanthrene | **-0.51** | -0.11 | 0.09 | **0.34** |
| Benzo[a]pyrene | **-0.41** | -0.12 | 0.08 | 0.13 |

**Table S12 Loading values (eighenvalues) in CHLs. Highest values in both positive and negative axis have been bolded.**

| Chlordanes | Liver Principal Components | | | | Muscle Principal Components | | | |
|---|---|---|---|---|---|---|---|---|
| | PC1 | PC2 | PC3 | PC4 | PC1 | PC2 | PC3 | PC4 |
| | 84.3% | 8.3% | 3.6% | 2.6% | 93.6% | 3.7% | 1.7% | 0.7% |
| Oxychlordane | **0.41** | **-0.36** | **0.36** | **-0.65** | 0.39 | **0.67** | **-0.62** | 0.0002 |
| Cis-chlordane | 0.40 | **-0.15** | **-0.83** | **-0.24** | 0.39 | **-0.68** | **-0.49** | **-0.37** |
| Trans-chlordane | 0.38 | **-0.62** | **0.22** | **0.58** | **0.42** | -0.06 | **0.30** | 0.16 |
| Heptachlor | 0.40 | **0.50** | **0.35** | **-0.17** | 0.41 | **0.25** | **0.50** | **-0.68** |
| Cis-nonachlor | **0.43** | **0.14** | -0.03 | **0.13** | **0.42** | -0.09 | 0.12 | **0.45** |
| Trans-nonachlor | **0.42** | **0.44** | -0.07 | **0.36** | **0.42** | -0.09 | 0.14 | **0.41** |

**Table S12 Following**

| Chlordanes | Fat Principal Components | | | |
|---|---|---|---|---|
| | **PC1** | **PC2** | **PC3** | **PC4** |
| | **79.9%** | **7.9%** | **6.9%** | **4.5%** |
| Oxychlordane | **0.42** | **0.38** | **-0.38** | 0.11 |
| Cis-chlordane | 0.37 | **0.35** | **0.72** | **0.44** |
| Trans-chlordane | 0.40 | 0.12 | **0.22** | **-0.86** |
| Heptachlor | **0.42** | 0.23 | **-0.53** | 0.16 |
| Cis-nonachlor | **0.44** | **-0.31** | 0.06 | 0.01 |
| Trans-nonachlor | 0.39 | **-0.75** | -0.003 | **0.17** |

**Table S13 Loading values (eighenvalues) in PCBs. Highest values in both positive and negative axis have been bolded.**

| PCB | Liver Principal Components | | | | Muscle Principal Components | | | |
|---|---|---|---|---|---|---|---|---|
| | **PC1 52.2%** | **PC2 15.1%** | **PC3 10.9%** | **PC4 7.7%** | **PC1 66.2%** | **PC2 20.2%** | **PC3 5.2%** | **PC4 3.4%** |
| PCB 47+48+65+75 | -0.19 | -0.20 | **-0.25** | 0.05 | **0.26** | -0.07 | -0.13 | **0.21** |
| PCB 99 | -0.28 | 0.13 | -0.004 | -0.03 | 0.25 | -0.17 | -0.04 | -0.05 |
| PCB 118 | 0.02 | -0.23 | 0.10 | **-0.68** | 0.11 | **0.34** | **-0.47** | **-0.42** |
| PCB 105+127 | -0.01 | **-0.41** | -0.06 | **-0.27** | 0.12 | **0.38** | **-0.26** | **-0.43** |
| PCB 137 | **-0.29** | 0.05 | -0.03 | 0.07 | **0.26** | 0.12 | -0.04 | 0.12 |
| PCB 138 | -0.28 | 0.00 | 0.23 | 0.00 | **0.27** | -0.03 | -0.16 | 0.02 |
| PCB 146 | -0.17 | **-0.45** | 0.02 | -0.01 | 0.19 | **0.31** | **-0.18** | 0.16 |
| PCB 153 | -0.26 | -0.19 | 0.08 | 0.07 | **0.26** | -0.14 | -0.05 | -0.005 |
| PCB 156 | **-0.29** | 0.10 | -0.04 | -0.15 | 0.24 | 0.20 | -0.01 | **0.30** |
| PCB 157 | -0.26 | **0.23** | **-0.20** | -0.17 | **0.26** | 0.05 | -0.11 | **0.29** |

**Table S13 Following**

| PCB | Fat Principal Components | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | **PC1 52.2%** | **PC2 15.1%** | **PC3 10.9%** | **PC4 7.7%** | | **PC1 66.2%** | **PC2 20.2%** | **PC3 5.2%** | **PC4 3.4%** |
| PCB 47+48+65+75 | **0.26** | -0.05 | -0.02 | **0.27** | PCB 163+164 | 0.25 | -0.18 | 0.03 | -0.12 |
| PCB 99 | 0.25 | -0.18 | -0.05 | -0.09 | PCB 170 | **0.26** | -0.11 | 0.07 | -0.01 |
| PCB 118 | 0.11 | **0.35** | **-0.47** | **-0.38** | PCB 180 | 0.25 | -0.18 | -0.07 | 0.03 |
| PCB 105+127 | 0.12 | **0.39** | **-0.25** | **-0.39** | PCB 183 | 0.24 | 0.19 | 0.03 | 0.18 |
| PCB 137 | **0.26** | 0.11 | -0.04 | 0.08 | PCB 190 | 0.23 | **-0.26** | -0.04 | -0.12 |
| PCB 138 | **0.27** | -0.03 | **-0.17** | -0.01 | PCB 193 | 0.14 | **-0.31** | 0.11 | **-0.58** |
| PCB 146 | 0.19 | **0.31** | **-0.17** | 0.10 | PCB 194 | 0.16 | **0.31** | **0.47** | -0.10 |
| PCB 153 | **0.26** | -0.15 | -0.06 | -0.05 | PCB 196 | 0.23 | **-0.23** | -0.13 | 0.09 |
| PCB 156 | 0.24 | 0.19 | -0.02 | **0.29** | PCB 203 | 0.17 | 0.30 | **0.45** | -0.11 |
| PCB 157 | **0.26** | 0.04 | -0.12 | **0.28** | PCB 205 | 0.24 | 0.02 | **0.41** | -0.11 |