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ENVIRONMENTAL CONTAMINANTS: PERSISTENT ORGANIC POLLUTANTS AND CONTAMINANTS OF EMERGING ARCTIC CONCERN

QANUILIRPITAA? 2017

Nunavik Inuit Health Survey

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RÉGIE RÉGIONALE DE LA SANTÉ ET DES SERVICES SOCIAUX DU NUNAVIK
NUNAVIK REGIONAL BOARD OF HEALTH AND SOCIAL SERVICES

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QANUILIRPITAA? 2017 HEALTH SURVEY

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In memory of Audrey Flemming and Linda Shipaluk

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LIST OF ACRONYMS

AFFFs	aqueous film forming foams
CHMS	Canadian Health Measures Survey
CI	confidence interval
CTQ	Centre de toxicologie du Québec
CV	coefficient of variation
DHA	docosahexaenoic acid
DPA	docosapentaenoic acid
EPA	eicosapentaenoic acid
FFQ	food frequency questionnaire
FTOHs	fluorotelomer alcohols
GM	geometric mean
IHS	Inuit Health Survey
INSPQ	Institut national de santé publique du Québec
IUPAC	International Union of Pure and Applied Chemistry
LOD	limits of detection
MRM	multiple reaction monitoring
n-3 PUFA	long-chain omega-3 polyunsaturated fatty acids
NQN	*Nutaratsaliit Qanuingisiarningit Niqituinnanut* – Pregnancy Wellness with Country Foods project
NRBHSS	Nunavik Regional Board of Health and Social Services
PBDEs	polybrominated diphenyl ethers
PCBs	polychlorinated biphenyls
PFAAs	perfluoroalkyl acids
PFASs	per- and polyfluoroalkyl substances
PFBA	perfluoro-n-butanoic acid
PFBS	perfluorobutane sulfonic acid
PFDA	perfluoro-n-decanoic acid
PFHxA	perfluoro-n-hexanoic acid
PFHxS	perfluorohexane sulfonic acid
PFNA	perfluoro-n-nonanoic acid

PFOA	perfluoro-n-octanoic acid
PFOS	perfluorooctanesulfonic acid
PFUdA	perfluoro-n-undecanoic acid
POPs	persistent organic pollutants
PPR	people per room
RBC	red blood cell
UNECE	United Nations Economic Commission for Europe
UNEP	United Nations Environment Programme
β-HCH	β-hexachlorocyclohexane
γ-HCH	γ-hexachlorocyclohexane
***p,p'*-DDE**	*p,p'*-dichlorodiphenyldichloroethylene
***p,p'*-DDT**	*p,p'*-dichlorodiphenyltrichloroethane

1 BACKGROUND OF THE *QANUILIRPITAA?* 2017 HEALTH SURVEY

The *Qanuilirpitaa?* 2017 Health Survey is a major population health survey conducted in Nunavik that involved the collection, analysis and dissemination of information on the health status of Nunavimmiut. The last health survey conducted prior to it in Nunavik dated from 2004. Since then, no other surveys providing updated information on the health of this population had been carried out. Thus, in February 2014, the Board of Directors of the Nunavik Regional Board of Health and Social Services (NRBHSS) unanimously adopted a resolution to conduct a new health survey in all 14 Nunavik communities, in support of the Strategic Regional Plan.

The general objective of the 2017 health survey was to provide an up-to-date portrait of the health status of Nunavimmiut. It was also aimed at assessing trends and following up on the health and health determinants of adult participants since 2004, as well as evaluating the health status of Nunavik youth. This health survey has strived to move beyond traditional survey approaches so as to nurture the research capabilities and skills of Inuit and support the development and empowerment of communities.

Qanuilirpitaa? 2017 included four different components: 1) an adult component to document the mental and physical health status of adults in 2017 and to follow up on the adult cohort of 2004; 2) a youth component to establish a new cohort of Nunavimmiut aged 16 to 30 years old and to document their mental and physical health status; 3) a community component to establish the health profiles and assets of communities in a participatory research approach; and 4) a community mobilization project aimed at mobilizing communities and fostering their development.

This health survey relied on a high degree of partnership within Nunavik (Nunavik Regional Board of Health and Social Services (NRBHSS), Makivik Corporation, Kativik Regional Government (KRG), Kativik Ilisarniliriniq (KI), Avataq Cultural Institute, Qarjuit Youth Council, Inuulitsivik Health Centre, Ungava Tulattavik Health Centre), as well as between Nunavik, the Institut national de santé publique du Québec (INSPQ) and academic researchers from three Canadian universities: Université Laval, McGill University and Trent University. This approach followed the OCAP principles of Ownership, Control, Access and Possession (First Nations Information Governance Centre, 2007)[1]. It also emphasized the following values and principles: empowerment and self-determination, respect, value, relevance and usefulness, trust, transparency, engagement, scientific rigour and a realistic approach.

TARGET POPULATION

The survey target population was all permanent Nunavik residents aged 16 years and over. Persons living full time in public institutions were not included in the survey. The most up-to-date beneficiaries register of all Inuit living in Nunavik, provided by the Makivik Corporation in spring 2017, was used to construct the main survey frame. According to this register, the population of Nunavik was 12 488 inhabitants spread out in 14 communities. The register allowed respondents to be selected on the basis of age, sex and coast of residence (Hudson coast and Ungava coast).

SURVEY FRAME

The survey used a stratified proportional model to select respondents. Stratification was conducted based on communities and age groups, given that one of the main objectives of the survey was to provide estimates for two subpopulations aged, respectively, 16 to 30 years and 31 years and over. In order to obtain precise estimates, the targeted sample size was 1 000 respondents in each age group. Assuming a 50% response rate, nearly 4 000 people were required to obtain the necessary sample size. From this pool, the number of individuals recruited from each community was proportionate to population size and took into account the number of days that the survey team

1. OCAP® is a registered trademark of the First Nations Information Governance Centre (FNIGC).

would remain in each community – a situation that imposed constraints on the number of participants that could be seen. Within each stratum, participants were randomly selected from the beneficiaries register. However, the individuals from the 2004 cohort, all 31 years old and over (representing approximately 700 individuals), were automatically included in the initial sample.

DATA COLLECTION

Data were collected from August 19, 2017 to October 5, 2017 in the 14 villages. The villages were reached by the *Amundsen*, a Canadian Coast Guard Icebreaker, and participants were invited on board the ship for data collection purposes.

Two recruitment teams travelled from one community to another before the ship's arrival. An Inuk assistant in each community helped: identify, contact and transport (if necessary) each participant; inform participants about the sampling and study procedures; obtain informed consent from participants (video) and fill in the identification sheet and sociodemographic questionnaire.

Data collection procedures for the survey included questionnaires, as well as clinical measurements. The survey duration was about four hours for each wave of participants, including their transportation to and from the ship. Unfortunately, this time frame was sometimes insufficient to complete the data collection process. This survey received ethical approval by the Comité d'éthique de la recherche du Centre Hospitalier Universitaire de Québec – Université Laval.

Aboard the ship, the survey questionnaires were administered by interviewers, many of whom were Inuit. Face-to-face interviews were conducted using a computer-assisted interviewing tool. If there were problems with the laptop connections, paper-form questionnaires were filled out. The questionnaires were administered in Inuktitut, English or French, according to the preference of the participants. Interviewers received training in administering the questionnaires prior to the start of the survey. The questionnaires were divided into five blocks: psychosocial interview (blocks 1 and 3), physical health and food security interview (block 2), food frequency questionnaire (block 4), and sociodemographic interview (block 5).

The survey also included a clinical component, with tests to document aspects of physical health, sampling of biological specimens (such as blood, oropharyngeal swabs, urine, stool, and vaginal swabs), spirometry, and an oral clinical exam. These sessions were supervised by a team comprised of nurses, respiratory therapists, dentists, dental hygienists and assistants, and laboratory technicians.

PARTICIPATION

There were a total of 1 326 participants, including 574 Nunavimmiut aged 16 to 30 years old and 752 Nunavimmiut aged 31 years and over, for total response rates of 30.7% and 41.5%, respectively. The participants' distribution between the two coasts (Ungava and Hudson) was similar. The distribution of men and women was unequal, with twice as many women (873) than men (453) participating in the survey. If the results obtained from this sample are to be inferred to the target population, survey weights must be used.

Overall, as compared to the 2004 survey, the response rate (i.e., the rate of participants over the total number of individuals on the sampling list) was lower than expected, especially among young people. This includes the refusal rate and especially a low contact rate. Several reasons might explain the low response rate, including the short time period available to contact individuals prior to the ship's arrival in the community and non-contact due to people being outside of the community or on the land. Nevertheless, among the individuals that were contacted (n = 1 661), the participation rate was satisfactory with an internal participation rate of 79.7% More details on the collection, processing and analysis of the data are given in the Methodological Report (Hamel, Hamel & Gagnon, 2020).

2 INTRODUCTION

The Inuit of Nunavik are exposed to persistent organic pollutants (POPs) that are carried from southern to northern latitudes by oceanic and atmospheric transport and are then biomagnified in Arctic food webs (AMAP, 2021). POPs are a group of synthetic industrial compounds highly resistant to metabolic degradation, making them bioaccumulative in the environment. After these stable chemicals travel North via wind and water currents, the low temperatures in the Arctic enhance the deposition of POPs in a process called "cold condensation" (Ottar, 1981) and slow down their degradation (Fliedner et al., 2012). These unique Arctic geoclimatic characteristics create a sink for POPs which can be remobilized with climate change and glacial melt (Ma et al., 2016; Pawlak et al., 2021). Additionally, POPs are biomagnified in Arctic food webs after redistribution in the environment, particularly in marine food webs. Since the late 1970s, high concentrations of POPs have been identified in certain fish, marine mammals, marine birds, and other organisms in Arctic regions, with especially elevated levels in predatory species (Braune et al., 2005; Letcher et al., 2010; Simond et al., 2017). Similarly, elevated blood POP concentrations have been found in Indigenous peoples of the Arctic who rely on these species for subsistence (Government of Canada, 2017).

Legacy POPs include the dirty dozen chemicals referred to in the Stockholm Convention, among which are polychlorinated biphenyls (PCBs) and organochlorinated pesticides. PCBs are a group of organochlorine compounds that were used in electrical apparatus for dielectric and coolant purposes. Chlorinated pesticides (dichlorodiphenyltrichloroethane (DDT), toxaphene, chlordane, etc.) are very efficient pesticides that were once used to control agricultural pests. Another group of POPs includes polybrominated diphenyl ethers (PBDEs), which are brominated organic compounds used in a variety of household items to increase products' flame ignition resistance. Due to their lipophilic properties, chlorinated pesticides, PCBs, and PBDEs accumulate in fatty tissues (Dudarev et al., 2019). This, along with their biomagnification properties, results in high concentrations of these POPs in predatory animals, particularly marine mammals (Dudarev et al., 2019; Walker et al., 2020). Per- and polyfluoroalkyl substances (PFASs) are a group of chemicals that are part of POPs and that represent a large family of synthetic compounds with various industrial and commercial applications. They are used for their ability to repel both oil and water (Buck et al., 2011). Perfluoroalkyl acids (PFAAs) are PFAS congeners that are exceptionally stable and highly mobile in the environment, and thus lead to elevated concentrations in Arctic air, snow, soil, water and sediment samples (MacInnis et al., 2017; Wong et al., 2018). Contrary to other POPs that are lipophilic, PFAAs accumulate in proteins, and have also been measured in many wildlife species consumed by Inuit populations living in the Arctic, such as marine mammals, fish, caribou and other species (AMAP, 2021; Muir et al., 2019; Ostertag et al., 2009).

In the 1990s and 2000s, international and national efforts were mobilized to reduce or ban many legacy POPs through domestic initiatives of chemical control and international treaties such as the United Nations Environment Programme (UNEP) Stockholm Convention on Persistent Organic Pollutants in 2004 and the United Nations Economic Commission for Europe (UNECE) POPs Protocol of the Convention on Long-range Transboundary Air Pollution in 1983 (Hillman, 1999; Magulova & Priceputu, 2016). Furthermore, perfluorooctanesulfonic acid (PFOS), the oldest of the PFAAs, has been banned under the Stockholm Convention since 2009, while perfluorooctanoic acid (PFOA) was included in the Convention only recently (May 2019) and perfluorohexanesulphonic acid (PFHxS) is still under review for its inclusion (UN Environment Programme 2019). The use of all these PFAAs has been restricted in the production of consumer goods in North America, with a few exceptions such as firefighting aqueous film forming foams (AFFFs) (Government of Canada, 2018; Houde et al., 2006; Muir et al., 2019; Paul et al., 2009). However, many unregulated imported products may contain long-chain PFAA congeners such as perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA) and perfluoroundecanoic acid (PFUdA), which, like PFOS, are very persistent in the environment (Longpré et al., 2020). PFOA and long-chain PFAAs are also degradation products of other more neutral PFASs such as fluorotelomer alcohols (FTOHs), which are still globally used in many consumer industrial products (e.g., paints, electronics, food packaging, waxes) (Dinglasan et al., 2004) and cosmetics (Whitehead et al., 2021). These long-chain PFAAs are considered chemicals of emerging Arctic concern.

It is important to note that although many legacy POP concentrations are decreasing in northern communities, there are still elevated levels of several legacy POP compounds in the Arctic environment and wildlife that could elicit downstream health impacts (AMAP, 2021; Government of Canada, 2017). For example, in 2016-2017, approximately 10% of Nunavik pregnant women who participated in the Nutaratsaliit Qanuingisiarningit Niqituinnanut—Pregnancy Wellness with Country Foods (NQN) project still had blood PCB concentrations above the critical concentration threshold proposed by the French Food Safety Agency based on sexual maturation in male rats (Adamou et al., 2020; AFSSA, 2010). Furthermore, increasing exposure to long-chain PFAAs in a sample of Nunavik pregnant women (NQN participants) was observed between 2016-2017 since the first time they were measured in 2011. The presence of PFAAs in the Arctic is most likely due to the environmental degradation of FTOHs (*AMAP Assessment 2016*, 2017). In 2016-2017, the overall concentration of PFAAs was twice as high in Nunavik pregnant women or the NQN project than among the general Canadian population (Caron-Beaudoin et al., 2020).

The half-life of many POPs in the human body is in the order of several years (Bu et al., 2015); therefore, blood concentrations tend to be reflective of lifetime exposures. Older individuals have been exposed to POP concentrations for a longer period (and thus, will have a greater body burden), and were also exposed to higher POP concentrations in their youth given that the environmental concentrations of many POPs have decreased in the last couple of decades (*AMAP Assessment 2016*, 2017). Indeed, the Santé Quebec Survey in 1992 and the *Qanuippitaa?* Nunavik Inuit Health Survey in 2004 (*Qanuippitaa? 2004*) documented greater concentrations of PFOS, PCBs, and PBDEs among older individuals (Dewailly et al., 2007). In 2004, differences in POP concentrations by gender varied by chemical. Hexachlorobenzene and toxaphenes levels were higher in women, whereas mirex and PFOS were higher in men (Dewailly et al., 2007).

As the Inuit traditional diet comprises large amounts of wild animals, Inuit face higher exposure to POPs compared to populations living in southern regions (AMAP, 2021). Indeed, coastal communities, and particularly Nunavik and Nunavut, are more highly exposed to POPs because of their consumption of marine species, notably marine mammals such as beluga and seal. This is in contrast to non-coastal communities, like those of the Northwest Territories, who consume more land mammals, such as caribou, and present lower exposure to POPs (Butler Walker et al., 2003; Curren et al., 2014; S. G. Donaldson et al., 2010; Muckle et al., 2001). Likewise, findings from the 2004 survey show higher concentrations of PCBs in Nunavimmiut living on the Hudson coast versus the Ungava coast, primarily due to the higher consumption of marine mammals and fish. In 2004, PFOS and PBDE-153 were also positively associated with the consumption of marine mammals and/or fish whereas PBDE-47 was not (Dallaire et al., 2009; Dewailly et al., 2007). The 2016-2017 NQN study that included 96 pregnant women in Nunavik explored the exposure determinants of several PFAAs including PFOS and long-chain PFAAs (Caron-Beaudoin et al., 2020). There was an association between omega-3/omega-6 polyunsaturated fatty acids (PUFA) ratios and higher PFAAs concentrations, suggesting that seafood and marine mammal consumption are important exposure sources. However, further studies are required to explore this in greater detail.

Other potential factors that may determine exposure to POPs include household dust and smoking status. Household dust is an important source of PFAAs and PBDEs due to their use in furniture as stain and fire-resistant compounds (Bost et al., 2016; D'Hollander et al., 2010b; Government of Canada, 2017). Thus, overcrowding and need of housing repairs could account for older furniture or increased household dust (Caron-Beaudoin et al., 2020). In another population, smoking was associated with lower concentrations of hexachlorobenzene and some PCBs, and higher concentrations of *cis*-chlordane and *trans*-nonachlor (Batterman et al., 2016). However, no association between smoking and exposure to POPs was reported among Nunavimmiut adults in 2004 (Dallaire, 2009; Dewailly et al., 2007).

In Nunavik, the majority of studies on POPs and health focused on children's health and/or the health impact of PCBs (Singh et al., 2014). Children in Nunavik with higher concentrations of PCBs in cord blood were more likely to have subtle neuromotor and visual recognition memory impairments, as well as symptoms of attention deficit/hyperactivity disorder (Boucher et al., 2012, 2014, 2016; Ethier et al., 2015). Evidence also suggests an association between exposure to PCBs and organochlorines during pregnancy and an increase in acute infections in infants (F. Dallaire et al., 2004, 2006). Similar neurodevelopmental and immune outcomes have been reported in other circumpolar populations highly exposed to POPs from diet (AMAP, 2021). Impacts on thyroid hormones are especially important during pregnancy because the fetus is dependent on the mother's thyroid hormones for brain development. Therefore, a study in Nunavik focused on 120 Inuit women of child-bearing age to assess the impact of POP compounds on the binding of thyroxine (the active thyroid hormone) to its "carrier protein" (transthyretin) across the placenta and brain (Audet-Delage et al., 2013). The study did not identify any associations between POP compounds and the circulating levels of transthyretin-bound thyroid hormones; however, it is possible that POPs could interfere with the binding of thyroxine to other transport proteins.

Exposure to POPs among adults has also been linked to cardiometabolic, thyroid, neurological, respiratory, and reproductive outcomes (Fong-McMaster et al., 2020; González, 2021; Guo et al., 2019; Lind & Lind, 2020). In Nunavik adults, results were a little more mixed. A study of 315 Inuit adults who participated in the 1992 Santé Quebec Survey reported an association between PCBs and *p,p'*-dichlorodiphenyldichloroethylene (*p,p'*-DDE) and increased risk of hypertension; however, *p,p'*-DDT and oxychlordane were associated with a decreased risk of hypertension (Valera et al., 2013). There was also evidence of a decreased thyroid hormone, total triiodothyronine, with exposure to PCBs and chlorinated pesticides, a decrease in the hormone free thyroxine, with hexachlorobenzene, and a decrease in total triiodothyronine and thyroid stimulating hormone and an increase in free thyroxine with PFOS plasmatic levels in *Qanuippitaa? 2004* (Dallaire et al., 2009). Studies regarding metabolic health were a little clearer. In 2004, plasma PFOS was associated with a decrease in triacylglycerol and the ratio of total cholesterol to high density lipoprotein cholesterol (good cholesterol) levels (Château-Degat et al., 2010), indicating changes in lipid profiles and potential cardiometabolic effects from PFOS exposure. Organochlorine exposure in 2004 was also associated with diabetes prevalence. Indeed, participants in the highest tertiles of PCBs and organochlorine pesticides plasma concentrations were respectively four times and two times more likely to have diabetes, compared to the lowest tertiles (Cordier et al., 2020). In the same study, a decrease in fasting insulin levels was observed in non-diabetic individuals with increasing PCBs and several organochlorine pesticides plasma levels. The associations were particularly relevant among those with a waist circumference above the median. This was in line with the results of the Inuit Health Survey 2007-2008 (IHS 2007-2008), which examined the relationship between diabetes and POPs in a study population of 2 595 adult Inuit in the Inuvialuit Settlement Region, Nunavut Territory, and Nunatsiavut and in which the authors reported 2 to 3-fold increased odds of self-reported diabetes with exposure to PCBs and *p,p'*-DDE (Singh & Chan, 2017). No studies with respect to POP exposure and respiratory and reproductive functions have been conducted yet in Nunavik.

The underlying mechanisms linking POPs to disease is thought to occur via endocrine disruption or immunotoxicity (Guo et al., 2019; Hertz-Picciotto et al., 2008). These compounds are able to cross the placental barrier and are also found in human milk (Guerranti et al., 2011; Zhang et al., 2021). The endocrine system is responsible for regulating the chemical messengers (i.e., hormones) that control various body functions. Some POPs can disrupt the endocrine pathways by mimicking hormones or interfering with the signalling process, leading to downstream adverse health effects (Routti et al., 2019; Sinclair et al., 2020; Zhu et al., 2020). Many POPs have also been found to induce insulin resistance and interfere with the metabolism of fats, carbohydrates, and proteins (Baillie-Hamilton, 2002; David O. Carpenter, 2008; Sharp, 2009). Evidence also points to an increase in chronic inflammation with excessive or prolonged exposure to POPs, whereby the natural defence mechanism of inflammation begins attacking nearby healthy body cells due to an excess of reactive oxygen species (i.e., oxidative stress) (Hertz-Picciotto et al., 2008; Mogensen et al., 2015; Zota et al., 2018). This may eventually lead to developmental adverse outcomes and chronic diseases later in life, including cardiometabolic (Ruiz-Castell et al. 2020) and respiratory (Fahy 2015) diseases.

OBJECTIVES

- To document blood levels of POPs among Nunavimmiut aged 18 years and over by age and sex in 2017;
- To compare these results to those of the 1992 Santé Québec Survey and the *Qanuippitaa?* 2004 Nunavik Health Survey, other Inuit populations in Canada, and the general Canadian population;
- To examine associations between potential contemporary determinants of exposure and blood concentrations of the aforementioned compounds among Nunavimmiut.

3 METHODOLOGICAL ASPECTS

STUDY POPULATION

A total of 1 326 individuals participated in the data collection process on board the CCGS *Amundsen*, and among them, 93.9% provided a blood sample. Of these participants, 500 were randomly selected and their plasma analyzed for POPs. The final study sample consisted of 500 participants aged 18 and over, including 6 pregnant women.

DATA COLLECTION AND LABORATORY ANALYSES

Blood sample collection and laboratory analyses

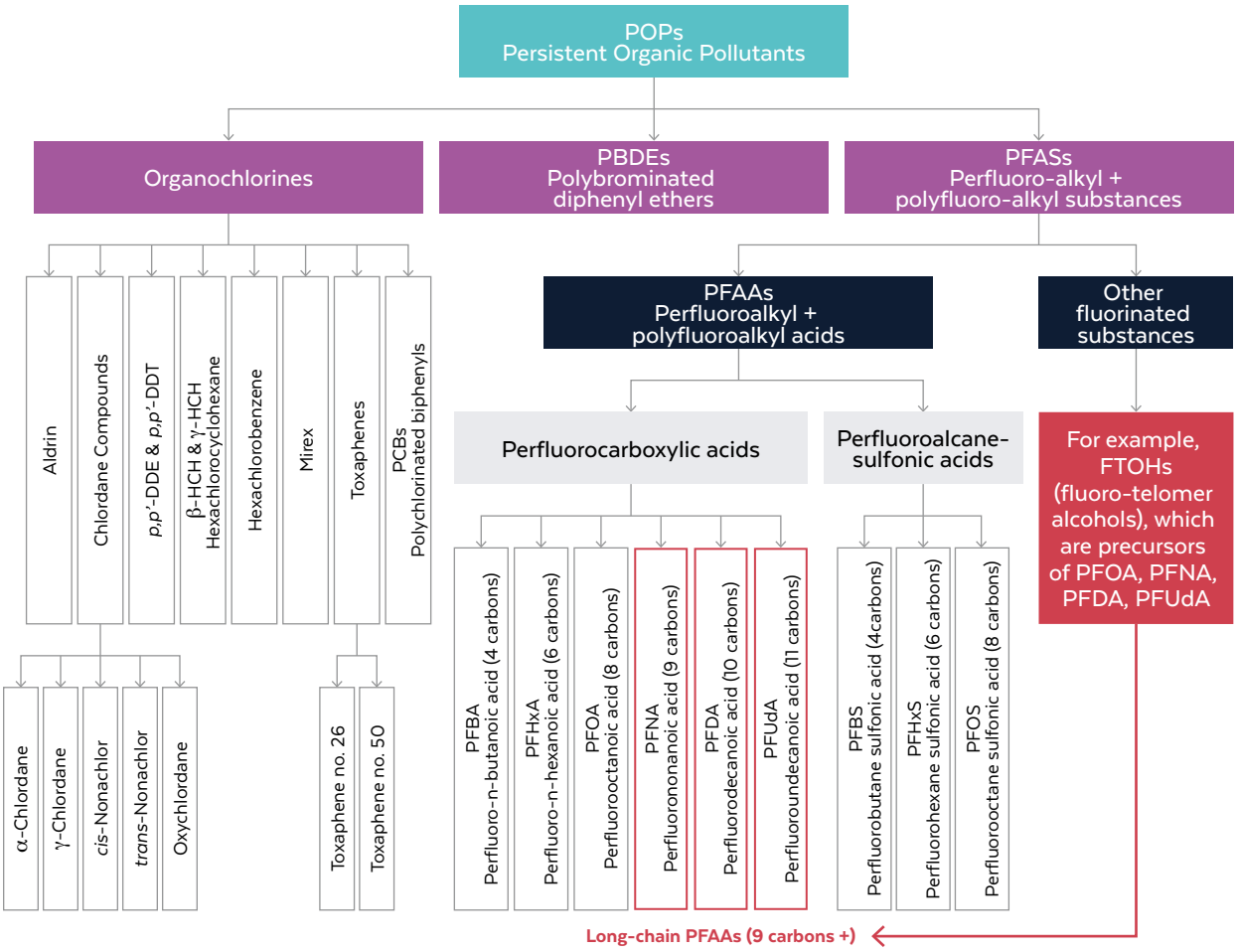
Blood samples were obtained through venipuncture performed by research nurses; blood was collected in K_2-EDTA vacutainers and processed within 90 min by the laboratory staff on board the *Amundsen*. Blood samples were centrifuged at 2000xg during 10 min and the plasma transferred into either a 2-mL polypropylene tube for storage at -20°C until time of analysis (PFASs), or into a 7-mL pre-cleaned glass vial for storage at -20°C until time of analysis (organochlorines, PCBs and PBDEs). A 600-µL red blood cell aliquot (pellet obtained following whole blood centrifugation) was transferred into a 2-mL polypropylene tube for storage at -80°C until time of PUFA analysis.

POPs analyses of Q2017 samples were carried out at the Centre de toxicologie du Québec (CTQ) of the Institut national de santé publique du Québec (INSPQ), which is accredited by the Canadian Association for Environmental Analytical Laboratories and is ISO 17025 accredited. The CTQ is a reference laboratory in interlaboratory comparison programs based on the excellence of analytical results obtained with conventional methods for POPs and metals measurements.

All POPs measured in the blood plasma of participants are shown in Figure 1. Organochlorines, including aldrin, α-chlordane, γ-chlordane, *cis*-nonachlor, *trans*-nonachlor, oxychlordane, *p,p'*-dichlorodiphenyldichloroethylene (*p,p'*-DDE), *p,p'*-dichlorodiphenyltrichloroethane (*p,p'*-DDT), β-hexachlorocyclohexane (β-HCH), γ-hexachlorocyclohexane (γ-HCH), hexachlorobenzene, mirex, toxaphene no. 26, toxaphene no. 50 and 24 congeners of polychlorinated biphenyls (PCB IUPAC #: 28, 52, 66, 74, 99, 101, 105, 118, 128, 138, 146, 153, 156, 163, 167, 170, 178, 180, 183, 187, 194, 201, 203 and 206) and polybrominated compounds (PBDE IUPAC #: 15, 17, 25, 28, 33, 47, 99, 100, 153; polybrominated biphenyl IUPAC # 153), were quantified in plasma samples using the CTQ's method E-446, described in Fisher et al. (2016). Briefly, compounds were extracted using a liquid-liquid extraction with a mixture of ammonium sulfate:denaturated alcohol:hexane (1:1:3). Thereafter, the extracts were concentrated, purified on Florisil and then analyzed by gas chromatography/mass spectrometry (Agilent Technologies) after a negative chemical ionization.

Nine PFASs—perfluoro-n-butanoic acid (PFBA), perfluoro-n-hexanoic acid (PFHxA), perfluoro-n-octanoic acid (PFOA), perfluoro-n-nonanoic acid (PFNA), perfluoro-n-decanoic acid (PFDA), perfluoro-n-undecanoic acid (PFUdA, perfluorobutane sulfonic acid (PFBS), perfluorohexane sulfonic acid (PFHxS) and perfluoro-1-octane sulfonic acid (PFOS)—were quantified in plasma samples using the CTQ's method E-501. After acidifying samples, compounds were extracted using a weak anion exchange solid-phase extraction on a 96-well plate. The extracts were evaporated to dryness, dissolved into the mobile phase and then analyzed by ultra performance liquid chromatography (Waters Acquity) coupled with tandem mass spectrometry (Waters Xevo TQ-S) in the multiple reaction monitoring mode with an electrospray ion source in the negative mode.

Figure 1 Relationship between persistent organic pollutant groups



Red blood cell (RBC) fatty acid composition was analyzed at the Laboratory of Nutritional Lipidomics of the University of Waterloo, Ontario, using a Varian 3900 gas chromatograph equipped with a 15 m DB-FFAP capillary column (df = 0.10 μm). Total RBC eicosapentaenoic acid (EPA), docosahexaenoic acid (DHA) and docosapentaenoic acid (DPA) were summed, expressed as a percent of total fatty acids, and categorized into quartiles (hereafter referred to as RBC long-chain omega-3 polyunsaturated fatty acids (n-3 PUFA)).

Dietary assessment and questionnaires

Dietary intake was assessed using a food frequency questionnaire (FFQ). The FFQ measured the frequency of intake for each item in the past three months, but serving sizes were not considered. Food items were then categorized into different groups: marine mammals, terrestrial animals, wild birds, fish, and seafood. In addition to the FFQ, questionnaires to document sociodemographic characteristics, hunting practices, housing conditions, and smoking status were administered by trained interviewers. Household overcrowding was quantified using the people per room (PPR) index; crowding occurs if there is more than one person per room. A list of variables used for bivariate analysis as well as the questionnaire from which they were obtained are presented in Table 1.

Table 1 List of variables used for bivariate analysis

Source	Variables
PHFSI (block 2)	Hunting frequency
PSI (block 3)	House in need of repairs, smoking status
FFQ (block 4)	Consumption of various country foods (marine mammals, fish, mollusks, seaweed, terrestrial animals and wild birds)
SDI (block 5)	Sex, age, ecological region, overcrowding

PSI = psychosocial interview; PHFSI = physical health and food security interview; FFQ = food frequency questionnaire; SDI = sociodemographic interview.

STATISTICAL ANALYSIS

All statistical analyses were performed with SAS® Studio, Version 3.8. (Cary, North Carolina, USA) and used weights to take into account sampling methodology and item non-response and thereby allow the results to be inferred to the target population (Hamel, Hamel & Gagnon, 2020). For all statistical analyses, a significance level of $\alpha < 0.05$ was used and variance was estimated with the balanced repeated replication method.

Descriptive statistics were presented as geometric means (95% confidence intervals) of chemical concentrations in plasma if the chemical was detected in more than 60% of the population. All POPs, with the exception of PFAAs compounds, were lipid-adjusted. Bivariate analysis was performed to assess POP concentrations according to sex, age, ecological region, housing conditions, smoking status, hunting frequency, consumption of marine mammals, terrestrial animals, wild birds, mollusks and seaweed, and RBC n-3 PUFA. For dichotomous independent variables, t-tests were used. For categorical independent variables with multiple categories, ANOVA were used, followed by post-hoc t-tests with Tukey-Kramer correction to consider unbalanced data. POP concentrations were compared with results from the 1992 Santé Quebec Health Survey and *Qanuippitaa?* 2004. Comparisons were also made with concentrations documented in the Inuit Health Survey 2007-2008 (IHS 2007-2008) conducted in the other regions of the Inuit Nunangat (Laird et al., 2013), and with the last cycle at which these concentrations were measured in the Canadian general population who participated in the Canadian Health Measures Survey (CHMS): Cycle 1 (2007-2008) for chlorinated pesticides, PCBs and PBDEs (Health Canada, 2020) and Cycle 5 (2017-2018) for PFAAs (Health Canada, 2019). When the 95% confidence intervals of geometric means did not overlap, the difference between geometric mean values was considered statistically significant. To estimate the reliability of an estimate, coefficients of variation (CV) were calculated as a percentage of the standard error. Proportions should be interpreted carefully if the CV is between 15% and 25%, and proportions have been shown for information only when the CV is above 25%.

4 RESULTS

4.1 OVERALL DISTRIBUTIONS AND CORRELATIONS BETWEEN POPS

Concentrations of 14 chlorinated pesticides and industrial compounds, 25 PCB congeners, nine polybrominated diphenyl ether congeners (PBDEs), and nine perfluoroalkyl acids (PFAAs) were determined in plasma samples from Nunavimmiut aged 18 and over (Table 2).

Of these POPs, nine chlorinated pesticides and other industrial compounds (*cis*-nonachlor, *trans*-nonachlor, oxychlordane, *p,p'*-DDE, β-HCH, hexachlorobenzene, mirex, and toxaphenes no.26 & 50), 16 PCB congeners, and six PFAAs [PFOA, PFHxS, and PFOS and long-chain PFAAs (PFNA, PFDA, PFUdA)] were detected in over 60% of the study population. No PBDE congeners were detected in more than 60% of the population.

Table 2 Overall distributions of plasma POP concentrations in Nunavimmiut aged 18 years and over, Nunavik, 2017

	n	% Detected	GM	25th	50th	75th	95th
Chlorinated pesticides/industrial compounds (μg/kg plasma lipids)							
Aldrin	499	0.0	<LOD	<LOD	<LOD	<LOD	<LOD
α-Chlordane	499	0.6	<LOD	<LOD	<LOD	<LOD	<LOD
γ-Chlordane	499	0.4	<LOD	<LOD	<LOD	<LOD	<LOD
cis-Nonachlor	500	96.0	9.0	4.4	9.5	19.8	70.8
trans-Nonachlor	500	99.6	60.0	27.2	63.2	134.8	527.0
Oxychlordane	500	99.8	31.1	13.9	31.4	66.2	323.4
p,p'-DDE	500	100	192.5	97.7	197.5	359.5	1044.3
p,p'-DDT	500	26.0	<LOD	<LOD	<LOD	6.2	17.7
β-HCH	500	83.0	4.4	1.5	3.8	8.9	30.7
γ-HCH	499	0.40	<LOD	<LOD	<LOD	<LOD	<LOD
Hexachlorobenzene	500	99.8	34.4	17.8	33.8	62.1	170.9
Mirex	500	83.6	6.2	2.5	5.7	14.5	66.9
Toxaphene no. 26	500	93.2	6.7	3.4	7.6	14.0	47.1
Toxaphene no. 50	500	96.4	10.9	5.4	12.4	23.5	74.5

	n	% Detected	GM	25th	50th	75th	95th
PCB congeners IUPAC # (μg/kg plasma lipids)							
28	500	0.6	<LOD	<LOD	<LOD	<LOD	5.8
52	499	0.0	<LOD	<LOD	<LOD	<LOD	<LOD
66	499	1.6	<LOD	<LOD	<LOD	<LOD	<LOD
74	500	34.0	<LOD	<LOD	<LOD	<LOD	26.2
99	500	78.0	10.8	4.8	11.1	22.4	64.0
101	500	18.6	<LOD	<LOD	<LOD	<LOD	9.4
105	500	56.6	<LOD	<LOD	<LOD	4.0	12.0
118	500	95.0	9.9	4.7	9.6	19.3	67.0
128	500	22.6	<LOD	<LOD	<LOD	<LOD	3.3
138	500	99.2	29.0	13.8	29.1	63.8	218
146	500	92.0	8.9	3.9	8.8	19.1	84.5
153	500	100	65.1	28.4	63.9	142.4	592.0
156	500	70.8	3.4	1.0	3.3	7.9	36.5
163	500	92.8	10.7	4.7	10.8	23.4	117.4
167	500	39.4	<LOD	<LOD	<LOD	3.3	9.1
170	500	91.6	9.8	3.8	9.8	24.0	111.5
178	500	76.4	4.1	<LOD	3.9	9.0	41.3
180	500	99.0	33.5	12.6	32.7	84.6	414.0
183	500	79.6	4.0	<LOD	4.1	8.6	31.4
187	500	97.2	16.0	7.0	16.3	34.9	131.0
194	500	77.0	5.8	<LOD	5.2	16.9	91.2
201	500	82.6	6.5	<LOD	6.2	17.7	88.1
203	500	70.0	3.7	<LOD	3.4	9.3	43.3
206	500	53.8	<LOD	<LOD	1.4	5.1	26.3
PCBs (Aroclor 1260)	500	89.8	493.9	216.0	482.3	1058.4	4096.7
PBDE congeners IUPAC # (μg/kg plasma lipids)							
15	499	0.0	<LOD	<LOD	<LOD	<LOD	<LOD
17	499	0.0	<LOD	<LOD	<LOD	<LOD	<LOD
25	499	0.0	<LOD	<LOD	<LOD	<LOD	<LOD
28	499	0.0	<LOD	<LOD	<LOD	<LOD	<LOD
33	499	0.0	<LOD	<LOD	<LOD	<LOD	<LOD
47	499	26.3	<LOD	<LOD	<LOD	3.6	11.7
99	499	5.0	<LOD	<LOD	<LOD	<LOD	<LOD
100	499	9.2	<LOD	<LOD	<LOD	<LOD	4.0
153	500	42.4	<LOD	<LOD	<LOD	6.7	14.8
Perfluoroalkyl acids (PFAAs) (μg/L)							
PFBA	500	8.4	<LOD	<LOD	<LOD	<LOD	0.09
PFHxA	500	0.2	<LOD	<LOD	<LOD	<LOD	<LOD
PFOA	500	100	1.1	0.7	1.0	1.5	2.4
PFNA	500	100	3.7	2.2	3.5	5.9	11.5
PFDA	491	100	0.7	0.4	0.7	1.2	2.7
PFUdA	500	100	0.7	0.4	0.8	1.2	2.6
PFBS	500	0.6	<LOD	<LOD	<LOD	<LOD	<LOD
PFHxS	500	100	0.6	0.4	0.6	1	2.4
PFOS	500	100	5.1	2.8	4.9	8.9	20.5

<LOD: Below the limit of detection.
GM (Geometric means) were calculated only if the chemical was detected in more than 60% of the population.
Chlorinated pesticides/industrial compounds, PCBs and PBDEs are adjusted for plasma lipids.
PCBs expressed as Aroclor 1260, calculated as the sum of PCB-138 and PCB-153 multiplied by 5.2.

The intercorrelations between the different POPs and groups of POPs detected in over 60% of the study population are presented in Table 3. Chlordane compounds correspond to the sum of oxychlordane, *cis*-nonachlor, and *trans*-nonachlor concentrations. The toxaphene group is the sum of toxaphene congeners 26 and 50. Total PCBs is the sum of all PCB congeners detected in at least 60% of Nunavimmiut. Total long-chain PFAAs is the sum of PFNA, PFDA, and PFUdA concentrations. Legacy POPs, including chlordane compounds, *p,p'*-DDE, β-HCH, hexachlorobenzene, mirex, toxaphenes, and PCBs, were strongly intercorrelated with coefficients ranging from 0.7 to 1.0. PFOS, PFHxS and long-chain PFAAs were moderately correlated to legacy POPs and other PFAAs with correlation coefficients ranging from 0.6 to 0.8, whereas correlations with PFOA were globally weaker (from 0.4 to 0.8). Long-chain PFAAs congeners (PFNA, PFDA, and PFUdA) were highly correlated with correlations ranging from 0.8 to 1.0 (not shown).

Table 3 Pearson correlation coefficients for different POPs and POP groups among Nunavimmiut aged 18 years and over, Nunavik, 2017.

	Chlordane compounds	*p,p'*-DDE	β-HCH	Hexachlorobenzene	Mirex	Toxaphenes	Total PCBs	PFOA	PFHxS	PFOS	Total long-chain PFAAs
Chlordane compounds	1										
***p,p'*-DDE**	0.9	1									
β-HCH	0.9	0.9	1								
Hexachlorobenzene	1	0.9	1	1							
Mirex	0.9	0.8	0.8	0.8	1						
Toxaphenes	0.9	0.9	0.9	0.9	0.7	1					
Total PCBs	0.9	0.9	0.9	0.9	1	0.8	1				
PFOA	0.5	0.5	0.5	0.5	0.5	0.4	0.6	1			
PFHxS	0.7	0.7	0.7	0.7	0.7	0.6	0.7	0.8	1		
PFOS	0.7	0.7	0.7	0.8	0.7	0.7	0.7	0.6	0.8	1	
Total long-chain PFAAs	0.7	0.7	0.7	0.7	0.6	0.7	0.7	0.6	0.7	0.8	1

Chlordane compounds: Sum of oxychlordane, cis-nonachlor, and trans-nonachlor; toxaphenes: Sum of toxaphene no. 26 and no. 50; Total PCBs: Sum of all PCBs detected in ≥60% of the population; Total long-chain PFAAs: Sum of PFNA, PFDA, and PFUdA.
All POP groups and/or compounds are logged. Chlordane compounds, *p,p'*-DDE, β-HCH, hexachlorobenzene, mirex, toxaphenes, and total PCBs are adjusted for lipids.
Long-chain PFAAs include PFNA, PFDA, and PFUdA.

4.2 MEAN CONCENTRATIONS OF POPS ACCORDING TO SEX AND AGE CATEGORIES

Concentrations of the different POPs were compared by age and sex groups (Table 4). All POP concentrations increased with age, such that Nunavimmiut aged 50 years and over had the highest levels with 2 to 10-fold increases compared to those aged 18 to 29 years. In general, concentrations of POPs were higher among younger men compared to younger women and among older women compared to older men, but these differences were not always statistically significant.

More specifically, women had higher levels of hexachlorobenzene and toxaphenes. Total long-chain PFAAs were also higher among women aged 30 to 49 and 50 years and over, but they were higher among men aged 18 to 29 years. Men had higher concentrations of mirex, PFOA, and PFHxS, and the differences between sex groups were more apparent in Nunavimmiut aged 18 to 29 and 30 to 49.

Table 4 POP plasma levels (GM and 95% CI) according to sex and age groups, Nunavik, 2017.

Contaminant and age	Total		Men		Women	
	GM	95% CI	GM	95% CI	GM	95% CI
Organochlorines						
Chlordane compounds						
18 – 29 years	52.8[a]	45.3 – 61.4	59.1[a]	47.8 – 73.2	47.0[a]	38.2 – 57.9
30 – 49 years	91.9[b]	77.6 – 108.9	80.3[b]	60.2 – 106.9	104.8[b]	87.7 – 125.2
50 years and over	299.6[c]	238.9 – 375.6	249.9[c]	179.7 – 347.5*	363.7[c]	271.2 – 487.9
Total	100.8	89.5 – 113.4	96.2	81.1 – 114.1	105.5	91.6 – 121.5
p,p'-DDE						
18 – 29 years	113.9[a]	100.2 – 129.5	125.6[a]	103.5 – 152.4	103.2[a]	88.0 – 120.9
30 – 49 years	180.0[b]	158.7 – 204.2	167.8[b]	135.9 – 207.2	192.6[b]	165.5 – 224.2
50 years and over	461.2[c]	381.2 – 558.1	*372.7*[c]	*283.6 – 489.9*	*579.4*[c]	*454.1 – 739.3*
Total	192.5	175.4 – 211.4	185.4	162.0 – 212.1	200.0	177.8 – 225.1
β-HCH						
18 – 29 years	2.4[a]	2.1 – 2.7	2.5[a]	2.1 – 3.0	2.3[a]	2.0 – 2.7
30 – 49 years	4.2[b]	3.7 – 4.8	3.7[b]	3.0 – 4.6	4.8[b]	4.1 – 5.6
50 years and over	11.2[c]	9.3 – 13.5	9.7[c]	7.4 – 12.8*	13.1[c]	10.4 – 16.6
Total	4.4	4.0 – 4.8	4.1	3.6 – 4.7	4.7	4.2 – 5.3
Hexachlorobenzene						
18 – 29 years	21.5[a]	19.3 – 23.9	22.4[a]	19.3 – 26.0	20.5[a]	17.6 – 23.9
30 – 49 years	33.1[b]	29.3 – 37.4	*28.3*[b]	*23.2 – 34.5*	*38.5*[b]	*33.4 – 44.4*
50 years and over	72.8[c]	61.5 – 86.3	*60.8*[c]	*47.7 – 77.5**	*88.4*[c]	*71.2 – 109.7*
Total	34.4	31.6 – 37.3	*31.6*	*28.1 – 35.7*	*37.4*	*33.8 – 41.3*
Mirex						
18 – 29 years	2.4[a]	2.1 – 2.8	2.8[a]	2.2 – 3.5	2.1[a]	1.8 – 2.5
30 – 49 years	6.1[b]	5.2 – 7.1	6.9[b]	5.4 – 8.9	5.4[b]	4.6 – 6.5
50 years and over	24.8[c]	20 – 30.7	26.3[c]	19.6 – 35.4	23.3[c]	17.2 – 31.5
Total	6.2	5.5 – 6.9	*6.9*	*5.9 – 8.2*	*5.5*	*4.8 – 6.3*
Toxaphenes						
18 – 29 years	10.5[a]	8.9 – 12.3	10.7[a]	8.6 – 13.3*	10.2[a]	8.3 – 12.6
30 – 49 years	17.2[b]	14.3 – 20.7	*13.2*[b]	*9.7 – 18.0*	*22.1*[b]	*18.4 – 26.6*
50 years and over	40.0[c]	31.6 – 50.6	33.8[c]	23.4 – 49.0*	47.8[c]	36.5 – 62.7
Total	17.7	8.9 – 12.3	*15.6*	*13.0 – 18.7*	*20.1*	*17.6 – 22.9*
Total PCBs						
18 – 29 years	108.5[a]	95.5 – 123.3	*124.7*[a]	103.0 – 151.0	*94.3*[a]	*79.9 – 111.3*
30 – 49 years	219.9[b]	190.5 – 253.9	234.9[b]	186.9 – 295.3	206.4[b]	174.5 – 244
50 years and over	851.8[c]	702.1 – 1033.4	802.1[c]	616.4 – 1043.8	908.3[c]	686.1 – 1202.5
Total	238.0	214.3 – 264.3	255.2	220.7 – 295.3	221.8	193.3 – 253.7
PFAAs						
PFOA						
18 – 29 years	0.9[a]	0.8 – 0.9	*1.2*[a]	*1.1 – 1.3*	*0.6*[a]	*0.6 – 0.7*
30 – 49 years	1.0[b]	0.9 – 1.1	*1.2*[b]	*1.1 – 1.3*	*0.8*[b]	*0.7 – 0.9*
50 years and over	1.6[c]	1.4 – 1.7	1.6[c]	1.3 – 1.8	1.6[c]	1.4 – 1.7
Total	1.0	1.0 – 1.1	*1.3*	*1.2 – 1.4*	*0.9*	*0.8 – 0.9*

Contaminant and age	Total		Men		Women	
	GM	95% CI	GM	95% CI	GM	95% CI
PFHxS						
18 – 29 years	0.4[a]	0.4 – 0.5	*0.6*[a]	*0.6 – 0.7*	*0.3*[a]	*0.3 – 0.3*
30 – 49 years	0.6[b]	0.5 – 0.7	*0.8*[b]	*0.7 – 0.9*	*0.5*[b]	*0.4 – 0.5*
50 years and over	1.2[c]	1.0 – 1.4	1.3[c]	1.0 – 1.5	1.1[c]	0.9 – 1.4
Total	0.6	0.6 – 0.7	*0.8*	*0.8 – 0.9*	*0.5*	*0.5 – 0.6*
PFOS						
18 – 29 years	3.7[a]	3.3 – 4.1	*4.1*[a]	*3.6 – 4.8*	*3.3*[a]	*2.8 – 3.8*
30 – 49 years	4.8[b]	4.3 – 5.4	4.6[b]	4.0 – 5.4	5.0[b]	4.3 – 5.9
50 years and over	8.8[c]	7.4 – 10.3	8.1[c]	6.5 – 10.1	9.5[c]	7.6 – 11.9
Total	5.1	4.7 – 5.5	5.1	4.6 – 5.7	5.0	4.5 – 5.6
Total long-chain PFAAs						
18 – 29 years	4.3[a]	3.9 – 4.6	*4.6*[a]	*4.1 – 5.2*	*3.9*[a]	*3.5 – 4.4*
30 – 49 years	4.8[a]	4.3 – 5.3	*4.1*[b]	*3.5 – 4.9*	*5.5*[b]	*4.8 – 6.2*
50 years and over	8.1[b]	7.0 – 9.3	*6.6*[c]	*5.4 – 8.1*	*10.1*[c]	*8.5 – 12*
Total	5.2	4.9 – 5.6	*4.9*	*4.4 – 5.4*	*5.6*	*5.2 – 6.1*

GM 95% CI: Geometric mean and its 95% confidence interval.
Chlordane compounds: Sum of oxychlordane, cis-nonachlor, and trans-nonachlor; toxaphenes: Sum of toxaphene no. 26 and no. 50; Total PCBs: Sum of all PCBs detected in ≥60% of the population; Total long-chain PFAAs: Sum of PFNA, PFDA, and PFUdA.
All POPs, except PFAAs μg/L, are adjusted for total plasma lipids μg/kg plasma lipids.
[a, b, c] Estimates with different superscripts are significantly different between age groups at $p < 0.05$. Estimates in italics are significantly different between sexes at $p < 0.05$.
* The coefficient of variation is greater than 15% and lower than or equal to 25%. The proportion should be interpreted carefully.
** The coefficient of variation is greater than 25%. The proportion is shown for information only.

4.3 COMPARISONS WITH OTHER SURVEYS IN NUNAVIK AND ELSEWHERE IN CANADA

Santé Québec Survey, 1992 and *Qanuippitaa?* 2004 (Nunavik)

There was a significant decrease in the plasma concentrations of all POPs measured in 1992, 2004 and 2017 (Table 5). In comparison to 2004, most legacy POPs decreased by approximately half in 2017. No information is available on PFAA congeners in 1992 and only PFOS was measured in 2004. PFOS plasma concentrations also decreased markedly (by 72%) in 2017 compared to 2004 (18 versus 5.1 μg/L).

Table 5 Plasma POP concentrations (μg/L) among Nunavimmiut aged 18 years and over in Nunavik (1992, 2004 and 2017)

	1992	2004	2017
	GM (95% CI)	GM (95% CI)	GM (95% CI)
Chlorinated pesticides and other industrial compounds			
Aldrin	<LOD	<LOD	<LOD
α-Chlordane	<LOD	<LOD	<LOD
γ-Chlordane			<LOD
Cis-nonachlor	0.23 (0.21 – 0.25)	0.11 (0.10 – 0.11)	0.06 (0.05 – 0.06)
Trans-nonachlor	1.1 (1.0 – 1.3)	0.72 (0.67 – 0.77)	0.38 (0.34 – 0.43)
Oxychlordane	0.79 (0.71 – 0.88)	0.41 (0.38 – 0.44)	0.20 (0.18 – 0.22)
p,p'-DDE	6.9 (6.3 – 7.5)	2.9 (2.7 – 3.0)	1.2 (1.1 – 1.4)
p,p'-DDT	0.21 (0.19 – 0.24)	0.07 (0.07 – 0.07)	<LOD
β-HCH	0.13 (0.12 – 0.14)	0.05 (0.05 – 0.05)	0.03 (0.03 – 0.03)
γ-HCH			<LOD
Hexachlorobenzene	0.94 (0.87 – 1.01)	0.35 (0.33 – 0.37)	0.22 (0.20 – 0.24)
Mirex	0.17 (0.15 – 0.18)	0.07 (0.06 – 0.07)	0.04 (0.04 – 0.04)
Toxaphene no. 26		0.08 (0.07 – 0.08)	0.04 (0.04 – 0.05)
Toxaphene no. 50		0.14 (0.13 – 0.15)	0.07 (0.06 – 0.08)
Polychlorinated biphenyls (PCBs) IUPAC #			
28			<LOD
52	0.06 (0.05 – 0.06)		<LOD
66			<LOD
74		0.05 (0.05 – 0.06)	<LOD
99	0.41 (0.36 – 0.46)	0.15 (0.14 – 0.16)	0.07 (0.06 – 0.08)
101	0.07 (0.06 – 0.07)	0.02 (0.02 – 0.02)	<LOD
105	0.07 (0.06 – 0.08)	0.03 (0.02 – 0.03)	<LOD
118	0.27 (0.24 – 0.30)	0.13 (0.12 – 0.14)	0.06 (0.06 – 0.07)
128		<LOD	<LOD
138	1.1 (0.97 – 1.2)	0.46 (0.43 – 0.49)	0.19 (0.17 – 0.21)
146		0.13 (0.12 – 0.14)	0.06 (0.05 – 0.06)
153	1.7 (1.5 – 1.9)	1.1 (0.99 – 1.1)	0.42 (0.37 – 0.46)
156	0.15 (0.13 – 0.17)	0.05 (0.04 – 0.05)	0.02 (0.02 – 0.02)
163		0.16 (0.15 – 0.17)	0.07 (0.06 – 0.08)
167		0.01 (0.01 – 0.02)	<LOD
170	0.33 (0.29 – 0.37)	0.16 (0.15 – 0.17)	0.06 (0.06 – 0.07)
178		0.05 (0.05 – 0.05)	0.03 (0.02 – 0.03)
180	0.98 (0.87 – 1.12)	0.56 (0.53 – 0.60)	0.21 (0.19 – 0.24)
183	0.12 (0.11 – 0.14)	0.06 (0.05 – 0.06)	0.03 (0.02 – 0.03)
187	0.41 (0.36 – 0.46)	0.23 (0.21 – 0.24)	0.10 (0.09 – 0.11)
194		0.10 (0.09 – 0.11)	0.04 (0.03 – 0.04)
201		0.10 (0.09 – 0.10)	0.04 (0.04 – 0.05)
203		0.05 (0.05 – 0.06)	0.02 (0.02 – 0.03)
206		0.03 (0.03 – 0.03)	<LOD
PCBs (Aroclor 1260)		7.7 (7.2 – 8.3)	3.2 (2.8 – 3.5)

	1992	2004	2017
	GM (95% CI)	GM (95% CI)	GM (95% CI)
Polybrominated diphenyl ethers (PBDEs) IUPAC #			
15			<LOD
17			<LOD
25			<LOD
28			<LOD
33			<LOD
47		0.04 (0.03 – 0.04)	<LOD
99		<LOD	<LOD
100		<LOD	<LOD
153		0.02 (0.02 – 0.02)	<LOD
Perfluoroalkyl acids (PFAAs)			
PFBA			<LOD
PFHxA			<LOD
PFOA			1.1 (1.00, 1.1)
PFNA			3.7 (3.4, 3.9)
PFDA			0.70 (0.65, 0.76)
PFUdA			0.69 (0.64, 0.76)
PFBS			<LOD
PFHxS			0.64 (0.60, 0.68)
PFOS		18 (18 – 19)	5.1 (4.7, 5.5)

GM (95% CI): Geometric mean and its 95% confidence interval.
<LOD: Below the limit of detection.
PCBs expressed as Aroclor 1260, calculated as the sum of PCB-138 and PCB-153 multiplied by 5.2.
All concentrations are unadjusted.

2007-2008 Inuit Health Survey

Table 6 compares plasma chlorinated pesticides and PCBs levels in Nunavik (*Qanuilirpitaa?* 2017) versus the three other regions of the Inuit Nunangat (IHS 2007-2008) (Laird et al., 2013). Concentrations of all POPs were higher in the adult population of the other regions in 2007-2008 compared to the Nunavik population in 2017. PFAAs levels were not measured in the IHS 2007-2008. PBDEs were detected in less than 60% of the Nunavik population in 2017.

Table 6 Plasma levels of chlorinated pesticides and PCBs (GM and 95% CI) among adults aged 18 years and over in the Inuit Nunangat from the Inuit Health Survey 2007-2008 and *Qanuilirpitaa?* 2017

	Inuit Health Survey 2007-2008	*Qanuilirpitaa?* 2017
	GM (95% CI)	GM (95% CI)
Chlordane compounds	**150.0 (138.0-159.0)**	**100.8 (89.5-113.4)**
p,p'-DDE & *p,p'*-DDT	**311.0 (292.0-323.0)**	**200.0 (182.7-218.9)**
Toxaphenes	**28.7 (26.3-30.3)**	**17.7 (15.7-19.9)**
PCBs (Aroclor 1260)	**1000.0 (949.0-1060.0)**	**493.9 (44.3-550.7)**
Total PCBs	**409.0 (389.0-430.0)**	**238.0 (214.3-264.3)**

GM (95% CI): Geometric mean and its 95% confidence interval.
Concentrations expressed on a lipid basis (µg/kg plasma lipids).
PCBs expressed as Aroclor 1260, calculated as the sum of PCB-138 and PCB-153 multiplied by 5.2.
Total PCBs: Sum of all PCBs detected in ≥60% of the population.
Significant differences were determined by a lack of overlap in the confidence intervals (in bold).

Non-Inuit Canadian population

Levels of chlorinated pesticides, PCBs and PBDEs in Nunavimmiut aged 18 years and over were compared to concentrations in the Canadian Health Measures Survey (CHMS): Cycle 1 (2007-2008) for adults aged 20 to 79 years (Table 7), as these chemicals were not available in later CHMS cycles. Concentrations of *cis*-nonachlor, *trans*-nonachlor, oxychlordane, hexachlorobenzene, toxaphenes 26 and 50, PCBs (Aroclor 1260), and PBDE 153 were detected at higher levels in Nunavik in 2017 than in the CHMS in 2007-2008. Indeed, despite the 10-year gap, *cis*-nonachlor, *trans*-nonachlor, oxychlordane, hexachlorobenzene, toxaphene no. 26, and toxaphene no. 50 levels were up to 7 to 10-fold higher in Nunavik. In contrast, PBDE 47 levels were over 3-fold higher in the Canadian population versus the population in Nunavik.

Table 7 Plasma levels of chlorinated pesticides, PBDEs, and PCBs (GM and 95% CI) among adults aged 18 years and over in Nunavik (2017) and adults aged 20 years and over from the general Canadian population (2007-2008).

Contaminant	CHMS 2007-2008		*Qanuilirpitaa?* 2017	
	GM	95% CI	GM	95% CI
Aldrin	<LOD		<LOD	
α-Chlordane	<LOD		<LOD	
γ-Chlordane	<LOD		<LOD	
cis-Nonachlor	**<LOD**		**9.0**	8.0 – 10.1
trans-Nonachlor	**6.0**	5.3 – 6.8	**60.0**	53.3 – 67.5
Oxychlordane	**4.2**	3.8 – 4.7	**31.1**	27.6 – 35.0
p,p'-DDE	152.1	127.0 – 182.0	192.5	175.4 – 211.4
p,p'-DDT	<LOD		<LOD	
β-HCH	**6.4**	4.8 – 8.6	**4.4**	4.0 – 4.8
Hexachlorobenzene	**9.1**	8.0 – 10.3	**34.4**	31.6 – 37.3
Mirex	**<LOD**		**6.2**	5.5 – 6.9
Toxaphene no.26	**<LOD**		**6.7**	6.0 – 7.6
Toxaphene no.50	**<LOD**		**10.9**	9.7 – 12.3
PCBs (Aroclor 1260)	**150.5**	131.8 – 172.0	**493.9**	443.0 – 550.7
PBDE 15	<LOD		<LOD	
PBDE 17	<LOD		<LOD	
PBDE 25	<LOD		<LOD	
PBDE 28	<LOD		<LOD	
PBDE 33	<LOD		<LOD	
PBDE 47	**10.0**	9.1 – 11.1	**3.2**	3.1 – 3.4
PBDE 99	<LOD		<LOD	
PBDE 100	<LOD		<LOD	
PBDE 153	**<LOD**		**4.0**	3.7 – 4.2

GM (95% CI): Geometric mean and its 95% confidence interval.
PCBs expressed as Aroclor 1260, calculated as the sum of PCB-138 and PCB-153 multiplied by 5.2.
Concentrations expressed on a lipid basis (μg/kg plasma lipids).
<LOD: Below the limit of detection.
Estimates in bold are significantly different across the CHMS and *Qanuilirpitaa?* 2017 at $p < 0.05$.

Concentrations of PFAAs were compared to those reported in the more recent cycle of CHMS (Cycle 5, 2017-2018) (Table 8). Plasma PFNA levels in Nunavimmiut were over 7-fold higher, plasma PFDA and PFUdA levels were over 3-fold higher, and plasma PFOS levels were 1.5-fold higher compared to those of the general Canadian population. Conversely, plasma levels of PFOA and PFHxS were significantly lower in Nunavik compared to the general Canadian population. Shorter chain PFAAs congeners, PFBA, PFHxA and PFBS were undetected in Nunavik and the general population alike.

Table 8 Plasma PFAAs levels (GM and 95% CI) among Nunavimmiut aged 18 aged years and over in Nunavik (2017) and in the general Canadian population (2017-2018).

Contaminant	CHMS 2017-2018		*Qanuilirpitaa?* 2017	
	GM	95% CI	GM	95% CI
PFBA	<LOD		<LOD	
PFHxA	<LOD		<LOD	
PFOA	**1.3**	1.2 – 1.4	**1.0**	1.0 – 1.1
PFNA	**0.5**	0.5 – 0.6	**3.7**	3.4 – 3.9
PFDA	**0.2**	0.2 – 0.2	**0.7**	0.6 – 0.8
PFUdA	**<LOD**		**0.7**	0.6 – 0.8
PFBS	<LOD		<LOD	
PFHxS	**1.0**	0.9 – 1.1	**0.6**	0.6 – 0.7
PFOS	**3.3**	2.9 – 3.7	**5.1**	4.7 – 5.5

GM (95% CI): Geometric mean and its 95% confidence interval.
Concentrations expressed in μg/L.
<LOD: Below the limit of detection.
Estimates in bold are significantly different across the CHMS and *Qanuilirpitaa?* 2017 at $p < 0.05$.

4.4 DETERMINANTS OF PLASMA POP LEVELS

Sociodemographic determinants

In 2017, Nunavimmiut living in Hudson Strait communities had the highest plasma levels of chlordane compounds, *p,p'*-DDE, β-HCH, and toxaphenes, but levels were comparable between Hudson Bay and Ungava Bay communities (Table 9). A trend was observed for the highest levels of total PCBs in Hudson Strait followed by Hudson Bay, although the differences were not statistically significant. Mirex and PFOS levels were higher in Hudson Strait and Hudson Bay, whereas total long-chain PFAAs concentrations were higher in Hudson Bay versus Ungava Bay.

Table 9 Plasma POP levels (GM and 95% CI) according to ecological region among Nunavimmiut aged 18 years and over, Nunavik, 2017

Contaminant	Hudson Bay	Hudson Strait	Ungava Bay
	GM (95% CI)	GM (95% CI)	GM (95% CI)
Organochlorines			
Chlordane compounds	90.7 (75.1 – 109.7)[a]	156.6 (127.7 – 192.1)*[b]	84.2 (70.5 – 100.5)[a]
p,p'-DDE	190.7 (162.2 – 224.2)[a]	241.0 (203.8 – 285.0)[b]	166.5 (146.4 – 189.3)[a]
β-HCH	3.7 (3.1 – 4.2)[a]	6.6 (5.5 – 8.0)*[b]	4.1 (3.6 – 4.8)[a]
Hexachlorobenzene	30.2 (26.3 – 34.5)[a]	45.0 (38.1 – 53.1)[b]	33.5 (29.6 – 37.8)[a]
Mirex	7.6 (6.3 – 9.1)[a]	7.3 (5.9 – 9.1)*[a]	4.3 (3.6 – 5.0)[b]
Toxaphenes	14.0 (11.6 – 16.8)[a]	27.8 (22.8 – 33.8)*[b]	17.2 (14.2 – 20.9)[a]
Total PCBs	247.9 (207.5 – 296.3)[ab]	285.0 (235.8 – 344.6)[a]	199.4 (171.1 – 232.5)[b]
PFAAs			
PFOA	1.1 (1.0 – 1.1)	1.0 (0.9 – 1.1)	1.1 (1.0 – 1.1)
PFHxS	0.6 (0.6 – 0.7)	0.7 (0.6 – 0.8)	0.6 (0.5 – 0.7)
PFOS	5.7 (5.0 – 6.4)[a]	6.1 (5.3 – 6.9)[a]	3.9 (3.5 – 4.4)[b]
Total long-chain PFAAs	5.8 (5.1 – 6.5)[a]	5.1 (4.5 – 5.7)[ab]	4.8 (4.4 – 5.3)[bc]

GM 95% CI: Geometric mean and its 95% confidence interval.
Chlordane compounds: Sum of oxychlordane, *cis*-nonachlor, and *trans*-nonachlor; Toxaphenes: Sum of toxaphene no. 26 and no. 50; Total PCBs: Sum of all PCBs detected in ≥60% of the population; Total long-chain PFAAs: Sum of PFNA, PFDA, and PFUdA.
Concentrations of PFAAs expressed in µg/L; concentrations of organochlorines expressed in µg/kg plasma lipids.
[a, b, c] Estimates with different superscripts are significantly different at $p < 0.05$.
* The coefficient of variation is greater than 15% and lower than or equal to 25%. The proportion should be interpreted carefully.
** The coefficient of variation is greater than 25%. The proportion is shown for information only.

There were no statistically significant differences in any POP groups by overcrowding status (Table 10). However, there were significantly higher plasma levels of all POPs, with the exception of *p,p'*-DDE, among Nunavimmiut living in a house requiring major repairs.

Table 10 Plasma POP levels (GM and 95% CI) according to housing conditions among Nunavimmiut aged 18 years and over, Nunavik, 2017.

Contaminant	Overcrowding Absent	Overcrowding Present	Minor/No housing repairs needed	Major housing repairs needed
	GM (95% CI)	GM (95% CI)	GM (95% CI)	GM (95% CI)
Organochlorines				
Chlordane compounds	109.9 (95.0 – 127.0)	100.1 (83.6 – 119.8)	**93.4 (82.5 – 105.9)**	**136.4 (101.2 – 183.8)****
p,p'-DDE	207.5 (184.7 – 233.1)	188.0 (162.8 – 217.2)	182.4 (165.3 – 201.4)	236.3 (183.0 – 305.1)*
β-HCH	4.6 (4.1 – 5.2)	4.3 (3.7 – 5.0)	**4.2 (3.8 – 4.6)**	**5.6 (4.3 – 7.3)***
Hexachlorobenzene	36.4 (32.6 – 40.5)	34.0 (29.8 – 38.9)	**32.4 (29.7 – 35.4)**	**44.3 (35.7 – 54.9)***
Mirex	6.9 (6.0 – 8.0)	5.7 (4.7 – 6.9)	**5.7 (5.0 – 6.4)**	**8.4 (6.0 – 11.7)***
Toxaphenes	19.3 (16.6 – 22.3)	17.3 (14.4 – 20.9)	**16.7 (14.7 – 19.0)**	**22.9 (17.4 – 30.2)****
Total PCBs	264.5 (231.3 – 302.6)	221.4 (187.2 – 261.9)	**220.5 (197.5 – 246.1)**	**313.1 (232.4 – 421.9)***
PFAAs				
PFOA	1.1 (1.0 – 1.1)	1.0 (0.9 – 1.1)	**1.0 (1.0 – 1.1)**	**1.2 (1.1 – 1.3)**
PFHxS	0.7 (0.6 – 0.7)	0.6 (0.5 – 0.7)	**0.6 (0.6 – 0.7)**	**0.8 (0.7 – 0.9)**
PFOS	5.3 (4.8 – 5.9)	5.0 (4.4 – 5.6)	**4.8 (4.4 – 5.3)**	**6.6 (5.6 – 7.8)**
Total long-chain PFAAs	5.4 (5.0 – 5.9)	5.2 (4.7 – 5.8)	**5.1 (4.7 – 5.4)**	**6.1 (5.3 – 7.2)**

GM 95% CI: Geometric mean and its 95% confidence interval.
Chlordane compounds: Sum of oxychlordane, *cis*-nonachlor, and *trans*-nonachlor; Toxaphenes: Sum of toxaphene no. 26 and no. 50; Total PCBs: Sum of all PCBs detected in ≥60% of the population; Total long-chain PFAAs: Sum of PFNA, PFDA, and PFUdA.
Concentrations of PFAAs expressed in μg/L; concentrations of organochlorines expressed in μg/kg plasma lipids.
Estimates in bold are significantly different at $p < 0.05$.
* The coefficient of variation is greater than 15% and lower than or equal to 25%. The proportion should be interpreted carefully.
** The coefficient of variation is greater than 25%. The proportion is shown for information only.
Overcrowding is defined using the persons per room (PPR) index. A household is considered overcrowded if the number of people in a household divided by the number of rooms (excluding the kitchen or bathrooms) plus 2 is greater than 1.
Minor repairs are defined as: missing or loose floor tiles, bricks or shingles, defective steps, railing or siding, etc., whereas major repairs are defined as: defective plumbing or electrical wiring, structural repairs to walls, floors or ceiling, etc.

Lifestyle and health determinants

There were no statistically significant differences in plasma POP levels by smoking status (Table 11).

Table 11 Plasma POP levels (GM and 95% CI) according to tobacco smoking status among Nunavimmiut aged 18 years and over, Nunavik, 2017.

Contaminant	Smoking status		
	Never smoked	Ex-smoker	Current smoker
	GM (95% CI)	GM (95% CI)	GM (95% CI)
Organochlorines			
Chlordane compounds	93.0 (58.1 – 148.6)**	111.9 (77.1 – 162.4)*	99.1 (87.5 – 112.2)
p,p'-DDE	171.2 (120.0 – 244.3)*	225.1 (172.5 – 293.7)*	190.3 (172.0 – 210.5)
β-HCH	4.3 (3.0 – 6.3)**	5.2 (3.8 – 7.1)*	4.3 (3.8 – 4.7)
Hexachlorobenzene	33.6 (24.0 – 47.2)*	37.3 (28.3 – 49)*	33.8 (30.9 – 36.9)
Mirex	5.6 (3.6 – 8.7)**	5.9 (4.1 – 8.6)*	6.2 (5.5 – 7.0)
Toxaphenes	18.6 (11.9 – 29.0)**	21.6 (14.7 – 31.6)*	16.9 (14.9 – 19.2)
Total PCBs	229.4 (148.7 – 353.7)	249.4 (177.9 – 349.5)*	234.1 (209.4 – 261.7)
PFAAs			
PFOA	1.2 (1.0 – 1.4)	1.2 (1.0 – 1.3)	1.0 (1.0 – 1.1)
PFHxS	0.7 (0.6 – 0.9)*	0.7 (0.6 – 0.9)	0.6 (0.6 – 0.7)
PFOS	5.8 (4.4 – 7.8)*	5.8 (4.6 – 7.4)	4.9 (4.5 – 5.3)
Total long-chain PFAAs	6.0 (4.8 – 7.6)	5.2 (4.3 – 6.3)	5.1 (4.8 – 5.5)

GM 95% CI: Geometric mean and its 95% confidence interval.
Chlordane compounds: Sum of oxychlordane, cis-nonachlor, and trans-nonachlor; Toxaphenes: Sum of toxaphene no. 26 and no. 50; Total PCBs: Sum of all PCBs detected in ≥60% of the population; Total long-chain PFAAs: Sum of PFNA, PFDA, and PFUdA. Concentrations of PFAAs expressed in μg/L; concentrations of organochlorines expressed in μg/kg plasma lipids. Estimates in bold are significantly different at $p < 0.05$.
* The coefficient of variation is greater than 15% and lower than or equal to 25%. The proportion should be interpreted carefully.
** The coefficient of variation is greater than 25%. The proportion is shown for information only.
Current smoker includes occasional and daily smokers; ex-smoker includes people who had smoked at least 100 cigarettes or more (about 4 packs) in their lifetime. All others were considered never smokers.

With the exception of mirex, the plasma concentrations of all POPs were significantly higher among Nunavimmiut who participated in summer hunting more than once a month compared to those who did so once a month or less (Table 12).

Table 12 Plasma POP levels (GM and 95% CI) according to hunting activities in the summer among Nunavimmiut aged 18 years and over, Nunavik, 2017.

Contaminant	Hunting frequency (summer)	
	≤1/month	>1/month
	GM (95% CI)	GM (95% CI)
Organochlorines		
Chlordane compounds	**81.1 (67.1 – 98.1)**	**115.5 (100.2 – 133.2)**
p,p'-DDE	**160.5 (137.7 – 187.0)**	**217.2 (194.6 – 242.4)**
β-HCH	**3.6 (3.1 – 4.2)**	**5.0 (4.4 – 5.6)**
Hexachlorobenzene	**28.6 (25.1 – 32.7)**	**38.7 (34.8 – 43.0)**
Mirex	5.5 (4.6 – 6.6)	6.6 (5.7 – 7.6)
Toxaphenes	**13.8 (11.5 – 16.6)**	**20.8 (18.0 – 24.1)**
Total PCBs	**203.5 (171.2 – 241.9)**	**260.8 (228.8 – 297.3)**
PFAAs		
PFOA	**0.9 (0.8 – 1.0)**	**1.2 (1.1 – 1.2)**
PFHxS	**0.5 (0.5 – 0.6)**	**0.7 (0.7 – 0.8)**
PFOS	**4.2 (3.7 – 4.7)**	**5.8 (5.3 – 6.4)**
Total long-chain PFAAs	**4.6 (4.2 – 5.1)**	**5.7 (5.2 – 6.2)**

GM 95% CI: Geometric mean and its 95% confidence interval.
Chlordane compounds: Sum of oxychlordane, *cis*-nonachlor, and *trans*-nonachlor; Toxaphenes: Sum of toxaphene no. 26 and no. 50; Total PCBs: Sum of all PCBs detected in ≥60% of the population; Total long-chain PFAAs: Sum of PFNA, PFDA, and PFUdA.
Concentrations of PFAAs expressed in μg/L; concentrations of organochlorines expressed in μg/kg plasma lipids.
Estimates in bold are significantly different at $p < 0.05$.
* The coefficient of variation is greater than 15% and lower than or equal to 25%. The proportion should be interpreted carefully.
** The coefficient of variation is greater than 25%. The proportion is shown for information only.

Dietary intake

Except in the case of PFOA and marine mammals, plasma POP levels were all significantly higher in Nunavimmiut who reported having consumed marine mammals and fish more frequently in the three months prior to the survey (Table 13). There was a 1.5 to 2-fold increase in chlordane compounds, *p,p'*-DDE, β-HCH, hexachlorobenzene, mirex, toxaphenes, and total PCBs in high consumers versus low consumers. There were also higher plasma levels of mirex, total PCBs and all PFAAs in high consumers versus low consumers of wild birds. None of the plasma POP levels differed by terrestrial animal consumption frequency.

Table 13 Plasma POP levels (GM and 95% CI) according to country food consumption among Nunavimmiut aged 18 years and over, Nunavik, 2017.

	Marine mammals (median: 1/week)		Fish and seafood (median: 1/week)		Terrestrial animals (median: 1/week)		Wild birds (median: 1-3/month)	
	Below median	Above median	Below median	Above median	Below median	Above median	Below median	Above median
Contaminant	GM (95% CI)	GM (95% CI)	GM (95% CI)	GM (95% CI)	GM (95% CI)	GM (95% CI)	GM (95% CI)	GM (95% CI)
Organochlorines								
Chlordane compounds	**71.6 (59.6-85.9)**	**136.4 (117.9-157.7)**	**71.7 (59.7-86.2)**	**130.6 (112.5-151.7)**	97.8 (80.1-119.4)*	100.7 (86.3-117.4)	80.5 (60.3-107.6)*	105.1 (92.4-119.6)
p,p'-DDE	**146.4 (126.5-169.3)**	**241.7 (214.8-271.8)**	**144.9 (125.1-167.8)**	**236.0 (208.9-266.7)**	182.2 (154.4-214.9)	194.0 (171.7-219.2)	163.3 (131.8-202.2)	196.5 (176.8-218.4)
β-HCH	**3.4 (2.9-3.9)**	**5.6 (4.9-6.4)**	**3.3 (2.9-3.9)**	**5.4 (4.8-6.2)**	4.3 (3.6-5.0)*	4.4 (3.9-5)	4.1 (3.3-5.0)	4.4 (4.0-5.0)
Hexachloro-benzene	**26.7 (23.6-30.2)**	**43.0 (38.3-48.3)**	**26.8 (23.6-30.4)**	**41.7 (37.2-46.8)**	33.1 (28.7-38.2)	34.8 (31.1-39)	29.4 (24-36)	35.4 (32.3-38.9)
Mirex	**4.6 (3.9-5.5)**	**7.5 (6.4-8.9)**	**4.5 (3.8-5.4)**	**7.4 (6.3-8.7)**	6.5 (5.3-8)	5.6 (4.8-6.5)	**4.0 (3.1-5.3)***	**6.5 (5.7-7.4)**
Toxaphenes	**12.7 (10.6-15.2)**	**24.5 (21.1-28.4)**	**13.3 (11.0-16.0)**	**22.6 (19.5-26.2)**	17.4 (14.3-21.1)*	18.0 (15.4-21.1)	16.3 (12.3-21.6)	18.2 (15.9-20.7)
Total PCBs	**179.2 (152.9-210.0)**	**292.2 (253.3-337.1)**	**174.6 (148.9-204.8)**	**289.3 (250.3-334.4)**	245.0 (202.2-296.8)	220.6 (192.6-252.6)	**175.3 (137-224.4)***	**246.9 (219.5-277.7)**
PFAAs								
PFOA	1.0 (0.9-1.1)	1.1 (1.0-1.2)	**1.0 (0.9-1.0)**	**1.1 (1.0-1.2)**	1.0 (1.0-1.1)	1.1 (1.0-1.1)	**0.9 (0.8-1.0)**	**1.1 (1.0-1.1)**
PFHxS	**0.6 (0.5-0.6)**	**0.7 (0.6-0.8)**	**0.6 (0.5-0.6)**	**0.7 (0.6-0.8)**	0.6 (0.6-0.7)	0.6 (0.6-0.7)	**0.5 (0.4-0.6)**	**0.7 (0.6-0.7)**
PFOS	**4.2 (3.7-4.7)**	**5.8 (5.2-6.5)**	**4.3 (3.9-4.9)**	**5.5 (5.0-6.2)**	4.8 (4.2-5.5)	5.1 (4.6-5.6)	**4.0 (3.4-4.7)**	**5.3 (4.8-5.7)**
Total long-chain PFAAs	**4.6 (4.2-5.1)**	**5.7 (5.2-6.2)**	**4.7 (4.2-5.2)**	**5.5 (5.1-6.0)**	5.2 (4.6-5.8)	5.1 (4.7-5.5)	**4.2 (3.6-4.8)**	**5.4 (5.0-5.8)**

GM (95% CI): Geometric mean and its 95% confidence interval.
Chlordane compounds: Sum of oxychlordane, *cis*-nonachlor, and *trans*-nonachlor; Toxaphenes: Sum of toxaphene no. 26 and no. 50; Total PCBs: Sum of all PCBs detected in ≥60% of the population; Total long-chain PFAAs: Sum of PFNA, PFDA, and PFUdA.
Concentrations of PFAAs expressed in μg/L; concentrations of organochlorines expressed in μg/kg plasma lipids.
Marine mammals: dried beluga meat (beluga *nikku*), beluga meat (fresh, cooked, frozen), beluga *mattaaq*, seal meat, seal liver, walrus meat (*igunak*); Fish and seafood: Arctic char, dried fish (*nikku, pitsik*), lake trout, brook or sea trout, salmon, pike or walleye, lake whitefish (coregone), sculpin (ugly fish), mollusk, seaweed; Terrestrial animals: dried caribou meat (caribou *nikku*), caribou meat (fresh, cooked, frozen), polar bear, muskox; Wild birds: ptarmigan, partridge, goose (Canada or white goose).
Estimates in bold are significantly different at $p < 0.05$.
* The coefficient of variation is greater than 15% and lower than or equal to 25%. The proportion should be interpreted carefully.
** The coefficient of variation is greater than 25%. The proportion is shown for information only.

All plasma POP levels were strongly and positively associated with RBC n-3-PUFA (Table 14). For example, plasma levels of chlordane compounds and toxaphenes were nearly 10-fold higher in the fourth quartile of RBC n-3-PUFA compared to the first quartile. Additionally, the increase across quartiles was stronger for PFOS and long-chain PFAAs compared to PFOA and PFHxS.

Table 14 Plasma POP levels (GM and 95% CI) according to RBC n-3 PUFA content among Nunavimmiut aged 18 years and over, Nunavik, 2017.

Contaminant	Quartile 1 RBC n-3 PUFA (2.8 – 5.7)	Quartile 2 RBC n-3 PUFA (5.7 – 7.4)	Quartile 3 RBC n-3 PUFA (7.4 – 9.2)	Quartile 4 RBC n-3 PUFA (9.2 – 16.2)
	GM (95% CI)	GM (95% CI)	GM (95% CI)	GM (95% CI)
Organochlorines				
Chlordane compounds	36.1 (29.0 – 44.9)[a]	72.3 (61.2 – 85.3)[b]	131.3 (109.5 – 157.4)[c]	300.5 (245.9 – 367.1)[d]
p,p'-DDE	98.6 (83.3 – 116.8)[a]	139.8 (121.5 – 160.9)[b]	229.3 (192.5 – 273.1)[c]	435.7 (368.8 – 514.7)[d]
β-HCH	1.8 (1.6 – 2.1)[a]	3.2 (2.8 – 3.7)[b]	5.3 (4.5 – 6.2)[c]	12.3 (10.4 – 14.6)[d]
Hexachlorobenzene	14.9 (13.1-17.0)[a]	26.2 (23.5 – 29.3)[b]	42.1 (36.8 – 48.3)[c]	85.0 (73.7 – 98.0)[d]
Mirex	2.9 (2.3 – 3.5)[a]	3.7 (3.1 – 4.4)[a]	8.4 (6.8 – 10.3)[b]	16.7 (13.3-21.0)[c]
Toxaphenes	5.6 (4.5-7.0)[a]	14.2 (11.9-17.0)[b]	23.6 (20.0 – 27.8)[c]	51.7 (43.1 – 61.9)[d]
Total PCBs	107.6 (90.0 – 128.7)[a]	153.2 (132.1 – 177.7)[b]	305.1 (251.6 – 369.9)[c]	643.1 (525.2 – 787.6)[d]
PFAAs				
PFOA	0.9 (0.8 – 0.9)[a]	0.9 (0.9-1.0)[a]	1.1 (1.0 – 1.2)[b]	1.4 (1.3 – 1.5)[c]
PFHxS	0.4 (0.4 – 0.5)[a]	0.5 (0.5 – 0.6)[b]	0.7 (0.6 – 0.8)[c]	1.1 (1.0 – 1.3)[d]
PFOS	2.8 (2.5 – 3.1)[a]	4.2 (3.8 – 4.8)[b]	5.6 (4.9 – 6.5)[c]	10.2 (8.9 – 11.6)[d]
Total long-chain PFAAs	3.0 (2.6 – 3.3)[a]	4.5 (4.1-5.0)[b]	5.9 (5.3 – 6.6)[c]	9.4 (8.5 – 10.5)[d]

GM (95% CI): Geometric mean and its 95% confidence interval.
Chlordane compounds: Sum of oxychlordane, *cis* – nonachlor, and *trans*-nonachlor; Toxaphenes: Sum of toxaphene no. 26 and no. 50; Total PCBs: Sum of all PCBs detected in ≥60% of the population; Total long-chain PFAAs: Sum of PFNA, PFDA, and PFUdA.
Concentrations of PFAAs expressed in μg/L; concentrations of organochlorines expressed in μg/kg plasma lipids.
[a, b, c] Estimates with different superscripts are significantly different at $p < 0.05$

5 DISCUSSION

ORGANOCHLORINES (INCLUDING PCBS) AND PBDES

Plasma levels of legacy POPs have decreased by approximately half since the *Qanuippitaa?* 2004 survey, and PBDEs were largely not detected in *Qanuilirpitaa?* 2017. Additionally, concentrations of POPs in Nunavik in 2017 were lower than the concentrations in the other regions of the Inuit Nunangat in 2007-2008. This highlights the importance of national and international regulations, such as the UNEP Stockholm Convention, that have played key roles in reducing and/or banning the production and use of many POP compounds. Notwithstanding these decreases, concentrations of *cis*-nonachlor, *trans*-nonachlor, oxychlordane, and toxaphenes remained 7 to 10-fold higher in 2017 than those of the general Canadian population measured in 2007-2008. Thus, continued reduction of POPs is necessary to protect the exceptional nutritional quality of country foods, and the health of adults and children in Nunavik.

As observed in previous Nunavik health surveys in 1992 and 2004 (Dewailly et al., 2007), older Nunavimmiut had higher plasma levels of all POPs, likely due to the long half lives of these compounds in the body (up to decades), higher exposure to POPs during their youth, and their greater consumption of country foods compared to younger Nunavimmiut. Residents of Hudson Strait communities had the highest concentrations of organochlorines, likely because of the higher consumption of marine mammals in this region (see the thematic report "Country and Market Food Consumption and Nutritional Status"). Predator marine mammals, such as beluga and seal, rely on subcutaneous fat reserves for insulation and metabolic fuel; however, their high fat content leads to the accumulation of lipophilic POPs in fatty tissues (Sonne et al., 2020). Relatedly, Nunavimmiut who frequently participated in hunting activities in the summer had higher levels of PCBs and other organochlorines. In addition to marine mammals, predatory fish, such as lake trout, also have higher concentrations of POPs due to their biomagnification properties (Walker et al., 2020). Lake trout was the second most consumed fish after Arctic char in Nunavik (see the thematic report "Country and Market Food Consumption and Nutritional Status"). The contribution of lake trout consumption to POP exposure remains to be investigated. Further evidence linking marine foods consumption and POP levels was observed in the strong associations between RBC n-3 PUFA, a biomarker of marine food consumption, and POP levels. Mirex and total PCBs were also greater in individuals with higher consumption of wild birds. Concentrations of organochlorines (including PCBs) have been detected in several bird species in the circumpolar region, such as Arctic murres in Newfoundland and gulls in Seymour Island and Arctic Norway (Braune et al., 2007; G. M. Donaldson et al., 1997; Fisk et al., 2005; Verreault et al., 2005). No studies have looked at birds in Nunavik specifically, but the aforementioned research provides evidence of POP contamination in bird species.

Differences in organochlorine compounds and total PCB levels across sex are in line with the findings from the *Qanuippitaa?* 2004 survey: hexachlorobenzene and toxaphenes levels were higher in women, and mirex levels were higher in men. Generally, higher POP levels were observed among younger men versus younger women and among older women versus older men. These results could be indicative of different dietary patterns by age and gender that influence POP levels. For example, RBC n-3 PUFA levels were higher in women versus men despite men reporting greater consumption of marine mammals and fish (see the thematic report "Country and Market Food Consumption and Nutritional Status"). Further multivariate analyses are needed to more precisely identify the factors responsible for different POP exposures among men and women in Nunavik.

Despite the discontinued use of PCBs and many PBDEs, they are still detected in residential dust worldwide (Klinčić et al., 2020; Whitehead et al., 2011). Although exposure determinants vary, higher dust PBDEs levels have been detected in urban homes and in those with more electronics (Klinčić et al., 2020). PCB dust levels, on the other hand, seem to be correlated with older houses, wood floors, and carpet pads, as well as with the type of carpet pads and wood floor finishing (Whitehead et al., 2014). Levels of PCBs and other organochlorines, except *p-p'* DDE, were elevated in Nunavimmiut living in a house needing major repairs, a proxy for older furniture use or excessive dust.

In contrast to other studies (Deutch et al., 2007), there were no differences in POP levels by smoking status, although cigarette smoking was frequent among Nunavimmiut in 2017, with 72% reporting daily smoking and 8% occasional smoking in the previous year.

PERFLUOROALKYL ACIDS

The present survey revealed a 72% decrease in plasma PFOS levels between 2004 (Dewailly et al., 2007) and 2017; however, PFOS concentrations in Nunavik remained 1.5-fold higher than those in the general Canadian population in 2017-2018. Additionally, PFNA, PFDA and PFUdA were 4 to 7-fold higher in residents of Nunavik in 2017 compared to the general Canadian population in 2017-2018.

Because only PFOS levels were measured in the *Qanuippitaa?* 2004 survey, additional information on temporal trends of plasma PFAA levels in Nunavik was obtained through different studies, including the NQN project conducted in collaboration with pregnant women in 2016-2017 (Caron-Beaudoin et al., 2020). Although there was a 24% to 28% decrease in older PFAAs congeners (PFOS, PFOA and PFHxS) in 2016-2017 compared to the levels observed in pregnant women recruited in 2012, concentrations of long-chain PFAAs congeners (PFNA, PFDA and PFUdA) increased by 13% to 21% between 2012 and 2016-2017. This increase in long-chain PFAAs exposure among Nunavimmiut was also documented in other circumpolar populations (AMAP, 2021). Indeed, PFAAs concentrations were also measured in Greenlandic Inuit women in 2011-2014 (Wielsøe et al., 2017). No confidence intervals were provided for the median PFAAs concentrations in the Greenlandic study, so it was not possible to compare statistical differences with Nunavik. Nevertheless, median PFOA, PFNA, and PFDA concentrations in Greenland appeared to be similar to the medians in Nunavik, whereas concentrations of PFUdA, PFHxS, and PFOS appeared higher than concentrations in Nunavik. Conversely, levels of PFHxS, PFOS, PFNA, PFDA and PFUdA in *Qanuilirpitaa?* 2017 were 3 to 6-fold higher than the levels observed in the Northwest Territories and Yukon (Garcia-Barrios et al., 2021). Long-chain PFAAs concentrations have been increasing in Arctic wildlife, including ringed seal, polar bears, and Arctic foxes (Muir et al., 2019), suggesting that consumption of country foods may be a source of exposure to these compounds among Nunavimmiut. The Northwest Territories and Yukon First Nations communities consume caribou, moose, whitefish, lake trout, and chinook, but no marine mammals (Garcia-Barrios et al., 2021), which could explain the differences in PFAAs levels.

As in the case of legacy POPs, the results of the present survey show that there were higher plasma levels of PFAAs in older Nunavimmiut compared to younger age groups. In addition, all PFAAs were moderately to strongly correlated with legacy POPs. PFOA was strongly correlated with only PFHxS, but moderately correlated with PFOS and total long-chain PFAAs. As such, the two congeners, PFOA and PFHxS, followed similar patterns. Plasma PFOA and PFHxS levels were higher in men versus women, in contrast to total long-chain PFAAs where plasma levels were higher in women. There were no differences in plasma PFOA and PFHxS levels by region; however, plasma PFOS and total long-chain PFAAs levels were higher in Hudson Bay (and Hudson Strait in the case of PFOS) compared to Ungava Bay. Likewise, there were no large differences in PFOA and PFHxS levels by country food consumption, although statistically significant differences were present; however, plasma PFOS and total long-chain PFAAs levels were higher in individuals with more frequent consumption of marine mammals, fish and seafood, and wild birds. PFAAs congener levels increased with higher quartiles of RBC n-3 PUFA, with 3 to 4-fold increases in PFOS and total long-chain PFAAs levels in the fourth RBC n-3 PUFA quartile versus the first, whereas a 2.75-fold and a 1.5-fold increase was observed for PFHxS and PFOA respectively. Taken together, these results indicate that marine food consumption is most likely a major contributor to PFOS and long-chain PFAAs exposure in Nunavik, whereas other sources of exposure could exist for PFOA and PFHxS.

In line with our current findings, PFOS and long-chain PFAAs were reportedly the main contributors of total PFAAs concentrations in marine and terrestrial wildlife (including reindeer, caribou, fish, seal, and beluga) throughout the circumpolar regions (Muir et al., 2019). Unlike lipophilic organochlorines, PFAAs congeners bind to proteins and phospholipids and are typically concentrated in the liver, kidney and blood of animals (Muir et al., 2019). In Nunavut in 2009, the highest PFAAs concentrations were identified in ringed seal liver and blood, caribou liver, polar bear meat and beluga meat (Ostertag et al., 2009). PFOS and PFNA were most commonly detected in these samples, with rare detection of PFOA. PFOS and long-chain PFAAs were also detected in adult thick-billed murres and northern fulmars and their eggs in Nunavut in 2008 (Braune et al., 2014; Braune & Letcher, 2013; Muir et al., 2019), albeit at lower concentrations compared to marine mammals, likely due to the shorter elimination half-life in birds (Butt et al., 2007). Further studies are required to explore PFAAs concentrations in country foods in Nunavik.

Of the older PFAAs congeners, PFOS and PFOA, PFOS has higher potential for bioaccumulation and persistence (D'Hollander et al., 2010a; Eriksson et al., 2016; Haukås et al., 2007; Müller et al., 2011; Xu et al., 2014), which explains the elevated concentrations of PFOS in Nunavik despite the ban on its use and production (UN Environment Programme 2019). Additionally, the long-range transport of volatile and non-bioaccumulative FTOHs in the Arctic (Wong et al., 2018), their biotransformation into PFOA and long-chain PFAAs (Martin et al., 2005; Muir et al., 2019), and the higher bioaccumulation potential of long-chain PFAAs compared to PFOA (D'Hollander et al., 2010a; Eriksson et al., 2016; Haukås et al., 2007; Müller et al., 2011; Xu et al., 2014) could lead to elevated concentrations of long-chain PFAAs in Arctic wildlife. This unique PFAAs plasma profile of higher concentrations of PFOS and long-chain PFAAs identified in this report mimics the profile identified in the NQN study, suggesting that FTOHs, particularly 8:2 FTOH, are important contributors to long-chain PFAAs in the Arctic (Caron-Beaudoin et al., 2020).

Other possible sources of exposure to PFAAs, particularly PFOA and PFHxS, include market foods, food packaging, local water sources, and consumer products (housing furniture, carpets, clothing, cosmetics, etc.) (Sunderland et al., 2019; Whitehead et al., 2021). Food is considered the dominant exposure pathway in non-Arctic communities (Haug et al., 2011), wherein PFAAs could be a food contaminant at the source of production or via contamination from PFAAs-treated food contact papers that migrate directly into some foods, such as butter, vinegar, and water/ethanol mixtures (Sunderland et al., 2019). A study in the U.S. found 46% of food contact papers and 20% of cardboard samples from a total of 400 samples contained detectable fluorine (Schaider et al., 2017). Many of the market foods available in Nunavik's stores require a lot of packaging since store-bought foods need to be flown into communities year round and shipped during the ice-free summer months (ITK, 2021), which involves additional handling (Canada, 2015). Cardboard is also commonly used to prepare and eat country foods in the North (Collings et al., 2016). It would be worthwhile to better document these potential exposure sources. Water can be an important source of PFAAs exposure in non-Arctic populations due to contamination of local water sources (Sunderland et al., 2019). Local water sources are unlikely to be an exposure source of concern in Nunavik, but further studies are required to confirm this.

In addition to food contamination, the indoor environment may account for up to half the total PFAAs intake (Haug et al., 2011; Sunderland et al., 2019). In this report, major house repairs needed was used as an indicator of older furniture use or excessive dust, both of which are sources of PFAAs (Whitehead et al., 2013; Zota et al., 2018). All PFAAs congeners were higher among individuals living in houses requiring major repairs, which may indicate PFAAs exposure via dust or higher PFAAs concentrations in individuals with lower socioeconomic status. The type of indoor ventilation could also influence the level of dust in a home (Degois et al., 2021). PFAAs measurement in dust samples could provide further information on this potential exposure source. There were no differences in PFAAs levels across smoking status, suggesting that smoking is unlikely an exposure source.

Further research with multivariate models is required to study exposure sources, country foods, market foods, cosmetics, and local water in relation to PFAAs and between genders, and to assess those with the largest contribution to PFAAs exposure. There is also a need to further study the impact of persistent elevated PFOS and increasing long-chain PFAAs exposure on immune, respiratory and cardiometabolic health in Nunavik and other circumpolar regions.

6 CONCLUSION

Our findings indicate that despite the decrease in legacy POP exposure by approximately half over the last two decades, concentrations of many POPs in Nunavik remain several fold higher than among the general Canadian population. Furthermore, evidence points to an increase in exposure to long-chain PFAAs, which are an emerging public health concern in the Arctic.

Older individuals had higher levels of all POPs compared to younger individuals. Additionally, younger men versus younger women and older women versus older men overall had higher concentrations of POPs, but this was not statistically significant across all POP groups. In general, Nunavimmiut in Hudson Strait had the highest concentrations of POPs, but this was also not consistent across all POP compounds/groups. All POPs were associated with marine mammal and/or fish/seafood consumption. Mirex, PCBs, and PFAAs levels were also associated with wild bird consumption. More studies are required to identify which species in particular contribute to POP exposure, as well as the extent to which market foods and food packaging, market products (including cosmetics for PFASs), housing conditions, and other possible local sources contribute to PFAAs exposures. Further studies need to explore the associations between PFAAs exposures and health impacts, particularly with respect to immune, endocrine and respiratory functions in Nunavik.

Assessing temporal trends of POPs in general as well as identifying sources of exposure to PFAAs and their possible health effects are required to advocate against the production and use of these persistent chemicals worldwide.

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APPENDIX
SUPPLEMENTARY TABLES

Supplementary Table 1 Limits of detection (LOD)

	2004	2017
Chlorinated pesticides and other industrial compounds		
Aldrin	0.01	0.01
α-Chlordane	0.005	0.005
γ-Chlordane		0.005
Cis-nonachlor	0.005	0.005
Trans-nonachlor	0.001	0.01
Oxychlordane	0.002	0.005
p,p'-DDE	0.015	0.02
p,p'-DDT	0.02	0.05
β-HCH	0.01	0.01
γ-HCH		0.01
Hexachlorobenzene	0.02	0.02
Mirex	0.01	0.01
Toxaphene no. 26	0.005	0.005
Toxaphene no. 50	0.005	0.005
Polychlorinated biphenyls (PCBs) IUPAC #		
28		0.05
52		0.3
66		0.03
74	0.01	0.03
99	0.01	0.03
101	0.01	0.03
105	0.01	0.01
118	0.01	0.01
128	0.01	0.01
138	0.01	0.01
146	0.01	0.01
153	0.01	0.01
156	0.01	0.01
163	0.01	0.01
167	0.01	0.01
170	0.01	0.01
178	0.01	0.01
180	0.01	0.01
183	0.01	0.01
187	0.01	0.01
194	0.01	0.01
201	0.01	0.01
203	0.01	0.01
206	0.01	0.01

	2004	2017
Polybrominated diphenyl ethers (PBDEs) IUPAC #		
15		0.03
17		0.03
25		0.03
28		0.03
33		0.03
47	0.01	0.03
99	0.03	0.02
100	0.02	0.03
153	0.02	0.03
Perfluoroalkyl acids (PFAAs)		
PFBA		0.08
PFHxA		0.08
PFOA		0.07
PFNA		0.10
PFDA		0.09
PFUdA		0.10
PFBS		0.07
PFHxS		0.06
PFOS	0.1	0.40

LOD are expressed in µg/L.

Supplementary Table 2 Pearson correlation coefficients for logged blood concentrations of PCBs

	PCB-99	PCB-118	PCB-138	PCB-146	PCB-153	PCB-156	PCB-163	PCB-170	PCB-178	PCB-180	PCB-183	PCB-187	PCB-194	PCB-201	PCB-203	Aroclor-1260*
PCB-99	1															
PCB-118	0.92	1														
PCB-138	0.95	0.94	1													
PCB-146	0.91	0.91	0.97	1												
PCB-153	0.92	0.90	0.98	0.99	1											
PCB-156	0.78	0.76	0.84	0.91	0.91	1										
PCB-163	0.89	0.89	0.95	0.99	0.98	0.92	1									
PCB-170	0.82	0.8	0.89	0.95	0.96	0.96	0.96	1								
PCB-178	0.85	0.82	0.89	0.95	0.94	0.96	0.96	0.96	1							
PCB-180	0.81	0.79	0.89	0.95	0.96	0.96	0.96	0.99	0.95	1						
PCB-183	0.95	0.89	0.96	0.96	0.96	0.89	0.94	0.92	0.93	0.91	1					
PCB-187	0.90	0.89	0.96	0.99	0.99	0.91	0.98	0.96	0.95	0.96	0.95	1				
PCB-194	0.72	0.69	0.79	0.88	0.88	0.96	0.89	0.96	0.94	0.96	0.85	0.89	1			
PCB-201	0.78	0.76	0.85	0.92	0.92	0.96	0.93	0.97	0.96	0.98	0.90	0.93	0.98	1		
PCB-203	0.77	0.74	0.83	0.90	0.90	0.97	0.90	0.95	0.95	0.95	0.89	0.91	0.98	0.97	1	
Aroclor-1260*	0.93	0.92	0.99	0.99	1	0.90	0.98	0.94	0.93	0.94	0.97	0.99	0.86	0.90	0.89	1

*PCBs expressed as Aroclor 1260, calculated as the sum of PCB-138 and PCB-153 multiplied by 5.2.

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