Science of the Total Environment 827 (2022) 154270

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Persistent organic pollutant exposures among Greenlandic adults in relation to lifestyle and diet: New data from the ACCEPT cohort

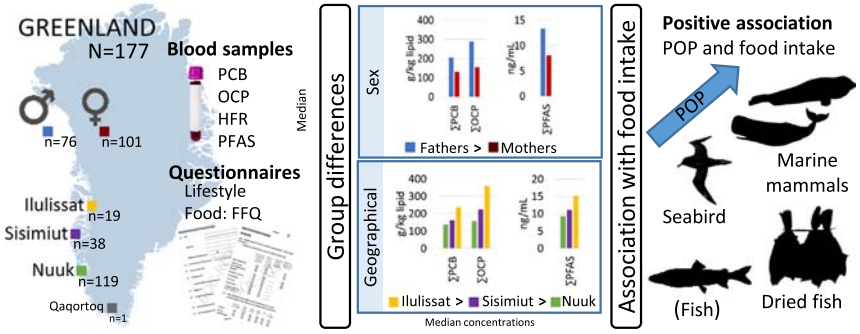
Maria Wielsøe [a,*], Manhai Long [a], Rossana Bossi [b], Katrin Vorkamp [b], Eva Cecilie Bonefeld-Jørgensen [a,c]

[a] Centre for Arctic Health & Molecular Epidemiology, Department of Public Health, Aarhus University, Bartholins Allé 2, 8000 Aarhus, Denmark
[b] Department of Environmental Science, Aarhus University, Frederiksborgvej 399, 4000 Roskilde, Denmark
[c] Greenland Centre for Health Research, University of Greenland, Manutooq 1, 3905 Nuussuaq, Greenland

HIGHLIGHTS

- Intra-individual decrease of POP concentrations in mothers from 2013–15 to 2019–20
- Higher POP concentrations in fathers than in mothers
- Geographical differences in POP concentration in the order Ilulissat > Sisimiut > Nuuk
- POPs associated positively with intake of marine mammals, seabirds, and dried fish

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 17 December 2021
Received in revised form 20 February 2022
Accepted 27 February 2022
Available online 1 March 2022

Editor: Adrian Covaci

Keywords:
Arctic
Organochlorine pesticides
Polychlorinated biphenyls (PCBs)
Per- and polyfluoroalkyl substances (PFAS)
Polybrominated diphenyl ethers (PBDEs)
Halogenated flame retardants (HFRs)

ABSTRACT

High concentrations of persistent organic pollutants (POPs) in blood of the Greenlandic population are well known. The exposure is mainly through traditional food intake, including marine mammals and seabirds.

The present study aimed to follow up on POP concentrations (organochlorine pesticides, polychlorinated biphenyls, per- and polyfluoroalkyl substances, and halogenated flame retardants (HFRs)) and relations to lifestyle and diet of the mothers included in the Greenlandic ACCEPT cohort (3–5 years after inclusion in 2013–15) and to include the children's fathers. This new data collection in 2019–20 included blood samples for measurement of POP concentrations and lifestyle and food frequency questionnaires from 101 mothers and 76 fathers aged 24–55 years living in Nuuk, Sisimiut, and Ilulissat, Greenland.

The mothers' intra-individual median percentage decrease in POP concentrations from inclusion to this follow-up (3–5 years later) was 16–58%, except for mirex (0% change). Median concentrations of POPs were 1.4–4.6 times higher in fathers than in mothers. The POPs differed by residential town with generally higher concentrations in Ilulissat compared to Sisimiut and Nuuk. We report, for the first time, novel HFRs in human samples from Greenland. However, concentrations were low and only dechlorane plus (with its anti-isomer) was detected in >50% of the samples.

Abbreviations: AA, arachidonic acid; anti-DP, anti-isomer dechlorane plus; AU, Aarhus University; BTBPE, 1,2-Bis(2,4,6-tribromophenoxy)ethane; DHA, docosahexaenoic acid; DP, dechlorane plus; DPTE, 2,4,6-Tribromophenyl-2,3-dibromopropyl ether; EH-TBB, 2-Ethylhexyl-2,3,4,5-tetrabromobenzoate; EPA, eicosapentaenoic acid; FA, fatty acid; FFQ, food frequency questionnaire; HCB, hexachlorobenzene; HFR, halogenated flame retardant; INSPQ, Institute National de Santé Publique du Québec; lipPOP, lipophilic persistent organic pollutant; LOD, limit of detection; LOQ, limit of quantification; n-3, n-3 polyunsaturated fatty acids; n-6, n-6 polyunsaturated fatty acids; OCP, organochlorine pesticide; p,p′-DDE, p,p′-dichlorodiphenyldichloroethylene; p,p′-DDT, p,p′-dichlorodiphenyltrichloroethane; PBB, polybrominated biphenyl; PBDE, polybrominated diphenyl ether; PCB, polychlorinated biphenyl; PFAS, per- and polyfluoroalkyl substance; PFBS, perfluorobutane sulfonate; PFCA, perfluorocarboxylic acids; PFDA, perfluorodecanoic acid; PFDoA, perfluorododecanoic acid; PFDS, perfluorodecane sulfonate; PFHpA, perfluoroheptanoic acid; PFHpS, perfluoroheptane sulfonate; PFHxA, perfluorohexanoic acid; PFHxS, perfluorohexane sulfonate; PFNA, perfluorononanoic acid; PFOA, perfluorooctanoic acid; PFOS, perfluorooctane sulfonate; PFOSA, perfluorooctane sulphonamide; PFSA, perfluorosulfonic acid; PFTeA, perfluorotetradecanoic acid; PFTrA, perfluorotridecanoicacid; PFUnA, perfluoroundecanoic acid; POP, persistent organic pollutant; *syn*-DP, syn-isomer dechlorane plus; β-HCH, β-hexachlorocyclohexan.

* Corresponding author at: Center for Arctic Health and Molecular Epidemiology, Department of Public Health, Aarhus University, Denmark.
E-mail addresses: mwielsoe@ph.au.dk (M. Wielsøe), ebj@ph.au.dk (E.C. Bonefeld-Jørgensen).

http://dx.doi.org/10.1016/j.scitotenv.2022.154270

Most POPs correlated positively with age and n-3/n-6 fatty acid ratio. The lipophilic POPs correlated positively with the percentage of life lived in Greenland, whereas few POPs correlated positively with BMI, income (personal and household), education, and alcohol intake.
The POPs generally associated positively with the intake of marine mammals, seabirds, and dried fish, while few POPs associated positively with Greenlandic fish intake. In contrast, POPs generally associated negatively with imported meat products intake.
The study findings may be of interest for future dietary recommendations in Greenland. We discuss the potential explanations for the findings and suggestions for future research.

1. Introduction

High concentrations of persistent organic pollutants (POPs) in the blood and tissues of people from the Arctic are well known. Global pollution and long-range transport are the main sources of POPs in the Arctic (Barrie et al., 1992; Heidam et al., 2004; Macdonald et al., 2000). The POP groups included in the present study are polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), halogenated flame retardants (HFRs) (such as polybrominated diphenyl ethers (PBDEs) and dechlorane plus (DP)), and per- and polyfluoroalkyl substances (PFAS). Due to their physical-chemical properties, these chemicals bio-accumulate and, in most cases, bio-magnify, in the Arctic marine food web and reach high concentrations in predatory species such as marine mammals (Borgå et al., 2004; Johansen et al., 2004).

POP exposure has been linked to a wide array of potential adverse health outcomes, including neurobehavioral, immunological, reproductive, endocrine, cardiovascular, and carcinogenic effects in humans (Weihe et al., 2016).

The Inuit population is at high risk of POP exposure due to their traditional food intake, including marine mammals (whales and seals) and seabirds. Consumption of traditional food is the main human exposure source to POPs in Greenland, even though PFAS and HFRs may also be present in imported food items, personal care products, other consumer products, and electronic devices (D'Hollander et al., 2010; Domingo and Nadal, 2017; Eriksson et al., 2013; Fujii et al., 2013; Yang et al., 2020). The dietary habits of the Greenlandic population have shifted from a diet mainly relying on traditional food, to a diet consisting of both traditional and imported foods (Knudsen et al., 2015; Larsen et al., 2019; Terkelsen et al., 2017; Wielsøe et al., 2021). Although it varies by the consumers' location, sex, and age, the traditional food today yields around 12–21% of the total food intake (Knudsen et al., 2015; Larsen et al., 2019; Terkelsen et al., 2017; Wielsøe et al., 2021). The dietary recommendations in Greenland take the high POP concentrations in some traditional food items into account and recommend a varied food intake including eating animals at the lower part of the marine food chain, frequent consumption of local fish, and less frequent consumption of marine mammals (Bjerregaard and Mulvad, 2012). However, the food recommendations and dietary choices in Greenland are complex due to the cultural role and nutritional benefits of traditional foods (Bjerregaard and Mulvad, 2012; PAME, 2021). Moreover, many imported food items, such as sweetened soft drinks, sweets, chips, and meat with a high content of saturated fat, can be nutrient-poor and may result in higher exposures to other environmental contaminants (Bjerregaard and Mulvad, 2012; Mulvad, 2018). Imported vegetables and fruit with high content of nutrients are also available, but mainly in the larger towns, and nutritious food can be expensive (Bjerregaard and Mulvad, 2012).

PCBs, PBDEs, and the OCPs of this study are, together with perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA), listed by the United Nations Stockholm Convention on POPs resulting in elimination (Annex A) or restricted use (Annex B), while perfluorohexane sulfonate (PFHxS), long-chain perfluorocarboxylic acids (PFCAs), and DP are under review (UNEP, n.d.). Reports show a decreasing trend for many of the lipophilic POPs (lipPOPs; PCBs and OCPs) and the regulated PFAS (PFOS and PFOA) in humans, reflecting effects of regulation and the dietary changes in Greenland (Abass et al., 2018; Long et al., 2021). However, the concentrations of currently unregulated PFAS (e.g. the PFCAs perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), and perfluoroundecanoic acid (PFUnA)) are still increasing in the Greenlandic population (Abass et al., 2018; Long et al., 2021). PBDE concentrations have generally been found to be low in the Greenlandic population compared to those of Europe and other Arctic regions (AMAP, 2016a; Hjermitslev et al., 2019), and no time trends are available for the Greenlandic population. The time trend of POPs is important to follow in the vulnerable Arctic populations. It also supports the evaluation of the effectiveness of the Stockholm Convention where human blood is one of the primary matrices for global monitoring of POPs (UNEP, n.d.).

The ACCEPT birth cohort established in 2010–2015 included pregnant women from all regions in Greenland with the overall objective to study lifestyle, food intake, and environmental contaminants and to investigate related health effects. We have previously reported regional differences in food intake, POP and metal concentrations in the blood of pregnant women, and effects of prenatal exposures on birth outcomes and child development (Bank-Nielsen et al., 2019; Hjermitslev et al., 2019; Knudsen et al., 2015; Kok Grouleff et al., 2021; Kornvig et al., 2021; Long et al., 2015; Terkelsen et al., 2017).

The purpose of this study was to provide updated biomonitoring data of POPs and some new chemicals (e.g. DP) in mothers of the ACCEPT birth cohort in samples from 2019 to 2020 with inclusion of the child's father. We assess the mother's intra-individual changes in POP concentrations from inclusion (2013–2015) to this follow-up 3–5 years later. The study examines sex and geographic differences in POP concentrations and investigates the relationship between contaminant concentrations in serum and lifestyle and dietary factors. To our knowledge, this study provides the most recent data on POP exposure of both men and women in Greenland.

2. Methods

2.1. Study population

The study population has been previously described (Wielsøe et al., 2021). This study is a follow-up with new data on the ACCEPT cohort established in Greenland during 2010–2015 (Knudsen et al., 2015; Terkelsen et al., 2017). In total, 614 Greenlandic pregnant women were recruited, 504 fulfilled the inclusion criteria ($\geq$18 years of age, having lived more than 50% of their lives in Greenland, and having at least one Inuit parent), and 478 completed their pregnancy with available birth outcome (Fig. S1).

During this follow-up study (May 2019 to January 2020), among the 295 live born singleton children aged 3–5 years (children of mothers recruited in 2013–2015), 150 fulfilled the follow-up inclusion criteria that the mothers had lived longest in the ACCEPT regions West or Disko Bay and currently lived in Nuuk, Sisimiut, or Ilulissat. The ACCEPT region West includes the towns Sisimiut, Maniitsoq, Nuuk, and Paamiut and the ACCEPT Disko Bay region includes the towns Qeqertarsuaq, Ilulissat, Qasigiannguit, and Kangaatsiaaq (Knudsen et al., 2015; Terkelsen et al., 2017). The child's biological father was included in the follow-up if possible. We contacted 133 mothers, and 102 agreed to participate (one family participated with two children from two independent pregnancies). Finally, the total study population at follow-up included 101 mothers, 76 fathers, and 102 ACCEPT children (Fig. S1). The participation rate for this follow-

up study was 76.6%, and those who did not wish to participate mainly gave lack of time as the reason.

After receiving a detailed description of the study, all participants gave written informed consent to participate. They were informed that they could withdraw their consent at any time in the process and that their participation was voluntary.

The families received two home visits from a health nurse visitor and project researchers, respectively. At the first visit, the health nurse interviewed the families and filled out the questionnaires. During the second visit, we collected biological samples (blood, hair, urine, and nails).

The study was approved by The Commission for Scientific Investigations in Greenland (KVUG 2019–04) and conducted in line with the Declaration of Helsinki.

2.2. Questionnaire data

The adult participants completed a self-administered questionnaire in Danish or Greenlandic with the possibility of assistance by the health nurse visitor if the participants were in doubt about the meaning of the questions and possible answers. Two independent persons double-entered data from the questionnaire into the EpiData Entry program, and if any discrepancy occurred between the two data-entries, it was solved by consultation of the original questionnaire.

The adult participants' questionnaire was divided into two sections; section 1 contained questions about demographics, lifestyle, and health, and section 2 was a food frequency questionnaire (FFQ) about food intake for the last 12 months.

Data extracted from section 1 included age, history of residences (in Greenland and outside of Greenland), ethnicity, educational level, income, alcohol intake, smoking history, drug (narcotic) use, body mass index (BMI) (from self-reported height and weight), and number of children.

Data from the FFQ (section 2 of the questionnaire) and method for calculation food frequency scores were previously published (Wielsøe et al., 2021). Briefly, food frequency scores (intakes per months) were calculated for seven traditional food groups (marine mammals, seabirds, fish, dried fish, shellfish, terrestrial animals, and berries) and seven imported food groups (meat products, carbohydrate foods, sauce, fruit, vegetables, fast food, and sweets & snacks) by summing the intake scores (times per months) from 42 traditional and 23 imported food items (Wielsøe et al., 2021).

At inclusion in the study during pregnancy (2013–2015), the mothers filled out similar questionnaires but at that time the questionnaires also contained information related to pregnancy lifestyle and diet, age, pre-pregnancy BMI, parity, alcohol intake (before and during pregnancy), and smoking history (never, former, current) (Terkelsen et al., 2017).

2.3. Chemical analyses

2.3.1. Measurement of lipophilic persistent organic pollutants

The chemical analysis of PCBs, OCPs, and HFRs was conducted at Centre de Toxicologie, Institute National de Santé Publique du Québec (INSPQ), a laboratory certified by the Canadian Association for Environmental Analytical Laboratories. The serum samples were analyzed for OCPs [aldrin, alpha-chlordane, cis-nonachlor, gamma-chlordane, hexachlorobenzene (HCB), mirex, oxychlordane, p,p′-dichlorodiphenyldichloroethylene (p,p′-DDE), p,p′-dichlorodiphenyltrichloroethane (p,p′-DDT), β-hexachlorocyclohexane (β-HCH), and trans-nonachlor], PCBs [28, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183, and 187], and HFRs [polybrominated biphenyl (PBB)-153 and PBDEs 15, 17, 25, 28, 33, 47, 99, 100, and 153].

Briefly, prior to extraction, 2 mL serum was spiked with internal standards ($^{13}C_{12}$-PCB141, $^{13}C_{12}$-PCB153, $^{13}C_{12}$-PCB180, $^{13}C_{12}$-PCB194, $^{13}C_6$-HCB, $^{13}C_6$-α-HCH, $^{13}C_{10}$-trans-nonachlor, $^{13}C_{10}$-oxychlordane, $^{13}C_{12}$-p,p′-DDE, $^{13}C_{10}$-Parlar26, $^{13}C_{10}$-Parlar50, $^{13}C_{12}$-PBDE77, 3,6-F_2-PBDE99, and PBDE101). The serum proteins were denatured followed by liquid-liquid extraction. The extracts were evaporated, reconditioned in hexane, and purified on activated florisil columns. The purified extracts were concentrated and analyzed using gas chromatography coupled to a mass spectrometer. Further details have been described elsewhere (Chevrier et al., 2013; Fisher et al., 2016). The limits of detection (LODs) are shown in Table S1.

In 99 samples (57 mothers and 42 fathers, randomly selected), additional HFRs were measured at the Department of Environmental Science, Aarhus University (AU), accredited for PBDEs in biological materials according to DIN EN ISO/IEC 17025. The measured HFRs included PBDEs [17, 28, 47, 66, 85, 99, 100, 153, 154, 183, 209], 2,4,6-tribromophenyl-2,3-dibromopropyl ether (DPTE), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE), and the two isomers of DP (*syn*-DP and anti-DP).

Briefly, prior to extraction, 1 mL serum was spiked with internal standards (^{13}C-PBDE209, ^{13}C-Syn-DP and ^{13}C-Anti-DP) and with ^{13}C-trans-chlordane and PCB198 for recovery determination. The samples were mixed with formic acid and water (*n*-hexan-extracted MilliQ), vortexed and ultrasound-treated, for protein denaturation. The analytes were extracted using solid-phase extraction on a C_{18}-column (Waters, Milford, MA, USA) conditioned with dichloromethane, methanol, methanol:water, water and eluted with *n*-hexane.

The eluates were filtrated over glass wool and Na_2SO_4, evaporated to dryness in silicone-coated vials (Vorkamp et al., 2014) and the compounds were repartitioned into 200 μL isooctane including the quantification standard PBDE71. The extracts were analyzed by gas chromatography and mass spectrometry with electron capture negative ionization (Vorkamp et al., 2015; Vorkamp et al., 2014), combining all analytes on a J&W Scientific DB-1 column (15 m, 0.25 mm inner diameter, 0.1 μm film thickness). The LODs are shown in Table S1.

The PCB, OCP, and HFR concentrations were corrected for total serum lipids (μg/kg lipid). Serum lipids were analyzed at INSPQ, Canada using a standard enzymatic procedure, and the total lipid concentration was calculated using the equation $TL = 1.677\ (TC - FC) + FC + TG + PL$, where TL is total lipids, TC is total cholesterol, FC is free cholesterol, TG is triglycerides, and PL is phospholipids, all expressed in units of g/L (Akins et al., 1989; Phillips et al., 1989).

2.3.2. Measurement of perfluoroalkyl substances

The chemical analysis of PFAS was performed at the Department of Environmental Science, AU. The serum samples were analyzed for perfluorobutane sulfonate (PFBS), perfluorohexane sulfonate (PFHxS), perfluoroheptane sulfonate (PFHpS), perfluorooctane sulfonate (PFOS), perfluorodecane sulfonate (PFDS), perfluorooctane sulfonamide (PFOSA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnA), perfluorododecanoic acid (PFDoA), perfluorotridecanoic acid (PFTrA), and perfluorotetradecanoic acid (PFTeA).

Before extraction, 100 μL serum was spiked with internal standards ($^{13}C_2$-PFHxA, $^{13}C_4$-PFOA, $^{13}C_5$-PFNA, $^{13}C_2$-PFDA, $^{13}C_2$-PFUnA, $^{13}C_2$-PFDoA, $^{13}C_8$-PFOSA, $^{18}O_2$-PFHxS, and $^{13}C_4$-PFOS). The analyses were performed using solid phase extraction followed by liquid chromatography–tandem mass spectrometry as described previously (Bjerregaard-Olesen et al., 2016b; Bonefeld-Jorgensen et al., 2014; Bossi et al., 2005). The limits of quantification (LOQ) are shown in Table S1.

2.3.3. Measurement of fatty acids

Fatty acid (FA) compositions of total plasma phospholipids were determined at Lipid Analytical Laboratories Inc., Guelph, Canada as previously described (Dewailly et al., 2001; Stark and Holub, 2004). Lipids were extracted from the plasma samples (Folch et al., 1957), and the plasma phospholipids were separated from the neutral lipids by thin-layer chromatography (Dewailly et al., 2001; Stark and Holub, 2004). The FAs methyl esters were prepared from the isolated phospholipid fraction (Morrison and Smith, 1964) and were analyzed using gas-liquid chromatography.

The results were expressed as the percentage of total FAs in plasma phospholipids. The sum percentage of the n-3 polyunsaturated FAs [C18:3n-3, C18:4n-3, C20:3n-3, C20:4n-3, C20:5n-3 eicosapentaenoic acid (EPA), C22:5n-3 and C22:6n-3 docosahexaenoic acid (DHA)] and the

sum percentage of the n-6 FAs [C18:2n-6, C18:3n-6, C20:2n-6, C20:3n-6, C20:4n-6 arachidonic acid (AA), C22:2n-6, C22:4n-6 and C22:5n-6] were calculated. The ratio between n-3 and n-6 (n-3/n-6) FAs was calculated as a strong indicator of marine food intake and gives the relative consumption of traditional marine food versus imported food.

2.4. Statistics

All statistical analyses were performed with SPSS software version 27 (SPSS Inc., Chicago, IL, USA). The statistically significant level was set to $p \leq 0.050$ and borderline significant level was set to $p \leq 0.080$.

We report data on individual compounds above LOD (lipPOPs) or LOQ (PFAS) in more than 50% of the samples (see detection frequencies for all compounds in Table S1). For samples with lipPOPs below LOD, we assigned the samples with values of LOD/2 for the statistical analysis. For samples with PFAS below LOQ, we assigned the samples with values of LOQ/2 for the statistical analysis.

We also performed analyses for the summed concentration of the compound groups as follows. ΣPCB: PCB 28, 52, 99, 101, 105, 118, 128, 138, 153, 156, 170, 180, 183, and 187; ΣOCP: aldrin, alpha-chlordane, cis-nonachlor, gamma-chlordane, HCB, mirex, oxychlordane, p,p′-DDE, p,p′-DDT, β-HCH, and trans-nonachlor; ΣPCB + OCP: ΣPCB and ΣOCP; ΣPFCA: PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, PFDoA PFTrA, and PFTeA; ΣPFSA (perfluorosulfonic acid): PFBS, PFHxS, PFHpS, PFOS, PFDS, and PFOSA, and ΣPFAS: ΣPFCA and ΣPFSA.

We checked the distribution of the continuous variables by Q-Q plots and when non-normal distribution was found, variables were ln-transformed to improve the normality.

Differences between two groups were tested with independent samples Student's *t*-test and linear regression upon adjustment for confounders. Differences between more than two groups were tested with ANOVA and ANCOVA upon adjustment for confounders. When we observed significant differences between groups with ANOVA or ANCOVA, Tukey HSD post hoc test was used to further reveal specific differences between the groups.

For categorical variables, Pearson's Chi-square test was used to test the difference between groups. Correlations between exposure variables and lifestyle and socioeconomic factors were analyzed with Spearman correlation.

Associations between exposure variables (POPs) and food intake were analyzed with linear regression, POPs as dependent variables and food intake as independent variable. Based on a priori knowledge of covariates suspected to be related to both food intake and POP concentrations (Agudo et al., 2009; Bjermo et al., 2013; Bjerregaard-Olesen et al., 2016a; Bjerregaard and Aidt, 2010; Dahl-Petersen et al., 2016; Deutch et al., 2003; Hjermitslev et al., 2019; Long et al., 2015; Tsai et al., 2018), the following variables were included in the models: age (continuous), town (categorical), sex (categorical), educational level (categorical), house income (categorical), and smoking history (categorical).

The analyses of exposure differences within mother-father pairs were restricted to mothers and fathers living together at the time of follow-up ($n = 73$). To assess the exposure differences within the individual mother-father pair, we subtracted the paternal concentrations from the maternal concentrations (mother's concentration – father's concentration). To assess exposure changes from inclusion at pregnancy (2013–2015) to follow-up for the mothers (3–5 years after inclusion: 2019–2020), the individual differences were calculated by subtracting the concentration measured at inclusion from the concentration measured at follow-up. Wilcoxon Signed Rank test (median equal to 0) was used to test mother-father-pair differences and changes from inclusion at pregnancy to follow-up (for mothers only) in exposure concentrations. Furthermore, we assessed the pairwise correlations of POP concentrations between mother-father-pairs and between inclusion and follow-up for the mothers using Pearson correlations. We also calculated partial Pearson correlations by adjusting for different predictors of the exposure concentrations. The correlations between the mother-father-pairs were adjusted for factors known to influence the POP concentrations (Agudo et al., 2009; Bjermo et al., 2013; Bjerregaard-Olesen et al., 2016a; Deutch et al., 2003; Hjermitslev et al., 2019; Long et al., 2015; Tsai et al., 2018): age difference between the parents (continuous), household income (categorical), percentage of life lived in Greenland (continuous), highest educational level (categorical), and smoking status for both the father and mother. The correlations between inclusion at pregnancy and follow-up of the mother were adjusted for the following factors known to influence the POP concentrations (Agudo et al., 2009; Bjermo et al., 2013; Bjerregaard-Olesen et al., 2016a; Deutch et al., 2003; Hjermitslev et al., 2019; Long et al., 2015; Tsai et al., 2018): the time from inclusion to follow-up (continuous), change in education level (categorical), change in household income (categorical), change in smoking status (categorical), change in BMI (continuous), and change in parity (categorical).

3. Results

3.1. Characteristics of the study population

Table 1 presents the characteristics of the participants (101 mothers/76 fathers). The mothers were younger than the fathers. Most of the participants lived in Nuuk (67.2%), while 21.5% and 10.7% lived in Sisimiut and Ilulissat, respectively. Due to the follow-up criteria, all mothers had lived longest in either Disko Bay or West region, while some fathers had lived longest in other regions (11.4%) or outside Greenland (7.1%). However, no differences between fathers and mothers were seen for the percentages of life lived in Greenland. Concerning the ethnic background, 80.0% were Inuit, 17.1% partly-Inuit, and 2.9% non-Inuit. An inclusion criterion for participating in the ACCEPT birth cohort was that the pregnant women were Inuit or partly-Inuit; thus, all non-Inuit participants in the present study were fathers (Table 1).

A high percentage of the study population was overweight (41.7%) or obese (34.7%) with similar distribution among fathers and mothers. A higher percentage of mothers had a university degree and had never used hash and drugs (narcotics), but less had a personal income of more than 250.000 DKK per year (Table 1). There were no significant differences between fathers and mothers regarding household income, alcohol consumption, smoking, or number of children (Table 1).

3.2. POP concentrations

Blood samples for POP measurements were available for 166 participants (95 mothers/71 fathers). Table 2 shows percentage above LOD, and concentrations of the measured PCBs, OCPs, HFRs and PFAS. The median concentrations of ΣPCB, ΣOCP, and Σ(PCB + OCP) were 147, 177, and 322 μg/kg lipid, receptively, and the median concentrations of ΣPFCA, ΣPFSA, and ΣPFAS were 2.89, 6.88, and 9.73 ng/mL, respectively (Table 2).

Only compounds detected in more than 50% of the samples were included in the analyses of individual compounds. The following compounds were detected in less than 50% of the samples and not included in analyses of individual compounds: PCB28, PCB52, PCB99, PCB101, PCB105, PCB128, aldrin, alpha-chlordane, gamma-chlordane, p,p′-DDT, PBB153, PBDE15, PBDE17, PBDE25, PBDE33, PBDE47, PBDE66, PBDE85, PBDE99, PBDE100, PBDE154, PBDE183, PBDE209, DPTE, EHTBB, BTBPE, *syn*-DP, PFBS, PFDS, PFOSA, PFHxA, PFHpA, PFDoA, PFTrA, and PFTeA (Table S1). PBDE28 and PBDE153 were detected in 50.5% and 76.8% of the samples analyzed at AU, respectively, but in less than 50% of samples in the chemical analyses at INSPQ (0.00–3.10% > LOD), thus, the data from AU were used in statistical analyses including PBDE28 and PBDE153. The measurements at INSPQ and AU are compared (for the samples measure above LOD at INSPQ) in Table S2. For these 10 samples, the concentrations were slightly higher in the measurements at INSPQ than at AU (Table S2).

3.2.1. Changes in POP concentrations from pregnancy to follow-up (mothers only)

All PCB, OCP (except mirex), and PFAS concentrations significantly decreased from inclusion at pregnancy to follow-up (Table 3). The median percentage of intra-individual decrease in concentrations from inclusion to follow-up was 20–32% for PCBs, 16–37% for OCPs (0% for mirex), and

Table 1
Characteristics of the study population.

		Fathers (N = 76)	Mothers (N = 101)	p-Value[a]	All (N = 177)
Age (years)	Mean (SD)	37.2 (7.5)	33.8 (4.8)	**<0.001**[b]*	35.3 (6.3)
	Median (IQR)	36.7 (11.4)	34.0 (7.5)		34.3 (8.9)
	Missing n (%)	0 (0%)	0 (0%)		0 (0%)
Current residential town					
Nuuk	n (%)	50 (65.8%)	69 (68.3%)	0.956[c]	119 (67.2%)
Sisimiut	n (%)	17 (22.4%)	21 (20.8%)		38 (21.5%)
Ilulissat	n (%)	8 (10.5%)	11 (10.9%)		19 (10.7%)
Qaqortoq	n (%)	1 (1.3%)	0 (0.0%)		1 (0.6%)
	Missing n (%)	0 (0%)	0 (0%)		0 (0%)
Region lived longest					
Disko Bay	n (%)	13 (18.6%)	25 (26.6%)	**<0.001**[c]*	38 (23.2%)
West	n (%)	44 (62.9%)	69 (73.4%)		113 (68.9%)
Other region in Greenland[d]	n (%)	8 (11.4%)	0 (0.0%)		8 (4.9%)
Outside Greenland	n (%)	5 (7.1%)	0 (0.0%)		5 (3.0%)
	Missing n (%)	6 (8%)	7 (7%)		13 (7%)
Percentage of life lived in Greenland (%)	Mean (SD)	89.7 (20.1)	95.0 (6.8)	0.503[b]	92.7 (14.3)
	Median (IQR)	97.8 (7.8)	97.5 (5.5)		97.5 (5.4)
	Missing n (%)	1 (1%)	1 (1%)		2 (1%)
Ethnic background[e]					
Inuit	n (%)	61 (80.3%)	79 (79.8%)	**0.021**[c]*	140 (80.0%)
Partly-Inuit	n (%)	10 (13.2%)	20 (20.2%)		30 (17.1%)
Non-Inuit	n (%)	5 (6.6%)	0 (0.0%)		5 (2.9%)
	Missing n (%)	0 (0%)	2 (2%)		2 (1%)
BMI (kg/m^2)	Mean (SD)	29.3 (4.9)	28.1 (5.3)	0.131[b]	28.6 (5.1)
	Median (IQR)	28.6 (6.4)	26.4 (6.3)		27.8 (6.3)
	Missing n (%)	12 (16%)	21 (21%)		33 (19%)
Underweight (<18.5)	n (%)	0 (0.0%)	0 (0.0%)	0.203[c]	0 (0.0%)
Normal (18.5–24.9)	n (%)	12 (18.8%)	22 (27.5%)		34 (23.6%)
Overweight (25.0–29.9)	n (%)	25 (39.1%)	35 (43.8%)		60 (41.7%)
Obese (≥30.0)	n (%)	27 (42.2%)	23 (28.7%)		50 (34.7%)
Highest education level					
Primary school	n (%)	16 (21.3%)	21 (21.2%)	**0.006**[c]*	37 (21.3%)
High School	n (%)	6 (8.0%)	8 (8.1%)		14 (8.0%)
Technical college	n (%)	41 (54.7%)	32 (32.3%)		73 (42.0%)
University	n (%)	12 (16.0%)	38 (38.4%)		50 (28.7%)
	Missing n (%)	1 (1%)	2 (2%)		3 (2%)
Personal income per year					
Under 100.000 DKK	n (%)	5 (6.8%)	15 (15.2%)	**0.014**[c]*	20 (11.6%)
100.000–250.000 DKK	n (%)	10 (13.5%)	20 (20.2%)		30 (17.3%)
Over 250.000 DKK	n (%)	56 (75.7%)	52 (52.5%)		108 (62.4%)
Don't know[f]	n (%)	3 (4.1%)	12 (12.1%)		15 (8.7%)
	Missing n (%)	1 (1%)	3 (3%)		4 (2%)
Household income per year					
Under 100.000 DKK	n (%)	2 (2.7%)	2 (2.0%)	0.311[c]	4 (2.3%)
100.000–250.000 DKK	n (%)	5 (6.7%)	9 (9.2%)		14 (8.1%)
Over 250.000 DKK	n (%)	65 (86.7%)	76 (77.6%)		141 (81.5%)
Don't know[f]	n (%)	3 (4.0%)	11 (11.2%)		14 (8.1%)
	Missing n (%)	1 (1%)	3 (3%)		4 (2.3%)
Current alcohol consumption					
0 drinks/week	n (%)	38 (53.5%)	34 (55.7%)	0.298[c]	72 (54.5%)
1–7 drinks/week	n (%)	20 (28.2%)	22 (36.1%)		42 (31.8%)
≥8 drinks/week	n (%)	5 (7.0%)	3 (4.9%)		8 (6.1%)
Don't know[f]	n (%)	8 (11.3%)	2 (3.3%)		10 (7.6%)
	Missing n (%)	5 (7%)	40 (40%)		45 (25.4%)
Smoking history					
Never smoker	n (%)	37 (48.7%)	49 (48.5%)	0.507[c]	86 (48.6%)
Former smoker	n (%)	20 (26.3%)	33 (32.7%)		53 (29.9%)
Current smoker	n (%)	19 (25.0%)	19 (18.8%)		38 (21.5%)
	Missing n (%)	0 (0%)	0 (0%)		0 (0%)
Hash use					
Have used hash	n (%)	45 (60.0%)	42 (42.0%)	**0.050**[c]*	87 (49.7%)
Never used hash	n (%)	28 (37.3%)	56 (56.0%)		84 (48.0%)
Don't know[f]	n (%)	2 (2.7%)	2 (2.0%)		4 (2.3%)
	Missing n (%)	1 (1%)	1 (1%)		2 (1%)
Drugs (narcotics) use					
Have used drugs (narcotics)	n (%)	17 (23.3%)	3 (3.1%)	**<0.001**[c]*	20 (11.8%)
Never used drugs (narcotics)	n (%)	55 (75.3%)	92 (95.8%)		147 (87.0%)
Don't know[f]	n (%)	1 (1.4%)	1 (1.0%)		2 (1.2%)
	Missing n (%)	3 (4%)	5 (5%)		8 (5%)
Number of children	Mean (SD)	2.5 (1.2)	2.4 (1.1)	0.723[b]	2.4 (1.1)
	Median (IQR)	2.0 (1.0)	2.0 (1.0)		2.0 (1.0)
	Missing n (%)	0 (0%)	0 (0%)		0 (0%)
1	n (%)	12 (15.8%)	19 (18.8%)	0.365[c]	31 (17.5%)
2–3	n (%)	55 (72.4%)	69 (68.3%)		124 (70.1%)
4 or more	n (%)	9 (11.8%)	13 (12.9%)		22 (12.4%)

[a] p-Value for statistical test between fathers and mothers.
[b] Students independent t-test.
[c] Chi-square test.
[d] Other region in Greenland include North (3 fathers), South (4 fathers), and East (1 father).
[e] Ethnic background include Inuit (both parents Greenlandic), Partly-Inuit (one Greenlandic parent), and Non-Inuit (no Greenlandic parents).
[f] The participants answered "Don't know" to the question. Bold text and * indicate significant difference ($p \leq 0.050$). N: total number of participant in the group. n (%): number of participants and valid percentages of the number of participant with information on the variable. IQR: Interquartile range. SD: Standard Deviation.

31–58% for PFAS, for both individual compounds and the summed compound groups (Table 3).

Pearson correlations for the mother's PCB, OCP, and PFAS concentrations between inclusion at pregnancy and this follow-up study were all positive, indicating positive correlation between the POP concentrations at the two time points. All the correlations were significant and strong with adjusted correlation coefficients from 0.404 to 0.897 (Table 3).

3.2.2. Differences in POP concentrations in fathers and mothers

All PCBs and OCPs were significantly higher in fathers than in mothers, except for PCB118 (Fig. 1, Table S3). For β-HCH and trans-nonachlor, the

Table 2
Detection frequencies and concentrations of persistent organic pollutant for the study population.

	Fathers (N = 76)[a]				Mothers (N = 101)[b]				All (N = 177)[c]			
	% > LOD[d]	Mean (SD)	Median (IQR)	Min-Max	% > LOD[d]	Mean (SD)	Median (IQR)	Min-Max	% > LOD[d]	Mean (SD)	Median (IQR)	Min-Max
Lipophilic POPs (μg/kg lipid)												
PCB 118	95.7%	11.5 (15.0)	9.10 (10.7)	1.00–110	92.5%	7.79 (5.90)	6.20 (7.60)	0.400–30.0	93.9%	9.40 (10.9)	7.40 (8.40)	0.400–110
PCB 138	100.0%	36.2 (37.4)	29.0 (30.5)	5.10–270	100.0%	19.1 (13.8)	16.0 (14.9)	1.40–75.0	100.0%	26.5 (27.9)	19.0 (22.0)	1.40–270
PCB 153	100.0%	78.2 (84.9)	61.0 (58.8)	9.90–630	100.0%	39.2 (28.6)	34.0 (30.0)	2.50–170	100.0%	55.9 (62.5)	41.0 (51.0)	2.50–630
PCB 156	87.1%	4.60 (4.14)	3.45 (4.25)	0.500–24.0	61.3%	2.11 (1.48)	1.80 (1.90)	0.400–7.60	72.4%	3.18 (3.17)	2.30 (2.80)	0.400–24.0
PCB 170	100.0%	13.5 (11.1)	10.0 (12.2)	1.80–66.0	96.8%	6.38 (4.17)	5.80 (5.00)	0.400–21.0	98.2%	9.44 (8.64)	6.90 (7.00)	0.400–66.0
PCB 180	100.0%	42.1 (37.4)	30.5 (34.5)	4.80–220	100.0%	20.2 (13.9)	17.0 (16.4)	1.90–68.0	100.0%	29.6 (28.7)	21.0 (24.0)	1.90–220
PCB 183	84.3%	4.05 (3.64)	3.40 (3.43)	0.500–23.0	63.4%	2.34 (1.67)	2.10 (2.00)	0.400–8.10	92.4%	3.07 (2.82)	2.50 (3.10)	0.400–23.0
PCB 187	100.0%	18.4 (16.6)	15.0 (17.0)	1.60–100	98.9%	9.68 (6.87)	8.30 (7.60)	0.500–35.0	99.4%	13.4 (12.8)	10.0 (11.7)	0.500–100
cis-Nonachlor	95.7%	14.2 (16.3)	12.0 (14.0)	0.300–110	98.9%	6.98 (5.66)	5.10 (7.00)	0.300–30.0	97.5%	10.2 (12.0)	6.90 (10.9)	0.300–110
HCB	100.0%	40.8 (48.9)	32.0 (33.3)	4.80–380	100.0%	26.7 (18.9)	22.0 (20.0)	3.80–110	100.0%	32.7 (35.6)	24.0 (28.0)	3.80–380
Mirex	87.1%	5.35 (5.41)	4.35 (4.05)	0.450–31.0	69.9%	3.00 (2.46)	2.30 (3.10)	0.400–12.0	77.3%	4.01 (4.15)	2.80 (30.6)	0.400–31.0
Oxychlordane	98.6%	33.7 (64.5)	23.5 (27.4)	0.350–530	98.9%	14.6 (12.8)	11.0 (13.9)	0.300–63.0	98.8%	22.8 (44.2)	14.0 (22.4)	0.300–530
p,p′-DDE	100.0%	174 (208)	135 (147)	11.0–1600	100.0%	95.3 (73.3)	72.0 (79.0)	6.10–350	100.0%	129 (151)	91.0 (119)	6.10–1600
β-HCH	74.3%	4.94 (7.20)	4.10 (4.80)	0.450–57.0	58.1%	2.72 (2.35)	2.20 (2.80)	0.400–14.0	65.0%	3.67 (5.14)	2.60 (3.70)	0.400–57.0
trans-Nonachlor	98.6%	78.1 (103)	62.0 (75.0)	0.500–760	100.0%	36.7 (30.8)	28.0 (36.0)	2.40–170	99.4%	54.5 (74.0)	37.0 (55.0)	0.500–760
PBDE28[e]	42.9%	1.50 (1.37)	0.884 (1.18)	0.360–7.53	56.1%	3.08 (4.54)	1.67 (2.13)	0.420–26.2	50.51%	2.41 (3.63)	1.00 (1.56)	0.36–26.2
PBDE153[e]	85.7%	2.72 (3.63)	1.97 (1.32)	0.610–24.8	70.2%	2.05 (1.27)	1.95 (1.57)	0.520–7.55	76.77%	2.34 (2.56)	1.95 (1.43)	0.52–24.8
Anti-DP	61.9%	2.46 (2.44)	1.77 (2.07)	0.490–10.4	54.4%	1.94 (2.07)	1.32 (1.38)	0.510–11.6	57.58%	2.16 (2.24)	1.42 (1.62)	0.49–11.6
ΣPCBs	n/a	249 (222)	206 (179)	48.4–1617	n/a	143 (79.6)	130 (86.4)	23.2–457	n/a	189 (165)	147 (138)	23.2–1617
ΣOCPs	n/a	356 (449)	290 (313)	31.4–3492	n/a	192 (141)	155 (163)	17.2–698	n/a	262 (321)	177 (233)	17.2–3492
Σ(PCB + OCP)	n/a	604 (664)	518 (483)	101–5109	n/a	335 (217)	278 (231)	40.3–1086	n/a	450 (482)	322 (367)	40.3–5109
PFAS (ng/mL)												
PFHxS	100.0%[d]	0.934 (0.804)	0.739 (0.539)	0.163–5.67	98.9%[d]	0.315 (0.199)	0.273 (0.197)	0.040–1.30	99.4%[d]	0.579 (0.625)	0.404 (0.477)	0.040–5.67
PFHpS	95.7%[d]	0.292 (0.250)	0.252 (0.194)	0.055–1.86	48.9%[d]	0.114 (0.078)	0.055 (0.099)	0.055–0.517	68.9%[d]	0.190 (0.194)	0.143 (0.190)	0.055–1.86
PFOS	100.0%[d]	10.0 (12.4)	7.49 (5.76)	1.75–103	100.0%[d]	5.221 (3.779)	4.12 (3.35)	1.23–29.8	100.0%[d]	7.27 (8.87)	5.44 (5.38)	1.23–103
PFOA	100.0%[d]	1.02 (0.369)	1.01 (0.570)	0.425–2.16	92.6%[d]	0.436 (0.225)	0.415 (0.251)	0.100–1.35	95.7%[d]	0.687 (0.414)	0.592 (0.559)	0.100–2.16
PFNA	100.0%[d]	1.37 (1.45)	1.15 (0.755)	0.350–12.0	97.9%[d]	0.786 (0.499)	0.679 (0.470)	0.135–3.77	98.8%[d]	1.04 (1.05)	0.819 (0.640)	0.135–12.0
PFDA	100.0%[d]	0.722 (1.17)	0.509 (0.487)	0.066–10.0	100.0%[d]	0.529 (0.376)	0.445 (0.377)	0.117–3.07	100.0%[d]	0.611 (0.821)	0.463 (0.435)	0.066–10.0
PFUnA	94.3%[d]	0.970 (1.77)	0.666 (0.812)	0.075–14.9	100.0%[d]	0.800 (0.626)	0.659 (0.678)	0.115–4.83	97.6%[d]	0.873 (1.25)	0.661 (0.715)	0.075–14.9
ΣPFCA	n/a	4.21 (3.90)	3.62 (1.88)	1.70–33.9	n/a	2.92 (1.52)	2.64 (1.38)	1.07–13.1	n/a	3.47 (2.86)	2.89 (1.89)	1.07–33.9
ΣPFSA	n/a	12.1 (13.1)	9.63 (6.07)	3.06–109	n/a	6.47 (4.02)	5.28 (3.57)	2.14–32.3	n/a	8.85 (9.45)	6.88 (6.07)	2.14–110
ΣPFAS	n/a	16.3 (16.9)	13.4 (8.07)	4.83–143	n/a	9.38 (5.47)	8.08 (4.79)	3.21–45.4	n/a	12.3 (12.3)	9.73 (8.02)	3.21–143

[a] n for the fathers: PCBs, OCPs, and PFAS, n = 70 (92.2%); HFRs, n = 42 (55.3%).
[b] n for the mothers: PCBs and OCPs, n = 93 (92.1%); HFRs, n = 57 (56.4%); PFAS, n = 94 (93.1%).
[c] n for all: PCBs and OCPs, n = 163 (92.1%); HFRs, n = 99 (54.9%); PFAS, n = 164 (92.7%).
[d] Percentage above LOQ was given for PFAS.
[e] PBDE28 and PBDE153 were measured at both INSPQ and AU, but the data from AU are displayed here, as all samples were below LOD in the measurement at INSPQ (see Table S1, see also Table S2, with the agreement between measurements at INSPQ and AU). n (%): number of participants with information and percentages of the total number of participant in the group (N). n/a: not applicable. Anti-DP: Anti-isomer dechlorane plus. HCB: Hexachlorobenzene. IQR: Interquartile range. OCP: organochlorine pesticides. p,p′-DDE: p,p′-dichlorodiphenyldichloroethylene. PBDE: Polybrominated diphenyl ether. PCB: polychlorinated biphenyl. PFAS: *per*- and polyfluoroalkyl substances. PFCA: perfluorocarboxylic acids. PFDA: perfluorodecanoic acid. PFHpS: perfluoroheptane sulfonate. PFHxS: perfluorohexane sulfonate. PFNA: perfluorononanoic acid. PFOA: perfluorooctanoic acid. PFOS: perfluorooctane sulfonate. PFSA: perfluorosulfonic acid. PFUnA: perfluoroundecanoic acid. SD: Standard Deviation. β-HCH: β-hexachlorocyclohexane.

Table 3
Changes and correlations of persistent organic pollutant concentrations for the mothers from inclusion to follow-up (3–5 years).

	Individual difference (follow-up – inclusion)						Unadjusted Pearson correlation between inclusion and follow-up			Adjusted Pearson correlation between inclusion and follow-up		
	n (%)	Change in concentration			Change in percentage			n	*r*	*p*-Value	n	*r*	*p*-Value
		Mean (SD)	Median (IQR)	*p*-Value	Mean (SD)	Median (IQR)	*p*-Value						
Lipophilic POPs (μg/kg lipid)													
PCB 118	94 (92.2%)	−2.44 (4.79)	−2.00 (4.08)	**<0.001***	−18.2% (47.4)	−22.9% (46.0)	**<0.001***	94 (92.2%)	0.829	**<0.001***	46 (45.1%)	0.849	**<0.001***
PCB 138	94 (92.2%)	−8.55 (13.4)	−6.00 (10.7)	**<0.001***	−24.8% (43.6)	−30.4% (33.8)	**<0.001***	94 (92.2%)	0.804	**<0.001***	46 (45.1%)	0.844	**<0.001***
PCB 153	94 (92.2%)	−19.8 (28.9)	−12.0 (23.5)	**<0.001***	−27.4% (35.4)	−30.2% (36.5)	**<0.001***	94 (92.2%)	0.828	**<0.001***	46 (45.1%)	0.866	**<0.001***
PCB 156	94 (92.2%)	−1.31 (2.00)	−0.800 (2.00)	**<0.001***	−24.7% (39.2)	−28.1% (54.6)	**<0.001***	94 (92.2%)	0.769	**<0.001***	46 (45.1%)	0.786	**<0.001***
PCB 170	94 (92.2%)	−3.23 (4.71)	−2.00 (3.50)	**<0.001***	−28.0% (30.7)	−32.2% (35.2)	**<0.001***	94 (92.2%)	0.842	**<0.001***	46 (45.1%)	0.875	**<0.001***
PCB 180	94 (92.2%)	−8.86 (13.9)	−5.00 (8.88)	**<0.001***	−24.5% (28.0)	−28.2% (35.9)	**<0.001***	94 (92.2%)	0.870	**<0.001***	46 (45.1%)	0.897	**<0.001***
PCB 183	94 (92.2%)	−0.882 (1.38)	−0.600 (1.50)	**<0.001***	−18.0% (48.9)	−25.7% (46.4)	**<0.001***	94 (92.2%)	0.811	**<0.001***	46 (45.1%)	0.814	**<0.001***
PCB 187	94 (92.2%)	−3.59 (5.80)	−2.30 (5.05)	**<0.001***	−22.4% (34.1)	−25.0% (32.7)	**<0.001***	94 (92.2%)	0.849	**<0.001***	46 (45.1%)	0.882	**<0.001***
cis-Nonachlor	94 (92.2%)	−2.07 (4.38)	−1.45 (3.73)	**<0.001***	−18.0% (49.1)	−21.3% (45.2)	**<0.001***	94 (92.2%)	0.839	**<0.001***	46 (45.1%)	0.873	**<0.001***
HCB	94 (92.2%)	−4.50 (14.2)	−4.15 (12.2)	**<0.001***	−9.40% (40.0)	−15.7% (47.9)	**0.002***	94 (92.2%)	0.785	**<0.001***	46 (45.1%)	0.814	**<0.001***
Mirex	94 (92.2%)	−0.0640 (1.33)	0.00 (1.02)	0.735	15.0% (65.0)	0.00% (34.2)	0.343	94 (92.2%)	0.856	**<0.001***	46 (45.1%)	0.866	**<0.001***
Oxychlordane	94 (92.2%)	−6.75 (12.3)	−3.40 (10.1)	**<0.001***	−24.1% (52.0)	−29.1% (45.0)	**<0.001***	94 (92.2%)	0.816	**<0.001***	46 (45.1%)	0.856	**<0.001***
p,p′-DDE	94 (92.2%)	−41.9 (66.6)	−28.0 (66.7)	**<0.001***	−23.6% (47.7)	−29.7% (42.1)	**<0.001***	94 (92.2%)	0.818	**<0.001***	46 (45.1%)	0.842	**<0.001***
β-HCH	94 (92.2%)	−1.38 (2.21)	−1.20 (2.05)	**<0.001***	−28.3% (59.7)	−36.9% (47.1)	**<0.001***	94 (92.2%)	0.673	**<0.001***	46 (45.1%)	0.691	**<0.001***
trans-Nonachlor	94 (92.2%)	−11.7 (24.9)	−6.65 (19.8)	**<0.001***	−18.0% (50.5)	−23.0% (47.4)	**<0.001***	94 (92.2%)	0.845	**<0.001***	46 (45.1%)	0.881	**<0.001***
ΣPCBs	94 (92.2%)	−45.8 (76.7)	−26.9 (65.6)	**<0.001***	−16.3% (30.2)	−19.8% (35.6)	**<0.001***	94 (92.2%)	0.829	**<0.001***	46 (45.1%)	0.855	**<0.001***
ΣOCPs	94 (92.2%)	−68.5 (121)	−49.6 (107)	**<0.001***	−20.3% (44.7)	−26.6% (39.1)	**<0.001***	94 (92.2%)	0.818	**<0.001***	46 (45.1%)	0.848	**<0.001***
Σ(PCB + OCP)	94 (92.2%)	−114 (193)	−78.5 (172)	**<0.001***	−18.9% (36.8)	−24.6% (35.5)	**<0.001***	94 (92.2%)	0.822	**<0.001***	46 (45.1%)	0.851	**<0.001***
PFAS (ng/mL)													
PFHxS	95 (93.1%)	−0.125 (0.212)	−0.111 (0.203)	**<0.001***	−22.0% (50.2)	−33.3% (44.2)	**<0.001***	95 (93.1%)	0.613	**<0.001***	47 (46.1%)	0.617	**<0.001***
PFHpS	95 (93.1%)	−0.046 (0.092)	−0.057 (0.102)	**<0.001***	−10.9% (72.5)	−30.6% (58.8)	**0.001***	95 (93.1%)	0.341	**0.001***	47 (46.1%)	0.420	**0.003***
PFOS	95 (93.1%)	−3.79 (3.42)	−3.24 (4.21)	**<0.001***	−41.6% (24.6)	−45.2% (30.7)	**<0.001***	95 (93.1%)	0.716	**<0.001***	47 (46.1%)	0.750	**<0.001***
PFOA	95 (93.1%)	−0.700 (0.907)	−0.508 (0.626)	**<0.001***	−49.3% (40.3)	−57.8% (32.9)	**<0.001***	95 (93.1%)	0.351	**<0.001***	47 (46.1%)	0.404	**0.005***
PFNA	95 (93.1%)	−0.405 (0.412)	−0.349 (0.440)	**<0.001***	−32.5% (26.4)	−34.1% (33.8)	**<0.001***	95 (93.1%)	0.668	**<0.001***	47 (46.1%)	0.686	**<0.001***
PFDA	95 (93.1%)	−0.314 (0.379)	−0.239 (0.395)	**<0.001***	−33.1% (27.1)	−38.3% (38.3)	**<0.001***	95 (93.1%)	0.715	**<0.001***	47 (46.1%)	0.755	**<0.001***
PFUnA	95 (93.1%)	−1.09 (1.43)	−0.506 (1.59)	**<0.001***	−42.6% (33.5)	−49.2% (46.8)	**<0.001***	95 (93.1%)	0.610	**<0.001***	47 (46.1%)	0.663	**<0.001***
ΣPFCA	95 (93.1%)	−2.67 (2.90)	−1.71 (2.75)	**<0.001***	−40.2% (26.0)	−41.7% (33.0)	**<0.001***	95 (93.1%)	0.446	**<0.001***	47 (46.1%)	0.536	**<0.001***
ΣPFSA	95 (93.1%)	−3.97 (3.66)	−3.48 (4.44)	**<0.001***	−36.9% (23.3)	−38.8% (29.8)	**<0.001***	95 (93.1%)	0.709	**<0.001***	47 (46.1%)	0.744	**<0.001***
ΣPFAS	95 (93.1%)	−6.64 (5.65)	−5.30 (6.99)	**<0.001***	−39.5% (22.3)	−42.4% (31.2)	**<0.001***	95 (93.1%)	0.651	**<0.001***	47 (46.1%)	0.697	**<0.001***

The analyses were conducted on mothers only. The individual differences were calculated by subtraction of the concentration from inclusion from the concentration at follow-up, thus a differences of zero (0) reflect a stable concentration at inclusion and follow-up, negative values reflect a decrease in concentration from inclusion to follow-up, and positive value reflect an increase in concentration from inclusion to follow-up. The individual differences were tested with a Wilcoxon Signed Rank test, with median equals 0. Bold text indicates significant difference ($p \leq 0.050$). Pearson correlations between concentration at inclusion and follow-up (3–5 years). Adjusted correlations is adjusted for time from inclusion to follow-up, change in education level, change in household income, change in BMI, and change in parity, change in smoking status. HCB: Hexachlorobenzene. IQR: interquartile range. n (%): number of participants with information and percentages of the total number of participant in the group (N). OCP: organochlorine pesticides. p,p′-DDE: p,p′-dichlorodiphenyldichloroethylene. PCB: polychlorinated biphenyl. PFAS: *per*- and polyfluoroalkyl substances. PFCA: perfluorocarboxylic acids. PFDA: perfluorodecanoic acid. PFHpS: perfluoroheptane sulfonate. PFHxS: perfluorohexane sulfonate. PFNA: perfluorononanoic acid. PFOA: perfluorooctanoic acid. PFOS: perfluorooctane sulfonate. PFSA: perfluorosulfonic acid. PFUnA: perfluoroundecanoic acid. r: Pearson correlation coefficient. SD: Standard Deviation. β-HCH: β-hexachlorocyclohexane.

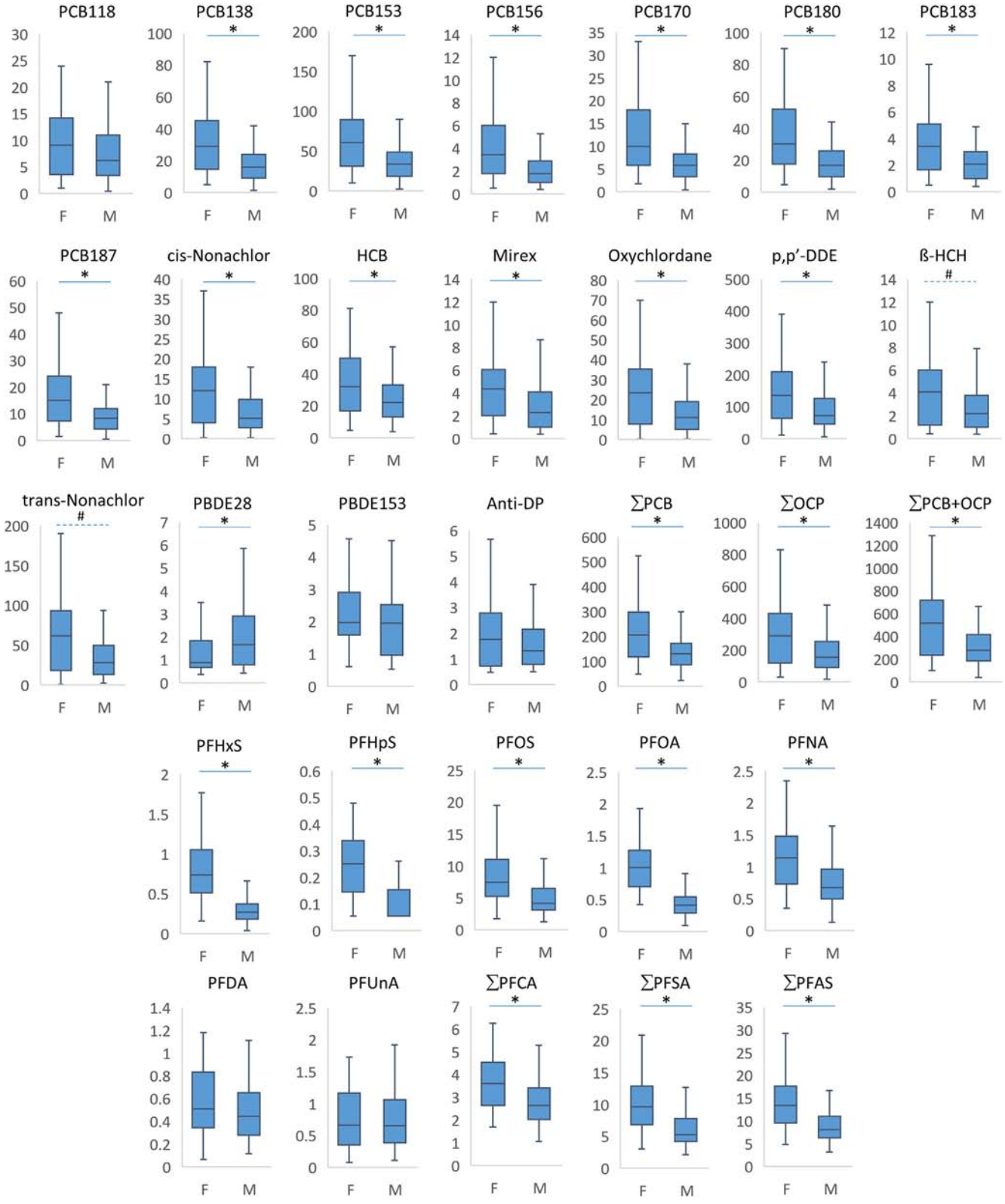


Fig. 1. Concentrations of persistent organic pollutant in fathers (F) and mothers (M). The lipPOPs concentrations are in μg/kg lipid and PFASs concentrations are in ng/mL. Differences between fathers (F) and mothers (M) were tested by linear regression with adjustment for age and town on ln-transformed variables. * and solid line indicate significant difference ($p \leq 0.050$) and # and dotted line indicate borderline significant difference ($p \leq 0.080$). The boxplots consist of the interquartile range (IQR) and the median (line inside the box), and whiskers are 1.5*IQR. Outliers are not included in the graphs (defined as values greater than 1.5*IQR). See Table S3 for concentrations and raw and adjusted *p*-values. PBDE28 and PBDE153 were measured at both INSPQ and AU, but the data from AU are displayed here, as most samples were below LOD in the measurement at INSPQ (see Table S1, see also Table S2, with the agreement between measurements at INSPQ and AU). Anti-DP: Anti-isomer dechlorane plus. F: Fathers. HCB: Hexachlorobenzene. M: Mothers. OCP: organochlorine pesticides. p,p′-DDE: p,p′-dichlorodiphenyldichloroethylene. PBDE: Polybrominated diphenyl ether. PCB: polychlorinated biphenyl. PFCA: perfluorocarboxylic acids. PFDA: perfluorodecanoic acid. PFHpS: perfluoroheptane sulfonate. PFHxS: perfluorohexane sulfonate. PFNA: perfluorononanoic acid. PFOA: perfluorooctanoic acid. PFOS: perfluorooctane sulfonate. PFSA: perfluorosulfonic acid. PFUnA: perfluoroundecanoic acid. β-HCH: β-hexachlorocyclohexane.

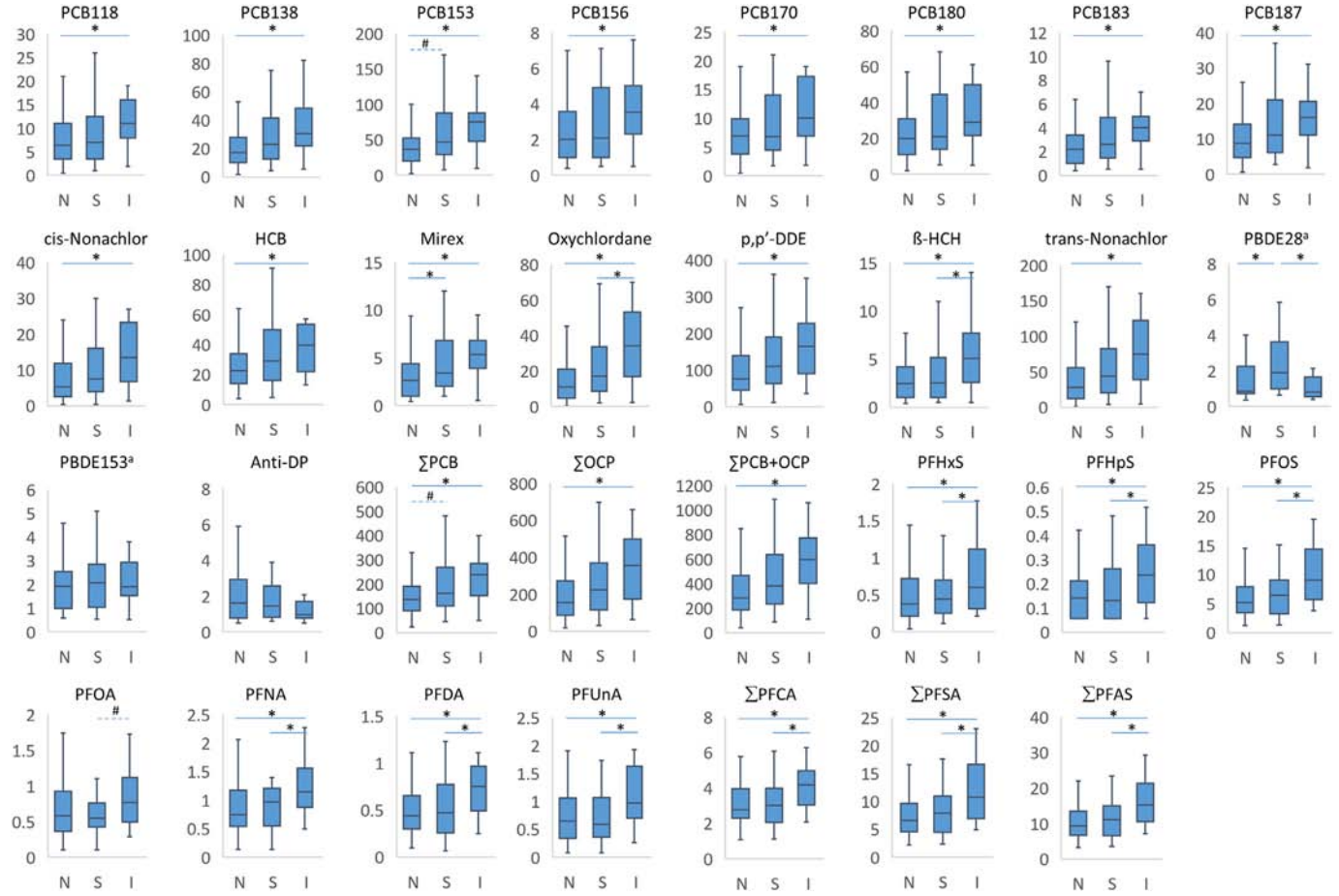


Fig. 2. Concentrations of persistent organic pollutants by residential town. The concentrations of the lipPOPs are shown in μg/kg lipid and concentrations of the PFASs are shown in ng/mL. Differences between towns (I: Ilulissat, N: Nuuk, and S: Sisimiut) were tested with ANCOVA on ln transformed variables adjusted for age and sex, and were all significant. Tukey HSD post hoc test were used to test the specific differences between towns. * and solid line indicate significant difference between the towns in the post hoc test ($p \leq 0.050$): # and dotted line indicate borderline significant difference between the towns in the post hoc test ($p \leq 0.080$); The boxplots consist of the interquartile range (IQR) and the median (line inside the box), and whiskers are 1.5*IQR. Outliers are not included in the graphs (defined as values greater than 1.5*IQR). See Table S5 for concentrations and raw and adjusted *p*-values. PBDE28 and PBDE153 were measured at both INSPQ and AU, but the data from AU are displayed here, as most samples were below LOD in the measurement at INSPQ (see Table S1, see also Table S2, with the agreement between measurements at INSPQ and AU). Anti-DP: Anti-isomer dechlorane plus. HCB: Hexachlorobenzene. OCP: organochlorine pesticides. p,p′-DDE: p,p′-dichlorodiphenyldichloroethylene. PBDE: Polybrominated diphenyl ether. PCB: polychlorinated biphenyl. PFCA: perfluorocarboxylic acids. PFDA: perfluorodecanoic acid. PFHpS: perfluoroheptane sulfonate. PFHxS: perfluorohexane sulfonate. PFNA: perfluorononanoic acid. PFOA: perfluorooctanoic acid. PFOS: perfluorooctane sulfonate. PFSA: perfluorosulfonic acid. PFUnA: perfluoroundecanoic acid. β-HCH: β-hexachlorocyclohexane.

difference was borderline significant in the adjusted analyses (Fig. 1, Table S3). The PCB and OCP median concentrations were 1.5–2.4 times higher in fathers than in mothers. For the HFRs, PBDE28 was higher in mothers than in fathers, whereas we found no differences for PBDE153 and anti-DP. Most PFAS (but not PFDA and PFUnA) were also significantly higher in fathers than in mothers (Fig. 1, Table S3). The PFAS median concentrations were 1.4–4.6 times higher in fathers than in mothers.

Within the study population, 73 mothers and fathers were living together, and 69 of these mother-father pairs had provided blood samples for POP measurements from both parents. Pearson pairwise correlations of the POP concentrations between mothers and fathers living together were generally positive, expect for PCB156 and the HFRs (Table S4). The adjusted correlations were significant for PCB118, PCB138, PCB153, all OCPs (except mirex), all PFAS (except PFHxS and PFOA) and borderline significant for PCB183 and PCB187 with adjusted correlation coefficients ranging from 0.247 to 0.513 (Table S4). The correlation for PBDE28 was significantly negative ($r = -0.354$) in the adjusted analyses (Table S4).

3.2.3. Differences in POP concentrations by residential town

We found significant differences among the residential towns for all PCBs, OCPs, and PFAS in the analyses adjusted for age and sex, and borderline significant difference ($p \leq 0.080$) for PFOA (Fig. 2, Table S5). Generally, the concentrations were lowest in Nuuk, higher in Sisimiut, and highest in Ilulissat. The adjusted post hoc test showed significantly higher concentrations in Ilulissat than Nuuk for all compounds (except for the HFRs and PFOA). PCB153 (borderline), mirex, and ΣPCBs (borderline) were also significantly higher in Sisimiut than Nuuk, and oxychlordane, β-HCH, and all PFAS (borderline for PFOA) were significantly higher in Ilulissat than Sisimiut (Fig. 2). PBDE28 was significantly higher in Sisimiut than in Nuuk and Ilulissat. The PBDE153 and anti-DP concentrations were not significantly different among the residential towns (Fig. 2, Table S5).

3.2.4. POP correlations with lifestyle and socioeconomic factors

Table 4 displays the correlations between the POP concentrations and lifestyle and socioeconomic factors. We observed significant or borderline significant correlations between the measured POP concentrations and age, BMI, n-3/n-6 FA ratio, educational level, personal income, household income, alcohol intake, and percentage of life lived in Greenland.

All PCBs, OCPs, and PBDE153 were significant positively correlated with age, with correlation coefficients ranging from 0.206–0.504 (Table 4). Most PFAS (except for PFDA, PFUnA, and ΣPFCA) were also significant positively correlated with age, however with weaker correlation coefficients (ranging from 0.163–0.230) than the lipPOPs.

Table 4
Spearman correlations between persistent organic pollutant concentrations and lifestyle and socioeconomic factors.

	Age (years) (n = 164)		BMI (kg/m^2) (n = 135)		n-3/n-6 ratio (indicator of marine food) (n = 161)		Educational level (categorical)[1] (n = 161)		Personal income (categorical)[2] (n = 160)		Household income (categorical)[3] (n = 160)		Alcohol intake (current, categorical)[4] (n = 120)		Smoking history (categorical)[5] (n = 164)		Life lived in Greenland (%) (n = 163)	
	r_s	p	r_s	p	r_s	p	r_s	p	r_s	p	r_s	p	r_s	p	r_s	p	r_s	p
Lipophilic POPs (μg/kg lipid)																		
PCB 118	0.305	**<0.001***	0.096	0.272	0.427	**<0.001***	0.055	0.490	0.089	0.266	0.066	0.406	0.055	0.543	−0.064	0.417	0.119	0.131
PCB 138	0.360	**<0.001***	0.057	0.514	0.340	**<0.001***	−0.012	0.885	0.092	0.249	−0.002	0.976	0.019	0.838	−0.004	0.957	0.117	0.138
PCB 153	0.359	**<0.001***	0.025	0.777	0.318	**<0.001***	−0.028	0.722	0.071	0.372	−0.010	0.903	0.007	0.941	−0.009	0.908	0.139	0.078#
PCB 156	0.504	**<0.001***	0.058	0.506	0.245	**<0.001***	0.079	0.320	0.139	0.080	0.048	0.545	0.004	0.967	−0.067	0.397	0.100	0.207
PCB 170	0.419	**<0.001***	−0.026	0.767	0.273	**<0.001***	0.032	0.686	0.094	0.240	−0.014	0.860	0.007	0.943	−0.034	0.665	0.101	0.203
PCB 180	0.401	**<0.001***	−0.037	0.676	0.275	**<0.001***	0.017	0.830	0.070	0.384	−0.040	0.614	0.007	0.942	−0.029	0.714	0.117	0.138
PCB 183	0.357	**<0.001***	0.046	0.603	0.348	**<0.001***	0.010	0.897	0.077	0.336	−0.020	0.801	−0.014	0.881	−0.032	0.687	0.150	0.056#
PCB 187	0.353	**<0.001***	−0.013	0.882	0.317	**<0.001***	−0.020	0.805	0.065	0.412	−0.015	0.850	−0.004	0.968	0.001	0.987	0.189	**0.016***
cis-Nonachlor	0.304	**<0.001***	0.066	0.451	0.395	**<0.001***	0.003	0.968	0.090	0.261	0.088	0.271	0.027	0.768	0.015	0.854	0.174	**0.027***
HCB	0.292	**<0.001***	−0.007	0.937	0.453	**<0.001***	0.018	0.817	0.122	0.125	0.086	0.282	0.104	0.260	0.052	0.509	0.127	0.107
Mirex	0.284	**<0.001***	−0.072	0.413	0.324	**<0.001***	−0.040	0.617	−0.004	0.961	−0.061	0.445	−0.010	0.915	0.040	0.609	0.256	**0.001***
Oxychlordane	0.266	**<0.001***	0.051	0.557	0.351	**<0.001***	−0.050	0.528	0.057	0.472	0.063	0.432	0.027	0.767	0.066	0.401	0.203	**0.010***
p,p′-DDE	0.294	**<0.001***	−0.007	0.938	0.378	**<0.001***	−0.035	0.656	0.090	0.259	0.005	0.952	0.061	0.509	0.059	0.456	0.103	0.192
β-HCH	0.277	**<0.001***	0.089	0.308	0.378	**<0.001***	0.013	0.867	0.149	0.061#	0.132	0.098	0.075	0.414	−0.002	0.977	0.140	0.075#
trans-Nonachlor	0.274	**<0.001***	0.051	0.558	0.365	**<0.001***	−0.030	0.704	0.068	0.393	0.064	0.425	0.023	0.801	0.037	0.641	0.196	**0.012***
PBDE28[a]	−0.128	0.206	−0.046	0.679	0.199	0.052#	0.014	0.889	−0.088	0.392	0.082	0.429	0.014	0.908	−0.132	0.194	0.061	0.550
PBDE153[a]	0.331	**0.001***	−0.009	0.935	0.158	0.124	0.252	**0.013***	0.040	0.699	−0.111	0.286	−0.154	0.199	−0.152	0.206	−0.193	0.055#
Anti-DP	0.110	0.278	−0.120	0.282	0.042	0.684	0.078	0.448	0.052	0.610	0.266	**0.025***	0.267	**0.024***	0.132	0.193	−0.246	**0.015***
ΣPCBs	0.350	**<0.001***	−0.004	0.967	0.312	**<0.001***	−0.023	0.777	0.061	0.447	−0.014	0.858	−0.007	0.943	−0.019	0.812	0.158	**0.045***
ΣOCPs	0.293	**<0.001***	0.017	0.845	0.360	**<0.001***	−0.030	0.708	0.085	0.285	0.037	0.644	0.046	0.619	0.054	0.491	0.150	0.056#
ΣPCB + OCP	0.317	**<0.001***	0.006	0.946	0.343	**<0.001***	−0.030	0.704	0.082	0.305	0.017	0.833	0.020	0.825	0.028	0.725	0.158	**0.045***
PFAS (ng/mL)																		
PFHxS	0.230	**0.003***	0.196	**0.023***	0.216	**0.006***	−0.073	0.357	0.150	0.059#	0.066	0.405	0.067	0.466	0.012	0.884	−0.068	0.387
PFHpS	0.163	**0.037***	0.148	0.088	0.178	**0.025***	0.022	0.779	0.165	**0.037***	0.013	0.870	0.045	0.628	−0.007	0.934	−0.046	0.561
PFOS	0.186	**0.017***	0.136	0.115	0.336	**<0.001***	−0.042	0.594	0.105	0.187	0.075	0.347	0.065	0.484	−0.022	0.775	0.038	0.628
PFOA	0.206	**0.008***	0.180	**0.036***	0.093	0.245	−0.046	0.563	0.190	**0.016***	0.048	0.551	0.096	0.297	0.033	0.677	−0.143	0.068#
PFNA	0.180	**0.021***	0.124	0.153	0.359	**<0.001***	0.014	0.857	0.129	0.103	0.077	0.334	0.107	0.244	−0.042	0.594	−0.004	0.955
PFDA	0.094	0.232	−0.026	0.768	0.415	**<0.001***	0.036	0.650	0.091	0.251	0.114	0.153	0.182	**0.046***	0.004	0.958	0.053	0.504
PFUnA	0.077	0.326	−0.119	0.169	0.393	**<0.001***	0.020	0.805	0.023	0.773	0.052	0.512	0.198	**0.030***	0.043	0.588	0.097	0.219
ΣPFCA	0.138	0.078#	0.043	0.618	0.353	**<0.001***	−0.006	0.939	0.111	0.161	0.077	0.330	0.155	0.092	0.024	0.763	−0.004	0.962
ΣPFSA	0.181	**0.020***	0.154	0.074#	0.313	**<0.001***	−0.043	0.592	0.117	0.141	0.072	0.364	0.072	0.434	−0.029	0.710	0.025	0.755
ΣPFAS	0.180	**0.021***	0.125	0.147	0.331	**<0.001***	−0.032	0.683	0.123	0.121	0.079	0.322	0.092	0.318	−0.019	0.807	0.014	0.855

Pooled data (mothers and fathers). Spearman correlation coefficients (r_s) with p-value for the correlation. Bold p-values and * indicate a significant correlation ($p \leq 0.050$), while # indicate a borderline significant correlation ($p \leq 0.080$). Some variables were categorical with the following categories: [1]Educational level (Primary School, High School, Technical college, and University), [2]Personal income/[3]Household Income (<100.000, 100.000–250.000, and >250.000 DKK/year), [4]Current alcohol intake (0 drinks/week, 1–7 drinks/week, and ≥8 drinks/week), [5]Smoking history (Never, Former, and Current).

[a] PBDE28 and PBDE153 were measured at both INSPQ and AU, but the data from AU are displayed here, as most samples were below LOD in the measurement at INSPQ (see Table S1, see also Table S2, with the agreement between measurements at INSPQ and AU). For personal income, household income, and alcohol intake the answer possibility "Don't know" was omitted in the analysis. Anti-DP: Anti-isomer dechlorane plus. HCB: Hexachlorobenzene. n: number of participants included in the analyses. OCP: organochlorine pesticides. p,p′-DDE: p,p′-dichlorodiphenyldichloroethylene. PBDE: Polybrominated diphenyl ether. PCB: polychlorinated biphenyl. PFAS: per- and polyfluoroalkyl substances. PFCA: perfluorocarboxylic acids. PFDA: perfluorodecanoic acid. PFHpS: perfluoroheptane sulfonate. PFHxS: perfluorohexane sulfonate. PFNA: perfluorononanoic acid. PFOA: perfluorooctanoic acid. PFOS: perfluorooctane sulfonate. PFSA: perfluorosulfonic acid. PFUnA: perfluoroundecanoic acid. β-HCH: β-hexachlorocyclohexane.

Table 5
Adjusted associations between persistent organic pollutant concentrations and intake of traditional food groups (times per month).

	Marine mammals (n = 110)	Seabirds (n = 106)	Greenlandic Fish (n = 142)	Shellfish (n = 148)	Dried Fish (n = 151)	Terrestrial animals (n = 145)	Berries (n = 141)	All traditional foods summed (n = 97)
	β (95%CI)	β (95%CI)	β (95%CI)	β (95%CI)	β (95%CI)	β (95%CI)	β (95%CI)	β (95%CI)
Lipophilic POPs (μg/kg lipid)								
PCB 118	**0.427 (0.274; 0.579)***	**0.333 (0.115; 0.550)***	**0.201 (0.015; 0.388)***	0.078 (−0.089; 0.245)	**0.345 (0.196; 0.493)***	0.081 (−0.078; 0.241)	0.055 (−0.081; 0.192)	**0.471 (0.240; 0.701)***
PCB 138	**0.330 (0.194; 0.466)***	**0.256 (0.047; 0.466)***	0.087 (−0.074; 0.248)	0.023 (−0.122; 0.169)	**0.242 (0.114; 0.369)***	0.058 (−0.075; 0.192)	0.019 (−0.100; 0.137)	**0.332 (0.134; 0.529)***
PCB 153	**0.339 (0.200; 0.478)***	**0.267 (0.053; 0.481)***	0.093 (−0.068; 0.255)	0.000 (−0.146; 0.146)	**0.251 (0.123; 0.378)***	0.060 (−0.075; 0.195)	0.021 (−0.098; 0.141)	**0.342 (0.141; 0.544)***
PCB 156	**0.156 (0.006; 0.307)***	0.156 (−0.068; 0.379)	−0.018 (−0.184; 0.148)	−0.090 (−0.240; 0.059)	0.131 (−0.001; 0.263)#	−0.028 (−0.170; 0.114)	−0.015 (−0.137; 0.107)	0.171 (−0.042; 0.383)
PCB 170	**0.257 (0.115; 0.400)***	**0.217 (0.000; 0.434)***	0.058 (−0.099; 0.215)	−0.056 (−0.197; 0.084)	**0.169 (0.045; 0.294)***	0.046 (−0.087; 0.180)	−0.003 (−0.119; 0.113)	**0.264 (0.064; 0.463)***
PCB 180	**0.259 (0.118; 0.400)***	**0.238 (0.023; 0.453)***	0.060 (−0.096; 0.215)	−0.072 (−0.214; 0.070)	**0.178 (0.053; 0.302)***	0.043 (−0.090; 0.175)	0.017 (−0.100; 0.135)	**0.255 (0.059; 0.450)***
PCB 183	**0.265 (0.115; 0.415)***	**0.255 (0.032; 0.478)***	0.032 (−0.137; 0.200)	−0.027 (−0.180; 0.126)	**0.193 (0.060; 0.326)***	0.025 (−0.115; 0.164)	−0.012 (−0.138; 0.114)	**0.253 (0.039; 0.468)***
PCB 187	**0.356 (0.208; 0.503)***	**0.294 (0.056; 0.531)***	0.084 (−0.085; 0.253)	−0.033 (−0.183; 0.116)	**0.256 (0.123; 0.389)***	0.056 (−0.089; 0.201)	0.006 (−0.121; 0.133)	**0.358 (0.144; 0.572)***
cis-Nonachlor	**0.527 (0.353; 0.701)***	**0.494 (0.199; 0.788)***	0.164 (−0.058; 0.386)	0.047 (−0.148; 0.241)	**0.451 (0.281; 0.621)***	0.103 (−0.089; 0.295)	0.005 (−0.152; 0.163)	**0.578 (0.304; 0.853)***
HCB	**0.344 (0.226; 0.462)***	**0.349 (0.170; 0.529)***	0.135 (−0.014; 0.283)#	0.027 (−0.106; 0.161)	**0.294 (0.181; 0.408)***	0.101 (−0.023; 0.225)	0.034 (−0.076; 0.144)	**0.329 (0.152; 0.507)***
Mirex	**0.389 (0.223; 0.555)***	**0.381 (0.127; 0.636)***	0.080 (−0.113; 0.273)	−0.099 (−0.274; 0.076)	**0.315 (0.168; 0.463)***	0.079 (−0.086; 0.244)	−0.037 (−0.184; 0.109)	**0.354 (0.110; 0.597)***
Oxychlordane	**0.564 (0.381; 0.747)***	**0.487 (0.182; 0.792)***	0.181 (−0.052; 0.414)	0.056 (−0.146; 0.259)	**0.485 (0.307; 0.663)***	0.151 (−0.048; 0.350)	−0.006 (−0.170; 0.158)	**0.661 (0.368; 0.955)***
p,p′-DDE	**0.389 (0.241; 0.537)***	**0.275 (0.045; 0.506)***	0.122 (−0.053; 0.298)	0.056 (−0.102; 0.213)	**0.302 (0.163; 0.440)***	0.086 (−0.063; 0.234)	0.044 (−0.084; 0.171)	**0.397 (0.177; 0.616)***
β-HCH	**0.339 (0.174; 0.505)***	**0.504 (0.241; 0.768)***	0.133 (−0.072; 0.339)	0.065 (−0.125; 0.255)	**0.335 (0.171; 0.500)***	0.084 (−0.094; 0.262)	0.005 (−0.149; 0.160)	**0.388 (0.140; 0.636)***
trans-Nonachlor	**0.570 (0.383; 0.757)***	**0.526 (0.210; 0.841)***	0.156 (−0.070; 0.381)	0.056 (−0.144; 0.256)	**0.479 (0.301; 0.657)***	0.131 (−0.069; 0.331)	−0.007 (−0.168; 0.154)	**0.639 (0.349; 0.929)***
PBDE28[a]	0.217 (−0.054; 0.487)	0.357 (−0.073; 0.790)	0.220 (−0.095; 0.534)	−0.038 (−0.314; 0.237)	**0.235 (0.008; 0.463)***	0.109 (−0.117; 0.334)	**0.308 (0.081; 0.535)***	0.244 (−0.121; 0.610)
PBDE153[a]	0.056 (−0.119; 0.231)	−0.036 (−0.280; 0.208)	**0.236 (0.056; 0.415)***	−0.051 (−0.215; 0.114)	0.031 (−0.108; 0.170)	0.012 (−0.119; 0.144)	−0.038 (−0.177; 0.101)	**0.262 (0.043; 0.481)***
Anti-DP	0.049 (−0.193; 0.290)	0.317 (−0.090; 0.725)	0.042 (−0.243; 0.326)	−0.136 (−0.376; 0.105)	0.044 (−0.164; 0.251)	0.050 (−0.154; 0.254)	0.019 (−0.181; 0.220)	0.070 (−0.251; 0.390)
ΣPCBs	**0.246 (0.143; 0.350)***	**0.196 (0.032; 0.361)***	0.052 (−0.072; 0.176)	−0.007 (−0.121; 0.108)	**0.183 (0.084; 0.281)***	0.036 (−0.066; 0.139)	0.016 (−0.077; 0.109)	**0.229 (0.078; 0.380)***
ΣOCPs	**0.401 (0.263; 0.540)***	**0.332 (0.110; 0.555)***	0.121 (−0.052; 0.293)	0.047 (−0.108; 0.202)	**0.324 (0.190; 0.458)***	0.094 (−0.051; 0.239)	0.025 (−0.101; 0.150)	**0.412 (0.202; 0.622)***
Σ(PCB + OCP)	**0.322 (0.204; 0.441)***	**0.266 (0.075; 0.458)***	0.085 (−0.061; 0.232)	0.020 (−0.114; 0.154)	**0.253 (0.138; 0.369)***	0.063 (−0.060; 0.185)	0.020 (−0.088; 0.129)	**0.316 (0.137; 0.494)***
PFAS (ng/mL)								
PFHxS	**0.180 (0.058; 0.302)***	**0.316 (0.127; 0.506)***	0.018 (−0.115; 0.152)	0.000 (−0.119; 0.119)	**0.161 (0.052; 0.270)***	0.067 (−0.049; 0.183)	0.020 (−0.083; 0.122)	0.088 (−0.076; 0.252)
PFHpS	**0.127 (0.002; 0.252)***	**0.295 (0.110; 0.479)***	0.085 (−0.051; 0.221)	0.059 (−0.069; 0.188)	**0.141 (0.025; 0.257)***	0.103 (−0.013; 0.219)	0.012 (−0.097; 0.121)	0.078 (−0.087; 0.243)
PFOS	**0.245 (0.139; 0.352)***	**0.288 (0.120; 0.456)***	0.084 (−0.044; 0.213)	0.017 (−0.103; 0.137)	**0.248 (0.143; 0.353)***	0.059 (−0.054; 0.172)	0.018 (−0.085; 0.121)	**0.170 (0.024; 0.316)***
PFOA	−0.018 (−0.132; 0.095)	0.050 (−0.114; 0.213)	0.082 (−0.030; 0.194)	0.020 (−0.081; 0.122)	0.031 (−0.064; 0.127)	0.008 (−0.091; 0.106)	0.054 (−0.031; 0.139)	−0.068 (−0.217; 0.082)
PFNA	**0.232 (0.131; 0.333)***	**0.298 (0.142; 0.454)***	**0.123 (0.004; 0.243)***	0.022 (−0.091; 0.136)	**0.225 (0.125; 0.324)***	0.081 (−0.024; 0.186)	0.055 (−0.041; 0.150)	**0.147 (0.009; 0.284)***
PFDA	**0.294 (0.187; 0.402)***	**0.296 (0.117; 0.475)***	0.118 (−0.012; 0.249)#	0.027 (−0.098; 0.152)	**0.274 (0.164; 0.383)***	0.070 (−0.048; 0.188)	0.048 (−0.060; 0.156)	**0.234 (0.078; 0.390)***
PFUnA	**0.461 (0.325; 0.597)***	**0.404 (0.172; 0.637)***	**0.186 (0.014; 0.358)***	−0.003 (−0.163; 0.157)	**0.343 (0.203; 0.483)***	0.074 (−0.081; 0.228)	0.039 (−0.096; 0.175)	**0.303 (0.083; 0.522)***
ΣPFCA	**0.169 (0.097; 0.241)***	**0.221 (0.104; 0.339)***	0.073 (−0.015; 0.161)	0.044 (−0.042; 0.131)	**0.177 (0.102; 0.253)***	0.053 (−0.025; 0.130)	0.037 (−0.038; 0.111)	**0.107 (0.009; 0.206)***
ΣPFSA	**0.203 (0.112; 0.294)***	**0.270 (0.125; 0.415)***	0.054 (−0.057; 0.164)	0.019 (−0.085; 0.124)	**0.207 (0.115; 0.299)***	0.050 (−0.047; 0.147)	0.017 (−0.073; 0.106)	**0.131 (0.007; 0.255)***
ΣPFAS	**0.192 (0.109; 0.275)***	**0.254 (0.120; 0.389)***	0.058 (−0.043; 0.160)	0.027 (−0.071; 0.125)	**0.198 (0.112; 0.283)***	0.050 (−0.039; 0.140)	0.023 (−0.061; 0.107)	**0.124 (0.011; 0.237)***

Pooled data (mothers and fathers). Linear regression analyses with POP as dependent variable and food intake (times per month) as independent variable with age, town, sex, education, house income, and smoking history as covariates. The analyses were performed with ln transformed variables. Unadjusted results are shown in Table S6. Bold text and * indicate a significant correlation ($p \leq 0.050$), while # indicate a borderline significant correlation ($p \leq 0.080$).

[a] PBDE28 and PBDE153 were measured at both INSPQ and AU, but the data from AU are displayed here, as most samples were below LOD in the measurement at INSPQ (see Table S1, see also Table S2, with the agreement between measurements at INSPQ and AU). Anti-DP: Anti-isomer dechlorane plus. HCB: Hexachlorobenzene. n: number of participants included in the analyses. OCP: organochlorine pesticides. p,p′-DDE: p,p′-dichlorodiphenyldichloroethylene. PBDE: Polybrominated diphenyl ether. PCB: polychlorinated biphenyl. PFAS: per- and polyfluoroalkyl substances. PFCA: perfluorocarboxylic acids. PFDA: perfluorodecanoic acid. PFHpS: perfluoroheptane sulfonate. PFHxS: perfluorohexane sulfonate. PFNA: perfluorononanoic acid. PFOA: perfluorooctanoic acid. PFOS: perfluorooctane sulfonate. PFSA: perfluorosulfonic acid. PFUnA: perfluoroundecanoic acid. β (95%CI): Beta coefficients from the linear regression analyses with 95% confidence interval. β-HCH: β-hexachlorocyclohexane.

Table 6
Adjusted associations between persistent organic pollutant concentrations and intake of imported food groups.

	Meat products (n = 157)	Carbohydrate foods (n = 160)	Sauce (n = 161)	Vegetables (n = 160)	Fruit (n = 157)	Fast food (n = 154)	Sweets & snacks (n = 157)	All imported food groups summed (n = 142)
	β (95%CI)	β (95%CI)	β (95%CI)	β (95%CI)	β (95%CI)	β (95%CI)	β (95%CI)	β (95%CI)
Lipophilic POPs (μg/kg lipid)								
PCB 118	**−0.427 (−0.625; −0.229)***	0.024 (−0.159; 0.208)	0.011 (−0.112; 0.134)	0.024 (−0.101; 0.150)	−0.002 (−0.142; 0.137)	−0.139 (−0.311; 0.033)	−0.005 (−0.174; 0.165)	**−0.371 (−0.672; −0.070)***
PCB 138	**−0.334 (−0.507; −0.161)***	0.032 (−0.121; 0.186)	0.006 (−0.098; 0.109)	0.026 (−0.081; 0.133)	0.015 (−0.102; 0.132)	−0.095 (−0.241; 0.052)	−0.043 (−0.186; 0.099)	**−0.286 (−0.544; −0.028)***
PCB 153	**−0.329 (−0.504; −0.153)***	0.024 (−0.133; 0.180)	−0.018 (−0.122; 0.087)	0.023 (−0.085; 0.131)	0.029 (−0.090; 0.148)	−0.099 (−0.247; 0.049)	−0.060 (−0.204; 0.085)	**−0.295 (−0.558; −0.031)***
PCB 156	**−0.275 (−0.458; −0.092)***	0.031 (−0.131; 0.193)	−0.042 (−0.149; 0.065)	0.033 (−0.077; 0.143)	0.052 (−0.070; 0.174)	−0.092 (−0.244; 0.060)	−0.071 (−0.221; 0.078)	−0.269 (−0.542; 0.004)#
PCB 170	**−0.270 (−0.444; −0.096)***	0.011 (−0.142; 0.163)	−0.026 (−0.127; 0.075)	0.039 (−0.066; 0.143)	0.042 (−0.074; 0.158)	−0.111 (−0.256; 0.034)	−0.060 (−0.201; 0.082)	**−0.262 (−0.522; −0.002)***
PCB 180	**−0.272 (−0.446; −0.099)***	0.032 (−0.121; 0.185)	−0.031 (−0.132; 0.070)	0.032 (−0.072; 0.137)	0.035 (−0.081; 0.151)	−0.108 (−0.253; 0.037)	−0.073 (−0.214; 0.068)	**−0.260 (−0.519; −0.000)***
PCB 183	**−0.322 (−0.502; −0.142)***	0.052 (−0.108; 0.212)	−0.003 (−0.110; 0.104)	0.010 (−0.101; 0.120)	0.033 (−0.089; 0.155)	−0.142 (−0.294; 0.010)#	−0.109 (−0.256; 0.038)	**−0.315 (−0.582; −0.048)***
PCB 187	**−0.332 (−0.517; −0.147)***	0.063 (−0.101; 0.227)	−0.015 (−0.125; 0.094)	0.019 (−0.095; 0.132)	0.034 (−0.091; 0.159)	−0.094 (−0.251; 0.063)	−0.081 (−0.233; 0.072)	−0.271 (−0.549; 0.008)#
cis-Nonachlor	**−0.480 (−0.719; −0.240)***	0.097 (−0.116; 0.309)	−0.001 (−0.143; 0.142)	0.049 (−0.098; 0.195)	0.018 (−0.145; 0.180)	−0.110 (−0.315, 0.094)	−0.061 (−0.259; 0.138)	−0.321 (−0.687; 0.045)
HCB	**−0.361 (−0.517; −0.205)***	0.054 (−0.089; 0.196)	0.023 (−0.073; 0.118)	0.018 (−0.079; 0.115)	0.000 (−0.109; 0.108)	−0.079 (−0.213; 0.055)	−0.008 (−0.140; 0.124)	−0.233 (−0.475; 0.008)#
Mirex	**−0.360 (−0.568; −0.152)***	0.134 (−0.051; 0.318)	0.013 (−0.110; 0.135)	0.009 (−0.119; 0.136)	0.019 (−0.122; 0.159)	−0.117 (−0.294; 0.060)	−0.169 (−0.339; 0.001)#	−0.310 (−0.625; 0.004)#
Oxychlordane	**−0.513 (−0.764; −0.262)***	0.088 (−0.136; 0.312)	−0.021 (−0.171; 0.129)	0.051 (−0.103; 0.206)	0.040 (−0.131; 0.211)	−0.092 (−0.307; 0.123)	−0.042 (−0.250; 0.168)	−0.328 (−0.715; 0.058)
p,p′-DDE	**−0.400 (−0.589; −0.212)***	0.034 (−0.134; 0.201)	−0.012 (−0.125; 0.102)	0.045 (−0.072; 0.161)	0.031 (−0.098; 0.159)	−0.088 (−0.248; 0.072)	−0.015 (−0.173; 0.142)	−0.269 (−0.554; 0.016)#
β-HCH	**−0.421 (−0.644; −0.198)***	0.078 (−0.117; 0.273)	0.020 (−0.112; 0.152)	0.043 (−0.094; 0.179)	0.004 (−0.147; 0.156)	−0.124 (−0.313; 0.065)	−0.028 (−0.211; 0.155)	**−0.347 (−0.679; −0.016)***
trans-Nonachlor	**−0.485 (−0.736; −0.234)***	0.118 (−0.105; 0.340)	−0.002 (−0.152; 0.147)	0.079 (−0.075; 0.232)	0.027 (−0.143; 0.198)	−0.115 (−0.329; 0.099)	−0.067 (−0.275; 0.140)	−0.319 (−0.703; 0.065)
PBDE28[a]	−0.143 (−0.474; 0.188)	0.121 (−0.140; 0.382)	−0.001 (−0.175; 0.173)	−0.083 (−0.258; 0.091)	0.000 (−0.200; 0.201)	0.023 (−0.226; 0.273)	−0.093 (−0.341; 0.155)	−0.014 (−0.450; 0.422)
PBDE153[a]	−0.124 (−0.317; 0.069)	0.003 (−0.154; 0.159)	−0.086 (−0.187; 0.015)	−0.043 (−0.149; 0.063)	0.053 (−0.071; 0.178)	−0.007 (−0.152; 0.137)	0.040 (−0.111; 0.192)	0.010 (−0.250; 0.270)
Anti-DP	0.050 (−0.244; 0.344)	0.158 (−0.067; 0.384)	−0.075 (−0.226; 0.075)	−0.017 (−0.170; 0.135)	0.019 (−0.157; 0.195)	0.064 (−0.148; 0.277)	0.049 (−0.176; 0.273)	0.075 (−0.300; 0.450)
ΣPCBs	**−0.240 (−0.376; −0.104)***	0.044 (−0.077; 0.164)	0.000 (−0.081; 0.081)	0.033 (−0.050; 0.116)	0.024 (−0.068; 0.115)	−0.081 (−0.195; 0.033)	−0.059 (−0.170; 0.052)	**−0.216 (−0.418; −0.014)***
ΣOCPs	**−0.390 (−0.574; −0.206)***	0.063 (−0.102; 0.227)	0.003 (−0.108; 0.114)	0.047 (−0.067; 0.161)	0.019 (−0.107; 0.145)	−0.088 (−0.245; 0.069)	−0.032 (−0.185; 0.122)	−0.270 (−0.550; 0.010)**
Σ(PCB + OCP)	**−0.318 (−0.476; −0.159)***	0.055 (−0.085; 0.196)	0.004 (−0.091; 0.099)	0.042 (−0.056; 0.139)	0.021 (−0.087; 0.128)	−0.084 (−0.218; 0.050)	−0.047 (−0.178; 0.084)	**−0.248 (−0.485; −0.010)***
PFAS (ng/mL)								
PFHxS	−0.149 (−0.299; 0.00)#	0.029 (−0.099; 0.157)	0.022 (−0.064; 0.108)	−0.023 (−0.112; 0.065)	0.041 (−0.052; 0.134)	0.005 (−0.115; 0.124)	0.070 (−0.046; 0.186)	0.010 (−0.203; 0.222)
PFHpS	**−0.202 (−0.357; −0.047)***	0.016 (−0.118; 0.151)	0.014 (−0.076; 0.105)	0.015 (−0.077; 0.106)	−0.008 (−0.109; 0.093)	−0.002 (−0.126; 0.123)	0.053 (−0.069; 0.175)	−0.055 (−0.280; 0.171)
PFOS	**−0.250 (−0.396, −0.105)***	0.002 (−0.128; 0.132)	0.005 (−0.083; 0.092)	−0.027 (−0.116; 0.063)	−0.007 (−0.104; 0.090)	−0.046 (−0.167; 0.075)	−0.004 (−0.124; 0.116)	−0.174 (−0.387; 0.040)
PFOA	−0.098 (−0.222; 0.026)	−0.049 (−0.159; 0.061)	−0.069 (−0.140; 0.002)#	−0.033 (−0.108; 0.042)	0.023 (−0.056; 0.103)	0.013 (−0.038; 0.168)	−0.021 (−0.120; 0.078)	−0.082 (−0.261; 0.097)
PFNA	**−0.256 (−0.392; −0.119)***	−0.005 (−0.127; 0.118)	−0.007 (−0.089; 0.075)	−0.055 (−0.138; 0.029)	0.008 (−0.084; 0.100)	−0.021 (−0.133; 0.092)	0.013 (−0.100; 0.126)	−0.126 (−0.330; 0.079)
PFDA	**−0.263 (−0.419; −0.107)***	0.043 (−0.094; 0.180)	0.025 (−0.067; 0.117)	−0.037 (−0.131; 0.057)	0.025 (−0.077; 0.128)	−0.014 (−0.143; 0.114)	0.035 (−0.092; 0.162)	−0.087 (−0.318; 0.145)
PFUnA	**−0.290 (−0.490; −0.090)***	0.109 (−0.066; 0.284)	0.072 (−0.044; 0.189)	0.031 (−0.087; 0.149)	0.075 (−0.053; 0.203)	−0.047 (−0.214; 0.120)	0.100 (−0.060; 0.260)	−0.023 (−0.313; 0.267)
ΣPFCA	**−0.183 (−0.288; −0.078)***	0.023 (−0.071; 0.116)	−0.006 (−0.068; 0.057)	−0.024 (−0.088; 0.040)	0.019 (−0.051; 0.088)	−0.002 (−0.088; 0.085)	0.015 (−0.072; 0.101)	−0.063 (−0.219; 0.094)
ΣPFSA	**−0.206 (−0.332; −0.079)***	0.014 (−0.099; 0.126)	0.012 (−0.063; 0.088)	−0.024 (−0.102; 0.053)	−0.003 (−0.087; 0.080)	−0.033 (−0.137; 0.071)	0.005 (−0.098; 0.109)	−0.128 (−0.314; 0.058)
ΣPFAA	**−0.198 (−0.316; −0.080)***	0.017 (−0.088; 0.122)	0.007 (−0.063; 0.078)	−0.025 (−0.097; 0.047)	0.002 (−0.076; 0.080)	−0.023 (−0.120; 0.074)	0.008 (−0.089; 0.105)	−0.109 (−0.283; 0.065)

Pooled data (mothers and fathers). Linear regression analyses with POP as dependent variable and food intake (times per month) as independent variable with age, town, sex, education, house income, and smoking history as covariates. The analyses were performed with ln transformed variables. Unadjusted results are shown in Table S7. Bold text and * indicate a significant correlation ($p \leq 0.050$), while # indicate a borderline significant correlation ($p \leq 0.080$).

[a] PBDE28 and PBDE153 were measured at both INSPQ and AU, but the data from AU are displayed here, as most samples were below LOD in the measurement at INSPQ (see Table S1, see also Table S2, with the agreement between measurements at INSPQ and AU). Anti-DP: Anti-isomer dechlorane plus. HCB: Hexachlorobenzene. n: number of participants included in the analyses. OCP: organochlorine pesticides. p,p′-DDE: p,p′-dichlorodiphenyldichloroethylene. PBDE: Polybrominated diphenyl ether. PCB: polychlorinated biphenyl. PFAS: *per-* and polyfluoroalkyl substances. PFCA: perfluorocarboxylic acids. PFDA: perfluorodecanoic acid. PFHpS: perfluoroheptane sulfonate. PFHxS: perfluorohexane sulfonate. PFNA: perfluorononanoic acid. PFOA: perfluorooctanoic acid. PFOS: perfluorooctane sulfonate. PFSA: perfluorosulfonic acid. PFUnA: perfluoroundecanoic acid. β (95%CI): Beta coefficients from the linear regression analyses with 95% confidence interval. β-HCH: β-hexachlorocyclohexane.

None of the lipPOPs correlated with BMI. The PFAS compounds PFHxS, PFOA, and ΣPFSA (borderline) were significantly and positively correlated with BMI (Table 4).

All PCBs, OCPs, PFAS (except PFOA), and PBDE28 (borderline) were significant positively correlated with n-3/n-6 FA ratio with correlation coefficients ranging from 0.178–0.453 (Table 4).

β-HCH (borderline), PFHxS (borderline), PFHpS, and PFOA correlated significantly and positively with personal income, while PBDE153 correlated significantly and positively with educational level and anti-DP correlated significantly and positively with household income (Table 4).

PFDA, PFUnA, and anti-DP showed a significant positive correlation with current alcohol intake, whereas no significant correlations between POPs and smoking history were observed (Table 4).

Several PCBs and OCPs correlated significantly and positively with the percentage of life lived in Greenland, while PBDE153 (borderline), anti-DP, and PFOA (borderline) correlated significantly and negatively with percentage of life lived in Greenland (Table 4).

3.2.5. Associations between POP concentrations and food intake

We studied the associations between POP concentrations and traditional Greenlandic and imported food intake. Data for the unadjusted analyses are shown in Tables S6–S7, being very similar to the adjusted data seen in Tables 5–6.

The majority of PCBs, OCPs, and PFAS were associated significantly and positively with the intake of marine mammals, seabirds, dried fish, and the summed traditional food group (Table 5). There were few significant positive associations between Greenlandic fish intake and PCB118, HCB (borderline), PBDE153, PFNA, PFDA (borderline), and PFUnA. PBDE28 concentrations were significantly associated with dried fish and berries, showing a positive association (Table 5).

The majority of PCBs and OCPs were associated negatively with intake of imported meat products and the summed imported food group, significantly or borderline significantly (Table 6). Furthermore, we found borderline significant negative associations between PCB183 and fast food intake and between mirex and sweets & snacks intake. We also observed significant negative associations between intake of imported meat products and the majority of PFAS. Furthermore, PFOA associated borderline negatively with sauce intake (Table 6).

4. Discussion

In the present study, we measured PCBs, OCPs, HFRs, and PFAS in Greenlandic mothers and fathers participating in this new ACCEPT follow-up data collection in 2019–2020. For the mothers, the individual concentrations of the POPs had decreased significantly from inclusion in the study to this follow-up 3–5 years later. We found higher concentrations in fathers than in mothers, and higher concentrations in Ilulissat than in Sisimiut and Nuuk. POP concentrations correlated with age, BMI, n-3/n-6 FA ratio, personal income, alcohol intake, and percentage of life lived in Greenland. The POPs were also positively associated with traditional food intake, especially intake of marine mammals, seabirds, dried fish, and the summed traditional food group. Furthermore, we observed negative associations between the POP concentrations and intake of imported meat and the summed imported food group.

4.1. Concentrations of POPs and comparison with other studies

Despite regulation of some POPs, they persist in the environment and wildlife with the consequence of continued human exposure through consumption of POP-containing food items. The temporal trend of POP concentrations in human blood is used to monitor the effectiveness of international regulations, such as the Stockholm Convention (UNEP, n.d.). In the Greenlandic population, the serum concentrations of the regulated POPs, including PCBs, OCPs, PFOS, and PFOA, decreased with up to 15% annually from 1994 to 2015 (Long et al., 2021). The POP concentrations found in the present study, including the unregulated PFAS, are generally lower than previously reported in Greenland (Bjerregaard et al., 2013; Deutch et al., 2007a; Deutch et al., 2007b; Deutch et al., 2004; Hjermitslev et al., 2019; Long et al., 2015; Long et al., 2021). Decreasing trends of regulated POPs were also observed in human samples from other Arctic areas (Abass et al., 2018; Berg et al., 2021; Nøst et al., 2019; Shu et al., 2019). For the unregulated PFAS, increasing trends were seen for PFNA in Yup'ik, US maternal blood from 2004—2006 to 2009—2012 (Abass et al., 2018). In Tromsø, Norway, the concentrations of PFHxS and PFHpS increased from 1986 to 2001 among 30-year old men and women, while decreasing from 2001 to 2007. The concentrations of PFNA, PFDA and PFUnA were increasing or stable over time (1986–2007) (Berg et al., 2021). In nursing Swedish first-time mothers, PFUnA increased throughout the study period (1996–2019), PFDA concentrations were stable, and in contrast, PFHxS and PFNA increased until 2009–2011 and 2008, respectively, and decreased afterwards (Gyllenhammar et al., 2020). Furthermore, concentrations of PFNA, PFDA, PFHxS, and PFHpA in Swedish pregnant women also significantly decreased (14–31%) from 2007 to 2010 (Shu et al., 2019).

The body burden of POPs in the Arctic population is high in comparison to non-Arctic populations (AMAP, 1998, 2003, 2009, 2016a, 2021). Generally, the highest concentrations of POPs in the Arctic populations have been measured in Greenland, the Faroe Islands, Nunavik (Canada), and the Chukotka district (Russia) (AMAP, 2021). Compared to other Arctic regions, the PCB and OCP concentrations in the present study were generally 1.5–25 times lower than concentrations seen in the Pechenga district of Murmansk Oblast, Russia (samples collected 2013–2014, men and women, 26–65 years) (Dudarev et al., 2016), but 2–16 times higher than the concentrations in Yukon, Canada (samples collected 2017–2018, men and women, >18 years) (Drysdale et al., 2021). For the PFAS, the concentrations of PFSAs were 7–30% higher in the present study than reported in men and women (18–88 years) from St. Lawrence Island, Alaska (samples collected 2013–2014) (Byrne et al., 2017) and pregnant women (15–39 years) from Nunavik, Canada (samples collected 2016–2017) (Caron-Beaudoin et al., 2020), while the PFCAs were 20–70% lower (Byrne et al., 2017; Caron-Beaudoin et al., 2020). The PFAS concentrations in the present study were comparable to concentrations reported in Yukon and the Northwest Territories, Canada (samples collected 2017–2018, men and women, 20–79 years) (Garcia-Barrios et al., 2021). The HFRs were generally below LOD in the present study; only PBDE28, PBDE153, and anti-DP were above LOD in more than 50% of the samples in the measurements conducted at AU. The PBDE concentrations in Greenland were lower than reported elsewhere in the Arctic (AMAP, 2016a; Byrne et al., 2017; Hjermitslev et al., 2019), however, considerable geographical differences in human PBDE concentrations are well-known, given national differences in use patterns of flame retardants in consumer products (Vorkamp, 2012). To our knowledge, we report for the first time DP concentrations in human samples from the Arctic region. However, compared to other geographical regions, the concentrations in the present study were 50% lower than reported in pregnant women from Wenling, China (2010 − 2011) (Yin et al., 2020), 30% lower than in maternal samples at delivery from Sherbrooke, Quebec, Canada (Zhou et al., 2014), but comparable with the concentrations in healthy blood donors from Munich, Germany (2013–2014) (Fromme et al., 2015). The anti-DP isomer had a higher detection frequency than the syn-DP isomer, which is consistent with higher concentrations of anti-DP in the commercial DP product and most environmental Arctic samples (Vorkamp et al., 2015).

We found that POP concentrations were higher in the fathers compared to the mother and in the participants living in Ilulissat compared to Sisimiut and Nuuk. Furthermore, POP concentrations correlated positively with age. These findings support the results from several previous studies in Greenland (Bjerregaard et al., 2013; Bonefeld-Jorgensen, 2010; Deutch et al., 2004; Hjermitslev et al., 2019; Long et al., 2015; Schæbel et al., 2017) and are in agreement with the higher intake of traditional food seen in fathers as well as in participants outside Nuuk (Wielsøe et al., 2021).

Only few studies have reported intra-individual time trends of POPs by using repeated sampling in the same individuals (Gribble et al., 2015;

Hagmar et al., 2006; Høyer et al., 2000; Nøst et al., 2013; Nøst et al., 2014; Stubleski et al., 2018; Sweeney et al., 2001; Wolff et al., 2000). The majority of studies report a decreasing trend in lipPOP concentrations for the same individual (Hagmar et al., 2006; Høyer et al., 2000; Nøst et al., 2013; Stubleski et al., 2018; Sweeney et al., 2001; Wolff et al., 2000), in agreement with the findings in the present study. Only mirex did not change significantly in the present study, which may be due to its currently slow decline in the environment, but it cannot be fully explained. Mirex is considered to be one of the most stable and persistent POPs with a half-life of up to 10 years in soil (ATSDR, 2020). Results from trend analyses indicate that the main decline of mirex in the Arctic biota occurred before 2000. Time-series starting after 2000, indicate a very slow decline of mirex in the Arctic biota with 1.9% annually (half-life of more than 30 years), whereas the mean annual change was 6.7% (half-life of 10 years) for time series starting prior to the year 2000 (AMAP, 2016b). However, slower declining rates after 2000 have also been seen for other POPs (AMAP, 2016b). In humans, Deutch et al. (2006) found that the estimated intake of mirex increase from 1970 to 2004, even though the intake of traditional food decreased. Bjerregaard et al. (2013) reported a decrease mirex (41%) in the general Greenlandic population from 1993 to 2009, less pronounced than for other OCPs included in the present study (54–56%) but similar with the decrease for ΣPCB (41%) and PCB153 (42%).

The intra-individual changes in PFAS concentrations have been less consistent. Decreasing individual concentrations of PFOS, PFOA, PFHxS, PFNA, and PFUnA were seen among the African American Gullah population in South Carolina, US, between 2003 and 2013 (Gribble et al., 2015). In Norway, PFOA and PFOS concentrations decreased as well after 2001, while PFNA, PFDA, and PFUnA increased in the same individuals throughout the study period 1976–2007 (Nøst et al., 2014). In the present study, we observed decreasing individual changes for all PFAS in the mothers from inclusion in the period 2013–2015 to this follow-up 3–5 years after giving birth (2019–2020). The divergence between the study results might be due to differences in study periods and exposure sources for the study populations.

The mother's food intake pattern at inclusion during pregnancy and at this follow up (3–5 years later) was generally similar, only few significant changes were observed, including a less frequent intake of seabirds and fruits and more frequent meat intake (Wielsøe et al., 2021). Decreasing trends of POP in Greenlandic marine food items are likely to explain the significant intra-individual decrease in serum POP concentrations observed in the present study. PCBs and OCP have decreased in Arctic biota (including Greenland) during the last 20–30 years (Rigét et al., 2019; Rigét et al., 2016), as well as in humans (Abass et al., 2018; Long et al., 2021). PFOS has also decreased in the Arctic biota, whereas the trend for other PFAS are less consistent (Rigét et al., 2019; Rigét et al., 2016). In Greenlandic ringed seal and polar bear, PFOS, PFOA, PFNA, PFDA and PFUnA generally peaked around year 2006 and decreased from 2006 to 2011 (Rigét et al., 2013).

4.2. Association between POP concentrations and food intake

POP concentrations and n-3/n-6 FA ratio correlated positively. The magnitude of the correlations was similar for PCBs, OCPs, and PFAS, indicating that marine food intake is a major contributor to the exposure to these chemical groups. This is in agreement with the results from the linear regression, showing significant positive associations between POPs and intake of marine mammals, seabirds, and dried fish. We found no significant correlations between HFRs and the n-3/n-6 FA ratio, and, in agreement, only few significant associations with the traditional marine food intake (PBDE153 associated positive with Greenlandic fish, PBDE28 associated positive with dried fish).

It is well known that the marine mammals contain high concentrations of the lipPOPs, especially whale and seal blubber (Johansen et al., 2004). Medium to high (5–500 ng/g Σ_{10}PCB) concentrations were also found in seabird muscle and liver (guillemot, eider, and kittiwake), while low (<5 ng/g Σ_{10}PCB) concentrations were found in terrestrial animals (hare, caribou, muskox, and lamb) (Johansen et al., 2004). The results are in agreement with the significant positive associations between lipPOP serum concentrations and intake of marine mammals and seabirds in the present study. Low to medium (<50 ng/g Σ_{10}PCB) concentrations have generally been found in muscle and liver from fish (Johansen et al., 2004), however, no studies have reported concentrations in dried fish. All lipPOP serum concentrations (except PBDE153 and anti-DP) were associated with intake of dried fish (discussed further below).

The literature on PFAS concentrations in traditional Greenlandic food items is scarce. Carlsson et al. have reported low concentrations in whale beef, seal beef, and narwhal mattak and non-detectable concentrations in fish fillets (fresh salmon, smoked salmon, and smoked halibut) (Carlsson et al., 2014). In agreement with these findings, we found a significant association of PFAS serum concentrations with intake of marine mammals (seal, whale, blubber, walrus, and polar bear). However, we also observed few positive associations with Greenlandic fish intake and several positive associations with dried fish intake. Our FFQ includes more Greenlandic fish species (cod, trout, Greenlandic halibut, Atlantic halibut, redfish, Atlantic salmon, Atlantic wolffish, and capelin) (Wielsøe et al., 2021) than those studied in Carlsson et al. (2014) (fresh salmon, smoked salmon, and smoked halibut), which also did not include dried fish. Human exposure routes to PFAS are not sufficiently understood, but fish, other seafood, and meat are often mentioned as the most important sources of exposure (EFSA, 2012). In Nunavut, Canada, traditional foods contributed more to the PFAS exposure than market foods, and caribou meat contributed with 43–75% of the daily PFAS dietary exposure (Ostertag et al., 2009). However, we did not observe any associations between PFAS concentration and intake of terrestrial animals (including grouse, Greenlandic lamb/sheep, caribou muskox, and hare) or with caribou alone (not shown). Our results call for further studies of PFAS concentration in Greenlandic food items.

The dried fish intake includes the summed intake of dried Greenlandic halibut, capelin, cod, wolffish, and other dried fish (Wielsøe et al., 2021). Dried cod was the most frequently consumed dried fish species with a mean intake of 1.97 times/months, followed by dried capelin (mean 0.93 times/months) and dried Greenlandic halibut (mean 0.63 times/months), while the mean intake of dried wolffish and other dried fish was less than 0.5 times/months (Wielsøe et al., 2021). No reports on lipPOP and PFAS concentrations in dried fish are available, however, in fresh fish, higher lipPOP concentrations have been found in Greenlandic halibut compared to capelin, cod and wolffish (Johansen et al., 2004). Thus, it might be expected that intake of dried Greenlandic halibut was more strongly associated with human serum POP concentrations than intake of dried capelin, cod, and wolffish. However, when looking into the association between the POP concentrations and intake of the specific dried fish species in the present study, the associations were strongest and significant for dried capelin, cod, and wolffish and non-significant for Greenlandic halibut and other dried fish (not shown). This unexpected result may be related to differences in the intake amount as we only measured the intake frequencies and a narrow distribution of dried halibut intake as the majority of the participants reported intake never (15%) or less than once a month (64%). Further studies should elucidate the associations including information on intake amounts and lipPOP and PFAS concentrations in dried fish.

The negative associations between POP concentrations and imported meat intake were unexpected, since reports suggest meat as an important contributor to both lipPOP and PFAS exposure in European and Asian populations (Barone et al., 2021; EFSA, 2012; Zhou et al., 2012). However, a high intake of a given food item relates to a lower intake of other food items. Thus, intake of other food items with higher POP concentrations may confound the negative associations between POP concentrations and imported meat intake. For instance, having a high intake of marine mammals and seabirds (with higher POP concentrations than imported meat (Johansen et al., 2004; Saktrakulkla et al., 2020; Voorspoels et al., 2008)) may correlate with a lower intake of imported meat. When adjusting further for intake of traditional foods, the association between POP concentrations and imported meat intake remained significant (not shown),

indicating that high intake of traditional food may not fully explain the negative association between imported meat intake and POP concentrations, thus other factors may affect the association as well. As the POP concentrations were not associated with any of the other imported food groups, the associations of POPs with imported meat intake drive the negative associations with the sum of all imported food groups.

4.3. Strengths and limitations of the study

One of the main strengths of our study is the inclusion of a large number of contaminants from diverse groups of chemicals. The chemical measurements was conducted in laboratories with a well-established quality assurance/quality control system, including the participation in proficiency testing schemes, such as the AMAP Ring Test for Persistent Organic Pollutants in Human Serum, ensure high quality of the measurements. However, we observed some disagreements between the measurements at INSPQ and AU for the PBDEs (PBDE47, PBDE99, and PBDE153) in some samples (Table S2). The disagreements are most likely due to differences in analytical methods and concentrations close to LOD. Thus, both measurements confirm low concentrations of PBDEs in the human serum samples from Greenland. Any imprecision in the measurements is likely to be non-differential and without any influence on the observed results.

Other strengths of the study are the inclusion of both men and women and the repeated measurements in the mothers providing longitudinal data not previously reported for Greenland. The majority of biomonitoring studies in Greenland include pregnant women or women in the childbearing age (AMAP, 2016a), and updated biomonitoring data on Greenlandic men have been missing in recent years. This study is strengthened by the inclusion of both sexes, although the relatively narrow age range (24–55 years) and few geographical locations (Nuuk, Sisimiut, and Ilulissat) may limit the generalizability and extern validity. The observed POP concentrations might have been higher for older age groups and participants from settlements at the north and east coast of Greenland. However, more than 40% of the adult population in Greenland are within the included age group and live in the included towns (Grønlands statistik, n.d.) and thus, the reported data reflect a large part of the general adult population in Greenland.

The information on lifestyle, socioeconomic factors, and food intake were self-reported and recall bias might affect the data. However, we believe that the possible recall would be non-differential, not affected by the individual's POP concentrations and, therefore, reducing the magnitude of the associations found. FFQs seem to be an appropriate dietary assessment method for the persistent compounds with long half-lives included in the present study. However, it would have been desirable to have more information on cooking procedures, total energy intake, and portion size. Furthermore, we cannot rule out the presence of residual confounding that might bias the results or potential confounders not taken into account. However, we have included the most important confounders, such as age, sex, and residential town.

5. Conclusion

Similarly to previous studies, we found that POP concentrations generally differed by sex (Fathers > Mothers) and geographical location (Ilulissat > Sisimiut > Nuuk). We report for the very first time novel and unregulated HFR (DPTE, EHTBB, BTBPE, syn-DP, and anti-DP) concentrations in Greenlandic human serum samples; however, only the concentration of anti-DP was above LOD in more than 50% of the samples. Similarly, the PBDE concentrations were lower than in other Arctic and European regions, whereas PCB, OCP and PFAS concentrations were comparable or higher, even though a decrease has been seen during recent years. We also reported, for the first time, intra-individual changes in POP concentrations in Greenlandic women, and found decreases of 16–58% from 2013—2015 to 2019—2020 (although 0% for mirex).

Our results indicate that intake of marine mammals, seabirds, and dried fish are the primary dietary exposure sources for the measured POPs. The findings may be of interest for future dietary recommendations in Greenland. However, further research should be undertaken to investigate and update the concentrations of POPs in the food items (especially data on dried fish), before specific recommendations can be given.

CRediT authorship contribution statement

Maria Wielsøe: Conceptualization, Methodology, Formal analysis, Writing - Original Draft, Visualization, Project administration. **Manhai Long**: Conceptualization, Methodology, Writing - Review & Editing, Supervision. **Rossana Bossi**: Investigation (PFAS measurements at AU), Writing - Review & Editing. **Katrin Vorkamp**: Investigation (HFR measurements at AU), Writing - Review & Editing. **Eva Cecilie Bonefeld-Jørgensen**: Conceptualization, Methodology, Writing - Review & Editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

The authors thank all the participating families. We gratefully acknowledge the health nurse visitors at “Sundhedsplejen” in Nuuk and Sisimiut and the staff at the clinical hospital laboratory at The Regional Hospital Sisimiut and Dronning Ingrid's Hospital in Nuuk, Greenland. The authors would also like to thank colleagues at the Centre for Arctic Health & Molecular Epidemiology, Department of Public Health, Aarhus University for their scientific support. We also thank Inga Jensen, Tobias Meulengracht, Annegrete Ljungqvist, and Birgit Groth at the Department of Environmental Science, Aarhus University, for technical assistance with the chemical analyses.

The establishment of the ACCEPT cohort was funded by the Danish Environmental Protection Agency (MST-112-00225 & MST-112-00289), and the follow-up was funded by the Danish Environmental Protection Agency (MST-113-00092) under the Danish Cooperation for Environment in the Arctic (DANCEA) programme.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.154270.

References

Abass, K., Emelyanova, A., Rautio, A., 2018. Temporal trends of contaminants in Arctic human populations. Environ. Sci. Pollut. Res. 25 (29), 28834–28850. https://doi.org/10.1007/s11356-018-2936-8.

Agudo, A., Goñi, F., Etxeandia, A., Vives, A., Millán, E., López, R., Amiano, P., Ardanaz, E., Barricarte, A., Dolores Chirlaque, M., Dorronsoro, M., Jakszyn, P., Larrañaga, N., Martínez, C., Navarro, C., Rodríguez, L., José Sánchez, M., José Tormo, M., González, C.A., 2009. Polychlorinated biphenyls in Spanish adults: determinants of serum concentrations. Environ. Res. 109 (5), 620–628. https://doi.org/10.1016/j.envres.2009.03.009.

Akins, J.R., Waldrep, K., Bernert, J.T., 1989. The estimation of total serum lipids by a completely enzymatic ‘summation’ method. Clin. Chim. Acta 184 (3), 219–226. https://doi.org/10.1016/0009-8981(89)90054-5.

AMAP, 1998. Assessment Report: Arctic Pollution Issues.

AMAP, 2003. AMAP Assessment 2002: Human Health in the Arctic.

AMAP, 2009. AMAP Assessment 2009: Human Health in the Arctic.

AMAP, 2016a. AMAP Assessment 2015: Human Health in the Arctic. . https://www.ncbi.nlm.nih.gov/pubmed/28156426.

AMAP, 2016b. AMAP Assessment 2015: Temporal Trends in Persistent Organic Pollutants in the Arctic.

AMAP, 2021. Human Health in the Arctic 2021. Summary for Policy-makers. T. Arctic Monitoring and Assessment Programme (AMAP), Norway.

ATSDR, 2020. Toxicological Profile for Mirex and Chlordecone.

Bank-Nielsen, P.I., Long, M., Bonefeld-Jorgensen, E.C., 2019. Pregnant Inuit women's exposure to metals and association with fetal growth outcomes: ACCEPT 2010(-)2015. Int. J. Environ Res Public Health 16 (7). https://doi.org/10.3390/ijerph16071171.

Barone, G., Storelli, A., Busco, A., Mallamaci, R., Storelli, M.M., 2021. Polychlorinated dioxins, furans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (dl-PCBs) in food from Italy: estimates of dietaryintake and assessment. J. Food Sci. https://doi.org/10.1111/1750-3841.15901.

Barrie, L.A., Gregor, D., Hargrave, B., Lake, R., Muir, D., Shearer, R., Tracey, B., Bidleman, T., 1992. Arctic contaminants: sources, occurrence and pathways. Sci. Total Environ. 122 (1), 1–74. https://doi.org/10.1016/0048-9697(92)90245-N.

Berg, V., Sandanger, T.M., Hanssen, L., Rylander, C., Nøst, T.H., 2021. Time trends of perfluoroalkyl substances in blood in 30-year old Norwegian men and women in the period 1986–2007. Environ. Sci. Pollut. Res. 28 (32), 43897–43907. https://doi.org/10.1007/s11356-021-13809-6.

Bjermo, H., Darnerud, P.O., Lignell, S., Pearson, M., Rantakokko, P., Nälsén, C., Enghardt Barbieri, H., Kiviranta, H., Lindroos, A.K., Glynn, A., 2013. Fish intake and breastfeeding time are associated with serum concentrations of organochlorines in a swedish population. Environ. Int. 51, 88–96. https://doi.org/10.1016/j.envint.2012.10.010.

Bjerregaard, P., Aidt, E.C., 2010. Levevilkår, livsstil og helbred - Befolkningsundersøgelsen i Grønland 2005-2009. https://www.sdu.dk/da/sif/rapporter/2010/levevilkaar_livsstil_og_helbred.

Bjerregaard, P., Mulvad, G., 2012. The best of two worlds: how the Greenland Board of Nutrition has handled conflicting evidence about diet and health. Int. J. Circumpolar Health 71, 18588. https://doi.org/10.3402/ijch.v71i0.18588.

Bjerregaard, P., Pedersen, H.S., Nielsen, N.O., Dewailly, E., 2013. Population surveys in Greenland 1993–2009: temporal trend of PCBs and pesticides in the general inuit population by age and urbanisation. Sci. Total Environ. 454, 283–288. https://doi.org/10.1016/j.scitotenv.2013.03.031.

Bjerregaard-Olesen, C., Bach, C.C., Long, M., Ghisari, M., Bech, B.H., Nohr, E.A., Henriksen, T.B., Olsen, J., Bonefeld-Jørgensen, E.C., 2016. Determinants of serum levels of perfluorinated alkyl acids in Danish pregnant women. Int. J. Hyg. Environ. Health 219 (8), 867–875. https://doi.org/10.1016/j.ijheh.2016.07.008.

Bjerregaard-Olesen, C., Ghisari, M., Bonefeld-Jorgensen, E.C., 2016. Activation of the estrogen receptor by human serum extracts containing mixtures of perfluorinated alkyl acids from pregnant women. Environ. Res. 151, 71–79. https://doi.org/10.1016/j.envres.2016.07.001.

Bonefeld-Jorgensen, E.C., 2010. Biomonitoring in Greenland: human biomarkers of exposure and effects - a short review. Rural Remote Health 10. https://doi.org/10.22605/RRH1362.

Bonefeld-Jorgensen, E.C., Long, M., Fredslund, S.O., Bossi, R., Olsen, J., 2014. Breast cancer risk after exposure to perfluorinated compounds in Danish women: a case-control study nested in the Danish National Birth Cohort. Cancer Causes Control 25 (11), 1439–1448. https://doi.org/10.1007/s10552-014-0446-7.

Borgå, K., Fisk, A.T., Hoekstra, P.F., Muir, D.C.G., 2004. Biological and chemical factors of importance in the bioaccumulation and trophic transfer of persistent organochlorine contaminants in arctic marine food webs. Environ. Toxicol. Chem. 23 (10), 2367–2385. https://doi.org/10.1897/03-518.

Bossi, R., Riget, F.F., Dietz, R., 2005. Temporal and spatial trends of perfluorinated compounds in ringed seal (Phoca hispida) from Greenland. Environ. Sci. Technol. 39. https://doi.org/10.1021/es0508469.

Byrne, S., Seguinot-Medina, S., Miller, P., Waghiyi, V., von Hippel, F.A., Buck, C.L., Carpenter, D.O., 2017. Exposure to polybrominated diphenyl ethers and perfluoroalkyl substances in a remote population of Alaska Natives. Environ. Pollut. 231 (Pt 1), 387–395. https://doi.org/10.1016/j.envpol.2017.08.020.

Carlsson, P., Herzke, D., Kallenborn, R., 2014. Polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and perfluorinated alkylated substances (PFASs) in traditional seafood items from western Greenland. Environ. Sci. Pollut. Res. 21 (6), 4741–4750. https://doi.org/10.1007/s11356-013-2435-x.

Caron-Beaudoin, É., Ayotte, P., Blanchette, C., Muckle, G., Avard, E., Ricard, S., Lemire, M., 2020. Perfluoroalkyl acids in pregnant women from Nunavik (Quebec, Canada): trends in exposure and associations with country foods consumption. Environ. Int. 145, 106169. https://doi.org/10.1016/j.envint.2020.106169.

Chevrier, C., Warembourg, C., Gaudreau, E., Monfort, C., Le Blanc, A., Guldner, L., Cordier, S., 2013. Organochlorine pesticides, polychlorinated biphenyls, seafood consumption, and time-to-pregnancy. Epidemiology 24 (2), 251–260. https://doi.org/10.1097/EDE.0b013e31827f53ec.

D'Hollander, W., de Voogt, P., De Coen, W., Bervoets, L., 2010. Perfluorinated substances in human food and other sources of human exposure. In: De Voogt, P. (Ed.), Reviews of Environmental Contamination and Toxicology Volume 208: Perfluorinated Alkylated Substances. Springer, New York, pp. 179–215 https://doi.org/10.1007/978-1-4419-6880-7_4.

Dahl-Petersen, I.K., Larsen, C.V.L., Nielsen, N.O., Jørgensen, M.E., Bjerregaard, P., 2016. Befolkningsundersøgelsen i Grønland 2014. Levevilkår, livsstil og helbred.

Deutch, B., Pedersen, H.S., Jørgensen, E.C., Hansen, J.C., 2003. Smoking as a determinant of high organochlorine levels in Greenland. Arch. Environ. Health 58 (1), 30–36. https://doi.org/10.3200/aeoh.58.1.30-36.

Deutch, B., Pedersen, H.S., Hansen, J.C., 2004. Dietary composition in Greenland 2000, plasma fatty acids and persistent organic pollutants. Sci. Total Environ. 331 (1), 177–188. https://doi.org/10.1016/j.scitotenv.2004.03.028.

Deutch, B., Dyerberg, J., Pedersen, H.S., Asmund, G., Moller, P., Hansen, J.C., 2006. Dietary composition and contaminants in north Greenland, in the 1970s and 2004. Sci. Total Environ. 370 (2–3), 372–381. https://doi.org/10.1016/j.scitotenv.2006.07.015.

Deutch, B., Dyerberg, J., Pedersen, H.S., Aschlund, E., Hansen, J.C., 2007. Traditional and modern Greenlandic food - dietary composition, nutrients and contaminants. Sci. Total Environ. 384 (1–3), 106–119. https://doi.org/10.1016/j.scitotenv.2007.05.042.

Deutch, B., Pedersen, H.S., Asmund, G., Hansen, J.C., 2007. Contaminants, diet, plasma fatty acids and smoking in Greenland 1999–2005. Sci. Total Environ. 372 (2–3), 486–496. https://doi.org/10.1016/j.scitotenv.2006.10.043.

Dewailly, E.E., Blanchet, C., Gingras, S., Lemieux, S., Sauvé, L., Bergeron, J., Holub, B.J., 2001. Relations between n-3 fatty acid status and cardiovascular disease risk factors among Quebecers. Am. J. Clin. Nutr. 74 (5), 603–611. https://doi.org/10.1093/ajcn/74.5.603.

Domingo, J.L., Nadal, M., 2017. Per- and polyfluoroalkyl substances (PFASs) in food and human dietary intake: a review of the recent scientific literature. J. Agric. Food Chem. 65 (3), 533–543. https://doi.org/10.1021/acs.jafc.6b04683.

Drysdale, M., Ratelle, M., Skinner, K., Garcia-Barrios, J., Gamberg, M., Williams, M., Majowicz, S., Bouchard, M., Stark, K., Chalil, D., Laird, B.D., 2021. Human biomonitoring results of contaminant and nutrient biomarkers in old crow, Yukon,Canada. Sci. Total Environ. 760, 143339. https://doi.org/10.1016/j.scitotenv.2020.143339.

Dudarev, A., Dushkina, E., Sladkova, Y., Chupakhin, V., Lukicheva, L., 2016. Exposure levels of persistent organic pollutants (POPs) among population of the Pechenga District in the Murmansk Region. Toxicol. Rev. 138, 2–9.

EFSA, 2012. Perfluoroalkylated substances in food: occurrence and dietary exposure. http://onlinelibrary.wiley.com/doi/10.2903/j.efsa.2012.2743/epdf.

Eriksson, U., Kärrman, A., Rotander, A., Mikkelsen, B., Dam, M., 2013. Perfluoroalkyl substances (PFASs) in food and water from Faroe Islands. Environ. Sci. Pollut. Res. Int. 20 (11), 7940–7948. https://doi.org/10.1007/s11356-013-1700-3.

Fisher, M., Arbuckle, T.E., Liang, C.L., LeBlanc, A., Gaudreau, E., Foster, W.G., Haines, D., Davis, K., Fraser, W.D., 2016. Concentrations of persistent organic pollutants in maternal and cord blood from the maternal-infant research on environmental chemicals (MIREC) cohort study. Environ. Health 15 (1), 59. https://doi.org/10.1186/s12940-016-0143-y.

Folch, J., Lees, M., Sloane Stanley, G.H., 1957. A simple method for the isolation and purification of total lipides from animal tissues. J. Biol. Chem. 226 (1), 497–509.

Fromme, H., Cequier, E., Kim, J.-T., Hanssen, L., Hilger, B., Thomsen, C., Chang, Y.-S., Völkel, W., 2015. Persistent and emerging pollutants in the blood of German adults: occurrence of dechloranes, polychlorinated naphthalenes, and siloxanes. Environ. Int. 85, 292–298. https://doi.org/10.1016/j.envint.2015.09.002.

Fujii, Y., Harada, K.H., Koizumi, A., 2013. Occurrence of perfluorinated carboxylic acids (PFCAs) in personal care products and compounding agents. Chemosphere 93 (3), 538–544. https://doi.org/10.1016/j.chemosphere.2013.06.049.

Garcia-Barrios, J., Drysdale, M., Ratelle, M., Gaudreau, É., LeBlanc, A., Gamberg, M., Laird, B.D., 2021. Biomarkers of poly- and perfluoroalkyl substances (PFAS) in sub-Arctic and Arctic communities in Canada. Int. J. Hyg. Environ. Health 235, 113754. https://doi.org/10.1016/j.ijheh.2021.113754.

Gribble, M.O., Bartell, S.M., Kannan, K., Wu, Q., Fair, P.A., Kamen, D.L., 2015. Longitudinal measures of perfluoroalkyl substances (PFAS) in serum of Gullah African Americans in South Carolina: 2003–2013. Environ. Res. 143, 82–88. https://doi.org/10.1016/j.envres.2015.03.012.

Grønlands statistik, .. . Retrieved 14. october 2021 from www.bank.stat.gl/.

Gyllenhammar, I., Benskin, J.P., Plassmann, M., Sandblom, O., Kallerman, P.H., Lampa, E., Ankarberg, E.H., 2020. Levels of perfluoroalkyl substances (PFAS) in individual serum samples from first-time mothers in Uppsala, Sweden: results from year 2017-2019, and temporal trends for the time period 1996-2019. Livsmedelsverket. https://www.diva-portal.org/smash/get/diva2:1509601/FULLTEXT01.pdf.

Hagmar, L., Wallin, E., Vessby, B., Jönsson, B.A.G., Bergman, Å., Rylander, L., 2006. Intra-individual variations and time trends 1991–2001 in human serum levels of PCB, DDE and hexachlorobenzene. Chemosphere 64 (9), 1507–1513. https://doi.org/10.1016/j.chemosphere.2005.12.054.

Heidam, N.Z., Christensen, J., Wåhlin, P., Skov, H., 2004. Arctic atmospheric contaminants in NE Greenland: levels, variations, origins, transport, transformations and trends 1990–2001. Sci. Total Environ. 331 (1), 5–28. https://doi.org/10.1016/j.scitotenv.2004.03.033.

Hjermitslev, M.H., Long, M., Wielsoe, M., Bonefeld-Jorgensen, E.C., 2019. Persistent organic pollutants in Greenlandic pregnant women and indices of foetal growth: the ACCEPT study. Sci. Total Environ. 698, 134118. https://doi.org/10.1016/j.scitotenv.2019.134118.

Høyer, A.P., Jørgensen, T., Grandjean, P., Hartvig, H.B., 2000. Repeated measurements of organochlorine exposure and breast cancer risk (Denmark). Cancer Causes Control 11 (2), 177–184. https://doi.org/10.1023/A:1008926219539.

Johansen, P., Muir, D., Asmund, G., Riget, F., 2004. Human exposure to contaminants in the traditional Greenland diet. Sci. Total Environ. 331 (1–3), 189–206. https://doi.org/10.1016/j.scitotenv.2004.03.029.

Knudsen, A.K., Long, M., Pedersen, H.S., Bonefeld-Jorgensen, E.C., 2015. Lifestyle, reproductive factors and food intake in Greenlandic pregnant women: the ACCEPT - sub-study. Int. J. Circumpolar Health 74, 29469. https://doi.org/10.3402/ijch.v74.29469.

Kok Grouleff, M., Wielsøe, M., Berthelsen, D., Mulvad, G., Isidor, S., Long, M., Bonefeld-Jørgensen, E.C., 2021. Anthropometric measures and blood pressure of Greenlandic preschool children. Int. J. Circumpolar Health 80 (1), 1954382. https://doi.org/10.1080/22423982.2021.1954382.

Kornvig, S., Wielsøe, M., Long, M., Bonefeld-Jørgensen, E.C., 2021. Prenatal exposure to persistent organic pollutants and metals and problematic child behavior at 3–5 years of age: a Greenlandic cohort study. Sci. Rep. 11 (1), 22182. https://doi.org/10.1038/s41598-021-01580-0.

Larsen, C.V.L., Hansen, C.B., Ingemann, C., Jørgensen, M.E., Olesen, I., Sørensen, I.K., Koch, A., Backer, V., Bjerregaard, P., 2019. Befolkningsundersøgelsen i Grønland 2018. Levevilkår, livsstil og helbred.

Long, M., Knudsen, A.K., Pedersen, H.S., Bonefeld-Jorgensen, E.C., 2015. Food intake and serum persistent organic pollutants in the Greenlandic pregnant women: the ACCEPT sub-study. Sci. Total Environ. 529, 198–212. https://doi.org/10.1016/j.scitotenv.2015.05.022.

Long, M., Wielsøe, M., Bonefeld-Jørgensen, E.C., 2021. Time trend of persistent organic pollutants and metals in Greenlandic Inuit during 1994–2015. Int. J. Environ. Res. Public Health 18 (5), 2774. https://doi.org/10.3390/ijerph18052774.

Macdonald, R.W., Barrie, L.A., Bidleman, T.F., Diamond, M.L., Gregor, D.J., Semkin, R.G., Strachan, W.M.J., Li, Y.F., Wania, F., Alaee, M., Alexeeva, L.B., Backus, S.M., Bailey, R.,

Bewers, J.M., Gobeil, C., Halsall, C.J., Harner, T., Hoff, J.T., Jantunen, L.M.M., Yunker, M.B., 2000. Contaminants in the Canadian Arctic: 5 years of progress in understanding sources, occurrence and pathways. Sci. Total Environ. 254 (2), 93–234. https://doi.org/10.1016/S0048-9697(00)00434-4.

Morrison, W.R., Smith, L.M., 1964. Preparation of fatty acids methyl esters and dimethylacetals from lipids with boron fluoride-methanol. J. Lipid Res. 5, 600–608.

Mulvad, G., 2018. Food security in Greenland. In: Exner-Pirot, H., Norbye, B., Butler, L. (Eds.), Northern and Indigenous Health and Health Care. University of Saskatchewan, Saskatoon, Saskatchewan (pp. Available from: openpress.usask.ca/northernhealthcare).

Nøst, T.H., Breivik, K., Fuskevåg, O.M., Nieboer, E., Odland, J., Sandanger, T.M., 2013. Persistent organic pollutants in Norwegian men from 1979 to 2007: intraindividual changes, age-period-cohort effects, and model predictions. Environ. Health Perspect. 121 (11–12), 1292–1298. https://doi.org/10.1289/ehp.1206317.

Nøst, T.H., Vestergren, R., Berg, V., Nieboer, E., Odland, J.Ø., Sandanger, T.M., 2014. Repeated measurements of per- and polyfluoroalkyl substances (PFASs) from 1979 to 2007 in males from Northern Norway: assessing time trends, compound correlations and relations to age/birth cohort. Environ. Int. 67, 43–53. https://doi.org/10.1016/j.envint.2014.02.011.

Nøst, T.H., Berg, V., Hanssen, L., Rylander, C., Gaudreau, E., Dumas, P., Breivik, K., Sandanger, T.M., 2019. Time trends of persistent organic pollutants in 30 year olds sampled in 1986, 1994, 2001 and 2007 in Northern Norway: measurements, mechanistic modeling and a comparison of study designs. Environ. Res. 172, 684–692. https://doi.org/10.1016/j.envres.2019.02.047.

Ostertag, S.K., Tague, B.A., Humphries, M.M., Tittlemier, S.A., Chan, H.M., 2009. Estimated dietary exposure to fluorinated compounds from traditional foods among Inuit in Nunavut,Canada. Chemosphere 75 (9), 1165–1172. https://doi.org/10.1016/j.chemosphere.2009.02.053.

PAME, 2021. MPA Information Brief: Indigenous Food Security in the Arctic. . https://oaarchive.arctic-council.org/bitstream/handle/11374/2662/MMIS12_2021_REYKJAVIK_PAME_Indigenous-Food-Security.pdf?sequence=1&isAllowed=y.

Phillips, D.L., Pirkle, J.L., Burse, V.W., Bernert, J.T., Henderson, L.O., Needham, L.L., 1989. Chlorinated-hydrocarbon levels in human-serum - effects of fasting and feeding. Arch. Environ. Contam. Toxicol. 18 (4), 495–500 <Go to ISI>://WOS:A1989AB29400005.

Rigét, F., Bossi, R., Sonne, C., Vorkamp, K., Dietz, R., 2013. Trends of perfluorochemicals in Greenland ringed seals and polar bears: indications of shifts to decreasing trends. Chemosphere 93 (8), 1607–1614. https://doi.org/10.1016/j.chemosphere.2013.08.015.

Rigét, F., Vorkamp, K., Bossi, R., Sonne, C., Letcher, R.J., Dietz, R., 2016. Twenty years of monitoring of persistent organic pollutants in Greenland biota.A review. Environ. Pollut. 217, 114–123. https://doi.org/10.1016/j.envpol.2015.11.006.

Rigét, F., Bignert, A., Braune, B., Dam, M., Dietz, R., Evans, M., Green, N., Gunnlaugsdóttir, H., Hoydal, K.S., Kucklick, J., Letcher, R., Muir, D., Schuur, S., Sonne, C., Stern, G., Tomy, G., Vorkamp, K., Wilson, S., 2019. Temporal trends of persistent organic pollutants in Arctic marine and freshwater biota. Sci. Total Environ. 649, 99–110. https://doi.org/10.1016/j.scitotenv.2018.08.268.

Saktrakulkla, P., Lan, T., Hua, J., Marek, R.F., Thorne, P.S., Hornbuckle, K.C., 2020. Polychlorinated biphenyls in food. Environ. Sci. Technol. 54 (18), 11443–11452. https://doi.org/10.1021/acs.est.0c03632.

Schæbel, L.K., Bonefeld-Jørgensen, E.C., Vestergaard, H., Andersen, S., 2017. The influence of persistent organic pollutants in the traditional Inuit diet on markers of inflammation. PLoS One 12 (5), e0177781. https://doi.org/10.1371/journal.pone.0177781.

Shu, H., Lindh, C.H., Wikström, S., Bornehag, C.-G., 2019. Temporal trends and predictors of perfluoroalkyl substances serum levels in Swedish pregnant women in the SELMA study. PLoS One 13 (12), e0209255. https://doi.org/10.1371/journal.pone.0209255.

Stark, K.D., Holub, B.J., 2004. Differential eicosapentaenoic acid elevations and altered cardiovascular disease risk factor responses after supplementation with docosahexaenoic acid in postmenopausal women receiving and not receiving hormone replacement therapy. Am. J. Clin. Nutr. 79 (5), 765–773. https://doi.org/10.1093/ajcn/79.5.765.

Stubleski, J., Lind, L., Salihovic, S., Lind, P.M., Kärrman, A., 2018. Longitudinal changes in persistent organic pollutants (POPs) from 2001 to 2009 in a sample of elderly Swedish men and women. Environ. Res. 165, 193–200. https://doi.org/10.1016/j.envres.2018.04.009.

Sweeney, A.M., Symanski, E., Burau, K.D., Kim, Y.J., Humphrey, H.E.B., Ann Smithci, M., 2001. Changes in serum PBB and PCB levels over time among women of varying ages at exposure. Environ. Res. 86 (2), 128–139. https://doi.org/10.1006/enrs.2001.4261.

Terkelsen, A.S., Long, M., Hounsgaard, L., Bonefeld-Jorgensen, E.C., 2017. Reproductive factors, lifestyle and dietary habits among pregnant women in Greenland: the ACCEPT substudy 2013–2015. Scand. J. Public Health https://doi.org/10.1177/1403494817714188 1403494817714188.

Tsai, M.-S., Miyashita, C., Araki, A., Itoh, S., Bamai, Y.A., Goudarzi, H., Okada, E., Kashino, I., Matsuura, H., Kishi, R., 2018. Determinants and temporal trends of perfluoroalkyl substances in pregnant women: the Hokkaido study on environment and children's health. Int. J. Environ. Res. Public Health 15 (5), 989. https://doi.org/10.3390/ijerph15050989.

UNEP, .. United Nations Environment Programme (UNEP): Stockholm Convention. Retrieved 21 July 2021 from http://www.pops.int/.

Voorspoels, S., Covaci, A., Neels, H., 2008. Dietary PCB intake in Belgium. Environ. Toxicol. Pharmacol. 25 (2), 179–182. https://doi.org/10.1016/j.etap.2007.10.013.

Vorkamp, K., 2012. Chapter 3D - brominated flame retardants: biomarkers and human biomonitoring. Biomarkers and Human Biomonitoring. vol. 1, 1, pp. 288–321. https://doi.org/10.1039/9781849733373-00288.

Vorkamp, K., Nielsen, F., Kyhl, H.B., Husby, S., Nielsen, L.B., Barington, T., Andersson, A.M., Jensen, T.K., 2014. Polybrominated diphenyl ethers and perfluoroalkyl substances in serum of pregnant women: levels, correlations, and potential health implications. Arch. Environ. Contam. Toxicol. 67 (1), 9–20. https://doi.org/10.1007/s00244-013-9988-z.

Vorkamp, K., Bossi, R., Rigét, F.F., Skov, H., Sonne, C., Dietz, R., 2015. Novel brominated flame retardants and dechlorane plus in Greenland air and biota. Environ. Pollut. 196, 284–291. https://doi.org/10.1016/j.envpol.2014.10.007.

Weihe, P., Debes, F., Halling, J., Petersen, M.S., Muckle, G., Odland, J.Ø., Dudarev, A., Ayotte, P., Dewailly, É., Grandjean, P., Bonefeld-Jørgensen, E., 2016. Health effects associated with measured levels of contaminants in the Arctic. Int. J. Circumpolar Health 75 (1), 33805. https://doi.org/10.3402/ijch.v75.33805.

Wielsøe, M., Berthelsen, D., Mulvad, G., Isidor, S., Long, M., Bonefeld-Jørgensen, E.C., 2021. Dietary habits among men and women in West Greenland: follow-up on the ACCEPT birth cohort. BMC Public Health 21 (1), 1426. https://doi.org/10.1186/s12889-021-11359-7.

Wolff, M.S., Zeleniuch-Jacquotte, A., Dubin, N., Toniolo, P., 2000. Risk of breast cancer and organochlorine exposure. Cancer Epidemiol. Biomark. Prev. 9 (3), 271–277.

Yang, C., Jílková, S.R., Melymuk, L., Harris, S.A., Jantunen, L.M., Pertili, J., Winn, L., Diamond, M.L., 2020. Are we exposed to halogenated flame retardants from both primary and secondary sources? Environ. Sci. Technol. Lett. 7 (8), 585–593. https://doi.org/10.1021/acs.estlett.0c00268.

Yin, J.-F., Li, J.-F.-T., Li, X.-H., Yang, Y.-L., Qin, Z.-F., 2020. Bioaccumulation and transfer characteristics of dechlorane plus in human adipose tissue and blood stream and the underlying mechanisms. Sci. Total Environ. 700, 134391. https://doi.org/10.1016/j.scitotenv.2019.134391.

Zhou, P., Zhao, Y., Li, J., Wu, G., Zhang, L., Liu, Q., Fan, S., Yang, X., Li, X., Wu, Y., 2012. Dietary exposure to persistent organochlorine pesticides in 2007 Chinese total diet study. Environ. Int. 42, 152–159. https://doi.org/10.1016/j.envint.2011.05.018.

Zhou, S.N., Siddique, S., Lavoie, L., Takser, L., Abdelouahab, N., Zhu, J., 2014. Hexachloronorbornene-based flame retardants in humans: levels in maternal serum and milk. Environ. Int. 66, 11–17. https://doi.org/10.1016/j.envint.2014.01.010.