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Associations between dietary profiles and perfluoroalkyl acids in Inuit youth and adults

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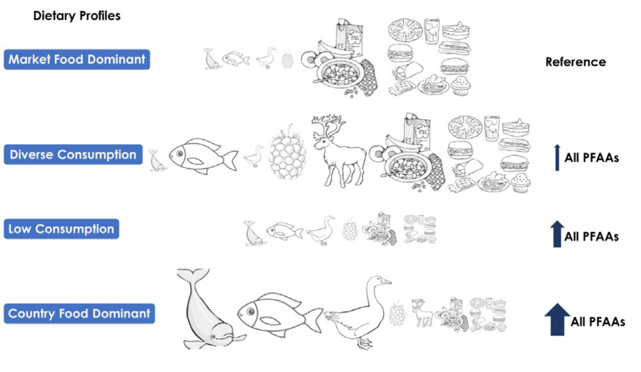
HIGHLIGHTS

- High concentrations of environmentally persistent perfluoroalkyl acids (PFAAs) were detected in Inuit populations.
- We examined associations between dietary profiles and PFAAs concentrations using data from a large survey in Nunavik.
- The strongest associations were between food profiles with frequent country food consumption and long-chain PFAAs.
- Dietary profiles with frequent consumption of marine mammals were also associated with higher concentrations of PFAAs.
- These results provide evidence for country foods as an exposure source of PFAAs in Arctic communities.

GRAPHICAL ABSTRACT



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ABSTRACT

Background: Perfluoroalkyl acids (PFAAs), a subset of perfluoroalkyl substances (PFAS), are synthetic chemicals used in industrial and consumer applications. They are exceptionally stable and highly mobile in the environment, and were detected in high concentrations in Arctic wildlife and Nunavik Inuit. The study's objective was to study the association between dietary profiles in Nunavik and plasma PFAAs concentrations.

Methods: The study used data from the *Qanuilirpitaa?* 2017 Nunavik Inuit Health Survey (Q2017) (N = 1172) on Inuit adults aged 16–80 years. Nine PFAAs congeners were measured in plasma samples (six were detected). Dietary profiles were identified using latent profile analysis. Two sets of dietary profiles were included; the first included market (store-bought) and country foods (harvested/hunted from the land), and the second included only country foods. Multiple linear regression models regressed log-transformed PFAAs concentrations against the dietary profiles, adjusting for sociodemographic variables.

Results: We identified statistically significant 24.54–57.55 % increases in all PFAAs congeners (PFOA, PFNA, PFDA, PFUnDA, PFHxS, and PFOS) in the dietary profile defined by frequent country food consumption compared to the dietary profile defined by frequent market food consumption. Individuals defined by low consumption of foods (related to food insecurity) had higher concentrations of six PFAAs compared to individuals with frequent market food consumption. The associations were stronger with profiles defined by more frequent country food consumption, and particularly those with increased marine mammal consumption. PFDA, PFUnDA, and PFOS were particularly associated

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with high country food consumption frequency, such that their concentrations increased by approximately 67–83 % compared to those reporting no or very little consumption of any country foods.

Conclusions: Increased country food consumption was strongly associated with higher PFAAs concentrations, particularly PFOS, PFDA, and PFUnDA. The results provide further evidence that the quality of country foods is being threatened by PFAAs contamination. Additional national and international regulations are required to protect the Arctic and its inhabitants from these pollutants.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are synthetic chemicals used for the water and oil resistant properties in various products, including textile stain repellents, grease-proof food contact-paper, and fire-fighting aqueous film–forming foams (AFFF) (Buck et al., 2011). PFAS are detected in the environment from industrial release and use and disposal of consumer products with PFAS. Some PFAS are precursors to the more persistent perfluoroalkyl acids (PFAAs). An example of PFAA-precursors are fluorotelomer alcohols (FTOH) (Buck et al., 2011; Dasu et al., 2022). Dubbed “forever chemicals”, PFAAs exhibit environmental persistence and some demonstrate bioaccumulation and biomagnification properties, deeming them important environmental and public health hazards (Brase et al., 2021; Dasu et al., 2022; Sunderland et al., 2019). These properties led to their national and international regulation, with perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) as well as their precursors heavily restricted under the Stockholm Convention, and perfluorohexane sulfonic acid (PFHxS) still under review for listing (Secretariat of the Stockholm Convention, 2019). Furthermore, certain long-chain PFAS compounds, namely perfluoroalkyl carboxylic acids (PFCA) with nine or more carbons (C9+) and including perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA) and perfluoroundecanoic acid (PFUnDA), and their precursors recently passed the Annex D requirements under the Stockholm Convention and were moved to the next stage for potential inclusion in the list (Health Canada, 2021; Secretariat of the Stockholm Convention, 2019).

PFAAs, which include PFOS, PFOA, PFHxS, and other long-chain PFCAs are detected in high concentrations in environmental samples throughout Arctic regions via long-range atmospheric and oceanic transport of precursors and PFAA from southern latitudes to northern latitudes (AMAP, 2021a). They further accumulate in Arctic marine and terrestrial food webs, and have been measured in many wildlife species, including polar bears, beluga whale, caribou, fish, and Arctic birds (AMAP Assessment 2016: Chemicals of Emerging Arctic Concern, 2017; Muir et al., 2019). The bioaccumulation potential of PFAAs is greater for PFOS and C9+ long-chain PFAAs (Buck et al., 2011; Eriksson et al., 2016; Haukås et al., 2007; Müller et al., 2011; Xu et al., 2014), making them particularly important in the Arctic context.

While overall PFAAs exposure is still a major concern, PFOA and PFOS concentrations in the Arctic have been decreasing over the last couple of decades, largely due to national and international regulations, such as the Stockholm Convention and industrial manufacturing shifts. However, evidence suggests an increase in human, animal and abiotic concentrations of C9+ long-chain PFAAs congeners over the last decade (Aker et al., 2021; Caron-Beaudoin et al., 2020; Muir et al., 2019).

Exposure to PFAAs has been associated with several health outcomes, including immunological outcomes (decreased vaccine response, increased respiratory and gastrointestinal infections, and immune hyperactive diseases like asthma and allergies (Fenton et al., 2020; Sinclair et al., 2020; Sunderland et al., 2019)), cardiometabolic outcomes (dyslipidemia, hypertension, glucose metabolism dysregulation, and diabetes (Canova et al., 2021; Gaston et al., 2020; Goodrich et al., 2021; Margolis and Sant, 2021; Meneguzzi et al., 2021; Sunderland et al., 2019)), and thyroid and reproductive endocrine disruption (Kim et al., 2018; Mokra, 2021; Wang et al., 2014; Yang et al., 2022). An understanding of potential exposure sources could help mitigate the risks associated with PFAAs exposure. While food is a major exposure source to PFAAs worldwide via food processing and packaging (Haug et al., 2010; Schaider et al., 2017; Susmann et al., 2019), previous studies suggest that among Inuit communities in the Arctic, country foods - wild foods hunted and harvested from the land, rivers and sea - may be more important PFAAs exposure sources (Aker et al., 2021; Byrne et al., 2017; Caron-Beaudoin et al., 2020; Long et al., 2012; Ostertag et al., 2009). However, few studies specifically examined the association between country food consumption and PFAAs biomarker concentrations (Caron-Beaudoin et al., 2020; Dallaire et al., 2009; Hu et al., 2018). Country foods are integral to Inuit culture, subsistence, food security, and nutrition, providing nutrients such as omega-3 polyunsaturated fatty acids, iron, selenium, vitamins D, B6 and B12, and proteins (Kenny et al., 2018; Lemire et al., 2015; Little et al., 2021). As such, the protection of country foods quality is paramount to maintaining the integrity of Inuit culture and health. Further studies documenting the contamination of country foods and their impact on Inuit exposure and health is essential to drive additional international actions and regulations.

The *Qanuilirpitaa?* 2017 Nunavik Inuit Health Survey (Q2017) is a population health survey conducted in the 14 villages of Nunavik, northern Quebec, Canada to assess the health status of Nunavimmiut (Inuit living in Nunavik) aged 16 and over. The survey included a food frequency questionnaire to identify country and market (i.e. store-bought) foods in the region, and measured plasma PFAAs concentrations in the large majority of the participants. Distinct dietary food profiles were identified in the Q2017 dataset in a previous study (Aker et al., in press). These profiles account for the correlations between individual food items or groups and provide an opportunity to study the associations between overall dietary patterns in a region and chemical exposure. In turn, the objective of the current study is to examine the associations between the dietary food profiles and exposure to PFAAs in Nunavik, and to disentangle the impact of country food versus market foods on PFAAs exposure.

2. Materials and methods

2.1. Study population

The study utilized sociodemographic, food frequency, and chemical biomarker data from the Q2017 survey. Details on the Q2017 survey have been described elsewhere (Hamel et al., 2020). Briefly, the survey implemented a stratified proportional model for respondent selection and targeted Nunavik permanent residents aged 16 years and older. People living in institutional settings or suffering from tuberculosis were excluded, and participants had to be able to come aboard the Amundsen, a Canadian Coast Guard Icebreaker, for the interviews and clinical tests. Stratification was based on age category (16–29, 30–49, and 50+ years) and communities (across three regions). A total of 1326 individuals participated in the survey aboard the Amundsen from August 19, 2017 to October 5, 2017 from 14 villages settled along the Ungava Bay, the Hudson Strait and the Hudson Bay (Fig. 1). Data was collected using questionnaires, clinical measurements, and human biological sampling of urine and blood.

Ethical approval was received from the Comité d'éthique de la recherche du Centre Hospitalier Universitaire de Québec - Université Laval (no. 2016–2499). The survey was conducted in close collaboration with several organisations from Nunavik (including but not limited to the Nunavik Regional Board of Health and Social Services, Kativik Regional Government, and Makivik Corporation) and governed by the OCAP® principles (Ownership, Control, Access, and Possession). The Data Management Committee (DMC), which was heavily represented by Inuit colleagues and

Fig. 1. Map of Nunavik.

partners, were integral to the discussion and interpretation of the Q2017 survey results.

2.2. Dietary profiles

Study participants were asked to complete a food frequency questionnaire (FFQ) to measure the consumption frequency of a range of market and country foods in the previous three months. Food items were rated on a scale of 1 (item consumed never or less than once a month) to 5 (item consumed daily).

Dietary profiles were assessed using latent profile analysis (LPA). Details on this analysis have been described in more detail previously (Aker et al., in press). Briefly, two types of dietary profiles were created. The first included all food items consumed, including both market and country food items and/or groups. The second dietary profile included only country food items to better understand what types of country food consumption profiles exist in the population. These models produced the following the profiles (Supplementary Figs. 1 & 2): Overall food profiles (market and country): 1) Market food dominant, 2) Country food dominant, 3) Diverse consumption, and 4) Low consumption; Country food profiles: 1) High consumption, 2) Moderate consumption, 3) Low consumption, and 4) No (/None) consumption.

The overall market and country food profiles were defined as follows. The *Market Food Dominant* profile included higher consumption frequency of fruits and vegetables, dairy products and refined grains; the *Country Food Dominant* profile included higher consumption frequency of marine mammals, fish and seafood, canned fish, legumes/nuts, processed meats, and non-nutrient dense foods (snacks and sugary foods); the *Diverse Consumption* profile included higher consumption frequency of caribou meat, *pitsik* (air dried fish, most often made of Arctic char), Arctic char, some beluga *mattaaq* (beluga skin and fat layer), fruits and vegetables, red meat, poultry, processed meats, dairy products, and non-nutrient dense foods; the *Low Consumption* profile had low consumption of all foods compared to other profiles but reported higher consumption frequency of marine mammal meats and organs, mollusks, wild birds and their eggs, canned fish, processed meats, and carbonated beverages.

The country food profiles were defined as follows. The *High* consumption profile included higher consumption frequency of marine mammal meats and organs (particularly seal meat and liver), wild birds and their eggs, and various fish and seafood items (Arctic char, lake trout, mollusks, seaweed); the *Moderate* consumption profile was defined by higher consumption frequency of caribou, *pitsik* (dried fish), Arctic char, wild berries and *suuvalik* (dish with fish roe, seal blubber or Crisco oil, and wild berries), and some consumption of beluga *nikku* (dried beluga meat) and beluga *mattaaq* (beluga skin and fat layer); the *Low* consumption profile had low consumption of all country foods compared to other profiles, and slightly higher consumption of beluga *misirak* (rendered beluga blubber); the *None* consumption profile reported no or very little consumption of any country foods.

2.3. Biomarker analysis

Research nurses collected blood samples were obtained through venipuncture and collected in K2-EDTA vacutainers. All samples were processed within 90 min on board the Amundsen. Blood samples were centrifuged at 2000 $\times g$ for 10 min and the plasma was transferred into a 2-mL polypropylene tube for storage at −20 °C until time of analysis.

Nine PFAAs congeners were quantified in plasma samples at the Centre de toxicologie du Québec (CTQ) of the Institut national de santé publique du Québec (INSPQ) (accredited by the Canadian Association for Environmental Analytical Laboratories and ISO 17025). PFAAs congeners included perfluoro-n-butanoic acid (PFBA), perfluoron-hexanoic acid (PFHxA), PFOA, PFNA, PFDA, PFUnDA, perfluorobutane sulfonic acid (PFBS), PFHxS and PFOS. Compounds were extracted using a weak anion exchange solid-phase extraction on a 96-well plate after acidifying the samples. The extracts were evaporated, dissolved into the mobile phase, and then analyzed using ultra performance liquid chromatography (Waters Acquity) coupled with tandem mass spectrometry (Waters Xevo TQ-S) with electrospray negative ionisation in multiple reaction monitoring mode. More details on the laboratory analysis can be found in a previously published article (Caron-Beaudoin et al., 2020).

Internal reference materials were used to control the quality of the analyses using the certified reference material SRM-1958 from the National Institute of Standards and Technology (NIST; Gaithersburg, MD) and in-house quality controls. The limits of detection (LOD) for the PFAAs congeners were 0.06 μg/L for PFHxS, 0.07 μg/L for PFOA and PFBS, 0.08 μg/L for PFBA and PFHxA, 0.09 μg/L for PFDA, 0.10 μg/L for PFNA and PFUnDA, and 0.40 μg/L for PFOS. Any biomarker concentrations detected at or below the LOD were replaced with the respective LOD/2.

2.4. Statistical analysis

Population descriptive analysis was first conducted and compared by overall and country food only profiles. PFAAs distributions were also calculated. All PFAAs concentrations were natural log transformed for further analysis to approach normality.

Multiple linear regression models were constructed to study associations between the two sets of dietary profiles and each of the PFAAs biomarker concentrations. The *Market Food Dominant* profile was the reference category for models with overall dietary profiles, and the *None* country food consumption profile was the reference category for models with country food profiles. Beta estimates were exponentiated to represent the percent change in PFAAs congener for the respective dietary profile compared to the reference category. The least square means of the PFAAs congeners and their associated 95 % confidence intervals (CI) for each dietary profile were also calculated. All models were adjusted for age, age squared (due to the non-linear association of age and certain PFAAs congeners (Aker et al., 2022)), sex, marital status, and education status (Supplementary Fig. 3). Only PFAAs congeners detected in at least 70 % of the population were included in models with dietary profiles. Waist circumference was further included in models in a sensitivity analysis. Waist circumference was categorized into quartiles by sex. We also replaced the age-squared covariate by an interaction term between age and sex to account for differences in PFAAs excretion in females via menstruation, pregnancy, and lactation (Macheka-Tendenguwo et al., 2018; Rickard et al., 2022; Upson et al., 2022).

All models applied survey weights calculated using sex, age, and ecological region distribution of the Nunavik population in 2017 to produce more representative population-level estimates (Hamel et al., 2020). The alpha level was set at 0.05. Models were conducted using SAS 9.4 (SAS Institute Inc., Cary, NC).

3. Results

A total of 1172 participants were included in the study (Table 1). Approximately 41.8 % of the participants were aged 16–29 years, whereas 24.1 % were aged 50–82 years, after weighting. Almost half the participants had household incomes less than $20,000 and the majority had up to a high school education (i.e. attended some or completed their high school education). Up to 14.3 % reported not eating for a whole day in the past three months due to a lack of resources. The distributions by covariates varied across the overall dietary profiles. Namely, males were more likely to have a *Country Food Dominant* dietary profile versus women (72.6 % versus 27.4 %). Individuals aged 16–29 and 50–82 years were also most likely to have a *Country Food Dominant* profile (51.3 % and 28.0 %, respectively), whereas individuals aged 30–49 years were more likely to have a *Market Food Dominant* (39.6 %). Participants in *Market Food Dominant* group were also more likely to have an income >$60,000 (16.2 %) and at least some college education (18.9 %). Conversely, participants in the *Country Food Dominant* group were also least likely to have waist circumferences in the third (10.9 %) or fourth (20.4 %) quartiles compared to all other profiles. Participants with a *Low Consumption* profile were most likely to report not eating for a whole day due to a lack of resources (20.5 %). The PFAAs congeners with the highest concentrations included PFOS (GM 4.90 μg/L), PFNA (GM 3.70 μg/L), and PFOA (1.02 μg/L) (Table 2). Shorter chain PFAAs congeners, PFBA, PFHxA, and PFBS, were largely undetected, and were not included in further analyses.

3.1. Overall food profiles

Individuals with a *Country Food Dominant* dietary profile had the highest concentrations of all detected PFAAs congeners, and these were statistically different compared to individuals with a *Market Food Dominant* profile (Table 3). In particular, individuals with a *Country Food Dominant* profile had considerably higher concentrations of PFNA [percent change in concentration (%Δ) 43.91, 95 % CI 20.39–72.02], PFDA (%Δ 49.53, 95 % CI 20.49–85.56), PFUnDA (%Δ 57.55, 95 % CI 27.35–94.90), and PFOS (%Δ 50.41, 95 % CI 21.65–85.97), compared to participants in the *Market Food Dominant* group. While participants with a *Diverse Consumption* dietary profile had higher concentrations of all PFAAs congeners compared to *Market Food Dominant*, only the association with PFOA was statistically significant (%Δ 10.69, 95 % CI 3.13–18.81). Participants with a *Low Consumption* profile also had higher concentrations of all PFAAs congeners compared to *Market Food Dominant* profiles, particularly PFDA (%Δ 21.81, 95 % CI 7.42–38.13) and PFUnDA (%Δ 27.04, 95 % CI 10.71–45.79).

3.2. Country food profiles

Country food dietary profiles *Low*, *Moderate*, and *High* had higher concentrations of all PFAAs congeners compared to the *None* country food dietary profile (Table 4). While differences in the percent change in chemical concentration were similar between *Low* and *Moderate* profiles (between %Δ 15.41 and 51.38), individuals with a *High* country food profile had much higher concentrations of PFAAs congeners compared to those in the *None* country food profile ((PFOA: %Δ 26.48, 95 % CI 10.14–45.24; PFNA: %Δ 49.53, 95 % CI 21.85–83.49; PFDA: %Δ 71.38, 95 % CI 33.56–119.93; PFUnDA: %Δ 82.98, 95 % CI 43.84–132.77; PFHxS: %Δ 45.98, 95 % CI 24.16–71.65); PFOS: %Δ 66.82, 95 % CI 31.03–112.39). Figs. 2 and 3 display the least square means derived from the adjusted regression models across the four categories.

There were no meaningful differences after including waist circumference as a covariate in the models (Supplementary Tables 3 and 4). The percent change in PFOA and PFNA were slightly increased by 1–4 % after including an interaction term between age and sex; however, no other models showed meaningful differences (Supplementary Table 5).

4. Discussion

Our study identified strong associations between having a dietary profile consisting of frequent country food consumption and higher levels of PFOA, PFNA, PFDA, PFUnDA, PFHxS, and PFOS, compared to people with a dietary profile of frequent market food consumption. After restricting the dietary profiles to country foods only, the models revealed stronger associations with increased frequency of country food consumption and PFAAs concentrations. PFDA, PFUnDA, and PFOS were particularly associated with increased country food consumption frequency, such that their concentrations increased by approximately 67–83 % in people

Table 1
Study population description for the 1172 participants of the Q2017 survey in Nunavik.

Characteristic	Total population (%)	Market food dominant (%)	Diverse consumption (%)	Country food dominant (%)	Low consumption (%)
Sex					
Female	49.6	53.2	52.9	27.4	51.7
Male	50.4	46.8	47.1	72.6	48.3
Age					
16–29 years	41.8	37.1	48.3	51.3	40.5
30–49 years	34.1	39.6	30.5	20.6	35.2
≥ 50 years	24.1	23.3	23.2	28.0	24.3
Region					
Hudson Strait	23.8	20.7	25.2	27.9	26.0
Hudson Bay	42.6	41.2	36.2	51.1	47.1
Ungava Bay	33.5	38.1	38.6	21.0	22.6
Marital status					
Married or living with partner	53.1	55.1	52.3	57.0	48.2
Single	46.8	44.8	47.7	43.0	51.8
Missing	0.04	0.1	–	–	–
Income					
<$20,000	46.6	39.4	50.1	55.8	50.8
$20,000–$59,999	29.0	32.3	30.2	23.2	24.8
>$60,000	11.4	16.2	9.4	7.8	6.6
Missing	13.1	12.1	10.2	13.2	17.7
Education					
<Grade 9	36.7	28.8	39.8	49.0	41.2
At least some high school	48.9	51.0	49.2	42.0	48.6
At least some college	12.6	18.9	10.3	7.3	6.6
Missing	1.7	1.3	0.7	1.6	3.6
Waist circumference[a]					
Q1	24.5	20.7	23.6	40.8	23.1
Q2	23.8	21.0	22.5	25.8	29.2
Q3	23.3	26.1	27.1	10.9	21.3
Q4	25.5	29.0	24.6	20.4	22.8
Missing	2.9	3.2	2.1	2.1	3.6
Food security					
Did not eat for a whole day due to lack resources	14.3	13.0	11.2	13.3	20.5
Missing	0.6	0.7	–	1.1	0.8

Analyses used weights to account for sampling methodology and item non-response and thereby allow the results to be inferred to the target population; variance was estimated with the balanced repeated replication method.

Percentage of the total population: Market Food Dominant 41.3 %; Diverse Consumption 23.4 %; Country Food Dominant 12.6 %; Low Consumption 22.6 %.

[a] Waist circumference quartiles based on sex. Females: $Q1 < 82$, $82 \leq Q2 < 94$, $94 \leq Q3 < 105$, $Q4 \geq 105$; Males: $Q1 < 79$, $79.5 \leq Q2 < 86.75$, $86.75 \leq Q3 < 103.5$, $Q4 \geq 103.5$.

with *High* country food consumption compared to people in the *None* country dietary profile. These results provide further evidence of country foods being a major exposure source of PFAAs in Nunavik, despite the lack of any identified local sites with direct use of PFAAs in the region. Thus, further national and international efforts must be enforced to protect the integrity of country foods from PFAAs contamination.

Exposure to PFOS in Nunavik decreased by 72 % compared to the first time they were measured in the Nunavik Inuit Health Survey in 2004; however PFOS concentrations remain 50 % more elevated compared to the Canadian general population (based on data from the Canadian Health Measures Survey 2016–2017; GM 3.3 μg/L) (Aker et al., 2021). Concentrations of other PFAAs congeners in this study are also markedly higher than other populations. PFNA, PFDA, PFUnDA, and PFOS are higher in Nunavik youth and adults (GM μg/L: PFNA 3.70; PFDA 0.66; PFUnDA 0.68; PFOS 4.90) versus the Canadian Health Measures Survey 2016–2017 (GM μg/L for 18+ years: PFNA 0.50; PFDA 0.20; PFUnDA <LOD; PFOS 3.30) (Health Canada, 2019) and the U.S. National Health and Nutrition Examination Survey (NHANES) 2017–2018 (GM μg/L for 20+ years: PFNA 0.42; PFDA 0.20; PFUnDA 0.13; PFOS 4.50) (Centers for Disease Control and Prevention (CDC), 2021). A previous study in Nunavik pregnant

Table 2
Distribution and concentration of PFAA congeners in the study population.

	N	% < LOD	GM (95 % CI) μg/L	25th μg/L	50th μg/L	75th μg/L	95th μg/L
PFBA	1168	77.6	<LOD	<LOD	<LOD	<LOD	0.14
PFHxA	1166	99.9	<LOD	<LOD	<LOD	<LOD	<LOD
PFOA	1172	0.1	1.02 (0.99–1.05)	0.73	1.01	1.39	2.19
PFNA	1172	0.1	3.70 (3.55–3.86)	2.30	3.63	5.79	10.95
PFDA	1164	0.9	0.66 (0.64–0.70)	0.38	0.65	1.12	2.47
PFUnDA	1172	1.4	0.68 (0.65–0.71)	0.40	0.71	1.15	2.54
PFBS	1149	99.8	<LOD	<LOD	<LOD	<LOD	<LOD
PFHxS	1172	0.1	0.62 (0.60–0.64)	0.38	0.60	0.97	2.33
PFOS	1172	0.1	4.90 (4.65–5.09)	2.75	4.58	8.29	20.07

GM: Geometric mean; 95 % CI: 95 % confidence intervals; LOD: limit of detection.

Table 3
Associations between PFAAs congeners and overall dietary profiles.

	Market food dominant	Country food dominant	Diverse consumption	Low consumption
PFOA				
%Δ in concentration (95 % CI)	Ref.	24.54 (10.62–40.19)*	10.69 (3.13–18.81)*	12.74 (5.48–20.51)*
p-Value		0.003	0.005	<0.001
PFNA				
%Δ in concentration (95 % CI)	Ref.	43.91 (20.39–72.02)*	7.18 (−2.89–18.30)	14.21 (2.83–26.85)*
p-Value		<0.0001	0.17	0.01
PFDA				
%Δ in concentration (95 % CI)	Ref.	49.53 (20.49–85.56)*	11.46 (−1.18–25.71)	21.81 (7.42–38.13)*
p-Value		0.003	0.08	0.002
PFUnDA				
%Δ in concentration (95 % CI)	Ref.	57.55 (27.35–94.90)*	11.40 (−2.10–26.77)	27.04 (10.71–45.79)*
p-Value		<0.0001	0.10	0.001
PFHxS				
%Δ in concentration (95 % CI)	Ref.	32.97 (15.33–53.32)*	8.70 (−1.16–19.53)	12.07 (1.92–23.24)*
p-Value		<0.0001	0.09	0.02
PFOS				
%Δ in concentration (95 % CI)	Ref.	50.41 (21.65–85.97)*	11.46 (−0.43–24.77)	18.91 (4.91–34.79)*
p-Value		0.0002	0.06	0.007

N = 1146 for all PFAAs congeners, except PFDA where N = 1139.
CI: confidence intervals.
%Δ in concentration: Exponentiated beta estimate representing the change in chemical biomarker concentration comparing each respective dietary profile to *Market Food Dominant.*
All models adjusted for age, age squared, sex, marital status, and education status.
Percentage of the total population: Market Food Dominant 41.3 %; Diverse Consumption 23.4 %; Country Food Dominant 12.6 %; Low Consumption 22.6 %.
* p value < 0.05.

women entitled *Nutaratsaliit qanuingisiarningit niqituinnanut* (NQN, i.e. Pregnancy wellness with country foods) and conducted in 2016–2017 identified similar trends (Caron-Beaudoin et al., 2020). Long-chain PFAAs with 9+ carbons (PFNA in particular) and PFOS are similarly elevated in other populations in Arctic regions, such as Alaska and Greenland (AMAP, 2021b; Byrne et al., 2017), and PFNA was also elevated in Indigenous communities in the Northwestern Territories, Canada (Garcia-Barrios et al., 2021). This PFAAs exposure signature points to the unique exposure sources that may exist in northern latitudes versus other population in southern latitudes. This is likely due to the bioaccumulative properties of these specific congeners in Arctic wildlife and the increased usage of FTOHs in products that get transported to the Arctic and degrade into PFOA and long-chain PFAAs congeners (Eriksson et al., 2016; Haukås et al., 2007; Muir et al., 2019; Müller et al., 2011).

Other studies also observed associations between PFAAs and seafood consumption. PFOA, PFNA, PFDA, PFUnDA and PFOS (among other PFAAs congeners and their precursors) concentrations were higher in men who participated in pilot whale harvesting compared to women and

Table 4
Associations between PFAAs congeners and country food profiles.

	None	Low	Moderate	High
PFOA				
%Δ in concentration (95 % CI)	Ref.	15.41 (8.07–23.24)*	19.82 (11.06–29.27)*	26.48 (10.14–45.24)*
p-Value		<0.0001	<0.0001	0.009
PFNA				
%Δ in concentration (95 % CI)	Ref.	23.78 (11.82–37.02)*	27.58 (13.32–43.63)*	49.53 (21.85–83.49)*
p-Value		<0.0001	<0.0001	0.0001
PFDA				
%Δ in concentration (95 % CI)	Ref.	49.63 (32.53–68.93)*	47.23 (25.95–72.11)*	71.38 (33.56–119.93)*
p-Value		<0.0001	<0.0001	<0.0001
PFUnDA				
%Δ in concentration (95 % CI)	Ref.	51.38 (32.92–72.41)*	46.98 (24.45–73.58)*	82.98 (43.84–132.77)*
p-Value		<0.0001	<0.0001	<0.0001
PFHxS				
%Δ in concentration (95 % CI)	Ref.	30.63 (19.38–42.93)*	29.26 (15.19–45.06)*	45.98 (24.16–71.65)*
p-Value		<0.0001	<0.0001	<0.0001
PFOS				
%Δ in concentration (95 % CI)	Ref.	41.72 (27.18–57.93)*	39.03 (19.53–61.71)*	66.82 (31.03–112.39)*
p-Value		<0.0001	<0.0001	<0.0001

N = 1146 for all PFAAs congeners, except PFDA where N = 1139.
CI: confidence intervals.
%Δ in concentration: Exponentiated beta estimate representing the change in chemical biomarker concentration comparing each respective dietary profile to *None*.
All models adjusted for age, age squared, sex, marital status, and education status.
Percentage of the total population: None 31.4 %; Low 42.1 %; Moderate 15.7 %; High 10.8 %.
* p value < 0.05.

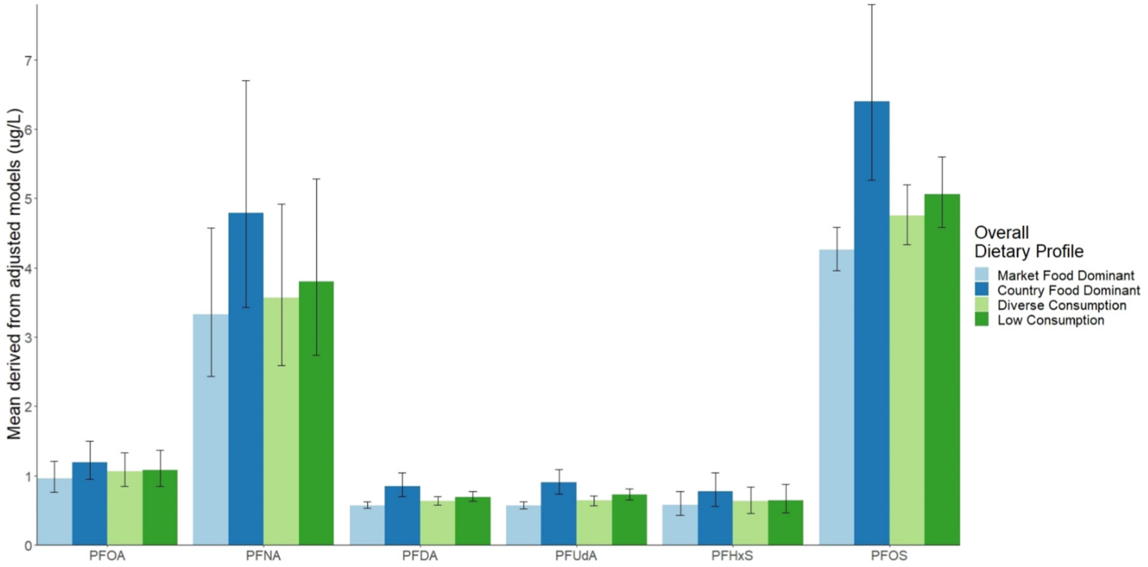

Fig. 2. Least square mean PFAA plasma concentrations derived from adjusted regressions for each PFAA by overall dietary profiles.

children in the Faroe Islands (Hu et al., 2018), and the study also pointed to a key exposure profile of long-chain PFAAs with carbon chain lengths 9–12 as effective tracers of seafood consumption (including marine mammals) in northern latitudes. Similarly, PFOS, PFNA, PFDA, PFUnDA, PFHxS, and PFOA (among other PFAAs congeners) were associated with fatty fish consumption in the Arctic Tromsø and Balsfjord municipalities in northern Norway (Averina et al., 2018). This is further evidenced by the fact that seafood was estimated to account for 93 % of PFUnDA and 81 % of PFOS daily intakes in Norway (Haug et al., 2010). Moreover, the NQN study with pregnant women in Nunavik identified a strong association between the omega-3/omega-6 polyunsaturated fatty acids ratio (indicative of marine mammal and seafood consumption) and PFNA, PFDA, PFUnDA, PFHxS, and PFOS (Caron-Beaudoin et al., 2020).

More specifically, evidence suggests that marine mammal consumption may be an important exposure source compared to other seafood items. Although the sample size was small ($n = 97$), the NQN study identified associations between marine mammal meat and organ consumption and PFNA, PFDA, PFUnDA, PFHxS, and PFOS among pregnant women in Nunavik (Caron-Beaudoin et al., 2020). The *High* country food dietary profile in the present study involving Inuit youth and adults was in part defined by frequent consumption of beluga meat, seal meat, and seal liver, all of which have been found to have high concentrations of PFNA, PFDA, PFUnDA and PFOS in a study in Nunavut that measured PFAAs concentrations in country food samples (Ostertag et al., 2009). In contrast to other persistent chemicals, PFAAs are not lipophilic and instead bind to proteins, leading to higher concentrations in the blood and liver (Kelly et al., 2009; Muir et al., 2019; Roth et al., 2020). Although the percent change in PFAAs concentrations in the *Moderate* country food dietary profile were slightly higher than those in the *Low* country food profile, the percent changes were quite similar in the *Moderate* and *Low* country food profiles. The *Moderate* country food dietary profile was defined by higher frequency consumption of caribou over marine mammals and caribou meat does not appear to be an important source of PFAAs exposure (Caron-Beaudoin et al., 2020; Ostertag et al., 2009), but this may be highly variable depending on the caribou herd and region in Nunavik (Committee on the Status of Endangered Wildlife in Canada (COSEWIC), 2018; Roos et al., 2021). For example, reindeer consumption in Norway was associated with higher dietary exposure to PFNA and PFUnDA (Averina et al., 2018). The *Diverse Consumption* overall dietary profile was also defined by more caribou consumption over marine mammals, and membership in this group was not

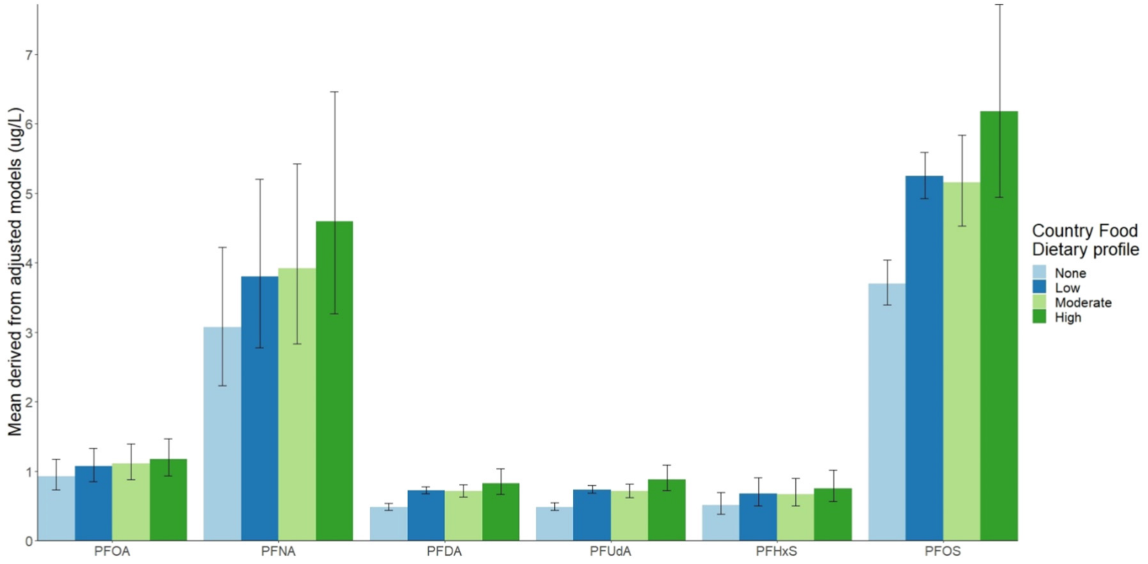

Fig. 3. Least square mean concentrations of PFAA in plasma derived from adjusted regressions for each PFAA by country food dietary profiles.

associated with higher PFAAs concentrations compared to those in the *Market Food Dominant* dietary profile. This adds further evidence to the role of different marine mammal parts as a more important exposure source, likely due to their position as apex animals in Arctic food webs. Country foods appear not to be as important of an exposure source for PFOA in the current study, nor in the Nunavik pregnant women study (Caron-Beaudoin et al., 2020) and Nunavut study (Ostertag et al., 2009). PFOA can be found from various exposure sources such as consumer goods and may not serve as effective tracers for specific exposure sources (Hu et al., 2018). Additionally, associations between PFNA and dietary profiles were not as strong as other long-chain PFAAs congeners, despite the elevated PFNA concentrations in Nunavik. Additional studies are required to further understand PFNA exposure sources.

Nunavimmiut in the *Low Consumption* overall dietary profile also had higher levels of all six PFAAs congeners compared to those in the *Market Food Dominant* profile, particularly long-chain PFAAs and PFOS pointing to their key exposure profile associated with seafood and marine mammal consumption. Although this group reported infrequent consumption of country foods compared to other dietary profiles, their diet was composed of more frequent country food consumption over market foods. Moreover, their country food consumption included higher consumption frequency of marine mammals, wild birds and their eggs. Seal liver, in particular, was one of the most frequently consumed country food items in Nunavimmiut with a *Low Consumption* profile, and PFAAs tend to accumulate in the liver due to PFAAs' binding to proteins (Forsthuber et al., 2020; Li et al., 2021; Pérez et al., 2013). PFAAs have also been detected in Arctic seabirds and their eggs (Braune and Letcher, 2013; Hitchcock et al., 2019; Jouanneau et al., 2021). Furthermore, the *Low Consumption* group was characterized by frequent consumption of canned fish, processed meats, and popcorn, which have all been previously identified as potential sources of PFAAs among store-bought foods (Averina et al., 2018; Callan et al., 2016; Eick et al., 2021; Hlouskova et al., 2013; Ramírez Carnero et al., 2021; Sunderland et al., 2019).

Interestingly, while Nunavimmiut with frequent country food consumption had higher PFAAs concentrations compared to those with lower or no country food consumption, participants in the *Market Food Dominant* overall dietary profile and *None* country food dietary profile still had elevated concentrations of C9+ long-chain PFAAs compared to the general populations in Canada (GM μg/L (95 % CI): PFNA 0.52 (0.47–0.58), PFDA 0.19 (0.17–0.21), and PFUnDA <LOD in the Canadian Health Measures Survey 2017) (Aker et al., 2022; Health Canada, 2019) and the US (GM μg/L (95 % CI): PFNA 0.69 (0.63–0.75), PFDA 0.19 (0.17–0.22), and PFUnDA <LOD in the National Health and Nutrition Examination Survey 2013–2014) (Centers for Disease Control and Prevention (CDC), 2021). PFNA and PFUnDA were still approximately six-fold higher in the *Market Food Dominant* compared to the general US and Canadian populations. This could point to the overall high environmental contamination of the region from long-range transfer of persistent contaminants and the unique Arctic conditions creating a sink for these chemicals (AMAP Assessment 2016: Chemicals of Emerging Arctic Concern, 2017; Muir et al., 2019; Müller et al., 2011). Other sources of exposure might also be contributing to higher levels of PFAAs in Nunavik, such as indoor dust (Björklund et al., 2009; Byrne et al., 2017; Harrad et al., 2019; Knobeloch et al., 2012; Winkens et al., 2018). PFOS and PFOA were the most detected congeners in household dust in an Alaskan study (Byrne et al., 2017), and several PFAAs congeners in dust (with the exception of PFNA) were associated with FTOH levels in air (Winkens et al., 2018). Country foods are frequently stored in and eaten on cardboard which may also explain some of the elevated concentrations (Zweigle et al., 2021). Houses with plastic floor materials (i.e. PVC or vinyl) were reported to have higher concentrations of PFOS, but this finding was not consistent with other congeners (Winkens et al., 2018). Additionally, some market foods are associated with elevated PFAAs concentrations, largely via migration of PFAAs from food packaging subjected to high temperatures (like in the case of microwave popcorn) or after prolonged contact of the food in the presence of emulsifiers, alcohol, or fatty foods (Begley et al., 2008; Ramírez Carnero et al., 2021; Sungur et al., 2014). For example, food contact products such as pizza boxes and fast food paper packaging use PFAAs as oil-repellant surfactants, and frequent consumption of pizza and fast food has been associated with higher concentrations of PFOA and PFNA (Averina et al., 2018; Sunderland et al., 2019; Susmann et al., 2019). Frozen ultra-processed foods are common in Nunavik (Nunavik Regional Board of Health and Social Services, personal communication, February 1, 2022), and exposure to PFAAs exposure could occur through contamination of the foods from PFAAs in the food packaging. This accumulating evidence supports the importance of food-related products as major exposure sources of PFAAs biomarker concentrations.

A strength of our study was the large representative sample size, our wide range of PFAAs congeners, and our inclusion of dietary profiles. Use of dietary profiles allowed us to better understand key population groups with higher PFAAs concentrations in Nunavik. Future studies will assess specific food items of concern while accounting for correlation between the food items. However, there were some limitations to our study. The food frequency questionnaire only provided data on frequency and not the quantity consumed, and not all foods were included in the questionnaire. Furthermore, the consumption of country foods is highly variable depending on the season and the year of the survey. An analysis of PFAAs concentrations in country foods collected throughout Nunavik will be conducted to address some of these limitations. While the present survey asked respondents on their consumption patterns in the three months prior (which should take into account the main hunting and harvesting season of the year), bias may still have been introduced. The survey also provided a cross-sectional view of the relationship between dietary factors and PFAAs concentrations Additional repeated measures of exposure may not have influenced the results much given the long plasma half-lives of PFAAs in humans; however, dietary patterns may have shifted over the years. There may have also been recall bias or respondent bias due to the length of the questionnaire.

5. Conclusions

As compared to frequent consumption of market foods, our study identified a strong association between frequent consumption of country foods and PFAAs biomarker concentrations, particularly PFOS and long-chain PFAAs congeners, PFNA, PFDA, and PFUnDA. The associations were stronger with increasing country food consumption and appeared to be driven by marine mammal consumption. The results of this study add further evidence that the quality of country foods in Arctic communities is threatened by chemical contamination and necessitate further international and national regulation to protect the exceptional quality of country foods.

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CRediT authorship contribution statement

Amira Aker: Conceptualization, Formal analysis, Methodology, Writing – original draft. **Pierre Ayotte:** Conceptualization, Validation, Data curation, Funding acquisition, Supervision, Writing – review &

editing. **Élyse Caron-Beaudoin:** Validation, Writing – review & editing. **Amila De Silva:** Validation, Writing – review & editing. **Sylvie Ricard:** Writing – review & editing. **Mélanie Lemire:** Conceptualization, Validation, Funding acquisition, Supervision, Writing – review & editing.

Data availability

The authors do not have permission to share data.

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.

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Appendix A. Supplementary data

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