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Current-use pesticide exposures in remote Inuit communities

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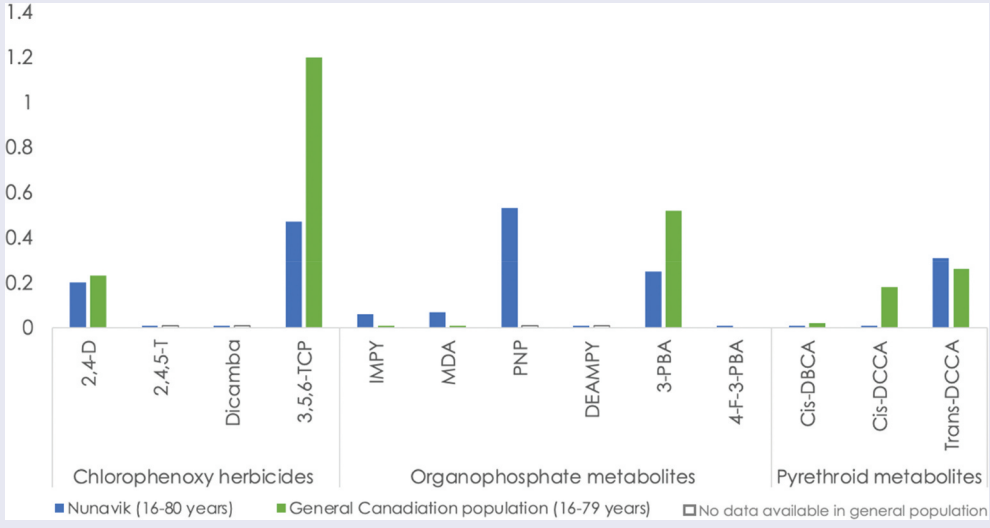
ABSTRACT

The global use of pesticides is increasing; however, few studies have examined the exposure of current-use pesticide exposure in Inuit populations. Some current use pesticides are also capable of long-range transport, potentially increasing exposures to northern populations. The study aim was to analyse pesticide (chlorophenoxy, organophosphates, and pyrethroid pesticide) biomarker levels in pooled samples from an Inuit population in Nunavik, Quebec. Thirty pooled samples from the Qanuilirpitaa? 2017 survey (Q2017) from individuals aged 16–80 years were included. Creatinine-adjusted arithmetic (AM) were compared by sex, age, and region sub-groups, and geometric mean concentrations (GM) were compared to those in the Canadian Health Measures Survey (CHMS). Most analysed pesticide biomarkers were detected, and PNP (a metabolite of methyl and ethyl parathion), trans-DCCA (a metabolite of pyrethroids), and 3,5,6-TCP (a metabolite of chlorpyrifos) had the highest concentrations. Concentrations in Q2017 were largely similar to or less than CHMS concentrations. Although not significant, there was a general increase in 2,4-D (a chlorophenoxy biomarker), 3,5,6-TCP, 3-PBA (a metabolite of pyrethroids), and trans-DCCA with increasing age. Concentrations were also somewhat higher in females versus males, but these were not significant. Environmental exposures to current use pesticides were detected in Nunavik and concentrations were similar to or less than those in the general Canadian population. Regular monitoring of current use pesticide exposures is recommended given the increasing global use of pesticides.



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Introduction

Northern populations were exposed to highly elevated concentrations of organochlorine pesticides due to the long-range transport of these persistent pesticides to the Arctic [1]. Organochlorines bioaccumulated in various species, and biomagnified in top predators [2,3],

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many of which are consumed by Inuit populations, including beluga and seal [1]. In 1993, breastmilk from Inuit women living in Nunavik, Quebec, had organochlorine pesticide concentrations up to ten times higher than concentrations in breastmilk from southern Quebec women [4,5]. However, since the phase-out and international regulation of organochlorine pesticides under the Stockholm Convention, concentrations in the environment and in biologic media have generally decreased [6]. A significant temporal decrease was also observed in children and maternal blood samples in Nunavik, Iceland, the Faroe Islands, and Greenland [7,8].

With the decreased global use of organochlorine pesticides, other types of pesticides have been used as replacements [9], including chlorophenoxy, organophosphates, and pyrethroid pesticides. Chlorophenoxy herbicides are used against broad-leaf plants. The most widely used chlorophenoxy herbicides are 2,4-dichlorophenoxyacetic acid (2,4-D), 2,4,5-trichlorophenoxyacetic acid (2,4,5-T), and dicamba, and non-occupational exposure to these chemicals have been linked to non-Hodgkin's lymphoma, soft-tissue sarcoma, wheeze, and adverse birth outcomes [10–13]. Organophosphates are anticholinesterase insecticides used to spray over cultivated areas, in landscape management, and to fight pest infestation [14]. Chlorpyrifos, diazinon and malathion are among the most widely used organophosphate insecticides used for agricultural purposes in Canada and elsewhere in the world [9,15]. They have been associated with neurotoxicity, respiratory, and cardiometabolic outcomes [16–18]. Pyrethroids are a group of synthetic organic insecticides derived from natural pyrethrins from chrysanthemum flowers [19]. Pyrethroids have been replacing organophosphate insecticides to manage insects on crops and for indoor and residential use [20]. Environmental exposures to pyrethroids have been associated with adverse reproductive, neurologic and developmental outcomes [21–23].

Although these newer pesticide formulations are meant to be less persistent and bioaccumlative compared to organochlorines, current-use pesticides are being detected in Arctic abiotic and biotic samples (e.g. char, polar bear, and seal samples) [24–27]. Chlorpyrifos, diazinon and a pyrethroid were detected in air, snow, terrestrial biota, freshwater, and marine water samples from the Canadian Arctic [25]. For example, chlorpyrifos was detected in 19% and tefluthrin (a pyrethroid pesticide) was detected in 59% of 68 air samples from the Canadian High Arctic station of Alert (Nunavut) between August 2006 and October 2009. Temporal trends of these contaminants are less clear, with some indication of an increase in certain current-use pesticides in surface water samples, including tefluthrin, but no significant change over time for others, including chlorpyrifos [5]. Concentrations of current-use pesticides generally decreased with latitude in a study examining pesticide levels in air across Europe and the European Arctic, with evidence of long-range transport for some pesticides, including 2,4-D [27]. Additionally, the global use of pesticides is increasing, with some estimates showing an 80% increase in the global use of pesticides from 1990 to 2017 [28]. The United States is currently the 1st largest producer of pesticides worldwide, followed by Brazil and China in 2nd and 3rd place, and Canada as the 6th largest producer [29]. This could potentially lead to elevated exposure to current-use pesticides worldwide, and more specifically, in Arctic populations despite their limited use of pesticides.

No studies have examined the level of current-use pesticide exposure in Arctic populations. Given the legacy of elevated exposure to persistent organic pollutants, including per- and polyfluoroalkyl substances (PFAS), in Arctic populations [30,31], it is essential to further understand potential exposures to seemingly "safer" or "less persistent" chemicals in these already systematically excluded populations. Although dietary exposure is the primary non-occupational exposure pathway of pesticides in non-Arctic communities [32,33], the unique conditions in the Arctic and long-range transfer of contaminants may put Arctic communities at higher risk of exposure. The aim of this study was to explore the concentration of several current-use pesticide biomarkers in pooled urine samples from a health survey conducted in Nunavik, Quebec, examine the concentrations by age, sex, and ecological region, and to compare the concentrations in Nunavik to those in the general Canadian population. Pooled samples are a cost-effective method for large-scale population testing and to identify potential high-exposed groups [34–36].

Methods

Study population

Data for this study were collected for the Qanuilirpitaa? Nunavik Inuit Health Survey 2017 (Q2017). Q2017 is a health survey conducted for and by the Inuit population of Nunavik, Quebec in Canada and covers all 14 communities across the three ecological regions (Ungava Bay, Hudson Bay and the Hudson Strait) (Figure 1). The survey targeted community members aged 16 years and over, and implemented a stratified proportional model to select respondents based on community and age groups. Details of this survey have been described elsewhere [37,38]. Briefly, the data were collected onboard the Amundsen,

Figure 1. Map of Nunavik, Quebec, Canada showing the 14 communities and the three ecological regions. Source: Makivvik corporation (http://www.makivik.org/wp-content/uploads/2013/02/nunavik1.gif).

a Canadian Coast Guard Icebreaker, from 19 August 2017 to 5 October 2017. The relatively low response rate (33%) was driven by noncontact in this remote population; however, among those contacted, 80% agreed to participate. The study was governed by the OCAP® principles (Ownership, Control, Access, and Possession) and was conducted in close collaboration with several organisations in Nunavik.

Biomarker analyses

Urine is a primary biomonitoring matrix for non-persistent chemicals with short half-lives, including organophosphate and pyrethroid pesticides, due to easy collection and non-invasive process [39,40]. To compare the results to other populations (particularly the general Canadian population) and apply standardised methods, we opted to measure current-use pesticides in urine samples. Of the 1326 individuals who participated in the survey, 1266 participants provided urine samples. Urine samples were collected in 60-mL polypropylene plastic jars, and were kept at room temperature until samples were transported to the laboratory onboard the ship. Laboratory personnel transferred 1.5-ml aliquots of urine into 2-mL polypropylene tubes and were stored at – 80°C.

A total of 30 pooled samples based on sex, age, and ecological region were used to analyse pesticide biomarkers (Supplementary Table S1). Pooled samples provide a cost-effective method for environmental chemical analyses while ensuring a sufficient sample volume. Each pool had a total volume of 15 mL of urine and were created by adding equal amounts of urine from participants by age group (16–19 years; 20–29 years; 30–39 years; 40–59 years; 60+), sex (male; female), and ecological region (Hudson Bay; Hudson Strait; Ungava Bay). Details of these pooled samples were described previously. [41] Individual samples were first thawed to room temperature and vortex mixed. The volume pipetted from each individual sample was equal to the volume of the pooled sample (15 mL) divided by the total number of participants in the subgroup. Pooled samples were prepared in 60-ml polypropylene containers and kept frozen at – 20°C until time of analysis. Two laboratory blanks consisting of polypropylene containers filled with ultrapure water

Table 1. Pesticide biomarkers included in the study alongside their corresponding pesticide parent and their limits of detection (LOD).

Biomarker	Acronym	Pesticide	LOD (µg/L)
Chlorophenoxy herbicides			
2,4-dichlorophenoxyacetic acid	2,4-D	2,4-dichlorophenoxyacetic acid	0.01
2,4,5-trichlorophenoxyacetic acid	2,4,5-T	2,4,5-trichlorophenoxyacetic acid	0.004
3,6-dichloro-2-methoxybenzoic acid	Dicamba	Dicamba	0.1
Organophosphate metabolites			
3,5,6-trichloro-2-pyridinol	3,5,6-TCP	Chlorpyrifos, Chlorpyrifos-methyl, Triclopyr	0.02
2-isopropyl-6-methyl-4-pyrimidinol	IMPY	Diazinon	0.01
Malathion dicarboxylic acid	MDA	Malathion	0.02
4-nitrophenol	PNP	Parathion, methyl parathion, o-ethyl o-(4-nitrophenyl) phenylphosphonothioate	0.08
2-diethylamino-6-methylpyrimidin-4-ol	DEAMPY	Pirimiphos	0.01
Pyrethroid metabolites			
3-phenoxybenzoic acid	3-PBA	Pyrethroid	0.04
4-fluoro-3-phenoxybenzoic acid	4-F-3-PBA	Pyrethroid	0.004
cis-3-(2,2-dibromovinyl)-2,2-dimethylcyclopropanecarboxylic acid	cis-DBCA	Pyrethroid	0.06
cis-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylic acid	cis-DCCA	Pyrethroid	0.09
trans-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylic acid	trans-DCCA	Pyrethroid	0.02

were prepared at the same time as the pools and stored under the same conditions.

All laboratory analyses (CHMS and Q2017) were completed at the Centre de Toxicologie du Québec (CTQ) facilities of the Institut national de santé publique du Québec (INSPQ). Pooled urine samples were analysed for 13 pesticide biomarkers (Table 1). This included two chlorophenoxy herbicides (2,4-dichlorophenoxyacetic acid (2,4-D) and 2,4,5-trichlorophenoxyacetic acid (2,4,5-T)), six organophosphate pesticide metabolites (3,5,6-trichloro-2-pyridinol (3,5,6-TCP), 2-isopropyl-6-methyl-4-pyrimidinol (IMPY), malathion dicarboxylic acid (MDA), 4-nitrophenol (PNP), 2-diethylamino-6-methylpyrimidin-4-ol (DEAMPY), and 3,6-dichloro-2-methoxybenzoic acid (Dicamba)), and five pyrethroid metabolites (3-phenoxybenzoic acid (3-PBA), 4-fluoro-3-phenoxybenzoic acid (4-F-3-PBA), cis-3-(2,2-dibromovinyl)-2,2-dimethylcyclopropanecarboxylic acid (cis-DBCA), cis-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylic acid (cis-DCCA), and trans-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylic acid (trans-DCCA)).

Briefly, urine samples (250 µL) were enriched with labelled internal standards. The urinary metabolites were then hydrolysed using a β-glucuronidase enzyme solution (6300 units/mL) in a pH 5.0 acetate buffer. The samples were extracted by solid phase extraction (SPE) on Strata-X cartridges (30 mg/3 mL; Phenomenex, Torrance, CA, USA). The extracts were evaporated to dryness, reconstituted in 100 µL of acetonitrile:methanol:water solution (14:31:55. v:v:v) and then analysed by Ultra Performance Liquid Chromatography (UPLC I-Class Acquity; Waters, Milford, MA, USA) with a tandem mass spectrometer (MS/MS, AB Sciex 7500 system; Concord, Ontario, Canada) in the multiple reaction monitoring mode (MRM). The analytical column used was an Acquity Premier BEH C18 with VanGuard Fit (100 mm x 2.1 mm; 1,7 µm) (Waters, Milford, MA, USA). The analytical method was described in more details in Larose et al [13]. The intra-day precision ranged from 1.3 to 8.5% and the inter-day precision ranged from 1.8 to 7.8%. The accuracy of the method ranged from 91.9 to 107.1%. The LODs were between 0.004 and 0.1 µg/L (Table 1). Note that the analytical method used in CHMS was specific to pyrethroids and, as such, achieved lower LODs. Whereas, the method used in Q2017 was a screening method of 13 compounds of different families of pesticides, resulting in higher LODs for some pesticides.

Creatinine was measured in urine pooled samples to account for urinary dilution. The creatinine in urine was measured with a DRITM Creatinine-Detect kit from Microgenics Corporation (Thermo Fisher Scientific; Waltham, MA, USA) by spectrophotometry at a wavelength of 510 nm with an Analyzer Indiko Plus (Thermo Fisher Scientific; Waltham, MA, USA). The reference materials used to control the quality of the analyses were the non-certified MASTM Urichem TRAK L1 and L2 controls from Thermo Fisher Scientific (Waltham, MA, USA).

Statistical analyses

The percentage of samples with levels above a biomarker's LOD were calculated in the overall pool and in groups by age, sex, and ecological region. The arithmetic mean (AM) with its corresponding 95% confidence intervals (CI), the geometric mean (GM) and its corresponding 95% CI, and the 25th, 50th, 75th, and 95th

percentiles were calculated for the overall pools. The GMs and AMs and their corresponding 95% CIs were calculated by age, sex, and ecological region. Means were not calculated if pesticide biomarkers were detected in less than 60% of pooled samples. Pooled sample concentrations below the LOD were attributed a value of LOD/2. All measurements were adjusted for creatinine to account for urine dilution. AMs, GMS, their associated 95% CIs, and coefficients of variation (CVs) were calculated using the sum of the survey weights of the individuals forming each pool.

GMs, and the 25th and 75th percentiles in Q2017 were compared to those from the Canadian Health Measures Survey (CHMS), a national survey of a representative sample to measure indicators of health and wellness. The CHMS cycle differed by pesticide biomarker depending on data availability [35]. 3,5,6-TCP and MDA were measured in CHMS Cycle 4 (2014–2015), pyrethroid metabolites were measured in CHMS Cycle 5 (2016–2017), and 2,4-D and IMPY were measured in CHMS Cycle 6 (2018–2019). The AM provides an accurate indication of the mean of the population in the case of pooled samples, whereas the GM can be biased. [42] As such, AMs were used to compare concentrations by age, sex, and ecological region in Nunavik. However, the GMs were used to compare concentrations with the CHMS since the AM from larger surveys with individual samples are likely to be biased. We also calculated the GMs for the sake of comprehensiveness since most studies in the literature present GMs. Means in Q2017 and CHMS were only compared if the CV was below 33%. Population GMs were considered significantly different if the 95% CI of the GMs did not overlap.

All analyses were performed using SAS software (SAS Institute Inc, Cary, NC, USA).

Results

With the exception of five biomarkers, pesticide biomarkers were detected in a large majority of Q2017 pooled samples (Table 2). The highest concentrations were that of PNP, trans-DCCA, and 3,5,6-TCP. PNP (a metabolite of methyl and ethyl parathion) and 3,5,6-TCP (a metabolite of chlorpyrifos) had the highest concentrations among organophosphate pesticides (AM: 0.55 µg/g creatinine, 95% CI: 0.49–0.61; AM: 0.49 µg/g creatinine, 95% CI: 0.42–0.55). Trans-DCCA had the highest concentration among pyrethroid metabolites (AM: 0.51 µg/g creatinine, 95% CI: 0.26–0.77), but this mean had a CV < 33.3 and should be interpreted with caution. GMs in CHMS were generally similar to or higher than those in Q2017. Of note, even though 3,5,6-TCP had one of the highest concentrations in Q2017, the GM was 2.5-fold lower than the GM in CHMS.

There was a general increase in 2,4-D, 3,5,6-TCP, 3-PBA, and trans-DCCA with increasing age (Figure 2), but only the difference between 2,4-D among those

Table 2. Overall concentrations of pesticide biomarkers in 30 pooled samples from Q2017 and comparison with CHMS concentrations. All concentrations adjusted for creatinine; units in µg/g creatinine.

			GM (95% CI)		P25		P75	
Variable	%>LOD	AM (95% CI)	Q2017	CHMS	Q2017	CHMS	Q2017	CHMS
Chlorophenoxy herbicides								
2,4-D[1]	100	0.22 (0.18-0.25)	0.20 (0.18-0.24)	0.23 (0.21-0.26)	0.15	0.12	0.26	0.40
2,4,5-T	0	a	a	-	<LOD	-	<LOD	-
Dicamba	3.5	a	a	-	<LOD	-	<LOD	-
Organophosphate metabolites								
3,5,6-TCP[2]	100	0.49 (0.42-0.55)	0.47 (0.42-0.52)	1.20 (0.99-1.40)	0.37	0.61	0.54	2.00
IMPY[1]	100	0.06 (0.05-0.08)	0.06 (0.04-0.07)	a	0.04	0.04	0.08	0.16
MDA[2]	98.9	0.11[b] (0.05-0.16)	0.07 b (0.05-0.10)	a	0.03	0.06	0.12	0.20
PNP	100	0.55 (0.49-0.61)	0.53 (0.46-0.60)	-	0.44	-	0.65	-
DEAMPY	31.6	a	a	-	<LOD	-	0.02	-
Pyrethroid metabolites								
3-PBA[3]	100	0.35[b] (0.23-0.48)	0.25 (0.18-0.33)	0.52 (0.42-0.64)	0.16	0.19	0.40	1.00
4-F-3-PBA[3]	89.6	0.01 (0.008-0.02)	0.009 (0.006-0.01)	a	0.006	0.002	0.01	0.01
Cis-DBCA[3]	4.4	a	a	0.02 (0.01-0.02)	<LOD	0.007	<LOD	0.04
Cis-DCCA[3]	51.6	a	a	0.18 (0.13-0.25)	<LOD	0.06	0.108	0.34
Trans-DCCA[3]	100	0.51[b] (0.26-0.77)	0.31 (0.23-0.42)	0.26 (0.19-0.36)	0.19	0.09	0.45	0.52

a < 60% pooled samples above LOD.
b 16.6< Coefficient of variation ≤ 33.3.
1. Data retrieved from CHMS Cycle 6 (2018–2019)
2. Data retrieved from CHMS Cycle 4 (2014–2015)
3. Data retrieved from CHMS Cycle 5 (2016–2017)
– Not measured in CHMS

aged 16–19 years and 60–80 years was statistically significant (Supplementary Table S2). Females had consistently slightly higher concentrations of 3,5,6-TCP, MDA, PNP, and 3-PBA compared to males (Figure 3), but none of these differences were significant (Supplementary Table S1). No clear patterns were evident by ecological region, except for slightly lower concentrations of 2,4-D, 3,5,6-TCP, and PNP in the Hudson Strait (Figure 4), but these were not significant (Supplementary Table S2).

Similar to Q2017, concentrations of 2,4-D and 3,5,6-TCP also increased with age in CHMS. Cis-DCCA and trans-DCCA levels in the youngest age group (16–19 years) were slightly lower than concentrations in those aged over 60 years (Table 3). 2,4-D concentrations in those aged 16–19 years and aged over 60 years had higher concentrations in Q2017 compared to those in CHMS, but again, these differences were not significant. Similar to Q2017, females in CHMS also had higher concentrations of pesticides compared to males, particularly 2,4-D, 3-PBA, cis-DBCA, cis-DCCA, and trans-DCCA (Table 4).

Discussion

Several current-use pesticides were detected in pooled samples from Inuit youth and adults in Nunavik, including organophosphate pesticides, chlorophenoxy herbicides, and pyrethroids. Chlorpyrifos (3,5,6-TCP), methyl and ethyl parathion (PNP), and a pyrethroid (trans-DCCA) metabolites had the highest urinary biomarker concentrations. There was some evidence of higher concentrations of the chlorpyrifos metabolite (3,5,6-TCP) and pyrethroid metabolites (3-PBA and trans-DCCA) with increasing age, and some evidence of higher pesticide concentrations among females versus males. However, most of these differences were not significant. Contrary to other persistent environmental contaminants, concentrations of most current-use pesticides in Q2017 pooled samples were similar to or lower than those in the general Canadian population (from CHMS).

Similar exposure trends were also detected in the U.S. Concentrations of the chlorophenoxy herbicide 2,4-D (GM 0.36 µg/g of creatinine, 95% CI 0.33–0.40 in 2015–2016), chlorpyrifos (3,5,6-TCP GM 1.26 µg/g of creatinine, 95% CI 1.18–1.35 in 2015–2016), parathions (PNP GM 0.61 µg/g of creatinine, 95% CI 0.57–0.66 in 2015–2016), and pyrethroids (3-PBA GM 10.78 µg/g of creatinine, 95% CI 10.72–10.84 in 2015–2016) in the National Health and Nutrition Examination Survey (NHANES) were higher than those in Nunavik adults [43]. 2,4,5-T and cis-DBCA were also not detected in the general population in NHANES in 2009–2010 (the latest available data). In contrast, IMPY, malathion, 4-F-3-PBA, and trans-DCCA were not detected in NHANES in 2015–2016, even though low concentrations were detected in Nunavik, albeit at low concentrations. Nunavik concentrations were also similar to or below concentrations in several other countries, including China, Germany, and Japan [23,32,33,44].

A study examining environmental pesticide exposures in eight countries observed similar pesticide exposure profiles, wherein 3,5,6-TCP, PNP, trans-DCCA, and 3-PBA also made up the largest relative abundance of urinary pesticide biomarkers (similar to the biomarkers of the current study) in study populations based in Vietnam, Korea, Japan, India, China, Greece, and the U.S. [33]. The higher concentration of 3-PBA and trans-DCCA

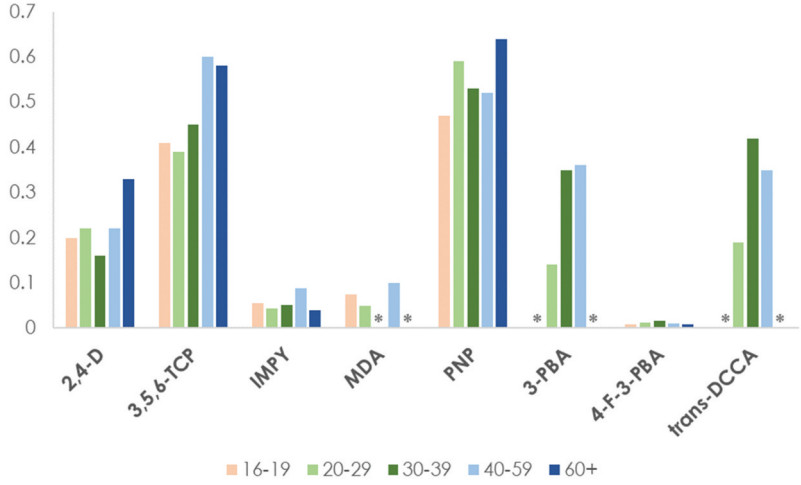

Figure 2. AMs of pesticide biomarkers by age group (years) in 30 pooled samples from Q2017 in Nunavik. All concentrations adjusted for creatinine; units in µg/g creatinine. *Coefficient of variation > 33.3; unreliable value.

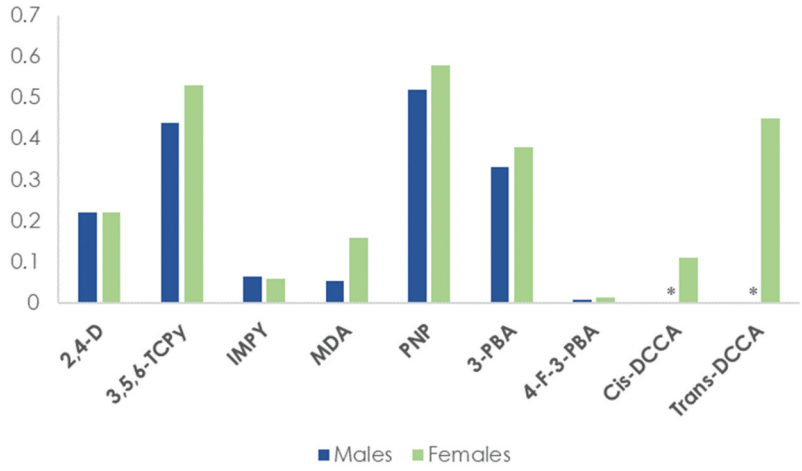

Figure 3. Arithmetic means of pesticide biomarkers by sex in 30 pooled samples from Q2017 in Nunavik. All concentrations adjusted for creatinine; units in µg/g creatinine. *Coefficient of variation > 33.3; unreliable value.

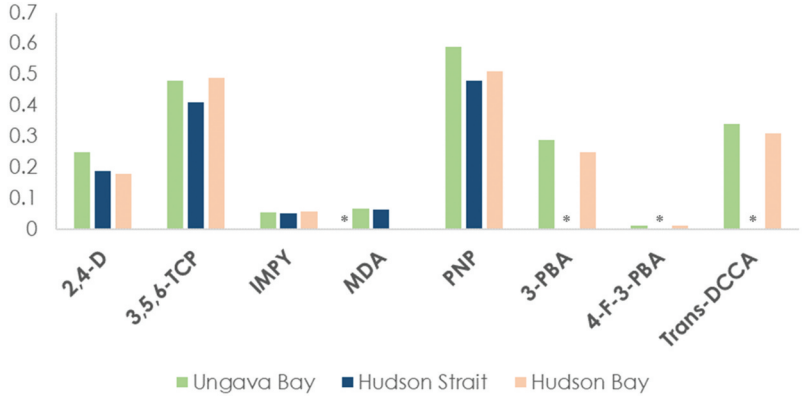

Figure 4. Arithmetic means of pesticide biomarkers by ecological region in 30 pooled samples from Q2017 in Nunavik. All concentrations adjusted for creatinine; units in µg/g creatinine. *Coefficient of variation > 33.3; unreliable value.

versus other pyrethroid metabolites may be indicative of permethrin and cypermethrin parent pesticide usage, which is consistent with reported pesticide use in agriculture-heavy nations [33].

The similarity or lower exposure levels and profiles in Nunavik and Canada indicates similar exposure pathways. Diet is usually the major source of pesticide exposure in non-occupationally exposed populations [44]. Despite the reliance on local country foods in Nunavik (which consists of harvested and hunted wild foods), the results may indicate similar overall patterns in some fruit and vegetable consumption. The consumption frequency of foods associated with increased 3-PBA differed by study population and country, however, fruits and leafy green vegetables, juice, and beans were among the common foods associated with 3-PBA urinary levels [32,44]. Associations between pesticide exposure and red meat and poultry varied by region, with some studies finding no association, and others detecting strong correlations [32]. Similarly, lower 3-PBA, cis-DCCA, and trans-DCCA levels were lower in populations consuming organic diets (uncommon in Nunavik) versus conventional diets [32]. Preparation methods also impacted exposures, such that while raw vegetables were associated with pyrethroid metabolite concentrations, cooked vegetable were not [32]. Residential use of pesticides was also associated with an increase in 3-PBA levels in some studies, but this finding was not consistent across studies [44]. Alternatively, the similar exposure concentrations in Arctic versus non-Arctic populations may point to efficient environmental degradation/metabolism of these pesticides if they were to reach the Arctic via long-range transport. For instance, significant trophic dilution was observed for several current-use pesticides, and no intertrophic level biomagnification was

Table 3. GMs and corresponding 95% confidence intervals by age group (years) for pesticide biomarkers in 30 pooled samples from Q2017 and individual samples from CHMS. All concentrations adjusted for creatinine; units in μg/g creatinine.

	16-19	20-29	30-39	40-59	60-80
Chlorophenoxy herbicides					
2,4-D					
Q2017	0.20 (0.16-0.24)	0.20[a] (0.11-0.35)	0.16 (0.12-0.20)	0.21 (0.15-0.30)	0.33 (0.29-0.38)
CHMS[1]	0.15 (0.12-0.20)	0.21 (0.15-0.30)	0.24 (0.18-0.30)	0.25 (0.21-0.29)	0.25 (0.21-0.30)
2,4,5-T					
Q2017	a	a	a	a	a
CHMS	–	–	–	–	–
Dicamba					
Q2017	a	a	a	a	a
CHMS	–	–	–	–	–
Organophosphate metabolites					
3,5,6-TCP					
Q2017	0.40 (0.34-0.48)	0.38 (0.34-0.43)	0.44 (0.36-0.53)	0.59 (0.44-0.78)	0.56 (0.40-0.78)
CHMS[2]	0.97 (0.83-1.10)	0.99 (0.73-1.30)	1.10 (0.91-1.30)	1.20 (0.92-1.50)	1.40 (1.20-1.60)
IMPY					
Q2017	0.05[b] (0.02-0.13)	0.04[b] (0.02-0.10)	0.05 (0.03-0.09)	0.09 (0.06-0.12)	0.04[b] (0.02-0.07)
CHMS[1]	a	a	a	a	a
MDA					
Q2017	0.06[b] (0.03-0.13)	0.05[b] (0.03-0.07)	c	0.08[b] (0.04-0.19)	c
CHMS[2]	a	a	a	a	a
PNP					
Q2017	0.47 (0.41-0.53)	0.57 (0.45-0.74)	0.52 (0.41-0.67)	0.48[b] (0.29-0.81)	0.63 (0.49-0.81)
CHMS	–	–	–	–	–
DEAMPY					
Q2017	a	a	a	a	a
CHMS	–	–	–	–	–
Pyrethroid metabolites					
3-PBA					
Q2017	c	0.12[b] (0.06-0.24)	0.28[b] (0.12-0.63)	0.34 (0.23-0.49)	c
CHMS[3]	0.33 (0.25-0.43)	0.48 (0.23-1.00)[c]	0.57 (0.38-0.85)[b]	0.51 (0.43-0.60)	0.55 (0.40-0.75)
4-F-3-PBA					
Q2017	c	c	0.015[b] (0.007-0.031)	0.009[b] (0.004-0.019)	0.008[b] (0.005-0.014)
CHMS[3]	c	c	c	c	c
Cis-DBCA					
Q2017	a	a	a	a	a
CHMS[3]	0.02 (0.01-0.03)[b]	0.02 (0.01-0.04)[b]	0.02 (0.01-0.02)	0.01 (0.01-0.02)	0.02 (0.02-0.03)
Cis-DCCA					
Q2017	c	–	–	–	c
CHMS[3]	0.12 (0.08-0.16)	0.18 (0.07-0.46)[c]	0.18 (0.11-0.30)[b]	0.17 (0.12-0.24)	0.19 (0.14-0.28)
Trans-DCCA					
Q2017	c	0.16[b] (0.07-0.33)	0.34[b] (0.15-0.77)	0.34 (0.24-0.48)	c
CHMS[3]	0.19 (0.14-0.27)	0.31 (0.12-0.83)[c]	0.28 (0.17-0.48)[b]	0.24 (0.17-0.33)	0.27 (0.20-0.37)

a > 40% below LOD.
b 16.6< Coefficient of variation ≤ 33.3.
c Coefficient of variation > 33.3.
1. Data retrieved from CHMS Cycle 6 (2018–2019)
2. Data retrieved from CHMS Cycle 4 (2014–2015)
3. Data retrieved from CHMS Cycle 5 (2016–2017)
– Not measured in CHMS

Table 4. GMs and corresponding 95% confidence intervals by sex for pesticide biomarkers in 30 pooled samples from Q2017 and individual samples from CHMS. All concentrations adjusted for creatinine; units in µg/g creatinine.

	Male	Female
Chlorophenoxy herbicides		
2,4-D		
Q2017	0.20 (0.16-0.26)	0.20 (0.16-0.26)
CHMS[1]	0.19 (0.17-0.22)	0.28 (0.25-0.32)
2,4,5-T		
Q2017	a	a
CHMS	–	–
Dicamba		
Q2017	a	a
CHMS	–	–
Organophosphate metabolites		
3,5,6-TCP		
Q2017	0.43 (0.40-0.48)	0.50 (0.40-0.62)
CHMS[2]	1.10 (0.92-1.40)	1.20 (1.00-1.50)
IMPY		
Q2017	0.06[b] (0.03-0.11)	0.06 (0.04-0.08)
CHMS[1]	a	a
MDA		
Q2017	0.04[b] (0.03-0.06)	0.10[b] (0.06-0.17)
CHMS[2]	a	a
PNP		
Q2017	0.49 (0.38-0.63)	0.57 (0.50-0.64)
CHMS	–	–
DEAMPY		
Q2017	a	a
CHMS	–	–
Pyrethroid metabolites		
3-PBA		
Q2017	0.20[b] (0.13-0.32)	0.30[b] (0.20-0.45)
CHMS[3]	0.38 (0.30-0.47)	0.71 (0.53-0.95)
4-F-3-PBA		
Q2017	0.007[b] (0.004-0.01)	0.01[b] (0.007-0.02)
CHMS[3]	c	c
Cis-DBCA		
Q2017	a	a
CHMS[3]	0.01 (0.01-0.02)	0.02 (0.02-0.02)
Cis-DCCA		
Q2017	a	0.08[b] (0.05-0.13)
CHMS[3]	0.14[b] (0.09-0.20)	0.23[b] (0.16-0.34)
Trans-DCCA		
Q2017	0.27[b] (0.16-0.46)	0.36 (0.25-0.50)
CHMS[3]	0.20 (0.15-0.28)	0.33[b] (0.22-0.50)

a > 40% below LOD.
b 16.6< Coefficient of variation ≤ 33.3.
c Coefficient of variation > 33.3.
1. Data retrieved from CHMS Cycle 6 (2018–2019)
2. Data retrieved from CHMS Cycle 4 (2014–2015)
3. Data retrieved from CHMS Cycle 5 (2016–2017)
– Not measured in CHMS

observed in Arctic food webs on a total body burden basis [25].

There was some evidence of higher pesticide biomarker concentrations by age and among females, albeit inconsistently, in Nunavik and the general Canadian population. Participants aged 40–59 and 60+ in Nunavik are more likely to consume country foods harvested or hunted from the land and less likely to consume market foods purchased at stores (including fruits and vegetables) [45], so it is unclear why older participants had

higher concentrations of 2,4-D, 3,4,5-TCP, PNP, 3-PBA, and trans-DCCA. This may point to bioaccumulation, although rates of bioaccumulation are low to moderate for these pesticides [46–48]. There may be other lifestyle or biological factors unaccounted for in the study. Some measurements were also too unreliable among those aged 60+ for a comprehensive analysis of pesticide exposure by age. Females are more likely to consume market foods and fruits and vegetables in Nunavik compared to males [45], and this may account for the differences observed. This is supported by the slightly lower concentrations of pesticide biomarkers among individuals living in the Hudson Strait, who generally consume a higher proportion of country foods over market foods [45].

Although 2,4,5-T use in Canada has been heavily restricted (and no longer registered for use in the U.S.), 2,4-D remains one of the top ten herbicides used in Canada. Chlorpyrifos was one of the most commonly used organophosphates worldwide, and among the top ten insecticides used in Canada [33,49]. However, it has been under extra scrutiny in recent years, and was banned in Canada in December 2023 [50]. It was banned for use in the European Union in 2020, and is currently under review to be included under the list of persistent organic pollutants of the Stockholm Convention, which may push for more uniform regulation globally. Parathion use was also heavily restricted in developed countries due to suspected adverse health effects, however, occupational and environmental exposures are still reported, suggesting its continued use [51]. A review found that pyrethroid biomarker concentrations have increased in recent years [44], indicating higher rates of use. They have a variety of uses, from organophosphate pesticide replacements for agriculture, livestock, forestry, and residential use [52]. Pyrethroids are also commonly used to impregnate mosquito nets and for medical applications [52].

The detectable concentrations of current-use pesticides in Nunavik and the anticipated increase in pesticide use in the future indicate the need for continued monitoring in Arctic populations. The lower levels of current-use pesticides compared to organochlorine pesticide levels are expected due to organochlorines bioaccumulation and biomagnification properties. Similar to this study, pooled samples could be used in the future as an efficient and cost-effective method of monitoring, given that current concentrations are relatively low. Environmental exposures, i.e. low and chronic exposure, to these pesticides at similar concentrations have been linked with a myriad of health outcomes in other populations [17,19,22,53,54]. It is important to understand their potential implications in northern populations who are still exposed to exceptionally high levels of persistent organic pollutants and heavy metals [30,55], which may increase northern populations' susceptibility to further environmental exposures. Further studies should consider including glyphosate in the list of monitored pesticides due to its extensive use worldwide and in Canada [56].

The purpose of this study was to document potential exposures to current use pesticides in an Arctic community, however the use of pooled samples has limitations. The lack of individual-level samples limits our statistical power and we had a limited number of pools which may have rendered outliers to be influential. However, we included CVs to account for the dispersion and to help in the interpretation of results with more confidence. Not all urinary biomarkers were measured in the CHMS, but we were able to compare concentrations in Nunavik to those in other countries, including the U.S. which tends to have similar concentrations to those in Canada. We were also unable to further explore associations between pesticide exposure and other lifestyle or socioeconomic factors (such as income or eating habits) or health outcomes. The use of single urine samples may underestimate pesticide exposure. The CHMS LODs for pyrethroids were 4–10 times lower than the LODs in Q2017 (with the exception of 4-F-3-PBA), which must be considered when comparing concentrations. However, the conclusions remain the same since the pyrethroid levels in Q2017 were lower than those in CHMS despite the higher LODs. There were multiple comparisons by age, sex, and region, and this may lead to an increased Type I error. Lastly, while some studies used to compare concentrations against those in Nunavik used first void urine samples, others did not specify the time of day urine was collected. Despite these limitations, our study was the first to document environmental exposures to pesticides in an Arctic community using a cost-effective and reliable method.

Conclusion

Environmental pesticide exposures were detected in Nunavik. Pesticides with the highest concentrations included metabolites of chlorpyrifos, parathion, and some pyrethroids, consistent with other studies conducted globally. The concentrations were largely similar to or lower than concentrations detected in the general Canadian population. The study points to the importance of regular monitoring of environmental contaminants in Arctic populations.

Disclosure statement

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