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Mechanistic polychlorinated biphenyl exposure modeling of mothers in the Canadian Arctic: the challenge of reliably establishing dietary composition

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ABSTRACT

Background: Traditional food (TF) consumption represents the main route of persistent organic pollutant (POP) exposure for indigenous Arctic Canadians. Ongoing dietary transitions away from TFs and toward imported foods (IFs) may contribute to decreasing POP exposures observed in these groups.
Methods: To explore this issue, we combined the global fate and transport model GloboPOP and the human food chain bioaccumulation model ACC-Human Arctic to simulate polychlorinated biphenyl (PCB) exposure in two indigenous Arctic Canadian communities from the Inuvik region, Northwest Territories and Baffin region, Nunavut. Using dietary survey information from initial (1996–98) and follow-up (2005–07) biomonitoring campaigns in Inuvik and Baffin, we simulated PCB exposures (PCB-118, -138, -153, and -180) for each individual study participant and also whole study populations.
Results: TF intake rates, particularly of marine mammals (MMs), were the most important predictors of modeled PCB exposure, while TF consumption did not associate consistently with measured PCB exposures. Further, reported mean TF intake increased from baseline to follow-up in both Inuvik (from 8 to 183 g d^{-1}) and Baffin (from 60 to 134 g d^{-1}), opposing both the expected dietary transition direction and the observed decrease in human PCB exposures in these communities (ΣPCB Inuvik: from 43 to 29 ng g $lipid^{-1}$, ΣPCB Baffin: from 213 to 82 ng g $lipid^{-1}$). However dietary questionnaire data are frequently subject to numerous biases (e.g., recall, recency, confirmation), and thus casts doubt on the usefulness of these data.
Conclusions: Ultimately, our model's capability to reproduce historic PCB exposure data in these two groups was highly sensitive to TF intake, further underscoring the importance of accurate TF consumption reporting, and clarification of the role of dietary transitions in future POP biomonitoring of indigenous Arctic populations.

1. Introduction

The exposure susceptibility of indigenous human populations throughout the Canadian Arctic to persistent organic pollutants (POPs) has been documented extensively (AMAP, 2004, 2009; Donaldson et al., 2010; NCP, 2003; Van Oostdam et al., 2005). Though these groups reside far from major industrial emissions sources, long-range transport and food chain bioaccumulation (AMAP, 2004, 2009; Brown and Wania, 2008; Czub et al., 2008; Donaldson et al., 2010; NCP, 2003; Van Oostdam et al., 2005; Wania, 2003) allow these chemicals to reach appreciable levels in Arctic marine mammals (Addison et al., 2014; Bentzen et al., 2008; Stern et al., 2005) and humans (AMAP, 2004, 2009; Curren et al., 2014a; Donaldson et al., 2010; Laird et al., 2013; NCP, 2003; Van Oostdam et al., 2005). Dietary intake of traditional food (TF) items is the main route of indigenous Arctic POP exposure, with consumption of lipid-rich blubber tissue from upper trophic level marine mammals (MMs) being particularly important; demonstrated through both empirical (AMAP, 2004, 2009; Dewailly et al., 1993, 1994; Donaldson et al., 2010; NCP, 2003; Van Oostdam et al., 2005) and modeling analyses (Czub et al., 2008; Quinn et al., 2012; Undeman et al., 2010). However the nutritional benefits of TFs are also substantial (Kuhnlein et al., 2004), and based on their significant contribution to dietary adequacy, the benefits of continued TF consumption by indigenous Arctic Canadians generally outweigh any contaminant exposure risks (Health Canada, 2007).

Concern over human POP exposure is due to these chemicals' known potential for deleterious health effects. Cardiovascular impairment, reproductive deficits, and immunosuppression have been documented

Abbreviations: POP, persistent organic pollutant; PCB, polychlorinated biphenyl; TF, traditional food; IF, imported food; MM, marine mammal; WCBA, women of childbearing age.

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in indigenous Arctic groups (AMAP, 1998, 2003, 2009), while recent studies from non-Arctic populations suggest potential links between POP exposure and metabolic syndromes (Lee et al., 2011). These results are of note as metabolic syndrome represents a growing health challenge for northern indigenous peoples (Hopping et al., 2010a; Young et al., 2007). Additionally, POPs have been linked to neurocognitive toxicity, especially during prenatal, postnatal, and early childhood stages (Boucher et al., 2010, 2012, 2014; Jacobson et al., 1990; Stewart et al., 2008; Walkowiak et al., 2001). For this reason, POP intakes by expectant, pregnant, and nursing mothers, and more widely women of childbearing age (WCBA), also represent a primary exposure issue (Binnington et al., 2014). The health consequences of POP exposure remain relevant in the Canadian Arctic as indigenous populations continue to exhibit measured POP concentrations greater than Southern comparator groups (Curren et al., 2014a). For example, (Laird et al., 2013) recently noted that 28% of female participants of childbearing age in the Inuit Health Survey exceeded 5 μg L^{-1} of total polychlorinated biphenyl (PCB) in blood, which was considered to be an exceedance of tolerable blood levels at the time.

Despite the fact that exposure concerns for POPs still persist among certain segments of Canadian indigenous Arctic populations, biomonitoring provides evidence that levels of POPs in Canadian indigenous Arctic populations have been decreasing over the past two decades (Armstrong et al., 2007; Dallaire et al., 2003; Donaldson et al., 2010; Muckle et al., 2001a, 2001b). Though this trend is in part due to decreasing levels of many POPs in both the Arctic environment (Hung et al., 2010) and relevant TF species (Braune et al., 2005; Riget et al., 2010), it has also coincided with a major dietary transition among circumpolar indigenous populations typified by intergenerational declines in the dietary proportion of TF (Kuhnlein et al., 2004; Popkin, 2006; Sharma, 2010). Decreasing overall intake of TF, and declining MM intake as a proportion of total TF consumption, have likely contributed to the trend of falling POP levels in some communities (Armstrong et al., 2007; Donaldson et al., 2010; Potyrala et al., 2008). Notably, MM TF intakes are typically greater among coastal residents of the Eastern Canadian Arctic than their Western counterparts (Armstrong et al., 2007; Kuhnlein and Chan, 2000; Potyrala et al., 2008). As a result, Eastern Arctic indigenous populations regularly exhibit higher blood (plasma) POP concentrations than those from the Western Arctic (Butler Walker et al., 2003; Donaldson et al., 2010; Muckle et al., 2001a).

Previous work by our group attempted to quantify the independent impacts of reduced global POPs emissions and dietary transition behavior on declining POP levels in a generic Arctic indigenous population using a deterministic modeling approach (Quinn et al., 2012). Specifically, we simulated human POP exposure through sequential calculations with the global chemical fate and transport model GloboPOP (Armitage et al., 2011; Wania and Mackay, 1995; Wania and Su, 2004) and the Arctic version of the food chain bioaccumulation model ACC-Human (Czub and McLachlan, 2007; Czub et al., 2008) to investigate a variety of hypothetical dietary transition scenarios varying in their degree of intergenerational dietary change and time of onset. Elements of this approach have been successfully evaluated using empirical PCB data in both environmental compartments (Armitage et al., 2011; Wania and Mackay, 1995; Wania and Su, 2004), and upper trophic level wildlife (Binnington and Wania, 2014; Czub et al., 2008). Ultimately, we estimated dietary change to result in anywhere from a 2 to 50-fold reduction in PCB exposure depending on assumed dietary variables. This was contrasted by a 6 to 13-fold decrease in human PCB levels attributed to declining PCB emissions.

In order to complement these generic scenarios with empirically observed trends of POP contamination in populations that had experienced a dietary transition, we sought out POP biomonitoring studies that had sampled the same Arctic indigenous population and collected TF intake information during two distinct time periods. Two Canadian POP biomonitoring studies performed in the Inuvik Region of the Northwest Territories (NT — Western Canadian Arctic) and Baffin Region of Nunavut (NU — Eastern Canadian Arctic) included baseline and follow-up convenience sampling of pregnant women several years apart; relevant details are outlined in the Methods Section 2.1 below. We also noted the need to expand our model framework to include Arctic organisms other than ringed seal and polar cod (Quinn et al., 2012) to more realistically reflect Northern TF consumption. As a first step, bioaccumulation modules for beluga whale (*Delphinapterus leucas*) and bowhead whale (*Balaena mysticetus*) were recently added to the ACC-Human Arctic model (Binnington and Wania, 2014).

The three main purposes of this study were: i) to expand our ACC-Human Arctic model framework to more closely match the breadth of TF items consumed in Canadian Arctic diets; ii) to evaluate the ability of our expanded model to reproduce historic PCB concentrations measured during Arctic human biomonitoring; and iii) to clarify the relative impacts of decreasing environmental POP levels and diminishing TF intake on declining indigenous Arctic Canadian human POP concentrations.

2. Methods

2.1. Details of the Arctic biomonitoring campaigns

The Northern Contaminants Program (NCP) of Indigenous and Northern Affairs Canada (INAC) was responsible for coordinating the two baseline and follow-up biomonitoring studies in the Inuvik, NT and Baffin, NU regions of the Canadian Arctic (Armstrong et al., 2007; Curren et al., 2014a, 2014b; Donaldson et al., 2010). The baseline studies, referred to as Inuvik-1 and Baffin-1, were performed between 1997–1999; the two corresponding follow-up campaigns, denoted as Inuvik-2 and Baffin-2, were conducted in 2005–2007. Expectant indigenous and non-indigenous mothers were recruited during their current pregnancy through the local hospitals serving each region, and provided signed informed consent prior to study participation. The research ethics boards of both participating regional hospitals, as well as the relevant territorial health authorities for the NT and NU approved the study protocols. The follow-up studies did not attempt to re-enroll participants from the baseline studies, as this would have required individuals to be pregnant during each study period. Thus, the follow-up campaigns represent subsequent measurement of distinct convenience sub-samples within the same greater maternal populations. Because the biomonitoring data were de-personalized, we cannot confirm whether any individuals participated in both studies. Demographic data for each of the four study populations are presented in Table 1, while average maternal intakes of different types of TF are presented in Table 2. Note that based on the limited number of non-indigenous participants in all four biomonitoring studies (32/298 participants — Table 1), we refer to these populations as “indigenous” throughout the results and discussion for convenience. These tables also list the assumptions that were necessary to facilitate individual and population modeling for each group of study participants, based on the specificity of interview data collected in each campaign. For example, while the studies collected information on the parity of each mother during her current pregnancy, they did not record the age at which mothers gave birth to each child. As age(s) at birth are required in our model, and can impact a women's lifetime POP exposure (Quinn et al., 2010; Verner et al., 2008), a value for this variable was assumed during calculations.

Curren et al. (2014a) provide details on the studies' sampling methodology. Briefly, maternal blood was collected late in the third trimester, specifically within one day of birth. Samples were obtained from the antecubital vein, and plasma was isolated by means of centrifugation prior to freezing and shipment to the Institut National de Santé Publique du Québec (INSPQ) in Québec City, Québec, Canada for POP identification and quantification via gas chromatography-mass spectrometry. PCB concentrations below the limit of detection (LOD) were substituted