



Effects of perfluorooctanesulfonate exposure on plasma lipid levels in the Inuit population of Nunavik (Northern Quebec) ^{☆, ☆, ☆}

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ABSTRACT

Background: Perfluorooctanesulfonate (PFOS) was used as a surfactant in various commercial products. In rodents, exposure to this compound induced various health effects, including hypolipidemia. In human populations, the potential toxicity of PFOS is not yet fully characterized, but indications of effects on lipids are reported. A recent study reported an increase in plasma cholesterol associated with exposure to perfluorinated compounds in humans exposed through drinking water, but similar effects were not reported in all exposed human populations. PFOS is widely distributed in the environment, including the arctic biota. The Inuit of Nunavik are exposed to environmental contaminants through the consumption of fish and game. This diet is also a source of ω -3-polyunsaturated fatty acids (n-3 PUFAs) that are known to lower plasma triacylglycerols.

Objective: This cross-sectional epidemiologic study aims at assessing the relationship between PFOS exposure and plasma lipids, while taking account of the concomitant hypolipidemic effect exerted by n-3 PUFAs.

Methods: Plasma concentrations of PFOS and lipids were assessed in Nunavik Inuit adults ($n=723$) in the framework of a large-scale environmental health study. Associations of exposure levels to age, gender and selected wild food consumption associated with n-3 PUFAs intake, as well as the exposure on lipid levels were investigated by multivariate linear modeling.

Results: In the Inuit population, PFOS exposure and n-3 PUFAs intake are related to traditional food consumption. Triacylglycerol and ratio of total cholesterol to high density lipoprotein cholesterol (HDL-C) levels were negatively associated with PFOS plasma levels, while HDL-C levels were positively associated, after adjustment for circulating levels of n-3 PUFAs and for the interaction between gender and PFOS plasma levels. Other plasma lipids, such as low density lipoprotein-cholesterol and non-HDL-C were not related to PFOS plasma concentrations.

Conclusion: The results of this study show a relationship between PFOS and plasma lipid levels in an environmentally exposed human population, and this effect appears distinct from that of n-3 PUFAs.

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Abbreviations: ALA, alpha linolenic acid; BMI_{std}, body mass index standardized for the Cormic index (sitting height/height); DHA, docosahexaenoic acid; EPA, eicosapentaenoic acid; FA, fatty acids; FID, flame ionization detector; HDL-C, high density lipoprotein cholesterol; IDF, international federation of diabetes; LC-MS-MS, liquid chromatography tandem mass spectrometry; n-3 PUFAs, Ω -3 polyunsaturated fatty acids; LDL-C, low density lipoprotein cholesterol; OA, oleic acid; PFC, perfluoroalkyl compound; PFOA, perfluorooctanoate; PFOS, perfluorooctanesulfonate; POSF, perfluorooctanesulfonyl fluoride; PPAR, peroxisome proliferator activated receptors; RBC, red blood cells; SE, standard error of the mean; TG, triacylglycerol; TC, total cholesterol

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1. Introduction

Perfluorooctanesulfonate (PFOS) is a fully fluorinated aliphatic molecule derived by electrochemical fluorination from perfluorooctanesulfonyl fluoride (POSF), or arising from the degradation or metabolism of other perfluoroalkyl compounds (PFCs) obtained through a similar process (3M, 1999; Olsen et al., 2005). PFOS and related compounds were mainly used as surfactants in a wide variety of commercial products, including oil and water repellents for textiles and food packaging, fire extinguishing foams and insecticides. They exhibit a remarkable resistance to environmental and metabolic degradation, due to the strength of the carbon–fluorine bonds, conferring to these molecules their unique properties (3M, 1999). Despite the production and use of PFCs for the past sixty years, concerns regarding environmental hazards related to these compounds arose only recently, and a growing body of literature is now building regarding human and wildlife exposure (Butenhoff et al., 2006; Calafat et al., 2007a; Fromme et al., 2007; Houde et al., 2006; Butenhoff, 2006 #1479; Kannan et al., 2004; Kannan et al., 2002; Karrman et al., 2006). Main sources of exposure to PFCs suggested in non-occupationally exposed human populations include inhalation of contaminated dust from indoor or outdoor air (Kubwabo et al., 2005; Shoeib et al., 2006; Shoeib et al., 2005) as well as dietary intake of wild fish (Falandysz et al., 2006; So et al., 2006), crustaceans and shellfish (Clarke et al., 2010) and different commercial foods (Begley et al., 2005; Clarke et al., 2010; Tittlemier et al., 2007). A recent analysis of NHANES data suggest that food is the primary source of PFOS exposure in American adults (Egeghy and Lorber, 2010).

Toxicological studies on adult laboratory animals (Cynomolgus primates, rats and mice) have shown that repeated dosing with PFOS induced several adverse health effects, among which metabolic wasting, hepatomegaly and decline in triacylglycerols (TG) and/or total cholesterol (TC) levels could be observed (Seacat et al., 2003; Seacat et al., 2002; Thibodeaux et al., 2003). However, the lipid lowering action was observed at high doses of PFOS and seemed not the earliest physiologic response after exposure (Seacat et al., 2003). In primates, Seacat and collaborators (2002) observed decreases in lipids levels only at higher administered doses (0.75 mg/kg/day) and mainly among males (Seacat et al., 2002). In rats, they detected lowering serum cholesterol after 14 weeks of exposure to 20 ppm PFOS in males only (Seacat et al., 2003). In humans, most epidemiological studies addressing the effects of PFCs were conducted in occupationally exposed populations with serum concentrations ranging 10–2000 ng/mL (Grice et al., 2007; Olsen et al., 2003; Olsen et al., 1999) and do not consistently report hypolipidemic effects, as those observed in animal studies. Moreover a recent epidemiological investigation of 46,294 adults (18 yr and above) residents of West Virginia, who drank PFOA-contaminated groundwater, reported a positive association between plasma PFOS levels (median=20 ng/mL) with all lipids, except high density lipoprotein cholesterol (HDL-C) which contradicts the effect observed in animal studies (Steenland et al., 2009). Those results were corroborated at least for non-HDL-C and total cholesterol by a cross-sectional study on the general US population (PFOS serum concentration median=21 µg/L) (Nelson et al., 2010).

The Inuit of Nunavik (Northern Quebec, Canada) are exposed to relatively high doses of several environmental contaminants through their traditional diet, which typically includes the consumption of fish and wild game, as well as marine mammal meat and fat (Dewailly et al., 1993). However, this diet is also an important source of long chain polyunsaturated fatty acids (n-3 PUFAs) such as eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA), which have been related to beneficial health effects, especially regarding the lowering of plasma lipids (Dewailly et al.,

2003; Holub, 1988; Simopoulos, 1991). PFOS is found in the arctic marine food chain, where it has shown a potential for bioaccumulation and biomagnification (Bossi et al., 2005; Haukas et al., 2007; Houde et al., 2006; Kannan et al., 2001; Powley et al., 2008), and its presence at northern latitudes suggests that it may undergo long range transport (Stock et al., 2007) and contaminate many of the Inuit's traditional food items. The objective of this cross-sectional epidemiologic study is to evaluate the effect of PFOS exposure on blood lipids in the Inuit population, while taking into account the already known hypolipidemic effect exerted by n-3 PUFAs found in traditional food items.

2. Materials and methods

2.1. Study design, data collection, sample size

A large-scale environmental health study was carried out in Nunavik during Fall 2004. The survey used a community-stratified random sampling of households, and targeted people aged 18–74 yrs old in order to allow a standard representation of the Nunavik population. Participants were contacted and given explanations on the projects' aim, tests and questionnaires before giving their written consent to participate. Among the 677 contacted households, 521 agreed to participate (household response rate of 77.8%), 1056 individuals signed a consent form and 917 people agreed to have blood samples taken for clinical and toxicological analyses (final participation rate of 67%). All communities were visited by the scientific icebreaker CCGS *Amundsen* and study participants were invited onboard for interviews and clinical sessions (Fig. 1). Participants answered questionnaires (some self administered, other assisted by trained interviewers) designed to collect information on socio-demographic characteristics, environmental factors, dietary and lifestyle habits, and underwent a series of clinical examinations and finally donated a blood sample for further toxicology and clinical biochemistry analyses. For the purposes of the current study, specific exclusion criteria were considered in order to avoid potential biases on the studied endpoints induced by pregnancy, non-fasting conditions (less than 8 h fasting) and non-Inuit ethnic origin ($n=194$). The final sub-sample on which analyses were conducted has a sample size of 723 and is representative of the adult, non-pregnant Inuit population of Nunavik.

2.2. Clinical examination and anthropometric assessment

After the interview session, participants underwent a clinical session, during which weight was measured on a digital scale (Tanita TBF-300) to the nearest 0.1 kg. Height was measured to the nearest centimeter using a rigid square measuring tape with the participant standing barefoot on a hard surface up against a wall and sitting height was assessed using a sitting height table. Anthropometrical measurements were transformed into body mass index (BMI in kg/m²) (Kuczmarski and Flegal, 2000).

Medical history and related medication use (including lipid-lowering drugs) were documented for all participants through a medical chart review.

2.3. Blood sampling, clinical biochemistry and analytical toxicology

Blood samples (60 ml) were collected through a venous catheter from an antecubital vein under fasting conditions (overnight fast, at least 8 h), for the determination of exposure to environmental contaminants as well as for the assessment of clinical biochemistry parameters. Blood was centrifuged within 3 h from collection and plasma was isolated, aliquoted and frozen

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