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Maternal serum concentrations of perfluoroalkyl acids in five international birth cohorts

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ABSTRACT

Background: Perfluoroalkyl acids (PFAAs) are persistent and bioaccumulating compounds, which are spread all over the globe. We aimed to compare the PFAA concentrations in serum from pregnant women in five birth cohorts from four countries (Denmark, China, Norway, and Greenland).
Methods: Serum samples were obtained from the following five birth cohorts including a total of 4718 pregnant women: the Danish National Birth Cohort (DNBC, years 1996–2002, Denmark), the Aarhus Birth Cohort (ABC, years 2008–2013, Denmark), the Shanghai Birth Cohort (SBC, years 2013–2015, China), the Northern Norway Mother-Child Contaminant Cohort (MISA, years 2007–2009, Norway), and the Greenlandic Birth Cohort (ACCEPT, years 2010–2013, Greenland). The samples were analyzed using liquid chromatography triple–quadrupole mass spectrometry. To ensure comparability, all samples except for the MISA samples were measured in the same laboratory. We adjusted the log-transformed PFAA concentrations for age and parity using analysis of covariance.
Results and discussion: The geometric mean (GM) of the summed concentrations of the seven most abundant PFAAs ($\sum$PFAA) was 35 ng/mL in the DNBC, 25 ng/mL in the SBC, 18 ng/mL in the ACCEPT, 12 ng/mL in the MISA cohort, and 12 ng/mL in the ABC. The DNBC concentration was highest presumably because these samples were taken in earlier years (i.e. 1996–2002) than the samples from the other cohorts (i.e. 2007–2015), and at a time when the production of PFAAs were at the highest. When excluding the DNBC samples, we found that the concentrations of all the perfluorinated sulfonic acids (PFSAs) and one of the four perfluorinated carboxylic acids (PFCAs) were highest in the Greenlandic women, whereas the other three PFCAs were highest in the Chinese women.
Conclusion: The concentration and composition of serum PFAAs were similar for the Danish ABC women and the Norwegian MISA women but were otherwise different across the cohorts. The different exposure profiles might partly be related to differences in lifestyle and diet. As the concentrations and compositional patterns vary between the countries, we suggest that the health implications associated with high PFAA exposure might also differ between the countries.

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1. Introduction

Perfluoroalkyl acids (PFAAs) are manmade substances which are used as surfactants in many products such as food paper and impregnation sprays for shoes, clothes, and furniture (Posner et al.,

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2013). They are very persistent and bio-accumulate in animals and humans. The PFAAs are spread around the globe via ocean currents and atmospheric transport (Prevedouros et al., 2006). They are resistant to metabolism, and the human excretion of the long-chain PFAAs (i.e. perfluorinated carboxylic acids [PFCAs] ≥ C8 and perfluorinated sulfonic acids [PFSAs] ≥ C6) is very slow with serum half-lives of 7.7–8.5 years for perfluorohexane sulfonate (PFHxS, C6), 5.4–6.7 years for perfluorooctane sulfonate (PFOS, C8), 2.3–3.5 years for perfluorooctanoic acid (PFOA, C8), and 4.5–12 years for perfluorodecanoic acid (PFDA, C10) (Krafft and Riess, 2015). PFAAs have been found in humans from around the globe including several European (Bjerregaard-Olesen et al., 2016a; Glynn et al., 2012; Nost et al., 2014; Schroter-Kermani et al., 2013; Ingelido et al., 2010; Grandjean et al., 2012; Kannan et al., 2004 Nost et al., 2014; Schroter-Kermani et al., 2013; Ingelido et al., 2010; Grandjean et al., 2012; Kannan et al., 2004), North American (Kannan et al., 2004; Gribble et al., 2015; Kato et al., 2011), South American (Kannan et al., 2004), Asian (Kannan et al., 2004; Okada et al., 2013; Harada et al., 2011; Jin et al., 2007), and Arctic populations (Long et al., 2012; AMAP, 2015).

In the FETOTOX project (http://fetotox.au.dk/), we have found that higher maternal PFAA concentrations were associated with a higher risk for cerebral palsy (Liew et al., 2014). We also found a higher risk for breast cancer in Danish and Greenlandic women with high PFAA serum concentrations (Bonefeld-Jorgensen et al., 2014, 2011). We did not find consistent evidence to suggest that PFAA exposure increases the risk for a longer time to pregnancy in women (Bach et al., 2015a,b), or attention deficit hyperactivity disease (ADHD) (Liew et al., 2015), autism spectrum disorders (ASD) (Liew et al., 2015), or lower birth weight in the newborn (Bach et al., 2016) although high maternal perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) concentrations have been associated with lower birth weight in other studies as recently reviewed (Bach et al., 2014). In addition, our studies on cell cultures showed that the PFAAs might affect the sex hormone receptors (Kjeldsen and Bonefeld-Jorgensen, 2013; Bjerregaard-Olesen et al., 2016b), the aryl hydrocarbon receptor (Long et al., 2013), the thyroid hormone levels (Long et al., 2013), and oxidative stress biomarkers (Wielsoe et al., 2015).

Few studies have measured and compared the PFAA concentrations and compositions in humans from different regions of the world. In the present study, we aimed to compare the PFAA concentrations and compositions in pregnant women from the following five birth cohorts that were a part of the international FETOTOX project (http://fetotox.au.dk/): The Danish National Birth Cohort (DNBC), the Aarhus Birth Cohort (ABC), the Shanghai Birth Cohort (SBC), the Northern Norway Mother-Child Contaminant Cohort (MISA), and the Greenlandic Birth Cohort (ACCEPT).

2. Material and methods

2.1. Participants

We included pregnant women from the Danish DNBC (n = 2139), the Danish ABC (n = 1533), the Chinese SBC (n = 448), the Norwegian MISA (n = 391), and the Greenlandic ACCEPT (n = 207) cohorts. All women provided a written consent that their blood samples and information could be used for research purposes.

2.1.1. The Danish National Birth Cohort (DNBC)

The establishment of the DNBC has been described previously (Olsen et al., 2001). Briefly, pregnant women from around Denmark were invited to participate at the first pregnancy visit with their general practitioner, typically during pregnancy weeks 6–12. Approximately 50% of all general practitioners in Denmark participated in the enrolment, and approximately 60% of the invited women agreed to participate. From the DNBC we selected two sample sets: Firstly, a sub-cohort (the life-style-during-pregnancy cohort (Kesmodel et al., 2010)) which included 1594 pregnant women (DNBC sample set 1). Secondly, we randomly selected 545 women (DNBC sample set 2) who were used as controls in two recent case-control studies investigating associations between PFAAs and cerebral palsy (Liew et al., 2014) and ADHD/ASD (Liew et al., 2015). There was an oversampling of women drinking alcohol during pregnancy (18% never drinkers compared to ~30% in the total DNBC cohort) in sample set 1 and an oversampling of women giving birth to boys (80%) in sample set 2 compared to the DNBC baseline. All 2139 women were included in the DNBC between years 1996 and 2002. The pregnant women gave two blood samples; one at the first pregnancy visit with the general practitioner during the first trimester and another around gestational week 24. When available, we analyzed the samples from the first visit (97%). The EDTA-treated blood samples were transported from the general practitioners to the DNBC biobank via ordinary mail without freezing or cooling during transport. The samples were processed at the biobank, and the serum was stored at −80 °C.

2.1.2. The Aarhus Birth Cohort (ABC)

Women who planned to give birth at Aarhus University Hospital (Denmark) were invited by letter to be included in the ABC. The ABC has been described in detail elsewhere (Mortensen et al., 2013). For the present study we randomly selected 1533 nulliparous pregnant women from 2853 eligible, who gave birth between years 2008 and 2013 as described previously (Bjerregaard-Olesen et al., 2016c). The pregnant women gave a blood sample around gestational week 12. The sample was processed within 2 h and the serum was stored at −80 °C. The PFAA data for the ABC have been used in other studies but with other purposes (Bjerregaard-Olesen et al., 2016a; Bach et al., 2016; Bjerregaard-Olesen et al., 2016c).

2.1.3. The Shanghai Birth Cohort (SBC)

Between year 2013 and 2015, SBC recruited 450 women in Xinhua Hospital in Shanghai who came for their first prenatal care. The following inclusion criteria were set for the pregnant women: (1) They were at least 20 years of age at inclusion, (2) they planned to receive prenatal care and give birth in one of nine participating hospitals, (3) they were registered residents of the Shanghai municipality, and (4) they lived in Shanghai in the past two years before inclusion, and did not plan to move out of the catchment area within the following two years.

At each trimester during pregnancy, a follow-up visit was scheduled. Blood and urine samples were collected at each visit. For the present study, we used serum samples taken around pregnancy week 15. Two samples were lost during PFAA analysis, and thus the final number of included women was 448.

2.1.4. The Northern Norway Mother-Child Contaminant Cohort (MISA)

A total of 2600 pregnant women from Finnmark, Troms, and Nordland in northern Norway were invited to participate in the MISA cohort during the recruitment period from May 2007 to June 2009. The birth cohort and the selection of the 391 women for the present study were described in detail previously (Veyhe et al., 2012). Briefly, 609 women responded to the invitation of which 52 refused further contact, 27 did not give a written consent, 15 did not donate a blood sample, and 124 did not participate in the follow-up at 6 weeks postpartum. The 391 included women gave a blood sample at the time of inclusion using a blood sample collection package. The serum samples were stored at −20 °C. The PFAA data from the Norwegian MISA cohort were recently published in

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