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Preliminary assessment of exposure to persistent organic pollutants among pregnant women in Puerto Rico

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Abstract

Background: Puerto Rico has over 200 hazardous waste sites, as well as higher rates of several adverse health outcomes compared to the mainland US. In response to concerns of potential links between environmental contaminant exposure and preterm birth, the Puerto Rico Testsite for Exploring Contamination Threats (PROTECT) birth cohort was formed. Biomarkers of exposure for several contaminants have been found to be elevated in this cohort compared to women of child-bearing age in the National Health and Nutrition Examination Survey (NHANES). However, exposure to persistent organic pollutants (POPs) has not been evaluated.

Methods: In this preliminary analysis, we measured four classes of POPs, including perfluoroalkyl substances (PFASs), polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyls (PCBs), and several persistent pesticides in serum samples collected at 16–20 weeks gestation from the first 48 women enrolled in PROTECT. We performed descriptive analyses for each analyte, assessed correlations between analytes using Spearman correlations, and compared serum levels in PROTECT women to levels in NHANES women aged 18–40.

Results: Several PFASs were detected in 96–100% of samples, with moderate to strong correlations between most PFASs (range r=0.44–0.88). BDE47, BDE153, PCB 138–158, PCB153 and p,p'-dichlorodiphenyldichloroethene (p,p'-DDE) were detected in the majority of samples, with strong correlations between PCBs and p,p'-DDE (range r=0.59–0.74). The median concentration for each analyte was lower than, sometimes by a factor of 4 (e.g. BDE47, p,p'-DDE), the median concentration reported in NHANES women aged 18–40.

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Conclusion: Although participants in the ongoing PROTECT birth cohort have higher exposure to many environmental contaminants, this preliminary study suggests that they have lower exposure to several POPS, specifically PCBs, OCPs, PFASs, and PBDEs.

Keywords

Puerto Rico; persistent organic pollutants; polychlorinated biphenyls (PCBs); organochlorine pesticides (OCPs); per- and polyfluoroalkyl substances (PFASs); polybrominated diphenyl ethers (PBDEs)

Introduction

Since the 1950s, the island of Puerto Rico has undergone rapid industrialization, resulting in a legacy of toxic spills, chemical waste, air pollution, and contamination of the surrounding environment (Gioda et al. 2007; Hunter and Arbona 1995; Padilla et al. 2011). Currently, Puerto Rico has over 200 hazardous waste sites, including 18 active Superfund sites (USEPA (EPA) 2018). In addition, Puerto Rico has higher rates of several adverse health outcomes, such as preterm birth (Martin et al. 2018), childhood asthma ((CDC) 2017b), autism (Cordero 2013), and obesity (Garza et al. 2011; Otero-Gonzalez and Garcia-Fragoso 2008; Rivera-Soto et al. 2010), compared to the mainland US. In response to concerns of potential links between environmental contaminant exposure and preterm birth, the Puerto Rico Testsite for Exploring Contamination Threats (PROTECT) birth cohort was formed as part of a NIEHS Superfund Research Program. Biomarkers of exposure for several contaminants have been found to be elevated among pregnant women in this cohort, including several phthalates (Cantonwine et al. 2014), phenols (Meeker et al. 2013), certain polycyclic aromatic hydrocarbons (Cathey et al. 2018), and certain metals, compared to female participants in the National Health and Nutrition Examination Survey (NHANES) aged 18–40. Notably, we recently reported that the antibacterial compounds triclosan and triclocarban were significantly higher in PROTECT women compared to NHANES (Ashrap et al. 2018). However, exposure to persistent organic pollutants (POPs), specifically polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), per- and polyfluoroalkyl substances (PFASs), and polybrominated diphenyl ethers (PBDEs), has not previously been evaluated within the PROTECT cohort.

POPs are toxic chemicals that resist degradation, and as a result, can be transported far from their point of release in the environment. POPs also bioaccumulate in wildlife and in humans, with wide-ranging adverse health effects ((EPA) 2017d; Fry and Power 2017). In response to concerns for environmental and human health, over 150 countries have signed the Stockholm Convention on Persistent Organic Pollutants since 2001, indicating agreement to reduce or eliminate the production, use, or release of specific POPs (House 2008; UNEP 2009), including the four classes of compounds discussed here (PCBs, OCPs, PFASs, and PBDEs) and many others. Importantly, the US signed the original Convention, but has not yet ratified the agreement.

PCBs were used as coolants, lubricants, and hydraulic fluids in heat transfer and electrical equipment, as well as plasticizers and dyes, and exposure to PCBs has been associated with cancer, as well as effects on immune function, birth outcomes, endocrine disruption, and

neurodevelopment ((EPA) 2017b). In 1979, production of PCBs was banned as a result of environmental and human health concerns, but PCB exposure continues to be widespread due to their persistence in the environment and presence in contaminated food ((CDC) 2017a; Xue et al. 2014).

OCPs include dichlorodiphenyltrichloroethane (DDT), lindane, dieldrin, hexachlorobenzene, chlordane, mirex, and others, many of which were used to control a range of insects. Human exposure to OCPs has been associated with a variety of adverse health effects, including cancer, neurotoxicity, and reproductive toxicity (Mrema et al. 2013). Again, due to environmental and health concerns they have been largely restricted or banned in the US. As a result, exposure to OCPs has been greatly reduced, but their environment persistence means that exposure to specific OC pesticides continues ((CDC) 2017a). In addition, DDT is still widely used in malaria-endemic areas (Mrema et al. 2013).

PFASs are man-made chemicals that are oil and water resistant and highly stable. Because of these properties, PFASs have been used in numerous consumer products, including stain repellants, textiles, cookware, food packaging, cleaning products, and fire-fighting foams (UEPA (EPA) 2018). Exposure to longer chain PFASs, such as perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS), has been associated with increased risk of cancer, low birth weight, and immune and endocrine system disruption (Bach et al. 2015; DeWitt et al. 2018; Grandjean et al. 2017; Rappazzo et al. 2017; Steenland et al. 2010). Although longer chain PFASs have been phased out of use in the US, use of shorter chain replacement PFASs has increased despite limited information on their toxicity (Birnbaum and Grandjean 2015; Brendel et al. 2018). Due to their persistence in the environment, PFASs have also become emerging drinking water contaminants in several communities in the US and worldwide (Banzhaf et al. 2017; Essumang et al. 2017; Guelfo and Adamson 2018; Hu et al. 2016).

Finally, PBDEs are a class of chemicals that have been widely used as flame retardants in a range of consumer products, including polyurethane foam furniture, textiles, and electronics. In human and experimental studies, PBDE exposure has been associated with cancer, neurotoxicity, reproductive toxicity, and thyroid hormone disruption ((EPA) 2017a). PBDEs were phased out of use in the US between 2004 and 2013, but exposure continues due to continued use of older PBDE-containing products, importation of PBDE-containing products from other countries, and persistence of these chemicals in the environment ((EPA) 2017c).

To our knowledge, no biomonitoring data exists on POP exposure in Puerto Rico. The goal of this preliminary analysis was to characterize exposure to the above chemicals among pregnant women in the PROTECT cohort, as identification of elevated exposures in this population is crucial to our overall research objectives. Specifically, understanding how POPs may contribute to the overall mixture of environmental contaminant exposures will aid our investigation of exposures, preterm birth, and child development.

Methods

Study population

The PROTECT project, which began in 2011, is an ongoing prospective birth cohort study in Northern Puerto Rico designed to investigate relationships between environmental exposures and adverse birth outcomes. The present analysis was conducted on 48 of the first PROTECT participants with sufficient serum sample volume availability, for whom recruitment and inclusion criteria have been previously described (Cantonwine et al. 2014; Meeker et al. 2013). Briefly, women were recruited early in pregnancy (<20 weeks gestation) in 2011 to 2013 from prenatal clinics and hospitals in Northern Puerto Rico, and were eligible to participate if they were between the ages of 18 and 40 years, had a singleton pregnancy, did not use oral contraceptives within three months before pregnancy or in vitro fertilization to get pregnant, and were free of known medical and obstetric complications. At the first study visit (16–20 weeks gestation), women provided a blood sample, urine sample, and demographic information via nurse-administered questionnaires. Although additional biological samples and questionnaire data were collected at subsequent prenatal study visits as previously described (Aker et al. 2016; Johns et al. 2015), because the chemicals of interest are biologically persistent they were measured only in samples collected at the first study visit for this preliminary analysis. The study was approved by the ethics committees of the University of Puerto Rico, Northeastern University, the University of Michigan School of Public Health, and the University of Georgia. All participants provided full informed consent prior to enrollment.

Measurement of persistent organic pollutants

Serum collected from participants at the first prenatal study visit was separated into aliquots, frozen and stored at −80°C, and then shipped on dry ice to laboratories at the Centers for Disease Control and Prevention (CDC) for analysis. Perfluoroalkyl substances (PFASs) were measured using on-line solid phase extraction coupled to isotope dilution-high performance liquid chromatography tandem mass spectrometry (SPE-HPLC-MS/MS) as previously described (Kato et al. 2011). Measured PFASs comprised linear and branched perfluorooctanoate isomers (n-PFOA and Sb-PFOA, respectively), linear and branched isomers of perfluorooctane sulfonate (n-PFOS and Sm-PFOS, respectively), perfluorooctane sulfonamide (FOSA), perfluorohexane sulfonate (PFHxS), perfluorononanoate (PFNA), perfluorodecanoate (PFDeA), 2-(N-ethyl-perfluorooctane sulfonamide) acetate (Et-FOSAA), and 2-(N-methyl-perfluorooctane sulfonamide) acetate (Me-FOSAA). Total PFOS was calculated by summing n-PFOS and Sm-PFOS isomers, while only n-PFOA was used in analyses as Sb-PFOA was detected in only one sample. Polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyls (PCBs), and persistent pesticides were measured using liquid/liquid extraction and gas chromatography isotope dilution high resolution mass spectrometry (GC-IDHRMS) as previously described (Jones et al. 2012). Eleven PBDE congeners were measured (BDE-17, 28, 47, 66, 85, 99, 100, 153, 154, 183, 209) as well as hexabromobiphenyl (PBB-153). Measured PCB congeners comprised PCB-28, 66, 74, 99, 105, 114, 118, 138–158, 146, 153, 156, 157, 167, 170, 178, 180, 183, 187, 189, 194, 196–203, 199, 206, and 209. Persistent pesticides included hexachlorobenzene (HCB), β−hexachlorocyclohexane (β-HCCH), γ-hexachlorobenzene (γ-HCCH, or lindane),

oxychlordane, trans-nonachlor, mirex, p,p'-dichlorodiphenyltrichloroethane (p,p'-DDT), o,p'-DDT, and p,p'-dichlorodiphenyldichloroethene (DDE, a DDT metabolite). Total cholesterol and total triglycerides were measured using commercially available test kits (Roche Diagnostics Corp.; Indianapolis, IN) and a Hitachi 912 chemistry analyzer (Hitachi; Tokyo, Japan). All values below their respective limit of detection (LOD) were replaced with the LOD/√2. Total PBDE, PCB, and pesticide concentrations were adjusted for serum total lipid concentration and reported in ng/g lipid weight.

Statistical Analysis

Descriptive analyses were performed for each analyte, including determination of the percent of serum samples with concentrations below the LOD for each chemical, means, and distribution percentiles, as well as Spearman correlations between chemicals detected in at least 50% of samples.

Results

The LODs, means, and distributions of all analytes detected in greater than 50% of serum samples are shown in Table 1. Several PFASs were highly detected, including the linear PFOA isomer, linear and branched PFOS isomers, PFNA, and PFHxS (96–100% above LOD). BDE47 and BDE153 were the only PBDEs detected in the majority of samples, while PCB 138–158 and PCB153 were the only commonly detected PCBs. Of the persistent pesticides that were measured, only p,p'-DDE was detected in the majority of samples. LODs and the number and percent of samples above the LOD for all other analytes are presented in Table 2.

The median concentration for each analyte was lower than, sometimes by a factor of 4, the median concentration reported in women aged 18–40 years participating in the National Health and Nutrition Examination Survey (NHANES) (Table 1). For example, the median BDE47 level in PROTECT was 4.7 ng/g lipid, while the median among women of child-bearing age in NHANES was 22.8 ng/g lipid. With the exception of PFOA, this trend was also evident, or even more pronounced, at the 95th percentile for many analytes. Specifically, the 95th percentile for p,p'-DDE among PROTECT women was 56 ng/g lipid but more than an order of magnitude higher in NHANES at 850 ng/g lipid. However, PBDEs, PCBs, and the pesticides measured in the current study have not been reported in NHANES since 2003–2004, 7 to 10 years before our samples were collected. On the other hand, NHANES PFAS levels were measured in samples collected during the same time frame as PROTECT samples.

Serum concentrations of analytes within each chemical group were significantly correlated (Table 2). For example, Spearman correlation coefficients for PFASs ranged from 0.44 to 0.88 ($p<0.05$ for all), and PCB138/158 and PCB153 were strongly correlated ($r=0.74$, $p<0.05$). p,p'-DDE was correlated with several analytes from other chemical groups, with significant correlation coefficients ranging from 0.29 to 0.38 for PFASs, 0.59 to 0.72 for PCBs, and 0.41 for BDE47. In addition, there were significant weak correlations between certain PCBs and PFASs: $r=0.29$ for PFDeA and PCB138/158; $r=0.33$ for PFHxS and PCB153.

Discussion

In this preliminary analysis, we characterized distributions of four classes of persistent organic pollutants among pregnant women in an ongoing birth cohort in Puerto Rico. Although this cohort has elevated exposure to several environmental contaminants, we found that serum concentrations of PCBs, OCPs, PFASs, and PBDEs were lower compared to levels among female NHANES participants aged 18–40 years. However, many of the chemicals discussed here have not been measured in NHANES since 2004 to 2010, and population levels have been shown to be declining (Olsen et al. 2012; Parry et al. 2018; Sjodin et al. 2004; Zota et al. 2013). As a result, we were not able to make direct comparisons to current concentrations among women in the US, with the exception of PFAS. Regardless, our findings suggest that our continuing research efforts should focus on contaminants known to be elevated in this population.

In comparison with other birth cohorts in the US, geometric mean concentrations of BDE47 and p,p'-DDE were higher among women from the Center for Health Assessment of Mothers and Children of Salinas (CHAMACOS) cohort in 2011 (post pregnancy), compared to median concentrations among pregnant PROTECT women, while concentrations of other analytes were similar (Sjodin et al. 2018). Pregnant women participating in the Health Outcomes and Measures of the Environment (HOME) Study in Cincinnati, OH from 2003–2006 had higher levels of several PFASs, BDE47, and BDE153 compared to PROTECT women (Braun et al. 2016; Vuong et al. 2015), although PROTECT samples were collected 5–10 years later. These comparisons are consistent with the reported NHANES comparisons and suggest that PROTECT women are not highly exposed to a number of POPs. In addition, the observed correlations of analytes within the same chemical group are as expected based on similar sources of exposure and metabolism within groups, and are consistent with previous reports (Bramwell et al. 2016; Gladen et al. 2003; Yorita Christensen and White 2011).

Given the numerous hazardous waste sites in northern Puerto Rico, it is unclear why PROTECT participants, and possibly the region's general population, have relatively low serum levels of several POPs, suggesting lower exposure. Interestingly, PCB levels were very low, despite reports of extensive PCB contamination in other regions of Puerto Rico (Alegria et al. 2016; Kumar et al. 2016). This suggests that PCB contamination may be variable around the island, and potentially lower in the areas where PROTECT women live in northern Puerto Rico. Given the tropical climate, it is also surprising that OCP levels were mostly below detection, suggesting that usage of other pesticide classes is likely. Another explanation for low serum PCB and OCP concentrations in this cohort is the relatively young age of PROTECT participants, with a mean age of 27, as many were not yet born when these chemicals were phased out of use in the 1970s. Regarding PFASs and PBDEs, it is possible that consumer products utilizing these chemicals were not widely distributed in Puerto Rico, despite their apparent ubiquity.

This exploratory study has several limitations, most notably the small sample size. As a result, our ability to detect statistically significant associations was limited, and we may have missed individuals or areas with higher exposure. With the exception of PFASs, the limited

volume of serum available for analyses led to relatively high limits of detection for many compounds, which may have affected our findings. In addition, blood samples were collected prior to the 2017 hurricane season, during which Hurricanes Irma and Maria likely spread existing environmental contamination on the Island, potentially leading to higher post-hurricane exposure.

Conclusion

Although participants in the ongoing PROTECT birth cohort have higher exposure to many environmental contaminants, this preliminary study suggests that they may have lower exposure to several PCBs, OCPs, PFASs, and PBDEs. Therefore, our continuing work in with this population will likely prioritize and focus on contaminants to which they have demonstrated increased exposure.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Abbreviations

AAA β-HCCH	Autoridad de Acueductos y Alcantarillados hexachlorocyclohexane
DDT	dichlorodiphenyltrichloroethane
DDE	dichlorodiphenyldichloroethene
Et-FOSAA	2-(N-ethyl-perfluorooctane sulfonamide) acetate
GC-IDHRMS	gas chromatography isotope dilution high resolution mass spectrometry
γ-HCCH	γ-hexachlorobenzene
HCB	hexachlorobenzene
LOD	limit of detection
Me-FOSAA	2-(N-methyl-perfluorooctane sulfonamide) acetate
NHANES	National Health and Nutrition Examination Survey
OCP	organochlorine pesticide
PBDE	polybrominated diphenyl ether

PCB	polychlorinated biphenyl
PFAS	per- and polyfluoroalkyl substance
PFDeA	perfluorodecanoate
PFHxS	perfluorohexane sulfonate
PFNA	perfluorononanoate
PFOA	perfluorooctanoic acid
PFOS	perfluorooctane sulfonate
FOSA	perfluorooctane sulfonamide
POP	persistent organic pollutant
PROTECT	Puerto Rico Testsite for Exploring Contamination Threats
SPE-HPLC-MS/MS	solid phase extraction, isotope dilution-high performance liquid chromatography tandem mass spectrometry

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Table 1.

Distributions of persistent organic pollutant concentrations in serum samples collected from 2011–2013 from a subset of PROTECT participants (n=48) and NHANES women aged 18–40 years.

					Percentiles							
Analyte	**LOD**[a]	**% >LOD**	**Mean**	**Min**	**25th**	**50th**	**75th**	**95th**	**NHANES 50th women 18–40**	**NHANES 95th women 18–40**	**NHANES n**	**NHANES cycle**
PFASs (μg/L)												
n-PFOA	0.1	100	1.16	0.20	0.60	0.90	1.13	3.10	1.10	2.90	375	2013–2014
n-PFOS	0.1	100	1.66	0.20	0.78	1.50	2.33	3.30	2.30	9.00	375	2013–2014
PFDeA	0.1	64.6	0.17	0.07	0.07	0.10	0.20	0.60	0.20	0.70	760	2009–2010
PFHxS	0.1	95.8	0.34	0.07	0.20	0.20	0.30	0.90	0.70	3.25	760	2009–2010
PFNA	0.1	97.9	0.42	0.07	0.20	0.30	0.50	1.00	0.70	2.03	760	2009–2010
Sm-PFOS	0.1	97.9	0.61	0.07	0.30	0.55	0.93	1.20	0.60	2.00	375	2013–2014
PBDEs (ng/g lipid)												
BDE153	2.3	70.8	5.55	1.27	2.08	3.87	6.57	12.8	4.90	36.6	363	2003–2004
BDE47	3.6	68.8	7.50	1.56	3.26	4.66	7.94	21.8	22.8	127	361	2003–2004
PCBs (ng/g lipid)												
PCB138–158	2.3	56.3	2.84	1.34	1.84	2.33	3.31	5.96	6.67	27.5	343	2003–2004
PCB153	2.3	77.1	3.64	1.41	2.23	3.15	3.82	9.12	8.23	31.7	342	2003–2004
Pesticides (ng/g lipid)												
p,p’-DDE	11.5	95.8	32.3	8.49	21.1	30.9	40.6	55.5	135	850	345	2003–2004

[a] median LOD; BDE153, PCB138–158, PCB153 range=1.4–3.3 ng/g lipid; BDE47 range=2.1–5.2 ng/g lipid; p,p’-DDE range=6.8–17 ng/g lipid.

Table 2.

Spearman correlations between chemicals (n=48)

	BDE47	BDE153	PFOA	PFOS	PFHxS	PFNA	PFDeA	SmPFOS	PCB138/158	PCB153	DDE
BDE47	1	0.35 *	0.13	0	0.01	0.08	−0.03	0.08	0.14	0.18	0.41 *
BDE153		1	−0.09	0.13	0.08	−0.07	−0.12	0.1	0.18	0.2	0.21
PFOA			1	0.44 *	0.55 **	0.71 **	0.63 **	0.57 **	0.08	0.06	0.15
PFOS				1	0.75 **	0.75 **	0.6 **	0.88 **	0.06	0.21	0.22
PFHxS					1	0.69 **	0.51 **	0.82 **	0.19	0.33 *	0.38 *
PFNA						1	0.85 **	0.7 **	0.14	0.26	0.29 *
PFDeA							1	0.56 **	0.29 *	0.28	0.23
SmPFOS								1	0.1	0.26	0.36 *
PCB138/158									1	0.74 *	0.59 *
PCB153										1	0.72 *
DDE											1

* p<0.05

** p<0.001