Environmental Research 133 (2014) 211–219

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

Prenatal exposure to persistent organic pollutants and polybrominated diphenyl ethers in 10 Caribbean countries

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ARTICLE INFO

Article history:
Received 28 February 2014
Received in revised form
6 May 2014
Accepted 20 May 2014
Available online 24 June 2014

Keywords:
Caribbean
Dioxins
Persistent organic pollutants
Polybrominated diphenyl ethers
Prenatal exposure

ABSTRACT

Prenatal exposures to legacy persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs) and dioxin-like compounds (DLC), as well as polybrominated diphenyl ethers (PBDE), were analyzed in pregnant women from 10 Caribbean countries. A total of 438 samples were collected and descriptive statistics calculated and compared to comparable Canadian Health Measure Survey (CHMS) and U.S. National Health and Nutritional Examination Survey (NHANES) datasets. Maternal POPs blood concentrations were found to be generally relatively low in the Caribbean samples compared with the U.S. and Canada datasets. Evidence of exposure to DLC and PBDE was established. DLC levels ranged from a geometric mean low of 3.96 pg/g lipids in Antigua and Barbuda to a high of 11.4 pg/g lipids in St. Lucia. Several of the PBDEs (15, 17, 25, 28, 33, 100) were detected in less than 60% of the country's samples. For PBDE-47, significantly higher levels were found in Bermuda, while Jamaica recorded a significantly low level. The overall calculated concentration of PBDE-47 for the Caribbean (5.33 μg/kg lipids) was significantly lower than the concentrations measured for the U.S. (10.83 μg/kg lipids) and Canada (23.90 μg/kg lipids). This study confirms that prenatal expose to multiple environmental chemicals is taking place in the Caribbean and highlights the need to implement surveillance programs that continuously monitor, intervene, and evaluate the levels of these toxic environmental contaminants to ensure that they are reduced as much as possible and that the health risk to humans, in particular the unborn child, are minimized.

1. Introduction

Concerns regarding the adverse health effects of prenatal exposures to environmental chemicals, especially those classed as persistent organic pollutants (POPs) and dioxin-like chemicals (DLCs), have been extensively examined and documented (Gascon et al., 2013; Mrema et al., 2013; Needham et al., 2005; Schell et al., 2006; Thundiyil et al., 2007; Wong et al., 2012). Given the danger these classes of chemicals poses to the environment and human health, the international community crafted the global Stockholm Convention on Persistent Organic Pollutants treaty which was adopted in 2001 and became effective in 2004 with the aim to eliminate or restrict the use of these classes of chemicals (UNDP, 2011). Given, however, that POPs by their chemical nature can persist for many decades in common environmental mediums such as air, water and soil, their potential to adversely affect human health can extend well beyond ban dates. All of these environmental toxicants have been found widespread in the environment with resultant human exposure and uptake confirmed (Jacobson and Jacobson, 1996; Rogan et al., 1986).

Widespread exposure to polybrominated diphenyl ethers (PBDEs) and human uptake has also been established (Frederiksen et al., 2009). Based on animal studies, PBDEs have been shown to cause endocrine disruption and neurodevelopmental problems (Fernie et al., 2005; Tomy et al., 2004). In a study of the effect of PBDE on pregnant women's thyroid function, results suggested that exposure to PBDEs is associated with thyroid dysfunction with potential implications for maternal health and fetal development (Chevrier et al., 2010).

Maternal blood levels of environmental chemicals have been found to be highly correlated with levels measured in the umbilical cord (Needham et al., 2011). Further, it has been found

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http://dx.doi.org/10.1016/j.envres.2014.05.021
0013-9351/

that the developing fetus is especially susceptible to the uptake of environmental chemicals since these toxicants readily cross the placenta from mother to baby (Barr et al., 2007).

Throughout the Caribbean region, POPs have been historically extensively used in agriculture and vector control programs as well as for other purposes. A UNEP (2002) survey of POPs pesticides usage in the Caribbean region found that while most countries have formally banned the use of the so-called legacy POPs pesticides, several Caribbean countries have acknowledged past use of legacy POPs such as dieldrin, DDT, toxaphene, and aldrin. These chemicals were used extensively to reduce insect pest populations, control vector borne diseases such as malaria and dengue, and in the agriculture sector, to reduce loss of crops and production as a result of various plant pathogens.

There is considerable evidence that POPs and DLCs circulate globally and their presence has been detected in humans from biological samples taken in different parts of the world (De Felip and Ingelido, 2004; Liberda et al., 2013; Van Oostdam et al., 2004). Further, because these chemicals are very persistent in the enviornment, biota, and humans, their concentrations will decrease only very slowly in populations that have been exposed over many years. Significant concentrations can persist in the environment that may cause local foods to be contaminated for long periods of time (Liberda et al., 2013). It is thus quite likely that people in the Caribbean have been exposed to these chemicals.

Biomonitoring studies done in North America and elsewhere have found nearly ubiquitous exposure to many chemicals (CDC, 2009; CHMS, 2010). Similar studies of the concentration levels of these chemicals have not, however, been systematically researched and documented in the Caribbean. As part of a Canadian Global Health Research Initiative's (GHRI) Teasdale-Corti Grant Programme funded research initiative, the Caribbean EcoHealth Programme (CEHP) launched a study entitled "Prenatal exposures to Persistent Organic Pollutants (POPs), heavy metals, and zoonoses" whose focus was to determine the level of prenatal exposures to persistent organic pollutants, other commonly used classes of pesticides, two heavy metals mercury and lead, and zoonotic infections (Forde et al., 2011). This study was the first to undertake a systematic examination geared at determining levels of human (as opposed to environmental) exposures to the above listed toxicants. This paper reports on the findings of the biomonitoring study for POPs and PBDE chemical contaminants for pregnant women who live in the 10 Caribbean countries where this research study was successfully executed.

2. Materials and methods

2.1. Ethics and governmental approvals

Ethics approval to conduct this study was first sought and obtained from the institutions of the two principle investigators of this study (Laval University, Canada, and St. George's University, Grenada). In addition to academic institutional ethics approvals, ethics approvals were also sought from each of the 10 Caribbean countries where this study was executed. In cases where a Caribbean country lacked a local ethics or institutional review board, other mechanisms were put in place such as asking that the Ministry of Health put together a committee to review this study and provide written approval before any samples were taken. Furthermore, in addition to institutional and country ethics approvals, governmental approval, typically through the Ministry of Health within each country, was also sought and obtained before commencement of the study.

2.2. Study protocols

Once ethical and governmental approvals were secured, local nurses and laboratory technicians were identified within each country with the assistance of the Ministry of Health and trained to recruit pregnant women to participate in this study, obtain their informed consent, and collect the samples in their respective countries. The locally trained laboratory technicians then processed all samples collected by the nurses according to a standardized protocol. Samples were initially stored at the main hospital located on each island and then prepared for shipment in this study's Atlantis Mobile Laboratory (AML). For shipping, the samples were packed by International Air Transport Association (IATA) certified technicians and then shipped in IATA certified boxes packed with dry ice to the CEHP's mobile Atlantis Mobile Laboratory (AML) facilities and the Laboratoire de Toxicologie of the Institut national de santé publique du Québec (INSPQ) located in Quebec City, Canada for analysis.

2.3. Study population and sampling

The recruitment and sampling procedure used in this study was based on the highly successful implementation of the Arctic Monitoring and Assessment Programme (AMAP, www.amap.no) (Van Oostdam et al., 2004). Following this protocol, pregnant and delivering women of ≥ 18 years coming to the main hospital or health clinics to receive prenatal care or deliver were invited to participate in this study by local nurses. Most samples were taken before delivery, however, in some cases these were taken after delivery. As in the AMAP protocol, the Caribbean prenatal exposures research protocol sets a goal of 50 mothers ≥ 18 years for each country.

2.4. Chemical analyses

Two laboratories were used in this study – a mobile laboratory facility called the Atlantis Mobile Laboratory (AML) and the Laboratoire de Toxicologie of the Institut national de santé publique du Québec (INSPQ) located in Quebec City, Canada. The AML was used to collect and prepare the samples for shipping to the INSPQ laboratory. The INSPQ laboratory is a certified laboratory compliant to ISO-CEI 17025, accredited since 2000. It is noted that the INSPQ laboratory is the reference laboratory for human toxicology in the Province of Quebec, Canada, and this laboratory participated in the Canadian quality assurance/quality control (QA/QC) program of the Canadian Northern Contaminants Program. The INSPQ laboratory also set up the external quality assessment scheme for the AMAP ring test for POPs in human serum. Additionally, the INSPQ laboratory was the same laboratory used to conduct the Canadian Health Measure Survey (CHMS) done in Canada.

Each maternal blood sample was analyzed for 35 organochlorine POPs, one DLCs, and 10 organobromine polybrominated diphenyl ethers (PBDE) chemical analytes (Table 1).

2.5. Analytical methods for POPs and PBDE

For all 10 participating Caribbean countries, blood serum samples (10 ml) were collected in vials containing EDTA, centrifuged (10 min, 5000 rpm), and then transferred to glass vials pre-washed with hexane. For the POPs analysis, samples were thawed overnight at 4 °C and a 2-ml aliquot was spiked with internal labeled standards and proteins denaturated with 2 ml formic acid and 2 ml of water. The extraction and purification steps of the procedure were conducted on a Rapid Trace Automated solid phase extraction (SPE) workstation (Caliper Life Science, Hopkinton, MA, USA). First, serum was extracted on an Oasis HLB SPE column as described by Sandau et al. (2003). The extracts were evaporated to dryness before they were dissolved in 0.5 ml of hexane. They were subsequently purified through a column containing 1 g activated Florisil (Thermo Fisher Scientific, Waltham, MA, USA). The fraction containing the PCBs and PBDEs was eluted using a mix of dichloromethane:hexane (1:3; 9 ml). A second fraction containing more polar organochlorines like dieldrin, endrin, and endosulfane was eluted using a mix of acetone: dichloromethane (2:98; 6 ml).

The first and second Florisil fractions were evaporated, taken up in 20 μL of hexane and analyzed by a gas-chromatography/mass spectrometry (GC–MS) system. The GC–MS system used was an Agilent 6890 Network gas chromatograph (Wilmington, DE, USA) equipped with an Agilent 7683 series automatic injector and an Agilent 5973 Network mass spectrometer. The GC was fitted with an Agilent 60 m DB-XLB column (0.25 mm i.d, 0.25 μm film thickness). The carrier gas was helium and the injection was 3 μL in splitless mode for the first fraction and 2 μL in splitless mode for the second fraction. The mass spectrometer was operated in selected ion monitoring (SIM), using negative chemical ionization (NCI) with methane (99.97%) as the reagent gas.

The third fraction containing polar metabolites of PCBs and halogenated phenolic compounds was evaporated, derivatized with diazomethane and cleaned up on an activated silica/acidic silica column. This fraction was evaporated, taken up in 20 μL of hexane being analyzed with the same GC–MS system described above.

2.6. Analytical methods for dioxin-like chemicals (DLCs).

In order to obtain a global measure of dioxin-like activity in serum that interacts with this signaling pathway, a luciferase reporter gene assay was used to