Chemosphere 176 (2017) 266–272

Contents lists available at ScienceDirect

Chemosphere

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

Levels and profiles of organohalogenated contaminants in human blood from Egypt

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HIGHLIGHTS

- PBDEs, PCBs and OCPs were determined in 67 human blood samples from Egypt.
- No significant differences between colorectal cancer patients and control group.
- ΣDDT and ΣOCPs increased significantly with age and BMI.
- PBDEs and PCBs in serum of Egyptians are among the lowest worldwide.

ARTICLE INFO

Article history:
Received 19 September 2016
Received in revised form
4 February 2017
Accepted 27 February 2017
Available online 1 March 2017

Handling Editor: Andreas Sjodin

Keywords:
Organohalogen contaminants
OCPs
PBDEs
PCBs
Egyptian blood
Colorectal cancer

ABSTRACT

Concentrations of polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyls (PCBs) and some organochlorine pesticides (OCPs) were determined in serum of Egyptian colorectal cancer patients (n = 35) and compared to a healthy control group (n = 32). p,p′-DDE (the major metabolite of DDT) was the most frequently detected contaminant with the highest concentration (median = 131 ng/g lw) in all studied serum samples. BDE-209 was the least frequently detected contaminant with a median concentration <0.3 ng/g lw. The contamination profile in patients and controls was almost identical with p,p′-DDE showing the highest median contribution (77%) and oxychlordane the lowest (1%). The low p,p′-DDT/p,p′-DDE ratio (3.7%) in serum implies bioaccumulation and past exposure to DDT (c.f. recent and ongoing intake). Statistical analysis revealed no significant differences ($P > 0.05$) between the levels of target contaminants in serum of patients and the control group. Gender, age and body mass index (BMI) were investigated as potential factors influencing serum contaminant levels. ΣDDT, hexachlorobenzene and pentachlorophenol concentrations showed significant positive associations with age and/or BMI of the participants. Comparison with other countries revealed concentrations of PBDEs and PCBs in Egyptian serum among the lowest reported worldwide.

1. Introduction

Organohalogen contaminants (OHCs) are a diverse group of chemicals including organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs). Despite their various applications, these compounds are highly lipophilic, persistent in the environment and capable of bioaccumulation and biomagnification along the food chain (UNEP, 2009; Xu et al., 2013). Moreover, numerous toxicological studies have associated different OHCs with adverse health effects including endocrine disruption, reproductive, developmental and neurological toxicity, type 2 diabetes and different types of cancer (Pi et al., 2016; Kuo et al., 2013; Gascon et al., 2013; Darnerud, 2003). Combined with their ubiquitous distribution in almost all biotic and abiotic environmental compartments, these features have led to global concern over the production and use of OHCs. As a result, most OCPs, PCBS and PBDEs are currently listed as persistent organic pollutants (POPs) under the UNEP Stockholm

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http://dx.doi.org/10.1016/j.chemosphere.2017.02.139
0045-6535/

Convention (UNEP, 2009).

While the regulatory actions have led to levelling off and even slight decline of the environmental concentrations of various OHCs in different regions, human exposure to these contaminants still occurs via direct and indirect pathways (Law et al., 2014). Several authors have documented the association between certain types of food (e.g. fish) and elevated serum levels of OCPs and PCBs (Bjermo et al., 2013; Rylander et al., 2012), while others established significant positive correlations between PBDEs in indoor dust and their serum levels (Whitehead et al., 2015). Recently, dermal uptake has been highlighted as a potential pathway of human exposure to PCBs and PBDEs (Abdallah et al., 2015).

Due to their relevance for public health and associations with various diseases, several countries have collected extensive data on the occurrence and levels of OHCs in their population (Aylward et al., 2014; Rawn et al., 2012, 2014; Thomas et al., 2006). However, very little is known about the concentrations of OHCs in the blood of the Egyptian population. Furthermore, epidemiological studies have raised concerns over the increased incidence of colorectal cancer among the Egyptian population, especially that patients under age 40 constitute 35.6% of all colorectal cancer cases in Egypt (Gado et al., 2014; Veruttipong et al., 2012). Few authors have investigated genetic, dietary and life-style factors as potential determinants for the increased incidence of colorectal cancer in Egypt. Abdel-Rahman et al. reported an association between polymorphisms in the gene for the DNA repair enzyme XRCC1 with increased risk of colorectal cancer among Egyptians. Interestingly, an association between the studied polymorphisms and early age of disease onset was observed (Abdel-Rahman et al., 2000). An epidemiological study of cancer of all organ sites in Gharbiah Province of Egypt from 1999 to 2002 revealed higher urban than rural incidences among both men and women for colorectal cancer (Dey et al., 2011). This was linked to dietary preferences of the urban population with higher contribution of meat and fat as opposed to vegetables. Dietary risk factors were further highlighted by the results of another study from Minia governorate, where the most significant dietary and lifestyle colorectal cancer risk factors were higher consumption of red meat, preserved food, artificial sweeteners and fast foods (Mahfouz et al., 2014). However, the relationship between exposure to chemical environmental contaminants and colorectal cancer in Egypt is yet to be fully explored. This may be partly attributed to the lack of information on the levels of different OHCs in the Egyptian population. Therefore, the aims of the current study are: (a) to provide first insights into the occurrence and levels of PBDEs, PCBs in addition to some OCPs in serum of Egyptian colorectal cancer patients, (b) to compare the levels of target OHCs in serum of patients to an age- and sex-matched control group, (c) to investigate possible correlations between the levels of OHCs in patients and their age, sex or body mass index (BMI) and finally (d) to compare the levels of target OHCs in the Egyptian serum with those reported from other countries.

2. Materials and methods

2.1. Target compounds

The OHCs investigated in this study include:

(a) Polychlorinated biphenyls (PCBs) no. 118, 138, 153, 170, 180.
(b) Polybrominated diphenyl ethers (PBDEs) no. 47, 99, 100, 153, 154, 183, 209
(c) Organochlorine pesticides, namely: pentachlorophenol (PCP), hexachlorobenzene (HCB), oxyhchlordane (OxC), 4,4′-dichlorodiphenyldichloroethylene (p,p′-DDE) and 4,4′-dichlorodiphenyltrichloroethane (p,p′-DDT).

2.2. Reagents and chemicals

Solvents and chemicals used for analysis were of HPLC grade (Sigma-Aldrich, St. Louis, MO 63103 USA). Oasis® HLB SPE cartridges (6 mL/500 mg, 30 μm) were obtained from Waters (Milford, MA 01757 USA), while empty polypropylene cartridges (3 mL) and corresponding frits were purchased from Supelco (Bellefonte, PA, USA). Individual standards of OCPs and PCBs were obtained from Dr.Ehrenstorfer Laboratories (Augsburg, Germany), while PBDEs standards were purchased from Wellington Laboratories (Guelph, ON, Canada).

2.3. Sample collection

Blood samples (5 mL, n = 35) were collected from colorectal cancer patients admitted to the South Egypt Cancer Institute (SECI) between October and December 2012, while control samples (n = 32) were obtained from the Upper Egypt Blood Bank from healthy donners at the same period. Informed consent was obtained from each participant following approval of the research protocol by the SECI ethics committee. Selection criteria included Egyptian nationality, living in the Upper Egypt region for the last 8 years and lack of occupational exposure to any of the studied chemicals. Exclusion criteria were family history of colorectal cancer, patients with a primary cancer other than colorectal cancer and severely ill cases. The controls were healthy, unrelated to the patients, from the same geographical area and with no family history of colorectal cancer. Demographic information on the participants' age, sex and BMI are provided in Table SI-1.

The serum was separated by centrifugation, transferred to clean tubes and kept frozen at −20 °C until analysis. Cholesterol and triglycerides were determined at the collection clinic using routine laboratory analysis then the value for total lipids (TL) was estimated according to the method of Phillips et al. (1989).

2.4. Sample extraction and clean-up

Sample extraction and clean-up were performed according to a previously reported method (Ali et al., 2013). Briefly, thawed and homogenized serum samples (~3 mL) were spiked with internal standards mixture (BDE-77, BDE-128, ^{13}C-BDE-209, PCB 143, ε-HCH), then mixed with 2 mL of milliQ-H_2O and 50 μL of formic acid prior to ultrasonication for 20 min. The equilibriated samples were loaded onto pre-conditioned Oasis HLB cartridges, rinsed with 4 mL of milliQ-H_2O and dried under vacuum. Target analytes were eluted with 10 mL of DCM:MeOH (4:1, v/v) and 2.5 mL of hexane. The eluate was dried under a gentle stream of N_2 and resolubilized in 500 μL of hexane:DCM (9:1, v/v).

The crude extract was cleaned-up over 800 mg of pre-washed silica topped with 100 mg of 10% acid silica packed in 3 mL polypropylene SPE cartridges. PCBs, PBDEs and DDTs were eluted first by 8 mL of hexane, followed by PCP in 10 mL of DCM. The 1st fraction was evaporated and redissolved in 80 μL of isooctane containing 100 pg/μL of PCB129 and ^{13}C-BDE-100 used as recovery determination (syringe) standards (RDS) for quality assurance/quality control (QA/QC) purposes. The 2nd fraction was derivatized by methylation and finally redissolved in 80 μL of iso-octane containing 100 pg/μL of PCB129 used as RDS. Further details are provided in the SI section.

2.5. Gas chromatography/mass spectrometry (GC/MS) analysis

Instrumental analysis of target analytes was performed using a FOCUS GC coupled to a DSQII mass spectrometer (Thermo Fisher Scientific, Austin, TX, USA). Separation of target analytes was