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Is the fact of parenting couples cohabitation affecting the serum levels of persistent organohalogen pollutants?

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ABSTRACT

Organohalogen compounds constitute one of the important groups of persistent organic pollutants (POPs). Among them, due to their long-term health effects, one should pay attention on organochlorine pesticides, polychlorinated biphenyls (PCBs) and perfluoroalkylated substances (PFASs). This paper is an attempt to answer the question about relation between the fact of cohabitation by couples expecting a child and the level of the organohalogen compounds in the blood serum of both parents. The study was done on a population of parent couples from Greenland, Poland and Ukraine, from whom blood samples were collected in order to establish the levels of marker organohalogen compounds. We selected, as the representative of these compounds, the most persistent metabolite of DDT, i.e. *p,p′*-DDE, the most frequently detected PCB congener – CB-153, and PFOS and PFOA as the representatives of PFASs. The results show that in case of all compounds under study the highest concentrations were present always in men in relation to the levels detected in the blood serum of their female partners, regardless of the country of origin of the couple. A positive correlation was noted between the concentrations of the studied compounds in the blood serum of men and women in parenting couples. In some cases these correlations were statistically significant, e.g. for concentrations of *p,p′*-DDE in pairs from Greenland and Ukraine, of CB-153 in pairs from Poland and Ukraine, and of PFOS for parents from Greenland and Poland, while for PFOA – only for couples from Greenland. The concentrations of the compounds included in the study were similar to the levels found in general population in other countries. Our results show that the exposure to POPs resulting from cohabitation plays a role in the general exposure to these compounds.

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Abbreviations: BMI, Body Mass Index; DDT, 1,1,1-trichloro-2,2-bis-(p-chlorophenyl) ethane; EFSA, European Food Safety Authority; HCH, hexachlorocyclohexane; ICC, intraclass correlation coefficient; LC, liquid chromatography; LC/MS/MS, liquid chromatography tandem mass spectrometry; LOD, limit of detection; OCs, organochlorine compounds; PCBs, polychlorinated biphenyls; *p,p′*-DDE, 1,1-dichloro-2,2-bis-(p-chlorophenyl) ethylene; PFASs, perfluoroalkylated substances; PFOA, perfluorooctanoic acid; PFOS, perfluorooctane sulfonate; POPs, persistent organic pollutants; SPE, solid phase extraction; TTP, time to pregnancy.

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Introduction

Organohalen compounds, being persistent organic pollutants (POPs), are a subject of keen interest of scientists, especially in the domain of chemical safety. Organohalen POPs include organochlorine compounds, such as polychlorinated biphenyls (PCBs) or DDT and its metabolites, as well as perfluoroalkylated substances (PFASs). Due to their similar physical and chemical properties, similar interaction with various elements of the environment, including the human organism, as well as their prevalence in the environment, they are a subject of environmental studies, monitoring of human exposure to food and air contaminants and studies of direct impact on the human health at various stages of development (Barr et al., 2007; Darnerud et al., 2006; Hernik et al., 2014; Góralczyk et al., 2010; Kucharska et al., 2011; Struciński et al., 2000, 2006; Tan et al., 2009).

Organochlorine compounds (OCs) found broad application as pesticides in crop protection. The most widely known organochlorine insecticides were: DDT and its metabolites, hexachlorocyclohexane (HCH), chlordane, endosulfan, metoxychlor, heptachlor and cyclodien pesticides – aldrin, dieldrin, endrin (McKinlay et al., 2008); their presence is still confirmed in biological monitoring (Czaja et al., 2006; Fernandez et al., 2008; Góralczyk et al., 2010; Hernik et al., 2014; Knutsen et al., 2011; Pulkrabová et al., 2009). Next to the above-mentioned pesticides, one should also list polychlorinated biphenyls (PCBs) which were widely used as dielectrics in large-capacity transformers and condensers, heat exchangers, hydraulic systems, as a part of lubricating oils and cooling/lubricating liquids, plasticizers for paints, inks, printing inks and copy paper, as flame retardants for plastics, as well as pesticide vectors (Ziets et al., 2008). Humans are exposed to these compounds throughout their whole life. The main exposure pathways are food and air. The share of these routes in the general exposure profile differs depending on the physical and chemical properties of the compound. Around 90% of organochlorine compounds (pesticides, PCBs) are taken in by humans with food, especially of animal origin, of which 70–80% of e.g. PCBs intake comes from fish and seafood, despite the withdrawal of these compounds from the market in Europe many years ago. Scandinavian research showed that daily intake of PCBs and organochlorine pesticides in food is respectively 615 and 523 ng (Darnerud et al., 2006). Due to their lipophilicity, these compounds may accumulate in human tissues. For example, in research conducted in the Czech Republic (Pulkrabová et al., 2009), samples of adipose tissue contained an average ΣDDT 615.6 ng g^{-1} lipid with a dominant *p,p′*-DDE metabolite, which confirms the distant and long-term exposure to this pesticide. Similar concentrations were found by Struciński et al. (2002) in the tissue of the mammary gland of women from Poland. This caused the concentration of *p,p′*-DDE to be accepted in numerous studies as the indicator of the exposure of humans to organochlorine pesticides (Crisp et al., 1998; Jönsson et al., 2004). In case of ΣPCBs, Fernandez et al. (2008) quantified the average concentration of seven indicator congeners of PCBs (CB-28, 52, 101, 118, 138, 153 and 180) in human adipose tissue to 625.5 ng g^{-1} lipid. The total concentration of these seven indicator congeners has been accepted in Europe as the marker of PCBs dietary exposure (Wingfors et al., 2006). The dominating congeners in the adipose tissue of general populations are always CB-138, 153 and 180. Similar results, together with a defined profile of occurrence of individual congeners, were achieved also by other authors (Covaci et al., 2008; Knutsen et al., 2011; Thomas et al., 2006). That is why in many studies it was accepted that the levels of congener CB-153 in the blood serum constitute a marker for human exposure to polichlorinated biphenyl (Crisp et al., 1998; Jönsson et al., 2004).

Perfluoroalkylated substances have high chemical and thermal stability. For this reason, they found application in many industries as oleophobic substances, textile impregnation coatings, sliding substances, protective anti-adhesive coatings, electric cable insulators, high-capacity fire suppressing foams, hydro-gels applied on open wounds, emulsifiers for cosmetics, as well as ingredients of fat-resistant food packaging (EFSA, 2012; Kucharska et al., 2011). Such a wide application of perfluoroalkylated substances resulted in their global emission and distribution to various parts of the environment, including the human organism. Among the PFASs, the most commonly applied ones were the perfluorooctanoic acid (PFOA) and the perfluorooctane sulfonate (PFOS). Due to their toxicological properties the restrictions on the use of PFOS and PFOA in Europe were introduced by Commission Regulation (2010). Similarly as in the case of OCs and PCBs, the basic source of intake of these substances by humans is the food (EFSA, 2012; Guerranti et al., 2013; Noorlander et al., 2011). In case of PFOS, for people from various age groups, the main source of exposure are fish and seafood (50–80%), fruits and processed fruit products (8–27%), as well as meat and meat products (5–8%). In case of PFOA, the main source of exposure are fruits and processed fruit products (18–39%), and fish and seafood (7.6–27%) (EFSA, 2008). Similar exposure profiles are mentioned also by other authors studying the exposure of various populations to these compounds (Guerranti et al., 2013; Yamaguchi et al., 2013; Vestergren et al., 2008). In contrast to OCs, the magnitude of exposure of humans to PFASs through diet, according the studies of some research centres, is influenced also by the drinking water (Noorlander et al., 2011). In Sweden, studies were carried out in the years 1987–2007 for the levels of PFOS and PFOA, among others, in the blood serum of healthy women born in the years 1934–1967 (Axmon et al., 2014). The median of PFOS concentrations in individual years were in the range between 10.2 and 35.5 ng mL^{-1}, with slight falling trend since 1998, when the highest level of this compound was noted. PFOA concentrations were found in the range between 1.78 and 5.51 ng mL^{-1}, without any discernible trend (Axmon et al., 2014). Other authors studying the levels of PFOA and PFOS in blood serum obtained varying results, depending on the geographical location (place of residence) of the sample donors. In the study of Liu et al. (2012), for the whole population studied a median of concentrations for PFOS was found to be 1.92 μg L^{-1} (for women = 1.20 μg L^{-1}, for men = 2.39 μg L^{-1}), and for PFOA all results were below 0.03 μg L^{-1}. Meanwhile, in similar studies performed in Greece, the detected levels were higher, while retaining the relation of the higher concentrations in the blood serum of men over women. For women, the PFOS median was equal to 7.03 ng mL^{-1}, and for men – 13.69 ng mL^{-1}, in case of PFOA the values were, respectively 1.70 ng mL^{-1} and 3.14 ng mL^{-1} (Vassiliadou et al., 2010). Much higher levels were found in Germany, where the range of concentrations for the general population was 6.2–130.7 μg L^{-1} with the median of 22.3 μg L^{-1}, and for PFOA – between 1.7 μg L^{-1} and 39.3 μg L^{-1}, with the median of 6.8 μg L^{-1}. In all samples studied both compounds were found (Midasch et al., 2006).

Numerous publications confirm that people who start to live together as a couple change their eating habits. These changes take various directions: either one of the partners adapts to the eating habits of the other, or a common diet is developed based on eating habits of both (Bove et al., 2003; Kremmer et al., 1998; Lewis et al., 2006; Louk et al., 1999; Nasuti et al., 2014). Additionally, the diet of couples expecting a baby tends to change. During this period couples avoid products which are popularly deemed as unhealthy, while increasingly and more frequently eating healthier products and those that are less burdensome for the organism. (Laroche et al., 2012; Verbeke and de Bourdeaudhuij, 2007). This approach varies depending on whether the couple expects their first or another child. In case of the first child, the change in diet is more pronounced and the couple tries to introduce as many products which according to contemporary dietary knowledge are beneficial to health as

possible. In case of couples expecting their second and later child the observance of the so-called "healthy diet" tends to be more liberal and either the couple uses the same diet as they used to, or just the woman makes certain modification to eliminate or introduce certain products (Nasuti et al., 2014).

Taking into account these sociological conditions and the knowledge about the impact of anthropogenic organohalogen compounds on the human health and development, an attempt was made to find out whether the fact of living together in couples expecting a child causes the exposure to POPs to influence the levels of these compounds in the blood serum of both partners. The aim of this paper is to find out whether dependencies exist between the recorded levels of *p,p′*-DDE, CB-153, and PFOS and PFOA in peripheral blood of women and men living in a couple.

Material and methods

Study population

The population enrolled to this study consisted of pregnant women and their male partners from Greenland, Poland and Ukraine and was described in details earlier (Toft et al., 2005) as the INUENDO cohort. Couples were recruited during their visits in the obstetric outpatients clinic of the Gynaecological and Obstetric Hospital (Warsaw, Poland), in the eight antenatal clinics in three maternity hospitals in Kharkiv, Ukraine, and from 19 municipalities representing natives (Inuits) from all regions in Greenland. For *p,p′*-DDE, CB-153, PFOS, and PFOA analysis blood samples were taken from the INUENDO biobank.

Assumptions of the INUENDO project for the entire cohort of women were such that at the same time it was filled questionnaire for time to pregnancy (TTP) and blood samples were collected. In contrast, the partners of examined women completed a questionnaire and if expressed their willingness additionally blood samples were collected from them. The present study included only those couples where blood samples from both partners were taken. The summary statistics describing the study population of the parenting couples are presented in Table 1. For the study population of parenting couples in the questionnaires, there was no information about the length of living together for 18 cases from Greenland, 22 from Poland and 20 from Ukraine.

Ethics

The study was approved by local ethical committees; Polish Bioethical Committee (approval no. 6/2002 of 3.07.2002), Ethical Committee for Human Research in Greenland (approval no. 2010-13) and the Commission on Ethics and Bioethics of Kharkiv National Medical University in Ukraine (protocol number 7, October 7 2009). All participating couples signed informed consent.

Table 1
Characteristics of the parenting couples.

Country		GL	PL	UA
Age (years ± SD)	Female	27 ± 6.1	29 ± 3.2	25 ± 4.8
	Male	31 ± 7.4	30 ± 4.0	28 ± 5.4
BMI (SD)	Female	24.5 (4.3)	21.6 (3.2)	22.1 (3.4)
	Male	25.9 (3.8)	25.8 (3.3.)	24.2 (3.4)
Years of living together[a]	0 to ≤1	117 (29.2%)	44 (18.9%)	82 (31.9%)
	<1 to ≤3	90 (22.4%)	86 (36.9%)	81 (31.5%)
	>3	194 (48.4%)	103 (44.2%)	94 (36.6%)

GL – Greenland, PL – Poland, UA – Ukraine.
[a] Number of subjects and in the brackets the percentage of the total study population.

Specimens collection

All blood samples were collected according to the unified procedure. They were drawn from a cubital vein into 10 mL vacuum tubes for serum collection without additives (Becton Dickinson, Meylan, France) and centrifuged after cooling to room temperature at 4000 × *g* for 15 min. Then sera were separated. Serum was transferred with ethanol rinsed Pasteur pipettes to ethanol rinsed brown glass bottles (Termometerfabriken, Gothenburgh, Sweden). A piece of aluminum foil was placed on top of the bottles which were then sealed. All sampling devices were prepared and sent from Lund University, Sweden to Greenland, Poland and Ukraine. The detailed procedure, including temperatures and further handling were described elsewhere (Axmon et al., 2006; Jönsson et al., 2004). The collected samples were delivered to the Division of Occupational and Environmental Medicine, Department of Laboratory Medicine in Lund University, Sweden where they were analyzed.

Analysis of p,p′*-DDE and CB-153*

The analysis of *p,p′*-DDE and CB-153 were performed by GC/MS. Aliquots of 1 mL serum was added with 50 μL internal standard methanol solution containing 20 ng of ^{13}C-labeled CB-153/mL. Then 2 mL formic acid:2-propanol (4:1) mixture was added to the serum samples. The sample was sonicated for 5 min. After 30 min. 2 mL water: 2-propanol (17:3) was added and the sample was sonicated again for 5 min. The *p,p′*-DDE and CB-153 were extracted from serum by solid phase extraction (SPE) using on-column degradation of the lipids. The detailed analytical procedure, including apparatus and analytical characteristics were described earlier by Janák et al. (1999) and Richthoff et al. (2003).

Analysis of PFOS and PFOA

The analysis of PFOS and PFOA were performed by LC/MS/MS. Aliquots of 100 μL serum were added with 10 μL glucuronidase, 10 μL ammonium acetate buffer, 10 μL methylumbelliferyl-β-D-glucuronide and 25 μL of a water:acetonitrile (50:50) solution containing ^{13}C- or ^{18}O-labelled internal standards for all evaluated compounds and digested at 37 °C for 90 min. The proteins were precipitated with 175 μL acetonitrile and vigorously shaking for 30 min. The samples were thereafter centrifuged at 4200 × *g* and 3 μL of the supernatant was injected on a LC. The detailed analytical procedure, including apparatus, analytical characteristics and quality control assurance were described earlier by Lindh et al. (2012).

Statistics

The R3.0.1 statistical software (R Core Team, 2013) was used for all calculations and graphical presentations. Spearman rank correlation coefficients between CB-153, *p,p′*-DDE, PFOS, and PFOA, and paired Wilcoxon test for analysing the differences between CB-153, *p,p′*-DDE, PFOS, and PFOA, concentrations in the sera of males and females were used. In all analyses the level of significance was set at 0.05. For each country expected level of pollutants in male and female partners was calculated separately using linear regression models of relationship between pollutant's level and person age and BMI. Next intraclass correlation coefficients (ICC) between residuals of fitted models for male and female partners were calculated. Results may be interpreted as correlation between level of pollutant in male and female partners adjusted for their age and BMI. To investigate if duration of marriage (partnership) influences strength of correlation we also calculated ICC in subgroups of partners living together less than 1 year, from 1 to 3 years and more than 3 years.

Table 2
Summary statistics for *p,p′*-DDE, CB-153 (ng g^{-1} lipid) and PFOS, PFOA (ng mL^{-1}) by sex and country.

Country	Compounds	*N*	Mean	SD	Min–Max	Median
GL	*p,p′*-DDE♀	419	399.8	396.1	5.3–3122.0	285.3
	p,p′-DDE♂		815.9	953.4	4.4–13,197.2	555.3
	CB-153♀	419	156.2	169.3	2.6–1403.7	100.6
	CB-153♂		319.4	394.3	5.1–5455.2	196.4
	PFOS♀	185	23.6	13.0	4.1–87.3	20.6
	PFOS♂		51.7	24.0	12.3–160.6	44.9
	PFOA♀	185	1.9	0.8	LOD[a]–4.6	1.8
	PFOA♂		4.8	1.6	LOD–35.0	4.5
PL	*p,p′*-DDE♀	255	426.9	269.4	32.5–1810.5	373.0
	p,p′-DDE♂		585.7	295.0	143.2–2094.2	528.1
	CB-153♀	255	12.0	9.0	2.1–74.8	10.7
	CB-153♂		19.4	13.6	3.3–129.1	16.8
	PFOS♀	188	8.4	3.0	1.6–21.3	7.9
	PFOS♂		18.6	5.7	8.2–40.2	18.6
	PFOA♀	188	2.9	1.3	LOD–9.8	2.7
	PFOA♂		5.3	2.1	16.0–4.9	4.9
UA	*p,p′*-DDE♀	277	788.9	544.0	147.1–5942.6	671.5
	p,p′-DDE♂		1170.8	967.4	257.1–11,790.9	930.0
	CB-153♀	277	32.8	35.9	3.3–533.4	27.7
	CB-153♂		55.9	72.1	5.5–953.8	44.3
	PFOS♀	189	5.4	2.2	1.2–17.5	5.1
	PFOS♂		8.1	3.7	2.8–28.8	7.6
	PFOA♀	189	1.0	0.6	<LOD–5.5	0.9
	PFOA♂		1.8	2.8	<LOD–35.0	1.3

♀ – female, ♂ – male, GL – Greenland, PL – Poland, UA – Ukraine.
[a] LOD – limit of detection, LOD for PFOS = 0.2 ng mL^{-1}, LOD for PFOA = 0.6 ng mL^{-1}.

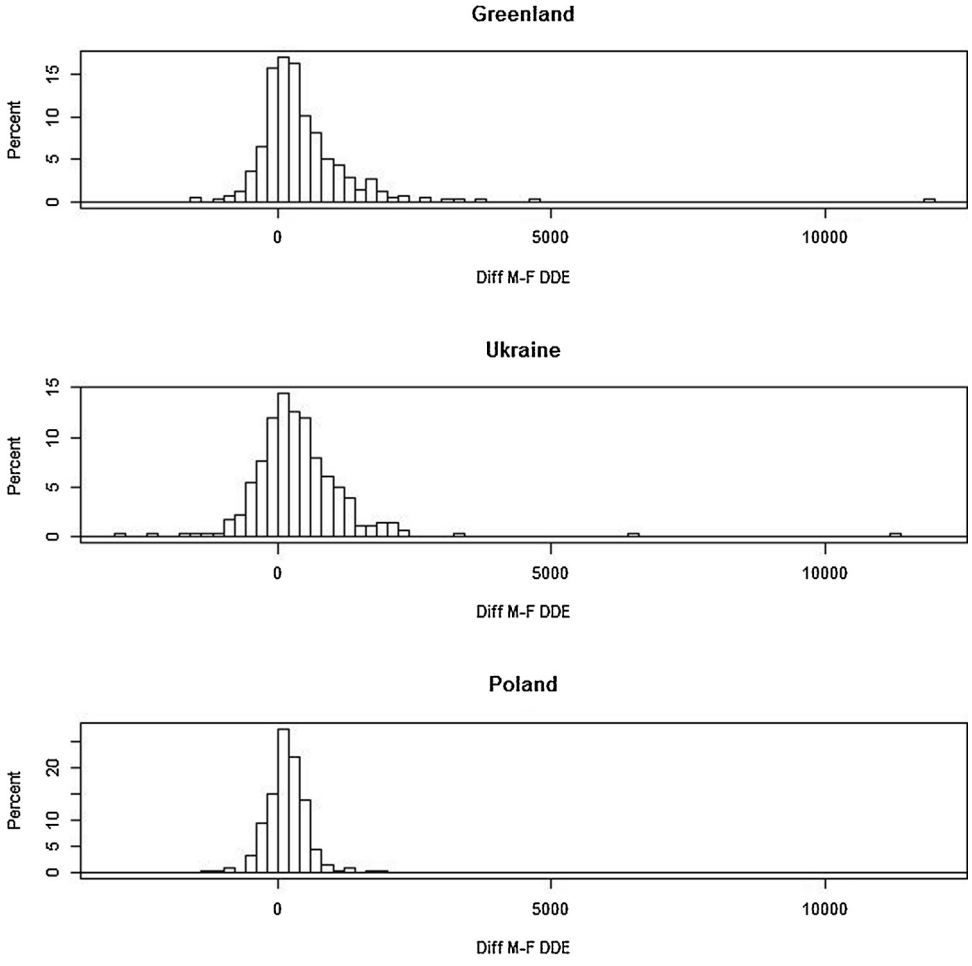

Fig. 1. Histogram of distributions of differences between concentrations of *p,p′*-DDE in blood serum of men and women living in parenting couples from Greenland, Poland and Ukraine.

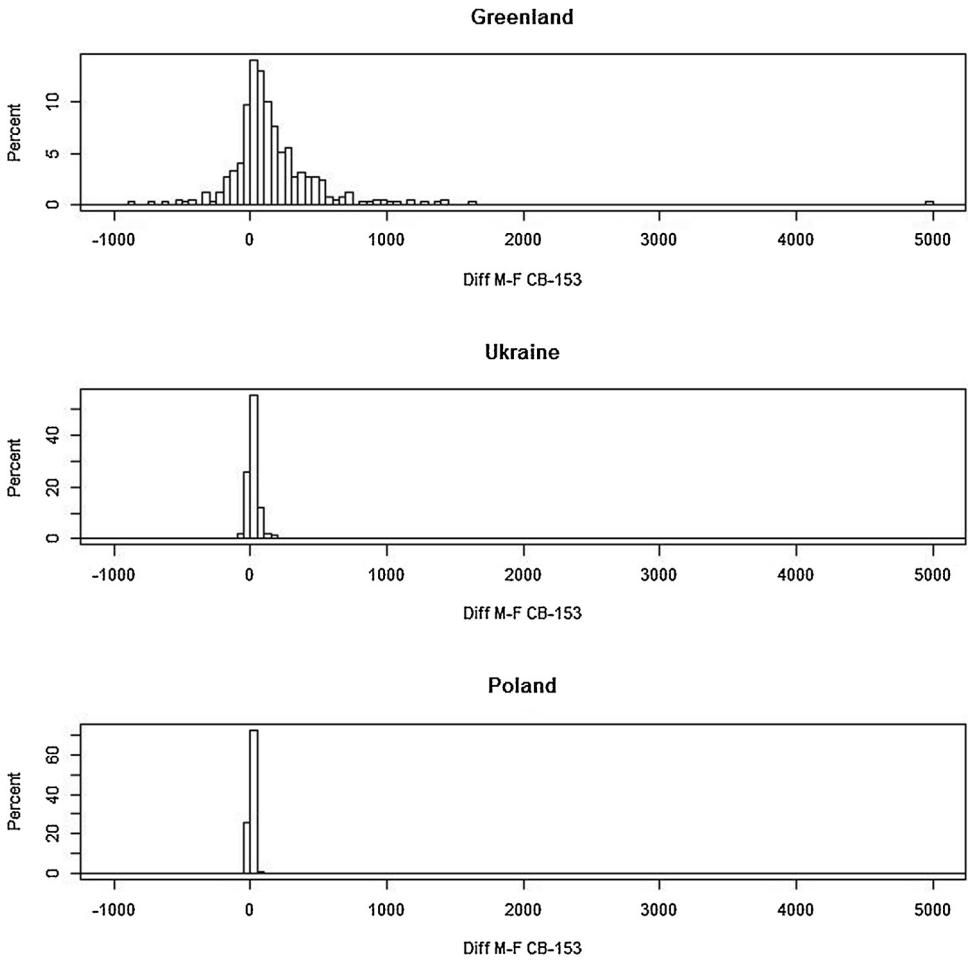

Fig. 2. Histogram of distributions of differences between concentrations of CB-153 in blood serum of men and women living in parenting couples from Greenland, Poland and Ukraine.

p,p′-DDE, CB-153, PFOA and PFOS were found above or equal to limit of detection (LOD) in all serum samples from Greenland and Poland. In Ukraine *p,p′*-DDE, CB-153, PFOS above LOD were in all serum samples and PFOA was above LOD in 92% serum samples.

Results

The summary statistics including serum *p,p′*-DDE, CB-153, PFOS and PFOA concentrations in couples from Greenland, Poland and Ukraine are presented in Table 2.

Due to strong lipophilicity of organochlorine compounds (*p,p′*-DDE and CB-153), their concentration in the blood serum is expressed in ng g^{-1} of extracted lipid to facilitate comparison with results obtained by other authors (Rylander et al., 2012). The results for PFOS and PFOA are presented in ng per 1 mL of blood serum. Concentrations of *p,p′*-DDE, CB-153, PFOS and PFOA presented in Table 2 show that in the studied population of parenting couples in Greenland, Poland and Ukraine the levels of these compounds were higher in men in comparison to the concentrations found in the blood serum of their female partners. The highest concentrations of *p,p′*-DDE were found in Ukrainian men, while the lowest – in men from Poland. In case of CB-153, the highest levels were found in the blood serum of men from Greenland, and the lowest in men from Poland. The concentration profiles for both these compounds in women were identical. These dependencies looked similar also for PFOS and PFOA, where the highest concentrations were found in men from Greenland, and the lowest in men from Ukraine. The differences in concentrations of the studied compounds in men and women were statistically significant in all cases ($p < 0.0001$). Figs. 1–4 present histograms of distributions of differences between the concentrations of the compounds found in the blood serum of men and women living in parenting couples. These distributions are presented separately for each compound and country of origin of the couples. The figures have a standardized scale, which allows for comparison. Majority of figures show a shift to the right, which is an indicator of higher concentrations of the compound in men. For samples from Greenland, for all compounds in question, the difference between the concentrations in men and women is significant, as indicated by the strong shift to the right. In case of Ukraine a strong shift is noted only for *p,p′*-DDE, and for Poland – for PFOS and PFOA. In other cases the differences between concentrations are insignificant, as indicated by results concentrated around zero.

Next, a study was made to find out whether a relation exists between the concentrations of compounds in question found in the blood serum of men and women from the same parenting couple. The intraclass correlation coefficient (ICC) was calculated for concentration levels of the compounds in question (*p,p′*-DDE, CB-153, PFOS and PFOA) in the blood serum of parenting couples standardized for their age and BMI, as presented in Table 3. The results in Table 3 show weak positive correlation between concentrations of the compounds in the blood serum of men and women forming parenting couples, but it is not statistically significant in all cases.

Significantly statistical intraclass correlation coefficient between the concentrations of *p,p′*-DDE were detected in the blood serum of women and men living in parenting couples from

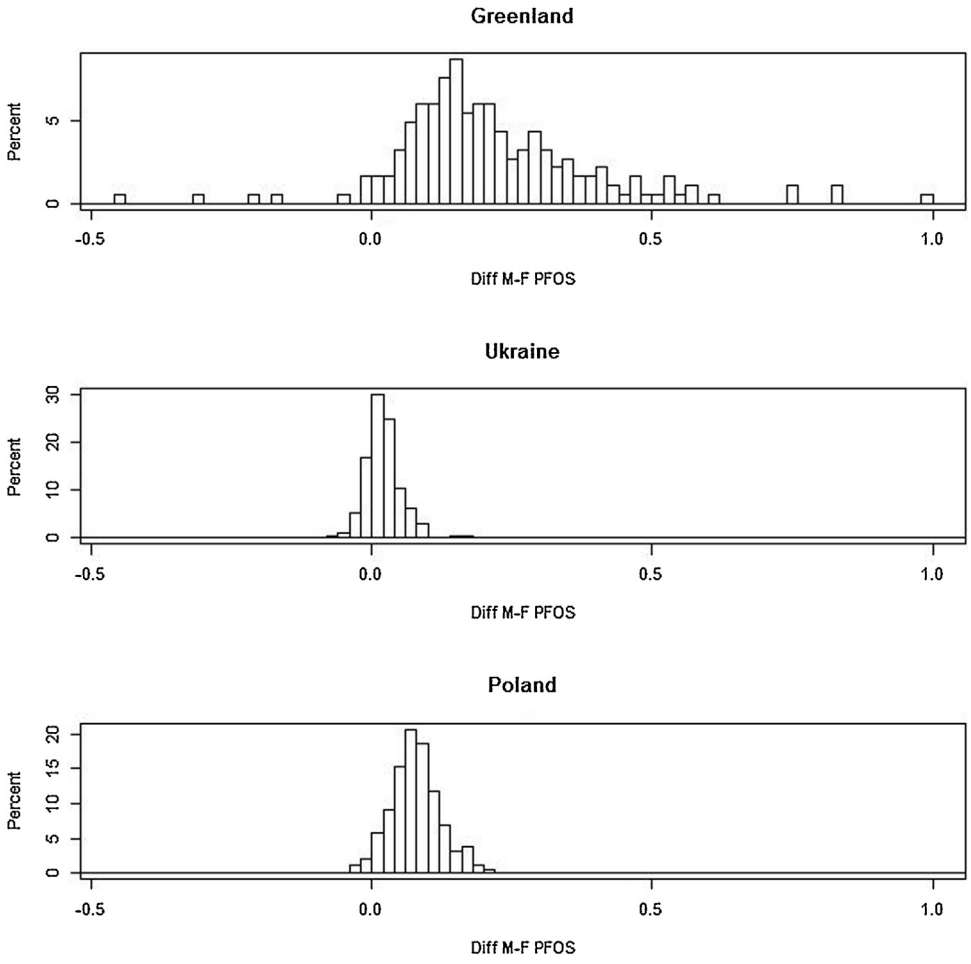

Fig. 3. Histogram of distributions of differences between concentrations of PFOS in blood serum of men and women living in parenting couples from Greenland, Poland and Ukraine.

Greenland and Poland. In case of CB-153, ICC was found in pairs from Greenland, while in case of PFOS such correlations were found only in population of parenting couples from Greenland and Poland. For PFOA, the correlation was found only in population of parenting couples from Poland. There was no correlation between the time of being together and ICC value.

Discussion

To date, the reports of concentrations of organochlorine compounds in the biological material collected from humans showed, in general, higher levels of these compounds in samples collected from men than those from women within the same population,

Table 3
Intraclass correlation coefficients between *p,p'*-DDE, CB-153, PFOS and PFOA in males and females, by country.

		Total			Length of living together (years)								
					0 to ≤1			<1 to ≤3			>3		
		ICC	95% C.I.	*p*	ICC	95% C.I.	*p*	ICC	95% C.I.	*p*	ICC	95% C.I.	*p*
GL	CB-153	0.232	(0.138–0.322)	<0.001	0.203	(0.018–0.375)	0.016	0.323	(0.122–0.499)	0.001	0.229	(0.089–0.361)	<0.001
PL		0.062	(−0.063 to 0.185)	0.166	−0.174	(−0.451 to 0.133)	0.868	0.029	(−0.187 to 0.242)	0.398	0.109	(−0.086 to 0.297)	0.136
UA		−0.023	(−0.143 to 0.098)	0.644	−0.150	(−0.364 to 0.078)	0.903	−0.024	(−0.253 to 0.207)	0.580	0.001	(−0.210 to 0.212)	0.496
GL	*p,p'*-DDE	0.242	(0.148–0.331)	<0.001	0.144	(−0.042 to 0.322)	0.064	0.357	(0.160–0.527)	<0.001	0.254	(0.115–0.383)	<0.001
PL		0.130	(0.006–0.251)	0.020	0.171	(−0.137 to 0.449)	0.136	0.123	(−0.094 to 0.328)	0.133	0.085	(−0.110 to 0.274)	0.196
UA		0.095	(−0.026 to 0.214)	0.062	−0.002	(−0.228 to 0.223)	0.508	0.142	(−0.091 to 0.361)	0.115	−0.055	(−0.263 to 0.157)	0.695
GL	PFOA	0.042	(−0.107 to 0.188)	0.291	0.304	(0.018–0.544)	0.019	−0.018	(−0.365 to 0.333)	0.539	0.003	(−0.204 to 0.210)	0.489
PL		0.211	(0.067–0.347)	0.002	0.094	(−0.259 to 0.424)	0.302	0.247	(−0.001 to 0.467)	0.025	0.194	(−0.036 to 0.405)	0.049
UA		0.045	(−0.100 to 0.188)	0.273	0.049	(−0.219 to 0.302)	0.370	0.199	(−0.091 to 0.457)	0.088	−0.006	(−0.264 to 0.253)	0.518
GL	PFAS	0.253	(0.109–0.386)	<0.001	0.275	(−0.014 to 0.521)	0.031	0.104	(−0.255 to 0.437)	0.286	0.284	(0.081–0.464)	0.003
PL		0.199	(0.054–0.335)	0.004	0.284	(−0.066 to 0.572)	0.054	0.220	(−0.030 to 0.443)	0.042	0.170	(−0.061 to 0.384)	0.073
UA		0.012	(−0.132 to 0.156)	0.434	0.019	(−0.244 to 0.278)	0.446	−0.037	(−0.318 to 0.250)	0.599	−0.072	(−0.324 to 0.190)	0.704

GL – Greenland, PL – Poland, UA – Ukraine.

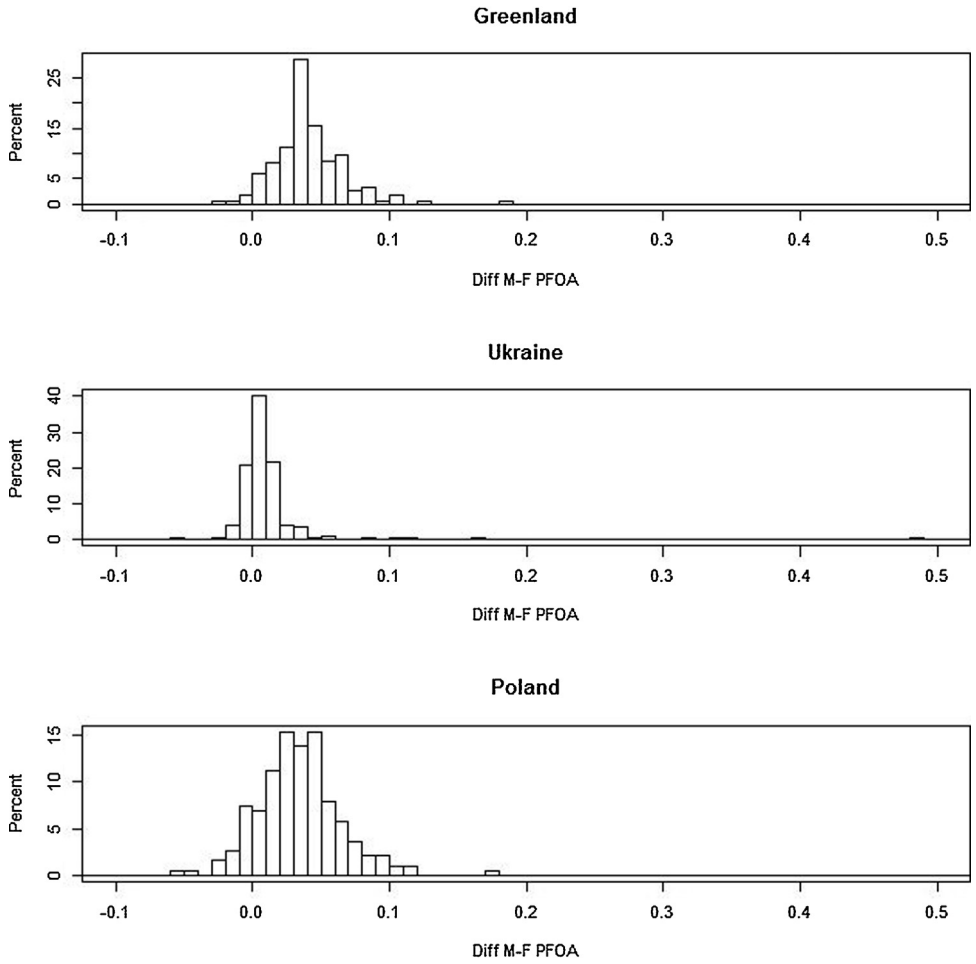

Fig. 4. Histogram of distributions of differences between concentrations of PFOA in blood serum of men and women living in parenting couples from Greenland, Poland and Ukraine.

regardless of the place of residence (Czaja et al., 2006; Gaffney et al., 2005; Salihovic et al., 2012; Thomas et al., 2006). For example, in the study of Gaffney et al. (2005) a higher concentration of organochlorine compounds was detected in the blood of men in relation to women living in various areas of the United States. Similar correlations were found also in the case of the INUENDO cohort, i.e. women and men inhabiting Greenland, Poland and Ukraine (Jönsson et al., 2004; Czaja et al., 2006). In our study, after the selection of only parenting couples from the whole INUENDO cohort, the above relations were also found. The levels of the studies organochlorine compounds were higher in men than in their female partners. For CB-153 these correlations were statistically significant in Poland and Ukraine, and for *p,p′*-DDE – for donors from Greenland and Ukraine (see Table 2). However, this relation is not a rule, as some authors cite higher concentrations in women than in men from the same population, which was reported, for example, in studies of volunteers from Spain. Their blood serum contained *p,p′*-DDE, for which the median in men was 168.0 ng g^{-1}, while in women – 191.0 ng g^{-1} (Arrebola et al., 2013).

The levels of PFOS and PFOA detected by numerous researchers in the blood serum indicate higher concentrations in men than in women. These differences were statistically significant regardless from the population from which the sample was taken. For example, Vassiliadou et al. (2010), studying the population of women and men from various regions of Greece, found that the average concentrations of PFOS in men: 13.63 ng mL^{-1} (Argolida) and 14.93 ng mL^{-1} (Athens), and for PFOA from 2.05 ng mL^{-1} (Argolida) and 3.88 ng mL^{-1} (Athens), while in women respectively 9.28 ng mL^{-1} and 7.49 ng mL^{-1}, and 1.92 ng mL^{-1} and 2.08 ng mL^{-1}. Similar results were noted by researchers from other centres, such as Italy – Ingelido et al. (2010), Germany – Fromme et al. (2007), or Ericson et al. (2007) from Spain. Similar correlation can be found in case of PFOS and PFOA concentrations in the blood serum of men and women from Greenland, Poland and Ukraine of the whole INUENDO cohort. Lindh et al. (2012) quantified PFOS and PFOA in the blood serum of men from Greenland, Poland and Ukraine, while Lyngsø et al. (2013) for women from the same cohort. After the isolation of parenting couples from the INUENDO cohort and comparison of concentrations of these compounds within the same couple, regardless of detection of positive correlation between concentrations in women and men, the profiles of the concentrations of the compounds in question in individual countries remained the same. In majority cases, higher concentrations of PFOA and PFOS were detected in men than in their female partners; in some instances these correlations were statistically significant (see Table 3).

Taking into account the fact that the majority of pairs included in this study, regardless of the place of residence, were of similar age (Jönsson et al., 2004) one should suppose that the differences of concentrations of the tested compounds between the populations in various regions of Europe resulted from differences in environmental pollution, including food, in a given region of Europe. Therefore, the assessment of human exposure to POPs, especially PFASs, should also take into account indoor dust as a potential source of exposure (Goosey and Harrad, 2011; Haug et al., 2011). Additionally, the detected levels of concentration could have been influenced rather by the differences in dietary habits

in individual countries, e.g. the higher share of fish in the diet in Greenland, rather than individual dietary preferences of individual couples. In the couples included in this study the concentrations of compounds in question were comparable to the levels measured in populations of Greenland, Poland and Ukraine, which was reported for whole populations included in INUENDO study (Jönsson et al., 2004; Lindh et al., 2012; Lyngsø et al., 2013). These results were also consistent with the concentrations reported by other authors (Kannan et al., 2004; Olsen et al., 2005).

Our estimates show that a weak correlation exists in parenting couples between concentrations of the studied compounds in both partners, but it is not statistically significant in all cases. A statistically significant positive intraclass correlation coefficients for PFOS and *p,p′*-DDE was found only for couples from Greenland and Poland, PFOA in Poland, and CB-153 in Greenland. Further analyses showed no association between duration of living together and ICC value (see Table 3). Similar correlations for *p,p′*-DDE and total PCBs were reported by Gaffney et al. (2005), who studied the population of men and women living in a selected area of the USA. It has to be stressed that couples included in our study were not asked about the frequency of eating together nor about the time they spent in their houses together, which would make it possible to estimate whether the house dust might to be considered as a potential source of exposure to these compounds for the tested couples. Still, the confirmation of these exposure profiles requires a more precise research, taking into account differences in metabolism of these compounds in both sexes (Knutsen et al., 2011; Rylander et al., 2012).

Conclusion

As far as we know, this is the first paper such study to suggest that living in one household is related to the level of exposure to certain organohalogen compounds, measured by their concentrations in the blood serum of the partners. The tests have shown that there is a relation between the concentrations of *p,p′*-DDE, CB-153, PFOS and PFOA in the blood serum of women and men living in parenting couples. The positive correlation between these concentrations is in some cases statistically significant, but not strong enough to confirm that the fact of living together, and – as a result – similar dietary model, is an important determinant shaping the levels of these compounds in blood serum. On these grounds it may be assumed that the concentrations of these compounds in men and women are mainly influenced by the place of residence.

Conflict of interest statement

The authors have no financial interest or other conflicts of interest in the publication of these results.

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