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Perfluorinated chemicals in blood serum of inhabitants in central Poland in relation to gender and age

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HIGHLIGHTS

- Concentrations of seven selected PFASs in serum samples of men and women were analysed.
- Higher concentrations were observed in five of seven PFASs in men than women.
- PFHxS and PFDoDA were statistically insignificant higher in women than in men.
- Hypothesis that concentrations of PFASs increase with age, regardless of sex, was not confirmed.

GRAPHICAL ABSTRACT

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ABSTRACT

The goal of this paper is to determine concentrations of seven selected perfluoroalkylated substances (PFASs): perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA), perfluorohexane sulfonic acid (PFHxS), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA) in the blood serum of men and women of reproductive age from the central region of Poland. The relation between sex of tested subjects and the levels of compounds in blood serum of humans will also be considered and analysed as an element of the risk assessment. The study was made on the blood serum samples collected from 253 women and 176 men of reproductive age between 20 and 44 years from Warsaw and surrounding areas. Higher concentrations of five (PFOS, PFOA, PFNA, PFDA, PFUnDA) from among seven selected PFASs were observed in men in comparison to women from the same populations. Only the concentrations of PFHxS and PFDoDA were slightly higher in women than in men. These differences were statistically significant in all cases, except for PFUnDA. The hypothesis that the concentrations of said compounds increase with age of the test subjects, regardless of gender has not been confirmed.

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1. Introduction

Perfluoroalkylated substances (PFASs) are listed among persistent organic pollutants (POPs), eliciting high interest of scientists especially in terms of biological response of the organism as a result of internal and external exposure. PFASs are characterized by high chemical and thermal stability, and by high surface activity. The terminology and classification of PFASs were proposed in 2011 by Buck et al. (2011). For this reason, they found broad use in various industries as oleophobic substances, impregnation agents for textiles, lubricants, etc. They are also used as elements of anti-stick coatings, dielectric insulators, fire suppressing foams, hydrogels applied on open wounds, emulsifiers in cosmetics and fat-resistant food packaging (EFSA, 2012; Kucharska et al., 2011; Prevedouros et al., 2006). Such a broad use of PFASs to 2010, resulted in their global distribution to the environment, including the human organism as demonstrated by publications describing the levels of tested compounds in various parts of the environment both in Poland and in other countries (Bossi et al., 2015; Falandysz et al., 2012; Rostkowski et al., 2006, 2009; White et al., 2015). Among the PFASs, the most widely used were: perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS). A series of tests on animals revealed the toxic effect these compounds have on the liver and lungs. Moreover, the scientists observed their reproductive, immunological toxicity, impact on the development and functioning of the endocrine system, their role in the development of neurobehavioural disorders, as well as weak genotoxicity and cancerogenicity (Eriksen et al., 2010; Kvist et al., 2012; Lau et al., 2007; Shi et al., 2009; Zhang et al., 2008). For this reason, PFOS and its salts were included as POPs into the Annex B of the Stockholm Convention, i.e. compounds the use of which is acceptable only in precisely defined areas. In 2010 the European Commission with its Regulation no. 757/2010 imposed restrictions for the use of PFOS in the territory of the European Union.

The PFASs have not been produced in Poland, while in one early of a few studies from Poland some of perfluorinated carboxylic and sulfonic acids have been determined at very low levels in samples of bottled mineral waters and in potable water (Rostkowski et al., 2008). Those compounds imported from abroad could be used somewhere in Poland as well as deposited from atmosphere because of global pollution. They were found in inland surface water from rivers and lakes and in Baltic coastal water and sediments in Poland but pollution was negligible (Falandysz et al., 2006a; Rostkowski et al., 2006, 2009). An important source of exposure of humans to PFSAs can be food. In a study in Poland as dietary source of several PFSA compounds was identified Baltic fish, waterfowl and game animal meat (Falandysz et al., 2006b, 2007; Waszak et al., 2014). According to the EFSA (2008) opinion the fish and seafood (50–80%), fruit and fruit products (8–27%) and meat and meat products (5–8%) contributed to PFOS exposure in diet, and in case of PFOA — fruit and fruit products (18–39%) and fish and seafood (7.6–27%). Similar dependencies are reported also by other authors studying the scope of exposure of various populations to these compounds (Berger et al., 2009; Delinsky et al., 2009; Guerranti et al., 2013; Yamaguchi et al., 2013; Vestergren et al., 2008). Similar dependencies are reported also by other authors studying the scope of exposure of various populations to these compounds (Guerranti et al., 2013; Yamaguchi et al., 2013; Vestergren et al., 2008). Fromme et al. (2009) assessed the total exposure of adults in Western Europe to PFASs from all potential sources (air, dust, food), and reported the following results: in case of PFOS, average (calculated on the basis of median or average concentration) 1.6 ng kg^{-1} bw day^{-1} vs. high (calculated on the basis of the highest concentration) 8.8 ng kg^{-1} bw day^{-1}, and in case of PFOA, it amounted respectively to 2.9 ng kg^{-1} bw day^{-1} and 12.6 ng kg^{-1} bw day^{-1}. The values were significantly lower than the tolerable daily intake (TDI) which for PFOS has been set at 150 ng kg^{-1} bw day^{-1}, and for PFOA at 1500 ng kg^{-1} bw day^{-1} (EFSA, 2008). Other PFAS compounds were found at significantly lower levels, sometimes below the limit of detection for a given analytical method. Similar intake of PFASs with food was reported by other authors (Ericson et al., 2009; Kärrman et al., 2009; Cornelis et al., 2012). The level of exposure of humans to PFAS is also influenced by the drinking water (Noorlander et al., 2011). According to Post et al. (2012) continued exposure to even relatively low concentrations in drinking water can substantially increase the total human exposure, with a serum:drinking water ratio of about 100:1.

Fish have been identified for some human populations as major source of PFSAs (Falandysz et al., 2006a,b; Denys et al., 2014; Heo et al., 2014). Another potential source of intake of these compounds by humans, still not well researched, is dust (D'Hollander et al., 2010; Eriksson and Kärrman, 2015). Dust as a significant source of exposure may be the case for North American and some Asian countries (Fromme et al., 2009).

Retrospective studies in Sweden, carried out between 1987 and 2007, focused on the levels of PFOS and PFOA in blood serum of healthy women born in the years 1934–1967 (Axmon et al., 2014). The values of the median for PFOS were within the concentration range between 10.2 and 35.5 ng mL^{-1}, with slight decreasing trends since 1998, when the highest level of this compound was reported. In case of PFOA, the concentrations were 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 the blood serum received various results, depending on the place of living of sample providers. In the study of Liu et al. (2012), the median of PFOS concentrations for the whole study group of volunteers from a selected province in China, representing cities, towns as well as rural areas amounted to 1.92 μg L^{-1} (1.20 μg L^{-1} for women and 2.39 μg L^{-1} for men), and for PFOA all results were below 0.03 μg L^{-1}. In similar study, conducted in Greece, the detected levels were higher, but retained the same relation, i.e. the concentrations in the serum of men were higher from those in the serum of women. The median of PFOS concentrations in women amounted to 7.03 ng mL^{-1}, and in men — to 13.69 ng mL^{-1}, in case of PFOA respectively 1.70 ng mL^{-1} and 3.14 ng mL^{-1} (Vassiliadou et al., 2010). Significantly higher levels were reported in Germany, where the range of PFOS concentrations in the blood serum of general population ranged between 6.2 and 130.7 μg L^{-1}, and of PFOA — between 1.7 μg L^{-1} and 39.3 μg L^{-1}. In all tested cases both compounds were detected (Midasch et al., 2006).

PFASs have a greater ability to bind with protein and less affinity with lipids, in contrast to other POPs. They have also higher ability of bioaccumulation in the food chain of humans, and the half-live elimination of these compounds from human organism takes between 4.5 and 5.4 years (Olsen et al., 2007). Such a long period of elimination from human organism, combined with the adverse toxicological characteristics of PFASs (EFSA, 2008) justifies investigation of their deposits in the human tissue as markers of internal exposure. Taking the above into account, an attempt was made to establish the concentrations of selected PFASs in the blood serum of women and men from Poland.

The goal of this paper was to determine concentrations and profiles of seven selected PFASs: perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA), perfluorohexane sulfonic acid (PFHxS), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), and perfluorododecanoic acid (PFDoDA) in the blood serum of men and women of reproductive age in Poland. The relation between gender and age and the detected levels of these compounds in human blood serum will also be considered and analysed as an element of the risk assessment.

2. Methods

2.1. Study population

The population enrolled to this study consisted of pregnant women and their male partners from Poland as a part of the INUENDO cohort

which included Greenlandic, Polish and Ukrainian population (Toft et al., 2005). Couples were recruited during their visits in the obstetric outpatients clinic of the Gynaecological and Obstetric Hospital (Warsaw, Poland). For PFASs analysis 176 male and 253 female blood samples were taken from the INUENDO biobank.

The study was approved by the Polish Bioethical Committee (approval no. 6/2002 of 3.07.2002). All participating couples signed informed consent.

The characteristics of the female and male cohorts are summarized in Table 1.

2.2. Specimens collection

All blood samples were collected according to a 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 Poland. The detailed procedure, including temperatures and further handling are described elsewhere (Axmon et al., 2006; Jönsson et al., 2005).

2.3. Analysis of PFASs

The analysis of PFASs was performed by LC/MS/MS in the Division of Occupational and Environmental Medicine, Department of Laboratory Medicine, Lund University, Sweden.

For the present study the following PFASs were selected: PFOS, PFOA, PFHxS, PFNA, PFDA, PFUnDA, and PFDoDA.

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 (UFLC^{XR} Shimadzu Corporation, Kyoto, Japan); LC/MS/MS using hybrid quadrupole linear ion trap mass spectrometry equipped with a Turbo Ion-Spray source (QTRAP 5500, Applied Biosystems). The quality of the measurements was controlled by analysing chemical blanks and in-house quality control samples, prepared from a large volume of serum spiked with small amounts of different PFASs. The analysis of PFASs was a part of the Round Robin inter-comparison programme (Professor Dr. med. Hans Drexler, Institute and Out-Patient Clinic for Occupational, Social and Environmental Medicine, University of Erlangen-Nuremberg, Germany) with results within the tolerance limits, determined as three times the standard deviation of the results from special reference laboratories. The detailed analytical procedure, including apparatus, analytical characteristics and quality control assurance was described earlier by Lindh et al. (2012).

Limits of detection (LOD) for the particular compounds were as follows: PFOS — 0.2 ng mL^{-1}, PFOA — 0.6 ng mL^{-1}, PFNA — 0.2 ng mL^{-1}, PFDA — 0.2 ng mL^{-1}, PFHxS — 0.06 ng mL^{-1}, PFUnDA — 0.3 ng mL^{-1} and PFDoDA — 0.07 ng mL^{-1}.

2.4. Statistics

The statistical analyses includes 429 samples (N = 253 for female, and N = 176 for male). Summary statistics for compounds with detection frequencies >30% were calculated by conventional methods, only when the frequency of detection was less than 30% the results were computed for each sample (Lubin et al., 2004).

Values of PFHxS, PFOA, PFOS, and PFNA were found in all serum samples. For other compounds, except PFUnDA and PFDoDA, when the concentrations were below the limit of detection the appropriate LOD values were applied for statistical calculations. Therefore values only for PFUnDA and PFDoDA were replaced with imputed values, where the minimum value is equal to, respectively 0.009 and 0.001.

The STATISTICA software rev. 9 was used for all calculations. The conformity of distribution of the results with the normal distribution was carried out with the help of chi-square test, K–S–L test and Shapiro–Wilk test. To establish the variance homogeneity, Hartley's *Fmax* test, the Levene's test and Broun–Forsythe test were used. In the statistical analysis also non-parametric tests were used: Mann–Whitney test, Wald–Wolfowitz run test and Kruskal–Wallis ANOVA test and the median test.

3. Results and discussion

The summary statistics including serum concentrations of PFASs in male and female from Poland are presented in Tables 2 and 3.

From among the seven analysed PFASs, the highest average concentrations were found in case of PFOS both in blood serum samples collected from men (18.49 ng mL^{-1}) and women (8.42 ng mL^{-1}). Higher average concentrations of all PFASs were noted in the blood serum of men rather than of women from the studied cohort, except

Table 1
Characteristics of the female and male cohorts.

Parameters	Male	Female
Age [years]		
Mean (SD)	30 (3.8)	29 (3.2)
Min–max	22–44	20–41
Median	29	28
BMI		
Mean (SD)	25.57 (2.9)	21.48 (2.9)
Min–max	18.51–38.51	15.43–40.75
Median	25.37	20.88

Table 2
Concentrations of PFASs for Polish females (n = 253) [ng mL^{-1}].

	PFHxS	PFOS	PFOA	PFNA	PFDA	PFUnDA	PFDoDA
Mean	2.34	8.42	2.83	0.67	0.24	0.14	0.06
SD	1.57	2.95	1.27	0.36	0.12	0.075	0.04
Min	0.34	1.61	0.67	0.20	<LOD[a]	0.01	<LOD
Max	10.16	21.28	9.83	2.75	0.91	0.72	0.45
Median	1.92	7.96	2.67	0.59	0.22	0.13	0.04
P 5%	0.75	4.38	1.27	0.29	<LOD	0.06	0.01
P 95%	5.46	13.35	4.87	1.38	0.45	0.24	0.11

[a] LOD — limit of detection (PFOS = 0.2 ng mL^{-1}, PFOA = 0.6 ng mL^{-1}, PFNA = 0.2 ng mL^{-1}, PFDA = 0.2 ng mL^{-1}, PFHxS = 0.06 ng mL^{-1}, PFUnDA = 0.3 ng mL^{-1} and PFDoDA = 0.07 ng mL^{-1}).

Table 3
Concentrations of PFASs for Polish males (n = 176) [ng mL^{-1}].

	PFHxS	PFOS	PFOA	PFNA	PFDA	PFUnDA	PFDoDA
Mean	1.22	18.49	5.12	1.29	0.40	0.17	0.04
SD	0.45	5.71	2.04	0.60	0.15	0.11	0.04
Min	0.12	8.20	0.94	0.47	<LOD[a]	0.02	<LOD
Max	3.78	40.14	12.56	5.32	1.09	0.69	0.44
Median	1.18	18.35	4.77	1.19	0.37	0.13	0.02
P 5%	0.66	9.60	2.41	0.66	0.21	0.06	<LOD
P 95%	2.01	29.17	8.80	2.14	0.69	0.34	0.08

[a] LOD — limit of detection (PFOS = 0.2 ng mL^{-1}, PFOA = 0.6 ng mL^{-1}, PFNA = 0.2 ng mL^{-1}, PFDA = 0.2 ng mL^{-1}, PFHxS = 0.06 ng mL^{-1}, PFUnDA = 0.3 ng mL^{-1} and PFDoDA = 0.07 ng mL^{-1}).

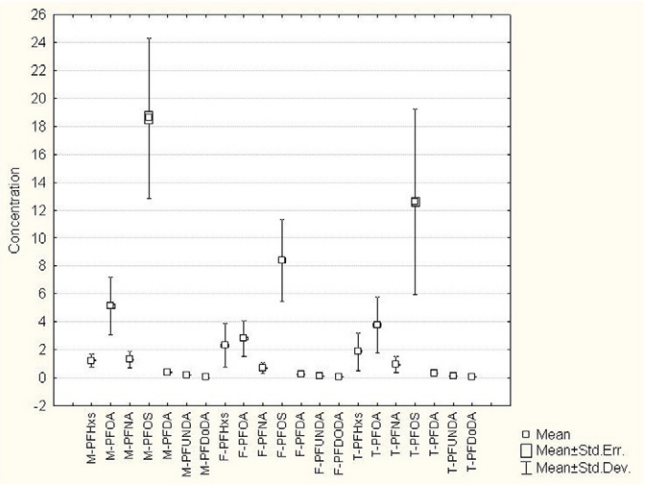

Fig. 1. Distributions of individual PFASs concentration in total population (T), and in males (M) and females (F).

for PFHxS and PFDoDA, the average concentrations of which were slightly higher in the blood serum of women. The average concentrations of these two compounds in the blood serum of women and men were, respectively, 2.34 ng mL^{-1} and 0.06 ng mL^{-1} vs. 1.22 ng mL^{-1} and 0.04 ng mL^{-1}. For all studied compounds, except for PFUnDA, the differences in concentrations in the blood serum of men and women were statistically significant with the assumed confidence interval $p = 0.05$. Due to the distribution of results proven by the chi-square test and confirmed by K–S–L and Shapiro–Wilk test as diverging from the standard distribution, the non-parametric tests were used to find out whether there are also statistically significant differences between concentrations of tested compounds in both sexes. For PFUnDA, this was not confirmed neither with Mann–Whitney test ($p = 0.0767$) nor with the median test ($p = 0.9226$). For the remaining compounds, the tests confirmed statistically significant differences between their concentrations. Fig. 1 shows the distribution of the tested compound concentrations in the total population treated as a whole and divided into sexes.

Similar relations were found also by other authors, who indicated higher average concentrations of PFASs in men in relation to the levels of these compounds in the blood serum of women. For example, Ericson et al. (2007) reported average concentrations e.g. PFOS 8.47 ng mL^{-1} in men vs. 6.81 ng mL^{-1} in women and respectively PFOA 2.02 ng mL^{-1} vs. 1.57 ng mL^{-1} in the full blood of Catalonians (Spain). Moreover, average concentrations of PFHxS in these studies were also higher in men (4.48 ng mL^{-1}) than in women (2.55 ng mL^{-1}), contrary to our results.

Table 4
Concentrations of PFASs for males and females in different age groups [ng mL^{-1}].

	PFHxS	PFOS	PFOA	PFNA	PFDA	PFUnDA	PFDoDA
Males							
≤25 years (n = 9)							
Mean (SD)	1.33 (0.52)	18.51 (6.00)	4.65 (2.20)	1.12 (0.35)	0.35 (0.10)	0.15 (0.08)	0.03 (0.02)
Min	0.62	8.41	1.48	0.50	0.20	0.06	0.012
Max	2.32	27.72	9.48	1.78	0.49	0.32	0.07
Median	1.25	19.12	4.36	1.20	0.37	0.13	0.04
26–29 years (n = 82)							
Mean (SD)	1.20 (0.47)	18.41 (5.50)	5.09 (2.25)	1.29 (0.57)	0.41 (0.14)	0.16 (0.10)	0.04 (0.05)
Min	0.13	8.36	0.94	0.48	<LOD[a]	0.05	0.01
Max	3.78	40.15	12.56	4.32	0.88	0.61	0.45
Median	1.19	18.45	4.75	1.20	0.39	0.13	0.02
30–35 years (n = 70)							
Mean (SD)	1.24 (0.46)	18.95 (6.17)	5.30 (1.89)	1.31 (0.70)	0.41 (0.17)	0.19 (0.13)	0.04 (0.03)
Min	0.43	8.20	2.14	0.52	<LOD	0.03	0.01
Max	2.84	32.82	9.48	5.33	1.09	0.70	0.12
Median	1.14	18.56	5.12	1.16	0.37	0.14	0.03
≥36 years (n = 15)							
Mean (SD)	1.27 (0.34)	17.53 (5.02)	4.39 (1.22)	1.29 (0.35)	0.43 (0.11)	0.19 (0.11)	0.03 (0.01)
Min	0.80	9.18	2.43	0.79	0.28	0.07	0.01
Max	2.21	26.65	7.46	2.11	0.61	0.42	0.05
Median	1.27	17.76	4.20	1.30	0.47	0.13	0.03
Females							
≤25 years (n = 20)							
Mean (SD)	2.12 (1.28)	7.47 (2.42)	2.70 (1.09)	0.76 (0.33)	0.26 (0.12)	0.16 (0.07)	0.06 (0.03)
Min	0.35	2.29	1.17	0.301	<LOD	0.03	0.02
Max	4.63	11.95	5.68	1.72	0.55	0.33	0.12
Median	1.87	7.20	2.39	0.72	0.23	0.14	0.06
26–29 years (n = 145)							
Mean (SD)	2.25 (1.50)	8.57 (3.26)	2.79 (1.23)	0.64 (0.37)	0.25 (0.13)	0.14 (0.06)	0.05 (0.03)
Min	0.53	1.61	1.61	0.20	<LOD	0.02	<LOD
Max	10.17	21.28	21.28	2.76	0.91	0.50	0.16
Median	1.78	7.90	7.90	0.57	0.22	0.14	0.05
30–35 years (n = 82)							
Mean (SD)	2.54 (1.71)	8.29 (2.43)	2.87 (1.33)	0.72 (0.35)	0.26 (0.12)	0.16 (0.09)	0.06 (0.06)
Min	0.53	2.53	0.67	0.20	<LOD	0.04	<LOD
Max	8.50	13.88	9.78	2.00	0.71	0.72	0.46
Median	2.06	8.10	2.78	0.65	0.24	0.13	0.05
≥36 years (n = 6)							
Mean (SD)	1.80 (0.71)	8.58 (2.93)	3.29 (2.18)	0.60 (0.25)	0.21 (0.09)	0.13 (0.07)	0.03 (0.02)
Min	1.00	4.62	1.49	0.38	<LOD	0.05	0.01
Max	2.97	10.99	6.17	1.01	0.38	0.25	0.05
Median	1.59	9.92	2.19	0.52	0.18	0.13	0.03

[a] LOD — limit of detection (PFOS = 0.2 ng mL^{-1}, PFOA = 0.6 ng mL^{-1}, PFNA = 0.2 ng mL^{-1}, PFDA = 0.2 ng mL^{-1}, PFHxS = 0.06 ng mL^{-1}, PFUnDA = 0.3 ng mL^{-1} and PFDoDA = 0.07 ng mL^{-1}).

Similar results of the dominating presence of PFOS and PFOA in human samples were obtained also by other authors who tested blood of populations of other countries, i.e. in Italy: Ingelido et al. (2010), Germany: Midasch et al. (2006) or Vassiliadou et al. (2010) in Greece.

Significantly lower concentrations for both compounds were found by Liu et al. (2012) in the blood serum of men from Chinese population. However, different results were obtained by Long et al. (2012), who – studying the levels of PFASs in the blood serum of Inuit from Greenland – noted higher median concentrations of PFOS and PFOA in women (19.5 ng mL^{-1} and 1.7 ng mL^{-1}) than in men (14.9 ng mL^{-1} and 0.94 ng mL^{-1}). Differing results were obtained in the case of Inuit from Greenland in the INUENDO project, where greater, statistically significant, concentrations of PFASs were detected in the serum of men in comparison to their female partners. In men, the median of PFOS = 44.9 ng L^{-1} and PFOA = 4.5 ng mL^{-1}, and for women 20.6 ng L^{-1} and 1.8 ng mL^{-1} respectively. A similar relationship but on much lower concentration levels of PFASs was found in the population of Ukraine. In the blood serum of men from Ukraine median equal 7.6 ng L^{-1} of PFOS and 1.3 ng L^{-1} of PFOA vs. 5.1 ng L^{-1} and 0.9 ng L^{-1} for women respectively (Góralczyk et al., 2015). Other authors did not report significant differences between PFAS concentrations in the studied men and women (Harada et al., 2004; Kubwabo et al., 2005; Olsen et al., 2004).

In order to examine possible gender driven relationships between the age and PFASs serum concentration the tested population was divided into 4 age groups and checked for differences in concentrations in age group and additionally searched for any relations that may be observed. The summary statistics for selected age groups are presented in Table 4.

The Kruskal–Wallis ANOVA test and median test for four age groups showed that there are statistically significant differences between the distributions of concentrations for PFNA, PFOS, PFDA and PFDoDA when viewing the results without differentiation into sexes. Fig. 2 shows the distribution of concentrations in four age groups.

Table 5
Results of Spearman's rank correlation between age, BMI and PFASs concentration.

	Spearman's rank order correlation					
	Male		Female		Male + female	
	Age	BMI	Age	BMI	Age	BMI
PFHxS	0.015	0.049	0.007	−0.094	−0.083	**−0.289**
PFOA	0.023	−0.120	0.102	**0.153**	**0.191**	**0.421**
PFNA	0.001	−0.018	0.117	0.063	**0.179**	**0.390**
PFOS	−0.018	−0.048	0.089	0.036	**0.164**	**0.465**
PFDA	0.016	**−0.185**	0.097	0.068	**0.160**	**0.323**
PFUnDA	0.129	−0.020	0.063	0.018	**0.110**	0.067
PFDoDA	0.061	−0.030	−0.73	−0.052	−0.089	**−0.231**

In bold indicated a statistically significant result ($p < 0.05$).

However, after differentiating the results according to age and sex, no statistically significant differences were observed for the tested substances. Using the Wald–Wolfowitz, Mann–Whitney and K–S–L tests revealed that the differences in concentrations of PFNA, PFOS in women and men in four age groups are statistically significant ($p < 0.05$). The relations between PFHxS, PFOA, PFDA and age have been confirmed in three age groups of men and women, except for the youngest group (up to 26 years of age). Such relation was not observed for PFUnDA in any of the age groups. At the same time, PFDoDA shows statistically significant relation in men and women in the age group 26 to 29 years and 30 to 35 years. The Spearman's rank test showed a weak positive correlation between age and PFHxS concentration at R = 0.015. However, it is impossible to confirm that the level of this substance increases in men with age, because this relation was not statistically significant (see Table 5). Taking into account all results without separation into sexes, it was observed that the increase of PFOA is accompanied with the increase of PFHxS. Despite confirming the above relation also with tau Kendall test and Gamma statistic (Gamma coefficients), due to lack of statistical significance of these

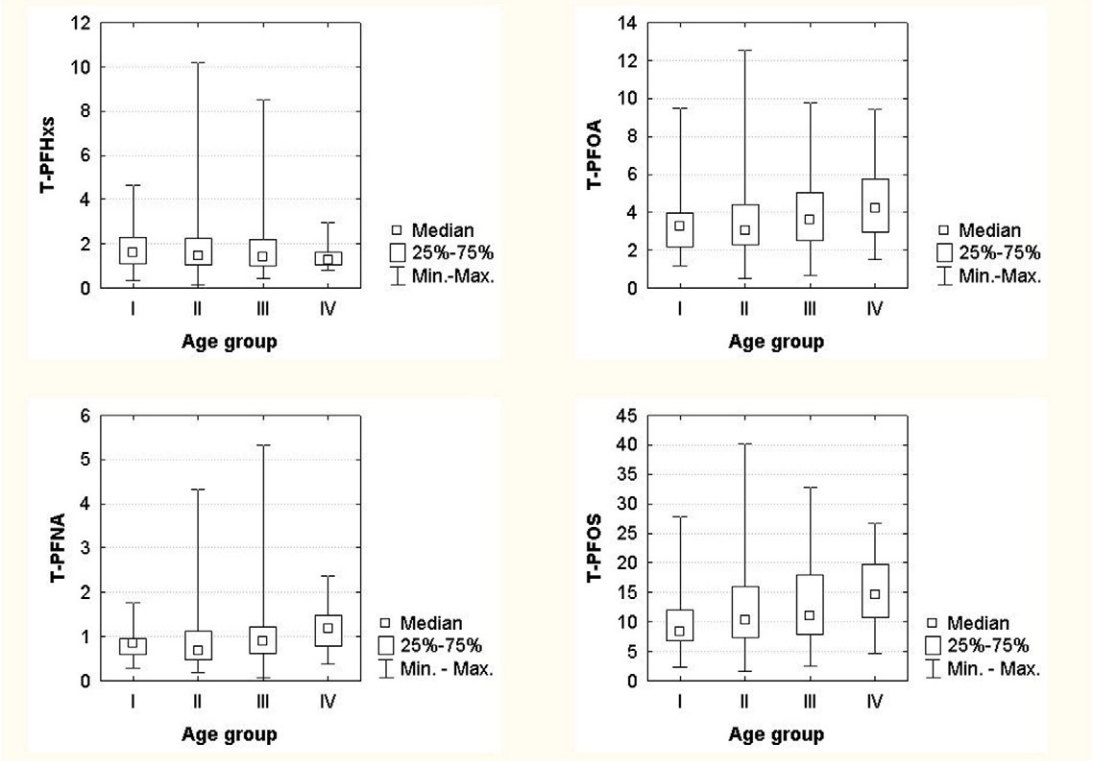

Fig. 2. Distributions of selected PFASs in four age groups in total population (T).

results, no correlation between the studied variables should be assumed. Similarly, the relation between concentrations and age were presented by Ericson et al. (2007), who reported slightly higher concentrations in the group of 55 ± 5 years of age in relation to younger groups of 25 ± 5 years old. In these studies, the average PFOS concentration amounted to 8.23 ng mL^{-1} in older and 7.06 ng mL^{-1} in younger humans, but these differences were also statistically insignificant. Lack of correlation between concentrations and the age was also presented by Axmon et al. (2014), Harada et al. (2004), Kannan et al. (2004), and Wilhelm et al. (2009).

It has to be stressed that the volunteers included in our study were not asked about the frequency of eating fish or seafood, nor about the time they spent in their houses. So it was difficult to interpret whether differences in the concentrations of test compounds may have resulted from various dietary habits, e.g. large or small intake of fish or rather of pollution of place where the donors resided. As shown by some authors food (fish) and dust are the main sources of human exposure to these compounds (D'Hollander et al., 2010; Eriksson and Kärrman, 2015; Falandysz et al., 2006b, 2007; Waszak et al., 2014).

The same statistical tests were performed to establish whether there is a relation between PFASs concentrations in blood serum of men and women and their BMI (see Table 5). The results show a weak of correlation between PFASs concentrations detected in the serum of men and women and their BMI.

Biomonitoring of the compounds adversely affecting human health is an essential tool for controlling risks arising from environmental pollution. This especially relates to compounds with proven or potential effects on the endocrine disruptors, which include PFASs. Checking the trends in the levels of these compounds in various parts of the environment, including in humans, is particularly important in the case of compounds that are banned for use because of their toxicological properties. This includes PFASs, which since 2010 are prohibited, while being ever-present in environmental samples. Constantly updated biomonitoring results are an important element used in both risk assessments together with toxicological data as points of departure (PoD) when determining the hazard quotient profiles (HQ) (Ludwicki et al., 2015) or for statistical models based on population data (age, gender) and biomarker levels of selected chemicals that can allow determination of the total concentration of certain environmental pollutants in the human body (Jain, 2015). This is particularly important in the case of PFASs, which have been shown diverse geographical distribution, as well as tissue distribution and different rates of excretion of individual PFASs in humans depending on their age and sex (Cho et al., 2015; Harada et al., 2004; Zhang et al., 2015).

4. Conclusions

This paper provides a comprehensive description of concentrations of seven selected PFASs in the blood serum of inhabitants of central Poland. Higher concentrations were observed in five (PFOS, PFOA, PFNA, PFDA, PFUnDA) of seven PFASs in men than in women of the same population. Only the concentrations of PFHxS and PFDoDA were slightly higher in women than in men. These differences were statistically insignificant in all cases apart from PFUnDA. Average concentrations and medians of all compounds were close to those reported in other European countries and lower than those reported in the Northern America. Moreover, no change was observed as to levels of concentration reported in earlier pilot studies performed in Poland on a selected group of 15 volunteers within the frames of WWF campaign (Struciński et al., 2006). The hypothesis assuming an increase of concentration with age of the studied volunteers, regardless of sex, was not confirmed.

Due to adverse health effects of exposure of humans to PFASs and introduction of limitations of their use in various industries throughout the European Union in 2010, the biomonitoring of these substances should be continued. These studies are especially important to discover whether the above-mentioned limitations resulted in the decrease of levels of individual perfluoroalkylated substances in the human organism. It also seems purposeful to perform tests to explain the mechanisms laying behind the differences in PFAS concentrations in men and women.

Conflict of interest statement

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

Abbreviations

BMI	body mass index
EFSA	European Food Safety Authority
HQ	hazard quotient
K–S–L test	Kolmogorov–Smirnov test and Lilliefors test
LC	liquid chromatography
LC/MS/MS	liquid chromatography tandem mass spectrometry
LOD	limit of detection
PFASs	perfluoroalkylated substances
PFDA	perfluorodecanoic acid
PFDoDA	perfluorododecanoic acid
PFHxS	perfluorohexane sulfonic acid
PFNA	perfluorononanoic acid
PFOA	perfluorooctanoic acid
PFOS	perfluorooctane sulfonate
PFUnDA	perfluoroundecanoic acid
PoD	point of departure
POPs	persistent organic pollutants
TDI	tolerable daily intake
WWF	World Wide Fund for Nature

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