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# Assessment of exposure to perfluoroalkyl substances (PFASs) in dogs by fur analysis☆

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ABSTRACT

Poly- and perfluoroalkyl substances (PFASs) are a large group of chemicals commonly used in various branches of industry, which may adversely affect the living organisms.

The aim of this study were to evaluate exposure of dogs to six selected PFASs: five perfluoroalkyl carboxylic acids (perfluorobutanoic acid - PFBuA, perfluoropentanoic acid - PFPeA, perfluorohexanoic acid - PFHxA, perfluoroheptanoic acid - PFHpA, perfluorooctanoic acid – PFOA) and perfluorooctane sulfonic acid (PFOS) through the analysis of fur samples. To our knowledge this is the first study concerning the use of fur samples to evaluation of exposure of domestic animals to PFASs. Relationship between PFASs concentration and age, gender and body weight of animals was also evaluated. Fur samples were collected from 30 dogs living in Olsztyn (Poland). All PFASs studied were detected in the canine fur samples.

The highest concentrations were observed in the case of PFOA and PFBuA, detected at concentrations in the range between 1.51 and 66.7 ng/g and 0.98–26.6 ng/g, respectively. During the present study generally no statistically significant differences dependent on gender, age and body weight of animals were found.

This study confirms the suitability of fur samples for biomonitoring of exposure to PFASs in domestic animals, what may be important in veterinary toxicology.

## 1. Introduction

Perfluoroalkyl substances (PFASs) are a large group of chemicals (more than 4000) built of linear or branched carbon chain, which may be fully or partially fluorinated (Buck et al., 2011). Due to their properties, and especially high thermal and chemical stability resulting from strong carbon–fluorine covalent bonds, PFASs are commonly used in the production of various everyday objects, including among others food containers, kitchenware, clothes cleaning products and electronical elements (Chohan et al., 2020).

PFASs may leach into the food of plant and animal origin, soil, water and air and therefore are now classified as the world's major environmental pollutants (Buck et al., 2011; Ghisi et al., 2019; Sunderland et al., 2019). The living organisms are mainly exposed to PFASs by eating contaminated food or drinking water (Sunderland et al., 2019). The exposure through breathing air and dust containing PFASs, hand-to mouth contact, dermal absorption or perinatal exposure (PFASs cross the placenta and may get into the fetal bloodstream) is also possible and has also some significance in toxicology (Liew et al., 2018; Poothong et al., 2019; Sunderland et al., 2019).

Till now PFASs have been detected in body fluids and tissues of humans and numerous species of domestic and wildlife animals (Guruge et al., 2008; Bost et al., 2016; Wang et al., 2018a, 2018b). It is also known that PFASs show multidirectional adverse effects on the living organisms. PFASs may penetrate across the brain barrier (Wang et al., 2018b) and strongly affect the nervous system (Mariussen, 2012). Moreover, exposure to PFASs causes negative effects in the gastrointestinal tract, as well as in the respiratory (Cui et al., 2009), reproductive (López-Arellano et al., 2019), immunological (Keil et al., 2008) and endocrine (Ballesteros et al., 2017) systems. Toxic properties of PFASs caused that many, as well as the biggest plastic producers introduced regulations restricting the use of these substances, especially long-chain

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PFASs, such as perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) (Wang et al., 2013; Ahrens and Bundschuh, 2014). Among others, PFOS and PFOA, as well as their derivatives have been included in the international Stockholm Convention to eliminate their global use (Qiao et al., 2019; Kwak et al., 2020).

It is relatively well known that pet animals, which live in the same conditions as humans and are exposed to similar environmental pollutants, may be treated as sentinels of human exposure to these pollutants. Such situation has been confirmed, among others, in the case of bisphenols benzophenone-type UV filters, triclosan and triclocarban (Karthikraj et al., 2020). Nevertheless information about a degree of exposure of pet animals to PFASs, contrary to humans, is relatively scanty. PFASs have been noted in serum of dogs in Japan (Guruge et al., 2008) and cats in the USA (Bost et al., 2016; Wang et al., 2018a). as well as in cat and dog feces in the USA (Ma et al., 2020). Moreover, it is known that one of the source of PFASs acting on pet animals may be the food package (Chinthakindi et al., 2021), Although it is known that pet animals are exposed to PFASs, the role of these substances as a toxin and disease factor in dogs is unknown.

Toxicological analysis of hair in humans or fur in animals is becoming increasingly important in the last years, due to ethically important non-invasive sample collection, as well as easy transport and storage of probes. Simultaneously it is known that toxicological analysis of hair is a good alternative for blood samples examinations, because it is characterized by similar reliability and sensitivity and allows to determine of integral exposition to environmental pollutants (Alves et al., 2015). Moreover, previous studies have shown that the evaluation of PFASs levels in hair and fur is a good method to determine a degree of exposure of the living organisms to these substances (Martín et al., 2019).

Therefore, the aim of this study was to evaluate the exposure of dogs to selected PFASs by evaluation of levels of these substances in fur samples. This investigation includes six substances from PFASs group. PFOA and PFOS are the most extensively used in the industry and their toxic influence on the living organisms is relatively well documented (Buck et al., 2011). Other substances included into this study, such as perfluorobutanoic (PFBuA), perfluoropentanoic (PFPeA), perfluorohexanoic (PFHxA) and perfluoroheptanoic (PFHpA) acids, are less investigated. Nevertheless they have been previously found in hair (Martín et al., 2019, 2016a; Ruan et al., 2019), as well as in other biological (Wang et al., 2018b; Martín et al., 2016b, 2016c) and environmental (Martín et al., 2019; Abril et al., 2020) samples and play important roles in toxicology. Moreover, according with Martín et al. (2019) PFASs of short chain (PFBuA, PFPeA and PFHxA) are less frequently detected than those of longer chain in hair samples but, when detected, they are present at higher concentrations than those of longer chain. Furthermore, because of the environmental concern of PFOS and PFOA, the 3M Company voluntarily phased out the production in 2000, and replaced them with shorter-chain chemicals (Ahrens and Bundschuh, 2014).

To knowledge of the authors, the present study is the first such comprehensive evaluation of a degree of dogs exposure to PFASs in Europe. This is also the first study showing PFASs levels in the canine fur as a method of the estimation of exposure of dogs to these substances.

## 2. Materials and methods

### 2.1. List of chemical reagents used in the study

All reagents were analytical grade unless otherwise specified. Acetic acid (HAc), ammonium acetate and sodium dodecylsulfate (SDS) were obtained from Panreac (Barcelona, Spain). HPLC-grade acetone, methanol and water were supplied by Romil (Barcelona, Spain). The PFASs PFBuA (98%), PFPeA (97%), PFHxA (≥97%), PFHpA (99%), PFOA (96%) and PFOS (≥98%) were supplied from Sigma-Aldrich (Steinheim, Germany). The internal standard (IS) perfluorooctanoic acid-$^{13}C_4$ (PFOA-$^{13}C_4$) (99%) was supplied by Cambridge Isotope Laboratories (MA, USA). Individual stock standard solutions were prepared at 1000 mg/L in MeOH and stored at −18 °C. Working solutions were prepared by dilution of the stock standard solutions in methanol.

### 2.2. Fur samples collection and preparation to analysis

Fur samples were collected from 30 female and male dogs of various breeds, age and body condition score (BCS) living in Olsztyn – the city in northeastern Poland with about 170 thousands of residents. Samples were collected from November to December of 2019. The exact data concerning dogs included in the study are summarized in Supplementary material – Table S1.

Additionally, to answer the questions if the PFASs levels in the fur correlate with factors such as age, body condition score or gender of animals dogs included into the study were split depending on gender for two groups: male (n = 9) and female (n = 21), depending on age – for young - aged up to 3 years (n = 9), middle-aged from 3 to 10 years (n = 13) and old – aged over 10 years (n = 8), and depending on body condition score (BCS), according to international canine body condition score system (Chun et al., 2019) on too thin animals (BCS points 1–3, n = 5), dogs with physiological weight (BCS points 4–5, n = 12) and overweight dogs (BCS points 6–9, n = 13).

Collection of fur samples was performed according to Act for the Protection of Animals for Scientific or Educational Purposes of 15 January 2015 (Official Gazette 2015, No. 266), applicable in the Republic of Poland. Based on this act, due to the fact that fur samples collection was performed during veterinary or beauty treatment and it was a non-invasive procedure, approval from the Bioethical Committee for the present study was not required.

About 2 g of fur were cut from the abdomen as close to the skin as possible. Immediately after collection fur samples were wrapped in aluminium foil and stored in the dark in room temperature until further studies (no longer than four weeks). During storage and analysis fur samples have not been in contact with plastic containers, wax paper, fabrics or other objects that could contaminate the samples with PFASs.

Before analysis exogenous substances and chemicals absorbed on the fur surface were removed through the fourfold washing: at first in ultrapure water, then with SDS (0.1%, w/v) and finally twice with ultrapure water. In each washing step the sonication of fur samples for 5 min was performed. Then fur samples were cut into small fragments with a length of about 2–3 mm. These fragments were dried and stored in aluminium foil at room temperature until further studies (no longer than three days).

### 2.3. Extraction of PFASs from fur samples and analysis of their levels

To determine PFASs levels in canine fur samples the method described previously by Martin et al. (Martín et al., 2016a) was used with slightly modifications. An aliquot of the fur sample (100 mg) containing the IS (12.5 ng) was weighed in an 10 mL screw-cap glass centrifuge tubes, and incubated with 2 mL of a mixture of methanol/HAc (85:15, v/v) at 38 °C for 12 h. After cooling to room temperature, 3 mL of acetone was added for the extraction of analytes. The mixture was sonicated for 15 min and centrifuged for 10 min at 2900 × *g*. The organic phase containing the analytes was separated into a clean tube and evaporated to dryness under a nitrogen stream at room temperature. The residue was reconstituted in 0.25 mL of methanol and filtered through a 0.22 μm nylon filter. A 10 μL aliquot of the extract was injected into the LC instrument.

Liquid chromatography-tandem mass spectrometry analysis was performed using a HALO C-18 Rapid Resolution (50 × 4.6 mm i.d., 2.7 μm particle size) column. Chromatographic separation was carried out by gradient elution with 10 mM ammonium acetate solution (solvent A) and methanol (solvent B) at a flow rate of 0.6 mL/min. The gradient program was as follows: 0–14 min, linear gradient from 28 to 70% of

solvent B, from 70% to 80% of solvent B in 5 min, and then increased to 100% in 6 min and held for 2 min. Column temperature was maintained at 30 °C. Two multiple reaction monitoring (MRM) transitions were selected for each analyze for quantification and confirmation of compounds. Mass spectrometer settings and validation parameters are summarized in Supplementary material Tables S2 and S3.

### 2.4. Quality assurance and quality control

To guarantee reliable and precise results a quality assurance/quality control (QA/QC) protocol was established. This protocol involves the use of control spiked samples, solvent (methanol) injections, standards containing a mixture of the target compounds (20 ng/mL) and procedural blanks (processed in the same way as the samples) into each analytical batch (15 samples). No quantifiable amounts of target compounds were detected in blank samples. Due to the lack of certified reference materials, in-house reference materials (prepared by spiking real samples at 10, 50 and 100 ng/g levels) are used to check for accuracy during validation and QA/QC (recovery rate range 85–103%).

### 2.5. Statistical analysis

The statistical analysis was performed using Student's t-test (Statistica 13.3, StatSoft, Inc., Cracow, Poland). The differences were considered statistically significant at $p < 0.05$.

## 3. Results

The presence of PFASs was noted in the fur samples from all dogs included into the study (Table S1, Table 1).

The highest levels were observed in the case of PFOA (Table 1). PFOA was present in all samples and its levels were highly diverse in the fur of particular dogs. PFOA levels fluctuated from 1.51 ± 0.05 ng/g to as high as 66.7 ± 1.96 ng/g. The other PFAS, observed in fur samples from all dogs, which levels were relatively high was PFBuA (Table S1, Table 1). The levels of PFBuA ranged from 0.98 ± 0.09 ng/g to 26.6 ± 3.07 ng/g (Table S1). In turn, the levels of PFPeA and PFHxA above the method quantification level (MQL) were evaluated in 29 dogs of the 30 animals included into the experiment. In one dog the levels of these substances were below the MQL (0.6 ng/g) (Table S1). In the remaining dogs the PFPeA levels fluctuated from 0.74 ± 0.15 ng/g to 15.6 ± 0.50 ng/g, and PFHxA levels from 0.65 ± 0.01 ng/g to 18.9 ± 3.04 ng/g (Table 1). The lower levels were noted in the case of PFHpA, which was below the MQL in five animals (Table S1). In remaining dogs PFHpA levels ranged from 0.65 ± 0.10 ng/g to 13.2 ± 0.19 ng/g (Table S1, Table 1). In turn, PFOS levels were the lowest and they were below the MQL in 5 animals, and in remaining dogs amounted to from 0.62 ± 0.09 ng/g and 0.62 ± 0.1 ng/g to 1.63 ± 0.14 ng/g (Table S1, Table 1).

During the present study, differences in levels of all PFASs between male and female dogs (Table 2) were not statistically significant (at $p < 0.05$). Similar situation was noted in the case of relationship between PFASs levels and age of animals (Table 2).

The levels of majority PFASs studied also did not depend on the weight of the animals. In the vast majority of cases differences in levels of PFOSs between animals belonging to various groups formed according to canine BCS scale were not statistically significant (at $p < 0.05$) (Table 2). The only exception was the level of PFHpA, which in overweight animals amounted to 3.18 ± 0.99 ng/g and was statistically significantly higher than that noted in dogs with correct weight (0.85 ± 0.25 ng/g) (Table 2).

**Table 2**
Comparison of concentration levels (ng/g) of PFASs in particular groups of dogs: male (n = 9), female (n = 21), young (aged up to 3 years, n = 9), middle-aged (from 3 to 10 years, n = 13), old (aged over 10 years, n = 8), too thin (BCS points 1–3, n = 5), with physiological weight (BCS points 4–5, n = 12) and too heavy (BSC points 6–9, n = 13). Values are presented as mean ± standard error of the mean (SEM).

| Groups of animals | PFASs | | | | | |
|---|---|---|---|---|---|---|
| | PFBuA | PFPeA | PFHxA | PFHpA | PFOA | PFOS |
| Male | 13.11 ± 2.79 | 5.51 ± 0.99 | 7.35 ± 1.73 | 1.44 ± 1.01 | 25.84 ± 5.63 | 0.77 ± 0.14 |
| Female | 10.45 ± 1.45 | 5.22 ± 0.90 | 7.48 ± 1.30 | 2.35 ± 0.66 | 19.17 ± 3.07 | 0.72 ± 0.10 |
| Young | 13.41 ± 2.33 | 6.02 ± 1.07 | 6.18 ± 1.20 | 1.11 ± 0.31 | 22.38 ± 6.01 | 0.74 ± 0.14 |
| Middle-aged | 10.78 ± 2.17 | 4.98 ± 1.14 | 8.32 ± 1.98 | 2.61 ± 0.96 | 21.52 ± 4.66 | 0.70 ± 0.15 |
| Old | 9.56 ± 2.28 | 5.64 ± 1.49 | 7.44 ± 1.82 | 2.30 ± 0.77 | 19.25 ± 2.97 | 0.78 ± 0.14 |
| BCS pt. 1-3 | 14.81 ± 2.95 | 5.14 ± 1.09 | 6.12 ± 1.69 | 1.71 ± 0.53 | 28.35 ± 10.47 | 0.68 ± 0.31 |
| BCS pt. 4-5 | 9.25 ± 1.73 | 3.99 ± 0.92 | 5.44 ± 1.02 | 0.85 ± 0.25* | 16.43 ± 2.85 | 0.76 ± 0.12 |
| BCS pt. 6-9 | 11.71 ± 2.27 | 6.56 ± 1.23 | 9.79 ± 1.96 | 3.18 ± 0.99* | 22.79 ± 4.16 | 0.84 ± 0.11 |

Statistically significant differences ($p \leq 0.05$) are marked with *.

## 4. Discussion

The presence of PFASs in fur samples of all animals included into this study confirms that these substances are widely distributed in the environment, and living organisms (including humans, wildlife and domestic animals) are exposed to their impact (Cai et al., 2012; Groffen et al., 2018; Toms et al., 2019). It is relatively well known that PFASs are present in the water, food products, soil, air and liquids and tissues of the living organisms around the world, and the degree of contamination of the environment with these substances clearly depends on the part of the world (Guruge et al., 2008; Martín et al., 2019). However, it should be underlined that information about the presence of PFASs in the environment in Poland is much more limited. Summary of previous studies concerning the distribution of PFASs included into the present study in Poland is presented in Table 3 and S4.

The levels of PFASs noted in the canine fur during the present study are relatively high in comparison to previous investigations on humans and wildlife animals in Poland (Table 3). It should be noted that Olsztyn city, where samples were collected, is not located in highly industrialized areas. However, some industrial plants (first of all rubber and furniture industry) exist in this city.

The concentration levels of PFASs observed in the present

**Table 1**
Concentration values (ng/g) and frequency of detection of PFASs in the analyzed fur samples (n = 30) – cumulative data.

| Compounds | Range (ng/g) | Arithmetic mean (ng/g)[a] | Geometric mean (ng/g)[a] | Median (ng/g)[a] | Frequency of detection (%) |
|---|---|---|---|---|---|
| PFBuA | 0.98–26.6 | 11.2 | 8.63 | 9.66 | 100 |
| PFPeA | <0.14–15.6 | 5.31 | 3.77 | 4.39 | 96.7 |
| PFHxA | <0.14–18.9 | 7.45 | 4.97 | 5.92 | 96.7 |
| PFHpA | <0.14–13.2 | 2.10 | 1.20 | 1.35 | 86.7 |
| PFOA | 1.51–66.7 | 21.2 | 16.0 | 17.4 | 100 |
| PFOS | <0.14–1.63 | 0.76 | 0.62 | 0.70 | 86.7 |

[a] Samples with PFASs levels < MQL are considered as MDL/square root of 2 (MQL: 0.90 ng/g for PFBuA and 0.60 ng/g for the rest of compounds); MDL: Method detection limit (0.30 ng/g for PFBuA and 0.20 ng/g for the rest of compounds)).

**Table 3**
Overview of biomonitoring studies of PFASs included into the present study in the living organisms in Poland.

| Matrix | PFOA | PFOS | PFHpA | PFHxA | PFPeA | References |
|---|---|---|---|---|---|---|
| **Human** | | | | | | |
| **Human blood serum** (ng/mL) – mean value or range depending on reference, F- female, M-male (in references, where gender is described) | 1.2–4.6 | 4.4–14 | 0–0.22 | n.d. | | Struciński et al. (2006) |
| | 5.3 ± 0.2 | 17.7 ± 0.5 | | | | Leter et al. (2014) |
| | 1,2–8.7 | 5.2–84 | 0.05–0.79 | <0.02–2.4 | | Falandysz et al. (2006b) |
| | 3.7 | 34 | 0.069 | 0.033 | | Falandysz et al. (2006a) |
| | 11-40(M)<br>9.7–34 (F) | 21-116 (M)<br>16-60 (F) | | | | Kannan et al. (2004) |
| | 2–16 | 8–40 | | | | Specht et al. (2012) |
| | 1.48–16(M) | 8.20–40.2(M) | | | | Lindh et al. (2012) |
| | 4.8 | 18.5 | | | | Toft et al. (2012) |
| | 1.5–16 (M), 0.5–9.8 (F) | 8.2–40.2 (M),<br>1.6–21.3 (F) | | | | Ludwicki et al. (2015) |
| | 0.94–12.56 (M),<br>0.67–9.83 (F) | 8.20–40.14 (M)<br>1.61–21.28 (F) | | | | Góralczyk et al. (2015a) |
| | 1.6–4.9 (M)<br><0.6–9.8 (F) | 8.2–40.2 (M)<br>1.6–21.3 (F) | | | | Góralczyk et al. (2015b) |
| | 1.5–4.3 (F) | 5.2–12.1 (F) | | | | Lyngsø et al. (2014) |
| | 1,34–2.51 (F) | 4.38–12.40 (F) | 0.02–0.60 (F) | | | Lenters et al. (2016) |
| | 4.81–5.89 (M) | 17.19–20.88 (M) | | | | Kvist et al. (2012) |
| **Wildlife animals** | | | | | | |
| **Whole blood** (ng/mL): | | | | | | |
| Baltic cod | 0.05–0.7 | 6.1–52 | <0.05–0.74 | <0.05–0.69 | | Falandysz et al. (2006b) |
| | 0.09 | 15 | 0.09 | 0.16 | | Falandysz et al. (2006a) |
| Eider duck | 0.06–0.1 | 12–38 | <0.05 | <0.05 | | Falandysz et al. (2006b) |
| | 0.1 | 22 | <0.05 | <0.05 | | Falandysz et al. (2006a) |
| **Whole blood** (pg/mL) | | | | | | |
| Velvet Scoter | 0.09–0.56 | 4.8–14 | <0.05 | <0.05 | | Falandysz et al. (2007) |
| Long-tailed Duck | 0.25–1.8 | 6.7–54 | <0.05 | <0.05 | | Falandysz et al. (2007) |
| Razorbill | <0.05–0.30 | 23–39 | <0.05 | <0.05 | | Falandysz et al. (2007) |
| Red-throated Diver | 0.17–0.85 | 40–200 | <0.05 | <0.05 | | Falandysz et al. (2007) |
| **Liver** (ng/g) | | | | | | |
| Beaver | 0.13 | 6.6 | 0.01 | <0.16 | | Falandysz et al. (2006a) |
| | 0.06–0.28 | 1.6–39 | <0.01–0.02 | 0.03–0.23 | | Falandysz et al. (2007) |
| | 0.51–0.87 | <0.126–8.45 | n.d. | n.d | n.d. | Surma et al. (2015) |
| White-tailed sea eagle | n.d. | <3.9–127 | | | | Kannan et al. (2002) |
| **Other tissues of beaver** (ng/g) | | | | | | |
| brain | 0.5–1.59 | <0.126–0.86 | n.d | n.d. | n.d. | |
| tail | 0.7–1.06 | <0.126–1.34 | n.d | n.d. | n.d. | |
| peritoneum | 0.66–1 | <0.126–4,64 | n.d. | n,d | n.d. | |
| subcutaneous adipose | 0.55–1.04 | <0.126–3.65 | n.d | n.d. | n.d. | Surma et al. (2015) |
| **Domestic animals** | | | | | | |
| **Whole blood** (ng/mL) | | | | | | |
| Cattle | <0.05–0.06 | 0.52 | <0.05 | 0.05 | | Falandysz et al. (2006a) |

n.d.: no detected.

investigation are extremely higher that those noted in the cattle in Poland (Table 3). It is in agreement with previous observations, conducted in Japan, where concentrations of PFOS and PFOA in dogs were several-fold higher than in farm animals (Guruge et al., 2008). It is probably connected with the fact that dogs live in the immediate vicinity of human, spend much of their lives indoors and are exposed to higher concentration of PFASs in toys, food, bedrolls and things used by humans. On the other hand, it is known that PFASs are present in household air and dust (Shoeib et al., 2011). The hypothesis about similar exposure of humans and dogs or cats to PFASs is confirmed by

studies performed in Japan and USA, in which considerable similarities between concentration of PFASs in blood serum of these mammal species have been noted (Taniyasu et al., 2003; Guruge et al., 2008; Wang et al., 2018a). Apart from household dust and air, the second most important source of PFASs may be connected with food. It is known that PFASs are present in human food package (Schaider et al., 2017), as well as in pet food and food package (Chinthakindi et al., 2021).

On the other hand, concentrations of PFASs noted in the present study greatly differed from results previously obtained in the blood serum of humans living in Poland (Table 3). These differences consisted in the higher concentration of PFOA, PFHxA and PFPeA and extremely lower levels of PFOS in fur samples collected from dogs. The reasons of these differences may be caused by various factors, including different places of residence, various evaluated matrix (blood serum in humans and fur in dogs) or other unidentified factors, such as using of particular PFASs to production of canine toys, cosmetics, food containers or bedrolls. It is known that in spite of the fact that hair analysis is becoming an increasingly common method of determining of exposure to PFASs, some differences in the concentration of these substances between serum and hair have been described. Namely, in the light of previous studies lower concentration of PFOA and PFOS (which are the most extensively studied), as well as other PFASs has been noted in hair compared to blood serum, and the degree of differences depend on the type of substance (Li et al., 2013; Wang et al., 2018c, 2018d).

Comparing the results of this study with previous investigations on pet animals, the differences are also visible. However, it should be underlined that previous data concerning PFASs levels in the accompanying animals are extremely scanty and they involve only PFAS, PFOA and PFHpA (Guruge et al., 2008; Bost et al., 2016; Wang et al., 2018a). Concentrations of PFASs noted in the present study are clearly differ from those noted in the canine blood serum in Japan, where concentration of PFOS and PFOA amounted to 12–57 ng/mL and 0.12–9.4 ng/mL, respectively (Guruge et al., 2008). Clearly differences are also visible between results obtained during the present study and studies on concertation of PFASs in cats in the USA (Bost et al., 2016; Wang et al., 2018a). Namely, Wang et al. (2018a), have reported that concentration of PFOA, PFOS and PFHpA in feline blood serum amounted to 3.10–13.5 ng/mL, 0.84–3.10 ng/mL and 0.12–0.50 ng/mL, respectively. Other studies have described the geometric mean of concentration of PFOA and PFOS at the levels of 3.28 ng/mL and 8.89 ng/mL (Bost et al., 2016). Discrepancies between the results obtained during the present study and above mentioned previous investigations concerning accompanying animals may result from significant differences in the environment, in which the animals lived. It should be pointed out that differences in PFASs concentration in humans between Poland (Table S4), the USA (Wang et al., 2018b) and Japan (Taniyasu et al., 2003) are also visible.

In the light of results obtained during the present study it may be assumed that exposure to PFASs has some importance in veterinary medicine of dogs as a cause of various diseases and disturbances in spite of the fact that all animals included into the present study were healthy. So far there are no studies on correlations between PFASs concentrations and risk of diseases in dogs. Previous studies have described correlations between exposure to PFASs and the increase in the risk of kidney diseases, respiratory disturbances and/or hyperthyroidism in cats (Bost et al., 2016; Wang et al., 2018a) In turn, in humans, the higher concentration of selected PFASs may be among others correlated with higher risk of cancer, disturbances in reproduction, endocrine disruption, diabetes, gastrointestinal diseases and/or hepatic steatosis (Kvist et al., 2012; Seo et al., 2018; Steenland et al., 2018; Jin et al., 2020). However, metabolism of PFASs and their adverse effects are differ in various species (Pizzurro et al., 2019) and it is not known whether similar influence of PFASs are present also in dogs.

Previous studies have reported that PFASs concentrations depend on gender of the individual (Calafat et al., 2007). The majority of studies have shown the higher concentration of PFASs in males, what is probably connected with the fact that females have a higher hormonal - dependent clearing rate. This fact has been confirmed in the case of PFOA on rats and dogs (Hanhijarvi, 1998; Kudo et al., 2002). Studies performed on humans are not conclusive in this term. Some of them have shown the higher concentration of PFASs in men (Kannan et al., 2004; Góralczyk et al., 2015a, 2015b), but other have no reported gender-dependent differences (Martín et al., 2019) or reported higher concentration of PFASs in females (Ruan et al., 2019). Some authors claim that the lower levels of PFASs in females than in males may be connected (besides mentioned above hormonal differences) with the fact that PFASs may be excreted through childbirth, and breastfeeding (Harada et al., 2005; Kim et al., 2019). During the present study PFAS levels in females were slightly lower that those noted in males, what confirms previous observations, but observed differences were not statistically significant.

The correlations between PFASs concentration and age or weight are also not clear. Some previous studies have shown such correlations (Guruge et al., 2008; Kim et al., 2019; Nguyen et al., 2019; Geiger et al., 2021), but the presence or absence of them clearly depends on the type of PFASs studied, part of the world and biological material investigated (Kim et al., 2019). In humans some studies have described that the concentration of PFAS is higher in children under the age of ten, decreases in adolescents and again increases after the age of twenty, and then the positive correlation between age and PFASs levels are visible (Kim et al., 2019). It is supposed that higher concentration of PFASs in early childhood is connected with exposure to these substances through gestation and breastfeeding (Kang et al., 2016; Kim et al., 2019). Decrease in the PFASs concentration in adolescents may be connected with intensively, rapid growth and “dilution” of these substances in the body (Kim et al., 2019). Further increase in the levels of PFASs in direct proportion to age in adults (in which the growth of organisms is completed) probably results from the ability of these substances to accumulation in tissues (Kim et al., 2019). In turn, the higher levels of PFASs in individuals with higher body weight, which have been observed both in humans and farm animals (Guruge et al., 2008; Jain and Ducatman, 2019). It may result from the activity of PFASs known to cause the liver steatosis, endocrinal disruption and obesity (Ballesteros et al., 2017) or on the other side from the fact that heavier individuals eat larger quantities of food, which seems to be a main source of PFASs getting inside the body (Sunderland et al., 2019). During the present study slight differences in the concentrations of PFASs between animals of different ages and with different weights have been noted. They seem to support mentioned above thesis concerning correlation between PFASs levels and age and/or weight, however the majority of differences in the concentration of PFASs in the particular groups of animals noted in the present study are not statistically significant.

It should be underlined that evaluation of the degree of exposure to PFASs through the hair analysis is a relatively new method, which until now has been used mainly in humans (Table S5), but it seems to be useful to evaluation of the degree of exposure of organisms to these substances (Gao et al., 2014; Jian et al., 2018). Probably PFASs get to the hair through the blood, secretion of sweat and/or sebum, as well as from the external environment, but the exact mechanism of PFASs migration to the hair is unknown (Jian et al., 2018). The major advantages of this method result from its non-invasive character and easiness of samples collection, storage and transport. On the other hand the exact determination of correlation between the levels of PFASs in canine fur with levels of this substances in blood serum in this animal species, as well as with PFASs concentration in household air and dust would help to better understand of all aspects connected with exposure of dogs to PFASs. Therefore further studies on these issues are needed.

## 5. Conclusion

The present study is the first application of the fur analysis to evaluate the exposure of dogs to PFASs. All PFASs studied in the present investigation were noted in the fur samples. The highest concentrations

were noted in the case of PFOA and PFBuA, detected at concentration in the range between 1.51 -66.7 ng/g and 0.98–26.6 ng/g, respectively. Generally no statistically significant differences dependent on gender, age and body weight of animals were found. Moreover, present study has confirmed that fur analysis may be used in the evaluation of exposure of dogs to PFASs, and as a non-invasive method, it may be important in veterinary medicine to evaluate the correlation between PFASs concentration and health status of dogs.

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## Author statement

Conceptualization - SG; Investigation - KM, JM, IA, JLS, EA; Methodology - KM, JM, IA, JLS, EA; Supervision - SG; Writing – original draft – KM, SG; Writing - review & editing – SG, JM, AR.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.117435.

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