DIETARY EXPOSURE TO PFOS AND PFOA IN THE AUSTRIAN POPULATION

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Introduction

Perfluoroalkylated substances (PFAS) are a group of fluorinated compounds, which are widely used in industrial and consumer applications including coating for textiles, carpets, packaging material, cleaning and floor polishing agents, paints, fire-fighting foams, and insecticides. Owing to their persistent and bioaccumulative properties, PFAS are found ubiquitously in the environment, food and tissues of animals and humans. Within the PFAS, perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) are the most important and extensively studied substances.

Several studies have indicated adverse health effects in experimental animals e.g. hepatotoxicity, developmental toxicity, neurobehavioral toxicity, immunotoxicity, reproductive toxicity, lung toxicity, hormonal effects, as well as a weak genotoxic and carcinogenic potential. In 2008, the European Food Safety Authority (EFSA) established a tolerable daily intake for PFOA of 1.5 µg/kg bw per day and for PFOS of 150 ng/kg bw per day[1].

According to Commission Recommendation on the monitoring of perfluoroalkylated substances in food (2010/161/EU[2]) monitoring of the presence of PFAS was performed during 2010 and 2011. In the present study, the occurrence of PFOS and PFOA in food originating from Austria was investigated for the first time. Based on occurrence and consumption, dietary intakes to PFOS and PFOA were estimated for various Austrian population groups. Finally, estimated intakes were compared to the respective tolerable daily intake (TDI).

Materials and methods

Sampling of meat, offal and fish was performed in slaughterhouses and aquaculture facilities by the veterinary authorities to allow clear assignment of origin. Vegetables, eggs or cheese from regional production were collected by official food inspectors in the provinces of Austria.

Analysis of perfluorinated compounds was performed at the Environment Agency Austria by means of liquid chromatography tandem mass spectrometry (LC-MS/MS) after extraction and clean-up. Briefly, 1 g of sample was spiked with an isotopically labelled surrogate standard, and extracted with acetonitrile for 30 min in an ultrasonic bath. The extract was centrifuged, and the supernatant was concentrated under a gentle stream of nitrogen. The extract was cleaned with dispersive graphitised carbon after addition of glacial acid and centrifuged again.
Quantitative analysis of PFOS and PFOA was performed using a HP1200 HPLC system (Agilent Technologies, Vienna, Austria) connected to a 4000 QTRAP triple quadrupole mass spectrometer (Applied Biosystems, Darmstadt, Germany) using MRM (multiple reaction monitoring) mode. Separation was achieved using a 200 x 2 mm Luna C18(2) column (Phenomenex, Aschaffenburg Germany) with 5 µm particle size. The mobile phase consisted of methanol and water modified with 10 mM ammonium acetate. The gradient elution started with 5% A for 2 min, followed by a 8 min linear gradient to 98% B, then 6.5 min hold at 98% B, and returned back to 5% A in 0.5 min (hold 8 min) at a flow rate of 250 µL/min. The injection volume was 10 µL and the column temperature was set to 25°C. Limit of detection (LOD) and limit of quantification (LOQ) were calculated for each sample. LODs for PFOA and PFOS ranged from 0.11 to 0.36 µg/kg and from 0.10 to 0.32 µg/kg, respectively. LOQs for PFOA and PFOS were in the range of 0.22 to 0.73 µg/kg and 0.2 to 0.92 µg/kg, respectively.

For dietary intake assessment, left-censored data were treated considering two scenarios. Lower bound (LB) concentrations were calculated by setting analytical results below LOD or LOQ at zero. For upper bound concentrations it was assumed that analytical results below LOD or LOQ are equal to the respective limit[3].

Dietary intakes were estimated using national food consumption data from a survey conducted within the scope of the Austrian Nutrition Report 2008[4]. Food consumption data of different population groups were used: females and men aged 19-65 years and children aged 6-15 years. Mean body weights were 63.6 kg for women, 81.5 kg for men and 39.7 kg for children.
A deterministic approach[5] was chosen to estimate dietary intake to PFOS and PFOA. Point estimates were calculated by multiplying the mean concentrations of food by the mean amount of food consumed by the respective population groups.

Results and discussion

Levels of PFOS and PFOA in food

A total of thirty seven samples of food originating from Austria from different food groups like cheese, eggs, fish, meat, liver and vegetables was analysed for PFOA and PFOS. PFOA (38%) was more frequently quantified in food samples than PFOS (16%). Measurable concentrations of PFOA were determined in hard cheese, hen´s eggs, fish, meat of pork and veal, venison, liver of pork, bacon, potatoes and sweet corn. In meat of lamb and poultry no PFOA was found. The highest concentrations were measured in meat of veal followed by hard cheese and hen´s eggs. The quantified results for PFOA ranged from 0.43 to 0.92 µg/kg. PFOS was only determined in fish and liver of pork and lamb with maximum concentration levels of 0.82 and 1.5 µg/kg, respectively. No quantified results were found in the other food groups. All details related to the occurrence of PFOA and PFOS in Austrian food are provided in Table 1.

Table 1: Mean and maximum concentrations of PFOA and PFOS in different food groups expressed in µg/kg

		PFOA				PFOS			
Food group	**N**	**N>LOQ**	**Mean LB**	**Mean UB**	**Maximum**	**N>LOQ**	**Mean LB**	**Mean UB**	**Maximum**
Hard cheese	3	2	0.41	0.48	0.77	0	0.00	0.10	<LOD
Hen´s eggs	6	2	0.20	0.36	0.76	0	0.00	0.13	<LOD
Fish	6	2	0.20	0.36	0.61	4	0.38	0.42	0.82
Meat	8	2	0.18	0.38	0.92	0	0.00	0.10	<LOD
Pork	3	1	0.17	0.37	0.50	0	0.00	0.10	<LOD
Lamb	2	0	0.00	0.20	<LOD	0	0.00	0.10	<LOD
Veal	3	1	0.31	0.51	0.92	0	0.00	0.10	<LOD
Poultry	4	0	0.00	0.25	<LOQ	0	0.00	0.10	<LOD
Venison	2	1	0.25	0.35	0.50	0	0.00	0.10	<LOD
Liver (pork, lamb)	2	1	0.27	0.37	0.54	2	1.21	1.21	1.50
Bacon	3	1	0.16	0.36	0.48	0	0.00	0.10	<LOD
Vegetables and vegetable products (potatoes, potato chips, sweet corn)	3	3	0.46	0.46	0.48	0	0.00	0.10	<LOD
Total	37	14	0.21	0.37	0.92	6	0.13	0.22	1.50

N=number of samples, LOQ=limit of quantification, LB=lower bound, UB=upper bound

Estimated dietary exposure

Table 2 shows the estimated dietary intake to PFOA and PFOS for different Austrian population groups. For children, the mean dietary exposure of PFOA ranged from 0.25 ng/kg bw per day (LB) to 0.55 ng/kg bw per day (UB). For PFOS, mean dietary exposure of 0.09 (LB) and 0.24 ng/kg bw per day (UB) was calculated. The mean

estimated dietary intake for women was in the range from 0.20 to 0.46 ng/kg bw per day (LB – UB) for PFOA (LB) and 0.07 to 0.20 ng/kg bw per day for PFOS. Men`s average intake to PFOA was between 0.22 ng/kg bw per day (LB) and 0.50 ng/kg bw per day (UB). For PFOS, mean dietary intake ranged from 0.05 ng/kg bw per day (LB) to 0.19 ng/kg bw per day (UB).
Results obtained in the current study are in line with several other exposure assessments which also reported dietary intake to PFOA and PFOS in a low ng/kg bw range[6-8].

Table 2: Estimation of PFOA and PFOS dietary intake through different foods based on the mean consumption and mean occurrence expressed in ng/kg bw/d (lower bound - upper bound)

	Consumption (g/d)			Daily intake (ng/kg bw/d)*					
				PFOA			PFOS		
Food group	**Children**	**Women**	**Men**	**Children**	**Women**	**Men**	**Children**	**Women**	**Men**
Hard cheese	2.1	4.9	5.3	0.02-0.03	0.03-0.04	0.026-0.031	0-0.005	0-0.01	0-0.01
Hen´s eggs	17.3	16.8	17.2	0.09-0.16	0.05-0.10	0.04-0.08	0-0.06	0-0.03	0-0.03
Fish	8.3	10.6	8.5	0.04-0.08	0.03-0.06	0.02-0.04	0.08-0.09	0.06-0.07	0.040-0.043
Meat (pork, lamb, veal)	19.7	26.9	54.1	0.09-0.19	0.07-0.16	0.12-0.25	0-0.05	0-0.04	0-0.07
Poultry	13.0	23.3	23.4	0-0.08	0-0.09	0-0.07	0-0.03	0-0.04	0-0.03
Venison	0.1	1.0	1.3	0.0005-0.001	0.004-0.005	0.004-0.005	0-0.0002	0-0.002	0-0.002
Liver (pork, lamb)	0.2	0.5	0.4	0.001-0.002	0.002-0.003	0.001-0.002	0.006	0.01	0.006
Bacon	1.9	2.0	5.3	0.01-0.02	0.005-0.01	0.01-0.02	0-0.005	0-0.003	0-0.006
Total	62.6	85.8	115.4	0.25-0.55	0.20-0.46	0.22-0.50	0.09-0.24	0.07-0.20	0.05-0.19

* When lower and upper bound of the range of daily intake are coincident, only one number is presented

In order to exclude the influence of left censored data, the lower bound exposure was used to determine the relative contribution of various food groups to the total dietary intake. The major contributor to PFOA exposure in adults is meat with 53% (men) and 37% (women). In children, meat and hen´s eggs contribute to nearly the same extent (35 and 36%, respectively) to PFOA intake. The contribution of hen´s eggs in men and women was 19 and 26%, respectively. Hard cheese contributed to a maximum of 15% to the estimated intake of PFOA, whereas the contribution of fish was 9% in men, 16% in women, and 17% in children, respectively. The other food groups (poultry, venison, liver and bacon) contributed to less than 5% each to the estimated dietary intake of PFOA (Figure 1).
Dietary intake of PFOS for all population groups was mainly due to the consumption of fish, accounting for 87% of the estimated intake in adults and to 93% in children. Liver contributed to 7% (children) and 13% (women, men) to the dietary intake of PFOS.

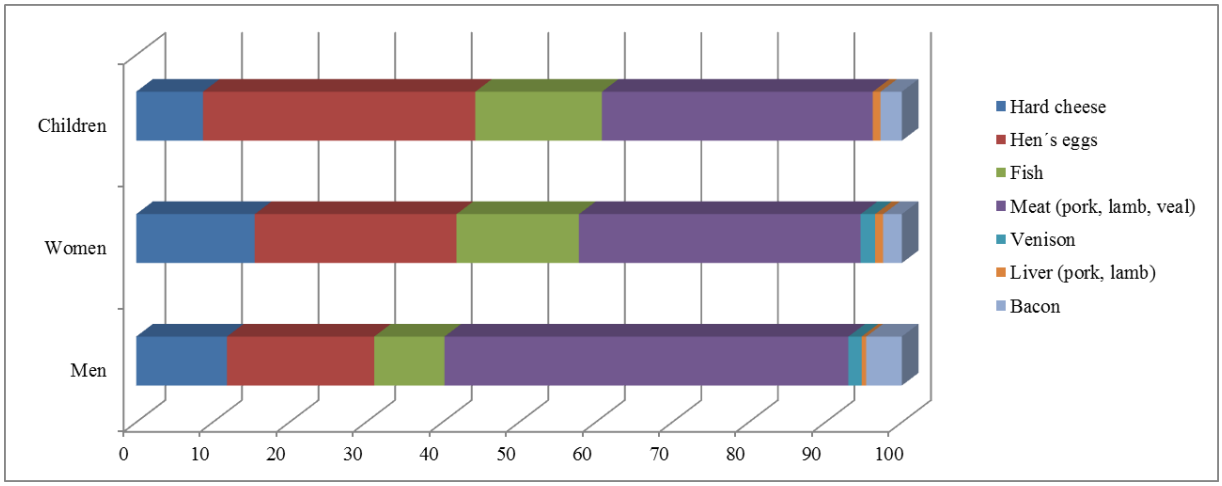

Figure 1: Contribution of various food groups (in per cent) to the total dietary intake of PFOA

Characterisation of health risks associated with the dietary exposure to PFOS and PFOA

Comparing the estimated dietary intake to the respective TDI revealed that intakes for PFOA and PFOS are well below the health-based guidance values in all population groups. Therefore it was concluded that it is unlikely that adverse effects are arising from the dietary exposure to PFOA and PFOS in the Austrian population.

The current intake levels appear to be low, however, the long half-life of PFOA and PFOS in the body must be taken into account. Even small amounts may contribute to an increase of the body burden. Hence, further monitoring of PFOA and PFOS is recommended.

Acknowledgements

The authors would like to thank the Austrian Federal Ministry of Health, Food Control Authority and Provincial Veterinary Services who participated in this monitoring programme.

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