INTAKE OF SELECTED PFAS IN THE ITALIAN GENERAL POPULATION

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Introduction

Perfluorinated chemicals (PFAS) are an emerging group of priority pollutants, whose main route of exposure in humans is represented by the dietary intake. The diversity of food habits and of the overall quality of the environment where food producing activities are in place, may determine qualitative/quantitative differences in the PFAS intake, on geo-referenced basis. Within the activity of the European Union granted PERFOOD project, the dietary intake of the Italian general population has been estimated and the preliminary results illustrated in this work.

Materials and methods

Italian food consumption rates. National food consumption data were recovered from the third nation-wide food consumption survey (2004–2006) of the Italian National Institute of Research on Food and Nutrition (INRAN)[1], and from the EFSA 2011 Comprehensive European Food Consumption Database in Exposure Assessment[2]. Individual food consumption rates were recorded for 3,323 subjects on a self-compiled diary over three consecutive days. Survey days were proportionally distributed among seasons: 25% in autumn, 25% in winter, 26% in spring and 24% in summer. Subjects were randomly selected to be representative of Italy's four main geographical areas (North-West, North-East, Centre, and South plus Sicily and Sardinia islands). The indicators of mean and high individual consumption (g $kg\text{-}bw^{-1}$ day^{-1}) refer to twenty large categories and 139 sub-categories of foods and beverages; they are expressed for the all population and for consumers only, for age and for sex categories. The PFAS dietary intake assessment was carried out for adults (N = 2,313; 18–64.9-year-old subjects) and children (N =193; 3-9.9-year-old subjects). The mean and 95^{th} percentile consumption values were used.

PFCs occurrence data. The following seven Perfluorinated compounds were selected for the assessment on the base of human biomonitoring data: Perfluorohexanesulfonic acid (PFHxS), Perfluorooctanesulfonic acid (PFOS), Perfluorohexanoic acid (PFHxA), Perfluorooctanoic acid (PFOA), Perfluorononanoic acid (PFNA), Perfluorodecanoic acid (PFDA), Perfluoroundecanoic acid (PFUdA). The PFAS concentrations in single food items and in composites were provided from the sampling and the analytical work framed within the PERFOOD project activities. The sampling campaigns were undertaken between June 2010 and 2011, following a market basket approach, at retailers' level. Only the data coming from the Italian samples were selected for this assessment. As a relevant number of the results was below the limits of quantification, it has been adopted the lower and the upper bound approaches (LB and UB) to express the associated analytical uncertainty.

Intake estimates. Intake data sets were evaluated with non-parametric statistics and characterized by several canonical descriptors. The deterministic approach was used: mean and 95^{th} percentile consumptions of the different food items were matched with mean LB and UB PFAS levels of the correspondent category. This lead to four different results (mean & LB, mean & UB, 95th percentile & LB, 95th percentile & UB). Adults and children were considered for the assessment. Due to the recorded differences in contamination between fresh and ground tap water supplies, we used the values referred to freshwater, as that more contaminated, under a conservative approach. Water values referred to potential hot spots were not considered in this preliminary assessment.

Results and discussion

The results of PFAS intakes *via* food for the two age ranges considered are shown in Tables 1 and 2.

Table 1. PFAS estimated dietary intake (ng kg-bw^{-1} day^{-1}) for adults (16–65 years) of the Italian general population .

PFOA		PFOS		PFHxA		PFNA		PFDA		PFUdA		PFHxS	
Mean consumption													
LB	0.14	LB	0.17	LB	0.03	LB	0.04	LB	0.02	LB	0.08	LB	0.03
UB	0.28	UB	0.24	UB	0.18	UB	0.19	UB	0.21	UB	0.36	UB	0.14
95° percentile consumption													
LB	0.48	LB	0.52	LB	0.10	LB	0.12	LB	0.05	LB	0.24	LB	0.05
UB	0.79	UB	0.74	UB	0.45	UB	0.47	UB	0.53	UB	1.05	UB	0.37

Table 2. PFAS estimated dietary intake (ng kg-bw^{-1} day^{-1}) for children (3–10 years) of the Italian general population.

PFOA		PFOS		PFHxA		PFNA		PFDA		PFUdA		PFHxS	
Mean consumption													
LB	0.21	LB	0.40	LB	0.08	LB	0.10	LB	0.04	LB	0.17	LB	0.03
UB	0.51	UB	0.50	UB	0.38	UB	0.40	UB	0.38	UB	0.70	UB	0.25
95° percentile consumption													
LB	0.74	LB	1.22	LB	0.30	LB	0.26	LB	0.11	LB	0.53	LB	0.10
UB	1.39	UB	1.48	UB	0.94	UB	0.91	UB	0.91	UB	2.01	UB	0.68

From the results shown in Table 1 and 2, it appears that the intake estimates fall well below the recommended TDIs of 150 ng kg-bw^{-1} day^{-1} and of 1,500 ng kg-bw^{-1} day^{-1} proposed by EFSA[3] for PFOS and PFOA, respectively. Such consideration is valid even under the worst case assumption in the most sensitive group (95th percentile, children). The relative small difference between the LB/UB estimates, considering the large number of left censored data, indicate the excellent quality of the analytical methods, where the Limit of Determinations for the different PFAS were tailored on intake estimates rather than on regulatory limits. The regional differences of PFAS intakes observed between the Northern, Central, and Southern Italy were comprised within a ± 15% of variation, and mainly addressable to the different food habits.

In the intake estimates, the uncertainties sourced from the lack of specification of some food items in the food consumption database. For instance, in the case of “miscellaneous”, marine salt, maize flour we did not recover the consumption data. For the contribution to the intake of the fish and fishery products category, farmed species, because those most consumed by the general population, were considered. For composite samples, the occurrence data of the main ingredient (i.e for pizza, snacks, biscuits = wheat flour) was assumed. Some low consumed food items were not included in the exposure assessment, such as: processed vegetables, spices and herbs; nuts, seeds, olives and their products, dried fruit; processed fruit in syrup, other vegetable oils, other fats, spirits and liquors. Possible losses/contributions to the PFAS contamination derived from cooking, packaging and serving were not accounted for.

The most contributing food categories to the mean dietary intake of PFOS and PFOA in adults are illustrated in Figure 1 and 2, respectively. It is worth noting that for PFOS the fish and fishery products category gives the largest contribution (46–65 %), while for PFOA, the most relevant category is the vegetables one. Drinking water accounts for <1–1 % and 10–12 % of the overall dietary intake for PFOS and PFOA, respectively.

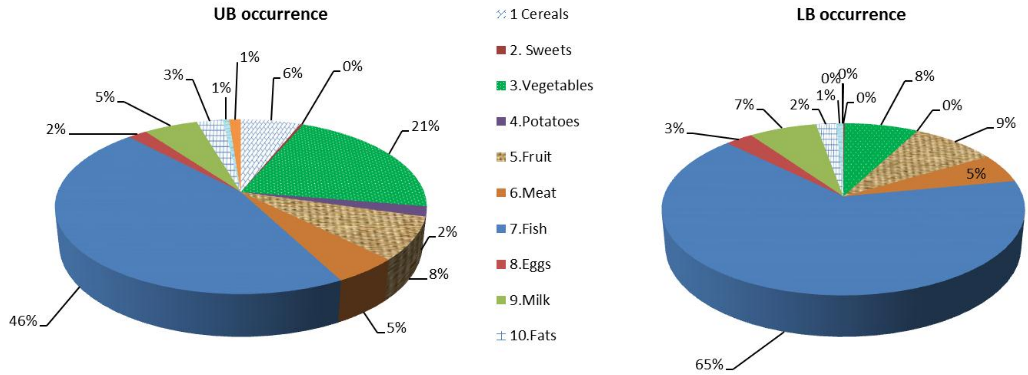

Figure 1. Average contribution of the different food groups to the total dietary intake of PFOS estimated for Italian adults using mean food consumptions.

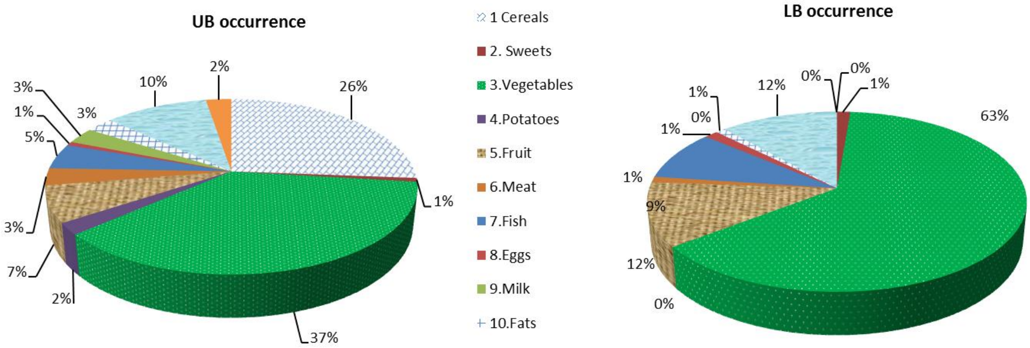

Figure 2. Average contribution of the different food groups to the total dietary intake of PFOA estimated for Italian adults using mean food consumptions.

The intakes estimated in the present evaluation are in general similar or lower than those recently reported in the literature [4-7] (Figure 3). The dietary intake of PFUdA for adults has been estimated to be 0.34 ng kg-bw^{-1} day^{-1} in Norwegian population[5]; such values are very similar to those estimated in this investigation (0.08–0.36 ng kg-bw^{-1} day^{-1} for Italian adults) (Table 1). Further work is in progress to provide dietary intake estimates in Italian hot spots and in sensitive groups such as fish and fishery products high consumers.

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References:
1. Leclerq C, et al. (2009) *Public Health Nutr.* 12(12):2504-32
2. EFSA **(**European Food Safety Authority) (2011) *The EFSA Journal* 9(3):2097.
3. EFSA **(**European Food Safety Authority) (2008)*The EFSA Journal* 653: 1-131.
4. Noorlander CW, et al. (2011) *J Agr Food Chem* 59: 7496–7505
5. Haug LS, et al. (2010) *Chemosphere* 80, 1137–1143
6. Clarke DB, et al. (2010) *Food Add Contam* 27 (4): 530–545
7. BfR (Federal Institute for Risk Assessment, Germany) (2008) Stellungnahme 004/2009
8. Cornelis C, et al. (2012) *Chemosphere* 86:308-314
9. Ericsson I, et al. (2008) *J Agric Food Chem* 56: 1787–1794

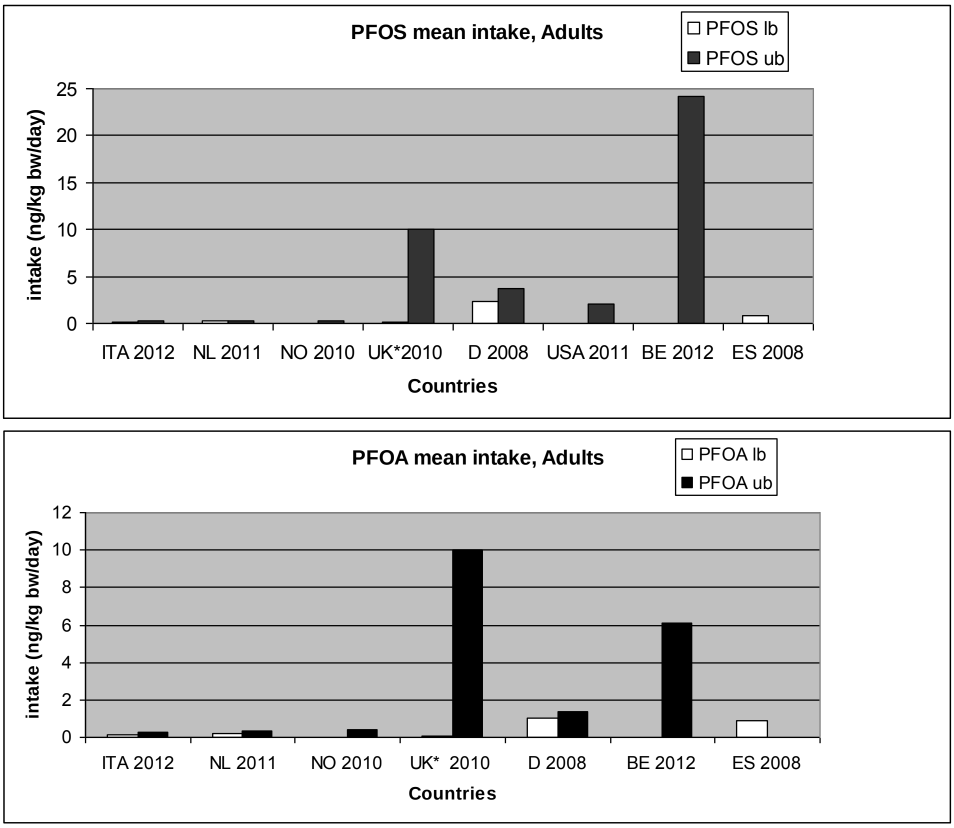

Figure 3. Comparison of PFOS and PFOA intake estimates, carried out in different Countries[4-7] in the last 5 years. UK* estimates referred to Consumers only.