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Two-year follow-up biomonitoring pilot study of residents' and controls' PFC plasma levels after PFOA reduction in public water system in Arnsberg, Germany

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ABSTRACT

Residents in Arnsberg, Germany, had been supplied by drinking water contaminated with perfluorooctanoate (PFOA). Biomonitoring data from 2006 evidenced that plasma PFOA concentrations of residents from Arnsberg were 4.5–8.3 times higher than those in reference groups. The introduction of charcoal filtration in July 2006 distinctly reduced PFOA concentrations in drinking water. Our one-year follow-up study showed a 10–20% reduction of PFOA plasma levels in residents from Arnsberg. Here we report the first results of the two-year follow-up study Arnsberg 2008. Additionally, the results of the two-year follow-up examination of the reference group are included. Paired plasma samples of 138 study participants (45 children, 46 mothers and 47 men) collected in 2006 and 2008 were considered in the statistical analyses. Within the two years plasma concentrations of PFOA, perfluorooctanesulfonate (PFOS) and perfluorohexanesulfonate (PFHxS) decreased in residents from Arnsberg and in control groups. The geometric means of PFOA plasma levels declined by 39% (children and mothers) and 26% (men) in Arnsberg and by 13–15% in the corresponding subgroups from the reference areas. For the population from Arnsberg a geometric mean plasma PFOA half-life of 3.26 years (range 1.03–14.67 years) was calculated. Our results confirm an ongoing reduction of the PFOA load in residents from Arnsberg. The decline of PFC levels in plasma of participants from the reference areas reflects the general decrease of human PFC exposure during the very recent years.

Introduction

Perfluorinated compounds (PFCs) are extensively used since the 1950s. They can be found worldwide in different environmental matrices, in wildlife and in humans. Long human plasma half-lives of some PFCs, like perfluorooctanoate (PFOA) and perfluorooctanesulfonate (PFOS), have been observed. Health concern is raised due to effects observed in animal studies, namely hepatotoxicity, carcinogenicity, reproductive and developmental toxicity (Lau et al., 2007). There are indications that a human PFC exposure may influence pregnancy outcome, however the results are inconsistent (Fei et al., 2007, 2008; Nolan et al., 2009). Existing information on the sources of the background exposure of the general population indicates a major role of dietary intake (Fromme et al., 2009). Additional exposure to PFOA via considerably contaminated drinking water has been observed in water districts near a chemical plant in Little Hocking, Ohio, USA (Emmett et al., 2006), in Arnsberg, Germany (Hölzer et al., 2008) and in Minnesota, USA (MDH, 2009). In Little Hocking and Arnsberg, biomonitoring studies showed a high internal PFOA load of residents, which was clearly related to the drinking water contamination (Emmett et al., 2006; Hölzer et al., 2008). In both locations, follow-up biomonitoring studies are ongoing to examine the trend of PFC concentrations in plasma, to calculate half-life elimination rates and to find out if the high PFOA exposure may cause health effects in the affected residents (Hölzer et al., 2009; Frisbee et al., 2009; Steenland et al., 2009; Bartell et al., 2010).

The drinking water contamination in Arnsberg was detected by Skutlarek et al. (2006). They reported high levels of PFOA in the rivers Rhine, Ruhr and Moehne (confluent of the river Ruhr, PFOA levels of up to 7070 ng/l), as well as in nearby public water supplies using river water to produce drinking water (500–640 ng/l). This environmental pollution was mainly caused by a so called soil improver mixed with industrial waste that was applied on agricultural areas on the upper reaches of the river Moehne (Skutlarek et al., 2006). 40,000 residents living in certain districts of Arnsberg

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had been constantly supplied by tap water that was markedly contaminated with PFOA. In July 2006, activated charcoal filters were installed that efficiently decreased PFOA concentrations in drinking water to levels predominantly under the limit of quantification (LOQ). More details on assessment and management of the PFC contamination in Arnsberg and the affected areas downstream have been summarized previously (Wilhelm et al., 2008).

In September and October 2006, our first biomonitoring study was accomplished to examine 90 children, 164 women and 101 men from Arnsberg who had been supplied by contaminated drinking water (Hölzer et al., 2008). The control group comprised 80 children, 153 mothers and 103 men from the neighboring towns Siegen and Brilon who received water with PFOA levels below the LOQ. In both locations, school beginners and their mothers were asked to participate. For the recruitment of male adults randomly selected residents were interviewed concerning their habits of drinking water consumption and those with highest intake were selected to participate. Geometric mean levels of PFOA plasma concentration of children, women and men from Arnsberg were 22.1 μg/l, 23.4 μg/l and 25.3 μg/l, respectively. They were increased 4.5–8.3 fold in comparison to PFOA levels in the control population. Consumption of PFC-contaminated tap water was a significant predictor of PFOA plasma concentrations (Hölzer et al., 2008). Our one-year follow-up examination conducted in Arnsberg in 2007 revealed a PFOA reduction of 10% (men), 17% (mothers) and 20% (children) during the first year (Hölzer et al., 2009).

Here we report the first results of the two-year follow-up study. Since PFC concentrations have been found to be declining in the general population in recent years (Olsen et al., 2008; Haug et al., 2009), we additionally included residents from the reference areas in the investigation. Moreover, we tried to identify factors influencing the amount of the decline of PFOA plasma concentrations.

Population, methods

Participants

The present survey is a follow-up examination of 138 individuals who already participated in the biomonitoring study in 2006. We considered data of 20–25 children, mothers and men each, from the target area and the reference areas, respectively. In Arnsberg, another analysis of plasma concentrations was offered to all 355 study participants from 2006. Those in each subgroup who first answered the invitation and agreed to a certain appointment were selected and taken in the pilot study group. For the recruitment of participants from the reference groups all 80 children and 80 mothers examined in 2006 were invited. Randomly selected, half of the men ($N = 50$) who took part in the study in 2006 were asked to participate again. All invitees from the control areas who agreed to join the examination were included in the pilot study.

The study was approved by the ethical commission of the Ruhr-University of Bochum and was conducted in accordance with the ethical principles for medical research involving human subjects as defined by the Declaration of Helsinki. Written informed consent was obtained from each subject or parent in the case of children. Information on demographic and occupational characteristics as well as health conditions and drinking water and food consumption had been surveyed during the first biomonitoring study. Within the follow-up study mail questionnaires for self-completion at home elicited information about actual body weight and body height, smoking and drinking habits, smoking exposure, actual drug intake and newly diagnosed diseases since the last study. On the day of the blood withdrawal trained interviewers accomplished a standardized questionnaire on drinking water consumption and food consumption habits since the last study and asked for acute diseases and drug consumption.

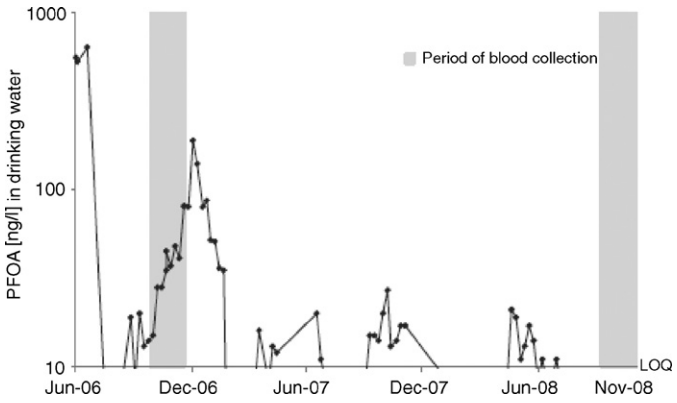

Fig. 1. PFOA concentrations in drinking water (frequency of analysis: weekly) in the waterworks Möhnebogen between May 2006 and November 2008 as published by the North Rhine-Westphalia State Agency for Nature, Environment and Consumer Protection, Germany (http://www.pft.lua.nrw.de/owl/GIS/exhibit/pft_tw.php?exhibit-use-local-resources; retrieved 1st December 2009). Measurement results below the limit of quantification (LOQ) of 10 ng/l are not drawn in the graph.

Sampling and chemical analysis

Blood samples were taken between 14 October and 11 November 2008. All materials used for venipuncture and processing had been tested for PFC contamination on the occasion of the first study without any findings. For PFC determinations 4.9 ml of blood were drawn into an EDTA tube and centrifuged the same day. The resulting plasma was stored at −20 °C and transported to the analytical laboratories in Erlangen, Germany, where analyses for PFOA, PFOS, perfluorohexanesulfonate (PFHxS), perfluorobutanesulfonate (PFBS), perfluorohexanoate (PFHxA) and perfluoropentanoic acid (PFPA) were performed using HPLC/tandem mass spectrometry. Handling of samples, assay procedure and quality control have been performed in the same way as in 2006 (Hölzer et al., 2008).

Calculation of PFOA intake via drinking water

The North Rhine-Westphalia State Agency for Nature, Environment and Consumer Protection, Germany, accomplished weekly analyses of PFOA concentration in drinking water in Arnsberg since May 2006 (Fig. 1). Each study participant's PFOA intake via drinking water between October 2006 and October 2008 was calculated. For each month in this period, arithmetic mean results of the PFOA levels in tap water (usually 4 measurements per month) were multiplied with the amount of individual drinking water consumption, which had been examined in detail in the interviews. For PFOA concentrations below the limit of quantification (LOQ) of 10 ng/l, a value of 5 ng/l was used in the calculations. In contrast to the weekly PFOA-analyses in Arnsberg's drinking water, the PFOA levels in drinking water in Siegen and Brilon have only been measured during each study period. PFOA has not been detected in any of the samples. To calculate the daily total PFOA exposure from all sources, the PFOA intake via drinking water was added to the amount of background exposure of the general population (1.6 ng/kg$_{\text{body weight}}$) as it was evaluated by Fromme et al. (2009).

Statistical analysis

All statistical analyses were carried out with SAS 9.2 (SAS Institute Inc., Cary, NC). A $p < 0.05$ was considered statistically significant. Plasma levels of fluorochemicals below the limit of