

PERFLUORINATED SUBSTANCES IN HOUSE DUST IN BAVARIA, GERMANY

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Introduction

Perfluorinated compounds (PFC) represent a large group of chemicals which are characterized by a fully fluorinated hydrophobic linear carbon chain attached to various hydrophilic heads. They can currently be detected in many environmental media and biota, as well as in human blood samples. Because of their persistence and their potential to accumulate they are of toxicological concern. The widespread distribution of various PFC and their corresponding degradation and metabolism products results in a very complex exposure situation. The contribution of single sources and pathways to the total exposure is currently not well defined. For the general population diet was estimated to be the dominant pathway, but for some subsets of the population a higher exposure could be observed due to environmental contamination¹.

It is well known that house dust is a receptor of and repository for chemicals in the home and thereby a potential source of exposure to them, especially for small children, who spend significant portions of their time on floors and engaging in frequent hand to mouth contact activities².

The present pilot study was conducted for the identification of the target substances in the indoor environment of apartments in a particular urban area. The aim was to measure the most significant PFC, perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA), to get a first overview of their presence in house dust in Germany.

Materials and Methods

Sampling sites

The concentrations of PFC in household dust were determined for a total of 12 residences. The apartments were built between 3 and 104 years before and had areas of 51 m² - 250 m².

Sample procedure and analysis

The dust samples were taken from the bags of the vacuum cleaners in regular use for cleaning the apartments. Particularly for determining substances in house dust it has become clear that sampling procedures and the preparation before conducting the analyses do definitely influence the results. The samples were split in two fractions. One fraction was not further prepared, but any larger dust particles, fibres and debris, which might otherwise distorted the results, were thus excluded. Whilst the other fraction were prepared sieved the dust to the fraction of < 63 µm. Afterwards the dust was put directly to clean aluminium foil and then transferred to glass bottles for storage.

Overall, 50 mg dust was solved in methanol. After addition of the internal standard (0.4 ng of perfluoro-1-[1,2,3,4-¹³C₄] octanesulfonat and 2.0 ng of Perfluoro-n-[1,2,3,4-¹³C₄] octanoic acid) the solutions were extracted by ultrasonic agitation and centrifuged at 20 000 g for 10 minutes. 500 µl of the supernatant was diluted with 500 µl water. For chromatographic separation 200 µl of the supernatant were injected into the HPLC system. The configuration of the column switching LC-MS/MS system is published elsewhere³. A triple-stage quadrupole mass spectrometer (API 3200 QTRAP™ Applied Biosystems, Darmstadt, Germany) equipped with an electrospray ionization source was used. The ionspray voltage was -4 500V, the source temperature 600°C, nitrogen was used as curtain and collision gas. Negative ions were analyzed by multiple reaction monitoring

(MRM) with a dwell time of 0.100 s for each transition. The compound specific parameters were obtained by infusion of the standards using the quantitative optimization function of the Analyst 1.4.1 software. Two transitions per analyte were monitored. Peak areas of the most sensitive transitions were used for quantitation.

Statistical analysis

For the estimation of correlations the non-parametric Spearman rank correlation coefficient was derived, the difference in median was tested with the Wilcoxon rank sum test.

Results and Discussion

The target substances could be quantified in all samples, with very high levels in two samples. The results of each sample are given in Figure 1. Median (range) PFOS and PFOA concentrations in the sieved fraction (< 63 µm) were 0.016 µg/g (0.003-0.342 µg/g) and 0.011 µg/g (0.002-0.141 µg/g), respectively. Significantly lower median concentrations were observed in the non sieved samples, with median values of 0.010 µg/g (PFOS) and 0.007 µg/g (PFOA), indicated that PFC were mainly associated with smaller particles. But between the sieved and non sieved dust fraction a significant correlation was observed for PFOS and PFOA ($r = 0.94$ and 0.91). Furthermore a significant correlation was found between the PFOS and PFOA concentrations in both fractions ($r = 0.86$ and 0.78).

Up to now PFC were measured in dust from homes in Canada, Japan and the US. In 16 Japanese houses concentrations between 0.011 and 2.500 µg PFOS/g dust (non sieved, only large particles removed) and between 0.070 and 3.700 µg PFOA/g dust were determined in dust collected from vacuum cleaner bags⁴. Median concentrations were 0.025 µg PFOS/g dust and 0.165 µg PFOA/g dust. A strong correlation was found between PFOS and PFOA ($r^2 = 0.99$), however the association dropped to $r^2 = 0.35$ when one outlier was removed. In another Japanese study, PFOS and PFOA were detected in all 7 collected dust samples (particle size of 75 µm to 1 mm) from 0.007-0.041 µg/g and 0.018-0.089 µg/g, respectively⁵.

Dust from vacuum cleaner bags from 67 Canadian homes was sieved to a size of < 150 µm⁶. The most frequently detected PFC were PFOS at < 0.002-5.065 µg/g (median: 0.038 µg/g) and PFOA at < 0.002-1.231 µg/g (median: 0.020 µg/g). All compounds were positively correlated with each.

Additionally, 112 dust samples collected in Ohio and North Carolina⁷. After sieving to < 150 µm, PFOS concentrations of < 0.009-12.1 µg/g (mean: 0.76 µg/g) and PFOA concentrations of 0.01-1.96 µg/g (mean: 0.30 µg/g) were found. No differences were observed between the two sampling regions, but a significant correlation was found between PFOS and PFOA ($r = 0.87$).

The mean PFOS concentration in samples collected from Canadian, German and Japanese homes appear to be very similar, whilst the mean PFOA concentration was nearly the same in Canada and Germany, but lower compared with the results from Japan. On the other hand, high concentrations were reported for both target analytes in the US study⁷. The reasons for these differences are yet unknown.

References

1. Fromme H, Schlummer M, Möller A, Gruber L, Wolz G, Ungewiß J, Böhmer S, Dekant W, Mayer R, Liebl B, Twardella D. *Environ Sci Technol* 2007; 41: 7928-7933.
2. Butte W, Heinzow B. *Rev Environ Contam Toxicol* 2002;175: 1-46.
3. Brink A., Lutz U, Völkel W, Lutz W. K. *J Chromatogr B Analyt Technol Biomed Life Sci* 2006; 830: 255-261.
4. Moriwaki H, Takata Y, Arakawa R J *Environ Monit* 2003; 5: 753-757.
5. Nakata A, Katsumata T, Iwasaki Y, Ito R, Saito K, Izumi S, Makino T, Kishi R, Nakazawa H. *Organohalogen Compounds* 2007; 69: 2844-2846.
6. Kubwabo C, Stewart B, Zhu J, Marro LJ. *Environ Monit* 2005; 7, 1074-1078.
7. Strynar MJ, Lindstrom AB. *Environ Sci Technol* 2008; 42: 3751-3756.

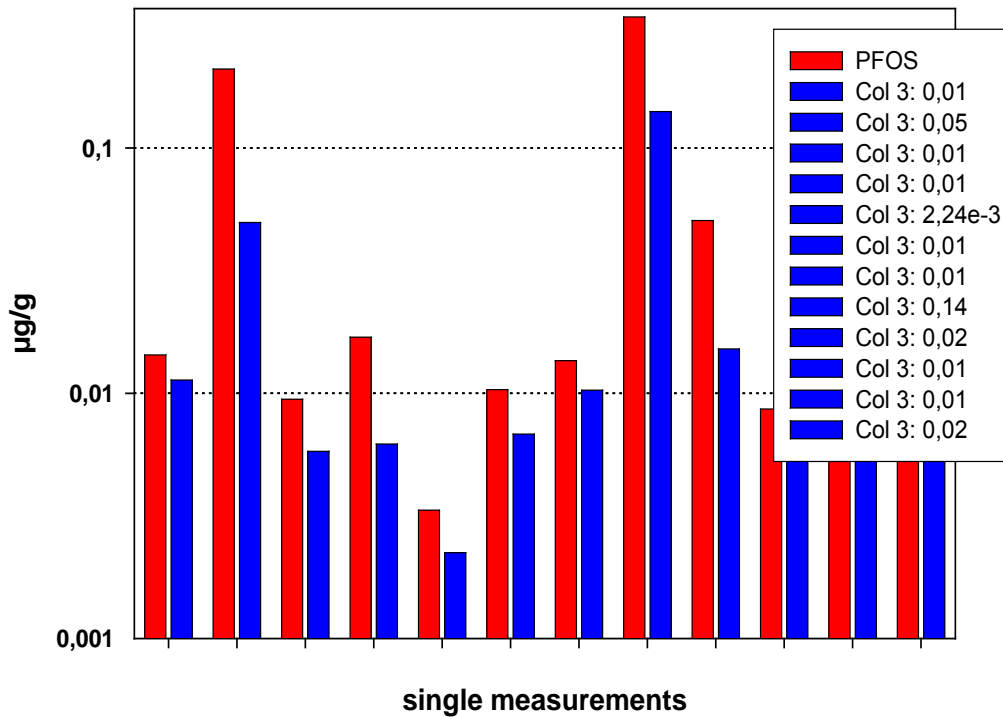


Figure 1: PFOS and PFOA in house dust samples (sieved to $<63 \mu\text{m}$)

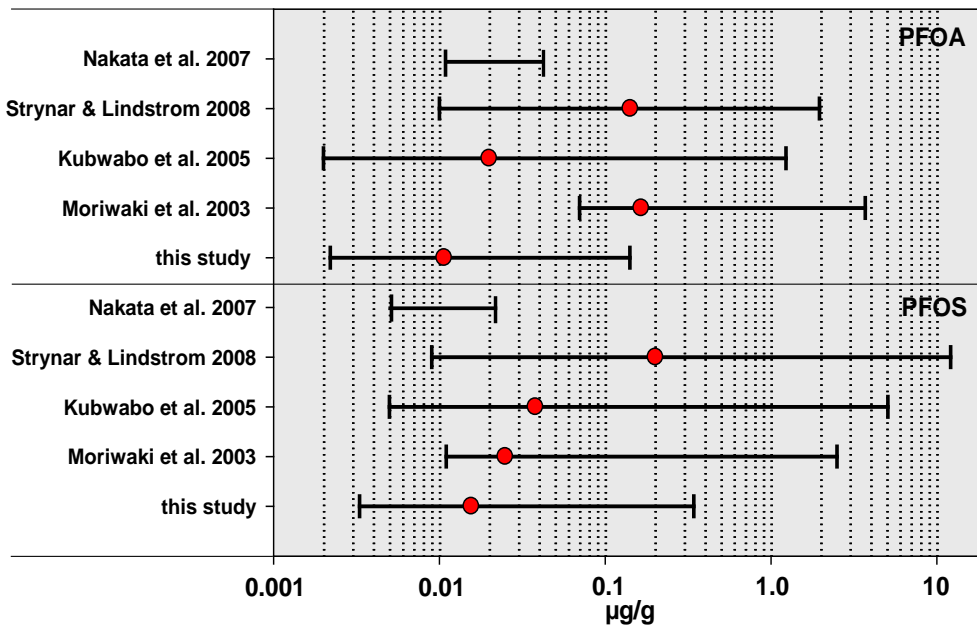


Figure 2: PFOS and PFOA concentrations in dust samples published so far in the scientific literature (minimum, median, maximum)