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Polyfluorinated compounds in ambient air from ship- and land-based measurements in northern Germany

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ABSTRACT

Neutral volatile and semi-volatile polyfluorinated organic compounds (PFC) and ionic perfluorinated compounds were determined in air samples collected at two sites in the vicinity of Hamburg, Germany, and onboard the German research vessel Atair during a cruise in the German Bight, North Sea, in early November 2007. PUF/XAD-2/PUF cartridges and glass fiber filters as sampling media were applied to collect several fluorotelomer alcohols (FTOH), fluorotelomer acrylates (FTA), perfluoroalkyl sulfonamides (FASA), and perfluoroalkyl sulfonamido ethanols (FASE) in the gas- and particle-phase as well as a set of perfluorinated carboxylates (PFCA) and sulfonates (PFSA) in the particle-phase. This study presents the distribution of PFC in ambient air of the German North Sea and in the vicinity of Hamburg for the first time. Average total PFC concentrations in and around Hamburg (180 pg m^{-3}) were higher than those observed in the German Bight (80 pg m⁻³). In the German Bight, minimum-maximum gas-phase concentrations of 17–82 pg m⁻³ for Σ FTOH, 2.6–10 pg m⁻³ for Σ FTA, 10–15 pg m⁻³ for Σ FASA, and 2–4.4 pg m^{-3} for Σ FASE were determined. In the vicinity of Hamburg, minimum-maximum gasphase concentrations of 32–204 pg m^{-3} for $\Sigma FTOH,$ 3–26 pg m^{-3} for $\Sigma FTA,$ 3–18 pg m^{-3} for $\Sigma FASA,$ and 2–15 pg m⁻³ for Σ FASE were detected. Concentrations of perfluorinated acids were in the range of $1-11 \text{ pg m}^{-3}$. FTOH clearly dominated the substance spectrum; 8:2 FTOH occurred in maximum proportions. Air mass back trajectories, cluster, and correlation analyses revealed that the air mass origin and thus medium to long range atmospheric transport was the governing parameter for the amount of PFC in ambient air. Southwesterly located source regions seemed to be responsible for elevated PFC concentrations, local sources appeared to be of minor importance.

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1. Introduction

In recent years, long chain perfluorinated carboxylates (PFCA) and -sulfonates (PFSA) such as perfluorooctanoate (PFOA) and perfluorooctane sulfonate (PFOS) have been found to be persistent, bioaccumulative, and entailing toxic properties (Beach et al., 2006; Giesy and Kannan, 2002; Haukas et al., 2007; Houde et al., 2006; Prevedouros et al., 2006). Furthermore, global distribution to remote regions caused by extensive industrial application and consumer use has been demonstrated for these classes of compounds (Bossi et al., 2005; Giesy and Kannan, 2001; Prevedouros et al., 2006; Smithwick et al., 2005; Taniyasu et al., 2003; Tao et al., 2006; Young et al., 2007).

The awareness of producers as well as political stakeholders for appropriate action concerning these compounds is reflected by voluntary commitments of the fluorochemical industry and regulation efforts of political institutions. Examples are the voluntary phase-out of PFOS-based compounds by its main producer 3M in 2002 or the commitment to reduce PFOA emissions by 95% until 2010 as well as several restrictions concerning manufacturing, marketing, and use of PFOS by the United States Environmental Protection Agency or the European Community. There are still many unknowns concerning environmental fate, distribution, and sources, especially in the perspective various polyfluorinated compounds which are also being used as PFOS and PFOA substitutes. Therefore, there is an ongoing need of decision makers for further information to evaluate polyfluorinated compounds properly.

The ubiquitous distribution of PFCA and PFSA pointed at two main transport pathways. Transport in the water phase, partly in relation to the global oceanic circulation system, has been demonstrated in various studies (Caliebe et al., 2004; McLachlan et al., 2007; So et al., 2007; Yamashita et al., 2008). However, this transport pathway does not explain the entire PFC burden of





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pristine ecosystems, especially of those that are not effected by marine influences. Due to their low volatility and efficient scavenging by wet and dry deposition, PFCA and PFSA are unlikely to travel long distances in the atmosphere (Hurley et al., 2004). However, an atmospheric transport with subsequent degradation of volatile and semi-volatile precursor compounds like fluorotelomer alcohols (FTOH), perfluorinated sulfonamides (FASA) and perfluorinated sulfonamido ethanols (FASE) to persistent PFCA and PFSA, was hypothesized to be a main transport mechanism (D'Eon et al., 2006; Ellis et al., 2004; Martin et al., 2006; Sulbaek Andersen et al., 2005). Because of their slow reaction with hydroxyl radicals, estimates of atmospheric lifetimes of more than 10-20 days for FTOH and more than 20–50 days for FASA in smog chamber studies (Ellis et al., 2003; Martin et al., 2006) and atmospheric residence times of more than 50 days for FTOH in field studies (Piekarz et al., 2007) indicate the possibility of regional and long range atmospheric transport. PFCA and PFSA were also determined in snow samples of the high Arctic which experienced contamination solely from atmospheric sources confirming the indirect precursor-based atmospheric transport and deposition of these compounds (Young et al., 2007).

Only a few studies examined the spatial distribution of polyfluorinated compounds in air (Barber et al., 2007; Jahnke et al., 2007b; Shoeib et al., 2006). Barber et al. (2007) found that PFC concentrations in air samples from northwest Europe were declining up to two orders of magnitude with decreasing degree of urbanization. Shoeib et al. (2006) determined FTOH and FASE in air masses of northern Canada and the North Atlantic and observed a widespread distribution and increasing concentrations of volatile PFC in air towards coastal areas of Europe and North America. Jahnke et al. (2007b) found decreasing concentrations of FTOH, FASA, and FASE towards the southern hemisphere along a northsouth transect from Bremerhaven, Germany to Cape Town, South Africa. The same authors analysed volatile polyfluorinated compounds in urban and remote air in Germany and observed no significant differences of PFC concentrations between those locations (Jahnke et al., 2007a). Concentrations of (semi-) volatile polyfluorinated substances covering four orders of magnitude have been determined at some selected locations in North America and Japan (Martin et al., 2002; Oono et al., 2008; Piekarz et al., 2007; Stock et al., 2004).

In this study, we present the spatial distribution of polyfluorinated compounds in air samples collected during a cruise in the German Bight (North Sea) on the research vessel Atair and at two permanent land-based sites in the vicinity of Hamburg, Germany. Samples were taken using high volume air samplers and analyzed for volatile and persistent PFC such as FTOH, FASA, FASE, PFCA, and PFSA. PFC concentrations were elucidated and the origin of PFC contamination was traced using air mass back trajectories, correlation-, and cluster analysis. Data presented in this study yield new information on the spatial atmospheric distribution of polyfluorinated compounds on a regional scale as well as the identification of source regions in the North Sea area.

2. Experimental

2.1. Chemicals

All chemicals, standard compounds, and gases were of high quality and purity. Details on chemicals concerning supplier and purity can be found in the supplemental information.

2.2. Sampling

Daily air samples were taken onboard the German research vessel Atair during a cruise from Hamburg to the German Bight, North Sea from October 30th to November 6th 2007 (Atair 155, Fig. 1). Samples were taken in parallel using two high volume air samplers (A and B) which were installed at the observation deck of the ship approximately 16 m above sea level. To ensure that ship exhausts were not sampled, samplers were controlled by a computer connected to the ship's meteorological system avoiding that sampling and thus ship-borne contamination was occurring with relative winds arriving from the rear of the ship.

Land-based sampling was conducted at two permanent sampling sites located in the vicinity of Hamburg with sampling



Fig. 1. Location of the two permanent sites Barsbuettel and GKSS and track of RV Atair during the cruise in the German Bight. Numbered dots mark the start point of each sample during the cruise. Samples were taken in between this and the following location. Sample 7 was taken while the ship was staying in the port of Hamburg situated in the city center.

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