



# Occurrence survey and spatial distribution of perfluoroalkyl and polyfluoroalkyl surfactants in groundwater, surface water, and sediments from tropical environments

Gabriel Munoz<sup>a</sup>, Pierre Labadie<sup>b</sup>, Fabrizio Botta<sup>c</sup>, François Lestremou<sup>c</sup>, Benjamin Lopez<sup>d</sup>, Emmanuel Geneste<sup>a</sup>, Patrick Pardon<sup>a</sup>, Marie-Hélène Dévier<sup>a</sup>, Hélène Budzinski<sup>b,\*</sup>

<sup>a</sup> Université de Bordeaux, EPOC, UMR 5805, LPTC, 351 Cours de la Libération, F-33400 Talence, France

<sup>b</sup> CNRS, EPOC, UMR 5805, LPTC, 351 Cours de la Libération, F-33400 Talence, France.

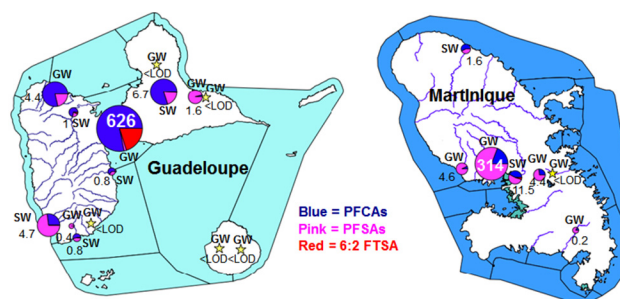
<sup>c</sup> INERIS, Parc Technologique Alata, BP2, 60550 Verneuil-en-Halatte, France

<sup>d</sup> BRGM, 3 avenue Claude Guillemin, BP 36009, 45060 Orléans Cedex 2, France

## HIGHLIGHTS

- The PFAS spatial distribution was investigated in tropical areas (Overseas France).
- Median  $\sum$  PFASs was  $1.8 \text{ ng L}^{-1}$  in surface water and  $0.56 \text{ ng L}^{-1}$  in groundwater.
- PFOS and PFOA prevailed in surface water Vs PFBS, PFHxS and PFHxA in groundwater.
- High 6:2 FTSA, PFBA, PFPeA and PFHxA levels were reported at several industrial sites.
- The repeated use of firefighting foams may partly explain the latter observation.

## GRAPHICAL ABSTRACT



Average  $\sum$ PFASs ( $\text{ng L}^{-1}$ ) and composition profile in Overseas France, illustrated for selected Guadeloupe and Martinique surface water (SW) and groundwater (GW) sampling sites

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## ABSTRACT

The occurrence and spatial distribution of 22 selected perfluoroalkyl and polyfluoroalkyl substances (PFASs) in surface water ( $n = 75$ ), groundwater ( $n = 80$ ) and surficial sediment ( $n = 15$ ) were investigated for the first time in the tropical areas constituted by the French Overseas Territories (French Guiana, Guadeloupe, Martinique, Mayotte and Reunion). Descriptive statistics and correlations between PFASs were evaluated through the use of specific statistical treatments to handle left-censored data (“non-detects”). PFASs were ubiquitous in these samples and detection frequencies as high as 79% for perfluorooctane sulfonate (PFOS) and 65% for perfluorooctanoate (PFOA) were reported in surface waters.  $\sum$  PFASs was in the range  $< \text{LOD} - 77 \text{ ng L}^{-1}$  (median =  $1.8 \text{ ng L}^{-1}$ ) in surface waters and  $< \text{LOD} - 638 \text{ ng L}^{-1}$  (median =  $0.56 \text{ ng L}^{-1}$ ) in groundwater. PFOS and PFOA dominated PFAS composition profiles in surface water, while shorter-chain compounds prevailed in groundwater, highlighting their higher transfer potential through infiltration processes and the possible influence of precursors. Elevated levels of 6:2 fluorotelomer sulfonate (6:2 FTSA) and short-chain perfluoroalkyl carboxylates were found in groundwater near several industrial facilities such as oil refineries or electricity power plants. This may be related to the existence of firefighting operations involving the use of aqueous film forming foams (AFFFs) that contain precursors to such compounds. These findings would also tend to confirm recent trends

\* Corresponding author.

E-mail address: [helene.budzinski@u-bordeaux.fr](mailto:helene.budzinski@u-bordeaux.fr) (H. Budzinski).

regarding the increasing proportion of C<sub>4</sub>- or C<sub>6</sub>-based perfluoroalkyl acids, their precursors, or even shorter-chain congeners at PFAS hot spots.

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## 1. Introduction

Perfluoroalkyl and polyfluoroalkyl substances (PFASs) are highly-fluorinated surfactants that have been used in a variety of formulations and consumer products, including textiles, food packagings, firefighting foams, and hydraulic fluids for aircrafts, as well as in industrial applications such as electronics and fluoropolymer manufacturing (Buck et al., 2011; Barzen-Hanson et al., 2017; Newton et al., 2017; Schaidler et al., 2017). In the early 2000s, the global extent of perfluorooctane sulfonate (PFOS) occurrence in biota was revealed, spurring research interest in the monitoring of this analyte and related compounds in all environmental media. PFOS was also documented in human tissues, blood or breast milk (e.g. Cariou et al., 2015; Olsen et al., 2017). Results from various lines of evidence suggested hepatic and developmental toxicity for some perfluoroalkyl acids (PFAAs) (Lau et al., 2007), as well as potential for bioaccumulation (Bertin et al., 2016; Dassuncao et al., 2017) and biomagnification (Martin et al., 2004; Houde et al., 2006).

In 2009, PFOS, its salts, and perfluorooctane sulfonyl fluoride were added to Annex B of the Stockholm Convention on persistent organic pollutants (UNEP-POPS-COP.4-SC-4-17). More recently, the watch list of priority substances under the European Union Water Framework Directive (2000/60/EC) was also extended to include PFOS and its derivatives (2013/39/EU). The use of PFOS is now restricted to a limited number of applications, and major manufacturers in the world are believed to have shifted to shorter-chain alternatives or precursors (Ahrens and Bundschuh, 2014). Other PFAAs of particular concern include perfluoroalkyl carboxylates (PFCAs), for instance perfluorooctanoate (PFOA) and longer chained analogs that are on track to be phased out globally (2010/2015 PFOA Stewardship Program). Environmental quality standards for inland waters have been proposed for PFOS in surface waters from European Union countries (e.g. 2013/39/EU), and the U.S. EPA has recently revised the Health Advisory Levels of PFOS and PFOA in drinking water to a more stringent guideline (Hu et al., 2016).

Comprehensive studies have examined PFAA surface water contamination at watershed scale (Möller et al., 2010; Campo et al., 2015; Lindim and Cousins, 2015), nationwide scale (Clara et al., 2009; Lam et al., 2014; Munoz et al., 2015; Valsecchi et al., 2015; Ahrens et al., 2016; Nguyen et al., 2016), or continent-wide scale (Loos et al., 2009; Gewurtz et al., 2013). In contrast, fewer studies have investigated PFAA groundwater contamination at large spatial scale (Loos et al., 2010; Lopez et al., 2015). A number of U.S. surveys have screened high profile sites located in the immediate vicinity of primary PFAS sources such as those impacted by firefighting activities. For instance, PFASs were previously documented in groundwater samples from military bases, due to the repeated use of aqueous film-forming foams (AFFFs) in firefighting training or fire emergency response (e.g., Moody et al., 2003; Backe et al., 2013). Even though PFAA production and use may be on the decline, some newly-identified AFFF fluorosurfactants, including zwitterionic, cationic and anionic polyfluoroalkyl surfactants, could biodegrade to PFAAs (e.g. Harding-Marjanovic et al., 2015; Mejia-Avenidaño et al., 2016). This could contribute to sustaining environmental levels of PFAAs through the degradation of precursors. Hence, monitoring data are still needed to more comprehensively characterize the occurrence of PFAAs and their precursors at background sites and other potentially impacted areas, including industrial sites (Zhang et al., 2016). Additionally, large-scale PFAS surveys have essentially focused on temperate and colder northern latitudes, while the available literature remains scarce regarding tropical environments (Lam et al., 2017).

The territories surveyed in the present study encompass both the tropical continental climates (French Guiana, Southern American continent) and tropical insular habitats from the Atlantic Ocean (Guadeloupe, Martinique) and Indian Ocean (Mayotte, Reunion). Although the present study was focused on the French Overseas, such territories have a type of tropical climates and habitats that can be deemed representative of this part of the world. Tropical areas are infrequently monitored compared to colder northern climates and the comprehensive database gathered under the framework of the present survey therefore constitutes an important contribution to the state-of-art. Only a few surveys have hitherto addressed pollution from organic contaminants in Overseas France. Although a water quality monitoring network was launched in 1993, only pesticides have been regularly investigated since 1999 and other organic micropollutants since 2006 (Bocquené and Franco, 2005; Cabidoche et al., 2009; Coat et al., 2011; Ramdine et al., 2012; Crabit et al., 2016; Dromard et al., 2017). Therefore, critical knowledge gaps still remain as regards the environmental fate of organic pollutants in these areas. With the exception of French Guiana, French Overseas Departments are characterized by peculiar hydrodynamics, with small-scale basins generally comprised of perennial and intermittent rivers, and soils often characterized by karstic or volcanic geological nature, wherein infiltration processes from rainfall or surface water may readily occur (Charlier et al., 2011; Lloret et al., 2011). In addition, although some legacy organic contaminants have been investigated (e.g.: pesticides, polycyclic aromatic hydrocarbons), little is still known about the occurrence of contaminants of emerging concern such as PFASs in these areas.

Inclusive of PFOS/PFOA, a target list of 22 PFASs (16 PFAAs + 6 PFAA-precursors) was established. 6:2 fluorotelomer sulfonate (6:2 FTSA) is a C<sub>6</sub>-based fluorotelomer that has been previously reported to occur at relatively high levels at AFFF-impacted sites (Backe et al., 2013; Ahrens et al., 2015; Field and Seow, 2017; Mejia-Avenidaño et al., 2017). Although 6:2 FTSA itself may not be a major AFFF component (Backe et al., 2013), it is the possible degradation intermediate of many other fluorotelomer precursors for which no standards are currently available (Liu and Mejia Avenidaño, 2013; Munoz et al., 2017), and can itself generate terminal perfluoroalkyl carboxylate products through degradation in natural or engineered environments (Wang et al., 2011). Given the recent regulations to ban PFOS/PFOA, long-chain analogs, and related long-chain PFAA precursors, the industry has essentially shifted to short-chain PFASs. The C<sub>6</sub>-based compounds, such as 6:2 FTSA, now dominate the trade (OECD, 2013). Taken together, these elements constitute the rationale for the inclusion of 6:2 FTSA under the list of target analytes. FOSA and four perfluorooctane sulfonamide derivatives were the other PFAA precursors included in the present survey. Such compounds have been shown to generate PFOS under microbially-mediated transformations (e.g. Mejia-Avenidaño and Liu, 2015; Zhang et al., 2017). Although N-alkyl perfluorooctane sulfonamido acetic acids (MeFOSAA, EtFOSAA) may arise from the degradation of volatile precursors, they have been shown to be relatively persistent metabolites (Zhang et al., 2017); additionally, an earlier survey in metropolitan France sediments showed relatively high levels of EtFOSAA compared to PFAAs at some specific sites (Munoz et al., 2015). Monitoring N-ethyl perfluorooctane sulfonamide (EtFOSA) appeared further relevant to the specific context of the present survey, given the vicinity of French Guiana with Brazil where EtFOSA has been used in insecticide formulations under the trade name of sulfuramid (OECD, 2013; Löfstedt-Gilljam et al., 2015).

Hence, the specific aims of the present study were three-fold: i) to document the occurrence and levels of PFAAs and selected precursors

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