



Endocrine disrupting compounds in gaseous and particulate outdoor air phases according to environmental factors



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HIGHLIGHTS

- EDCs in the Paris area were similar to Western countries except PAHs and PBDEs.
- PCBs and PAEs showed an urban signature.
- AP, PAH and PBDE sources were more distributed along a urban-suburban transect.
- EDCs prevailed in gaseous phase whatever the season except DEHP and heavy PAHs.

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ABSTRACT

This study investigated, for the first time in France, the spatial and temporal patterns of 55 endocrine disrupting chemicals (EDCs) in ambient air at three sites (urban, suburban and forest) under two climatic periods (warm/cold) for 2 successive years. All EDCs, except tetrabromobisphenol A (TBBPA), were encountered with various frequencies of up to 100%. Phthalate diesters (PAEs) were the most abundant chemicals with total concentrations as the sum of compounds, ranging from 10 to 100 ng m⁻³ of total air, followed by alkylphenols (APs) and polycyclic aromatic hydrocarbons (PAHs), which were both approximately 1 ng m⁻³. Polychlorinated biphenyl (PCBs) and bisphenol A (BPA) concentrations were notably lower (approximately 0.1 ng m⁻³). Air concentrations, depending on the considered compounds, were from 1.2 to 2 times higher in the urban than the suburban area and from 2 to 5 times higher in the urban than the forest site. PAH emissions were higher in the cold period, due to combustion processes. This finding is contrary to the other EDCs that are more abundant in the summer and governed by volatilisation. Most of the EDCs were largely distributed in the gaseous phase (>80% in the summer). The octanol/air partition coefficient (K_{OA}) and vapour pressure (V_p) were relevant parameters for predicting EDC partitioning and direct relationships ($p < 0.001$) were observed i) between log K particle/gas partitioning (log K_p) and log K_{OA} and ii) between EDC ratios in the gaseous phase and log vapour pressure (log V_p).

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1. Introduction

The continuous release of a number of endocrine disrupting chemicals (EDCs), such as bisphenol A (BPA), tetrabromobisphenol

A (TBBPA), phthalate diesters (PAEs), alkylphenols (APs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and polycyclic aromatic hydrocarbons (PAHs), to the environment generates new concerns regarding human exposure to low doses of toxic mixtures and possible subsequent effects on health (Benachour et al., 2007; Woodruff, 2011).

The emission sources of these chemicals have greatly increased and diversified over the past decades (EEA, 2013); and therefore,

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they have appeared as major air contaminants, particularly because of their semi-volatile properties (Rudel and Perovich, 2009). Thus, PAEs, APs, PCBs and PBDEs that are not chemically bound to matrices might become easily dispersed by volatilisation (Palm et al., 2002; Fujii et al., 2003; Liang and Xu, 2014).

PAEs can be emitted into the atmosphere from commonly used products, plastics, detergents, building materials or furniture (Wormuth et al., 2006). To date, despite a wide use of BPA in polycarbonate and epoxy resin production, emission sources and subsequent impacts upon ambient air quality remain poorly documented (Huang et al., 2012).

APs are by-products of alkylphenol ethoxylates largely used in household detergents. The European directive 2003/53/EC that came into force in January 2005 restrained the use of products containing more than 0.1% nonylphenols (NP) or nonylphenol ethoxylates. However, alkylphenol ethoxylates are still applied as dispersing agents in pulp and paper production, emulsifying agents in latex paints, glue and pesticides or in the preparation of anti-oxidants, curing agents and heat stabilisers for polymer resins and may be emitted to the environment from building materials (Saito et al., 2004).

PBDEs, a major class of flame retardants used in a number of manufactured goods to reduce their flammability, were composed of three major commercial formulations: penta-BDE, octa-BDE and deca-BDE (de Wit et al., 2010). Since 2003, penta- and octa-BDE were banned of use in all applications for the European Union market (Directive 2003/11/EC). Deca-BDE was registered under the "Registration, Evaluation, Authorisation and Restriction of Chemicals" (REACH) in 2010 and thus its uses have been restricted to applications such as automotive equipment, textiles and construction and suppressed for electric and electronic equipment (Deca-BDE, 2012). Tetrabromobisphenol A (TBBPA) is used as a reactive or additive flame retardant in polymers such as ABS, epoxy and polycarbonate resins, polystyrene, phenolic resins or adhesives (Who, 1995).

Chlorinated compounds such as hexachlorobenzene (HCB) and PCBs have been classified as persistent organic pollutants in the 1998 Aarhus Protocol (Aarhus, 24 June 1998), in the framework of the Geneva Convention on long-range transboundary air pollution. Dispersive uses of PCBs were banned in France by the Decree of 8 July 1975 and their uses were restricted to in-service equipment by a second decree in February 1986. Finally, in February 2003, a decree on the elimination of all equipment containing PCBs before the end of 2010 has been published. An unknown stock of PCBs is within now buried landfills and other waste-handling facilities as well as diffuse sources such as electrical wiring and paints (Diamond et al., 2010). Most recently, new "non-aroclor" PCBs were detected as byproduct of diarylide yellow (Rodenburg et al., 2010) and other pigment production (Anezaki et al., 2015).

Although HCB production has ceased since 1993, this compound is still being generated inadvertently as a by-product and/or an impurity in several chemical processes (Bonnomet et al., 2011) and pentachlorobenzene (PeCB) occurs as a by-product of HCB degradation (Brignon, 2005).

PAHs are continuously produced by combustion processes, mainly from domestic heating and automobile traffic (Katsoyiannis et al., 2011; Tobiszewski and Namieśnik, 2012). Despite PAHs are not considered as POPs, they were included to the Aarhus protocol. The objective was to reduce their emissions from stationary sources below their levels in 1990. For the emission inventories, the four indicator compounds shall be used: benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, and indeno(1,2,3-cd)pyrene.

Consequently, EDCs might undergo local or long range transport and their residence time in the air depends on climatic conditions and pollutant partitioning between the gaseous and particulate

phases (Wania, 2003).

This study investigated for the first time in France, the atmospheric fate of a wide variety of EDCs ($n = 55$) including PCBs, PBDEs, PAHs, APs, PAEs, HCB, PeCB, BPA and TBBPA, and considering both their gaseous and particulate phases. The target compounds were selected according to suspicion of endocrine disrupting properties, to their classification as POPs and semi-volatile organic compounds and to their environmental occurrence. The concentration levels for the different EDC families were first considered, in order to highlight the general trends of EDC distribution in France and to compare with the contamination in other countries. As EDCs result from various sources, the spatial variations between three sites located respectively in urban, suburban and forest areas were studied. In addition, temporal variations between two contrasted seasons with and without domestic heating and between two successive years were investigated. Next, the particle/gas partitioning was examined on the basis of compound molecular properties. Last, relationships between outdoor temperature, suspended particles $<10\ \mu\text{m}$ (PM₁₀) and EDC air concentrations were explored at the most polluted site.

2. Material and methods

2.1. Sampling schedule

The three sampling sites belonged to the local air monitoring network of the Ile-de-France district (Airparif) (Fig. Supplementary Material – SM 1).

Two sampling campaigns were carried out at the following 3 sampling sites in the Paris region: urban centre residential (population density: $20\ 000\ \text{inhab km}^{-2}$) – Paris 13th (75013), suburban residential area (population density $4400\ \text{inhab km}^{-2}$) – Lognes (77185 – Seine et Marne) and forest area (population density: $87\ \text{inhab km}^{-2}$) – Fontainebleau (77186 – Seine et Marne). The prevailing wind direction was W/NW. Because of logistical constraints, only two sites per year were simultaneously sampled: first, Paris/Fontainebleau (distance between the 2 sites, 56.5 km) and then Paris/Lognes (distance 20 km).

The first campaign was carried out for two periods: summer 2010 and winter 2010/2011 at the urban (Paris 13th) and the forest (Fontainebleau) sites. The second campaign was carried out in summer 2011 and winter 2011/2012 and was performed at the urban and the suburban sites i.e., Paris and Lognes.

Each sampling period consisted of three successive and individual samples over 14 d, accounting for 42 sampling days. Sampling conditions are presented in Table SM.1. Air sample volumes ranged from 276 to 555 m³.

2.2. Sample collection and treatment

The particulate phases were sampled onto quartz fibre filters (porosity $0.3\ \mu\text{m}$ supplied by Whatman) and gaseous phases onto XAD-2 resin (20–60 mesh). The resin from Sigma–Aldrich (Saint-Quentin Fallavier, France) was cleaned and dried under vacuum, and 75 g were introduced into two brass cartridges (2 cm diameter; 20 cm length). The filters were put on open aluminium filter holders (Ecomesure, Janvry, France) and used in total suspended particle mode (Granier and Chevreuil, 1997). The sampler (filter and resin) was connected to a sampling pump (Busch, model SV1005GP) with a paddle dryer combined with a timer and an air flow meter (Hivolin, Germany). To secure the trapping of all EDCs, air speed through the system was $<170\ \text{cm s}^{-1}$ as stated in the IEPA method (IEPA/APC/85-003, 1985). The air flow in the sampler remained constant, from 1.2 to 1.8 m³ h⁻¹ (Table SM.1). The outdoor temperatures and the concentrations of particulate matter $<10\ \mu\text{m}$

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