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# Fate and spatial variations of polybrominated diphenyl ethers in the deposition within a heavily urbanized area: case of Paris (France)

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## ABSTRACT

In this paper, we present the first results on the geochemical cycle of PolyBrominated Diphenyl Ethers (or PBDE) in the Paris Region (France). In order to provide information about the distribution and mobility of eight PBDE congeners, we first determined the level of contamination of different environmental compartments: i.e. atmosphere, soils and waters. Atmospheric PBDE deposition was estimated from a site located in the centre of Paris. Surface soils (0–10 cm) were collected from multiple wooded, rural and urban locations through the Paris Region (12,000 km<sup>2</sup>). To complete our investigation, we measured PBDE concentrations/contents in the runoff from an urban catchment and settleable particles from the Seine River. Hence, gained results showed that in the superficial soils, the highest concentrations of highly brominated congeners were measured in the vicinity of the most urbanized areas whereas less brominated congeners were widespread in the whole Paris Region. This could be explained by the higher affinity of highly brominated congeners for the solid phase substrata coupled with the fact that the atmospheric deposition occurred mainly through particle deposition (close to 90% of the total atmospheric deposition). To the opposite, the less brominated congeners from the superficial soils were readily transferred to the dissolved phase of runoff and could reach more distant terrestrial and aquatic systems. Finally, a mass transfer was established at the scale of the Paris metropolitan city (105 km<sup>2</sup>). It showed that the cycle of PBDE in this particular urban area is highly dynamic with multiple sources and sinks, and rapid transfers between the ecosystem compartments.

Key words | atmospheric deposition, Paris Region, PBDE, soil contamination, surface waters, transfer

## INTRODUCTION

From an ecological point of view, the emission of anthropogenic pollutants is of increasing concern. Polybrominated diphenyl ethers (or PBDE) are emerging xenobiotics that are commonly added to building materials, automotive parts, plastics, textiles, foam furnishings and electronic equipments as flame retardants. The PBDE were showed to be carcinogenic and potential endocrine disruptors (neuro-development toxicants) (Branchi *et al.* 2003; Costa & Giordano 2007; Talsness 2008). Since they are volatile, PBDE can spread out in the ecosystems: in air, in soils, in aquatic systems and their living organisms. Recent studies

showed that the transport and removal of PBDE from the atmosphere and their subsequent transfer to the surface aquatic environments are strongly influenced by their partitioning between gaseous, aqueous and particulate phases (Cetin & Odabasi 2008; Oram *et al.* 2008). Throughout the world, the fate of PBDE in urban regions has given rise to numerous studies (Hoh & Hites 2005; Harrad & Hunter 2006; Li *et al.* 2008). Yet, in France, the partitioning and transfers of these xenobiotics between environmental compartments are very poorly known (no study found to our knowledge). The lack of data is all

the more manifest when considering the Paris Region. A region that displays multiple sources of PBDE coupled with a high density of population. Hence, our study aimed at: i) determining the PBDE concentrations in the atmosphere, waters and soils from the Paris Region, ii) assessing the PBDE partitioning and mobility between the different phases (gaseous, dissolved and particulate) and iii) providing information about the PBDE amounts and fluxes in the Paris metropolitan city area.

## MATERIAL AND METHODS

### Sample collection

The Paris metropolitan city, with a population of 2.2 million people, is the most densely populated city in the EU (20,900 inhab. km<sup>-2</sup>). Paris metropolitan city is located in the Île-de-France Region (Paris Region in this paper) which includes 11.6 million inhabitants while it represents less than 2% of the French territory (12,011 km<sup>2</sup>). In this area, the Seine River with its tributaries carries most of the waterway traffic and conveys most of the water-discharged pollutants. The pollution primarily originated from the multiple human activities: the heavy road traffic, fuel facilities, industrial and domestic heating, factories, waste incineration plants, etc.

During the March-May 2008 period, atmospheric samples for PBDE measurements in the gaseous and the particulate phases were collected on a weekly basis. A sampling station was set up on the roof of the Pierre and Marie Curie University (PMCU, 48°50'48" N–2°21'23" E) in the 5th district of Paris. The PMCU station has been used as a reference site for atmospheric deposition in previous works on PAHs (Garban *et al.* 2002) and PCBs (Chevreuil *et al.* 1996). Located downtown, it is representative of the Parisian urban pattern with heavy automobile traffic and domestic heating. Using a high volume pump, air was drawn through a pre-cleaned quartz fibre filter (Ø 110 mm, 0.7 µm pore size) and two pre-cleaned polyurethane foam plugs, PUFs (Ø 50 mm, density 30 kg m<sup>-3</sup>) connected in series. About 700 m<sup>3</sup> of air were collected for each sample. Exposed PUFs and filters (folded in aluminum foil) were kept frozen until further analyses.

Meanwhile, to evaluate the resulting contamination of the terrestrial environment, multiple samples (N = 120)

of superficial soils (10 cm long cores of about 600 kg m<sup>-3</sup> dry density) were collected in the Paris Region at various locations. To lower the heterogeneity and improve representativeness, all samples were prepared by mixing together three different soils collected within 10–100 m distance one from another. The homogenised samples were freeze dried, tested for their organic matter compositions and then stored in dark conditions until analysis.

Concomitantly to the atmosphere and superficial soils samplings, the total atmospheric deposition (wet + dry) (N = 8) and runoff waters (N = 13) were sampled to study the deposition and post-depositional fate of PBDE. The atmospheric deposition and runoff waters were collected in the eastern suburbs of Paris, ~10 km from the PMCU. This site is typical of dense urban areas within Paris conurbation (Zgheib *et al.* 2008). The collection system for the atmospheric deposition consisted of a 1 m<sup>2</sup> surface stainless steel funnel attached to a 30 L pre-cleaned stainless steel drum. Runoff waters (the catchment area of the watershed was ~230 ha) were collected using refrigerated automatic samplers (Bühler 1029) equipped with ultra-cleaned glass jars (all the glassware was thoroughly washed with detergent, rinsed with ultrapure water and precombusted for 5 h at 500°C). Within 2 h of collection, the dissolved phase (including the free, colloidal and the hydrosoluble fractions of PBDE) was separated from the particles by filtration on a precombusted quartz fibre filters (Ø 90 mm, 0.7 µm pore size).

Finally, in order to probe PBDE transfers towards the sediments, traps for settleable particles (N = 3) were deployed in the Seine River over a period of 4 weeks (October–November 2008). They provided settleable PBDE-loaded particles from the Seine River at 3 different locations downstream from Paris (at Austerlitz, Asnières and Epinay).

### Sample treatments and analyses

In order to extract the PBDE, PUFs were ultra-sonicated for 15 min in a mixture of dichloromethane (DCM), n-heptane and acetone (10:5:5 v/v), kept in the extraction solvent overnight and ultra-sonicated for an additional 15 min. Filters and solid samples (particles and surface soils) were extracted in a mixture of DCM and methanol (7:1 v/v)

using focused microwave-assisted extractions in open cells (SOXWAVE, Prolabo, France). The PBDE in the dissolved phase were trapped by passing through a C-18 SPE cartridge. They were subsequently eluted using a mixture of DCM and ethyl acetate (4:1 v/v). The sample amounts extracted for PBDE analyses in the soils and the dissolved phases were 1-3 g and 500–2,000 mL, respectively. The obtained extracts were then cleaned-up using silica gel columns.

Eight PBDE congeners among most produced ones (BDE-47, -99, -100, -153, -154, -183, -205 and -209) were analyzed by gas chromatography/mass spectrometry in electron impact ionization mode (GC/MS-EI) equipped with a fused silica capillary column (10 m × 0.25 mm I.D., film thickness 0.1 μm; Rtx-5) and using helium as a carrier gas (1.8 mL min<sup>-1</sup>). The mass detector was operated in selective ion monitoring mode for the [M + n]<sup>+</sup> and [M + n - 2Br]<sup>+</sup> ions.

The relative standard deviation (SD), calculated on five replicate samples of about 50 pg L<sup>-1</sup>, was lower than 15%. The mean recovery and variance associated with the accuracy for PBDE determinations were 90 ± 30% and were regularly checked, using a reference material (NIST-1944) as certified reference material. Procedural blanks were routinely extracted, cleaned, concentrated and analyzed identical to the samples to assess possible contamination. Finally, the detection limits, defined as 3.3 times the standard deviation of blanks, were usually 0.2–0.5 pg for BDE-47, -99, -100, -153, -154 and -183, and 4–20 pg for BDE-205 and -209.

## RESULTS AND DISCUSSION

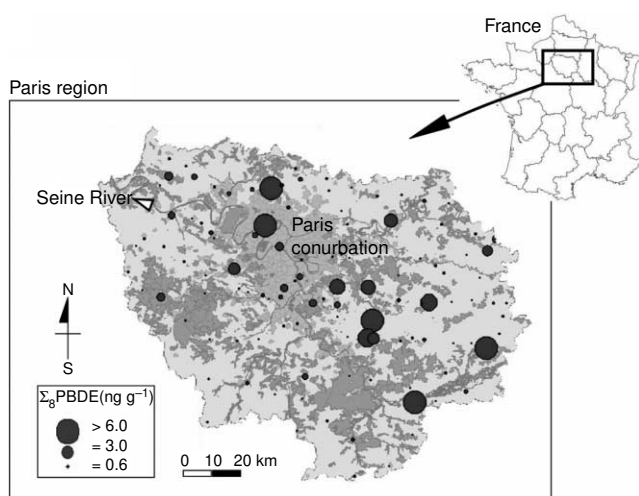
### PBDE in the atmosphere of Paris

In the ambient air of Paris, Σ<sub>8</sub>PBDE (sum of all tested congeners) averaged (± SD) 55 ± 12 pg m<sup>-3</sup> (60% particle-bound) (Table 1). These values are in the high range of concentrations reported in the literature (Lee *et al.* 2004; Chen *et al.* 2006; Cetin & Odabasi 2007, 2008). BDE-47, -99 and -209 constituted 25 → 90% of Σ<sub>8</sub>PBDE. The contributions of BDE-209 were 10–70% in the gaseous, 15–90% in the particulate and 10–80% in the dissolved phase of the total deposition (Table 1). PBDE were mostly found in particles: logK<sub>D</sub>(part./gas) = 7.9 ± 0.6 and logK<sub>D</sub>(part./disso.) = 5.4 ± 0.6 (with K<sub>D</sub> = [BDE]<sub>part.</sub> : [BDE]<sub>gas/diss.</sub>; K<sub>D</sub>(part./gas) in m<sup>3</sup> kg<sup>-1</sup> and K<sub>D</sub>(part./disso.) in L kg<sup>-1</sup>). Therefore, dry deposition (the deposition of atmospheric solids was calculated according to Zhang *et al.* 2001; 0.15 ± 0.02 g m<sup>-2</sup> d<sup>-1</sup>) appeared to be the main pathway for atmospheric PBDE to reach the surface ecosystems.

The annual atmospheric deposition of Σ<sub>8</sub>PBDE for the Paris metropolitan city (105 km<sup>2</sup> of surface) was roughly estimated to be 10 kg (close to 90% as particles). This was about half the total BDE-209 emissions to the atmosphere according to regional-scale estimations (about 20 kg; European Commission 2004). The gaseous (0.7 ± 0.5 cm s<sup>-1</sup> of depositional velocity) and dissolved (about 600 mm y<sup>-1</sup> of rainfall amount) depositions were comparable: each constituted less than 10% of the total deposited amount

**Table 1** | Atmospheric concentrations and deposition fluxes (average ± SD) of measured PBDE congeners at the Paris metropolitan site (N = 8)

Congener	Concentrations			Deposition fluxes		
	Gas (pg m <sup>-3</sup> )	Particles (ng g <sup>-1</sup> , d.w.)	Dissolved (ng L <sup>-1</sup> )	Gas (ng m <sup>-2</sup> d <sup>-1</sup> )	Particles (ng m <sup>-2</sup> d <sup>-1</sup> )	Dissolved (ng m <sup>-2</sup> d <sup>-1</sup> )
BDE-47	2.0 ± 0.6	270 ± 150	0.4 ± 0.2	1.2 ± 0.4	40 ± 20	0.6 ± 0.3
BDE-100	0.7 ± 0.4	90 ± 60	0.3 ± 0.1	0.4 ± 0.3	15 ± 10	0.4 ± 0.2
BDE-99	3 ± 1	380 ± 200	0.8 ± 0.5	1.8 ± 0.6	60 ± 30	1.3 ± 0.8
BDE-154	0.2 ± 0.1	37 ± 3	0.22 ± 0.09	0.12 ± 0.06	6 ± 1	0.4 ± 0.2
BDE-153	4 ± 2	46 ± 4	0.2 ± 0.1	2.4 ± 1.0	7 ± 1	0.3 ± 0.2
BDE-183	0.9 ± 0.6	120 ± 10	0.27 ± 0.06	0.6 ± 0.4	19 ± 2	0.5 ± 0.1
BDE-205	4.0 ± 0.5	14 ± 2	0.8 ± 0.6	2.4 ± 0.3	2.2 ± 0.3	1.4 ± 1.0
BDE-209	6 ± 3	590 ± 230	3 ± 2	4 ± 2	90 ± 40	5 ± 3
Σ <sub>8</sub> PBDE	21 ± 8	1,700 ± 800	6 ± 3	13 ± 5	270 ± 100	9 ± 5



**Figure 1** | Distribution and concentrations of  $\Sigma_8$ PBDE within the superficial soils of the Paris Region (N = 120).

of PBDE. BDE-209 was the main deposited congener ( $2-5 \text{ kg y}^{-1}$ ) followed by BDE-99 ( $1-3 \text{ kg y}^{-1}$ ) and BDE-47 ( $1-2 \text{ kg y}^{-1}$ ).

### PBDE in the soils of the Paris Region

In the superficial soils of the Paris Region,  $\Sigma_8$ PBDE ranged between  $0.2$  and  $43.9 \text{ ng g}^{-1}$  (Figure 1). The average (and mean) concentration was  $1.8$  (and  $0.7$ )  $\text{ng g}^{-1}$  and was comparable to the available data from the literature (Hassanin *et al.* 2004; Harrad & Hunter 2006; Zou *et al.* 2007). As for PBDE in the atmosphere, BDE-209 was predominant in superficial soils (55% of  $\Sigma_8$ PBDE (59% as mean)) followed by BDE-47 (9% (8% as mean)) and

BDE-99 (11% (9% as mean)). According to these figures, the amount of  $\Sigma_8$ PBDE in the superficial soils of Paris was between 7 and 14 kg ( $500-1,300 \text{ kg}$  for the whole Paris Region). This roughly represented the equivalent of one year of urban atmospheric  $\Sigma_8$ PBDE deposition.

Due to the anthropogenic nature of PBDE sources (de Wit 2002),  $\Sigma_8$ PBDE levels increased in relation to urbanization (Table 2). As a matter of fact, no significant difference in the average (and mean)  $\Sigma_8$ PBDE concentrations was observed between forested  $1.20$  ( $0.59$ )  $\text{ng g}^{-1}$  and rural  $1.93$  ( $0.66$ )  $\text{ng g}^{-1}$  domains. However, it reached up to  $2.24$  ( $1.08$ )  $\text{ng g}^{-1}$  in highly urbanized regions. Along with  $\Sigma_8$ PBDE increase, a significant increase of the contribution of BDE-209 was observed (from 53 to 65%), whereas BDE-47 and BDE-99 contributions remained relatively stable (6–7 and 6–9%, respectively). This observation indicated that the different PBDE congeners either originated from different urban sources and/or exhibited different geochemical behaviors (e.g. trapping and mobilization). To illustrate, the distribution patterns of BDE-47 and 99 strongly contrasted with that of BDE-209: the former exhibited localized maxima of concentrations that could be located far from urbanized areas whereas the BDE-209 displayed a notable urban signature.

### PBDE in runoff and settleable particles

The observed distributions of PBDE in the superficial soils of the Paris Region clearly indicated that the different PBDE congeners had distinct origins and/or exhibited

**Table 2** | Summary statistics for PBDE concentrations in the principal categories of soils available in the Paris Region; [n] number of samples, average (mean)  $\pm$  SD, range (minimum-maximum)

Congener	Wooded soils ( $\text{ng g}^{-1}$ , d.w.) [13]	Agricultural soils ( $\text{ng g}^{-1}$ d.w.) [59]	Urban soils ( $\text{ng g}^{-1}$ d.w.) [48]	Paris Region* ( $\text{ng g}^{-1}$ , d.w.) [120]
BDE-47	$0.22$ ( $0.04$ ) $\pm$ $0.37$ 0.028–1.198	$0.27$ ( $0.04$ ) $\pm$ $1.05$ 0.015–7.307	$0.15$ ( $0.07$ ) $\pm$ $0.20$ 0.021–1.038	$0.23$ ( $0.05$ ) $\pm$ $0.71$ 0.015–7.307
BDE-99	$0.17$ ( $0.05$ ) $\pm$ $0.26$ 0.020–0.893	$0.46$ ( $0.05$ ) $\pm$ $1.93$ 0.007–13.167	$0.22$ ( $0.07$ ) $\pm$ $0.38$ 0.014–1.813	$0.34$ ( $0.06$ ) $\pm$ $1.22$ 0.007–13.167
BDE-209	$0.38$ ( $0.31$ ) $\pm$ $0.25$ 0.067–0.952	$0.57$ ( $0.36$ ) $\pm$ $0.084$ 0.070–5.568	$1.50$ ( $0.70$ ) $\pm$ $2.97$ 0.099–16.77	$0.72$ ( $0.42$ ) $\pm$ $1.15$ 0.067–16.77
$\Sigma_8$ PBDE	$1.20$ ( $0.59$ ) $\pm$ $1.42$ 0.225–5.112	$1.93$ ( $0.66$ ) $\pm$ $5.93$ 0.242–43.934	$2.24$ ( $1.08$ ) $\pm$ $3.24$ 0.324–17.971	$1.83$ ( $0.73$ ) $\pm$ $4.33$ 0.242–43.934

\*PBDE concentrations for the whole Paris Region were calculated using the spatial areas and measured concentrations relative to the different soil occupations (IUARIF data).



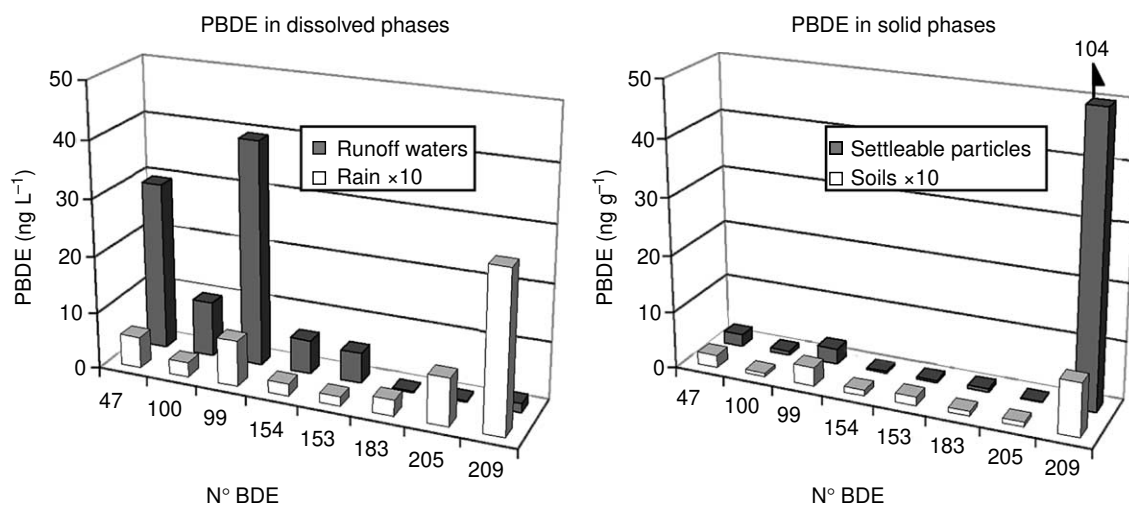
different post-depositional fates. To test this observation, we measured the concentrations of PBDE in runoff and settleable particles from the Seine River. In the dissolved phase of runoff, the average  $\Sigma_8$ PBDE concentrations were more than one order of magnitude higher than in the dissolved phases of the total atmospheric deposition:  $95 \pm 20$  vs.  $6 \pm 3 \text{ ng L}^{-1}$ . This discrepancy mainly resulted from the relative enrichment with BDE-47 and BDE-99: their sum accounted for 50–80% of  $\Sigma_8$ PBDE. This observation confirmed that both congeners could readily be transferred from the surface soils to the dissolved phase of aquatics systems during storm events (Figure 2).

Average  $\Sigma_8$ PBDE concentrations in the settleable particles from the Seine River were more than one order of magnitude higher than in the superficial soils of the Paris Region:  $110 \pm 50$  vs.  $1.83 \pm 4.33 \text{ ng g}^{-1} \text{ d.w.}$ , respectively. In fact, BDE-209 high partition coefficient ( $K_{ow} = 10^{6.5}$ ; MacGregor & Nixon 1997) and binding affinity to solids likely made this congener to constitute  $93 \pm 2\%$  of  $\Sigma_8$ PBDE in settleable particles. These observations suggest that, unlike the BDE-47 and BDE-99, the main pathway for BDE-209 to reach the aquatic systems could consist of the leached particles. This was in accordance with the results by Oram *et al.* (2008) stating that runoff from local tributaries represented the largest contributor to BDE-209 loads in the San Francisco Bay, CA.

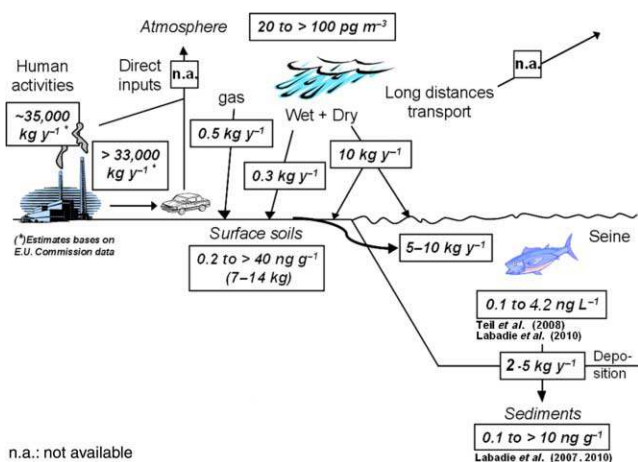
### PBDE cycle in the Paris metropolitan city area

The amounts and fluxes of  $\Sigma_8$ PBDE were investigated for the Paris metropolitan city area according to the measured concentrations from the collected samples and the available data from the literature (Figure 3). Since samples were representative of  $\Sigma_8$ PBDE levels for a limited spatial area and time period, the displayed cycle only provides indicative data. In 2001, about 8,300 tons of  $\Sigma_8$ PBDE were produced in the EU (EU data, 2004). More than 90% ( $>7,600$  tons) of the produced  $\Sigma_8$ PBDE consisted of BDE-209 that could ubiquitously be found in mass-consumption products (like clothing, furniture, electronic equipment, etc.). Reported to the Paris metropolitan population, these figures roughly represented  $35,000 \text{ kg y}^{-1}$  for  $\Sigma_8$ PBDE and  $33,000 \text{ kg y}^{-1}$  for BDE-209.

Despite the lack of data about the direct  $\Sigma_8$ PBDE inputs to the atmosphere and their subsequent long range transport, measurements indicated that the atmospheric  $\Sigma_8$ PBDE deposition accounted for  $<0.1\%$  of the PBDE production. This observation suggested that only a minor fraction of the produced  $\Sigma_8$ PBDE could reach the atmosphere and/or that the atmospheric  $\Sigma_8$ PBDE inputs were readily transported away from sources. Several researches dedicated to the transport of PBDE in the UE atmosphere clearly highlighted the long distance dissemination potential of less brominated congeners and the enrichment with highly brominated ones near urban areas (Law *et al.* 2006;



**Figure 2** | Concentrations of PBDE: (left) in the dissolved phases of rainfall and runoff, (right) in the superficial soils from the Paris Region and settleable particles from the Seine River.



**Figure 3** | Incomplete cycle of  $\Sigma_8$ PBDE in the Paris area (105 km<sup>2</sup>). Given data are only indicative of  $\Sigma_8$ PBDE amounts and fluxes on a seasonal time scale (2007–2008).

Wilford *et al.* 2008). Therefore, and as meteorological data indicated (the characteristic time for airborne particle concentration variations  $\sim 1$  week; Tombette & Sportisse 2007), it is likely that most of the anthropogenically produced PBDE hardly accumulated into the Parisian atmosphere and was transported away. Only a small fraction of highly brominated congeners could be deposited via airborne particles in the immediate vicinity of the urban sites.

With regard to the terrestrial  $\Sigma_8$ PBDE, we used the measured concentrations and mean rainfall amount, to estimate the amount of  $\Sigma_8$ PBDE transferred from the surface soils to the dissolved phase of runoff in the Paris metropolitan city area. Calculations indicated that a total of  $5-10 \text{ kg y}^{-1}$  ( $4-7 \text{ kg y}^{-1}$  as dissolved) of  $\Sigma_8$ PBDE may have been transferred from the surface soils to the dissolved phase of runoff. This was more than half the  $\Sigma_8$ PBDE amounts of the superficial soils (i.e. 7–14 kg). Hence, the lifespan for the PBDE within the soils of Paris metropolitan city is thought to be short (less than a few years) and its transfer to the aquatic system to be fast. However, since BDE-47, -99 and BDE-209 displayed different vectors of transport (dissolved phase and particles, respectively) their dissemination ranges and half-lives in the soils, sediments and aquatic systems should be different. To illustrate, due to the faster deposition of the conveyed particles, the impregnation of terrestrial and aquatic ecosystems with BDE-209 is expected to primarily occur near the emission sources. This was confirmed by our previous observations (Table 2)

showing that BDE-47 and -99 were widespread in the soils of Paris Region whereas the BDE-209 was preferentially found close to urban areas (the acknowledged areas of usage and emissions).

Once into the riverine ecosystems, PBDE can be transported in the forms of dissolved and colloidal species or suspended particles. They can also be deposited at the sediment-water interface by the settleable particles. From the measured concentrations of  $\Sigma_8$ PBDE of settleable particles and the annually deposited amounts of sediments (25,000–40,000 tons according to the French Ministry of ecology and sustainable development),  $2-5 \text{ kg y}^{-1}$  of  $\Sigma_8$ PBDE reach the sediments. Despite the uncertainties surrounding calculations, this estimation suggested that a significant proportion of  $\Sigma_8$ PBDE (20%-all) may be trapped into sediments. However, as a result of remobilization mechanisms (e.g. via BDE-209 degradation and production of less hydrophobic congeners and/or solid phase dissolution followed by the upward diffusion of the solubilized molecules), sediments may also serve as a long term sources of PBDE for the water column and developing aquatic organisms.

## CONCLUSIONS

For the first time, the cycle of PBDE was investigated in the urban environment of the Paris Region. Gained results showed that the different studied PBDE congeners exhibited contrasted geochemical behaviors affecting their mobilities and spatial distributions. Despite the fact that sources of PBDE could not be clearly identified (provisional results highlighted high BDE-209 concentrations in the Parisian wastewater particles), multiple lines of evidence indicated that highly brominated PBDE concentrated in environments near urbanized areas (on the scale from meters to hundreds of meters). To the opposite, less brominated PBDE were widespread in the whole Paris Region. Finally, we showed that the geochemical cycles of PBDE in the Paris metropolitan city area is highly dynamic. It presented pathways of rapid transfers (e.g. wet and dry depositions, runoff leaching) and short characteristic times for PBDE in the terrestrial environments (less than a few years).

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