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## Meta-analysis of environmental contamination by phthalates.

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<b>Abstract:</b>	<p><b>Introduction:</b> Phthalate Acid Esters (PAE), commonly named Phthalates, are toxics classified as endocrine-disrupting compounds; they are primarily used as additives to improve the flexibility in polyvinyl chloride.</p> <p><b>Occurrence:</b> Many studies have reported the occurrence of phthalates in different environmental matrices, however none of these studies has yet establish a complete overview for those compounds in the water cycle within an urban environment. This review summarizes PAE concentrations for all environmental media throughout the water cycle, from atmosphere to receiving waters. Once the occurrences of compounds have been evaluated for each environmental compartment (urban wastewater, wastewater treatment plants, atmosphere and the natural environment), data were reviewed in order to identify the fate of PAE in the environment and establish whether geographical and historical trends exist. Indeed, geographical and historical trends appear between Europe and other countries such as USA/Canada and China, however they remain location-dependent.</p> <p><b>Discussion:</b> This study aimed at identifying both the correlations existing between environmental compartments and the processes influencing the fate and transport of these contaminants in the environment. In Europe, the concentrations measured in waterways today represent the background level of contamination, which provides evidence of a past diffuse pollution. In contrast, an increasing trend has actually been observed for developing countries, especially for China.</p>
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# Meta-analysis of environmental contamination by phthalates.

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Abstract

*Introduction:* Phthalate Acid Esters (PAE), commonly named Phthalates, are toxics classified as endocrine-disrupting compounds; they are primarily used as additives to improve the flexibility in polyvinyl chloride.

*Occurrence:* Many studies have reported the occurrence of phthalates in different environmental matrices, however none of these studies has yet establish a complete overview for those compounds in the water cycle within an urban environment. This review summarizes PAE concentrations for all environmental media throughout the water cycle, from atmosphere to receiving waters. Once the occurrences of compounds have been evaluated for each environmental compartment (urban wastewater, wastewater treatment plants, atmosphere and the natural environment), data are reviewed in order to identify the fate of PAE in the environment and establish whether geographical and historical trends exist. Indeed, geographical and historical trends appear between Europe and other countries such as USA/Canada and China, however they remain location-dependent.

*Discussion:* This study aimed at identifying both the correlations existing between environmental compartments and the processes influencing the fate and transport of these contaminants into the environment. In Europe, the concentrations measured in waterways today represent the background level of contamination, which provides evidence of a past diffuse pollution. In contrast, an increasing trend has actually been observed for developing countries, especially for China.

*Keyword:* phthalates, DEHP, review, state-of-the-art.

## Introduction

Most of studies, published during the last three decades, have reported several categories of man-made chemicals, classified as “endocrine-disrupting

1 compounds” (EDCs). Among the most frequently cited EDCs, phthalates are of  
2 particular concern due to their ubiquity and generally higher levels found in  
3 environment comparatively to other EDCs (Sanchez-Avila et al. 2009; Fauser et  
4 al. 2003; Staples et al. 1997; Fatoki and Mathabatha 2001). The predominant use  
5 of Phthalic Acid Esters (PAE) is for improving flexibility in polyvinyl chloride  
6 (PVC) (Giam et al. 1984). For instance, the amount of DEHP in PVC depends on  
7 plastic composition. Some products can contain up to 50%, but typically there will  
8 be approximately 30% DEHP in most PVC products (Ranke 2005). Moreover,  
9 their applications extend to industrial and/or domestic sectors, depending on their  
10 molecular weight (Table 1). Low molecular weight, especially Dimethyl phthalate  
11 (DMP) and Diethyl phthalate (DEP) have therefore been incorporated into  
12 cosmetics, fragrance and other personal care-products. Besides, Di-n-butyl  
13 phthalate (DnBP) is also used in epoxy resins, cellulose esters and special  
14 adhesive formulations. Additionally, high molecular weight and branching alkyl  
15 chain PAEs, primarily Butylbenzyl phthalate (BBP), Di-n-octyl phthalate (DnOP)  
16 and Di-(2-ethylhexyl) phthalate (DEHP) are being incorporated into food-  
17 packaging, building materials, home furnishing, clothing and medical products  
18 (Liu et al. 2010; Staples et al. 1997; Jackson and Sutton 2008; Cespedes et al.  
19 2004). Despite their various uses, PAE have in recent studies leveled off in  
20 American, Canadian and European production, mainly as a result of highly  
21 restrictive regulations (EU 2005, 2004, 2007, 2008; CPSI 2008), while worldwide  
22 output has been stagnating to 6,000,000 tons in 2006 (Peijnenburg and Struijs  
23 2006). This difference may stem from the increase in consumption from emerging  
24 nations, such as Brazil, Russia, India and China. The European and North  
25 American productions have however evolved because of the replacement of  
26 DEHP by heavier and more stable phthalates (Di-iso-nonyl phthalate and Di-iso-  
27 decyl phthalate) therefore less subject to release into the environment.

34 Despite their societal and economical benefits, PAE consumption has lead to the  
35 release of PAE into the environment, where they are now ubiquitous and can be  
36 found in air, water, sediments and soils. This study focuses more specifically on 6  
37 most commonly studied congeners of this family (Table 1): Dimethyl phthalate  
38 (DMP), Diethyl phthalate (DEP), Di-n-butyl phthalate (DnBP), Butylbenzyl  
39 phthalate (BBP), Di-n-octyl phthalate (DnOP) and Di-(2-ethylhexyl) phthalate  
40 (DEHP). Most studies in the literature examine the occurrence and behavior of  
41 these six compounds in various environmental matrices (surface water,  
42 wastewater, atmosphere and treated water) given that these compounds are  
43 regularly found in urban and environmental compartments at significant levels  
44 (tens or more  $\mu\text{g.l}^{-1}$ ) (Peijnenburg and Struijs 2006; Vethaak et al. 2005; Cespedes  
45 et al. 2004; Fauser et al. 2003; Abb et al. 2009).

## 49 Strategy of this review

54 Most of the literature on PAEs in the environment, as published since 1990  
55 (Staples et al. 1997; Vethaak et al. 2005; Deblonde et al. 2011) have been  
56 concerned with one or more compartments, primarily downstream compartments  
57 such as WWTP effluent and receiving waters, but none of these works has dealt  
58 with all environmental compartments, in particular urban wastewater, nor with  
59 PAE behavior throughout the whole water cycle. As similarly done for the meta-  
60 analysis of environmental contamination by alkylphenols (Bergé et al. 2012a), the

1 purpose of this review is to collect a substantial dataset for all compartments (Fig.  
2 1) from the atmosphere (gaseous phase and rain water) and extending to urban  
3 areas (industrial, residential and man-made wastewater, WWTP influent and  
4 effluent, WWTP sludge and stormwater) and the natural environment (surface  
5 water, sediments and soils). Unfortunately, the set of congeners reported in the  
6 various papers has not been consistent and equivalent between compartments.  
7 Some papers have reported on as few as one or two congeners (DEHP, DnBP or  
8 DEP), while others have reported on four or six (DMP, DEP, DnBP, BBP, DEHP  
9 and DnOP). In this review, PAE refers systematically to the sum of these six  
10 congeners. From the database we built, statistical calculations have been derived  
11 for certain sample sets. The year of sample extraction was not specified in all  
12 references, so it was assumed that the samples were extracted on the article  
13 publication date. It was also decided that this review focused exclusively on  
14 environmental contamination and not on biota or bioaccumulation processes.  
15 Moreover, the concentration ranges reported in all tables correspond to minimal  
16 and maximal concentrations of each compound for each compartment. The other  
17 values represent median concentrations calculated by authors. This review is also  
18 aimed at determining whether temporal and/or geographical trends can be drawn  
19 in PAE contamination as observed for alkylphenols (Bergé et al. 2012a). To  
20 achieve this objective, the data were examined from geographical perspective.  
21 Recommendations for further research will also be provided.  
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## 26 **ATMOSPHERE**

### 27 **Air contamination**

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34 PAE in the atmosphere or in urban areas are generated by various emission  
35 sources, including volatilization from materials, industrial processes, waste  
36 combustion and wastewater treatment processes (Salapasidou et al. 2011). It has  
37 been calculated that approximately 4% of the quantity of DEHP can evaporate  
38 from materials such as floorings, rain clothes, toys, soles of shoes (Ranke 2005).  
39 DEHP was indeed detected in aerosols emitted from the aeration tank of a WWTP  
40 ranging from 71.1 to 228 ng/m<sup>3</sup> (Lepri et al. 2000). As shown in Table 2, a small  
41 number of air samples (10 references in all) has been reported for PAE. From data  
42 collected, PAE contamination globally lies in the 1-50 ng.m<sup>-3</sup> range for all  
43 congeners, except for DEHP presenting higher levels (up to 3,640 ng.m<sup>-3</sup>). The  
44 analysis of this database reveals that European samples were consistently above  
45 the values reported in the world (USA, China, etc.), and DEHP was the most  
46 abundant phthalate in air, with concentrations ranging from 0.08 to 3,640 ng/m<sup>3</sup>  
47 (Müller et al. 2003). Recent studies conducted by Tlili et al. (2010) and Teil et al.  
48 (2006), in the Parisian area (France), underscored that phthalates are preferentially  
49 associated with the gaseous phase rather than aerosols. For instance, Tlili et al.  
50 (2010) reported that between 60% to 70% against 40% to 30%, respectively, of  
51 phthalates are associated to gaseous phase. In addition, the same authors reported  
52 that the phthalates exhibiting short alkyl chains and high vapor pressures  
53 (especially, DMP and DEP) are predominant in the gaseous phase and inversely  
54 for the others compounds. Study conducted by Xie et al. (2005) showed that the  
55 air-sea exchange was preferentially dominated by the deposition, especially for  
56 DnBP (-60 to -686 ng/m<sup>2</sup>/d) and BBP (-4 to -28 ng/m<sup>2</sup>/d). Additionally to this  
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1 latter point, the fluxes calculated for DEHP (-95 to 686 ng/m<sup>2</sup>/d) highlighted a  
2 more complicated mechanism. Moreover, DnBP and DEHP total (dry and wet)  
3 depositions, to the Great Lakes (Canada), provided by Eisenreich et al. (1981)  
4 were estimated at 3.7 and 16 tons per year, respectively. This indicates that the  
5 atmosphere could be one of the major contamination sources of PAEs. However,  
6 according to Staples et al. (1997) and, more recently to Xie et al. (2007),  
7 phthalates are subject to photo-degradation, and therefore generally do not persist  
8 in the atmosphere. Authors reported photo-degradation half-lives of common  
9 phthalates ranging from 0.3 to 15 days. A study led by Salapasidou et al. (2011)  
10 underlined that concentrations of DEHP were significantly higher on urban-traffic  
11 areas (4.63 to 45.0 ng/m<sup>3</sup>; median 19.4 ng/m<sup>3</sup>) than on urban-industrial site (up to  
12 6.50 ng/m<sup>3</sup>; median 2.80 ng/m<sup>3</sup>) implying generally an input from vehicular  
13 emissions. Similarly, Wang et al. (2008) reported that concentrations were about  
14 3.5 higher above urban sites than above suburban sites. These authors attributed  
15 this difference to both many point-sources and environmental recycling.  
16 Additionally, Dargnat (2008) underlined the presence of seasonal variability  
17 above the Parisian area, with smaller concentrations for spring and during  
18 summer. This was previously observed by Guidotti et al. (2000) in Italy and Teil  
19 et al. (2006) in France above the same area. Due to the limited number of studies  
20 available, no distinct temporal or geographical trend could be drawn.  
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## 25 **Rain water**

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29 A few measurements of PAE in rain water have been undertaken (see Table 3, 10  
30 references in all). Outdoor levels of DEHP lie on the order of 0.02 to 39.0 µg.l<sup>-1</sup>  
31 and 0.03 to 11.0 µg.l<sup>-1</sup> for DnBP. At the scale of Europe, data have shown that  
32 PAE concentrations in rain water seem to decline, which implies that the  
33 environment in European countries is exposed to decreasing contamination. In  
34 addition, data show that PAE concentrations in rain water are more important in  
35 Northern European countries than Southern European countries, which implies  
36 that the fate of PAE in the atmosphere could be governed by atmospheric currents.  
37 A similar phenomenon has been reported for alkylphenols (Bergé et al. 2012a).  
38 Dargnat (2008) emphasized that passive volatilization from buildings, in urban  
39 areas, was the prevailing source of phthalate in rain water. Author therefore  
40 concluded that phthalate uses in building materials constitutes a significant source  
41 of contamination. Finally, Vethaak et al. (2005), in the Netherlands, reported that  
42 most of phthalates present in rain water were at concentrations comparable to  
43 those in surface water.  
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## 48 **URBAN AREAS**

### 49 **Sewer contamination**

#### 50 *Industrial, man-made and residential wastewater*

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60 As reported in Table 4 and Figure 2, only a few measurements of PAE have been  
61 performed in wastewater (4-5 references for each type of wastewater). Industrial  
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1 DEHP levels were in the 0.01 to 4,400  $\mu\text{g}\cdot\text{l}^{-1}$  range (median: 34.6  $\mu\text{g}\cdot\text{l}^{-1}$ ), i.e. twice  
2 as low as levels in residential wastewater (3.30-160  $\mu\text{g}\cdot\text{l}^{-1}$ ; median: 61.3  $\mu\text{g}\cdot\text{l}^{-1}$ )  
3 and in man-made wastewater (0.60-470  $\mu\text{g}\cdot\text{l}^{-1}$ ; median: 66.0  $\mu\text{g}\cdot\text{l}^{-1}$ ). In Europe and  
4 North America, it has been reported that high DEHP and other phthalate  
5 concentrations were measured in industrial wastewater. In the USA, for instance,  
6 DEHP levels of 4,400  $\mu\text{g}\cdot\text{l}^{-1}$  were measured in untreated industrial wastewater  
7 (Clark et al. 2003). Similarly, Jackson and Sutton (2008) quoted DEHP, DEP and  
8 DnBP levels reaching 2,700, 100 and 120  $\mu\text{g}\cdot\text{l}^{-1}$  in industrial laundry and adhesive  
9 manufacturer wastewaters. Both studies led by Vethaak et al. (2005) and Sanchez-  
10 Avila et al. (2009) showed that levels between 45 and 100  $\mu\text{g}\cdot\text{l}^{-1}$  have also been  
11 measured in residential wastewater in the Netherlands and Spain. Some authors  
12 mentioned that the high concentrations measured in Maresme wastewater  
13 (Catalonia, Spain) and Parisian sewer network (France) would point out that PAE  
14 are still being produced and used in industrial, household and agricultural  
15 formulations despite the implementation of European regulations (EU 2005, 2004,  
16 2007) restricting their use within the EU to levels  $< 0.1\%$  (Sanchez-Avila et al.  
17 2009; Bergé et al. 2012c).

### 21 *Wastewater Treatment Plant Influent*

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25 As opposed to industrial, man-made and residential wastewater, a large number of  
26 WWTP samples have been analyzed for PAEs (see Table 5, i.e. 13 references).  
27 Moreover, according to these concentrations, the PAE median levels reported for  
28 WWTP influent were in the same order of magnitude than residential and man-  
29 made wastewater but in the lower range of industrial wastewater (PAE: 50.7  $\mu\text{g}\cdot\text{l}^{-1}$   
30 in WWTP influent vs 72.6, 42.8 and 139.9  $\mu\text{g}\cdot\text{l}^{-1}$  in these types of wastewater,  
31 respectively, Fig. 2) (Jackson and Sutton 2008; Sanchez-Avila et al. 2009; Bergé  
32 et al. 2012b). The variations in PAE concentrations among WWTP influent have  
33 been further investigated using published data, which have provided sufficient  
34 data for geographical areas including Europe and North America (Fig. 3).  
35 Surprisingly, no data was available for Chinese plants. This figure also points out  
36 the 10<sup>th</sup> and 90<sup>th</sup> percentiles (i.e. “the whiskers”) as well as the 25<sup>th</sup> and 75<sup>th</sup>  
37 percentiles (“boxes”); the medians are also highlighted. PAE median  
38 concentrations in samples from North America and Europe were quite similar:  
39 49.5 and 51.8  $\mu\text{g}\cdot\text{l}^{-1}$ , respectively. It is also interesting to note that European  
40 countries display only a few outliers to the high side (see Table 5). In Spain and  
41 France, these outlier concentrations typically exceed 160 and 100  $\mu\text{g}\cdot\text{l}^{-1}$ , i.e. 2-3  
42 times the average level, respectively. These outliers are not yet explained,  
43 although the presence of untreated loaded industrial effluent could be  
44 hypothesized. Additionally, the database analysis indicates that the PAE  
45 concentrations measured in French influent were quite similar to those measured  
46 in Europe except in Spain, where DEHP concentrations in influent were  
47 significantly lower: i.e. 5 times the average level (Reyes-Contreras et al. 2011;  
48 Martin-Ruel et al. 2010; Bergé et al. 2012b). This difference could be due to the  
49 lower DEHP consumptions in Spain where this congener was found in lower  
50 levels than in other European countries.  
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## Wastewater Treatment Plant Effluent

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4 Understanding the biodegradation processes of phthalic compounds proves to be a  
5 critical factor in predicting the fate of these compounds in the environment. From  
6 an overall standpoint, the removal of PAE by conventional activated sludge  
7 WWTPs has been well documented (Dargnat et al. 2009; Tan et al. 2007; Barnabé  
8 et al. 2008; Marttinen et al. 2003). Most of studies reported removal up to 90%.  
9 Additionally, recent studies conducted by Bergé et al. (2012b) and Gasperi et al.  
10 (2010) explained that biofiltration coupled to physicochemical lamellar  
11 clarification could be a promising alternative to activated sludge tanks for today  
12 plant built in large urbanized areas where the building pressure makes available  
13 land scarce. A large number of samples from WWTP effluent have been analyzed  
14 with respect to PAE (see Table 6). Generally speaking, effluent concentrations lie  
15 in the range of 0.02-49.9  $\mu\text{g.l}^{-1}$  (mean: 0.80  $\mu\text{g.l}^{-1}$ ) for DEP and 0.02-69.0  $\mu\text{g.l}^{-1}$   
16 (mean: 2.44  $\mu\text{g.l}^{-1}$ ) for DEHP, with PAE ranges extending from 0.07 to 108  $\mu\text{g.l}^{-1}$ .  
17 As with WWTP influents, no clear difference has been observed for PAE  
18 concentrations in final effluent whatever the location (Fig. 3). Median PAE  
19 concentrations in the samples from North America and Europe were quite similar,  
20 lying in the 4.85-5.35  $\mu\text{g.l}^{-1}$  range. As previously mentioned for WWTP influent,  
21 European effluent has exhibited a few high outliers. In Spain, these outlier  
22 concentrations typically exceeded 50  $\mu\text{g.l}^{-1}$ ; i.e. 10 times the average level. The  
23 little availability of phthalate data before 2000 does not allow defining a trend of  
24 these compounds in the discharges of WWTP. Surprisingly, the PAE median  
25 concentration measured in Spanish WWTP effluent exceeded that in WWTP  
26 influent (de los Rios et al. 2012; Bizkarguenaga et al. 2012; Sanchez-Avila et al.  
27 2009; Sanchez-Avila et al. 2012). However, no explanation could be brought by  
28 authors to elucidate this difference, but the WWTP efficiency could however be  
29 subject to questioning.  
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36 According to our review, PAE can display moderate (50%) to high (94%) removal  
37 rates. In addition, a study conducted by Vogelsang et al. (2006) explained that  
38 chemical and biological treatment eliminates from 50-60% of phthalates in a  
39 Norwegian WWTP. In Spain, Sanchez-Avila et al. (2009) reported PAE removal  
40 efficiency of 68%. Finally, a study led by Marttinen et al. (2003) explained that  
41 DEHP has been removed from wastewater with an efficiency of 94%. In addition,  
42 both studies conducted by Fauser et al. (2003) and in EU (2008) reported similar  
43 removal efficiencies for DEHP. Ordinarily, phthalates are removed by different  
44 processes occurring within WWTP, including solid settlement, sorption to sludge,  
45 volatilization, biodegradation, hydrolysis and/or photolysis (Rogers 1996).  
46 Sorption on sludge is considered to be one of the major pathways for PAE  
47 removal in WWTP. Moreover, even if DEHP may be considered inherently  
48 biodegradable under aerobic conditions (Staples et al. 1997), it was not removed  
49 by the biological pathway. A study conducted by Gavala et al. (2004) showed that  
50 enzymatic pretreatment increased DEHP biodegradation rate in secondary sludge.  
51 Furthermore, it was showed that recirculating sludge and thus retaining specific  
52 micro-organisms could stabilize removal capacity up to 86% whereas continuous  
53 flow had a varying removal of 77-88% (Oliver et al. 2007, 2005). Alternatively,  
54 Vogelsang et al. (2006) and Staples et al. (1997) reported that DEP, a soluble and  
55 biodegradable compound under aerobic conditions, has been removed during  
56 biological treatment process, although it has not been significantly eliminated by  
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1 any chemical or mechanical sedimentation processes. As a general rule, in WWTP  
2 effluent, DEHP is expected to be the most abundant congener. Phthalates'  
3 distribution is however lightly different, compared to WWTP effluents, with a  
4 higher proportion of DnBP and DEP (Fig. 4).  
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## 6 **Wastewater Treatment Plant Sludge**

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9 As mentioned above, sorption on sludge is considered to be one of the major  
10 pathways for PAE removal in WWTP (Kinney et al. 2006; Cai et al. 2007;  
11 Barnabé et al. 2008; Clara et al. 2010) (Fig. 5). A large number of samples have  
12 been analyzed for PAEs (see Table 7, i.e. 30 references). In accordance with its  
13 predominance in wastewater, Harrison et al. (2006) and Tan et al. (2007) showed  
14 that DEHP was the most abundant phthalate in sludge (Fig. 4). In addition, most  
15 studies have reported significant DEHP levels in sludge, i.e. in the range of 150-  
16 600 mg/kg.dw. For example, levels up to 3,514 mg/kg.dw in digested sludge from  
17 Spain (Abad et al. 2005), 661 mg/kg.dw in digested sludge from Sweden  
18 (Sweetman 1994) and 578 mg/kg.dw in digested sludge from USA (Staples et al.  
19 1997) have been recorded. Additionally, a recent study conducted by Clara et al.  
20 (2010) explained that PAE sorption importance and phthalate removals via sludge  
21 increased with the molecular weight, and is therefore depending on the molecular  
22 weight (Table 1). As a complement, authors reported that proportional mass  
23 fraction removed with the sludge amounts to 3% for DMP, 1% for DEP, 76% for  
24 DnBP, 21% for BBP and 78% for DEHP. Generally speaking, sludge contents lie  
25 in the range of 0.02-2.00 mg/kg.dw (mean: 0.19 mg/kg.dw) for DMP, 0.01-11.0  
26 mg/kg.dw (mean: 0.45 mg/kg.dw) for DEP and 0.32-3,514 mg/kg.dw (mean: 60.3  
27 mg/kg.dw) for DEHP. It is interesting to note that certain phthalates display only a  
28 few outliers to the high side. For DnBP and BBP, the outlier contents typically  
29 exceed 260 and 35 mg/kg.dw, respectively, i.e., 490 and 150 times the average  
30 level (0.53 and 0.23 mg/kg.dw, respectively). The little availability of phthalate  
31 removal efficiencies in biosolids suggested that heat drying and anaerobic  
32 digestion were less effective at reducing phthalates, particularly DEHP, than  
33 composting. As a complement, both studies conducted by Gibson et al. (2005) and  
34 Williams (2007) reported that composting removes between 64 and 70% of  
35 DEHP, respectively. Based on our database, PAE contents are statistically close  
36 regardless of the location considered, especially between European and Chinese  
37 samples, and moreover DEHP contents have been decreasing in both  
38 anaerobically-digested sludge and final sludge.  
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## 47 **Stormwater**

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50 Although it has been established that stormwater is responsible for the spread of  
51 pollutants, especially in urban areas, data concerning the emission, occurrence and  
52 fate of PAE in stormwater remains poorly reported (Björklund et al. 2009; Rule et  
53 al. 2006; Makepeace et al. 1995; Clara et al. 2010; Pitt et al. 1999; Zgheib et al.  
54 2012). According to these authors, phthalates in stormwater originated from  
55 plasticizers in PVC, paints, building materials, etc. For instance, wastewater from  
56 car washes can also make a significant contribution to the emission of DEHP  
57 (Vikelseo et al. 1998). Interestingly, PAEs are not chemically bound to the  
58 material and may migrate from products during use and disposal (Cadogan et al.  
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1993). DEHP has been measured at concentrations between 0.45 and 24.0  $\mu\text{g}\cdot\text{l}^{-1}$  in urban stormwater in Austria (Clara et al. 2010). DEHP was also measured in all urban and suburban samples between 3 and 58  $\mu\text{g}\cdot\text{l}^{-1}$  (Zgheib et al. 2012). Such levels were higher to those previously reported for stormwater in Sweden (5  $\mu\text{g}\cdot\text{l}^{-1}$ , (Björklund et al. 2009)) and in London (0.75 to 1.25  $\mu\text{g}\cdot\text{l}^{-1}$ , (Rule et al. 2006)). Finally, screening performed in the 1990s revealed higher concentrations of phthalates in stormwater compared to results from studies performed in 2000s, from less than ten times higher for DnOP and DEHP (Makepeace et al. 1995) up to 140 times higher for BBP (Pitt et al. 1999; Björklund et al. 2009). The higher concentrations measured in previous studies may be explained by the difference between phthalate uses a decade ago and the current situation.

## NATURAL ENVIRONMENT

### Surface water

Surface water is commonly considered as the natural compartment most affected by human pressures, since this water is subjected to the discharges of treated and/or untreated wastewater and/or stormwater (Lin et al. 2009). Therefore, throughout the world, the occurrence and fate of PAE in surface water have been well documented, as demonstrated by the 28 references pertaining to surface water contamination (see Table 8). First, close attention must be paid when comparing concentrations reported in literature, since industrialized and urbanized watershed have been monitored (Vitali et al. 1997; Tan 1995; Yuan et al. 2002; Long et al. 1998; Sha et al. 2007; Zhu and Qiu 2011; Dargnat 2008). Globally and at the world scale, PAE contamination in surface water varies from few  $\mu\text{g}\cdot\text{l}^{-1}$  to several tens of  $\mu\text{g}\cdot\text{l}^{-1}$ . The variations in PAE concentrations in surface water can be examined using published data, which provide sufficient details for determining the statistical distribution of concentrations. Figure 3 summarizes data for three geographical areas, namely Europe, North America and China. The median DEHP concentrations for Europe and China are very similar, i.e. 1.05 and 1.11  $\mu\text{g}\cdot\text{l}^{-1}$ , respectively; the median concentration in the North American samples is relatively lower (0.27  $\mu\text{g}\cdot\text{l}^{-1}$ ). Moreover, median PAE concentrations in the North American samples (0.29  $\mu\text{g}\cdot\text{l}^{-1}$ ) are notably lower than the Chinese (1.24  $\mu\text{g}\cdot\text{l}^{-1}$ ) and European samples (1.18  $\mu\text{g}\cdot\text{l}^{-1}$ ). It is also interesting to note that all geographical areas displayed a few high outliers. In United Kingdom (Long et al. 1998), these outlier concentrations typically exceeded 15  $\mu\text{g}\cdot\text{l}^{-1}$  (i.e., 12 times the average level). In addition, in China (Sha et al. 2007), they were above 28  $\mu\text{g}\cdot\text{l}^{-1}$  (i.e., 20 times the average level).

It is obvious from the database that PAE concentrations in European surface water first increased before exhibiting a decrease due to tighter regulations (EU 2005, 2004, 2007). It would be appropriate to treat these datasets as two separate time series (Fig. 6). The first series contains the samples from European countries before adoption of the Water Framework Directive (Thüren 1986; Ernst et al. 1988; Fatoki and Vernon 1990; Law et al. 1991; Vitali et al. 1997; Long et al. 1998; Belfroid et al. 1999), all of them exhibit PAE concentrations from 0.04 to 15.8  $\mu\text{g}\cdot\text{l}^{-1}$ . The second series is composed of samples from European countries after the WFD implementation (Penalver et al. 2001; Fromme et al. 2002; Vethaak

1 et al. 2005; Bendz et al. 2005; Prieto et al. 2007; Sanchez-Avila et al. 2012),  
2 revealing lower concentrations, i.e. of about  $1.16 \mu\text{g.l}^{-1}$  for the sum of PAE. The  
3 PAE concentrations plotted as a function of time for these two series are shown in  
4 Figure 6. In contrast, concentrations measured in the Chinese surface water have  
5 been increasing for the last decade (see Table 8). This is the result of the constant  
6 increase of industrialization, consumptions of phthalates and absence of regulation  
7 in China and more generally in developing countries (Zhu and Qiu 2011; Zeng et  
8 al. 2008; Sha et al. 2007; Li et al. 2006; Chen et al. 2012).  
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10 Most studies focusing on surface water analyze only the dissolved phase, hence  
11 only a few results are available for suspended solids (Table 9; four references  
12 available). According to these four studies, levels of phthalates in suspended  
13 solids range from a few to 630 mg/kg.dw (Table 9). Such contamination levels  
14 may be explained by the high hydrophobicity of DEHP and DnBP (with respect to  
15 their Log  $K_{ow}$ ; see Table 1), which leads to sorption and accumulation of these  
16 compounds on suspended solids (Gounaris et al. 1993; Sha et al. 2007). For less  
17 hydrophobic compounds, contents are lower, i.e. about between 0.05 and 0.20  
18 mg/kg for all compounds, except DEHP (0.70-630 mg/kg). Finally, suspended  
19 solids may, however, play a key role in aquatic systems since, under low-flow  
20 conditions, particles can settle and contribute to sediment formation, thus yielding  
21 contaminant stocks (Fig. 1 and Fig. 5). This phenomenon has been demonstrated  
22 for some heavy metals, pesticides and PCBs by Zgheib et al. (2012).  
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## 27 Sediments

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31 Several sediment samples have been collected from rivers and lakes across  
32 Europe, North America and developing countries (particularly China and India)  
33 (Vitali et al. 1997; Fromme et al. 2002; Yuan et al. 2002; Vethaak et al. 2005; Sha  
34 et al. 2007; Zeng et al. 2008; Liu et al. 2010). The amount of DnBP and DEHP  
35 measured in these samples (see Table 10; 23 references) varied from 0.01 to 115  
36 mg/kg.dw (median, 0.44 and 1.90 mg/kg.dw, respectively). Generally speaking,  
37 DMP and DEP were not detected in sediments (Fig. 4), mainly due to their low  
38 hydrophobicity. In addition, a recent study led by Liu et al. (2010) reported that  
39 DEP is easily degraded in top material and cannot be eluted in deep sediments.  
40 Moreover, under anaerobic conditions, DnBP and BBP are easily degraded  
41 whereas DEP and DEHP were poorly removed (Yuan et al. 2002). From the  
42 exhaustive data in literature, it is clear that PAE are often present in sediment  
43 regardless of the location (Fig. 2). The variation in phthalate contents among  
44 sediments was examined using published data, which provided sufficient details  
45 for determining the statistical distribution of these concentrations. Huang et al.  
46 (2005) examined the evolution of DEHP sediment content along a river transects  
47 densely populated. They revealed the existence of dynamic processes occurring in  
48 surface water, such as sorption to settleable particles and atmospheric  
49 volatilization governing the fate of phthalates in the environment and could lead  
50 to the formation of sinks in sediments and atmospheric compartment.  
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56 A comparison between Tables 9 and 10 indicates that the PAE contents found in  
57 sediments are close to those found in suspended solids (Fig. 5). As previously  
58 mentioned and highlighted in Figure 1, sediments and suspended solids are linked  
59 through sedimentation during low-flow periods and through re-suspension during  
60 high-flow periods (Mitsunobu and Takahashi 2006). Presumably, the high  $K_{ow}$  of  
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1 certain phthalates explains their sorption to particles in water, which in turn settle  
2 to form sediment (Staples et al. 1997). Additionally, this phthalate accumulation  
3 in sediments is fed and promoted by anthropogenic suspended solids, such as  
4 particles released by municipal WWTP and untreated water (Srivastava et al.  
5 2009; Huang et al. 2005).

## 6 7 **Soils**

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10 Several measurements of phthalates in soils have been undertaken (see Table 11,  
11 12 references); these have mainly concerned agricultural and urban soils. DEHP  
12 and DnBP levels ranged, respectively, from 0.02 to 264 mg/kg.dw (median: 3.33  
13 mg/kg.dw) and 0.01 to 30.1 mg/kg.dw (median: 0.96 mg/kg.dw) for both types of  
14 soils. A study performed by Michael et al. (1984) highlighted that these two  
15 compounds could enter soils via irrigation and pesticide application. In addition,  
16 both studies conducted by Wang et al. (2003) and Dolgen et al. (2007) reported  
17 that sewage sludge application could also lead to soil contamination,  
18 phytotoxicity, and could cause the accumulation of phthalates in the food supply.  
19 Some studies have observed high biodegradation rates of DnBP and DEHP in  
20 soils (Juneson et al. 2001; DiGennaro et al. 2005). Biodegradation however is not  
21 the only pathway for eliminating DEHP and DnBP in soils. Other processes, such  
22 as hydrolysis and photolysis may affect DEHP and DnBP concentrations and  
23 occurrence rates. These processes however are recognized to be less important  
24 than biodegradation (Yan et al. 1995). A study led by Chang et al. (2004)  
25 explained that optimal PAE degradation is enhanced when DnBP and DEHP are  
26 present simultaneously. It may be due to the large carbon source and energy  
27 provided by both compounds. In addition, in specific physicochemical conditions,  
28 DEP biodegradation can lead to the formation of DMP and its monoester  
29 (Cartwright et al. 2000). Interestingly and according to the available literature on  
30 soils (Table 11), similar contents for agricultural and urban soils have been  
31 observed. This homogeneity is quite surprising but could be explained by a global  
32 contamination through atmospheric deposits (Fig. 1). Nonetheless, a direct  
33 comparison of soil contamination is rather difficult and subtle, since various  
34 processes may be occurring. The differences or homogeneity observed can in fact  
35 reflect different inputs, environmental factors, including oxygen availability and  
36 nutrient amounts in soil, and sources or either different dynamic pollutants in the  
37 soils (Semple et al. 2001; Namkoong et al. 2002).

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40 Compost particle sizes have been investigated by several studies (Amellal et al.  
41 2001; Delhomenie et al. 2002; Chang et al. 2009). Authors explained that the  
42 smaller the particle was, the higher the specific surface was and the lower the  
43 porosity was. This favored the microorganism settlement and therefore promoted  
44 microbial degradation activity. Finally, the use of straw compost in remediating  
45 contaminated soil by phthalates is therefore effective to solid wastes recycling.  
46 The variation in PAE contents among soils was examined using published data,  
47 which provided sufficient details for determining the statistical distribution of  
48 these contents. Figure 3 shows the data from Europe and China. Soils in China  
49 (3.09 mg/kg.dw) seem to be slightly more contaminated than European soils (0.17  
50 mg/kg.dw). This difference may be linked to several factors: i) greater use or  
51 consumption of phthalate-containing goods in China; ii) a higher contamination of  
52 air in China, and iii) an absence of pertinent regulations on sludge amendment.  
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## DISCUSSION AND RECOMMENDATIONS FOR FURTHER STUDY

It is now obvious from the concentrations and contents measured in the different environmental compartments that phthalates are ubiquitous environmental contaminants. Their concentrations and trends in most environmental compartments however remain location-dependent. This review has highlighted some geographical disparities, especially between Europe, North America and China. In Europe, historical trends were also highlighted. In pursuit of the WFD implementation, more specific data for most congeners are needed in upstream compartments to enable source elucidation and to elaborate potential source control action. It is obviously important to measure all industrially-significant phthalates across all samples. It is also important to monitor chemicals used to replace phthalates after the implementations of restrictive regulations worldwide, such as Di-iso-nonyl phthalate (DiNP) and Di-iso-decyl phthalate (DiDP). Finally, phthalate metabolites have to be investigated, which are supposed to be the active molecule of the estrogenic effect (Rais-Bahrami et al. 2004).

As mentioned above, DEHP is always present in all environmental compartments. It is moreover the abundant phthalate in all matrices, particularly in solid matrices, such as sludge and suspended solids (Fig. 4). The result is that what is left over from many industrial and commercial products of this compound leads to the contamination of the wastewater of urban areas.

In Europe, regulations banning products containing certain phthalates (DEHP, DnBP and BBP) have been implemented since 2000. It is therefore relevant to evaluate the effectiveness of these regulations by means of reliable and accurate measurements of the PAE contamination levels for sediments, soils and surface water as a function of time. Indeed, each day without consistent restrictions means that phthalates are produced in huge quantities and it can subsequently result on a widespread use in a lot of products leading to uncontrolled environmental discharges and dissemination. Recent observations in Europe have however shown that PAE concentrations have not been increasing. Moreover, a decreasing trend has actually been observed just over the last few years. Nevertheless, it is important to track these changes closely. As previously noted however, sediments and atmosphere behave like major sinks for PAE in the environment. The role of atmosphere has to be better understood since atmosphere seems to play a key role in PAE widespread. Recent measurements in China, whatever the media, have indicated increasing phthalate contents and concentrations. In addition, the environmental data exhibit a large spatial variability, reflecting differences in phthalate contaminations in various areas of China, which might be caused by uneven development of heavy industry, the imbalance of enforcement or the specific geographical location. Finally, even if trends show decreases across industrialized countries, especially in Europe and North America, the levels being recovered are still significant (up to  $100 \mu\text{g}\cdot\text{l}^{-1}$ ) and still contribute to build the background level. A similar trend has been observed for alkylphenols (Bergé et al. 2012a). In addition, and based on the data collected, there are a few samples with very high levels as compared to the average (see the high outliers in all environmental media). A closer attention needs to be paid to these data – why are

1 these few samples so highly contaminated? Is it related to industrial activities or  
2 to unknown diffuse sources? Understanding these outliers may contribute to  
3 elucidate the mechanisms by which these samples have become contaminated.

4 The mechanisms involving these contaminants between the environmental  
5 compartments cannot be easily distinguished; possibilities include:  
6 biodegradation, volatilization, sorption to biomass, and particle sedimentation.  
7 The three latter mechanisms may be important in explaining the distribution  
8 changes throughout the whole water cycle from wastewater to the natural  
9 environment. This change is featured by an increase of DnBP and a decrease of  
10 DEHP and DEP (Fig. 4). Research on these mechanisms, particularly the  
11 processes acting in WWTP, is required in order to improve better understanding  
12 and to quantify what proportion is being volatilized into the atmospheric  
13 compartment and how much is adsorbed to biomass. The latest research on  
14 WWTP has shown high efficiency of the processes acting on sewage treatment  
15 plants as well as an insignificant contribution from discharges (Dagnat et al.  
16 2009; Bergé et al. 2012b; Martin-Ruel et al. 2010). Additionally, latest research  
17 showed that DEHP removal was dependent on various parameters, but that the  
18 type of biomass could significantly enhance removal (Oliver et al. 2007, 2005).  
19 The removal of phthalates from wastewater can, however, be improved by adding  
20 tertiary treatment such as nanofiltration, reverse osmosis, ozone oxidation, UV  
21 irradiation or activated carbon filters to existing processes, in spite of the high cost  
22 of these processes (Bodzek et al. 2004; Agenson et al. 2003; Verliefde et al.  
23 2007; Oh et al. 2006).

24 A serious element dictating the fate of micro-pollutants in urban areas is their  
25 release into the environment during storms. During wet-weather periods, the  
26 operations of wastewater treatment plants are modified and may influence the  
27 quality of effluent discharged into the environment (Bergé et al. 2012a; Gilbert et  
28 al. 2011; Bergé et al. 2012b). Additionally, combined sewer overflow discharges  
29 can happen (Zgheib et al. 2012; Gasperi et al. 2008). During wet-weather events,  
30 the re-suspension of in-sewer deposits provides for additional pollution at the  
31 WWTP. This additional contamination can then increase the load of phthalates  
32 and disturb usual plant operations and, in return, efficiency. Finally, nowadays, no  
33 information is available to evaluate the runoff contribution to wastewater.

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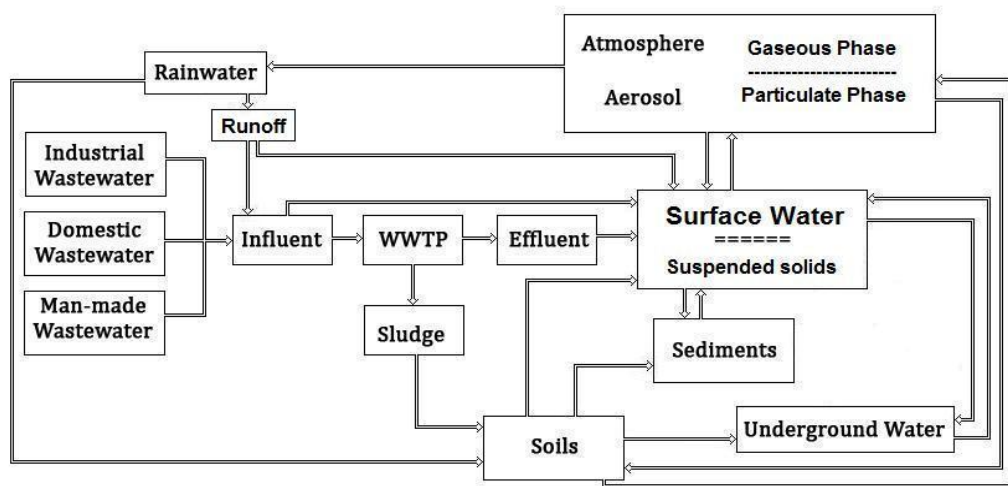


Figure 1: Interactions between the various environmental compartments – Cited from Bergé et al. (2012a)

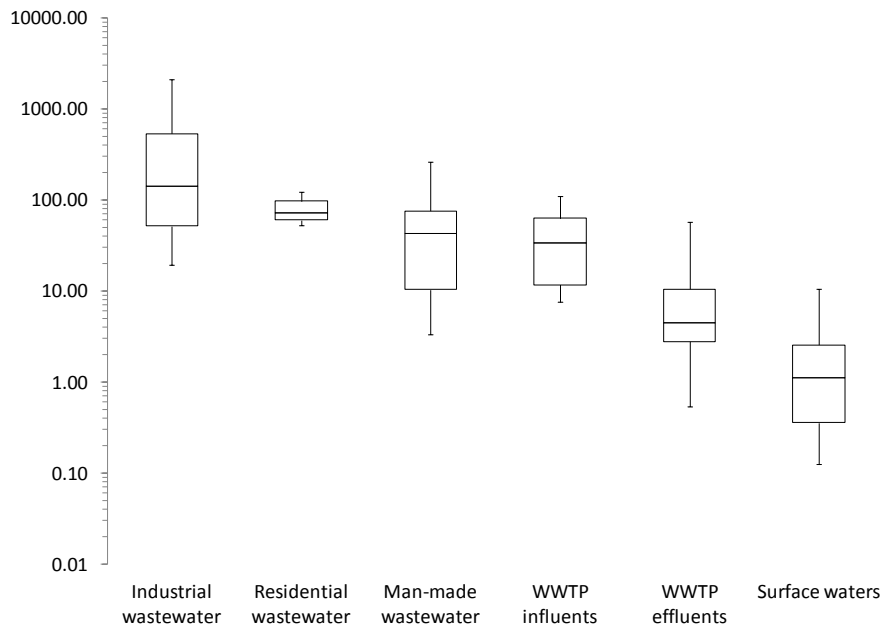


Figure 2: Total PAE (DMP + DEP + DnBP + BBP + DEHP + DnOP) concentrations (in  $\mu\text{g.l}^{-1}$ ) in liquid matrices (WWTP: Wastewater Treatment Plant - global scale)

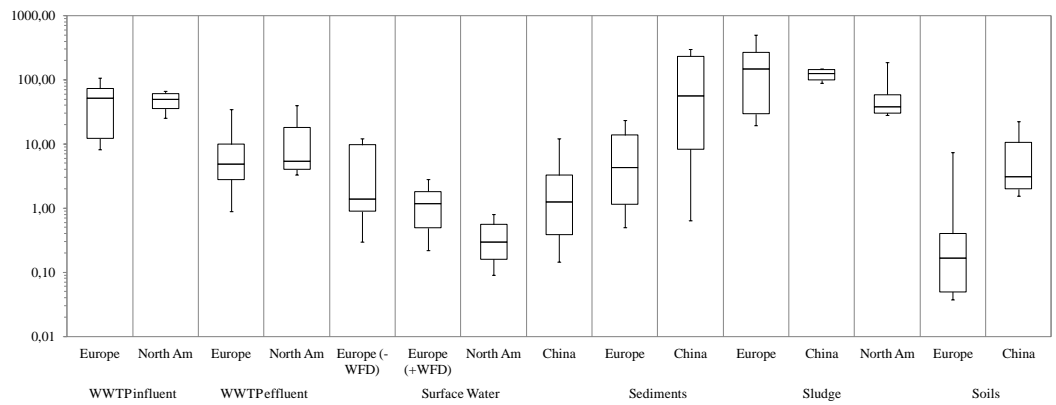


Figure 3: Distributions of total PAE (DMP + DEP + DnBP + BBP + DEHP + DnOP) concentrations ( $\mu\text{g.l}^{-1}$ ) and contents ( $\text{mg/kg.dw}$ ) in WWTP influent and effluent, surface water, sediments, sludge and soils from various locations (WFD: Water Framework Directive; North Am: North America)

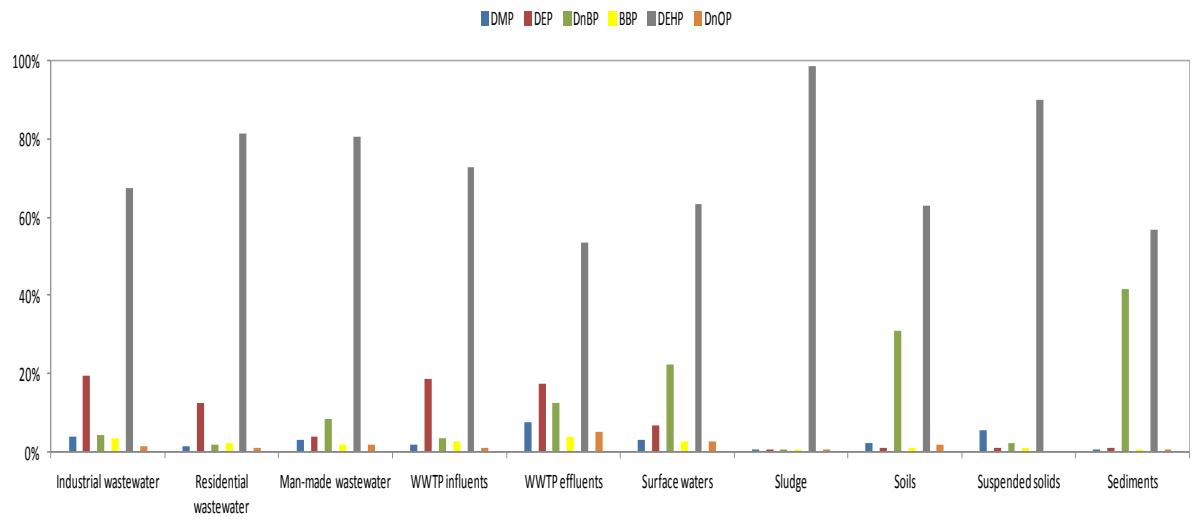


Figure 4: Evolution in the distribution of DMP, DEP, DnBP, BBP, DEHP and DnOP through the environmental cycle (WWTP: Wastewater Treatment Plant)

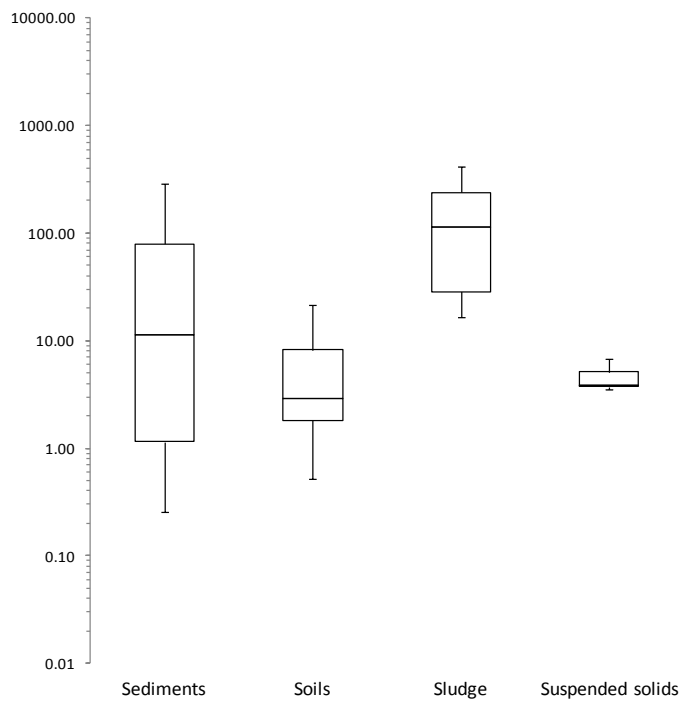


Figure 5: Total PAE (DMP + DEP + DnBP + BBP + DEHP + DnOP) contents (in mg/kg.dw) in solids matrices - global scale

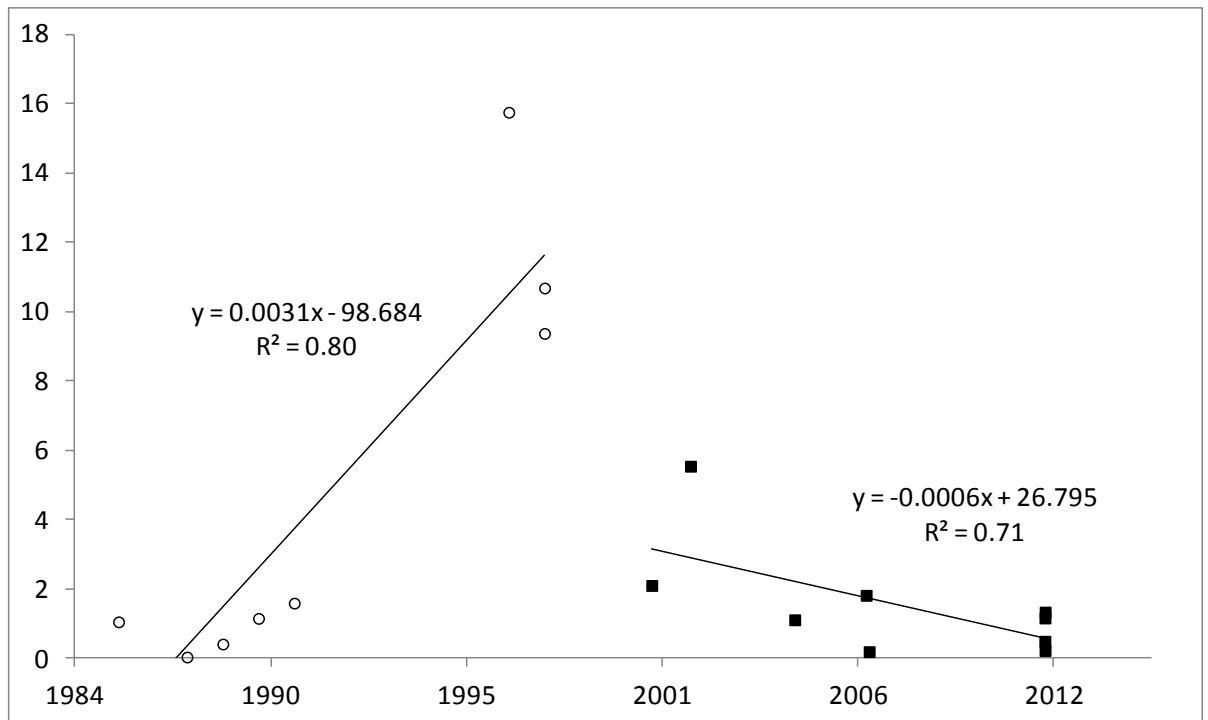


Figure 6: Historical trend lines for the median total PAE (DMP + DEP + DnBP + BBP + DEHP + DnOP) concentrations (in  $\mu\text{g.l}^{-1}$ ) in European surface water. The dots represent samples collected before implementation of the Water Framework Directive, whereas the squares depict samples collected after its implementation

Table 1: Physicochemical properties of PAE

Compound	Formula	MW (g/mol)	Water solubility at 20 °C (mg/l)	Log K <sub>ow</sub>	Log K <sub>oc</sub>	Log K <sub>d</sub>	H (Atm.m <sup>3</sup> /mol)
DMP	C <sub>10</sub> H <sub>10</sub> O <sub>4</sub>	194.2 <sup>c</sup>	4,20 <sup>c</sup>	1.61 <sup>c</sup>	1.74 <sup>c</sup> 1.90- 2.56 <sup>b</sup>		1.22 E <sup>-07c</sup>
DEP	C <sub>12</sub> H <sub>14</sub> O <sub>4</sub>	222.2 <sup>c</sup>	1,10 <sup>c</sup>	2.42 <sup>c</sup> 2.47- 2.51 <sup>b</sup>	1.84 <sup>c</sup> 2.85- 3.24 <sup>b</sup>		2.66 E <sup>-07c</sup> 7.80 E <sup>-07b</sup>
DnBP	C <sub>16</sub> H <sub>22</sub> O <sub>4</sub>	278.4 <sup>c</sup>	11.2 <sup>c</sup>	4.57 <sup>c</sup>	3.14 <sup>c</sup> 4.17 <sup>b</sup>		8.83 E <sup>-07c</sup>
BBP	C <sub>19</sub> H <sub>20</sub> O <sub>4</sub>	312.4 <sup>c</sup>	2.70 <sup>c</sup>	4.84 <sup>c</sup>	4.23 <sup>c</sup> 3.95 <sup>b</sup>	3.55 <sup>c</sup>	7.61 E <sup>-07c</sup>
DEHP	C <sub>24</sub> H <sub>38</sub> O <sub>4</sub>	390.6 <sup>c</sup>	0.003 <sup>c</sup>	7.50 <sup>c</sup>		4.12 <sup>c</sup> 4.18 <sup>a</sup>	1.71 E <sup>-05c</sup>
DnOP	C <sub>24</sub> H <sub>38</sub> O <sub>4</sub>	390.6 <sup>c</sup>	0.0005 <sup>c</sup>	8.06 <sup>c</sup>	4.94 <sup>c</sup> 5.68 <sup>b</sup> 5.71 <sup>a</sup>	4.46 <sup>c</sup>	1.03 E <sup>-04c</sup>
a: (Lützhof et al. 2008); b: (IPCS 2003); c: (Staples et al. 1997)							



Table 2: Atmospheric contamination by PAE (in ng/m<sup>3</sup>)

Location	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
USA			0.60-5.00				(Atlas and Giam 1988)
Sweden			0.23-49.9		0.28-77.7		(Thüren and Larsson 1990)
China		56.0	32.0	1,910	550		(Wang et al. 2002)
China			224		56.0		(Wang et al. 2002)
USA					5.00-132		(Clark et al. 2003)
Denmark			1.50-2,480		5.30-3,640		(Müller et al. 2003)
France	0.10-21.2	1.70-24.6	2.90-59.3	0.50-12.2	3.40-25.7	<log-1.10	(Teil et al. 2006)
Netherlands			2.00-70.0		0.70-333		(Peijnenburg and Struijs 2006)
Norwegian Sea	0.01-0.22	0.18-0.90	0.16-0.43	0.02-0.07	0.08-0.46		(Xie et al. 2007)
France	1.11	3.53	1.09	0.21	1.66	0.08	(Tlili et al. 2010)
Greece			1.20-3.36	0.11-0.80	<log-6.50	<log-0.11	(Salapavidou et al. 2011)
Greece			0.43-2.40	0.04-0.98	4.63-45.0		(Salapavidou et al. 2011)
Min	0.01	0.18	0.16	0.02	0.08	0.08	n = 10
Max	21.2	56.0	2,480	1,910	3,640	1.10	
Med	2.93	8.70	2.50	0.48	19.7	0.24	

Table 3: Rain water concentrations of PAE (in  $\mu\text{g.l}^{-1}$ )

Location	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
USA		0.02-0.10	0.03-0.06	0.02-0.07	0.02-0.10		(Ligocki et al. 1985)
Sweden			<log-0.50		0.01-0.43		(Thüren and Larsson 1990)
Canada			0.50-11.0		7.00-39.0		(Makepeace et al. 1995)
Denmark			1.30		32.0		(Kjohlholt et al. 1997)
Denmark			<log-1.60		1.30-30.0		(Boutrup and Plesner 2001)
Netherlands	0.01-0.02	0.24-0.43	0.28-0.88	0.14-0.26	0.69-1.70	0.04-0.25	(Vethaak et al. 2005)
France	0.12	0.33	0.59	0.08	0.42	0.01	(Teil et al. 2006)
France	0.07-0.11	0.14-0.25	0.10-0.16	0.02-0.06	0.36-0.85	<log-0.03	(Dargnat 2008)
Norway					5.00		(Björklund et al. 2009)
Min	0.01	0.02	0.03	0.02	0.02	0.01	n = 9
Max	0.12	0.43	11.0	0.26	39.0	0.25	
Med	0.06	0.33	0.61	0.16	0.77	0.05	

Table 4: Industrial, man-made and residential wastewater concentrations (in  $\mu\text{g.l}^{-1}$ )

Location	Type	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
USA	Textile		3.20					(IPCS 2003)
France	Textile					282		(INERIS 2007)
France	Laundry					470		(INERIS 2007)
USA	Diaper service		<loq-1.00	12.0	0.20	0.63	<loq-0.15	(Jackson and Sutton 2008)
USA	Pet wash		1.30	0.76	2.30	6.50	1.60	(Jackson and Sutton 2008)
USA	Veterinary clinic		<loq-51.0	<loq-18.0	<loq-7.10	<loq-11.0	<loq-7.70	(Jackson and Sutton 2008)
USA	Hospital		<loq-1.00	<loq-0.36	0.82	2.70	<loq-0.15	(Jackson and Sutton 2008)
USA	Medical clinic		<loq-0.98	0.66	0.74	1.00	<loq-0.15	(Jackson and Sutton 2008)
Spain	Industrial-Residential	0.28-4.94	27.5-192	<loq-33.6	<loq-5.70	9.88-287		(Sanchez-Avila et al. 2009)
Netherlands	Residential	0.39-6.20	4.10-44.0	0.38-51.0	0.56-4.90	13.0-100	0.26-2.40	(Vethaak et al. 2005)
USA	Residential		4.00	<loq-0.36	0.76	9.10	0.60	(Jackson and Sutton 2008)
USA	Residential		9.10	<loq-0.34	1.00	3.30	<loq-0.14	(Jackson and Sutton 2008)
Spain	Residential	0.45-1.12	44.0-45.9		0.23-0.29	19.5-20.2		(Sanchez-Avila et al. 2009)
France	Sewer network		5.23-17.7	0.60-3.31	0.46-3.91	39.48-161		(Bergé et al. 2012c)
USA	Industrial activities					0.01-4,400		(Clark et al. 2003)
USA	Manufactured factory		60.0					(IPCS 2003)
Netherlands	Industrial effluents	<loq-1.30	0.35-5.20	0.69-21.0	0.17-1.30	1.00-150	0.01-2.80	(Vethaak et al. 2005)
France	Paper pulp					61.0		(INERIS 2007)
France	Dyeing					41.0		(INERIS 2007)
France	Leather					68.0		(INERIS 2007)
France	Painting					1,300		(INERIS 2007)
France	Chemistry					280		(INERIS 2007)
USA	Plastic manufacturer		<loq-1.00	0.36	2.30	49.0	10.0	(Jackson and Sutton 2008)
USA	Paper manufacturer		<loq-1.00	<loq-0.36	0.14	6.80	<loq-0.15	(Jackson and Sutton 2008)
USA	Beverage manufacturer		<loq-20.0	<loq-7.1	<loq-2.70	<loq-4.10	<loq-2.90	(Jackson and Sutton 2008)
USA	Adhesive manufacturer		<loq-100	120	39.0	47.0	<loq-15.0	(Jackson and Sutton 2008)
USA	Industrial laundry		<loq-24.0	86.0	95.0	2,700		(Jackson and Sutton 2008)
USA	Residential laundry		16.0	<loq-3.6	<loq-1.40	66.0	13.0	(Jackson and Sutton 2008)
Spain	Industrial effluents	0.15-7.05	22.2-53.6		<loq-10.0	8.00-36.4		(Sanchez-Avila et al. 2009)
Man-made wastewater	Min	0.28	1.30	0.76	0.20	0.63	0.15	n = 4
	Max	4.94	192	33.6	5.70	470	7.70	
	Med	2.61	3.20	6.90	1.50	66.0	1.60	
Residential wastewater	Min	1.00	0.35	0.34	0.23	3.30	0.07	n = 4
	Max	1.12	45.0	51.0	4.90	161	2.40	
	Med	1.06	9.48	1.29	1.57	61.3	0.60	
Industrial	Min	0.15	0.35	0.36	0.14	0.01	0.01	n = 5
	Max	7.05	100	120	95.0	4,400	15.0	

wastewtaer	Med	1.96	10.0	2.20	1.83	34.6	0.80	
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Table 5: WWTP influent concentrations of PAE (in  $\mu\text{g.l}^{-1}$ )

Location	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
Finland	<log-1.00	6.00-11.0	6.00-8.00	3.00-5.00	28.0-66.0	<log-1.00	(Marttinen et al. 2003)
Finland	<log-1.00	5.00-74.0	3.00-9.00	<log-3.00	28.0-122	<log-2.00	(Marttinen et al. 2003)
Finland		<log-10.0	4.00-6.00	<log-2.00	83.0	<log-1.00	(Marttinen et al. 2003)
Denmark			<log-1.03	0.05-0.97	13.1-44.3	0.22-0.79	(Fauser et al. 2003)
Sweden		0.19	0.15		0.27		(Bendz et al. 2005)
Australia		0.39	0.14	0.06	20.3		(Tan et al. 2007)
France					55.0		(INERIS 2007)
Spain		0.68	0.62		1.90		(Regueiro et al. 2008)
USA		<log-10.0	<log-3.60	14.0	33.0	4.20	(Jackson and Sutton 2008)
USA		<log-10.0	<log-3.60	1.90	9.20	<log-1.5	(Jackson and Sutton 2008)
Denmark					8.10-31.0		(Seriki et al. 2008)
Canada					70.0		(Barnabé et al. 2008)
Canada					41.0		(Barnabé et al. 2008)
France	0.82	7.71	1.10	1.12	22.5	0.10	(Dargnat et al. 2009)
Spain	0.60	50.7	46.8	0.67	47.9		(Sanchez-Avila et al. 2009)
Austria	<log-2.40	0.77-9.20	<log-8.70	0.31-3.20	3.40-34.0	<log-1.10	(Clara et al. 2010)
Austria	0.26-0.41	1.20-2.00	<log-0.47	<log-0.11	4.40-8.80		(Clara et al. 2010)
Austria	0.43-0.81	2.20-2.70	0.15-0.41	0.11-0.26	4.10-13.0	<log-0.10	(Clara et al. 2010)
France					52.8		(Martin-Ruel et al. 2010)
Spain		1.05-2.59	0.20-0.55		4.23-4.65		(Reyes-Contreras et al. 2011)
Spain		1.90-3.98	0.12-0.20		7.50-9.91		(Reyes-Contreras et al. 2011)
Spain	<log-7.47	2.11-5.76	0.23-1.99	0.25-0.31		0.24-5.91	(Bizkarguenaga et al. 2012)
Spain	0.31-9.13	0.14-5.91	<log-0.76			0.27	(Bizkarguenaga et al. 2012)
Spain	0.36-93.3	0.12-4.34	0.59-2.24			0.22-0.25	(Bizkarguenaga et al. 2012)
Spain	3.94-10.6	<log-0.96	0.23-1.54			<log-3.54	(Bizkarguenaga et al. 2012)
France		7.00-36.0	1.86-6.01	0.97-2.29	32.4-71.9		(Bergé et al. 2012b)
Min	0.26	0.19	0.14	0.06	0.27	0.10	n = 16
Max	93.3	74.0	46.8	14.0	122	5.91	
Med	0.89	9.81	1.86	1.29	38.10	0.50	

Table 6: WWTP effluent concentrations of PAE (in  $\mu\text{g.l}^{-1}$ )

Location	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
Finland			2.00		2.00	<loq-1.00	(Martinen et al. 2003)
Finland	<loq-1.00	<loq-4.00	<loq-6.00	<loq-1.00	2.00-8.00		(Martinen et al. 2003)
Finland					4.00		(Martinen et al. 2003)
Denmark			0.18-2.50	0.06-0.27	0.11-2.65	0.01-0.03	(Fauser et al. 2003)
Netherlands	<loq-0.32	0.30-0.93	0.42-0.84	0.07-0.29	0.47-2.40	<loq-0.02	(Vethaak et al. 2005)
Sweden		0.02	0.03		0.02		(Bendz et al. 2005)
Norway		7.90-9.90			0.50		(Vogelsang et al. 2006)
Australia		0.02	0.05	0.02	0.36		(Tan et al. 2007)
France		0.43			0.55		(INERIS 2007)
Spain		<loq-1.0	<loq-0.36	0.84	2.90		(Regueiro et al. 2008)
USA		<loq-1.0	0.57	0.74	1.00		(Jackson and Sutton 2008)
USA		<loq-1.0	5.50	<loq-0.14	0.21		(Jackson and Sutton 2008)
Denmark					0.30-6.10		(Seriki et al. 2008)
Canada					54.0		(Barnabé et al. 2008)
France	0.08	0.78	0.15	0.30	5.42	0.02	(Dargnat et al. 2009)
Spain	0.13	49.8		0.01	9.43		(Sanchez-Avila et al. 2009)
Austria	<loq-0.19	<loq-1.10	<loq-2.40	0.09-1.40	0.08-6.60	<loq-0.26	(Clara et al. 2010)
Austria		<loq-0.10			<loq-0.28		(Clara et al. 2010)
Austria					<loq-1.30		(Clara et al. 2010)
France					4.20		(Martin-Ruel et al. 2010)
Spain	<loq-7.18	0.23-0.40	0.22-7.92		0.39-0.41	0.21-30.8	(Bizkarguenaga et al. 2012)
Spain	6.25-11.7	0.20-1.02	<loq-10.9			0.22-0.27	(Bizkarguenaga et al. 2012)
Spain	2.07-11.2	0.36-1.06	0.21-1.34		0.31	0.23-0.29	(Bizkarguenaga et al. 2012)
Spain	7.97-13.4	0.15-0.51	0.20-1.90	1.14		3.26-9.06	(Bizkarguenaga et al. 2012)
Austria					<loq-6.60		(Clara et al. 2012)
Spain		15.7-40.9	3.33-58.9		4.17-69.0		(de los Rios et al. 2012)
Spain		0.03-3.21			1.72-9.22		(Sanchez-Avila et al. 2012)
France		0.46-6.77	0.01-0.93	0.01-0.21	0.95-6.43		(Bergé et al. 2012b)
Min	0.08	0.02	0.01	0.01	0.02	0.01	n = 20
Max	13.4	49.8	58.9	1.40	69.0	30.8	
Med	0.34	0.80	0.57	0.18	2.44	0.24	

Table 7: PAE in WWTP sludge contents (in mg/kg.dw)

Location	Type	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
Canada	Final					3.00-176		(Webber and Lesage 1989)
Canada	Final					1.60-245		(Webber and Nichols 1995)
Germany	Final			9.70		8.00		(Petrovic and Barcelo 2000)
Austria	Final			<loq-0.69		<loq-47.0		(Gangl et al. 2001)
China	Final	0.02-2.00	0.01-11.0	0.04-3.70	0.02-35.0	<loq-108	<loq-6.60	(Cai et al. 2007)
Norway	Final					27.0-115		(Jaganyi 2007)
Sweden	Final					25.0-661		(Jaganyi 2007)
Denmark	Final					3.90-170		(Jaganyi 2007)
Denmark	Final					3.46-40.6		(Seriki et al. 2008)
Australia	Primary					134		(Tan et al. 2007)
Austria	Primary	0.07-0.09	0.04-0.09	0.27-0.85	0.14-0.38	20.0-27.0	0.13-0.26	(Clara et al. 2010)
USA	Secondary					3.46-31.7		(Kinney et al. 2006)
Sweden	Digested					25.0-661		(Sweetman 1994)
Finland	Digested					163		(Martinen et al. 2003)
USA	Digested					3.33		(Kinney et al. 2008)
USA	Amended					163-578		(Staples et al. 1997)
Denmark	Amended			0.02-260		3.00-170		(Torslov et al. 1997)
Germany	Amended					170		(Schnaak et al. 1997)
Germany	Amended					1.50-5.10		(Schaecke and Kape 2003)
UK	Amended	0.30	0.02	0.39	0.20	0.32-0.55	0.57	(Gibson et al. 2005)
Spain	Amended					2.00-3,514		(Abad et al. 2005)
Denmark	Amended		1.52	1.30	1.17		1.24	(Laturus and Gron 2007)
Denmark	Amended		0.91	0.56				(Laturus and Gron 2007)
Spain	Amended					13.0-345		(Aparicio et al. 2009)
Norway	Compost					1.00-140		(Paulsrud et al. 2000)
Taiwan	Aerobically					143		(Cheng et al. 2001)
Taiwan	Anaerobically					105-153		(Cheng et al. 2001)
Germany	Spin-dried			0.20-1.70		28.0-154		(Fromme et al. 2002)
France	Spin-dried					72.1		(Dargnat et al. 2009)
Austria	Excess	<loq-0.06	0.06-0.13	0.64-1.20	0.12-0.25	22.0-29.0	0.06-0.12	(Clara et al. 2010)
Canada	Press-filtered					80.0-90.0		(Barnabé et al. 2008)
USA						<loq-58.3		(Clark et al. 2003)
Germany						0.90-110		(Fragermann 2003)
USA						<loq-310		(EPA 2009)
	Min	0.02	0.01	0.04	0.02	0.32	0.08	n = 29
	Max	2.00	11.0	260	35.0	3,514	6.60	
	Med	0.19	0.45	0.53	0.23	60.3	0.20	

Table 8: Surface water concentrations of PAE (in  $\mu\text{g}\cdot\text{l}^{-1}$ )

Location	Type	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
USA	River					0.50-1.00		(Sheldon and Hites 1979)
USA	Estuary					0.21-0.77		(Ray et al. 1983)
USA	River					0.01-0.70		(DeLeon et al. 1986)
Sweden	River					0.30-1.80		(Thüren 1986)
Germany	River					0.03-0.04		(Ernst et al. 1988)
UK	River					0.40-1.90		(Fatoki and Vernon 1990)
UK	Bay					0.98-2.20		(Law et al. 1991)
Malaysia	Estuary					3.10-64.3		(Tan 1995)
Italy	River					0.30-31.2		(Vitali et al. 1997)
UK	River					0.74-18.0		(Long et al. 1998)
Netherlands	Estuary				<loq-0.50	0.04-1.90		(Belfroid et al. 1999)
Spain	River		0.60	0.40		1.10		(Penalver et al. 2001)
Spain	Fishing port	1.60	1.40	1.30	0.50	2.10		(Penalver et al. 2001)
Spain	Industrial port	2.10	1.80	1.90	1.10	3.20		(Penalver et al. 2001)
Japan	Bay	0.01-0.09	<loq-0.31	0.01-0.54	<loq-0.06			(Suzuki et al. 2001)
Taiwan	Bay		<loq-2.50	1.00-13.5		<loq-18.5		(Yuan et al. 2002)
Germany	River			0.50		0.08-10.0		(Fromme et al. 2002)
Netherlands	River				0.01-1.00			(Vethaak et al. 2002)
Netherlands	Sea				0.01-1.80			(Vethaak et al. 2002)
China	River			3.39-12.6		<loq-1.90		(Zhang and Chen 2003)
Netherlands	River	0.01-0.19	0.07-2.30	0.07-3.10	0.01-1.80	0.90-5.00	<loq-0.08	(Vethaak et al. 2005)
Sweden	River		0.01-0.03	0.02-0.06		0.01-0.04		(Bendz et al. 2005)
China	River	<loq-1.40	0.26-1.28	0.05-3.91			0.10-0.80	(Li et al. 2006)
Spain	Estuaries	0.01-0.18	0.31-1.31	0.25-0.58	0.01-0.03	0.32-0.46	0.08-0.09	(Prieto et al. 2007)
Spain	Sea	0.01	0.03	0.08	0.01	0.06		(Prieto et al. 2007)
China	River	0.10-0.25	0.16-0.44	4.28-21.0		0.34-24.0	<loq-0.79	(Sha et al. 2007)
China	Tributaries	<loq-0.58	0.01-1.09	9.24-26.0		3.91-31.8	<loq-7.10	(Sha et al. 2007)
USA	River			0.14-4.14	0.04-0.35			(Solis et al. 2007)
USA	River			0.16-1.36	0.07-0.14			(Solis et al. 2007)
Spain	Estuary	0.21-0.28	0.07	1.25-1.26	0.05	0.22	0.03	(Prieto et al. 2008)
Spain	Sea	0.22-0.25	0.04	0.25-0.40	0.06	0.17	0.03	(Prieto et al. 2008)
China	River	<loq-0.09	0.02-0.32	0.94-3.60				(Zeng et al. 2008)
France	River		0.07-0.18	0.07-0.32		0.16-0.31		(Dargnat et al. 2009)
China	River					0.62-15.2	0.04-0.21	(Zhu and Qiu 2011)
Iran	River	0.87	0.67					(Hadjmohammadi et al. 2011)
Iran	Sea	0.49	0.52					(Hadjmohammadi et al. 2011)
Canada	Sea			0.18-3.00		0.01-0.95		(Keil et al. 2011)
USA	Sea					0.06-0.64		(Keil et al. 2011)
China	River			0.11-0.29		<loq-0.84		(He et al. 2011)
Spain	Sea	<loq-0.14	0.02-0.48		<loq-0.10	0.03-0.62		(Sanchez-Avila et al. 2012)



Spain	Port	<loq-0.01	0.02-0.87		<loq-0.80	0.06-5.97		(Sanchez-Avila et al. 2012)
Spain	River-sea		0.07-0.16		<loq-0.08	0.02-0.21		(Sanchez-Avila et al. 2012)
Spain	River		0.05-0.28		<loq-0.02	0.12-4.98		(Sanchez-Avila et al. 2012)
China	River	0.03-1.45	0.03-0.71	0.02-1.35	0.01-0.86	0.02-5.58	0.04-0.12	(Shi et al. 2012)
China	River	0.02-0.13	0.01-0.09	0.06-7.19	0.02-0.07	0.23-28.4	0.01-0.34	(Zhang et al. 2012)
Min		0.01	0.01	0.01	0.01	0.01	0.01	n = 33
Max		2.10	2.50	26.0	1.80	64.3	7.10	
Med		0.05	0.11	0.37	0.04	1.00	0.04	

Table 9: PAE contents in suspended solids (in mg/kg.dw)

Location	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
Netherlands	<loq-16.0	<loq-2.69	<loq-4.10	<loq-3.00	<loq-19.0		(Vethaak et al. 2005)
Netherlands					0.79-11.4		(Peijnenburg and Struijs 2006)
Netherlands					0.70-14.6		(Peijnenburg and Struijs 2006)
Netherlands					0.97-19.3		(Peijnenburg and Struijs 2006)
China	0.49-3.01	<loq-0.13	17.6-57.8		5.40-630		(Sha et al. 2007)
France					0.91-25.1		(Gasperi et al. 2008)
Min					0.70		n = 4
Max	16.0	2.69	57.8	3.00	630		
Med	0.22	0.04	0.09	0.02	4.23		

Table 10: PAE contents in sediments (in mg/kg.dw)

Location	Type	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
USA	River					0.04-16.0		(Ray et al. 1983)
UK	River					1.20		(Preston and Al-Omran 1989)
Malaysia	River			0.25		0.49-15.0		(Tan 1995)
Singapore	Bay					0.89-2.79		(Chee et al. 1996)
Italy	River			<loq-0.03		0.06-0.49		(Vitali et al. 1997)
UK	River					0.84-115		(Long et al. 1998)
Germany	River			0.45		0.21-8.44		(Fromme et al. 2002)
Taiwan	River		0.10-1.10	0.30-30.3	<loq-1.80	0.50-23.9		(Yuan et al. 2002)
China	River			0.02-0.05		0.03-0.05		(Zhang and Chen 2003)
Netherlands	River	<loq-2.50	0.07-1.20	0.03-1.00	<loq-0.06	0.10-7.60	<loq-0.55	(Vethaak et al. 2005)
Spain	River		<loq-0.24	0.02-0.79	0.50-0.95	10.1-16.8		(Cortazar et al. 2005)
France	River					0.55		(Quenea et al. 2005)
Taiwan	River					<loq-8.25		(Huang et al. 2005)
Japan	River					1.00-2.00		(Yuwatini et al. 2006)
Netherlands	River			0.09		4.30		(Peijnenburg and Struijs 2006)
China	River	0.14-0.42		18.1-34.1		9.29-50.7		(Sha et al. 2007)
China	Tributaries	<loq-1.04	<loq-0.01	3.63-72.2		5.35-259		(Sha et al. 2007)
China	Urban lake	0.04	0.13	0.28	0.03	1.30	0.02	(Zeng et al. 2008)
Taiwan	River		0.60	0.40		<loq-46.5		(Huang et al. 2008)
Taiwan	River			0.04-1.88				(Huang et al. 2008)
China	River	0.01-0.41	0.55-6.81	0.50-155		0.40-324	0.01-1.19	(Wang et al. 2008)
India	Urban River	<loq-0.02		<loq-0.02		<loq-0.02		(Srivastava et al. 2009)
India	Rural River	<loq-0.01	<loq-0.01	<loq-0.02		<loq-0.01		(Srivastava et al. 2009)
India	Urban River	<loq-0.05	<loq-0.04	<loq-0.04		<loq-0.32	<loq-0.05	(Srivastava et al. 2009)
Taiwan	River					0.10-20.2		(Lin et al. 2009)
France	River					0.91-26.6		(Gasperi et al. 2009)
China	River	0.02	0.18	0.10	0.02	0.39		(Liu et al. 2010)
China	River	0.03	0.26	0.04	0.01	0.22		(Liu et al. 2010)
	Min	0.01	0.01	0.02	0.01	0.01	0.02	n = 23
	Max	2.50	1.20	72.2	1.80	115	0.55	
	Med	0.02	0.20	7.45	0.02	10.2	0.04	

Table 11: PAE contents in soils (in mg/kg.dw)

Location	Type	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
USA	Urban					nd-1.20		(Russel and Mc Duffie 1986)
Denmark	Amended					0.03-0.04		(Vikelsøe et al. 2002)
China	Urban	<loq-0.20	<loq-2.61	<loq-1.66		0.20-7.11		(Hu et al. 2003)
China	Urban	<loq-0.02	<loq-0.05	0.34-1.66		0.22-0.74	0.09	(Ma et al. 2003)
UK	Rural			0.01		0.02-0.08	0.01	(Gibson et al. 2005)
UK	Amended			0.01		0.32-0.55		(Gibson et al. 2005)
China	Vegetable	<loq-0.07	<loq-1.77	<loq-20.6	<loq-1.48	2.82-25.1	<loq-0.92	(Cai et al. 2005)
Netherlands				0.01		0.03		(Peijnenburg and Struijs 2006)
China	Urban	<loq-0.07	<loq-0.25	0.28-3.82	<loq-0.06	0.17-6.49	<loq-0.17	(Li et al. 2006)
Denmark	Agricultural		0.01	0.07		0.18		(Laternus and Gron 2007)
Denmark	Compost		0.46	0.29		12.2	1.24	(Laternus and Gron 2007)
China	Agricultural			2.75-29.4		0.49-7.99		(Xu et al. 2008)
China	Agricultural	<loq-0.16	<loq-0.18	<loq-2.74	<loq-1.58	0.11-29.4	<loq-0.08	(Zeng et al. 2008)
China	Roadsides	0.02-0.35	0.01-0.20	0.29-30.1	<loq-1.58	1.41-264	<loq-2.31	(Zeng et al. 2009)
China	Resident	0.01-0.13	0.01-0.10	0.21-7.49	<loq-0.16	1.40-97.2	<loq-0.08	(Zeng et al. 2009)
China	Parks	0.02-0.10	0.01-0.07	0.21-7.49	<loq-0.16	0.89-154	<loq-0.03	(Zeng et al. 2009)
China	Urban					0.04		(Zhou and Liu 2010)
	Min	0.01	0.01	0.01		0.02	0.01	n = 13
	Max	0.35	2.61	30.1	1.58	264	2.31	
	Med	0.08	0.09	0.96	0.05	3.33	0.09	