Meta-analysis of environmental contamination by phtalates

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Meta-analysis of environmental contamination by phthalates.

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Abstract

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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.

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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

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

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

Strategy of this review

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

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

ATMOSPHERE

Air contamination

PAE in the atmosphere or in urban areas are generated by various emission sources, including volatilization from materials, industrial processes, waste combustion and wastewater treatment processes (Salapasidou et al. 2011). It has been calculated that approximately 4% of the quantity of DEHP can evaporate from materials such as floorings, rain clothes, toys, soles of shoes (Ranke 2005). DEHP was indeed detected in aerosols emitted from the aeration tank of a WWTP ranging from 71.1 to 228 ng/m^3 (Lepri et al. 2000). As shown in Table 2, a small number of air samples (10 references in all) has been reported for PAE. From data collected, PAE contamination globally lies in the 1-50 ng.m^{-3} range for all congeners, except for DEHP presenting higher levels (up to 3,640 ng.m^{-3}). The analysis of this database reveals that European samples were consistently above the values reported in the world (USA, China, etc.), and DEHP was the most abundant phthalate in air, with concentrations ranging from 0.08 to 3,640 ng/m^3 (Müller et al. 2003). Recent studies conducted by Tlili et al. (2010) and Teil et al. (2006), in the Parisian area (France), underscored that phthalates are preferentially associated with the gaseous phase rather than aerosols. For instance, Tlili et al. (2010) reported that between 60% to 70% against 40% to 30%, respectively, of phthalates are associated to gaseous phase. In addition, the same authors reported that the phthalates exhibiting short alkyl chains and high vapor pressures (especially, DMP and DEP) are predominant in the gaseous phase and inversely for the others compounds. Study conducted by Xie et al. (2005) showed that the air-sea exchange was preferentially dominated by the deposition, especially for DnBP (-60 to -686 ng/m²/d) and BBP (-4 to -28 ng/m²/d). Additionally to this

latter point, the fluxes calculated for DEHP (-95 to 686 ng/m²/d) highlighted a more complicated mechanism. Moreover, DnBP and DEHP total (dry and wet) depositions, to the Great Lakes (Canada), provided by Eisenreich et al. (1981) were estimated at 3.7 and 16 tons per year, respectively. This indicates that the atmosphere could be one of the major contamination sources of PAEs. However, according to Staples et al. (1997) and, more recently to Xie et al. (2007), phthalates are subject to photo-degradation, and therefore generally do not persist in the atmosphere. Authors reported photo-degradation half-lives of common phthalates ranging from 0.3 to 15 days. A study led by Salapasidou et al. (2011) underlined that concentrations of DEHP were significantly higher on urban-traffic areas (4.63 to 45.0 ng/m^3; median 19.4 ng/m^3) than on urban-industrial site (up to 6.50 ng/m^3; median 2.80 ng/m^3) implying generally an input from vehicular emissions. Similarly, Wang et al. (2008) reported that concentrations were about 3.5 higher above urban sites than above suburban sites. These authors attributed this difference to both many point-sources and environmental recycling. Additionally, Dargnat (2008) underlined the presence of seasonal variability above the Parisian area, with smaller concentrations for spring and during summer. This was previously observed by Guidotti et al. (2000) in Italy and Teil et al. (2006) in France above the same area. Due to the limited number of studies available, no distinct temporal or geographical trend could be drawn.

Rain water

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

URBAN AREAS

Sewer contamination

Industrial, man-made and residential wastewater

As reported in Table 4 and Figure 2, only a few measurements of PAE have been performed in wastewater (4-5 references for each type of wastewater). Industrial

DEHP levels were in the 0.01 to 4,400 µg.l^{-1} range (median: 34.6 µg.l^{-1}), i.e. twice as low as levels in residential wastewater (3.30-160 µg.l^{-1}; median: 61.3 µg.l^{-1}) and in man-made wastewater (0.60-470 µg.l^{-1}; median: 66.0 µg.l^{-1}). In Europe and North America, it has been reported that high DEHP and other phthalate concentrations were measured in industrial wastewater. In the USA, for instance, DEHP levels of 4,400 µg.l^{-1} were measured in untreated industrial wastewater (Clark et al. 2003). Similarly, Jackson and Sutton (2008) quoted DEHP, DEP and DnBP levels reaching 2,700, 100 and 120 µg.l^{-1} in industrial laundry and adhesive manufacturer wastewaters. Both studies led by Vethaak et al. (2005) and Sanchez-Avila et al. (2009) showed that levels between 45 and 100 µg.l^{-1} have also been measured in residential wastewater in the Netherlands and Spain. Some authors mentioned that the high concentrations measured in Maresme wastewater (Catalonia, Spain) and Parisian sewer network (France) would point out that PAE are still being produced and used in industrial, household and agricultural formulations despite the implementation of European regulations (EU 2005, 2004, 2007) restricting their use within the EU to levels < 0.1% (Sanchez-Avila et al. 2009; Bergé et al. 2012c).

Wastewater Treatment Plant Influent

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

Wastewater Treatment Plant Effluent

Understanding the biodegradation processes of phthalic compounds proves to be a critical factor in predicting the fate of these compounds in the environment. From an overall standpoint, the removal of PAE by conventional activated sludge WWTPs has been well documented (Dargnat et al. 2009; Tan et al. 2007; Barnabé et al. 2008; Marttinen et al. 2003). Most of studies reported removal up to 90%. Additionally, recent studies conducted by Bergé et al. (2012b) and Gasperi et al. (2010) explained that biofiltration coupled to physicochemical lamellar clarification could be a promising alternative to activated sludge tanks for today plant built in large urbanized areas where the building pressure makes available land scarce. A large number of samples from WWTP effluent have been analyzed with respect to PAE (see Table 6). Generally speaking, effluent concentrations lie in the range of 0.02-49.9 $\mu g.l^{-1}$ (mean: 0.80 $\mu g.l^{-1}$) for DEP and 0.02-69.0 $\mu g.l^{-1}$ (mean: 2.44 $\mu g.l^{-1}$) for DEHP, with PAE ranges extending from 0.07 to 108 $\mu g.l^{-1}$. As with WWTP influents, no clear difference has been observed for PAE concentrations in final effluent whatever the location (Fig. 3). Median PAE concentrations in the samples from North America and Europe were quite similar, lying in the 4.85-5.35 $\mu g.l^{-1}$ range. As previously mentioned for WWTP influent, European effluent has exhibited a few high outliers. In Spain, these outlier concentrations typically exceeded 50 $\mu g.l^{-1}$; i.e. 10 times the average level. The little availability of phthalate data before 2000 does not allow defining a trend of these compounds in the discharges of WWTP. Surprisingly, the PAE median concentration measured in Spanish WWTP effluent exceeded that in WWTP influent (de los Rios et al. 2012; Bizkarguenaga et al. 2012; Sanchez-Avila et al. 2009; Sanchez-Avila et al. 2012). However, no explanation could be brought by authors to elucidate this difference, but the WWTP efficiency could however be subject to questioning.

According to our review, PAE can display moderate (50%) to high (94%) removal rates. In addition, a study conducted by Vogelsang et al. (2006) explained that chemical and biological treatment eliminates from 50-60% of phthalates in a Norwegian WWTP. In Spain, Sanchez-Avila et al. (2009) reported PAE removal efficiency of 68%. Finally, a study led by Marttinen et al. (2003) explained that DEHP has been removed from wastewater with an efficiency of 94%. In addition, both studies conducted by Fauser et al. (2003) and in EU (2008) reported similar removal efficiencies for DEHP. Ordinarily, phthalates are removed by different processes occurring within WWTP, including solid settlement, sorption to sludge, volatilization, biodegradation, hydrolysis and/or photolysis (Rogers 1996). Sorption on sludge is considered to be one of the major pathways for PAE removal in WWTP. Moreover, even if DEHP may be considered inherently biodegradable under aerobic conditions (Staples et al. 1997), it was not removed by the biological pathway. A study conducted by Gavala et al. (2004) showed that enzymatic pretreatment increased DEHP biodegradation rate in secondary sludge. Furthermore, it was showed that recirculating sludge and thus retaining specific micro-organisms could stabilize removal capacity up to 86% whereas continuous flow had a varying removal of 77-88% (Oliver et al. 2007, 2005). Alternatively, Vogelsang et al. (2006) and Staples et al. (1997) reported that DEP, a soluble and biodegradable compound under aerobic conditions, has been removed during biological treatment process, although it has not been significantly eliminated by

any chemical or mechanical sedimentation processes. As a general rule, in WWTP effluent, DEHP is expected to be the most abundant congener. Phthalates' distribution is however lightly different, compared to WWTP effluents, with a higher proportion of DnBP and DEP (Fig. 4).

Wastewater Treatment Plant Sludge

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

Stormwater

Although it has been established that stormwater is responsible for the spread of pollutants, especially in urban areas, data concerning the emission, occurrence and fate of PAE in stormwater remains poorly reported (Björklund et al. 2009; Rule et al. 2006; Makepeace et al. 1995; Clara et al. 2010; Pitt et al. 1999; Zgheib et al. 2012). According to these authors, phthalates in stormwater originated from plasticizers in PVC, paints, building materials, etc. For instance, wastewater from car washes can also make a significant contribution to the emission of DEHP (Vikelsoe et al. 1998). Interestingly, PAEs are not chemically bound to the material and may migrate from products during use and disposal (Cadogan et al.

1993). DEHP has been measured at concentrations between 0.45 and 24.0 µg.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 µg.l^{-1} (Zgheib et al. 2012). Such levels were higher to those previously reported for stormwater in Sweden (5 µg.l^{-1}, (Björklund et al. 2009)) and in London (0.75 to 1.25 µg.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 µg.l^{-1} to several tens of µg.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 µg.l^{-1}, respectively; the median concentration in the North American samples is relatively lower (0.27 µg.l^{-1}). Moreover, median PAE concentrations in the North American samples (0.29 µg.l^{-1}) are notably lower than the Chinese (1.24 µg.l^{-1}) and European samples (1.18 µg.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 µg.l^{-1} (i.e., 12 times the average level). In addition, in China (Sha et al. 2007), they were above 28 µg.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 µg.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

et al. 2005; Bendz et al. 2005; Prieto et al. 2007; Sanchez-Avila et al. 2012), revealing lower concentrations, i.e. of about 1.16 $\mu g.l^{-1}$ for the sum of PAE. The PAE concentrations plotted as a function of time for these two series are shown in Figure 6. In contrast, concentrations measured in the Chinese surface water have been increasing for the last decade (see Table 8). This is the result of the constant increase of industrialization, consumptions of phthalates and absence of regulation in China and more generally in developing countries (Zhu and Qiu 2011; Zeng et al. 2008; Sha et al. 2007; Li et al. 2006; Chen et al. 2012).

Most studies focusing on surface water analyze only the dissolved phase, hence only a few results are available for suspended solids (Table 9; four references available). According to these four studies, levels of phthalates in suspended solids range from a few to 630 mg/kg.dw (Table 9). Such contamination levels may be explained by the high hydrophobicity of DEHP and DnBP (with respect to their Log K_{ow}; see Table 1), which leads to sorption and accumulation of these compounds on suspended solids (Gounaris et al. 1993; Sha et al. 2007). For less hydrophobic compounds, contents are lower, i.e. about between 0.05 and 0.20 mg/kg for all compounds, except DEHP (0.70-630 mg/kg). Finally, suspended solids may, however, play a key role in aquatic systems since, under low-flow conditions, particles can settle and contribute to sediment formation, thus yielding contaminant stocks (Fig. 1 and Fig. 5). This phenomenon has been demonstrated for some heavy metals, pesticides and PCBs by Zgheib et al. (2012).

Sediments

Several sediment samples have been collected from rivers and lakes across Europe, North America and developing countries (particularly China and India) (Vitali et al. 1997; Fromme et al. 2002; Yuan et al. 2002; Vethaak et al. 2005; Sha et al. 2007; Zeng et al. 2008; Liu et al. 2010). The amount of DnBP and DEHP measured in these samples (see Table 10; 23 references) varied from 0.01 to 115 mg/kg.dw (median, 0.44 and 1.90 mg/kg.dw, respectively). Generally speaking, DMP and DEP were not detected in sediments (Fig. 4), mainly due to their low hydrophobicity. In addition, a recent study led by Liu et al. (2010) reported that DEP is easily degraded in top material and cannot be eluted in deep sediments. Moreover, under anaerobic conditions, DnBP and BBP are easily degraded whereas DEP and DEHP were poorly removed (Yuan et al. 2002). From the exhaustive data in literature, it is clear that PAE are often present in sediment regardless of the location (Fig. 2). The variation in phthalate contents among sediments was examined using published data, which provided sufficient details for determining the statistical distribution of these concentrations. Huang et al. (2005) examined the evolution of DEHP sediment content along a river transects densely populated. They revealed the existence of dynamic processes occurring in surface water, such as sorption to settleable particles and atmospheric volatilization governing the fate of phthalates in the environment and could lead to the formation of sinks in sediments and atmospheric compartment.

A comparison between Tables 9 and 10 indicates that the PAE contents found in sediments are close to those found in suspended solids (Fig. 5). As previously mentioned and highlighted in Figure 1, sediments and suspended solids are linked through sedimentation during low-flow periods and through re-suspension during high-flow periods (Mitsunobu and Takahashi 2006). Presumably, the high K_{ow} of

certain phthalates explains their sorption to particles in water, which in turn settle to form sediment (Staples et al. 1997). Additionally, this phthalate accumulation in sediments is fed and promoted by anthropogenic suspended solids, such as particles released by municipal WWTP and untreated water (Srivastava et al. 2009; Huang et al. 2005).

Soils

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

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

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 $\mu g.l^{-1}$) 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

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

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

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

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Figure

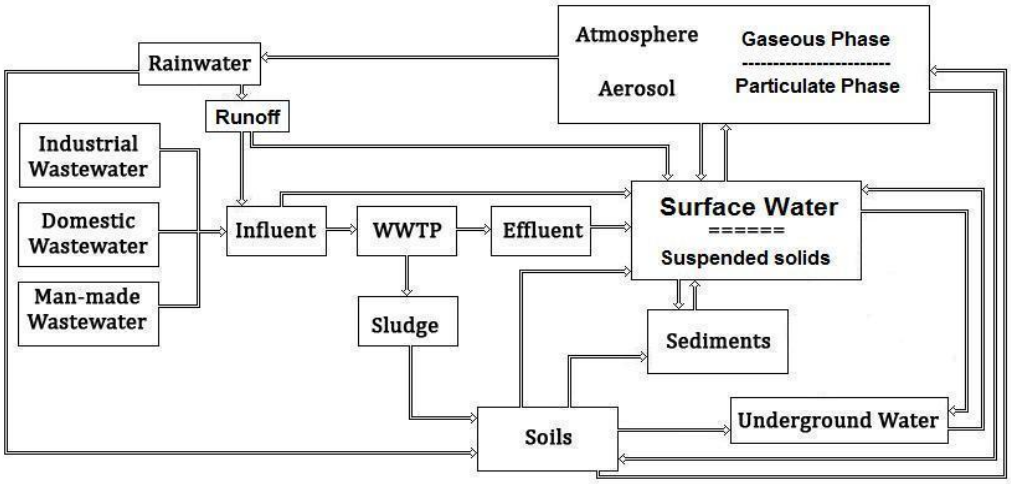

Figure 1: Interactions between the various environmental compartments – Cited from Bergé et al. (2012a)

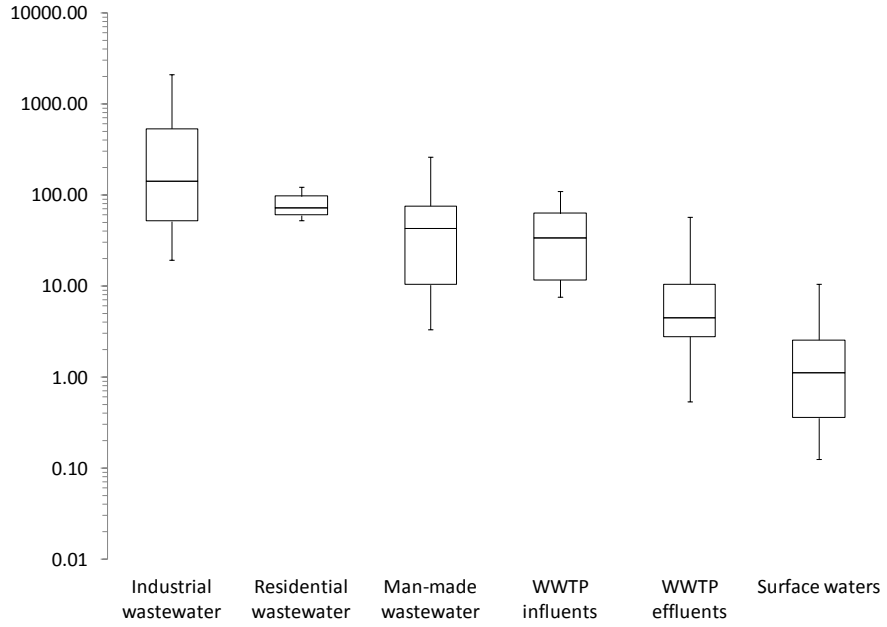

Figure 2: Total PAE (DMP + DEP + DnBP + BBP + DEHP + DnOP) concentrations (in µg.l^{-1}) in liquid matrices (WWTP: Wastewater Treatment Plant - global scale)

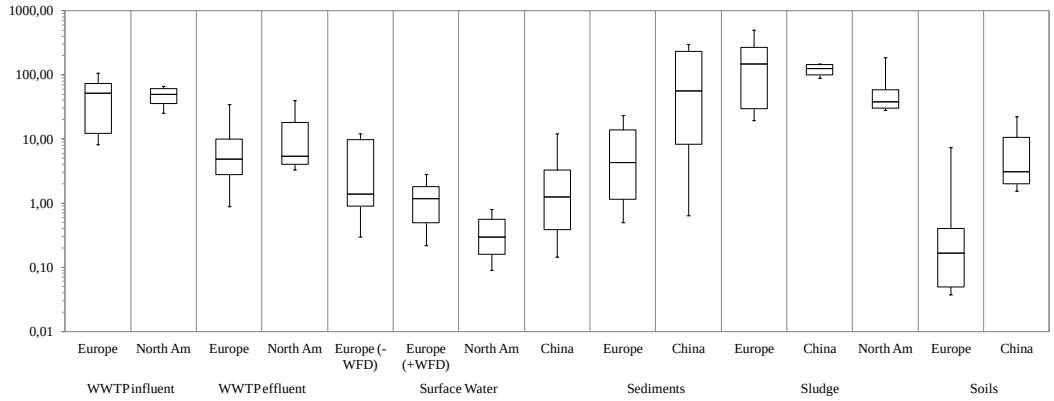

Figure 3: Distributions of total PAE (DMP + DEP + DnBP + BBP + DEHP + DnOP) concentrations (µg.l^{-1}) and contents (mg/kg.dw) in WWTP influent and effluent, surface water, sediments, sludge and soils from various locations (WFD: Water Framework Directive; North Am: North America)

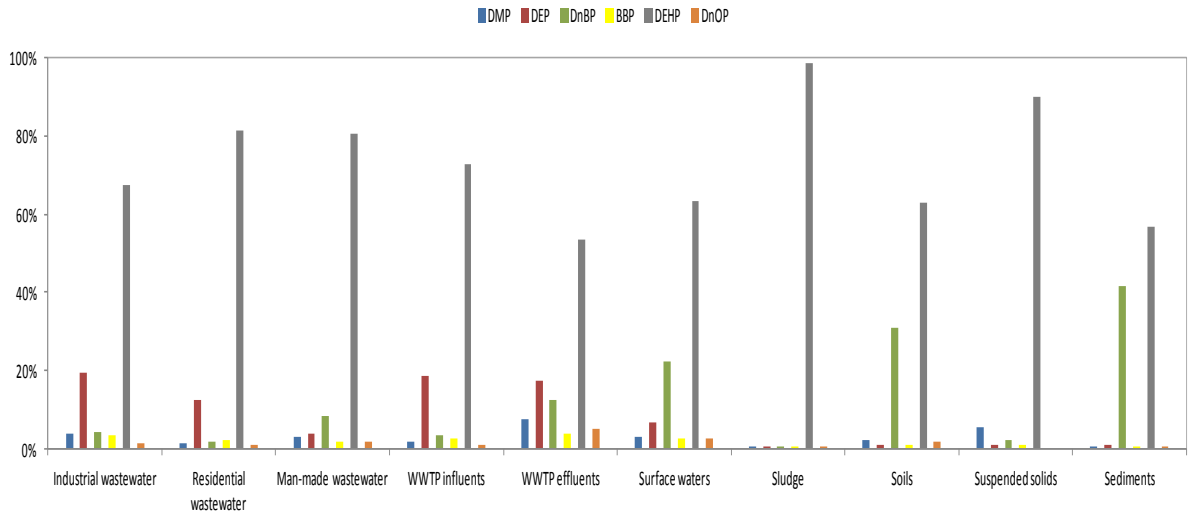

Figure 4: Evolution in the distribution of DMP, DEP, DnBP, BBP, DEHP and DnOP through the environmental cycle (WWTP: Wastewater Treatment Plant)

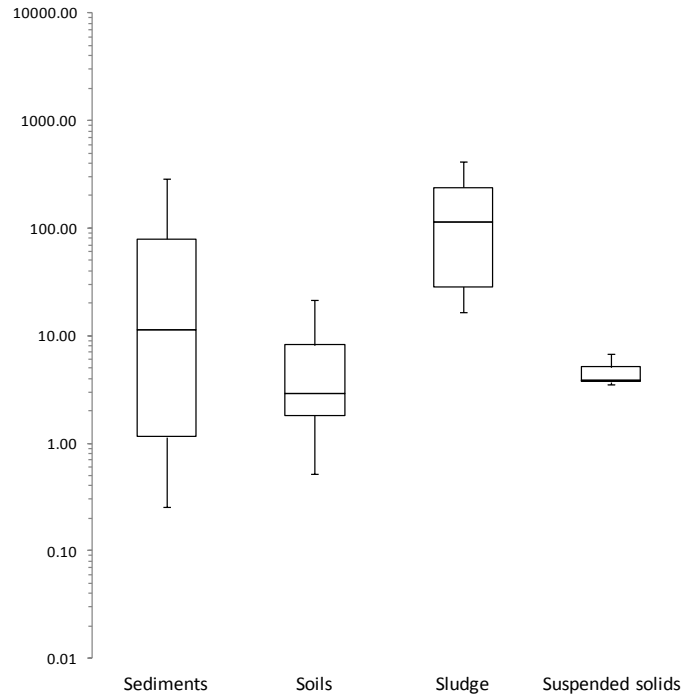

Figure 5: Total PAE (DMP + DEP + DnBP + BBP + DEHP + DnOP) contents (in mg/kg.dw) in solids matrices - global scale

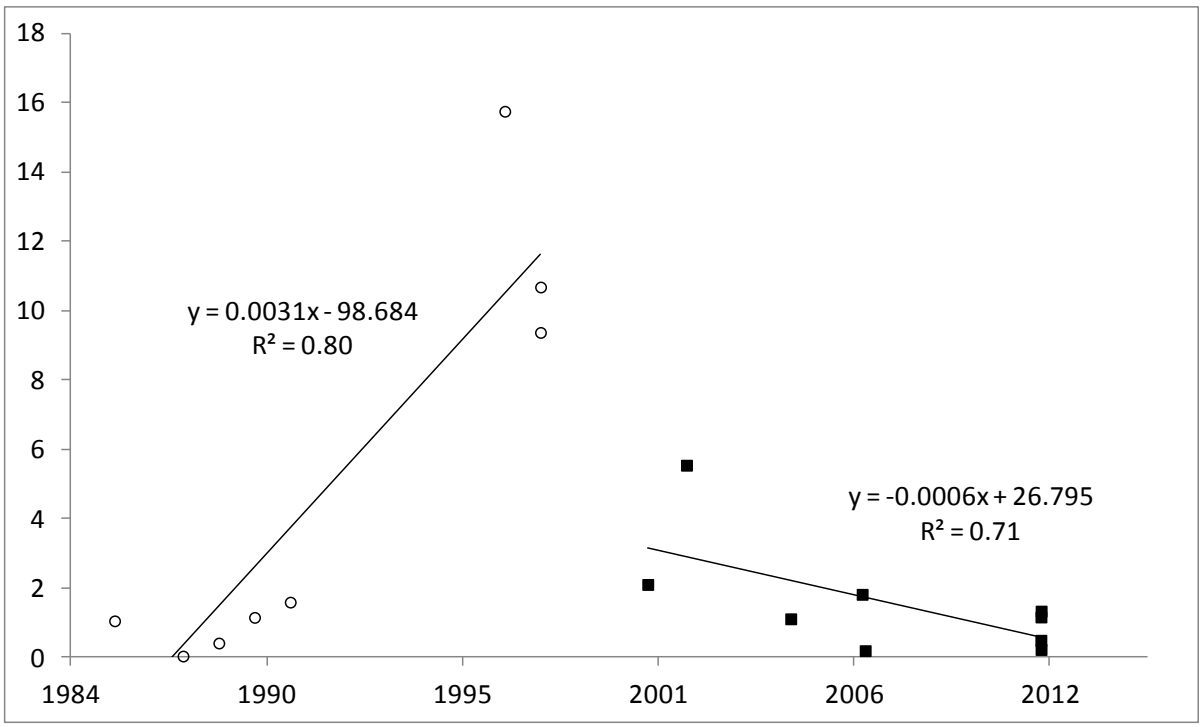

Figure 6: Historical trend lines for the median total PAE (DMP + DEP + DnBP + BBP + DEHP + DnOP) concentrations (in $\mu g.l^{-1}$) in European surface water. The dots represent samples collected before implementation of the Water Framework Directive, whereas the squares depict samples collected after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

Table 1: Physicochemical properties of PAE

Compound	Formula	MW (g/mol)	Water solubility at 20 °C (mg/l)	Log K_{ow}	Log K_{oc}	Log Kd	H ($Atm.m^3/mol$)
DMP	$C_{10}H_{10}O_4$	194.2[c]	4,20[c]	1.61[c]	1.74[c] 1.90-2.56[b]		1.22 E^{-07}[c]
DEP	$C_{12}H_{14}O_4$	222.2[c]	1,10[c]	2.42[c] 2.47-2.51[b]	1.84[c] 2.85-3.24[b]		2.66 E^{-07}[c] 7.80 E^{-07}[b]
DnBP	$C_{16}H_{22}O_4$	278.4[c]	11.2[c]	4.57[c]	3.14[c] 4.17[b]		8.83 E^{-07}[c]
BBP	$C_{19}H_{20}O_4$	312.4[c]	2.70[c]	4.84[c]	4.23[c] 3.95[b]	3.55[c]	7.61 E^{-07}[c]
DEHP	$C_{24}H_{38}O_4$	390.6[c]	0.003[c]	7.50[c]		4.12[c] 4.18[a]	1.71 E^{-05}[c]
DnOP	$C_{24}H_{38}O_4$	390.6[c]	0.0005[c]	8.06[c]	4.94[c] 5.68[b] 5.71[a]	4.46[c]	1.03 E^{-04}[c]
a: (Lützhoft et al. 2008); b: (IPCS 2003); c: (Staples et al. 1997)							

Table 2: Atmospheric contamination by PAE (in ng/m^3)

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	<loq-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	<loq-6.50	<loq-0.11	(Salapasidou et al. 2011)
Greece			0.43-2.40	0.04-0.98	4.63-45.0		(Salapasidou 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 µg.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			<loq-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			<loq-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	<loq-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 µg.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 efluents	<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	

Table 5: WWTP influent concentrations of PAE (in µg.l^{-1})

Location	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
Finland	<loq-1.00	6.00-11.0	6.00-8.00	3.00-5.00	28.0-66.0	<loq-1.00	(Marttinen et al. 2003)
Finland	<loq-1.00	5.00-74.0	3.00-9.00	<loq-3.00	28.0-122	<loq-2.00	(Marttinen et al. 2003)
Finland		<loq-10.0	4.00-6.00	<loq-2.00	83.0	<loq-1.00	(Marttinen et al. 2003)
Denmark			<loq-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		<loq-10.0	<loq-3.60	14.0	33.0	4.20	(Jackson and Sutton 2008)
USA		<loq-10.0	<loq-3.60	1.90	9.20	<loq-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	<loq-2.40	0.77-9.20	<loq-8.70	0.31-3.20	3.40-34.0	<loq-1.10	(Clara et al. 2010)
Austria	0.26-0.41	1.20-2.00	<loq-0.47	<loq-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	<loq-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	<loq-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	<loq-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	<loq-0.96	0.23-1.54			<loq-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 µg.l^{-1})

Location	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
Finland			2.00		2.00	<loq-1.00	(Marttinen et al. 2003)
Finland	<loq-1.00	<loq-4.00	<loq-6.00	<loq-1.00	2.00-8.00		(Marttinen et al. 2003)
Finland					4.00		(Marttinen 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-032	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		(Marttinen 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	(Laturnus and Gron 2007)
Denmark	Amended		0.91	0.56				(Laturnus 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 µg.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		(Vikelsoe 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		(Laturnus and Gron 2007)
Denmark	Compost		0.46	0.29		12.2	1.24	(Laturnus 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	