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REVIEW ARTICLE

Meta-analysis of environmental contamination by alkylphenols

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Abstract Alkylphenols and alkylphenol ethoxylates (APE) are toxics classified as endocrine-disrupting compounds; they are used in detergents, paints, herbicides, pesticides, emulsifiers, wetting and dispersing agents, antistatic agents, demulsifiers, and solubilizers. Many studies have reported the occurrence of alkylphenols in different environmental matrices, though none of these studies have yet to establish a comprehensive overview of such compounds in the water cycle within an urban environment. This review summarizes APE concentrations for all environmental media throughout the water cycle, from the atmosphere to receiving waters. Once the occurrence of compounds has been assessed for each environmental compartment (urban wastewater, wastewater treatment plants [WWTP], atmosphere, and the natural environment), data are examined in order to understand the fate of APE in the environment and establish their geographical and historical trends. From this database, it is clear that the environment in Europe is much more contaminated by APE compared to North America and developing countries, although these APE levels have been decreasing in the last decade. APE concentrations in the WWTP effluent of developed countries have decreased by a factor of 100 over the past 30 years. This study is aimed at identifying both the correlations existing between environmental compartments and the processes that influence the fate and transport of these contaminants in the environment. In industrial countries, the concentrations observed in waterways now represent the background level of contamination, which provides evidence of a past diffuse pollution in these countries, whereas sediment analyses conducted in developing countries show an increase in APE content over the last several years. Finally, similar trends have been observed in samples drawn from Europe and North America.

Keywords Nonylphenol · Nonylphenol ethoxylate · Review · State-of-the-art

Introduction

Over the last three decades, a large number of studies have highlighted several categories of man-made compounds, classified as “endocrine-disrupting compounds” (EDC). Among the more frequently mentioned EDC, alkylphenols and their ethoxylates (APE) are of particular concern due to their ubiquity (Jobling and Sumpter 1993; Loos et al. 2007; Wang et al. 2010). The predominant uses of APE are found in pulp and paper production, textile manufacturing, and production of crop protection chemicals (Melcer et al. 2006). Moreover, their applications extend to the agricultural, industrial, and household sectors, by being incorporated into detergents, paints, herbicides, pesticides, emulsifiers, wetting and dispersing agents, antistatic agents, demulsifiers, and solubilizers (Fiege et al. 2000; Fu et al. 2007; Krogh et al. 2003; Langford and Lester 2002; Loos et al. 2007; Lorenc and Scheffer 2003;

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Shang et al. 1999; Ying et al. 2002). Among common APE applications, nonylphenol ethoxylates (NPE) account for approximately 80 % of total use, while octylphenol ethoxylates represent most of the remaining 20 % (Melcer et al. 2006; Renner 1997). According to Ayorinde et al. (1999), NPE are commercially available as ethoxylate mixtures with a chain length ranging either from 5 to 25 ethoxylate units (CH_2–CH_2–O–) (centered around 11 or 14 units) or from 15 to 40 ethoxylate units (centered around 25 units). Like most commercial chemical mixtures, the ethoxylate chain compositions determine the physicochemical properties of the mixture; NPE with 4 or 5 ethoxylate units are used as soluble cleaners in oil, while those with 8 or 9 ethoxylate units serve as the basis for high-performance cleaning agents found in the textile industry and those with 13 or 15 ethoxylate units are used for the preparation of emulsifiers, solvents, and pesticides (Berryman et al. 2003; Maguire 1999). Despite their various uses, NPE have in recent studies leveled off in American, Canadian, and European production, mainly as a result of highly restrictive regulations (EU 2003 18th June; USEPA 2010 August), while worldwide output had been increasing from 300,000 t in 1994 to 500,000 t in 2002 (Berryman et al. 2003; Ying et al. 2002). This difference may stem from the increase in consumption on the part of emerging nations, such as Brazil, Russia, India, and China.

Despite its societal benefits, APE consumption has lead to the release of APE into the environment, where they are now ubiquitous and can be found in air, water, sediments, and soils. This study focuses more specifically on five congeners of this family: nonylphenol diethoxylate (NP_2EO), nonylphenol monoethoxylate (NP_1EO), nonylphenoxy (ethoxy) acetic acid (NP_2EC), nonylphenoxy acetic acid (NP_1EC), and lastly nonylphenol (NP). These five compounds result from the degradation of long-chain congeners and are more persistent and toxic in the environment than their precursors (Soares et al. 2008). As a consequence, most studies in the literature examine the occurrence and behavior of these five compounds in various environmental matrices (surface water, wastewater, atmosphere, and treated water) given that the compounds are regularly found in urban and environmental compartments at significant levels (tens or more micrograms per liter) (Rudel et al. 2003; Stasinakis et al. 2008; Vethaak et al. 2005; Wilson et al. 2001).

Strategy of this review

Most of the literature on APE in the environment, as published since 2000 (Soares et al. 2008; Ying et al. 2002), have been concerned with one or more compartments, primarily downstream compartments such as wastewater treatment plant (WWTP) effluent and receiving waters, but none of these works have dealt with all environmental compartments, in particular urban wastewater, nor with APE behavior throughout the water cycle. 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 and residential wastewater, WWTP influent and effluent, and 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 (NP, $NP_{1–2}EO$, or $NP_{1–2}EC$), while others have reported on four or five (NP, $NP_{1–2}EO$, and $NP_{1–2}EC$). From the database we built, statistical calculations have been derived for certain sample sets. The year of sample extraction was not specified in all papers, so it was assumed that the samples were extracted on the paper's publication date. It was also decided by this study's authors that the review was to focus solely 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 the authors. This review is also aimed at determining whether temporal and/or geographical trends can be observed in APE contamination. To achieve this objective, the data were

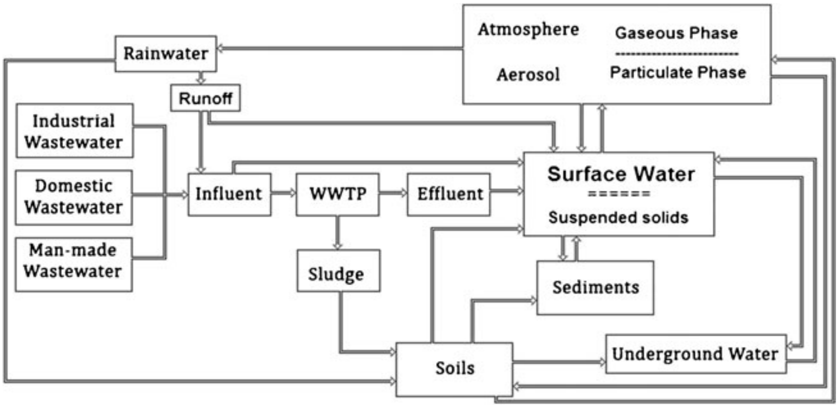


Fig. 1 Interactions between the various environmental compartments

examined from a geographical perspective. Recommendations for further research will also be provided.

Atmosphere

Air contamination

APE in the atmosphere of urban areas are generated from various emission sources, including road traffic, industrial processes, waste incineration, wastewater treatment processes, and domestic heating (Berkner et al. 2004). Furthermore, it has been reported that NP was detected in aerosols emitted from the aeration tank of a WWTP (Cincinelli et al. 2003). As shown in Table 1, a small number of air samples (eight references in all) have been analyzed for APE. The analysis of this database reveals that the North American samples were consistently above the values reported elsewhere in the world (Japan, Greece, Italy, etc.), and NP was the abundant alkylphenol in air, with concentrations ranging from 0.10 to 95.0 ng/m^3. Both of the studies conducted by Dachs et al. (1999) and Berkner et al. (2004) underscored that NP was preferentially associated with the gaseous phase more so than aerosols (between 90 and 98 % vs. 10 and 2 %, respectively): Dachs et al. (1999) reported NP concentrations in the gaseous phase ranging from 0.50 to 70.4 ng/m^3, while concentrations for aerosols varied between 0.90 and 4.60 ng/m^3. In addition, Berkner et al. (2004) suggested that a heavily industrialized and populated urban zone would emit more pollution than a rural zone. According to Xie et al. (2004), another source of potential contamination could be the emissions from surface water through water–air exchange processes. High concentrations of NP were indeed measured in rivers and estuaries; moreover, due to its vapor pressure and to the Henry's Law constant (Table 2), NP was found in the gaseous phase of the atmosphere (Dachs et al. 1999; Salapasidou et al. 2011; Van Ry et al. 2000). Keep in mind that the photochemical degradation of NP occurs with a half-life of approximately 10 to 15 h (Warhurst 1995). Due to the limited number of studies available, no distinct temporal or geographical trend could, therefore, be drawn.

Rain water

A few measurements of APE in rain water have been undertaken (see Table 3). Outdoor levels of NP lie on the order of 0.03–9.20 and 0.44–3.10 µg/l for NPE. NP concentrations in rain water and snow were found at levels between 0.03 and 1.20 µg/l, thus suggesting that wet deposition must be considered as a major source of NP in the environment (Björklund et al. 2009; Fries and Püttmann 2004). At the scale of Europe, data show that NP concentrations in rain water are on the decline, which implies that the environment in European countries is exposed to decreasing contamination. In Germany, for example, it has been reported that rain water is more highly contaminated in urban and suburban areas than in rural or remote zones (0.53 vs. 0.10 ng/m^3) (Fries and Püttmann 2004).

Bressy et al. (2011) extrapolated AP fluxes, on a yearly scale, from the total mass of AP measured in bulk atmospheric deposition, building runoff, road runoff, and stormwater on an urban catchment. According to two different hypotheses, the first based on an extrapolation of the number of rainy days and the second on an extrapolation of total rainfall, the NP fluxes ranged from 44.0 to 84.0 µg/m^2/year for atmospheric deposition and from 100 to 190 µg/m^2/year for stormwater. These authors, therefore, concluded that local human activity (runoff) constitutes a significant source

Table 1 Atmospheric contamination by APE (in nanograms per cubic meter)

Location	NP	NP$_1$EO	NP$_2$EO	NP$_1$EC	NP$_2$EC	Reference
USA	0.50–70.4					Dachs et al. (1999)
USA	0.90–4.60					Dachs et al. (1999)
USA	<loq–81.0					Van Ry et al. (2000)
USA	45.0–95.0					Wilson et al. (2001)
Italy	<loq–10.0	<loq–14.0				Cincinelli et al. (2003)
Japan	<loq–53.0					Saito et al. (2004)
Germany	0.15–1.00					Berkner et al. (2004)
Germany	0.10–0.30					Berkner et al. (2004)
Germany	0.20–0.50					Berkner et al. (2004)
North Sea	0.03–0.11	0.01–0.03				Xie et al. (2006)
Greece	1.60–16.5	0.29–12.8				Salapasidou et al. (2011)
Greece	2.50–10.9	<loq–2.41				Salapasidou et al. (2011)
Min	0.10	0.03				$n=8$
Max	95.0	22.2				
Med	4.60	3.33				

Table 2 Physicochemical properties of APE

Compound	Formula	MW (g/mol)	Water solubility at 20 °C (mg/l)	Log K_{ow}	Log K_{oc}	Log K_d	H (Pa m^3/mol)
NP	$C_{15}H_{24}O$	220	5.43[a] 5.40[e]	4.48[b] 4.80[e]	5.39[c] 5.22[f] 4.70–5.60[g]	2.70–3.82[d]	3.00–4.00[e]
NP_1EO	$C_{17}H_{28}O_2$	264	3.02[a]	4.17[b]	5.46[c]	2.88–3.44[d]	–
NP_2EO	$C_{19}H_{32}O_3$	308	3.38[a] 3.28[e]	4.21[b]	5.18[c]	2.04–3.26[d]	–
NP_1EC	$C_{17}H_{26}O_3$	278	–	4.12	–	–	–
NP_2EC	$C_{19}H_{30}O_4$	322	–	–	–	–	–

[a] Ahel and Giger (1993a)

[b] Ahel and Giger (1993b)

[c] Ferguson et al. (2001a)

[d] Clara et al. (2007)

[e] Dachs et al. (1999)

[f] Isobe et al. (2001)

[g] Sekela et al. (1999)

of contamination. Moreover, since AP were mainly found in the dissolved fraction (between 65 and 85 %), runoff treatment devices based on settling are unlikely to be very efficient. Similarly, Björklund (2010) estimated in Sweden fluxes of 110 µg/m^2/year for NP and their ethoxylates.

Urban areas

Sewer contamination

Industrial, man-made, and residential wastewater

As reported in Table 4, only a few measurements of APE have been performed in wastewater (two to five references for each type of wastewater). Industrial NP levels were in the 0.13–200 µg/l range (median, 10.0 µg/l), i.e., twice as high as levels in residential wastewater (1.25–76.0 µg/l; median, 5.09 µg/l). Both the absolute level and proportion of NPE were greater, i.e., on the order of 0.26 to 70,000 µg/l (median, 240 µg/l) for industrial wastewater and 0.24 to 193 µg/l (median, 63.0 µg/l) for domestic water. In Europe, it has been reported that high NP and NPE concentrations are measured in industrial wastewater. In Finland, for instance, NP and NPE levels of 100 to 200 and 30,000 to 70,000 µg/l, respectively, were measured in untreated wastewater (OSPAR 2001). This same report explained that NPE and NP levels reaching 2,270 and 400 µg/l have also been measured in untreated industrial wastewater in the Netherlands. Some authors mentioned that the high concentrations obtained in Maresme wastewater (Catalonia, Spain) would indicate that AP and APE are still being produced and used in industrial, household, and agricultural formulations despite the adoption of Directive 2003/53/EC (EU 2003 18th June) restricting their use within the EU to levels <0.1 % (Sanchez-Avila et al. 2009).

Table 3 Rainwater concentrations of APE (in micrograms per liter)

Location	NP	NP_1EO	NP_2EO	NP_1EC	NP_2EC	Reference
Germany	0.03–0.95					Fries and Püttmann (2004)
Netherlands	0.41	0.36–0.99			0.08–0.28	Vethaak et al. (2005)
UK	1.75					Rule et al. (2006)
Belgium	0.53					Michalowicz and Duda (2007)
Germany	0.25					Michalowicz and Duda (2007)
Sweden	0.10–1.20	1.10	2.00			Björklund et al. (2009)
France	0.05–0.34					Bressy et al. (2011)
Min	0.03	0.44				n=6
Max	1.75	3.10				
Med	0.41	1.98				

Table 4 Industrial, man-made, and residential wastewater concentrations of APE (in micrograms per liter)

Location	Type	NP	NP$_1$EO	NP$_2$EO	NP$_1$EC	NP$_2$EC	Reference
USA	Diaper service	<loq–0.25					Jackson and Sutton (2008)
USA	Pet wash	<loq–2.50					Jackson and Sutton (2008)
USA	Veterinary clinic	<loq–2.50					Jackson and Sutton (2008)
USA	Hospital	<loq–2.50					Jackson and Sutton (2008)
USA	Medical clinic	<loq–1.25					Jackson and Sutton (2008)
Spain	Industrial/residential	<loq–193	7.95–505	24.9–193			Sanchez-Avila et al. (2009)
Switzerland	Residential	14.0					OSPAR (2001)
Netherlands	Residential	2.10–170					OSPAR (2001)
Netherlands	Residential	0.24–19.0	0.82–125		0.27–13.0		Vethaak et al. (2005)
USA	Residential	<loq–2.50					Jackson and Sutton (2008)
Bosnia	Residential	0.46–4.40	5.00–392		<loq–3.20	<loq–4.37	Terzic et al. (2008)
Spain	Residential	29.0	134	40.9			Sanchez-Avila et al. (2009)
Finland	Industrial effluent	100–200	30,000–70,000				OSPAR (2001)
Netherlands	Industrial effluent	<loq–400	<loq–2,270				OSPAR (2001)
Netherlands	Industrial effluent	0.44–39.0	0.26–2,300				Vethaak et al. (2005)
USA	Plastic manufacturer	<loq–2.50					Jackson and Sutton (2008)
USA	Paper manufacturer	<loq–0.25					Jackson and Sutton (2008)
USA	Beverage manufacturer	<loq–2.40					Jackson and Sutton (2008)
USA	Adhesive manufacturer	<loq–0.28					Jackson and Sutton (2008)
USA	Industrial laundry	<loq–6.25					Jackson and Sutton (2008)
USA	Residential laundry	<loq–2.50					Jackson and Sutton (2008)
Bosnia	Industrial effluent	<loq–105	9.50–750		<loq–9.50		Terzic et al. (2008)
Spain	Industrial effluent	8.60–79.8	8.82–398	17.5–698			Sanchez-Avila et al. (2009)
Man-made wastewater	Min	0.25	32.9				n=2
	Max	193	698				
	Med	16.7	140				
Residential wastewater	Min	0.24	1.09				n=5
	Max	170	400				
	Med	5.09	63.0				
Industrial wastewater	Min	0.25	0.26				n=5
	Max	400	70,000				
	Med	10.0	240				

Wastewater treatment plant influent

As opposed to industrial, man-made, and residential wastewater, a large number of WWTP samples have been analyzed for APE (see Table 5, i.e., 21 references). Moreover, according to these concentrations, the APE median levels reported for WWTP influent were significantly lower than industrial, man-made, and residential wastewater (APE, 25.7 µg/l in WWTP influent vs. 160, 32.9, and 40.0 µg/l in these types of wastewater, respectively (Fig. 2)) (Jackson and Sutton 2008; Sanchez-Avila et al. 2009). These differences between WWTP influent and residential wastewater can be explained by processes occurring within the sewer network. The first such process is the sorption of compounds on suspended solids, which may thereafter settle in the sewer network, hence leading to a decrease in concentration. In general, NP is considered a hydrophobic compound with a high Log K_{ow}, i.e., approximately 4.5, thus making it poorly soluble. As such, NP is preferentially associated with suspended solids (John et al. 2000; Langford and Lester 2002; Soares et al. 2008). In addition, the K_{d} values provided by Clara et al. (2007) (Log K_{d} ranges between 2.0 and 3.9 depending on APE) confirm the strong affinity of these compounds to the particulate phase. The second process is the degradation of APE. As suggested by Ahel et al. (1994b), John and White (1998), and Giger et al. (2009), biodegradation can actually occur within the sewer network in spite of anaerobic conditions. Moreover, Lian et al. (2009) advanced the hypothesis that, in Chinese wastewater flowing long distances in the sewer network before reaching the WWTP, a large

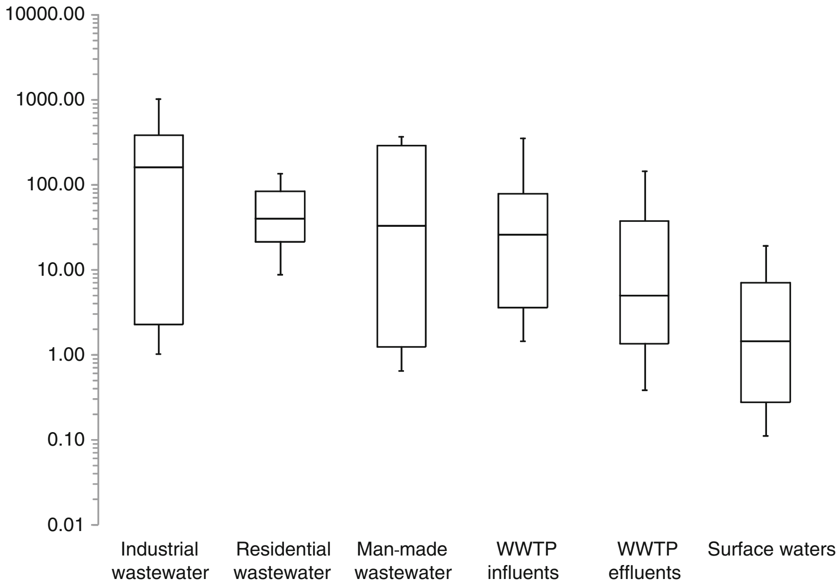
Table 5 WWTP influent concentrations of APE (in micrograms per liter)

Location	NP	NP_1EO	NP_2EO	NP_1EC	NP_2EC	Reference
Switzerland	110–430	230–570		80.0–270		Ahel et al. (1994a)
Spain	40.0–343					Sole et al. (2000)
Germany	2.13					Körner et al. (2000)
Japan		<loq–938		<loq–25.8		Fujita et al. (2000)
Spain	1.00–80.0			1.00–65.0		Petrovic et al. (2002)
Canada	<loq–51.8	<loq–4.00	<loq–1.80	<loq–27.0	<loq–19.0	Berryman et al. (2003)
Japan	0.50–20.0	0.80–17.0	0.50–11.0	0.08–0.78	0.10–4.70	Okayasu et al. (2003)
China	9.30					Shao et al. (2003)
Japan	1.00			0.10	0.20	Isobe and Takada (2004)
Japan	0.30–10.0					Nakada et al. (2006)
Australia	9.61					Tan et al. (2007)
USA	<loq–5.00					Jackson and Sutton (2008)
USA	<loq–1.25					Jackson and Sutton (2008)
Spain		1.47				Cespedes et al. (2008)
Spain	7.23	7.96				Cespedes et al. (2008)
Spain	17.5	8.19				Cespedes et al. (2008)
Spain	5.59	10.3				Cespedes et al. (2008)
Spain	10.4	5.41				Cespedes et al. (2008)
Denmark	1.10–8.40					Seriki et al. (2008)
China	4.18–18.7	16.0–60.6	2.40–66.0			Lian et al. (2009)
Spain	102	128	345			Sanchez-Avila et al. (2009)
Spain	9.70	171	188			Vega-Morales et al. (2010)
France	15.7	9.00	2.90		2.30	Martin-Ruel et al. (2010)
France	0.80	0.97	0.98			Gilbert et al. (2011)
France	4.08–10.6					Bergé et al. (2012)
Min	0.30	1.48				n=20
Max	430	964				
Med	9.61	17.5				

proportion of APE with long chains can thus biodegrade into short chains or carboxylate byproducts. This distribution evolves according to the season. NP and $NP_{1-3}EO$, therefore, represent 42 % of the total APE compounds during the summer vs. just 23 % in winter (Loyo-Rosales et al. 2007). This difference has been attributed to a higher biodegradation rate of the APE with long chains in pipes in the summer compared with winter, as a result of higher temperatures. The third process involves losses due to volatilization inside the sewer network, according to Ney (1990). With respect to both its vapor pressure and the Henry's Law constant (see Table 2), NP is considered a semivolatile organic compound, which tends to transfer into the vapor phase. This process is enhanced by the temperature and movements in sewer pipes. Moreover, infiltration and exfiltration are interactive processes that can dynamically affect the quality of wastewater during its transit period in the network.

The variations in APE concentrations among WWTP influent has been further investigated using published data, which have provided sufficient details for determining the statistical distribution of concentrations. Figure 3 shows data collected for geographical areas including Europe, North America, and China. This figure 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. APE median concentrations in samples from North America and Europe were quite similar: 15.5 and 15.9 µg/l, respectively. The median concentration in Chinese samples was higher (39.5 µg/l). It is also interesting to note that European countries display only a few outliers to the high side. In Spain, these outlier concentrations typically exceed 500 µg/l, i.e., 30 times the average level. The reason for these outliers is not yet clear, although the presence of heavily loaded industrial effluent could be hypothesized. The database analysis indicates that the APE concentrations measured in Chinese influent were above those measured in Europe and North America. This difference may be tied to several factors: (1) greater use or consumption of NP-containing goods in China; (2) a higher proportion of industrial influent in wastewater; and (3) an absence of pertinent regulations.

Fig. 2 Total APE (NP + NPE) concentrations (in micrograms per liter) in liquid matrices (*WWTP* wastewater treatment plant—global scale)



Wastewater treatment plant effluent

Understanding the biodegradation processes of nonylphenolic compounds proves to be a critical factor in predicting the fate of these compounds in the environment. From an overall standpoint, the removal of APE at conventional activated sludge WWTP has been well documented (Ahel et al. 1994a; Giger et al. 2009). A large number of samples from WWTP effluent have been analyzed with respect to APE (see Table 6). Generally speaking, effluent concentrations lie in the range of 0.03–200 µg/l (mean, 1.42 µg/l) for NP and 0.10–1,179 µg/l (mean, 4.54 µg/l) for the sum of NPE, with APE ranges extending from 5.02 to 1,181 µg/l. As opposed to WWTP influent, no clear difference has been observed for APE concentrations in final effluent anywhere in the world (Fig. 3). Median APE concentrations in the samples from North America, China, and Europe were all quite similar, lying in the 4.67–6.12 range. The 10th percentile concentration for China, however, was significantly higher than the North American and European values (3.84 µg/l). This difference points to a smaller disparity in contamination levels in China than in Europe and North America. Furthermore, this difference may result from the disparities between measurements conducted in the 1980s and more recent studies. In plotting a linear regression for this database, as separated into two periods (i.e., before and after the implementation of the European Water Framework Directive (WFD); EU 2000), the concentrations over time display a good linear fit. The implementation of a new regulation clearly influences the levels observed in wastewater, with the system reacting quite rapidly: European samples before the WFD implementation were on a positive slope, whereas post-implementation samples showed no slope.

As previously mentioned for WWTP influent, European effluent has exhibited a few high outliers. In Spain, these outlier concentrations typically exceed 400 µg/l. In general, however, APE concentrations in the WWTP effluent of developed countries have decreased by a factor of 100 over the last 30 years (Ahel et al. 1994a; Jonkers et al. 2009). According to our review, APE can display moderate (50 %) to high (90 %) removal rates. In addition, Ahel et al. (1994a) estimated that at least 60 to 65 % of all nonylphenolic compounds entering the WWTP are discharged into receiving waters alongside the effluent, with 51 % being released as metabolic products after undergoing mechanical and biological treatments (Ahel et al. 1994a; Giger et al. 2009; John and White 1998). These authors have all reported that, during biological treatment, APE can be degraded aerobically and anaerobically in generating shorter-chain APE and AP, both of which are resistant to any further degradation under anaerobic conditions (Birkett and Lester 2003; Ying 2006; Ying et al. 2002). As a general rule, in WWTP effluent, NPEC are expected to be the most abundant group, thus indicating that NPEC were the main metabolites of NPEO during biological degradation processes (Ahel et al. 1994a; Giger et al. 2009; Loyo-Rosales et al. 2007). The increased proportion of NPEC in WWTP effluent (Fig. 4) highlights the preference for long-chain ethoxylate compounds in this degradation pathway (Thiele et al. 1997). Moreover, McAdam et al. (2010) observed negative removal rates for short-chain ethoxylates ($NP_{1-2}EO$) and carboxylates ($NP_{1-2}EC$) in WWTP.

Wastewater treatment plant sludge

As mentioned above, sorption on sludge is considered to be one of the major pathways for APE removal in WWTP (Shao et al. 2003; Soares et al. 2008). A large number of sludge samples have been analyzed for APE (see Table 7). Most studies have reported significant levels in sludge, i.e.,

Table 6 WWTP effluent concentrations of APE (in micrograms per liter)

Location	NP	NP$_1$EO	NP$_2$EO	NP$_1$EC	NP$_2$EC	Reference
Switzerland	5.00–11.0	30.0–65.0	47.0–77.0			Ahel and Giger (1985)
USA		<loq–0.08	<loq–0.15		<loq–0.16	Clark et al. (1992)
Switzerland	10.0–200	10.0–620				Ahel et al. (1994a)
USA	0.80–15.1					Lee and Peart (1995)
Canada	1.00–2.00					Lee and Peart (1995)
Taiwan	1.60	9.60	15.3	19.2	99.2	Ding and Tzing (1998)
USA	16.0	5.50	0.80			Rudel et al. (1998)
Germany		0.32–1.57				Körner et al. (2000)
Spain	6.00–289					Sole et al. (2000)
Japan	<loq–1.70	<loq–60.0		<loq–1,119		Fujita et al. (2000)
Germany	0.25–2.30					Spengler et al. (2001)
Germany	0.03–0.77					Kuch and Ballschmiter (2001)
Japan	0.08–1.24	0.20–2.96				Isobe et al. (2001)
Spain	<loq–154			4–105		Petrovic et al. (2002)
Canada		<loq–3.10	<loq–1.40	<loq–20	<loq–15	Berryman et al. (2003)
China	1.50					Shao et al. (2003)
Germany	0.24	0.18	0.21	0.80		Jahnke et al. (2004)
Japan	0.10			2.90	2.00	Isobe and Takada (2004)
Norway	0.05–1.31					Johnson et al. (2005)
Netherlands	0.55–1.50	1.90–2.20				Vethaak et al. (2005)
Japan	0.50–1.10					Nakada et al. (2006)
Belgium	0.25–0.73	0.20–1.30	0.05–2.10	0.01–4.50	0.02–0.9	Loos et al. (2007)
USA	<loq–0.25					Jackson and Sutton (2008)
Denmark	<loq–0.60					Seriki et al. (2008)
Spain		3.85				Cespedes et al. (2008)
Spain	0.38	4.98				Cespedes et al. (2008)
Spain	2.07	3.63				Cespedes et al. (2008)
Spain	1.42	3.81				Cespedes et al. (2008)
Spain	0.33	4.45				Cespedes et al. (2008)
China	<loq–0.44	<loq–4.58		<loq–16.8		Lian et al. (2009)
Spain	21.9	47.7	12.6			Sanchez-Avila et al. (2009)
Greece	0.85–37.9	0.05–1.30	0.05–1.00			Arditsoglou and Voutsa (2010)
Spain	18.7	279	154			Vega-Morales et al. (2010)
Spain	17.5	274	162			Vega-Morales et al. (2010)
France	1.30	0.47	0.95		2.20	Martin-Ruel et al. (2010)
France	0.15	0.10	0.15			Gilbert et al. (2011)
France	0.31–1.36					Bergé et al. (2012)
Min	0.03	0.10				$n=32$
Max	200	1,179				
Med	1.28	4.50				

in the range of 1,000–2,000 mg/kg dry weight (dw). For example, levels of up to 2,530 mg/kg dw in digested sludge from Switzerland (Giger et al. 1984) and 1,800 mg/kg dw in final sludge from the USA (Pryor et al. 2002) have been recorded. Table 7 shows that the NP levels observed after anaerobic digestion were, in general, significantly higher than those after any other form of treatment (Kinney et al. 2006). Giger et al. (1984) were among the first to propose that the hydrophobic NP$_1$EO and NP$_2$EO compounds associated with wastewater solids were degraded biologically to NP in anaerobic digestion. This finding was confirmed by Minamiyama et al. (2006). Moreover, the lowest APE concentrations result from compost, presumably as a result of prolonged residence time under aerobic biodegradation at elevated temperatures during the curing treatment (Gibson et al. 2007; Kinney et al. 2006). The average congener distribution in

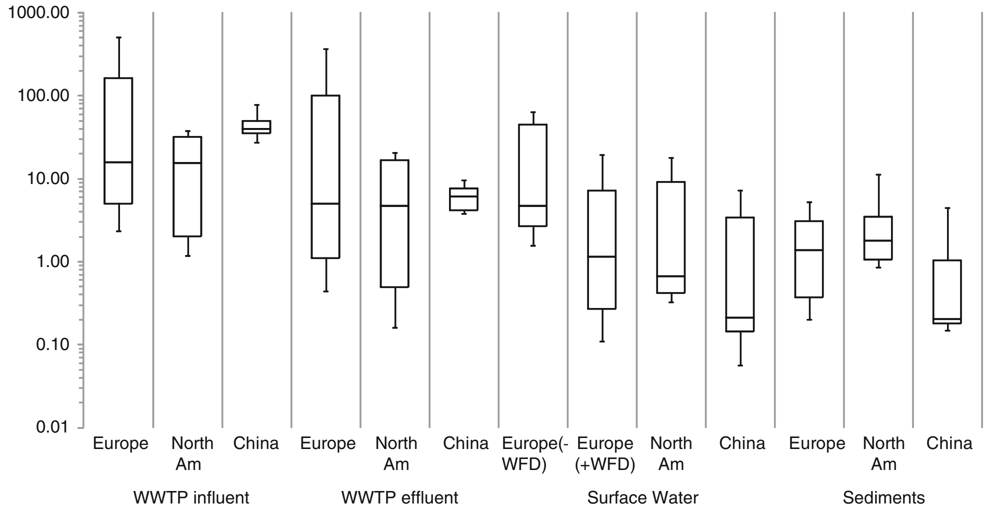


Fig. 3 Distributions of total APE (NP + NPE) concentrations (in micrograms per liter) and contents (in milligrams per kilogram) in WWTP influent and effluent, surface water, and sediments from various locations (*WFD* Water Framework Directive, *North Am* North America)

sludge samples is provided in Fig. 4; this information should not be interpreted too literally since, in a number of studies (Table 7), only a few congeners were measured. The most notable missing congeners are NPEC, which has rarely been measured in sludge. Interestingly, we can note an inverse distribution in sludge and WWTP effluent. It is clear from these data that APE contents are statistically close regardless of the location considered and moreover that NP 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 APE in stormwater remains poorly reported (Björklund et al. 2009). Several studies have recently identified stormwater as a potential source of organic pollutants in surface water (Björklund et al. 2009, 2010). NP in stormwater originates from paints and lacquers, motor oils, and concrete building materials (Björklund 2010). NP have been measured at concentrations of between 0.10 and 1.20 μg/l in urban stormwater in Sweden (Björklund et al. 2009). According to Bressy et al. (2011), APE levels range from 0.15 to 0.75 μg/l for building runoff, 1.50–1.90 μg/l for road runoff, and 0.16–0.92 μg/l for stormwater. Moreover, Bressy et al. (2011) underscored enrichment from the atmosphere to runoff due to the local contributions to building runoff (70 %) and road runoff (87 %). Also, AP are in fact degradation products of the APE found in detergents, paints, plastics, and construction materials (Soares et al. 2008; Ying et al. 2002).

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 (Musolff et al. 2010).

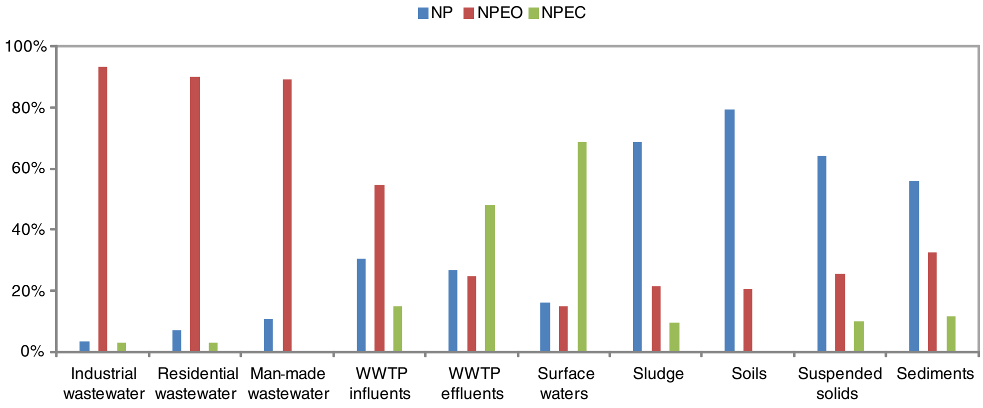


Fig. 4 Evolution in the distribution of NP, NPEO, and NPEC through the environmental cycle (*WWTP* wastewater treatment plant)

Table 7 APE in WWTP sludge contents (in milligrams per kilogram dw)

Location	Type	NP	NP$_1$EO	NP$_2$EO	NP$_1$EC	NP$_2$EC	Reference
Spain	Primary sludge	0.90–194	<loq–525	<loq–325			Gonzalez et al. (2010)
Spain	Primary sludge	0.80	2.60	2.90			Vega-Morales et al. (2010)
Spain	Secondary sludge	<loq–194	<loq–285	<loq–344			Gonzalez et al. (2010)
USA	Secondary sludge	2.18–491	3.96–79.4	0.79–89.0			Kinney et al. (2006)
Finland	Anaerobically digested	640–2,200					Brunner et al. (1988)
Taiwan	Anaerobically digested	244					Cheng et al. (2001)
Taiwan	Anaerobically digested	267					Cheng et al. (2001)
China	Anaerobically digested	149–167	27.6–30.0	9.50–11.0			Lian et al. (2009)
Spain	Anaerobically digested	<loq–358	<loq–288	<loq–294			Gonzalez et al. (2010)
Spain	Aerobically digested	2.40–260	3.80–63.6	<loq–32.6			Gonzalez et al. (2010)
Switzerland	Digested sludge	450–2,530					Giger et al. (1984)
UK	Digested sludge	238					Gibson et al. (2005)
Switzerland	Final sludge	540–1,000					Ahel et al. (1994a)
Germany	Final sludge	128					Jobst (1995)
Austria	Final sludge	0.46–65.0	0.15–23.0				Gangl et al. (2001)
USA	Final sludge	1,100–1,800					Pryor et al. (2002)
Germany	Final sludge	0.04–650					Fragermann (2003)
Denmark	Final sludge	0.20–46.2					Seriki et al. (2008)
USA	Compost	5.38	0.72				La Guardia et al. (2001)
USA	Compost	172	2.55				La Guardia et al. (2001)
Spain	Compost	32.8–829	4.30–85.9	<loq–47.2			Gonzalez et al. (2010)
USA	Lime stabilization	820	81.7	23.5			La Guardia et al. (2001)
USA	Heat treatment	496	33.5	7.36			La Guardia et al. (2001)
China	Excess sludge	3.10–22.3	3.40–38.7	5.50–32.8			Lian et al. (2009)
Spain	Lagoon sludge	33.2–199	13.5–145	<loq–29.8			Gonzalez et al. (2010)
Spain	Dehydrated sludge	35.1–135					Gonzalez et al. (2010)
Spain	Mixed sludge	3.20–191	<loq–25.8	<loq–25.6			Gonzalez et al. (2010)
France	Treated sludge	9.90	8.90	6.50		18.0	Martin-Ruel et al. (2010)
Canada		137–470					Lee and Peart (1995)
USA		100–500					Lee and Peart (1995)
Denmark		0.02–0.13	0.30–67.0				Torslov et al. (1997)
Switzerland		44.0					Bätscher et al. (1999)
Germany		25.5					Petrovic and Barcelo (2000)
Germany		3.60–21.3					Schaecke and Kape (2003)
France		200–500					Dubroca et al. (2003)
Australia		0.35–513					Langdon et al. (2011)
Min		0.02	0.15				*n*=24
Max		2,530	850				
Med		128	40.2				

Throughout the world, therefore, the occurrence and fate of APE in surface water have been well documented, as demonstrated by the 41 references pertaining to surface water contamination (Table 8). First, close attention must be paid when comparing concentrations reported in the literature, since high spatial and seasonal variations have been noticed (Cailleaud et al. 2007; Micic and Hofmann 2009). The variations in APE concentrations in surface water can be examined using published data, which provide a source of sufficient details for determining the statistical distribution of concentrations. Figure 3 summarizes the data for three geographical areas, namely, Europe, North America, and China. The median NP concentrations for North America and China are very similar, i.e., both at 0.21 µg/l; the median concentration in the European samples is relatively higher (0.44 µg/l). Moreover, median APE concentrations in

Table 8 Surface water concentrations of APE (in micrograms per liter)

Location	Type	NP	NP$_1$EO	NP$_2$EO	NP$_1$EC	NP$_2$EC	Reference
Spain	River		<loq–4.80	<loq–1.90			Guardiola et al. (1991)
USA	River		<loq–0.60	<loq–1.20	<loq–20.0	<loq–11.8	Naylor et al. (1992)
Switzerland	River	0.30–45.0	3.00–69.0	0.30–30.0	1.00–45.0	2.00–71.0	Ahel et al. (1994b)
UK	River	<loq–12.0					Blackburn and Waldock (1995)
Switzerland	Estuary	0.70–26.0	2.80–41.0		29.0–48.8		Ahel et al. (1996)
UK	River	<loq–2.00					Janbakhsh (1996)
Canada	River–lake	0.21	1.30	1.40			Bennie et al. (1997)
USA	River	0.12–0.29	0.04–0.37				Naylor (1998)
China	River	0.60	0.40	0.90		<loq–0.14	Ding and Tzing (1998)
Austria	River	<loq–0.57	<loq–0.71	<loq–0.11			Guelden et al. (1999)
USA	River	<loq–1.19					Snyder et al. (1999)
Belgium	River	0.16–0.71	<loq–2.45				Ghijsen and Hoogenboezem (2000)
Switzerland	River	0.03–0.48			0.50–3.00		Ahel et al. (2000)
Japan	River	0.11–3.08					Tsuda et al. (2000)
USA	Bay	0.20	0.15	0.32			Ferguson et al. (2001b)
Japan	River	0.08–1.08					Isobe et al. (2001)
Germany	River	0.01–0.13					Kuch and Ballschmiter (2001)
Italy	Lagoon		1.10–38.5		0.60–102		Petrovic et al. (2001)
Germany	River	<loq–2.72	<loq–3.27	<loq–0.84			Fromme et al. (2002)
Netherlands	River	<loq–6.30					Vethaak et al. (2002)
USA	River	0.10–0.50	0.02–0.42	<loq–1.50			Rice et al. (2003)
South Korea	Lake–river	<loq–15.8					Li et al. (2004)
Spain	River	<loq–15.0	<loq–31.0		<loq–35.0		Petrovic and Barcelo (2004)
Italy	River	0.10–1.60					Vitali et al. (2004)
China	River	0.11–0.30					Jin et al. (2004)
Netherlands	River	0.11–4.10	0.18–8.70				Vethaak et al. (2005)
Portugal	River	<loq–25.5					Quiros et al. (2005)
Spain	River	0.15–37.3	0.28–10.0				Cespedes et al. (2005)
Switzerland	River	0.03–0.33					Voutsa et al. (2006)
Spain	Estuary–sea	0.05–0.31	0.05–0.41				Prieto et al. (2007)
Canada	Lake	<loq–0.29	<loq–0.28	<loq–33.0	<loq–2.49	<loq–9.67	Mayer et al. (2007)
China	River–bay	0.02–28.6					Fu et al. (2007)
Belgium	River	0.63		0.22	6.37	2.48	Loos et al. (2007)
France	River	<loq–0.38	<loq–0.14	<loq–0.24	0.25–1.40	<loq–0.14	Cailleaud et al. (2007)
China	River	<loq–0.69	<loq–0.55	<loq–1.38	<loq–3.84	<loq–9.59	Yu et al. (2009)
Switzerland	River	0.19–0.30	0.01–0.15		0.09–0.50		Jonkers et al. (2009)
Greece	River	0.15–0.39	0.07–0.34	0.02–0.07			Arditsoglou and Voutsa (2010)
Spain	River	<loq–0.53		<loq–0.27	<loq–6.11	<loq–30.6	Brix et al. (2010)
France	River	0.06–0.43	0.01–0.08	0.02–0.41			Cladière et al. (2010)
Singapore	River	0.11–3.91	0.05–5.89				Xu et al. (2011)
Min		0.01	0.03				*n*=41
Max		45.0	215				
Med		0.33	0.99				

the North American samples (0.67 µg/l) are notably higher than the Chinese samples (0.21 µg/l) yet remain much lower than the European samples (2.45 µg/l). This difference can be explained by a former contamination in the case of the European media. It is also interesting to note that all geographical areas displayed a few high outliers. In Spain, these outlier concentrations typically exceeded 35 µg/l, i.e., 80 times the average level.

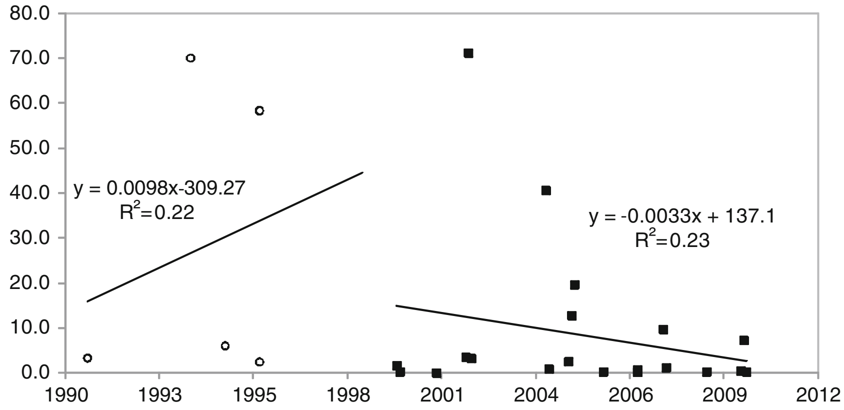


Fig. 5 Historical trend lines for the median total APE concentrations (in micrograms per liter) in European surface water. The *dots* represent samples collected before the implementation of the WFD, whereas the *squares* depict the samples collected after its implementation

It is obvious from the database that APE concentrations in European surface water first increased before undergoing a decrease due to the tighter regulations (EU 2003 18th June; USEPA 2010 August). It would be appropriate to treat these datasets as two separate time series. The first series contains the samples from European countries before the adoption of the WFD (Ahel et al. 1994b, 1996; Guardiola et al. 1991; Janbakhsh 1996; Guelden et al. 1999), all of which exhibit APE concentrations from 0.16 to 45.0 μg/l for NP and from 2.45 to above 100 μg/l for NPE. The second series is composed of samples from European countries after the WFD implementation (Cailleaud et al. 2007; Cespedes et al. 2005; Cladière et al. 2010; Fromme et al. 2002; Vethaak et al. 2005; Vitali et al. 2004), all of which reveal lower concentrations, i.e., of about 0.43 μg/l for NP and 0.66 μg/l for NPE. The APE concentrations plotted as a function of time for these two series are shown in Fig. 5. As a matter of fact, Quednow and Püttmann (2009) studied the influence of this European Directive (EU 2003 18th June) on NP concentrations and loads found in Rhine River tributaries within Germany. Their results indicated a sharp decrease in concentrations and loads in all tributaries as a result of the lower inputs imposed by use restrictions due to the implementation of European Directives. This decrease is in good agreement with the projected decrease for WWTP influence issued 10 years ago.

Owing to their physicochemical properties, APE have also contaminated suspended solids. From data presented in the literature and in focusing on developing countries, it is clear that the APE contents of suspended solids have been increasing rapidly in Chinese surface water. The initial results found in China in 1998 (Ding and Tzing 1998) were indeed in the same range of current concentrations as those in European rivers (0.60 μg/l for NP and approximately 1.00 μg/l for NPE). However, more recent studies performed in China (Fu et al. 2007; Jin et al. 2004; Yu et al. 2009), South Korea (Li et al. 2004), and Singapore (Xu et al. 2011) have revealed higher levels due to increased industrialization and APE consumption in these countries over the last decade (NP ranges from 0.02 to 28.6 μg/l in Chinese rivers, up to 15.8 μg/l in Korean rivers, and from 0.11 to 3.91 μg/l in Singapore's rivers). As demonstrated by Jonkers et al. (2001), aerobic biodegradation

Table 9 APE in suspended solid contents (in milligrams per kilogram dw)

Location	NP	NP$_1$EO	NP$_2$EO	NP$_1$EC	NP$_2$EC	Reference
Japan	1.58–5.50					Isobe et al. (2001)
USA	3.58–8.39	2.03–18.3	1.09–18.2			Ferguson et al. (2001a)
China	1.00–16.6					Li et al. (2004)
Netherlands	0.08–120	0.09–95.0				Jonkers et al. (2005a)
Netherlands	0.02–20.0	0.09–88.0				Jonkers et al. (2005b)
Netherlands	<loq–4.10	<loq–22.0				Vethaak et al. (2005)
Italy	0.24–7.32					Patrolecco et al. (2006)
France	0.29–7.11	0.04–0.69	0.02–0.76	0.02–0.51		Cailleaud et al. (2007)
Europe	0.02–0.18	0.02–0.12	<loq–0.10			Micic and Hofmann (2009)
Greece	0.41–4.98	0.96–17.9	0.39–5.75			Arditsoglou and Voutsa (2010)
Min	0.02	0.02				*n*=10
Max	120	95.0				
Med	0.85	0.34				

plays a key role in the fate of NPE and NP in surface water. Due to high oxygen concentrations in these waters, the aerobic pathway is in fact being favored over the anaerobic pathway. As a consequence, carboxylate derivates (NP_1EC and NP_2EC) are predominant in surface water, with concentrations ranging from below 1.00 μg/l to more than 100 μg/l, even for recent studies (Fig. 4).

Most studies focusing on surface water analyze just the dissolved phase, hence only a few results are available for suspended solids (Table 9; nine references available). According to these studies, the levels of NP and NPE in suspended solids range from 0.02 to 120 mg/kg dw and from 0.02 to 95.0 mg/kg dw (Table 9), respectively. Such contamination levels may be explained by the high hydrophobicity of NP and NPE (with respect to their Log K_{ow}; see Table 2), which leads to sorption and accumulation of these compounds on suspended solids and sediments (Navarro et al. 2009). Despite their contamination, suspended solids only account for 10 to 20 % of the total load in surface water (Cladière et al. 2010; Isobe et al. 2001). 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.

Case study of the Glatt River

Among the rivers studied throughout the world, Switzerland's Glatt River receives the most attention. EAWAG (the Swiss Federal Institute of Aquatic Science and Technology) has actually been investigating concentrations of NP, NPEO, and NPEC on this river since 1983 (Ahel et al. 1994b, 1996, 2000; Giger et al. 2009; Jonkers et al. 2009; Voutsa et al. 2006). Thanks to these five studies, Glatt River concentrations are available from 1983 to 2006 (Fig. 6 and Table 9). As an initial observation, regardless of the sampling campaign, between 1983 and 2006, NPEC concentrations were always higher than those for NPEO and NP. This predominance highlights the importance of the aerobic biodegradation pathway for NPEO in surface water. As regards NP, Ahel et al. (1994b) and (1996) reported concentrations ranging from 0.30 to 45.0 μg/l (with a median of around 2.00 μg/l), while more recent studies (Jonkers et al. 2009; Voutsa et al. 2006) indicated significantly lower concentrations, i.e., in the range of about 0.03–0.30 μg/l, with a median of 0.10 μg/l. Similarly, concentration decreases have been noticed for NPEO and NPEC (Fig. 7). These decreases may be correlated with the gradual restriction on the use of APE in Switzerland over the period from 1972 to 2005. While protocol no. 5 adopted on 22 July 1972 authorized the importation and stocks of NP, which probably led to high consumption of these compounds during the 1980s, the ordinance enacted on 18 May 2005 prohibited all use of NP and its ethoxylates. The impact of this latter ordinance on Glatt River concentrations is visible in particular on NP concentrations. As shown in Fig. 7, median NP concentrations remained relatively constant between 1998 and 2004 (Ahel et al. 2000; Voutsa et al. 2006), while they had dropped to a level three times lower by 2006 (Jonkers et al. 2009) (i.e., 0.25 and 0.20 μg/l in 1998 and 2004, respectively, vs. 0.06 μg/l in 2006). In addition to this use restriction, the improvement in WWTP effluent quality has played a critical role in the decreases observed on the Glatt River. Ahel et al. (1994a) reported NP concentrations in WWTP effluent of between 10.0 and 200 μg/l in 1984, whereas, more recently,

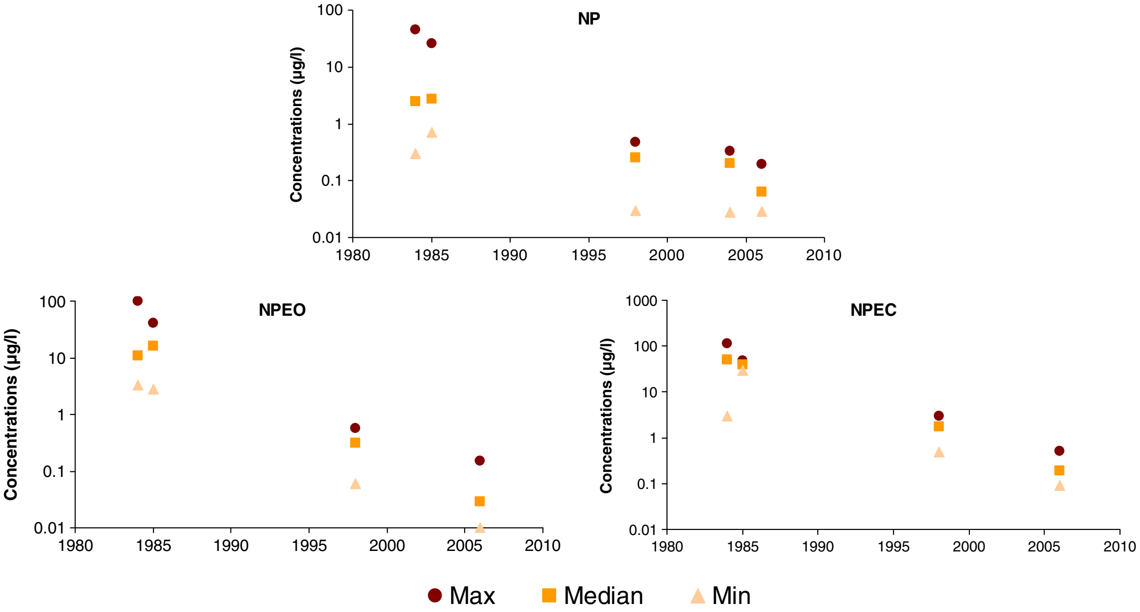


Fig. 6 Historical trend lines for the concentrations (in micrograms per liter) of NP, NPEO, and NPEC in the Glatt River

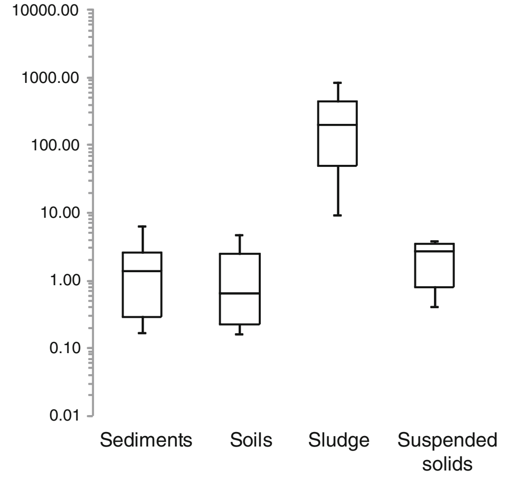


Fig. 7 Total APE (NP + NPE) contents (in milligrams per kilogram) in solid matrices—global scale

Jonkers et al. (2009) measured concentrations from <0.03 to 0.28 μg/l in 2006, i.e., about 850 times lower. The contribution of WWTP effluent to Glatt River contamination has thus declined, and this factor can explain the concentration decreases found in the river over the past 20 years.

Sediments

Several sediment samples have been collected from rivers and lakes across Europe, North America, and developing countries. The amount of NP measured in these samples (see Table 10; 36 references) has varied from 0.02 to 72.0 mg/kg dw (median, 0.62 mg/kg dw). Similarly, NPE levels range from 0.01 to 11.7 mg/kg dw (median, 0.66 mg/kg dw). From the exhaustive data in the literature, it is apparent that APE is often present in sediment regardless of the location. The variation in APE contents among sediments was examined using published data, which provided sufficient details for determining the statistical distribution of these concentrations. Figure 3 shows the data from three geographical areas: Europe, North America, and China. While APE median contents in North America and Europe were similar, at 1.79 and 1.37 mg/kg, respectively, sediments in China seem to be slightly less contaminated than European sediments (0.21 mg/kg). This difference may be explained by the fact that sediment contamination in Europe and North America has resulted from a much longer exposure period due to the past use history of these compounds. It is interesting to note that the contents measured in sediments depend on both the depth and location from where individual samples were extracted. Figure 8 synthesizes all of the data obtained in sediments between 1987 and 2009. Two different profiles for APE accumulation in European sediments are distinguishable, with a clear decrease after 2000, i.e., once the WFD was implemented (EU 2000). Moreover, Jin et al. (2007) measured APE as a function of depth in dated sediment cores, which served to indicate the biodegradation processes. Kannan et al. (2003a) reported NP values vs. distance in sediment between the upstream and downstream stretches of the river; this work revealed the existence of dynamic processes occurring in surface water, such as sorption to settleable particles and atmospheric volatilization.

A comparison between Tables 9 and 10 indicates that the APE contents found in sediment are close to those found in suspended solids. As previously mentioned and highlighted in Fig. 1, sediments and suspended solids are linked through sedimentation during low-flow periods and through resuspension during high-flow periods. Presumably, the high K_{ow} of NP explains its sorption to particles in water, which in turn sink to form sediment. Additionally, this NP accumulation in sediments is fed and promoted by anthropogenic suspended solids, such as particles released by municipal WWTP (Kannan et al. 2003b). Though Ferguson et al. (2001b) showed that NP with long ethoxylate chains were present in coastal sediments, they also revealed that NP, NP_1EO, and NP_2EO accounted for more than 90 % of all measured compounds ($NP_{1-15}EO$). This predominance of short NPE may stem from the anaerobic conditions present in sediment, with the exception of the first few millimeters (Gejlsbjerg et al. 2003). Anaerobic biodegradation leads to the breakdown of long NPE and, consequently, to the production of short NPE such as NP_2EO and NP_1EO along with NP (John and White 1998). These latter are persistent in sediments since they are poorly degraded under anaerobic conditions. Finally, as previously stated, NP is biodegraded, thus resulting in the formation of an alcohol and hydroquinone, only under aerobic conditions (Giger et al. 2009). Furthermore, Ying et al. (2003) demonstrated that, in marine sediments and after 70 days of anaerobic treatment, no NP degradation was observed.

Soils

A few measurements of APE in soils have been undertaken (see Table 11; nine references available); these have mainly concerned agricultural and urban soils. NP levels ranged from 0.01 to 9.00 mg/kg dw (median, 1.62 mg/kg dw) for both types of soils. The degradation and persistence of NP in soils depend on factors such as oxygen availability and microbial activity. Some studies have observed a rapid degradation of NP in soils (Hesselsoe et al. 2001; Kirchmann et al. 1991). Biodegradation, however, is not the only pathway for eliminating NP in soils. Other processes, such as volatilization and sorption, may affect NP concentrations and occurrence rates (Soares et al. 2008; Trocme et al. 1988). Interestingly and according to the available literature on soils (Table 11), similar contents for agricultural and urban soils have indeed been observed. This homogeneity is quite surprising. Nonetheless, a direct comparison of soil contamination is rather difficult and subtle, since

Table 10 APE in sediment contents (in milligrams per kilogram dw)

Location	Type	NP	NP$_1$EO	NP$_2$EO	NP$_1$EC	NP$_2$EC	Reference
Germany	River		0.80	0.70			Marcomini et al. (1987)
Spain	River		2.40	1.20			Grifoll et al. (1990)
Italy	Lagoon		<loq–0.08	<loq–0.02			Marcomini et al. (1990)
Switzerland	River		0.10–8.85	<loq–2.72			Ahel et al. (1991)
USA	River	<loq–2.96					Naylor et al. (1992)
Canada	Lake	0.17–72.0	<loq–3.80				Bennie et al. (1997)
USA	Lake	<loq–37.0					Bennett and Metcalfe (1998)
UK	River–estuary	0.10–15.0					Blackburn et al. (1999)
UK	Estuary	0.03–9.05	0.12–3.97				Lye et al. (1999)
South Korea	Bay	0.11–3.89					Khim et al. (1999)
Japan	Bay	<loq–5.54					Yamashita et al. (2000)
Japan	Bay	0.03–13.0					Isobe et al. (2001)
USA	Bay	<loq–13.7	2.12	1.02			Ferguson et al. (2001b)
France	River	0.02–3.00					Fernet et al. (2003)
USA	River	<loq–1.08	<loq–0.32	<loq–0.20			Ferguson et al. (2001a)
Italy	River	0.04–0.60					Guidotti (2004)
Japan	River	0.05–0.80	<loq–2.60				Mibu et al. (2004)
Netherlands	River	<loq–3.80	<loq–2.80				Vethaak et al. (2005)
Spain	Estuary	0.14–1.10	<loq–1.51				Cortazar et al. (2005)
Japan	Bay	<loq–4.56					Hashimoto et al. (2005)
France	Coastal area	0.05–15.0					Quenea and Abarnou (2005)
China	River	0.06–0.57					Chen et al. (2005)
Spain	River	0.54					Lacorte et al. (2006)
South Korea	Bay	0.09					Koh et al. (2006)
Japan	Bay	0.06–0.56					Chen et al. (2006)
China	Coast	0.29–1.21					Chen et al. (2006)
Taiwan	River	0.13–0.19					Cheng et al. (2006)
China	Bay	0.08–0.70					Peng et al. (2007)
Japan	Bay	<loq–20.7					Kurihara et al. (2007)
USA	River	<loq–1.75	<loq–1.75	<loq–0.69			Mayer et al. (2007)
France	River	1.67	0.23	0.17	0.20		Cailleaud et al. (2007)
South Korea	Bay	0.11–0.47					Moon et al. (2009)
China	River	0.34–1.03	0.42–2.38	0.53–6.17	0.13–0.95	1.61–2.23	Yu et al. (2009)
China	Sea	<loq–1.00					Wang et al. (2010)
Spain	River	<loq–0.08		<loq–0.03	<loq–0.24	<loq–0.53	Brix et al. (2010)
China	River delta	0.11–16.2					Gong et al. (2011)
Min		0.02	0.01				$n=35$
Max		72.0	11.7				
Med		0.62	0.66				

various processes may be occurring. The differences or homogeneity observed can in fact reflect different inputs and sources or else different dynamic pollutants in the soils.

Urban soils are usually directly contaminated by industries or detergent use (by households or municipal services) (Weltin et al. 2002), whereas agricultural soils are mainly contaminated by sewage sludge amendment (Andreu et al. 2007). When applied to soils, sewage sludge tends to form heterogeneous aggregates with less pronounced aerobic conditions, and moreover, degradation is recognized as being slow (Hesselsoe et al. 2001). Soils may also become less aerobic following intense application of sewage sludge due to increased biological oxygen demand and reduced oxygen diffusion (Sjöström et al. 2008). Sludge layer depth, oxygen availability, and contaminant bioavailability are, therefore, of major importance to NP degradation in soils (Soares et al.

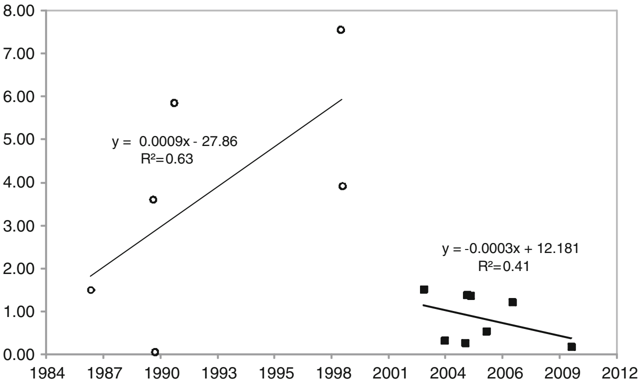


Fig. 8 Historical trend lines for the median total APE (NP + NPE) contents in European sediments (in milligrams per kilogram dw). The *dots* represent samples collected before the implementation of the WFD, whereas the *squares* depict the samples collected after its implementation

2008). Lastly, when NPE were applied to soils, a significant amount of these compounds was transformed into NP, thus resulting in an increase of NP abundance (Fig. 4) in soils and, as a consequence, of the NP proportion capable of reaching aquifers (Montgomery-Brown and Reinhard 2003). While land application is a major route in the environmental fate of sewage sludge (Table 11), a decrease in APE contents appears between 1989 and 2008 in European countries (Fig. 9). The most likely cause of this decrease would be the strengthening of European regulations. The policy applicable to sewage sludge in agriculture (EU 1986, 1991) calls for promoting its use, provided compliance with all pertinent requirements in terms of monitoring, treatment, and quality.

Discussion and recommendations for further study

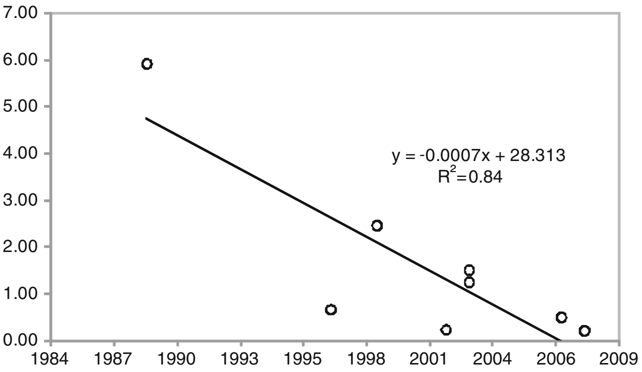


Fig. 9 Historical trend line for total median APE (NP + NPE) contents in European soils (in milligrams per kilogram)

It is now obvious from the levels observed in the various environmental compartments that APE are ubiquitous environmental pollutants. Their concentrations in most environmental compartments, however, remain location-dependent. This review has emphasized some disparities, especially between Europe, North America, and China. Furthermore, more specific data for most congeners are needed in upstream compartments to enable source elucidation. It is naturally important to measure all industrially significant congeners across all samples.

In Europe and North America, namely, Canada, regulations banning products containing NP have taken effect since 2000. It is, therefore, relevant to evaluate the effectiveness of these regulations by means of reliable and accurate measurements of the APE contamination level for sediments and surface water as a function of time. Recent observations in Europe have shown that both APE concentrations and contents have not been increasing; moreover, a decreasing trend has actually been observed over just the last few years. Nevertheless, it is important to track these changes closely. In fact, indications already point to declining APE levels in Switzerland's Glatt River over the past few decades as a result of the regulations now being implemented in this country. As previously noted, however,

Table 11 APE in soil contents (in milligrams per kilogram dw)

Location	Type	NP	NP_1EO	NP_2EO	NP_1EC	NP_2EC	Reference
Germany	Agricultural soil	4.70	1.20				Marcomini et al. (1989)
Denmark		0.30–1.00					Torslov et al. (1997)
Denmark		0.01	0.01				Jensen and Krogh (1997)
Denmark	Amended soil	<loq–4.90					Vikelsoe et al. (1999)
Germany	Urban soil	0.22					Weltin et al. (2002)
Sweden	Urban soil	0.05–9.00					Sternbeck et al. (2003)
Denmark	Urban soil	1.40–1.60					Falkenberg et al. (2003)
Spain	Amended soil	0.14–0.50	<loq–0.33				Andreu et al. (2007)
UK	Amended soil	0.18–0.23					Sjöström et al. (2008)
Min		0.01	0.01				*n*=9
Max		9.00	1.20				
Med		1.62	0.46				

sediment behaves like a major sink for APE in the environment. In contrast, concentrations in the environment and contents measured in Chinese sediments are lower than those reported in Europe, yet the trend lines are moving in opposite directions. Recent measurements in Chinese sediments have indicated increasing NP content. Finally, even if trends show decreases across industrialized countries, especially in Europe and North America, the levels being recovered are still significant and add to the background level. This latter point highlights the potential existence of diffuse contamination sources that have not yet been well identified, perhaps as the result of enhanced NP availability from sediments and deposition from the atmosphere.

The mechanisms moving these compounds between the environmental compartments where they are eventually released cannot be easily distinguished; possibilities include particle sedimentation, sorption to biomass, volatilization, and biodegradation. The latter mechanism may be important in explaining the distribution changes throughout the water cycle from wastewater to the natural environment. Research on these mechanisms, particularly the processes acting in a WWTP, is required. Even though the portion of APE sorbed to sludge and biodegraded by microorganisms is known with accuracy, it would still be useful to quantify how much is being volatilized into the atmospheric compartment and how much is adsorbed to biomass.

The latest research on surface water and WWTP has shown high efficiency of the processes acting on sewage treatment plants as well as an insignificant contribution from discharges (Bergé et al. 2012; Cladière et al. 2010; Gilbert et al. 2011; Martin-Ruel et al. 2010; Vega-Morales et al. 2010). The removal of NP and NPE from wastewater can, however, be improved by adding tertiary treatment such as UV, ozonation, and activated carbon filters to existing processes, despite these processes being expensive and failing to resolve the main problem (i.e., NP and NPE accumulation in sludge and, more generally, in solid matrices) (Johnson et al. 2005; Johnson and Sumpter 2001; Spengler et al. 2001). Sludge removal is a worldwide issue with a wide range of disposal routes depending on the type of sludge. Contaminated sludge spread on agricultural soils can raise several serious agroecosystem disturbances, such as leaching into aquifers.

A critical component dictating the fate of contaminants in urban areas is their release into the environment during storms. During wet-weather periods, the operations of WWTP are modified and may influence the quality of effluent discharged into the environment (Gilbert et al. 2011). During rainfall events, the resuspension of in-sewer deposits provides for additional pollution at the WWTP; this additional pollution can then increase the load of alkylphenols and disrupt typical plant operations.

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