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Review

Occurrence, fate and effects of Di (2-ethylhexyl) phthalate in wastewater treatment plants: A review

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ABSTRACT

Phthalates, such as Di (2-ethylhexyl) Phthalate (DEHP) are compounds extensively used as plasticizer for long time around the world. Due to the extensive usage, DEHP is found in many surface waters (0.013–18.5 μg/L), wastewaters (0.716–122 μg/L), landfill leachate (88–460 μg/L), sludge (12–1250 mg/kg), soil (2–10 mg/kg). DEHP is persistent in the environment and the toxicity of the byproducts resulting from the degradation of DEHP sometime exacerbates the parent compound toxicity. Water/Wastewater treatment processes might play a key role in delivering safe, reliable supplies of water to households, industry and in safeguarding the quality of water in rivers, lakes and aquifers. This review addresses state of knowledge concerning the worldwide production, occurrence, fate and effects of DEHP in the environment. Moreover, the fate and behavior of DEHP in various treatment processes, including biological, physicochemical and advanced processes are reviewed and comparison (qualitative and quantitative) has been done between the processes. The trends and perspectives for treatment of wastewaters contaminated by DEHP are also analyzed in this review.

1. Introduction

In recent years, the worldwide increase of urbanization and industrialization has led to the release of some complex and toxic organic compounds into the environment. The presence of the contaminants called “emerging” contaminants (ECs), such as phthalates, has been reported in many countries (Melo-Guimarães et al., 2013). Di (2-ethylhexyl) phthalate (DEHP) is a typical phthalate, widely used in industry for the production of polymeric products (97% of DEHP), such as polyvinylchloride (PVC) (Kroschwitz and Howe-Grant, 1999; Nalli et al., 2006; Yuwatini et al., 2013) and non-polymeric products, such as solvents in adhesives, inks, cosmetics, munitions, perfumes, paints, additives in hair-sprays and insect repellents, and lubricating oil (Huang et al., 2008a). DEHP is a colorless chemical substance, oily liquid, with vapor pressure of 3.4×10^{-5} at 20 °C and 4.7×10^{-4} Pa at 40 °C. Its Henry's law constant is about 4.43 Pa m^3/mol at 25 °C (Magdouli et al., 2013), and its solubility in water fluctuate between 0.6 and 1300 μg/L (20–25 °C). Many studies demonstrated that DEHP has adverse effects in environment, even at low concentrations (Clausen et al., 2004; Staples et al., 1997; Shi et al., 2012). Nowadays, environmental protection agencies of various countries such as, Canada, USA, and EU (European union) have classified DEHP as priority substance and restrict its use in products in direct contact with humans such as toys, domestic teethers, food wraps, medical devices (Wilkinson and Lamb, 1999). Sewage effluents and sludge have been identified as major routes that bring DEHP into the environment. Several papers analyzed physicochemical properties, degradation pathway, and behavior of DEHP in environmental media (Cousins et al., 2003; Liang et al., 2008; Magdouli et al., 2013; Peterson and Staples, 2003; Staples et al., 1997) or discussed environmental toxicity of DEHP (Kamrin, 2009). However, according to the knowledge of the authors, critical review of the treatment behavior of DEHP in wastewater treatment plants (WWTPs), and quantitative and qualitative comparison of wastewater treatment processes performance in removing DEHP was never performed. This paper gives an overview of the use and release of DEHP into the environment and their adverse effects. Moreover, the fate and behavior of DEHP in various wastewater treatment processes, including biological, physicochemical, conventional and advanced processes are also reviewed. Comparison (qualitative and quantitative) is carried out between the processes and the trends and perspectives for treatment of wastewaters contaminated by DEHP are also analyzed in this review.

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Nomenclature

ATSDR	Agency for Toxic Substances and Disease Registry
CNEM	China National Environmental Monitoring
DBP	Di-n-butyl phthalate
DEHP	Di (2-ethylhexyl) phthalate
DEP	Diethyl phthalate
EFGR	European Union Food Grade Regulation
GFIDMD	German Federal Institute for Drugs and Medical Devices
KEMI	Swedish Chemicals Inspectorate
MDR	Multidrug Resistance Gene
MEHP	Mono-(2-ethylhexyl) phthalate
OSHA	Occupational Safety and Health Administration
PAEs	phthalate acid esters
RBC	Rotating Biological Contactor
DHHS	US Department of Health and Human Services
USEPA	US Environmental Protection Agency
WWTPs	Wastewater Treatment Plants

2. Worldwide usage of DEHP

DEHP is extensively used as a plasticizer in polymeric products. The content of DEHP in PVC fluctuates between 10 and 40%, depending on the type of products (Horn et al., 2004; Lassen et al., 2009). Industrial hoses may contain 10–15% of DEHP by weight, while some types of flexible PVC films contain more than 30–40% of DEHP (Kroschwitz and Howe-Grant, 1999; Mersiowsky et al., 2001). Table 1 presents the production or usage of DEHP around the world. EU is the most significant worldwide producer (46%) and consumer (37%) of plasticizers. In China, DEHP was accounted for 80% of PAEs production (Meng et al., 2014). From 80's to beginning of 21th century, the worldwide production of phthalates, including DEHP increased from 2 to 5.5 million tons (Liang et al., 2008). Nowadays, the worldwide production of DEHP is decreasing (2 million tons per year) (Chan et al., 2007a; Koch et al., 2003). In Western Europe, the production of DEHP dropped from 595,000 tons to 221,000 tons between 1997 and 2004 (Roslev et al., 2007). The decrease of DEHP production is mainly related to the strict regulation against DEHP utilization as a plasticizer.

3. Occurrence of DEHP in the environment

DEHP has been found in many environmental matrices, such as surface waters, groundwater, drinking waters, wastewaters, landfill leachate, sludge, soil, and sediments. Table 2 presents DEHP concentrations detected around the world. Indoor and outdoor end products (such as, PVC) are important sources of DEHP pollution in water and soil. DEHP can be primarily released from indoor and outdoor products to the soil compartments, and a fraction may ultimately be distributed to the air or surface water compartments. According to Marttinen et al. (2003a), articles containing phthalate-plasticized material (such as, DEHP) might lose about 1% of their phthalate content by year when in contact with liquids. Mersiowsky (2002) estimated that around 5% of DEHP are being released from PVC flooring material every year. During the whole life cycle of products containing DEHP, around 72% of DEHP is released into the soil, 21% ended up in water, and the rest 7% is evaporated into the air (Rank, 2005). Moreover, DEHP is released into the environment during its production, transportation, manufacturing, and improper disposal (De Moura Carrara et al., 2011; Leah, 1977). Leah (1977) estimated that about 2–4.5% of the total Canadian supplies of phthalates were lost into the environment during production and processing.

3.1. DHEP in surface water, groundwater, drinking water, wastewater and landfill leachate

The concentrations of DEHP in surface water range from undetected to 97.8 μg/L (Table 2). For instance, the maximum concentration reported by Yuan et al. (2002) in Taiwan rivers was 18.5 μg/L. In Germany, the maximum DEHP concentration detected in surface water was 97.8 μg/L (Fromme et al., 2002). DEHP is also present in groundwater, at concentrations ranging from undetected to 5.661 μg/L. The maximum concentration of 5.661 μg/L was found in Spain (López-Roldán et al., 2004). The distribution of DEHP in groundwater is affected by the surface water concentration (Zhang et al., 2009). DEHP has been detected in drinking water around the world (Germany and Poland 0.05–0.06 μg/L, Greece 0.93 μg/L, USA 0.55 μg/L; China 3.47 μg/L) (Psillakis and Kalogerakis, 2003; Kavlock et al., 2006; Huerta-Fontela and Ventura, 2008; Liu et al., 2013). The highest concentration was detected in China. The concentrations of DEHP in wastewater are relatively higher, ranging between 0.716 and 400 μg/L (Lin et al., 2009; Mersiowsky, 2002). According to our knowledge, the maximum concentration detected in municipal wastewater was 400 μg/L (Taiwan) (Cheng et al., 2010). In landfill leachates, the concentrations of DEHP fluctuate between 88 and 460 μg/L (Jonsson et al., 2003). Atmospheric deposition also brings DEHP in environment (Kavlock et al., 2006). In Canada, around 48 tons/year of DEHP were deposited in the Great Lakes only by atmospheric deposition (Eisenreich et al., 1981). DEHP concentrations in aquatic media can vary from one country to another. This could be mainly related to the intensity of usage of DEHP in the countries, and the existing regulations. In some countries like China, it has been reported illegal addition of DEHP to clouding agents in foods and beverages (Yen et al., 2011). This can contribute to increase the concentration of DEHP in environmental media. Moreover, the high concentrations of DEHP detected in some wastewaters could be linked to the fact that in many countries, industrial and municipal wastewaters, as well as landfill leachate are mixed in sewage networks. Treatment at source of some effluents before releasing in the sewage networks could be an interesting option.

Table 1
Worldwide production and/or usage of phtalates and DEHP.

Countries	Year	Production and/or usage	References
European Union	–	Phtalates: 1 000 000 tons/year	Lin et al., 2009
		DEHP: 500 000 tons/year	Magdouli et al., 2013
Western Europe	1997 2004	DEHP: 595 000 tons DEHP: 221 000 tons	Roslev et al., 2007
Canada	1991	DEHP: 10 000 tons PVC: 49%	Minister of Supply and Services Canada, 1994
Germany	–	PAEs: 240 000 tons/year DEHP: 144 000/year	Kurane, 1997 Magnusalnervik et al., 1997 Nakamiya et al., 2005
Japan	–	PAEs: 340 000 tons/year DEHP: 204 000 tons/year	Kurane, 1997 Magnusalnervik et al., 1997 Nakamiya et al., 2005
Sweden	1989	DEHP: 2.4 kg/person Population: ≈8 400 000	Ejlertsson et al., 1996
USA	2006	DEHP: 45 000–230 000 tons	Tecnon Orbichem, 2007
China	2006–2007	DEHP: 305 000–340 000 tons	Tecnon Orbichem, 2007

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