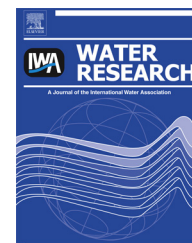


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Fate of 1,4-dioxane in the aquatic environment: From sewage to drinking water

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ABSTRACT

Potential health effects of 1,4-dioxane and the limited data on its occurrence in the water cycle command for more research. In the current study, mobility and persistence of 1,4-dioxane in the sewage-, surface-, and drinking water was investigated. The occurrence of 1,4-dioxane was determined in wastewater samples from four domestic sewage treatment plants (STP). The influent and effluent samples were collected during weekly campaigns. The average influent concentrations in all four plants ranged from $262 \pm 32 \text{ ng L}^{-1}$ to $834 \pm 480 \text{ ng L}^{-1}$, whereas the average effluent concentrations were between $267 \pm 35 \text{ ng L}^{-1}$ and $62,260 \pm 36,000 \text{ ng L}^{-1}$. No removal of 1,4-dioxane during water treatment was observed. Owing to its strong internal chemical bonding, 1,4-dioxane is considered non-biodegradable under conventional bio-treatment technologies. The source of increased 1,4-dioxane concentrations in the effluents was identified to originate from impurities in the methanol used in the postanoxic denitrification process in one of the STPs. In view of poor biodegradation in STPs, surface water samples were collected to establish an extent of 1,4-dioxane pollution. Spatial and temporal distribution of 1,4-dioxane in the Rivers Main, Rhine, and Oder was examined. Concentrations reaching 2200 ng L^{-1} in the Oder River, and 860 ng L^{-1} in both Main and Rhine River were detected. The average monthly load of 1,4-dioxane in the Rhine River was calculated to equal to 172 kg d^{-1} . In all rivers, concentration of 1,4-dioxane increased with distance from the spring and was found to negatively correlate with the discharge of the river. Additionally, bank filtration and drinking water samples from two drinking water facilities were analyzed for the presence of 1,4-dioxane. The raw water contained 650 ng L^{-1} – 670 ng L^{-1} of 1,4-dioxane, whereas the concentration in the drinking water fell only to 600 ng L^{-1} and 490 ng L^{-1} , respectively. Neither of the purification processes employed was able to reduce the presence of 1,4-dioxane below the precautionary guideline limit of 100 ng L^{-1} set by the German Federal Environmental Agency.

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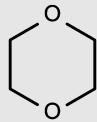
1. Introduction

1,4-Dioxane is a polar cyclic diether, commonly used as an industrial solvent in the production of adhesives, paint strippers, dyes, degreasers, fabric cleaners, paper, electronics, and pharmaceuticals (Tanabe et al., 2006). In the past, 1,4-dioxane was mainly associated with its use as a solvent stabilizer especially for 1,1,1-trichloroethane (1,1,1-TCA). This application was discontinued in the 1990s, when 1,1,1-TCA was banned by the Montreal Protocol, due to its ozone depleting properties. Moreover, 1,4-dioxane is unintentionally formed during several chemical processes used to produce soaps, polyester, and plastics. According to the European Chemical Substances Information System, 1,4-dioxane is a high production volume chemical, meaning production exceeds 1000 tons per year in at least one member country. The European Union Risk Assessment Report from 2002, states that the only production site in Europe; BASF SE in Ludwigshafen, Germany produced 2000–2500 tons of 1,4-dioxane in 1997. Currently, European Chemicals Agency lists three registrants/suppliers in Europe: BASF SE and Merck KGaA in Germany and Sustainability Support Services (Europe) AB in Sweden with a total amount of 1,4-dioxane exceeding 100 tonnes per year.

In recent years international concern has risen about the ubiquitous presence of 1,4-dioxane in the environment and the adverse health effects to its exposure. United States Environmental Protection Agency (USEPA) and the International Agency for Research on Cancer assigned 1,4-dioxane to group B2 as a possible human carcinogen. Toxicological studies revealed an increased incidence of nasal cavity and liver carcinomas in rats, liver carcinomas in mice, and gall bladder carcinomas in guinea pigs (Zenker et al., 2003). Since then, a number of international regulatory guidelines emerged for 1,4-dioxane. World Health Organization suggested a 50 $\mu\text{g L}^{-1}$ drinking water threshold value for 1,4-dioxane, whereas the USEPA National Center for Environmental Assessment proposed a health-based advisory level of 3 $\mu\text{g L}^{-1}$ in the tap water (Mohr, 2010). According to the USEPA Integrated Risk Information System, cancer development could occur in 1 out of 1,000,000 people exposed to a concentration of 350 ng L^{-1} in drinking water over a lifetime. As a result, Unregulated Contaminant Monitoring Regulation from 2012, proposed a minimum reporting level for 1,4-dioxane at 70 ng L^{-1} . In its Drinking Water Regulation (2001), the German Federal Environmental Agency suggested a precautionary guideline limit for weak or non genotoxic compounds such as 1,4-dioxane at 100 ng L^{-1} in drinking water.

The physiochemical properties of 1,4-dioxane govern its high mobility and persistence in the environment (Table 1). Based on Henry's law constant ($4.88 \times 10^{-6} \text{ atm} \times \text{m}^3 \times \text{mol}^{-1}$) and the indefinite solubility in water, volatilization of 1,4-dioxane from water is expected to occur slowly (NICNAS, 1998). The low octanol water coefficient ($\log K_{oc} = -0.27$) is in accordance with its high mobility in soil and leaching to groundwater as well as no significant adsorbance to suspended sediments. The moderate vapor pressure and Henry's law constant implies slow volatilization from moist soil; however fast volatilization from dry soils. When 1,4-dioxane enters the atmosphere it is subjected to photo oxidation

Table 1 – Physico-chemical properties and structure of 1,4-dioxane.

Property	Value	Reference	Structure
Molecular weight (g/mol)	88.1	Budavari et al. (1989)	
Density (g/cm^3)	1.033	Keith and Waters (1985)	
Boiling point ($^{\circ}\text{C}$ at 760 mmHg)	101.1	Verschueren (1983)	
Water solubility (at 25 $^{\circ}\text{C}$; g/L)	Miscible	Budavari et al. (1989)	
Vapor pressure (mmHg at 20 $^{\circ}\text{C}$)	30	Verschueren (1983)	
Henry's law constant ($\text{atm} \times \text{m}^3/\text{mol}^{-1}$)	4.88×10^{-6}	Howard (1990)	
Partition coefficient ($\text{Log } P_{ow}$)	-0.27	Howard (1990)	
$\log P_{oc}$	0.54	Howard (1990)	

with hydroxyl radical ($^{\bullet}\text{OH}$) with a half-life of less than 7 h (NICNAS, 1998). Based on the aforementioned properties, removal of 1,4-dioxane from wastewater is expected to be difficult, increasing the possibility of surface, groundwater, as well as drinking water contamination.

Data on the present dispersion of 1,4-dioxane in the environment in Europe and around the world are not readily available. Already, three decades ago 1,4-dioxane was first found as a water contaminant in the US (Kraybill, 1978; Burmaster, 1982; Hartung, 1989). Most recent work focussed on distribution of 1,4-dioxane in polluted ground waters (Isacson et al., 2006; Chiang et al., 2008). Also, in Canada, groundwater contamination with the ether was documented (EC, 2002; Lesage et al., 1990). In Europe, the presence of 1,4-dioxane was confirmed in surface waters in Germany, Netherlands, and in the United Kingdom (EC, 2002; Gelman Sciences, 1989). Moreover, 1,4-dioxane was detected in municipal landfill leachate in Sweden as well as in the industrial wastewater from polyester resin producing company (Paxéus, 2000; Romero et al., 1998). Extensive research on the distribution and occurrence of 1,4-dioxane has been carried out in Japan. Numerous studies confirmed the presence of 1,4-dioxane in landfill leachate (Yasuhara and Tanaka, 2003; Fujiwara et al., 2008), effluents from sewage plants (Abe, 1999; Tanabe et al., 2006), surface and groundwater (Abe, 1999; Kawata et al., 2003; Kawata and Tanabe, 2009). Other studies focused on the investigation of 1,4-dioxane in non-ionic surfactants and cosmetics. Fuh et al. (2005) determined that 22% of daily use cosmetics in Taiwan contained between 4.2 ppm and 41.1 ppm of 1,4-dioxane. Black et al. (2001) reported up to 1410 ppm of 1,4-dioxane in ethoxylated raw materials and up to 279 ppm in cosmetic finished products. Numerous studies on the presence of 1,4-dioxane in cosmetic products have been conducted by independent consumer organizations in both US and Europe.

In the presented study, the occurrence and distribution of 1,4-dioxane in the aquatic environment in Germany and Poland is investigated. The focus of the current study was to examine the mobility of 1,4-dioxane from wastewater to surface water, bank filtered groundwater, and finally to drinking

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