The Use of an Ultrasonic Field in Support of Classical Methods of Oxidising Component Leached from Microplastics in Bottom Sediments

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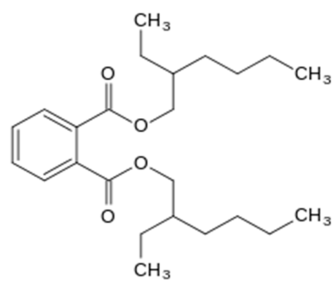
Table S1. World production and consumption of DEHP

Location	Year	Production/use thousand tons/year	Literature
EU	–	500	Magdouli et al., 2013
Western Europe	1997	595	Roslev et al., 2007
	1999	476	
	2004	221	
Canada	1991	10	Minister of Supply and Services Canada, 1994
Japan	–	204	Magnusalnervik et al., 1997
Germany	–	144	Magnusalnervik et al., 1997
USA	2006	45-230	Tecnon Orbichem, 2007
China	2006–2007	305-340	Tecnon Orbichem, 2007

Table S2. Physicochemical properties of DEHP [Peijnenburg and Struijs, 2006; Net et al., 2015]

Physicochemical properties	*Unit*	*DEHP*	*Structural formula*
Chemical formula		$C_{24}H_{38}O_4$	
Molecular weight	g/mol	390.54	
Vapor pressure at 200°C	hPa	1.6	
Vapor pressure at temperature 20°C	Pa	$3.4 \cdot 10^{-5}$	
Water solubility	mg/dm³	$2.49 \cdot 10^{-3}$	
Color		colorless	
Physical state at temperature 20°C		oily liquid	
Log K_{OW}		7.73	
Log K_{OA}		10.53	
Log K_{AW}		–2.8	
K_{OC}	dm³/kg	87 420–$51 \cdot 10^{4}$	
pKa (strongly alkaline environment)		–6.7	
H	Pa·m³/mol	4.43	
Density	g/cm³	0.98	
Boiling point	°C	385	
Melting temperature	°C	–55	
Flash-point	°C	193	
Temperature of self-ignition	°C	400	



K_{OA}–octanol-air partition, K_{OW}–octanol–water partitioning, K_{AW}–air–water partitioning, K_{OC}–organic carbon partitioning, pK_a–dissociation constant, H–Henry's constant.

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Table S3. Occurrence of di(2-ethylhexyl) phthalate in various environmental components

Matrix	Type	Location	DEHP [ng/dm³]				Literature
			min	*max*	*average*	*median*	
Drinking water	tap water	Spain	-	-	445	-	Blanchard et al., 2013
		China	-	-	12, 487	-	Tang et al., 2012
		France	-	-	350	-	Blanchard et al., 2013
	bottled water	Spain	ND	1520	-	-	Guart et al., 2014
		Hungary	16	1700	-	-	Keresztes et al., 2013
		Canada	360	460	-	-	Cao et al., 2008
		Egypt	ND	298	104	67	Zaki et al.,
Underground water		Spain	-	5661	-	-	López-Roldán et al., 2004
Rain		Netherlands	0.69	1.7	-	-	Vethaak et al.., 2005
Air		Shanghai, China	0.150	0.368	-	-	Wang et al., 2006
		Zhejiang, China	0.097	0.301	-	-	Wang et al., 2006
		Guangdong, China	0.112	0.250	-	-	Wang et al., 2006

ND – no data

Table S4. Occurrence of di(2-ethylhexyl) phthalate in surface water

Type	Location	DEHP [μg/dm³]				Literature
		min	*max*	*average*	*median*	
River	Veldwachters, South Africa	80	431	-	-	Olujimi et al., 2012
	Klang, Malaysia	3.1	64.3	-	-	Tan, 1995
	Yangtze River, China	12.07	2127	-	-	Zhang et al., 2018
	Rhône, France	0.04	0.407	-	-	Paluselli et al., 2017
	Somme, France	5.16	20.76	10.23	8.88	Net et al., 2014
	Jiulong, China	0.62	12.43	-	-	Li et al., 2017
	Yangtze River, China	0.01	54.7	-	-	Wang et al., 2008a
	Dam, Japan	ND	3.60	-	-	Suzuki et al., 2001

	Dommel, the Netherlands	0.9	5	-	3.2	Vethaak et al., 2005
	Kaweri, India	ND	0.822	-	-	Selvaraj et al., 2015
	Ebro, Muga, Fluvia, Ter, Besos, Llobregat, Spain	0.12	4.98	1.19	-	Sanchez-Avila et al., 2012
	Ebro, Spain	0.136	32.86	2.28	-	Catalan Water Agency, 2011
	Wangyang, China	0.26	0.94	0.48	0.43	Zhang et al., 2015
	Rivers of the Eastern Cape, South Africa	0.06	2306	-	-	Fatoki andNoma, 2002
	Rhine, Germany	25	36	-	-	Fromme et al., 2002
	Supraśl, Polska	-	8.67	1.04	-	GIOŚ, 2016
	Horodnianka, Poland	-	1.63	0.81	-	GIOŚ, 2016
		-	-	1.78	-	GIOŚ, 2016
	Czarna, Poland	-	0.66	0.24	-	GIOŚ, 2016
	Liza, Poland	-	0.98	0.23	-	GIOŚ, 2016
	Mała Panew, Poland	-	0.40	-	-	GIOŚ, 2016
	Łukawica, Poland	-	-	0.3	-	GIOŚ, 2016
	Biała, Poland	-	-	3.22	-	GIOŚ, 2016
Lake	15 lakes in the city of Guangzhou, China	0.237	1.60	-	-	Zeng et al., 2008a
	Tuczno, Poland	-	-	0.29	-	GIOŚ, 2013
		-	-	0.65	-	GIOŚ, 2014
	Kłeckie, Poland	-	-	0.65	-	GIOŚ, 2014
	Skąpe, Poland	-	-	0.405	-	GIOŚ, 2014

ND – no data

Table S5. Occurrence of di(2-ethylhexyl) phthalate in bottom sediments

Type	Location	DEHP [mg/kg d.w.]				Literature
		min	*max*	*average*	*median*	
River	Meuse, Rhine, Scheldt, the Netherlands	0.123	7.6	0.6	-	Vethaak et al., 2005
	Wangyang, China	0.161	0.465	0.307	0.304	Zhang et al., 2015
	Yellow River, China	6.35	259	-	45.7	Sha et al., 2007
	Klang, Malaysia	0.49	15.0	-	1.63	Tan, 1995
	Houjing, Taiwan	0.10	20.22	7.0	-	Lin et al., 2009
	Yangtze River, China	13.9	322	-	144	Wang et al., 2008b
	Pearl River, China	0.415	29.5	-	1.46	Liu et al., 2014
	Ogun, Nigeria	0.02	0.82	0.172	0.05	Adeniyi et al., 2011
	Trent, UK	0.84	31.0	-	-	Long et al., 1998
	Aire, UK	7.89	115.2	26.63	-	Long et al., 1998

	Love, Salt, Canon, Taiwan	0.40	34.8	5.02	-	Chen et al., 2013
	Jiulong, China	0.053	1.28	0.35	0.19	Li et al., 2017
	Odra, Poland	-	-	1.19	-	GIOŚ, 2016
	Parsęta, Poland	-	-	1.94	-	GIOŚ, 2016
Lake	Wu, China	-	-	202.3	-	Wang et al., 2008b
	East, China	-	-	323.5	-	Wang et al., 2008b
	Wielkie Dąbie, Poland	-	-	4.17	-	GIOŚ, 2016
				<0.1		GIOŚ, 2013
	Lubinieckie, Poland	-	-	1.20	-	GIOŚ, 2015

GIOŚ - Chief Inspectorate for Environmental Protection in Poland

Table S6. Occurrence of di (2-ethylhexyl) phthalate in soil

Type	Location	DEHP [mg/kg d.w.]				Literature
		min	*max*	*average*	*median*	
Area agricultural	Guangzhou, China	0.180	29.37	4.09	0.308	Zeng et al., 2008b
	Spain	1	63	-	-	Plaza-Bolanos et al., 2012
	Shijiazhuang, China	0.066	0.263	0.143	0.132	Zhang et al., 2015
	Nanjing, China	0.240	4.18	0.184	0.194	Wang et al., 2015
	Yangtze River Delta, China	ND	9.19	0.546	0.349	Sun et al., 2013
	Amursko-Sungarian Plain, China	0.077	0.583	0.222	0.203	Wang et al., 2017
	Xinjiang, China	104	149	128.7	-	Guo and Wu, 2011
	France	-	-	0.242	-	Tran et al., 2015
	Czech Republic	0.16	1.02	-	-	Dankova et al., 2012
	Roskilde, Denmark	0.012	1.9	-	-	Vikelsøe et al., 2002
	UK	0.022	0.075	-	-	Gibson et al., 2005
	China	ND	6.22	0.821	0.562	Niu et al., 2014
Urban area	Guangzhou, China	1.41	264	63	31.9	Zeng et al., 2009
	Tianjin, China	0.028	4.17	0.618	0.099	Kong et al., 2012
	Scotland, Great Britain	0.025	1.596	0.162	0.110	Rhind et al., 2013
Recreation area	Novi Sad, Serbia	0.164	2.04	0.844	-	Škrbić et al., 2016
	Tianjin, China	0.043	0.801	0.179	0.125	Zhao et al., 2018
	Guangzhou, China	0.892	154	29.4	10.4	Zeng et al., 2009

ND–no data

Cavitation bubbles appear as a result of local disruption of the medium under the influence of high tensile forces. During the operation of the ultrasonic field, cavitation bubbles increase their radius twice or more. Subsequently, they collapse rapidly, resulting in the release of a large amount of energy, resulting in a local increase in temperature and pressure. A sharp increase in pressure inside the bubble causes them to crack and the formation of a hydrodynamic shock wave. Extreme conditions are the reason for the thermal breakdown of water molecules, during which hydroxyl radicals are formed, which are the main source of so-called sonochemical reactions (reactions 1–13). This phenomenon significantly accelerates the course of chemical and physical reactions [Miłowska, 2007; Bogacki and Naumczyk, 2009].

$$H_2O +))) \longrightarrow HO\cdot + H\cdot \quad (1)$$

$$O_2 +))) \longrightarrow 2O\cdot \quad (2)$$

$$HO\cdot + O\cdot \longrightarrow HOO\cdot \quad (3)$$

$$O\cdot + H_2O\cdot \longrightarrow 2HO\cdot \quad (4)$$

$$H\cdot + O_2\cdot \longrightarrow HOO\cdot \quad (5)$$

$$OH\cdot + H\cdot \longrightarrow H_2O\cdot \quad (6)$$

$$2HO\cdot \longrightarrow H_2O\cdot + O\cdot \quad (7)$$

$$HOO\cdot + HO\cdot \longrightarrow O_2 + H_2O \quad (8)$$

$$2HO\cdot \longrightarrow H_2O_2 \quad (9)$$

$$2HOO\cdot \longrightarrow H_2O_2 + O_2 \quad (10)$$

$$H\cdot + H_2O_2 \longrightarrow HO\cdot + H_2O \quad (11)$$

$$HO\cdot + H_2O_2 \longrightarrow HOO\cdot + H_2O \quad (12)$$

$$2H\cdot \longrightarrow H_2 \quad (13)$$

Fenton's reaction is a complex process that involves many side reactions. In soil, bottom sediments, these reactions include basic radical initiation (reactions (14) and (15)), propagation (reactions (16) and (17)) and termination (reactions (18)–(20)), as well as reactions with intermediates (reactions (21)–(28)), with carbonate and bicarbonate ions (reactions (29)–(34)) and with natural organic matter (NOM) (reactions (35)) [Yap et al., 2011 ; Gan and Ng, 2012; Cheng et al., 2016].

$$H_2O_2 + Fe^{2+} \longrightarrow Fe^{3+} + HO\cdot + OH^- \quad (14)$$

$$Fe^{3+} + H_2O_2 \longrightarrow Fe^{2+} + HO_2\cdot + H^+ \quad (15)$$

$$HO\cdot + RH \longrightarrow R\cdot + H_2O \quad (16)$$

$$R\cdot + H_2O_2 \longrightarrow ROH + HO\cdot \quad (17)$$

$$HO\cdot + Fe^{2+} \longrightarrow Fe^{3+} + OH^- \quad (18)$$

$$HO\cdot + H_2O_2 \longrightarrow H_2O + HO_2\cdot \quad (19)$$

$$HO\cdot + HO\cdot \longrightarrow H_2O_2 \quad (20)$$

$$Fe^{2+} + HO_2\cdot \longrightarrow Fe^{3+} + HO_2^- \quad (21)$$

$$Fe^{3+} + HO_2\cdot \longrightarrow Fe^{2+} + O_2 + 2H^+ \quad (22)$$

$$HO_2\cdot + H_2O_2 \longrightarrow H_2O + OH\cdot + O_2 \quad (23)$$

$$HO_2\cdot + HO_2\cdot \longrightarrow H_2O + O_2 \quad (24)$$

$$O_2\cdot^- + Fe^{3+} \longrightarrow Fe^{2+} + O_2 \quad (25)$$

$$O_2\cdot^- + Fe^{2+} + 2H^+ \longrightarrow Fe^{3+} + H_2O_2 \quad (26)$$

$$HO\cdot_2 + O_2\cdot^- + H_2O \longrightarrow H_2O_2 + O_2 + OH^- \quad (27)$$

$$HO\cdot + HO_2\cdot \longrightarrow H_2O + O_2 \quad (28)$$

$$CO_2(g) + H_2O \longleftrightarrow H_2CO_3(aq) \quad (29)$$

$$H_2CO_3(aq) \longleftrightarrow HCO_3^- + H^+ \quad (30)$$

$$HCO_3^- + OH\cdot \longrightarrow CO_3^- + OH^- \quad (31)$$

$$CO_3^- + H_2O_2 \longrightarrow HO_2\cdot + HCO_3^- \quad (32)$$

$$2CO_3^- \longrightarrow \text{products} \quad (33)$$

$$CaCO_3(s) + H^+ \longleftrightarrow Ca^{2+} + HCO_3^- \quad (34)$$

$$NOM + HO\cdot \longrightarrow \text{products} \quad (35)$$

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