



Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

The occurrence and source identification of bisphenol compounds in wastewaters

Marjeta Česen^{a,b}, Kaja Lenarčič^c, Vesna Mislej^d, Meta Levstek^e, Ana Kovačič^{a,b}, Bernardka Cimrmančič^f, Nataša Uranjek^g, Tina Kosjek^{a,b}, David Heath^b, Marija Sollner Dolenc^c, Ester Heath^{a,b,*}

^a Department of Environmental Sciences, Jožef Stefan Institute, Jamova cesta 39, 1000 Ljubljana, Slovenia

^b Jožef Stefan International Postgraduate School, Jamova cesta 39, 1000 Ljubljana, Slovenia

^c Department of Pharmaceutical Chemistry, Faculty of Pharmacy, University of Ljubljana, Aškerčeva cesta 7, 1000 Ljubljana, Slovenia

^d JP VODOVOD-KANALIZACIJA d.o.o., Central Wastewater treatment plant Ljubljana, Vodovodna c. 90, 1000 Ljubljana, Slovenia

^e Wastewater Treatment Plant Domžale-Kamnik, Študljanska cesta 91, 1230 Domžale, Slovenia

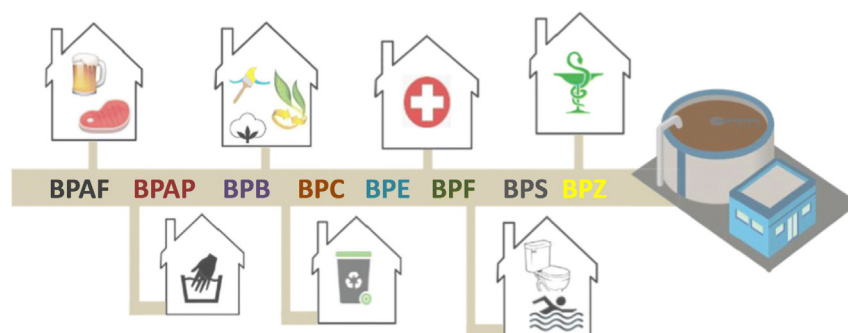
^f Wastewater Treatment Plant Novo mesto, Šmarješka cesta 10, Novo Mesto, Slovenia

^g Wastewater Treatment Plant Šaleška dolina, Primorska cesta 8, Šoštanj, Slovenia

HIGHLIGHTS

- BPs were measured in different WWs including inflows, influents and effluents.
- Most BPs were >LOD in analysed WW.
- BP removal efficiencies were in general >96%.
- BPZ was most commonly detected and BPC was quantified in WW for the first time.
- Food processing and textile cleaning facilities represent major sources of BPs.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 30 August 2017

Received in revised form 23 October 2017

Accepted 24 October 2017

Available online xxxxx

Editor: D. Barcelo

Keywords:

Bisphenols

Wastewater

Wastewater treatment plant

Source identification

Removal efficiency

ABSTRACT

This study reports the occurrence of eight bisphenols (BPs): bisphenol AF (BPAF), bisphenol AP (BPAP), bisphenol B (BPB), bisphenol C (BPC), bisphenol E (BPE), bisphenol F (BPF), bisphenol S (BPS) and bisphenol Z (BPZ) in wastewaters (WWs). Sample preparation involved pre-concentration with SPE cartridges (Oasis HLB), followed by derivatization using *N*-(*tert*-butyldimethylsilyl)-*N*-methyltrifluoroacetamide with 1% *tert*-butyldimethylchlorosilane. Chemical analysis was based on gas chromatography–mass spectrometry. A validated method with limits of detection (LODs) at ng L⁻¹ range was applied to WWs collected at five Slovene wastewater treatment plants (WWTPs) and WW inflows from industrial, commercial and residential sources entering the sewerage systems of two catchments (Domžale-Kamnik (DK) and Ljubljana (LJ)). The presence of all BPs was confirmed in three inflows in DK and two inflows in the LJ catchments. High cumulative concentrations of all BPs were determined in WW from food processing facilities (LJ: 3030 ng L⁻¹ and DK: 599 ng L⁻¹). A high detection frequency was observed in the WW from two textile cleaning companies (6 BPs for LJ and 8 BPs for DK). The analysis of WW from WWTPs revealed that only BPF (36.7 ng L⁻¹) and BPS (40.6 ng L⁻¹) were >LODs in the influents, whereas other BPs were detected also in the effluents. BPZ was found in the highest concentration (403 ng L⁻¹ at WWTP-DK). WW collected at this WWTP also contained the highest amount of BPE (238 ng L⁻¹). Although BPs removal could not be directly compared between the WWTPs, with the exception

* Corresponding author at: Jožef Stefan Institute, Jamova 39, 1000 Ljubljana, Slovenia.

E-mail address: ester.heath@ijs.si (E. Heath).

<https://doi.org/10.1016/j.scitotenv.2017.10.252>

0048-9697/© 2017 The Author(s). Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

Please cite this article as: Česen, M., et al., The occurrence and source identification of bisphenol compounds in wastewaters, Sci Total Environ (2017), <https://doi.org/10.1016/j.scitotenv.2017.10.252>

of BPAP and BPB in the case of two smaller WWTPs (6.39%–43.2%) bisphenols were in general highly removed ($\geq 96.2\%$). Finally, levels of BPC > LOD are reported for first time (WWTP in the DK catchment: 1.01 ng L^{-1} – 11.8 ng L^{-1} ; LJ inflow from food processing plant up to 2560 ng L^{-1}).

© 2017 The Author(s). Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

1. Introduction

Bisphenol A (BPA) is a monomer used in the production of polycarbonates and epoxy resins commonly applied in food contact materials, digital media (CDs and DVDs), electronic equipment, medical devices, dental fillings, thermal receipts, water pipes and toys, etc. (Chen et al., 2016). The potential adverse effects on human health and regulation of BPA production led to the use of structurally related BPA alternatives (BPs) (Belfroid et al., 2002; Goldinger et al., 2015; Usman and Ahmad, 2016).

Data on the physico-chemical properties of BPs is limited (see Supplementary Information 1; SI-1; Table S2), but still could be used to explain their environmental fate. Based on their Henry's Law constants (K_H), their presence in the gas phase is negligible (Chen et al., 2016). The octanol-water partition coefficient ($\log K_{ow}$) of BPs is from 1.25–7.17. Bisphenols with $\log K_{ow} \leq 4$, such as bisphenol B (BPB), bisphenol E (BPE), bisphenol F (BPF) and bisphenol S (BPS) are expected mostly in the water phase. Other BPs such as bisphenol AF (BPAF), bisphenol AP (BPAP), bisphenol C (BPC), bisphenol P (BPP) and bisphenol Z (BPZ) have $\log K_{ow}$ of 4–6 and have the potential to adsorb to soil and sediment and to accumulate into the fat tissues of living organisms (Caballero-Casero et al., 2016; Chen et al., 2016).

A review by Jiang et al. (2013) reports numerous studies on the global occurrence of BPA in surface waters (SWs) and wastewaters (WWs), whereas there is only limited information available about other BP compounds. For example, only five published studies describe the presence of BPAF, BPAP, BPC, BPB, BPF, BPFL, BPS, BPZ, tetrachlorobisphenol A (TCBPA) and tetrabromobisphenol A in SWs in China and BPS and BPF in SWs from Japan, Korea and India (Jin and Zhu, 2016; Liu et al., 2016; Song et al., 2012; Yamazaki et al., 2015; Yang et al., 2014). Also, WW as a matrix is poorly investigated in terms of the presence of BPs. BPAF, BPB, BPE, BPF, BPS and BPZ were targeted in WW at wastewater treatment plants (WWTPs) in China, BPAF, BPAP, BPB, BPF, BPP, BPS and BPZ in WWs at WWTPs in India and chlorobisphenol A (CBPA), dichlorobisphenol A (DCBPA), trichlorobisphenol A (TrCBPA) and TCBPA in WW effluents in Spain (Ballesteros et al., 2006; Karthikraj and Kannan, 2017; Sun et al., 2017). To our knowledge, Ballesteros et al. (2006) is the only published study addressing the presence of chlorinated BPs in Europe, however their limits of quantification (LOQs) were so high (100 – 250 ng L^{-1}), that the reported values of all BPs were <LOQs. In addition to this study, a local screening project conducted in Norway was also reported, where the occurrence of BPA, BPAF, two structural isomers of BPF (2,2- and 4,4-dihydroxy bisphenols), BPS and bisphenol BP (BPBP) were determined in WWTP effluents, leachates, sediments and biota from Oslofjord and Mjøsa (Thomas et al., 2014).

Besides the occurrence of BPs in the WWTP influents and effluents, it is also important to assign point sources that contribute to the overall contamination with these compounds. Sun et al. (2017) reported BPAF and BPE as having low removal during conventional WW treatment. In such cases, it seems reasonable to search for the main inputs of contamination, which can be achieved by the collection and analysis of WWs originating from various facilities (e.g. inflows from industrial facilities and hospitals) entering the sewerage system. The aim of this study was to assess the occurrence and removal of BPAF, BPAP, BPF, BPE, BPB, BPC, BPS and BPZ in WWs collected at five Slovene WWTPs with varying treatment technologies. In addition, we attempted to identify possible sources of target BPs in two different WWTP catchments in Slovenia: Ljubljana (LJ) and Domžale-Kamnik (DK).

2. Materials and methods

2.1. Materials

BPAF (>99%), BPAP (>99%), BPC (>99%), BPE (>98%), BPF (>98%), BPS (>98%) and BPZ (>98%) were obtained from Sigma-Aldrich (St. Louis, USA), while BPB (>99.8%) was purchased from Dr Ehrenstorfer (Augsburg, Germany). Structural formulas, CAS numbers and structural names are presented in SI-1; Table S1. BPF refers to 4,4-dihydroxy substituted BP. Deuterated bisphenol A (BPA-d16), purchased from Sigma Aldrich (USA), was used as a surrogate standard. The silylating agent *N*-(*tert*-butyldiethylsilyl)-*N*-methyltrifluoroacetamide with 1% *tert*-butyldimethylchlorosilane (MTBSTFA with 1% TBDMCS; 95%) was purchased from Sigma Aldrich (St. Louis, USA). Dichloromethane (DCM), ethyl acetate (EtAc), methanol (MeOH) and purified water were obtained from J. T. Baker (Deventer, Netherlands) and were all of analytical grade purity. Stock solutions ($\approx 10 \mu\text{g L}^{-1}$) of each compound were prepared in MeOH, while calibration standards were prepared through serial dilutions of the stock solutions.

2.2. Sampling

Eighteen samples of WW influents and effluents ($V = 2 \text{ L}$; $n = 1$) were collected at five WWTPs during August and October 2015 (Table 1). At the WWTP Domžale-Kamnik (WWTP-DK), an additional four samples containing WW influent mixed with the WW from cistern trucks from surrounding industries were collected. Full details are provided in with additional information given in SI-2 (Tables S4–S5).

In addition, twenty-five samples ($V = 2 \text{ L}$; $n = 1$) of various potential source WWs were also sampled, i.e. the inflows of various industrial and other facilities entering into the sewerage system of the LJ and DK treatment plant catchments (SI-2; Fig. S1 and Table S3). The identity of these sources cannot be disclosed, therefore only the type of WW is shown (Table 2). These sources included a pre-treated WW (e.g. neutralisation, settlement, chemical precipitation) from two food and beverage producers (F), various industrial sources (I), health centres, hospitals and retirement homes (H), pharmaceutical companies (P), textile cleaning companies (C), landfill sites (L), swimming pool and communal sewerage canals (other sources, O; SI-2; Fig. S1 and Table S3). All samples were stored at -20°C prior to analysis. Additional sample information is given in SI-2; Table S3.

2.3. Sample preparation

Samples were first filtered through glass-microfibre filters (Macherey Nagel, Düren, Germany) and then through cellulose nitrate filters ($0.45 \mu\text{m}$, Sartorius Stedim Biotech, Göttingen, Germany). A surrogate standard was then added to give a final concentration of 25 ng L^{-1} and the samples extracted using solid-phase extraction (SPE) cartridges with divinylbenzene-*N*-vinylpyrrolidone copolymer sorbent (Oasis HLB Waters, Massachusetts, USA; 60 mg, 3 mL). The cartridges were conditioned with 3 mL of DCM, EtAc and MeOH and equilibrated with 3 mL of purified water. The samples were then extracted at a flow-rate of 3 mL min^{-1} using a Supelco Vacuum Manifold (Bellefonte, USA). After loading, the sorbents were dried under vacuum (-1.33 kPa , 30 min). Elution was performed with 3 mL of MeOH (3 aliquots of 1 mL). The remaining solvent was removed under a gentle stream of nitrogen. Derivatization was performed with $30 \mu\text{L}$ of MTBSTFA with 1% TBDMCS in $220 \mu\text{L}$ EtAc for 16 h at 60°C .

Download English Version:

<https://daneshyari.com/en/article/8862482>

Download Persian Version:

<https://daneshyari.com/article/8862482>

[Daneshyari.com](https://daneshyari.com)