



## Sandy beaches as hotspots of bisphenol A

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### ABSTRACT

Bisphenol A (BPA) poses a serious environmental threat and health concern. This study presents the global monitoring of BPA on oceanic sandy beaches. According to monitoring results, many beach sands contain a harmful concentrations of BPA. Likewise, styrene oligomers (SOs), anthropogenic chemicals derived from polystyrene plastics, show similar concentrations as BPA. This study shows a strong, positive correlation between BPA and SOs. The results indicate that probably BPA-containing materials including micro- and nano-plastics can be an important source of BPA to the sand beaches. Therefore, BPA presents potential health risks to people spending considerable time on the beach.

### 1. Introduction

Currently, bisphenol A (BPA) constitutes some of the most widely used and applied material. Since its development as a synthetic estrogen in the 1890s (vom Saal and Hughes, 2005; Rochester, 2013), BPA has become one of the most highly produced chemicals (Flint et al., 2012; Staples et al., 2018) and is an important commercial component of consumer products, such as can liners, plastic bottles, food containers, and thermal papers (Geens et al., 2012; European Parliament, 2019). However, BPA in the environment has been shown to be an endocrine disruptor over the last 30 years (Kang et al., 2006; Crain et al., 2007; Vandenberg et al., 2009; Zalko et al., 2011).

Reports of the ill effects of BPA vary among studies. BPA is typically considered to have adverse human health effects as an endocrine disruptor and is also associated with environmental pollution in ecosystems (Kang et al., 2006; Crain et al., 2007; Vandenberg et al., 2009; Zalko et al., 2011; Dodson et al., 2012; Flint et al., 2012; Geens et al., 2012; Rochester, 2013; Staples et al., 2018; European Parliament, 2019). For these reasons, BPA has been previously measured in water, soil, air, seawater, sewage sludge, foodstuff, and sediment (Kang et al., 2006; Fu and Kawamura, 2010; Zalko et al., 2011; Flint et al., 2012; Careghini et al., 2015; Giulivo et al., 2016; Staples et al., 2018). Specifically, biomonitoring studies have reported the global ubiquity of BPA at parts

per billion (ppb) levels in human tissue and fluids, including urine, blood, and other bodily fluids (Vandenberg et al., 2010; TOXNET, 2019). Concerns have been raised over long-term exposure to BPA with a focus on the potential risks to human health and the environment (TOXNET, 2019). However, the potential harm from BPA has emerged as a contentious issue that has led to disagreements regarding its low-dose effects and exposure period (Vandenberg et al., 2009). As such, environmental monitoring has gained importance in determining the environmental fate of BPA and risk assessment. Therefore, BPA has recently become the subject of major monitoring efforts (Kang et al., 2006; Careghini et al., 2015; Staples et al., 2018).

In general, human exposure to endocrine disruptors occurs through skin, mouth, and through inhalation, and in the case of BPA, human exposure occurs mainly through the diet (Vandenberg et al., 2009; Geens et al., 2012). However, data on the contribution of the skin and oral routes are lacking (Dodson et al., 2012). Furthermore, BPA can be readily absorbed by the skin in human (Zalko et al., 2011). In particular, the contribution of non-dietary sources of BPA exposure, such as BPA transport through human skin that may occur on sandy beaches, is not well understood. There is a lack of monitoring data regarding BPA in sandy beaches where all or part of human skin may be exposed.

In particular, BPA has attracted the attention of regulatory agencies across the globe due to its toxicity and widespread use. The U.S. Food

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and Drug Administration (FDA) no longer approves the use of BPA in baby bottles, sippy cups, and baby formula packaging (TOXNET, 2019). The European Chemicals Agency (ECHA) reclassified BPA as a chemical of very high concern due to its endocrine disruptive properties (ECHA, 2017). In addition, in December 2014, the European Food Safety Authority (EFSA) reduced the tolerable daily intake (TDI) for BPA from 50 to 4  $\mu\text{g kg}^{-1}$  per bodyweight day (Ćwiek-Ludwicka, 2015). Thus, legal measures have been taken to further strengthen the BPA exposure limits.

This study is the first report on the global BPA concentrations in sand samples from beaches around the world. In order to investigate the BPA concentrations in the oceanic beaches, we collected sand and seawater samples from oceanic sandy beaches around the world. In this study, a correlation analysis was performed to understand the source of BPA.

## 2. Experimental

### 2.1. Sampling and sample preparation

We have already conducted an extensive study to develop sampling and sample preparation procedures to identify and measure the concentrations of BPA including styrene oligomers (SOs) found in regional seawater and sand samples (Kwon et al., 2014 & 2015). SOs concentrations from the same sampling sites were obtained from previous study (Kwon et al., 2015), and compared to the BPA concentrations obtained in order to determine the correlation between these two associated contaminants.

In this study, seawater and sand samples were collected from selected coastal regions, which included 19 nations, 26 sampling sites (Table 1). Each sample consisted of about 100 g of beach sand by wet weight and about 2 L of seawater. The sites range from England (study No. 1 in Table 1) to Costa Rica (study No. 26). The sampling locations are detailed further in Supplementary information Figure S1 (Kwon et al., 2014 & 2015).

Further details on the sampling methods in the field and sample preparation in the laboratory have been provided in previous studies (Kwon et al., 2014 & 2015). Briefly, almost all sampling was accessible

by walking. In each coastal region, seawater and sand samples were collected at depths of approximately 30 cm from the surface water and surface sand, respectively. The seawater sample in the field was subjected to cotton plug filtration and immediately extracted four times, with a total of 100 mL dichloromethane by shaking for 10 min. Sand sample was stored in a glass container and delivered in the laboratory. After dehydration by freeze-drying overnight in the laboratory, the sand sample was accurately weighed to 5.0 g using a balance and spiked with surrogate prior to extraction. Other procedures for sampling and preparation used in this study are described in details in the previous studies (Kwon et al., 2014 & 2015).

### 2.2. Analytical methods and QA/QC

BPA (>99%) was obtained from Daejung Chemicals & Metals Co., LTD. (Siheung-si, Gyeonggi-do, Korea). The measurement of BPA is based upon gas chromatography/mass spectrometry using HP 6890 GC with a JEOL Auto MS-II equipped with a 30 m  $\times$  0.32 mm i. d. (0.25  $\mu\text{m}$  film thickness) DB-1 capillary column (J&W Scientific, Folsom, CA), with which it was possible to identify and measure the concentrations of BPA and SOs. Further details of the analytical methods have been provided in previous studies (Kwon et al., 2014 & 2015).

Data quality assurance and quality control included standard solution injection every 20 sample in order to monitor changes in the sensitivity of the instrument. BPA recoveries were tested for artificial seawater and sand media based on triplicate analysis of matrix spiked and extracted with the same analytical procedure reported in the previous studies (Kwon et al., 2014 & 2015). The recoveries of BPA on the real sand and seawater samples were 82–120%. No equipment or material composed of plastic materials such as polystyrene (PS) plastic were adopted in this analysis.

### 2.3. Physicochemical property estimation of BPA

To investigate the source of BPA, this study examined its physicochemical properties, which used the computational method based on

**Table 1**

Concentrations of bisphenol-A (BPA) found in beach sand and seawater samples and styrene oligomers (SOs) found in beach sand samples collected from beaches around the world. Detailed sampling locations are shown in Fig. S1 (Kwon et al., 2015).

No.	Country	Sample numbers	BPA concentration in sand, $\mu\text{g kg}^{-1}$	Standard deviation ( $\pm$ ) for BPA in sand, $\mu\text{g kg}^{-1}$	BPA in seawater, $\mu\text{g L}^{-1}$	SOs on sand, $\mu\text{g kg}^{-1}$ (Kwon et al., 2015)
1	England	4	159.1	97.0	1.2	389.6
2	Portugal	2	577.8		0.9	274.2
3	Spain	4	668.5	543.0	1.2	1,143.3
4	Tunisia	5	1,097.2	697.0	29.8	92.0
5	Slovenia	1	22.0		1.1	30.8
6	Italy	8	23.2	11.0	0.7	37.5
7	Croatia	1	305.0		0.2	15.4
8	Greece	9	215,133.3	192,987.0	1.0	31,400.0
9	India	3	316.4	17.2	3.9	940.4
10	Sri Lanka	2	277.4		6.0	237.6
11	Malaysia	4	15,708.3	12,498.0	2.1	18,924.8
12	Philippines	9	3,275.0	2,739.0	31.8	153.2
13	Taiwan	19	2,878.4	1,789.0	359.0	150.4
14	China	6	1,935.6	1,304.0	49.6	1,408.2
15	Korea	8	83.7	35.6	72.9	57.9
16	Okinawa (JPN)	14	94.2	39.0	1.3	69.2
17	Honshu (JPN)	34	318.1	149.0	25.3	335.3
18	Hokkaido (JPN)	11	260.2	197.0	61.0	100.0
19	Guam (USA)	5	4,489.4	2,798.0	57.2	1,402.7
20	Washington (USA)	5	439.1	312.0	10.3	505.8
21	San Francisco (USA)	9	653.0	319.0	9.5	816.1
22	Los Angeles (USA)	5	423.6	318.9	4.6	2,906.8
23	Massachusetts (USA)	1	909.1		2.4	7,368.3
24	Florida (USA)	4	44,688.5	29,874.0	0.4	23,507.0
25	Puerto Rico (USA)	20	514.6	297.0	102.9	352.0
26	Costa Rica	5	75,168.8	22,589.0	17.9	26,277.4

Estimation Programs Interface (EPI) Suite version 4.11. More detailed EPI methods are presented in Table 2.

### 3. Results and discussion

BPA appears to be ubiquitous in sandy beaches around the world (Table 1). In this study, sandy beaches are a newly identified reservoir of BPA chemicals, and the concentrations of BPA in the sand samples are significantly higher than those in the seawater samples. Table 1 shows the BPA concentrations in sand and seawater samples collected from beaches around the world. The results show that the BPA concentrations are typically much higher in sand than in the neighboring seawater. The BPA concentration in the beach sand samples range from 22 to 215,133  $\mu\text{g kg}^{-1}$ , and had a mean concentration of  $4,247 \pm 12,839 \mu\text{g kg}^{-1}$ , with high variability. The BPA concentrations in Greece are high. In contrast, the mean BPA concentration in seawater is relatively low at  $32.9 \pm 71.9 \mu\text{g L}^{-1}$  compared to the concentrations of BPA in the sand samples. In particular, the BPA concentrations in sandy samples collected from beaches around the world are significantly higher than be reported from the previous monitoring results (details will be described later).

As shown in Table 1, the sandy beaches of Greece have the highest concentrations of BPA at  $215,133 \mu\text{g kg}^{-1}$ , whereas those of Slovenia and Italy have relatively low levels. In these sandy beaches, converging sand surfaces trap and retain BPA chemicals at levels up to  $10^2$  ppm. The concentrations of BPA in the sandy beach areas found in this study are significantly higher than previously reported monitoring data (Kang et al., 2006; Crain et al., 2007; Vandenberg et al., 2009; Fu and Kawamura, 2010; Zalko et al., 2011; Dodson et al., 2012; Flint et al., 2012; Rochester, 2013; Careghini et al., 2015; Giulivo et al., 2016; Staples et al., 2018), with the exception of specific sewage sludge (details will be described later), reported by a study carried out in a wastewater treatment plant (Flint et al., 2012). Thus, to our knowledge, to date, the levels of BPA in sandy beaches have been underestimated when compared with those in other environmental samples.

Since BPA was detected in water in the late 1990s, BPA monitoring results have been reported for multiple environmental matrices, including water, soil, sediment and air (Kang et al., 2006; Crain et al., 2007; Vandenberg et al., 2009; Zalko et al., 2011; Dodson et al., 2012; Flint et al., 2012; Geens et al., 2012; Giulivo et al., 2016; Staples et al., 2018; European Parliament, 2019). For example, in Asia, Europe, and North America, BPA concentrations in the surface waters of fresh and marine and other observed environmental samples, such as suspended

solids, soils, sediments, landfill leachate, treatment plants, biota, and wildlife, ranged from undetectable to  $805 \mu\text{g L}^{-1}$  and undetectable to  $17, 200 \mu\text{g L}^{-1}$  (ppb levels) (Flint et al., 2012; Staples et al., 2018). In addition, atmospheric concentrations of BPA were reported to range from 1 to  $17,400 \text{pg m}^{-3}$  (Fu and Kawamura, 2010).

To date, the highest level of BPA reported is  $3.2 \times 10^7 \mu\text{g kg}^{-1}$  dry weight (ppb levels) in sewage sludges from wastewater treatment plants (Flint et al., 2012). This concentration in sewage sludge is approximately 149 times higher than that in the Greek sand in this study. Unlike the highly polluted sewage sludge samples (Flint et al., 2012), BPA has been found in common soil and sediment environmental samples ranging from  $0.24$  to  $20,136 \mu\text{g kg}^{-1}$  dry weight (Careghini et al., 2015; Giulivo et al., 2016; 21) which is approximately 11 times lower than that found in the Greek sand in this study (Table 1). The level of BPA in the sandy beaches of Greece is roughly two or four orders of magnitude higher than those in common environmental soils and sediments, suggesting that the risk of human exposure of BPA in sandy beaches is much higher.

Nevertheless, with the exception of specific sludge, the source of BPA is poorly understood in sandy beaches. Well-known sources of BPA include polycarbonate (PC) plastic, epoxy resins for can coating, antioxidants for processing polyvinyl chloride (PVC) plastic, dental sealants, thermal receipts, and others (TOXNET, 2019). Primarily, BPA is used in the production of PC plastics (Staples et al., 2018). BPA leaches into water from PC plastic increasing the concentration and becoming more hydrolyzed over time with changes in water temperature and at basic pH (Sajiki and Yonekubo, 2003). However, these sources cannot significantly contaminate sandy beaches. The presence of a PC plastic on sandy beaches has never been reported, to the best of our knowledge. Hence, more research is needed to understand the sources of BPA in beach sand.

To investigate the source of BPA, this study examined its physicochemical properties (Table 2). The characteristics of BPA are a moderate water solubility ( $120\text{--}172.7 \text{mg L}^{-1}$ ), octanol-water partition coefficient ( $\log K_{ow} = 3.2\text{--}3.4$ ), and extremely low vapor pressure ( $2.27 \times 10^{-7} \text{mmHg}$ ). If released into the environment, BPA is not expected to volatilize from both wet and dry particle such as sands and is expected to have low mobility. Based on monitoring results (Table 1) and estimated physicochemical properties (Table 2), BPA is expected to be adsorbed to the sand surface, and thus to contaminate beach sands.

SOs, by-products of PS plastic that exhibit characteristics similar to the physicochemical properties of BPA (Table 2), were monitored in sand samples collected at different geographical locations throughout the world. The same sand samples used for BPA measurement were

**Table 2**  
Summary of physicochemical properties<sup>a</sup> of bisphenol A (BPA) and styrene oligomers (SOs) (Kwon et al., 2019).

Chemical Name (abb. Name)	Solubility mg/L	Vapor pressure mmHg	LogK <sub>ow</sub> <sup>b</sup>	Henry's constant atm·m <sup>3</sup> /mole	LogK <sub>oc</sub> <sup>c</sup>	LogBCF <sup>d</sup>
Bisphenol A (BPA)	120–172.7	2.27E-07	3.32–3.64	9.16E-12	3.095–4.576	1.858–2.237
Styrene monomer (SM)	160.01–343.7	5.05	2.89	2.81E-03	2.560	1.613
1,3-diphenyl propane (SD-1)	53.8	1.92E-03	3.43	0.26–0.85E-03	2.976–4.483	1.930
2,4-diphenyl-1-butene (SD-2)	0.2–0.6	0.82E-03	5.64	0.46E-03	4.697–4.895	3.389
cis-1,2-diphenyl cyclobutene (SD-3)	0.9	0.39E-03	5.46	0.46–4.95E-04	4.738–4.744	3.271
trans-1,2-diphenyl cyclobutene (SD-4)						
2,4,6-triphenyl-1-hexene (ST-1)	0.1–0.9E-03	3.34E-07	8.26	0.60E-04	6.765–7.168	3.508
1a-phenyl-4a-(1-phenylethyl)tetralin (ST-2) <sup>e</sup>	0.2–3.2E-03	1.95E-07	7.62	0.59E-04	6.613–6.773	3.821
1a-phenyl-4e-(1-phenylethyl)tetralin (ST-3) <sup>e</sup>						
1e-phenyl-4a-(1-phenylethyl)tetralin (ST-4) <sup>e</sup>						
1e-phenyl-4e-(1-phenylethyl)tetralin (ST-5) <sup>e</sup>						
1e,3e,5a-triphenylcyclohexane (ST-6) <sup>f</sup>	0.3–1.3E-03	1.25E-07	8.08	0.14–7.04E-05	6.803–7.012	3.596
1e,3e,5e-triphenylcyclohexane (ST-7) <sup>f</sup>						

<sup>a</sup> All data produced by the US EPA EPI suite program and its database (DB) (Card et al., 2017; Kwon et al., 2019).

<sup>b</sup> The differential solubility of the compound between one being 1-octanol and the other being water.

<sup>c</sup> The organic carbon adsorption in predicting the amount of a compound that is able to be absorbed by soil.

<sup>d</sup> The bioconcentration factor in predicting a compound's tendency to build up in marine organisms.

<sup>e</sup> In the EPI Suite, these compounds exhibit the same physico-chemical properties because they appear in the same simplified molecular input line entry system (SMILES) code as isomers.

<sup>f</sup> In the EPI Suite, these compounds exhibit the same physico-chemical properties because they appear in the same simplified molecular input line entry system (SMILES) code as isomers.

monitored.

The concentration ranges of SOs are summarized in Table 1, together with the BPA data. A positive correlation between BPA and SOs is evident in Fig. 1. The SOs originated from PS plastic (Kwon et al., 2014). SOs do not exist naturally in the environment but are eluted from PS plastics (Kwon et al., 2014 & 2015; Eo et al., 2018; De-la-Torre et al., 2020). Some amount of micro-PS and other micro-sized plastic particles can remain buried in beach sand for long periods (Corcoran et al., 2009; Li et al., 2016). This suggests that the source of SOs in beach sand may be significantly associated with the presence of BPA. In this study, the global levels of BPA in sandy beaches ( $14,247 \pm 12,839 \mu\text{g kg}^{-1}$ ) show very large variations, implying that BPA is sensitive to changes in environmental conditions, i.e., weather such as precipitation. Based on both monitoring results and physicochemical properties, this result suggests that BPA can leach out of BPA-containing materials into the sand, like SOs in PS plastics.

Direct sources of BPA could be PVC, polyethylene (PE), polyester, and polypropylene (PP) plastics commonly observed in sand beaches (Corcoran et al., 2009; Law and Thompson, 2014; Li et al., 2016), rather than PC plastic. BPA has been used as an antioxidant and/or as a stabilizer at amounts of 0.05–3% (weight/weight) in PE, polyester, PVC, and PP plastics (Hermabessiere et al., 2017; Li et al., 2016; TOXNET, 2019), ranging from 100 to 310,000 parts per million (ppm). BPA can leach out slowly, as most of it is not chemically bound (Hermabessiere et al., 2017). This process is well-known but has been mostly overlooked as a potential source of BPA contamination.

In particular, another possible source of BPA could be personal care and cleaning products as well as household products such as sunscreen, body lotion, soaps, and so on. Many of the contents of these consumer products are mostly 10 ppm (weight/weight) and can be a source of BPA reaching up to 100 ppm (Dodson et al., 2012). The release of BPA from plastic containers including personal care products dumped in sandy beaches can accelerate leaching through the fragmentation of larger plastics by runoff, tides, winds, catastrophic events, and human activities including recreational and tourist behavior on the sandy beaches (Corcoran et al., 2009; Hirai et al., 2011; Kwon et al., 2015; Hermabessiere et al., 2017; Pinon-Colin et al., 2018). In particular, once on the sandy beach, abandoned BPA-containing materials can be actively transported by human activities, concentrating it in beachy sand around the world. As an example of typical human activity, plastic materials containing personal care products such as sunscreens, oils, lotions, and so on, which are often used on sandy beaches, are discarded on the beach, from which high concentrations of BPA can be transferred to sand.

Furthermore, given that BPA concentrations up to  $730 \text{ ng g}^{-1}$  have been measured in plastic debris (Hirai et al., 2011), plastic debris as a vector can serve as an indirect source of BPA in sandy beaches. However, this level is low when compared to the value found in this study. Considering that BPA is used in a variety of plastic products, alternatives to BPA must be identified and used in the future.

The high BPA concentrations found in sandy beaches in this study should be a global concern. As sandy beaches usually have no direct BPA sources, such as nearby factories, the high concentrations in beach sand indicate a direct relationship between BPA concentrations and other human activities. High concentrations of BPA from land-based sources in heavily populated cities worldwide have been observed in enclosed sandy beaches in marine environments (Corcoran et al., 2009; Li et al., 2016). Therefore, beach sand serves as a hotspot for high concentrations of BPA, and stricter measures are required to prevent further accumulation of BPA in sandy beaches.

#### 4. Conclusions

This study shows that sandy beaches around the world are heavily contaminated with BPA. This is of particular interest from a public health standpoint because the high concentrations of BPA can pose a

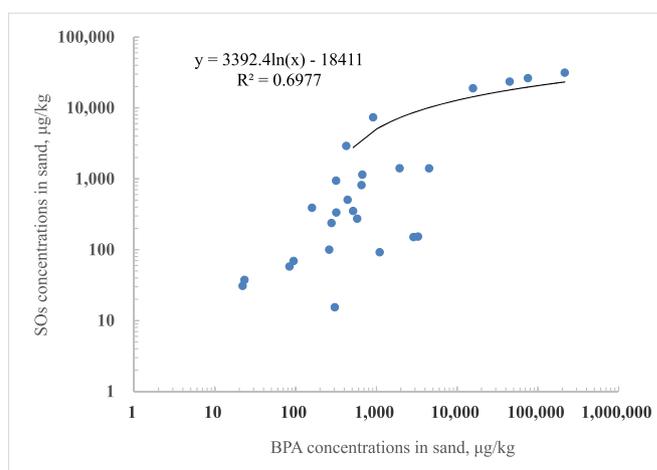


Fig. 1. Positive correlation\* between the concentrations of styrene oligomers (SOs), originated from polystyrene (PS) plastics, and bisphenol-A (BPA). All data used in Fig. 1 is presented in Table 1. In the figure, both axes are plotted on a logarithmic scale. \* The software used in this study is Microsoft® Excel® 2016 (16.0.5017.1000).

high risk to humans and ecosystems. BPA, with a relatively low molecular weight, can penetrate the skin and trans-epidermal penetration rates are high (Zalko et al., 2011). People can be exposed to BPA through skin exposure to beach sand. Therefore, at a sandy beach, BPA is one of the most common chemicals that can directly penetrate human skin.

#### Credit author statement

Bum Gun Kwon contributed the coordination of this project, total preparation and complete writing of this manuscript for submission, field sampling, and chemical analysis. K. Saïdo contributed the coordination of this project, field sampling and chemical analysis. Seon-Yong Chung contributed field sampling and discussions.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envres.2020.110175>.

## Data and materials availability

All data is available in the main text or the supplementary materials. All data, code, and materials used in the analysis must be available in some form to any researcher for purposes of reproducing or extending the analysis.

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