



Review

Occurrence of bisphenol S in the environment and implications for human exposure: A short review



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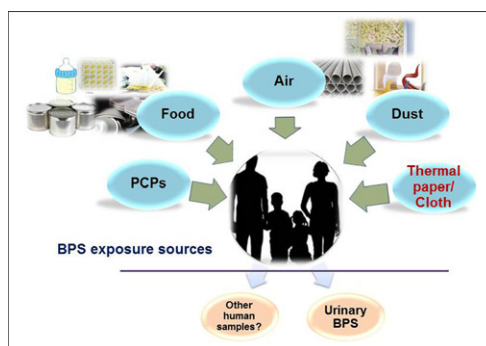
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HIGHLIGHTS

- BPA was gradually substituted by BPS in consumer products worldwide.
- The concentration ratios of BPS to BPA were higher in the aquatic environment than in other environmental media.
- Food is the dominant source for general population exposure to BPS.

GRAPHICAL ABSTRACT



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ABSTRACT

As a substitute of bisphenol A (BPA), bisphenol S (BPS) has been applied in consumer products present in our daily lives. With a similar chemical structure as BPA, BPS has also been demonstrated as an exogenous endocrine disrupting chemical. Compared with a large number of studies on BPA, investigation on BPS has remained limited. In this study, we reviewed the literature of BPS mainly published during 2010–2017, including its environmental distributions, toxicities, and human exposure. The data demonstrated that BPS is now ubiquitous in the environment and found worldwide, but generally with concentration levels lower than BPA in various environment media, including water, sediment, sludge, indoor dust and air, consumer products, and human urine. However, we found that the concentration levels of BPS in aquatic environments, especially water samples, were almost comparable or equal to that of BPA. Our summary also indicated that process speed of substituting BPA with BPS in consumer products in the U.S. was relatively faster than other countries. In addition, we summarized the toxicities of exposure to BPS both *in vivo* and *in vitro* experiments. The current data supports that exposure to BPS may have adverse effects on reproductive systems, endocrine systems, and nervous systems in animals and humans, and may trigger oxidative stress. The occurrence of BPS was frequently reported in human urine, but rarely in other human samples. The current research indicates that food is the dominant source for human exposure to BPS, and the contribution of personal care product usage is low. The occurrence of BPS and their metabolites in the human body and the guidelines for BPS exposure merit further investigation.

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1. Introduction

Many synthetic substances released into the environment were initially thought to be relatively harmless, but over time, scientists began to realize their adverse effects on the environment and human health. Some of these substances may be banned or limited, then are often replaced by other substitutes. A typical example is bisphenol A (BPA). BPA is one of the most widely used industrial additives, which is used as a monomer in the production of polycarbonate plastics, epoxy resins, and phenolic resin in the manufacture of food and beverage packaging material or containers, can linings, baby feeding bottles, toys, sealants, eyeglass lenses, several paper consumer products, and so on (Huang et al., 2012; Vandenberg et al., 2007; Žalmanová et al., 2016). The global production of BPA is about 8 million tons per year (Paul et al., 2008). Consequently, BPA is ubiquitously present in the global environment.

BPA has been demonstrated as a typical exogenous endocrine disrupting chemical (Bin Shao, 2016; Eladak et al., 2015). It may change human hormones (Huang et al., 2012) and may lead to adverse effects, especially significant negative impacts on reproductive abilities, such as female infertility (Diamanti-Kandarakis et al., 2012), male low sperm quality (Li et al., 2011), low sperm count (Meeker et al., 2010), and sex hormone concentration changes (Rochester, 2013). On October 18, 2008, Canada was the first country worldwide to view BPA as a toxic chemical and announced the prohibition of BPA usage in baby bottles, and in October 2010, Canada also restricted BPA usage in all food packaging and containers (Government of Canada, 2010). Similarly, the European Union believed that BPA in bottles can induce baby precocious puberty, and banned the BPA usage in baby bottles on March 1, 2011 (The European Commission, 2011). As a result of the restrictions on the production and use of BPA, an alternative, bisphenol S (BPS) has become one of the major substitutes for BPA added into products.

Bisphenol S, with a much similar chemical structure to that of BPA (Table 1), contains a sulfone group with strong electron-absorbing ability and two hydroxyl groups, so it is stronger than other bisphenols in term of acidity, with more stability than BPA. For example, BPS is more resistant to heat and sunlight than BPA, and the biodegradability of bisphenol analogues in seawater was ranked as Bisphenol F (BPF, another BPA alternative) > BPA > BPS (Danzi et al., 2009; Ike et al., 2006; Kuruto-Niwa et al., 2005). Since bisphenol S is often used as an intermediate for the production of epoxy resins and polycarbonate plastics, it exists in our consumer products. Therefore, humans are widely exposed to BPS, similar to BPA. The annual manufactured or imported rate of BPS was as high as 1000–10,000 tons in the European Economic Area reported by European Chemicals Agency (ECHA, 2015). As a main substitute of BPA, BPS is considered safer than BPA, which partly may be due to the

lack of sufficient data to support risk assessment, especially its toxicities at low dose exposure. Although data are limited, a few studies have documented that BPS may be equally or more harmful than BPA in some investigated endpoints (Rochester and Bolden, 2015; Žalmanová et al., 2016). For example, BPS had the same qualitative effects on estrogen receptor and androgen receptor activities in the range of BPA, but BPS exhibited the greatest changes in efficacy on 17α -hydroxyprogesterone in all tested bisphenol analogues (Rosenmai et al., 2014). Those raised the concern on the safety of BPS as a “safe substitution”.

In the present study, we review the reports of BPS mainly published after 2010, including its environmental occurrence, human exposure, and toxicities. We aim to give a comprehensive distribution of BPS in the environment, and figure out the potential sources and routes for human exposure to BPS.

2. Physicochemical properties of BPS

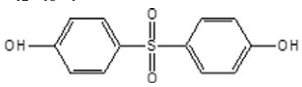
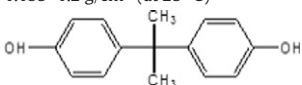
The basic physical and chemical properties of BPS, as well as BPA, are shown in Table 1. BPS is a commonly used name for 4,4'-Sulfonyldiphenol (CAS NO. 80-09-1), with chemical formula as $(\text{HOOC}_6\text{H}_4)_2\text{SO}_2$ and molar mass of 250.27 g/mol. Generally, BPS is white colorless solid under ambient conditions (density: 1.3663 g/cm^3), and is molten at $240\text{--}250 \text{ }^\circ\text{C}$. BPS is freely soluble in aliphatic hydrocarbons, soluble in ethanol, isopropyl alcohol, acetonitrile, and acetone, and slightly soluble in aromatic hydrocarbons. The reported water solubility of BPS is 1100 mg/L at $20 \text{ }^\circ\text{C}$.

The value of octanol-water partition coefficient ($\log K_{ow}$) reflects the lipid solubility of the pollutants, characterizing the ability of pollutants to migrate, transform, and distribute in the environment. Compounds with low K_{ow} values are considered to be relatively hydrophilic, so they usually have a high water solubility, and the adsorption coefficient (K_{oc} values) in soil or sediments and bioconcentration factors in aquatic organisms are correspondingly small. BPS has $\log K_{ow}$ value 1.65 (<5.0), which suggests its tendency to dissolve into water. It takes time for BPS degradation in the environment (Danzi et al., 2009; Ike et al., 2006). For example, BPS is more persistent in sediments (half-life $t_{1/2} = 135$ days) than in soil ($t_{1/2} = 30$ days) and water ($t_{1/2} = 15$ days) (Chen et al., 2016).

3. Potential sources of BPS in the environment

Bisphenol S was first synthesized as a kind of dye in 1869 and is used as the substitute of BPA introduced into consumer products in the 2000s (Glausiusz, 2014). As the usage of BPS is not regulated, it is difficult to specify the products containing and leaching BPS. Generally, BPS is

Table 1
Physical and chemical properties of BPS and BPA.

	Parameter	Value	Reference
BPS	CAS no.	080-09-1	
	Molecular weight	250.27 g/mol	
	Formula	C ₁₂ H ₁₀ O ₄ S	
	Chemical structure		
	Boiling point	240–241 °C	(Qiu, 2016)
	Melting point	245–250 °C	
		240.5	(Bjornsdotter et al., 2017)
	pKa	8	(Bjornsdotter et al., 2017)
	Density	1.366	
	Water solubility	1100 mg/L (at 20 °C)	
	pH	6.6–7.0 (100 g/L, H ₂ O, 20 °C)	
	Stability	Stable, strong oxidizing agent	
	Log Kow	1.65	(Chen et al., 2016)
		1.2	(Bjornsdotter et al., 2017)
	CF ^a	3.535 (L/kg wet weight)	(Chen et al., 2016)
	BAF ^b	3.535 (L/kg wet weight)	(Chen et al., 2016)
	Half-life (day)		(Chen et al., 2016)
	Sediment	135	
	Soil	30	
	Water	15	
BPA	CAS no.	080-05-7	
	Molecular weight	228.29 g/mol	
	Formula	C ₁₅ H ₁₆ O ₂	
	Specific gravity	1.195–1.2 g/cm ³ (at 25 °C)	
	Chemical structure		
	Boiling point	220 °C (at 4 mm Hg)	(Muhamad et al., 2016)
	Melting point	153 °C	(Muhamad et al., 2016)
		158–159 °C	(Ghazali and Wan, 2015)
	pK _a	10.1	(Muhamad et al., 2016)
	Water solubility	120 mg/L (at 25 °C)	(Zhu and Zuo, 2013)
	Vapor pressure	5.3 × 10 ⁻⁶ Pa (at 25 °C)	(Cao, 2012)
		87 Pa (at 190 °C)	(Ghazali and Wan, 2015)
	BCF	172.7 (L/kg wet weight)	(Chen et al., 2016)
	BAF	172.8 (L/kg wet weight)	(Chen et al., 2016)
	Log Kow	3.43	(Chen et al., 2016)
		3.32	(Muhamad et al., 2016)
	Half-life (day)		(Chen et al., 2016)
	Sediment	337.5	
	Soil	75.0	
	Water	37.5	

^a Bioconcentration factor.^b Bioaccumulation factor.

used as a monomer in synthetic polymers as well as in epoxy, so it is frequently found in some daily consumer products, such as plastics, food cans linings and packaging, baby bottles and toys, dental materials, personal care products (PCPs) (Liao and Kannan, 2014a), various papers (Liao et al., 2012b), and so on. For example, BPS was the main analogue added into thermal papers as a color developer to replace BPA. In addition, the recycling of paper and plastic, especially thermal papers, is a special source of BPS in related consumer products.

As BPS is easy to phase-out in consumer products, all products mentioned above might be the potential sources for BPS in the environment. The “leached” BPS could enter and degrade in air, particle/dust, soil, water, sediment, biota, and foodstuffs, which causes the BPS pollution in the environment, and finally induces human exposure.

4. Occurrence of bisphenol S in the environment

By far, BPS was ubiquitous in environment. The occurrence of BPS was reported in indoor dust (Liao et al., 2012c; Wang et al., 2015), fluvial water (Yang et al., 2014a), surface water (Yamazaki et al., 2015), sewage sludge (Lee et al., 2015; Song et al., 2014; Yu et al., 2015), and marine sediment (Liao et al., 2012d; Yang et al., 2014a). Furthermore, BPS was also found in human urine (Asimakopoulos et al., 2016; Liao et al.,

2012a; Xue et al., 2015; Yang et al., 2014c), generally with concentration levels lower than, but in the same order of magnitude as that of BPA. We did not find much BPS data regarding other human matrices, such as breast milk, placental tissues, fetal plasma, amniotic fluid, semen plasma, or saliva. We summarize the results of these studies of BPS in the following sections for each environmental medium.

4.1. BPS in water, sediment, and sludge

As sediment and water are identified as important reservoirs of contaminants, monitoring BPS occurrence in those samples is important to track the current and historical usage of BPS, and evaluate and predict the effects of BPS to the surrounding environment. A group of studies have determined BPS levels in surface water and sediment, as well as sewage sludge worldwide (Tables 2 and 3).

Several studies of BPS occurrence in aquatic environments both for water and sediment were conducted in China (Jin and Zhu, 2016; Yang et al., 2014a). As the concentration of BPS was generally low in environment, Yang et al. (2014a, 2014b, 2014c) had developed a sensitivity method to simultaneously analyze BPS and other analogues by liquid chromatography-electrospray tandem mass spectrometry (LC-MS/MS) (Yang et al., 2014a). With that method, they reported the concentrations

of BPS were 0.29 to 19.0 ng/L and 0.12–0.22 ng/g dry weight (dw) in river water and their sediment, respectively, in Hangzhou Bay, Zhejiang Province. Recently, another study investigated several BPA analogues in 46 water and 42 sediment samples from Liaohe River Basin (including Liaohe River and Hunhe River) and Taihu (Jin and Zhu, 2016). BPS was found in all water samples, contributing second to the total analogues amount, just next to BPA and BPF. The concentrations of BPS were 0.28–67, 0.22–52, and 0.61–46 ng/L in waters of Taihu Lake, Liaohe River, and Hunhe River, respectively. For sediment, concentrations of BPS were lower than those of BPA and BPF. For the first time, the field sediment-water partitioning coefficients ($\log K_{oc}$) were estimated based on concentrations of bisphenol analogues in those aquatic environment samples, and the respective values of BPS, BPA, and BPF were 3.5, 3.8, and 4.7 in Taihu Lake. For Liaohe River Basin, the partitioning coefficient was only calculated for BPA, and was 3.0 and 3.1 in Liaohe River and Hunhe River, respectively. This study offered an important parameter, $\log K_{oc}$, to understand the fate of BPS in the aquatic environment.

In 2016, bisphenol analogues were determined again in water and sediment samples of Taihu Lake (Liu et al., 2017b). As shown in Tables 2 and 3, the concentrations of BPS, BPA, and BPF were all significantly increased in both water and sediment compared with those in 2013 (Jin and Zhu, 2016), especially for sediment samples. The concentrations of target contaminants were approximately one order of magnitude higher than previous studies. For example, the median concentration of BPS was 2.0 ng/L and 0.071 ng/g dw in water and sediment of Taihu Lake in 2013, and the values increased to 6.4 ng/L and 0.57 ng/g dw in 2016, respectively. In addition, BPA was the dominant compound among all target bisphenols in samples collected in 2013, but BPF was the main congener in samples collected in 2016. The time trend of concentrations and profiles of bisphenol analogues in Taihu Lake indicated that BPA was quickly replaced by BPS and BPF in the productions in the research area in China.

In the global range, BPS was reported ubiquitous in aquatic environments, such as water and sediments, as well as wastewater sludge. In 2015, the occurrence of BPS was determined in surface waters sampled from four Asian countries: China, India, Japan, and Korea (Yamazaki et al., 2015). In this study, a higher level of BPS in water was found in India (26.5 ng/L, median) than the other three countries (not detected (ND) to 3.4 ng/L, median), indicating the high use of BPS in the sampling area in India. Another comprehensive study determined that BPS and other BPA analogues were present in sediments collected from the rivers near the major industrial areas of the United States (U.S.), Japan, and South Korea (Liao et al., 2012d). BPS was detected in 28.5% of 172 sediment samples, and the average and the highest concentration was 12.4 and 1970 ng/g (dw), respectively. In addition, because wastewater

treatment plants may be significant sources of BPS discharge to the environment, BPS in sewage sludge was determined in 76 sludge samples from 74 sewage treatment plants collected by the U.S. Environmental Protection Agency (EPA) in 2006–2007 (Yu et al., 2015). The concentration levels of BPS in sludge ranged from <1.79 to 1480 ng/g dw, detectable in 84% of the total samples (Yu et al., 2015). Similar studies of sludge were also conducted in Korea and China (Lee et al., 2015; Song et al., 2014). The compositions of bisphenol analogues were significantly different in sludge from China and Korea. The median concentration of BPS (4.34 ng/g dw) was on the same order of magnitude of those of BPA (9.38 ng/g) and BPF (1.97 ng/g) in sludge from China (Song et al., 2014). However, the level of BPS (3.80 ng/g) was two orders of magnitude lower than those of BPA (275 ng/g) and BPF (249 ng/g) in sludge samples from Korea (Lee et al., 2015).

Overall, BPS was frequently detected in freshwater and sewage sludge, but rarely found in marine surface sediment before 2013. The concentration levels of BPS in aquatic environments were lower than those of BPA, at the level of 0.1–10 ng/L in river waters, 0.01 ng/g dw in sediments, and 1.0 ng/g dw in sewage sludge. The temporal and spatial distribution of occurrence of BPS in the water, sediment, and sludge sampled worldwide indicated that BPS is now widely used all over the world and its usage is increasing. However, the speed of replacement BPA by BPS in products or usage of BPS for other purposes are not similar in different countries. In addition, the reports above also reflected the fact that BPF may be used more than BPS as the concentrations and detection frequencies of BPF were similar or much higher than those of BPS in some water and sediment samples.

4.2. Bisphenol S in indoor dust

Indoor dust is a potential source for human exposure to environmental contaminants. The contaminants in indoor dust can enter the human body through inhalation, ingestion, or dermal absorption. Almost all of the new or traditional organic pollutants were reported sufficient in indoor dusts, including bisphenol analogues (Vandenberg et al., 2007) (Table 4).

It was reported that BPS was 100% detected in 156 indoor dust samples collected from three Asian countries (China, Japan, and Korea) and the U.S., with concentrations ranging from 0.0008 to 26.6 $\mu\text{g/g}$ (0.34 $\mu\text{g/g}$, geometric mean) (Liao et al., 2012c). Although on the same order of magnitude, concentration levels of BPS in dusts from U.S. (0.63 $\mu\text{g/g}$, median) and Japan (0.81 $\mu\text{g/g}$) were higher than those from China (0.17 $\mu\text{g/g}$) and Korea (0.36 $\mu\text{g/g}$) (Liao et al., 2012c). In another study, Wang et al. (2015) compared the occurrence of bisphenol analogues in 388 indoor dust samples from 12 countries in the global range

Table 2
Occurrence of bisphenol analogues in water: Concentrations (ng/L, median or mean) and detection frequency (100%).

Country	N ^a	Sampling location (sample number)	Year	BPA	BPS	BPF	Ref
Japan	18	Edogawa river (3)/Arakawa river (2)/Tamagawa river (3)/Tokyo bay (10)	2012–2013	12 (–) ^b	3.4 (–)	215 (–)	(Yamazaki et al., 2015)
China	6	Pearl river (3)/West river (3)		22.2 (–)	nd ^c (–)	277 (–)	
Korea	10	Han river (4)/Nakdong river (3)/Yeongsan river (3)		63 (–)	nd (–)	nd (–)	
India	14	Cooum river (5)/Puzhal river (2)/Adyar river (3)/Buckingham canal (2)/Korttalaiyar river (2)		380 (–)	26.5 (–)	nd (–)	
China	5	Hangzhou Bay (5)	2012	11.40 (–)	0.51 (–)	nd (–)	(Yang et al., 2014a)
China	13	Liaohe river (13)	2013	29 (100) ^d	8.9 (100)	nd (0)	(Jin and Zhu, 2016)
	10	Hunhe river (10)		42 (100)	8.4 (100)	nd (0)	
	23	Taihu lake (23)		7.9 (100)	2.0 (100)	0.50 (87)	
China	26	Taihu lake (26)	2016	23 (100)	6.4 (100)	29 (100)	(Liu et al., 2017b)

^a N: sample number.

^b “–”: not detected.

^c nd = not detectable.

^d detection frequency (100%).

Table 3

Occurrence of bisphenol analogues in sediment and sludge: Concentrations (ng/g, median or mean, dry weight) and detection frequency (100%).

Sample	Country	N	Year	BPA	BPS	BPF	Ref	
Sediment	U.S.	82	1998–2012	1.49 (74)	nd ^a (16 ^b)	1.44 (59)	(Liao et al., 2012d)	
	Japan	56	2012	8.30 (100)	nd (46)	3.57 (89)		
	Korea	34	2008	6.02 (85)	nd (29)	nd (27)		
	All	172		3.94 (85)	nd (29)	2.76 (62)		
	China	5	2012	9.09 (–)	0.07 (–)	0.60 (–)	(Yang et al., 2014a)	
	China	23	2013	0.72 (100)	0.07 (57)	0.47 (91)	(Jin and Zhu, 2016)	
		12		0.11 (67)	nd (8.3)	nd (8.3)		
		7		0.93 (100)	nd (14)	nd (43)		
		China	24	2016	6.00 (100)	0.57 (100)	5.1 (100)	(Liu et al., 2017b)
	Sludge	China	52	2010–2011	9.38 (77)	4.34 (83)	1.97 (64)	(Song et al., 2014)
Korea		40	2011	275 (98)	3.80 (70)	249 (75)	(Lee et al., 2015)	
U.S.		76	2006–2007	222 (100)	5.8 (84)	8.16 (68)	(Yu et al., 2015)	

^a nd = not detectable.^b Detection frequency (100%).

(Wang et al., 2015). For those dust, 284 were collected from family homes, and 104 were collected from other microenvironments, such as laboratories, offices, cars, air conditioners, and e-waste workshops. For house dust, BPS was found in all samples from Japan ($n = 14$) and Romania ($n = 23$), and detection frequencies ranged from zero (China) to 86% (Greece) in samples from other 10 countries. The highest BPS levels in house dusts were found in samples from Greece (0.86 $\mu\text{g/g}$, median), followed by Japan (0.16 $\mu\text{g/g}$) (Wang et al., 2015). The authors indicated that the different detection frequencies and concentration levels of BPS in the 12 countries were partly attributed to the dust

collection time. For example, the indoor dusts of China were collected 2010–2011 when BPA was just banned for use in baby bottles in China, so both the concentrations and detection frequencies of BPS in samples from China were low. However, for microenvironment dusts, BPA and BPS were found in all 104 analyzed samples. The concentrations of BPA in the same microenvironment dusts from different countries were significantly different, but the concentrations of BPS were almost on the same levels, such as those in offices from South Korea (for BPA: 2.4 $\mu\text{g/g}$, BPS: <0.002 $\mu\text{g/g}$, median) and Pakistan (for BPA: 0.09 $\mu\text{g/g}$, for BPS: <0.002 $\mu\text{g/g}$) (Wang et al., 2015).

Table 4Concentrations of bisphenol analogues in indoor dust ($\mu\text{g/g}$, median or mean) and indoor air (ng/m^3 , median or mean), and the detection frequency (100%).

Country	Sample location	N	year	BPA	BPS	BPF	BPAF	Ref
<i>Indoor dust</i>								
China	Home	34	2012–2014	0.330 (100 ^a)	<0.002 (0)	<0.001 (53)	0.0019 (100)	(Wang et al., 2015)
Colombia		42		0.120 (100)	0.0024 (62)	0.033 (90)	0.0022 (98)	
Greece		28		1.500 (100)	0.860 (86)	0.780 (82)	0.0025 (79)	
India		35		0.130 (100)	0.0042 (60)	0.0067 (77)	0.0015 (83)	
Japan		14		1.700 (100)	0.160 (100)	0.230 (93)	0.0041 (100)	
South Korea		16		0.720 (100)	0.0036 (50)	1.000 (100)	0.0030 (94)	
Kuwait		17		0.250 (100)	0.020 (68)	0.022 (89)	0.0025 (100)	
Pakistan		22		0.066 (100)	0.0018 (50)	0.050 (100)	0.0013 (77)	
Romania		23		0.600 (100)	0.082 (100)	0.0020 (61)	0.00039 (74)	
U.S.		22		1.500 (100)	<0.002 (18)	0.200 (100)	0.0014 (100)	
Vietnam		12		0.230 (100)	<0.002 (33)	0.057 (92)	0.0011 (92)	
Saudi Arabia		29		0.650 (100)	0.028 (63)	0.073 (100)	0.0022 (89)	
All		284		0.440 (100)	0.0032 (100)	0.036 (83)	0.0018 (73)	
South Korea	Lab	11	2012–2014	1.400 (100)	<0.002 (100)	2.300 (100)	0.012 (100)	(Wang et al., 2015)
	Office	14		2.400 (100)	<0.002 (100)	7.300 (100)	0.0054 (100)	
Kuwait	Car	15		0.610 (100)	0.0060 (100)	0.0048 (73)	0.0012 (53)	
Pakistan	Car	6		0.500 (100)	0.0064 (100)	<0.001 (17)	0.0011 (67)	
	Office	24		0.090 (100)	<0.002 (100)	0.0065 (54)	0.0015 (71)	
Saudi Arabia	Air conditioners	12		0.510 (100)	0.016 (100)	0.030 (83)	0.0021 (75)	
	Car	10		0.380 (100)	0.0024 (100)	0.029 (100)	0.0014 (90)	
Vietnam	E-waste work shop	4		0.980 (100)	<0.002 (100)	0.032 (100)	0.0012 (100)	
	Public area	8		0.180 (100)	<0.002 (100)	<0.001 (13)	0.0022 (100)	
All		104		0.480 (100)	<0.002 (100)	0.021 (72)	0.0016 (99)	
U.S.	Home	38	2006–2010	1.60 (100)	0.63 (100)	0.049 (68)	0.00035 (0)	(Liao et al., 2012c)
Japan		22	2012	2.70 (100)	0.81 (100)	0.057 (82)	0.00035 (9.1)	
Korea		41	2012	3.26 (100)	0.36 (100)	0.45 (98)	0.0048 (76)	
China		55	2010	0.63 (96)	0.17 (100)	0.038 (56)	0.00035 (0)	
All		156		1.58 (99)	0.36 (100)	0.096 (7)	0.00035 (21)	
<i>Indoor air</i>								
U.S. ^b	Parking garages	3	2014	0.24 (100)	0.07 (0)	0.13 (33.3)	0.03 (0)	(Xue et al., 2016)
	Auto repair shops	4		0.99 (100)	0.07 (25)	1.49 (75)	0.03 (0)	
	Cars	7		0.75 (100)	0.07 (14.3)	1.66 (85.7)	0.03 (0)	
	Barber shops	5		0.60 (100)	0.07 (40)	1.35 (80)	0.03 (0)	
	Public places	13		0.70 (92.3)	0.07 (38.5)	0.55 (61.5)	0.03 (0)	
	Homes	26		0.59 (100)	0.07 (15.4)	0.78 (69.2)	0.03 (3.85)	
	Labs	12		0.07 (41.7)	0.07 (0)	0.7 (75)	0.03 (8.33)	
	Offices	13		0.29 (76.9)	0.07 (0)	0.13 (46.2)	0.03 (7.69)	
All		83		0.48 (86.8)	0.07 (15.7)	0.81 (66.3)	0.03 (3.61)	

^a Detection frequency (100%).^b Sample was indoor air.

In addition to indoor dust, bisphenol analogues in indoor air were also investigated. For the first time, Xue et al. (2016) reported the occurrence of BPS and BPF in indoor air (Xue et al., 2016). Their results indicated that BPA was the main analogue in indoor air (0.48 ng/m³, median concentration; 86.8%, detection frequency), followed by BPF (0.81 ng/m³; 66.3%), BPS (0.07 ng/m³; 15.7%), and BPAF (0.03 ng/m³; 3.61%). They also found a positive correlation between concentrations of BPA and BPF, while BPS concentrations were negatively associated with BPA or BPF. These results indicated that BPS may substitute BPA and BPF in some products in indoor environment, consistent with the negative correlation between concentrations of BPS and BPA reported in thermal receipt papers (Liao et al., 2012b).

The results of above studies in indoor dust were consistent with those of BPS occurrence in sediment and water samples, in which the BPS concentrations and detection frequencies were highly dependent on sampling time and location. In addition, the distribution of bisphenol analogues in dust and indoor air also demonstrated that use of BPF in consumer products may be more frequent than BPS.

4.3. Bisphenol S in foodstuffs

Epoxy resins are applied as inner coatings for metallic food cans and packaging containers, so BPS may leach from these packaging materials and contaminate specific foods during storage. Similar to BPA, by far, dietary is considered as the dominant source for human exposure to BPS (Liao and Kannan, 2013). The survey of occurrence of BPS in dietary is necessary.

Bisphenol analogues were determined by HPLC-MS/MS in foodstuffs collected from the U.S. and China (Table 5). BPS was detected in 21% of 267 foodstuffs, including beverages, vegetables, fruit, cereal, and meat collected from Albany, New York, U.S. (Liao and Kannan, 2013), and concentrations of BPS in most samples were below the detection limit. BPS was frequently found in meat, with a detection frequency of 43%.

The average and median concentration of BPS in 267 food samples in the U.S. was 0.13 and 0.005 ng/g (fresh weight). For the foodstuffs in China, the occurrence of BPS was almost similar to that of the U.S., with detection frequency of 23% in 289 samples and concentrations ranging from ND to 42.3 ng/g (fresh weight) (Liao and Kannan, 2014b). The results from the two studies also indicated that canned foods usually contained higher concentrations of bisphenol analogues in comparison to foods in glass, paper, or plastic containers. This conclusion was also demonstrated by a well-designed survey to assess food contamination of bisphenol analogues resulting from packing material. Seven paired beverage samples, respectively packed in cans and plastic bottles, and five canned foods were collected from local supermarkets in Beijing (Yang et al., 2014b). In this study, BPS was only found in coconut juice, corn, and pork samples (Table 5), and the concentrations of BPS were almost similar in all coconut juices packed with different materials (0.036 ng/mL in cans and 0.019 ng/mL in plastic bottles). However, BPS levels of canned food (canned corn and pork) were approximately one order of magnitude higher than those of the canned beverage (canned coconut juice) (Yang et al., 2014b). Therefore, the concentrations of BPS in food were not only depending on packing materials, but also the characteristics of the food. In addition, the low detection frequency of BPS in those beverages (<20%) indicated that BPS may not have been widely added to beverage packaging materials at that time in Beijing.

The limited reports of BPS in food samples indicated that contamination of BPS in foodstuffs was not serious compared with BPA and BPF, with slightly higher concentrations and detection frequencies in canned food.

4.4. Bisphenol S in consumer products

BPS was also analyzed in some consumer products in our daily life, such as various paper products and PCPs. As a major alternative to BPA in thermal paper, BPS is used as a color developer which has lots

Table 5
Occurrence of bisphenol analogues in foodstuffs: Concentrations (ng/g, median or mean, fresh weight) and detection frequency (100%).

Country	Year	Sample	N	BPS	BPA	BPF	BPP	BPAF	Ref			
U.S.	2008–2012	Beverage	31	<0.01 (3.2 ^a)	<0.01 (13)	<0.05 (0)	<0.025 (3.23)	<0.01 (6.5)	(Liao and Kannan, 2013)			
		Dairy products	29	<0.01 (14)	<0.01 (48)	<0.05 (17)	<0.025 (0)	<0.01 (10)				
		Fats and oils	5	<0.01 (0)	0.721 (80)	<0.05 (60)	<0.025 (0)	<0.01 (20)				
		Fish and seafood	23	<0.01 (26)	0.369 (74)	<0.05 (17)	<0.025 (0)	<0.01 (4.4)				
		Cereals	48	<0.01 (15)	0.185 (56)	<0.05 (2.1)	<0.025 (4.2)	<0.01 (2.1)				
		Meat	51	<0.01 (43)	0.431 (76)	<0.05 (7.8)	<0.025 (3.9)	<0.01 (7.8)				
		Fruits	20	<0.01 (25)	<0.01 (45)	<0.05 (0)	<0.025 (0)	<0.01 (30)				
		Vegetables	45	<0.01 (22)	0.556 (67)	<0.05 (18)	<0.025 (4.4)	<0.01 (16)				
		Other	15	<0.01 (6.7)	0.186 (53)	<0.05 (13)	<0.025 (13)	<0.01 (20)				
		All	267	<0.01 (21)	0.125 (57)	<0.05 (10)	<0.025 (3.4)	<0.01 (10)				
		China	2014	Coconut juice	1 ^b	0.036 (–) ^f	0.23 (–)	nd (–)		–	0.052 (–)	(Yang et al., 2014b)
					1 ^c	0.019 (–)	12 (–)	0.39 (–)		–	0.013 (–)	
				Canned foods	1 ^d	0.26 (–)	6.5 (–)	4.3 (–)		–	0.070 (–)	
		1 ^e	0.22 (–)	30 (–)	35 (–)	–	nd (–)					
China	2012	Cereals	39	<0.01 (23)	0.383 (69)	<0.05 (13)	<0.025 (5.1)	<0.01 (2.6)	(Liao and Kannan, 2014b)			
		Meat	20	<0.01 (30)	0.098 (55)	<0.05 (25)	<0.025 (15)	<0.01 (10)				
		Fish and seafood	11	0.145 (73)	4.46 (100)	<0.05 (46)	<0.025 (0)	<0.01 (36)				
		Eggs	11	<0.01 (0)	0.967 (64)	<0.05 (27)	<0.025 (9.1)	<0.01 (36)				
		Milk products	17	<0.01 (12)	0.942 (82)	<0.05 (53)	<0.025 (5.9)	<0.01 (0)				
		Bean products	27	<0.01 (15)	0.147 (63)	<0.05 (3.7)	<0.025 (11)	<0.01 (7.4)				
		Fruits	20	<0.01 (15)	0.530 (85)	<0.05 (10)	<0.025 (0)	<0.01 (25)				
		Vegetable	42	<0.01 (31)	0.224 (60)	<0.05 (26)	<0.025 (17)	<0.01 (21)				
		Cookies	26	<0.01 (35)	0.668 (73)	<0.05 (7.7)	<0.025 (3.8)	<0.01 (3.8)				
		Beverages	42	<0.01 (0)	7.84 (75)	<0.05 (0)	<0.025 (0)	<0.01 (0)				
		Cooking oils	11	<0.01 (9.1)	0.724 (55)	<0.05 (46)	<0.025 (0)	<0.01 (9.1)				
		Condiments	48	<0.01 (17)	0.005 (25)	<0.05 (10)	<0.025 (2.1)	<0.01 (15)				
		Other	13	<0.01 (15)	0.097 (54)	<0.05 (23)	<0.025 (7.7)	<0.01 (15)				
		All	289	<0.01 (23)	0.212 (61)	<0.05 (19)	<0.025 (6.9)	<0.01 (13)				

^a Detection frequency (100%).

^b Coconut juice packed in poly (ethylene terephthalate) or PET.

^c Coconut juice packed in can.

^d Sample is corn (solid content).

^e Sample is pork luncheon meat.

^f Not detected.

of commercial uses, such as receipt papers, airplane boarding passes, luggage tags, and tickets (Liao et al., 2012b; Mendum et al., 2011).

It was reported that BPS was detected in all 103 thermal receipt papers collected from China, Korea, Japan, and the U.S., and the median concentration of BPS in these receipt papers was as high as 7440 $\mu\text{g/g}$ (Liao et al., 2012b). At the same time, BPA was also found in receipt papers from all these countries except Japan. In Japan, both concentrations and detection frequencies of BPS in receipt papers were significantly higher than those of BPA, indicating a phased out trend of BPA in Japan. They indicated that this trend could also be verified by the significant negative correlation between the concentrations of BPS and BPA in 103 thermal receipt samples ($r = -0.55, p < 0.0001$). The thermal papers with a high concentration of BPS had a low or non-detectable level of BPA (Liao et al., 2012b). In addition to receipts, BPS was also frequently found in other paper products, such as paper currencies, tickets, and airplane boarding passes (Liao et al., 2012b). Based on the two paper studies, the authors indicated that due to the extremely high concentration of BPS (5000 $\mu\text{g/g}$, median) in receipts, the BPS in thermal receipt papers may be an important source for other paper products. On one hand, BPS can migrate from thermal receipt papers to other paper products by contact. For example, it was reported that the main source of BPS in currency bills was the thermal receipt paper due to their frequent contact (Liao et al., 2012b). On the other hand, paper products are usually made from recycled paper which may also include thermal receipt paper (Liao and Kannan, 2011; Liao et al., 2012b). Another study also reported the occurrence of bisphenol analogues in house waste papers from Danish households, such as thermal paper receipts, non carbon copy paper, and conventional printer paper, and BPS was found in 73% of the waste papers (Pivnenko et al., 2015). The highest concentration of BPS was also found in thermal paper receipts, with a median value of 7800 $\mu\text{g/g}$ (Pivnenko et al., 2015). A much similar study was also conducted in Brazil (Rocha et al., 2015). In 2015, 190 receipts were randomly collected from different locations in Brazil (such as supermarkets, restaurants, banks) and measured bisphenol analogues in them. BPS was found in 6.3% ($n = 12$) of the 190 samples, with concentration ranging from 11 to 22 mg/g, but BPA was not presented in these 12 samples (Rocha et al., 2015). The results reflected that BPS was also gradually replacing BPA in Brazil nowadays.

We summarized these recent reports of occurrence of BPS and BPA in thermal papers in Fig. 1. As shown, BPS now is used as the color developer instead of BPA in thermal papers all over the world. Concentrations of BPS were almost similar or higher to that of BPA in thermal papers collected from the U.S. and Europe. As the high concentration of BPS in thermal papers, the related products become an important source of BPS to the environment.

In addition to various paper products, bisphenol analogues were also determined in PCPs collected from China and the U.S. (Liao and Kannan, 2014a). BPS and BPA were only detected in <20% PCP samples, with concentration levels usually below the limit of detection. Whether for BPS or BPA, there was no significant difference of concentrations between the two countries. The low concentration and detection frequency of BPS indicated that BPS contamination was not serious in PCPs.

4.5. Bisphenol S in human urine

Similar to BPA, urinary BPS may be used as a biomarker to reflect the human exposure level to BPS from various potential sources. Only limited studies have reported the occurrence of BPS in human urine (Table 6).

In 2012, for the first time, BPS was reported present in human urine (Liao et al., 2012a). In that study, several bisphenol analogues were determined by HPLC-MS/MS in 315 urine samples collected from the U.S. and seven Asian countries, including China, India, Japan, Korea, Kuwait, Malaysia, and Vietnam. BPS was found in 81% of the total samples, and the concentrations of BPS were from <0.02 to 21.0 ng/mL, with a geometric mean value of 0.17 ng/mL. The concentrations of BPS in urine from Japan and the U.S. were relatively higher than other countries, which was consistent with the fact that the levels and detection frequencies of BPS in paper products and dust samples from the two countries were higher than those in other target countries (Liao et al., 2012a; Liao et al., 2012b; Liao et al., 2012c). This phenomenon indicated that the process speed of substituting BPA with BPS in products in Japan and the U.S. was relatively faster than other target countries (Liao et al., 2012a; Liao et al., 2012b; Liao et al., 2012c). In another study in 2012–2013, the relationships between childhood obesity and their urinary concentrations of several endocrine disruptors were investigated,

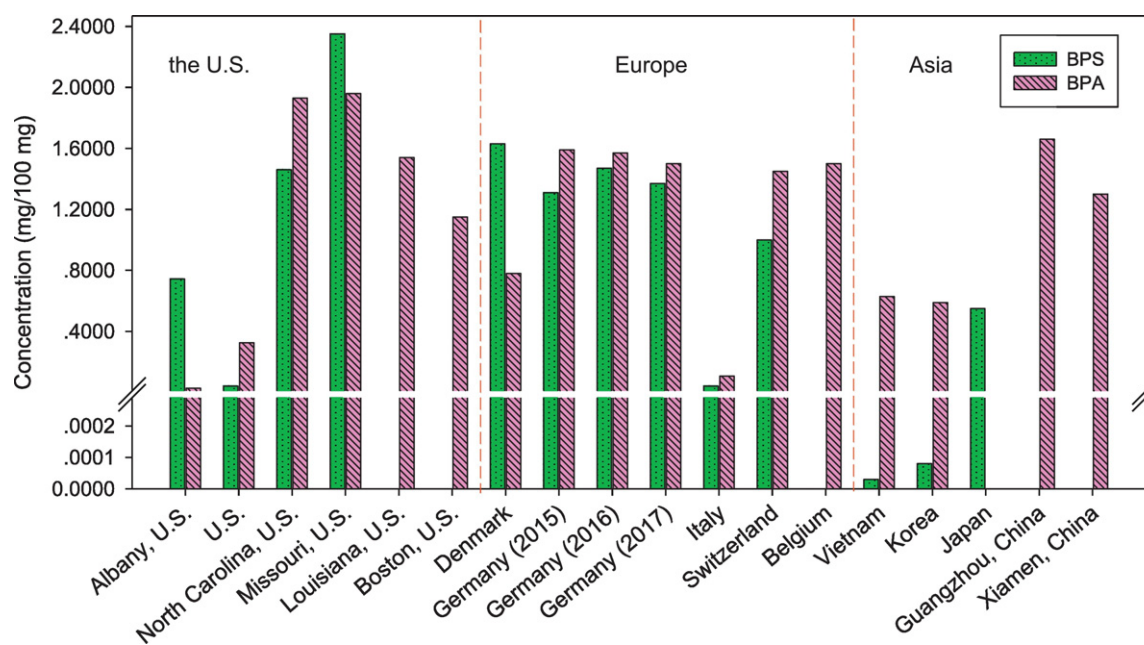


Fig. 1. BPS concentration in thermal papers from different countries (mg/100 mg). (Data sources: Albany NY and other cities, the U.S., Vietnam, Korea and Japan (Liao et al., 2012b); Missouri, the U.S. (Hormann et al., 2014); North Carolina, the U.S. (Thayer et al., 2016); Louisiana, the U.S. (Babu et al., 2015); Boston, the U.S. (Mendum et al., 2011); Denmark (Pivnenko et al., 2015); Germany (Martin and Thomas, 2017); Italy (Russo et al., 2017); Switzerland (Goldinger et al., 2015); Belgium (Geens et al., 2012); Guangzhou, China (Fan et al., 2015); Xiamen, China (Gao et al., 2013)). *Except for data of Italy, Louisiana and Missouri, the U.S., Belgium, and Xiamen, China are the mean value, others are the median value.

Table 6
Occurrence of bisphenol analogues in human urine: Concentrations (ng/mL, median, or geometric mean) and detection frequency (100%).

Country	N	Year	BPA ^a	BPS ^b	BPF	BPAF	Ref
China	94	2013	0.886 (— ^c)	0.029 (—)	0.228 (—)	0.018 (—)	(Yang et al., 2014c)
U.S.	31	2010–2011	—	0.299 (97 ^d)	—	—	(Liao et al., 2012a)
China	89 ^a (116 ^b)		1.10 (90)	0.226 (82)	—	—	(Liao et al., 2012a; Zhang et al., 2011)
Japan	36 (36)		0.84 (100)	1.18 (100)	—	—	
Korea	33 (32)		2.00 (97)	0.030 (42)	—	—	
Kuwait	30 (32)		1.24 (81)	0.172 (70)	—	—	
Malaysia	29 (29)		1.00 (97)	0.071 (76)	—	—	
India	38 (21)		1.59 (100)	0.072 (76)	—	—	
Vietnam	29 (30)		1.42 (100)	0.160 (100)	—	—	
U.S.	122	2009	1.3 (98)	0.1 (73)	0.4 (75)	<0.1 (2)	(Ye et al., 2015)
	43	2010	2.0 (98)	0.1 (65)	<0.1 (44)	<0.1 (0)	
	95	2011	0.8 (99)	0.1 (63)	<0.1 (42)	<0.1 (1)	
	141	2013	0.6 (97)	0.2 (74)	<0.1 (50)	<0.1 (1)	
	42	2014	0.5 (74)	0.2 (74)	0.3 (88)	<0.1 (0)	
U.S.	100	2009–2012	0.72 (95)	0.13 (78)	0.08 (55)	—	(Zhou et al., 2014)
Saudi Arabia	130	2014	2.01 (—)	4.92 (—)	2.16 (—)	1.10 (—)	(Asimakopoulos et al., 2016)
India	76	2012–2013	5.08 (99)	0.04 (70)	—	—	(Xue et al., 2015)

^a Data were acquired from reference (Zhang et al., 2011).

^b Data were acquired from reference (Liao et al., 2012a).

^c “—”: not detected.

^d Detection frequency (100%).

and it was reported that the concentrations of BPS in 76 children from India (0.01–12.2 ng/mL) were relatively higher than the values of children and adolescents (age < 19) in the U.S. and seven Asian countries in Liao's study (0.02–2.39 ng/mL) (Liao et al., 2012a; Xue et al., 2015). In addition, a study has reported the occurrence of bisphenol analogues in urine collected from a cohort of residents living near a bisphenol AF manufacturing plant in south China (Yang et al., 2014c). The authors found the concentration levels of BPS in these populations were from ND to 2.51 ng/mL (Yang et al., 2014c), in the ranges of those in the U.S. and Asian countries (Liao et al., 2012a). A recent study from the U.S., which developed a sensitivity method for the determination of bisphenols in urine with small sample volume (100 μ L), also reported that BPA (0.72 ng/mL, median) and BPS (0.13 ng/mL) were the dominant bisphenol analogues in urine samples collected between 2009

and 2012 (Zhou et al., 2014). Furthermore, scientists also tried to find the relationship between BPS exposure and body oxidative stress (Asimakopoulos et al., 2016). By measuring 130 urine samples from the Saudi Arabian region, they found that concentrations of BPS were higher than those of BPA, and both of them were significantly associated with concentrations of an oxidative stress biomarker, 8-hydroxy-2'-deoxyguanosine (8-OHdG) (Asimakopoulos et al., 2016).

In addition to space distribution, the time trend of BPS in human urine was also investigated. In a recent study, four bisphenol analogues, BPA, BPS, BPF, and BPAF were determined in 616 adults urine samples collected from the U.S. at eight time points from 2000 to 2014 (Ye et al., 2015). The occurrence of BPS in urine samples well reflected the usage history of BPS in the U.S. (Ye et al., 2015). For examples, BPS was not found in urine samples collected before 2009. Although always

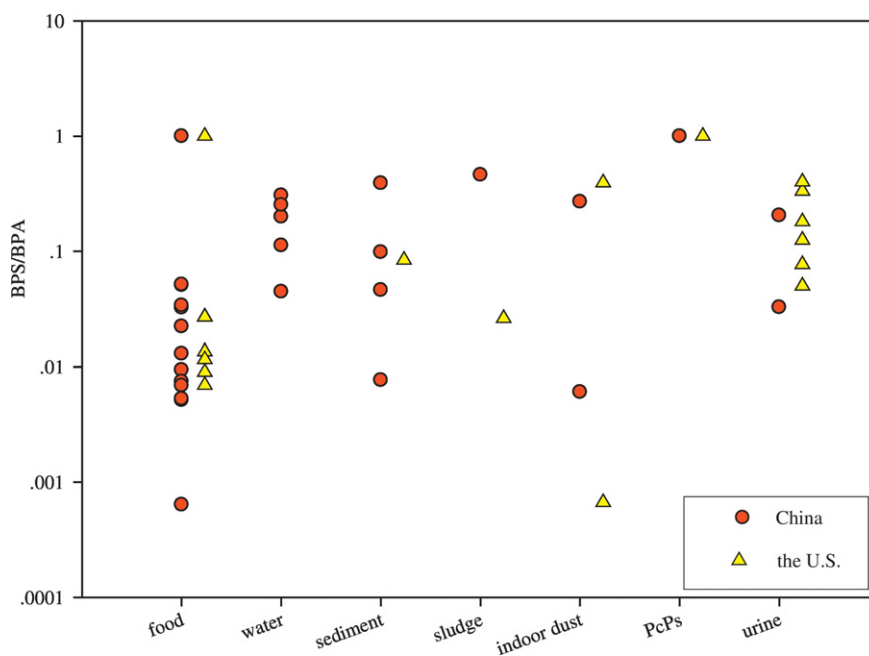


Fig. 2. The ratio of concentration of BPS to BPA in the environment and human urine samples from China and the United States. (Data sources: China: food (Liao and Kannan, 2014b); water (Jin and Zhu, 2016; Yamazaki et al., 2015; Yang et al., 2014a); sediment (Jin and Zhu, 2016; Yang et al., 2014a); sludge (Song et al., 2014); indoor dust (Liao et al., 2012c; Wang et al., 2015); PCPs (Liao and Kannan, 2014a); urine (Liao et al., 2012a; Yang et al., 2014c; Zhang et al., 2011); United States: food (Liao and Kannan, 2014a); sediment (Liao et al., 2012d) sludge (Yu et al., 2015); indoor dust (Liao et al., 2012c; Wang et al., 2015); PCPs (Liao and Kannan, 2014a); urine (Ye et al., 2015; Zhou et al., 2014)).

lower than BPA, concentration of BPS gradually increased from 0.1 to 0.2 ng/mL since 2009, while the concentration of BPA decreased from 1.3 to 0.5 ng/mL, as well as its detection rate reduced from 98% to 74% during the same period (Ye et al., 2015). Meanwhile, BPA was gradually replaced by BPS in the U.S. As indicated in their research, in 2006, a large U.S. thermal receipt paper producer announced the cessation of the use of BPA in paper products manufacture, replaced by BPS for production (Raloff, 2010). In addition, the U.S. Food and Drug Administration banned the use of BPA in baby bottles and children's drinking cups in 2012 (FDA, 2012).

The above studies indicated that urinary BPS could be used as the biomarker to reflect BPS human exposure status directly and reflect the replacement of BPA by BPS in productions indirectly. Further, BPS metabolites in human urine were rarely reported and an increasing concentration of BPS metabolites in human urine is expected in future studies.

The research on occurrence of BPS was limited in other human specimens other than human urine. One recent study reported that BPS was found in four maternal and seven cord sera samples collected from China, with low concentrations ranging from <0.03–0.12 ng/mL (Liu et al., 2017a). However, BPA was frequently found in human blood and breast milk (Azzouz et al., 2016; Lin et al., 2017; Mendonca et al., 2014). Considering the close chemical structure of BPS to BPA and the toxicities of BPA, further studies should be conducted to investigate BPS in other human specimens to understand the bioavailability, distribution, or toxicities of BPS in the human body.

As BPS is a substitute for BPA added in products, we assumed the concentration ratio of BPS to BPA could be used as a simple factor to reflect the substitution degree, and to track the fate of BPS and BPA in the environment. We calculated the concentration ratio of BPS to BPA based on data reported in each environmental medium for China and the U.S. as an example (Fig. 2). As predicted, the concentrations of BPS in all environmental media were generally lower than that of BPA, usually one to two orders of magnitude lower in the two countries. However, the levels of BPS in aquatic environments and indoor dusts, especially water and sludge, were almost comparable or equal to that of BPA, indicating the gradual use exchange from BPA to BPS in the U.S. and China. In addition, considering the relatively shorter half-life of BPS than BPA in water and sediment (Chen et al., 2016), the ratio here may be underestimated, which indicates that the exchange of BPA to BPS in products or the use of BPS for other purposes is happening more quickly and at a larger scale than we expected.

5. Human exposure to BPS

The ubiquity of BPS in the environment indicates that people are exposed to BPS on a daily basis. The potential sources and routes for human exposure to BPS are shown in our Graphical Abstract. As shown, BPS can enter the human body by ingestion (food or dust), inhalation (air or particle), and dermal absorption (dust or PCPs). The handling of thermal papers and clothes are relatively special routes of human exposure to BPS (Liao and Kannan, 2014a; Liao et al., 2012b; Liao and Kannan, 2013; Liao et al., 2012c). The daily exposure of BPS was estimated by directly calculating from BPS in potential environmental sources or back-calculating from BPS in human urine in several countries (Table 7).

Human exposure to BPS through indoor dusts has been investigated worldwide. To compare exposures from different countries, the estimated daily intake (EDI) of BPS based on concentrations in dusts collected from 12 countries were calculated by Wang et al. (2015). The higher EDIs by dust ingestion were found for dusts collected from households in Korea ($1.07E-03$ ng/kg_bw/day), Saudi Arabia (0.01 ng/kg_bw/day), and Kuwait ($5.94E-03$ ng/kg_bw/day), and the EDIs were much lower for dusts from other microenvironments (max: $1.41E-04$ ng/kg_bw/day) (Wang et al., 2015). This may be related to the long time people spent in the home environment every day. In

addition, the authors also calculated the EDIs of BPS via dust inhalation in different age groups for populations of all 12 countries. They found that the EDIs were generally decreasing with increasing age, thus infants were the highest exposed group (Wang et al., 2015). Similarly, another study in U.S. also reported that toddlers had higher EDIs of BPS than other age groups (Liao et al., 2012c). For example, in U.S., the maximum EDI (based on median concentration) was found in toddlers of 3.09 ng/kg_bw/day, and in decreasing order, 2.20, 0.85, 0.53, and 0.42 ng/kg_bw/day for infants, children, teenagers, and adults, respectively. The high EDIs for infants and toddlers may be due to their low body weight and frequent hand to mouth contact (Liao et al., 2012c). However, although both of the two studies reported the high levels of BPS in indoor dusts, the contributions of EDIs from dust ingestion or inhalation to total exposure doses were not high (Liao et al., 2012c; Wang et al., 2015).

Personal exposure to BPS from dermal absorption was also investigated. EDI by contact products was estimated based on the BPS concentration in various paper products (Liao et al., 2012b). BPS exposure from thermal paper contact accounted for 88% of the total dose from all paper products. The EDIs of BPS from thermal receipt papers for occupationally exposed individuals were significantly higher than those of the general population, with a median value of 21,804 and 291 ng/day, respectively (Liao et al., 2012b). Hand contact exposures of BPS from tickets (0.766 ng/day for all population), airplane boarding passes (0.401 ng/day for all population) and paper currencies (respective values of 0.0175 and 0.0017 ng/day for occupational and general populations) were also studied (Liao et al., 2012b). In addition, the EDIs of BPA and BPS by dermal absorption from handling of paper receipts was also estimated for the population in Brazil (Rocha et al., 2015). The EDIs of BPA and BPS for the occupational population (median: 71,040 ng/day) who had contact with thermal paper on a daily basis were much greater than those of the general population (median: 142 ng/day) (Rocha et al., 2015).

Furthermore, based on urinary concentrations, the total exposure doses of BPS were estimated for the general population in the U.S. and seven Asian countries (Liao et al., 2012a). The levels of EDIs in Japan (1.67 µg/day, median), China (0.339 µg/day), the U.S. (0.316 µg/day) and Kuwait (0.292 µg/day) were higher than other target countries. In addition, there was no difference for EDIs among different age groups, but a slightly higher value was observed for male (0.348 µg/day) than that of female (0.186 µg/day) (Liao et al., 2012a).

The current research indicates that food may be the dominant source for human exposure to BPS. The study of Wang et al. (2015) emphasized that, for the population in China and U.S., the EDIs of total bisphenol analogues via diets accounted for >90% of the total EDIs when considering both dust and diet (Wang et al., 2015). Additionally, the use of PCPs may also contribute certain amounts to the total EDIs of BPS. It was reported that the geometric mean EDIs of all bisphenol analogues via PCP dermal exposure was 0.072 and 0.120 µg/day for Chinese and U.S. adult women, respectively (Liao and Kannan, 2014a). Furthermore, a recent study found that textiles and baby clothing contained a certain amount of BPS, and newborns can be affected by skin absorption. Dermal BPS exposure doses from textiles ranged from 8.20 pg/kg_bw/d for 6–12 months old infants to 10.1 pg/kg_bw/d for newborns (<1 month) (Xue et al., 2017). However, depending on the data of Table 7, the contribution of PCPs or textiles to total BPS exposure is low.

6. Toxicities and potential adverse health effects of BPS

Compared to numerous studies on the toxicities of BPA, effects of BPS exposure are less understood. A previous review in 2015 indicated that BPS was an endocrine disruptor and may have potential health hazards almost in the same order of magnitude as BPA (Rochester and Bolden, 2015). Scientists found that the isopropylation of 4-hydroxy in BPS can trigger estrogenic activity, thus the two terminal hydroxyl groups in the *para* position are crucial for estrogenic activities of BPS (Kuruto-Niwa et al., 2005). After the review (Rochester and Bolden,

Table 7
Daily exposure doses of BPS estimated from potential sources in several countries (mean/median, ng/day).

Country	Total	Dust	Paper	Food	PCPs	Indoor air	Ref.
China	339/40.6 ^a	7.56/0.063	— ^b	19.40	4.16/72 ^c	–	Dust (Liao et al., 2012c; Wang et al., 2015); food (Liao and Kannan, 2014b); PCPs (Liao and Kannan, 2014a); total (Liao et al., 2012a; Yang et al., 2014c)
India	84/56	0.126	–	–	–	–	Dust (Wang et al., 2015); total (Liao et al., 2012a; Xue et al., 2015)
Japan	1670	35.91/5.04	319.61	–	–	–	Dust (Liao et al., 2012c; Wang et al., 2015); paper (Liao et al., 2012b); total (Liao et al., 2012a)
Korea	23	15.75/0.13	0.0465	–	–	–	Dust (Liao et al., 2012c; Wang et al., 2015); paper (Liao et al., 2012b); total (Liao et al., 2012a)
Kuwait	292	0.63	–	–	–	–	Dust (Wang et al., 2015); Total (Liao et al., 2012a)
Malaysia	122	–	–	–	–	–	Total (Liao et al., 2012a)
Pakistan	–	0.063	–	–	–	–	Dust (Wang et al., 2015)
Saudi Arabia	6888	0.63	–	–	–	–	Dust (Wang et al., 2015); Total (Asimakopoulos et al., 2016)
Vietnam	217	0.063	0.01743	–	–	–	Dust (Wang et al., 2015); paper (Liao et al., 2012b); total (Liao et al., 2012a)
U.S.	316	27.3/0.08	24.0577/1.996 ^d	8678	4.16/120 ^e	1.13	Dust (Liao et al., 2012c; Wang et al., 2015); paper (Liao et al., 2012b); food (Liao and Kannan, 2013); PCPs (Liao and Kannan, 2014a); total (Liao et al., 2012a)
Brazil	–	–	GP ^e : 1421/OP ^f : 71,040	–	–	–	Paper (Rocha et al., 2015)
Colombia	–	0.126	–	–	–	–	Dust (Wang et al., 2015)
Italy	–	–	GP: 24.4/OP: 15,600	–	–	–	Paper (Russo et al., 2017)
Greece	–	27.2	–	–	–	–	Dust (Wang et al., 2015)
Romania	–	1.95	–	–	–	–	Dust (Wang et al., 2015)

^a X/X: data from different studies; the calculation method is shown in Supporting information in Table S1.

^b Data not available.

^c Exposure dose for all bisphenol analogues including BPS.

^d Exposures from various papers other than thermal paper.

^e The estimated median daily intake of BPS from handling of thermal receipts for the general population (GP).

^f The estimated median daily intake of BPS from handling of thermal receipts for the occupational population (OP).

2015), several new studies were conducted to study the toxicities of BPS.

6.1. BPS toxicity *in vitro* experiments

BPS exposure may cause oxidative stress. The mechanism of oxidation of BPA and its analogues was described for the first time by Macczak et al. (2017). Red blood cells and bisphenols, including BPA, BPAF, and BPS, were incubated at a concentration of 0.1 to 500 µg/mL for 1, 4, or 24 h to assess the effect on oxidative stress parameters. Compared with BPA and BPAF, BPS did not cause significant changes in oxidative stress parameters, and showed weaker oxidative potential in this experiment (Macczak et al., 2017). A enhanced reactive oxygen species level (106.9%) was observed when cells exposed 4 h to BPS at the levels of 250 µg/mL. Although the oxidative potential of BPS was weak in this study, a biomonitoring study conducted in Saudi Arabia found a significant positive association between concentrations of BPS and 8-OHdG in human urine (Asimakopoulos et al., 2016). BPS may disrupt the function of the endocrine system, and then affect the reproductive system (Žalmanová et al., 2016). For example, significantly negative effects were found between the meiotic maturation of pig oocyte and BPS exposure even at a low concentration (3 nM) (Zalmanova et al., 2017).

The metabolites of BPS may also affect the endocrine activities. In 2016, Skledar et al. used two test systems, yeast cells (evaluating estrogenic and androgenic activities), and GH3.TRE-Luc reporter cell line (measuring thyroid hormone activity) to explore the effects of BPS and its two main metabolites, BPS glucuronide and hydroxylated BPS 4-(4-hydroxy-benzenesulfonyl)-benzene-1,2-diol (BPSM1), on endocrine activity (Skledar et al., 2016). The results indicated that BPS and BPSM1 were weak agonists of the estrogen receptor, with EC50 values 8.4×10^{-5} M and 6.7×10^{-4} M, respectively. BPSM1 also had a weak antagonistic thyroid hormone activity (IC50 4.3×10^{-5} M). But BPS glucuronide has no activity on estrogen, androgen, or thyroid hormone receptors, so glucuronidation seems to be the most important pathway for BPS metabolism and detoxification (Skledar et al., 2016).

Furthermore, several studies also indicated that exposure to BPS may induce obesity. For example, BPS exposure could modulate

macrophage phenotype. After treatment with low-concentration of BPS (10^{-10} M), the macrophages increased the secretion of pro-inflammatory cytokine (TNF-α), reduced anti-inflammatory cytokine (IL-10), and induced macrophage proinflammation (Zhao et al., 2017). In another study, Boucher et al. incubated human preadipocytes at a BPS concentration ranging from 0.1 nM to 25 µM. They found that the 25 µM BPS treatment group increased the expression of several key adipogenic markers (e.g., lipoprotein lipase and adipocyte protein 2) both in mRNA and protein levels, and induced lipid accumulation. This effect was possibly mediated through a peroxisome proliferator-activated receptor-γ approach. This was the first study demonstrating the effect of BPS on lipid accumulation and differentiation in human preadipocytes (Boucher et al., 2016).

6.2. BPS toxicity *in vivo* experiments

Zebrafish is a hermaphrodite organism which is a valuable biological model species for a variety of toxicology analyses. Previous zebrafish exposure experiments indicated that BPS had adverse effects on the endocrine system (Ji et al., 2013; Naderi et al., 2014). In 2015, Kinch et al. (2015) found that low-dose BPA or BPS (0.0068 µM) exposure may affect the development of the hypothalamus and cause zebrafish larvae hyperactive behavior. Also, they found the expression of aromatase in the synthesis of estradiol in zebrafish was changed after BPS exposure. In 2016, Qiu et al. (2016) studied the effect of BPS on the reproductive neuronal endocrine system during zebrafish embryonic and larval development period, and explored the potential mechanisms connected with estrogen receptors, thyroid hormone receptors, and aromatase approach. They reported that exposure to a low level of BPS (100 µg/L) in the early development stage (25 h postfertilization in zebrafish embryos) could add numbers of Gonadotropin-Releasing Hormone 3 neurons in hypothalamus and increase reproduction-related gene (e.g., *kiss1*, *gnrh3* and *erα*) expression, similar to the results of BPA at the same concentration (Qiu et al., 2016).

The BPS toxicities were also investigated in rodent experiments. In 2016, Ullah et al. used *in vivo* and *in vitro* experiments of rats demonstrated that BPS may cause oxidative stress and have anti-androgenic

characteristics (Ullah et al., 2016). In the study, adult male rats were chronically exposed to different levels of BPS (0, 1, 5, 25, 50 mg/kg_bw/day) and adult male rat testicular slices was also treated with various doses of BPS (0, 0.5, 1, 10, and 100 ng/mL) *in vitro* assay. An obvious increasing in testicular reactive oxygen and lipid peroxidation was found in high level BPS treated testicular tissue, but a decreasing concentration of plasma and testosterone was observed *in vivo*. The results indicated that exposure to high concentrations of BPS can induce oxidative stress in the testis and perform anti-androgenic characteristics which may alter adult reproductive function (Ullah et al., 2016). In another study, CD-1 mice were incubated from gestational day 8 to postnatal day 19 at a BPS dose of 200 µg/kg_bw/day, and the expression of estrogen-responsive genes both in the ovary and uterus were changed (Hill et al., 2017). It was noteworthy that the females responded abnormally to an estrogen challenge, suggesting exposures to BPS during the perinatal period may heighten responses of the uterus and reduce responses in the ovaries with an estrogen challenge. Additionally, they also found that BPS exposure would interfere with the development of the reproductive tract in female mouse (Hill et al., 2017). In addition, the effects of BPS exposure on maternal behavior and brain in mice may not be terminated by the F1 generation. Catanese et al. evaluated the maternal behaviors, and expression of estrogen receptor and tyrosine hydroxylase immunoreactive cells in two brain regions critical for maternal care in two generation of CD-1 mice (Catanese and Vandenberg, 2017). They found that the expression of estrogen receptor in the caudal medial preoptic area (cMPOA) was significantly increased when female F0 were exposed to a BPS dose of 200 µg/kg_bw/day, but the same phenomenon was not found in F1 generation. They demonstrated that the effect of BPS on maternal behavior and brain in mice depended on the exposure dose and time, and generation. They also indicated that the pregnancy and lactation periods were the window of vulnerability to BPS exposure for mothers (Catanese and Vandenberg, 2017).

In summary, BPS and its metabolites are endocrine disruptors, which were demonstrated by both *in vitro* and *in vivo* exposure studies. In addition, exposure to BPS may trigger oxidative stress and obesity. Therefore, exposure to BPS from potential sources might have adverse effects on human health.

7. Conclusion and perspective

In this short review, we summarized the recent reports of occurrence of BPS in different environment media, and found that BPS is now ubiquitously detected worldwide. Although the concentrations of BPS were generally lower than those of BPA in each environmental medium, with the prohibition use of BPA, the levels of BPS will continuously increase in the environment, which has already been reflected by the comparable concentrations of BPS and BPA in water and sludge samples from the U.S. and China. In addition, BPS was detected in human urine samples all over the world, indicating that human are widely exposure to BPS. Similar to BPA, food, indoor dust, PCPs, and especially thermal papers are the potential sources for human exposure to BPS, and food is the dominant source. Although scientists have found some toxicities of BPS by *in vivo* and *in vitro* experiments, no suggested daily exposure dose or oral maximum allowable dose of BPS was derived for human exposure by far. However, considering the much similar chemical structure of BPS to BPA, we suspect that long time exposure to BPS may have adverse effects on the development of reproductive and nervous systems for biota and humans. As the prenatal period was sensitive to BPS exposure, we suggest pregnant women and babies should pay special attention to avoid exposure to BPS (e.g., reducing the use of plastic products, PCPs, canned food, and dermal contact with thermal receipt papers). Finally, the further investigation of BPS and its metabolites should pay attention to their occurrence in human specimens other than urine, endpoints regarding human exposure, fate, and bioavailability in the environment.

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