Determination and risk characterization of polycyclic aromatic hydrocarbons of tea by using the Margin of Exposure (MOE) approach

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Abstract Tea is one of the most frequently consumed drinks due to its favourite taste and the health benefit. Tea is produced by several processes and drying is very important step to develop the flavour and destroys the enzymes in tea. However, during drying tea, polycyclic aromatic hydrocarbons some of which are carcinogen and genotoxin are naturally produced. The risk of PAHs by drinking tea was characterized by determining contents of 4 PAHs in tea. 4 PAHs including Benz(a)anthracene (BaA), Chrysene (CHR), Benzo(b)fluoranthene (BbF) and Benzo(a)pyrene (BaP) were investigated by GC–MS in total 468 tea products, which were contaminated up to 4.63 ng g⁻¹. Mate tea was the most highly contaminated by BaA, CHR, BbF and BaP and followed by Solomon's seal and Chrysanthemum. The Margin of Exposures calculated by the concentration of BaA, CHR, BbF and BaP and consumption amount of tea were higher than 10,000, and the risk of PAHs in tea were low concern to public health.

Keywords Polycyclic aromatic hydrocarbons (PAHs) · Margin of Exposure (MOE) · Tea · Risk characterization · Determination

Introduction

Tea is one of the most frequently consumed drinks and is made of vegetable substances and tea plant like green and mate. Tea can be classified into a variety of types according to production processes like fermentation and heating (Abd EL-Aty et al., 2014). Tea usually refers to green tea and black tea, which are produced from the leaves of the plant *camellia sinensis* (Wang et al., 2011). Tea is not only highly consumed due to its favoured taste but also due to its health benefits. Tea, especially green tea, possesses catechins, a group of polyphenols, including (–)-epigallocatechin-3-gallate (EGCG), (–)-epigallocatechin (EGC), (–)-epicatechin (EC), and (–)-epicatechin gallate (ECG), giving it antiproliferative, antimutagenic, antioxidant, antibacterial, antiviral, and chemopreventive properties (Banerjee et al., 2010; Butt and Sultan, 2009). In Korea, teas are categorized into three types according to the production processes: leached, liquid and solid tea. Leached tea, including green, black and oolong tea is manufactured by drying and frying plant-derived raw materials like plants sprout, leaves, flowers, stalks, fruits, or gains and consumed as liquid by dipping them in water and filtering the extracts. Liquid tea is made to liquid by treating the plant-derived raw materials through extraction and drunk itself. Solid tea is a powder or other form produced by using the plant-derived raw material as a main material and consumed with water (MFDS, 2016). The drying process is one of the most important processes in making tea. The heat destroys the enzymes and reduces moisture levels. The drying step also affects the flavor of the tea (Özcan et al., 2005; Venskutonis, 1997; Vieira et al., 2010). Several methods such as smoke, steam and sun-light are used to dry tea, and direct exposure to flames is sometimes used for rapid drying. However, in these drying processes, high

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levels of Polycyclic aromatic hydrocarbons (PAHs) are unintentionally produced (Adisa et al., 2015; Pincemaille et al., 2014; Vieira et al., 2010).

PAHs are chemicals containing more than two aromatic rings. They are produced from the pyrolysis of organic matter in free-radical polymerization processes (Longwell, 1982; Richter and Howard, 2000). Food can be contaminated with PAHs from the environment, but PAHs also can be produced in food during cooking and processing food with heating, drying, baking, frying, ohmic-infrared cooking, roasting, grilling, and smoking (Boffetta et al., 1997; Lijinsky, 1991; Singh et al., 2016). With the exception of tobacco smoke, people are mostly exposed to PAHs through food consumption (Zelinkova and Wenzl, 2015). PAHs intake form reactive intermediates, and which interact with DNA and induce mutation and cancer (IARC, 2010; Lagerqvist et al., 2011; Ramesh et al., 2004, Szeliga and Dipple, 1998). Some PAHs among the more than 100 known PAHs are mutagenic and carcinogenic (Kao et al., 2014), and the US Environmental Protection Agency (EPA) (1983) classified 16 PAHs as the indicators based on carcinogenicity to control their release into the environment. The European Commission (2005) selected benzo(a)pyrene (BaP) as an indicator for PAHs for food and set maximum levels of BaP in several categories. Furthermore, The European Commission (2011) has recently added maximum levels of sum of 4 PAHs, benz(a)anthracene (BaA), Chrysene (CHR), benzo(b)fluoranthene (BbF), and benzo(a)pyrene (BaP) from 1 to 50 µg/kg in different foods, since the European Food Safety Authority (EFSA) (2008) showed that BaP was not enough to be an indicator for PAHs.

In order to analyze PAHs in food, several methods have been published. Liquid–liquid extraction for extracting PAHs from sample, saponification for reducing the lipid contents, and solid-phase extraction for removing polar compounds have been adopted to isolate PAHs from various food matrices. Following these, liquid chromatography coupled with fluorescence, ultraviolet–visible or mass spectrometry and gas chromatography coupled with mass spectrometry have been used to qualify and quantify PAHs. FLD detection has been used more than FLD detection, since FLD detection is more selective and sensitive than UV detection. However, FLD detection still has some disadvantages including impossibility of using isotopically labeled surrogates. Therefore GC–MS is frequently applied to recent research. (Jira et al., 2008; Plaza-Bolaños and Frenich, 2010; Veyrand et al., 2007; Yu et al., 2012).

PAHs are found in various kinds of food and some surveys have been conducted to confirm levels of PAHs in edible oils, barbecued meat products. Edible oils are easily contaminated with PAHs due to their strong lipophilic properties (Balenovic et al., 1995; Gertz and Kogelheide,

1994) and barbecued meats contain higher levels of PAHs due to the incomplete combustion of drips from meat on charcoal (Lee et al., 2016a).

The risk of exposure to PAHs via drinking tea has been assessed to ensure the safety of tea. The majority of research has used the Toxic Equivalency Quotient (TEQ) concept adopted by the US EPA to estimate the intake of PAHs (Nisbet and Lagoy, 1992). The PAHs contents were converted to BaP equivalent contents (TEQ_{BaP}) by multiplying toxic equivalency factors (TEFs) and the exposure to PAHs was shown by the exposure to BaP (Alomirah et al., 2010; Jiang et al., 2015; Martí-Cid et al., 2008; Zhao et al., 2014). However, the TEQ concept should not be used for PAHs as they do not have the same toxicological mechanism in recent research, and the European Food Safety Authority (EFSA) proposed using the concentration of a mixture of PAHs and new toxicological value to assess the risk of PAHs in food (EFSA, 2008).

In 2005, a Margin of Exposure (MOE) approach was adopted by the Joint FAO/WHO Expert committee on Food Additives (JECFA) to characterize the risk of genotoxic and carcinogenic substances (JECFA, 2005; Lee et al., 2016b). MOE uses the toxicological data, BMDL₁₀, which is the benchmark dose of lower confidence limit to increase the amount of animals bearing tumor by 10%, since genotoxic carcinogens do not have a linear dose–response relationship and it is impossible to establish health guide value. The exposure should be minimized to As Low As Reasonably Achievable (ALARA). However, managers who try to ensure food safety cannot get any information from ALARA. Instead, the risk managers set priority lists by comparing MOE values and an appropriate reference point. An MOE of 10,000 or higher in general would be interpreted as low concern to public health (Benford et al., 2010a). Only a few studies have been conducted to estimate the risk of PAHs in food since the MOE approach was recommended for them (Benford et al., 2010b). This research was conducted to evaluate contamination levels of 4 PAHs in highly consumed tea products in the Korean market and to characterize the risk to the public health by using the MOE approach. Furthermore, the risks assessed by the TEQ concept and the MOE approach were compared.

Materials and methods

Chemicals and materials

4 PAHs (BaA, CHR, BbF, and BaP) and 2 deuterated PAHs (d12-CHR and d12-BaP) were purchased from Supelco (Bellefonte, PA, USA) for standards and internal standards, respectively. The 4 PAHs and 2 deuterated PAHs in methylene chloride were prepared at 100 µg/mL of stock

standard solutions and these were diluted using methylene chloride to 400 µg/L. 5 calibration standards of 0.2, 0.5, 1, 2 and 5 µg/L with internal standards of 4 µg/L were prepared for leached tea and solid tea. For liquid tea, calibration standards of 0.1, 0.2, 0.4, 1 and 2 µg/L were prepared with internal standards of 0.4 µg/L. HPLC grade solvents were used. Potassium hydroxide and sodium sulfate were purchased from Wako (Osaka, Japan) and ethyl alcohol, *N,N*-dimethylformamide, ethyl acetate, and *n*-hexane were obtained from Merck (Darmstadt, Hesse, Germany). Methylene chloride was purchased from Burdick and Jackson (Muskegon, MI, USA) and water was distilled and deionized using a Milli-Q System (Bedford, MA, USA). Silica SPE cartridges of 1 g were purchased from Waters Corporation (Milford, MA, USA) and SPE cartridges of 50 mg for use with PAHs were purchased from Supelco (Bellefonte, PA, USA). To evaporate solutions, a rotary evaporator (Eyela, Tokyo Rikakikai Co. Ltd., Japan) and a nitrogen evaporator (Oa-SYS Heating Device 5085, Organomation Associates. Inc., USA) were used.

Sampling

468 tea products including 174 leached teas, 155 liquid teas and 139 solid teas were collected from Korean offline markets in 2015. Each tea product was selected according to consumption data originating from the Korea National Health and Nutrition Examination Survey (KNHANES) and relating to the contents of PAHs from other research.

Sample extraction and clean-up

Leached tea and solid tea (powdered leached tea)

Each sample was homogenized using a blender (Mix-h03, Tongyang magic, China) and 5 g was weighed into a round-bottom flask. 200 µL of the internal standards d12-CHR and d12-BaP were added to the sample, and the mixture was saponified with 100 mL of 1 M potassium hydroxide in ethanol under reflux at 80 °C for 3 h. After allowing to cool 50 mL of *n*-hexane were added to the flask through a reflux condenser. The solution collected was transferred to another separatory funnel. Afterwards, the flask was rinsed with 50 mL of ethanol and *n*-hexane (1:1, v/v) and the rinsed solution was added to the separatory funnel. After the funnel was shaken, the hexane layer was transferred into another separatory funnel. The remaining water was extracted twice with *n*-hexane. The combined hexane layers were washed three times with 50 mL of distilled deionized water. Hexane layer was dried by anhydrous sodium sulfate and concentrated to 2 mL using the evaporator. The concentrate was loaded onto a silica cartridge previously activated by 10 mL of methylene

chloride and 20 mL of *n*-hexane and the cartridge was eluted with 10 mL of *n*-hexane and 20 mL of a mixture of *n*-hexane and dichloromethane (3:1, v/v). The solution eluted was concentrated to 2 mL under nitrogen at 20 psi and 40 °C. The concentrate was loaded onto a SPE-PAHs cartridge previously activated with 1 mL of *n*-hexane, and was purified by elution of 1 mL of *n*-hexane followed by 3 mL of ethyl acetate. The solution eluted was concentrated under nitrogen and was dissolved in 200 µL of methylene chloride for analysis by GC–MS.

Solid tea (processed) and liquid tea (preserved by sugar)

5 g of a homogenized sample was weighed in a separatory funnel and the internal standards 200 µL of d12-CHR and d12-BaP of 4 µg/L were added. 50 mL of a *N,N*-DMF and water solution (9:1, v/v) and 100 mL of *n*-hexane were added. The funnel was shaken vigorously. After equilibration the *N,N*-DMF and water layer (9:1, v/v) was transferred to another separatory funnel, and the hexane layer was extracted twice with 25 mL of the *N,N*-DMF and water solution (9:1, v/v). After the *N,N* and DMF-water layers were collected in a funnel, 100 mL of sodium sulfate solution (1%) and 50 mL of *n*-hexane were added and shaken vigorously. The hexane layer was separated into another separatory funnel and the *N,N* and DMF-water solution was extracted twice with 50 mL of *n*-hexane. The combined hexane layer was rinsed three times with 40 mL of distilled deionized water, and evaporated to 2 mL using a rotary evaporator, after being dried with anhydrous sodium sulfate. The concentrate was purified by silica cartridge as the aforementioned procedures for Leached tea and solid tea (powdered leached tea) and the final elute dried under nitrogen was dissolved in methylene chloride for analysis by GC–MS.

Liquid tea

A 100 g sample was weighed and 200 µL of internal standards were added to the sample. The sample was diluted with 100 mL of *n*-hexane or 50 mL *n*-hexane in a separatory funnel, and was washed three times with 50 mL of distilled deionized water. The hexane layer was concentrated and purified using the same procedures used for solid tea (processed) and liquid tea (preserved by sugar) for GC–MS analysis.

GC–MS analysis of PAHs

PAHs were determined using a gas chromatographer (Agilent Technology 7890A, USA) with a MS detector (Agilent Technology 5975C, USA) using a DB-5 ms column (30 m length × 0.25 mm inner diameter × 0.25 µm film thickness, Agilent Technology, USA). After being

held at 80 C° for 1 min, the oven temperature was increased to 245 C° at a rate of 4 C°/min, The temperature was raised to 270 C° at a rate of 30 C°/min and held for 10 min. The carrier gas, helium, was held at a constant flow of 1.5 mL/min. The injector was set at 320 C°, and the source and the quadrupole were at 250 and 180 C°, respectively. Split less mode was used for injection with 1 µL of analyte. The mass spectrometer was operated with the electron ionization (EI) of 70 eV and selective ion monitoring (SIM) modes. The retention times of BaA, CHR, BbF, BaP, d12-CHR, and d12-BaP were 40.63, 40.81, 45.22, 46.51, 40.67, and 46.41, respectively. The Quantifying ions were 228 for BaA and CHR, 252 for BbF and BaP, 240 for d12-CHR, and 264 for d12-BaP, and the two qualifying ions were 229 and 226 for BaA and CHR, 253 and 250 for BbF, and BaP, 241 and 236 for d12-CHR, and 265 and 260 for d12-BaP, respectively.

Quality control

The performance parameters of the methods used were verified to assure a quality of research according to guidelines recommended by Magnusson and Qrnemark (2014). Green tea, mixed tea and herbal tea were selected to validate representative tea products of leached tea, liquid tea and solid tea, respectively. Specificity, detection limit, quantification limit, linearity, and accuracy including precision and recovery, were obtained as the performance parameters. Specificity was confirmed by differentiating standard peaks from noise peaks in samples spiked with standards and internal standards. Limit of detection (LOD) was derived by multiplying the standard deviation calculated by analyzing 7 independent samples of 1 ng/L for leached, solid, and liquid tea (preserved by sugar) and samples of 0.4 ng/L for liquid tea by 3. Limit of Quantification (LOQ) was simply calcu-

regression equation of peak areas and PAHs concentrations, which range from 10 to 250 µg/L for leached, solid and liquid tea (preserved by sugar) to 50–1000 µg/L for liquid tea. Accuracy, including recovery and precision, was estimated by analyzing 5 samples with 4 standards and 2 internal standards. Recovery was calculated by comparing the fortified PAHs amount and the measured PAHs amount. Precision of repeatability was determined by the relative standard deviation (RSD) acquired on the accuracy tests.

Exposure estimation and Risk Characterization

The PAHs concentrations in teas and tea consumption amount were used to estimate the exposure to PAHs. For the tea consumption data, results of the Korea National Health and Nutrition Examination Survey (KNHANES) were used. The statistical approach recommended by GEMS/Food was applied to assume the concentration of not detected (ND) samples (GEMS/Food-EURO, 1995). When the proportion of ND samples was less than 60%, the values of the ND samples were assumed to be half of LOD, and when the proportion of ND samples were between 60 and 80% with over 25 quantified data or higher than 80%, the values are substituted to be zero and the value of LOD as lower-bounder (LB) and upper-bounder (UB), respectively. Total PAHs contents and total TEQ_{BaP} of 4 PAHs were estimated by summing every 4 PAHs concentrations and every TEQ_{BaP} of 4 PAHs according to the TEQ approach by multiplying BaA, CHR and BbF with their TEF of 0.1, 0.01, and 0.1, respectively (Nisbet and Lagoy, 1992).

The daily dietary exposure to 4 PAHs was estimated by multiplying concentration of total PAHs (or total TEQ_{BaP}) with daily consumption of tea and dividing with average body weight (Eq. 1).

Daily dietary exposure (ng kg⁻¹ b.w.day⁻¹)

$$= \frac{\text{total concentration of 4 PAHs(or TEQ}_{\text{BaP}}) \left(\frac{\text{ng}}{\text{g}} \right) \times \text{daily consumption} \left(\frac{\text{g}}{\text{day}} \right)}{\text{average body weight(kg)}} \quad (1)$$

lated by multiplying the LOD by 3. Linearity was verified by obtaining the determination coefficient (R^2) of the calibration curve. Calibration curves were obtained by calculating a

Equation (2) shows how the MOE is calculated.

$$\text{Margin of Exposure} = \frac{\text{BMDL}_{10} \left(\frac{\text{ng}}{\text{kg b.w.day}} \right)}{\text{The estimated daily dietary exposure} \left(\frac{\text{ng}}{\text{kg b.w.day}} \right)} \quad (2)$$

The MOE was calculated with the BMDL_{10} and the daily dietary exposure. The BMDL_{10} was obtained by calculating the 95% lower confidence limit of a dose showing a 10% incidence response collectively. BMDL_{10} of $70,000 \text{ ng kg}^{-1} \text{ b.w}^{-1} \cdot \text{day}^{-1}$ for BaP and $340,000 \text{ ng kg}^{-1} \text{ b.w}^{-1} \cdot \text{day}^{-1}$ for 4 PAHs were conservatively used (EFSA, 2008).

Results and discussion

Quality control

Specificity was confirmed by isolating standard and internal standard peaks of monitoring fragment ions from noise peaks in representative tea samples. The peaks of standards and internal standards were separated enough to isolate them from noise peaks in leached, solid and liquid teas (Fig. 1). The LOD and LOQ for 4 PAHs ranged from 0.01 to 0.32 and from 0.03 to 0.99 according to representative samples, and the calibration curve showed good linearity

Fig. 1 Chromatograms of samples fortified with 4 PAHs and isotope-labelled 2 PAHs at selected ion monitoring mode. (A) The quantifying ions of 4 PAHs in leached tea, (B) in solid tea, (C) in liquid tea

Table 1 Linearity, detection limit, recovery and repeatability of 4 PAHs in tea

PAHs	Conc. (ng g ⁻¹)	Linear equation	LOD (ng g ⁻¹)			LOQ (ng g ⁻¹)			Recovery (%)			RSD (%)		
			Leached tea	Liquid tea	Solid tea	Leached tea	Liquid tea	Solid tea	Leached tea	Liquid tea	Solid tea	Leached tea	Liquid tea	Solid tea
BaA	1 ^a (0.4) ^b	$y = 0.0057x - 0.0618^c$	0.32	0.01	0.07	0.96	0.03	0.21	105.0	102.5	100.8	1.78	0.00	0.44
	2 (1)	(R ² = 0.9997)							74.7	108.4	99.8	4.06	1.24	0.98
	10 (2)	$y = 0.0077x - 0.0396^d$							113.0	110.4	111.9	3.47	1.41	0.56
CHR	1 (0.4)	(R ² = 0.9996)												
	2 (1)	$y = 0.0066x - 0.0322^c$	0.30	0.01	0.14	0.90	0.03	0.42	109.0	108.0	94.4	3.24	1.04	0.95
	10 (2)	(R ² = 0.9999)							82.2	90.2	84.4	13.21	1.21	0.26
BbF	1 (0.4)	$y = 0.0120x + 0.2220^d$							95.7	87.0	94.5	0.58	0.70	0.97
	2 (1)	(R ² = 0.9979)												
	10 (2)	$y = 0.0038x - 0.0048^c$	0.33	0.01	0.20	0.99	0.03	0.60	75.4	106.5	87.8	0.73	1.29	1.25
BaP	1 (0.4)	(R ² = 0.9999)							93.5	74.0	73.5	5.28	0.00	3.47
	2 (1)	$y = 0.0076x + 0.0872^d$							97.2	71.7	76.3	6.29	2.98	2.02
	10 (2)	(R ² = 0.9993)												
BaP	1 (0.4)	$y = 0.0060x - 0.0030^c$	0.32	0.01	0.25	0.96	0.03	0.75	99.2	100.0	96.4	1.80	0.00	2.02
	2 (1)	(R ² = 0.9999)							82.8	73.4	76.4	14.36	0.75	1.17
	10 (2)	$y = 0.0100x + 0.0120^d$							92.7	73.3	87.4	2.34	0.61	0.81
		(R ² = 0.9997)												

^aConcentration of 4 PAHs fortified in leached tea and solid tea^bConcentration of 4 PAHs fortified in liquid tea^cCalibration curve for leached tea and solid tea^dCalibration curve for liquid tea

Table 2 Contents of 4 PAHs in various teas consumed in several countries

Type	Teas	No.	BaA (ng g ⁻¹)		CHR (ng g ⁻¹)		BbF (ng g ⁻¹)		BaP (ng g ⁻¹)		Total (ng g ⁻¹)		Country (references)
			Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	
Leached tea	Green	49	0.40	ND–4.74	2.52	ND–8.58	3.80	ND–9.88	4.12	ND–9.98	10.83	2.25–23.00	Korea (this study)
	Solomon's seal	20	1.83	ND–5.33	3.90	ND–8.27	6.22	1.37–14.90	5.39	1.17–9.16	17.34	5.32–28.60	
	Cassia seed	13	0.18	ND–1.24	0.94	ND–4.28	1.43	ND–5.31	1.99	ND–4.98	4.54	ND–14.14	
	Black	21	1.65	ND–6.75	3.04	0.94–7.78	2.89	ND–7.66	1.53	ND–4.49	9.11	3.31–20.08	
	Chrysanthemum	13	0.42	ND–1.90	2.19	ND–4.75	4.96	1.36–9.56	5.02	1.90–9.68	12.59	3.37–22.03	
	Mate	10	6.51	1.71–8.91	9.35	2.66–15.22	8.19	4.17–12.39	9.40	3.76–12.45	33.45	12.30–44.25	
	Corn	9	0.69	ND–5.23	1.24	ND–3.57	3.67	ND–8.68	3.04	1.17–8.43	8.65	1.52–18.96	
	Buckwheat	9	0.39	ND–1.82	1.83	ND–3.67	3.52	ND–8.28	3.31	ND–8.14	9.05	ND–20.99	
	Burdock	8	0.29	ND–1.18	1.31	ND–2.83	3.93	ND–8.19	2.44	ND–3.76	7.97	2.83–12.60	
	Matrimony vine	7	ND	ND	0.72	ND–1.81	ND	ND	0.36	ND–1.27	1.07	ND–2.89	
Liquid tea	Dandelion	5	0.60	ND–1.77	4.45	2.56–6.96	3.54	1.25–9.22	3.88	ND–9.35	12.46	7.26–22.06	USA (Adisa et al. 2015)
	Brown rice	5	0.64	ND–1.73	2.19	ND–4.23	1.54	ND–3.55	3.12	1.87–4.51	7.49	3.93–9.65	
	Mulberry leaves	5	ND	ND	1.20	ND–2.19	2.68	1.97–3.77	0.23	ND–1.17	4.12	1.97–7.14	
	Green	13	0.07	0.06–0.13	ND	ND	0.01	ND–0.06	0.01	ND–0.10	0.08	0.06–0.30	
	Barley	37	0.07	0.06–0.13	ND	ND	0.03	ND–0.07	0.01	ND–0.11	0.11	0.06–0.30	
	Black	26	0.08	0.06–0.14	ND	ND	0.01	ND–0.06	0.01	ND–0.11	0.10	0.06–0.31	
	Preserved fruit	40	ND	ND	0.03	ND–0.74	0.04	ND–0.29	0.05	ND–0.34	0.12	ND–0.74	
	Ginger	11	ND	ND	ND	ND	ND	ND	0.03	ND–0.29	0.03	ND–0.29	
	Korean raisin	10	0.11	0.07–0.13	ND	ND	0.05	0.04–0.08	0.05	ND–0.11	0.22	0.13–0.31	
	Corn	7	0.11	0.08–0.13	ND	ND	0.06	0.04–0.07	0.08	ND–0.10	0.24	0.19–0.30	
Solid tea	Jujube	6	ND	ND	ND	ND	0.04	ND–0.26	ND	ND	0.04	ND–0.26	USA (Adisa et al. 2015)
	Burdock	5	0.07	0.06–0.07	ND	ND	ND	ND	0.01	ND–0.04	0.08	0.06–0.11	
	Adlay	22	0.19	ND–0.64	0.16	ND–0.86	0.16	ND–1.59	ND	ND	0.51	ND–2.47	
	Black	22	0.10	ND–0.59	0.06	ND–0.73	0.03	ND–0.72	ND	ND	0.19	ND–1.32	
	Ginger	22	0.23	ND–0.62	0.21	ND–0.71	0.03	ND–0.72	ND	ND	0.48	ND–1.32	
	Herbal	60	0.24	ND–0.95	0.24	ND–1.50	0.13	ND–1.89	ND	ND	0.65	ND–5.48	
	Green	13	0.25	ND–1.78	3.44	1.01–5.11	3.26	ND–5.94	1.47	ND–3.04	8.42	4.57–12.70	
	Earl	5	5.40	1–11	135.80	95–240	3.80	1–8	7.60	2–14	152.60	104–273	
	Fine	3	5.00	5–5	127.67	50–214	3.33	1–5	6.33	3–10	140.67	54–234	
	Black	6	5.80	2–18	136.83	57–365	4.20	1–14	7.33	2–29	152.50	63–426	
Leached tea	Chinamom and Nutmeg	5	1.25	1–2	40.20	27–55	1.33	1–2	2.20	2–3	44.20	30–59	USA (Adisa et al. 2015)
	Green	9	3.00	1–6	94.11	17–240	2.83	1–5	6.50	2–12	103.78	17.263	

Table 2 continued

Type	Teas	No.	BaA (ng g ⁻¹)		CHR (ng g ⁻¹)		BbF (ng g ⁻¹)		BaP (ng g ⁻¹)		Total (ng g ⁻¹)		Country (references)
			Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	
'Leached tea	Green	11	12.31	1.8–40.4	25.84	6.7–61.5	11.45	2.2–33.4	9.12	1.6–32.6	58.72	12.3–167.9	Germany (Ziegenhals et al. 2008)
	Mate	8	147.05	38.4–374.6	276.20	81.9–746.3	105.21	34.1–258.1	106.55	24.8–236.5	635.01	1846–1615.5	
	Black	11	5.36	1.3–13.1	10.08	3.4–18.1	4.27	1.5–8.1	4.94	0.8–14.1	24.65	9.0–44.6	
	Herbal/fruit	7	2.44	1.2–3.5	7.57	4–11.6	2.43	1.3–4.5	1.67	0.8–3.1	14.11	7.8–21.5	
	White	3	38.23	14.1–80.1	49.50	19–95.1	25.93	14.5–44.6	15.17	11.4–19	128.83	59.0–238.8	
Leached tea	Black	26	30.60	1.1–360	24.08	2–220	23.80	1.2–240	25.62	0.6–330	105.95	4.9–1200	Several countries (Schulz et al. 2015)
	Green	15	22.33	0–130	22.96	0–120	19.49	0–85	16.78	0–97	81.73	0–430	
	Oolong	6	18.20	5.7–30	16.70	6.6–28	13.83	4.4–23	10.47	2.7–19	59.17	21–99	
	Pu erh	3	57.67	43–72	55.33	46–73	31.33	26–36	30.33	25–37	176.67	140–220	
	White	3	16.03	5.1–23	14.50	8.5–20	11.67	7–16	5.20	2.4–6.8	47.33	23–65	
	Lapsang souchong	3	466.67	240–620	546.67	270–700	111.33	64–150	346.67	280–460	1433.33	1000–1700	
	Green	2	8.60	6.99–10.2	19.65	18.2–21.1					28.25	25.5–31.3	
Leached tea	Oolong	1	4.43		5.96				5.10		15.49		China (Lin et al. 2005)
	Black	1	175.00		241.00		37.60		39.70		493.3		
	Puerh	1	13.40		23.00		8.03		7.84		52.27		
	Brick	1	31.90		40.30		12.90		14.60		99.7		
	Jasmine	1	67.30		45.90		54.00		28.10		195.3		
	Kuding	1	7.70		17.40						25.1		
	Sage	5	12.93	0.31–45.19	2.54	0.17–6.02	1.37	0.06–3.12	0.78	0.03–1.38	17.62		
	Melissa	5	0.45	0.08–0.71	3.22	0.64–8.18	0.81	0.57–1.08	0.29	0.19–0.38	4.77		
	Marjoram	5	1.38	0.1–3.31	7.87	2.12–10.64	1.77	0.76–4.15	0.97	0.41–2.09	11.99		
	Rosmary	4	10.43	2.88–24.55	12.24	1.24–39.1	1.17	0.39–2.03	0.68	0.14–1.52	24.52		
Leached tea	Wild thyme	5	1.19	0.11–2.19	1.29	0.23–2.46	0.47	0.29–0.73	0.49	0.12–1.76	3.44		Syria (Krajić and Odeh 2013)
	Mint	5	1.34	0.22–4.24	2.94	0.24–7.7	0.6	0.28–0.98	0.49	0.02–1.59	5.37		
	Holzhock	4	0.94	0.32–2	2.25	1.01–4.45	2.1	0.29–5.06	0.22	0.02–0.39	5.51		
	Chamomile	5	1.67	0.17–4.04	1.25	0.47–2.25	0.9	0.11–1.83	0.17	0.07–0.32	3.99		
	Damask rose	4	0.34	0.18–0.76	1.16	0.55–2.14	0.65	0.16–1.15	0.41	0.19–0.99	2.56		
	Roselle	5	0.27	0.1–0.62	0.78	0.04–1.54	0.17	0.04–0.28	0.08	0.03–0.13	1.3		
	Yerba mate	8	72.83	24.5–99.9	121.44	42.3–169	51.03	13.3–76.3	37.13	8.03–53.3	282.42	88.1–397.7	
	Leached tea												
	Yerba mate	50	28.7		67.6		21.9		26.9		145.1		
	Leached tea												

Table 3 Estimation of dietary exposure to each 4 PAHs for whole population

Type	Teas	Daily consumption (g day ⁻¹)		Estimated exposure (ng kg ⁻¹ b.w. day ⁻¹)							
		Mean	P95 ^a	BaA				CHR			
				Mean		P95		Mean		P95	
				LB ^b	UB ^c	LB	UB	LB	UB	LB	UB
Leached tea	Green	0.04	–	4.80×10^{-4}	4.80×10^{-4}			1.79×10^{-3}	1.79×10^{-3}		
	Solomon's seal	0.02	–	6.48×10^{-4}	6.48×10^{-4}			1.36×10^{-3}	1.36×10^{-3}		
	Chrysanthemum	0.04×10^{-2}	–	0.05×10^{-4}	0.05×10^{-4}	–	–	0.15×10^{-4}	0.15×10^{-4}	–	–
	Mate	0.04	–	4.47×10^{-3}	4.47×10^{-3}	–	–	6.42×10^{-3}	6.42×10^{-3}	–	–
Liquid tea	Green	11.84	–	1.42×10^{-2}	1.42×10^{-2}	–	–		2.03×10^{-3}	–	–
	Barley	0.87	–	1.05×10^{-3}	1.05×10^{-3}	–	–		1.49×10^{-4}		
	Black	0.66	–	9.06×10^{-4}	9.06×10^{-4}	–	–		1.13×10^{-4}		
	Preserved fruit	0.12	–	1.85×10^{-4}	7.41×10^{-4}	–	–	3.29×10^{-4}	3.29×10^{-4}		
Solid tea	Adlay	0.09	–	4.01×10^{-4}	4.01×10^{-4}	–	–	4.79×10^{-4}	4.79×10^{-4}		
	Black	0.03	–	1.03×10^{-4}	1.03×10^{-4}	–	–	0.87×10^{-4}	0.87×10^{-4}		
	Ginger	0.02	–	0.99×10^{-4}	0.99×10^{-4}	–	–	1.20×10^{-4}	1.20×10^{-4}		
	Herbal	0.04	–	1.99×10^{-4}	1.99×10^{-4}	–	–	2.54×10^{-4}	2.54×10^{-4}		
Total	Green	0.04	–	4.80×10^{-4}	4.80×10^{-4}	–	–	2.36×10^{-3}	2.36×10^{-3}		
					2.38×10^{-2}			1.32×10^{-2}	1.55×10^{-2}		

Type	Teas	Daily consumption (g day ⁻¹)		Estimated exposure (ng kg ⁻¹ b.w. day ⁻¹)							
		Mean	P95 ^a	BbF				BaP			
				Mean		P95		Mean		P95	
				LB	UB	LB	UB	LB	UB	LB	UB
Leached tea	Green	0.04	–	2.63×10^{-3}	2.63×10^{-3}			2.83×10^{-3}	2.83×10^{-3}		
	Solomon's seal	0.02	–	2.13×10^{-3}	2.13×10^{-3}			1.85×10^{-3}	1.85×10^{-3}		
	Chrysanthemum	0.04×10^{-2}	–	0.34×10^{-4}	0.34×10^{-4}			0.34×10^{-4}	0.34×10^{-4}		
	Mate	0.04	–	5.62×10^{-3}	5.62×10^{-3}			6.45×10^{-3}	6.45×10^{-3}		
Liquid tea	Green	11.84	–	2.03×10^{-3}	2.03×10^{-3}			6.09×10^{-3}	6.09×10^{-3}		
	Barley	0.87	–	5.97×10^{-4}	5.97×10^{-4}			4.48×10^{-4}	4.48×10^{-4}		
	Black	0.66	–	2.26×10^{-4}	2.26×10^{-4}			3.40×10^{-4}	3.40×10^{-4}		
	Preserved fruit	0.12	–	4.12×10^{-4}	4.12×10^{-4}			4.73×10^{-4}	4.73×10^{-4}		
Solid tea	Adlay	0.09	–	5.87×10^{-4}	5.87×10^{-4}			2.47×10^{-4}	2.47×10^{-4}		
	Black	0.03	–	0.98×10^{-4}	0.98×10^{-4}			0.31×10^{-4}	1.39×10^{-4}		
	Ginger	0.02	–	0.89×10^{-4}	0.89×10^{-4}			0.14×10^{-4}	0.93×10^{-4}		
	Herbal	0.04	–	2.40×10^{-4}	2.40×10^{-4}			0.96×10^{-4}	2.20×10^{-4}		
Total	Green	0.04	–	2.29×10^{-3}	2.29×10^{-3}			1.17×10^{-3}	1.17×10^{-3}		
				1.70×10^{-2}	1.70×10^{-2}			2.01×10^{-2}	2.04×10^{-2}		

^aDaily intake of teas at the 95th percentile^bLower bound: the left censored data are regarded as zero^cUpper bound: the left censored data are regarded as LOD

with a correlation coefficient of over 0.99. The recoveries of 4 PAHs were 74.7–113.0% for BaA, 82.2–109.0% for CHR, 71.7–106.5% for BbF and 73.3–100.0% for BaP and the RSD ranged from 0.00 to 4.06% for BaA, from 0.26 to 13.21% for CHR, from 0.00 to 6.29% for BbF and from

0.00 to 14.36% for BaP (Table 1). Recovery should be between 40 and 120% and RSD should be lower than 15% at 10 ng g⁻¹, 20% at 2 ng g⁻¹, 22% at 1 ng g⁻¹, 25% at 0.4 ng g⁻¹, respectively, according to the criteria of the Association of Official Agricultural Chemists (AOAC)

Table 4 Estimation of dietary exposure to each 4 PAHs for drinker only

Type	Teas	Daily consumption (g day ⁻¹)		Estimated exposure (ng kg ⁻¹ b.w. day ⁻¹)					
		P95 ^a		BaA		CHR			
		Mean	P95 ^a	Mean	P95	Mean	LB	UB	P95
Leached tea	Green	2.80	7.50	3.36 × 10 ⁻²	3.36 × 10 ⁻²	9.01 × 10 ⁻²	1.25 × 10 ⁻¹	1.25 × 10 ⁻¹	3.36 × 10 ⁻¹
	Solomon's seal	7.60	24.00	2.46 × 10 ⁻¹	2.46 × 10 ⁻¹	7.78 × 10 ⁻¹	5.18 × 10 ⁻¹	5.18 × 10 ⁻¹	1.63
	Chrysanthemum	1.50	3.00	1.83 × 10 ⁻²	1.83 × 10 ⁻²	3.65 × 10 ⁻²	5.69 × 10 ⁻²	5.69 × 10 ⁻²	1.14 × 10 ⁻¹
	Mate	2.80	7.50	3.13 × 10 ⁻¹	3.13 × 10 ⁻¹	8.37 × 10 ⁻¹	4.49 × 10 ⁻¹	4.49 × 10 ⁻¹	1.20
	Green	357.40	960.00	4.29 × 10 ⁻¹	4.29 × 10 ⁻¹	1.15	–	6.13 × 10 ⁻²	–
Liquid tea	Barley	356.00	800.00	4.27 × 10 ⁻¹	4.27 × 10 ⁻¹	9.61 × 10 ⁻¹	–	6.11 × 10 ⁻²	–
	Black	274.90	600.00	3.77 × 10 ⁻¹	3.77 × 10 ⁻¹	8.23 × 10 ⁻¹	–	4.72 × 10 ⁻²	–
	Preserved fruit	41.00	73.50	6.32 × 10 ⁻²	2.53 × 10 ⁻¹	1.13 × 10 ⁻¹	1.13 × 10 ⁻¹	1.13 × 10 ⁻¹	2.02 × 10 ⁻¹
Solid tea	Adlay	20.00	40.00	8.92 × 10 ⁻²	8.92 × 10 ⁻²	1.78 × 10 ⁻¹	1.06 × 10 ⁻¹	1.06 × 10 ⁻¹	2.13 × 10 ⁻¹
	Black	17.30	42.00	5.93 × 10 ⁻²	5.93 × 10 ⁻²	1.44 × 10 ⁻¹	5.04 × 10 ⁻²	5.04 × 10 ⁻²	1.22 × 10 ⁻¹
	Ginger	10.10	22.10	5.02 × 10 ⁻²	5.02 × 10 ⁻²	1.10 × 10 ⁻¹	6.06 × 10 ⁻²	6.06 × 10 ⁻²	1.33 × 10 ⁻¹
	Herbal	23.00	31.20	1.14 × 10 ⁻¹	1.14 × 10 ⁻¹	1.55 × 10 ⁻¹	1.46 × 10 ⁻¹	1.46 × 10 ⁻¹	1.98 × 10 ⁻¹
	Green	2.80	7.50	3.36 × 10 ⁻²	3.36 × 10 ⁻²	9.01 × 10 ⁻²	1.65 × 10 ⁻¹	1.65 × 10 ⁻¹	4.43 × 10 ⁻¹
Total		2.25	2.44	5.47	5.81	1.79	1.96	1.96	5.00
Type	Teas	Daily consumption (g day ⁻¹)		Estimated exposure (ng kg ⁻¹ b.w. day ⁻¹)					
		P95 ^a		BbF		BaP			
		Mean	P95 ^a	Mean	P95	Mean	LB	UB	P95
Leached tea	Green	2.80	7.50	1.84 × 10 ⁻¹	1.84 × 10 ⁻¹	4.93 × 10 ⁻¹	1.98 × 10 ⁻¹	1.98 × 10 ⁻¹	5.31 × 10 ⁻¹
	Solomon's seal	7.60	24.00	8.11 × 10 ⁻¹	8.11 × 10 ⁻¹	2.56	7.03 × 10 ⁻¹	7.03 × 10 ⁻¹	2.22
	Chrysanthemum	1.50	3.00	1.28 × 10 ⁻¹	1.28 × 10 ⁻¹	2.55 × 10 ⁻¹	1.29 × 10 ⁻¹	1.29 × 10 ⁻¹	2.58 × 10 ⁻¹
	Mate	2.80	7.50	3.93 × 10 ⁻¹	3.93 × 10 ⁻¹	1.05	4.51 × 10 ⁻¹	4.51 × 10 ⁻¹	1.21
	Green	357.40	960.00	6.13 × 10 ⁻²	6.13 × 10 ⁻²	1.65 × 10 ⁻¹	1.84 × 10 ⁻¹	1.84 × 10 ⁻¹	4.94 × 10 ⁻¹
Liquid tea	Barley	356.00	800.00	2.44 × 10 ⁻¹	2.44 × 10 ⁻¹	5.49 × 10 ⁻¹	1.83 × 10 ⁻¹	1.83 × 10 ⁻¹	4.12 × 10 ⁻¹
	Black	274.90	600.00	9.43 × 10 ⁻²	9.43 × 10 ⁻²	2.06 × 10 ⁻¹	1.41 × 10 ⁻¹	1.41 × 10 ⁻¹	3.09 × 10 ⁻¹
	Preserved fruit	41.00	73.50	1.41 × 10 ⁻¹	1.41 × 10 ⁻¹	2.52 × 10 ⁻¹	1.62 × 10 ⁻¹	1.62 × 10 ⁻¹	2.90 × 10 ⁻¹
Solid tea	Adlay	20.00	40.00	1.30 × 10 ⁻¹	1.30 × 10 ⁻¹	2.61 × 10 ⁻¹	5.49 × 10 ⁻²	5.49 × 10 ⁻²	1.10 × 10 ⁻¹
	Black	17.30	42.00	5.64 × 10 ⁻²	5.64 × 10 ⁻²	1.37 × 10 ⁻¹	1.78 × 10 ⁻²	8.01 × 10 ⁻²	4.32 × 10 ⁻²
	Ginger	10.10	22.10	4.50 × 10 ⁻²	4.50 × 10 ⁻²	9.86 × 10 ⁻²	6.93 × 10 ⁻³	4.68 × 10 ⁻²	1.52 × 10 ⁻¹
	Herbal	23.00	31.20	1.38 × 10 ⁻¹	1.38 × 10 ⁻¹	1.87 × 10 ⁻¹	5.52 × 10 ⁻²	1.26 × 10 ⁻¹	7.49 × 10 ⁻²
	Green	2.80	7.50	3.36 × 10 ⁻²	3.36 × 10 ⁻²	9.01 × 10 ⁻²	1.65 × 10 ⁻¹	1.65 × 10 ⁻¹	4.43 × 10 ⁻¹

Table 4 continued

Type	Teas	Daily consumption (g day ⁻¹)		Estimated exposure (ng kg ⁻¹ b.w. day ⁻¹)														
		P95 ^a		BbF				BaP										
		Mean		Mean		P95		Mean		P95								
				LB	UB	LB	UB	LB	UB	LB	UB							
Green	2.80	7.50			1.60 × 10 ⁻¹		1.60 × 10 ⁻¹		4.28 × 10 ⁻¹		4.28 × 10 ⁻¹		8.16 × 10 ⁻²		8.16 × 10 ⁻²		2.19 × 10 ⁻¹	
			2.59	2.59	6.64	6.64	2.37	2.54	6.19	6.52								
Total																		

^aDaily intake of teas at the 95th percentile^bLower bound: the left censored data are regarded as zero^cUpper bound: the left censored data are regarded as LOD

(Taverniers et al., 2004). All performance parameters satisfied the criteria to control the quality of analytical results.

Occurrence of 4 PAHs in teas

Table 2 shows the average and the range of concentrations of BaA, CHR, BbF, BaP and the sum of these 4 PAHs in teas. Contents lower than the LOQ were shown as ND. Leached teas contained high levels of PAHs, and liquid and solid tea contain low levels of PAHs except for solid green tea. The average levels of PAHs in teas were 0.47 ng g⁻¹ of BaA, 1.18 ng g⁻¹ of CHR, 1.56 ng g⁻¹ of BbF, 1.42 ng g⁻¹ of BaP, resulting in 4.63 ng g⁻¹ of the average sum of 4 PAHs. The average BaP levels were 11.36 ng g⁻¹ in leached tea, 0.11 ng g⁻¹ in liquid tea and 1.25 ng g⁻¹ in solid tea, and the average sum of 4 PAHs levels were 3.69 ng g⁻¹ in leached tea, 0.03 ng g⁻¹ in liquid tea and 0.14 ng g⁻¹ in solid tea. The most contaminated tea was mate tea of leached tea with a total of 33.45 ng g⁻¹ for all 4 PAHs, which is about 7.5 fold higher than the average total concentration of all 4 PAHs in teas. Solomon's seal tea was the second most highly contaminated with a total of 17.34 ng g⁻¹ for all 4 PAHs, followed by chrysanthemum and dandelion teas. The average levels of 4 PAHs in liquid and solid teas were very low under 0.65 ng g⁻¹ except for solid green tea. These differences can be affected by different processing procedures used to manufacture these teas. Liquid tea is extracted from the leached tea with water, and solid tea, except for green tea, is extracted from leached tea, dried, and pulverized. PAHs are so lipophilic that they would not be extracted into water by extracting leached tea when making liquid and solid tea (Lin and Zhu, 2006; Vieira et al., 2010). Meanwhile, solid green tea is only made by pulverizing leached tea. Therefore, solid green tea contains high amounts of PAHs. Table 4 shows the PAHs content of leached teas. The PAHs contents of Korean leached teas were lower than those of other countries. Mate teas consumed in Germany and Brazil contained higher levels of 4 PAHs than other teas (Kamangar et al., 2008; Ziegenhals et al., 2008).

Selection of consumption data of tea from KNHANES

Tea consumption data of the whole population and tea drinkers only were produced from the second and third programs of KNHANES IV (2007–2009) and the first program of KNHANES V (2010–2012). The second and the third programs in 2008 and 2009 surveyed 9308 and 10,078 sample populations, respectively, and the first program in 2010 chose 8473 sample populations (CDC, 2008, CDC, 2009, CDC, 2010). Green, solomon's seal, chrysanthemum and mate tea in the leached tea variety

Table 5 Estimation of dietary exposure to total PAHs

Type	Teas	Estimated exposure (ng kg ⁻¹ b.w. day ⁻¹)							
		Whole population				Drinker only			
		Mean		P95 ^a		mean		P95	
		LB ^b	UB ^c	LB	UB	LB	UB	LB	UB
Leached tea	Green	7.74×10^{-3}	7.74×10^{-3}	–	–	5.42×10^{-1}	5.42×10^{-1}	1.45	1.45
	Solomon's seal	5.99×10^{-3}	5.99×10^{-3}	–	–	2.28	2.28	7.19	7.19
	Chrysanthemum	0.88×10^{-4}	0.88×10^{-4}	–	–	3.32×10^{-1}	3.32×10^{-1}	6.63×10^{-1}	6.63×10^{-1}
	Mate	2.30×10^{-2}	2.30×10^{-2}	–	–	1.61	1.61	4.30	4.30
Liquid tea	Green	2.23×10^{-2}	2.44×10^{-2}	–	–	6.74×10^{-1}	7.36×10^{-1}	1.81	1.98
	Barley	2.09×10^{-3}	2.24×10^{-3}	–	–	8.55×10^{-1}	9.16×10^{-1}	1.92	2.06
	Black	1.47×10^{-3}	1.59×10^{-3}	–	–	6.13×10^{-1}	6.60×10^{-1}	1.34	1.44
	Preserved fruit	1.38×10^{-3}	1.96×10^{-3}	–	–	4.71×10^{-1}	6.68×10^{-1}	8.45×10^{-1}	1.20
Solid tea	Adlay	1.73×10^{-3}	1.73×10^{-3}	–	–	3.84×10^{-1}	3.84×10^{-1}	7.68×10^{-1}	7.68×10^{-1}
	Black	3.14×10^{-4}	4.27×10^{-4}	–	–	1.81×10^{-1}	2.46×10^{-1}	4.39×10^{-1}	5.98×10^{-1}
	Ginger	3.22×10^{-4}	4.01×10^{-4}	–	–	1.63×10^{-1}	2.03×10^{-1}	3.56×10^{-1}	4.44×10^{-1}
	Herbal	7.96×10^{-4}	9.13×10^{-4}	–	–	4.58×10^{-1}	5.25×10^{-1}	6.21×10^{-1}	7.12×10^{-1}
	Green	6.29×10^{-3}	6.29×10^{-3}	–	–	4.40×10^{-1}	4.40×10^{-1}	1.18	1.18
Total		7.35×10^{-3}	7.68×10^{-2}			9.00	9.54	22.9	24.0

^aDaily intake of teas at the 95th percentile^bLower bound: the left censored data are regarded as zero^cUpper bound: the left censored data are regarded as LOD

were surveyed in KNHANES and the consumption amounts were obtained. Meanwhile, cassia seed, black, corn, buckwheat, burdock, matrimony vine, dandelion, brown rice and mulberry leaves tea were not included in KNHANES and the consumption amounts were not obtained. Therefore, green, solomon's seal, chrysanthemum and mate tea were selected for obtaining estimate exposure to PAHs. For the liquid tea variety, the consumption amounts of green, barley, black and preserved fruit tea were obtained from KNHANES and used for exposure estimation. Ginger, Korean raisin, corn, jujube and burdock tea were monitored but not used for risk assessment. Consumption amounts for all solid teas such as adlay, black, ginger, herbal and green tea were obtained from KNHANES and used for risk assessment. The consumption amounts of all teas at 95th percentile are zero because Koreans consume tea a lot in a single time, but not frequently.

Statistical treatment of not-detected samples

The percentages of ND sample of BaA, CHR, BbF and BaP in all leached teas were less than 60%, therefore, the values of ND samples were substituted with half of the LOD. Meanwhile, CHR for green, barley and black tea in the liquid tea variety were not detected at all, and were replaced with 0 for LB and LOD for UB. The percentage of

sample containing BaA, BbF and BaP were less than 60% and replaced with half of the LOD. BaA for preserved fruit tea was changed to 0 for LB and the LOD for UB, and half of the LOD was used for the other 3 PAHs for preserved fruit tea. For solid tea, BaP for black, herbal and ginger were substituted with 0 for LB and the LOD for UB, and half of the LOD was used for the other 3 PAHs. All samples of green tea contained all 4 PAHs.

Exposure estimation

The estimated exposure to PAHs through drinking tea was obtained by using the contents of 4 PAHs that are statistically treated and the tea consumption amount obtained from KNHANES. Table 3 shows the estimated exposure to each of the 4 PAHs for the whole population.

On average, people were exposed to BaA, CHR, BbF and BaP through tea consumption from 2.32×10^{-2} (LB) to 2.38×10^{-2} (UB) ng kg⁻¹ b.w⁻¹. day⁻¹, 1.32×10^{-2} (LB) to 1.55×10^{-2} (UB) ng kg⁻¹ b.w⁻¹. day⁻¹, 1.70×10^{-2} ng kg⁻¹ b.w⁻¹. day⁻¹ and 2.01×10^{-2} (LB) to 2.04×10^{-2} (UB) ng kg⁻¹ b.w⁻¹. day⁻¹, respectively. Meanwhile, people only consuming tea were exposed on average to BaA from 2.25 (LB) to 2.44 (UB) ng kg⁻¹ b.w⁻¹. day⁻¹, CHR from 1.79 (LB) to 1.96 (UB) ng kg⁻¹ b.w⁻¹. day⁻¹, BbF of 2.59 ng kg⁻¹ b.w⁻¹. day⁻¹ and BaP from 2.37 (LB) to 2.54 (UB) ng kg⁻¹ b.w⁻¹. day⁻¹

(Table 4). The estimated mean exposures to all 4 PAHs were from 7.35×10^{-3} (LB) to 7.68×10^{-2} (UB) $\text{ng kg}^{-1} \text{ b.w}^{-1} \text{ day}^{-1}$ for the whole population and from 9.00 (LB) to 9.54 (UB) $\text{ng kg}^{-1} \text{ b.w}^{-1} \text{ day}^{-1}$ for tea drinkers only (Table 5). People were mainly exposed to PAHs by mate leached tea (30.0–31.1%) and liquid green tea (30.3–31.8%). Meanwhile, Solomon's seal leached tea mainly contributed to exposure to PAHs to tea drinkers only (23.9–25.3%). Mate leached tea and liquid green tea are main sources of exposure to PAHs for the whole population because mate tea is highly contaminated with PAHs and liquid green tea is highly consumed. Solomon's seal tea is highly consumed but not often. As the result of exposure estimation, people are mostly exposed to PAHs by liquid tea, because people consume a number of teas. Therefore, liquid tea must be carefully produced so as not to be contaminated with PAHs, although liquid tea is not highly contaminated with PAHs.

Risk characterisation and uncertainty

The MOEs of the total of 4 PAHs in teas for the whole population were 4.63×10^6 (LB) and 4.43×10^6 (UB). And those for tea drinker only were 3.78×10^4 (LB) and 3.57×10^4 (UB). The MOEs were over 10,000, and risk of exposure to all 4 PAHs in consuming these teas are of "low concern from a public health point of view" (EFSA, 2005). The priority of the risk of PAHs in teas is low and risk managers should therefore focus on other hazards that are of high concerns to the public in food.

The exposures to PAHs would be under- or over-estimated because of the uncertainty of PAHs contents caused by the left censored data. The consumption data of teas obtained from 24 h recall survey would also lead to the uncertainty of exposure. The risk assessment of low concern to public health could be changed when specific sensitive groups are exposed to PAHs and non-dietary sources are considered. Importantly, tea contains numerous beneficial elements like catechins, so the further study is needed to understand the risk with benefits of drinking tea.

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