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Date:	18th, March 2016	

STATEMENT on Polycyclic Aromatic Hydrocarbons (PAHs)

EC Regulation 2015/1933 introduces new limits in relation to the PAHs for some categories of products for which the practical application involves some interpretative difficulties if not connected to the history of this Regulation.
The classifications "Tea and herbal infusion" were included in a first draft of the Regulations in 2014 project (see attached working document SANCO / 12420/2014 points (5) and Annex 6.1.13) .
The comments received from various associations had as a result an updated version in 2015 (see attached working document SANCO / 10381/2015), where tea and herbal teas are no longer mentioned.
In support of that decision there are also data of a scientific publication, where experimental results show that PAHs are not extracted in water (see attached document, requested by the European Tea Committee (ETC) and the European Association of Herbal Infusions (EHIA).
In addition to the above, the associations are of the opinion that herbal teas are not included in the category "dried herbs" or "dried spices" because these groups of products are belonging to the category “Herbal infusion”.
It could be also considered that the consumer exposure is referred to the product "consumed / ready to drink", that is the brew, the infusion or the water-alcohol preparation.
In the end we believe that in virtue of different PAH extraction mechanisms in liquid solutions, products intended for infusion or extraction are not affected by this Regulation.
For what concerns botanicals for food supplements and herbs for direct ingestion, the situation is different and well-defined (point 6.1.14 Dried herbs), for which the limits in the Regulation are clearly stated.

Martin Bauer S.p.a.

This statement replaces all our previous editions.
This statement is transmitted electronically and is valid without signature.

Martin Bauer Plantextrakt Finzelberg

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Version 1
Page 1 of 25
Issued: 18.03.2016

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EUROPEAN
COMMISSION

Brussels, XXX
SANCO/12420/2014
[…](2014) XXX draft

COMMISSION REGULATION (EU) No …/..

of XXX

amending Regulation (EC) No 1881/2006 as regards maximum levels for polycyclic aromatic hydrocarbons in foodstuffs

(Text with EEA relevance)

COMMISSION REGULATION (EU) No …/..

of XXX

amending Regulation (EC) No 1881/2006 as regards maximum levels for polycyclic aromatic hydrocarbons in foodstuffs

(Text with EEA relevance)

WORKING DOCUMENT – DOES NOT NECESSARILY REPRESENT THE VIEWS OF THE COMMISSION SERVICES

THE EUROPEAN COMMISSION,

Having regard to the Treaty on the Functioning of the European Union,

Having regard to Council Regulation (EEC) No 315/93 of 8 February 1993 laying down Community procedures for contaminants in food[1], and in particular Article 2(3) thereof,

Whereas:

(1) Commission Regulation (EC) No 1881/2006[2] sets maximum levels for polycyclic aromatic hydrocarbons (PAHs) in food

(2) According to that Regulation, maximum levels for PAHs must be safe and as low as reasonably achievable (ALARA) based upon good manufacturing, drying and agricultural/fishery practices. In 2011, data for smoked fish have shown that lower maximum levels were achievable. Nevertheless, adaptations of smoking technology were necessary in some cases. Therefore transition period of three years was granted before the lower maximum levels become applicable as from 1 September 2014.

(3) *Katsuobushi* is a traditional Japanese food product made from bonito. Its manufacturing process involves filleting, boiling and deboning followed by smoking/drying process over combusting woods. Recent evidence has been provided by the Japanese authorities demonstrating that, despite the application of good smoking practices to the extent possible, the lower levels for PAHs are not achievable. Therefore it is appropriate to provide that the maximum levels applicable before 1 September 2014, should continue to apply to *Katsuobushi.*

(4) The product name "*Sprotid*" is a general traditional name in Estonia for a product which traditionally can contain both sprat (*Sprattus sprattus)* and Baltic herring (*Clupea harengus membras*) depending on the season and availability. Both fishes are of comparable size and are classified as small scale fish. The label of "*Sprotid*" mentions if the product contains sprats or Baltic herring or a mixture, with the ratio of each fish species present. The smoking procedure for this small Baltic herring is the

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[1] OJ L 37, 13.2.1993, p. 1.

[2] Commission Regulation (EC) No 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs (OJ L 364, 20.12.2006, p. 5).

same as the one applied to sprats and consequently levels of PAHs in small Baltic herring are similar to those found in smoked sprat. Therefore it is appropriate to establish he same maximum level for small Baltic herring as for smoked sprats and canned smoked sprats.

(5) High levels of PAH have been found in some food supplements. The presence of high levels in certain food supplements have been linked to the presence of botanical ingredients. Also high levels of PAH have been found in herbs and certain spices as well in tea and herbal infusions. The source of the presence of high levels of PAH in these products have been identified to be the bad drying practices and these high levels are avoidable by applying good practices. It is therefore appropriate to establish a maximum level for PAH in these products which is achievable by applying good drying practices and which ensures a high level of human health protection;

(6) The measures provided for in this Regulation are in accordance with the opinion of the Standing Committee on Plants, Food and Feed,

HAS ADOPTED THIS REGULATION:

Article 1

The Annex to Regulation (EC) No 1881/2006 is amended in accordance with the Annex to this Regulation.

Article 2

This Regulation shall enter into force on the twentieth day following that of its publication in the *Official Journal of the European Union.*

It shall apply from

This Regulation shall be binding in its entirety and directly applicable in all Member States.

Done at Brussels,

For the Commission
The President
Jean-Claude JUNCKER

EUROPEAN
COMMISSION

Brussels, XXX
SANCO/12420/2014
[...](2014) XXX draft

ANNEX 1

ANNEX

to the

Commission Regulation (EU) No .../...

amending Regulation (EC) No 1881/2006 as regards maximum levels for polycyclic aromatic hydrocarbons in foodstuffs

ANNEX

to the

Commission Regulation (EU) No .../...

amending Regulation (EC) No 1881/2006 as regards maximum levels for polycyclic aromatic hydrocarbons in foodstuffs

WORKING DOCUMENT – DOES NOT NECESSARILY REPRESENT THE VIEWS OF THE COMMISSION SERVICES

The Annex to Regulation (EC) No 1881/2006 is amended as follows:

(1) Section 6: *Polycyclic aromatic hydrocarbons* is replaced by the following

"Section 6:Polycyclic aromatic hydrocarbons

Foodstuffs		Maximum levels (µg/kg)	
6.1	Benzo(a)pyrene, benz(a)anthracene, benzo(b)fluoranthene and chrysene	Benzo(a)pyrene	Sum of benzo(a)pyrene, benz(a)anthracene, benzo(b)fluoranthene and chrysene(45)
6.1.1	Oils and fats (excluding cocoa butter and coconut oil) intended for direct human consumption or use as an ingredient in food	2,0	10,0
6.1.2	Cocoa beans and derived products	5,0 µg/kg fat	30,0 µg/kg fat
6.1.3	Coconut oil intended for direct human consumption or use as an ingredient in food	2,0	20,0
6.1.4	Smoked meat and smoked meat products	2,0	12,0
6.1.5	Muscle meat of smoked fish and smoked fishery products(25)(36), excluding fishery products listed in points 6.1.6 and 6.1.7. The maximum level for smoked crustaceans applies to muscle meat from appendages and abdomen(44). In case of smoked crabs and crab-like crustaceans (*Brachyura* and *Anomura*) it applies to muscle meat from appendages.	2,0	12,0

6.1.6	Smoked sprats and canned smoked sprats(25)(47) (*sprattus sprattus*); Smoked Baltic herring ≤ 12 cm length and canned smoked Baltic herring ≤ 14 cm length(25)(47) (*Clupea harengus membras*); Katsuobushi (dried bonito, *Katsuwonus pelamis);* bivalve molluscs (fresh, chilled or frozen)(26); heat treated meat and heat treated meat products(46) sold to the final consumer	5,0	30,0
6.1.7	Bivalve molluscs(36)(smoked)	6,0	35,0
6.1.8	Processed cereal-based foods and baby foods for infants and young children(3)(29)	1,0	1,0
6.1.9	Infant formulae and follow-on formulae, including infant milk and follow-on milk(8)(29)	1,0	1,0
6.1.10	Dietary foods for special medical purposes(9)(29) intended specifically for infants	1,0	1,0
6.1.11	Food supplements	10,0	50,0
6.1.12	Dried herbs and spices	10,0	50,0
6.1.13	Tea and herbal infusions	10,0	50,0

(45) Lower bound concentrations are calculated on the assumption that all the values of the four substances below the limit of quantification are zero.

(46) Meat and meat products that have undergone a heat treatment potentially resulting in formation of PAH, i.e. only grilling and barbecuing.

(47) For the canned product the analysis shall be carried out on the whole content of the can. As regards the maximum level for the whole composite product Art. 2(1)(c) and 2(2) shall apply."

EUROPEAN
COMMISSION

Brussels, XXX
SANCO/10381/2015
[…](2015) XXX draft

COMMISSION REGULATION (EU) No …/..

of XXX

amending Regulation (EC) No 1881/2006 as regards maximum levels for polycyclic aromatic hydrocarbons in cocoa fibre, banana chips, food supplements, dried herbs and spices

(Text with EEA relevance)

COMMISSION REGULATION (EU) No …/..

of XXX

amending Regulation (EC) No 1881/2006 as regards maximum levels for polycyclic aromatic hydrocarbons in cocoa fibre, banana chips, food supplements, dried herbs and spices

(Text with EEA relevance)

WORKING DOCUMENT – DOES NOT NECESSARILY REPRESENT THE VIEWS OF THE COMMISSION SERVICES

THE EUROPEAN COMMISSION,

Having regard to the Treaty on the Functioning of the European Union,

Having regard to Council Regulation (EEC) No 315/93 of 8 February 1993 laying down Community procedures for contaminants in food[1], and in particular Article 2(3) thereof,

Whereas:

(1) Commission Regulation (EC) No 1881/2006[2] sets maximum levels for polycyclic aromatic hydrocarbons (PAHs) in food

(2) According to that Regulation, maximum levels for PAHs must be safe and as low as reasonably achievable (ALARA) based upon good manufacturing, drying and agricultural/fishery practices.

(3) Cocoa fibre is a specific cocoa product produced from the shell of cocoa bean and contains therefore higher levels of PAHs than the cocoa products produced from the cocoa nibs. The cocoa and cocoa fibres are intermediate products in the food chain and used as an ingredient in the preparation of low calorie, high fibre foods. It is appropriate to establish a specific level of PAHs for cocoa fibre and derived products on fresh weight basis given their low fat content.

(4) Banana chips are widely used in cereals and confectionery as well as eaten alone as snacks. Recently, high levels of PAHs have been found in banana chips. The finding of high levels of PAHs is related to the frying of these banana chips in coconut oil. Therefore it is appropriate to establish a maximum level for these banana chips corresponding to the maximum level established for PAHs in coconut oil intended for direct human consumption or use as an ingredient in food.

(5) High levels of PAHs have been found in some food supplements. The presence of high levels in certain food supplements have been linked to the presence of or derived from botanical ingredients. Also high levels of PAHs have been found in dried herbs and spices. The source of the presence of high levels of PAH in these products has been identified to be the bad drying practices and these high levels are avoidable by

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[1] OJ L 37, 13.2.1993, p. 1.

[2] Commission Regulation (EC) No 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs (OJ L 364, 20.12.2006, p. 5).

applying good practices. It is therefore appropriate to establish a maximum level for PAH in these products which is achievable by applying good drying practices and which ensures a high level of human health protection. The traditional smoking method used for the production of Pimentón de la Vera (smoked paprika) and the traditional drying method for the production of black cardamom results in high levels of PAHs. Given that the consumption of these spices is low and to enable these traditionally smoked products to remain on the market, it is appropriate to exempt these spices from the maximum level.

(6) The measures provided for in this Regulation are in accordance with the opinion of the Standing Committee on Plants, Food and Feed,

HAS ADOPTED THIS REGULATION:

Article 1

The Annex to Regulation (EC) No 1881/2006 is amended in accordance with the Annex to this Regulation.

Article 2

The maximum levels of polycyclic aromatic hydrocarbons (PAHs) laid down in points 6.1.12, 6.1.13, 6.1.14 and 6.1.15 in the Annex to Regulation (EC) No 1881/2006 shall apply from 1 January 2016. Foodstuffs not complying with these maximum levels which are lawfully placed on the market prior to 1 January 2016 may continue to be marketed after that date until their date of minimum durability or use-by-date.

Article 2

This Regulation shall enter into force on the twentieth day following that of its publication in the *Official Journal of the European Union.*

This Regulation shall be binding in its entirety and directly applicable in all Member States.

Done at Brussels,

For the Commission
The President
Jean-Claude JUNCKER

EUROPEAN
COMMISSION

Brussels, XXX
SANCO/10381/2015
[...](2015) XXX draft

ANNEX 1

ANNEX

to the

Commission Regulation (EU) No .../...

amending Regulation (EC) No 1881/2006 as regards maximum levels for polycyclic aromatic hydrocarbons in cocoa fibre, banana chips, food supplements, dried herbs and spices

ANNEX

to the

Commission Regulation (EU) No .../...

amending Regulation (EC) No 1881/2006 as regards maximum levels for polycyclic aromatic hydrocarbons in cocoa fibre, banana chips, food supplements, dried herbs and spices

WORKING DOCUMENT – DOES NOT NECESSARILY REPRESENT THE VIEWS OF THE COMMISSION SERVICES

Section 6: "*Polycyclic aromatic hydrocarbons*" of the Annex to Regulation (EC) No 1881/2006 is amended as follows:

(1) The point 6.1.2 is replaced by the following:

"6.1.2	Cocoa beans and derived products with the exception of the products referred to in 6.1.11	5,0 µg/kg fat as from 1.4.2013	35.0 µg/kg fat as from 1.4.2013 until 31.3.2015 30,0 µg/kg fat as from 1.4.2015

(2) The following points 6.1.11, 6.1.12, 6.1.13, 6.1.14 and 6.1.15 are added:

6.1.11	Cocoa fibre and derived products intended for use as an ingredient in food	3.0	15.0
6.1.12	Banana chips	2.0	20.0
6.1.13	Food supplements containing and/or derived from botanical ingredients	10,0	50,0
6.1.14	Dried herbs	10,0	50,0
6.1.15	Spices with the exception of Pimentón de la Vera (traditionally smoked paprika) and cardamom	10,0	50,0

Food Additives & Contaminants: Part A, 2014
http://dx.doi.org/10.1080/19440049.2014.952785

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Occurrence of 15 + 1 EU priority polycyclic aromatic hydrocarbons (PAH) in various types of tea (*Camellia sinensis*) and herbal infusions

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(*Received 4 March 2014; accepted 4 August 2014*)

For the analysis of 15 + 1 EU priority PAH in tea and herbal infusions, an online-SPE-LVI-GC-MS method was developed. This method includes sample extraction of the tea and herbal infusions with saponification followed by an automated SPE clean-up step. For brews a liquid–liquid extraction with cyclohexane was performed before an automated SPE clean-up. Gas chromatographic separation was done using an Agilent J&W Select PAH (15 m × 0.15 mm × 0.10 µm) column, which allows the separation of the three benzofluoranthenes as well as triphenylene from chrysene. Method performance criteria such as method linearity, limit of quantitation (LOQ) and repeatability were determined and demonstrated that the method was fit for purpose. The method was used to analyse 15 + 1 EU priority PAH in 91 tea and herbal infusion samples. The levels of PAHs ranged from below 0.5 (LOQ) to 460 µg kg^{-1}, with a median of 4.7 µg kg^{-1} and a mean of 39 µg kg^{-1} for BaP, and from below 1.0 (LOQ) to 2700 µg kg^{-1}, with a median of 39 µg kg^{-1} and a mean of 250 µg kg^{-1} for total PAH, which were in good agreement with other studies reported in the literature. For the brews prepared under normal house preparation (20 g material in 2 L boiling tap water for 10 min), no total 15 + 1 PAH could be detected above the LOQ. With an extended brewing time of 30 min, a transfer rate between 0.25% and 0.52% could be determined, which results in no exceeding of the maximum limits given by the European Union directive for drinking water (EU Council Directive 98/83/EC).

Keywords: polycyclic aromatic hydrocarbons (PAH); GC-MS; *Camellia sinensis*; green tea; black tea; white tea; oolong; pu erh; lapsang souchong; herbal infusions

Introduction

Polycyclic aromatic hydrocarbons (PAH) comprise a large group of more than several hundred chemical compounds containing two or more fused aromatic rings. They originate from incomplete combustion of organic material and are found in various types of food, either by uptake from the environment or from generation during processing procedures such as grilling, frying, smoking or drying (Larsson et al. 1983; Guillen 1994; European Commission 2002; Šimko 2005; Jira et al. 2008; Perelló et al. 2009). Many of them are known to be carcinogenic and genotoxic. Therefore, their presence in food is a potential concern for consumer health. The two most important groups of PAH monitored worldwide are the 16 EPA PAH, listed by the USEPA, and the 15 + 1 EU priority PAH, defined by the European Union (USEPA 1999; Wenzl et al. 2006; EFSA 2008a). In the past, benzo[a]pyrene was thought to be a suitable marker for the occurrence of carcinogenic PAH and therefore maximum limits were set for certain food categories under EU law (European Commission 2005). However, in 2008 EFSA published a report on suitable indicators for the occurrence of toxic PAH in food. EFSA's CONTAM Panel concluded that benzo[a]pyrene (BaP) was not a suitable marker for the occurrence of carcinogenic PAH in food and recommended instead a group of PAH4 (benzo[a]pyrene, chrysene, benzo[a]anthracene and benzo[b]fluoranthene) in order to protect consumer health better (EFSA 2008a). As a consequence, the EU regulation was amended in August 2011, giving an individual maximum level (ML) for benzo[a]pyrene as well as a sum ML for the PAH4 in different types of food categories (Regulation (EU) No. 835/2011; European Commission 2011a).

Tea is the second most consumed beverage after water, with 4 million tonnes consumed worldwide in 2010 (FAO 2012). Herbal and fruit infusions have been drunk for centuries, especially for their medicinal properties. Nowadays, they have become popular beverages in a well-balanced diet because in contrast to most other beverages they have a low sugar and calorie content, are caffeine free (exceptions are infusions with mate, guarana or cola nut), and offer a vast range of different flavours. For many years the consumption of herbal and fruit infusions in the EU has been highest in Austria, Germany, France and the UK (EHIA 2012a). Since adults should drink a minimum of 2 L (EFSA 2010) preferably of low caloric fluids per day (Bundesministerium für Gesundheit Austria 2011; Deutsche Gesellschaft für Ernährung 2011; Schweizerische Gesellschaft für Ernährung 2011), tea and

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herbal and fruit infusions are a healthy choice to achieve this aim. The consumption of tea and herbal and fruit infusions is recommended by official bodies across Europe to maintain fluid balance or to prevent dehydration (Bundesministerium für Gesundheit Austria 2011; Ruxton & Hart, 2011; Schweizerische Gesellschaft für Ernährung 2011). As well as providing hydration, the health benefits of tea have been linked to its flavonoid polyphenol content, which accounts for approximately 30% of the dry weight of the tea leaf (Yamanishi 1995). Epidemiological studies have associated tea consumption with the prevention of several chronic and degenerative diseases including cancer (Dufresne & Farnworth 2001; Wu et al. 2003; Basu & Lucas 2007; Arab et al. 2009; Thielecke & Boschmann 2009; Yuan et al. 2011). Herbal and fruit infusion ingredients are of special scientific interest due to many being rich in polyphenols and are studied for their antioxidative properties (Heck & de Mejia 2007; Büyükbalci & El 2008; Winterhalter et al. 2010; Pękal et al. 2011).

Teas are produced from the tender leaves and shoots of *Camellia sinensis* L. Kuntze. Green tea is produced by steaming or roasting freshly harvested leaves to prevent oxidation reactions. To produce black tea, freshly harvested leaves are withered to reduce the moisture content, then rolled and crushed to release polyphenol oxidase which initiates oxidation reactions (known as aeration or formerly as fermentation), and finally dried (fired). Oolong teas undergo a partial oxidation of the leaf and are classed as semi-aerated teas (Yamanishi 1995). Lapsang souchong is a Chinese tea that is roasted and smoked over resin-rich wood (Adrian et al. 1983), which gives this tea its typical smoky flavour. White tea is derived from the gently plucked tender shoots of *Camellia sinensis*; it does not undergo an aeration but is only minimally processed and dried (ETC 2012). The first steps of the manufacture of pu erh tea are similar to those of green tea, though the aeration process in some cases may not be as complete for pu erhs. Next the tea leaves are ‘aged’ or post-aerated in different ways depending on the type of pu erh that is produced. This may take up to several weeks (Lu & Hwang 2008; Ahmed et al. 2010). Herbal and fruit infusions are prepared from dried plants or parts of plants (e.g. fruits, flowers, leaves, roots, bark) that do not originate from the tea bush, i.e. *C. sinensis* L. Kuntze. Herbal and fruit infusions are intended for use by brewing with freshly boiled water. In Europe more than 400 different plant parts are used as ingredients for herbal and fruit infusions (EHIA 2012b). These plants are either globally cultivated or wild gathered and originate from countries all over the world. After the raw materials have been harvested they are gently dried, sorted, often cut and finally ready for consumption as a mono-product or blend of herbs and/or fruits. The manufacturing process for the herbal infusion yerba mate (*Ilex paraguariensis*) slightly differs from the above procedure. After harvesting, mate leaves and stems are blanched at high temperatures by flash heating over wood or propane fire for several seconds. This processing deactivates specific enzymes. Afterwards the leaves are dried slowly often using wood smoke (Heck & de Mejia 2007). The special processing gives mate its typical bitter aromatic and slightly smoky flavour.

PAH may be present in tea and herbal and fruit infusions either by uptake from the environment (through contaminated soil, water or air) or due to processing procedures (e.g. drying or roasting) where PAH are formed and accumulated on the surface of the plant material.

Published studies about the PAH content in tea leaves have focused mainly on the group of EPA PAH or only BaP since this was previously thought of as a good marker substance. Reported PAH contents in tea leaves are in the range of 497–1162 μg kg^{-1} (n = 4; Fiedler et al. 2002), 13–7536 μg kg^{-1} (n = 11; Schlemitz & Pfannhauser 1997), 48–1703 μg kg^{-1} (n = 34; Ciemniak 2005), 323–8800 μg kg^{-1} (n = 8; Lin et al. 2005), 257–7970 μg kg^{-1} (n = 4; Lin et al. 2006) for the EPA PAH, and 3–20 μg kg^{-1} for BaP (n = 10; Speer 1987), where n is the total number of analysed samples. The EFSA report is based on a data collection of PAH in different foods consumed in the EU and also comprises data for the Codex food category 14.1.5, coffee, tea etc., showing a range of above 1 μg kg^{-1} (LOD) up to 101 μg kg^{-1} for BaP in this category (n = 99), but without specifying data for only tea or herbal and fruit infusions (EFSA 2008b). One published study focused on the amount of EU priority PAH in 40 teas and herbal and fruit infusions, ranging from 14 to 2662 μg kg^{-1} (Ziegenhals et al. 2008). The transfer of PAH from tea leaves into the infusion has been reported too, indicating that a maximum of 11% of the total PAH present in the tea is transferred into the infusion. These studies were all focused on the group of EPA PAH (Bishnoi et al. 2005; Zuin et al. 2005; Lin et al. 2006; Viñas et al. 2007). Two fruit-tea infusions being investigated for the presence of EU priority PAH have been found to be free of PAH (Germán-Hernández et al. 2013).

The aim of the present study was to determine the content of the 15 + 1 EU priority PAH in the dry matter and brew of different types of teas as well as herbal and fruit infusions. Due to the poor availability of data on the EU priority PAH in tea infusions, the transfer of this PAH group was also determined for some selected tea samples.

Material and methods

Materials

The 57 teas used in this study were selected to include samples from all the major tea-producing countries and

Table 1. Major matrix types of herbal and fruit infusions.

Matrix type				
Fruit	Acidic	Volatile oil rich	Roasted/ smoked	Herb
Apple	Hibiscus	Peppermint	Yerba mate	Nettle
Rosehip		Fennel		Lime flowers
Elderberry		Lemon peel		Chamomile
				Rooibos

each of the main commercial tea types (green, black, white, oolong, pu erh). All samples were the typical leaf grades for the type, either whole leaf or fannings. Samples from lapsang souchong were also included in the study as these are expected to contain higher levels of PAH compounds.

The 36 herbal and fruit infusions were selected to represent the major matrix types (Table 1). Samples from mate were also included as products at risk for high PAH content due to their special processing procedure.

The isotope-labelled (deuterated) stock solution containing d10-fluoranthene, d12-CHR, d12-BaP, d12-BgP and d14-DiP at a concentration of 100 µg ml^{-1} in toluene was purchased from Campro Scientific (Berlin, Germany). The native stock solution containing the EU priority PAH at a concentration of 50 µg ml^{-1} in toluene was obtained from NEOCHEMA (Bodenheim/Mainz, Germany). From the stock solutions, an internal standard solution with a concentration of 0.5 µg ml^{-1} for each deuterated PAH in cyclohexane was prepared as well as a working solution containing deuterated and native PAH at a concentration of 0.01 µg ml^{-1} in a mixture of 90% ethyl acetate and 10% cyclohexane.

All solvents were purchased of analytical quality from VWR International (Darmstadt, Germany) as well as potassium hydroxide. SPE cartridges (Supelclean ENVI-Chrom P SPE tubes 6 ml, 0.5 g phase) were obtained from Sigma-Aldrich (Taufkirchen, Germany). The column used for gas-chromatographic separation (Agilent J&W Select PAH, 15 m × 0.15 mm × 0.10 µm) was purchased from Agilent Technologies (Waldbronn, Germany). The polyamide filters (CHROMAFIL AO-45/25, 0.45 µm pore size) were obtained from Macherey-Nagel (Düren, Germany).

Sample preparation

Between 1.5 and 2.0 g of the homogenised samples were weighed and an internal standard solution containing a mixture of deuterated PAHs was added, followed by 10 ml methanolic potassium hydroxide solution and 5 ml cyclohexane. The samples were shaken for 10 min on a shaker and then put in an ultrasonic bath for 30 min at 60°C to enhance the extraction step. After centrifugation, about 1.8 ml of the organic phase was filtered through a membrane filter into a vial. Further sample clean-up was carried out using an online SPE-GC-MS system.

The infusions were prepared closely to conditions of normal home preparation by infusing 20 g of the homogenised sample in 2 L boiling tap water for 10 min, filtered and cooled to ambient temperature. The larger initial weight and volume of boiling tap water with regard to normal home preparation was chosen in order to facilitate laboratory practice. A volume of 100 µl of the internal standard solution was added to the 500 ml infusion, followed by two extractions with 50 ml cyclohexane. The combined organic phases were concentrated to a volume of 2 ml and further purified with SPE using an online SPE-GC-MS system.

The SPE cartridges were conditioned with a 2.5 ml of a mixture of 90% ethyl acetate and 10% cyclohexane, followed by 2.5 ml of cyclohexane. After application of the sample extract, the cartridge was washed with 7.5 ml of a mixture of 10% ethanol and 90% cyclohexane. Elution of the PAH containing fraction was carried out with 4 ml of a mixture of 90% ethyl acetate and 10% cyclohexane. The volume was concentrated to an extract of approximately 1 ml and analysed by GC-MS.

Instrumentation

A gas chromatograph 7890A (Agilent Technologies) equipped with a UNIS 2100 PTV injection port (JAS) and a MPS2 XL Dual Rail Autosampler (Gerstel, Mühlheim an der Ruhr, Germany) was used for sample clean-up and measurement. The GC was coupled to an Agilent 5975C inert mass spectrometer (Agilent Technologies) operating in EI mode at 70 eV and SIM mode. Ion source and transfer line temperature were maintained at 300°C. Helium was used as carrier gas with a flow of 1.2 ml min^{-1}. The injection was a large volume injection (LVI) of 50 µl. The applied injector program was 50°C (hold 0.02 min), to 380°C (hold 12 min) at 720°C min^{-1} and 2.0 min splitless. Solvent vent mode was used with a vent flow of 40.0 ml min^{-1}. For gas chromatographic separation, an Agilent J&W Select PAH (15 m × 0.15 mm × 0.10 µm) column was used. The applied GC temperature programme was optimised for both separation and total run time as: 70°C (hold 0.4 min), to 180°C (hold 1 min) at 70°C min^{-1}, to 230°C (hold 7 min) at 7°C min^{-1}, to 280°C (hold 7 min) at 50°C min^{-1}, to 350°C (hold 3.6 min) at 30°C min^{-1}.

Results and discussion

The applied online-SPE-GC/MS method for the determination of PAH in food was validated in different food matrices, including a dry powder. The linearity of the method was determined at a minimum of five levels in duplicate and found to have high accuracy for all target analytes (coefficient of determination between 0.995 and 1.000; Table 2). The investigation for repeatability of the method was performed seven-fold. The repeatability RSD was found to be between 3.7% and 11.5% for all EU priority PAH and therefore considered satisfactory. The LOQ was calculated from multiple analysis of material which was spiked at the level of the target LOQ. For the lower volatile EU priority PAH a target LOQ of 0.5 µg kg^{-1} could be achieved considering the requirements of Commission Regulation (EU) No. 836/2011 (European Commission 2011b) which demand an LOQ equal or below 0.9 µg kg^{-1} for BaP, CHR, BaA and BbF. The PAH with a higher molecular weight are generally discriminated with GC-MS (Gómez-Ruiz & Wenzl 2009), therefore an increased LOQ of 1 µg kg^{-1} was reached for them. With the help of a spiked material the recovery of the applied method was also investigated. The values ranged from 71% to 113%, which was satisfactory; only 5-methylchrysene showed a higher recovery of 128%. The overall results of the validation indicate that the applied method is suitable for the analysis of the EU priority PAH in tea as well as herbal and fruit infusions.

In the first step of the study, the 91 tea and herbal and fruit infusion samples were analysed using the online-SPE-GC/MS method. Figure 1 shows a representative chromatogram of a peppermint sample with higher PAH levels. Accurate quantification of individual PAH requires chromatographic separation and/or differentiation by mass ions, so the selection of an appropriate capillary column is one of the most critical aspects. Due to identical mass ions and similar physico-chemical properties some PAH with regard to co-elution in the stationary phase are challenging to analyse, among them the pair of chrysene and triphenylene and the group of benzofluoranthene isomers (Bordajandi et al. 2008; Gómez-Ruiz & Wenzl 2009). The use of an Agilent J&W Select PAH column for gas chromatographic separation allows a satisfactory separation of chrysene from triphenylene, which is a frequent interfering PAH as well as the three benzofluoranthenes in one GC run (Figure 2a and b).

Tables 3 and 4 show the content of the 15 + 1 EU priority PAH in the analysed 91 tea and herbal and fruit infusion samples. The content of the 15 + 1 EU priority PAH ranged from non-quantifiable to 2700 µg kg^{-1}. The median of total PAH was found to be 39 µg kg^{-1} and the mean was 250 µg kg^{-1}. The amount of BaP ranged from non-quantifiable to 460 µg kg^{-1}. The median content of BaP was 4.7 µg kg^{-1} and the mean was 39 µg kg^{-1}. The sum of PAH4 (BaP, BaA, BbF and CHR) ranged from non-quantifiable to 1700 µg kg^{-1} with a median of 24 µg kg^{-1} and a mean of 140 µg kg^{-1}. This has a high correlation with the amounts of total PAH and the BaP content in the tea and herbal and fruit infusion samples. In

Table 2. Method performance data for the determination of PAH with online-SPE-GC-MS.

Compound	Abbreviation	MW	Method linearity r^2	r mean (µg kg^{-1})	r, RSD (%)	LOQ (µg kg^{-1})	Recovery (%)
Benzo[a]pyrene	BaP	252	0.998	8.1	3.9	0.5	97
Benzo[a]anthracene	BaA	228	0.999	5.1	6.1	0.5	109
Chrysene	CHR	228	0.997	4.3	7.4	0.5	98
Benzo[b]fluoranthene	BbF	252	1.000	6.1	4.2	0.5	97
Benzo[k]fluoranthene	BkF	252	0.999	6.0	3.7	0.5	107
Indeno[1,2,3-cd]perylene	IcP	276	0.999	9.2	6.0	0.5	95
Dibenzo[a,h]anthracene	DhA	278	0.995	9.5	6.1	0.5	82
Benzo[g,h,i]perylene	BgP	276	1.000	2.3	6.0	0.5	71
Dibenzo[a,l]pyrene	DlP	302	0.998	2.8	4.6	1.0	102
Dibenzo[a,i]pyrene	DiP	302	0.996	1.7	10.1	1.0	83
Dibenzo[a,h]pyrene	DhP	302	0.998	2.4	11.5	1.0	98
Dibenzo[a,e]pyrene	DeP	302	0.999	2.2	7.7	1.0	108
Cyclopenta[c,d]pyrene	CPP	226	0.998	3.0	3.7	1.0	93
5-Methylchrysene	5MC	242	1.000	2.6	3.9	1.0	128
Benzo[c]fluorene	BcL	216	0.999	2.3	7.6	1.0	113

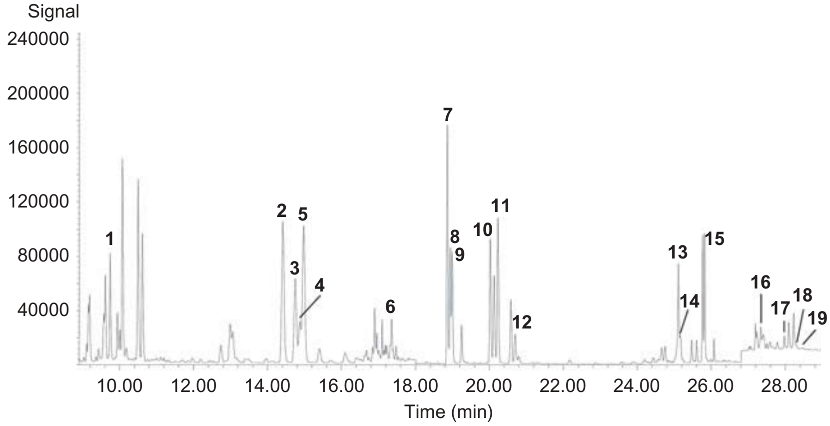

Figure 1. TIC of a peppermint sample, naturally loaded with 215 µg kg^{-1} total PAH on an Agilent J&W Select PAH (15 m × 0.15 mm × 0.10 µm). Peak identification: 1, BcL; 2, BaA; 3, CPP; 4, triphenylene; 5, CHR; 6, 5MC; 7, BbF; 8, BkF; 9, benzo[j]fluoranthene; 10, benzo[e]pyrene; 11, BaP; 12, perylene; 13, IcP; 14, DhA; 15, BgP; 16, DlP; 17, DeP; 18, DiP; and 19, DhP.

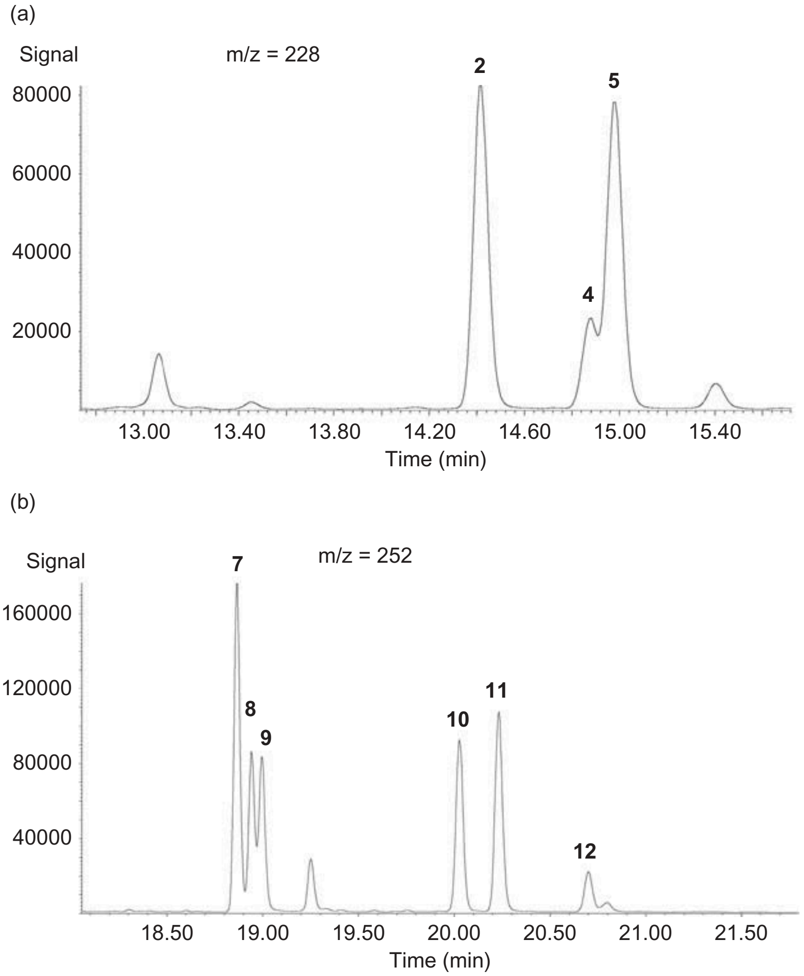

Figure 2. SIM chromatograms of a peppermint sample, naturally loaded with 215 µg kg^{-1} total PAH on an Agilent J&W Select PAH (15 m × 0.15 mm × 0.10 µm). Details and peak identification: (a) $m/z = 228$; 2, BaA; 4, triphenylene; and 5, CHR; and (b) $m/z = 252$; 7, BbF; 8, BkF; 9, benzo[j]fluoranthene; 10, benzo[e]pyrene; 11, BaP; and 12, perylene.

Table 3. PAH content in various types of teas (μg kg^{-1}, *LOQ) (n = 57).

	BaA	CHR	BbF	BkF	BjF	BaP	IcP	DhP	BgP	DlP	DiP	DhA	DeP	CPP	5MC	BcL	Σ PAH	Σ PAH4
Black	43	43	44	17	23	28	24	1.1	29	2.0	1.1	3.0	2.7	3.6	3.0	4.4	270	160
Black	37	36	39	13	20	23	16	< 1.0*	21	1.2	< 1.0*	1.8	1.6	6.7	3.7	4.0	220	140
Black	13	15	15	6.3	9.9	11	8.7	< 1.0*	13	< 1.0*	< 1.0*	1.0	< 1.0*	1.2	< 1.0*	1.1	95	54
Black	5.1	6.7	5.7	2.5	3.7	3.8	3.3	< 1.0*	3.4	< 1.0*	< 1.0*	< 0.5*	< 1.0*	1.9	< 1.0*	< 1.0*	36	21
Black	3.1	5.3	3.8	2.1	2.5	2.0	1.9	< 1.0*	1.6	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	22	14
Black	4.4	6.7	4.1	2.1	2.5	2.0	1.6	< 1.0*	1.2	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	25	17
Black	38	31	32	16	23	31	20	< 1.0*	23	< 1.0*	< 1.0*	1.7	< 1.0*	17	2.2	10	250	130
Black	9.1	10	8.4	3.3	4.2	4.7	4.0	< 1.0*	4.4	< 1.0*	< 1.0*	< 0.5*	< 1.0*	4.2	< 1.0*	1.2	54	32
Black	9.2	11	7.5	3.0	5.2	4.1	3.1	< 1.0*	3.6	< 1.0*	< 1.0*	< 0.5*	< 1.0*	2.4	< 1.0*	2.8	52	32
Black	2.1	4.7	5.1	1.9	2.1	1.4	1.5	< 1.0*	1.2	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	20	13
Black	9.0	8.9	8.3	3.4	4.9	5.7	4.2	< 1.0*	6.2	< 1.0*	< 1.0*	< 0.5*	< 1.0*	2.2	< 1.0*	1.2	54	32
Black	41	31	40	16	19	32	21	1.2	27	1.8	< 1.0*	2.7	2.1	6.7	< 1.0*	4.6	250	140
Black	6.9	9.3	4.9	2.1	3.8	3.7	2.1	< 1.0*	3.0	< 1.0*	< 1.0*	< 0.5*	< 1.0*	3.2	< 1.0*	< 1.0*	39	25
Black	7.9	10	5.7	2.8	4.4	3.7	2.3	< 1.0*	2.6	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	1.0	40	27
Black	5.3	6.7	4.2	2.1	2.7	2.2	1.6	< 1.0*	1.7	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	27	18
Black	3.5	5.5	3.2	1.9	2.0	1.5	1.6	< 1.0*	2.9	< 1.0*	< 1.0*	< 0.5*	< 1.0*	2.3	< 1.0*	< 1.0*	24	14
Black	2.0	4.2	1.8	1.0	0.9	0.9	1.1	< 1.0*	1.8	< 1.0*	< 1.0*	< 0.5*	< 1.0*	1.8	< 1.0*	< 1.0*	16	8.9
Black	4.6	8.3	3.9	2.2	2.1	2.1	1.8	< 1.0*	2.7	< 1.0*	< 1.0*	< 0.5*	< 1.0*	4.2	< 1.0*	< 1.0*	32	19
Black	1.1	2.0	1.2	< 0.5*	0.6	0.6	< 0.5*	< 1.0*	0.5	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	6.0	4.9
Black	4.8	7.2	4.7	1.3	2.2	2.5	1.9	< 1.0*	5.5	< 1.0*	< 1.0*	< 0.5*	< 1.0*	1.6	< 1.0*	< 1.0*	32	19
Black	360	220	240	120	190	330	150	1.1	210	1.8	2.5	9.8	4.7	370	2.8	30	2200	1200
Black	8.0	12	6.5	2.9	4.3	3.6	2.8	< 1.0*	3.9	< 1.0*	< 1.0*	< 0.5*	< 1.0*	1.4	< 1.0*	< 1.0*	45	30
Black	170	120	120	55	95	160	60	< 1.0*	76	1.6	< 1.0*	4.1	1.5	70	< 1.0*	16	950	570
Black	2.7	3.6	3.2	2.1	1.9	2.9	1.9	< 1.0*	4.8	< 1.0*	< 1.0*	< 0.5*	< 1.0*	1.5	< 1.0*	< 1.0*	25	12
Black	2.3	3.9	3.3	2.2	1.7	1.9	1.8	< 1.0*	1.9	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	19	11
Black	2.5	4.0	3.2	2.1	2.1	1.7	1.3	< 1.0*	1.0	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	18	11
Green	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	< 0.5*
Green	130	120	85	36	63	97	53	1.1	84	1.5	1.9	6.1	3.2	170	1.6	15	870	430
Green	53	32	32	16	22	34	15	< 1.0*	27	< 1.0*	< 1.0*	1.8	< 1.0*	84	< 1.0*	3.4	320	150
Green	11	16	14	6.1	8.0	7.1	5.8	< 1.0*	7.1	< 1.0*	< 1.0*	0.87	< 1.0*	3.7	< 1.0*	2.6	82	48
Green	7.7	12	9.1	5.4	5.2	6.5	4.5	< 1.0*	7.8	< 1.0*	< 1.0*	< 0.5*	< 1.0*	8.6	< 1.0*	1.5	68	35
Green	5.5	9.3	7.9	4.0	4.5	4.3	3.8	< 1.0*	5.5	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	45	27
Green	6.7	13	20	7.7	9.8	10	10	< 1.0*	16	1.3	< 1.0*	0.77	< 1.0*	33	< 1.0*	1.3	130	50
Green	9.6	5.8	5.3	3.3	3.4	5.9	2.3	< 1.0*	3.7	< 1.0*	< 1.0*	< 0.5*	< 1.0*	15	< 1.0*	1.2	56	27
Green	1.7	2.3	1.1	< 0.5*	0.6	0.9	< 0.5*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 0.5*	< 1.0*	1.3	< 1.0*	< 1.0*	7.9	6.0
Green	38	33	30	12	21	24	16	< 1.0*	20	< 1.0*	< 1.0*	1.6	1.2	23	< 1.0*	7.1	230	130

(*continued*)

Table 3. Continued.

	BaA	CHR	BbF	BkF	BjF	BaP	IcP	DhP	BgP	DlP	DiP	DhA	DeP	CPP	5MC	BcL	Σ PAH	Σ PAH4
Green	37	34	26	12	20	22	14	< 1.0*	19	1.0	< 1.0*	1.1	1.2	19	1.3	7.1	220	120
Green	2.4	31	27	8.2	12	13	10	< 1.0*	13	< 1.0*	< 1.0*	1.5	1.4	5.9	1.4	3.2	130	73
Green	6.5	7.0	5.9	1.9	2.9	4.6	2.9	< 1.0*	4.1	< 1.0*	< 1.0*	< 0.5*	< 1.0*	3.3	< 1.0*	< 1.0*	39	24
Green	18	14	20	9.0	13	18	11	< 1.0*	13	< 1.0*	< 1.0*	0.9	< 1.0*	25	< 1.0*	2.2	140	70
Green	7.8	15	9.1	3.9	5.6	4.4	4.2	< 1.0*	5.5	< 1.0*	< 1.0*	< 0.5*	< 1.0	4.5	< 1.0*	1.4	61	36
Oolong	18	12	14	8.8	8.6	13	8.0	< 1.0*	11	< 1.0*	< 1.0	0.88	< 1.0	4.8	< 1.0*	1.4	100	57
Oolong	6.5	7.6	4.4	2.2	4.0	2.7	1.7	< 1.0*	2.0	< 1.0*	< 1.0*	< 0.5*	< 1.0	1.3	< 1.0*	< 1.0*	32	21
Oolong	29	28	20	9.2	10	12	6.6	< 1.0*	8.8	< 1.0*	< 1.0*	0.8	< 1.0	2.4	1.6	2.4	130	89
Oolong	20	19	16	5.6	10	11	6.3	< 1.0*	8.5	< 1.0*	< 1.0*	0.7	< 1.0	4.7	1.1	1.3	100	66
Oolong	30	27	23	8.6	13	19	11	< 1.0*	14	1.3	< 1.0*	0.9	< 1.0	9.8	< 1.0*	2.6	160	99
Oolong	5.7	6.6	5.6	2.1	4.1	5.1	3.3	< 1.0*	4.6	< 1.0*	< 1.0*	< 0.5*	< 1.0	1.0	< 1.0*	< 1.0*	38	23
Pu Erh	43	47	26	14	18	25	17	< 1.0*	17	1.1	< 1.0*	1.5	< 1.0	4.1	2.6	8.9	230	140
Pu Erh	72	73	36	19	25	37	23	< 1.0	23	1.2	< 1.0*	1.8	1.0	1.1	2.6	17	330	220
Pu Erh	58	46	32	14	23	29	18	< 1.0*	20	1.1	< 1.0*	1.5	1.3	1.1	1.5	14	260	170
White	23	20	16	5.9	8.8	6.4	5.1	< 1.0*	7.4	< 1.0*	< 1.0	< 0.5*	< 1.0	6.9	< 1.0*	1.6	100	65
White	20	15	12	4.4	6.8	6.8	4.9	< 1.0*	7.2	< 1.0*	< 1.0*	5.4	< 1.0	6.5	< 1.0*	4.0	93	54
White	5.1	8.5	7.0	2.3	3.5	2.4	2.7	< 1.0*	3.0	< 1.0*	< 1.0*	< 0.5*	< 1.0	< 1.0*	< 1.0*	< 1.0*	35	23
Lapsang souchong	540	700	120	50	65	280	47	2.6	49	6.1	2.8	13	1.0	130	430	93	2500	1600
Lapsang souchong	620	670	150	54	75	300	44	1.8	46	4.5	2.3	13	8.8	160	480	66	2700	1700
Lapsang souchong	240	270	64	91	29	460	25	< 1.0*	31	2.6	1.2	5.9	6.1	21	410	44	1700	1000

Table 4. PAH content in various types of herbal and fruit infusions (μg kg^{-1}, *LOQ) (n = 36).

	BaA	CHR	BbF	BkF	BjF	BaP	IcP	DhP	BgP	DlP	DiP	DhA	DeP	CPP	5MC	BcL	Σ PAH	Σ PAH4
Apple	1.8	2.1	1.6	0.8	1.2	1.4	1.2	< 1.0*	1.3	< 1.0*	< 1.0*	< 0.5*	< 1.0*	1.7	< 1.0*	< 1.0*	13	6.9
Apple	8.5	8.2	5.5	2.9	4.1	5.3	3.1	< 1.0*	3.4	< 1.0*	< 1.0*	< 0.5*	< 1.0*	11	< 1.0*	1.6	54	28
Apple	1.3	1.1	0.6	< 0.5*	0.5	0.9	0.8	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 0.5*	< 1.0*	1.3	< 1.0*	< 1.0*	6.5	3.9
Camomile	1.4	3.4	1.4	0.55	1.0	0.6	0.8	< 1.0*	1.5	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	11	6.8
Camomile	1.3	3.7	1.1	< 0.5*	0.9	< 0.5*	0.7	< 1.0*	0.7	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	8.3	6.1
Camomile	1.2	6.0	1.5	0.6	1.3	< 0.5*	0.8	< 1.0*	1.0	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	12	8.7
Elderberries	0.7	1.4	0.6	< 0.5*	< 0.5*	< 0.5*	0.8	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	3.5	2.7
Elderberries	< 0.5*	0.7	0.5	< 0.5*	< 0.5*	< 0.5*	0.6	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	1.8	1.2
Elderberries	0.7	0.9	0.7	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	2.3	2.3
Fennel	< 0.5*	0.7	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	0.7	0.7
Fennel	2.0	4.2	2.1	0.7	1.3	0.8	1.2	< 1.0*	1.0	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	13	9.1
Fennel	< 0.5*	1.1	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	1.1	1.1
Hibiscus	0.7	1.2	0.7	< 0.5*	0.5	< 0.5*	0.7	< 1.0*	0.6	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	4.4	2.6
Hibiscus	2.1	3.2	1.8	1.0	1.6	1.3	1.5	< 1.0*	1.5	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	14	8.4
Hibiscus	0.7	1.1	0.7	< 0.5*	0.5	0.5	0.7	< 1.0*	0.8	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	5.0	3.0
Lemon peel	0.7	1.1	0.9	< 0.5*	0.5	0.6	0.8	< 1.0*	1.2	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	5.8	3.3
Lemon peel	< 0.5*	0.6	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	0.6	0.6
Lemon peel	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	< 0.5*
Lime	3.1	4.7	2.4	1.1	1.5	1.5	1.1	< 1.0*	1.1	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	17	12
Lime	4.9	11	9.5	3.6	4.5	3.7	5.7	< 1.0*	5.1	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	1.3	49	29
Lime	3.0	5.4	3.8	1.7	2.0	1.9	2.1	< 1.0*	1.9	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	22	14
Mate	240	260	110	74	98	200	180	2.5	180	5.9	3.6	11	10	230	2.8	12	1600	810
Mate	310	420	250	140	200	380	300	6.0	310	24	12	32	20	71	19	19	2500	1400
Mate	200	280	220	98	140	240	220	2.0	270	12	6.8	17	11	80	15	11	1800	940
Nettle	5.5	5.7	3.5	1.5	1.9	1.9	1.4	< 1.0*	1.5	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	23	17
Nettle	1.1	1.7	2.4	0.8	< 0.5*	1.3	1.1	< 1.0*	0.7	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	9.1	6.5
Nettle	3.2	6.2	10	3.8	5.0	7.1	12	< 1.0*	16	1.9	< 1.0*	1.0	1.7	< 1.0*	1.0	< 1.0*	69	27
Peppermint	1.2	4.1	1.8	0.8	1.1	0.6	0.8	< 1.0*	0.9	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	11	7.7
Peppermint	0.6	1	< 0.5*	< 0.5*	< 0.5*	< 0.5*	0.9	< 1.0*	0.6	< 1.0*	< 1.0*	< 0.5*	1.4	< 1.0*	< 1.0*	< 1.0*	4.5	1.6
Peppermint	32	21	25	14	14	24	24	< 1.0*	18	2.3	1.1	2.7	2.4	32	< 1.0*	2.4	220	100
Rooibos	1.2	1.3	< 0.5*	< 0.5*	< 0.5*	0.7	0.5	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 0.5*	< 1.0*	1.1	< 1.0*	< 1.0*	4.8	3.2
Rooibos	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	< 0.5*
Rooibos	0.9	1.6	0.9	< 0.5*	< 0.5*	0.7	< 0.5*	< 1.0*	1.0	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	5.1	4.1
Rose hip	0.7	1.3	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 0.5*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	2.0	2.0
Rose hip	2.7	3.3	1.6	0.8	1.1	1.4	0.8	< 1.0*	1.0	< 1.0*	< 1.0*	< 0.5*	< 1.0*	< 1.0*	< 1.0*	< 1.0*	13	9.0
Rose hip	13	14	9.1	4.5	5.4	7.9	5.1	< 1.0*	5.9	< 1.0*	< 1.0*	0.6	< 1.0*	< 1.0*	< 1.0*	2.1	68	44

three samples, one green, one roiboos and one lemon peel infusion, no total 15 + 1 PAH were detected above the LOQ. The three lapsang souchong and the three mate samples were among the samples with the highest amounts of PAH detected. All findings are in good agreement with the published literature (Schlemitz & Pfannhauser 1997; Fiedler et al. 2002; Lin et al. 2005, 2006; Ziegenhals et al. 2008). In the report from EFSA on data collection for PAH in food (EFSA 2008b), in the category coffee and tea the median of BaP was determined as 1.90 μg kg^{-1}, the mean as 7.81 μg kg^{-1} and the maximum as 101 μg kg^{-1} (n = 99), and therefore a bit lower than the values resulting from the outcome of this study.

The significantly higher presence of PAH in lapsang souchong and mate could be explained by their special processing procedures, which are also described in the Introduction. Lapsang souchong is roasted and smoked over resin-rich wood (Adrian et al. 1983) giving the tea its typical smoky flavour. Yerba mate is first blanched at high temperatures resulting in deactivation of specific enzymes, and then dried slowly often using wood smoke. This procedure gives mate its typical bitter aromatic and slightly smoky flavour (Heck & de Mejia 2007).

With respect to total market share, lapsang souchong is mostly sold in tea speciality shops in Europe. Its market share of the total EU tea market is considerably below 1%. In 2011 about 2400 tons of mate were imported to the EU. If this amount is infused (3 g mate per 200 ml water) a theoretical consumption of 0.318 L mate for each EU inhabitant results for 2011 (EURОstat 2011). Without Spain, which counts for 42.8% of the market for mate within the EU, the estimated EU per capita mate consumption was only 0.201 L, i.e. just one cup, in 2011. For Spain, a theoretical per capita consumption of 1.482 L mate can be calculated (EURОstat 2011). In conclusion, both lapsang souchong and mate per capita consumption in the EU are very low. Nevertheless, due to the particular processing of smoking and roasting, these two products were included into the study as potential worst-case representatives.

The presence of total EU priority PAH content (median value) in different types of tea and herbal and fruit infusion samples was compared with results published (Table 5) (Ziegenhals et al. 2008). In general an increase in total PAH content can be confirmed in the following order: herbal and fruit infusions (8.3 μg kg^{-1}, n = 33) < black (38 μg kg^{-1}, n = 26) < green (82 μg kg^{-1}, n = 15) < white (93 μg kg^{-1}, n = 3) < oolong (100 μg kg^{-1}, n = 6) < pu erh (260 μg kg^{-}1, n = 3) < mate (1800 μg kg^{-1}, n = 3), lapsang souchong (2500 μg kg^{-1}, n = 3). The median value for herbal and fruit infusions is a bit lower than the reported value by Ziegenhals et al. (2008), but the number of samples included in this study was significantly higher. The values found for black and green tea are in good correlation, whereas the amounts for white tea and mate were a bit lower than those published. For these two groups the number of analysed samples in the study was too small to draw a general conclusion. For the first time values for the total EU priority PAH content in oolong, pu erh and lapsang souchong are presented. The presence of high amounts of PAH in lapsang souchong due to a special manufacturing process has already been discussed. As the processing procedures for oolong and pu erh differ at decisive steps compared with black and green tea, these findings could be explained by this fact. However, the number of analysed samples was also too small to draw a final conclusion, meaning more studies would be needed.

Table 5. Comparison of median values of total EU priority PAH (μg kg^{-1}) from the literature (Ziegenhals et al. 2008) and the present study in different types of teas, fruit and herbal infusions.

	Median value Σ total PAH (μg kg^{-1})	
Type of tea	Ziegenhals et al.	Present study
Herbal/fruit	22 (n = 7)	8.3 (n = 33)
Black	33 (n = 11)	38 (n = 26)
Green	72 (n = 11)	82 (n = 15)
White	156 (n = 3)	93 (n = 3)
Oolong		102 (n = 6)
Pu Erh		261 (n = 3)
Lapsang souchong		2530 (n = 3)
Mate	873 (n = 8)	1823 (n = 3)

Sixteen tea and nine herbal and fruit infusion samples were taken forward to the infusion stage. The samples selected were those that contained the highest levels of PAH in the dry material and included samples of the different manufacturing types (except white) and herbal and fruit infusion matrices. If more than one sample of the same herbal or fruit infusion material contained PAH, the sample with the highest level was selected.

The brewing procedure was designed as close as possible to normal household preparation (20 g of sample, 2 L of boiling tap water, brewing time 10 min) in order to get a realistic view on the consumer exposure. During a brewing time of 10 min no transfer of PAH could be observed (results not shown). Therefore, an additional brewing time of 30 min was tested, which does not represent normal practice, but as a worst-case scenario. The results are shown in Table 6. Only in one sample of a black tea were PAH quantified at levels between 0.005 and 0.011 μg l^{-1}, indicating a transfer rate between 0.25% and 0.52% of the PAH present in the sample. In tea and herbal and fruit infusion samples of a comparable high amount of PAH, lapsang souchong and mate, no transfer of PAH above the LOQ was observed.

Table 6. PAH content in various types of tea, herbal and fruit brews (µg l^{-1}, *LOQ) (n = 25).

	BaA	CHR	BbF	BkF	BjF	BaP	IcP	DhP	BgP	DlP	DiP	DhA	DeP	CPP	5MC	BcL
	brew (µg l^{-1})															
Apple	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Camomile	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Fennel	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Hibiscus	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Lime	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Mate	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Nettle	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Peppermint	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Rose hip	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Pu Erh	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Black	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Black	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Black	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Black	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Black	< 0.005*	< 0.005*	0.006	< 0.005*	0.005	0.009	0.007	< 0.005*	0.011	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Black	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Green	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Green	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Green	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Green	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Oolong	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Oolong	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Lapsang souchong	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Lapsang souchong	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*
Lapsang souchong	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*	< 0.005*

Some studies have been published about the transfer of PAH into the tea brew, most focusing on the EPA PAH and one investigating the presence of EU priority PAH in two fruit infusions (Germán-Hernández et al. 2013). Different tea brew preparing procedures have been used as 1 g tea in 100 ml boiling water for 5 min (Zuin et al. 2005) as well as differing times (Lin et al. 2006), 20 g tea in 500 ml boiling water and different times (Lin et al. 2005), 10 g tea in 100 ml boiling water for 10 min (Bishnoi et al. 2005), 1.75 g tea (bag) in 100 ml boiling water for 5 min (Viñas et al. 2007), and 1.5 g fruit infusion (bag) in 100 ml boiling water for 10 min (Germán-Hernández et al. 2013). It can be observed that for the low molecular weight PAH that are not part of the EU priority PAH and consist of two to three aromatic rings, a transfer between 1.4% and 44.5% (Lin et al. 2006) or between 3.8% and 50.6% (Lin et al. 2005) in the infusion was found. These PAH possess a higher water solubility and are therefore more dominant in the aqueous tea brew than the higher molecular weight PAH which have a higher octanol coefficient (log K_{OW}) and a low aqueous solubility (Ma et al. 2010). For individual PAH, also varying transfer rates in different types of teas were reported (17.1–44.5% for naphthalene, 12.9–32.5% for fluorene, 2.2–16.9% for phenanthrene; Lin et al. 2006) indicating more factors than the water solubility and the log K_{OW} being involved in the transfer of PAH in tea infusions. For the PAH with a higher molecular weight, which are also included in the group of EU priority PAH, transfer percentage rates between non-detectable and 3% (Lin et al. 2006), non-detectable and 1.2% (Lin et al. 2005) were observed, which is slightly higher, but in general in line with the observations of this study. Absolute values for PAH being part of the EU priority PAH are reported to be between non-detectable up to 0.051 µg l^{-1} in some mate infusions (brewing procedure 1 g sample in 100 ml boiling water, brewing time 5 min; Zuin et al. 2005), non-detectable up to 6.5 µg l^{-1} in non-specified teas (brewing procedure 10 g sample in 100 ml boiling water, brewing time 10 min; Bishnoi et al. 2005) and non-detectable in two fruit infusions (brewing procedure 1.5 g sample (bag) in 100 ml boiling water, brewing time 10 min; Germán-Hernández et al. 2013). However, without details about the contamination grade in the samples in µg kg^{-1}, a calculation of the transfer rate was not possible. Further investigations are needed to understand completely all factors that have an influence on the transfer of PAH, like processing procedures, pollution history, interaction with other ingredients and leaf size. In order to evaluate the consumer risk, the EU directive for drinking water with a maximum level of 10 ng l^{-1} for BaP and a sum maximum level of 100 ng l^{-1} for the following five PAH, BbF, BkF, BaP, BgP and IcP was taken into account (European Commission 1998). Due to the low transfer percentage of the EU priority PAH, it could be concluded that there is no risk for the consumer of tea infusions.

Conclusions

For the described online-SPE-LVI-GC-MS method, good validation data with regard to linearity, repeatability, LOQ and recovery were obtained and therefore found to be fit for purpose for the determination of PAH in tea and herbal and fruit infusions. Ninety-one teas and herbal and fruit infusions were analysed with this method. The results, showing a median of 4.7 µg kg^{-1} and a mean of 39 µg kg^{-1} for BaP, and a median of 39 µg kg^{-1} and a mean of 250 µg kg^{-1} for total PAH, are in good correlation with other studies described in the literature, as well as the outcome of the EU monitoring programme by EFSA. Two types of samples, lapsang souchong and mate, showed the highest amount of PAH, which could be explained by their special processing procedures. Twenty-five samples were selected for the infusion stage of this study in order to determine the transfer of EU priority PAH in the infusion under the normal household preparation. No PAH were quantified above the LOQ of 0.005 µg l^{-1}. An extension of the brewing time to 30 min as a worst-case scenario led to a PAH level in one sample between 0.005 and 0.011 µg l^{-1}, indicating a maximal transfer rate of 0.52%. Higher transfer rates have been reported in literature, but for the low molecular weight EPA PAH, which also possess a higher water solubility. The maximum level of 10 ng l^{-1} for BaP and the sum maximum level of 100 ng l^{-1} for the following 5 PAH, BbF, BkF, BaP, BgP and IcP, given by the EU directive for drinking water (European Commission 1998) was not exceeded. In summary, it can be concluded from this study that although the level of EU priority 15 + 1 PAH in teas and herbal and fruit infusions samples is in some cases relatively high, there is no risk for consumers. This is because in general no transfer of PAH in the infusions was observed. Only in one experiment where the infusion was prepared under worst-case conditions was a low transfer of PAH from the dry leaves into the brew measured. However, it should be highlighted that even in this case the level detected was significantly below the maximum level for PAH in drinking water set under the respective EU regulation.

Acknowledgements

The authors are especially grateful to the members of the European Tea Committee (ETC) and European Herbal Infusions Association (EHIA) Working Group Transition Rates of Contaminants in Hamburg, Germany, who provided expertise input for product-specific information on tea as well as herbal and fruit infusions. All samples analysed in this study were made available by companies organised under the roof of ETC and EHIA.

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