Food Additives & Contaminants: Part B
Surveillance

ISSN: 1939-3210 (Print) 1939-3229 (Online) Journal homepage: http://www.tandfonline.com/loi/tfab20

Polycyclic aromatic hydrocarbons in commercial brands of dry whole soybeans for direct human consumption

Laura Pereira Garcia, Fernanda M.L. Gomes, Silvia Tfouni, Eduardo Vicente, Geovana D. Savi, Karolina Santos & Vildes M. Scussel

To cite this article: Laura Pereira Garcia, Fernanda M.L. Gomes, Silvia Tfouni, Eduardo Vicente, Geovana D. Savi, Karolina Santos & Vildes M. Scussel (2016): Polycyclic aromatic hydrocarbons in commercial brands of dry whole soybeans for direct human consumption, Food Additives & Contaminants: Part B, DOI: 10.1080/19393210.2016.1240244

To link to this article: http://dx.doi.org/10.1080/19393210.2016.1240244

FOOD ADDITIVES & CONTAMINANTS: PART B, 2016
http://dx.doi.org/10.1080/19393210.2016.1240244

Polycyclic aromatic hydrocarbons in commercial brands of dry whole soybeans for direct human consumption

Laura Pereira Garcia[a], Fernanda M.L. Gomes[b], Silvia Tfouni[b], Eduardo Vicente[b], Geovana D. Savi[a], Karolina Santos[a] and Vildes M. Scussel[a]

[a]Laboratory of Mycotoxicology and Food Contaminants, Food Science and Technology Department, Center of Agricultural Sciences, Federal University of Santa Catarina, Florianopolis, Brazil; [b]Food Science and Quality Center, Food Technology Institute, Campinas, Brazil

ABSTRACT
A total of 13 polycyclic aromatic hydrocarbons (PAHs) were surveyed in dry whole soybeans (*Glycine max* L.) sold for human consumption. The analysed samples were commercialised in vegetarian shops and food stores in Southern Brazil regions. The determination of PAHs levels was carried out by high performance liquid chromatography (HPLC)with fluorescence detection. PAHs were present in 89.7% of the samples. Chrysene (Chy) and 5 methyl chrysene (5MeChy) were the main PAHs detected (76.9% and 71.8%, respectively) followed by dibenzo(ai)pyrene (DaiP), however in a much less percentage of samples (23.1%). Apart from those PAHs, also benzo (a)antracene (BaA), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF) and benzo(a)pyrene (BaP) were found. The ΣPAH levels in the positive samples ranged from 0.80 to 38.78 μg kg^{-1}, with exception of one sample that reached 204.46 μg kg^{-1} (ΣPAHs of six compounds). Despite the present contamination there are no maximum limits set for PAHs in soybeans.

ARTICLE HISTORY
Received 28 March 2016
Accepted 13 September 2016

KEYWORDS
Polycyclic aromatic hydrocarbons; PAHs; soybean; *Glycine max* L; Brazil

Introduction

Soybean (*Glycine max* L.) is an important crop for the food industry, mainly for oil production, however it is also utilised as protein source for vegetarian consumers. Although they can get protein from different vegetables, soybean protein is the most preferred either itself as whole beans or as protein meal (Gomes et al. 2009; Garcia et al. 2014). Soybean meal (proteins) from oil industries is further processed for human consumption (texturised vegetable protein) or utilised unprocessed for animals as feed ingredient, including husk. As far as food consumption, such as whole, oil, protein or husk soybeans and safety are concerned, they should be free of contaminants like pesticides, polycyclic aromatic hydrocarbons – PAHs, heavy metals and mycotoxins (Bohn et al. 2014; Garcia et al. 2014; Savi et al. 2014).

PAHs are a group of organic molecules with two or more condensed aromatic rings, produced during incomplete combustion processes at high temperatures from 400°C to 800°C and are widely distributed in nature (air, soil, plants, water). They also can be produced during food heating (Vives et al. 2001). Some of them (> molecular mass) are highly carcinogenic (EFSA 2008a; Alomirah et al. 2011; Xia et al. 2013; Rengarajan et al. 2015). Cereals (maize, wheat, rice) and pulses (beans, peas, lentils) have been reported to be contaminated with PAHs (Camargo & Toledo 2002; Falco et al. 2003; Martorell et al. 2010; Camargo et al. 2011). Furthermore, they can be found in roots of soybean, wheat and carrot, as well as in sunflower, soybean and virgin olive oils (Teixeira et al. 2007; Camargo et al. 2011; Rojo Camargo et al. 2012; Yin et al. 2013; Pongpiachan 2015). Reinik et al. (2007) and Garcia et al. (2014) report them contaminating food mainly by environmental exposure (soil, water or atmosphere contaminated), produced during processes (high temperature of drying, roasting, baking, among others) and packaging (food contact materials). Their formation depends, apart from the temperature intensity, also on grain characteristics as type, size, shape and chemical composition (Scussel et al. 2014).

The extraction step of PAHs from food has mainly been carried out by Soxhlet liquid–liquid extraction, pressurised liquid extraction or ultrasound extraction, whereas gel permeation chromatography or solid phase extraction (SPE) have been applied in the clean-up step. The determination of the different PAH compounds may be performed by high performance liquid chromatography (HPLC) coupled to fluorescence (FLD) and ultraviolet (UV) light detection or gas and liquid chromatography with mass spectrometric detection (Rey-Salgueiro et al. 2009; Martorell et al. 2010; Plaza-

CONTACT Vildes M. Scussel ✉ vildescussel_2000@yahoo.co.uk; vildes@cca.ufsc.br

Bolaños et al. 2010; Pongpiachan 2015). In this study, the SPE technique was applied to achieve an adequate limit of detection. Also, analysis was performed by FLD detection, since it is more selective and sensitive than UV detection.

Maximum limits (MLs) for PAHs were established by European Commission regulation EC n° 1881/2006 for several food products and updated in 2011 (EC n°835/2011). The 2011 revision introduced new MLs for the sum of the 4 main PAHs (BaP, chrysene – Chy, benzo(b) fluoranthene – BbF and benzo(a)antracene – BaA). Apart from this MLs for benzo(a)pyrene (BaP) only were maintained. For oils and fats the MLs are 2.0 and 10.0 μg kg^{-1} for BaP and PAH4, respectively. In addition, MLs for processed cereal-based foods and baby foods for infants and young children have been set at 1.0 μg kg^{-1}, for both BaP and PAH4 (EC 2011a). Other food commodities, such as smoked meat, fish and mollusks, seafood, cocoa butter and chocolates have MLs. However, soybeans are not covered by regulations. Considering the lack of information on safety regarding PAHs in dry whole soybeans, the objective of this study was to determine the content of a total of 13 PAHs in commercialised soybeans intended for direct human consumption.

Materials and methods

Chemicals

The 13 total recommended warning PAHs were purchased from Sigma Aldrich (St Louis, MO, USA): BaA, Chy, BbF, BaP, benzo(j)fluoranthene-BjF, benzo(k)fluoranthene-BkF, dibenzo(ae)pyrene-DaeP, dibenzo(ah) anthracene-DahA, dibenzo(al)pyrene-DalP, indeno(cd) pyrene-IcdP, 5-methylcriseno-5MeChy, dibenzo(ai)pyrene-DaiP and Dibenzo(ah)pyrene-DahP. Acetonitrile and n-hexane were obtained from Panreac (Castellar del Vallès, Barcelona, Spain). Water was obtained from a Milli-Q system on 18.2 MΩ/cm (Millipore, Bedford, MA, USA).

Equipment

The determination of PAHs was carried out by HPLC (Shimadzu, Kyoto, Japan) with a quaternary pump model LC-20AT, degasser model DGU-20A5, autosampler (30 μl loop), model SIL-20A, column oven model CTO-20A and FLD model RF-10Axl. The C_{18} chromatographic column (250 cm x 4.6 mm, 5 μm particle size, model 201 TP54) was obtained from Vydac (Hesperia, CA, USA). Other materials used were SPE columns (Sep-Pak Vac Silica, 500 mg, 55–105 mM particle size), nylon membrane filters (0.45 μm pore size, 13 mm diameter) and nylon syringe filter (0.45 mm pore size, 4 mm diameter) from Waters (Milford, MS, USA).

Samples

Dry soybean samples sold in packs (0.5–1 kg) and bulk (25–60 kg), commercialised in vegetarian shops and food stores of Rio Grande do Sul (RS) and Santa Catarina (SC) states (Southern Brazil) were collected randomly. However, those samples were produced in different Brazilian regions: Sao Paulo-SP, Parana-PR, RS and SC states. They were stored in a refrigerator (4 ± 1°C) before analysis in the Laboratory of Mycotoxicology and Food Contaminants (LABMICO), Food Science and Technology Department, Center of Agricultural Sciences at the Federal University of Santa Catarina (UFSC), Florianopolis. Table 1 shows sample origin, produced and commercialised, planting conditions (traditional and organic) and dimensions (length, width and thickness) characteristics. Dry soybean dimensions were determined by measuring diameter and length of individual beans (n = 50) according to Siqueira et al. (2011). By means of a caliper rule data were collected (mm) in the orthogonal axes (*length*/major axis, *width*/middle axis and *thickness*/minor axis) and their sizes calculated as proposed by Mohsenin (1986).

PAH analysis

The method applied was of Rey-Salgueiro et al. (2009). Briefly, soybean samples (0.5 kg) were milled in an industrial blender jar and thoroughly mixed. A representative subsample of 50 g was collected and small portions were taken and weighed (1 ± 0.0001 g) for PAH extraction with hexane (10 min at 35°C) followed by centrifugation (4000 rpm for 10 min). The procedure was repeated 3 times, the supernatants collected (in total 30 ml) and partially evaporated in a rotary evaporator (ca. 3 ml), filtered, washed 3 times and reduced to 1 ml with a nitrogen stream. Extract clean up was carried out through SPE. The SPE column was solvent conditioned, the sample extract was loaded and the PAHs eluted with hexane. After complete extract evaporation, the residue was redissolved in acetonitrile (500 μl), filtered and collected in amber vials for injection in the HPLC system. The mobile phase gradient started with 70–75% of acetonitrile/water (20 min), followed by 75–100% acetonitrile (15 min) and kept 100% acetonitrile isocratic up to (55 min) at a flow rate of 1 ml/min. The changing wavelength programme of excitation/emission applied for the fluorescence detection was: 0.01 min (274/414 nm) for BaA, Chy and 5MChr; 16.70 min (312/507 nm) for BjF;

Table 1. Soybean (*Glycine max* L.) sample characteristics and PAHs levels in different origins and types.

			Collected		Planting condition			Dimension**(mm)			PAHs[a] ($\mu g\ kg^{-1}$)***							
Sold	Produced	Brand	P[b]	B[c]	O[d]	C[e]	Mean	Range	SD[f]	RSD%[g]	BaA[h]	Chy[i]	5MeChy[j]	BbF[k]	BkF[l]	BaP[m]	DaiP[n]	Σ[o]
RS[p]	RS	NB*	NA[t]	L[u]	NA	C	7.10	6.81–7.40	0.22	3.16	ND	4.05	ND[w]	ND	ND	ND	0.18	4.23
		A	PE	NA	NA	C	7.31	6.20–7.86	0.41	5.56	ND	2.55	7.33	ND	ND	ND	ND	9.88
		B	PE	NA	NA	C	7.09	6.72–7.73	0.29	4.09	ND	ND	0.16	ND	ND	ND	2.72	2.88
		B	PE	NA	NA	C	6.83	6.13–7.58	0.42	6.24	ND	1.98	5.49	ND	ND	ND	0.35	7.82
		NB	NA	L	NA	C	7.38	7.10–7.80	0.17	2.27	ND	3.81	ND	ND	ND	ND	ND	3.81
		NB	NA	L	NA	C	7.48	6.40–8.65	0.53	7.15	ND	0.80	ND	ND	ND	ND	ND	0.80
		NB	NA	L	NA	C	6.56	6.27–6.86	0.17	2.54	ND	16.75	18.24	ND	ND	ND	0.23	35.22
		C	PE	NA	O	NA	6.75	5.82–7.33	0.45	6.65	ND	0.9	0.58	ND	ND	ND	ND	1.48
		C	PE	NA	O	NA	6.96	6.47–7.52	0.27	3.89	ND	ND	ND	ND	ND	ND	ND	ND
		NB	NA	L	NA	C	6.77	6.21–7.30	0.27	3.93	ND	9.48	9.74	ND	ND	ND	8.06	27.28
		A	PE	NA	NA	C	6.77	5.90–7.43	0.43	6.50	ND	6.88	8.72	ND	ND	ND	ND	15.60
	SC[q]	F	PE	NA	NA	C	6.49	5.71–7.46	0.44	6.79	ND	ND	ND	ND	ND	ND	ND	ND
	SP[r]	NB	NA	L	NA	C	6.72	5.51–8.01	0.78	11.67	ND	14.30	5.43	ND	ND	ND	ND	19.73
		NB	NA	L	NA	C	7.05	6.32–7.91	0.51	7.24	ND	8.16	14.74	ND	ND	ND	6.48	29.38
		NB	NA	L	NA	C	7.10	6.27–8.10	0.43	6.08	ND	4.14	12.30	ND	ND	ND	0.23	16.67
		D	PE	NA	NA	C	7.31	6.84–7.83	0.27	3.63	ND	5.02	3.95	ND	ND	ND	ND	8.97
		E	PE	NA	NA	C	NR[v]	NR	NR	NR	ND	ND	0.79	ND	ND	ND	ND	0.79
SC	RS	A	PE	NA	NA	C	7.29	6.42–7.82	0.31	4.26	ND	ND	1.25	ND	ND	ND	6.62	7.87
		G	PE	NA	NA	C	7.58	7.12–9.06	0.50	6.57	ND	18.45	7.75	ND	ND	ND	1.82	28.02
		C	PE	NA	O	NA	5.56	4.88–5.92	0.27	4.90	ND	ND	ND	ND	ND	ND	ND	ND
	PR[s]	H[6]	PE	NA	O	NA	7.19	6.2–7.73	0.36	5.07	ND	6.29	0.18	ND	ND	ND	ND	6.47
		H	PE	NA	NA	C	7.15	6.10–7.60	0.43	6.07	ND	15.51	2.73	ND	ND	ND	ND	18.24
		NB	NA	L	NA	C	7.68	6.84–8.70	0.48	6.24	ND	ND	ND	ND	ND	ND	ND	ND
		NB	NA	L	NA	C	7.35	6.91–8.14	0.29	4.01	ND	23.53	15.25	ND	ND	ND	ND	38.78
		I	PE	NA	NA	C	7.67	7.13–8.35	0.47	6.13	ND	19.39	2.70	ND	ND	ND	ND	22.09
		NB	NA	L	NA	C	6.41	5.69–7.27	0.43	6.74	ND	7.97	1.37	ND	ND	ND	ND	9.34
		NB[7]	NA	L	NA	C	NR	NR	NR	NR	ND	3.75	ND	ND	ND	ND	ND	3.75
	SC	NB	NA	L	NA	C	6.62	5.60–7.51	0.50	7.61	ND	3.63	4.92	ND	ND	ND	ND	8.55
		NB	NA	L	NA	C	7.50	7.02–8.66	0.42	5.62	ND	9.65	11.20	ND	ND	ND	ND	20.85
		NB	NA	L	NA	C	6.81	6.07–7.64	0.44	6.41	ND	2.04	3.39	ND	ND	ND	ND	5.43
		NB	NA	L	NA	C	7.26	6.11–8.05	0.50	6.83	ND	ND	1.79	ND	ND	ND	ND	1.79
		J[4]	PE	NA	NA	C	7.38	7.10–8.00	0.47	6.13	ND	ND	2.81	ND	ND	ND	ND	2.81
		K	PE	NA	NA	C	7.40	6.71–8.43	0.44	5.97	0.5	9.25	ND	ND	ND	ND	ND	9.75
		F	PE	NA	NA	C	7.30	6.32–7.72	0.34	4.66	ND	14.46	ND	ND	ND	ND	ND	14.46
		L[5]	PE	NA	O	NA	6.19	5.60–6.83	0.32	5.09	ND	1.72	ND	ND	ND	ND	ND	1.72
	SP	NB	P	NA	NA	C	6.05	5.60–6.40	0.24	3.91	ND	1.29	10.71	ND	ND	ND	ND	12.00
		NB[3]	P	NA	NA	C	NR	NR	NR	NR	58.75	103.89	16.10	18.52	3.71	3.49	ND	204.46
		M	PE	NA	NA	C	7.37	6.90–7.96	0.26	3.49	0.65	31.04	4.56	1.36	ND	ND	ND	37.61
		I	PE	NA	NA	C	7.45	7.12–8.78	0.40	5.35	ND	18.03	1.1	ND	ND	ND	ND	19.13
Total samples: 39								**PAH/total sample:**			03/39	30/39	28/39	02/39	01/39	01/39	09/39	NA

[a]polycyclic aromatic hydrocarbons, [b]pack (PE: polyethylene), [c]bulk (loose), [d]organic, [e]commercial, [f]standard deviation, [g]relative standard deviation, [h]benzo(a)anthracene, [i]chrysene, [j]5-methylchrysene, [k]benzo(b) fluoranthene, [l]benzo(k)fluoranthene, [m]benzo(a)pyrene, [n]dibenzo(al)pyrene, [o]sum of total PAHs detected levels/sample, [p]Rio grande do Sul, [q]Santa Catarina, [r]São Paulo, [s]Paraná, [t]not applicable, [u]loose, [v]not registered, [w]not detected (*[1]in bulk,[2]organic [government certified])[3] match and peeled, [4]non-trangenic, [5]claims to be organic, not government certified though, [6]organic [IDB seal], [7]barbecue flavoured*)

*brand not specified

**width/middle/length axes – formula: bean diameter = (length x width x middle)

*** Benzo(j)fluoranthene (BjF); dibenzo(al)pyrene (DalP); dibenzo(ah)anthracene (DahA); indeno(cd)pyrene (IcdP); dibenzo(ae)pyrene (DaeP); dibenzo(ah)pyrene (DahP) were below LOQ.

18.20 min (290/430 nm) for BbF, BkF, BaP, DalP and DahA; 32.40 min (300/500 nm) for IcpD; 34.90 min (297/403 nm) for DaeP; 45 min (304/457 nm) for DaiP and DahP. For quality control of the routine analytical process, samples were analysed on different days and studies on PAH recovery were performed to demonstrate the viability of the method. This work was performed in the LABMICO Laboratory at UFSC and Food Science and Quality Center at ITAL (Food Technology Institute), accredited by MAPA (Ministry of Agriculture and Food Supplies) and the National Institute of Metrology, Quality and Technology (INMETRO), following ISO/IEC 17.025 (2005) procedures. Measurement uncertainty (data not shown in the table) was performed according to Commission Regulation No. 836/2011 (EC 2011b).

Results and discussion

Sample characterisation

The sample characteristics regarding soybean origin (where they were grown, produced and sold), the planting conditions (traditional or organic) and grain dimensions (format: width, length and thickness) are given in Table 1. As far as the origin of the soybeans sold in both Southern Brazil states is concerned, most of the soybeans sold in RS were produced in that state, followed by SP. Otherwise, the ones sold in SC were produced among the 4 states PR/RS/SC/SP. Regarding planting conditions, most of the soybean samples were from conventional crops, being only 11% and 9% from organic crops sold in RS and SC state vegetarian shops, respectively. Those samples were produced among the Southern region states surveyed (PR, RS, SC) only.

The grain dimension characteristics showed that soybean samples had different sizes, which may lead to different pyrolys reaction, with variation on PAHs formation and levels (Nazzal 2008). Part of them was of small size (mean: 6.1 mm – min 5.6; max 6.6) mainly from RS state and large (mean: 7.2 mm – min 6.6; max 7.7) from PR, SP, SC states, similar to the main two varieties utilised for region environment adequacy cultivation in Brazil. Previously, a study of Nazzal (2008) showed that the increase of the particle grain size caused a decrease in the derived hydrocarbon gases. On the other hand, decreasing the particle grain size used in the study caused an increase in the PAH concentration in the derived oil, which has been suggested as a result of the aromatisation of the derived oils by secondary decomposition reactions.

PAH method validation

Calibration curves with mean correlation coefficients of 0.994 were obtained for all PAHs. Recoveries at concentrations of 0.26, 45 and 132 µg kg^{-1}, determined in triplicate, were 73% to 101% with SDs ranging from 4.3% to 10.3%. The limits of quantification (LOQs) were calculated as the lowest concentrations used in the calibration curves. These data were 1.0 µg kg^{-1} for BjF/IcdP and 0.1 µg kg^{-1} for the other PAHs. From these data it was concluded that the applied method was valid.

PAH levels

In 39 soybean samples surveyed and sold in both (RS and SC) states, 7 PAHs (BaA, Chy, 5MeChy, BbF, BkF, BaP and DaiP) were detected (Table 1). Chy and 5MeChy were the main PAHs, with 76.9% and 71.8% positive samples, respectively, followed by DaiP (23.1%). Chy levels ranged from 0.8 to 31.04 µg kg^{-1}, with only 1 exception of a quite high level (103.89 µg kg^{-1}), which was from SP growers. 5MeChy levels ranged from 0.16 to 18.24 µg kg^{-1} and DaiP from 0.18 to 8.06 µg kg^{-1}. BkF and BaP were also registered, however at the lowest concentrations. Figure 1 shows the different types of PAHs detected in the same soybean sample, from which it is possible to observe that most of the positive samples were contaminated by 2 PAHs (50%) followed by 3 to 6 PAHs (30%). It is important to highlight that there is no ML established for PAH contamination in soybeans. However, taking into account the ML for BaP in processed cereals-based foods and oils and fats, 1 (2.5%) sample did not comply with current regulations for both products. On the other hand, taking to account the ML for the sum of PAH4 in processed cereals-based foods and oils and fats, 72% and 26% samples do not comply with current regulations, respectively. This occurred mainly at high Chy contamination.

ΣPAHs levels per sample ranged from 0.79 to 38.78 µg kg^{-1} (Table 1), except 1 sample with Chy of 103.89 µg kg^{-1}, which reached a ΣPAHs of 204.46 µg kg^{-1} with 6 compounds (BaA, Chy, 5MeChy, BbF, BkF and BaP at 58.75, 103.89, 16.10, 18.52, 3.71 and

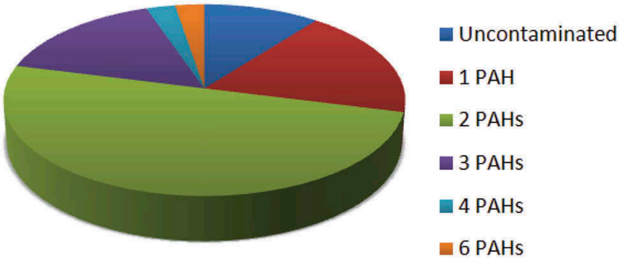

Figure 1. Number of polycyclic aromatic hydrocarbons found per soybean sample.

3.49 µg kg^{-1}, respectively). It was the most contaminated sample, grown in SP state (Southeastern region) and sold in the SC state market. This was probably due to bean crop grown under contaminated environment (air, soil, water, industry chimneys) or near industrial area with high car traffic roads (Camargo & Toledo 2002; Camargo et al. 2011; Garcia et al. 2014; Scussel et al. 2014, 2015). PAH formation is also reported due to processing, packaging and thermal processes like drying, smoking, baking, roasting, grilling, frying and others. Smoke drying of oilseeds, drying degrees and refining processes greatly varied PAHs content among different batches. In a study of Rojo Camargo et al. (2012) direct smoke drying of soybean seeds presented 10–208 µg kg^{-1} and 26–316 µg kg^{-1} PAH levels in 2007 and 2008. In our study, only 10.3% of the dry soybeans samples did not show any PAHs contamination (< LOQ).

In addition, Paraíba et al. (2010) reported PAHs in maize grown in soils treated with sewage sludge. The highest contamination (mean: 3 µg kg^{-1}) was of phenanthrene (Phe), a low molecular mass PAH, which was not included in the current survey. Falco et al. (2003) analysed several foods, among them beans, rice and lentils and reported contamination ranging from 0.11 to 4.28 µg kg^{-1} with the sum of PAHs reaching 2.89 µg kg^{-1}, where the PAH with the highest contamination was also Phe. Martorell et al. (2010) reported PAHs in beans, lentils, peas and chickpeas with sum levels varying from 0.75 to 2.30 µg kg^{-1}. On the other hand, Camargo and Toledo (2002) did not detect PAHs in beans, however in rice and pea, the mean sum was 0.13 and 1.31 µg kg^{-1}, respectively. The highest rice contamination was of BaP (0.13 µg kg^{-1}) and in peas of BbF (0.48 µg kg^{-1}).

There are no MLs for soybean established, either in the country or elsewhere. MLs set for other food and especially for fat and oils (10 µg kg^{-1} for the PAHs4 only.). Despite of not being detected in the soybean samples, all PAHs4 and no BaP levels were rather high if compared to those for high lipid content extracts (oils and fats). Indeed, soybean is a pulse also high in lipids, utilised for oil extraction. In a study of Camargo et al. (2011), the presence of PAHs in different brands of soybean oils available on the Brazilian market was detected in all 42 samples with mean summed PAH levels ranging from 10.4 to 112.0 µg kg^{-1}. In addition, Tfouni et al. (2014) showed that different brands of oil blends (mainly a mixture of olive and soybean oils) were contaminated with mean sums ranging from 2.59 µg kg^{-1} to 85.30 µg kg^{-1} and 50% of the samples were not in accordance with MLs for BaP and PAH4 established in the Brazilian and/or European regulations for this food. Regarding to dietary intake of food contaminants like PAHs, EFSA (2008b) summarised that the overall dietary exposure, assuming a person of 60 kg, is about 235 ng day^{-1} (BaP), 641 ng day^{-1} (PAH2: BaP, Chy), 1168 ng day^{-1} (PAH4: PAH2, BaA, BbF), and 1729 ng day^{-1} (PAH8: PAH4, BkF, benzoperylene-BPY, DahA, IcdP). Therefore, the relationship between their quantities contained in food products as well as nutritional habits should be taken into account

Despite the samples that were mostly contaminated (79.4%), half of them had only 2 PAHs per sample (Figure 1). That should be of concern for vegans as they focus on healthy food and more often consume soybean protein. The source of that contamination could be either from field exposure (atmosphere and roads contamination deposition) or from the drying process applied post-harvest (high temperature and smoke contact). Therefore, methods of PAH reduction need to be surveyed worldwide, taking into account prevention of contamination of food with PAH as smoking and direct drying processes (Codex 2009).

Conclusion

In dry soybean samples sold for direct human consumption PAHs were detected in 89.7% of them, mainly Chy, 5MeChy and DaiP. The highest contamination and PAH number per sample (total: 6) were of soybeans grown and produced in SP and PR states, sold in SC market, followed by the ones grown in RS state and sold in the same state. Regarding organic *versus* commercial soybean planting conditions, no PAHs contamination differences were observed. It is important to emphasise that, despite of contaminants presence in the soybean samples surveyed for human consumption, there is no regulation. In order to reduce PAH contamination and formation, the strategies should be through (a) choosing soybean planting areas away from factories smoke chimneys or heavy traffic roads; (b) applying electricity oven instead of wood, as the heat source, in the drying process. Although it is known that their high temperatures (despite the heat source) can induce PAHs formation and/or (c) applying mild temperature during the drying process, if possible.

Acknowledgement

The authors thank CAPES (Coordination for the Improvement of Higher Education Personnel) sponsors for providing a grant to Laura Pereira Garcia.

Disclosure statement

No potential conflict of interest was reported by the authors.

References

Alomirah H, Al-Zenki S, Al-Hooti S, Zaghloul S, Zawaya W, Ahmed N. 2011. Concentration and dietary exposure to polycyclic aromatic hydrocarbons from grilled and smoked foods. Food Control. 22:2028–2035.

Bøhn T, Cuhra M, Traavik T, Sanden M, Fagan J, Primicerio R. 2014. Compositional differences in soybeans on the market: glyphosate accumulates in Roundup Ready GM soybeans. Food Chem. 153:207–215.

Camargo M, Toledo M. 2002. Avaliação da Contaminação de Diferentes Grupos de Alimentos por Hidrocarbonetos Policíclicos Aromáticos. Braz J Food Technol. 5:19–26.

Camargo MCR, Antoniolli PR, Vicente E, Tfouni SA. 2011. Polycyclic aromatic hydrocarbons in Brazilian commercial soybean oils and dietary exposure. Food Additives Contaminants: Part B. 4:152–159.

Codex Alimentarius Commision. 2009. Code of practice for the reduction of contamination of food with polycyclic aromatic hydrocarbons (PAH) from smoking and direct drying processes CAC/RCP 68-2009. Rome: Food and Agriculture Organization of the United Nations; p. 1–15.

EC - European Commission. 2011a. Commission regulation (EU) no 835/2011 of 19 August 2011 amending regulation (EC) no 1881/2006 as regards maximum levels for polycyclic aromatic hydrocarbons in foodstuffs. Off J Eur Comm. L215:7–8.

EC - European Commission. 2011b. Commission Regulation (EU) No 836/2011 of 19 August 2011 amending Regulation (EC) No 333/2007 laying down the methods of sampling and analysis for the official control of the levels of lead, cadmium, mercury, inorganic tin, 3-MCPD and benzo (a)pyrene in foodstuffs. Off J Eur Union. L215:9.

EFSA - European Food Safety Authority. 2008a. Food Consumption, Database. Available from: http://www.efsa.europa.eu/EFSA/ScientificPanels/efsa_locale-1178620753812_DATEX.htm.

EFSA - European Food Safety Authority. 2008b. Scientific opinion of the panel on contaminants in the food Chain on a request from the European Commission on polycyclic aromatic hydrocarbons in food. EFSA J. 724:1–114.

Falco G, Domingo JL, Llobet JM, Teixido A, Casas C, Muller L. 2003. Polycyclic aromatic hydrocarbons in foods: human exposure through the diet in Catalonia, Spain. J Food Prot. 66:2325–2331.

Garcia LP, Gonçalves BL, Panho G, Scussel VM. 2014. Hidrocarbonetos policíclicos aromáticos em alimentos: uma revisão. Pubvet. 8:2292–2450.

Gomes DP, Kronka AZ, Barrozo LM, Da Silva RP, Souza AL, Silva BMS, Panizzi RC. 2009. Efeito da colhedora, velocidade e ponto de coleta na contaminação de sementes de soja por fungos. Rev Bras Sementes. 31:160–166.

ISO/IEC 17.025. 2005. General requirements for the competence of testing and calibration laboratories. 2nd ed. Geneva: International Organization for Standardization/International Electro technical Commission; p. 31.

Martorell I, Perelló G, Martí-Cid R, Castell V, Llobet JM, Doming JL. 2010. Polycyclic aromatic hydrocarbons (PAH) in foods and estimated PAH intake by the population of Catalonia, Spain: temporal trend. Environ Int. 36:424–432.

Mohsenin NN. 1986. Physical properties of plant and animal materials. New York: Gordon and Breach Publishers; p. 841.

Nazzal JM. 2008. The influence of grain size on the products yield and shale oil composition from the pyrolysis of sultani oil shale. Energ Convers Manage. 49:3278–3286.

Paraíba LC, Queiroz SCN, Maia AHN, Ferracini VL. 2010. Bioconcentration factor estimates of polycyclic aromatic hydrocarbons in grains of corn plants cultivated in soils treated with sewage sludge. Sci Total Environ. 408:3270–3276.

Plaza-Bolaños P, Frenich AG, Vidal JLM. 2010. Polycyclic aromatic hydrocarbons in food and beverages. Analytical methods and trends. J Chromatogr. 1217:6303–6326.

Pongpiachan S. 2015. A preliminary study of using polycyclic aromatic hydrocarbons as chemical tracers for traceability in soybean products. Food Control. 47:392–400.

Reinik M, Tamme T, Roasto M, Juhkam K, Tenno T, Kiis A. 2007. Polycyclic aromatic hydrocarbons in meat products and estimated PAH intake by children and the general population in Estonia. Food Addit Contamin. 24:429–437.

Rengarajan T, Rajendran P, Nandakumar N, Lokeshkumar B, Rajendran P, Nishigaki I. 2015. Exposure to polycyclic aromatic hydrocarbons with special focus on cancer. Asian Pac J Trop Biomed. 5:182–189.

Rey-Salgueiro L, Martínez-Carballo E, García-Falcón MS, González-Barreiro C, Simal-Gándara J. 2009. Occurrence of polycyclic aromatic hydrocarbons and their hydroxylated metabolites in infant foods. Food Chem. 115:814–819.

Rojo Camargo MC, Antoniolli PR, Vicente E. 2012. Evaluation of polycyclic aromatic hydrocarbons content in different stages of soybean oils processing. Food Chem. 135:937–942.

Savi GD, Piacentini K, Scussel VM. 2014. Mycoflora and occurrence of deoxynivalenol in whole wheat grains (Triticum aestivum L.). Vol. 7. from. Southern Brazil: Food Addit Contam; 232–237.

Scussel VM, Christ D, Valmorbida R, Kreibich HH, Silva J, Savi G 2015. Características microscópicas de deterioração de grãos de soja por fungos e enzimas. VII Congresso Brasileiro de Soja, Sessão Biotecnologia; 22–25 Jun; Florianopolis; p. 41.

Scussel VM, Garcia L, Tfouni SAV, Leonel B. 2014. A secagem de grãos e os hidrocarbonetos policíclicos aromáticos (HPAs). In: Lorini I, Oliveira C, editores. Anais da VI Conferência Brasileira de Pós-colheita e o VIII Simpósio Paranaense de Pós-colheita. Maringa: Parana; p. 214–223.

Siqueira VC, Resende O, Chaves TH. 2011. Contração volumétrica dos frutos de pinhão-manso durante a secagem em diversas temperaturas. Rev Bras Armazen. 36:171–178.

Teixeira VH, Casal S, Oliveira MBPP. 2007. PAHs content in sunflower, soybean and virgin olive oils: evaluation in commercial samples and during refining process. Food Chem. 104:106–112.

Tfouni SAV, Padovani GR, Reis RM, Furlani RPZ, Camargo MCR. 2014. Incidence of polycyclic aromatic hydrocarbons in vegetable oil blends. Food Control. 46:539–543.

Vives I, Grimalt JO, Guitart R. 2001. Los hidrocarburos aromáticos policíclicos y lasaludhumana. Apunt Cienc Tecnol. 3:45–51.

Xia Z, Duan X, Tao S, Qiu W, Liu D, Wang Y, Wei S, Wang B, Jiang Q, Lu B, et al. 2013. Pollution level, inhalation exposure and lung cancer risk of ambient atmospheric polycyclic aromatic hydrocarbons (PAHs) in Taiyuan, China. Environ Pollut. 173:150–156.

Yin X, Liang X, Xu G, Zhan X. 2013. Effect of phenanthrene uptake on membrane potential in roots of soybean, wheat and carrot. Environ Exper Bot. 99:53–58.