Determination and Occurrence of Bisphenol A, Bisphenol A Diglycidyl Ether, and Bisphenol F Diglycidyl Ether, Including Their Derivatives, in Canned Foodstuffs' from the Czech Retail Market

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Abstract

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A several-year survey (2000–2006) documents a continuing occurrence of bisphenol A (BPA), bisphenol A diglycidyl ether (BADGE), and bisphenol F diglycidyl ether (BFDGE), including their derivatives, migrating from packaging into food. A wide range of bisphenols levels (from traces up to hundreds μg/kg) in canned foodstuffs available at the Czech retail market was found. An analytical procedure suitable for routine monitoring of bisphenols in various matrices was validated. Crude extracts (obtained by dichloromethane extraction in ultrasonic bath) were purified by gel permeation chromatography (GPC), identification/quantification was carried out by HPLC/FLD method. Optimised procedure allowed to measure trace levels of the target analytes (LODs – 3 μg/kg) with good repeatability (RSDs – 3% at level 100 μg/kg) and recoveries exceeding 75%.

Keywords: bisphenols; hydrolysis and chlorohydroxy derivatives; migration; can lacquers; foodstuff; contaminants

The use of thin plastic coatings on the interior surface of metal cans is the most common way to avoid their corrosion and the contamination of food with dissolved metals ions. In addition, the coating helps to prevent the canned foods from becoming tainted. Epoxy-based or PVC organosol resins are used for this purpose. Epoxy resins are commonly synthesised from bisphenol A (BPA, CAS 80-05-7) and they also contain bisphenol A diglycidyl ether (BADGE, CAS 1675-54-3). For PVC organosol resins production, additives such as BADGE and bisphenol F diglycidyl ether (BFDGE, CAS 2095-03-6) are required to prevent the thermal degradation of the polymer by hydrochloric acid formed during the curing process. Residual BPA, BADGE, and BFDGE monomers can migrate

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into the preserved food, the formation of various chlorohydroxy compounds such as BADGE·HCl, BADGE·2HCl, BFDGE·HCl and BFDGE·2HCl also occurrs. Moreover, the remaining epoxy groups can be hydrolysed, which leads to the formation of mono- and dihydrolysed products such as BADGE·H_2O, BADGE·$2H_2O$, BADGE·H_2O·HCl, BFDGE·H_2O, BFDGE·$2H_2O$, and BFDGE.H_2O·HCl. The structures of BPA, BADGE, BFDGE and their reaction products are illustrated in Figure 1. Similarly as in the case of other food contaminants, the occurrence of migrants in human diet is associated with consumers' health concerns. The recent evaluation of bisphenols migration based on new toxicological data is reflected in the new European Union legislation (valid from January 1st, 2006) established in Commission Regulation (EC) No. 1895/2005 on the restriction of the use of certain epoxy derivatives in materials and articles intended to come into contact with food, in which specific migration limits (SMLs) in Annex I for two BADGE groups are defined: (a) sum of migrations of BADGE including its hydrolysis derivates: 9 mg/kg in food or food simulant, and (b) sum of migrations of BADGE chlorohydroxy derivatives: 1 mg/kg in food or food simulants. The use of BFDGE and novolac glycidyl ethers (NOGE) is no more permitted, but existing stocks are allowed to be sold. This important change from the previous provisional SMLs of 1 mg/kg in foodstuffs or in food simulants specified in Annex I and II of Commission Directive No. 2002/16/EC (on the use of certain epoxy derivatives in materials and articles intended to come into contact with foodstuffs) for BADGE and BFDGE including their hydrolysis and chlorohydroxy derivatives significantly modifies both the food inspection strategy and the demands on the analytical procedures used. In any case, the monitoring of the above mentioned migrants in food supply is an important strategy aimed at the control of regulations implementation and, consequently, a measure protecting consumers' health.

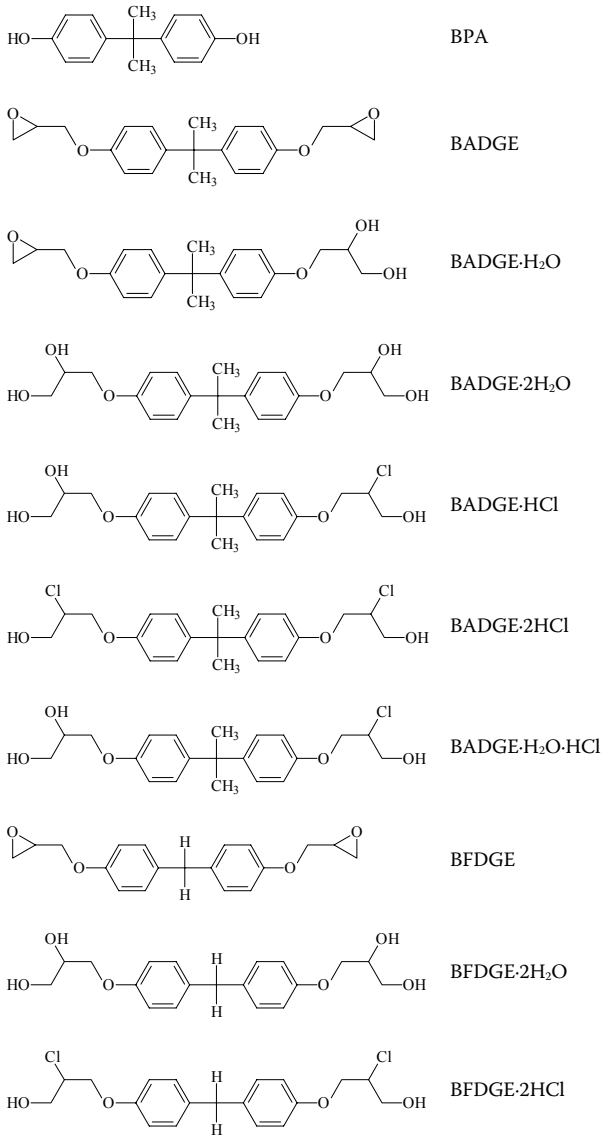

Figure 1. Chemical structures of BPA, BADGE, BFDGE, and its investigated derivatives

Following the first report updating the issue of bisphenols migration from can lacquers into preserved food (Biedermann *et al.* 1996), numerous studies were carried out to monitor some of the mentioned migrants in canned food samples like various fish and seafood in oil (Biedermann *et al.* 1996, 1997; Bronz *et al.* 1997; Roubtsova *et al.* 1997; Biedermann & Grob 1998; Rauter *et al.* 1999; Simoneau *et al.* 1999; Hammarling *et al.* 2000; Lintschinger & Rauter 2000; Theobald *et al.* 2000; Berger *et al.* 2001), and also various vegetables and fruits (Biedermann *et al.* 1999a, b; Lintschinger & Rauter 2000). In a comprehensive survey published in the late 90s, the migration of bisphenols from various can lacquers and their distribution between the solid and liquid portions of the can content (Summerfield *et al.* 1998) were reported.

Various chromatographic methods applicable for the analysis of BPA, BADGE, BFDGE and their reaction products in food simulants were published. Most of those used reversed phase high performance liquid chromatography (RP-HPLC) on C18 and/or C8 silica stationary phases with

acetonitrile/water mixtures as the mobile phase. Fluorescence detection (FLD) was used as a method of the first choice in many studies (Biedermann *et al.* 1996; Biedermann & Grob 1998). As far as more selective detection/confirmation of target analytes was required, mass spectrometry (MS) coupled with either RP-HPLC (Simal Gandara *et al.* 1992; Berger & Oehme 2000; Berger *et al.* 2001) or gas chromatography, GC (Roubtsova *et al.* 1997; Salafranca *et al.* 1999) was applied in laboratories concerned with food chemical safety. While employing RP-HPLC separation step, lipids must be removed from the crude extract prior to injection due to their very high capacity factor values on such columns; in the case of normal phase HPLC (NP-HPLC), direct injection of "oily" samples is possible (no sample preparation is needed), under this set-up the affinity of lipids to polar stationary phases is low, thus their elution occurs close to the dead volume (Biedermann *et al.* 1996; Biedermann & Grob 1998; Uematsu *et al.* 1998). Considering a wide range of physical-chemical properties of BADGE, BFDGE, and their derivatives, it is not surprising that more analytical strategies had to be combined to enable the examination of various canned foods for the whole set of these migrants (Biedermann *et al.* 1999a, b). In the first step, RP-HPLC/FLD was utilised to identify the apparently contaminated samples, in the next phase the confirmation was carried out based on acetylation followed by analysis using NP-HPLC/FLD. In the case of inconsistency of the results obtained by these two approaches, NPLC fractions were collected and analysed by GC-MS. Lintschinger and Rauter (2000) used RP-HPLC with a binary mobile phase system consisting of methanol/water and acetonitrile/water to separate BADGE, BFDGE, and their derivatives. However, this approach did not enable a sufficient resolution BADGE·H_2O and BADGE·H_2O·HCl, an isocratic method of methanol/water was required to complete the separation. An improved resolution of all compounds concerned was later demonstrated in RP-HPLC system with water/methanol gradient elution (Leepipatpiboon *et al.* 2005).

Within this study, the results are reported of bisphenols monitoring in various categories of canned food obtained from the Czech retail market during the sampling in the years 2000–2006. For this purpose, the validation of a simple analytical procedure suitable for the analysis of a wide range of food matrices was carried out.

MATERIAL AND METHODS

Chemicals. Certified standards of bisphenol A (BPA) – 99%, bisphenol A diglycidyl ether (BADGE) – 97%, bisphenol A glycidyl (2,3-dihydroxypropyl) ether (BADGE·H_2O) – 95%, bisphenol A bis(2,3-dihydroxypropyl) ether (BADGE·2H_2O) – 95%, bisphenol A (3-chloro-2-hydroxypropyl) (2,3-dihydroxypropyl) ether (BADGE·H_2O·HCl) – 95%, bisphenol A glycidyl (3-chloro-2-hydroxypropyl) ether (BADGE·HCl) – 90%, bisphenol A bis(3-chloro-2-hydroxypropyl) ether (BADGE·2HCl) – 95%, bisphenol F diglycidyl ether (BFDGE) – 95%, bisphenol F bis(2,3-dihydroxypropyl) ether (BFDGE·2H_2O) – 95%, bisphenol F bis(3-chloro-2-hydroxypropyl) ether (BFDGE·2HCl) – 95% were obtained from Fluka (Switzerland). All bisphenol F compounds were mixtures of three isomers: ortho-ortho, ortho-para, para-para. Stock solutions were prepared in the concentration of 1 mg/ml acetonitrile. Standard mixtures for calibration were obtained by appropriate dilution in 40% acetonitrile to the concentrations of 2, 5, 10, 20, 50, and 100 ng/ml (i.e. contamination level 10 to 500 µg/kg according to the sample preparation). Distilled water was purified using Milli-Q Purifier (Millipore, USA). Acetonitrile (gradient grade for chromatography, Merck, Germany), cyclohexane, and dichloromethane (for organic trace analysis, Merck, Germany) were used as supplied. Sodium sulphate, anhydrous (Penta Praha, Czech Republic) was heated at 500°C for 4 h and stored in a tightly capped bottle up to 2 weeks.

Sample preparation. All samples of various canned food in differently shaped packages were purchased on the retail market in the Czech Republic. The whole content of packaging was homogenised using a laboratory blender (Waring, USA). 1.25 g of the sample was blended with 10 g of anhydrous sodium sulphate and the flowing powder obtained was extracted with 30 ml of dichloromethane in ultrasonic bath (Sonorex Super 510H, Bandelin electronics, Germany) for 5 minutes. The crude extract was filtered under vacuum using Büchner funnel and the remaining sample in the flask was rinsed up with approx. 30 ml of dichloromethane. The combined extracts were evaporated to dryness using rotary vacuum evaporator Büchi B-178 (Büchi, Switzerland). The residue was transferred to 25 ml volumetric flask and made up with dichloromethane:cyclohexane (1:1, v/v) mixture. 2 ml aliquot of this extract was

cleaned-up by gel permeation chromatography (GPC) using Bio-Beads S-X3 column (column dimensions – 500 × 8 mm i.d., styrene-divinylbenzene copolymer Bio-Beads S-X3, 40–80 µm, Bio Rad, USA) under the following conditions: mobile phase – dichloromethane:cyclohexane (1:1, v/v), flow rate – 0.6 ml/min, injection loop volume – 2 ml, collected fraction 12.5–25 ml. After evaporation of this eluate to dryness using rotary vacuum evaporator, the residue was dissolved in 0.5 ml of acetonitrile and analysed by HPLC-FLD (i.e. 0.2 g of original matrix in 1 ml).

Separation, detection, quantification. HPLC system consisted of Hewlett Packard 1100 Series chromatograph (degasser, quaternary pump, autosampler, fluorescence detector) and the separation column LiChroCART 250-4 (250 mm × 4 mm i.d., LiChrospher 100 RP-18e, 5 µm, coupled with a guard column 4 mm × 4 mm i.d., Merck, Germany). 20 µl of acetonitrile sample solutions were injected into HPLC system and separated under the following conditions: mobile phase flow rate 1 ml/min, gradient elution [A – acetonitrile, B – water; 0–1 min: 40% A, 1–12 min: 40–100% A, 12–13 min: 100% A, 13–15 min: 100% A at the flow rate of 2 ml/min (column flushing), 15–20 min: 40% A at the flow rate of 1 ml/min (reconditioning)]. The column oven temperature was held at 40°C, fluorescence detection was carried out using 233 nm as the excitation and 310 nm as the emission wavelengths. An external calibration method using the calibration curve in the concentration range of 2–100 ng/ml (see chemicals) was used for quantification.

Method validation. For the in house method validation, a spiked sample of pork luncheon meat with only a background contamination (analytes at detection limits) was used. Acetonitrile solution of bisphenols standard mixture (100 µl of concentration 25 µg/ml) was thoroughly incorporated into 25 g of finely homogenised matrix to obtain the contamination level of 100 µg/kg of each individual analyte. The sample prepared in this way was stored at 4°C for 4 days before analysis.

RESULTS AND DISCUSSION

In the following paragraphs, the results are presented as achieved in our study concerned with selected migrants potentially occurring in canned food. Both the analytical issues and the data generated by the implemented procedure are discussed below.

Validation of analytical procedure. Obviously, the contents of the canned foods can largely differ in their characters ranging from almost homogeneous material (e.g. pates, spreads) to the coarse suspensions and mixtures consisting or solid and liquid/semiliquid components (sardines in oil, olives etc.). Therefore, to characterise the extent of migration from the internal coatings into the particular type of canned food, thorough homogenisation of the whole content was carried out prior to the sampling of a representative aliquot for further analysis. Among the solvents conceivable for the isolation of a wide range of analytes involved in this study, dichloromethane

Table 1. Method performance characteristics (n = 6)

Compound	Recovery (%)	CV_r(%)	LOD (µg/kg)	LOQ (µg/kg)
BPA	83	3.0	3	10
BADGE	80	2.4	3	10
BADGE·H_2O	81	2.7	3	10
BADGE·2H_2O	75	2.8	3	10
BADGE·HCl	81	3.0	3	10
BADGE·2HCl	82	2.6	3	10
BADGE·H_2O·HCl	79	2.6	3	10
BFDGE	78	2.5	3	10
BFDGE·2H_2O	77	2.4	3	10
BFDGE·2HCl	92	2.3	3	10

Values measured for spiked pork luncheon meat to the level 100 µg/kg (see experiment)

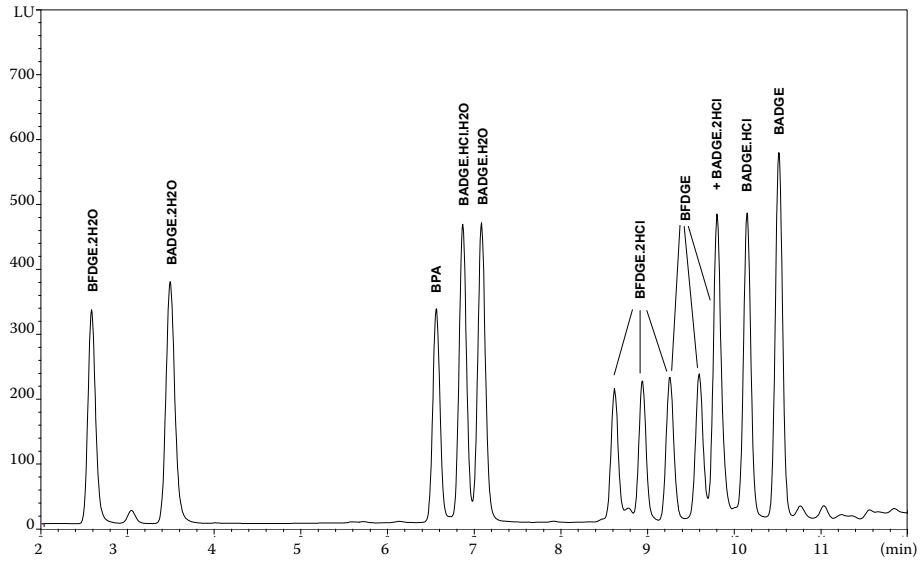

Figure 2. Chromatogram of bisphenols standard mixture (concentration 100 ng per ml, 20 µl injected)

was identified as suitable for an effective multi-analyte/multimatrix approach. In the next step, purification strategy was considered since many matrix co-extracts were contained in the crude dichloromethane extract. Because of a relatively wide range of properties (namely polarities) of the target analytes, a less selective gel permeation chromatographic separation was chosen to minimise the losses during the clean-up. It should be noted that this technique is particularly suitable for the purification of extracts obtained from fatty samples. Regarding the determinative step, relatively common equipment such as HPLC coupled with FLD was shown to be adequate for unbiased identification/quantification of bisphenol levels in various matrices. Moreover, the application of liquid chromatography significantly facilitates the sample preparation (no derivatisation is needed) which results in a cost-effective analytical procedure. The performance characteristics of the method developed are summarised in Table 1, acceptable recoveries not below 75% prove the sufficient extraction efficiency of dichloromethane. Also the repeatability of the measurements and the detection limits attainable by this method meet the requirements for a reliable analysis of the complete list of the bisphenols investigated including all more polar derivatives. As regards the calibration (linearity range), very good correlation coefficients ($R^2 \geq 0.999$) for all compounds in the concentration range followed (2–100 ng/ml) were achieved. In Figure 2, HPLC/FLD chromatogram

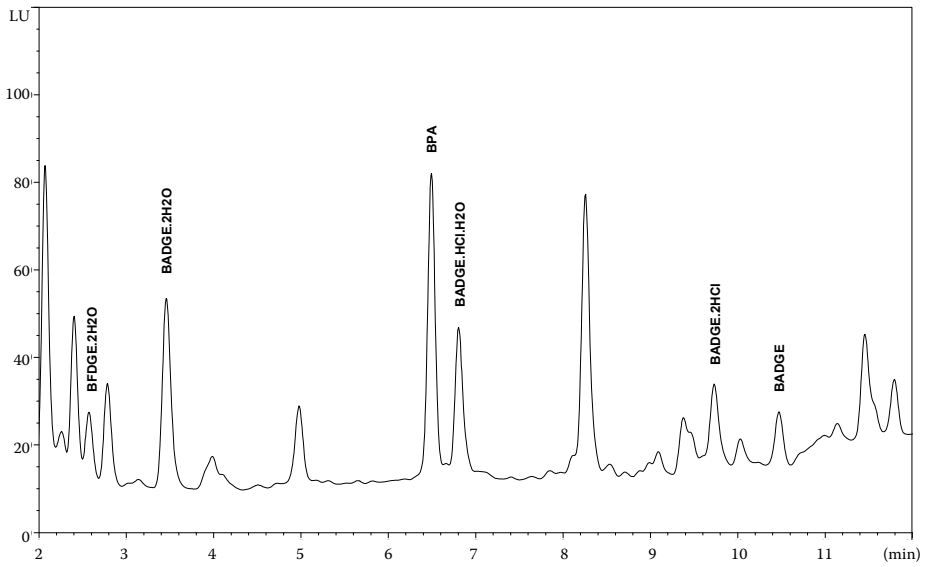

Figure 3. Example of HPLC/FLD analysis of contaminated canned tuna fish (sum of bisphenols 200 µg/kg)

Table 2. External quality control: results of proficiency testing achieved within FAPAS®

Date	Matrix	Analyte	Result# (µg/kg)	Assigned value* (µg/kg)	Z-score*
Feb-03	Sunflower oil	BADGE	340	390	–0.7
		BADGE·H_2O	160	230	–1.5
		BADGE·HCl	480	540	–0.7
		BADGE·H_2O·HCl	200	280	–1.4
Nov-03	Olive oil	BADGE	524	798	–2.1
		BADGE·HCl	116	143	–0.9
Oct-04	Sunflower oil	BADGE·HCl	61	90	–1.5
		BADGE·2HCl	405	477	–0.8
		BADGE·H_2O·HCl	349	491	–1.6
Oct-05	Sunflower oil	BADGE	1731	1483	1.1
		BFDGE	1218	949	1.8

#results were corrected for recovery as required; *for detailed description and definitions see reference (Thompson & Wood 1993)

of bisphenols standard mixture is presented. As can be seen, the elution bands of BFDGE·2HCl and BFDGE clusters (in both cases consisting of three isomers with the probable elution order on reverse phase: p,p'; o,p'; o,o' – (Biedermann *et al.* 1997) are not fully resolved due to the close retention times. Also, BADGE·2HCl is coeluted with the third BFDGE isomer (o,o' configuration). These problems, in some cases potentially complicating the data evaluation, were earlier discussed also by other authors (Lintschinger & Rauter 2000). The calculation of the proportional contributions to the responses of individual analytes in the corresponding isomeric clusters is based on the specific response factors. The trueness of the data generated by the optimised procedure was verified within laboratory proficiency testing FAPAS – Food Analysis Performance Assessment Scheme® (organised by CSL York, UK). As shown in Table 2, satisfactory results were achieved during four rounds.

Survey of bisphenols occurrence in canned food. In Table 3, mean levels are summarised of bisphenols determined in the selected samples from the Czech retail market obtained within the survey realised in the years 2001–2006. The spectrum of target bisphenols was gradually expanded in accordance with the commercial availability of analytical standards. While at the beginning, in the

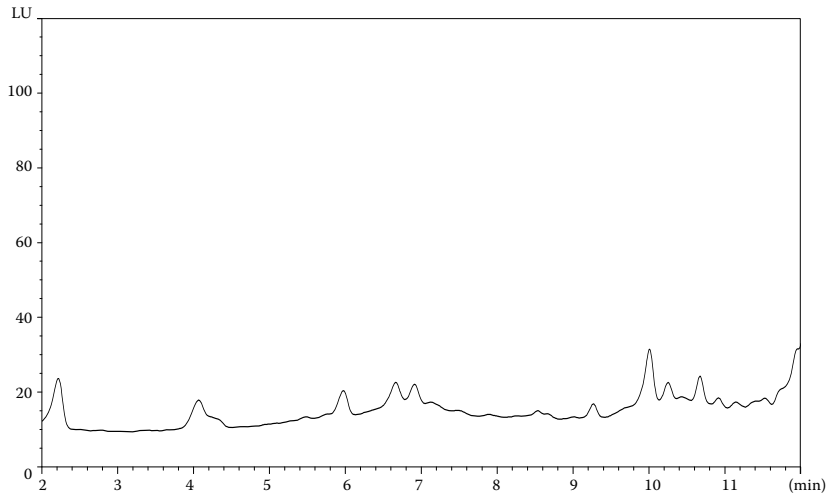

Figure 4. Example of HPLC/FLD analysis of blank oil sample

Table 3. Bisphenols in selected products from Czech retail market: survey 2001–2006 (levels in µg/kg)

Period	Analyte	Sardines in oil	Mackerel in oil	Tuna fish	Cod liver	Luncheon meat	Pate (pork)
		median (min/max)					
2001	*n*	1	1	3	2	3	4
	A	< 10	37.4	9.2 (< 3/10.9)	< 3 (< 3/< 3)	< 3 (< 3/< 3)	24.4 (< 3/70.7)
	B	32.3	7.8	< 3 (< 3/< 3)	<3 (< 3/< 3)	10.5 (< 3/12.1)	< 3 (< 3/17.8)
	C	48.4	20.5	7.6 (< 3/19.6)	41.4 (39.1/43.6)	10.4 (<3/58.0)	32.4 (< 3/95.4)
	D	24.4	30.4	15.6 (< 3/24.5)	11.7 (< 3/23.4)	< 3 (< 3/< 3)	42.7 (< 3/79.5)
2002	*n*	2	3	3	2	1	3
	A	16.6 (< 3/33.1)	< 3 (< 3/34.5)	8.2 (< 3/138.4)	69.5 (45.0/94.0)	18.5	< 3 (< 3/25.8)
	B	166.6 (< 3/332.2)	37.4 (< 3/4872.2)	43.2 (10.6/221.1)	342.0 (319.0/364.9)	384.3	106.9 (50.6/700.5)
	C	58.1 (< 3/116.2)	50.8 (< 3/277.9)	36.6 (26.3/38.5)	119.9 (105.4/134.4)	11.2	89.8 (74.7/110.8)
	D	< 3 (< 3/< 3)	< 3 (< 3/< 3)	< 3 (< 3/< 3)	< 3 (< 3/< 3)	< 3	< 3 (< 3/11.3)
2003	*n*	3	3	2	1	2	4
	A	31.9 (< 3/73.5)	< 3 (< 3/24.5)	87.6 (65.3/109.9)	< 3	30.9 (10.7/51.1)	< 3 (< 3/7.2)
	B	280.9 (256.0/388.0)	312.3 (240.5/513.8)	74.7 (65.5/83.9)	135.6	101.0 (83.6/118.4)	158.7 (40.1/287.0)
	C	81.6 (72.9/101.2)	72.7 (< 3/101.0)	35.4 (8.9/61.9)	< 3	24.8 (< 3/49.6)	17.7 (< 3/461.9)
	D	21.8 (< 3/114.1)	27.2 (< 3/78.1)	11.3 (< 3/22.6)	< 3	15.3 (6.4/24.1)	55.7 (31.4/500.7)
2004	*n*	5	1	1	1	5	x
	A	100.8 (10.1/203.0)	102.3	39	28.8	10.8 (< 3/19.0)	x
	B	318.0 (34.9/787.9)	440.7	107.4	389.4	155.1 (84.7/407.3)	x
	C	61.3 (< 10/99.9)	33.4	58.6	161.1	68.6 (< 3/146.9)	x
	D	29.4 (< 3/80.0)	30.6	15	47.2	< 3 (< 3/< 3)	x
2005	*n*	6	1	1	x	x	4
	A	77.4 (< 10/151.0)	40.3	63.5	x	x	< 10 (< 10/< 10)
	B	267.5 (< 10/420.7)	165.6	44.7	x	x	28.4 (17.6/101.7)
	C	45.0 (< 10/150.8)	8.0	16.0	x	x	7.0 (< 10/20.6)
	D	< 3 (< 3/15.5)	31.0	<3	x	x	< 3 (< 3/< 3)
2006	*n*	4	1	3	1	x	1
	A	124.5 (69.7/200.9)	100.8	35.7 (15.9/66.6)	93.3	x	20.4
	B	197.5 (108.9/227.8)	285.9	159.9 (113.2/288.9)	245.1	x	100.1
	C	94.2 (53.6/124.0)	98.9	45.9 (18.8/54.5)	170.7	x	97.3
	D	< 3	< 3	< 3 (< 3/48.9)	< 3	x	< 3

n – number of analysed products of the same sort; A – BPA; B – ∑ BADGE [H_2O]; C – ∑ BADGE [HCl]; D – ∑ BFDGE; x – an analyte/sample combination was not analysed in this period – Sums are in accordance with Commission Regulation (EC) No 1895/2005: ∑ BADGE [H_2O]: in 2001 = BADGE; in 2002–2006 = BADGE + BADGE·H_2O + BADGE·2H_2O; ∑ BADGE [HCl]: in 2001 = BADGE·HCl + BADGE·HCl·H_2O; in 2002–2006 = BADGE·HCl + BADGE·2HCl + BADGE·HCl·H_2O; ∑ BFDGE: in 2001–2002: BFDGE; in 2003–2006 = BFDGE + BFDGE·2HCl + BFDGE·2H_2O

year 2001, only BPA, BADGE, BADGE·H_2O·HCl, BADGE·HCl, and BFDGE could be determined, in 2002 also BADGE·H_2O, BADGE·$2H_2O$, and BADGE·2HCl were added onto the list of target analytes. In 2003–2005, the full set of compounds mentioned in Experimental was measured. Typical chromatograms obtained in the real samples analysis are shown in Figures 3 and 4. The highest overall contamination did not commonly exceed hundreds of μg/kg and slightly decreased during the monitoring period. In view of the recent legislation, no significant contamination was found. With both BADGE [H_2O] and BADGE [HCl] groups the findings were fairly below SMLs 9 mg/kg and 1 mg/kg, respectively (see Introduction). The occurrence of BFDGE compounds was very rare. While up to the end of 90s several hundreds, exceptionally thousands, of μg/kg of bisphenols were found many times, recently hundreds of μg/kg appeared to be the maximum.

CONCLUSIONS

The validated analytical procedure based on dichloromethane extraction, GPC clean-up, and HPLC/FLD determination is suitable for the routine analysis of migrating bisphenols in a wide spectrum of food matrices. The examination of a large range of real samples showed a satisfactory situation in the overall contamination of canned food as regards the recent legislative requirements. In spite of this fact, regular monitoring of bisphenols in the food supply should be carried out.

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