

Revision of analytical strategies to evaluate different migrants from food packaging materials

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In order to carry out a compilation of the available information on analytical methods for determination of some model migrants in selected foodstuffs, an extensive bibliographic review was done.

Model migrants of different chemical structures, polarities, lipophilicity and molecular weight have been selected: styrene, bisphenol A, 1-octene, limonene, di-isopropyl naphthalene (DIPN), lauro lactam, triacetin, tri-*n*-butylacetyl citrate (ATBC), butylated hydroxytoluene (BHT) and Triclosan.

Suggestions regarding the analytical procedure more convenient to determine the 10 model migrants studied in this review are given.

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Introduction

Lately, food packaging has gained a widespread importance in food safety due to the possibility of migration of chemicals from food contact materials. The term 'migration' usually describes a diffusion process, which may be strongly influenced by an interaction of the packaging material with the food (Arvanitoyannis & Bosnea, 2004).

Although much work has been undertaken to help to reduce lab testing required through simplifying analytical procedures or use of mathematical modeling, within different EU projects during last years, further work was proposed before finally agreed models could be put forward for its adoption (Gilbert & Rossi, 2000).

The current approach for the authorization and control of substances used in food contact materials is cautious in estimating the potential exposure of the consumer to these substances. Approaches, which take better account of the actual exposure of the consumer to food contact materials in risk assessment, are under discussion.

To estimate dietary exposure to a substance migrating from a food packaging material, information is needed on the types of food packaged, the nature of the packaging material, migration data, packaging usage factors and food consumption (ILSI Europe, 2001).

Migration testing using food simulants is the normal procedure for checking compliance of a food packaging material against specific migration limits (SMLs). However, as Feigenbaum *et al.* (2002) point out, this is not practical for several reasons. Industries that put packaged foods or materials intended for food contact on the market do not know the identities of the potential migrants; sometimes, those who manufacture and sell these raw materials do not know the processing conditions or the final application; and even if the identities of the migrants were known it may be difficult to analyze them.

Therefore, the use of mathematical modeling to predict migration, which can reduce the amount of tests to be undertaken, has been recently introduced into legislation. Practical examples for the application of this new concept are described in the Practical Guide (European Commission, 2003a).

Within this framework, the project FOODMIGROSURE (QLRT-2001–2390) (Modeling migration from plastic materials into foodstuffs as a novel and cost efficient tool for estimation of consumer exposure from food contact

materials) aims to find tools that allow evaluate consumer exposure to these chemicals. The above mentioned project carried out the selection of these model migrants being based on several approaches: the measurement of the substance on foodstuffs should be possible at low detection limits; migrants should be stable in foods; should cover a range of molecular weights and chemical groups and should be relevant to food contact materials, including environmental aspects.

Once model migrants of different chemical structures, polarities, lipophilicity and molecular weight have been selected (styrene, bisphenol A, 1-octene, limonene, diisopropyl naphthalene (DIPN), lauro lactam, triacetin, tri-*n*-butylacetyl citrate (ATBC), butylated hydroxytoluene (BHT) and triclosan, a bibliographic review for the determination of these substances in different matrices was made.

It is essential to have suitable analytical methods to determine these substances in different foodstuffs at low concentrations. However, to avoid any double development it is necessary to study, the state of the art of analytical methods used to evaluate migrants from packaging materials, previous to validate new methods.

In line with this, the main goal of this review was to compile the current available methods and to define the guidelines for the suitable routine analysis of 10 model migrants, target within the FOODMIGROSURE project.

Chemical and physical information of model migrants

Styrene, also known as styrol, vinylbenzene, etc. is a monomer used in the manufacture of numerous types of plastics. Styrene monomer can be in direct contact with food when polymers such as polystyrene and acrylonitrile–butadiene–styrene are used as packaging materials (Silva, De Carvalho, & Cardeal, 2000). It is a low molecular weight substance and a volatile compound.

Bisphenol A (other names are: diphenylolpropane, Ipopnox 198, Parabis, etc.) is a monomer used to produce polycarbonate plastics (Mountfort, Kelly, Jickells, & Castle, 1997). It is also used as a raw material in the synthesis of bisphenol A diglycidyl ether (BADGE) (Simal Gandara, Paz Abuín, Paseiro Losada, & Simal Lozano, 1993) and as an antioxidant or inhibitor in the manufacture and processing of polyvinyl chloride (PVC) (López-Cervantes & Paseiro-Losada, 2003). To modify the polyethylene polymers' properties, 1-octene is incorporated to these materials as a co-monomer.

Limonene is very commonly found in recovered PET (polyethylene terephthalate) and arises from earlier contact with soft or fruit-flavored drinks (Franz, 2002; Nerín *et al.*, 2003). It is a volatile compound that is known as a precursor to off-flavor compounds in foods and it can be absorbed also by other packaging materials as low density polyethylene (LDPE) and polycarbonate (PC) (Van Willige, Linssen, Legger-Huysman, & Voragen, 2003). Hence, the

determination of this substance could be used to ensure that a recycled plastic material is free of contaminants from previous contents.

Diisopropyl naphthalenes (DIPN) are widely employed as solvents in the paper industry in the manufacture of carbonless copy paper and thermal paper, and also for the production of dielectric fluid and thermal oil substituting polychlorinated biphenyls. The presence of this substance in foodstuffs may possibly occur due to the migration from the paperboard packaging, made from recycled fibers originating from feedstock containing carbonless copy paper (Bocacci, Chiaccherini, & Gesumundo, 1999).

In food packaging, nylon 12 is an important polyamide, which is produced by polycondensation of lauro lactam. During its production, the monomer lauro lactam as well as lower oligomers will remain in the polyamide and may migrate into foods (Stoffers, Brandl, Linssen, & Franz, 2003).

Triacetin, also known as glycerol triacetate (GTA) is an antifungal agent used in the perfumery and pharmaceutical industries, although it is also used as a plasticizer in cellulose.

Acetyl tributyl citrate (ATBC) is the most widely used plasticizer in cling-films made of polyvinylidene chloride (also known as the trade name of 'Saran') (Castle, Gilbert, Jickells, & Gramshaw, 1988). This type of material is broadly used in microwave oven, especially in home-use applications (Badeka, Pappa, & Kontominas, 1999).

The phenolic compound, butylated hydroxytoluene (BHT), is used as a food additive, but it has been also used as a common antioxidant in rubber and petroleum products and lately in plastics (Tombesi & Freije, 2002). BHT-impregnated film has shown to retard lipid oxidation of a packaged oatmeal cereal through its migration from the product via an evaporation/sorption mechanism (Lee, Shin, Han, Lee, & Giacin, 2004). It is a small molecule which exhibits high mobility and therefore has a tendency to migrate rapidly from packaging materials into foods (Wessling, Nielsen, Leufvén, & Jägerstad, 1998).

Triclosan has been widely used recently as a broad spectrum antimicrobial and antifungal agent. As an additive in plastics it prevents the growth of microorganisms, avoiding the formation of stains and odors and, therefore, allows extending the polymer's shelf life. For this reason, its inclusion on food contact polymeric materials has been considered (Sanches-Silva, Sendón-García, López-Hernández, & Paseiro-Losada, 2005).

These substances cover a wide range of different technical applications in the plastic materials as monomers (styrene, bisphenol A, 1-octene, lauro lactam) and additives (triacetin, ATBC, BHT). All of them, except DIPN, are included in the Synoptic Document (European Commission, 2003b). For this reason, they could be used in the

manufacture of plastics intended to come into contact with foodstuffs. Some of them have a specific migration limit (SML) like lauro lactam, 1-octene, BHT and bisphenol A, while those which have not been given a SML, are: substances with an ADI (acceptable daily intake) or TDI (tolerable daily intake) established, like triacetin; substances without ADI or TDI but their use could be accepted like triclosan; monomers that could be used but if its

residue it is reduced as much as possible (styrene) and substances for which enough data are not available (limonene).

Moreover, they have different chemical structures (Table 1), polarities, and also their molecular weights are very different, from 104 uma (styrene) to 402 uma (ATBC). Their solubility varies between triacetin (very soluble in water) and DIPN (almost insoluble in water); some of them are considered volatile compounds (1-octene, styrene) while others have very high boiling points. All the physico-chemical properties considered significant, since could help to approach an analytical method for their determination, are summarized in Table 2. All data were collected from different databases (Budavari, 2001; EPI Suite; Lide, 1997; Properties of Organic Compounds, 1996), and those data that were not available from the literature were estimated using the Epiwin software ver. 3.11.

Sample preparation approaches

Migrants determination in polymeric materials

In general, the first stage in the determination of the migrants in polymers is their separation from the matrix.

When the migrant is a volatile compound, such as styrene, the headspace GC technique is very suitable for the analysis and it has been used in the EU project Specific Migration, by heating the polystyrene dissolved in dimethylacetamide at 90 °C for 120 min. This methodology is also used by Simal-Gandara, Sarria-Vidal, Koorevaar *et al.* (2000) in the analysis of 1-octene, and in the analysis of limonene in LDPE, PC and PET absorbed from orange juice (Nerín *et al.*, 2003; Van Willige, Linssen, Meinders, van der Stege, & Voragen, 2002; Van Willige *et al.*, 2003).

Otherwise, if the migrant is not a volatile compound it is necessary to use a liquid extraction step. It is widely accepted that the solvent used should both dissolve the target compound well and also swell or dissolve the polymer matrix. Polymer swelling data are readily available in the literature, but the solubilities of the selected substances in plausible extraction solvents are only incompletely documented and, pending determination, they must be estimated on the basis of the nature of analyte and the extracting solvent. Several combinations solvent/analyte/polymer has been used. When the polymer is dissolved in a solvent, the polymer is then usually reprecipitated by addition of a solvent in which the polymer is insoluble.

Discontinuous extraction. Van Willige *et al.* (2003) utilized hexane for the extraction of limonene, and Nerín *et al.* (2003) dichloromethane or ethanol shaking the sample for 50 h; a liquid extraction with toluene from PP or a mixture of toluene and *m*-cresol from other packaging materials is also used by Caner (2004) for the analysis of

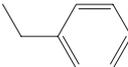
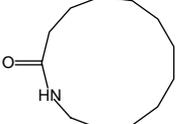
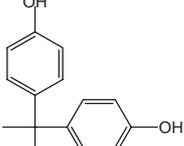
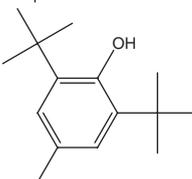
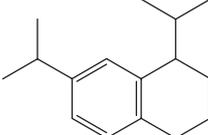
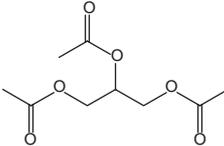
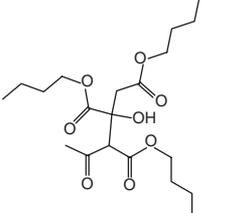
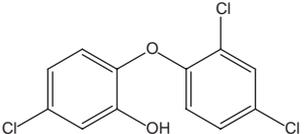
Table 1. Chemical structures	
Migrant	Chemical structure
Styrene	
Lauro lactam	
Limonene	
1-octene	
Bisphenol A	
BHT	
Diisopropylnaphthlene	
Triacetin	
Tri- <i>n</i> -butylacetyl citrate	
Triclosan	

Table 2. Chemical and physical information

Migrant	Ref. no.	CAS no.	Formula	MW	Mp (°C)	Bp/Fp (°C)	Density (g/cm ³)	Log Kow	Solubility	Spectral data
Styrene (ethenylbenzene)	24610	100-42-5	C ₈ H ₈	104	−31	145–146/32	0.9	2.95	Slightly soluble in water; soluble in alcohol, ether, acetone, and carbon disulfide; water solubility: 310 mg/L	MS peaks: 104(100) 103(41) 78(32) 51(28) 77(23) 105(12) 50(12) 52(11) UV max: 289 nm (cyhex)
Lauro lactam (aza-2-cyclotridecanone SML=5 mg/kg)	19490	947-04-6	C ₁₂ H ₂₃ NO	197	149–153	–	n.a.	2.92	Water solubility: 290 mg/L	MS peaks: 30(100) 55(98) 41(96) 100(64) 98(64) 73(53) 86(51) 43(46) 72(45) 44(44)
Limonene (1-methyl-4-(1-methylethenyl)cyclohexene)	63970	138-86-3	C ₁₀ H ₁₆	136	−95	178/46	0.84	4.57	Insoluble in water; soluble in carbon tetrachloride; miscible in ethanol and diethyl ether; water solubility: 7.6 mg/L	MS peaks: 68(100) 67(64) 93(60) 39(58) 41(46) 79(41) 43(41) 27(40)
1-Octene (SML=15 mg/kg)	22660	111-66-0	C ₈ H ₁₆	112	−101	121/–	0.71	4.57	Insoluble in water; slightly soluble in carbon tetrachloride; soluble in diethyl ether, acetone, benzene; very soluble in organic solvents; miscible in ethanol; water solubility: 4.1 mg/L	MS peaks: 43(100) 41(82) 55(80) 56(67) 70(54) 29(44)
Bisphenol A (2,2'-bis(4-hydroxyphenyl) propane SML=3 mg/kg)	13480	80-05-7	C ₁₅ H ₁₆ O ₂	228	153	250/–	n.a.	3.32	Insoluble in water; soluble in acetic acid; very soluble in ethanol, diethyl ether; benzene, alkali; water solubility: 120 mg/L	UV max: 177 nm (hp) MS peaks: 213(100) 228(26) 119(25)

Table 2 (continued)										
Migrant	Ref. no.	CAS no.	Formula	MW	Mp (°C)	Bp/Fp (°C)	Density (g/cm ³)	Log Kow	Solubility	Spectral data
BHT (2,6-di-tert-butyl- <i>p</i> -cresol)	46640	128-37-0	C ₁₅ H ₂₄ O	220	71	265/	0.89	5.10	Insoluble in water, alkali; soluble in ethanol, acetone, benzene, petroleum ether; water solubility: 0.6 mg/L	UV max: 279, 227 nm (MeOH) MS peaks: 205(100), 220(27), 57(27)
Diisopropyl naphthalene bis (1-methylhexyl) naphthalene (DIPN)		38640-62-9	C ₁₆ H ₂₀	212	n.a.	n.a.		6.08 ^a	Water solubility: 0.11 mg/L	UV max: 283, 277, 227 nm (Iso) n.a.
Triacetin (Glycerol triacetate (GTA))	57760	102-76-1	C ₉ H ₁₄ O ₆	218	−78	259/138	1.16	0.25	Slightly soluble in water, ligroin; very soluble in acetone; miscible in ethanol, diethyl ether, benzene, chloroform; water solubility: 58000 mg/L	MS peaks: 43(100), 103(44), 145(34)
Tri- <i>n</i> -butylacetyl citrate (1,2,3-Propanetricarboxylic acid, 2-(acetyloxy)-, tributyl ester (ATBC))	93760	77-90-7	C ₂₀ H ₃₄ O ₈	402	> 330	170/−	1.05	4.29 ^a	Water solubility: 5 mg/L	MS peaks: 185(100), 129(57), 259(54), 43(54)
Triclosan (2,4,4'-Trichloro-2'-hydroxydiphenyl ether)	93930	3380-34-5	C ₁₂ H ₇ Cl ₃ O ₂	289	56–58	−/223	1.58	4.76	Soluble in acetone and isopropanol; water solubility: 17 mg/L	n.a.
n.a., not available. ^a Estimated with epiwin ver. 3.11.										

limonene. Avison, Gray, Davison, and Taylor (2001) extracted limonene using methanol from a polymer infused with this substance.

In the extraction of ATBC from sealing resins, hexane is the chosen solvent by Hirayama, Tanaka, Kawana, Tani, and Nakazawa (2001).

Acetonitrile (Monteiro, Nerín, Rubio, & Reyes, 1998; Vargo & Olson, 1985) was selected for the extraction of BHT. Instead of this, Wessling *et al.* (1998) used a double extraction with heptane and then a concentration step.

Usually, extraction procedures are carried out with hand shaking. Nevertheless, sometimes ultrasonics (Marque, Feigenbaum, Dainelli, & Riquet, 1998) or maceration (Monteiro *et al.*, 1998) were employed to improve the process.

Continuous extraction. Gramshaw and Vandenburg (1995) used dynamic headspace to extract styrene from thermoset polyester passing through the U-tube nitrogen at 25 ml min⁻¹ and heating the oven at 200 °C. To analyze styrene dimers and trimers Soxhlet extraction with dichloromethane was used followed by size exclusion chromatography to clean-up the dichloromethane extracts. The Soxhlet extraction is also used by Summerfield and Cooper (2001) to determine DIPN from paper and board employing dichloromethane and ethanol as solvents, and Guerra, Marín, Sánchez, and Jiménez (2002) extracted ATBC from PVC refluxing cyclohexane during 5 h. To extract BHT incorporated in a HDPE film Lee *et al.* (2004) utilized 100 ml of acetonitrile and 24 h of Soxhlet extraction.

Extraction with solvents that swell and/or dissolve both migrant and polymer

Snyder and Breder (1985) determined residual styrene monomer from polystyrene by dissolving a portion of the material in tetrahydrofuran (THF) followed by SEC prior the analysis by RP-HPLC.

For the analysis of Bisphenol A in baby bottles Biles, McNeal, Begley, and Hollifield (1997) dissolved the polymer with methylene chloride in an ultrasonic bath and then the polymer was precipitated by adding methanol. Similar procedure was used by Mountfort *et al.* (1997) but employing dichloromethane to dissolve and propan-2-ol to precipitate the polymer, afterwards hexane was added and supernatant was taken to dryness and dissolved in the HPLC mobile phase. Residual bisphenol A in polycarbonates was determined by Howe and Borodinsky (1998) dissolving the polymer in chloroform and extracting the solution with 0.01 M sodium hydroxide.

Triantafyllou, Akrida-Demertzi, and Demertzis (2002) extracted limonene from recycled PET with a mixture of 1,1,1,3,3,3-hexafluoro-2-propanol (HFIP) and 2-propanol.

For the determination of DIPN from paper and board Summerfield and Cooper (2001) immersed those materials

in absolute ethanol in an ultrasonic bath when the analysis method chosen was HPLC.

Supercritical fluid extraction (SFE). Nerín *et al.* (2003) analyzed limonene using CO₂ modified with toluene as SFE fluid. ATBC was extracted also by this system (Cano, Marín, Sánchez, & Hernandis, 2002; Guerra *et al.*, 2002).

Microwave-assisted extraction (MAE). Cano *et al.* (2002) extracted ATBC by a microwave sample preparation system from an ethanol dissolution of the sample.

Migrants determination in food simulants

Official aqueous simulants

Varne and Breder (1981) analyzed styrene in distilled water by static headspace GC. But usually, the aqueous simulants are directly injected when analysis is carried out with RP-HPLC technique. Extracts of bisphenol A in water and different concentrations of ethanol in water (Biles *et al.*, 1997) and in acetic acid (Lopez-Cervantes & Paseiro-Losada, 2003) were directly injected for its analysis.

Nevertheless, sometimes a concentration step or a change of solvent step is needed prior to chromatographic analysis once the solvent must be compatible with the chromatography conditions used.

For volatiles substances some specific techniques has been applied: Silva *et al.* (2000) employed SPME for the determination of styrene and Tombesi and Freije (2002) for the analysis of BHT. BHT has also been determined by solid-phase extraction using a silica C18 cartridge (Dopico-Garcia, López-Vilariño, & González-Rodríguez, 2005). Linssen, Janssens, Roozen, and Posthumus (1993) used thermal desorption extraction (DTD) for the determination of limonene.

Fatty food simulants

The official fatty food simulant is rectified olive oil, but sometimes alternatives simulants are used as synthetic mixtures of triglycerides (e.g. Myglyol) sunflower oil or corn oil. When tests with the food oils are not possible, the European legislation allows substitute tests using isooctane, 95% ethanol and modified polyphenylene oxide (MPPO, Tenax is the registered trademark).

Due to the complexity of olive oil, migration tests are time consuming and difficult to conduct and therefore to approach this problem, many methods of extracting migrants from this simulant were investigated. An exception is when the substance is a volatile compound; an example is the determination of styrene using the static headspace (Specific Migration). Some of the methodologies adopted were:

Extraction with solvents. Lopez-Cervantes and Paseiro-Losada (2003) used acetonitrile 90% (v/v) for the extraction of bisphenol A.

Continuous steam distillation. Styrene migration into olive oil was determined by Jickells, Gancedo, Nerín, Castle, and Gilbert (1993) extracting the sample with hexane, using the Linkens-Nikerson apparatus.

SEC (size exclusion chromatography). This procedure, based on the separation of the components using a column and further collection of the eluted fraction of interest, was used by *Stoffers et al.* (2003) in the migration evaluation of lauro lactam.

The methodologies applied to simulants, as the synthetic mixtures of triglycerides and sunflower oil, are very similar to those used when olive oil is the fatty food simulant selected. In the analysis of bisphenol A from Miglyol an extraction step in methanol is required prior to its analysis by HPLC (*Biles et al.*, 1997) or dissolution in chloroform (*Howe & Borodinsky*, 1998).

Solid phase extraction with a Tenax adsorption column followed by extraction with hexane has been employed to determine ATBC in these simulants by *Van Lierop and Van Veen* (1988).

In order to overcome the analytical difficulties found in extracting migrants from olive oil, solvents have been used as substitutes. Commonly, extraction of the migrant from some of these simulants (e.g. ethanol 95%, isooctane) does not present a major problem. So, more undemanding techniques may be applied such as:

Simulant evaporation to dryness followed by dissolution in an appropriate solvent. This method was applied when migration of lauro lactam (*Stoffers et al.*, 2003) and BHT (*Wessling et al.*, 1998) were evaluated. Methanol was the selected solvent in the dissolution step for the first substance (*Stoffers et al.*, 2003) and heptane for the last one (*Wessling et al.*, 1998).

Extraction with solvents. Diethyl ether is used to extract styrene from Tenax (*Specific migration*). Absolute ethanol was the selected solvent to extract DIPN (*Summerfield & Cooper*, 2001; *Triantafyllou et al.*, 2002), methanol was used to extract BHT according the IUPAC method 2.432 (*Dieffenbacher*, 1998); *Yankah, Ushio, Ohshim, and Koizumi* (1998) used acetonitrile to extract BHT.

Migrants determination in foodstuffs

Due to the complexity of food samples, most extraction procedures performed in simulants, especially in aqueous simulants, are not suitable to obtain a suitable analytical method for evaluate migration in food. Although there is a lack of bibliographic support, the few available information related with the study of migration into foodstuffs reveals that scientific community approaches the problem in two different ways.

There are approaches that try to apply the most undemanding technique. This, commonly coincides with the one used with simulants, usually fat simulants. These methodologies include:

Extraction with solvents

Discontinuous. This procedure is one of the most widely applied when migration on food is evaluated, especially nonfatty food, although has also been applied to fatty foods.

ATBC from non fatty foods (such as jelly and candy) was extracted by *Lau and Wong* (1996) with cyclohexane–dichloromethane by shaking the sample during 2 h and finally dried with sodium sulphate. *Summerfield and Cooper* (2001) determined DIPN in fatty food (pastry and cake) by extraction with acetonitrile. Nevertheless, dry food was extracted by the same authors with absolute ethanol in an ultrasonics bath. *Yankah et al.* (1998) also determined BHT on smoke flavored sausage by extraction with acetonitrile. Good recoveries were achieved. *Munguia-Lopez and Soto-Valdez* (2001) extracted bisphenol A from the liquid portion of tuna and jalapeno peppers also with acetonitrile. *Jordan, Margaría, Shaw, and Goodner* (2002) utilized methylene chloride to extract limonene from kiwi fruit puree and commercial kiwi essence. *Sanches-Silva et al.* (2005) extracted triclosan from orange juice, chicken meat and cheese using hexane, this organic phase was evaporated in a rotary evaporator and then the residue extracted with acetonitrile.

Nevertheless, a detail that must be had in account when food is the study matrix, and it is the importance of the homogenization step as well as phases separation and filtration steps. These are critical in chromatographic analysis and establish the column and equipment life as well as the suitability for routinely use of the method.

Continuous. To extract styrene from cooked pork meat *Gramshaw and Vandenburg* (1995) used a Likens-Nickerson extractor with pentane for 1 h. The pentane extracts were then evaporated using a Kuderna-Danish evaporator prior the analysis. Otherwise, *Van Lierop and Van Veen* (1988) determined ATBC migration in fatty food extracting samples with diethyl ether in a Soxhlet apparatus for 6–7 h. Soxhlet apparatus was also used by *Badeka et al.* (1999) to extract this compound from cooked meat of different fat contents but followed by a saponification with KOH, which decomposes the ATBC in citric acid and L-butanol, and this last one is quantified by GC.

In the other hand, there are papers which describe newer extraction methodologies to overcome this difficulty. Although being, in general, time consuming and laborious approaches. Some of these alternatives approaches are:

Solid phase extraction (SPE). The use of a column which allows the separation of the substances of interest it is helpful when working samples have a complex composition.

Mountfort et al. (1997) analyzed an infant feed regarding bisphenol A content. After mixing some ammonia drops and ethanol to the sample, hexane was added. The lower layer was passed through an SPE cartridge and bisphenol A was eluted with water and methanol. *Yoshida, Horie, Hoshino, and Nakazawa* (2001) homogenized the solid portion in cans of vegetables and fruits with acetonitrile and sodium sulphate (avoids the formation of an emulsion) followed by an extraction with hexane. The acetonitrile layer was evaporated to dryness and dissolved with acetone–heptane

(2.5:97.5). This solution was applied to a Sep-Pak Florisil cartridge and bisphenol A eluted with acetone/heptane (20:80). Good recoveries and relative standard deviation were achieved. Otherwise, the aqueous portions of those food samples were applied to an OASIS HLD extraction cartridge. Bisphenol A was eluted with methanol. The eluate was evaporated to dryness and applied to a Sep-Pak Florisil cartridge. Kang and Kondo (2002) also determined bisphenol A but in a coffee sample. After treatment, sample was passed through an ISOLUTE multimode cartridge. Bisphenol A was eluted with acetonitrile/water (40:60, v/v). Limonene was determined in water (Linssen *et al.*, 1993) and cheese (Valero, Sanz, & Martínez-Castro, 2001) purging the sample with purified nitrogen using a Tenax TA to trap the substance of interest. For the determination of ATBC in fatty food Van Lierop and Van Veen (1988) extracted a mixture of the fatty food and dry sodium sulphate with diethyl ether in a Soxhlet apparatus for 6–7 h. After evaporation of ether, the solution was passed through a Tenax absorption column, heated at 210 °C in an oil-bath.

Size exclusion chromatography (SEC). Castle *et al.* (1988) determined ATBC (acetyl tributyl citrate) in food (cheese, fruit, vegetables, soups, cakes, puddings and meal dishes) using this procedure. Prior to SEC clean-up, homogenized food was bended with acetone/hexane (1:1). Extraction was repeated and then, once the extracts were combined, were evaporated to dryness and the residue re-dissolved in dichloromethane–cyclohexane (1:1). The column used was a Biobeads S-X3, the mobile phase was dichloromethane–cyclohexane (1:1) flowing at 3.0 ml/min.

Gel permeation chromatography (GPC). ATBC content on fatty food (bacon and cheese) was determined by Lau and Wong (1996) applying this method. First, sample was homogenized with cyclohexane–dichloromethane (1:1) for 2 h in an automatic shaker. The extract (5 ml) was dried with anhydrous sodium sulphate, evaporated to dryness and re-dissolved with the same solution. Then, a gel-permeation chromatography was made to clean-up. First 90 ml of the eluent were discarded and the following 40 ml collected and evaporated to 2 ml by heating.

Headspace (static and dynamic). When the migrant is a volatile substance this is an effective analytical procedure. This technique has been applied to the determination of styrene in coffee and tea by Varner and Breder (1981), in sliced potatoes with grated cheese and minced beef fried with tomato sauce by Jickells *et al.* (1993). Otherwise Gramshaw and Vandenburg (1995) could not analyze this compound in meat by dynamic headspace due to the water present in the meat condensing in the trap. The dynamic procedure was applied for the determination of 1-octene from pure beef fat (Umano & Shibamoto, 1987) and menhaden fish oil (Horiuchi, Umano, & Shibamoto, 1998) and for the

determination of limonene from cheese (Péres, Denoyer, Tournayre, & Berdagué, 2002; Valero *et al.*, 2001).

Solid phase microextraction (SPME). This technique is very useful when the substance is volatile. Silva *et al.* (2000) determined styrene in drinking water using a fiber with 85 µm polyacrylate coating; Andrés, Cava, and Ruiz (2002) determined 1-octene in the ham by this technique using a 75 µm carboxen-poly(dimethylsiloxane) coating fiber. Péres, Viallon, and Berdagué (2001) determined limonene using also this procedure attempting four different fibers: 100 µm dimethylsiloxane (DVB); 85 µm polyacrylate (PA); 65 µm PDMS/divinylbenzene (PDMS/DVB); 75 µm Carboxen/DVB.

Review of chromatographic methods for migrants determination

A large variety of chromatographic methods were found in the literature. The following revision was sorted by chromatographic technique and by detector.

High performance liquid chromatography (reversed phase)

Fluorescence detector

Bisphenol A was analyzed employing a C18 column with acetonitrile/water as mobile phase. Fluorescence conditions of λ_{ex} 275 nm and λ_{em} 300 nm were employed (Kang & Kondo 2002; Simal-Gandara, Sarria-Vidal, & Rijk, 2000; Simal Gandara *et al.*, 1993). Other authors used similar chromatographic conditions but changing Fluorescence the wavelengths, λ_{ex} 225 nm and λ_{em} 310 nm (Munguia-Lopez & Soto-Valdez, 2001) or λ_{ex} 225 nm and λ_{em} 305 nm (Lopez-Cervantes & Paserio-Losada, 2003). Different detection conditions (λ_{ex} 235 nm and λ_{em} 317 nm) were used for the analysis of this compound by Howe and Borodinsky (1998) and heating the chromatographic column. Similar wavelengths were used by Biles *et al.* (1997) but with a mobile phase of methanol/water/acetonitrile. To carry out the analysis, Mountfort *et al.* (1997) employed a C8 column with methanol/water as mobile phase with similar fluorescence conditions (λ_{ex} 285 nm and λ_{em} 300 nm).

Summerfield and Cooper (2001) analyzed DIPN using an ODS column with a mobile phase of water/acetonitrile and λ_{ex} : 232 nm and λ_{em} : 338 nm.

Yankah *et al.* (1998) determined BHT extracted from lipids and smoked-flavored sausages using a RP-18 column and mixture of water/acetic acid/acetonitrile as mobile phase working with an λ_{ex} 280 nm and λ_{em} 310 nm followed by a GC/MS confirmation.

UV detector

Tawfik and Huyghebaert (1998) determined styrene using a C18 column and employing a mobile phase of acetonitrile/water and setting the detector to 245 nm. Bisphenol A was analyzed utilizing a C18 column with acetonitrile/water as mobile phase, setting the detector to 228 nm (Yoshida *et al.*, 2001) or to 225 nm

(Lopez-Cervantes & Paseiro-Losada, 2003). Similar column was used by Howe and Borodinsky (1998) but with a different mobile phase (0.1% acetic acid in water/acetonitrile). This analysis was carried out heating the column to 60–65 °C and setting the UV detector to 280 nm.

Stoffers *et al.* (2003) determined lauro lactam utilizing an ODS column and using water/methanol as mobile phase and fixing the wavelength in 207 nm.

The IUPAC method for BHT uses a Silica RP18 column, a mobile phase of water/acetic acid/acetonitrile and a wavelength of 280 nm. An alternative method comprises just a change in the mobile phase composition: methanol and potassium dihydrogen phosphate buffer 0.02 M (Dieffenbacher, 1998). Lee *et al.* (2004) used methanol and water as mobile phase setting the detector to 280 nm to analyze this compound extracted from HDPE with this antioxidant incorporated.

Sanches-Silva *et al.* (2005) analyzed triclosan using a C18 column with a gradient of acetonitrile, water, using three different wavelengths (205, 235 and 280 nm).

MS detector

Lopez-Cervantes and Paseiro-Losada (2003) confirmed the identity of bisphenol A using this detector with APCI (Atmospheric Pressure Chemical Ionization) in negative mode with identical chromatographic conditions as for the quantification by UV. To determine lauro lactam (dimer and trimer) Stoffers *et al.* (2003) also used APCI. The column was an ODS and water/methanol was the mobile phase. Moreover, this detector (in negative mode) is one of the selected by Sanches-Silva *et al.* (2005) to confirm triclosan identity extracted from food samples.

High performance liquid chromatography (normal phase)

Fluorescence detector

Mountfort *et al.* (1997) utilized for the analysis of bisphenol A, a cyano phase column with hexane/2-propanol/dichloromethane as mobile phase and with the same fluorescence conditions as for the reversed phase HPLC.

Gas chromatography

GC-FID

This technique has been used to determine styrene, 1-octene, DIPN, lauro lactam, ATBC and limonene. To determine styrene, Gramshaw and Vandenburg (1995) used a BP-1 column setting the oven to 200 °C, and injecting in a cold on-column mode, while Silva *et al.* (2000) used an HP-5 column and employing lower T^a (from 50 to 150 °C), after SPME extraction.

In the analysis of 1-octene from a liquid paraffin matrix carried out by Simal-Gandara, Sarria-Vidal, Koorevaar *et al.* (2000), a PLOT fused silica column with a film of PoraPLOT Q was used, injecting the sample in unsplit headspace mode, setting the oven from

70 to 200 °C. Otherwise, Umamo and Shibamoto (1987) used a DB-WAX column with an oven program from 40 to 200 °C.

Boccacci *et al.* (1999) determined DIPN extracted from food samples using an OVI column and with hydrogen as carrier gas (oven T^a from 40 to 200 °C) followed by confirmation by GC-MS. Triantafyllou *et al.* (2002) used a DB-1 column and the same carrier gas (oven T^a from 60 to 270 °C) followed also by confirmation by GC-MS.

Stoffers *et al.* (2003) analyzed lauro lactam using a DB-624 column and hydrogen as carrier gas (oven T^a max.: 240 °C) after be separated by SEC. Hirayama *et al.* (2001) determined ATBC extracted from sealing resins using an HP-5 column, injecting the sample in split mode and heating the oven from 150 to 250 °C. When this compound was extracted by Cano *et al.* (2002) from different samples by MAE (microwave-assisted extraction) and by SFE, it was determined using an analogous column but setting the oven T^a from 110 to 260 °C.

For the analysis of limonene, Linssen *et al.* (1993) used a thermal desorption/cold trap device for transferring the volatile compounds from Tenax to the column (DB-1). The oven program achieved to 250 °C. Triantafyllou *et al.* (2002) analyzed limonene in recycled PET using a DB-1 column with an oven temperature from 50 to 280 °C. Furthermore, this compound was analyzed by Van Willige, Linssen, and Voragen (2000) using the large volume injection (LVI), injecting 30 μ l of sample; the desolvation column was a MEGA and the analytical column was a DB-1701.

GC-MS

Gramshaw and Vandenburg (1995) investigated the presence of styrene in cooked pork meat using a BP-1 column holding the oven to 50 °C. When the sample is polystyrene, this technique is also used in the EU project Specific Migration, but employing a HP-5MS column and heating the oven from 40 to 180 °C, and with helium as carrier gas. If the sample is Tenax, the same project used different oven temperatures (from 60 to 100 °C) and a different column (DB17) (Specific Migration).

For the identification of bisphenol A this technique was chosen by Biles *et al.* (1997), using an Rtx-5 capillary column and setting the oven temperature from 100 to 280 °C. Analogous conditions were used by other authors (Lopez-Cervantes & Paseiro-Losada, 2003; Munguia-Lopez & Soto-Valdez, 2001).

In the determination of 1-octene after a SPME extraction, Andrés *et al.* (2002) used a HP-5 column for the separation with a temperature gradient from 40 to 200 °C. Instead of a HP-5, a DB-WAX column was used for the separation with a similar oven program (Horiuchi *et al.*, 1998; Umamo & Shibamoto, 1987).

A ZB-5MS column was employed to determine limonene with helium as carrier gas. The analysis was carried out with an oven program from 40 to 250 °C (Avison *et al.*, 2001; Bentivenga, D'Auria, De Luca, De Bona, & Mauriello,

Table 3. Suggestions to prepare analytical methods to determine migrants in foods

Migrant	Type of food	Sample preparation	Analysis procedure
Styrene	Nonfatty	Separation by volatilization. Extraction or dilution with polar solvents.	GC-FID or MS. Column type apolar, with medium to low phase ratio (β), static or dynamic headspace.
	Fatty	Separation by volatilization. Extraction with polar solvents not miscible with fat.	HPLC-UV or FI. Column type C18. Mobile phase: acetonitrile, Detection at. $\lambda=247$ nm by UV, $\lambda_{ex}=250$ nm, $\lambda_{em}=305$ nm by fluorescence.
Bisphenol A	Nonfatty	Extraction or dilution with polar solvents.	HPLC-UV or FI or APCI (–).
	Fatty	Extraction with polar solvents not miscible with fat.	Column type C18. Mobile phase: acetonitrile/water, Detection at. $\lambda=227$ nm by UV, $\lambda_{em}=280$ or 225 nm, $\lambda_{em}=307$ nm by fluorescence and 227 (m/z) by MS.
1-octene	Nonfatty	Separation by volatilization.	GC-FID or MS.
	Fatty	Separation by volatilization.	Column type apolar, with medium to low phase ratio (β), static or dynamic headspace.
Limonene	Nonfatty	Separation by volatilization. Extraction or dilution with polar solvents (acetonitrile, methanol or isopropanol).	GC-FID or MS.
	Fatty	Separation by volatilization. Extraction with polar solvents not miscible with fat (acetonitrile, methanol or isopropanol).	Column type polar, with medium to low phase ratio (β), static or dynamic
DIPN	Nonfatty	Extraction or dilution with polar solvents.	GC-FID or MS. Column type apolar, with medium to high phase ratio (β), on column or splitless injection mode.
	Fatty	Extraction with polar solvents not miscible with fat.	HPLC-UV or FI or APCI (+) or (–). Column type C18. Mobile phase: acetonitrile/water.
Lauro lactam	Nonfatty	Extraction with weakly or nonpolar solvents and concentration step. Solvent changes if HPLC is used. Extraction/dilution with polar solvents.	GC-FID or MS.
	Fatty	Extraction with weakly nonpolar solvents followed by SEC-GPC clean-up and concentration step. Solvent changes if HPLC is used. Extraction with polar solvents not miscible with fat.	Column type apolar, with medium to low phase ratio (β), split injection mode. HPLC-APCI (+). Column type C18. Mobile phase: acetonitrile/water, Detection at. 198 (m/z).
Triacetin	Nonfatty	Extraction with polar to nonpolar solvents.	GC-FID or MS. Column type apolar, with medium phase ratio (β), split injection mode.
	Fatty	Extraction with polar solvents not miscible with fat.	HPLC-APCI. Column type C18. Mobile phase: acetonitrile/water, Detection at. 159 (m/z).
ATBC	Nonfatty	Extraction with weakly or nonpolar solvents and concentration step. Solvent changes if HPLC is used. Extraction/dilution with polar solvents.	GC-FID or MS.
	Fatty	Extraction with weakly or nonpolar solvents followed by SEC-GPC clean-up and concentration step. Solvent changes if HPLC is used. Extraction with polar solvents not miscible with fat.	Column type apolar, with medium to low phase ratio (β), split injection mode. HPLC-APCI (+) or (–). Column type C18. Mobile phase: acetonitrile/water, Detection at. 259 (m/z) by APCI (+) or 341 (m/z) APCI (–).
BHT	Nonfatty	Extraction or dilution with polar or on polar solvents.	GC-FID or MS.
	Fatty	Extraction with polar solvents not miscible with fat.	Column type apolar, with medium phase ratio (β), split or splitless injection mode. HPLC-UV or APCI (–). Column type C18. Mobile phase: acetonitrile/water, Detection at. $\lambda=217$ nm by UV and 219 (m/z) APCI (–).
Triclosan	Nonfatty	Extraction or dilution with polar or nonpolar solvents.	HPLC-UV or APCI (–).
	Fatty	Extraction with polar solvents not miscible with fat.	Column type C18. Mobile phase: acetonitrile/water, Detection at. $\lambda=229$ nm by UV and 289 (m/z) APCI (–).

2001; Péres, 2001, 2002; Triantafyllou *et al.*, 2002). Analogous oven program was also used by other authors but carrying out the separation in a different column, a cross linked phenyl–methyl siloxane column, (Jordan *et al.*, 2002), a homemade column (FFAP/OV-1) (Valero *et al.*, 2001), and a methyl silicone or phenyl/methyl silicone column (Nerín *et al.*, 2003).

DIPN extracted from paper and board was analyzed by Summerfield and Cooper (2001) using also a HP5-MS column and employing a ramp temperature from 40 to 300 °C. Boccacci *et al.* (1999) investigated its presence in foodstuffs using a SPB-608 column with helium as carrier gas (oven T^a from 50 to 240 °C). Triantafyllou *et al.* (2002) also confirmed its presence using a HP-5MS column (oven T^a from 60 to 270 °C).

Castle *et al.* (1988), after clean-up samples by SEC, determined ATBC using a BP-5 column, by on column injection and with helium as carrier gas (oven T^a from 70 to 210 °C). Other authors also used similar column and an oven program that heated from 60 to 300 °C (Van Lierop & Van Veen, 1988) or from 100 to 300 °C (Lau & Wong, 1996). Otherwise Hirayama *et al.* (2001) used a DB-1701 column and a ramp temperature from 50 to 280 °C to confirm the identity of this substance.

Wessling *et al.* (1998) separated BHT using a 50% phenyl-505 methyl polysiloxane column and helium as carrier gas employing a ramp temperature from 100 to 290 °C. Yankah *et al.* (1998) used a Quadrex 65 HT column to confirm its identity.

After a SPME extraction, Tombesi and Freije (2002) determined BHT using a HP-5 column, with helium as carrier gas and with a ramp temperature from 40 to 250 °C.

Sanches-Silva *et al.* (2005) confirmed triclosan identity using a DB-5 column and a temperature gradient from 160 to 260 °C.

Concluding remarks

The literature on this subject is, most of the times, scant, and has placed more emphasis on results concerning migration in simulants and amount in polymers than on food analysis. Moreover, analytical methods seem to be still in the research and development stage, and no particular set of methods has attracted widespread agreement. No two studies on the use of compounds appear to have used the same procedure to determine a given compound in a given type of sample.

Some of the protocols that have been used are not described in the literature in sufficient detail to allow confident replication by other laboratories. This is a common situation in the early stage of the development of the analytical methodology.

In general, it is not at present possible to draw up reliable analytical protocols for the determination of all the selected compounds in food. For most migrant/food combinations, any protocol drawn up on the basis of the literature and general analytical principles and experience is necessarily a

tentative proposal requiring exhaustive evaluation. However, in view of the bibliographic data available some guidelines could be drawn.

Bisphenol A, BHT and triclosan can be extracted from foods with polar solvents immiscible with fat, although BHT with some difficulty. All the migrants exhibit good properties in the ultraviolet region and furthermore bisphenol A shows very good response in fluorescence. Therefore, HPLC with UV-FI detection seems an appropriate determination technique. HPLC coupled with APCI-MS detector can be also used. GC-(FID or MS) is also a common possible technique for most of them.

Triacetin is a compound soluble in water and not very soluble in oils. It can be extracted from foods with polar solvents immiscible with fat. It can be determined by GC-(FID or MS) or by HPLC only with an APCI (+)-MS detector because it does not show interesting properties in fluorescence or ultraviolet region.

Lauroctam and ATBC have low solubility in water and they present similar difficulties to the first group. They do not show interesting UV and fluorescence properties and they need to be determined by GC-(FID or MS) or by HPLC with only an APCI-MS detector.

Styrene, 1-octene and limonene can be separated from food matrix by volatilization or distillation and it seems the better way to remove them from other food components. Analysis by static or dynamic headspace-GC (FID or MS) is an excellent and common determination for the three compounds. Other ways are also possible as extraction from nonfatty foods with weakly polar solvents immiscible with water, or using polar solvents immiscible with fat to extract from fatty foods. Styrene shows a very good response by fluorescence and ultraviolet and can be determined by HPLC using those detectors.

Table 3 summarized proposals found more suitable for the separation and quantification of these substances in foodstuffs.

For all migrants and depending of each combination analyte/food matrix, more purification steps could be necessary, usually using SPE or liquid–liquid extraction, again.

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