

Introduction

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Food contact applications represent an important part of the paper and board packaging sector. CEPI's activity in this field aims at ensuring that this sector can continue to supply its products in a market that is increasingly controlled by regulation, food scares and customer demands. CEPI's view is to protect businesses, while increasing consumer safety.

In the EU, food contact materials and articles shall comply with the framework Regulation 1935/04. More specifically, the Regulation's main principle is that they should not "transfer their constituents to food in quantities which could endanger human health or bring about an unacceptable change in the composition or a deterioration of the organoleptic characteristics of the foods".

Therefore, the focus is on the transfer of constituents, verified via the so-called "migration tests". These tests imply the use of food simulants that mimic the behaviour of a certain food (or a class of foods) with respect to its capability to absorb the substances that may be transferred (may migrate) from the packaging.

In many cases, the real behaviour of a material/article may be over- or under-estimated by carrying out tests with the use of simulants. Consequently, Correction Factors should be used so that the migration test results are aligned with the real situation and to ensure that the safety assessment is precise.

In other words, Correction Factors aim at relating migration levels into food with migration levels obtained with the food simulants or extraction solvents used in simplified and standardised test procedures. The use of Correction Factors is envisaged in the EU legislation.

CEPI, with DG Sanco's guidance, funded a research work on Correction Factors carried out by Laurence Castle, a well-known and estimated scientist, active in the field of food contact materials since many years. Laurence works at FERA, the UK Food and Environment Research Agency.

The report includes a review of published research works along with any unpublished reports on the topic of Correction Factors provided for this review by CEPI. The aim was to relate chemical migration levels from paper and board into foods, with the migration or extraction values obtained using food simulants or solvents, as well as to propose a set of Correction Factors, where justified. The CEPI Food Contact Group has given its support to this work by providing comments in the report's review phase.

The set of information contained in this work will be one of the tools to be used in the discussion with EU authorities on a specific, EU-harmonised regulation on paper and board for food contact that CEPI is currently advocating for.

Systematic derivation of Correction Factors (CFs) to relate chemical migration levels from paper and board into foods, with the migration or extraction values obtained using food simulants or solvents

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SUMMARY

The concept of Correction Factors (CFs) was introduced during the Biosafepaper project. Correction Factors aim to relate migration levels into food with migration levels obtained with the food simulants or extraction solvents used in rapid, simplified and standardised test procedures such as those described in CEN standards. Correction Factors are directly analogous to the reduction factors described in EU legislation on plastics for food contact.

This work has reviewed published work along with any unpublished reports on the topic of Correction Factors provided for this review by CEPI. The aim was to relate chemical migration levels from paper and board into foods, with the migration or extraction values obtained using food simulants or solvents. The work has proposed a set of Correction Factors where justified and has identified any data gaps and the further work that would be needed to fill such gaps.

The review and the report is organised in chapters according to the different categories of food contact that P/B is used for; (i) Fruits and vegetables (fresh, whole); (ii) Frozen foods; (iii) Fatty contact; (iv) Dry foods (long-term ambient storage); (v) Baking papers; (vi) Hot and cold water including filters; and lastly, (vi) the special case of Inorganic Substances.

Fruits And Vegetables (Fresh, Whole)

- Using Tenax under test conditions of 10 days at 40°C and a Correction Factor of 10 (CF=10) would provide a good estimate for the migration into the whole fresh products.
- This would be conservative considering that depending on cooking and eating habits, concentrations may be reduced by processing operations such as washing, peeling and/or cooking.
- The data set is limited for this food category but given the limited migration seen, further is not recommended as a priority.

Frozen Foods

- Using Tenax under test conditions of 10 days at 5°C and no Correction Factor (CF=1) would be appropriate both for chilled and frozen storage of foods.
- This is because:- a) the standard test conditions of 10 days at 5°C are directly comparable to the storage life and temperature of most chilled foods; b) the test temperature of 5°C is higher than the storage temperature of frozen foods (-20°C) and this compensates for the fact that the simulant is used for only 10 days whereas the food itself may be stored frozen for up to one year.
- The data set is limited and dominated by migration studies of small, non-polar molecules with a relatively high vapour pressure. If it could be assured that such volatiles were not present in P/B food packaging materials, a Correction Factor of 10 or more would be appropriate.

Fatty Contact

- Tenax cannot be considered a good simulant for P/B intended for contact with fatty foods. Isooctane or ethanol should be used instead, although ethanol may not be suitable for P/B treated with fluorochemicals.
- For solid foods that have fatty substances on the surface (as in the meaning of EU 10/2011) the Factor as specified in that directive should be used to allow for the less complete contact the food makes compared with the (liquid) organic solvents (CF=DRF).

- For specific applications where the fraction of the food that makes fatty contact with the P/B may be grossly underestimated by the fractional Correction Factors in 10/2011 (e.g. take-away pizza) a higher Correction Factor could be argued case-by-case.
- The data set is very limited. More test results and a further consideration on the definition and characteristics of fatty foods is needed.

Dry Foods (Long-Term Ambient Storage)

- The use of Tenax with no Correction Factor (CF=1) is convenient. But care is needed to ensure Tenax does mimic the foodstuff for the migrant(s) of interest.
- Special care is needed if accelerating the tests by raising the temperature above the intended conditions of use.
- The extensive transfer that can occur both into food and into Tenax means that a compositional analysis and the assumption of total mass transfer may be more convenient and justified.

Baking Papers

- Baking papers should be tested using the CEN test for fatty contact (ethanol and/or isooctane extraction) with no Correction Factor (CF=1). Ethanol is not likely to be appropriate if the baking paper is treated with fluorochemicals.
- The paper should also be tested at high temperatures, using Tenax, as a check against the formation and subsequent migration of thermal decomposition products.
- This would be conservative considering that, depending on cooking conditions, concentrations in the food may be reduced by volatilisation.

Hot And Cold Water Including Filters

- EN 645:1993 and EN 647:1993 should be used to test P/B intended for contact with cold- or hot-liquids respectively.
- The test results for the EN method should be expressed on an area-related basis (not on the mass of paper extracted) and the results calculated for the area: mass ratio of the intended application. No Correction Factor should be used (CF=1).

Inorganic Substances

- For P/B used for food packaging (excluding e.g. filtration and liquids or solid foods with an external aqueous liquid phase, unless the P/B is coated) it does not seem necessary to test for the migration of inorganics.
- This conclusion does not apply to any inorganic substances that may be used in nano form.
- It is recommended to evaluate whether or not an acidic extraction medium should be added as a CEN standard.

All food classes and overall conclusion and recommendation

- The full utility of Correction Factor (in their fullest sense) will not be realised unless the control of P/B moves away from simplistic migration limit values and uses instead, exposure concepts to better estimate, describe and control consumer exposure and therefore safety. To use the legal maxim, hard cases make bad law - an extreme case is a poor basis for a general law that would cover a wider range of less extreme cases.
- Since certain classes of substances can migrate extensively, and if they cannot be excluded from P/B, then it is recommended that migration modelling work be conducted with a view to deriving

some substance-specific CFs for important combinations of substance (class) / food (category) / t/T (conditions).

INTRODUCTION

The concept of Correction Factors (CFs) was introduced during the Biosafepaper project which was funded by the European Union under contract QLK1-2001-00930 and by a consortium of 16 paper and board making companies. The Biosafepaper project researched into the application of bioassays for the safety assessment of paper and board for food contact [Severin *et al.* 2005; Bradley *et al.* 2008; Bradley *et al.* 2010; Honkalampi-Hämäläinen *et al.* 2010] but Correction Factors have a wider applicability since they aim to relate migration levels into food with migration levels obtained with the food simulants or extraction solvents used in rapid, simplified and standardised test procedures such as those described in CEN standards.

Correction Factors are directly analogous to the reduction factors described in EU legislation on plastics for food contact. There it is stated that food simulant D2 (vegetable oil) shall be used to simulate foods which contain free fats at the surface [EU 10/2011]. However, the regulation further states that the result obtained with food simulant may in certain cases significantly overestimate migration into food (and) in these cases the result in food simulant is corrected by a reduction factor. Simulant D2 reduction factors are allocated to different broad food categories and are in the range of 2 to 5 depending on how closely or not, simulant D2 and the associated time and temperature migration test conditions used, do really simulate (mimic) that category of fatty food.

This work has reviewed published studies along with any unpublished reports on the topic of Correction Factors provided for this review by CEPI. The aim was to relate chemical migration levels from paper and board into foods, with the migration or extraction values obtained using food simulants or solvents. If and where they are found to exist, these relationships are the quantitative evidence underpinning numerical Correction Factors. The work aimed to propose a set of Correction Factors where justified and to identify any data gaps and further work that would be needed to fill such gaps.

EXTRACTION / MIGRATION TEST METHODS AVAILABLE FOR P/B

There are currently 4 key CEN methods published that describe extraction or migration procedures for paper and board intended to come into contact with foodstuffs. Their purpose is to place the P/B in contact with a simulant or an extraction solvent under defined test conditions, so that the simulant/solvent can be analysed subsequently for any chemical substances of interest.

Table 1. Test simulations described in CEN standards for P/B

Standard #	Title	Purpose
EN 645:1993	Preparation of a cold water extract	Cold-water extractables
EN 647:1993	Preparation of a hot water extract	Hot-water extractables
EN 14338:2003	Conditions for determination of migration from paper and board using modified polyphenylene oxide (MPPO) as a simulant [‡]	Migration simulant for dry foods
EN 15519:2007	Preparation of an organic solvent extract	Migration simulant for fatty foods *

[‡] Note that MPPO has now been re-named as PPO in the Plastics Regulation 10/2011. The more common name of Tenax will be used generally in this report.

* It is stated that: ‘This European Standard specifies a test method for the assessment of substitute tests performed with volatile test media for the determination of migration from paper and board intended to come

into contact with fatty foodstuffs at all temperatures and for any period of time. At the time that this European Standard was prepared, the EU directives for plastics coming into contact with food required use of isooctane or 95 % v/v aqueous ethanol.

There are other CEN standards that describe extraction or leeching tests using solvents or other extraction media but they are not considered pertinent here. They are listed below for completeness:

EN 1104:2005: Paper and board intended to come into contact with foodstuffs - Determination of the transfer of antimicrobial constituents.

EN 14719:2005: Pulp, paper and board - Determination of the Diisopropylnaphthalene (DiPN) content by solvent extraction.

EN 646:2006: Paper and board intended to come into contact with foodstuffs - Determination of colour fastness of dyed paper and board.

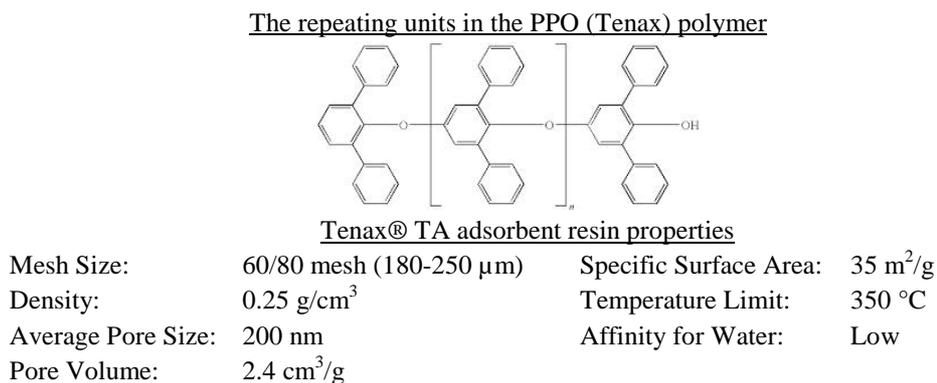
EN 648:2006: Paper and board intended to come into contact with foodstuffs - Determination of the fastness of fluorescent whitened paper and board.

The simulants / extraction solvents water, isooctane and 95% ethanol need little introduction. They represent the full range of polarity and will extract the most polar (e.g. ionic salts, polar substances such as acrylamide or epichlorohydrin) up to the most non-polar substances (e.g. mineral hydrocarbons or DiPN).

Tenax requires a little more introduction. PPO as it is now termed in the Plastics Regulation 10/2011, is described in that regulation simply as: Food simulant E; poly(2,6-diphenyl-p-phenylene oxide), particle size 60-80 mesh, pore size 200 nm.

Poly(2,6-diphenyl-p-phenylene oxide) (PPO) is a porous polymer resin made by oxidative polymerisation of 2,6-diphenylphenol. It is generally known by its trademark Tenax. Tenax is a polymeric material that absorbs a wide variety of organic substances. It has a largely hydrocarbon character but also has aromatic character (allowing pi-pi interactions and polarisability) and also has a polyether character. Consequently it is a good absorbent for both polar and non-polar substances. It has a very high specific surface area, a good temperature resistance, and its sorption capacity is largely unaffected by moisture. The mesh size fraction 60/80 corresponds to a particle size range of ca. 180 to 250 micrometers so it is a fine powder. P/B is a web of tangled fibres that are flat or cylindrical (depending on the wood species) with a fibre width of ca. 20 to 50 micrometers. Tenax will make dry but intimate contact with the P/B surface and will fill most surface irregularities, but the powder is too large to penetrate into the fibre network.

Figure 1. Physical and chemical properties of PPO (aka Tenax)



Tenax was first specified in EU regulations on plastic materials and articles intended to come into contact with food, as a simulant suitable for testing at high temperatures, to supplement tests with the volatile fatty food simulants isooctane and/or 95% ethanol. The main concept behind food simulants is that they should be capable of representing a broad category (type) of foods and should be easy to use and analyse in migration tests. Tenax was subsequently specified as a simulant for dry foods and at that time the test conditions intended to simulate long-term storage of foods in contact with plastics at ambient temperature were 10 days at 40°C. The aim was to have an accelerated test that could be conducted in a reasonable period of time, without having to wait the whole shelf-life of a product which could be one year or more. The test temperature of 40°C was intended to compensate for the shorter time but this was found to be inadequate so the new plastics regulation changed the test conditions to 10 days at 60°C to better simulate long-term storage at ambient temperatures [EC 2011].

Since in the absence of specific and detailed regulation at the EU level on P/B, Tenax had become the presumptive standard for testing P/B too. Some researchers have now also adopted the new test condition of 60°C rather than 40°C although as we will see later, the higher temperature of 60°C can cause atypical behaviour especially the migration of less volatile substances (vapour pressure is critical for migration into dry foods) that would not migrate to any significant extent at ambient temperatures. Such “atypical behaviour” could in principle lead to an under- or overestimation of migration when using Tenax, depending on the exact characteristics of the chemical involved. This is one further reason for the need to consider the applicability and the numerical values of Correction Factors for Tenax.

Other simulants for dry foods have been explored, such as Porapak, silica gel, activated carbon (charcoal), or even foods themselves such as semolina, rice or sugar. In many cases these reports present no data at all for migration into foods and the conclusions (on the suitability or otherwise of these alternative simulants for dry foods) seem to be based simply on the convenience of the analysis and the attractive kinetic curves that can be generated rather than any evidence that as a simulant they really do simulate migration into foods. None has been found to be better than Tenax and they are not considered further here.

SUBSTANCES THAT MAY MIGRATE FROM P/B

Substances that may migrate from P/B to food have a variety of possible sources:

- Wood constituents (e.g. naturally-occurring rosin acids, esters)

- Process aids that are not intended to remain in the P/B (e.g. defoaming agents, dewatering aids). Some being fibre-retentive, some being non-fibre-retentive
- Additives that are intended to remain in the P/B (e.g. coatings, grease-proofing, whiteners)
- Recycling-derived contaminants
- Other sources especially printing inks and lacquers but also adhesives used to fabricate the finished food contact material or article
- General (environmental) contaminants (e.g. Pb, Cd)

Virtually all of the very recent literature has been concerned with only two of these bulleted sources, migration from substances originating from inks and adhesives, either directly from the printed and fabricated food packaging or indirectly from the use of recovered fibres that can contain chemical residues from inks (including thermal papers and carbon-less copy papers) and adhesives. Paradoxically, virtually no literature is used here on migration of the other bulleted ‘classes’ and, put simply, this is because generally they do not migrate to concentrations that are of interest to researchers, funding bodies and publications. So little useable quantitative information is available for them. It could be argued that this review is therefore unbalanced, being dominated by chemicals that are not from P/B per se. But this is the reality of P/B food contact materials.

FOOD CATEGORIES TO BE CONSIDERED

The following description of the uses of paper and board [CEPI and CITPA 2012] sets the scene for the food contact applications (categories) that need to be considered here.

“ Paper and board has a long and successful history of safe use in the food industry in a wide range of applications. These include applications where intimate contact is involved, such as tea bags, baking papers, and filters, and direct contact packaging such as butter wrapping, sugar bags, and cartons for dry and frozen foods. In addition it has a very wide range of uses in transport and distribution packaging.

Although there is a wide range of applications, the proportion of packaging made from uncoated and untreated paper and board and coming into direct contact with food bought by the end-consumer was estimated to be less than 3.5% (3.39% is the actual value quoted) of all direct contact food packaging in the EU-15 in 2000 (Ref. 1). Compared to other direct contact food packaging materials, for example plastics (estimated proportion around 70%), this estimate of 3.5%, less than 0.9 kg paper per person per year, is relatively low and thus consumer exposure is similarly low. In addition direct contact is primarily with dry food (approximately 50%) and with food that is to be peeled or washed (approximately 30%) so only the remaining 20% is for contact with moist and/or fatty food.

Uncoated and untreated paper and board is not suitable to pack food with very high moisture content (for example liquid food or wet chilled products), since exposure to high moisture will cause disintegration of the material. For these food types coated paper and board is commonly used, and in the great majority of applications direct food contact is with a plastic layer. The proportion of packaging made from coated paper and board was estimated to be 17% (equivalent to 4.4 kg per person per year) of all direct contact food packaging in the EU-15 in 2003 (Ref. 2). Between 70-80% of all coated paper and board that is in direct contact with food is liquid packaging board (used for instance in cartons for milk and beverages) and 75% of this has aluminium foil as a barrier layer in the laminate structure that will prevent migration from the paper and board. If liquid packaging board containing an aluminium layer is excluded, the proportion of coated paper and board packaging used for food contact is 7.6% (equivalent to 1.93 kg per person per year). ”

With this background, this report has been organised in chapters according to the different categories of food contact that P/B is used for; (i) Fruits and vegetables (fresh, whole); (ii) Frozen foods; (iii) Fatty contact; (iv) Dry foods (long-term ambient storage); (v) Baking papers; (vi) Hot and cold water including filters; and lastly, the special case of (vii) Inorganic Substances.

SEARCH AND SELECTION CRITERIA

The project scope of work stated that CEPI would provide “published work plus unpublished reports” to Fera for review. One report was provided [Gonzalez and Cooper 2012].

The ISI Web of Knowledge and the Internet (Google) were searched using combinations of the words; paper, board, migration, food, simulant, transfer, survey, packaging, contact. The cut-off date for the literature that was searched and used was 1st January 2014.

Promising references that were cited in the publications selected (see below) were inspected (abstracts only) to reveal any further information sources.

The publication (often just the abstract) was inspected to see if it could fulfil this requirement:

- Migration data into foods and food simulants,
- from the same well characterised and commercially-relevant P/B samples,
- with compositional information (exhaustive solvent extraction),
- ideally with migration measurements at different time points,
- preferably for intrinsic substances rather than substances spiked-in,
- with good AQA (analytical quality assurance) and
- with not too many non-detects.

As a basic minimum, publications had to have migration data for foods along with simulant or solvent extraction data for the same P/B sample(s).

In all cases this work has collected the published data, assessed its methodological quality and then assessed the data for the purpose of deriving Correction Factors. Where text taken from the original papers has been used here directly, then the attribution should be clear by way of a [reference]. Should any text or conclusions or theories etc. seem copied but unattributed, this is inadvertent and if notified we will remedy this.

INTRINSIC OR SPIKED SUBSTANCES ?

To determine the relationship between food and simulant migration data, the analyte of interest must be present at a high enough concentration in the P/B to result in measurable migration. Many published studies on so-called intrinsic substances, are incomplete or inconclusive because too many data points (sometimes even all) are ‘non-detects’. As a consequence, many workers have elected to spike the P/B with one or more substances as model migrants. Typically a range of substances will be added to the P/B to allow the migration of chemicals covering a range of polarities and volatilities to be studied. When spiking a matrix in this way, problems may occur with inhomogeneous

incorporation of substances throughout the matrix. Therefore the homogeneity of incorporation should be taken into consideration when assessing the uncertainty of the migration results. In addition, the way in which these substances integrate into the P/B matrix may not be the same as if they were bound to the matrix during paper-making; i.e. those present as a result of using recovered fibres or chemicals added intentionally to the P/B to perform a specific function. If the spiked substances are not incorporated into the matrix in the same way (being more- or less- tightly bound for example) then migration may not occur at the same rate. Such uncertainties need to be considered when using the migration results for spiked P/B samples.

SHELF-LIFE CONSIDERATIONS

There are a number of factors that will affect the actual amount of a given substance that will migrate into foods or food simulants. Material type, food type, nature of the migrant, contact time, contact temperature and extent of contact (direct/indirect, point contact/continuous contact) may all influence the extent of the migration that will occur under real storage conditions of foods. Many products packaged in direct contact with paper and board matrices are dry foodstuffs which are stable commodities with long shelf lives of a year or more. For the strict derivation of Correction Factors, to relate migration values between simulant data and food data, then the migration test conditions investigated should be defined to result in the worst foreseeable migration, i.e. the highest concentration of the migrant in the foodstuff. We will return to this point.

FRUITS AND VEGETABLES (FRESH, WHOLE)

FV#1. A study funded by CEPI set out to investigate the migration of 8 recycling-related contaminants from 10 P/B samples into six types of fruits and vegetables [Gonzalez and Cooper 2012]. The substances were diisopropyl naphthalene (DiPN), benzophenone (BP), diisobutyl phthalate (DiBP), di-n-butyl phthalate (DBP), di-(2-ethylhexyl) phthalate (DEHP), di-2-ethylhexyl adipate (DEHA), diethylene glycol dibenzoate (DEGDB) and vanillin (VAN). The fruits were strawberries, blueberries and grapes. The vegetables were Brussels sprouts, potatoes and carrots. The P/B samples were supplied by CEPI. For reasons of low composition and/or low migration, there was no complete data set for any of the combinations.

Table 2. Substances monitored by Gonzalez and Cooper 2012

Substance	MW (g/mol)	Boiling point (°C)	Log Pow *
VAN Vanillin (4-hydroxy-3-methoxy-benzaldehyde)	152	285	1.19
BP Benzophenone	182	306	3.38
DiPN Diisopropylnaphthalenes, isomeric mix	212	295	5.65
DiBP Diisobutyl phthalate	278	327	4.11
DBP Dibutyl phthalate	278	340	4.45
DEGDB Diethylene glycol dibenzoate	314	>230 (dec) 408 calc	3.2
DEHA Di(2-ethylhexyl) adipate	370	417	8.1
DEHP Di(2-ethylhexyl) phthalate	390	386	7.5

* Log Pow is the log of the partition coefficient of the substance between octanol and water. Experimental or estimated values from ChemSpider at <http://www.acdlabs.com/>

Concentrations in the P/B samples were determined by extraction into acetone followed by gas chromatography-mass spectrometry analysis. All of the selected substances were present in the P/B samples and so no additional impregnation was required thereby avoiding any uncertainty associated with homogeneity and type of incorporation into the matrix.

Small boxes were made by folding the board samples and the food was placed in the box. The food was totally enclosed by the 6-sided box. The food-in-boxes were not overwrapped in aluminium foil. Exposure times and temperatures (see Table) were selected to represent the worst foreseeable contact conditions and the migration tests were set up to mimic the partial contact that typically occurs between fruit and vegetables and the materials in which they are packed. No tests with food simulants were conducted and the intention was to compare the migration results with the concentrations (on an area-related basis) in the paperboard. Although the experimental design allowed for the extraction data to be compared directly with the migration data this was not possible for all substances. Some were present at too low concentrations in the P/B samples for migration to be detectable in the exposed foodstuffs. This was the case for BP, DEHA and DEGDB.

For the DEHP and DBP the concentrations detected in the control food samples (i.e. those that had not been exposed to the P/B samples) were too high and too variable for any usable migration data to be derived. Similarly, for the carrots the DiBP levels were too high and too variable for any usable migration data to be derived.

Data for DiBP migration for all foodstuffs except carrots (see above) were used to estimate the relationship between the extraction and migration data. From these data the authors concluded that it

may seem appropriate to derive a Correction Factor of 20 to be applied to extraction data to provide an estimation of the migration that will occur into the foodstuff. For each of the combinations of P/B and foodstuff investigated this would still result in an overestimation of the actual migration that would take place, thereby providing a margin of safety for the consumer.

Data for DiPN migration were also used to estimate the relationship between the extraction and migration data. From this data the authors concluded that it may seem appropriate to derive a Correction Factor of 5 to be applied to extraction data to provide an estimation of the migration that will occur into the foodstuff. For each of the combinations of P/B and foodstuff investigated this would still result in an overestimation of the actual migration that would take place, thereby providing a margin of safety for the consumer.

There was no detectable migration of BP, DEHA or DEGDB into any of the foods. The authors estimated the limit of detection (LoD) for these three substances and then placed an upper limit on the percentage migration, assuming that migration was at the LoD (a conservative assumption). The LoDs were 1, 10 and 20 $\mu\text{g}/\text{dm}^2$ for benzophenone, DEHA and DEGDB, respectively. The result of these calculations (Table) did not provide any information useful for derivation of Correction Factors.

Table 3. Migration expressed as a percentage of the total content of the P/B sample [Gonzalez and Cooper 2012]

	CEPI 2 GD board	CEPI 4 Corrugated board	CEPI 7 GD hard sized board
DiBP in the board	46 $\mu\text{g}/\text{dm}^2$	47 $\mu\text{g}/\text{dm}^2$	12 $\mu\text{g}/\text{dm}^2$
Strawberries (6 days at 23°C)	1.7%	not tested	1.5%
Blueberries (10 days at 23°C)	0.6%	0.8%	not tested
Grapes(10 days at 23°C)	0.9%	1.2%	1.3%
Brussels sprouts (15 days at 23°C)	2.4%	4.2%	0.7%
Potatoes (30 days at 23°C)	1.9%	2.7%	not tested
DiPN in the board	239 $\mu\text{g}/\text{dm}^2$	172 $\mu\text{g}/\text{dm}^2$	30 $\mu\text{g}/\text{dm}^2$
Strawberries (6 days at 23°C)	2.8%	not tested	8.1%
Blueberries (10 days at 23°C)	1.8%	2.9%	not tested
Grapes(10 days at 23°C)	2.4%	2.9%	7.9%
Brussels sprouts (15 days at 23°C)	3.4%	4.4%	12.0%
Potatoes (30 days at 23°C)	0.7%	1.0%	not tested
Carrots (16 days at 23°C)	4.9%	not tested	12.6%
BP in the board	11 $\mu\text{g}/\text{dm}^2$	15 $\mu\text{g}/\text{dm}^2$	3.5 $\mu\text{g}/\text{dm}^2$
not detected in any of the 5 foods	nd (<9.1%)	nd (<6.7%)	nd (<28.6%)
DEHA in the board	7.9 $\mu\text{g}/\text{dm}^2$	12 $\mu\text{g}/\text{dm}^2$	6.3 $\mu\text{g}/\text{dm}^2$
not detected in any of the 5 foods	nd (<100%)	nd (<83%)	nd (<100%)
DEGDB in the board	95 $\mu\text{g}/\text{dm}^2$	74 $\mu\text{g}/\text{dm}^2$	20 $\mu\text{g}/\text{dm}^2$
not detected in any of the 5 foods	nd (<21%)	nd (<13.5%)	nd (<100%)

FV#2. In a second study funded by CEPI [Fera 2009] four P/B samples were impregnated with *o*-xylene, acetophenone, dodecane, benzophenone, DiPN and DiBP and migration into fruits and vegetables was determined. Extraction data for the impregnated P/B samples was compared with migration data using the food simulant Tenax and with migration data for 5 types of fruits and vegetables (mushrooms, apples, potatoes, bananas and raisins).

Table 4. Model substances used in Fera 2009

Substance	Molecular weight (g/mol)	Boiling point (°C)	Log Pow *
<i>o</i> -Xylene	106	144	3.12
Acetophenone	120	203	1.58
<i>n</i> -Dodecane	170	216	2.92
Benzophenone	182	306	3.38
Diisopropyl naphthalenes, isomeric mix	212	295	5.65
Diisobutyl phthalate	278	340	4.11

* Log Pow is the log of the partition coefficient of the substance between octanol and water. Experimental or estimated values from ChemSpider at <http://www.acdlabs.com/>

Exposure of the P/B samples to the food simulant Tenax was carried out in accordance with EN 14338:2003. In short, 0.6 dm² of P/B was exposed to 2.4 g of Tenax for 10 days at 40°C. Ethanol extracts of the exposed Tenax were analysed by gas chromatography-mass spectrometry.

The food samples were packaged in the impregnated P/B samples, loosely overwrapped in aluminium foil to mimic stacking and allowed to stand at room temperature (20°C) for 5 days for the perishable products and for 10 days at 40°C for the raisins. The percent migration is shown in the Table. For some combinations of analyte and foodstuff the calculated migration was > 100%. This was said to be due to the inhomogeneity of the incorporation, estimated to be up to 30%.

Table 5. Migration of model substances into foods expressed as a percentage of the concentration in the paper/board samples [Fera 2009]

Paper/board sample	<i>o</i> -Xylene	Acetophen.	<i>n</i> -Dodecane	Benzophen.	DiPN	DiBP
Corrugated board migration						
into mushrooms (5 days at 20°C)	< LoD	4.7%	0.01%	4.3%	0.62%	0.47%
into raisins (10 days at 40°C)	1.0%	5.6%	0.32%	30%	(183%)	74%
into Tenax (10 days at 40°C)	< LoD	11%	(166%)	17%	(108%)	53%
Bread bag migration						
into mushrooms (5 days at 20°C)	0.67%	1.6%	0.01%	2.4%	0.32%	0.38%
into apples (5 days at 20°C)	< LoD	2.5%	0.12%	0.19%	19%	13%
into potatoes (5 days at 20°C)	0.67%	0.56%	0.03%	4.2%	1.4%	0.56%
into raisins (10 days at 40°C)	< LoD	2.3%	0.12%	7.3%	14%	8.0%
into Tenax (10 days at 40°C)	< LoD	2%	16%	9%	16%	19%
Fruit board migration						
into mushrooms (5 days at 20°C)	0.03%	7.8%	0.31%	2.9%	0.21%	0.58%
into apples (5 days at 20°C)	0.03%	2.6%	1.3%	0.11%	15%	3.2%
into potatoes (5 days at 20°C)	0.10%	0.30%	0.05%	0.72%	0.17%	0.39%
into raisins (10 days at 40°C)	0.08%	1.0%	0.64%	7.1%	42%	19%
into Tenax (10 days at 40°C)	13%	17%	(266%)	25%	79%	75%
Grey board migration						
into mushrooms (5 days at 20°C)	0.27%	10%	0.28%	3.7%	0.13%	0.35%
into apples (5 days at 20°C)	0.02%	3.4%	1.1%	0.10%	9.4%	1.9%
into potatoes (5 days at 20°C)	0.08%	0.22%	0.03%	0.80%	0.11%	0.21%
into raisins (10 days at 40°C)	0.27%	3.1%	1.6%	14%	52%	21%
into Tenax (10 days at 40°C)	23%	21%	81%	22%	47%	44%

Migration into foods - The migration results were attributed to a combination of three factors: (i) the extent of direct contact made between the P/B and the food (most important for the rather non volatile substances); (ii) the extent of indirect contact and the surface area of the food (important for the semi-volatile substances); (iii) the polarity of the food surface (which sets the affinity of each substance for that food).

So taking apples as an example, the authors noted that the skin can have a waxy surface with a high affinity for non-polar analytes and the pair of least polar substances, DiPN and DiBP, had the highest migration from all three P/B types tested with apples. Migration of DiPN and DiBP into apples was highest for the bread bag which was a flexible paper and the experimental set-up used meant that it made almost continuous direct contact with the whole apple surface. Migration of benzophenone, which has a boiling point between that of DiPN and DiBP (306, 295 and 340°C respectively) but is more polar than both (log Pow values of 3.38, 5.65 and 4.11 respectively) migrated far less into apples than did DiPN and DiBP.

In contrast, benzophenone was the substance that migrated to the largest extent into potatoes. The authors noted that potato tubers have a less waxy skin than apple fruits, resulting in a higher uptake of the more polar benzophenone. Similarly, of the six substances used, benzophenone along with acetophenone had the highest migration levels into the mushrooms. This was rationalised because the mushroom surface has a relatively polar surface and, especially for acetophenone (the most volatile excluding *o*-xylene), because migration is likely to include vapour phase transfer and mushrooms have a large surface area because of their gills.

Migration into raisins was generally much higher and much more uniform (across the 6 substances) than into the fresh fruits and vegetables. Raisins are not a fresh fruit but are preserved by drying and they are approximately 80% carbohydrate by weight. The relatively high and uniform migration into raisins can be explained partly by the longer storage time and the higher temperature used (10 days at 40°C compared with 5 days at room temperature for the fresh fruits and vegetables) and partly since raisins have a smaller particle size and are soft and malleable, so the contact with P/B was more extensive.

Migration into Tenax - The migration results for exposure of the impregnated P/B samples to Tenax for 10 days at 40°C are also given in the Table. Allowing for the occasional outliers, looking both across the six substances and down the four P/B types, these results are much more uniform than the migration results into foods. This was expected by the authors because Tenax is a fine powder that makes uniform and intimate contact with the P/B samples and because Tenax has a high affinity for organic compounds.

Comparing migration into foods and into Tenax - To allow some general conclusions to be drawn, the Table summaries the range and the average level of migration of the set of six substances from the four P/B types. There were no obvious trends between the four different P/B samples in that migration levels seemed to depend more on the nature of the model substance and the nature of the food rather than on the detailed characteristics of the P/B. Probably the most important factor with respect to the P/B is the grammage since this determines the flexibility and therefore to what extent the P/B can mould to the shape of the packed food item and give extensive contact. This being so, data for the four P/B types are gathered together and averaged to provide a simple overview in the Table of migration levels into the different foods and how they compare with the simulant Tenax.

Table 6. Summary of the migration of the impregnated model substances [Fera 2009].

	Mushrooms		Apples		Potatoes		Raisins		Tenax	
	range	mean.	range	mean	range	mean	range	mean	range	mean
Corrugated board	0-4.7%	1.7%	nt	nt	nt	nt	0.3-100%	35%	0-100%	48%
Bread bag	0-2.4%	0.9%	0-19%	5.8%	0-4.2%	1.2%	0-14%	5.3%	0-20%	10%
Fruit board	0-7.8%	2.0%	0-15%	3.8%	0-0.7%	0.3%	0-42%	12%	13-100%	51%
Grey board	0-10%	2.5%	0-9.4%	2.6%	0-0.8%	0.2%	0.3-52%	15%	21-81%	39%
Mean of all		2%		4%		0.6%		17		37%

Migration levels into the foods or simulant are expressed as a percentage of the starting amount in the P/B and are the range and the simple arithmetic mean of the results for the 6 model substance. nt = not tested

According to the authors [Fera 2009] for the foods with contact conditions of 5 days at room temperature, the standard test with Tenax according to EN 14338:2003 (i.e. 10 days at 40°C) overestimates migration by a factor of 9 for apples, by a factor of 21 for mushrooms, and by a factor of 62 for potatoes. For raisins (contact conditions of 10 days at 40°C) the overestimation is much smaller but Tenax still overestimates migration by a factor of 2.2. and so to cover all events then a Correction Factor of < 9 would be required.

Migration of substances intrinsic to the paper/board One concern with the experimental design used by [Fera 2009] and by others, is that the spiked P/B may not be representative of 'real' migration of intrinsic migrants. Analysis of the P/B samples as received detected DiPN and DiBP. These two substances are commonly found in recycled P/B. The intrinsic levels of DiPN and DiBP were much lower than the levels of these two model substances that were subsequently spiked in. The levels in the bread bag paper were considered too low for migration tests but the other three P/B types were used 'as received' in migration studies using Tenax and raisins and for which the results are summarised in the Table. The Table also gives the migration results for the P/B samples into which DiPN and DiBP were impregnated. Considering the results for Tenax first, migration (as a percentage of the P/B content) of the spiked DiPN and DiBP was more extensive than of the intrinsic substances, by a factor of 2 to 3. This suggests that the intrinsic substances are more strongly bound into the paper matrix than the substances that were spiked in and simply allowed to equilibrate within the P/B matrix. The effect was even larger for migration into raisins for which the impregnated/intrinsic ratio was 24 for DiPN and 25 for DiBP. The fact that about the same ratio was seen for both DiBP and DiPN, suggests that the effect is due to a stronger interaction with the paper matrix and is not due to the special case where DiPN can occur in recycled P/B in microcapsules coming unbroken from carbonless copypapers and consequently with lowered migration properties. For intrinsic DiPN and DiBP the ratio of migration into Tenax compared to raisins, for the same storage time and temperature conditions, was 12.6 and 12.0 for the two substances respectively.

Table 7. Comparison of migration of intrinsic and impregnated substances from paper/board samples [Fera 2009]

	Tenax		Tenax		Raisins		Raisins	
	Intrinsic		Impregnated		Intrinsic		Impregnated	
	DiPN	DiBP	DiPN	DiBP	DiPN	DiBP	DiPN	DiBP
Corrugated board	37%	21%	100%	53%	3.5%	2.4%	100%	74%
Bread bag	<LoD	<LoD	(16)	(19)	<LoD	<LoD	(14)	(8)
Fruit board	36%	18%	79%	75%	3.1%	0.9%	42%	19%
Grey board	29%	16%	47%	44%	1.6%	1.2%	52%	21%
Average	34%	18%	75%	57%	2.7%	1.5%	65%	38%
Ratio IMP / INT	-	-	2.2	3.2	-	-	24	25

Migration levels are expressed as a percentage of the starting concentration in the P/B. () bracketed data not used in the calculation of the average since there is no corresponding value to calculate the Ratio IMP / INT

The studies described here demonstrate the difficulties in carrying out comprehensive studies using intrinsic contaminants, i.e. concentrations in the P/B samples are usually too low to give measureable migration levels and intrinsic contaminants can be the same as contaminants already present in the foodstuff [Gonzalez and Cooper 2012]. On the other hand, studies using impregnated P/B have their pitfalls too, i.e. inhomogeneity of incorporation and that migration of the substances added may be higher than that of intrinsic migratable substances.

Conclusions. Comparing migration into Tenax under standard test conditions (10 days at 40°C), using the results from the Fera project, overestimated the migration into the fresh fruit and vegetables by a factor of 9 to 62 and the application of a Correction Factor of 10 to the Tenax migration data would provide a good estimate for the migration into the food. This would also accommodate the more limited data from [Gonzalez and Cooper 2012] that had composition/migration factors in the range of 5 to 20.

Conclusions. For raisins (contact conditions of 10 days at 40°C for both raisins and Tenax) the overestimation is much smaller but Tenax still overestimates migration by a factor of 2.2. However considering the long shelf-life of raisins no Correction Factors would seem to be appropriate. As described, raisins are a dried fruit and further discussion is presented in the dry foods section.

Limitations. These conclusions on Correction Factors for fresh fruits and vegetables do not take account of any reduction in concentrations that may be brought about by processing operations such as washing, peeling and/or cooking.

Recommendations. None.

FROZEN FOODS

FF#1. In kinetic migration studies funded by CEPI and aimed at deriving diffusion and partition coefficients for use in migration modelling, Zülch and Piringer [2010] presented some migration results into frozen foods that are of value here. The P/B samples used were of food-grade quality, unprinted and supplied by members of the CEPI Food Contact group. The target substances varied in their molecular weights (in the range 178–390 Da) as well as their chemical properties, especially in their polarity. Whereas diisobutyl phthalate (DiBP), benzyl-2-naphthyl ether (BENE), di(2-ethylhexyl) phthalate (DEHP), and diisopropyl naphthalene (DiPN) represent the non-polar, hydrophobic compounds, benzophenone (BP) and triclosan (TRICL) are polar migrants.

Migration into foods was in the range of 0.3 to 3% of the initial content in the P/B. Migration into Tenax was higher in the range 0.5 to 9%. BP was atypical in having much lower migration (percentage) levels although the data were limited. Similarly, butter behaved differently to the other foods studied and gave higher migration levels, 3 to 8%. The result of 26.3% migration of DiBP into butter is considered to be questionable and is not used here.

Table 8. Migration of intrinsic substances into frozen foods stored for 90 days at -18°C [Zülch and Piringer 2010]

	DiBP	BP	BENE
Wheat flour	2 %	0.04 %	0.3 %
Chocolate	1.3 %		0.4 %
Egg pasta	nt		
Sponge biscuits	nt		
Puff pastry	1.7 %		
Raspberries	2.2 & 3.8 %		
Tenax	3.8 %	0.08 %	0.5 %

Table 9. Migration of spiked substances into frozen food at -18°C [Zülch and Piringer 2010]

	BP	DiBP	DiPN	DEHP	PHEN	TRICL
Butter; 60 days at -18°C	2.7 %	26.3 %	8.2 %	2.7 %	4.1 %	5.1 %
Tenax; 30 or 60 days at -18°C	<0.4 %	0.6 %	9.4 %	<1 %	2.1 %	<0.4 %

FF#2. A study of benzophenone migration from cartonboard assessed the relative migration levels seen under different contact conditions of retail foods packaged in printed cartonboard [Anderson and Castle 2003]. Frozen foods packaged in printed cartonboard were purchased from retail stores and stored at -20°C. The stored samples were over-wrapped in aluminium foil to limit any inter-sample migration. Once the best-by date was reached or after 1 month from purchase (whichever came first), the food was removed from the packaging and the food and the packaging P/B was analysed. Shelf-stable and chilled foods were studied too. For direct contact and room temperature storage, the average mass fraction migration of benzophenone was 16.1%. The average migration at room temperature storage but with indirect contact was 6 fold lower at 2.7%. The average migration with direct contact but with chilled or frozen storage was again 6 fold lower at 2.6%. Thus, the benefits of a lower storage temperature were judged to be the same as the benefits of making only indirect contact. Finally, the average migration with both indirect contact and with chilled or frozen storage was only 0.4% and this was 40 times lower than the room temperature/direct contact average. According to the authors, this shows that to a good approximation, the attenuation effects of indirect

contact and of low temperature storage are cumulative; with a 6 fold reduction for indirect contact compared with direct contact, a 6 fold reduction for chilled/frozen storage compared with ambient storage, and a 6 x 6 or ca. 40 fold reduction for the two contact conditions combined.

FF#3. In their tests that had as the main focus migration into fruits and vegetables, [Gonzalez and Cooper 2012] conducted additional experiments to study migration into frozen fish fingers (sticks of coarsely-minced fish with a battered-breadcrumb coating) using migration conditions of 4 months at -18°C. In this case the exposure time (4 months) was much shorter than the product shelf life (typically up to 2 years) and so this had to be considered when interpreting the findings. For DiPN, 3.5% of the extraction concentration migrated into the fish fingers. They assumed that equilibrium had not been reached in the migration kinetic and that instead a linear (i.e. conservative) migration rate would be followed until the end of the product shelf-life. In this way they estimated that the migration after a 2 year exposure period could be 21% of the extraction level. Thus a Correction Factor of 5 could be applied to the extraction data to provide an estimation of the migration that could occur into the foodstuff at the end of its shelf-life.

FF#4. In a systematic evaluation of chemical migration during low temperature storage of packaged foodstuffs, five model migrants were incorporated into inks used to print cartonboard [Bradley *et al.*, 2001]. The substances were limonene, acetophenone, cyclohexylbenzene, dimethylphthalate and benzophenone. They were incorporated at 0.1% each into a paste ink and the ink was printed at 2 g/m² onto cartonboard (uncoated, 3 g/dm²) by a sheet-fed offset lithographic process. The paste ink was based on drying oils such as linseed and tung which dry by oxidation and absorption and so did not require heating to dry.

Table 10. Model substances used (in order of boiling point) [Bradley et al. 2001]

Substance	Boiling point at STP, °C	Molecular weight	Solubility in water, g/L at 25°C	log P _{ow}	Incorp. (µg/dm ²)
LIM Limonene	176	136	0.014	4.57	9
ACP Acetophenone	202	120	6.1	1.58	40
CHB Cyclohexylbenzene	240	160	0.0053	4.81	30
DMP Dimethyl phthalate	282	194	4.0	1.60	44
BP Benzophenone	305	182	0.14	3.18	50

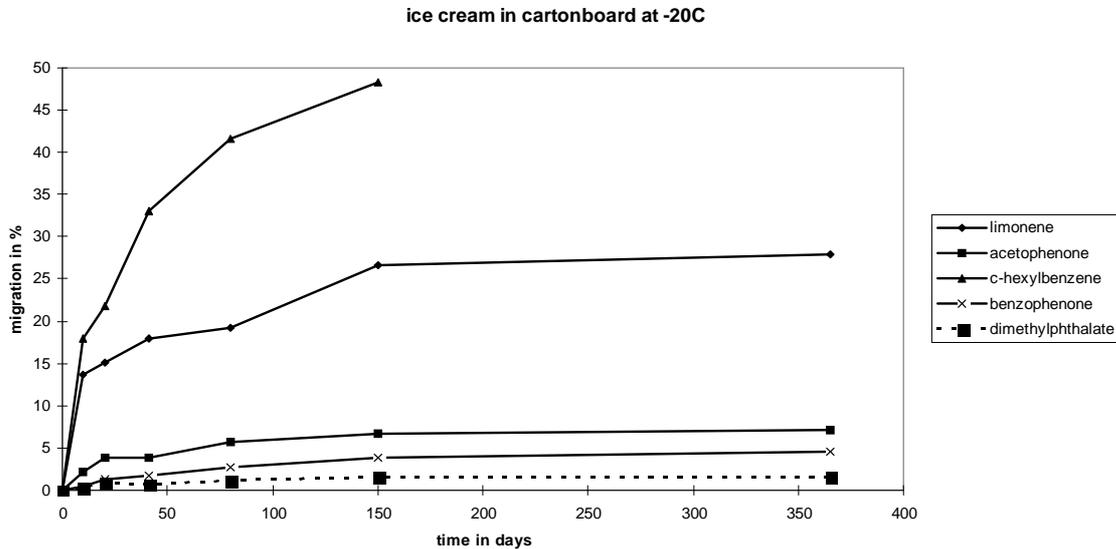
The cartonboard was formed into boxes (3.5-4.0 dm² surface area) using a hot-melt adhesive and used to pack sausage rolls (fatty puff pastry filled with seasoned sausage meat; 180 g) or fish fingers (sticks of coarsely-minced fish with a battered-breadcrumb coating; 250 g) or ice-cream (14 x 9 x 3 cm block). The packed cartons were stored at -20°C for up to one year. The cartons were not over-wrapped with aluminium foil. Migration studies using the simulants distilled water, 95% ethanol and Tenax at -20°C and 5°C were also conducted, in tightly-sealed migration cells that prevented any loss of substances other than by migration to the simulant.

Migration into foods stored at -20 °C

Ice cream: The fastest migration was seen for cyclohexylbenzene which rose to 50% transfer after 150 days storage in the cartonboard at -20°C. No data point is shown for this substance at the one year timepoint because according to the study report, those samples were spiked with cyclohexylbenzene

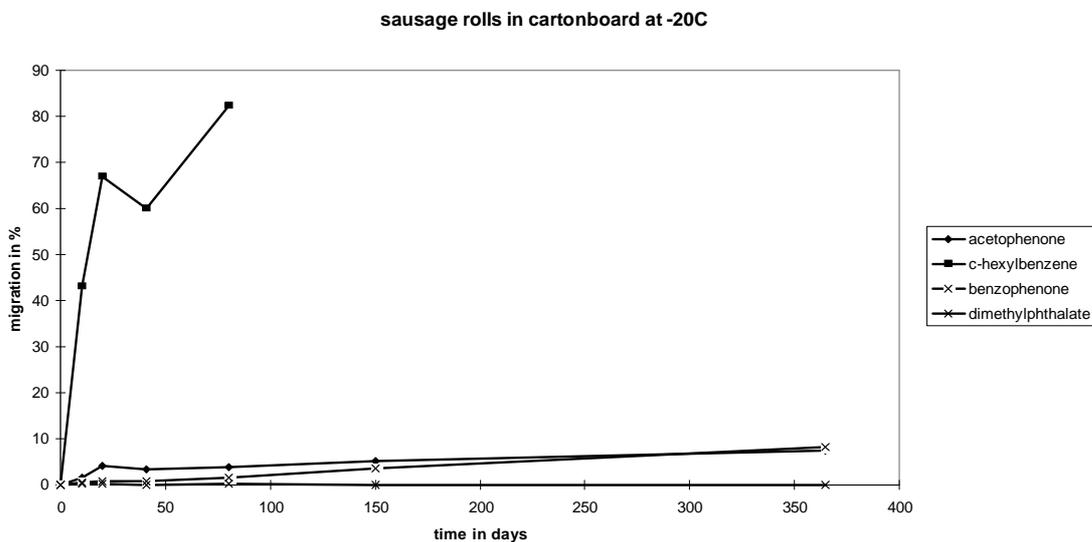
by mistake, instead of with internal standard, and the 1 year exposure was not repeated. The next fastest migrant was the other non-polar hydrocarbon, limonene, which migrated up to 25%. The more polar acetophenone, benzophenone and dimethylphthalate migrated to much lower levels (<5%).

Figure 2. Migration of model substances from P/B into ice cream [Bradley et al., 2007]



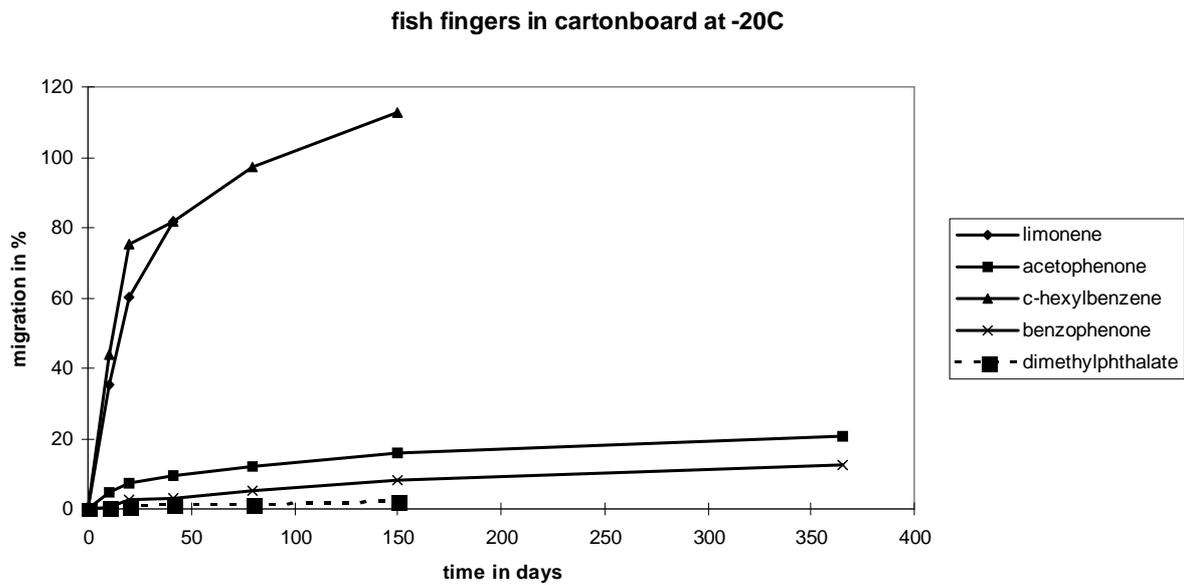
Sausage rolls: Again the non-polar cyclohexylbenzene had the fastest migration, rising rapidly to approximately 70% migration after 20 days. Acetophenone and benzophenone showed a slow but progressive migration to about 8% after one year. Dimethylphthalate stayed under 1%.

Figure 3. Migration of model substances from P/B into sausage rolls [Bradley et al., 2007]



Fish fingers: The two substances that showed the fastest migration into fish fingers were the non-polar hydrocarbons limonene and cyclohexylbenzene. They rose steeply to more than 80% migration after 40 days. The migration levels of acetophenone, benzophenone and dimethylphthalate followed the same pattern observed above, with levels after one year of acetophenone (21%) > benzophenone (12%) > dimethylphthalate (ca. 5%, extrapolated).

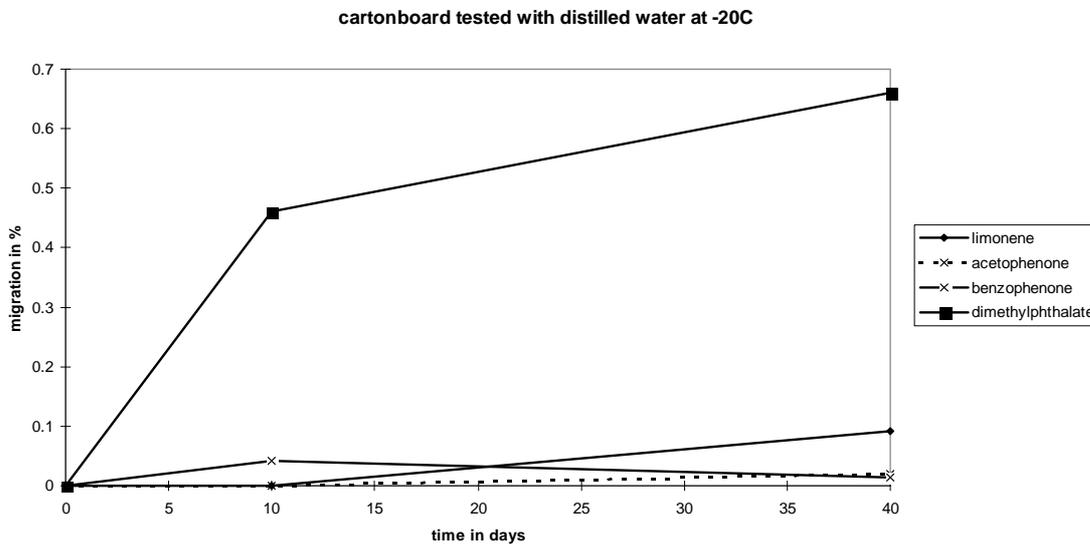
Figure 4. Migration of model substances from P/B into fish fingers [Bradley et al., 2007]



Migration into simulants

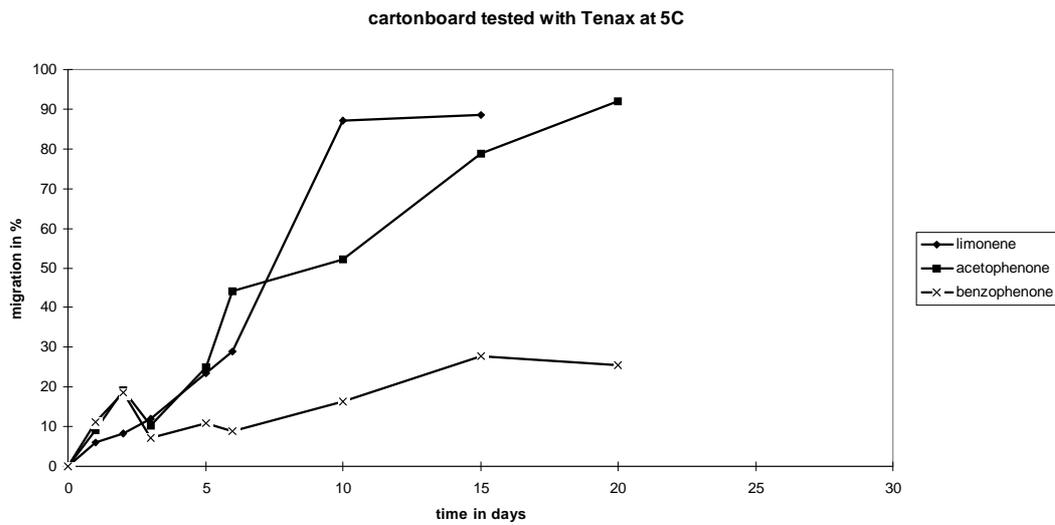
Ice: Migration levels were much lower with just 0.6% of dimethylphthalate migrating after 40 days and less than 0.1% of acetophenone, benzophenone and limonene transferring during the same period. The cartonboard was placed in contact with a pre-formed solid ice surface, which would have given less intimate contact than if liquid water had been used and then frozen *in situ*.

Figure 5. Migration of model substances from P/B into distilled water [Bradley et al., 2007]



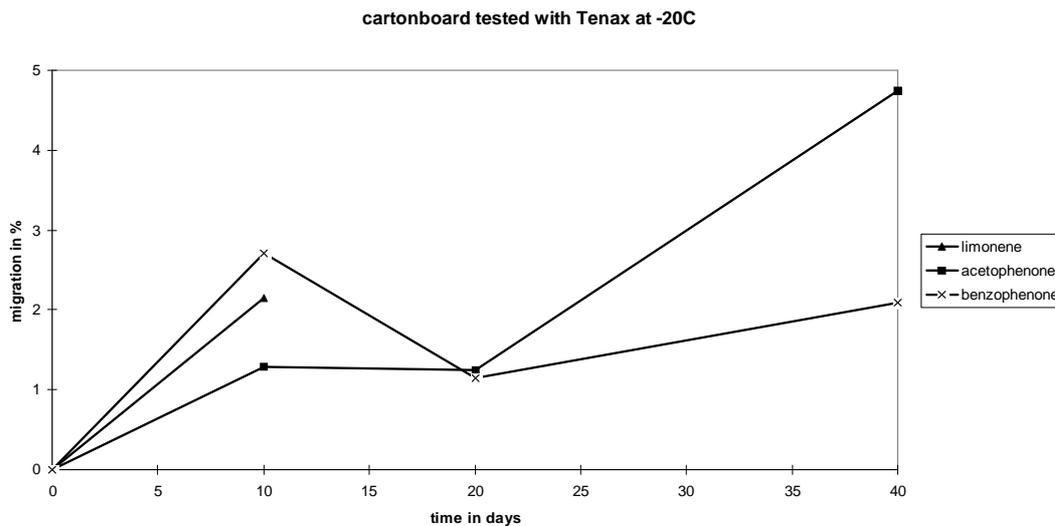
Tenax at 5°C: Migration of limonene, acetophenone and benzophenone into Tenax was also quite facile, at 90%, 50% and 20% respectively after 10 days at 5°C. Migration of acetophenone continued to rise and reached 90% after 20 days.

Figure 6. Migration of model substances from P/B into Tenax at 5°C [Bradley et al., 2007]



Tenax at -20°C. Migration values for Tenax at -20°C were 10-fold lower than for the same substances migrating at 5°C, with no migration exceeding 5% (acetophenone after 40 days).

Figure 7. Migration of model substances from P/B into Tenax at -20°C [Bradley et al., 2007]



The study authors [Bradley et al., 2001] discussed the results for benzophenone most, because it was for this substances that most information is available. Based on the benzophenone data, the best simulant and allied time/temperature conditions were considered to be Tenax (10 days at 5°C) for the cartonboard used to pack ice cream, sausage rolls or fish fingers, both for chilled storage at 5°C and for long-term frozen storage at -18°C.

The study authors then took the comparison further and listed the data available for all the model substances using the best simulant and allied time/temperature conditions selected from the benzophenone results alone, to see how good the fit was for the model substances as a whole. Although the data set was incomplete, it was concluded that using Tenax for 10 days at 5°C gave an acceptable approximation of migration into the frozen foods during long-term storage conditions.

Table 11. Migration levels (%age) from cartonboard into foods compared with migration into the best simulant for each food identified in [Bradley et al., 2001]

	LIM	ACP	CHB	BP	DMP
Ice cream at -20°C (365 days)	27 %	7 %	>48 %	5 %	2 %
Sausage roll at -20°C (365 days)	na	8 %	85 %	8 %	1 %
Fish fingers at -20°C (365 days)	100 %	21 %	100 %	12 %	5 %
Tenax at 5°C (10 days)	88 %	51 %	na %	16 %	na

Conclusions. These four studies demonstrate that migration of low molecular weight substances into frozen foods can be quite extensive. A test protocol for P/B using short-duration contact such as 10 days with the simulant Tenax at 5°C would be appropriate both for chilled and frozen storage of foods. This is because:- a) the standard test conditions of 10 days at 5°C are directly comparable to the storage life and temperature of most chilled foods; b) the test temperature of 5°C is higher than the storage temperature of frozen foods (-20°C) and this compensates for the fact that the simulant is used for only 10 days whereas the food itself may be stored frozen for up to one year.

Limitations. These results are skewed by the migration results of limonene (C₁₀H₁₆) and cyclohexylbenzene (C₁₂H₁₆). Being small, non-polar molecules with a relatively high vapour pressure compared to their molecular weight, they migrated extensively into frozen foods. They were incorporated into the P/B via a printing ink. If it could be assured that such low MW hydrocarbons were not present in P/B food packaging materials, the body of evidence indicates that a Correction Factor of 10 or more would be appropriate, applied either to the P/B composition measured by solvent extraction or by extensive migration into Tenax (e.g. 10 days at 40°C or 60°C).

Recommendations. None

FATTY CONTACT

Drawing parallels with EU regulations on plastics, there it is stated that food simulant D2 (vegetable oil) shall be used for foods which contain free fats at the surface [EU 10/2011]. However, the regulation further states that the result obtained with food simulant may in certain cases significantly overestimate migration into food (and) in these cases the result in food simulant is corrected by a reduction factor. Simulant D2 reduction factors are in the range of 2 to 5 and four typical food categories are in the Table below.

Table 12. Examples of simulant D2 reduction factors from Regulation 10/2011

Specific assignment of food simulants according to Food Category [EU 10/2011]	Simulant D2 reduction
Description of food	
Pastry, cakes, bread, dough and other bakers' wares, fresh; with fatty substances on the surface	X/3
Chocolate, chocolate-coated products, substitutes and products coated with substitutes	X/3
Fried potatoes, fritters and the like	X/5
Sandwiches, toasted bread pizza and the like containing any kind of foodstuff; with fatty substances on the surface	X/5

P/B materials are not very suitable for direct fatty contact - foods with free oils or fat on the surface - unless they are treated in some way, otherwise the oils and fats may soak into the P/B. Some notable exceptions may be fast-foods such as burgers or pizza where the main contact is with the bread bun or pizza base and any contact with the fatty food component is incidental, transient (short-term) and affecting only a small proportion (percentage) of the total packaging surface area. Other exceptions could be P/B bags and boxes used for transportation and short-term contact with cakes and pastries with fatty fillings or toppings.

A broad range of fluorochemicals may be used to impart oil and water barrier properties to paper and paperboard food packaging and for example several per- and poly-fluorinated chemicals are listed in BfR Recommendation 36. They are used in a limited number of special applications representing a low share of the total volumes of food contact paper and board. Three migration studies of fluorochemicals have been conducted over the past decade by the US-FDA.

FC#1. In early work, US-FDA researchers examined popcorn bags taken from the US market [Begley *et al.* 2005]. Bags selected for migration tests showed C6, C8, C10 and C12 fluorochemicals on the food-contact side as evidenced by a single-sided extraction test using 50/50 water/ethanol. The cleaned bags (11.7 dm² food-contact surface area) were filled with 40 g Miglyol (a synthetic triglyceride oil), folded, held closed with plastic clamps and microwaved for 2 min. It was reported that 1.4 mg/kg (4 mg/dm² paper) fluorotelomer had already migrated to the popcorn oil before microwaving. Microwaving caused an additional 2.1 mg/kg migration into the simulant Miglyol. Subsequent analysis of these bags for fluorotelomer additives suggests these particular test bags had the fluorotelomer coating/additive containing perfluorooctanoic acid primarily on the outside of the bags.

FC#2. The FDA researchers reported follow-up work on the migration of fluorochemical paper additives in 2008 [Begley *et al.* 2008]. Microwaved popcorn contained 3.2 mg/kg of fluorochemical after popping and butter contained 0.1 mg/kg after stored wrapped for 40 days at 4°C. Migration into

butter was higher than into other foods/simulants and this was attributed to it being a water-in-oil emulsion. Emulsions (80% oil and 20% water) containing non-ionic or ionic emulsifiers, oil (Miglyol) with and without added emulsifiers and two foods, butter and chocolate spread were studied. Migration into soy lecithin and polysorbate 60 emulsions (ionic and non-ionic emulsifiers, respectively) was similar in magnitude to butter whereas migration to a pure food oil was very low. The authors concluded that a pure food oil would significantly underestimate and, therefore, not be a good choice as a simulant for migration testing from fluorochemical- treated paper into some oily foods.

When tested using Tenax in contact with fluorochemicals-treated paper at 40°C for 32 days, no migration was detectable. The authors speculated that the lack of migration may be because these fluorochemicals have extremely low vapour pressures, which makes it difficult for them to transfer to Tenax [Begley *et al.* 2008].

FC#3. In their most recently-published work [Xu *et al.* 2013] the research group of Begley focussed on seven perfluorocarboxylic acids (PFCAs) in two commercially available food contact papers: a di-perfluoro-alkyloxy-amino-acid and a perfluoroalkyl phosphate surfactant. The migration from two commercial packages into five food simulants was also evaluated. All seven PFCAs were detected in the range of 0.7 to 2.2 mg/kg of paper, while three perfluoroalkyl sulphonates were under the LoD. Results from migration tests showed that migration depends on paper characteristics, time and food simulant. The percentage of migration after 10 days at 40°C ranged from 4.8% to 100% for the two papers and different food simulants.

Further comparison for all simulants shows that the migration into ethanol was greatest for all of the PFCAs except PFHxA (C₆) and PFHpA (C₇) into acetic acid. Generally, the migration into the aqueous simulants is greater than that into oily simulants except for the less water-soluble PFUnDA (C₁₁) and PFDoDA (C₁₂). The authors concluded that this study on commercially available FCMs is important in expanding the findings of Begley *et al.* (2008), that the addition of emulsifiers can dramatically influence the migration of fluorinated substances. In addition, this research has shown that the properties of the P/B, such as the substances used or the method used in treating the paper, may influence the migration of PFCAs into a variety of simulants. Therefore, while models may be used as a general guide for PFCA migration, testing of specific products is necessary to evaluate PFCA migration.

However, although the FDA researchers found that migration was nearly always highest into ethanol, the relevance of ethanol as a simulant for P/B treated with fluorochemicals has been questioned. In reporting on the Biosafepaper project, one sample contained perfluorinated chemicals (used to provide grease-resistance) and this treatment was said by the manufacturer to be incompatible with ethanol and so an alternative fat simulant would have to be used for testing [Bradley *et al.* 2008] No evidence was provided in the publication to support that statement by the manufacturer.

Conclusions. It is clear that Tenax cannot be considered a good simulant for P/B intended for contact with fatty foods. EN 15519:2007 describes the preparation of an organic solvent extract. of P/B. It states that “This European Standard specifies a test method for the assessment of substitute tests performed with volatile test media for the determination of migration from P/B intended to come into contact with fatty foodstuffs at all temperatures and for any period of time. At the time that this

European Standard was prepared, the EU directives for material coming into contact with food required use of isooctane or 95 % v/v aqueous ethanol.” This being so it is concluded that for contact with fatty foods the CEN test should be conducted. For solid foods that have fatty substances on the surface (as in the meaning of EU 10/2011) the Correction Factor as specified in the directive should be used to allow for the less complete contact the food makes compared with the (liquid) organic solvents. For specific applications where the fraction of the food that makes fatty contact with the P/B may be grossly underestimated by the fractional Correction Factors in 10/2011 (e.g. take-away pizza) a higher Correction Factor could be argued case-by-case.

Limitations. Only 3 studies and all on perfluorochemicals have been considered here. If CEPI members have information on migration into fatty foods and their simulants then they should kindly provide it.

Recommendations. None.

DRY FOODS (LONG-TERM AMBIENT STORAGE)

DF#1. Following-up the finding of migration levels in retail foods, Italian workers looked into the migration of DiPN from spiked paperboard into four dry foodstuffs that were; husked rice, wheat semolina pasta, egg pasta and maize flour [Bocacci *et al.* 1999]. The foods were said to had a high specific surface area but the particle size was not given. The egg pasta and wheat had 2.4% and 2.8% fat but the fat content of the other foods was not reported. Disks of paper (2 g/dm^2) were spiked with DiPN (liquid, not CCP microcapsules) and the food was placed in contact with the paper with a surface area : mass ratio of $20 \text{ dm}^2/\text{kg}$ in a sealed Petri dish. Migration tests were also conducted using a metal grid inserted between the paper and the foodstuff to study purely non-contact (gas-phase) migration. The tests were conducted for up to 60 days. The temperature was not given in the publication and it is assumed here that it was room temperature.

Migration was quite rapid with detectable migration after 3 days and with extensive migration (4 to 53% transfer, depending on the food) after 60 days of contact. The authors claimed that migration into the foodstuffs did not occur if the DiPN content was below 20 mg/kg in the paper and suggested that this was a migration threshold level [Bocacci *et al.* 1999]. This claim of a 'no migration' threshold has been repeated by others but in fact there is no evidence of a discontinuous composition-migration relationship. Rather, it was simply the case that the limit of determination for food analysis was $50 \mu\text{g/kg}$ (which for the 7 DiPN isomers monitored would be $<10 \mu\text{g/kg}$ for each isomer in food) and migration was not zero but simply not detectable when the DiPN content of the paper was low.

Table 13. Contact migration (percentage transfer of DiPN) from the spiked paper [Bocacci *et al.* 1999]

Day	20 mg/kg spiked paper				46 mg/kg spiked paper				146 mg/kg spiked paper			
	Semo	Pasta	Rice	Flour	Semo	Pasta	Rice	Flour	Semo	Pasta	Rice	Flour
3	0%	0%	16%	10%	3%	4%	7%	7%	4%	4%	4%	6%
9	0%	0%	17%	11%	3%	4%	8%	5%	5%	5%	6%	7%
15	0%	0%	16%	17%	3%	5%	8%	5%	4%	5%	5%	7%
24	0%	0%	18%	15%	3%	5%	10%	21%	4%	6%	6%	17%
40	0%	0%	18%	17%	4%	6%	14%	50%	4%	6%	10%	31%
60	0%	0%	18%	21%	5%	8%	14%	53%	4%	6%	13%	46%

Regarding the non-contact migration tests where a metal grid was used to keep an air gap between the P/B and the food, the authors concluded that this process did not depend on the characteristics of the foodstuff (i.e. specific surface area and lipid content) in that all the foodstuffs showed similar migration patterns and low DiPN concentrations (less than 0.5 mg/kg) when an air space over the P/B existed.[Bocacci *et al.* 1999].

Table 14. Non-contact migration (gas-phase only) [Boccacci *et al.* 1999]

Day	146 mg/kg spiked paper			
	Semo	Pasta	Rice	Flour
3	3%	4%	5%	4%
9	3%	7%	7%	4%
15	4%	7%	7%	7%
24	5%	8%	7%	7%
40	7%	9%	7%	8%
60	7%	8%	7%	7%

DF#2. For a study of phthalates in P/B and their migration into Tenax and sugar, P/B was obtained from packaging manufacturers [Aurela *et al.* 1999]. Eighteen out of the 29 samples studied had phthalate contents exceeding 5 mg/kg. The most common phthalates were DEHP and DBP. In most cases the origin of the phthalates was assumed to be the offset printing applied to the samples but for two of the highest the source was the adhesive used to make the paper stock into bags. DBP and DiBP were determined in sugar (white beet sugar, grain size 0.35 ± 0.41 mm) before and after packaging. Migration ranged from 57 to 74% of the original content in the packaging after 4 months storage at room temperature. The sugar packages were also tested using Tenax as a food stimulant. The results indicated that 69-91% of the original content of the phthalates migrated into Tenax after 10 days at 40°C.

Paper and board consists mainly of fibres and fillers. The cellulose fibres have repeat units of glucose residues and are very hydrophilic but are largely non-ionic. Sugar is also polar. It is a polyol with the simple empirical formula $C_6H_{12}O_6$ and the closest simple analogy would be methanol (CH_3OH) or ethylene glycol (CH_2OH)₂. DBP and DiBP ($C_{16}O_4H_{22}$) are not completely non-polar with a log Pow of ca. 4.6 and are freely soluble in alcohols. The weight of the paper bag was 8.7 g whereas the weight of the packed sugar was more than 100-times greater at 1000 g. Given enough time, the phthalates clearly partitioned extensively from the P/B into the sugar. Although not originating from the paper (being in the glue and/or the ink, and possibly subject to set-off too) the phthalate would equilibrate amongst the packaging components (paper, ink layer, glue layer) over time and so this publication is considered relevant.

DF#3. In Industry-sponsored work to develop rapid methods of test for P/B, Summerfield and Cooper [2001] followed the migration of DiBP, DBP and DiPN from three P/B samples into sugar, rice, cake, pastry, flour and Tenax. When stored at 20°C for 6 months, the migration of DiBP, DBP and DiPN into flour, rice and sugar was in the range of 5 to 41% and the average transfer rate was 19%. Quick tests were developed using Tenax under accelerated conditions of 4 h at 80°C to cover room temperature storage with dry foods and fatty dry foods.

DF#4. In a report that summarised the large body of work by project partners in the P/B WorkPackage of the 'Recyclability' EU project (coordinated by the Fraunhofer Institute) some general observations were made on migration kinetics and thermodynamics of substances in- and from P/B

[Castle 2004]. P/B samples that were supplied by the Industry were spiked with an agreed set of model compounds. The following findings and conclusions are drawn from that report.

Concerning the partitioning of the substances between the P/B and the gas phase. As expected because the boiling point of many of the substances was above 200°C, they partitioned strongly into the condensed phase of the P/B. The equilibrium concentration in the headspace gas phase depended on the boiling point of the substances and the test temperature. There were no significant differences noted between the different P/B samples tested. That is, different compositions of the P/B samples made no significant difference in the P/B - air distribution coefficients. This means that after allowing for the grammage of the samples (mass per unit area) they behaved essentially identically. The shape of the adsorption isotherms was classical. An important finding was that the linear range of the adsorption isotherms covered the range of concentration of migrants in P/B that occur in practice. In this linear range, the value of the equilibrium coefficient between board and air, $K_{B/A}$, is approximately constant [Castle 2004].

Concerning the migration kinetics. All the migration tests indicated just how rapid migration is from the P/B materials. The kinetics depended on the grammage of the P/B sample – tissue paper for example equilibrated very fast – and also on the nature (molecular size and volatility) of the substance. The rate of migration into Tenax was nearly always greater than into the dry foodstuffs cookies, flour, milk powder, noodles, salt, semolina, soup powder, sugar and icing sugar, when tested under the same conditions. Similarly, the final equilibrium partitioning concentration between impregnated board and Tenax simulant, $K_{B/T}$ was always at least one-order of magnitude less than the corresponding partitioning between board and foodstuff, $K_{B/F}$, indicating again that Tenax is a good simulant insofar as it is more severe than food [Castle 2004].

Effect of paper/board composition. There was no significant difference between the migration properties of P/B samples with different compositions. So, for example, the rate and extent of migration from clay coated board was the same as from the uncoated board [Castle 2004].

Effect of temperature on partition coefficients. The equilibrium partition coefficient of substances between P/B and Tenax was almost independent of temperature but there was evidence that it decreased slightly with increasing temperature e.g. at 60°C compared to 23°C. This means that the substances had a slightly greater affinity for the board relative to Tenax, at the higher temp. Two possible explanations were proposed for this. First, that hemicellulose polymers pass their glass transition temp at 40-50°C and melt, so increasing the adsorption affinity of the board. Second, that the water adsorbed on the fibre surface evaporates as the temperature was raised, changing the polarity of the surface and leading to an increase in adsorption affinity. The experimental evidence did not allow any firm decision to be made between these two possibilities. Given that sorption isotherms and migration behaviour seemed to be insensitive to the composition of the P/B samples, the first explanation seems less likely. The second explanation is consistent with the general every-day observation that P/B (especially 'brown' grades) release a faint smell when wetted – desorption due to wetting [Castle 2004]. See also DF#13 on the effect of relative humidity on benzophenone migration.

Different migration behaviour of DiPN. 'Native' DiPN (i.e. that intrinsic to a recycled board) displayed kinetic behaviour that differed from DiPN that was incorporated into samples by solvent impregnation. This is the same phenomenon as reported by Bradley *et al.* 2001 in FF#4. The difference in behaviour was interpreted as the 'native' DiPN being present mainly as the encapsulated form, as microcapsules from the recycling of carbonless copy papers. For its intended use in

carbonless copy papers, the capsule wall needs to be essentially impermeable to the capsule contents. Undamaged capsules of the DiPN can survive the recycling process [Castle 2004]. So encapsulated DiPN does not migrate (or migration is extremely low and so not measurable) whereas DiPN in the free form (i.e. released from broken capsules or spiked-in for model experiments) can migrate. This behaviour is unique to the potential contaminants PCBs (polychlorinated biphenyls), DiPN and HTP (hydrogenated terphenyls), which can arise from pressure-sensitive carbonless copy papers.

Applicability of Tenax as a simulant. In comparisons between Tenax and foodstuffs the partition coefficients between paper or board and Tenax were always approximately one order of magnitude smaller than the partition coefficients between paper or board and real foodstuffs. From these results the study authors concluded that Tenax is a suitable food simulant for testing migration from paper or board samples into foodstuffs - fulfilling the general safety requirement that the migration determined using Tenax is always higher than into foodstuffs.

Correction factors. At equilibrium, migration levels were typically in the range 50-100% (worst-case, migration losses from the P/B) or in the range 20-100% (measured in the food/simulant). Migration to Tenax was higher than to foods, with Correction Factors of 1-5. However, migration into food was also simply a factor of 1-5 less than the initial content in the P/B. Thus, for intimate contact with dry foods/simulants (e.g. powdered) at room temperature and above, the authors concluded that there seems little point in determining migration to a simulant and then applying a reduction factor. Determining the initial content and applying a similar factor or modelling is both more straightforward and quicker [Castle 2004].

But for contact temperatures at ca. 40°C and higher and for long-term storage at ambient temperature, in the authors view [Castle 2004] there was no strong need to apply kinetic migration models since the migration level is in the first place determined not by kinetics but by thermodynamics and most of the substances studied reached equilibrium in foods and in Tenax.

DF#5. Following up their earlier work, Italian workers reported on the contamination of dry foods with trimethyldiphenylmethanes (TMPMs) by migration from recycled P/B packaging [Sturaro *et al.* 2006]. Being essentially a small survey of retail foods with little compositional information provided, the results have limited value in the derivation of Correction Factors but they do serve to highlight the problems of deriving any conclusions that can hold generally for different classes of potential migrants.

The authors commented that the main solvents for the dyes in CCPs are hydrogenated terphenyls, diarylethanes, alkylnaphthalenes, cyclohexane, and dibutylphthalate. These are often diluted with odourless kerosene [CDC 2000]. Seven food samples, namely three types of egg pasta, three brands of rice and a brand of barley, all contained low levels of TMPMs and most of the foods also showed the presence of DiPN. But food such as sugar and salt did not show any evident contamination, confirming previous their findings. Our comment here is that this is not surprising given that the CCP solvents are non-polar hydrocarbons (diisopropylnaphthalenes = C₁₆H₁₈; hydrogenated terphenyls = C₁₈H₂₂; trimethyldiphenylmethanes = C₁₆H₂₀) whereas sugar is polar (a polyol) and is freely water-soluble (even water miscible) and common salt NaCl is ionic and also is freely water soluble. So these hydrocarbon CCP solvents have virtually no affinity for polar food or food ingredients such as sugar and salt. But this is in contrast to the findings of [Aurela *et al.* 1999], where migration of DBP and

DiBP into sugar ranged from 57 to 74% of the original content in the packaging after 4 months storage at room temperature, because the phthalates are more polar than the CCP solvents.

DF#6. In kinetic migration studies funded by CEPI and aimed at deriving diffusion and partition coefficients for use in migration modelling, Zülch and Piringer [2010] presented some migration results into ambient-storage foods that are of value here. The P/B samples used were of food-grade quality, unprinted and supplied by members of the CEPI Food Contact group. The authors emphasised a characteristic behaviour of the migration from P/B as being the dependency on the polarity of the migrant in comparison with the property of the fibres (with their polar hydroxyl groups on the surface) and the food / simulant. The derivation of the one- and two-layer migration models is beyond the scope of the work here but some informative migration results were reported in this paper. Importantly, these were industry-supplied samples and the substances monitored were intrinsic to the P/B and not spiked-in. Migration of particularly DiBP into several dry foods from different papers, under long-term ambient storage conditions was rather extensive even after just 2 months (60 days) (Table).

Table 15. Migration of intrinsic substances into food stored for 60 days at 22°C [Zülch and Piringer 2010]

	60 days at 22°C	
Wheat flour	Abeitic acid	6.2 %
Sponge biscuits	Diisobutylphthalate	38 %
Chocolate	Diethylene glycol dibenzoate	4.4 %
Chocolate	Dipropylene glycol dibenzoate	4.7 %
Wheat flour	Diisobutylphthalate	31 %
Chocolate	Diisobutylphthalate	60 %
Egg pasta	Diisobutylphthalate	35 %
Sponge biscuits	Diisobutylphthalate	31 %
Puff pastry	Diisobutylphthalate	not tested
Tenax	Diisobutylphthalate	44 %

DF#7. In follow-up work published 3 years later, this research group revisited the migration data of [Zülch and Piringer 2010] to give a better explanation of their adoption of a two-layer migration model, particularly in case of low temperatures (<40°C) and migrants with high molecular weight. (>350 Da). The dataset used was scattered and not especially coherent but overall, data covering 16 hydrocarbons (aliphatic and aromatic) and 11 oxygenated molecules present in recycled P/B were used [Hauder *et al.*2013].

They concluded that vapour pressure rather than simple molecular weight (size) was a determining factor - although they are of course related. However, vapour pressure alone was an incomplete parameter since for example they found it necessary to postulate a stronger interaction between the oxygen-containing structure and also the oxygen-containing board surfaces via hydrogen bonding or other dipole interactions in order to explain and fit the migration data. So taking a specific example, they described how comparing benzophenone with di-2-ethylhexyl phthalate, the latter has a non-polar aliphatic chain with a low interaction with the fibre matrix. Therefore, despite of the higher vapour

pressure of benzophenone, its effective diffusion coefficient can be comparable with that of di-2-ethylhexyl phthalate.

They also concluded that in the two-layer model, spiking P/B samples with too high concentrations of test migrants leads to too high diffusion coefficients in the surface (B2) layer because the adsorption isotherm is linear only in a relative small range of low concentrations.

They also re-affirmed the importance of the humidity during contact with certain foodstuffs as it can influence the migration behaviour.

DF#8. A survey was reported on the occurrence of photo-initiators and amine synergists in cartonboard packaging on the German market and their migration into the packaged foodstuff [Jung *et al.* 2013]. Retail foods (n=310) packaged directly or indirectly in printed P/B were collected during 2008 to 2011 from the German market. Shelf-stable foods were stored at room temperature prior to analysis. Eight samples were additionally stored until the best before date was reached and then analysed. Four cartonboard samples for migration tests with Tenax were obtained directly from a manufacturer.

Table 16. Photoinitiators and amine synergists monitored by [Jung *et al.* 2013]

BP	benzophenone	MOBB	methyl-o-benzoylbenzoate (MOBB)
HBP	hydroxybenzophenone	EHDAB	2-ethylhexyl-4-(dimethylamino)-benzoate
MBP	4-methylbenzophenone	EDAB	ethyl-4-dimethylaminobenzoate
PBP	4-phenylbenzophenone	HCHPK	1-hydroxycyclohexylphenylketone
DMABP	4-dimethylaminobenzophenone	MK	4,4'-bis(dimethylamino) benzophenone (Michler's ketone)
		MEK	4,4'-bis(diethylamino) benzophenone (Michler's ethylketone)

For the retail samples, the mean surface to mass ratio was 34 dm²/kg which is similar to the findings of Anderson and Castle (2003) who found a similar distribution of the surface to mass ratio and a mean ratio of 30 dm²/kg for 143 cartonboard packages. The median value of the packaging mass to food mass ratio was 93 g/kg (a 1:11 w/w ratio) [Jung *et al.* 2013].

Storage of samples until the best before date showed that HCHPK, BP and MBP migrated very easily via the gas phase. In contrast, PBP and MEK migrated only very slowly or not in quantifiable amounts into the foodstuffs. These data revealed that high concentrations in the packaging material do not necessarily correlate with high transfer rates. A possible explanation for these substance-specific observations was proposed to be the vapour pressures of the photo-initiators and amine synergists which lay in the order EDAB > MBP > BP > DMABP > HCHPK > MOBB > MK > EHDAB > HBP > PBP >> MEK. This means that, for example, MBP is more volatile than MEK and thus migrates very easily via the gas phase, while MEK does not [Jung *et al.* 2013].

DF#9. A research cluster headed by Koni Grob studied the saturated and aromatic mineral oil hydrocarbons in paperboard food packaging and estimated the long-term migration into foods [Lorenzini *et al.* 2010]. The paperboard from 102 products sampled in 2009 from the Italian and the Swiss markets were analysed for the potentially relevant MOSH and MOAH content.

They reasoned that migration is restricted to components of sufficient volatility to evaporate from the packaging material and recondense in the food. The range of hydrocarbons potentially migrating into dry food was investigated, focusing on the first step of the transfer process, i.e. evaporation: the loss of hydrocarbons from paperboard was monitored over 180 days. So for example, migration of MOSH into rice packed for 6 months reached 86% and exposure of the same type of board to indoor air during for 6 months caused a very similar percentage, 77% of the MOSH and MOAH fraction to evaporate. The same was true for DiPN.

The authors concluded that the migration process depends on the vapour pressure of the substances and their partitioning between the packaging material and the food,. Other factors come into play in so far as they may allow- or prevent evaporative losses (a box standing alone on a shelf or boxes stacked together) and if the vapour phase transfer is impeded or blocked by an internal bag. Since migration can be fast and is driven towards equilibrium with the food, and since the mass of the food exceeds that of the packaging material by a factor typically in the range of 5 to 25, the authors concluded that most of the hydrocarbons may end up in the food fairly independently of this partitioning coefficient.

DF#10. This same research cluster headed by Koni Grob then studied in depth, the migration kinetics of mineral oil hydrocarbons from recycled paperboard to two examples of dry foods [Lorenzini *et al.* 2013]. In the two food models tested, migration of mineral oil from recycled paperboard during 1 year at 20°C reached a MOSH concentrations of 20 mg/kg in muesli (45% of the potential) and 14 mg/kg in pasta (53% of the potential). The migration to food was fast with 5 mg/kg MOSH migrated into pasta after 1 week at 20°C and 10 mg/kg after 2 weeks. Storage at 30°C (said to be not uncommon in summer time in many warehouse storage facilities) caused a transfer of 15 mg/kg (57% of the potential) to the pasta in about 1 month. Migration at refrigerator temperature (4°C) was lower and there was selective migration of the more volatile hydrocarbons. Conversely, at 30°C migration was about 50% higher than at 20°C and the migrating species were skewed towards slightly higher molecular weight hydrocarbons.

The authors stated that the concentrations and transfer percentages were even higher if packs were kept inside corrugated cardboard boxes for the whole period. MOSH, MOAH and DiPN migration depends on how the pack is positioned since boxes standing alone on a shelf lose volatile hydrocarbons into the ambient air whereas such loss is negligible for products packed into larger cardboard boxes or stacked on pallets.

They concluded that data obtained from accelerated migrations tests (i.e. using Tenax 40°C and 60°C) are difficult to translate to long-term migration at ambient temperature, since the higher temperatures not only accelerates the hydrocarbons that do migrate at ambient temperature (mainly the fraction below carbon-number C24) but the higher temperatures also promotes migration of larger hydrocarbons that do not migrate significantly at ambient temperature [Lorenzini *et al.* 2013].

DF#11. In perhaps the most comprehensive investigations reported to date, Biedermann *et al.* studied migration from recycled paperboard into six test foods after 2, 4 and 9 months of storage at ambient temperature [Biedermann *et al.* 2013]. Strengths of this work are the wide-ranging nature of the chemicals monitored and the fact that the paper was commercially available and not spiked. There

was good AQA (analytical quality assurance) and any background levels in the foods were subtracted, and there was reasonable agreement in the amounts determined in the foods and the amounts lost from the paperboard (mass-balance). Limitations for interpretation here (not weaknesses *per se*) are that no comparable tests were conducted using simulants (it will be assumed that simulants would have given 100% migration) and most crucially, the test packs were separately overwrapped in aluminium foil to force migration inwards to the food and present a worst-case. The substances monitored were MOSH, MOAH, ink photoinitiators, phthalates, and DiPN. Several packaging formats were studied including the use of internal barrier layers (outside the scope of discussion here) but the food was also packed directly in contact with the board.

The authors reported that after 9 months storage at ambient temperature, foods directly packed in a P/B box absorbed on average 69% of MOSH (maximum 81%) and 50% of MOAH < n-C24 from the paperboard. More than half of the migration occurred in the first 2 months. The differences between the various foods was small, mostly less than a factor of 2 between the extremes. Migration seemed to be influenced more by the porosity of the food than by the fat content so, for instance, MOSH migration into oatmeal was markedly higher than into fatty biscuits (they were chocolate-coated). Nine photoinitiators were detected in the paperboard, of which eight migrated into the packed food at up to 24 %. Several plasticisers were present in the recycled paperboard, but only butyl phthalates showed significant migration. After 9 months, up to 40% of DiBP and 20% of DBP migrated into the food. See the Table below.

For photoinitiators, benzophenone was present at the highest levels in the food with a 10–24% transfer of the content in the paperboard. More than 20% of 4-methylbenzophenone migrated into the biscuits and oatmeal. Noodles were the exception: they contained only 6 µg/kg of benzophenone (maximum concentration 12 µg/kg). For methyl-2-benzoylbenzoate, migration still corresponded to 14% (biscuits, 8 µg/kg). 2-Isopropyl-9H-thioxanthene-9-one, with a boiling point of 399°C, migrated up to 8 µg/kg (polenta), at a rate similar to benzophenone, which according to the authors shows that gas-phase transfer cannot be predicted on the basis of boiling point and gas chromatographic retention alone.

Table 17. Migration into foods packed directly in recycled paperboard and stored at ambient temperature for up to 9 months overwrapped with aluminium foil [Biedermann *et al.* 2013]

	months	MOSH	MOAH	DiPN	DiBP	DBP
Biscuits	2	41 %	33 %	37 %	41 %	50 %
	4	56 %	45 %	46 %	39 %	64 %
	9	69 %	58 %	49 %	100 %	100 %
Polenta	2	46 %	32 %	28 %	49 %	33 %
	4	60 %	41 %	39 %	58 %	38 %
	9	77 %	53 %	41 %	80 %	71 %
Noodles	2	22 %	17 %	15 %	22 %	0 %
	4	39 %	30 %	29 %	22 %	22 %
	9	66 %	50 %	37 %	33 %	29 %
Rice	2	42 %	34 %	33 %	13 %	11 %
	4	65 %	48 %	44 %	13 %	11 %
	9	70 %	49 %	43 %	13 %	11 %
Breadcrumbs	2	35 %	26 %	26 %	28 %	22 %
	4	43 %	30 %	26 %	33 %	22 %
	9	49 %	35 %	26 %	49 %	29 %
Oatmeal	2	57 %	42 %	37 %	24 %	109 %
	4	71 %	53 %	48 %	49 %	44 %
	9	81 %	56 %	46 %	65 %	58 %

* Table is adapted from Biederman *et al.* 2013

DF#12. In the most recent publication from the Zurich control lab, migration testing for long-term storage at ambient temperature with Tenax was applied to a recycled paperboard [Zurfluh *et al.* 2013]. Test conditions were 10 days at 60°C (taken from the Plastics Regulation 10/2011) as well as the 'old' test conditions of 10 days at 40°C. The results were compared with the migration into six dry foods; biscuits (18% fat), polenta, noodles, rice, breadcrumbs and oatmeal, stored for up to 9 months. The packs were wrapped into aluminium foil to simulate storage in transport boxes where evaporation of volatile substances outwards into the atmosphere is virtually blocked.

The Table below summarises and then recalculates the data from [Zurfluh *et al.* 2013]. The first part gives the mass of MOSH and MOAH (range and average) migrated into the six food types after 9 months ambient storage along with the migration results for Tenax. The second part of the Table takes these values and calculates the corresponding Correction Factors (CF) as the range and the average of the ratio of migration into Tenax and foods (Tenax/Food). A CF value greater than 1 means that Tenax under the applied t/T conditions, overestimates migration into food.

Table 18. MOSH and MOAH migration from simulation for 10 days at 40°C or 60°C compared with migration into packs with six foods stored for 9 months. Results adapted from Zurfluh et al. 2013.

Migration values	Foods Range	Foods Average	Tenax 10 days at 40°C	Tenax 10 days at 60°C
MOSH	3.2 - 5.2 mg	4.3 mg	7.6 mg	9.0 mg
MOAH	0.59 - 0.98 mg	0.8 mg	1.7 mg	1.4 mg

CF values	Range CF Tenax 10 days at 40°C	Av. CF Tenax 10 days at 40°C	Range CF Tenax 10 days at 60°C	Av. CF Tenax 10 days at 60°C
MOSH	1.46 - 2.38	1.77	1.73 - 2.81	2.10
MOAH	1.73 - 2.88	2.13	1.43 - 2.37	1.75

Simulation at 40°C overestimated the maximum migration of the hydrocarbons into foods with CFs (rounded to one-decimal place) in the range of 1.5 to 2.9 and averages of 1.8 and 2.1 for MOSH and MOAH respectively. Simulation at 60°C overestimated the maximum migration of the hydrocarbons into foods with CFs in the range of 1.4 to 2.8 and averages of 2.1 and 1.8 for MOSH and MOAH respectively. Two main causes for this overestimation were suggested by the authors to be: (i) at 60°C, migration reached beyond n-C35, whereas it ends at about n-C24 in reality. (ii) Tenax is a far stronger adsorbent than foods, resulting in almost complete extraction. The authors concluded further that, even the 9-month migration results are over-estimated.

It is noteworthy however that because the migration into foods was extensive, the numerical values of CF are compressed. Migration of the low molecular weight fractions of MOSH and MOAH (i.e. that \leq C24) into Tenax were in all cases $>85\%$ of the total content in the P/B. The corresponding percentages for foods can be calculated from the respective CFs. In such cases, for migration into dry foods that predominantly or exclusively occurs through the gas phase, where the packs are wrapped with aluminium foil to blocked any possible evaporation of volatile substances outwards into the atmosphere, and where the migration at the end of the product shelf life is the end result to be simulated, rather than when the product would typically be eaten, then almost by default the CF values will become compressed and approach unity.

DF#13. Paper and board is an inhomogeneous fibrous network consisting mainly of cellulose, hemicelluloses and lignin. Also, various additives, fillers and coatings may be added during production. The cellulose fibres consist of repeat units of glucose residues and are therefore very hydrophilic and mainly non-ionic. The lignin consists of a network of repeated phenolic ring units with both hydrophilic and hydrophobic domains, both of which are anionic in nature. This gives a total negative charge to the surface of paperboard, which will retain positively charged migrants and also migrants containing aromatic rings, because of pi-electron interaction with the lignin surface [Castle 2004]. This being so it may be anticipated that the interactions of substances with P/B and their subsequent migration, may be influenced by the water content of the P/B.

Researchers at the Danish National Food Institute studied the effect of relative humidity on the migration of benzophenone from P/B into Tenax [Barnkob and Petersen 2013]. The paperboard used was 270 gsm and it was printed. The ink was not described further. The benzophenone was present at 1 mg/dm² from a uv-cured over-varnish. The varnish was at 9 gsm and was uv-cured but otherwise the nature of the varnish (a polymeric film) was not considered further in the migration modelling. The P/B was used 9 months after printing and it was assumed that the benzophenone had fully

equilibrated, but the location - the partitioning of the benzophenone between the P/B, the ink and the varnish - was not established. After 30 days at 34°C, the migration of benzophenone was 4.8 times higher at a RH (relative humidity) of ca. 67%, and 7.3 times higher at a RH of > 73%, compared with migration at RH of ca. 44 %. Both the diffusion coefficient and the partition coefficient, between paperboard and Tenax, decreasing with increasing relative humidity. The researchers measured the moisture sorption isotherms relating the equilibrium moisture content of the P/B but they did not consider the possible effects of RH on the Tenax. Tenax is considered to have a low affinity for water and it is generally considered that its sorption capacity (sorption of chemicals from gas streams which was and is the main scientific use of Tenax) is largely unaffected by moisture.

The findings relate to only one sample of printed and varnished P/B and only for benzophenone and only for Tenax at 34°C. Clearly with food contact the RH would be moderated if not controlled by the water activity of the packed food. The authors also note that they had found reference to only one earlier study of the effect of RH on migration from P/B which measured six substances, including benzophenone, migrating into Tenax at 20% RH and 90% RH at room temperature, but that earlier study did not find any conclusive evidence for an effect.

Nonetheless, if experiments confirm the general validity of this finding [Barnkob and Petersen 2013] then the numerical relationship between migration into foods and Tenax could be further complicated by RH effects.

Discussion on migration into dry foods (long-term ambient storage)

The CEPI / CITPA estimate is that fully 50% of their P/B used in direct contact, is primarily used with dry foods [CEPI and CITPA 2012] and so these applications merit a fuller discussion than afforded the other food contact categories dealt with here.

For migration to occur into truly dry foods, where there is no contact of the packaging with a liquid food phase and only marginal point contact, it is generally accepted that a vaporisation step must occur whereby the substance diffuses [Biran *et al.* 1979; Schwöpe *et al.* 1988; Schwöpe *et al.* 1994] across the gap between the pack surface and the solid food surface. To aid understanding, and building on the work of others, the migration process was broken down into five steps by [Castle 2004] who reasoned that the nature of the chemicals that migrate and the extent of their migration, is controlled by a combination of all five steps.

Step 0. Composition of the packaging The substance must of course be present (as a paper-making chemical or as a contaminant) in the packaging material. The propensity to migrate could be proportional to the concentration in the paper or board.

Step 1. Mobility in the packaging material The substance must be mobile enough to diffuse through the packaging material to appear at the food contact surface. The rate of diffusion will determine how rapidly the surface is replenished with migrant. Paper and board are rather open and porous structures and this step is not likely to be rate-limiting.

Step 2. Volatilisation The substance must have a discernible vapour pressure under the conditions (temperature) of use of the pack. It must volatilise into the interspatial gap between the solid (rigid) food surface or food particles and the pack surface.

Step 3. Absorption onto the food surface The substance must have an affinity for the food surface causing it to condense from the vapour phase, onto and into the food.

Step 4. Mobility in the food The substance must be mobile enough in the food matrix to diffuse away from the surface into the bulk of the food. This is necessary to avoid a concentration build-up at the food surface which would slow subsequent migration.

These ideas have been echoed by others, most recently including [Zurfluh *et al.* 2013] who stated that “Migration into dry foods predominantly or exclusively occurs through the gas phase, which comprises three steps: evaporation (partitioning with air), diffusion through air and recondensation/absorption into the food. The proportion of the migrated substance depended on the nature of the paperboard, the fat content of the food as well as the chemical nature and volatility of the migrant. The highest migration was observed for foods with the highest fat content”. Zurfluh *et al.* 2013 were concerned mainly with migration of the very non-polar mineral hydrocarbons. The findings of [Aurela *et al.* 1999] on extensive migration of the DBP and DiBP from P/B to sugar, can expand this conclusion to encompass moderately non-polar substances such as the phthalate esters.

This returns us to the point made in the introductory sections. For the strict derivation of Correction Factors, to relate migration values between simulant data and food data, then the migration test conditions investigated should be defined to result in the worst foreseeable migration, i.e. the highest concentration of the migrant in the foodstuff.

The view of the control authorities is that simulation is supposed to somewhat exceed real migration to provide the safety margin covering extraordinary samples or configurations. Food control is the traditional tool for providing food safety in many areas including FCMs, direct food additives, residues, contaminants etc. This involves measuring against limit values based on deterministic, conservative (max) assumptions. In the case of FCM this means control using migration values rather than making an estimate of consumer exposure. The policy is that consumers should be able to eat as much as they like (and even more than is physiologically possible, long-term) with all the food eaten at the end of its shelf-life after contact under the most unfavourable circumstances of temperature, SA:M ratio etc.

Conclusions. There is ample evidence that, unless the composition of P/B can be controlled to effectively exclude substances that have a significant vapour pressure, low affinity for the P/B matrix and higher affinity for the packed food, then no Correction Factor (i.e. CF=1) can be supported for P/B intended for packing dry foods for long-term ambient storage. The extensive transfer that can occur both into food and the food simulant Tenax, effectively compresses the CFs into the region of 1-2. This would be both unreliable - if each and every packed food must comply with a migration limit value - and would also be of limited usefulness anyway.

Limitations. None.

Recommendation. The full utility of Correction Factor (in their fullest sense) will not be realised unless the control of P/B moves away from simplistic migration limit values and uses instead, exposure concepts to better estimate, describe and control consumer exposure and therefore safety.

Recommendation. Since certain classes of substances can migrate extensively, if they cannot be excluded from P/B (see above) then it is recommended that the migration modelling work conducted at Fabes be explored with a view to deriving some substance-specific CFs for important combinations of substance (class) / food (category) / t/T (conditions).

BAKING PAPERS

BP#1. The Scientific Committee on Food evaluated the composition and migration data for butyltins from baking papers [SCF 2002]. The use of organotin compounds, as catalysts for making the silicones used to provide grease-resistance, has long been discontinued for the European market but the findings remain informative. The SCF concluded that, considering the harsh contact conditions during baking for e.g. cookies - with intimate contact between a hot fatty food and the baking paper - the propensity of different organotin compounds to migrate is about equal. From the results presented it can be concluded that migration was virtually complete since the concentration ratio of migration/composition was in the range of 1-2% which would equate to about total migration from baking paper in mass ratio contact of 1-2 gram of paper in contact with 100 g cookies which seems plausible for a thin (low grammage) baking paper.

BP#2. In their investigations of the migration of per- and polyfluorinated compounds from paper based packaging into real food and food simulants, researcher at the Fraunhofer Institute reported that migration into polar and non-polar liquids such as methanol and olive oil, over- or under- estimated the migration into foods such as butter and that Tenax was a suitable food simulant especially at elevated temperatures. Specifically, at baking temperatures, migration values of certain volatile fluorotelomer alcohols (FTOHs) indicated clearly that the chemical used to treat the P/B decomposed to generate further migration. This net production of FTOHs was confirmed in kinetic studies at 60, 120 and 220 °C [Müller *et al.* 2012].

Conclusions. Taking these findings into consideration, it is concluded that baking papers should be tested using the CEN test for fatty contact (ethanol and/or isooctane extraction) and should also be tested at high temperatures, using Tenax, as a check against the formation and subsequent migration of thermal decomposition products at baking temperatures. Concerning the choice between ethanol or isooctane, ethanol is not likely to be appropriate if the baking paper is treated with fluorochemicals. No CFs can be recommended for either simulant test so CF should be 1.

Limitations. It is possible that during the use of baking papers, volatile substances may be lost to the oven and not persist in the food, and that non-volatile fat-soluble substances may dissolve in any cooked-out fat and so be discarded rather than consumed with the food. These attenuation arguments are not allowed for here and it is recommended that they should not be used.

Recommendations. None.

HOT AND COLD WATER INCLUDING FILTERS

P/B finds only very limited applications in contact with liquids or foods that have a free aqueous phase on the surface. Applications include tea bags and filter papers. Two European Standards describes the preparation of a cold or a hot water extract for investigations of certain extractives in paper and board intended to come into contact with foodstuffs. These are EN 645:1993 (cold) and EN 647:1993 (hot) respectively. These CEN standards aim to achieve exhaustive extraction and they seem to be well suited for the purpose.

Conclusions. EN 645:1993 and EN 647:1993 should be used to test P/B intended for contact with cold- or hot- liquids respectively. No CF should be used (CF=1).

Limitations. P/B may be used in contact with foods that are not 'liquid' but are moist and so which may 'wet' the paper slightly. In those cases an exhaustive extraction by total immersion in cold or hot water can be expected to give migration/extraction that is too high. No body of data could be found to support any general CFs to fit these cases (although see the recommendation below).

Recommendations. The CEN standards call for extraction of 10 g of P/B using 200 mL of cold or hot water. Thee extract is subsequently analysed for the substance(s) of interest. If the grammage and the contact area : food mass ratio are known for the specific application(s) intended then this 10 g:200 mL ratio could be varied up or down to reflect better the actual application with foodstuffs.

INORGANIC SUBSTANCES

Potential inorganic substances migrating from P/B may be heavy metals (present as contaminants), metals from processing aids (e.g. aluminium from retention aids), inorganic pigments, chromium(VI) in baking papers, and the elements present in clay coatings [Castle *et al.* 1997; Damant and Castle 1999].

Virtually all studies have concentrated on the potential migration, indeed extraction, of heavy metals as potential contaminants of P/B and there are two CEN methods specifically for such testing purposes. These are EN 12497:2005 for the determination of mercury and EN 12498:2005 for the determination of cadmium and lead, in an aqueous extract of paper and board. The extraction procedures to be used are EN 645:1993 (preparation of a cold water extract) or EN 647:1993 (preparation of a hot water extract) depending on the intended use of the P/B for e.g. cold- or hot-water filtration.

For migration into solid foodstuffs, Parry and Aston measured the element content of virgin P/B (20 samples), recycled P/B (22 samples) and recovered feedstock (4 samples) for their content of 64 elements [Parry and Aston 2004]. Based on these results, ten foodstuffs packaged in recycled P/B were tested for migration. The packaging was irradiated in a nuclear reactor to produce radionuclides of the elements of interest and then placed in contact with the food. The use of neutron activation to form the radioisotopes in the packaging materials *in situ* allowed even low levels of migration of elements commonly found in foods (and therefore giving a high background level using other techniques) to be measured. The migration test applied for food types with a long shelf life (salt, rice, oats, tea, cannelloni, custard powder) was 90 days at 20°C with interim measurements being taken at 1, 8, 21, 30 and 60 days. For hot foods (pizza, French fries) the tests were for 2 hours at 100°C.

The only elements found to migrate in static tests were zinc and iron, at 4.8 and 2.6 µg/dm² respectively. For samples of rice, oats and custard powder (each dry foods) and for French fries (fatty) the migration was less than 1% of the content in the P&B. When Tenax was tested as a food simulant, migration was again not detectable in the large majority of cases and never more than 1% transfer. One sample of fruit (mushroom) was tested. It was observed that the mushrooms spoiled after 8 days at ambient temperature, they went slimy, and that this gave rise to much higher migration of 12.5% of the zinc available in the recycled paper packaging.

It was considered possible that any potential for migration could be due to physical abrasion and the introduction of fibres into the food during agitation of the packaging. This was investigated by shaking the samples of powdered food for 30 min at the end of the 90-day migration test. This was carried out using a mechanical shaker with 600 oscillations/min, so the samples were shaken a total of 18,000 times. Shaking had no effect on migration, except in one case of custard powder where migration occurred at 0.25% transfer. It was therefore concluded that physical abrasion does not play an important factor in the migration process and any migration that occurs is no more significant than that found in the static tests [Parry and Aston 2004].

Conclusions. Considering the range of elements detected in P&B samples [Castle *et al.* 1997; Damant and Castle 1999] their migration into dry foods and fatty foods does not seem to be an issue of concern. With the exception of paper intended for e.g. filtration, P/B that is not protected by some form of barrier layer is not suitable for contact with liquids or with solid foods that have an external

aqueous liquid phase. This being the case, for P/B used for food packaging, it does not seem necessary to test for the migration of inorganics. Compositional limits may be desirable for GMP (Good Manufacturing Practice) reasons but migration testing and Correction Factors do not seem necessary.

Limitations. This conclusion does not apply to any inorganic substances that may be used in nano form.

Limitations. EN 646:2006 describes a method of test for the colour fastness of dyed paper and board. The word 'dye' infers an organic colourant but the test may also be used for P/B coloured with inorganic pigment particles. The simulants / extractants used in that test are not considered here, since the test is visual and does not give results in concentration units.

Recommendation. Given the recent re-evaluation and consequent lowering of health-based guidance values for certain elements of interest (Al, Pb, Cd) it would be sensible to re-examine the elemental composition of current P/B production samples to establish where they are in relation to new (and significantly lower) limits on certain metals.

Recommendation. Depending on the data on the elemental composition of current production P/B (see above) it may be desirable to evaluate whether the lack of an acidic extraction medium as a CEN standard is a weakness or not.

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GLOSSARY AND INDEX OF TERMS USED *

* Where used on two or more occasions and not spelled-out in the text

ACP	acetophenone
BP	benzophenone
CCP	carbonless copy papers
CEPI	Confederation of European Paper Industries
CF	Correction Factor
DBP	di-n-butyl phthalate
DEGDB	diethylene glycol dibenzoate
DEHA	di-2-ethylhexyl adipate
DEHP	di-(2-ethylhexyl) phthalate
DiBP	diisobutyl phthalate
DiPN	diisopropyl naphthalene
DMABP	4-dimethylaminobenzophenone
DMP	dimethyl phthalate
EDAB	ethyl-4-dimethylaminobenzoate
EHDAB	2-ethylhexyl-4-(dimethylamino)-benzoate
HBP	hydroxybenzophenone
LIM	limonene
LoD	limit of detection
Log Pow	log of the partition coefficient of the substance between octanol and water
MBP	4-methylbenzophenone
MEK	4,4'-bis(diethylamino) benzophenone (Michler's ethylketone)
MK	4,4'-bis(dimethylamino) benzophenone (Michler's ketone)
MOAH	mineral oil aromatic hydrocarbon
MOSH	mineral oil saturated hydrocarbon
MPPO	modified polyphenylene oxide
P/B	paper / board
PFCAs	perfluorocarboxylic acids
PPO	poly(2,6-diphenyl-p-phenylene oxide)
RH	relative humidity
t	time
T	temperature
US-FDA	United States Food & Drug Administration
VAN	vanillin