

1 **The use of Tenax[®] films to demonstrate the migration of chemical**
2 **contaminants from cardboard into dry food.**

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19 **The use of Tenax[®] films to demonstrate the migration of chemical**
20 **contaminants from cardboard into dry foodstuffs.**

21 Contaminants in food packaging are a challenge of our time since the packaging
22 material itself has been found to represent a source of food contamination
23 through the migration of substances from the packaging. Before first use,
24 packaging materials destined for the packaging of dry foods can be evaluated by
25 performing migration experiments with the simulant for dry foods: Tenax[®]. This
26 simulant is commercially available as a powder that is more difficult to handle
27 during the migration experiments. In this contribution a Tenax[®] film was
28 developed. The film can be applied to the cardboard surface to conduct the
29 migration test. After the migration is completed, the film can be easily extracted
30 to determine the concentration of the contaminants in the film. Finally, the
31 performance of the Tenax[®] film was compared to the conventional Tenax[®]
32 powder for the evaluation of 15 model migrants.

33 Keywords: Tenax[®]; film; food simulant; migration testing; food contact material

34

35

36 **Introduction**

37 Recent improvements in food production and distribution have led to an
38 increased sophistication of food packaging. Unfortunately, non-inert materials, like
39 paper and board, can be a direct source of chemical contaminants. Chemical partitioning
40 from the packaging into the food is known as migration. In 1999, Boccacci et al.
41 showed that diisopropylnaphtalene (DiPN) was transferred from cardboard to rice, pasta
42 and maize flour already after three days at ambient temperature (Boccacci et al. 1999).
43 Inevitably, several alerts for food contamination caused by migration have been
44 reported in the past. Examples are the presence of photo-initiator 2-
45 isopropylthioxanthenone (2-ITX) in instant baby milk and photo-initiator 4-
46 methylbenzophenone (4-MBP) in breakfast cereals, leading to the temporary
47 withdrawal of these products from the market (IBFAN 2005, EFSA 2009).

48 Therefore, a careful evaluation of food contact materials and their interactions
49 with food is needed to ensure consumers' safety. Evaluation of the migration of
50 chemical contaminants in food is necessary but challenging due to the complexity of the
51 matrix and the wide variety of foods that need to be analysed. If a food contact material
52 is not yet in contact with food, the contact material can be checked for compliance using
53 food simulants. According to the European Regulation No 10/2011, the official simulant
54 for dry food is poly(2,6-diphenylphenylene oxide), also known under its commercial
55 name Tenax[®] (Regulation (EU) N° 10/2011).

56 Numerous studies have been reported on the migration of contaminants from
57 FCM into Tenax[®]. A few examples are the migration of phthalates (Aurela et al. 1999),
58 diisopropylnaphthalene (Summerfield & Cooper 2001), dihydroabietic and abietic acids
59 (Ozaki et al. 2006), alkylbenzenes (Aurela et al. 2001) and fatty acid esters (Richter et

60 al. 2009). Furthermore, Cannelas et al. demonstrated the use of Tenax[®] in the
61 determination of non-intentionally added substances in acrylic adhesives (Cannelas et
62 al. 2012) and Zurfluh et al. studied the use of Tenax[®] for the analysis of mineral oil
63 saturated hydrocarbons (MOSH) from recycled paperboard (Zurfluh et al. 2013).
64 Furthermore, Triantafyllou (Triantafyllou et al. 2007), Bradley (Bradley et al. 2014) and
65 Nerín (Nerín et al. 2007a) demonstrated the use of Tenax[®] for a kinetic evaluation of
66 certain migrants.

67 Before use, Tenax[®] is usually cleaned multiple times by soxhlet and dried
68 afterwards. Next, Tenax[®] is applied on the food contact material surface for a certain
69 contact time and contact temperature. According to the internationally recognised
70 standard on overall migration from plastic materials and articles in contact with
71 foodstuffs, 4 gram of Tenax[®] per square decimetre of surface area of the test specimen
72 is required to cover the food contact surface sufficiently (EN 1186-13:2002).
73 Nevertheless, it was demonstrated that 1 gram of Tenax[®] is equally sufficient to cover a
74 0.15 dm² circular sample (Jakubowska et al. 2014). After the migration is completed,
75 Tenax[®] is removed from the food contact material surface and extracted. Extracts are
76 usually analysed using gas or liquid chromatography. The use of the Tenax[®] powder as
77 a simulant for dry foodstuffs is rather inconvenient since the Tenax[®] powder has to be
78 entirely collected in a recipient prior to contaminant extraction. Hence, the concept of a
79 practical Tenax[®] film was introduced.

80 In 2010, Alfeeli et al. developed thin Tenax[®] films as adsorbent material for
81 micro preconcentration applications (Alfeeli et al. 2011). Tenax[®] was dissolved in
82 dichloromethane and used to coat the embedded high-aspect-ratio three-dimensional
83 micro pillars of the micro preconcentrator at room temperature or at frozen conditions
84 (Alfeeli et al. 2011). Furthermore, Alfeeli et al. concluded that there was no difference

85 in the adsorption properties between the powder and the film form (Alfeeli et al. 2010).

86 Next, in 2013, Tenax[®]-coated silica nanoparticles were incorporated as an
87 adsorbent bed in silicon based micro-thermal preconcentrator chips (Akbar et al. 2013).

88 Tenax[®] was dissolved in dichloromethane and again evaporated to leave a thin film of
89 the polymer adsorbent on the cavity surfaces of the micro-thermal preconcentrator chips
90 (Akbar et al. 2013).

91 Recently, Tenax[®] was combined with the zeolite material ZSM-5 in order to
92 form a thin film microextraction device that was used as a novel alternative tool for
93 headspace volatile organic compound extraction and preconcentration (Goda et al.
94 2014). Tenax[®] was dissolved in chloroform and used to apply thin flat films via dip
95 coating. The coating took place at room temperature in ambient air (Goda et al. 2014).

96 In this contribution, Tenax[®] films were constructed by dissolution in
97 chlorinated solvents, followed by evaporation under air. The Tenax[®] films were
98 evaluated for their application in compliance testing of cardboard food contact
99 materials.

100

101 **Materials and methods**

102

103 ***Chemicals***

104 Neat certified standards of acetophenone (AP, 99%) benzophenone (BP, purity 99.9%),
105 benzyl butyl phthalate (BBP, 98%), dibutyl phthalate (DBP, 99%), dibutyl sebacate
106 (DBS, 97%), 2,6-diisopropylnaphthalene (DiPN, CPR), 2,2-dimethoxy-2-phenyl
107 acetophenone (DMPA, 99%), trans,trans-1,4-diphenyl-1,3-butadiene (DPBD, 98%),
108 2,6-di-tert-butylphenol (DTBP, 99%), 2-ethylhexyl-4-dimethylaminobenzoate (EDB ,
109 98%), 4-methylbenzophenone (MBP, 99.9%), methyl stearate (MS, 99%), naphthalene

110 (NPT, 99%), 4-phenylbenzophenone (PBZ, 99.8%) were purchased from Sigma-
111 Aldrich (Bornem, Belgium). 2-Isopropyl-9H-thioxanthen-9-one (ITX, 100%) was
112 supplied by Rahn (Zürich, Switzerland). Chemical structures are given in Figure 1.
113 Tenax[®] (60/80 Mesh) was also purchased from Sigma-Aldrich (Bornem, Belgium).
114 Acetonitrile, chloroform, dichloromethane and methanol were purchased from Biosolve
115 (Valkenswaard, Netherlands). All solvents were HPLC-grade.
116 All stock solutions were prepared in methanol at a concentration of 1 mg mL⁻¹ and
117 stored at -20°C for 6 months.

118 ***Development of the Tenax[®] film***

119 Prior to its use, Tenax[®] was cleaned by soxhlet extraction with acetonitrile for at least
120 6h, followed by drying in an oven (BINDER, Bohemia, NY). Next, Tenax[®] was
121 dissolved in chloroform or dichloromethane in concentrations ranging from 0.00625 up
122 to 0.05 g mL⁻¹. After filtration using a 0.45 µm PVDF filter (GRACE) and a 1-0.45 µm
123 glass fibre + PVDF prefilter (CHROMAFIL[®]), a fixed volume (2, 4 or 6 mL) was
124 brought into a glass recipient and the solvent was evaporated at room temperature in air.
125 A petri dish with a diameter of 58 mm was chosen as a recipient. To construct visibly
126 flat and smooth films, any form of air circulation such as fumehoods was avoided.

127 ***Set-up migration experiment***

128 Since a petri dish was chosen as a recipient, the Tenax[®] films could remain fixed on the
129 glass to perform migration testing using a previously described set-up by Jakubowska et
130 al. (Jakubowska et al. 2014). The Tenax[®] film was brought in contact with 0.15 dm² of
131 spiked blank circular cardboard sample. The cardboard was spiked on the side of the
132 cardboard intended for printing by homogeneously dotting 100 µL of a solution with a
133 concentration of 25 µg mL⁻¹ of the following contaminants: AP, BP, BBP, DBP, DBS,

134 DiPN, DMPA, DPBD, DTBP, EDB, ITX, MBP, MS, NPT and PBZ. A smaller petri
135 dish with a diameter of 52 mm was used to cover the sample and the complete set was
136 wrapped in aluminium foil. Migration conditions were according to European
137 Regulation No 10/2011 (Regulation (EU) No 10/2011) for long-term storage of dry
138 foodstuffs at room temperature, resulting in a migration of 10 days at 60°C. Afterwards,
139 both film and cardboard were analysed and the concentration of the contaminants was
140 determined.

141 Additionally, a conventional migration experiment with 1 gram Tenax[®] powder was
142 conducted by bringing spiked blank cardboard (100 µL of a AP, BP, BBP, DBP, DBS,
143 DiPN, DMPA, DPBD, DTBP, EDB, ITX, MBP, MS, NPT and PBZ solution with a
144 concentration of 25 µg mL⁻¹) in contact with Tenax[®]. Prior to its use, Tenax[®] was
145 cleaned by soxhlet extraction. After the migration (10 days at 60°C) was completed,
146 Tenax[®] and cardboard were analysed and the rate of migration was determined.

147

148 *Analysis of contaminants*

149 *Analysis of contaminants in the cardboard:*

150 Cardboards were cut into pieces and extracted using 20 mL acetonitrile in 100 mL
151 flasks for 24h at 70°C (Sanches-Silva et al. 2008). Analyses were carried out using GC-
152 MS. Quantification was done using a five point calibration curve ranging from 0.625 to
153 2.5 µg per cardboard circle, external calibration. The calibration curve was constructed
154 in the matrix. Hence, the cardboard was enriched with 100 µL of the contaminant
155 solution (0, 6.25, 12.55, 18.75 and 25 µg mL⁻¹). The enriched cardboards were also kept
156 in petri dishes for 10 days at 60°C to encounter the loss of substances due to
157 evaporation. Afterwards, cardboard samples were extracted prior to calibration.

158

159 *Analysis of contaminants in Tenax[®] film:*

160 Tenax[®] films were cut into pieces and extracted with 20 mL acetonitrile in 100 mL
161 flasks for 24h at 70°C. Analyses were carried out using GC-MS. Quantification was
162 done using a five point calibration curve ranging from 0.625 to 2.5 µg per film, external
163 calibration. The calibration curve was constructed in the matrix. Hence, after bringing 4
164 mL of a 0.05 g Tenax[®] mL⁻¹ in the petri dish, the 'calibration-film' was spiked with 100
165 µL of a contaminant solution (0, 6.25, 12.55, 18.75 and 25 µg mL⁻¹) before evaporating
166 into a Tenax[®] calibration-film. The calibration-films were also placed in the oven for 10
167 days at 60°C prior to extraction for calibration purposes.

168

169 *Analysis of contaminants in Tenax[®]*

170 After the migration experiments, the simulant Tenax[®] was analysed and the
171 concentration of the model migrants was determined. A previously optimised method
172 by Van Den Houwe et al. (Van Den Houwe et al. 2014) was used. Tenax[®] was extracted
173 twice with 20 mL of acetonitrile. Both extracts were combined and further diluted to 50
174 mL. For the quantification, a five point calibration curve in the matrix was constructed
175 using 1.0 g Tenax[®] spiked at a concentration ranging from 0.625 µg g⁻¹ up to 2.5 µg g⁻¹.
176 Calibration samples were also kept in the migration oven for 10 days at 60°C before
177 being extracted.

178 ***GC-MS analysis***

179 GC-MS analyses were carried out using a Agilent 7683 Automatic Liquid
180 sampler (Agilent Technologies, Palo Alto, USA). The GC-MS analyses were performed
181 on an Agilent 6890N gas chromatograph coupled to an Agilent 5973N single quad mass
182 selective detector. 2 µL was injected into the GC-MS system in split injection mode
183 (split ratio 3,0:1). A VF-5ms column (Factor four, Agilent, California, VS) of 30 m (Ø

184 0.25 mm and film thickness of 0.30 μm) was used. Helium was delivered as carrier gas
185 at a constant pressure of 14.65 psi with an initial flow of 1.3 mL min^{-1} .

186 The temperature gradient started at 80°C (held for 2 min) and rose with 20°C
187 min^{-1} to reach 290°C, which was held for 10 min. The total run time was 22.5 min.
188 Temperatures of injection port, ion source, quadrupole and interface were set at 160,
189 230, 150 and 280°C, respectively. For quantification of compounds, the mass
190 spectrometer was operated in selective ion monitoring (SIM) mode (100 ms dwell
191 times). Table 1 presents the specific m/z ratios and the retention times.

192 Instrument control and data acquisition were performed by Agilent software
193 (Enhance Chemstation 2004, Agilent).

194 **Results and discussion**

195 *Selection of contaminants*

196 The contaminants were selected based on a literature search on contaminants in paper
197 and board. It was found that photo-initiators and phthalates are one of the most common
198 groups of compounds present in paper and board food packaging (Aurela et al. 1999;
199 Jakubowska et al., 2014; Jung et al., 2013; Summerfield et al. 2001; Van Den Houwe et
200 al., 2014). Hence, two phthalates (BBP and DBP) and six photo-initiators (BP, DMPA,
201 EDB, ITX, MBP and PBZ) were selected. Futhermore, the following contaminants were
202 also frequently studied: AP, DBS, DiPN, DPBD, DTBP, MS and NPT (Aurela et al.,
203 2001; Bradely et al., 2014 and 2015; Isella et al., 2013; Nérin et al., 2007b; Poças et al.,
204 2011; Sanches Silva et al., 2006 and Triantafyllou et al., 2007).

205

206 *Optimisation of the production of the Tenax[®] film*

207 Among different organic solvents that were tested, Tenax[®] or poly(2,6-
208 diphenylphenylene oxide), was best dissolved in chloroform or dichloromethane

209 (DCM). Since it was practically not feasible to dissolve Tenax[®] at higher concentrations
210 than 0.05 gram per millilitre chloroform, solutions ranging from 0.00625 g mL⁻¹ to 0.05
211 g mL⁻¹ were used to construct films of 25, 50, 100, 150 and 200 mg by evaporating 4
212 mL of a Tenax[®] solution in a petri dish (Figure 2). In parallel, Tenax[®] films of 100, 200
213 and 300 mg were constructed by evaporating different volumes (2, 4 and 6 mL) of a
214 0.05 g mL⁻¹ chloroform solution (Figure 3). Tenax[®] films lighter than 200 mg were too
215 thin and broke easily when removed from the petri dish. Hence, films containing 200
216 mg and 300 mg Tenax[®] were found suitable to proceed.

217 Next, the performance of 200 mg and 300 mg films (evaporation of 4 mL and 6 mL of a
218 0.05 g mL⁻¹ solution) was compared for Tenax[®] films constructed from chloroform and
219 Tenax[®] films constructed from DCM. The results for ITX, BBP and NPT are given in
220 Figure 4. The results, an average of triplicate experiments, demonstrated that the choice
221 of dissolution solvent, nor the weight of the film had an important impact on the
222 migration performance, for all compounds investigated. Additionally, 200 mg films
223 were more homogenous compared to 300 mg films because less evaporation time was
224 needed. Hence, 200 mg films constructed by evaporation of 4 mL of a chloroform
225 solution (0.05 g mL⁻¹) were chosen for further experiments.

226

227 ***Impact of the amount of Tenax[®] in a conventional migration experiment***

228 In the conventional migration experiment 1 gram Tenax[®] is brought into contact with
229 0.15 dm² of cardboard sample (Jakubowska et al., 2014). The optimised Tenax[®] film
230 only contained 200 mg Tenax[®]. Hence, the conventional migration from the selected
231 compounds in 1 gram Tenax[®] powder was compared to the migration in 0.2 gram
232 Tenax[®] powder. The results for ITX, BBP and NPT are shown in Figure 5. Despite the
233 fact that the cardboard surface is barely covered using 0.2 gram Tenax[®], the migration

234 results of the selected contaminants towards 0.2 gram Tenax[®] are comparable with the
235 migration results towards 1 gram Tenax[®] for all contaminants investigated. Hence, the
236 performance of the optimised Tenax[®] films can be compared with the conventional
237 migration experiment using 1 gram Tenax[®] powder.

238

239 ***Evaluation performance Tenax[®] film***

240 Finally the performance of the Tenax[®] film was compared to the performance of the
241 conventional 1 gram Tenax[®] powder for two available blank cardboards (cardboard A
242 and B) destined for the packaging of dry foodstuffs. Hence, the spiked cardboards were
243 each brought into contact with a Tenax[®] film for 10 days at 60°C. In parallel, spiked
244 cardboard was brought into contact with 1 gram Tenax[®] powder for the same contact
245 time and temperature. The experiment was performed in triplicate, using calibration
246 curves constructed in the matrix. The results for ITX, BBP and NPT are given in figure
247 6. Except for NPT, all compounds showed a comparable or even a higher migration
248 towards the Tenax[®] powder for cardboard A. Since different cardboards have different
249 properties, different migration results were observed for both migration into the
250 conventional 1 gram Tenax[®] powder and the 200 mg Tenax[®] film. Nevertheless,
251 cardboard A is rather rough and probably a more intense contact is established between
252 the cardboard and the Tenax[®] powder compared to the contact with the Tenax[®] film.
253 For cardboard B; BP, DTBP, EDB, ITX and MBP have a higher migration potential for
254 the Tenax[®] powder, while DiPN, MS, NPT and PBZ migrate more towards the designed
255 Tenax[®] film . Probably the effect of a more intense contact between the cardboard and
256 the powder is lost because cardboard B was found to be much smoother compared to
257 cardboard A. Hence, the migration in Tenax[®] powder compared to the migration in the

258 Tenax[®] film is influenced by the surface properties of the cardboard with a more
259 favourable migration in Tenax[®] powder for rough surfaces.

260 In 2016, Van Den Houwe et al. performed migration experiments for the
261 migration of several photo-initiators from cardboard in cereals after a contact of 6
262 months at room temperature (Van Den Houwe et al., 2016). About 5.4% BP migrated in
263 cereals after a six months contact between the cereals and the cardboard at room
264 temperature (Van Den Houwe et al., 2016). The percentage of migration of BP in the
265 Tenax[®] films was 26.1% using a smooth cardboard and 14.1% using a more rough
266 cardboard to establish the migration contact for 10 days at 60°C. Although the
267 migration rate in the Tenax[®] film (14.1% and 26.1%) is less compared to the migration
268 in the powder form (38.8% and 36.9%), the Tenax[®] film migration rate might be better
269 estimate of the migration in dry foods. Indeed, the migration of photo-initiators in other
270 dry foodstuffs such as bread crumbs, pasta and rice was more realistically simulated by
271 the use of the Tenax[®] film in comparison with the conventional migration (Table 2).
272 However, a higher migration potential of photo-initiators towards rice was indicated in a
273 rice matrix (Table 2). These results are of great consistency with the market survey
274 performed in 2014 where significant amounts of photo-initiators were only found in
275 several rice samples (Van Den Houwe et al., 2014).

276 Nevertheless, Tenax[®] films were fast and easily applied to the cardboard
277 surfaces and ensure the entire collection of Tenax[®] adsorbent prior to extraction,
278 making the developed Tenax[®] films worthy for further optimisation and investigation.

279

280 **Conclusion**

281 The use of Tenax[®] films as a food simulant for dry foodstuffs for the migration of 15
282 chemical contaminants from cardboard was evaluated. Initially, the construction of the

283 Tenax[®] films was optimised. Films with less than 200 mg Tenax[®] showed severe
284 drawbacks, but 200 mg Tenax[®] films constructed from a Tenax[®]-chloroform solution
285 were found suitable. The optimised films were brought into contact with two types of
286 cardboard enriched with 15 chemical contaminants relevant for paper and board food
287 contact materials.

288 Higher migration rates were observed towards conventional Tenax[®] powder compared
289 to the migration in the designed Tenax[®] films. This tendency was explained by a more
290 intense contact between the cardboard and the conventional Tenax[®] powder.
291 Nevertheless, the designed Tenax[®] film is fast and easy applicable and can open new
292 perspectives in the domain of testing food contact materials intended for contact with
293 dry foodstuffs, since preliminary experiments have demonstrated that the migration rate
294 is more realistic when compared to the migration in dry foodstuffs.

295

296

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300

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382

383 *Overview of the figures*

384 **Figure 1**

385 Chemical structures of the chemical contaminants.

386 **Figure 2**

387 Tenax[®] films constructed from different chloroform solutions with a weight of a) 25
388 mg, b) 50 mg, c) 100 mg, d) 150 mg and e) 200 mg.

389 **Figure 3**

390 Tenax[®] films constructed from a 0.05 g mL⁻¹ chloroform solution with a weight of a)
391 100 mg, b) 200 mg and c) 300 mg.

392 **Figure 4**

393 Comparison of the performance of 200 mg and 300 mg Tenax[®] films constructed from
394 DCM and chloroform, relative to the 200 mg film constructed from chloroform.

395 **Figure 5**

396 Comparison of the migration towards 1 gram and 0.2 gram Tenax[®] powder, relative to 1
397 gram Tenax[®].

398 **Figure 6**

399 Comparison of the performance of Tenax[®] films and Tenax[®] powder for cardboard A,
400 resulting in a higher migration rate towards the Tenax[®] powder for ITX, a comparable
401 migration rate for BBP and a higher migration rate towards the Tenax[®] film for NPT.

402

403 *Overview of the tables*

404 **Table 1**

405 Overview of the specific m/z ratios and retention times of the selected chemical
406 contaminants.

407 **Table 2**

408 Comparison of the conventional migration and the migration in Tenax[®] films with the
409 actual migration in dry foodstuffs for several photo-initiators (average of 3 replicates).

410

411 *Overview of supplementary data*

412 **Figure 1**

413 Comparison of the performance of 200 mg and 300 mg Tenax[®] films constructed from
414 DCM and chloroform, relative to the 200 mg film constructed from chloroform.

415 **Figure 2**

416 Comparison of the migration towards 1 gram and 0.2 gram Tenax[®] powder, relative to 1
417 gram Tenax[®].

418 **Figure 3**

419 Comparison of the performance of Tenax[®] films and Tenax[®] powder for cardboard A,
420 resulting in a) a higher migration rate towards the Tenax[®] powder, b) a comparable
421 migration rate and c) a higher migration rate towards the Tenax[®] film.

422 **Figure 4**

423 Comparison of the performance of Tenax[®] films and Tenax[®] powder for cardboard B
424 resulting in a) a higher migration rate towards the Tenax[®] powder, b) a comparable
425 migration rate and c) a higher migration rate towards the Tenax[®] film.

426

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429 **Table 1**

N°	Chemical contaminant	Abbreviation	Specific m/z ratio	Retention time (min)
1.	Acetophenone	AP	105	4.76
2.	Naphthalene	NPT	128	5.97
3.	2,6-Di-tert-butylphenol	DTBP	191	8.44
4.	Benzophenone	BP	105	9.36
5.	2,6-Diisopropyl-naphthalene	DiPN	197	9.95
6.	4-methylbenzophenone	MBP	119	10.16
7.	2,2-Dimethoxy-2-phenyl acetophenone	DMPA	151	10.87
8.	Dibutyl phthalate	DBP	149	11.21
9.	Trans,trans-1,4-diphenyl-1,3-butadiene	DBPD	206	11.78
10.	Methyl stearate	MS	74	11.96
11.	Dibutyl sebacate	DBS	241	12.22
12.	2-Ethylhexyl-4-dimethylaminobenzoate	EDB	165	12.80
13.	Benzyl butyl phthalate	BBP	149	13.22
14.	2-Isopropyl-9H-thioxanthen-9-one	ITX	239	13.55
15.	4-Phenylbenzophenone	PBZ	181	13.94

430

431 **Table 2**

	Conventional migration in Tenax [®] powder (%) (10 days at 60°C)	Migration in Tenax [®] film (%) (10 days at 60°C)	Migration in dry foodstuffs (%) (6 months at room temperature)			
			Cereals	Bread crumbs	Pasta	Rice
BP	50.5	26.1	5.4	5.11	5.98	17.58
EDB	69.4	23.4	8.9	11.08	17.81	36.77
ITX	54.7	21.7	7.4	8.14	14.57	30.78
MBP	50.2	17.1	5.4	14.01	16.98	42.87

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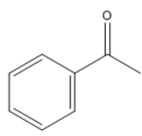
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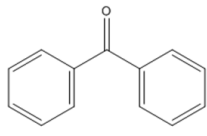
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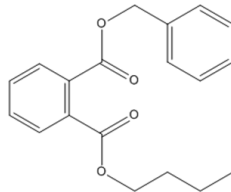
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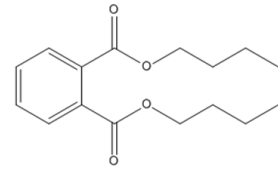
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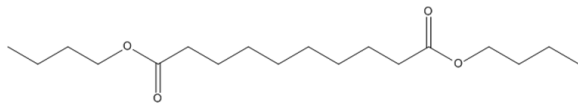
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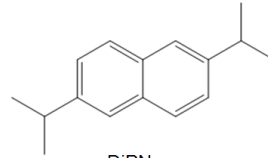
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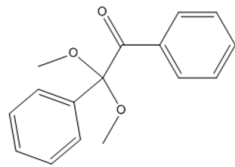
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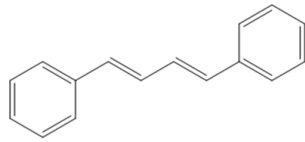
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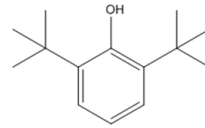
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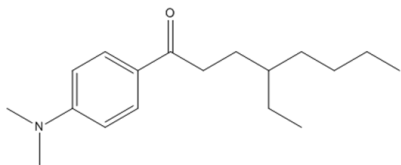
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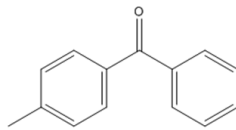
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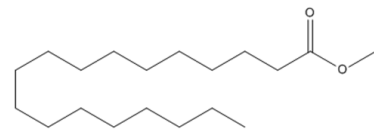
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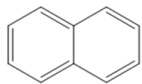
EDB



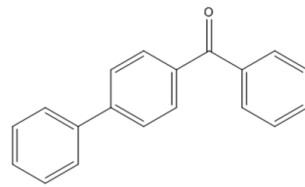
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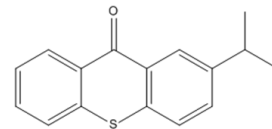
MS



NPT



PBZ



ITX

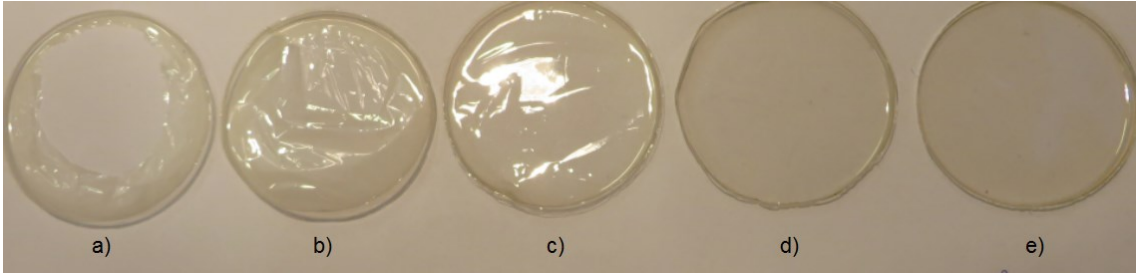
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442 Fig1

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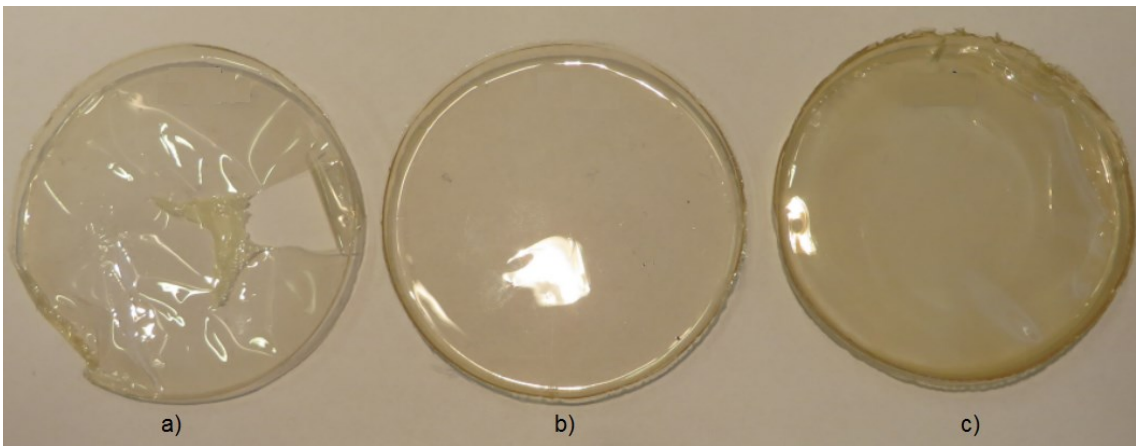
448 Fig 2

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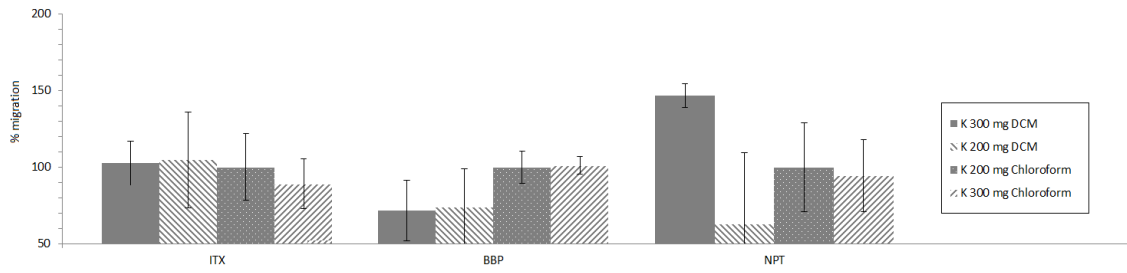
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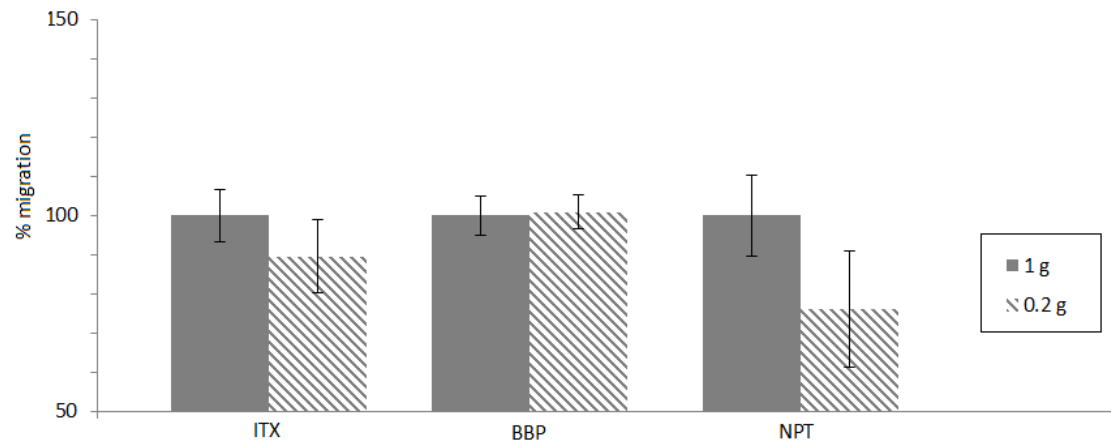
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460 Fig 4

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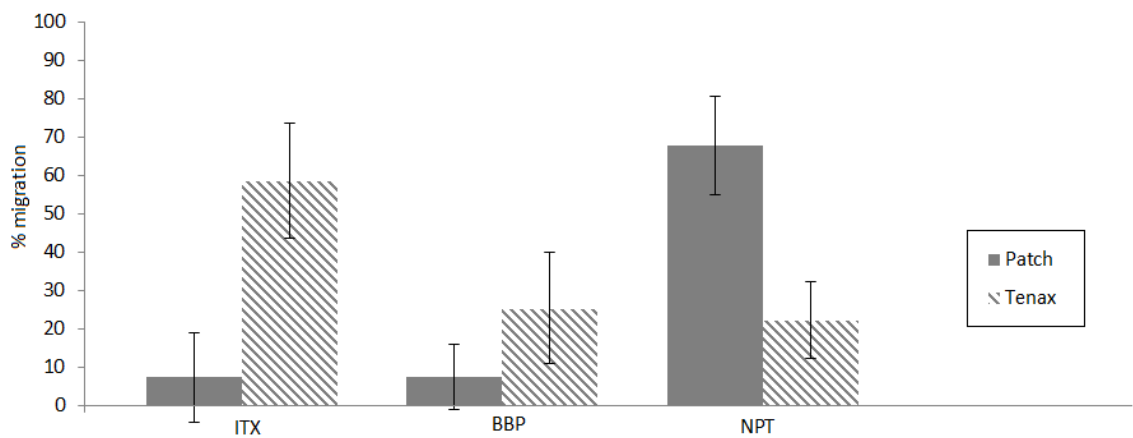
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463 Fig 5

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468 Fig 6