Comparison of 3 analytical techniques for the extraction and determination of 5 possible contaminants in food contact recycled materials

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Abstract. Climate change and ecological crises have become pressing matters in Europe and the world. In an attempt to put a halt to the massive production of packaging waste, the European Standardization Organisation (CENELEC) has developed harmonized standards to promote reuse, recycling, and other forms of recovering packaging waste. However, when implemented in the food and beverage industry, possible contaminants present in these recycled packaging materials could compromise public health. The present study is part of an ongoing research aiming to develop fast, simple, and reliable analytical methods to identify such contaminants in recycled materials intended to come in contact with foodstuffs. To this end, three extraction methods were developed and assessed regarding their effectiveness and accuracy in isolating a mixture of five possible contaminants [Benzophenone (BP), 2,6-Diisopropylnaphthalene (2,6-DiPN), 2,7-Diisopropylnaphthalene (2,7-DiPN), o-Terphenyl (o-TPH), and m-Terphenyl (m-TPH)] in samples of recycled paperboard materials: the Soxtec Extraction (SE), the Ultrasound-Assisted Extraction (UAE), and the Head Space Solid Phase Micro Extraction (HS-SPME). The average recovery rates (ARRs) of the contaminants were satisfactory; the substances were recovered at rates between 61-100% using the SE method, at 100-120% using the UAE method, and at 98-107% using the HS-SPME method. All methods performed well in terms of precision; all produced an overall repeatability relative standard deviation (RSD) of below 2.5% and all HorRat values were well within the acceptable limits, ranging from 0.04 to about 0.1. However, taking into consideration the ANOVA analysis, the method that stood out and altogether demonstrated the best analytical results was the HS-SPME.

Keywords: contaminants; recycled paperboard; extraction techniques; GC/MS

1 Introduction

In recent years, EU waste legislation has actuated considerable improvements in waste management. Full implementation of that legislation, however, is of the highest importance if the EU is to reap the environmental and economic benefits of the circular

economy; the transition to such an economy calls for extensive modifications from product design, production, and supply, to novel business and market models. In addition, the rise of environmental awareness has inevitably prompted industries to increase their demand for and development of alternative packaging materials, including fiber-based packaging, aiming to encourage consumers to make more eco-friendly choices. In the circular economy, "waste" from one process becomes the input for another and so new ways of turning waste into resources and prolonging the life of products are being implemented throughout the supply chain. Recovered fibers constitute a significant source of raw materials for the paper industry given the fact that paper packaging is largely manufactured with the use of recycled paper. In 2020, around 74% of all paper and board consumed in Europe was recycled. However, due to the Covid-19 crisis, among other factors, the consumption and recycling of paper and board seem to have recently taken a heavy drop. More specifically, in comparison to the base year of the Declaration (2015), the amount of paper and board collected and recycled decreased by 3.1 million tonnes [1].

As highlighted in several studies, recycling flaunts both environmental and economic assets in terms of technological and system-level perspectives [2–7]. However, when it comes to the production of packaging designated for food contact, the use of recovered fibers must be carefully assessed. Recycled paper and board have been used primarily as outer packaging of food products as they are infamous sources of non-volatile compounds and external contaminants. The basic source of these contaminants is usually the "raw" recovered material, which, during the recycling process, is being treated with various chemicals, many of which are not intended to come into contact with foodstuff and can exceed acceptable levels. Such well-known migrants include mineral oils, photoinitiators, phthalates, and per- and polyfluorinated substances [8–12]. Under appropriate conditions, these contaminants could migrate from and through the packaging into the food [13]. Even though the recycling of paper and board is essential for a greener future, the safe use of paper and board for food contact materials (FCM) remains a challenge; the migration of substances should be restricted to quantities harmless to human health [14]. To this end, in Europe, as well as the US, the same level of safety for chemicals migrating into foods is considered a requisite for all recycled and virgin materials alike [15–18]. In an attempt to minimize the risks, many food companies either discontinued using recycled paper/paperboard and retracted to virgin fibers or utilized additional functional barriers in an attempt to curb the migration from recycled materials into foodstuff [8, 19].

Numerous studies have been carried out on the migration of substances from packaging materials to foods. By and large, the results of these studies demonstrate that the interaction between the packaging and food products is influenced by a lot of factors, such as the structure of the packaging samples, the chemical nature of the migrants, and the time and temperature conditions of the migration tests [12, 14, 18, 20–36]. Consequently, a cautious selection of packaging materials is entailed in order to avoid any adverse effects on the quality, safety, and shelf stability of foodstuffs. To this end, the Committee of Ministers of the Council of Europe adopted in October 2020 new, updated guidelines known as "Resolution CM/Res (2020) 9 on the safety and quality of materials and articles for contact with food". These guidelines, in conjunction with the

supplementary Technical Guide, supersede the previously in force "Policy statement concerning paper and board materials and articles intended to come into contact with foodstuffs" (Version 4 dated 12.02.2009) [37–39].

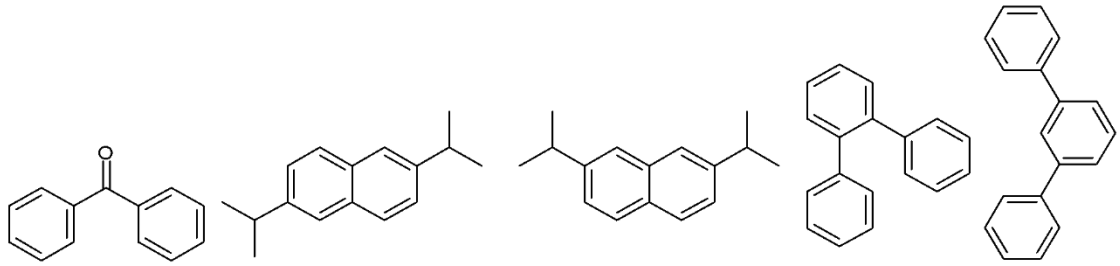
To extensively promote manufacturing and policy decisions favoring harmless alternatives over hazardous chemicals in food packaging, all concerning substances, as well as the amounts thereof, should be identified. So far, studies on determining contaminants in food packaging have focused mainly on monomers and a few additives used in high concentrations, such as phthalate plasticizers [34, 36, 40]. However, even low amounts of these hazardous substances in food contact materials (FCMs) could cause health damage, even though the material as such complies with all legal requirements. Prior to being identified, the substances in question should first be efficiently extracted from the packaging material. The most common extraction and clean-up techniques that have been tested on paper and board FCMs; include HS-SPME [41–46], Soxhlet extraction [43, 47, 48], reflux distillation [49–52], and UAE [53]. The choice of extraction method is of utmost significance in order to achieve accurate and reliable results.

The current study focuses on evaluating the extraction efficiency of three commonly used techniques to determine five potential contaminants present as residues in commercially recycled paperboards intended for food packaging. The selection of the contaminants was based on their prospective presence and frequency in materials containing recycled fibers. The main objective was to identify the most appropriate technique meeting the criteria of being rapid, straightforward, and sensitive for extracting and determining these compounds at low concentrations.

2 Experimental procedures

2.1 Chemicals

All chemicals used were of high analytical grade and purchased from Sigma-Aldrich, Fluka, Supelco, and Merck. Standard solutions were prepared of the five following possible contaminants: Benzophenone (BP), 2,6-Diisopropylnaphthalene (2,6-DiPN), 2,7-Diisopropylnaphthalene (2,7-DiPN), o-Terphenyl (o-TPH), and m-Terphenyl (m-TPH) (Scheme 1). A mixture of these substances was used as the standard solution for the contamination of the paperboards.



1 Benzophenone **BP** | **2** 2,6-Diisopropylnaphthalene **2,6DiPN** | **3** 2,7-Diisopropylnaphthalene **2,7DiPN** | **4** *o*-Terphenyl ***o*-TPH** | **5** *m*-Terphenyl ***m*-TPH**

The initial concentrations of the selected substances were 1 g L-1, from which two final standard solutions of 10 mg L-1 and 1 mg L-1 of all substances were prepared.

2.2 Paper samples

The paper samples used in this study were the same as in previous work [46, 54]. Three of them were made of 100% recycled pulp matter (R1, R2, R3) and one was of 0% recycled material (V). The absorption capacity of each type of paperboard is expected to vary therefore known volumes of the standard solution were used during contamination as a correction factor.

More specifically, the materials used were:

R1: "Triplex R Kraft", Basis Weight: 440 g/m^2, thickness: 541 μm.

R2: "Duplex R", Basis Weight: 400 g/m^2, thickness: 485 μm.

R3: "Triplex BR", Basis Weight: 440 g/m^2, thickness: 541 μm.

V: "Bl. Kraft + CTMP", Basis Weight: 273 g/m^2, thickness: 486 μm.

2.3 Analytical Methods

Three commonly used extraction methods were optimized and evaluated in this study: the Soxtec extraction (SE), the ultrasound-assisted extraction (UAE), and the headspace solid phase microextraction (HS-SPME). A gas chromatography-mass spectrometry (GC/MS) analysis superseded each extraction to determine the contaminants.

For the identification and quantification of the selected substances, as well as the assessment of their recovery rates, several contamination tests were conducted on the paperboard samples. The ones consisting of recycled material were thoroughly cleansed beforehand with dichloromethane in ultrasonic baths. All four paperboard samples were then thoroughly saturated with a range of solutions of the standard substances (20-20.000 μg/l) and standard reference curves were plotted for each contaminant.

2.4 Soxtec Extraction (SE)

The contaminants were extracted from the paperboard samples with the use of a semi-automatic solvent extractor (SER 148, VELP Scientifica Srl, Italy). Prior to the analysis, all parameters were optimized through testing according to previous work [54]. The paperboard samples, each weighing 1.0 g, were consecutively placed in extraction thimbles and treated with 50 ml dichloromethane (DCM) at 110ºC for 2h. The thimbles were then set in rinse position for an extra 1.5h and, after the samples were dried with the use of synthetic air, they were redissolved in 1.0 ml of acetonitrile. An aliquot of 1.0 μl of the final solution was transferred into the GC/MS injection port. All analyses were performed in triplicate.

2.5 Ultrasound-Assisted Extraction (UAE)

An Elmasonic S10 H ultrasonic bath (Elma GmbH, Singen, Germany) was utilized for this analysis. The parameters of the liquid-phase extraction were the same as in previous work [53].

2.6 Head Space – Solid Phase Micro Extraction (HS-SPME)

For the identification and quantification of the substances under study, paperboard samples were steeped in 1 ml of the 1 mg L-1 standard solution. Approximately 100 mg of the saturated paperboard samples were weighed, cut into pieces (5 x 5 mm), and placed in glass vials of 10 ml volume.

The extraction of volatile compounds from the headspace was performed using a 65 μm Polydimethylsiloxane / Divinylbenzene (PDMS/DVB) fiber (Supelco, USA) [41]. The vials were placed in an oil bath at 135°C (higher bath temperatures produced adverse recovery results) for 15 minutes until equilibrium and after that, the PDMS/DVB fiber was inserted and exposed to the headspace for 30 minutes. The fiber was then transferred to the GC instrument for the analysis of the substances. All experimental tests were carried out in triplicate.

Additionally, all samples were saturated with aliquots ranging from 30 to 750 μg kg-1 of the standard solutions of the contaminants and subjected to HS-SPME analysis under the same conditions as described above in order to plot the necessary reference curves.

2.7 GC/MS

All chromatographic analyses were conducted on a HP 6890 GC system coupled to a HP 5973MS detector. Additional components consisted of a nonpolar HP-5 (5%-phenyl)-methylpolysiloxane column (60 m × 0.25 mm i.d., with 1 μm film thickness) (J&W Scientific, Folsom, USA) and high purity He (carrier gas) released at a flow rate of 0.8 mL/min. The temperature programming used was the same as in previous work [54]. Finally, the identification of the compounds was performed using a mass spectral library (Wiley 7, NIST 2005) and through comparing the linear retention indices (Kovats indices) to reference standards and/or published data [55, 56].

3 Statistics

The software used for all statistical analyses was the SAS 9.4 (SAS Institute Inc., Cary, NC, USA).

4 Results and discussion

4.1 Recoveries

According to the updated SANTE Directive [57] the recovery rates of the substances should span from 70 to 120% in all contaminated samples and the relative standard deviations should not surpass 20%. The limits of detection (LODs) for the standard solutions of the 5 contaminants were determined at a signal-to-noise ratio of 3. The LODs and the limits of quantification (LOQs) for the concentrations tested ranged from 0.005 to 0.5 mg kg-1, and from 0.1 to 1 mg kg-1, respectively.

Soxtec Extraction (SE)

The SE method recovery rates for all concentrations of the 5 substances ranged between ~53% and ~100%, whilst the average recovery rates (ARRs) ranged from ~61% and ~100% (Table 1). In addition, the standard deviation values (SD) were all well below 20% in all cases. Even though the RRs of o-TPH were found to be a bit lower than the acceptable lower limits (<70%), all in all the SE can be considered a reliable method for the analysis of these contaminants that may be present in recycled paperboard.

Table 1. Paper recovery rates (RRs) and standard deviation (SD) of the 5 contaminants at three concentrations (125 μg kg^{-1}, 300 μg $kg^{-1,}$ and 500 μg kg^{-1}) using the SE method.

Contaminant	**Recovery**							
	125μg kg^{-1}		300μg kg^{-1}		500μg kg^{-1}		ARR*	SD (%)
	RR (%)	SD (%)	RR (%)	SD (%)	RR (%)	SD (%)	(%)	
BP	73.24%	1.46%	62.68%	0.89%	73.17%	0.91%	69.70%	6.07%
2,6-DiPN	86.52%	1.58%	83.50%	1.35%	100.29%	1.14%	90.10%	8.95%
2,7-DiPN	84.16%	1.53%	78.20%	1.38%	99.10%	1.25%	87.15%	10.76%
o-TPH	53.36%	1.38%	60.59%	1.42%	68.55%	1.39%	60.83%	7.60%
m-TPH	69.88%	0.79%	75.90%	0.72%	86.31%	0.77%	77.36%	8.31%

*ARR: Average Recovery Rate

Ultrasound – Assisted Extraction (UAE)

The UAE method recovery rates for all concentrations of the 5 substances ranged between ~94% and ~126%, while the average recovery rates (ARRs) ranged from ~100% and ~120% (Table 2). In addition, the standard deviation values (SD) were all well below 20% in all cases. Therefore, the UAE constitutes a reliable method for the analysis of these contaminants that may be present in recycled paperboard.

Table 2. Paper recovery rates (RRs) and standard deviation (SD) of the 5 contaminants at three concentration levels (125 μg kg^{-1}, 300 μg $kg^{-1,}$ and 500 μg kg^{-1}) using the UAE method.

Contaminant	**Recovery**							
	125μg kg^{-1}		300μg kg^{-1}		500μg kg^{-1}		ARR*	SD (%)
	RR (%)	SD (%)	RR (%)	SD (%)	RR (%)	SD (%)	(%)	
BP	122.30%	1.72%	109.50%	1.32%	94.56%	1.29%	108.79%	13.88%
2,6-DiPN	100.40%	1.56%	99.54%	1.35%	99.02%	1.21%	99.65%	0.70%
2,7-DiPN	102.20%	1.60%	101.55%	1.58%	100.61%	1.15%	101.45%	0.80%
o-TPH	120.60%	1.82%	116.17%	1.79%	113.75%	1.47%	116.84%	3.48%
m-TPH	125.50%	2.20%	118.09%	1.54%	116.33%	1.11%	119.97%	4.87%

*ARR: Average Recovery Rate

Head Space – Solid Phase Extraction (HS-SPME)

The HS-SPME method recovery rates for all concentrations of the 5 substances ranged between ~96% and ~110% (Table 3). Even though at concentrations 125 and 300 μg kg-1 the contaminant o-TPH was not recovered, it was completely recovered at concentration 500 μg kg-1. According to the results, the HS-SPME method can be reliably used for the recovery of all 5 contaminants.

Table 3. Paper recovery rates (RRs) and standard deviation (SD) of the 5 contaminants at three concentration levels (125 μg kg^{-1}, 300 μg $kg^{-1,}$ and 500 μg kg^{-1}) using the HS-SPME extraction method.

Contaminant	**Recovery**							
	125μg kg^{-1}		300μg kg^{-1}		500μ kg^{-1}		ARR*	SD (%)
	RR (%)	SD (%)	RR (%)	SD (%)	RR (%)	SD (%)	(%)	
BP	98.26%	0.98%	99.35%	0.95%	96.54%	2.04%	98.05%	0.62%
2,6-DiPN	105.38%	1.25%	106.44%	1.26%	105.83%	2.39%	105.88%	0.65%
2,7-DiPN	100.73%	0.98%	100.86%	1.09%	97.26%	2.60%	99.62%	0.90%
o-TPH	n.d.	n.d.	n.d.	n.d.	104.23%	2.86%	104.23%	2.86%
m-TPH	106.13%	1.07%	109.53%	1.36%	106.40%	2.87%	107.35%	0.97%

*ARR: Average Recovery Rate

4.2 Accuracy of methods

The accuracy of the methods was expressed as approximate trueness since Certified Reference Materials (CRMs) were not used in this study. The concentrations were greater than 10 μg kg-1, so the acceptable range was from -20% to + 10%. The trueness of the methods was evaluated at 125 μg kg-1. Altogether, twenty trials were performed. The results of the tests are given in Table 4. In bold are the acceptable values.

Table 4. Approximate values of the trials at 125 μg kg^{-1} for each extraction technique.

Contaminant	**UAE**	**SE**	**HS-SPME**
	125μg kg^{-1}	125μg kg^{-1}	125μg kg^{-1}
BP	153	92	**122.83**
2,6-DiPN	**126**	**108**	**131.72**
2,7-DiPN	**128**	**105**	**125.91**
o-TPH	151	67	-
m-TPH	157	87	**132.66**

According to the results, of the three methods, the HS-SMPE has the highest accuracy at 125 μg kg-1 as 4 out of 5 contaminants are within limits, while in the UAE and SE methods, only two out of 5 are within limits.

4.3 Precision of methods

The methods' precision was evaluated using the Horwitz Ratio (HorRat) equation [58]. This equation is a precision assessment criterion that finds wide applicability in certifications and international standards such as ISO 17025. For a method to be considered precise, it should meet the following criteria:

- HorRat ≤ 0.5: excellent precision
- HorRat ≥ 2: unacceptable precision

A method can be accepted for values between these limits with appropriate substantiation.

HorRat values of each method were calculated by assessing the relative standard deviation (RSD) at a concentration of 125 μg kg-1. The maximum theoretically calculated RSD at said concentration was estimated to be RSD= 21,88. The experimental %RSD and HorRat values are given in Table 5.

Table 5. Experimental %RSD and HorRat values for the UAE, SE, and HS-SPME methods at a concentration of 125μg kg^{-1}.

	RSD			HorRat		
	UAE	**SE**	**HS-SPME**	**UAE**	**SE**	**HS-SPME**
BP	1.51	2.05	1.08	0.07	0.09	0.05
2,6-DiPN	1.50	1.92	1.08	0.07	0.09	0.05
2,7-DiPN	1.68	1.76	0.97	0.08	0.08	0.04
o-TPH	1.61	2.43	-	0.07	0.11	-
m-TPH	1.83	1.19	1.03	0.08	0.05	0.05

The RSD and HorRat values indicate that the HS-SPME method produced more precise results than the other two methods tested.

4.4 ANOVA analysis

The analysis of variance (ANOVA) was performed on a total of 20 spiked samples to assess the aberration between the average values of the 4 out of 5 studied compounds at a concentration of 125 μg kg-1.

For the BP compound, the HS-SPME method almost coincided with the theoretical approach; 99% of HS-SPME values appeared to be better distributed than the corresponding distribution of the range of 25% of the theoretical values. The UAE and Soxtec methods showed statistically significant differences but had better distribution than the theoretical.

For the 2,6-DiPN compound, the UAE seems to coincide with the theoretical approach; 99% of the UAE values appeared to be better distributed than the corresponding distribution of the range of 25% of its prices theoretical. HS-SPME has a small price range, which is almost within the range of 25% of the theoretical values. The means are significantly different prices and intermediaries. Although SE shows statistically significant differences from the theoretical, it seems that the price distribution is better. The range of 99% of SE is within the 99% distribution range of theoretical values.

For the 2,7-DiPN compound, the UAE and HS-SPME seem to coincide statistically with the theoretical approach; 99% of the values of the two methods seem to be better distributed than the corresponding distribution of the range of 25% of the theoretical values. However, the SE analysis presented statistically significant differences.

For the m-TPH compound, none of the methods is grouped by the theoretical approach. Only the HS-SPME method seemed to demonstrate the smallest statistical differences. The value distribution range is very small and is almost within the distribution range of 25% of the theoretical prices, which is also the case for the SE method.

The statistical approaches confirmed that the most reliable results were obtained when using the HS-SMPE method.

5 Conclusions

All methods tested performed well in terms of precision; all showed an overall repeatability relative standard deviation (RSD) of below 2.5%. Additionally, the HorRat values of all methods were within the limits for performance acceptability, ranging from 0.04 to about 0.1. However, taking into consideration the ANOVA analysis, the method that stood out and altogether demonstrated the best analytical results was the HS-SPME.

The proposed method could be used for the determination of these compounds as undesirable impurities in paperboards classified as food-grade materials. The HS-SMPE analysis, coupled with the GC/MS method, is not only simple and rapid but also sensitive. It is therefore suitable for the routine analysis of the most common organic compounds found in recycled paperboard. The integration of such a simple and reliable analytical technique in the line of food packaging production could be an important asset to both the producers and the promotion of circular economy in general. Not only could the proposed HS-SMPE analysis be used to identify and prevent potential risks prior to the formation of the final packaging products, it would also ensure the distribution of safe food packaging without putting financial strain on the producers. Finally, acknowledging the risk-free products, consumers would feel more open to the use of recycled materials in food packaging and together with the producers they could fully embrace the benefits of circular economy.

5.1 Correction factors

After determining that HS-SPME was the most reliable for this specific procedure, correction factors were computed so as to attain optimal accuracy of the results. The said factors were calculated for each individual contaminant by estimating the mean value of 20 spiked samples at a concentration of 125 μg kg-1 and then dividing the resulting value by the value of the concentration (Table 6).

Table 6. Correction factors estimated at a concentration of 125 μg kg^{-1}.

Contaminant	Correction factor
BP	1.02
2,6-DiPN	0.95
2,7-DiPN	0.99
o-TPH	-
m-TPH	0.94

In addition, three commercial food-grade paperboard samples were tested (R1, R2, R3) in order to verify the applicability of the method for the simultaneous determination of all 5 contaminants. The corrected values of the contaminants were evaluated by multiplying their concentrations by the corrected factors estimated above (Table 7). As seen in Table 7, the deviation of the corrected value of the contaminants' concentration compared to the one obtained using the proposed methods is low, which indicates the high accuracy of the methods. However, by implementing the said correction factors to all

contaminants the overall accuracy of the methods, as well as the reliability of the results are increased.

Table 7. Concentrations found in three commercial food-grade paperboard samples and their corrected values.

Contaminant	R1			R2			R3		
	Found (μg kg^{-1})	SD	Corrected (μg kg^{-1})	Found (μg kg^{-1})	SD	Corrected (μg kg^{-1})	Found (μg kg^{-1})	SD	Corrected (μg kg^{-1})
BP	23	1	24	48	1	49	21	1	21
2,6-DiPN	250	1	238	420	17	399	174	3	165
2,7-DiPN	141	2	140	202	3	200	93	3	92
o-TPH	11	2		33	1		11	2	
m-TPH	18	2	17	13	1	12	12	2	12

Declarations

Conflict of interest The authors declare that they have no conflict of interest.

Research involving human participants and/or animals This article does not contain any studies with human or animal subjects.

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