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RESEARCH ARTICLE

Occurrence of phthalates and 2,6-diisopropylnaphthalene in dry foods packed in cellulosic materials

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Abstract
Paper and cardboard for food packaging are non-inert materials that can transfer chemical compounds by migration into food. Phthalates, namely dibutyl phthalate (DBP), diisobutyl phthalate (DIBP), bis(2-ethylhexyl) phthalate (DEHP) and 2,6-diisopropylnaphthalene (DIPN) are some of the compounds that can migrate into food. In this study, 7 dry foods [wheat flour (2), powdered chocolate (2), cornstarch, toast and biscuit] in paper packaging were evaluated for the presence of DBP, DIBP, DEHP and DIPN by gas chromatography coupled with mass spectrometry (GC-MS). Only two food samples (powdered chocolate and toast) did not contain any of the studied substances. In the other dry food samples, 1, respectively 2 of the investigated phthalates were detected. DEHP was found in the highest concentration (ranging from 1.56 to 3.85 mg kg^{-1}), followed by DIBP (ranging from 0.36 to 1.51 mg kg^{-1}). The values exceeded the migration limits established by Mercosur and European Union legislation by 2.5 to 5 times, indicating the need to improve process control and to adopt stricter good manufacturing practices, in order to avoid exposure of the population to these substances.

Keywords Phthalates · Migration · Dry food · Cellulose packaging · Food contact material

1 Introduction

Food packaging is defined as food contact material (FCM) and is used for the protection and preservation of food (Marangoni Júnior et al. 2020a, b). In addition to packaging, other materials may come into direct or indirect contact with food, such as transport containers, processing machines, and kitchen utensils (Vandermarken et al. 2019). Different substances can migrate from FCMs into food, thereby contaminating it (Blanco-Zubiaguirre et al. 2020; Marangoni Júnior et al. 2022).

Phthalates are potential contaminants that can be present in FCMs (e.g., equipment and cellulosic packaging) and migrate into the food. Regarding cellulosic materials, phthalates are often incorporated unintentionally due to the possibility and permission of using recycled pulps in the manufacture of packaging (Poças and Hoog 2007). Phthalates are a group of organic lipophilic chemicals used primarily as plasticizers to increase flexibility of polymer products including food packaging, bottled water, wall coverings, wires, toys, blood bags, grocery bags, garbage bags etc. (Ahmad et al. 2021; Cheshmazar et al. 2021). In addition, phthalates may be present in printing inks, lacquers and adhesives of such packages. DBP, DIBP and DEHP are some of these phthalates that can migrate into food (Geueke et al. 2018). Contamination can occur by set-off, which is defined as the transfer of components from the external layer (printed surface) of a cellulosic packaging material to the inner side (surface to come into contact with food) during storage of printed substrates in reels or stacks (Lemos et al. 2017; Nerín et al. 1993; Asensio et al. 2019).

In addition to phthalates, recycled cellulosic materials may contain DIPN (Jaén et al. 2021). DIPN is mostly found in packaging that has been produced from recycled office papers, since DIPN is used as solvent in carbonless copy paper in substitution to polychlorinated biphenyls (Sturaro

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et al. 1994; Mariani et al. 1999; Singh et al. 2018). In this context, since they have no other uses in the food sector, they are excellent markers for migration from recycled paper and board (Biedermann and Grob 2012; Conchione et al. 2020). DIPN or phthalates can migrate into food through direct contact or even through gaseous transport (Geueke et al. 2018; Poças et al. 2011a). According to Coltro and Machado (2020), based on analyses of samples contaminated with different levels of DIPN, 12 mg kg^{-1} is the maximum concentration of DIPN in cellulose packaging to reach the specific migration limit of 0.01 mg kg^{-1} or to be detected.

In order to ensure food safety, FCMs must comply with the regulations agreed among the countries of Mercosur (Southern Common Market), which incorporate Resolution RDC no. 88 of 29 June 2016, issued by the Brazilian National Health Surveillance Agency (Anvisa) of the Brazilian Ministry of Health (2016). This resolution includes a positive list of components for materials, packages and cellulose equipment intended to come into contact with food, specifying maximum specific migration limits (SML) for several substances, including inorganic contaminants, when the material includes recycled fibers in its production. This resolution establishes for the most common phthalates used in packaging and/or printing inks the same SML as those adopted by the European Union, which are 0.3 mg kg^{-1} for DIBP, 0.3 mg kg^{-1} for DBP (however, the sum of DIBP and DBP must not exceed 0.3 mg kg^{-1}), 1.5 mg kg− 1 for DEHP, and DIPN must be non-detectable.

Several studies on DBP and DIPN occurrence and migration into food packed in cellulose packages have been conducted in the US and Europe. Zhang et al. (2008) detected no contamination by DIPN in 41 U.S. food samples, but DBP was detected in 2 U.S. market samples at concentrations ranging from < 0.01 to 0.13 mg kg^{-1}, whereas in 4 samples of imported food concentrations ranged from < 0.01 to 0.81 mg kg^{-1}. A review performed by Serrano et al. (2014) on phthalate monitoring in food have found DEHP in high concentrations in meat, fat and dairy products and may contribute to exposure to phthalates in the population´s diet. In a study conducted in Portugal by Poças et al. (2011b) on consumer exposure to phthalates from cellulose packaging, 21 packaging materials and 5 food items were evaluated. None of them contained DBP, but DIBP and/or DEHP were detected in all 5 food items. The DIBP amount was approximately 0.15 mg kg^{-1} in cake mix and tea, and 0.37 mg kg^{-1} in wheat flour. DEHP was found in concentrations of 0.06 mg kg^{-1} in cake mix, 0.2 mg kg^{-1} in stocks and 2.2 mg kg^{-1} in butter. In Belgium, phthalates were found in food and packaging materials: Fierens et al. (2012) investigated 400 items of food and packaging sold on the Belgian market. DEHP was found with the highest concentrations and was detected in 81% of the products evaluated, followed by DIBP (75%) and DBP (69%). DEHP concentrations in cereal and cereal products group ranged from undetectable to 1.07 mg kg^{-1}, DIBP ranged from undetectable to 1.05 mg kg^{-1}, and DBP ranged from undetectable to 0.06 mg kg^{-1}.

Generally, the food industry is responsible for ensuring safety and quality of food in order to prevent damage to consumers' health, both regarding long-term and short-term effects. Cellulose packaging is often used as transport packaging, secondary and mainly primary packaging in the case of dry food, such as flour, chocolate, cereals, toast and pasta. In a previous study, we evaluated 20 samples of cellulose packaging for dry foods available on the Brazilian market and tested for the migration of phthalates (DBP, DIBP and DEHP) and DIPN into fatty food simulant. 50% of the cellulosic packaging samples showed no migration of phthalates and DIPN, while 20% showed migration of DIBP, 15% migration of DBP and 40% migration of DEHP (Coltro et al. 2021). Based on this study, 7 dry foods packed in cellulosic packaging evaluated regarding migration of phthalates and DIPN were selected to investigate the occurrence of these contaminants in the food itself.

Therefore, the current study aimed to determine and quantify the presence of DIPN and the phthalates DBP, DIBP and DEHP in dry foods [wheat flour (2), powdered chocolate (2), cornstarch, toast and biscuit] packaged in cellulose commonly available on the Brazilian market. As far as we know, this is the first study on phthalate contamination in dry foods packaged in cellulosic packaging on the Brazilian market.

2 Materials and methods

2.1 Reagents

The following reagents were used in this study: 2,6-diisopropylnaphthalene – DIPN, CAS number 24157-81-1 (99% purity, Sigma Aldrich, USA); dibutyl phthalate – DBP, CAS number 84-74-2 (99% purity, Sigma Aldrich, USA); diisobutyl phthalate – DIBP, CAS number 84-69-5 (99% purity, Sigma Aldrich, USA); bis(2-ethylhexyl) phthalate – DEHP, CAS number 117-81-7 (99% purity, Sigma Aldrich, USA); bis(2-ethyhexyl) adipate - DEHA, CAS number 103-23-1 (98% purity, Sigma Aldrich, USA), used as internal standard; Dichloromethane (p.a., Merck KGaA, Germany); n-heptane (p.a., Synth, Brazil). A stock solution of 1,000 mg kg^{-1} DEHA in n-heptane was used to prepare the working solutions of 100 and 10 mg kg^{-1} DEHA in n-heptane.

Table 1 Types of dry food evaluated and their respective packages

Sample description	Primary packaging	Secondary packaging
White wheat flour A	White paper, printed	-----
White wheat flour B	White paper, printed	-----
Powdered chocolate A	Plastic bag (BOPP/PE)*	Cardboard, printed
Powdered chocolate B	White paper	Cardboard, printed
Cornstarch	White paper	Cardboard, printed
Toast	Corrugated paperboard, white	BOPP/BOPP, printed
Biscuit with sugar crystals	Plastic bag (BOPP/BOPP)	Cardboard, printed

* *BOPP/PE: Bioriented polypropylene/polyethylene*

2.2 Food samples

Several types of dry food commonly packed in cellulose, either primary or secondary packaging, and sold commercially in supermarkets, were purchased in the retail market in Campinas, São Paulo, Brazil from 2016 to 2019. In this study, 7 food samples, each one from at least 3 different production batches, were evaluated as follows:

- 2 brands of wheat flour,
- 2 brands of powdered chocolate,
- 1 brand of cornstarch, toast and biscuit.

As shown in Table 1, all samples were stored in primary paper packaging, except one of the powdered chocolate samples (packed in plastic bag of BOPP/PE, inside a printed cardboard) and biscuit sample (packed in plastic bag of BOPP/BOPP, inside a printed cardboard). Therefore, most products had a direct contact with the cellulose packaging.

2.3 Food extraction

In order to eliminate possible phthalate contamination of the glassware used in the assay, all materials were previously cleaned as follows: washing with ultra-purified water (Milli-Q Direct, Millipore, US) and drying in an oven (Fanem Ltda., SP, Brazil), model 002 CB. After drying, they were washed with acetone and allowed to evaporate in an exhaust hood. Before mounting the extractor, they were washed with methanol and hexane and again allowed to evaporate in the fume hood. Before using the other glassware, such as volumetric flasks and beakers, washing with hexane was performed, followed by evaporation in the fume hood. Besides, a blank of the solvents used in the assays was injected into the GC-MS chromatograph to confirm the absence of the phthalates and DIPN under study.

Extraction of the substances from the dry food was performed according to the method used by Zhang et al. (2008). Different production batches of each dry food were mixed to obtain a homogeneous sample of each type of food evaluated, which was stored in glass jars. About 5 g of the homogenized food sample was extracted with 10 mL of dichloromethane (except for biscuit, which was extracted with 20 mL), with the aid of ultrasound instrument (Cole-Parmer, Illinois, USA), model 8846-40, for 30 min. After this time, the extract was vacuum filtered with Whatman GF/A filter paper. Then 2 mL of this extract was evaporated under nitrogen flow until complete drying and 2 mL of a 10 mg kg^{-1} DEHA solution in n-heptane, used as internal standard, was added. The extract was filtered through a 45-µm filter and injected in the GC-MS chromatograph.

2.4 Chromatographic conditions

For determination of target substances in samples of dry food, GC-MS analyses were performed using a HP Hewlett Packard 6890 (Palo Alto, CA, USA) interfaced with HP Hewlett Packard 5973 mass spectrometer with electron ionization (70 eV). A HP5/MS capillary column (Agilent, 30 m length, 0.25 mm I.D., 0.25 µm film thickness) was used with a helium flow of 2.014 mL min^{-1}. Samples of 1 µL were injected manually using a split ratio of 2:1. The injector temperature was set to 250 °C. The initial temperature of the oven was set at 140 °C, followed by an increase to 200 °C at a flow rate of 5 °C min^{-1} and, finally, at 10 °C min^{-1} to reach the final temperature of 280 °C. Total run time was 40 min. Full scan mass spectra were acquired using a mass range of 50–550 m/z at 1.6 scans s^{-1} and a 1.90-min solvent delay. The temperature for the ion source was set to 230 °C. DEHA at a concentration of 10 mg kg^{-1} was used as internal standard. Chromatograms and spectra were recorded and processed using the MSD ChemStation E.02.02.1431 software for GC–MS (Agilent). The analyses were performed in triplicate for each sample.

Phthalates and DIPN in the food samples were identified by matching retention time and mass spectra with those in a NIST Mass Spectral Library 2.0. Phthalates and DIPN annotations were accepted with a spectral match score higher than 60 and RI-deviation lower than 8 min. The area of each peak, after being recognized, was normalized to the total area for each chromatogram.

2.5 Method validation

The method was validated according to selectivity, linearity range, detection limit, quantification limit, recovery, precision, and accuracy (Ribani et al. 2004; Ribeiro and Ferreira 2008).

Table 2 Analytical method validation parameters*

** Results for triplicates, except for LOD / LOQ (results from seven replicates), LOD = detection limit; LOQ = quantification limit*

Parameter	DIPN	DIBP	DBP	DEHP
Working range (mg kg^{-1})	0.1–5.0	0.1–5.0	0.1–5.0	0.1–5.0
LOD (mg kg^{-1})	0.004	0.003	0.006	0.016
LOQ (mg kg^{-1})	0.012	0.011	0.018	0.050
Accuracy (%)	-5.4–7.2	0.0–5.6	-3.1–11.8	-1.7–9.1
Precision (%)	0.7–18.0	0.2–23.0	2.5–7.2	0.0–23.0
Recovery (%)	86.3 ± 9.6	84.3 ± 12.1	109.4 ± 11.2	128.4 ± 24.7

2.5.1 Linearity range

The calibration curves were built with five points ranging from 0.1 to 5 mg kg^{-1} and 1.0 mg kg^{-1} of DEHA (internal standard) taking into account the ratio of phthalate area per internal standard area and the respective concentration of phthalate solutions. The coefficients of linear and angular correlation were calculated with a linear regression model.

2.5.2 Detection limit (LOD) and quantification limit (LOQ)

Three analytical curves were constructed on different days. Taking into account the average peak areas of the substances under study, the detection and quantification limits were determined by means of analytical curves using the following equations: LOQ = 10 * linear coefficient error/angular coefficient and LOD = 3.3 * linear coefficient error/angular coefficient (Ribani et al. 2004).

2.5.3 Precision and accuracy

Two analytical curves were built with concentrations ranging from 0.1 to 5 mg kg^{-1} of phthalates and DIPN and 1.0 mg kg^{-1} of DEHA (internal standard). Each curve was obtained by a different analyst. The intraday repeatability was estimated from the relative standard deviation (RSD) among the replicates of the points of one curve. The intermediate precision was obtained by calculating the relative standard deviation but considering both analytical curves. The accuracy was assessed via calculation of the relative error (RE), expressed in percentage (Inmetro 2018).

2.5.4 Recovery

The recovery was estimated from the analysis of food samples fortified with known quantities of the substances of interest. For this purpose, wheat flour was assumed as standard for dry food samples. One of the wheat flour samples was fortified with the most critical situation, i.e. low concentration level (0.1 mg kg^{-1}), by pipetting 50 µL of a 10 mg kg^{-1} phthalate and DIPN fortification solution in 5 g of wheat flour. After the organic solvent had evaporated at room temperature, the samples were analyzed as described above employing a 10 mg kg^{-1} DEHA solution as internal standard. The recovery rate was obtained via the ratio between the concentrations obtained in the tests and the expected concentration, as represented by the standard solution.

3 Results and discussion

3.1 Method validation

The analytical curves obtained in GC-MS were constructed from the ratio of phthalate areas or DIPN and internal standard area (DEHA) vs. the concentration of solutions of 5 different concentrations and showed good linearity, with correlation coefficients values > 0.99 for all analytes. Table 2 shows the method validation parameters including accuracy, precision, LOQ and LOD obtained for each substance analyzed.

The obtained recovery values for the analyzed substances at 0.1 mg kg^{-1} concentration level ranged from 84 to 109%, except DEHP that showed values in the order of 130%. The recovery values are within the range and acceptable for these analytes, according to the acceptance criteria for recovery established by Inmetro, which ranges from 80 to 110% (Inmetro 2018). The high recovery value for DEHP is probably due to the fact that it is present in most environments and very difficult to control, so this value is acceptable. Resolution RDC no. 88/16 (Anvisa) (Brazil 2016) sets a SML of 1.5 mg kg^{-1} for DEHP, 0.3 mg kg^{-1} for DIBP and DBP and absence of DIPN into food or food simulants. According to the data shown in Table 2, this analytical method has a LOD well below the SML values and is therefore adequate for these assays.

3.2 Evaluation of food samples

The retention times of the different studied substances by GC-MS method ranged from 9.2 to 24.1 min (Table 3). Identification of the substances detected in the samples was made by comparison of the retention times and mass spectra of the compounds studied in this work with analytical spectra standards from National Institute of Standards and Technology (NIST) library available at the GC-MS. In addition to the 3 evaluated phthalates, the chromatograms

Table 3 Compounds identified in foods analyzed by GC-MS

Sample	Substance	CAS number	Retention time (min)
Wheat flour A	Diisobutyl phthalate (DIBP)	84-69-5	11.6
	Bis-(2-ethylhexyl) phthalate (DEHP)	117-81-7	24.1
Wheat flour B	Diisobutyl phthalate (DIBP)	84-69-5	11.6
	Bis-(2-ethylhexyl) phthalate (DEHP)	117-81-7	24.1
Powdered chocolate A	----	----	----
Powdered chocolate B	Bis-(2-ethylhexyl) phthalate (DEHP)	117-81-7	24.1
Cornstarch	Diisobutyl phthalate (DIBP)	84-69-5	11.6
	Dibutyl phthalate (DBP)	84-74-2	13.4
Toast	----	----	----
Biscuit with sugar crystals	Diisobutyl phthalate (DIBP)	84-69-5	11.6

Table 4 Phthalate concentration in analyzed foods

Sample	Fat content (%)*	Concentration (mg kg^{-1})			
		DIPN	DIBP	DBP	DEHP
Wheat flour A[a]	1.6	n.d.	1.13 ± 0.19	n.d.	3.85 ± 4.19
Wheat flour B[b]	n.i.	n.d.	1.18 ± 0.20	n.d.	1.56 ± 0.14
Powdered chocolate A[b]	3.5	n.d.	n.d.	n.d.	n.d.
Powdered chocolate B[a]	10.0	n.d.	n.d.	n.d.	1.83 ± 0.37
Cornstarch[b]	n.i.	n.d.	1.51 ± 0.05	0.26 ± 0.02	n.d.
Toast[b]	12.0	n.d.	n.d.	n.d.	n.d.
Biscuit with sugar crystals[b]	2.0	n.d.	0.36 ± 0.04	n.d.	n.d.

Results expressed as mean ± standard deviation

n-values (a) duplicates and (b) triplicates

n.i. = non-indicated and n.d. = non detected

** According to label content of the samples*

showed several characteristic peaks of the analyzed foods, such as flavoring agents (vanillin and dodecanoic acid, 1-methylethyl ester), caffeine, sitosterol, essential fatty acid (9,12-octadecadienoic acid), cholesterol, etc. Only the powdered chocolate A and toast samples showed no contamination from any of the studied substances.

All the other food samples showed contamination by phthalates. DIBP was detected in 4 food samples (wheat flour A, wheat flour B, cornstarch and biscuit), DEHP was detected in 3 food samples (wheat flour A, wheat flour B and powdered chocolate B) and DBP was detected only in the cornstarch sample. These substances showed peak area ranging from 0.1 to 4% of total area, indicating product contamination.

The concentration of the target substances determined in the food samples is shown in Table 4. DIBP was detected in 4 food samples, where in both wheat flour samples the concentration of DIBP was approximately 1.2 mg kg^{-1} and in cornstarch the concentration was 1.5 mg kg^{-1}, and these values are 4 to 5 times higher than the SML established for DIBP in Resolution RDC no. 88/2016. In the biscuit sample, the DIBP concentration of 0.4 mg kg^{-1} was slightly above SML. In powdered chocolate samples and toast, DIBP concentration was lower than the LOQ (0.011 mg kg^{-1}). DBP was determined only in the cornstarch sample (0.3 mg kg^{-1}), whereas the concentration in the other food samples was lower than the LOQ (< 0.018 mg kg^{-1}). DEHP was determined in 3 food samples at a concentration of 3.8 mg kg^{-1} in the wheat flour A, 1.6 mg kg^{-1} in wheat flour B and 1.8 mg kg^{-1} in the powdered chocolate B, which are 2.5, 1.1 and 1.2 x higher than the SML established for DEHP in Resolution RDC no. 88/2016, respectively, while in the other samples the values were lower than the LOQ (0.050 mg kg^{-1}).

Due to the higher chemical affinity between phthalates and fatty products, migration of phthalates is more likely to occur into fattier foods, such as powdered chocolate A (3.5% fat content) and powdered chocolate B (10% fat content). In fact, when chocolate is in direct contact with the cellulosic packaging (powdered chocolate B) the presence of DEHP occurred. On the other hand, the chocolate sample that was not in direct contact with the cellulosic packaging (powdered chocolate A) did not show DEHP. This result can be attributed to the direct contact of the product with the plastic packaging (BOPP/PE). This result corroborates the findings from Jickells et al. (2005), which observed the primary packaging made of plastic material acts as a migration barrier to phthalates (when present in the material) from the secondary cellulosic packaging into the product. DEHP was also detected in higher proportion in high fat wheat flour A

(1.6% fat content) than wheat flour B, which had no fat indication in its composition, even with both samples of wheat flour packed in paper primary packaging.

The occurrence of DIBP was of the same order of magnitude for both wheat flour samples, regardless of the fat content of the products. DIBP also occurred in non-fat cornstarch. The primary packaging of all these foods was paper, being printed packaging for wheat flour and non-printed packaging for cornstarch. This last food sample was the only one that showed DBP migration, which could be attributed to set-off effect since DBP is mainly associated with inks and printing activities that could be transferred by contact from a printed surface to a non-printed packaging surface and then migrate into food.

In cases where food was contaminated by phthalates, the determined values were higher than the SML established by Resolution RDC no. 88/16 (Brazil 2016). This is concerning, as SML are established from toxicological studies and aim to protect consumers' health.

The food packaging evaluated in this study were analyzed for specific migration of phthalates into n-heptane fatty food simulant in a previous study (Coltro et al. 2021). The results showed that the migration of phthalates into the fatty food simulant was lower than the values quantified in the dry foods of the present study. In other words, the concentrations of DIBP in wheat flour, cornstarch and biscuit, DEHP in the powdered chocolate B and wheat flour and DBP in the cornstarch were higher than the migration values of their respective packages into the fatty simulant. Therefore, packaging is not the main source of contamination by phthalates in the different types of food evaluated, thus corroborating the results obtained by Holderbeke et al. (2014) and Park et al. (2016). Storage containers, processing equipment and filling lines are examples of food contact articles important for food production that can be a source of food contamination by phthalates and other chemicals (Muncke et al. 2020).

The presence of DIPN was not detected in any of the dry food samples evaluated in our study. These results corroborate the non-detection of DIPN migration from the packaging materials into the respective samples, reported in a previous study (Coltro et al. 2021). Therefore, all analyzed samples are considered safe in relation to the presence of this substance, as the cellulosic packaging was not a source of DIPN contamination for the evaluated foods. In contrast to Sturaro et al. (1994), where cellulosic packaging material was a source of DIPN contamination for rice, however, the detected concentration was not significant from a toxicological point of view. Furthermore, the authors emphasized that DIPN contamination can be avoided when food is packed in plastic bags, or by using an inner plastic bag in cardboard boxes.

The results for DIBP and DEHP migration of our study are of the same magnitude as those found in Belgium by Holderbeke et al. (2014). However, DBP migration was approximately 2 x higher. The values determined in the present study are higher than the values found in studies conducted in other European countries and in the USA (Nerín et al. 1993; Fierens et al. 2012; Poças and Hoog 2007; Poças et al. 2011b; Serrano et al. 2014; Zhang et al. 2008). According to Serrano et al. (2014), except for vegetables, the concentration of phthalates in food generally decreases after cooking, i.e., although DEHP is present in all raw foodstuffs evaluated, the percentage decreased to 65.4% after cooking.

Nevertheless, the results of our study indicate that better control is required in production processes in Brazil and more stringent good manufacturing practices to avoid food contamination and consumer exposure to these substances need to be adopted.

4 Conclusion

Seven food samples from different production batches were analyzed for 3 phthalates and DIPN in order to evaluate if these substances contaminate food in Brazil. Two food samples (powdered chocolate A and toast) did not contain any of the studied substances. The powdered chocolate A was packed in plastic primary packaging (BOPP/PE), which provides a better barrier than paper packaging, demonstrating the importance of the type of material used as primary packaging to reduce potential substance transfer from the secondary packaging into food. For the other food samples, 1 respectively 2 of the studied phthalates were detected; which may have migrated from phthalate-containing materials used in production and, to a lesser extent, from packaging. DEHP was found at the highest concentration among the samples, followed by DIBP. Since the determined values were > 2.5 to 5 of the migration limits established by the legislation, these results indicate the need for better process control and adoption of more stringent good manufacturing practices.

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Declarations

Competing interest The authors declare no competing financial/non-financial interests.

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