Migration of Phthalates from Cellulose Packaging into Food Simulant: Assessment of Different Levels of Contaminants

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Since paper recycling operates with market-collected paper scrap, this may lead to undesirable chemicals migrating from the wastepaper into the recycled material. Therefore, this study is based on the assessment of several additives inadvertently included in cellulose packaging that may migrate to foodstuff, considering that toxicological effects of some of these additives may persist in recycled cellulose fibers used to produce new food packaging. The purpose of this study is to determine migration of dibutyl phthalate (DBP), diisobutyl phthalate (DIBP), bis(2-ethylhexyl) phthalate (DEHP) and 2,6-diisopropylnaphthalene (DIPN) into fatty food simulant (n-heptane) by means of gas chromatography with flame ionization detector (GC-FID). For this purpose, commercial samples of cellulose food packages are contaminated with different concentrations of these additives, and migration into the food simulant is evaluated after the samples remain in contact with the food simulant for a prolonged time at maximum temperatures of 40 °C. The results of the assessment show that an amount ranging from 0.1–0.3% of phthalates and DIPN migrate from the fortified package into the fatty food simulant. The maximum levels of migration of these additives into the recycled cellulose packaging in order to comply with the specific migration limits for contact with foodstuff have been estimated.

1. Introduction

Paper and cardboard are used mostly to pack dry food and as secondary packaging. Moreover, coated or waxed paper and cardboard can be used to pack wet and fatty foodstuff. Technically, paper fibers can be recycled up to seven times. Recycled material is often preferred over paper and paperboard made from fresh fibers since recycling reduces waste, thus saving raw materials and energy, besides other environmental benefits.[1]

In the paper recycling stream, it is not common practice to sort the collected material into food-grade and non-food grade streams before recycling. After sorting recovered materials into different technical grades, they are mixed with water in a hydra pulper to produce a pulp. In this process, non-fibrous parts – textiles, tapes, etc. are removed. The pulp is then ground in a dispenser, water is removed in drum filters and fibers are cleaned via chemical, thermal and/or mechanical treatments. Bleaching and deinking can be applied to improve the appearance of the final product. Next, the recycled fibers are mixed with fresh fibers to preserve quality, and this mix is then processed in a paper machine to produce the final material.[1]

Taking the whole process into account, it can be concluded that chemicals from diverse origins are usually present in wastepaper and may eventually be carried over to the recycled material. Such chemicals include additives used in production and intended to be retained in the paper product, such as fillers, coatings, biocides etc. Besides that, paper is usually printed, dyed, glued and/or labeled, which may account for the existence of printing inks, adhesives, solvents, plasticizers, pigments etc. in the wastepaper.[1]

Therefore, various substances are added as additives to the packaging material during the manufacturing process to improve its characteristics, while other substances are unintentionally added due to the incorporation of recycled pulp.[2,3] In both cases, these substances should be considered as potential migrants.

The transfer of components from the packaging material to the packaged food is named migration. It may occur from the primary and secondary packaging material, even though in the latter case there is no direct contact with the food. In the case of primary packaging, substances in the packaging material may migrate into the food either via direct contact or indirectly through the gas phase between the packaging surface and the food surface as shown in **Figure 1**a,b.[4]

Migration may also come from the secondary packaging, which is usually made of cardboard or corrugated cardboard, although there is no direct contact with the food. Therefore, volatile and semi-volatile organic substances in these materials can permeate through the primary packaging and reach the food. Likewise, substances on the outer surface of the packaging, such as components of printing inks and varnishes, can permeate the cellulose matrix of the packaging and reach the food or else via setoff when the cardboard is wound and stored, so that the printed outer face touches the inner (non-printed) face, with the possibility of migration of components from the printing ink to the

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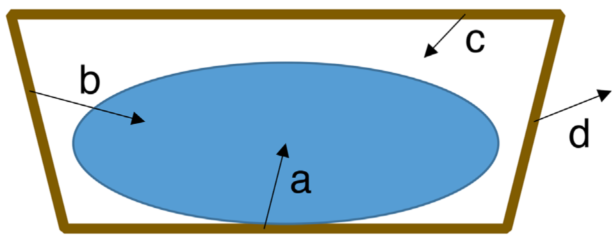

Figure 1. Mass transfer mechanisms in the packaging: (a) from the matrix fiber by direct contact; (b) from the packaging fiber by indirect contact; (c) from the outer surface through the matrix fiber; (d) from the fiber of the matrix to the external environment. Adapted with permission.[4] Copyright 2011, Elsevier.

Table 1. Maximum specific migration limits (SML) for cellulose materials intended to be in contact with foodstuff in packaging produced with inclusion of recycled fibers.

Substance	SML [mg kg^{-1}]	Remark
Benzophenone	0.6	–
Bisphenol A	0.6	a)
Di-ethylhexyl phthalate (DEHP)	1.5	–
Di-n-butyl phthalate (DBP)	0.3	The sum of DBP and DIBP cannot exceed 0.3 mg kg^{-1}
Diisobutyl phthalate (DIBP)	0.3	
4,4′ bis (dimethylamine) benzophenone	<0.01	a)
2,6-diisopropylnaphthalene (DIPN)	ND b)	–
Primary aromatic amines	ND b)	a)

a) Verification of specific migration of these compounds is required only for cellulose materials in contact with aqueous or fatty foods. b) ND = Cannot be detected.[5]

inner face and then to the packed food. Besides, a fraction of the substances may also be transferred from the cellulose matrix to the external environment. These mechanisms are shown in Figure 1c,d.[4]

In order to regulate the packaging market and to ensure safety both of consumers and of food, national and international health regulatory agencies have developed technical regulations specific for packaging materials. In Brazil, the Brazilian National Health Surveillance Agency (ANVISA) of the Ministry of Health (MH) regulates food-contact packaging, equipment and utensils. These regulations are harmonized among MERCOSUR (Southern Common Market) countries, so packaging materials can be freely traded among Argentina, Brazil, Paraguay and Uruguay.

On June 29, 2016, ANVISA published Resolution RDC no. 88/16 in Brazil. This resolution incorporates MERCOSUR GMC Resolution no. 40/15 into the Brazilian national legal system, which approves the technical regulation on materials, packaging and cellulose equipment intended for food contact and other arrangements.[5,6] This Resolution includes a positive list of components for food-contact materials, packaging, and cellulose equipment. It also specifies that food-contact cellulose material that includes recycled fibers in its production must comply with the maximum limits for specific migration (SML) of certain chemicals (**Table 1**) and of inorganic elements.

The approval to use recycled paper in food-contact packaging is of concern from the point of view of health of the Brazilian population, since recycling cellulose material is a traditional activity in Brazil. In 2011, 34.5% of cellulose scrap was used to manufacture food packaging, while 15.6% was used to produce corrugated paper sheets. Corrugated paper is one of the most widely used recycled packaging materials in the country; it accounts for 64.5% of recycled scrap in Brazil.[7] The use of scrap to manufacture food packaging is expected to increase after the approval of Resolution RDC no. 88/16, which may include the use of lower quality scrap.

Phthalates are the most frequent contaminants in cellulose packaging. Several studies report that di-2-ethylhexyl phthalate – DEHP – can produce toxic and adverse effects, particularly in animal or human tissues and organs, such as the pituitary gland, liver or testicles.[8–11]

A review on food monitoring and epidemiology by Serrano et al. identified 17 studies on concentrations of phthalate in food in North America, Asia and Europe between 1990 and 2013, and three studies on epidemiological associations.[12] In these studies, the authors observed high concentrations of DEHP in poultry, cooking oils and cream-based dairy products (≥300 µg kg^{-1}), which contribute significantly to exposure in epidemiological studies.

Yang et al. evaluated migration of phthalates from plastic packages into 283 convenience foods. DEHP was detected in samples of high-fat food ranging from below the limit of detection to 5.23 mg kg^{-1}. DBP ranged from 0.51 mg kg^{-1} in meat to 2.54 mg kg^{-1} in cake. The authors also found that the content of phthalates in convenience foods near their expiration date was much higher than the content of phthalates in recently manufactured and packed foods.[13]

Vandermarken et al. assessed estrogenic compounds in virgin and recycled paperboard used for dry food packaging. The authors found a relationship between estrogenic activity and the recycling rate of the paperboard. Recycled non-coated printed paperboard samples showed the highest migration levels, i.e., 0.77 mg kg^{-1} for DEHP, 0.26 mg kg^{-1} for DBP, 0.10 mg kg^{-1} for BBP and 0.84 mg kg^{-1} for DINCH. DBP migration showed values very close to the SML of 0.3 mg kg^{-1}, whereas the others showed migration quite lower than the SML.[14]

Asensio, Peiro and Nerín assessed migration of compounds from several types of cardboard cups used in coffee vending machines. They identified and quantified several substances from the cardboard material, from the plastic coating (LDPE) and from printing inks (set-off migration) of the cardboard cups. Besides compounds listed in Regulation no. 10/2011 that migrated at levels lower than the SML, the authors also detected migration of several non-listed and non-authorized compounds.[15]

Alp and Yerlikaya assessed migration of phthalate esters (DEHP, DBP, BBP, DINP, DIDP and DNOP) into samples of seafood packed in polypropylene (PP), polyvinyl chloride (PVC), tin cans and glass containers. The results indicated migration of phthalate esters from PVC, PP and tin cans. Only glass jars with no metal lids with PVC seals had no migration of phthalates. Among the phthalates, DEHP was quantified in high levels (up to 830.30 ng kg^{-1}) in most of the samples.[16]

Therefore, a case study was performed to assess management practices for recycling paper in order to produce food packages. The purpose of this case study was to assess migration of DIBP, DBP, DEHP and DIPN from cellulose packaging into fatty food simulant (n-heptane) according to Resolution RDC no. 88/16. For that reason, cellulose packages contaminated with different levels of these substances were evaluated, and an estimate was made of the concentration of these additives in the cellulose packaging that was enough to comply with the SML.

2. Materials and Methods

2.1. Reagents and Materials

The following reagents were used in this study: 2,6-diisopropylnaphthalene – DIPN, CAS number 24157-81-1 (Sigma-Aldrich, 99%); diisobutyl phthalate – DIBP, CAS number 84-69-5 (Sigma-Aldrich, 99%); dibutyl phthalate – DBP, CAS number 84-74-2 (Sigma-Aldrich, 99%); bis(2-ethylhexyl) phthalate – DEHP, CAS number 117-81-7 (Sigma-Aldrich, 99%) and n-heptane p.a. (Synth), used as fatty food simulant. Commercial samples of cellulose packaging (cardboard with grammage of 256 ± 2 g m^{-2}) were acquired in the market to be used to assess specific migration of phthalates and DIPN into fatty food simulant.

2.2. Equipment

The following instruments were used: Sartorius analytical balance, model MSU225-1CE-DU, with 0.0001-g resolution; Fisatom rotary evaporator, model 450–5; Fisatom 826T vacuum pump and Fisatom 860 refrigerated heating bath. Chromatographic analyses were performed using a gas chromatograph with flame ionization detector (GC-FID), model 7890A manufactured by Agilent Technologies, operating with DB1 capillary column (30 m long × 0.25 mm internal diameter × 0.25 µm film thickness).

2.3. Specific migration from cellulose package into fatty food simulant

Square specimens (5 cm × 5 cm) were cut out from samples of cellulose packages (cardboard), and then contaminated with 18, 36 and 54 mg of DIPN and phthalates kg^{-1} packaging material by pipetting 115, 230 and 345 µL of 100 mg kg^{-1} DIPN, DIBP, DBP and DEHP solution, respectively, in n-heptane. After fully drying in an exhaust chapel, the specimens were dipped in n-heptane food simulant at a ratio of 0.3 mL cm^{-2} of the surface under analysis. Both sides of the material were considered in the calculations. Three specimens were placed in contact with 50 mL of fatty food simulant (n-heptane) at 20 °C ± 1 °C for 30 min + 1 min for the contact condition of long-term storage at maximum temperatures of 40 °C. A minimum of twelve specimens were analyzed, totaling an area of 600 cm^2 (considering both sides of the material).[5] A blank sample was also prepared. At the end of the contact time, the specimens were removed; the food simulant volume was reduced to 2 mL and then injected in triplicate into the GC-FID. The chromatographic conditions followed the method developed by Coltro et al.[17]

Equation 1 was applied in order to calculate specific migration of phthalates and DIPN in mg kg^{-1}:

$$SM = \frac{m\ x\ S}{A\ x\ M} \tag{1}$$

Where SM represents the specific migration of substance or element per kilogram of food, expressed in mg kg^{-1}; m is the mass of substance or element in the migration extract, expressed in mg; A is the total area of the sample in contact with simulant, expressed in dm^2; S/M is the ratio of the contact area of the cellulose material (S) to the mass of food (M), expressed in dm^2 kg^{-1}. When the mass of food is unknown, the mass of water corresponding to the volume of the package, expressed in kg, is used. When the real S/M ratio for a cellulose material is unknown, the S/M ratio = 6 dm^2 kg^{-1} should be used.[5]

2.3.1. Statistical analysis

Variance analysis was applied to the results, and Tukey test was used to determine statistically significant differences ($p < 0.05$) among averages using Excel + Action 2.9 software program.

3. Results and Discussion

Phthalates have been regularly quantified in food packages made of recycled paper and paperboard. Such phthalates typically come from printing inks, lacquers and adhesives included in the paper.[18,19] Some of the phthalates that are often identified are DEHP, DBP, DIBP, BBP and DEP, and their migration contributes to increase levels of phthalate in food.[1,12,18,20] Migration of diisopropylnaphthalene – DIPN – from recycled paper and paperboard packaging has been also measured in several studies.[1,21] The source of DIPN is the incorporation of recycled office paper into food packaging since DIPN is employed as solvent in carbonless copy paper, which has replaced formerly used polychlorinated biphenyls.[22]

However, the existence of such substances in cellulose packages in the Brazilian market has never been assessed because the legislation authorized the use of recycled cellulose packaging for contact with food only in 2018. Therefore, in order to evaluate migration of DIBP, DBP, DEHP and DIPN from cellulose packaging into fatty food simulant (n-heptane) according to Resolution RDC no. 88/16, fortified samples at three concentration levels (low, medium and high) were placed in contact with food simulant which was afterwards injected in triplicate in GC-FID (gas chromatography with flame ionization detection). GC is one of the most widely used methods to determine migration of phthalates. The chromatograms showed peaks at 14.0, 14.7, 15.2 and 18.5 min, corresponding to retention times of DIPN, DIBP, DBP and DEHP, respectively.

The study was conducted with fatty food simulant (n-heptane) since this is the most critical simulant for migration of these substances due to the chemical affinity between these additives and the simulant.

Migration of the additives under study into fatty food simulant was detected at all concentrations of the samples of fortified cellulose packaging under assessment (**Table 2**). The results demonstrated that approx. 0.1% to 0.3% of the additives added to the sample migrated into the fatty food simulant when comparing the concentration of additives that migrated into food simulant and the concentration of additives added to the sample of cardboard.

Table 2. Migration of phthalates and DIPN from cellulose packaging into n-heptane food simulant at 20 °C, for 30 min (mg kg^{-1}).

Fortified sample [mg kg^{-1}]	DIPN Mean ± SD Migrated (%)	DIBP Mean ± SD Migrated (%)	DBP Mean ± SD Migrated (%)	DEHP Mean ± SD Migrated (%)
18	0.023 ± 0.002[a]	0.045 ± 0.003[b]	0.040 ± 0.003[b]	0.050 ± 0.007[c]
	0.1	0.3	0.2	0.3
36	0.057 ± 0.006[a]	0.081 ± 0.009[b]	0.078 ± 0.005[b]	0.103 ± 0.013[c]
	0.2	0.2	0.2	0.3
54	0.095 ± 0.007[a]	0.118 ± 0.005[b]	0.114 ± 0.005[b]	0.139 ± 0.010[c]
	0.2	0.2	0.2	0.3

Mean ± standard deviation of four experimental determinations. Different letters in the same line indicate significant differences ($p < 0.05$).

Table 3. Estimation of maximum concentration of additives in cellulose packaging to reach the specific migration limit – SML (mg kg^{-1}).

Additive	Concentration	
	In the cellulose packaging	SML
DIPN	12	0.01
DIBP	71	0.15[a)]
DBP	73	0.15[a)]
DEHP	285	1.5

[a)] Estimated 50% SML for DIBP and DBP, since the sum of both cannot exceed 0.3 mg kg^{-1}.

For all concentrations evaluated, DIBP and DBP displayed the same migration rate, which is probably due to the similarity of the chemical structures of these additives. DIPN and DEHP showed the lowest and highest migration rates, respectively (no significant difference at 95% confidence, Tukey test), probably due to the rigidity of the two aromatic rings of DIPN ($C_{10}H_6R_2$, with R = $CH(CH_3)_2$) and the linear chain of DEHP ($C_6H_6(C = OOR)_2$, with R = $CH_2CH(CH_2CH_3)(CH_2)_3CH_3$) as opposed to the volume of DIBP ($C_6H_6(C = OOR)_2$, with R = $CH_2(CH_3)_3$) and DBP ($C_6H_6(C = OOR)_2$, with R = $(CH_2)_3CH_3$) molecules.

The SML set forth by Resolution RDC no. 88/16 for DIPN is undetectable.[5] According to EU Regulation no. 10/2011, "undetectable" refers to the detection limit of 0.01 mg kg^{-1}.[23] Migration of DIPN from the fortified samples with the lowest concentration of additives (18 mg kg^{-1}) was 0.023 mg kg^{-1}, which exceeds the SML for this substance. This implies restrictions to use these cellulose packages for contact with fatty foods in respect to this requirement. This result corroborates the results obtained by Zhang, Noonan and Begley and Jickells et al., who detected migration of DIPN into food when concentration of DIPN in paper reached 20 mg kg^{-1}.[24,25]

Table 2 shows that the migration levels of lighter phthalates (DIBP and DBP) into food simulant were lower than migration levels of heavier phthalate (DEHP). These results confirm the results obtained by Poças et al., who studied migration of several phthalates with different molecular sizes from paper and paperboard into solid food simulant.[4] According to the authors, the variation of relative concentration of phthalates as a function of their molecular sizes can be described by a second order polynomial function. The explanation for this behavior is based on two aspects: (1) molecules with higher boiling points migrated less due to their larger molecule size, and (2) the lighter molecules evaporated to the surrounding air at larger rates, resulting in higher losses from the system.

The results are also in accordance with the results obtained by Biedermann-Brem et al. who performed a migration experiment with recycled paperboard containing 7.5 mg kg^{-1} DIBP. Migration was 0.76 mg kg^{-1}, i.e., 10% of the paperboard content.[26] Therefore, considering an internal bag limiting the migration to 1% of the content in the paperboard and a standard mass ratio of food to paperboard of 10, migration is reduced to 0.008 mg kg^{-1}.

Park et al. assessed 19 samples of paper cups in which they detected five phthalates, among them DBP at 0.07 to 3.14 mg kg^{-1} and DEHP at 0.45 to 58.56 mg kg^{-1}. The migration levels of these phthalates into n-heptane used as food simulant stored in paper cups at 25 °C for up to 120 min were: 0.023 to 0.032 mg kg^{-1} for DBP, and <LOQ to 0.015 mg kg^{-1} for DEHP. Therefore, migration levels of DBP were higher than migration levels of DEHP despite the higher level of DEHP quantified in the samples of paper cups.[27] These results are opposite to those obtained in this study, since the migration levels of DBP found in this study were lower than migration levels of DEHP.

Concentrations of DIPN from recycled carbonless copy paper are usually in the range of 10 to 50 mg kg^{-1} in recycled paperboard.[24] According to Pivnenko et at., a range of concentrations of phthalates was quantified in paper, being DIBP the highest one (up to 120 mg kg^{-1}).[28] A study carried out by German authorities detected presence of phthalates up to 35 mg kg^{-1} in board and wastepaper from offices, mostly papers containing relatively high amounts of glue. This denotes that adhesives are the primary source of phthalates in paper to be recycled.[29]

For the phthalates assessed in this study, all mean values of migration observed are below the SML set forth by Resolution RDC no. 88/16 for these additives. [5] Therefore, for maximum concentrations of 54 mg of DIBP, DBP and DEHP kg^{-1} of cellulose packaging, there are no restrictions to use cellulose packages for contact with fatty foods in respect to this requirement.

Linear regression and/or second order polynomial function were applied to the results of DIPN and phthalates migrated into fatty food simulant shown in Table 2, as follows: DIPN: $y = 0.002x – 0.0137$ ($R^2 = 0.9999$); DIBP: $y = 0.002x + 0.0085$ ($R^2 = 0.9999$); DBP: $y = 0.002x + 0.0036$ ($R^2 = 0.9999$); DEHP: $y = -3E\text{-}05x^2 + 0.0043x – 0.0192$ ($R^2 = 1$).

Applying these regressions to the results allowed estimating the concentration of these additives in the cellulose packaging to comply with the SML, as shown in **Table 3**. These concentrations correspond to maximum contamination by additives of cellulose packaging to be approved for food contact, according to the requirements for SML.

The SML for DIBP and DBP was estimated as 50% of the value established by the legislation, since these additives have similar migration rates and the sum of both cannot exceed the SML of 0.3 mg kg^{-1}.

The results suggest potential for contamination of food by DIPN above the 10 mg kg^{-1} level of cellulose packaging, contamination by DIBP and DBP above the 70 mg kg^{-1} level of cellulose packaging, and contamination by DEHP above the 300 mg kg^{-1} level of cellulose packaging. As the SML of DEHP is much higher than that of the other substances, the relative concentration of DEHP is higher as well.

The fact that cellulose material may possibly contain maximum contamination of 300 mg kg^{-1} for DEHP and yet comply with the SML of 1.5 mg kg^{-1} is positive from the point of view of consumer safety. DEHP is the phthalate most used as a plasticizer due to its excellent performance and reduced cost, which increases the possibility of this phthalate being present in waste cellulose material.

4. Conclusion

Migration of DIBP, DBP and DEHP from samples of cellulose packaging contaminated with up to 54 mg kg^{-1} was lower than the SML set forth by Resolution RDC no. 88/16; therefore there are no restrictions to use these packages for contact with fatty foods in respect to this requirement. Migration of DIPN from samples of cellulose packaging contaminated with 18 mg kg^{-1} was higher than the SML, with restrictions to use these packages for contact with fatty foods in respect to this requirement.

Between 0.1% and 0.3% of the concentration of phthalates added to cellulose packaging migrated to the n-heptane fatty food simulant, which reduces concerns about possible contamination in cellulose packaging that includes recycled fibers. The maximum concentration of these additives in the cellulose packaging so that they comply with the SML was estimated to be 10 mg kg^{-1} level for DIPN, 70 mg kg^{-1} level for DIBP, and DBP and 300 mg kg^{-1} level for DEHP. These results indicate that good control of cellulose scrap to produce recycled food-contact packaging can ensure compliance with the specific migration limits established by the current legislation.

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Figure 1 was adapted from Food Control, 22, M. F. F. Poças, J. C. Oliveira, J. R. Pereira, R. Brandsch, T. Hogg, Modelling migration from paper into a food simulant, 303–312, Copyright 2011, with permission from Elsevier.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

additives, cellulose, food packaging, migration, phthalate

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