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Analysis of Phthalate Migration from Plastic Containers to Packaged Cooking Oil and Mineral Water

Qian Xu,*,†,‡,§ Xueyan Yin,†,§ Min Wang,†,§ Haifeng Wang,# Niping Zhang,†,§ Yanyan Shen,†,§ Shi Xu,† Ling Zhang,† and Zhongze Gu*,‡,§

†Ministry of Education Key Laboratory of Environmental Medicine and Engineering, Southeast University, Nanjing 210009, China, ‡State Key Laboratory of Bioelectronics, Southeast University, Nanjing 210096, China, §Suzhou Key Laboratory of Environment and Biosafety, Suzhou 215123, China, and #School of Food Science and Engineering, Nanjing University of Finance and Economics, Nanjing 210003, China

The migration of phthalates (PAEs), a class of typical environmental estrogen contaminants in food, from food packaging to packaged food attracts more and more attention worldwide. Many factors will affect the migration processes. The purpose of this study was to evaluate PAE migration from plastic containers to cooking oil and mineral water packed in authentic commercial packaging and stored under various conditions (different storage temperatures, contact times, and storage states (static or dynamic state)) and to identify a potential relationship between the amount and type of PAEs migrated and the lipophilic character of the food matrix. The samples were analyzed by a novel method of liquid chromatography combined with solid-phase extraction by an electrospun nylon 6 nanofibers mat, with PAE detection limits of 0.001 μg/L in mineral water and 0.020 μg/L in cooking oil, respectively. The results demonstrated that the cooking oil was a more suitable medium for the migration of PAEs from packages into foodstuffs than mineral water. Scilicet, the migration potential of the PAEs into foodstuffs, depends on the lipophilic characteristics of the food matrix. The results also demonstrated that migrations were more significant at higher temperature, longer contact time, and higher dynamic frequency; thus, the migration tests should be evaluated with consideration of different storage temperatures and contact times. Mathematical models with good logarithmic relationships were established to demonstrate the relationship between the PAE migration and food/packaging contact time for different storage temperatures. These established mathematical models would be expected to become a set of practical tools for the prediction of PAE migration.

KEYWORDS: Migration; cooking oil; mineral water; PAEs; mathematical modeling

INTRODUCTION

Because foods are the major sources of exposure to contaminants, the contamination of foodstuffs with undesirable substances migrated from the packaging materials receives more attention in recent years. The common food-packaging materials are considered as contaminant sources for a variety of substances. Different materials are used to package foodstuffs. The main focus of this paper is on plastic packaging materials, due to their wide use. Plastic food packages may contain additives used to minimize degradation during processing, to facilitate processing, and to increase stability during storage (*1*). Additives, such as antioxidants, dyes, pigments, antifogging agents, stabilizers, and plasticizers, are generally present at low levels but may migrate into the packaged food and then be ingested by the consumers (*2*, *3*).

Phthalates (PAEs) are widely used as additives in plastic products. Higher molecular weight PAEs, such as di-2-ethylhexyl PAEs (DEHP), are primarily used as plasticizers, whereas lower molecular weight PAEs, such as diethyl PAEs (DEP), di-*n*-butyl PAEs (DBP), and butyl benzyl PAEs (BBP), are widely used as solvents to hold color and scent (*4*). Because of their estrogenic properties and potential negative impact on human health, human exposure to PAEs migrated from plastic food containers has gained an increased concern.

Food packaging can interact with the packaged foodstuffs by migration processes, which mainly depend on the chemical and physical nature of the food contact materials and foodstuffs, the surface area of the packaging material in contact with the foodstuff, the time and temperature of the contact, and the type of packaging material (*5*). For a certain type of packaging materials, for example, plastic food packages, the evaluation of the migration characteristic of PAEs from the plastic food packages to the foodstuff should take into consideration the dynamic frequency of interaction between food packaging and the packaged foodstuff during storage, contact time, storage temperature, and the lipophilic character of the food matrix. Most migrants have a greater migration rate into oily and fatty media due to their

*Corresponding authors [(Q.X.) e-mail q_xu@163.com; (Z.G.) e-mail gu@seu.edu.cn].

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organic nature (*6*). Cooking oil and mineral water are considered to be representative fatty and aqueous foods, respectively. Migration of phthalates from plastic containers into soft drinks and mineral water has been published (*7*). Migration of PVC plasticizers/additives from the gaskets of metal closures was reported (*8*). Migration of plasticizers from PVC gaskets of lids for glass jars (*9*) and migration of PAEs to infant food packed in recycled paperboard were also studied (*10*). In most of the studies, olive oil was used to simulate fatty foods, and a storage condition of 10 days at 40 °C was used to simulate long-term storage under nominal conditions. To our knowledge, no report on PAE migration from plastic containers to cooking oil has been published to date.

Because the actual contaminant levels migrated from food contact materials need to be assessed over time, the use of food simulants might lead to an underestimation of actual migration into food (*11*, *12*). Therefore, in this study, we evaluated authentic commercial samples stored under different conditions.

Due to the low abundance of contaminants and the complexity of the food sample matrices, sample extraction and cleanup prior to analysis are necessary and critical. Although liquid−liquid extraction is employed by most laboratories, solid-phase extraction (SPE) of PAEs was proposed in recent years due to its low consumption of organic solvents, simplicity, high recovery, high preconcentration factors, and ease of automation and operation (*13*−*18*). To achieve higher extraction efficiency of target analytes, the choice of suitable adsorbents for SPE is very important. Recently, nanofibers as adsorbents for SPE have attracted much attention (*19*−*23*). Compared with conventional microscale adsorbents, nanofibers have high surface-to-volume and length-to-diameter ratios that can achieve larger specific surface and more active sites to enable more efficient adsorption. Accordingly, the attachment of the target molecules would be facilitated, and therefore a small amount of nanofibers is sufficient for the extraction process, which greatly reduces the use of desorption solvent. Nanofibers can be easily produced by a process commonly known as electrospinning (e-spinning) (*24*). Electrospun nanofibers are usually obtained in the form of a membrane or so-called "mat". The major advantages of the membrane or mat are large medium cross-sectional area and decreased back pressure, which allow for sample processing at higher flow rates, easier processing of large volume of samples, and achievement of better enrichment coefficient and lower detection limits for targeted analytes.

A novel SPE procedure using a nylon 6 electrospun nanofiber mat as sorbent, coupled with HPLC-UV for simultaneous determination of six PAEs in marketed milk, has been developed and validated in our laboratory. Compared with the literature reported methods, the new method has lower detection limits, better reproducibility, and higher recoveries. Moreover, PAEs in marketed milk samples can be completely extracted by only 2.5 mg of a nylon 6 nanofiber mat, and the consumption of organic solvent is also minimized (*22*). The feasibility of using this method for the determination of PAEs in other food types is under further investigation.

The purpose of this work is to determine the level of each of the eight PAEs present in cooking oil and mineral water packaged in plastic containers using HPLC-UV coupled with the nylon 6 electrospun nanofiber mat SPE procedure, as well as to evaluate the migration rates and establish mathematical models for different storage conditions. The eight PAEs are dimethyl PAEs (DMP), diethyl PAEs (DEP), butyl benzyl PAEs (BBP), di-*n*-butyl PAEs (DBP), di-(2-ethylhexyl) PAEs (DEHP), dioctyl PAEs (DOP), diisononyl PAEs (DINP), and diisodecyl PAEs (DIDP). Evaluations are performed for different storage temperatures, contact times, and dynamic frequencies to study the migration under these conditions. As mentioned previously, migration experiments were performed on authentic commercial samples in this study to examine the possible transfer rate and migration characteristics of PAEs from plastic containers into the food matrix.

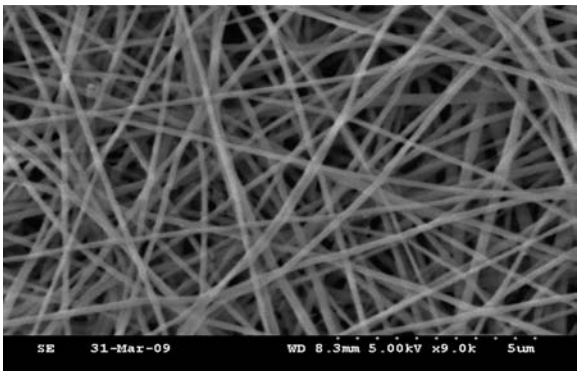

Figure 1. Scanning electron microscope images of nylon 6 nanofibers.

MATERIALS AND METHODS

Reagents and Chemicals. Cresol and formic acid were analytically pure and purchased from Chemical Reagent Factory, Shanghai, China. HPLC-grade methanol, acetone, and acetonitrile were purchased from Tedia Co. Inc., USA. Alcohol absolute was analytically pure and purchased from Yasheng Chemical Co., Wuxi, China. DMP, DEP, BBP, DBP, DEHP, DOP, DINP, and DIDP standards were purchased from Lingfeng Chemical Co., Shanghai, China. Nylon 6 material was purchased from Debiochem, China. The water used throughout the experiments was double distilled.

A standard stock solution containing eight PAEs was prepared in methanol with a concentration of 100 μg mL^{-1} for each PAE and stored under refrigerated conditions. Fresh working solutions were prepared daily by serial dilutions of the stock solution with purified water. In all cases, the methanol concentration in the solution before SPE was restricted to <1.00% (v/v).

Instrumentation. The HPLC used in this study has an LC-10AD pump and an SPD10-Avp UV detector and utilizes CLASS-VP software package for data analysis (Shimadzu, Japan). A reversed-phase C18 column (150 mm × 4.6 mm i.d., particle size = 5 μm, Dikma, China) was used for separation at 30 °C. The eight PAEs were chromatographically separated by running a linear gradient of acetonitrile in water (60−100% acetonitrile for 0−15 min followed by 100% acetonitrile for 15−35 min) for 40 min. The flow rate, injection volume, and detection wavelength were 1.0 mL min^{-1}, 20 μL, and 230 nm, respectively.

Preparation of Nylon 6 Nanofiber Mat. Nylon 6 nanofibers were fabricated by electrospinning according to the reported procedure (*25*). In brief, an appropriate amount of nylon 6 was dissolved in a composite solvent of formic acid and *m*-cresol (6:4, v/v). This solution was loaded into a 5 mL glass syringe. The glass syringe was fitted to a stainless needle (0.5 mm in diameter) with a flat tip connected to the anode. The aluminum foil collecting screen was connected to the cathode, and the distance between the needle tip and the grounded aluminum foil collection screen was approximately 20 cm. A voltage of 15 kV (DW-P403-1AC high-voltage generator, Dongwen Factory, Tianjing, China) was applied between the tip and the aluminum foil. The moving rate of the plunger was fixed at 0.5 mL h^{-1} and controlled by a syringe pump (model TCI-I, SLGO, Beijing, China). A dense membrane of nylon 6 nanofibers with thickness in the range of 120−150 μm was collected on the aluminum foil after 10 h of spinning. A scanning electron microscope (Hitachi S-3000N, Japan) was utilized to characterize the nylon 6 nanofibers (**Figure 1**)

Sample Preparation. Extraction was performed as follows: a homemade SPE device is shown in **Figure 2**. One piece of nylon 6 nanofiber mat was accurately cut into a circular shape with diameter of approximately 20 mm and attached tightly to the filter. The nanofiber mat was preconditioned with 200 μL of acetone and 200 μL of water twice each, and then with 200 μL of methanol and 200 μL of water once each.

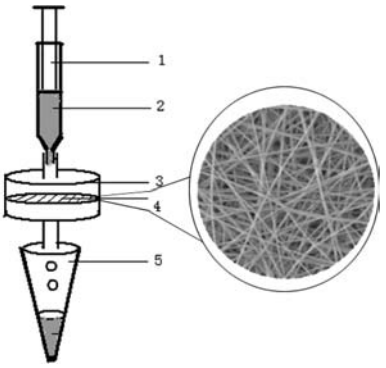

Figure 2. Fiber filter solid-phase extraction device: gastight syringe (1); sample solution (2); filter (3); electrospun nylon 6 nanofiber mat (4); collecting tube (5).

Table 1. Migration Test Conditions

food item	storage state	storage temperature (°C)	storage time (days)
cooking oil	static	20, 40, 60	2, 5, 10, 15, 30, 45, 60
	dynamic	20	2, 5, 10, 15, 30, 45, 60
mineral water	static	4, 20, 40	2, 5, 10, 15, 30, 45, 60
	dynamic	20	2, 5, 10, 15, 30, 45, 60

Fifty milliliters of mineral water samples was passed through the nylon 6 nanofibers mat at an appropriate flow rate. The analytes were subsequently eluted with 100 μL of acetone, which was fully evaporated with a gentle N_2 flow at room temperature and reconstituted in 50 μL of mobile phase. The extracted analytes were then analyzed by HPLC-UV with an injection volume of 20 μL.

Five milliliters of cooking oil was extracted with 5 mL of 85% alcohol and shaken for 10 min. The extraction was repeated once. The alcohol phases were separated by centrifugation (2000 rpm for 10 min), pooled, and fully evaporated with a gentle N_2 flow at room temperature. The residue obtained was reconstituted with 10 mL of double-distilled water containing 1% methanol and passed through the nylon 6 nanofiber mat at an appropriate flow rate. The analytes were subsequently eluted with 100 μL of acetone, which was fully evaporated with a gentle N_2 flow at room temperature and reconstituted in 50 μL of mobile phase. The extracted analytes were then analyzed by HPLC-UV with an injection of 20 μL.

Plastic containers of samples were cut into small pieces of equal sizes (about 5 × 5 mm), which were then mixed together; 0.2 g of the cut pieces was dissolved in 50 mL of *n*-hexane and then extracted ultrasonically for 30 min. The supernatant was directly injected into HPLC.

Blank Controls. Due to the ubiquitous presence of PAEs in the environment and samples, the analysis of these compounds was complicated by the lack of appropriate blanks. Therefore, special precaution was taken regarding the experimental control: laboratory glassware was consecutively rinsed with ethyl acetate, iso-octane, and purified water twice each before use. Because the content levels of PAEs were unknown in commercial test samples, we used cooking oil and mineral water of the same brand name and lot number to minimize sample to sample variability. Prior to the migration tests, the PAE levels in the cooking oil and mineral water samples were measured and used as the baseline values for blank controls.

Migration Tests. The migration tests were performed over a period of 2 months at static state (20, 40, and 60 °C) and dynamic state (20 °C) on authentic commercial samples to obtain representative migration information (**Table 1**). The dynamic frequency was 50 times/min for 5 min daily for 2 months. The objective of these tests was to study the migration of PAEs from plastic containers into cooking oil and mineral water under different storage states (static and dynamic state), storage temperatures, and contact times. In addition, cooking oil and mineral water were representative fatty and aqueous foods, and PAEs might exhibit different migration rate and characteristics when interacting with these two different media. All of the experiments were conducted simultaneously, and each test was run in triplicate.

Table 2. Linear Range, Correlation Coefficient (*r*), Repeatability (RSD), and Limit of Detection (LOD) of Methods in Different Samples (n = 6)

sample	analyte	linear range (ng mL^{-1})	linearity (*r*)	repeatability (RSD %)	LOD (ng mL^{-1})	recovery (%)
mineral water	DMP	0.01−20.0	0.9995	5.13	0.001	94.78
	DEP	0.01−20.0	0.9994	4.72	0.002	93.24
	BBP	0.02−20.0	0.9994	5.87	0.005	90.56
	DBP	0.02−20.0	0.9997	5.93	0.005	88.37
	DEHP	0.06−20.0	0.9993	6.38	0.008	96.28
	DOP	0.06−20.0	0.9998	6.92	0.020	100.39
	DINP	0.06−20.0	0.9995	5.76	0.020	97.62
	DIDP	0.06−20.0	0.9996	3.82	0.020	98.34
cooking oil	DMP	0.10−100.0	0.9994	5.72	0.020	92.13
	DEP	0.10−100.0	0.9995	6.03	0.030	94.08
	BBP	0.30−100.0	0.9997	6.58	0.080	89.68
	DBP	0.30−100.0	0.9993	7.03	0.080	85.92
	DEHP	0.30−100.0	0.9995	5.34	0.100	101.03
	DOP	0.50−100.0	0.9995	5.82	0.150	99.72
	DINP	0.50−100.0	0.9996	4.93	0.150	96.75
	DIDP	0.50−100.0	0.9997	4.61	0.150	95.48

RESULTS AND DISCUSSION

Selection of Foodstuffs. According to European Union (EU) regulations (*26*, *27*), foods are mainly divided into water foods, alcohol foods, acidic foods, and fatty foods. Cooking oil was chosen because it has a high fat content. In addition, it is an essential food in people's daily lives. Mineral water has served as a model foodstuff because it is a simple matrix and does not contain endogenous hormones, like, for example, dairy products. Moreover, consumption of mineral water is increasing worldwide. The components of target compounds present in these two matrices were totally different, thus providing interesting contrast against each other.

Evaluation of Analytical Method. Identification of the target compounds was accomplished by comparing the retention times against those of known standards. Linear range, limit of detection, repeatability, and recovery were obtained and are shown in **Table 2**. Example HPLC chromatograms of the samples are shown in **Figure 3**.

For the cooking oil samples, good linearity was achieved in the ranges of 0.10−100.0 ng mL^{-1} for DMP and DEP, 0.30−100.0 ng mL^{-1} for BBP, DBP, and DEHP, and 0.50−100.0 ng mL^{-1} for DOP, DINP, and DIDP with correlation coefficients (*r*) ranging from 0.9993 to 0.9997. The LODs based on signal-to-noise ratio (S/N ≥ 3) for the eight PAE compounds ranged from 0.020 to 0.150 ng mL^{-1}. The repeatability of the method was determined by spiking the test samples with PAE standards at the concentration of 0.5 ng mL^{-1} for each analyte. The RSD was <7.03% for these compounds.

For the mineral water sample, the detection limit of the target compounds ranged from 0.001 to 0.020 ng mL^{-1} (S/N ≥ 3). The method precisions were evaluated by six repetitive analyses of 50 mL of mineral water sample containing standard PAEs at the concentration of 0.5 ng mL^{-1} for each analyte. The RSDs were below 6.92%, and recoveries were above 88.37% for all compounds. The linearity study was carried out at six concentration levels in the ranges of 0.01−20.0 ng mL^{-1} for DMP and DEP, 0.02−20.0 ng mL^{-1} for BBP and DBP, and 0.06−20.0 ng mL^{-1} for DEHP, DOP, DINP, and DIDP. Coefficients of correlation (*r*) ranged from 0.9993 to 0.9997.

Migration Tests. The EU legislation on food contact materials is based on the assumption that migration estimation should be conservative, and thus actual migration values should be

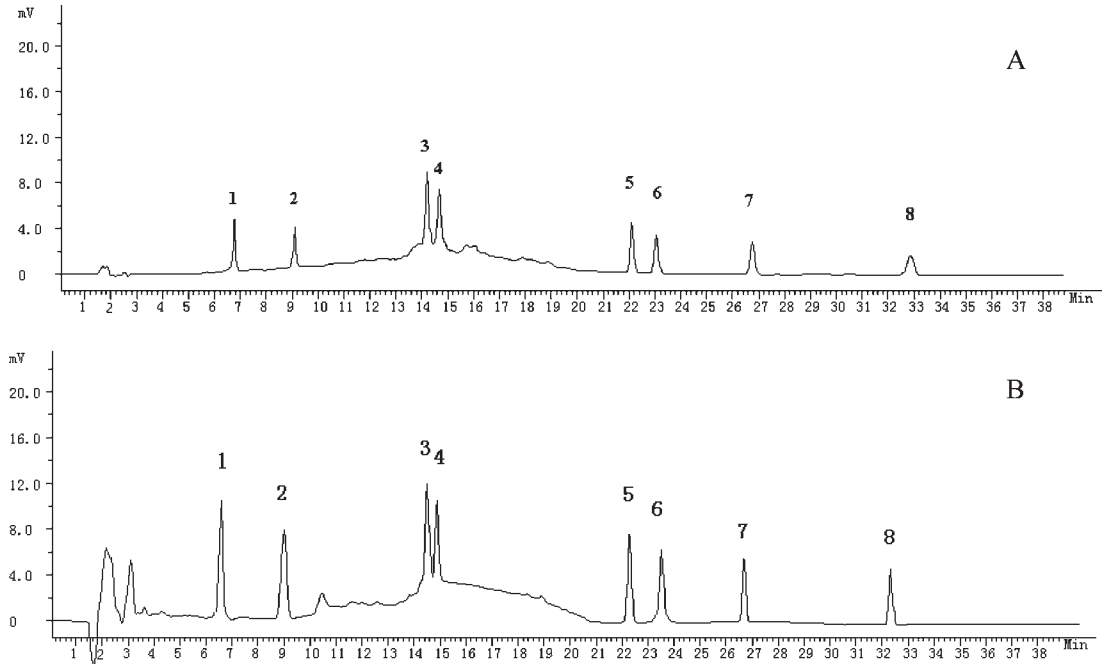

Figure 3. HPLC chromatogram of mineral water (**A**, spiked with 0.5 ng mL^{-1} of each PAE standard) and cooking oil (**B**, spiked with 1.0 ng $mL^{-1}$of each PAE standard). Peaks: 1, DMP; 2, DEP; 3, BBP; 4, DBP; 5, DEHP; 6, DOP; 7, DINP; 8, DIDP.

Table 3. Overview on the PAE Content (ng/mL) of Samples as Blank Value

blank sample	DMP	DEP	BBP	DBP	DEHP	DOP	DINP	DIDP
mineral water	0.23	0.58	1.25	5.62	0.93	3.58	2.84	2.36
cooking oil	3.18	6.72	10.83	14.93	17.47	4.81	3.02	4.70

overestimated to ensure consumer health (*28*). According to the current EU legislation, the migration test carried out at 40 °C for 10 days is the most stringent for any foodstuff stored at room temperature and should thus yield the highest degree of migration. We questioned these popular time/temperature conditions for migration tests and studied the migration of PAEs from plastic packages into foodstuffs stored at room temperature (20 °C) and 40 and 60 °C for a period of 2 months. Storage state was also taken into consideration to examine the difference between the samples stored in the static and dynamic states. Some of the conditions were more stringent than those required by EU legislation.

In the current study, mineral water and cooking oil samples were analyzed with HPLC-UV after SPE by nylon 6 nanofiber mat. Food samples were tested for levels of PAE contaminants migrated from plastic containers. The PAE levels in the samples before the migration test were measured and used as values for blank controls. In the majority of the blank controls, the target compounds could be detected and quantified, suggesting that the contaminants may come from food processing prior to packaging or migrate from packaging before the migration experiments. A detailed overview of the results for blank controls is given in **Table 3**. These results indicate that DBP had shown relatively high abundance in both food matrices and thus was considered as the major contaminant for data analysis in the subsequent experiments. BBP, DEHP, and DOP had also shown considerable levels in the blank controls.

The migration of PAEs is summarized in **Figure 4** for the cooking oil and mineral water stored at 20 °C for a period of 2 months. Samples were tested for PAE level after storage of 2, 5, 10, 15, 30, 45, and 60 days. The results demonstrated a rather rapid migration process of the PAEs. PAEs reached significant levels

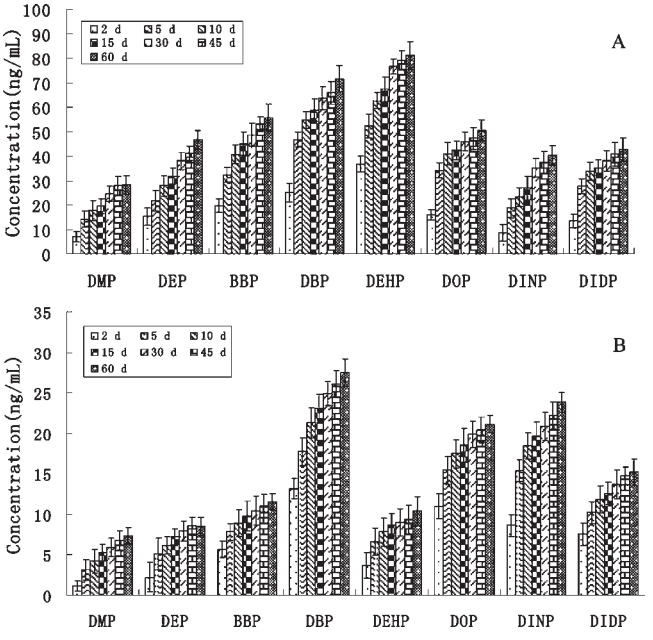

Figure 4. Migration of PAEs into cooking oil (**A**) and mineral water (**B**) at 20 °C for 2 months (n = 3).

after 10 days of migration. For the major PAEs, an equilibrium corresponding to a saturation of the food phase had already been reached after 1 month. On the one hand, it was noteworthy that the content of PAEs in the cooking oil was always higher than that in mineral water, which was confirmed throughout the whole migration experiment (DBP, for example, as shown in **Figure 5**). On the other hand, one should also note that under the applied migration conditions the migration potential of the PAEs into foodstuffs depended on not only the lipophilic characteristic of the food matrix but also the molecular structure of the PAEs. For example, DBP and DINP showed greatest migration potential in the mineral water, whereas DEHP and DBP displayed the greatest degree of migration in the cooking oil.

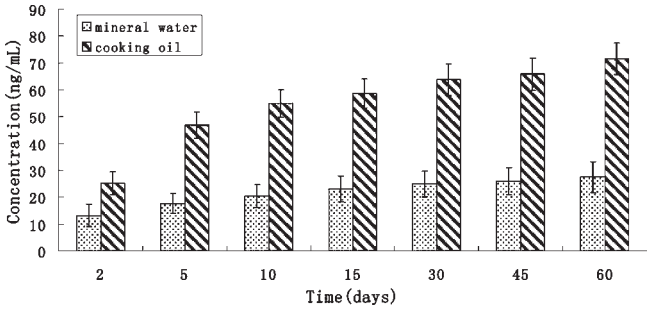

Figure 5. Comparison of DBP migration in two different food matrices ($n = 3$).

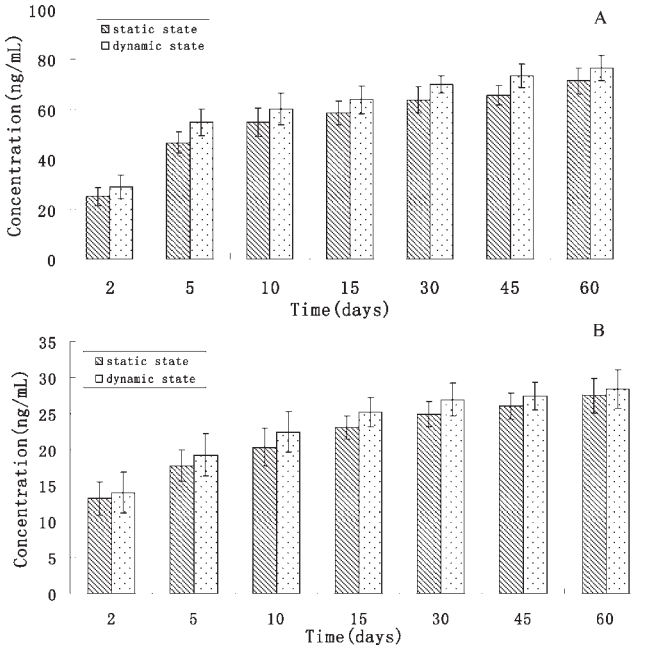

Figure 6. Migration of DBP into cooking oil (**A**) and mineral water (**B**) at 20 °C in static and dynamic state for 2 months ($n = 3$).

The corresponding values for PAEs migrated under static or dynamic state are shown in **Figure 6** (using DBP as the example). Oscillation facilitated contact between the food and the packaging and thus promoted the transfer of PAEs from the packaging materials into foodstuffs.

The migration curves for DBP at 20, 40, and 60 °C, respectively, are shown in **Figure 7**. Under the accelerated storage condition of 60 °C, the migration of DBP was approximately 25% more significant than that at 40 °C and 50% more significant than that at 20 °C, indicating the migration of PAEs from plastics into samples was facilitated by the elevation of storage temperature.

For our migration experiments, we chose the food itself as the test medium to ensure an authentic migration process relevant to this particular type of food matrix. Migration conditions were also carefully selected for the storage time (2 months), storage temperature (20, 40, and 60 °C), and storage state (static and dynamic). In this experiment, all samples contacted plastic packages directly. Migration was expressed as a percentage of the initial PAE concentration present in the plastic packages according to eq 1.

$$\text{migration}\ (\%) = \frac{C_{\text{foodstuff}}}{C_{\text{plastic,initial}}} \times 100 \qquad (1)$$

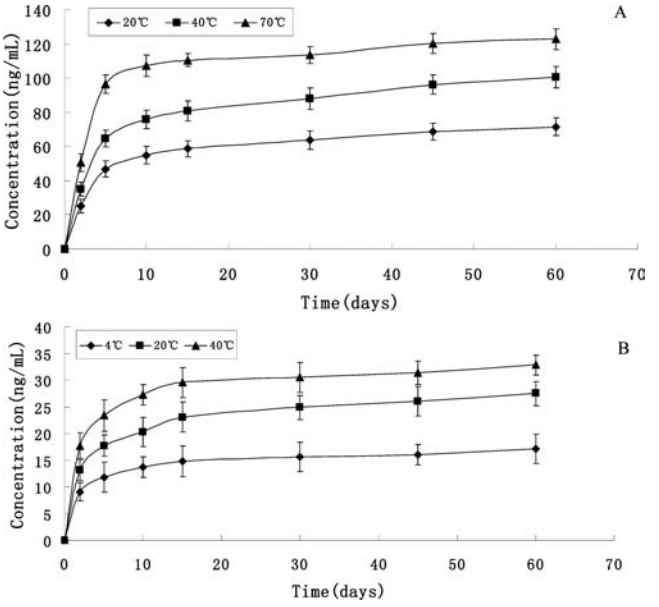

Figure 7. Migration of DBP into cooking oil (**A**) and mineral water (**B**) at 20, 40, or 60 °C for 2 months ($n = 3$).

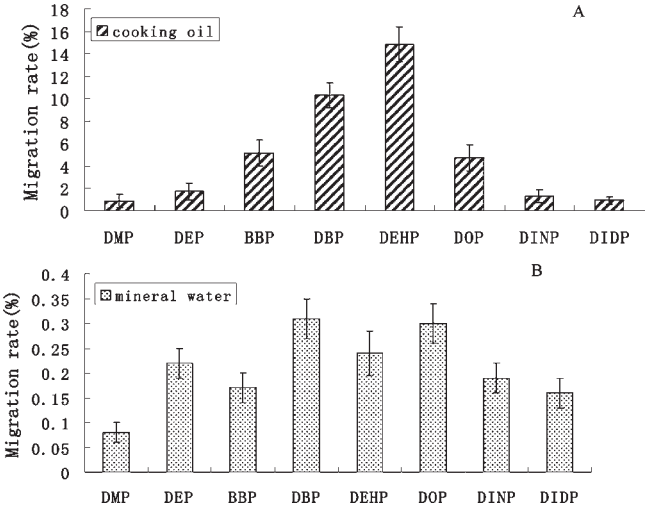

Figure 8. Migration of PAEs at 20 °C with migration values in percent after 2 months in cooking oil (**A**) and mineral water (**B**) ($n = 3$).

The target compound migration values in different sample matrices are shown in **Figure 8**. DBP and DEHP displayed great transfer potential in both cooking oil and mineral water. However, migration of DMP, DINP, and DIDP occurred only to a smaller extent. As we can see, lipids were more suitable media for the transfer of PAEs from plastic packages into foodstuffs than aqueous. The migration values of target compounds were all below 0.35% in mineral water, whereas they were in the range of 1−14% in cooking oil.

Figure 8 has shown that PAE migration into foodstuffs is never negligible. Therefore, a mathematical model should be established to predict the migration of PAEs from plastic packages into foodstuffs over time. In our case, we used SPSS (Statistical Product and Service Solutions) software to establish the nonlinear relationship between PAE levels present in foodstuffs and storage time under different temperature conditions, taking DBP as an example. The fitting curves of DBP level versus time at 20 °C for cooking oil and mineral water samples are shown in **Figure 9**

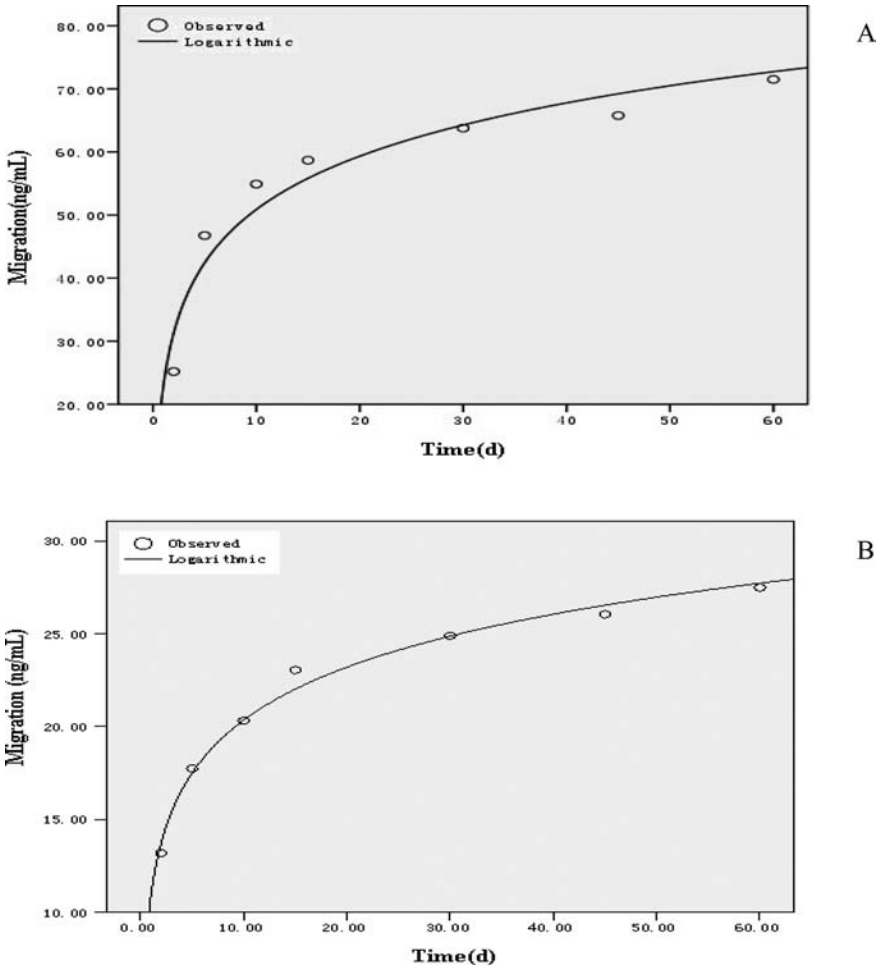

Figure 9. Fitting curve of time−migration relationship at 20 °C for DBP in cooking oil (**A**) and mineral water (**B**) samples.

Table 4. Regression Equations and Correlation Coefficients (r) of Migration of DBP at Different Temperatures in Samples

sample	temperature (°C)	regression eq[a]	correlation coefficient (r)
mineral water	4	$Y = 2.233 \ln X + 8.089$	0.985
	20	$Y = 4.115 \ln X + 10.869$	0.989
	40	$Y = 4.223 \ln X + 16.291$	0.949
cooking oil	20	$Y = 12.193 \ln X + 22.804$	0.935
	40	$Y = 17.001 \ln X + 30.058$	0.951
	60	$Y = 18.265 \ln X + 54.523$	0.907

[a] X, storage time (days); Y, concentration of DBP in food matrix.

(fitting curves at other temperatures are similar to the ones in **Figure 9**). For all of the fitting curves, logarithmic equations displayed the best fit based on the criteria of correlation coefficient values (i.e., closest to 1.0). The logarithmic equations for all of the temperature conditions are presented in **Table 4**

The data obtained from the current study are valid only for PAEs evaluated in this study, but they confirm the validity of using mathematical models to predict the migration of chemicals from packaging into foodstuffs. The mathematical modeling of PAE migration in this study was established for different temperature conditions and can be used as a practical tool by the food industry for the prediction of PAE migration. The manufacturers and food inspectors of plastic packaging materials will thus be able to comply with the regulations and confirm food safety without the need of expensive and time-consuming tests in the chemical enforcement laboratories.

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