Journal of Oleo Science
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doi : 10.5650/jos.ess20054
J. Oleo Sci. **69**, *(8) 851-858 (2020)*

JOURNAL OF OLEO SCIENCE
JOS

Phthalate and Polycyclic Aromatic Hydrocarbon Levels in Liquid Ingredients of Packaged Fish Sold in Turkish Markets

Sezer Kıralan*

Department of Food Engineering, Balıkesir University, Balıkesir 10145, TURKEY

Abstract: Phthalates (PAEs) and polycyclic aromatic hydrocarbons (PAHs) are ubiquitous contaminants in environment and foodstuffs. The objective of this study was to investigate the contamination possibility of phthalates and PAHs in packaged and canned fishes. For this purpose, tuna, salmon, sardine and mackerel canned and packaged with different liquid ingredients (water, olive oil, sunflower oil, mixture of sunflower and canola oil) attained from local markets in Turkey in 2019, were analyzed for presence of diethylhexyl phthalate (DEHP), dibutyl phthalate (DBP), butyl benzyl phthalate (BBP), diisononyl phthalate (DINP), diisodecyl phthalate (DIDP) and benzo(a)anthracene (BaA), benzo[a]pyrene (BaP), benzo(b)fluoranthene (BbF), chrysene (Chr). The instrumental analyses were performed by gas chromatography-mass spectrometry (GC-MS) and high-performance liquid chromatography fluorescence detection (HPLC-FLD). In all analyzed samples, the levels of DBP, BBP, DINP and DIDP were less than their LOQ, so these phthalates were not quantified. The highest DEPH content was found 650 µg/kg in sample 2 (tuna in olive oil, packaged in plastic package). The highest sum of PAH 4 concentration was 9.97 µg/kg in sample 4 (salmon canned in sunflower oil). Some samples (19 samples) were free for all analyzed PAEs and PAHs. All levels of these persistent organic pollutants were lower than regulation limits of Turkey and EU.

Key words: phthalates, polycyclic aromatic hydrocarbons, fish

1 Introduction

Over the last decades, persistent organic pollutants (POP) such as polycyclic aromatic hydrocarbons (PAHs) and phthalic acid esters (PAEs) have attracted public attention due to their possible harmful effects to human health[1−5]. The POPs which have genotoxic and carcinogenic impacts might be unintentionally present in the food chain[6, 7].

Most of PAHs and PAEs are found in vegetable oils and foods with high fat content because of their lipophilic nature[8−10]. These are common organic contaminants in environment but presence of them in food products is not only a consequence of the environmental contamination but also a consequence of the processing conditions[11−13] and contact with packaging material[14, 15]. Contamination of foods with PAHs may occur in different ways such as soil, water, oil[16], solvents used for oil extraction[17] and drying process of oil seeds[18]. The major occurrences of PAEs in foods are due to the contact with various plastic food containers and contamination from the process of food production[9, 19]. PAHs and PAEs have been detected in various food products such as fruit, bread, cheese, milk and especially foods with high-fat content[20−23].

Some of PAHs and PAEs are showing toxic and carcinogenic properties to human health[24, 25]. Because of their potential hazards and carcinogenic properties to human health, scientific and public worldwide organizations have been concerned with the concentrations of these POPs in food and food related products[7, 19]. In 2011, Commission

Abbreviations: BaA: Benzo(a)anthracene, BaP: Benzo[a]pyrene, BbF: Benzo(b)fluoranthene, Chr: Chrysene, EFSA: European Food Safety Authority, HPLC: High performance liquid chromatography, HPLC-FLD: High performance liquid chromatography fluorescence detection, PAH: Polycyclic aromatic hydrocarbons, SMLs: Special migration limits, LOD: Limit of detection, LOQ: Limit of quantification, GC-MS: Gas chromatography mass spectrometry, DEHP: diethylhexyl phthalate, DBP: dibutyl phthalate, BBP: butyl benzyl phthalate, DINP: diisononyl phthalate, DIDP: diisodecyl phthalate, DEHP: diethylhexyl phthalate, DBP: dibutyl phthalate, BBP: butyl benzyl phthalate, DINP: diisononyl phthalate, DIDP: diisodecyl phthalate, POP: persistent organic pollutants.

*Correspondence to: Sezer Kıralan, Department of Food Engineering, Balıkesir University, Balıkesir 10145, TURKEY
E-mail: sezertrhn@gmail.com
Accepted April 15, 2020 (received for review February 27, 2020)
Journal of Oleo Science ISSN 1345-8957 print / ISSN 1347-3352 online
http://www.jstage.jst.go.jp/browse/jos/ http://mc.manusriptcentral.com/jjocs

Regulation (EU) 835/2011 indicated the maximum limitations for the sum content of four PAHs (PAH4) which is the most suitable indicator of PAHs in food[26]. The sum of PAH4, has been listed in the Commission Regulation (EU) 835/2011 at the level of 10 mg/kg in oils and fats[26]. The maximum limit of benzo (a) pyrene has been reported as 2 μg/kg in oils and fats[27]. Some of the phthalates are restricted in the EU and listed in EU 2011/10, due to their potential health impact on humans[28]. In EU 2011/10 the values for DBP, BBP, DEHP, and sum of DINP and DIDP in food contact materials are listed as 0.3 mg/kg, 30 mg/kg, 1.5 mg/kg and 9 mg/kg, respectively[28].

Fish is an important source of nutrients such as protein and fatty acids (omega-3) for human diet that reduce the risk of some diseases (heart disease, stroke, and preterm delivery)[29, 30]. There are many ways for the preservation of fish, but canning is the most useful way of preserving sea products[31]. Vegetable oils are used in many foods as ingredient especially in canned fished products. Olive oil and sunflower oil are mainly used as liquid ingredients in canned fish products. A mixture of different vegetable oils such as sunflower and soy oil, can be used as vegetable oil in canned fishes. Vegetable oil is the most important ingredient in canned fish due to its direct consumption of the fish product. Fish, in particular is of great importance in Mediterranean diet due to its important nutrients. The occurrence of metals in canned fish in different regions of the world have been reported in the literature[31–37]. However, there is little information about concentrations of PAHs in canned fish[35, 38]. Canned and packed fish products may also be contaminated with phthalates. To the best knowledge, no current study has come across involving the phthalate presence in canned or packaged fish. Thus, this study aimed to report the levels of PAHs and phthalates in ingredients (oil, water, souse) of packaged fishes sold in Turkish markets.

2 Experimental

2.1 Sample collection

In October 2019, twentynine fish samples including tuna, salmon, sardine mackerel packaged with plastic package, can and glass bottle were collected from local markets and supermarkets in Turkey and analyzed in November 2019. Table 1 contains information regarding the fish species, packaging material and liquid ingredient.

2.2 Reagents

The phthalate standards for benzyl butyl phthalate (BBP, 99%), di (2-ethylhexyl) phthalate (DEHP, 98%), dibutyl phthalate (DBP, 99%), diisononyl phthalate (DINP, 98%), diisodecyl phthalate (DIDP, 99%) and 2,6-Di-*tert*-butyl-4-methylphenol (BHT, 99%) and PAH standards (benz (a) anthracene, chrysene, benzo (b) fluoranthene, benzo (a) pyrene) were purchased from Dr. Ehrenstorfer GmbH (Germany). BHT is an antioxidant and it was used as internal standard in this study. All the organic solvents such as acetonitrile and *n*-hexane were of HPLC grade and obtained from Merck (Darmstadt, Germany).

2.3 Sample preparation

Due to the cross contamination from plastic laboratory materials and equipment, only glass laboratory materials and equipment were used in this study for sample preparation. During phthalate analyses, *n*-hexane was injected after calibration curve analyses and between every samples for preventing potential contamination with samples.

i. *Sample preparation of phthalates*: Sample preparation was performed according to the previous study with some modifications[39]. 1 g oil or water was weighed with a glass pasteur pipette in a 10 mL glass tubes. 10 μL of the internal standard (BHT) was added and diluted to about 10 mL with acetonitrile. After homogenization by vortex (IKA-Werke, Staufen, Germany), the samples were centrifuged (Universal 32/R Hettich Zentrifugen, Germany) at 2500 rpm for 10 min. The upper phase part separated from the oil was transferred to another glass with glass pasteur pipette and then the phase was dried down under nitrogen at 40℃ until approximately 1 mL remains at the bottom of the tube. After a waiting period for 1 hour, the extract was transferred to vials and analyzed by the GC-MS.

   Preparation of calibration curve for the five phthalates and internal standard (BHT) was performed according to the previous method as described by authors[12]. The known chemical properties as well as the mass parameters of the five phthalates and internal standard (BHT) are shown in Table 2. Phthalates were quantified based on the peak areas compared with those of external standards.

ii. *Sample preparation of PAHs*: Preparation and analytical methods for the determination of PAH of canned and packaged fishes were performed according to the previous method as described by ISO 2006[40]. Each sample (2 g) was placed in centrifuge tube and then 10 mL acetonitrile: aceton (60/40%) was added into the test tube. The mixture was was centrifuged at 6000 rpm for 5 min. The upper phase was placed in C18 tube and sample tube was hand shaken vigorously. A further centrifugation was performed at 6000 rpm for 5 min. Then 7 mL of elute was evaporated under nitrogen. The concentrate was dissolved in 0.7 mL acetonitrile: acetone (60/40%) and transferred to the HPLC sample vial. PAHs were quantified based on the peak areas com-

Table 1 Sampling numbers, species, packaging materials and liquid ingredients of fish samples.

No	Packaging material	Fish species	Canned in
1	Plastic package	Tuna	Water
2	Plastic package	Tuna	Olive oil
3	Plastic package	Salmon	Olive oil
4	Can	Salmon	Sunflower oil
5	Can	Sardine	Sunflower oil
6	Can	Mackerel	Sunflower oil
7	Can	Mackerel	Sunflower oil
8	Can	Sardine with tomato souse	Sunflower oil
9	Can	Hot tuna	Sunflower oil
10	Can	Tuna with mayonnaise	Olive oil
11	Can	Tuna with mustard souse	Sunflower oil
12	Can	Tuna with barbeque souse	Sunflower oil
13	Can	Tuna with jalapeno souse	Sunflower oil
14	Can	Tuna	Sunflower oil
15	Can	Tuna	Sunflower oil
16	Can	Tuna	Sunflower oil
17	Can	Tuna	Sunflower oil
18	Can	Tuna	Sunflower oil
19	Can	Tuna	Sunflower oil
20	Can	Tuna	Sunflower oil
21	Can	Tuna	Sunflower oil
22	Can	Tuna	Sunflower oil
23	Can	Tuna	Sunflower oil, canola oil
24	Can	Tuna	Sunflower oil, canola oil
25	Can	Tuna	Sunflower oil, canola oil
26	Can	Tuna	Sunflower oil, canola oil
27	Can	Tuna	Water
28	Can	Tuna	Olive oil
29	Glass bottle	Tuna	Olive oil

Table 2 CAS numbers, retention times, sim ions and time windows for the phthalates and internal standard (BHT) in SIM mode.

Compounds	Cas no	Retention time (min)	SIM ion (m/z)	Time window (min)
BHT (IS)	128-37-0	8.099	205[1], 145, 177, 220	4.00-10.00
DBP	84-74-2	11.354	223[1], 150, 205	10.00-12.50
BBP	85-68-7	13.748	238[1], 91, 150, 206	12.50-14.00
DEHP	117-81-7	14.750	279[1], 150, 167	14.00-15.50
DINP	68515-48-0	16.074	307[1], 150, 167	15.50-18.00
DIDP	68515-49-1	17.909	293[1], 150, 167	15.50-18.00

[1] Quantitative ion.

Table 3 Method performance parameters of five PAE and four PAH.

Analyte	Linear Equation	R^2	LOD (μg/kg)	LOQ (μg/kg)	RSD (%)	Recovery (%)
PAE						
DBP	y =1300x-1593	0.999	0.06	0.09	11	94
BBP	y=4137x-4493	0.999	1.97	2.28	10	87
DEHP	y=3006x-3097	0.998	0.10	0.23	7	87
DINP	y=6823x-8488	0.997	1.37	1.75	4	100
DIDP	y=6849x-8525	0.997	1.20	1.40	3	92
PAH						
BaA	y= 5.18x-0,20	0.999	0.15	0.41	4	80
Chr	y= 0.91x-0	0.999	0.12	0.35	8	83
BbF	y= 5.20x-0,06	0.998	0.12	0.34	3	86
BaP	y= 13.08x-0,59	0.999	0.13	0.37	5	79

pared with those of external standards.

2.4 Analytical Methods

2.4.1 Instruments and conditions for the determination of phthalates

GC-MS analysis was performed by an Agilent system (GC 6890, MS 5973, Santa Clara, CA, USA) which is a gas phase chromatograph coupled to a mass spectrometer with a single quadruple type analyzer. The chromatograph was equipped with a HP-5MS capillary column (30 m length × 0.25 mm ID × 0.25 μm film thickness, Agilent Technologies, Santa Clara, CA, USA). The flow rate of helium gas (purity ≥ 99.999%) was 1.0 mL/min. v1 μL sample was injected in splitless mode through the injection port at 280℃. The temperature of the GC–MS interface was 280℃. Oven temperature program started at 80℃ for 1 min, and then followed by an increase of 15℃/min to 280℃, and held for 15 min. The full scan electron impact data were obtained as electron impact energy of 70 eV, and source temperature of 230℃. Selected-ion monitoring (SIM) data were collected during the GC-MS analysis.

2.4.2 Instruments and conditions for the determination of PAHs

The analytical determination of PAHs was carried out using an HPLC (Agilent 1100, Agilent Technologies, Santa Clara, CA, USA) equipped with a fluorescence detector. A PAH column (Zorbax Eclipse PAH, 4.6 mm length × 50 mm ID × 1.8 μm film thickness, Agilent Technologies, Santa Clara, CA, USA) at 18 ℃ was used for separation. Injection volume was 25 μL. The mobile phase of gradient program was composed of water (A) and acetonitrile (B). The solvent system was started with 60% acetonitrile (B) and 40% water (A) and continued with a gradient program to obtain 60% (B) at 1.5 min, 90% (B) at 7 min, 100% (B) at 13 min, 60% (B) at 14, 60% (B) at 15 min, 60% (B) 16.5 min until the end of the run with the flow rate of 0.8 mL/min. Excitation (260 nm) and emission (375, 420 and 460 nm) wavelengths channels were used.

3 Results and Discussion

3.1 Method Validation

PAEs (DBP, BBP, DEHP, DINP and DIDP) and PAHs (BaP, BaA, BbF and Chr) were analyzed in this study and detailed information is presented in Tables 2 and 3. The internal calibration and external calibration method were used for the quantification of PAEs and PAHs respectively. Limit of detection (LOD) and limit of quantification (LOQ) were defined, respectively, as the signal corresponding to 3 and 10 times the noise ratio, determined experimentally from fortified samples. As shown in Table 3, a good linearity was achieved in all cases with correlation coefficients of PAHs and PAEs, ranged between of 0.997-0.999 and 0.998-0.999, respectively. Limit of detection (LOD) and limit of quantification (LOQ) values for PAH were ranged between 0.12-0.15 and 0.34-0.41 μg/kg, respectively. For PAE, LOD and LOQ values ranged between 0.06-1.97 and 0.09-2.28 mg/kg, respectively (Table 3). The recoveries of five PAEs and four PAHs were between 87%-100% and 79%-86%, respectively. The relative standard deviation (RSD) values were less than 20% for all the tested concentrations of PAEs and PAHs.

3.2 Phthalate levels of water and vegetable oils in canned and packaged fish samples were collected from the Turkish market

Twentynine samples of canned and packaged fish were collected in duplicate packages in different Turkish markets. They were analyzed for the five phthalates. Table 4 summarizes the results of phthalate concentrations of vegetable oils and water in canned and packaged fish samples.

Concentrations of DBP, BBP, DINP and DIDP were below

Table 4 Concentrations of PAEs and PAHs in fish samples (μg/kg).

	PAEs					PAHs				
Sample	DBP	BBP	DEHP	DINP	DIDP	BaP	BaA	BbF	Chr	Total PAH
1	**	**	**	**	**	**	**	**	**	**
2	**	**	650 ± 20	**	**	**	**	**	**	**
3	**	**	350 ± 40	**	**	**	**	**	**	**
4	**	**	**	**	**	1.27 ± 0.01	2.35 ± 0.64	1.5 ± 0.28	4 ± 0.28	9.12 ± 0.06
5	**	**	500 ± 20	**	**	**	**	**	**	**
6	**	**	550 ± 30	**	**	0.44 ± 0.13	1.18 ± 0.03	0.64 ± 0.30	1.36 ± 0.04	3.61 ± 0.26
7	**	**	460 ± 10	**	**	**	**	**	**	**
8	**	**	300 ± 30	**	**	**	**	**	**	**
9	**	**	**	**	**	**	**	**	**	**
10	**	**	**	**	**	0.39 ± 0.04	1.48 ± 0.17	0.17 ± 0.07	1.69 ± 0.08	4.26 ± 0.01
11	**	**	**	**	**	**	**	**	**	**
12	**	**	**	**	**	**	**	**	**	**
13	**	**	**	**	**	**	**	**	**	**
14	**	**	**	**	**	**	**	**	**	**
15	**	**	**	**	**	**	**	**	**	**
16	**	**	**	**	**	**	**	**	**	**
17	**	**	**	**	**	**	**	**	**	**
18	**	**	**	**	**	**	**	**	**	**
19	**	**	**	**	**	**	**	**	**	**
20	**	**	630 ± 20	**	**	**	**	**	**	**
21	**	**	**	**	**	**	**	**	**	**
22	**	**	**	**	**	**	**	**	**	**
23	**	**	**	**	**	**	**	**	**	**
24	**	**	**	**	**	**	**	**	**	**
25	**	**	**	**	**	**	**	**	**	**
26	**	**	**	**	**	**	**	**	**	**
27	**	**	**	**	**	**	**	**	**	**
28	**	**	**	**	**	**	**	**	**	**
29	**	**	130 ± 18	**	**	**	**	**	**	**

**: Values are below LOQ.

LOQ in all analyzed samples. DEHP were detected in 8 samples among 29 from minimum LOQ to 650 μg/kg. The concentration of DEHP in all samples were out of substance migration (SML) limit (1.5 mg/kg) for this phthalate as set by EU 2011/10[28].

As shown in Table 1, different type of fishes (tuna, salmon, sardine, mackerel) were packaged in different type of oil (olive oil, sunflower oil, oil mixture (sunflower and canola oil)) and water. The concentration of DEHP varied depending on the nature of the fish material. As seen in Table 4, DEHP was detected in tuna (sample no: 2, 20, 29), salmon (sample no: 3), mackerel (sample no: 6, 7), and sardine (sample no: 5, 8) samples. DEHP was detected in all vegetable oil of sardine and mackerel samples ranging from 300 μg/kg to 550 μg/kg. So, probably the detected concentrations of DEHP in oil of sardine and mackerel samples were due to the fish type. A previous study reported the presence of microplastic and mesoplastics in canned sardines and sprats originating from 13 countries[35]. Polypropylene and polyethylene terephthalate were the most abundant plastic polymers in canned sardines and sprats and particle size range was between 190 and 3800 μm with an average ± SD size of 1149 ± 936 μm. Results from a study involving Atlantic chub mackerel (*Scomber colias*) from coast of the Canary Islands showed the presence of microplastic particles in the digestive tract of fish and from the 120 examined fish gastrointestinal tracts, 78.3% contained some type of microplastics and 17.5% contained plastic fragments[41].

The types of the liquid ingredient of canned fishes are shown in Table 1. Although oil was the main ingredient of packed fishes, water was also used in light products. Olive oil (samples 2, 3, 10, 28 and 29), sunflower oil (sample 4-9 and 11-22) and mixture of sunflower and canola oils (samples 23, 24, 25, 26) were used as an ingredient of packaged fish products. As shown in Table 4, samples 1 and 27 were free from phthalates which are packaged in water. DEHP was only detected in oily media due to its lipolytic nature. In this study, DEHP was detected in olive oil (samples 2, 3, 29) and sunflower oil (samples 5, 6, 7, 8, 20), while its concentration was below LOQ in mixture of sunflower oil and canola oil. The maximum and minimum concentration, of DEHP in olive oil and sunflower was 130-650 μg/kg and 300-630 μg/kg, respectively (Table 4). The occurrence of phthalates in vegetable oils was reported in many studies[19, 42, 43] evidencing that olive oils were contaminated with phthalates at high amounts. Results from a previous study revealed the average concentrations for DEHP of 1,262 μg/kg in olive oil and 134 μg/kg in sunflower oil. These oils were collected in Italian markets and olive-derived oils were more contaminated with PAEs than the other analysed vegetable oils[43].

According to Tables 1 and 4 shows the packaging materials and phthalate concentrations of oil samples respectively, 3 samples were packaged in plastic package, 1 sample was packaged in a glass bottle and 25 samples were packed in a metal can. Results indicated that, DEHP was detected in 2 of 3 samples packaged in a plastic package, 4 of 25 samples packaged in a metal can and 1 sample packaged in glass bottle. One of the plastic package samples was packed with water and no DEHP was detected in this sample; however, DEHP was present in all oil samples in a plastic package. Also, the highest DEHP level was measured in sample 2 which was packed in plastic package. The author reported that phthalate migration level to food in plastic packaging material was more than food in can[44]. In a previous study the concentrations for DMP and DEHP in oil tuna cans were reported as 6.6 ng/L and 2668 ng/L, respectively[44]. In this study results showed that DMP and BBP were detected trace levels in oil tuna cans. Also, in another study from Italy DMP concentration was reported as 1.9 μg/g in mayonnaise of codfish[45].

3.3 PAH levels of water and vegetable oils in canned and packaged fish samples were collected from the Turkish market

PAHs were present in 3 of 29 samples and total PAH4 concentration ranged from minimum LOQ to 9.12 μg/kg in all analyzed samples. The concentration of BaP ranged from minimum LOQ to 1.27 μg/kg, while minimum and maximum BaP concentrations were 0.39 μg/kg and 1.27 μg/kg in sample 10 and sample 4, respectively. BaA and BbF concentrations ranged from minimum LOQ to 2.35 μg/kg and minimum LOQ to 1.5 μg/kg, respectively. Chr was the predominant PAH in all analyzed samples and Chr concentration ranged from minimum LOQ to 4.0 μg/kg in all samples. Total PAH4 concentrations were 3.61, 4.26 and 9.12 μg/kg in sample 6 (mackerel canned in sunflower oil), sample 10 (tuna with mayonnaise canned in olive oil) and sample 4 (salmon canned in sunflower oil), respectively.

The results of PAH4 in vegetable oil of different types of fish samples were not necessarily associated with plastic packaging, fish type or oil type. PAHs were detected only in 3 of 29 samples and these samples are mackerel canned in sunflower oil, tuna with mayonnaise canned in olive oil and salmon canned in sunflower oil. PAHs were not detected in vegetable oils of fish samples packaged with plastic package and glass bottle, so it was concluded that packaging material was not associated with PAH migration. Also, PAHs were detected in vegetable oils of different species of fish (mackerel, salmon, tuna) and one of these samples was canned in olive oil and two of them canned in sunflower oil. The difference of PAH concentration in oil samples may be due to environmental contamination or contamination from the oil and sauce[34]. Furthermore, it is a fact that fish can be easily contaminated with PAHs by environment[35, 38]. In a previous study, the reported concentrations of sum of 16 PAHs in different brands of canned fishes (mackerel, sardine and tuna) collected from different Nigerian cities ranged from 174.6 μg/kg to 592.5 μg/kg. BaP was detected in 48% of analysed samples and BaP concentrations of these samples were higher than the maximum tolerable limit of 5 μg/kg[33]. In another study, the authors reported PAH concentrations of vegetable oils from canned tunas. The highest concentration of BaP of 1.9 μg/kg in olive oil from canned tuna, while none of analysed samples exceeded the 2 μg/kg of legal limit of BaP[38].

4 Conclusions

This study aimed to provide information on phthalate and PAH levels in liquid ingredients (water and vegetable oil) of different packaged fish species (tuna, salmon, sardine, mackerel) marketed in the Turkish markets. The results showed that only DEHP was detected in vegetable oil of fish samples as a phthalate while DBP, BBP, DINP and DIDP were not detected in all analyzed samples. DEHP levels (130-650 μg/kg) of analyzed samples did not exceed maximum residue levels according to both to Turkish and EU regulations. Furthermore, similar results were observed for PAH concentrations in vegetable oil of fish samples and their concentrations were under the legal limits of Turkey and EU. This study demonstrates that different type of packaged and canned fish samples consumed in Turkey are safe according to Turkish and EU limits for phthalates and PAHs; however, the quality parameters should always be monitored for ensuring a safe production of packed fish products.

Acknowledgements

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

Conflicts of Interest

The author declare no conflict of interest.

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