



Microplastics in house dust from 12 countries and associated human exposure



Junjie Zhang^{a,b}, Lei Wang^{a,*}, Kurunthachalam Kannan^{b,*}

^a Ministry of Education, Key Laboratory of Pollution Processes and Environmental Criteria, Tianjin Key Laboratory of Environmental Remediation and Pollution Control, College of Environmental Science and Engineering, Nankai University, Tianjin 300350, China

^b Wadsworth Center, New York State Department of Health, and Department of Environmental Health Sciences, School of Public Health, State University of New York at Albany, Empire State Plaza, P.O. Box 509, Albany, NY 12201-0509, United States

ARTICLE INFO

Handling Editor: Adrian Covaci

Keywords:

Microplastics
Indoor dust
Polyethylene terephthalate
Polycarbonate
Human exposure

ABSTRACT

Global marine pollution by microplastics (MPs) has received considerable attention in recent years. Nevertheless, little is known about the occurrence of MPs in indoor environments. A novel analytical method was used to quantitatively determine polyethylene terephthalate (PET)- and polycarbonate (PC)- based MPs in 286 indoor dust samples collected from 12 countries. PET-based MPs were detected in all dust samples at concentrations of 38–120,000 µg/g (median: 5900 µg/g), whereas PC-based MPs were measured at < 0.11–1700 µg/g (median: 8.8 µg/g). Significant positive correlations were found between the concentrations of terephthalic acid (a PET monomer) and PET as well as between bisphenol A (a PC monomer) and PC. Based on the concentrations of MPs measured in indoor dust, the median daily intake of PET-based MPs calculated for infants was in the range of 4000–150,000 ng/kg-bw/day.

1. Introduction

Approximately 8300 million metric tons of plastics have been produced worldwide since 1950 (Geyer et al., 2017). Microplastics (MPs) refer to plastics with a size of < 5 mm (Arthur, 2008) that originate from primary sources (e.g., plastic beads and synthetic fibers deliberately manufactured in microscopic size) and secondary sources (e.g., environmental degradation of large-sized plastic pieces that yield MPs) (Rillig, 2012). The most commonly manufactured plastics are polypropylene (PP), polyethylene (PE), polyethylene terephthalate (PET), polystyrene (PS), polyurethane (PUR), polyvinyl chloride (PVC), and polycarbonate (PC) (Li et al., 2016). Following substantial production and usage for decades, aquatic pollution caused by MPs has received considerable attention in recent years (Dris et al., 2016; Qiu et al., 2015; Su et al., 2016; Teng et al., 2019; Zhang et al., 2017). However, limited information is available about the occurrence of MPs in the indoor environment (Cai et al., 2017; Dris et al., 2017; Liu et al., 2019). Furthermore, quantitative assessment of human exposure to MPs is limited.

PET is produced through the esterification of terephthalic acid (TPA) with ethylene glycol, which is chiefly used in textile industry and in bottle production for food, water, and soft-drink packaging (ECI, 2016). The annual global production of PET (Fig. 1) was 53.3 million

metric tons in 2016 (ECI, 2016). PC is a polymer that consists of building blocks of bisphenol A (BPA) linked by carbonate groups. PC plastic is mainly used in medical care, electric, electronic, construction, optical, automobile, and packaging applications (ECI, 2017). The annual production of PC (Fig. 1) was approximately 4.4 million metric tons in 2016 (ECI, 2017). A few studies have reported the toxic effects of PET- and PC-based MPs. For instance, PET instigated irritation, discomfort, tearing, and blurring of vision when contacted with the eyes (TOXNET, 2019). PC-based nanoplastics increased the degranulation of primary neutrophils *in vitro* (Greven et al., 2016). PET degradation in the environment and human bodies can yield TPA (Nayebare et al., 2018; Yoshida et al., 2016). Although TPA is reported to be relatively less toxic (McLellan and Bhat, 2012), high production and usage of PET and the resulting exposure to TPA remains a concern. BPA is utilized in the production of epoxy resins and additives (Groshart et al., 2001). As a weak estrogen mimic, it is reported to occur widely in the environment (Le et al., 2008; Liao et al., 2012; Wang et al., 2015). Leaching from PC plastics is an important source of human exposure to BPA. For example, concentrations of BPA in human urine increased significantly following the use of PC bottles for over a week (Carwile et al., 2009).

A novel analytical method based on alkali-assisted thermal depolymerization coupled with high-performance liquid chromatography-tandem mass spectrometry (HPLC-MS/MS) was developed for accurate

* Corresponding authors.

E-mail addresses: wang2007@nankai.edu.cn (L. Wang), Kurunthachalam.Kannan@health.ny.gov (K. Kannan).

<https://doi.org/10.1016/j.envint.2019.105314>

Received 9 June 2019; Received in revised form 4 November 2019; Accepted 4 November 2019

Available online 19 November 2019

0160-4120/ © 2019 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

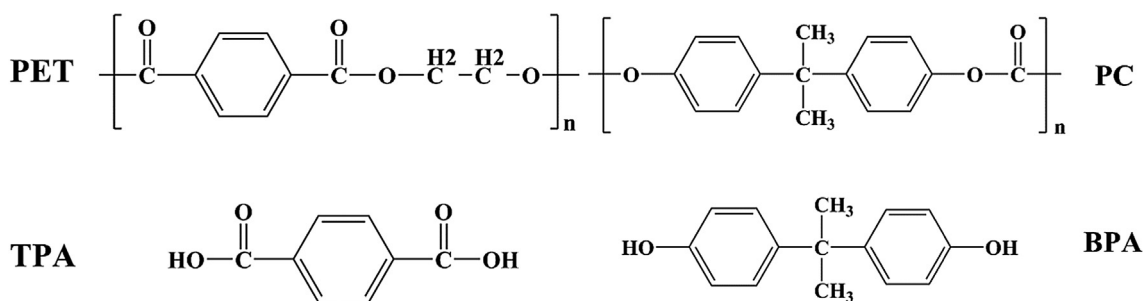


Fig. 1. Chemical structures of polyethylene terephthalate (PET) and polycarbonate (PC) and their monomers, terephthalic acid (TPA) and bisphenol A (BPA).

quantification of PET- and PC-based MPs (Wang et al., 2017). In this study, the occurrence of PET- and PC-based MPs was determined in 286 indoor dust samples collected from 12 countries, with the aim to quantitatively measure PET- and PC-based MPs and their monomers, TPA and BPA, respectively, in indoor dust and to assess the contribution of dust ingestion to MP exposure in humans.

2. Materials and methods

2.1. Chemicals and reagents

TPA was purchased from Toronto Research Chemicals, Inc. (North York, ON, Canada). BPA and D₄-TPA (99%) were purchased from Sigma-Aldrich (St. Louis, MO, USA). ¹³C₁₂-BPA (99%) was obtained from Cambridge Isotope Laboratories (Andover, MA, USA). HPLC grade methanol and water were supplied by J.T. Baker (Phillipsburg, NJ, USA). 1-Pentanol was purchased from Fisher Scientific (Pittsburgh, PA, USA). PET (3–5 mm) and PC (3 mm) particles were purchased from Goodfellow Cambridge Ltd (Huntingdon, England). Oasis HLB (200 mg, 6 mL) solid-phase extraction (SPE) cartridges were purchased from Waters (Milford, MA, USA).

2.2. Sample collection

From 2010 to 2014, 286 indoor dust samples were collected from 12 countries, comprising China ($n = 39$), Colombia (45), Greece (26), India (33), Japan (5), Kuwait (18), Pakistan (25), Romania (21), Saudi Arabia (30), South Korea (16), the USA (10), and Vietnam (18). Further details of sampling locations can be found in the [Supplementary Information \(Table S1\)](#). Floor-dust samples from bedrooms or living rooms were collected from each country by using a vacuum cleaner or by directly sweeping the floor with a nylon brush (only for the samples collected from China and India). Multiple dust samples were collected from each house or apartment and pooled into a single sample. After sampling, the dust samples were wrapped in aluminum foil and stored in sealed containers. All samples were sieved through a 150- μ m sieve, and samples below 150 μ m in size were collected, homogenized and stored at 4 °C until analysis. The field blanks were prepared by exposing aluminum foil to air during sampling (from three randomly selected bedrooms).

2.3. Sample extraction

Dust samples were depolymerized and extracted as described earlier (Wang et al., 2017). Dust samples (50 mg; spiked with 500 ng D₄-TPA and 200 ng ¹³C₁₂-BPA) were weighed and placed in a 100-mL round-bottom flask, and 0.1 g of KOH and 20 mL of 1-pentanol were added. The mixture was digested by heating at 135 °C in a stirring heating mantle for 30 min. Then the mixture was allowed to cool down at room temperature, and the pentanol solution was transferred into a 50-mL PP tube. The flask was rinsed twice with 10 mL of HPLC grade water each time, and the rinsate was transferred into the PP tube. The

depolymerized products of PET- and PC-based MPs were extracted from pentanol by shaking the PP tube at 180 strokes per minute for 5 min in an orbital shaker (Eberbach Corp., Ann Arbor, MI, USA) followed by centrifugation at 1620g for 5 min (Eppendorf Centrifuge 5804, Hamburg, Germany). The upper organic phase of pentanol was transferred to another tube to which 20 mL of HPLC grade water was added, and the extraction was repeated. The aqueous layer (water solution) that contained TPA and BPA was combined to a total volume of 50 mL with HPLC grade water. An aliquot of 10 mL of the solution was purified by passing through a SPE cartridge (detailed information is provided in Text S1).

The dust samples were also analyzed separately to determine the concentrations of freely available TPA and BPA. Briefly, 50 mg of dust sample was weighed and transferred into a 15 mL PP conical tube. After spiking with 250 ng of D₄-TPA and 50 ng of ¹³C₁₂-BPA, the samples were extracted with 5 mL of methanol by shaking in an orbital shaker for 30 min. The mixture was centrifuged at 2880g for 5 min, and the supernatant was transferred into a new PP tube. The extraction step was repeated twice with 5 mL of methanol, and the extracts were combined and concentrated to approximately 1 mL under a gentle nitrogen stream. The solution was diluted to 5 mL with a solvent mixture of methanol and HPLC grade water at 2:8 ratio (v/v). One milliliter of the diluted solution was centrifuged at 9030g for 5 min and then transferred into an amber glass vial for HPLC-MS/MS analysis.

2.4. Instrumental analysis

TPA concentrations in extracts were determined with a Shimadzu Prominence LC-20 AD HPLC (Shimadzu, Kyoto, Japan) interfaced with an Applied Biosystems API 3200 electrospray triple quadrupole mass spectrometer (ESI-MS/MS; Applied Biosystems, Foster City, CA, USA). An analytical column (Ultra Biphenyl USP L11 3 μ m, 100 $\times$ 2.1 mm column, Restek Corporation, Bellefonte, PA, USA) connected to a Javelin guard column (Betasil® C18, 20 $\times$ 2.1 mm) was used for LC separation (Table S2). BPA was determined with an Agilent 1100 HPLC (Agilent Technologies Inc., Santa Clara, CA, USA) interfaced with an Applied Biosystems API 2000 electrospray triple quadrupole mass spectrometer (ESI-MS/MS; Applied Biosystems, Foster City, CA, USA). Betasil® C18 (100 $\times$ 2.1 mm) column (Thermo Electron Corporation, Waltham, MA, USA) connected to a Javelin guard column (Betasil® C18, 20 $\times$ 2.1 mm) was utilized for LC separation (Table S2). The negative ion multiple reaction monitoring mode was applied. Table S3 presents detailed information of MS/MS parameters used in the analysis.

2.5. Quality assurance and quality control (QA/QC)

The field blanks did not contain free BPA, PC, free TPA and PET at concentrations above the method quantification limits (MQLs). Recovery experiments were conducted using soil dispersed on floors that were swept three times or vacuumed three times separately. The recoveries of PET and PC through the sweeping method of sampling were $92.3 \pm 5.9\%$ and $75.8 \pm 15.0\%$, respectively. The recoveries of

PET and PC by vacuum cleaner method of sampling were $64.0 \pm 7.7\%$ and $44.7 \pm 21.4\%$, respectively (Fig. S1). Lower recoveries of PET and PC in vacuum cleaner collected dust were due to the sorption of target chemicals to the vacuum cleaner bags.

Recoveries of TPA and BPA in spiked dust matrices were $105 \pm 5.9\%$ and $92.6 \pm 10.7\%$, respectively (Table S4). Procedural blanks were analyzed with every batch of 15–20 samples. The mean value of TPA found in procedural blanks analyzed through depolymerization methods was $2.6 \pm 0.62 \mu\text{g/g}$, whereas that through the methanol-extraction method was $0.14 \pm 0.01 \mu\text{g/g}$. BPA was not found in procedural blanks at concentrations above the MQLs. Background subtraction was performed in the quantification of TPA concentrations in samples. Depolymerization efficiency was tested by fortifying PET and PC particles in indoor dust and analyzed the above method. The recoveries of PET and PC fortified into samples were $> 95\%$. The round-bottom flasks were muffled at 450°C for 12 h and rinsed with acetone and methanol prior to use (to prevent carryover; this was required due to high concentrations of PET found in indoor dust). Duplicate analysis of randomly selected samples showed a coefficient variation of $< 10\%$ for TPA and BPA. The MQLs were 0.05 and $0.10 \mu\text{g/g}$ for free and depolymerized BPA, respectively (Table S4), as calculated from the lowest acceptable calibration standard and a nominal sample weight of 0.05 g. Due to the high background levels of TPA, MQLs of free and depolymerized TPA were 0.18 and $4.6 \mu\text{g/g}$, respectively, which were based on the mean concentrations in procedural blanks and standard deviation measured at low concentrations (Armbruster and Pry, 2008). The MQLs of PET and PC were 5.3 and $0.11 \mu\text{g/g}$, as calculated by the corresponding MQLs of depolymerized TPA and BPA, respectively (Wang et al., 2017). A midpoint calibration standard was injected as a check for instrumental drift in sensitivity after every 20 samples, and a pure solvent was injected as a check for carry-over of the target chemicals from sample to sample. Instrumental calibration was verified by the injection of 10 calibration standards, and the regression coefficient of the calibration curve (r) was > 0.99 . TPA and BPA concentrations in the fourth extraction with methanol for 15 randomly selected dust samples were $< 1\%$ of the concentrations found in the first three extractions, thereby indicating that the three extraction cycles almost completely extracted the target chemicals. Extracts were diluted and reanalyzed for those samples with concentrations above the upper limit of the calibration curve. The concentrations of PET- and PC-based MPs were calculated by the $(C_{\text{depolymerization}} - C_{\text{free}})$ of TPA and BPA, divided by 0.90 and 0.86, respectively (Wang et al., 2017).

2.6. Statistical analysis

Statistical analyses were performed using Origin version 9.0 (OriginLab, Northampton, MA, USA) and SPSS 16 (IBM, Armonk, NY, USA) software. Concentrations below the MQLs were substituted with a value equal to the MQLs divided by the square root of 2 to calculate the mean. Prior to one-way ANOVA and Pearson correlation analysis, the data were ln-transformed to meet the normality assumptions. The probability value of $p < 0.05$ was set for statistical significance.

The daily intake (EDI) of PET- and PC-based MPs and their corresponding monomers in toddlers (1–5 years), children (6–11 years), teenagers (12–19 years), and adults (≥ 20 years) through indoor dust ingestion was calculated as shown in Eqn 1:

$$\text{EDI} = \frac{C \times m}{BW} \quad (1)$$

where m is the indoor dust ingestion rate (g/day), BW is the body weight (kg), and C is the concentration of MPs in dust. The mean dust ingestion rates used in this calculation were 0.03 g/day for infants (< 1 year) and 0.06 g/day for toddlers (1–5 years), children (6–11 years) and teenagers (12–19 years), and 0.03 g/day for adults (≥ 20 years) (USEPA, 2011). The average body weights reported for infants, toddlers, children, teenagers, and adults in western countries (United

States, Colombia, Greece, and Romania) were 7.0, 15, 32, 64, and 80 kg, respectively (USEPA, 2011), whereas the corresponding values for Asian countries (China, South Korea, Japan, India, Pakistan, Kuwait, Vietnam, and Saudi Arabia) were 5, 19, 29, 53, and 63 kg, respectively (Guo and Kannan, 2011; Liao et al., 2012).

3. Results

The concentrations of PET and PC were corrected by the recoveries of different sampling methods. Details of the uncorrected data are shown in Table S5 in the supplementary material.

3.1. PET-based MPs and TPA in indoor dust

PET-based MPs were found in all 286 dust samples at concentrations in the range of 29–110,000 $\mu\text{g/g}$. The median concentrations of PET-based MPs in indoor dust varied by two orders of magnitude among the countries studied, in the following order: South Korea (25,000 $\mu\text{g/g}$) $>$ Japan (23,000 $\mu\text{g/g}$) $>$ Saudi Arabia (13,000 $\mu\text{g/g}$) $>$ Greece (9700 $\mu\text{g/g}$) $>$ Romania (9100 $\mu\text{g/g}$) $>$ the United States (8900 $\mu\text{g/g}$) $>$ Kuwait (8600 $\mu\text{g/g}$) $>$ Vietnam (3900 $\mu\text{g/g}$) $>$ China (3700 $\mu\text{g/g}$) $>$ Pakistan (1900 $\mu\text{g/g}$) $>$ India (1600 $\mu\text{g/g}$) $>$ Colombia (1000 $\mu\text{g/g}$) (Fig. 2). One dust sample from China contained the highest concentration of PET at 120,000 $\mu\text{g/g}$, an amount which was 12% of the total mass of dust.

Free TPA was also found in most dust samples, except for one sample from Vietnam, two samples from Pakistan, and 10 samples from Colombia. The median concentrations of TPA in dust samples from the 12 countries ranged from 2.0 $\mu\text{g/g}$ (Pakistan) to 34 $\mu\text{g/g}$ (Japan) (Fig. S2). The highest TPA concentration of 200 $\mu\text{g/g}$ was found in a dust sample from India (Table 1). A significant positive correlation was presented between the concentrations of TPA and PET-based MPs ($r = 0.512$, $p = 0.000$) (Fig. 3). The median concentration ratios of TPA to PET-based MPs in dust were 0.0005–0.0040.

3.2. PC-based MPs and BPA in indoor dust

The detection rate (DR) of PC-based MPs in dust samples was 99.3% with a concentration range of < 0.11 –1700 $\mu\text{g/g}$ (Table 1). Among the 12 countries studied, dust samples from Saudi Arabia (2.5–190 $\mu\text{g/g}$, median: 45 $\mu\text{g/g}$) and South Korea (6.7–140 $\mu\text{g/g}$, median: 38 $\mu\text{g/g}$) contained the highest concentrations of PC (Fig. 2). Relatively high concentrations of PC-based MPs were also found in dust samples from India and Vietnam. The annual production of PC plastics was 2.0, 1.5, and 0.9 million metric tons in Asia, Europe, and the United States, respectively (ECI, 2017).

Free BPA was also found in dust samples at concentrations of < 0.05 –36 $\mu\text{g/g}$. Except for China (95%), Vietnam (94%), Colombia (93%), India (82%), and Pakistan (76%), the DR of BPA in dust from the remaining seven countries was 100%. A significant positive correlation ($r = 0.677$, $p = 0.000$) was found between the concentrations of free BPA and PC-based MPs in indoor dust (Fig. 3). The median concentration ratios of BPA to PC-based MPs in dust samples were 0.06–0.34.

3.3. Factors that affect MP concentrations in indoor dust

Dust samples collected from homes in Pakistan were stratified as rural ($n = 13$) and urban ($n = 12$). Concentrations of PC-based MPs in rural homes were significantly lower than those in urban homes ($p < 0.05$, one-way ANOVA). Similarly, concentrations of PET-based MPs in dust from urban homes were higher than those from rural homes ($p = 0.069$, one-way ANOVA). A significant positive correlation was found between the concentrations of MPs and gross domestic product (GDP) per capita of the 12 countries ($r = 0.771$, $p = 0.003$ for PET and GDP per capita; $r = 0.597$, $p = 0.041$ for PC and GDP per capita) (Fig.

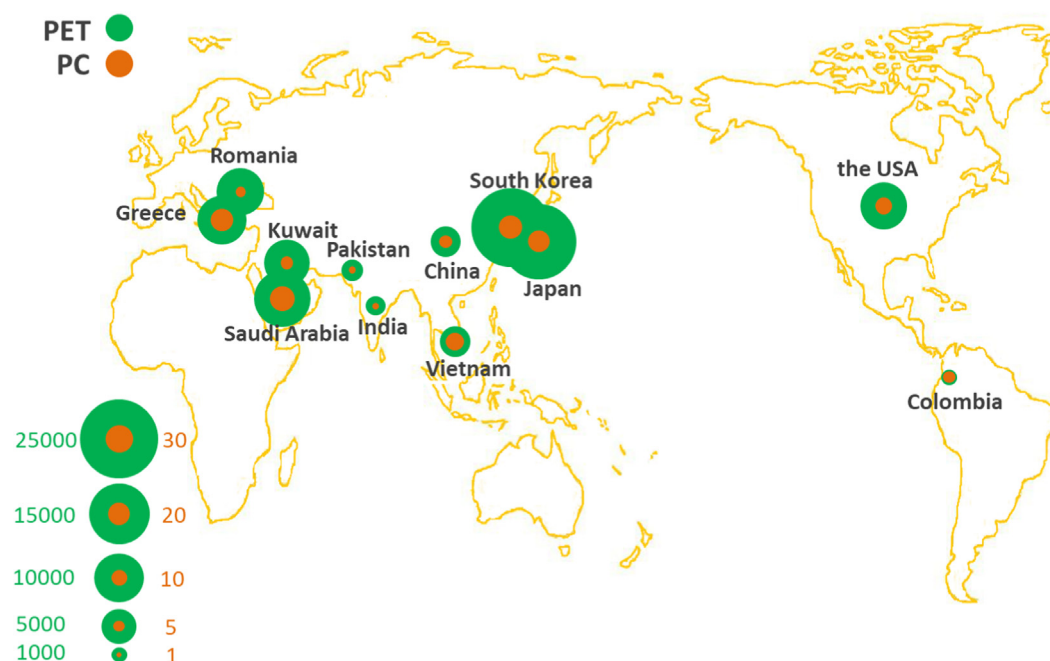


Fig. 2. Distribution of polyethylene terephthalate (PET) and polycarbonate (PC)-based microplastics in indoor dust collected from 12 countries (median concentration, $\mu\text{g/g}$).

S3). A wide range of environmental contaminants have been identified in indoor dust collected from the United States (Guo and Kannan, 2011; Johnson-Restrepo and Kannan, 2009; Kannan et al., 2010; Qian, 2013; Tran et al., 2015; Tue et al., 2013; Wan et al., 2015; Wang et al., 2013; Wang et al., 2012; Wang et al., 2015; Wang et al., 2016; Zhu and Kannan, 2018), among which PET-based MP showed the highest concentrations (Fig. 4). In addition, a significant positive correlation was found between the median concentrations of TBBPA and MPs ($r = 0.858$, $p = 0.000$) and SPAs and MPs ($r = 0.822$, $p = 0.001$) (Fig. S4) in indoor dust.

3.4. Human exposure to MPs

Dust ingestion is an important route of human exposure to several environmental pollutants (USEPA, 2011). The median EDI ($\text{EDI}_{50\text{th}}$) values of PET- and PC-based MPs in the 12 countries were in the ranges of 360–150,000 and 0.88–270 ng/kg-bw/day , respectively. The $\text{EDI}_{50\text{th}}$ of MPs decreased with the increasing age (Table 2). The $\text{EDI}_{50\text{th}}$ values for infants were tenfold higher than those for adults. The $\text{EDI}_{50\text{th}}$ values of MPs through dust ingestion for all age groups in Saudi Arabia, South Korea, and Japan were significantly higher than those in Colombia, Pakistan, and India. The $\text{EDI}_{50\text{th}}$ of free TPA through dust ingestion ranged from 11 to 200 ng/kg-bw/day for infants and 0.96 to 9.4 ng/kg-

Table 1

Concentrations of free terephthalic acid (TPA) and free bisphenol A (BPA), and polyethylene terephthalate (PET) and polycarbonate (PC)-based microplastics in indoor dust from 12 countries ($\mu\text{g/g}$).

		TPA	PET	BPA	PC			TPA	PET	BPA	PC
China n = 39	mean	18	9200	1.1	18	Pakistan n = 25	mean	8.1	4400	0.49	6.0
	median	14	3700	0.61	6.6		median	2	1900	0.26	3.4
	min	2.1	38	< 0.05	0.38		min	< 0.18	45	< 0.05	< 0.11
	max	79	120,000	5.2	170		max	47	38,000	3.1	49
Colombia n = 45	mean	9	3900	1.3	11	Romania n = 21	mean	7.8	14,000	3.7	15
	median	3	1000	0.75	5.6		median	4.5	9100	2.6	7.2
	min	< 0.18	140	< 0.05	0.65		min	1.8	2200	0.9	0.98
	max	94	22,000	5.1	110		max	54	73,000	12	110
Greece n = 26	mean	12	15,000	6.4	34	Saudi Arabia n = 30	mean	16	20,000	4.1	58
	median	8.3	9700	5.7	21		median	9	13,000	3.9	45
	min	3.6	2300	2.1	4.9		min	1.9	1500	0.57	2.5
	max	76	80,000	22	110		max	56	52,000	17	190
India n = 33	mean	12	2000	0.71	20	South Korea n = 16	mean	20	27,000	8.2	54
	median	3.1	1600	0.31	1.8		median	20	25,000	7.5	38
	min	0.5	55	< 0.05	< 0.11		min	7.2	6900	1.8	6.7
	max	200	6800	7.1	530		max	45	70,000	17	140
Japan n = 5	mean	27	20,000	12	63	USA n = 10	mean	11	14,000	3.1	87
	median	34	23,000	6.4	19		median	6.7	8900	2.8	20
	min	5.4	6700	3.3	< 0.11		min	0.72	1600	0.95	1.3
	max	47	36,000	36	180		max	34	44,000	8.2	400
Kuwait n = 18	mean	11	10,000	2.1	6.9	Vietnam n = 18	mean	31	11,000	2.1	120
	median	8.1	8600	1.3	6.9		median	17	3900	0.77	22
	min	3.6	1400	0.13	0.54		min	< 0.18	630	< 0.05	1.6
	max	25	31,000	9	13		max	180	42,000	13	1700

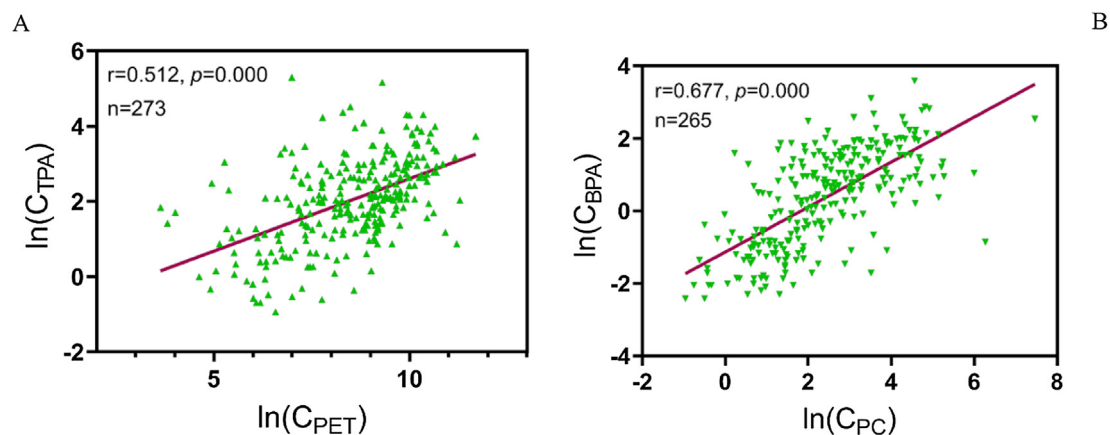


Fig. 3. Correlation between concentrations of polyethylene terephthalate (PET)-based microplastics and terephthalic acid (TPA) (A) and polycarbonate (PC)-based microplastics and bisphenol A (BPA) (B) in indoor dust samples from 12 countries.

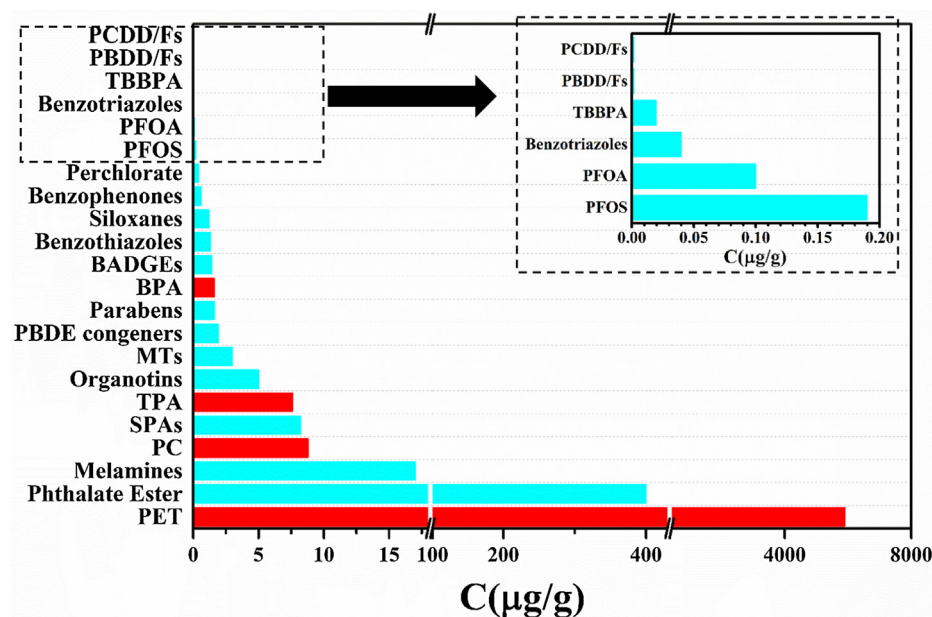


Fig. 4. Comparisons of the median concentrations of polyethylene terephthalate (PET) and polycarbonate (PC)-based microplastics in indoor dust samples measured in this study compared with those of other chemicals previously measured in indoor dust collected from Albany, NY, United States, by our research group (PCDD and PCDFs, polychlorinated dibenzo-p-dioxins and dibenzofurans, respectively; PBDD and PBDFs, polybrominated dibenzop-dioxins and dibenzofurans, respectively; TBBPA, tetrabromobisphenol A; PFOA, perfluorooctanoic acid; PFOS, perfluorooctanesulfonate; BADGEs, bisphenol A diglycidyl ether; PBDEs, polybrominated diphenyl ethers; BHT metabolites, metabolites of 2,6-di-tert-butyl-4-hydroxytoluene; SPAs, synthetic phenolic antioxidants).

Table 2

Median estimated daily intakes (EDI, ng/kg-bw/day) of polyethylene terephthalate (PET) and polycarbonate (PC)-based MPs via house-dust ingestion for different age groups in 12 countries.

	PET					PC				
	infants	toddlers	children	teenagers	adults	infants	toddlers	children	teenagers	adults
China	22,000	12,000	7600	4100	1700	40	21	13	7.5	3.2
Colombia	4000	3800	1700	890	360	25	22	11	5.1	2.1
Greece	42,000	39,000	19,000	9100	3600	89	83	38	19	7.8
India	10,000	5100	3400	1800	770	11	5.8	3.8	2.1	0.88
Japan	140,000	73,000	48,000	27,000	11,000	120	60	40	22	8.9
Kuwait	52,000	27,000	17,000	9700	4100	43	22	14	7.8	3.4
Romania	39,000	36,000	17,000	8400	3400	31	29	13	6.7	2.7
Pakistan	11,000	5900	3900	2200	890	20	11	6.9	3.8	1.6
Saudi Arabia	78,000	42,000	27,000	15,000	6300	270	140	92	51	21
South Korea	150,000	80,000	52,000	28,000	12,000	220	120	78	43	18
USA	38,000	36,000	17,000	8300	3300	87	81	38	19	7.6
Vietnam	23,000	12,000	8100	4400	1900	130	69	45	25	10

bw/day for adults in all 12 countries, whereas the EDI_{50th} of free BPA ranged from 1.5 to 44 ng/kg-bw/day for infants and 0.13 to 3.6 ng/kg-bw/day for adults.

4. Discussion

Quantitative information that pertains to the mass concentration of MPs in indoor dust is scarce. Very high concentrations of PET-based MPs were found at 37–120 000 µg/g in indoor dust, which can be explained by the use of PET in textile fibers (65%) and packaging (30%) (ECI, 2016). The concentrations of PET-based MPs in indoor dust were one to three orders of magnitude higher than those of PC-based MP analyzed by alkali-assisted thermal depolymerization coupled with HPLC-MS/MS. The annual global production of PET is five- to ten-fold higher than that of PC (ECI, 2016, 2017). A positive correlation was reported between the concentrations of PET-based MPs and the amount of fibers present in indoor dust (Liu et al., 2019). These results suggest that PET fibers are important MPs present in indoor environments.

Economic development and human activity are important factors in the distribution of PET- and PC-based MPs in indoor dust. Positive correlations were found between the concentrations of MPs and GDP per capita of various countries. Plastics consumption in the United States (an example of a developed country) and India (an example of a developing country) in 2014 was 95 and 8 kg/capita, respectively (Plastics Insight, 2015). In addition, concentrations of PET- and PC-based MPs in the rural area of Pakistan were lower than those in the urban area. China is the largest producer of PET plastics globally, with an annual production of 18.9 million metric tons in 2016 (ECI, 2016). The highest concentration of PET-based MPs was found in a dust sample from China.

Human exposure to MPs has generated widespread concern in recent years. One study indicated that plastic ingestion via mussel consumption (123 particles/year/capita) was considerably lower than that of fiber exposure from dust fallout in homes (13,731–68,415 particles/year/capita) (Catarino et al., 2018). Sea salt intake was shown to contribute to MPs ingestion at 0–36,135 particles/year/capita (Peixoto et al., 2019). By contrast, higher contribution from dust ingestion to MPs exposure was estimated in this study, indicating that indoor dust is a very important pathway of MPs exposure in humans.

TPA is the building block of PET, and more than 93% of TPA was used in the manufacture of PET (Plastics Insight, 2017). Similarly, more than 70% of BPA production was utilized in the manufacture of PC plastics (Groshart et al., 2001). MPs exposure may be an important vector for human exposure to monomers, such as TPA and BPA. A significant correlation existed between the concentrations of TPA and PET-based MPs and between BPA and PC-based MPs, indicating that PET- and PC-based MPs are important sources of TPA and BPA in indoor dust. Several studies have reported leaching of TPA and BPA from PET and PC plastic bottles, respectively (Montuori et al., 2008; Le et al., 2008), as well as the increasing urinary BPA concentrations in humans with continuous use of PC bottles for over a week (Carwile et al., 2009). In addition to monomers, several additives, such as phthalates and TBBPA can leach from MPs (Hahladakis et al., 2018). Thus MPs can be vectors of exposure to several contaminants in humans.

Although this study describes indoor dust as an important source of MP exposure in humans for the first time, there are a few limitations that deserve attention. Several other important MPs, such as PP, PE, and PUR should be also quantified in dust in the future. The shape and size of MPs are also important factors in the exposure risk assessment, especially of nanosized MPs. In addition, it is difficult to compare the mass concentrations of MPs reported in this study with the number of fibers or particles reported in previous studies (for dust and other matrices), because of the high heterogeneity of microplastics morphology reported in different studies (Gatidou et al., 2019), and the conversion between mass concentration from abundance has not been established. Furthermore, dust samples were sieved through a 150-µm sieve which

would considerably underestimate the actual concentrations of MPs present in dust. Therefore our exposure estimates are an underestimate of the actual dose of MPs ingestion in humans. It should be noted that, the sampling methods might affect the distribution of MPs measured in dust. Further studies with larger sample size are needed to describe global distribution and human exposure doses of MPs in indoor dust.

5. Conclusions

PET- and PC-based MPs are ubiquitous in indoor dust from different countries and are found at notable concentrations in some cases. This indicates that MPs are global indoor pollutants. Dust ingestion is an important exposure route for MPs in humans. Exposure to PET- and PC-based MPs via dust ingestion in infants is significantly higher than those in adults. The toxicological significance of micro- and nano-plastics has been reported in recent years. This study provides information on the mass concentrations of typical MPs and assesses their intake via dust ingestion, which are the basis for assessing risks of MPs to human health.

Declaration of Competing Interest

None.

Acknowledgements

We thank Drs. A. G. Asimakopoulou (Greece), A. Covaci (Belgium), B. Gevao (Kuwait), B. Johnson-Restrepo (Colombia), T. A. Kumosani (Saudi Arabia), G. Malarvannan (Belgium), T. B. Minh (Vietnam), H.-B. Moon (South Korea), H. Nakata (Japan), and R. K. Sinha (India) for providing dust samples. The samples were analyzed at Wadsworth Center, New York State. This work was supported in part by the National Natural Science Foundation of China (Grant No. 41722304), the 111 Program of the Ministry of Education, China (Grant No. T2017002), the Chinese Scholarship Council (Grant No. CSC 201806200120), and the National Institute of Environmental Health Sciences of the National Institutes of Health (Award No. U2CES026542-01). The content is the sole responsibility of the authors and does not necessarily represent the official views of the National Institutes of Health. All authors declare no conflicting interests.

Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2019.105314>.

References

- Armbruster, D.A., Pry, T., 2008. Limit of blank, limit of detection and limit of quantitation. *Clin. Biochem. Rev.* 29 (Suppl 1), S49–S52.
- C.B.J. Arthur H. Bamford Proceedings of the international research workshop on the occurrence, effects and fate of microplastic marine debris. National Oceanic and Atmospheric Administration; 2008.
- Cai, L., Wang, J., Peng, J., Tan, Z., Zhan, Z., Tan, X., Chen, Q., 2017. Characteristic of microplastics in the atmospheric fallout from Dongguan city, China: preliminary research and first evidence. *Environ. Sci. Pollut. Res. Int.* 24, 24928–24935.
- Carwile, J.L., Luu, H.T., Bassett, L.S., Driscoll, D.A., Yuan, C., Chang, J.Y., Ye, X., Calafat, A.M., Michels, K.B., 2009. Polycarbonate bottle use and urinary bisphenol A concentrations. *Environ. Health Perspect.* 117, 1368–1372.
- Catarino, A.I., Macchia, V., Sanderson, W.G., Thompson, R.C., Henry, T.B., 2018. Low levels of microplastics (MP) in wild mussels indicate that MP ingestion by humans is minimal compared to exposure via household fibres fallout during a meal. *Environ. Pollut.* 237, 675–684.
- Dris, R., Gasperi, J., Mirande, C., Mandin, C., Guerrouache, M., Langlois, V., Tassin, B., 2017. A first overview of textile fibers, including microplastics, in indoor and outdoor environments. *Environ. Pollut.* 221, 453–458.
- Dris, R., Gasperi, J., Saad, M., Mirande, C., Tassin, B., 2016. Synthetic fibers in atmospheric fallout: a source of microplastics in the environment? *Mar. Pollut. Bull.* 104, 290–293.
- Gatidou, G., Arvaniti, O.S., Stasinakis, A.S., 2019. Review on the occurrence and fate of microplastics in Sewage Treatment Plants. *J. Hazard Mater.* 367, 504–512.

- Geyer, R., Jambeck, J.R., Law, K.L., 2017. Production, use, and fate of all plastics ever made. *Sci. Adv.* 3, e1700782.
- Greven, A.C., Merk, T., Karagoz, F., Mohr, K., Klapper, M., Jovanovic, B., Palic, D., 2016. Polycarbonate and polystyrene nanoplastic particles act as stressors to the innate immune system of fathead minnow (*Pimephales promelas*). *Environ. Toxicol. Chem.* 35, 3093–3100.
- Groshart, C.P., Okkerman, P.C., Pijnenburg, A.M.C.M. Chemical study on Bisphenol A, 2001.
- Guo, Y., Kannan, K., 2011. Comparative assessment of human exposure to phthalate esters from house dust in China and the United States. *Environ. Sci. Technol.* 45, 3788–3794.
- Hahladakis, J.N., Velis, C.A., Weber, R., Iacovidou, E., Purnell, P., 2018. An overview of chemical additives present in plastics: Migration, release, fate and environmental impact during their use, disposal and recycling. *J. Hazard. Mater.* 344, 179–199.
- Johnson-Restrepo, B., Kannan, K., 2009. An assessment of sources and pathways of human exposure to polybrominated diphenyl ethers in the United States. *Chemosphere* 76, 542–548.
- Kannan, K., Takahashi, S., Fujiwara, N., Mizukawa, H., Tanabe, S., 2010. Organotin compounds, including butyltins and octyltins, in house dust from Albany, New York, USA. *Arch. Environ. Contam. Toxicol.* 58, 901–907.
- Le, H.H., Carlson, E.M., Chua, J.P., Belcher, S.M., 2008. Bisphenol A is released from polycarbonate drinking bottles and mimics the neurotoxic actions of estrogen in developing cerebellar neurons. *Toxicol. Lett.* 176, 149–156.
- Li, W.C., Tse, H.F., Fok, L., 2016. Plastic waste in the marine environment: a review of sources, occurrence and effects. *Sci. Total. Environ.* 566–567, 333–349.
- Liao, C., Liu, F., Guo, Y., Moon, H.B., Nakata, H., Wu, Q., Kannan, K., 2012. Occurrence of eight bisphenol analogues in indoor dust from the United States and several Asian countries: implications for human exposure. *Environ. Sci. Technol.* 46, 9138–9145.
- Liu, C., Li, J., Zhang, Y., Wang, L., Deng, J., Gao, Y., Yu, L., Zhang, J., Sun, H., 2019. Widespread distribution of PET and PC microplastics in dust in urban China and their estimated human exposure. *Environ. Int.* 128, 116–124.
- McLellan, C.J., Bhat, V.S., 2012. Toxicological review and oral risk assessment of terephthalic acid (TPA) and its esters: a category approach AU – Ball, Gwendolyn L. *Crit. Rev. Toxicol.* 42, 28–67.
- Montuori, P., Jover, E., Morgantini, M., Bayona, J.M., Triassi, M., 2008. Assessing human exposure to phthalic acid and phthalate esters from mineral water stored in polyethylene terephthalate and glass bottles. *Food Addit. Contam. Part A* 25, 511–518.
- Nayebare, S.R., Karthikraj, R., Kannan, K., 2018. Analysis of terephthalate metabolites in human urine by high-performance liquid chromatography-tandem mass spectrometry (HPLC-MS/MS). *J. Chromatogr. B Analyt. Technol. Biomed. Life Sci.* 1092, 473–479.
- Peixoto, D., Pinheiro, C., Amorim, J., Oliva-Teles, L., Guilhermino, L., Vieira, M.N., 2019. Microplastic pollution in commercial salt for human consumption: a review. *Estuar. Coast. Shelf Sci.* 219, 161–168.
- Plastics Insight, 2015. Market by country. <https://www.plasticsinsight.com/export-market/explore-your-market/by-country-profile/> (accessed date: 09 September 2019).
- Plastics Insight, 2017. Purified Terephthalic Acid (PTA) Properties, Production, Price, and Market. <https://www.plasticsinsight.com/resin-intelligence/resin-prices/purified-terephthalic-acid-pta/> (accessed date: 09 September 2019).
- Qian, W., 2013. Sources and Pathways of Human Exposure to Perfluoroalkyl Substances and Perchlorate. State University of New York at Albany, Albany, NY.
- Qiu, Q., Peng, J., Yu, X., Chen, F., Wang, J., Dong, F., 2015. Occurrence of microplastics in the coastal marine environment: First observation on sediment of China. *Mar. Pollut. Bull.* 98, 274–280.
- Rillig, M.C., 2012. Microplastic in terrestrial ecosystems and the soil? *Environ. Sci. Technol.* 46, 6453–6454.
- Su, L., Xue, Y., Li, L., Yang, D., Kolandhasamy, P., Li, D., Shi, H., 2016. Microplastics in Taihu Lake, China. *Environ. Pollut.* 216, 711–719.
- Teng, J., Wang, Q., Ran, W., Wu, D., Liu, Y., Sun, S., Liu, H., Cao, R., Zhao, J., 2019. Microplastic in cultured oysters from different coastal areas of China. *Sci. Total. Environ.* 653, 1282–1292.
- The Essential Chemical Industry (ECI), 2016. Polyesters; <http://www.essentialchemicalindustry.org/polymers/polyesters.html> (accessed date: 09 September 2019).
- The Essential Chemical Industry (ECI), 2017. Polycarbonates. <http://www.essentialchemicalindustry.org/polymers/polycarbonates.html> (accessed date: 09 September 2019).
- TOXNET, 2018. Polyethylene terephthalate. <https://toxnet.nlm.nih.gov/cgi-bin/sis/search/a?dbs+hsdb:@term+@DOCNO+7712> (accessed date: 09 September 2019).
- Tran, T.M., Abualnaja, K.O., Asimakopoulos, A.G., Covaci, A., Geva, B., Johnson-Restrepo, B., Kumosani, T.A., Malarvannan, G., Minh, T.B., Moon, H.B., Nakata, H., Sinha, R.K., Kannan, K., 2015. A survey of cyclic and linear siloxanes in indoor dust and their implications for human exposures in twelve countries. *Environ. Int.* 78, 39–44.
- Tue, N.M., Suzuki, G., Takahashi, S., Kannan, K., Takigami, H., Tanabe, S., 2013. Dioxin-related compounds in house dust from New York State: occurrence, in vitro toxic evaluation and implications for indoor exposure. *Environ. Pollut.* 181, 75–80.
- USEPA, 2011. Exposure Factors Handbook. United States Environmental Protection Agency.
- Wan, Y., Wu, Q., Abualnaja, K.O., Asimakopoulos, A.G., Covaci, A., Geva, B., Johnson-Restrepo, B., Kumosani, T.A., Malarvannan, G., Moon, H.B., Nakata, H., Sinha, R.K., Minh, T.B., Kannan, K., 2015. Occurrence of perchlorate in indoor dust from the United States and eleven other countries: implications for human exposure. *Environ. Int.* 75, 166–171.
- Wang, L., Asimakopoulos, A.G., Moon, H.B., Nakata, H., Kannan, K., 2013. Benzotriazole, benzothiazole, and benzophenone compounds in indoor dust from the United States and East Asian countries. *Environ. Sci. Technol.* 47, 4752–4759.
- Wang, L., Liao, C., Liu, F., Wu, Q., Guo, Y., Moon, H.B., Nakata, H., Kannan, K., 2012. Occurrence and human exposure of p-hydroxybenzoic acid esters (parabens), bisphenol A diglycidyl ether (BADGE), and their hydrolysis products in indoor dust from the United States and three East Asian countries. *Environ. Sci. Technol.* 46, 11584–11593.
- Wang, L., Zhang, J., Hou, S., Sun, H., 2017. A simple method for quantifying polycarbonate and polyethylene terephthalate microplastics in environmental samples by liquid chromatography-tandem mass spectrometry. *Environ. Sci. Technol. Lett.* 4, 530–534.
- Wang, W., Abualnaja, K.O., Asimakopoulos, A.G., Covaci, A., Geva, B., Johnson-Restrepo, B., Kumosani, T.A., Malarvannan, G., Minh, T.B., Moon, H.B., Nakata, H., Sinha, R.K., Kannan, K., 2015. A comparative assessment of human exposure to tetrabromobisphenol A and eight bisphenols including bisphenol A via indoor dust ingestion in twelve countries. *Environ. Int.* 83, 183–191.
- Wang, W., Asimakopoulos, A.G., Abualnaja, K.O., Covaci, A., Geva, B., Johnson-Restrepo, B., Kumosani, T.A., Malarvannan, G., Minh, T.B., Moon, H.B., Nakata, H., Sinha, R.K., Kannan, K., 2016. Synthetic phenolic antioxidants and their metabolites in indoor dust from homes and microenvironments. *Environ. Sci. Technol.* 50, 428–434.
- Yoshida, S., Hiraga, K., Takehana, T., Taniguchi, I., Yamaji, H., Maeda, Y., Toyohara, K., Miyamoto, K., Kimura, Y., Oda, K., 2016. A bacterium that degrades and assimilates poly(ethylene terephthalate). *Science* 351, 1196.
- Zhang, K., Xiong, X., Hu, H., Wu, C., Bi, Y., Wu, Y., Zhou, B., Lam, P.K., Liu, J., 2017. Occurrence and characteristics of microplastic pollution in Xiangxi Bay of Three Gorges Reservoir, China. *Environ. Sci. Technol.* 51, 3794–3801.
- Zhu, H.K., Kannan, K., 2018. Distribution profiles of melamine and its derivatives in indoor dust from 12 countries and the implications for human exposure. *Environ. Sci. Technol.* 52, 12801–12810.