RESEARCH ARTICLE

Insights into the efect of crystallinity on the sorption of organic pollutants to microplastics

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Abstract

The environmental behavior of microplastics (MPs) has attracted global attention. Research has confrmed that MPs can strongly absorb almost every kind of pollutant and can serve as vectors for pollutant transport. In this research, the sorption isotherms of six organic pollutants with diferent structure on four virgin plastic particles with diferent crystallinity were determined. Results indicated that the hydrophobicity (K_{OW}) of organic pollutants and the crystallinity of MPs were the two key factors that afected the sorption process of organic pollutants on MPs. Strong correlations were observed between K_{OW} and the partition coefficient. Hydrophobic partition was one of the major mechanisms regardless of the type of organic chemical (hydrophobic, polar, or dissociable). What is more, the infuence of the crystallinity of MPs on the sorption process increased with increasing hydrophobicity of the chemical. Combining this result with analyzing the related literature on the efect of crystallinity, it was concluded that the efect of crystallinity on the sorption of chemicals with strong hydrophobicity was obvious, whereas this efect was negligible for chemicals with weak hydrophobicity. The infuence of the crystallinity of MPs on sorption could even exceed the infuence of MPs type, so crystallinity should be considered carefully when discussing the sorption capacity of MPs. This study enhances the understanding of the sorption of organic pollutants by MPs.

Keywords Microplastics · Organic pollutants · Crystallinity · Sorption · Hydrophobic partition

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Highlights

- Sorption of six organic chemicals on four virgin plastic particles was investigated.
- Hydrophobic partition was one of the major mechanisms for most organic chemicals.
- K_{OW} and the crystallinity were the two key factors affecting the sorption process on MPs.
- The degree to which crystallinity afected the sorption varied with hydrophobicity.
- Crystallinity should be considered when discussing the sorption capacity of MPs.

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Introduction

In the past decades, plastic products have been widely used due to their durability, versatility, and low cost, so the global generation of plastic waste has increased rapidly. In 2018, the global total plastic waste reached 380 million tons (Liu et al. [2021\)](#page-8-0). However, on a global scale, only 9% of plastic products are recycled, about 12% are incinerated, and the remaining 79% are discharged into the environment (Pico et al. [2019](#page-8-1)).

After being discharged into the environment, plastics break down continuously into microsized or nanosized particles known as microplastics (MPs). The large amount of plastic entering the environment poses a threat to human and environmental health (Enfrin et al. [2020;](#page-8-2) Kogel et al. [2020](#page-8-3); Luo et al. [2020](#page-8-4), [2023](#page-8-5); Rai et al. [2021](#page-8-6)). Recently, MPs have been found in marine organisms such as fish, mussels, and seabirds (Herzke et al. [2016](#page-8-7); Garnier et al. [2019](#page-8-8); Hermabessiere et al. [2019](#page-8-9); Liu et al. [2020](#page-8-10)) and in various environmental compartments, including sediments, lakes, rivers, oceans, and urban wastewater (Carr et al. [2016](#page-7-0); Wang et al. [2017a,](#page-8-11)

[2017b](#page-8-12); Hossain et al. [2023](#page-8-13)). Therefore, the environmental behavior of MPs has attracted global attention (Zhou et al. [2020](#page-9-0); Feng et al. [2023](#page-8-14)).

Research has confrmed that MPs can strongly adsorb almost every kind of pollutant, including heavy metals, organic pollutants, pharmaceutical products, and personal care products, because of their large surface area and high hydrophobicity. MPs can therefore serve as vectors for pollutant transport (Avio et al. [2015;](#page-7-1) Guan et al. [2020;](#page-8-15) Menendez-Pedriza and Jaumot [2020;](#page-8-16) Wang et al. [2020](#page-8-17); Atugoda et al. [2021](#page-7-2); Chang et al. [2022](#page-7-3); Liu et al. [2022](#page-8-18); Luo et al. [2023\)](#page-8-5). The sorption of chemical pollutants on MPs has been investigated intensively. The adsorption process may be infuenced by three factors, namely, pollutant, MPs, and aquatic background characteristics, such as pH, ion strength, and dissolved organic matter.

For heavy metals, electrostatic interaction and surface complexation control the adsorption of heavy metals onto MPs. Therefore, heavy metal characteristics such as ionic radius, charge number, and complexing capability and MP characteristics such as the surface potential and functional groups present at the surface exert a considerable efect regardless of the crystallinity of MPs (Tourinho et al. [2019](#page-8-19); Zou et al. [2020\)](#page-9-1).

Hydrophobicity (K_{OW}) is a good predictor of sorption of hydrophobic organic pollutants on MPs, and strong correlations between K_{OW} and the partition coefficient (K_{D}) have been observed (Karapanagioti and Klontza [2008;](#page-8-20) Velez et al. [2018;](#page-8-21) Jiang et al. [2021](#page-8-22)). Given that the dominant interaction is related to hydrophobic partition, the water chemistry in terms of, for instance, pH and salinity slightly infuences the sorption process of organics to MPs. However, pH and salinity may strongly afect the sorption process for hydrophilic or ionizable organic pollutants because the corresponding sorption mechanisms include electrostatic forces aside from hydrophobic partition. With regard to the effect of the properties of MPs, most studies have focused on surface area, particle size, aging, type of plastics, and crystallinity. Some studies have found that polythene has the strongest sorption capacity among MPs, and others have reported that the order of sorption capacity is polystyrene>PVC>polythene (Huffer and Hofmann [2016](#page-8-24); O'Connor et al. 2016; Wu et al. [2019](#page-9-2); Tang et al. [2020](#page-8-25); Bao et al. [2021\)](#page-7-4). In short, no agreement currently exists about which type of MP has the strongest adsorption capacity despite numerous studies conducted.

In addition, consensus about the efect of the crystallinity of MPs on the sorption of organic contaminants is lacking. In accordance with the regularity of molecular chain arrangement, polymers can be divided into three categories: crystalline, semicrystalline, and amorphous. Generally, glassy polymers have rigid and condensed structures,

whereas the amorphous polymers have relatively expansive fexible structures. Hydrophobic organic contaminants can easily partition in amorphous polymers. Some studies have found that crystallinity considerably afects the sorption of organic pollutants onto MPs, whereas others have indicated that the efect is negligible (Teuten et al. [2009;](#page-8-26) Liu et al. [2019b\)](#page-8-27).

The key objectives of this study are to explore the origin of these contradictions and gain insights into the efect of crystallinity on the sorption of organic pollutants by MPs. Six organic chemicals that signifcantly difered in structure, hydrophobicity, ionization, and polarity were selected. Four plastic particles, namely chlorinated polyethylene (CPE) and three polyethylene plastic particles (i.e., LPE, LPE₂, and HPE), were adopted as target sorbents. The degree of crystallinity of HPE is higher than the crystallinity of LPE and $LPE₂$ which have a similar degree of crystallinity. CPE is an amorphous polymer. The batch sorption isotherms of the target sorbates onto MPs with diferent crystallinity were investigated and modeled, and the efects of particle size were studied. Moreover, the studies in this area were collected and analyzed, and their results were combined with those of the current study to determine the specifc efect of crystallinity.

Materials and methods

Chemicals and materials

All chemicals purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China) were of analytical grade or high purity. The relevant physicochemical properties of the chemicals are shown in Table [1.](#page-2-0)

Four sorbents (CPE, HPE, LPE, and LPE₂) were utilized in this study. HPE was purchased from Cospheric LLC (USA), and the others were bought from Haoying Plastic Material Corporation (Dongguan, China). Before the particles were used in experiments, they were made to pass through a 280-μm sieve (60 mesh).

MP characterization

The morphologies of the MPs were characterized using a scanning electron microscope and an X-ray photoelectron spectrometer. The elemental compositions, surface area, purity, and molecular weight of the MPs were determined. Crystallinity was determined based on the thermogravimetric and diferential scanning calorimeter (DSC) curves. The details of the characterization of MPs have been provided in our previous research (Zou et al. [2020](#page-9-1)).

Adsorption experiments

In the adsorption experiment, 0.01 mol/L of sodium chloride and calcium chloride were dissolved in distilled water as the background solution, and 200 mg/L of sodium azide was added to inhibit the bacteria. The stock solution of the involved chemicals with a concentration range of 0.001–100 mg/L was prepared with this background solution. About 10 mg of MPs were placed in a 42-mL brown glass bottle, and the prepared stock solution of organic pollutants with diferent concentrations (0.001–100 mg/L) was added. The glass bottles were mixed on a turntable at room temperature at 20 rpm. After equilibration, a 2 mL solution was obtained and fltered to determine the concentration of organic pollutants in the aqueous solution. The detailed analytical methods are given in Supplementary data. Control experiments in which MPs were not added were conducted, and the other processes were similar to those described above. The concentration of organic pollutants measured in the control experiment was adopted as the initial concentration. In accordance with mass conservation, the amount of chemicals on the MPs was determined by the diference in aqueous concentrations between the control and equilibrium

experiments. The medium conditions involved in this paper are relatively simple, so duplicate samples were obtained in all the experiments. And the results showed that in most cases the standard deviation of parallel samples was less than 5%. The pH of the medium was measured, and it was between 7.0 and 7.6, staying almost the same over the course of the experiments. In the case of CPE, the pH was a little higher than the other MPs involved here, likely because of its relatively strong nucleophilic ability which has been con-firmed in our previous research (Zou et al. [2020\)](#page-9-1).

Data analysis

It was reported that the Freundlich model ft the sorption isotherms of pollutants on microplastics well. And here it was used to ft the data. The Freundlich model is described as $q = K_F \times C_W^n$, where *q* (mmol/kg) and C_W (mmol/L) are the concentrations of organic pollutants sorbed on MPs and in the aqueous solutions at equilibrium, respectively; K_F (mmol¹⁻ⁿ/Lⁿ/kg) is the Freundlich affinity coefficient; and *n* is the exponential index. The MPs-water distribution coefficient (K_D) is also important in research on sorption, and it is expressed as $K_{\text{D}} = \frac{q}{c_{\text{w}}}$.

^an-octanol-water partition coefficient of organic pollutants, fromSchwarzenbach et al. [2003](#page-8-28)

b Saturated solubility of the organic pollutants in aqueous solution (25 ℃)

c Source:<https://pubchem.ncbi.nlm.nih.gov>

Table 1 The main physical and chemical properties of organic pollutants

Table 2 The major physical and chemical properties of the MPs

Results and discussion

Summary of the physicochemical properties of MPs

In this study, we selected four sorbents (CPE, HPE, LPE, and $LPE₂$), and three of them (i.e., CPE, HPE, and LPE) were used in our previous research (Zou et al. [2020](#page-9-1)), where the properties of CPE, HPE, and LPE were characterized and shown in detail. The corresponding characterization results of the sorbent $LPE₂$ are given in Fig. S1, and the major chemical and physical properties of the MPs closely related with this research are listed in Table [2.](#page-3-0) In brief, the purity of all sorbents was high. HPE had the highest crystallinity (77.9%), and LPE and $LPE₂$ had similar but relatively

low crystallinity (38.1% and 45.8%, respectively). CPE is an amorphous substance. The surface morphologies of CPE, LPE, and $LPE₂$ were similar and in all cases rough, possibly because they were commercial materials. Their diameters (between 150 and 250 μm) were also similar. HPE had uniform spherical pellets with relatively smooth surfaces, and the diameter was 27–45 μm. All the sorbents displayed similar surface areas.

Sorption of organic contaminants on MPs

The sorption isotherms of six organic chemicals on HPE and CPE are shown in Fig. [1](#page-3-1)a and b. For LPE and LPE $_2$, DNT, NAPH, and PHEN were selected as representative sorbates

Fig. 1 Sorption comparison of different organic pollutants on MPs. Error bars, in most cases smaller than the symbols, represent standard deviations of duplicate samples

Table 3 Parameters of Freundlich model obtained from sorption isotherms of organic pollutants on MPs with diferent crystallinity

Sorbent	Sorbate	K_F (mmol ¹⁻ⁿ L ⁿ /kg)	n	R^2	$\text{Log } K_{\text{D}}$
CPE	DNT	22 ± 2	0.67 ± 0.01	0.977	2.42
	NAPH	874 ± 14	0.82 ± 0.02	0.850	3.45
	PHEN	2950 ± 122	0.63 ± 0.01	0.983	4.83
	TeCB	71276 ± 581	1.06 ± 0.05	0.994	4.66
	DCP	28 ± 2	0.83 ± 0.03	0.997	1.82
	TRI	6.9 ± 0.3	0.62 ± 0.05	0.923	1.84
HPE	DNT	9.54 ± 0.09	0.52 ± 0.02	0.827	2.35
	NAPH	$180 + 23$	0.73 ± 0.03	0.963	2.93
	PHEN	1061 ± 57	0.93 ± 0.02	0.912	3.23
	TeCB	6519 ± 31	1.11 ± 0.03	0.922	3.49
	DCP	7.2 ± 0.2	0.66 ± 0.02	0.731	1.83
	TRI	8.7 ± 0.9	0.63 ± 0.04	0.928	1.91
LPE	DNT	1.3 ± 0.3	0.40 ± 0.02	0.953	2.35
	NAPH	2580 ± 28	1.06 ± 0.03	0.939	3.35
	PHEN	$697 + 24$	0.58 ± 0.06	0.947	4.18
LPE ₂	DNT	13 ± 3	0.73 ± 0.07	0.976	2.16
	NAPH	1915 ± 21	1.08 ± 0.03	0.963	3.12
	PHEN	$358 + 89$	0.50 ± 0.04	0.981	4.13

a KD was averaged from the KD obtained at diferent initial organic pollutant concentrations

(shown in Fig. [1](#page-3-1)c and d). The sorption data were modeled with the Freundlich model, and the corresponding coefficients obtained are listed in Table [3.](#page-4-0) As shown in Table [3,](#page-4-0) the *n* values for NAPH, PHEN, and TeCB were close to 1, and the *n* values for the other chemicals were much smaller than 1. This indicates that hydrophobic partition was the major sorption mechanism of NAPH, PHEN, and TeCB. For the other chemicals, other interactions aside from partition occurred.

The K_D in Table [3](#page-4-0) can directly reflect the sorption strength of sorbates on MPs, and the sequence was consistent with the order of hydrophobicity (usually quantifed by means of K_{OW} , listed in Table [1](#page-2-0)). This observation agrees with the findings of previous studies (Wang et al. [2020;](#page-8-17) Jiang et al. [2021\)](#page-8-22). The adsorption of organic pollutants on MPs is known to be mainly related to hydrophobicity. Therefore, K_{OW} and MPs–water distribution coefficient K_{D} were fitted (shown in Fig. [2](#page-4-1)) to further study the infuence of hydrophobicity on the sorption process, and the corresponding equations are listed in Table S1. Because of the deviation of DCP (seen from Fig. [2\)](#page-4-1), it was not included in the $K_{\text{OW}}-K_{\text{D}}$ fitting.

The correlation between K_{OW} and K_{D} was extremely strong for all sorbents tested. This indicates the outstanding effect of K_{OW} . The chemicals examined in this study

Fig. 2 The correlation between the K_{OW} and the K_{D} of the organic chemicals

included a polycyclic aromatic hydrocarbon, a nitroaromatic hydrocarbon, a phenol, a chlorinated benzene, and a pesticide. Hydrophobicity played a crucial role in the sorption of most organic pollutants by MPs. But it is worth noting that the K_D of DCP on CPE and HPE deviated obviously from the ftting curve compared with other sorbate-sorbent combinations. This is most likely because of its dissociation. The p*K*a of DCP was 7.85, so it is mainly present in its non-dissociated state and partly in its anionic state at the pH of the test medium (about 7.6) (Wen et al. [2022](#page-8-29)). According to the calculation, about 58.5% of DCP is present in its non-dissociated state at pH 7.6. Then, the K_D of DCP was recalculated on the basis of the non-dissociated concentration of DCP. It was found that the log K_D increased approximately from 1.8 to 2.0 and still deviated obviously from the ftting curve. In general, the surface of MPs is electronegative especially for CPE (Zou et al. [2020](#page-9-1)). Therefore, it was speculated that the electrostatic repulsion between the anionic state (about 41.5% at pH 7.6) and the electronegative surfaces of CPE and HPE further weakened the adsorption of DCP on MPs. This observation was in agreement with previous research about the sorption of dissociating organic pollutants on MPs (Wang et al. [2015](#page-8-30); Li et al. [2018](#page-8-31)). It was reported that the

electrostatic repulsion of anionic perfuorooctanesulfonate molecules at the surface of PE reduced its partition on PE and thereby decreased its K_D (Wang et al. [2015\)](#page-8-30).

Infuence of the crystallinity of MPs

To explore the efect of crystallinity, we selected DNT, NAPH, and PHEN as representatives and investigated their sorption on four MPs (i.e., CPE, HPE, LPE, and LPE₂) with diferent crystallinity. As shown in Fig. [3](#page-5-0), the sequence of the sorption strength of sorbates on MPs was consistent with the order of crystallinity. CPE, which was an amorphous substance, had the strongest sorption capacity among the four MPs, while HPE, being the MPs with the highest crystallinity, had the weakest sorption capacity. LPE and $LPE₂$ with low crystallinity had a moderate sorption affinity to the sorbates. Furthermore, the sorption strengths of the different sorbents dramatically difered for PHEN and were discernibly diferent for NAPH. However, this diference seemed negligible for DNT. Considering that their surface areas were relatively close, the diference in their adsorption capacity possibly resulted from crystallinity.

Fig. 3 Sorption comparison of DNT, NAPH, and PHEN on MPs with diferent crystallinities. Error bars, in most cases smaller than the symbols, represent standard deviations of duplicate samples

Fig. 4 Correlation between the crystallinity of MPs and the relevant distribution coefficient K_D

On the basis of the analysis above, we ftted crystallinity and K_D (Fig. [4\)](#page-6-0) and found that for PHEN and NAPH, the correlation between crystallinity and K_D was good, indicating that crystallinity played a crucial role in the sorption by MPs, especially for strong hydrophobic PHEN. However, no correlation was found between crystallinity and K_D for DNT, indicating the insignificance of crystallinity.

Research has shown that amorphous polymers are conducive to the difusion and partition of organic chemicals (Andrady [2017](#page-7-5); Zuo et al. [2019\)](#page-9-3). According to the discussion above, the main mechanism of the sorption of organic pollutants on MPs is hydrophobic partition. Therefore, crystallinity showed the greatest infuence on the strong hydrophobic pollutants such as PHEN and moderate infuence on the moderate hydrophobic pollutants such as NAPH. Nevertheless, for pollutants with weak hydrophobicity such as DNT, in addition to hydrophobic partition, a variety of sorption mechanisms are generally involved such as electrostatic interaction. Therefore, the infuence of crystallinity weakened correspondingly.

In addition, particle size may have had an efect because the particle size of the four sorbents was diferent. Another sorbent with the same physical and chemical properties as HPE, except for particle size (212–240 µm), was compared with HPE $(27-45 \mu m)$ to exclude the possible effect of particle size. The sorption isotherms of the two sorbents with diferent diameters for DNT and PHEN are displayed in Fig. S2. Particle size exerted a negligible efect on the sorption process although a large diference in particle size existed between the two sorbents, which was consistent with the results observed in Fig. [3](#page-5-0). CPE with a diameter of 150–250 μm showed higher sorption capacity than HPE with a diameter of $27-45$ µm and LPE and LPE₂ with a diameter of 150–250 μm. After excluding the possible causes of surface area and particle size, the crystallinity of MPs was

Table 4 Summary of the related literature on the efect of crystallinity on sorption of organic chemicals on MPs

Pollutants	MPs	Effect of crystallinity Reference	
Sulfadiazine, amoxicillin, tetracycline, ciprofloxacin, trimethoprim	PE, PS, PP, PA, and PVC No effect		Li et al. (2018)
Progesterone	HDPE, PP, PS, LDPE	No effect	Siri et al. (2021)
Tetracycline	PE, PP, PS	No effect	Xu et al. (2018)
Ciprofloxacin	PS and PVC	No effect	Liu et al. $(2019b)$
Perfluorooctanesulfonate and perfluorooctanesulfonamide	PE, PVC, PS	Slight effect	Wang et al. (2015)
diethyl phthalate and dibutyl phthalate	PVC, PE, PS	Slight effect	Liu et al. $(2019a)$
Phenanthrene, naphthalene, lindane, 1-naphthol	PE, PS, PPO	Strong effect	Guo et al. (2012)
Polycyclic aromatic hydrocarbons, polybrominated diphe- nyl ethers, alkanes, organophosphorous pesticides	PDMS, LDPE, HDPE, UHMWPE, PS, PP, PVC	Strong effect	O'Connor et al. (2016)
Phenanthrene	PEP, POM, PE, PP	Strong effect	Karapanagioti and Klontza (2008)
Phenanthrene	PBAT, PE, PS	Strong effect	Zuo et al. (2019)
Phenanthrene and pyrene	POM	Strong effect	Ahn et al. (2005)
Pesticide: S-metolachlor	PE, PP, PVC	Strong effect	Jiang et al. (2021)
Toluene, O-xylene tetrachloroethylene	HDPE, PVC	Strong effect	Teuten et al. (2009)
Aliphatic and aromatic organic	PA, PE, PVC, PS	Strong effect	Huffer and Hofmann (2016)
PCBs, DDE, and nonylphenols	PP	Strong effect	Mato et al. (2001)

PE polyethylene, *HDPE* high-density polyethylene, *LDPE* low-density PE, *PP* polypropylene, *PVC* polyvinyl chloride, *PPO* polyphenylene oxide, *PS* polystyrene, *PET* polyethylene terephthalate, *PEP* plastic eroded pellets, *POM* polyoxymethylene, *PDMS* polydimethylsiloxane, *UHMWPE* ultra-high molecular weight polyethylene, *PA* polyamide, *PBAT* polybutylene adipate terephthalate

found to have a considerable effect on the sorption of organic pollutants by MPs, especially for the organic chemicals with a large K_{OW} .

Insights into the efect of crystallinity

According to the discussion above, the more hydrophobic the pollutants are, the larger the efect of crystallinity on sorption. For pollutants with weak hydrophobicity, the infuence of crystallinity weakens correspondingly.

To further confrm our inference, we analyzed related literature on the efect of crystallinity, as shown in Table [4](#page-6-1). We found that studies where the concerned sorbates had weak hydrophobicity and dissociation ability, such as antibiotics and pesticides, concluded that crystallinity has a negligible infuence on sorption. In studies where the target chemicals were strongly hydrophobic, such as polycyclic aromatic hydrocarbons and chlorobenzenes, crystallinity was considered a predominant factor.

This discovery further confrms our preliminary conclusion. The degree to which crystallinity afected the sorption varied with the hydrophobicity of organic pollutants. In addition, it may explain the inconsistencies in the sequence of the sorption capacity of diferent types of MPs obtained in diferent studies. Possibly, crystallinity was disregarded in these studies. The sorption capacity of MPs can be very diferent due to diferent crystallinity even though some MPs have the same monomer.

Conclusions

In this study, it was indicated that the K_{OW} of organic pollutants and the crystallinity of MPs were two key factors that afected the sorption process of organic pollutants on MPs. Hydrophobic partition was one of the major mechanisms regardless of whether the organic chemicals were hydrophobic, polar, or dissociable. The general fnding is the larger the value of K_{OW} , the stronger the sorption. In addition, the degree of the effect of crystallinity varied with the hydrophobicity of the sorbates. The infuence of crystallinity on the sorption process increased with increasing hydrophobicity of the chemical. In summary, this study answered the two questions raised at the beginning of this paper: frstly, for the sorption of chemicals with strong hydrophobicity, the efect of crystallinity is obvious, whereas this effect is difficult to observe for chemicals with weak hydrophobicity. Therefore, some studies have concluded that the effect of crystallinity is prominent, whereas others have reported that crystallinity has no effect on sorption. This might also be due to the diferent chemicals studied. Secondly, the infuence of the crystallinity of MPs is sometimes even greater than the infuence of MPs type, so crystallinity should be considered

carefully when discussing the sorption capacity of MPs. The study enhances our understanding of the sorption of organic pollutants by MPs.

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Author contribution All authors contributed to the study conception and design.

Dongmei Zhang: Conceptualization; supervision; writing, original draft; reviewing and editing

Zining Zhang: Investigation, validation and data curation Hui Liu: Investigation and validation Jiying Zou: Investigation and paper revision Xiuping Liu: Investigation and validation Longyu Yin: Investigation and validation Ya-nan Zhang: Paper revision Jiao Qu: Project administration, supervision and reviewing Willie J.G.M. Peijnenburg: Reviewing and editing

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Data Availability The data that support the fndings of this study are available on request from the corresponding author.

Declarations

Ethical approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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