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MINI REVIEW ON RECENT ADVANCES OF THE ADSORPTION MECHANISM BETWEEN MICROPLASTICS AND EMERGING CONTAMINANTS FOR CONSERVATION OF WATER

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Abstract

Pharmaceuticals and microplastics have long been identified as water pollutants. Pollutants, including pharmaceutical compounds, have been shown to be transported by microplastics (MPs). In this mini-review, adsorption mechanism between microplastics and emerging contaminants were highlighted. Polyethylene is a non-polar, semi-cystalline microplastic with a density of 240 to 244 kg/m^3. Besides, Ibuprofen adsorption onto microplastics is pH dependent. Non-polar or neutral compounds that are homogeneous and extremely hydrophobic in nature interact with non-polar and weakly polar plastics such as Polypropylene and Polyethylene. Furthermore, Molecular dynamic (MD) simulation can be employed to study the mechanism of interaction between MPs and contaminants. As a result, some studies show a complex interaction between polyethylene (PE) and certain contaminants, with no significant differences in adsorption energies, but sulfamethazine molecules effectively adsorbed on the MPs surface. In summary, this mini review shed lights on the insights of adsorption mechanism between these compounds.

***Keywords**: Conservation of water; Microplastics; Pharmaceutical compounds; Adsorption,*

Introduction

Water is one of the most vital resources for sustainable living and should be conserved wisely. Water conservation has recently faced substantial obstacles due to the emergence of recalcitrant water pollutants. Polyethylene microplastic (PE MP) is found all over the continent. Hundreds of studies on the consequences of PE MP on the environment, particularly the biosphere, have been published. Polyethylene (PE) can be found in a variety of products, including containers and plastic bottles. In Europe, for example, skin care products contain at least 6% of PE [1]. This research provides an overview of microplastic classifications and identifications, followed by a discussion of emerging contaminants, most notably ibuprofen (IB) and its factors. The adsorption mechanism of emerging contaminants onto microplastics is also covered in this study. Finally, near the end of the study, the research on molecular dynamics simulation is discussed, and a summary of the literature review on the adsorption process between microplastics and emerging contaminants is obtained.

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Microplastics

Classifications

MPs have two types: primary and secondary MPs. The primary microplastics are obtained from manufacturing, processing, and packaging, whereas secondary MPs are derived from mechanical impact or chemical processes of larger particles through degradation or fragmentation [2]. Both primary and secondary MPs generated have the ability to efficiently absorb pollutants in the environment because MPs have a strong hydrophobicity added with a large specific surface area due to their small size [3]. The groups contain oxygen will increase the polarity of the MPs, resulting in a decrease in the hydrophobicity of the MPs for adsorption. In addition, the crystallinity of MPs has been shown to influence their sorption rate and capabilities in previous works of literature. Furthermore, the glass transition temperature (T_g) influences the crystallinity of the MPs, which in turn affects the adsorption capacity as well [4]. Table 1 shows the MPs' characteristics classification.

Table 1. MPs characteristics [2, 5, 6].

MP	Polarity	Crystallinity	T_g (°C)	Density (kg/m³)	Degradation time (year)
PA	Strongly polar	Semi-crystalline	-60	460 to 1350	
PVC	Polar	Amorphous	60 to 100	530 to 1450	< 100
PET	Polar	Semi-crystalline	73 to 78	1314 to 1323	< 450
PS	Non-polar	Amorphous	90	370 to 143	< 500
PE	Non-polar	Semi-crystalline	-110	240 to 244	< 1000
PP	Non-polar	Semi-crystalline	-49 to 20	220 to 240	20 to 30

Notes: PA - Polyamide; **PVC** - Polyvinyl Chloride; **PET** - Polyethylene Terephthalate; **PS** - Polystyrene; **PE** - Polyethylene; **PP** - Polypropylene;

Identification

Although developments are established in the detection and characterisation of other emerging pollutants, further work on MPs' identification in environmental contexts is needed to give consistent and credible data on their existence and predicament in the environment. MPs in environmental contexts have been studied using a variety of methods. In this study, FTIR spectra, SEM+EDX and BET analyzer, will be used to identify and characterize the PE MP according to shape, number, size and chemical composition of the MPs.

Polyethylene Microplastics. In general, polyethylene is not toxic when it comes into contact. Indeed, if a small amount of PE is ingested by humans or animals, it can cause severe damage to the internal organs, potentially leading to death [7]. Therefore, in this study, polyethylene microplastics will be chosen as the adsorbent to be studied due to lack of its investigation.

Emerging contaminants

Due to their physicochemical properties and complex environmental characteristics, data on the toxicity of emerging contaminants is still lacking [8]. However, earlier research has shown that these emerging contaminants are harmful to biological organisms, indicating that these contaminants are toxic. The risks, according to Hlavínek and Žižlavská [9], include acute and chronic toxicity, hazards to human health, toxicological effects, and the development of resistance in microbes. A list of emerging contaminants observed in the water ecosystem, along with examples is shown in Table 2.

Table 2. Emerging contaminants found in water ecosystem.

References	Emerging contaminants	Example
[10]	3D printing technologies	Caprolactam, Lactide, Irganox 1076
[9]	Analgesics	Phenazone, Codeine
[8]	Antibiotics	Trimethoprim, Sulfamethoxazole, Ciprofloxacin
[8]	Antiepileptics	Carbamazepine
[8]	Anti-inflammatory	Diclofenac, Naproxen
[11]	Beta Blocker	Atendol, Propanolol, Sotalol
[12]	Biocides	Parabens, Neonicotinoids
[8]	Cosmetics	Galaxolide, Tonalide
[8]	Disinfectants	Triclosan
[9]	EDC	EDC EE2, E2, E1
[10]	Flame retardants	PBB, PBT, TBCT
[10]	Nanosprays	MgO, ZnO, TiO_2
[13]	Pesticides	Atrazine, Diazinon, Terbutrin
[12]	Synthetic musk	Galaxolide, Tonalide
[12]	UV filters	Benzophenone-3, Homosalate, Octocrylene

Notes: **EDC**, Endocrine Disruption Chemicals, **PBB**, Pentabromobenzene; **PBT**, Pentabromotoulene; **TBCT**, Tetrabromo-o-chlorotoluene

Pharmaceutical Contaminants

The presence of pharmaceuticals in the surroundings can be categorized into two sources, primary source and secondary source [14]. The primary source of pharmaceutical waste is pharmaceuticals disposed of due to expired prescriptions and/or consumed and excreted by humans through the toilet for both hospitals and residences, whereas the secondary source includes inappropriate management of outdated and unused pharmaceuticals, spillages during production, packaging, and transportation. All of these sources are then passed through sewage treatment plant (STP) before being discharged into the water environment. Non-steroidal anti-inflammatory drugs (NSAIDs) group is one of the pharmaceutical emerging contaminants.

Ibuprofen

Ibuprofen (IB) is one of the world's most sold over the counter and supplied drugs. The consumption, disposal and manufacturing of IB can be said as major pathways for the entry of IB into the environment. As a result, IB and its derivatives are introduced into the food web and biomagnified [15]. IB is a white solid crystalline material in room condition and has a chemical structure of 2-(4-isobutylphenyl) propionic acid and 206.28 g/mol of molecular weight. Fig. 1 shows the IB’s powder and 3D molecular structure. The colours of grey, red and white indicate the carbon, oxygen and hydrogen atoms, respectively.

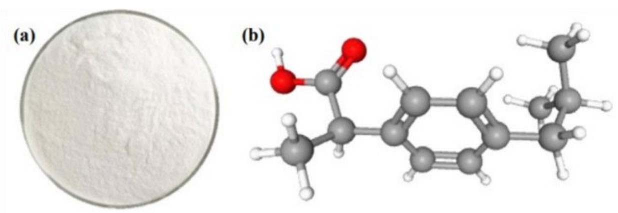

Fig. 1. Ibuprofen (a) powder and (b) molecular structure

The majority of current research involving pharmaceutical adsorption behavior on MPs has centred on batch adsorption studies in the lab, and only limited studies have looked into the adsorption of IB onto microplastics at the molecular level in depth. Furthermore, the number of researches done on the adsorption of IB onto PE MP is not abundantly available. Therefore, in this study, IB was chosen as the adsorbate. Besides, the adsorption of NSAIDs onto MPs are pH dependent. The MPs are able to adsorb the NSAIDs via hydrophobic interactions at pH = 2.0 due to its surface area. The most significant characteristic determining the plastics' sorption capacity seemed to be rapid biofouling, which was found in all specimens.

Adsorption of Emerging Contaminant onto Microplastic

Mechanism of Interactions

Emerging contaminants interact with MPs through a variety of methods. According to *W. Wang et al.* [16], several main mechanisms which these contaminants could interact with the MPs are pore-filling interaction, electrostatic interaction, π–π interactions and hydrophobic interaction. Fig. 2 depicts the potential mechanisms for MPs and contaminants to interact. Above the blue dotted lines indicates the known adsorption mechanisms, while the dotted green line-boxed mechanisms are applicable to a wide range of systems. In contrast, the plausible but unverifiable mechanisms are provided below the blue dotted line.

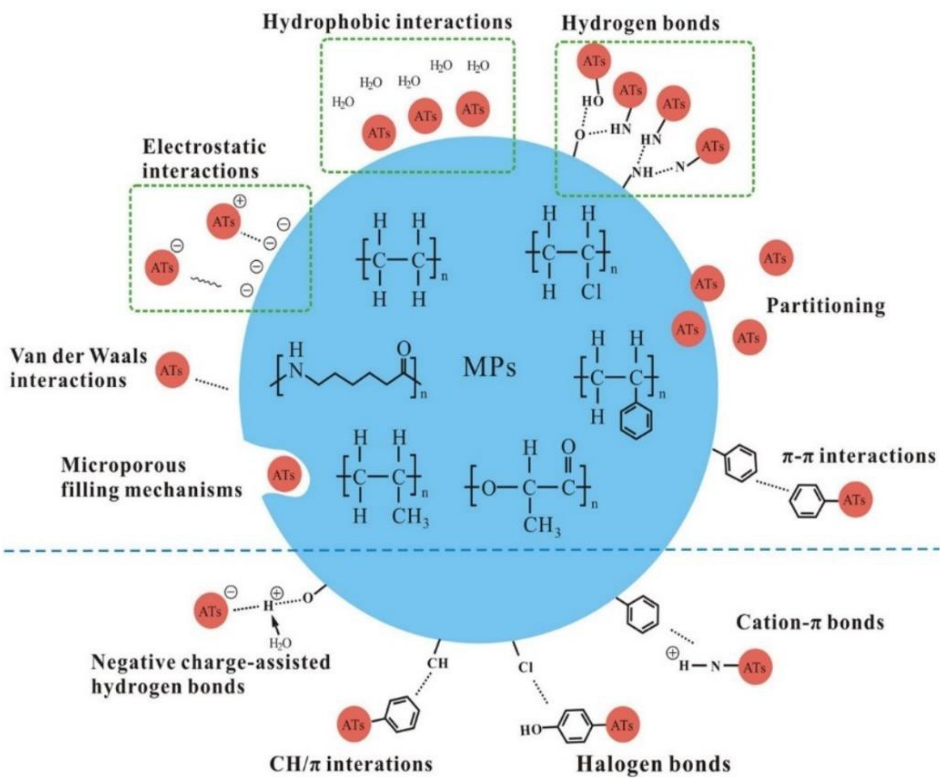

Fig. 2. The possible interactions of contaminants at the interface of MPs.
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However, the nature of the MPs' polymer, the qualities of the contaminants, and ambient parameters, such as the pH and salinity of the medium chosen, can influence these interactions. Different groups will 12 establish different interactions with the MP's polymer. Several researches have been carried out in order to identify the sorption mechanism between MPs and contaminants. Table 3 depicts the sorption of emerging contaminants on MPs from previous studies.

Table 3. Sorption mechanism for emerging organic contaminants on MPs

Groups	References	Contaminants	MP	Sorption mechanism
Antibiotics	[17]	Carbamazepine Triclosan	PE	Hydrophobic interaction
	[18]	Tetracycline	PE PP PS	Hydrophobic interaction π–π interactions
	[19]	Tylosin	PE PP PS PVC	Hydrophobic interaction Electrostatic interaction
Endocrine Disrupting Chemicals (EDCs)	[17]	EE2	PE	Hydrophobic interaction
Polycyclic Aromatic Hydrocarbons (PAHs)	[20]	Phenanthrene	PE PS PVC	Hydrophobic interaction
	[21]	Pyrene	PE PS PVC	Hydrophobic interaction
Other	[22]	Propranolol Sertraline	PE	Hydrophobic interaction

Notes: **EE2** - 17α-Ethinyl estradiol

Hydrophobic Interaction

Non-covalent forces like hydrophobic interactions cause non-polar components to cluster in polar media such as water. Non-polar plastics such as PE and PP, as well as weakly polar plastics such as polystyrene (PS), interact through non-polar or neutral molecules that are homogeneous and highly hydrophobic in nature. Hydrophobic interaction is a common interaction between MPs and organic contaminants in an aqueous phase. According to *X. Guo et al.* [23], organic contaminants such as phenanthrene, lindane, and naphthalene adsorb to PE owing to hydrophobic interactions. Due to hydrophobic interactions also, PS microplastics shows a high adsorption affinity of neutral triclosan molecules [24].

Hydrogen Bond Interactions

A hydrogen bond is formed when a hydrogen atom and a strongly electronegative atom collides with another electronegative atom with a single pair of electrons that produces dipole-dipole attraction. There are various studies conducted that show the presence of hydrogen bond interaction between MPs and organic contaminants. Hydrogen bond interactions are formed mostly between polar PA and hydrophilic organic contaminants. Several researchers have proven the claim. In 2019, Liu and co-workers [25] managed to provide evidence of hydrogen bond interaction in the pharmaceutical's adsorption onto PA MP.

Electrostatic Interaction

Oppositely charged solutes and sorbent molecules interact via electrostatic interactions. When both pharmaceutical contaminants and MPs have 14 opposite electric charges, electrostatic interaction takes place as it is more prominent. When the positive and negative charges are opposite, electrostatic sorption occurs, and likewise. Charge at the surface of particles is closely related to pH. The situation where the adsorbent's surface exhibits zero net electrical charges, its pH, is known as the point of zero charges (pH_{PZC}). Under environmental circumstances, the plastics's pH_{PZC} indicates that the most of microplastics's surfaces have a net negative charge, which is why the value is lower than pH_{PZC} [26].

Factor Affecting Adsorption

pH. The pH of a solution may change a compound's physicochemical and biological characteristics, affecting its equilibrium condition, chemical reactivity, and toxicity [27]. Abnormal pH values ranging from 2 to 10 in natural water sources can also occur, widening the range of environmental factors influencing pharmaceutical contaminants' behaviour and fates [28]. *T. Wan et al.* [29] agree that pH has an effect on adsorption rate. The antibiotic sulfamethoxazole showed strong adsorption by all forms of microplastics that are mostly neutral in the pH range of 3 to 6 because of its hydrophobic interactions with the non-polar polymeric surface. Above that pH range, the microplastics's electronegativity increases as this chemical becomes anionic, inhibiting adsorption due to electrostatic repulsion. Both antibiotics have a high adsorption rate in low pH environments.

Salinity. As the salt concentration rises, salinity reduces a non-electrolyte substance's solubility in the presence of ions. Besides, as the solubility of weakly non-polar and polar organic compounds in aqueous medium decreases, inorganic salts can alter the sorption equilibrium of that molecules favoring the organic state. According to some research, salinity appears to have no influences on the adsorption of certain pharmaceutical contaminants on microplastics. The gap in adsorption of sulfamethoxazole and tetracycline was negligible over a range of Sodium Chloride (NaCl) concentrations from 0.05 to 3.5%, according to *B. Xu et al.* [30]. As a result, electrostatic interactions and salinity are unlikely to have played a significant role in sorption.

Molecular Dynamics Simulation

Understanding the sorption mechanism of contaminants on MPs can aid in understanding and predicting the adsorption behaviour of MPs. A molecular dynamics (MD) simulation will be used to study the mechanism of interaction between MPs and contaminants during the sorption process. Up until now, researchers have yet to fully explore the adsorption of molecular dynamic simulation, particularly in relation to the interaction of microplastics with pharmaceutical pollutants. The MD calculation done by *H. Li et al.* [3] shows that the PE's surface adsorbed the three pesticides, namely buprofezin, difenoconazole, and imidacloprid. The adsorption simulation achieved equilibrium within 200 ps. From the simulation results, the PE and pesticide molecules show a complex interaction with no significant differences in adsorption energies. Therefore, the result was concluded to tally with the adsorption thermodynamics data. Table 4 shows the past pieces of literature done with MD simulation on the adsorption of Ethylene Carbonate (EC) onto MP.

Table 4. Studies done for the adsorption of emerging contaminants on MP via MD simulation

References	MP	Contaminants	Findings
[3]	PE	Buprofezin Difenoconazole Imidacloprid	• The MD calculations revealed that the PE's surface adsorbed the three pesticides and achieved equilibrium within 200 ps. • The three pesticides' adsorption energies do not differ significantly. • Within the error range, the results of the theoretical simulation correspond with adsorption thermodynamic data.
[5]	PE PP PS PET PVC	Sulfamethazine	• Sulfamethazine molecules effectively adsorbed on the MPs surface. • The sorption capacity was shown to be directly correlated to the interaction energies between MP and sulfamethazine, as revealed by the isotherm experimental results. • The system's temperature and energy variations were less than 10%, suggesting that the simulation system had attained equilibrium.
[31]	PE	Tetracyclines	• When compared to the layer model of PE and tetracycline prior to MD simulation, the molecule of tetracycline positioned itself towards the PE's surface and distorted after MD simulation.

Conclusions

This mini review demonstrates the importance of water conservation especially from the recalcitrant emerging contaminants such as microplastics and pharmaceutical compounds. The mechanistics overview from experimental and computational point of view of the adsorption between microplastics and pharmaceutical compounds such as Ibuprofen is paramount in order to understand the behavior for successful water conservation process design in future.

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