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CURRENT STATE OF KNOWLEDGE ABOUT PLASTICS' ABILITY TO SORPTION

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Abstract

Plastic pollution is and will be a problem for humanity to deal with for decades to come. The main cause for concern is plastic debris of microsizes, that has been detected in many worrying locations, e.g. human body. Microplastic can carry many substances. It's possible for many substances, including toxins, to accumulate on plastic surface. This review pertains to the compilation of the newest scientific information regarding plastic's ability to sorption. The methodological approach as well as mathematical models used in studies pertaining to this topic have been demonstrated. The type of plastic, the matrix and state of microplastic sample have been described to be affecting sorption on plastic. Some problems with methodology in compiled sorption studies have been outlined. It was concluded that more research is needed to be done to fully grasp this topic.

Keywords: plastic pollution, microplastic, sorption on plastic surface

1. OMNIPRESENCE OF PLASTIC

In recent years, the problem of plastic pollution has become increasingly worrying. In 2022, there were 400 million tonnes of plastic waste produced, with 90% of it consisting of fossil-based polymers [1]. This is an all-time-high for the plastic industry. Such a high number could be even higher as in 2022, many regulatory acts were implemented that were supposed to slow down and minimize plastic manufacturing and usage [2, 3]. China, the rest of South and East Asia (except Japan), and the United States of America are among regions characterized by the highest plastic production [1]. There is no denying the fact that we are surrounded by an unimaginable mountain of plastic waste.

Focusing merely on big, macroplastic is insufficient to understand the whole issue of plastic pollution. Many recent scientific articles have been focused on investigating smaller bits of plastic in the environment. There are no standardized IUPAC definition for these particles, however the term

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'microplastic' (MP) is most commonly used to describe the plastic debris of diameters ranging from 5000 nm to 1000 nm. Plastic particles smaller than this range are referred to as 'nanoplastics' (NPs), and those bigger – as 'macroplastics' [4, 5].

The smaller bits of plastic fragments can be released into the environment in two distinct ways. The primary way is to emit microplastic in the shape and size they have been manufactured as, e.g. by using personal care products that have exfoliating plastic beads. MPs released by the result of weathering and mechanical fragmentation of larger plastic waste already present in the environment, are referred to as secondary microplastics [6].

In Figure 1, the sizes of macro-, micro- and nanoplastics as well as information related to the sources of MPs are depicted.



Fig. 1. The size ranges of different types of plastic debris and main ways of microplastic emission (made using images from freepik.com, based on information found in [6])

Many scientists are focused on the comprehensive study of plastic and microplastic pollution. Today what can be stated as a fact is that plastic is everywhere. Studies have confirmed the occurrence of MP in many locations that should not contain any traces of plastic materials [7, 8]. There are three places that can be accounted as noteworthy when it comes to omnipresence of plastic in the world. The first place pertains to aquatic animals. Studies have confirmed the presence of microplastic in fish, plankton,

and various others marine creatures [8–10]. Many tonnes of improperly disposed plastic waste ends up in Earth's waters every year [11]. This causes seawater to contain high concentrations of MP and NP, which are susceptible to intake by aquatic animals due to their respiration and feeding [4, 12]. Many plastic materials can be characterized as resistant to degradation, causing plastic waste to linger in the environment for hundreds of years [7]. Additionally, when MPs and NPs enter the body of a living being, these plastic fragments can find their way inside of the cells where they can be stored [13]. This creates an effect of accumulation of plastic in the food chain, as plastic amassed by lower trophic level organisms is transferred to higher trophic level organisms upon consumption [8]. The second noteworthy place is food. As has been stated, drinking water, as well as many other food products like honey, fruit, and meat contain varying levels of MP and NP in them [7, 14]. This has led to consumption being one of the main ways of humans' exposure to plastic [5, 7]. The third crucial place is human body. Microplastics have already been detected in human lungs, blood, and placenta [15–17]. Plastic particles can carry many substances with them, mainly those that are used in their manufacturing processes like plasticizers (e.g. bisphenol-A), UV filters (heavy metals among others) and dyes (such as azo dyes) [7, 18]. However, plastic has an ability to sorption – to accumulate on its surface various substances and become their carrier [19]. As for now a worrying scenario is possible in which microplastic particles could mobilize significant amounts of environmentally-present toxins and transport them into bodies of living beings [20, 21].

2. METHODOLOGY OF ADSORPTION STUDIES

Experiments investigating the adsorption process occurring on the plastic's surface are largely using similar methodology [22–24]. These studies are performed by putting a weighed sample of plastic material with a known amount of investigated substance into a vessel containing aqueous matrix. Throughout an allotted amount of time, *e.g.* 96 hours, at certain intervals, measurements of the amount of free substance in the solution are performed. Using such data relevant parameters can be calculated and plotted into the graphs. Then through trial and error, the obtained values are fitted to various kinetics and adsorption isotherm models, allowing to explain recorded sorption processes by mathematical equations [23–25]. Figure 2 shows a graphical representation of this methodology.



Fig. 2. Most commonly used methodology in studies focused on plastic's ability to focus (used graphs are a part of publication authored by Gong et al., cited as [24])

2.1. Kinetics models

The pseudo-first-order (PFO) and pseudo-second-order (PSO) kinetics models can be counted as the most prevalent models used in the studies focused on sorption on plastic surfaces. The first operates on the premise that the rate of sorbate uptake in time is directly corelated to the disparity between sorbate concentration and the amount of sorbate adsorbed in time. The PSO kinetics model however takes into an account limitations present towards the final stages of adsorption [26].

Both of these models are a simplification of a Langmuir kinetics model and are dependent on the initial amount of sorbate in the solution. PFO and PSO have some physical meanings connected to their mathematical equations. PFO best describes kinetics of adsorption processes happening in the presence of vast excess of sorbate or on the surface of a material of little adsorption sites. In contrast, pseudo-second-order model most often describe adsorption of much smaller amounts of sorbate or on adsorbate of many sorption sites. Thus, pseudo-first-order kinetics model is more appropriate towards describing physical adsorption, while pseudo-second-order model better suits chemical adsorption [27, 28].

Kinetics model	Nonlinear equation	Linear equation
Pseudo-first-order	$q_t = q_e \left(1 - e^{-kt} \right)$	$\ln(q_e - q_t) = lnq_e - kt$
Pseudo-second order	$q_t = \frac{q_e^2 k t}{1 + q_e k t}$	$\frac{t}{q_t} = \frac{1}{kq_e^2} + \frac{1}{q_e}$
Elovich	$\frac{dq_t}{dt} = \alpha \; e^{-\beta q_t}$	$q_t = \frac{1}{\beta} \ln\left[t + \frac{1}{\alpha\beta}\right] - \frac{1}{\beta} \ln(\alpha\beta)$
Intraparticle diffusion	-	$q_t = kt^{1/2}$
Boyd's	-	$Bt = 2\pi - \frac{\pi^2 F}{3} - 2\pi \left(1 - \frac{nF}{3}\right)^{\frac{1}{2}}; (F \le 0.85)$ $Bt = -\ln\frac{\pi^2}{6}(1 - F); (0.86 \le F)$

Table 1. Linear and nonlinear equations of described kinetics models [18]

 q_t – amount of sorbate that has undergone sorption [mg/L];

 q_e – adsorption capacity at equilibrium [mg/L];

k – rate constant [unit dependent on the kinetics model]; t – time [h];

 α – initial sorption rate constant [mg g⁻¹ h⁻¹]; β – desorption rate constant [g/mg];

B – Boyd's constant [1/h]; n – number of occupied active sites;

F – fraction of sorbate adsorbed at time to sorbate adsorbed at infinite time.

Some works do include usage of other kinetics models, such as Elovich, intraparticle diffusion and Boyd's models in their calculations [23, 24]. Elovich kinetic model estimates exponential decrease of rate of the adsorption process as the amount of sorbate accumulated on the adsorbate surface increases. It is mostly deployed to further understand adsorption processes chemical in nature [29].

Intraparticle diffusion model (IP), also known as Weber and Morris kinetics model, as well as Boyd's model are most commonly used in identifying the rate limiting step of sorption processes. Adsorption can be described by four elementary mechanisms: the transfer of sorbate to adsorbate as soon as the latter enters the matrix, the attachment of sorbate to the active adsorption sites, the movement of sorbate molecules to the pores of adsorbent, and the diffusion of sorbate from matrix to the adsorbate surface. The first two are too quick in nature, thus they are not considered in kinetic calculations, while the latter two are called intraparticle diffusion and film diffusion respectively. Throughout the whole sorption process both could be the limiting step of adsorption. Fitting the experimental data both of these models could be used to determine the limiting step of sorption process [26, 28, 29].

In Table 1, mathematical equations of described kinetics models are included.

2.2. Isotherm models

While in the case of kinetics there could be some generalizations made about used mathematical models, regarding the isotherm models no such assumptions can be made. In most works, the linear, Langmuir, and Freundlich isotherm models are used. Linear model is based on Henry's law and it is fitted the best towards systems of low initial amount of sorbate molecules leading to low amount of occupied adsorption sites on the adsorbate surface. In some cases, it has been suggested that the Langmuir model approximates to linear model under certain conditions [30]. Langmuir isotherm model describes relation of the area of adsorbate covered by sorbate molecules with the partial pressure or concentration of sorbate at a fixed temperature. This model is based upon four assumptions: adsorbent surface is homogenous, sorbate molecules attach themselves to adsorbent at distinct active sites, only one molecule can be adsorbed to one active site and adsorbed molecules do not interact with each other. Langmuir model is usually fitted towards sorption that is chemical in nature [31]. Freundlich model is an empirical model, mostly having little physical meaning behind it. In many works it has been deployed to investigate sorption processes that take place on the heterogenous adsorbent or those that are physical in nature [30].

In their calculations researchers also try other isotherm models like Temkin and Dubinin-Radushkevich with varying levels of success [23]. Temkin isotherm presumes that sorption process is multi-layer and Temkin equation takes into an account interactions between adsorbent surface and sorbate molecules. This model excludes borderline high and low concentration values of the sorbate substance and assumes that the heat of adsorption of molecules in one layer would decreases linearly [32]. Dubinin-Radushkevich model is based upon Polanyi's potential theory, which estimates that in sorption system potential energy of molecules present are not affected by temperature and increase towards the surface of adsorbent. According to this theory, the potential energy in cracks and pores of adsorbent material is the highest. In Dubinin-Radushkevich model the distribution of these high energy places conforms to the Gaussian energy distribution [30].

In Table 2, the equations of described isotherm models are included with descriptions of used mathematical symbols.

Table 2. Mathematical ec	juation presenti	ng described son	rption isotherm	models [30]
		<u> </u>		

Isotherm model	Equation
Linear	$q_e = K_d C_e$
Langmuir	$q_e = \frac{q_m K_L C_e}{1 + K_L C_e}$
Freundlich	$\log q_e = \log K_F + \frac{1}{n} \log C_e$
Temkin	$q_e = \frac{RT}{b} \ln(AC_e)$
Dubinin-Radushkevich	$q_e = q_{mD-R} e^{-K_{DR} \varepsilon^2}$
	$\varepsilon = RT \ln \frac{C_s}{C_e}$

 $\begin{array}{l} q_e - \text{adsorption capacity at equilibrium [mg/L]; } K_d - \text{partition coefficient [L/g];} \\ C_e - \text{sorbate concentration at equilibrium [mg/L]; } q_m - \text{maximum adsorption capacity [mg/g];} \\ K_F - \text{Freundlich constant [L}^{1/n} \text{ mg}^{1-1/n} \text{ g}^{-1}]; n - \text{constant;} \\ R - \text{gas constant (8.3145 J mol^{-1} K^{-1}); } T - \text{temperature [K]; } A \text{ [L/g], } b \text{ [J/mol]} - \text{constants;} \\ q_{mD-R} - \text{maximum adsorbed amount [mg/g]; } K_{DR} - \text{Dubinin-Radushkevich constant [mol^2 kJ^{-2}];} \\ \varepsilon - \text{adsorption potential [kJ/mol]; } C_s - \text{solubility of sorbate [mg/L].} \end{array}$

2.3. Fitting the data

To verify which model of kinetics and isotherm fits the data the best, various statistical parameters and tests are deployed. Many papers include usage of coefficient of correlation (R^2), sum-of-squared errors and chi-square test [29].

In Table 3, the effects of calculations from few of compiled studies were displayed with values of coefficient of correlation.

Table 3. Calculations results of kinetics and isotherm fittings (used abbreviations: PE – polyethylene, PS – polystyrene, PVC – polyvinyl chloride, PP – polypropylene, PLA – polylactic acid, PBS – polybutylene succinate, PA – polyamide; PSO – pseudo-second-order model; IP – intraparticle diffusion model; PFO – pseudo-first-order model).

Sorbate	Adsorbent	Sorption conditions	Kinetics model	R^2	Isotherm model	R^2	Ref.	
fipronil	PE		PSO	0.996	Langmuir	0.999	[24]	
	PS			0.993		0.997		
	PVC	0.01M CaCl ₂ , room temperature		0.998		0.998		
	PP			0.998		0.999		
	PLA		IP	0.997	Enoundlich	0.999		
	PBS		PSO	0.994	Freundlich	0.998		
	PE		PSO	0.999	Linear	0.983	[33]	
triadimefon	PVC			0.992		0.999		
	PBS	PBS buffer,		0.995		0.983		
difenoconazole	PE	temperature		0.998		0.993		
	PVC			0.992		0.991		
	PBS			0.997		0.991		
sulfamethoxazole		0.01M CaCl ₂ , 0.02% NaN ₃ , room temperature	0.01M CaCl ₂ , 0.02% NaN ₃ , room temperature		0.999	Freundlich	0.977	[34]
propranolol	PE			PSO	0.988	Linear	0.986	
sertraline					0.980	Freundlich	0.995	
	PP		not calculated		Langmuir	0.990	[35]	
	PS					0.968		
ciprofloxacin	PVC				Freundlich	0.998		
	PE					0.994		
	PA	ultrapure water, room temperature			Langmuir	0.980		
trimethoprim	PP				Freundlich	0.964		
	PS					0.992		
	PVC				Langmuir	0.944		
	PE				Freundlich	0.986		
	PA					0.985		

trimethoprim	PP	seawater, room temperature	not calculated		Langmuir	0.917	[35]
	PS				Linear	0.927	
	PVC					0.961	
	PE				Langmuir	0.989	
	PA				Freundlich	0.962	
	PS	saline water, room	PFO	0.943	not calculated		[36]
okadaic acid	PET			0.992			
	PP	temperature	PSO	0.999			
4-chlorophenol	PE	Danube river water, room temperature	PSO -	0.946	0.983		[37]
	PP			0.982	Langmuir	0.969	
	PLA			0.998		0.994	
	PE			0.993	Langmuir	0.999	
2,4- dichlorophenol	PP			0.992		0.988	
ucniorophenoi	PLA			0.999		0.939	
2,4,6- trichlorophenol	PE			0.993		0.992	
	PP			0.999	Langmuir	0.991	
	PLA			0.999		0.977	
pentachlorophenol	PE			0.999	Freundlich	0.996	
	PP			0.995	Langmuir	0.990	
	PLA			0.999	Freundlich	0.993	

3. WHAT AFFECTS THE SORPTION PROCESSES ON THE PLASTIC SURFACE?

Compiled studies investigated sorption on plastic surface of various environmentally present substances, like pesticides, pharmaceuticals, aromatic compounds and even toxins of microbial origin [23, 28–30]. Although any generalizations of adsorption on microplastic cannot be made yet, gathered works show some interesting trends, when it comes to what and what not affects the sorption processes on the plastic's surface.

3.1. Plastic material

The plastic material used during performed experiments has a lot to do with interactions present during sorption processes. In their works, researchers try to use many distinct types of polymers, as each of them express characteristic physical and chemical properties, differentiating it among others [35]. Based solely on their polymeric chain, we can characterize polymers as aliphatic (such as polyethylene, polypropylene or polyvinyl chloride) or aromatic (like polystyrene

and polyethylene terephthalate). Aliphatic and aromatic plastic can interact with sorbate molecules in different ways. In the case of the first type, these interactions are mostly based upon van der Waals forces, while in the case of the latter type – at the base of their interactions with sorbed substances can lay hydrophobicity, planarity and π - π interactions [41].

Polymeric materials can also be categorized based on their biodegradability, that is, an ability to be broken down, due to the influence of microorganisms, into base, primary substances [42]. Over the past decade, there has been a growing demand for plastic materials that are more ecological and less persistent in the environment [43]. There are already some published works focused on finding these new materials [44, 45]. There are however some concerns that can rise from usage of such plastics. Mainly they have to do with the fact that, even with their quick decomposition, biodegradable polymeric materials still produce micro- and nanofragments into the environment [44, 46]. Although more ecofriendly than conventional polymers, these plastic particles can also be persistent if improperly disposed [47]. During their existence in the environment, they can behave similarly to their nondegradable counterparts, adversely affecting the ecosystem [48, 49].

Some of the compiled works were focused on finding differences in sorption processes happening on the surface of nondegradable (e.g. polyethylene PE, polystyrene PS, polypropylene PP) and biodegradable (polybutylene succinate PBS and polylactic acid PLA) polymers. In the case of work by Jiang et al. in 2020, PBS was characterized to have a higher adsorption capacity than PE and PVC (around 2-fold compared to PE and 7-fold compared to PVC) [33]. Higher sorption capacity were also noted in the other works, more notably Concha-Graña et al. [50] and Zuo et al. [51]. However, in the paper authored by Tubić et al., PLA was shown to have lesser capacity of sorption in comparison to PP (in the case of 2,4-dichlorophenol 45.48 μ g/g to 68.98 μ g/g respectively) [37]. There is not enough data available currently to make any generalizations on biodegradability of plastic material having an effect on its sorption capacity can be made. There was however an additional notion presented by researchers pertaining to the effect of the matrix on biodegradable plastic ability to sorption.

3.2. The effect of the matrix

pH level, salinity and the amount of dissolved organic matter can be accounted as the most prevalent parameters of the matrix that can have an effect on the sorption processes happening on the plastic surface [23, 33].

pH level is an amount of hydrogen or hydronium ions in the solution. It is used to describe acidity of a matrix. An increase or decrease in pH can cause changes to both sorbate and sorbent. Sorbed molecule can change its form from ionic to non-ionic and vice versa. This affects the sorption as plastic's hydrophobic nature makes substances similar to it more prone to accumulate on its surface [19]. In Figure 3 the structural changes of ibuprofen due to pH level changes, one of the investigated substances, are presented. Additionally, pH can change the surface charge of microplastic, which also has an effect on the sorption [34].



Fig. 3. Changes to ibuprofen structure due to fluctuation in pH level [52]

Salinity is an amount of salts that are dissolved in the solution. An increase or decrease of cations and anions present in the matrix can have an effect on electrostatic interactions between microplastic and sorbed substances. The dissolved organic matter is mostly made up from humic and fulvic acids. While present in the matrix, they can create complex compounds with sorbate molecules, thus lowering the amount of free, sorbable substance. Additionally, they can be sorbed to plastic surface, therefore blocking active adsorption sites [25, 41].

In the compiled studies it has been shown that increase in salinity and in the amount of dissolved organic matter in the matrix hinders the sorption on the plastic surface [23, 33]. However, a study by Jiang et al. showed that sorption on the surface of biodegradable polymer is not significantly affected by matrix parameters with similar findings present in a study done by Zuo et al. [33, 51]. As of now, there is unfortunately too little scientific data yet available on this topic to fully grasp the effect of the matrix on the sorption processes that take place on the surface of biodegradable plastic. This research gap is needed to be addressed before these new, biodegradable plastics are allowed to the market.

3.3. Environmental weathering

Most compiled studies use pristine, commercially bought microplastic. However, the usage of those materials has been criticized in the scientific community, as those plastic sample are not similar in many ways to MPs present in the environment [53].

Pristine microplastic is most often of sphere, bead or tube-like shape, which are round and dull. While in the ecosystem, MPs are constantly weathered by various substances, sun, water, atmosphere gases, wind, temperature and microbiota. Due to this aging, environmental microplastic is of variety of rugged shapes and of different surface [54]. This weathering can lead to creation of microcanals and pores of bigger size leading to environmental MPs being characterized by having more free surface, thus being able to sorb bigger amounts of sorbate molecules [55, 56]. Aging of microplastic, especially due to sun radiation, can lead to creation of new, reactive function groups, an example of which is shown in Figure 4. Their occurrence on plastic surface causes decrease in its sorption capacity towards hydrophobic organic substances and increase in ability to sorb molecules of ionic nature [39].



Fig. 4. Simplified photoaging of polyethylene [57]

4. METHODOLOGICAL PROBLEMS OF CURRENT STUDIES ON PLASTICS' ABILITY TO SORPTION

Researchers who authored compiled research, do their best to use proper samples and proper methodology, however currently performed research has some substantial limitations. It needs to be underlined that sorption processes that take place on the plastic surface are affected by numerous variables. Experiments performed using described methodology are needed to be carefully planned out, to ensure that acquired data is applicable.

Firstly, the choice of the used matrix as well as investigated substance is crucial. There are more and more works done on sorption processes that use environmental matrix. *e.g.* water from local rivers and lakes [23, 37, 50]. However, very little data is available on plastic ability to sorb in multi-sorbate systems, that is in more akin to real-like systems in which more than one investigated substance is present. In a work of Ho et al., in which such a system was investigated, there has been made claims that in presence of many substances, competitive sorption takes place, leading to decrease in the amount of each sorbate on plastic surface [58]. This notion at current state of knowledge cannot be accepted or denied and this research gap is needed to be filled.

Secondly, the selection of researched polymeric material is imperative. As previously stated, current knowledge about biodegradable plastic ability to sorb, and the differences between them and their nondegradable counterparts, is abysmal. Researchers do need to identify the limitations of data predictability from experiments performed using only pristine microplastics. To battle discrepancies between commercially bought and environmental MPs, the accelerated aging of plastic samples is performed. Done in special apparatus, it is based mostly on sun and temperature weathering

[23, 25, 50, 59]. However, there is an argument that could be made against such a practice. Relying on accelerated aging in order to really age plastic samples and get significant, outstanding results, conditions so far from natural can be created that plastic samples aged in them cannot be used to properly portray the environmental MPs [60]. Researchers can always opt to age their samples in real-time, over the course of e.g. four months [61]. Lastly, the choice of used isotherm and kinetics models can have a limiting effect on use of acquired data to predict sorption processes on plastic surface. To use said models some assumptions, that sometimes cannot be met in real-life, are needed to be taken into an account [29, 31].

Described methodology can be viewed as too far from real-like conditions, to which experiments should strive to be akin. Ideally, scientists should be able to acquire environmental samples, in which the amount of plastic as well as contents of plastic's surface could be determined. Current analytical technology blocks such methodology to be used globally, however in the future such research could potentially be carried out. Pyrolysis gas chromatography mass spectroscopy has already found use in determining the type of plastic in a real-like sample like human blood [16]. Using this method, gas obtained during the pyrolysis of the sample is pumped into the gas chromatography and then analysed [47]. In a work done by Reichel et al. it has been showed that a modification of this technique can lead to properly assessing the contents of plastic surface present in the sample as well as the type of plastic itself. By firstly subjecting the sample to the thermal desorption, before the pyrolysis such analytical information can be acquired [62]. In the future we can hope that by modifying current techniques or developing entirely new methods of analysis scientists will be able to more reliable quantify plastic amount in real-life samples.

Additionally, to ensure applicability and use of acquired data in predicting microplastic effect on human life desorption studies are needed to be performed. Based on compiled research, not every study did contain and take into the account desorption, which could be used to properly quantify the leeching of sorbed substances into living beings. Some works did actually considered this aspect of sorption processes [51, 58, 63, 64]. Most often these tests are done by mimicking what would happen to the plastic sample if it was ingested. Special substances are added (e.g. sodium taurocholate) or special matrixes are used that imitate conditions present in the gut [34, 39, 65].

5. SUMMARY

The aim of this study was to gather and compile the most recent scientific data pertaining to the plastic's ability to sorption. Such reviews are important as plastic pollution are going to a problem for decades to come and spreading knowledge helps to generate new ideas, that can ultimately tackle this problem. There has been a lot of work poured into a better understanding of plastic's ability to sorption. However, there is still much unknown regarding this topic. Sorption on plastic surface is affected by many distinct factors, like type of plastic material, the parameters of the matrix and the state of microplastic. This makes it hard to generalize this process in any way at the current state of affairs. Additionally, there are some methodological problems that are still needed to be addressed properly to actually use this studies to predict MPs' fate in the environment. However, much needed steps towards understanding the effect of plastic pollution on the ecosystem and human life were and are taken. With much more research some of the still troubling notions could be addressed and problems with plastic could be ultimately tackled down.

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