Journal of Hazardous Materials 429 (2022) 128399

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Research Paper

Ranking of potential hazards from microplastics polymers in the marine environment

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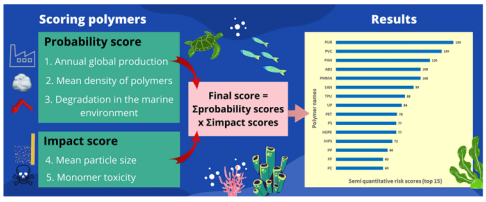
HIGHLIGHTS

- A semi-quantitative approach to rank potential hazards from microplastics has been developed.
- Probability scores are based on the global waste generation of polymers, mean density, and degradability.
- Impact scores are assigned according to the mean particle size of polymers/ monomer toxicity.
- PUR, PVC, PAN, ABS, PMMA, SAN, TPU, UP, PET, PS, and HDPE are found to be top-ranking polymers of concern (descending order).

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Teresa A.P. Rocha-Santos

Keywords:
Risk ranking
Microplastics
Marine pollution
Polymer and monomer
Toxicity

ABSTRACT

Microplastics (MPs) have been detected globally in the marine environment. MP polymers of various kinds have different toxicity potentials when decomposed into monomers. Also, the toxicity of MPs is influenced by the particle size distribution of MPs. Based on these parameters, a semi-quantitative risk assessment model has been developed in this study to rank MP polymers of potential health concern emerging from marine exposure pathways. A screening strategy was used to categorize three probability factors and two impact factors and calculate the final risk scores. Four different scenarios were assessed to investigate the influence of risk factors on the model output. The screening strategy prioritised PUR, PVC, PAN, ABS, PMMA, SAN, TPU, UP, PET, PS, and HDPE as the top-ranking polymers of concern (descending order). The sensitivity analysis revealed parameters that influenced the final risk score were hazard score based on monomer classification (RF5 coefficient +0.60)>

Abbreviations: ABS, Acrylonitrile-butadiene-styrene terpolymer; CLP, Classification, Labelling and Packaging; EP, Epoxy resins; EPS, Expandable polystyrene; EPA, Environmental Protection Agency; EU, European Union; EVA, Ethylene vinylacetate; GHS, Global Harmonized System; GIT, Gastrointestinal tract; HIPS, High-impact polystyrene; HDPE, High-density polyethylene; LDPE, Low-density polyethylene; LLDPE, Linear-low-density polyethylene; MF, Melamine-formaldehyde resin; MPD-I, Poly(m-phenyleneisophthalamide); MP, Microplastic; NP, Nano-plastic; PAA, Polyacrylic acid; PA, Polyamide; PA6, Polyamide 6-nylon 6; PA66, Polyamide 6.6–Nylon 6.6; PA11/12, Polyamide 11 and 12–Nylon 11 and 12; PAN, Polyacrylonitrile; PBT, Polybutylene terephthalate; PC, Polycarbonate; PE, Polyethylene; PET, Polyethylene terephthalate; PF, Phenol formaldehyde resins; PLA, Polylactic acid; PMMA, Poly(methyl methacrylate); PMP, Primary microplastic; POM, Polyoxy-methylene; PP, Polypropylene; PPE / PPO, Polyphenylene ether - also called polyphenylene oxide; PPD-T, Poly(p-phenyleneterephthalamide); PPS, Polyphenylene sulphide; PS, Polystyrene; PTFE, Polytetrafluorethylene; PUR, Polyurethane; PVAc, Polyvinyl acetate; PVC, Polyvinyl chloride; PVDF, Polyvinylidene fluoride; RA, Risk assessment; RF, Risk factor; SAN, Styrene acrylonitrile copolymer; SEM, Scanning electron microscope; SMP, Secondary microplastic; TPU, Thermoplastic polyurethanes; UF, Urea-formaldehyde resin; UP, Unsaturated polyester; UV, Ultraviolet.

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https://doi.org/10.1016/j.jhazmat.2022.128399
Received 11 December 2021; Received in revised form 27 January 2022; Accepted 28 January 2022
Available online 1 February 2022

particle size distribution of MPs (RF4 +0.54)> annual global waste generation (RF1 +0.52)> status of degradation in the marine environment (RF3 +0.32)> mean density of polymers (RF2 +0.16). The outcome of this study can inform the scientific community and the policymakers for better management of MPs where regulation and guidelines need to be considered.

1. Introduction

Since the large-scale manufacture of plastics in the 1950s, plastic generation has been steadily increasing every year. In 2013, the estimated plastic waste in marine water was 268,950 tons and 5.25 trillion particles (Eriksen et al., 2014). Up until 2015, recovery has not yet reached 50% of global plastic disposal (Geyer, Jambeck and Law, 2017). Most of the largest contributing rivers to the world's marine plastic pollution come from Asia, while a few come from East Africa and the Caribbean (Ritchie, 2021). In 2019, Asia accounted for 81% of the annual global marine plastic release, with the top 10 discharging rivers all in Asia (Philippines, India and Malaysia) and most of the top 50 discharging rivers in Asia (Ritchie, 2021). In contrast, Europe discharged only 0.6% (Ritchie, 2021) of global plastic waste into the oceans. It is reported that China, the United States, Germany, Brazil, and Japan (Ritchie, 2021) were the largest contributors to plastic generation, while the United States, China, Japan, Brazil, and Indonesia ranked at the top in terms of plastic waste by coastal population in 2010 (Ritchie, 2021). However, comparing plastic generation per person in 2010, Kuwait, Antigua and Barbuda, St. Kitts and Nevis, Guyana, and Barbados ranked top (Ritchie and Roser, 2018). Mismanaged plastic waste includes littered and inadequately disposed plastics, which are at high risk of being transported by wind or tide into the ocean or carried to shorelines through inland waterways. Ritchie (2021) predicted that by 2025, mismanaged plastic waste would affect most heavily China, Indonesia, the Philippines, Vietnam, India, Nigeria, Bangladesh, Thailand, Egypt, and Sri Lanka (Fig. 1). In 2021, the Philippines (Ritchie, 2021) accounted for 36% of the annual global marine plastic input, 6.52% (4.03 million tons in total and 3.5 kg of plastic per capita) of global annual mismanaged plastic waste, and a 7.17% probability of the global mismanaged plastic waste emitted to oceans. The densely populated small islands and coastal areas have 7 of the top 10 worst plastic polluted rivers of the world. It is reported within the EU (Ritchie, 2021) countries with a more comprehensive plastic waste management and disposal infrastructure on average disposed of less than 0.1 kg of plastic per person. For example, Ireland represented only 0.01% of the annual global plastic waste emitted to the marine environment as mismanaged plastic waste, with a total of 2675 tons of mismanaged plastic waste annually and an average of 0.02 kg of plastic per person annually, with a 2.97% probability of mismanaged plastic waste entering oceans (Ritchie, 2021).

Microplastic (MP) is defined as solid plastic pellets less than 5 mm in the largest dimension (Novotna et al., 2019), while the smallest size (<100 nm or <1 μm, the definition is unclear) is referred as nanoplastics (NPs) (Koelmans et al., 2019a, 2019b). Both MPs and NPs are emerging pollutants in freshwater bodies (including lakes, rivers) and marine systems. MP includes primary microplastic (PMP), which is intentionally manufactured at the microscale and used in consumer products (personal care products and industrial scrubbers), and secondary microplastic (SMP), which is weathered from larger plastic items (plastic film, fishing nets, textile fibres, and household items) (Novotna et al., 2019). Regulations are being implemented to reduce the level of exposure to PMP and SMP worldwide. However, since plastic production has increased and most of the MPs released in marine environments are SMPs with low biodegradability, it is estimated that by 2050, there will be 3.5 trillion tons of macro-plastics and 2.5 million tons of MPs in marine surface water (Lebreton et al., 2017). Based on the polymer type, the plastic global production scale is in the order polypropylene (PP) > linear-low-density polyethylene (LLDPE) and low-density polyethylene (LDPE) > PP and fibre > high-density polyethylene (HDPE) > polyethylene terephthalate (PET) > polyethylene (PE) > additives > polystyrene (PS) (Geyer et al., 2017).

Due to their smaller size and higher volume-surface area ratios, MPs are capable of adsorbing or leaching toxic substances such as persistent

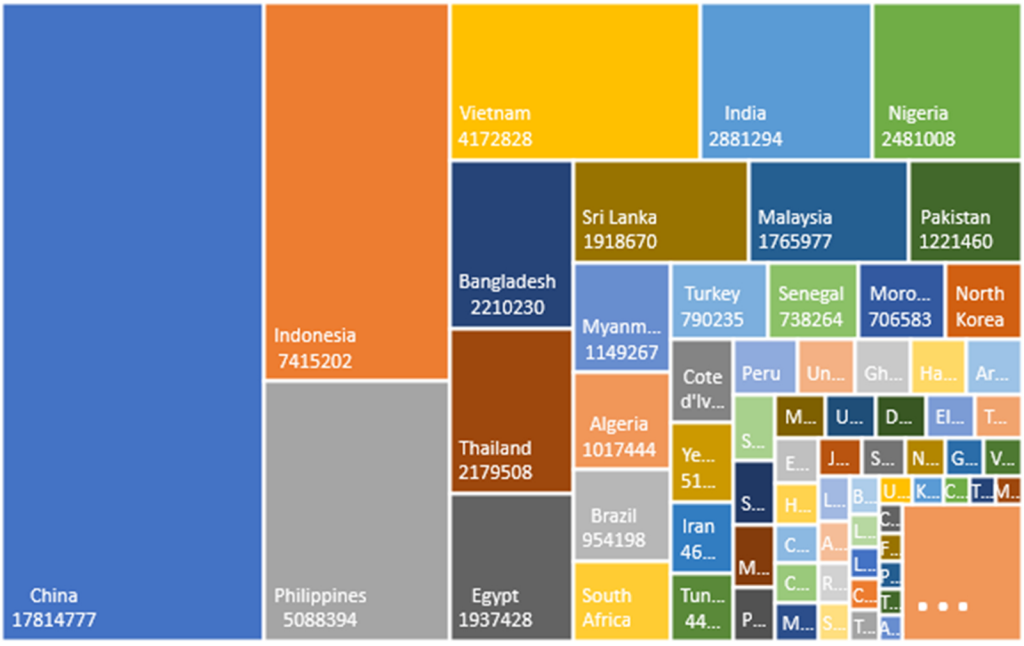

Fig. 1. Estimated total mismanaged plastic waste in 2025.
Data source: Ritchie and Roser (2018).

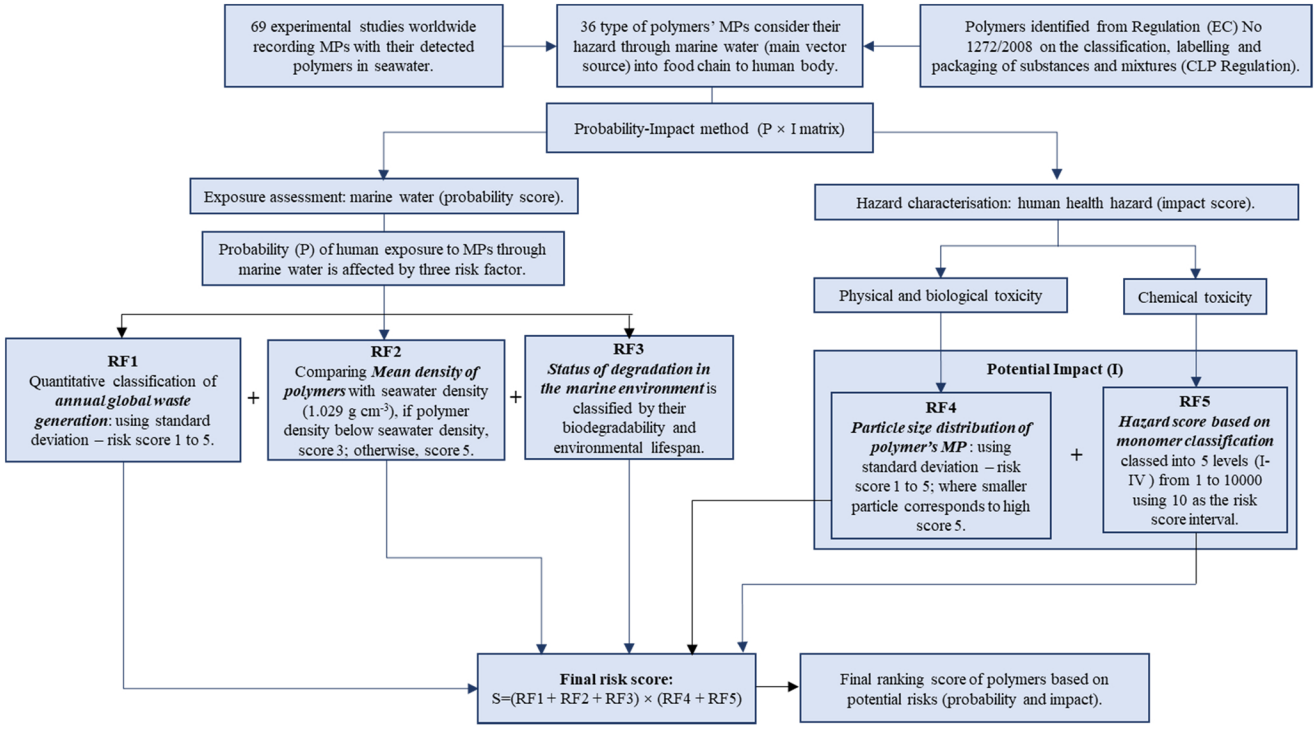

Fig. 2. Schematic presentation of the risk screening method for microplastics in marine water.

organic pollutants or metal particles, and even biofilms of pathogenic bacteria may develop on the surface of particles (Xu et al., 2018). Humans may be exposed to MPs through water, air, soil and food pathways (Revel et al., 2018). Among the potential pathways, direct gastrointestinal (GIT) ingestion, pulmonary inhalation and dermal infiltration are the predominant pathways of human exposure, followed by indirect pathways, including bioaccumulation of MPs through the food chain (Raamsdonk et al., 2020). MP particles themselves can cause wounds or blockages to the gastrointestinal tract, leading to more serious consequences (Revel et al., 2018). In addition, MPs may cause potential human health concerns based on their polymer types. Risk assessment (RA) of MPs is complex because even though the polymers that form MPs are regulated and approved, this does not mean that MPs have the same potential hazard and exposure routes (Bergmann et al., 2015). Lithner et al. (2011) have developed a hazard ranking model for 55 thermoplastic and thermoset polymers based on the EU regulation (EC: European Commission) No 1272/2008 and Labelling (CLP: Classification, Labelling and Packaging) regulation (European Parliament and Council, 2008) on hazard classes and categories of chemicals which are hazardous to the environment and human health. As a result, different polymers will also have different hazard classes when broken down into micro/nano-particles.

As constructed by Codex Alimentarius in 2011 (FAO and WHO, 2021), the steps involved in risk assessment are hazard identification, exposure assessment, hazard characterization, and risk characterisation. At the microscopic scale, structure determines the behavior of a substance (Bergmann et al., 2015). MPs are not on the molecular scale; hence chemical risk assessment models of plastic polymers cannot be applied to MPs. To the authors' knowledge, to date, no study has reported a multi-parameter (probability-impact matrix) ranking strategy of polymers, which is an important aspect of risk assessment/analysis. Qualitative/semi-quantitative risk assessment models have been used in the past to assess the environmental and health hazards of plastic polymers (Lithner et al., 2011). In this research, a similar approach is adopted to rank MP in the marine environment. It is essential to conduct a RA and rank potential hazards from MPs based on their polymer classification in the marine environment. The greatest potential human exposure to MPs is through the food chain, in particular through shellfish, finfish and sea salt. The predominant vector source is via marine water, hence risk ranking of polymers in marine water is necessary to establish which MPs are likely to dominate.

Hence, the overall objective of this study was to rank potential hazards from MPs based on their polymer classification by polymer generation, characterisation of MP density and degradability, pre-characterized toxicity of MP monomers and particle size distribution in marine water using a semi-quantitative RA method.

2. Materials and method

2.1. Data extraction, collation, and harmonisation

Web of Science and Scopus were searched for data up to the end of May 2021 by using the keyword 'microplastic', together with all references cited in each of those selected papers. Beach and seabed sediments have been excluded from this study since their locations are at the intersection of terrestrial and marine environments with contamination from MPs. Airborne MPs may enter the atmosphere and get captured in water droplets as part of the hydrologic cycle and return to terrestrial or marine environments, while others deposit into marine water or sediments (Barletta et al., 2019). Food, including shellfish and fish (Barletta et al., 2019), is an important potential route of human exposure to MPs. MPs can be found in marine water at random depths where MPs may be conferred to humans through the food chain (Barletta et al., 2019). Seafood is the most relevant and fundamental human exposure route to MPs from marine waters (Novotna et al., 2019). Therefore, only peer-reviewed experimental studies on MPs in marine water were considered for this current research.

2.2. Baseline model

A qualitative/semi-quantitative RA model is created for this study using @RISK 7.6 software (PALISADE corporation, New York, USA), an add-in to Microsoft Excel 2016. The model combines qualitative and quantitative RA models by assigning a score to risk factors (RF) and

Table 1
(a) Annual global production, recovery, waste generation, and recovery rates of the selected 36 polymers; (b) Data classification system for risk factor 1 (RF1) using mean and standard deviation of annual global plastic waste generation data; (c) Classification method of RF1.

(a)

Number	Polymer	Abbreviation	Global production (million tonnes year^{-1})	Recovery (million tonnes year^{-1})[a]	Global waste generations (million tonnes year^{-1})	Recovery rate (%)	Reference
1	Low-density polyethylene	LDPE	35.84	3.92	31.92	10.94	Geyer et. (2017); Gnanou and Fontanille (2008)
2	Polypropylene	PP	68	13.00	55.00	19.12	Geyer et. (2017); Lithner et al. (2011)
3	High-density Polyethylene	HDPE	52	12.00	40.00	23.08	Geyer et. (2017); Lithner et al. (2011)
4	Polyethylene terephthalate	PET	33	1.00	32.00	3.03	Geyer et. (2017); Lithner et al. (2011)
5	Polystyrene (general purpose)	PS	25	8.00	17.00	32.00	Geyer et. (2017); Lithner et al. (2011)
6	Polyurethane	PUR	27	11.00	16.00	40.74	Geyer et. (2017); Lithner et al. (2011)
7	Linear-low-density polyethylene	LLDPE	28.16	3.08	25.08	10.94	Geyer et. (2017); Lithner et al. (2011)
8	Polyvinyl chloride	PVC	38	23.00	15.00	60.53	Geyer et. (2017); Lithner et al. (2011)
9	Styrene acrylonitrile copolymer	SAN	0.5	0.16	0.34	32.00 (Same as PS)[b]	Lithner et al., (2011)
10	Acrylonitrile-butadiene-styrene terpolymer	ABS	6.6	2.11	4.49	32.00 (Same as PS)[b]	Lithner et al., (2011)
11	Phenol formaldehyde resins	PF	> 5[c]	–	5.00	0.00	Lithner et al., (2011)
12	High-impact polystyrene	HIPS	6.4	2.05	4.35	32.00 (Same as PS)[b]	Meira and Kiparissides, 2007
13	Expandable polystyrene	EPS	> 4[c]	1.28	2.72	32.00 (Same as PS)[b]	Lithner et al. (2011)
14	Ethylene vinylacetate	EVA	2.6	–	2.60	0.00	China Chemical Reporter (2003)
15	Polycarbonate	PC	2.15	–	2.15	0.00	Lithner et al. (2011)
16	Polyacrylonitrile	PAN	3	0.96	2.04	32.00 (Same as SAN)[b]	Lithner et al. (2011)
17	Unsaturated polyester	UP	> 2[c]	–	2.00	0.00	Gnanou and Fontanille (2008)
18	Polyoxymethylene	POM	1.7	–	1.70	0.00	Beydoun and Klankermayer (2020)
19	Polymethyl methacrylate	PMMA	1.8	0.18	1.62	10.00	Lithner et al. (2011); Stark (2018)
20	Polyamide 6-nylon 6	PA6	1.17	–	1.17	0.00	Lithner et al. (2011)
21	Urea-formaldehyde resin	UF	1	–	1.00	0.00	Gnanou and Fontanille (2008)
22	Polyvinyl acetate	PVAc	2.5	1.51	0.99	60.53 (Same as PVC)[b]	Gnanou and Fontanille (2008)
23	Epoxy resins	EP	0.8	–	0.80	0.00	Gnanou and Fontanille (2008)
24	Polyamide 6.6–Nylon 6.6	PA66	0.59	–	0.59	0.00	Lithner et al. (2011)
25	Polybutylene terephthalate	PBT	0.48	0.01	0.47	3.03 (Same as PET)[b]	Lithner et al. (2011)
26	Polyphenylene ether - also called polyphenylene oxide	PPE / PPO	0.35	–	0.35	0.00	Lithner et al. (2011)
27	Polyacrylic acid	PAA	0.3	–	0.30	0.00	Gnanou and Fontanille (2008)
28	Melamine-formaldehyde resin	MF	0.25	–	0.25	0.00	Gnanou and Fontanille (2008)
29	Polyamide 11 and 12–Nylon 11 and 12	PA11/12	0.2	–	0.20	0.00	Lithner et al. (2011)
30	Thermoplastic polyurethanes	TPU	0.32	0.13	0.19	40.74 (Same as PUR)[b]	Lithner et al. (2011)
31	Poly(m-phenyleneisophthalamide)	MPD-I	0.18	–	0.18	0.00	Lithner et al. (2011)
32	Poly(p-phenyleneterephthalamide)	PPD-T	0.1	–	0.10	0.00	Lithner et al. (2011)
33	Polytetrafluorethylene	PTFE	0.098	–	0.10	0.00	Lithner et al. (2011)
34	Polylactic acid	PLA	0.06	–	0.06	0.00	Avérous (2008)
35	Polyphenylene sulphide	PPS	0.05	–	0.05	0.00	Lithner et al. (2011)
36	Polyvinylidene fluoride	PVDF	0.02	–	0.02	0.00	Lithner et al. (2011)

(b)

Data classification system for risk factor 1 (RF1)	**Annual global waste generation (million tonnes year^{-1})**
Mean	**7.44**
Standard deviation (SD)	**13.28**
Segregation intervals for data classification	**Threshold values for scoring (million tonnes year^{-1})**
Min	0.02
Mean - 0.45 × sd	1.46

(*continued on next page*)

Table 1 (*continued*)

(a)

Number	Polymer	Abbreviation	Global production (million tonnes year^{-1})	Recovery (million tonnes year^{-1})[a]	Global waste generations (million tonnes year^{-1})	Recovery rate (%)	Reference
Mean - 0.15 × sd				5.45			
Mean + 0.15 × sd				9.43			
Mean + 0.45 × sd				13.42			
Max				55			

(c)

Risk factor 1 (RF1)				
Classes	***Classification***	***Annual global waste generation (million tonnes year^{-1})***		***Score***
1	Very low	≥0.02	<1.46	1
2	Low	1.46	<5.45	2
3	Moderate	5.45	<9.43	3
4	High	9.43	<13.42	4
5	Very high	13.42	≤55.00	5

[a] In the absence of data, considered no recovery as the worst-case scenario.
[b] Based on homologous or chemically similar polymers.
[c] If it is >X it has been considered as X.

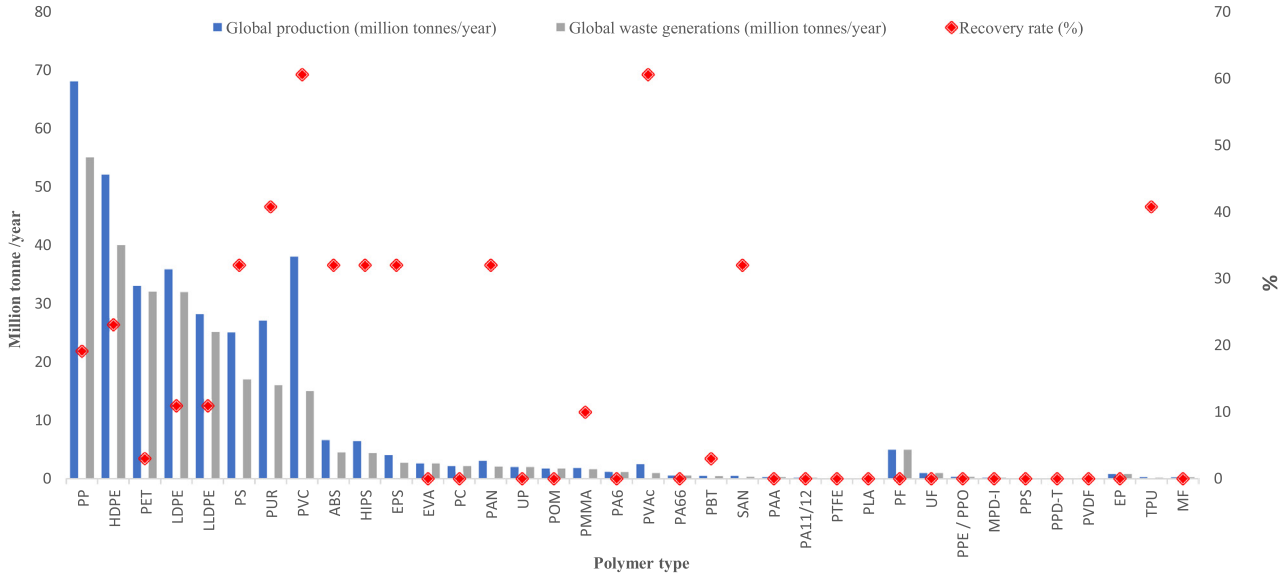

Fig. 3. Annual global production, waste generation, and recovery rates of selected polymers, listed in Table 1.

calculating a final risk score (S). The probability-impact (P × I matrix) method is often used for screening hazards for RA (Nag et al., 2020). The probability (P) scores of human exposure to MPs through marine water sources were derived by three risk factors in this model, including annual global waste generation (RF1), the mean density of polymers (RF2), and status of degradation in the marine environment (RF3). In addition, the impact (I) scores are given based on the physical and biological toxicity (measured by the particle size distribution of MP, RF4) and the potential chemical toxicity (measured by hazard score of polymers based on monomer composition, RF5). A schematic presentation of the ranking method is provided in Fig. 2.

2.2.1. *Annual global waste generation (RF1)*

The annual global waste generation of polymers directly contributes to plastic pollution ultimately released into oceans (Novotna et al., 2019). Plastic release plays a major role in dictating the abundance of MPs in oceans, which greatly affects the probability of human exposure to MPs. Meanwhile, the source of MPs from different marine sites selected for different research studies is vast and complex; hence the MP concentration of specific polymer types in oceans is uncertain. Therefore, in this study, the annual global waste generation of a specific polymer was selected as the first parameter (RF1) of the model. This study assumes that the data presented in Table 1 do not change as technology improves. Also, it is presumed that polymers with similar chemical properties or structures are recycled at equivalent rates. In the absence of data, no recovery was considered for the rest of the polymers as a worst-case scenario. As shown in Table 1a and Fig. 3, the global annual production, recovery, waste generation, and recovery rates are listed for the 36 polymers considered in the study. Annual global waste generation data is divided into five equal interval segments using mean and standard deviation (0.3 as the coefficient factor, resulting in data segregation by 'Mean - 0.45 × sd', 'Mean - 0.15 × sd', 'Mean + 0.15 × sd', 'Mean + 0.45 × sd') to get an equal distribution for the data classification system of RF1 (Table 1b). Then, RF1 are classified into 5

Table 2
(a) Mean density of the selected 36 polymers; (b) Data classification system of risk factor 2 (RF2) using mean density data of polymers.

(a)

Number	Polymers	Mean density (g cm^{-3})	Reference
1	LDPE	0.93	Andrady (2017)
2	PP	0.90	Oni et al. (2020)
3	HDPE	0.96	Oni et al. (2020)
4	PET	1.38	Oni et al. (2020)
5	PS	1.05	Chubarenko and Stepanova (2017)
6	PUR	1.31	UL Prospector (2021)
7	LLDPE	0.93	Andrady (2017)
8	PVC	1.38	Wagner and Lambert (2018)
9	SAN	1.08	SpecialChem (2021)
10	ABS	1.07	SpecialChem (2021)
11	PF	1.20	ChemicalBook (2016)
12	HIPS	1.08	SpecialChem (2021)
13	EPS	0.03	Nabizadeh et al. (2019)
14	EVA	0.94	SpecialChem (2021)
15	PC	1.22	Wagner and Lambert (2018)
16	PAN	1.18	Liu et al. (2019)
17	UP	1.12	Dmitri_kopeliovich (2012)
18	POM	1.41	Wagner and Lambert (2018)
19	PMMA	1.18	SpecialChem (2021)
20	PA6	1.14	Andrady (2017)
21	UF	1.20	Wypych (2016)
22	PVAc	1.29	Polymer Properties Database (2019)
23	EP	1.21	Qianming et al. (2008)
24	PA66	1.31	SpecialChem (2021)
25	PBT	1.31	SpecialChem (2021)
26	PPE / PPO	1.06	SpecialChem (2021)
27	PAA	1.30	Liu et al. (2019)
28	MF	1.50	ChemicalBook (2016)
29	PA11/12	1.04	SpecialChem (2021)
30	TPU	1.23	UL Prospector (2021)
31	MPD-I	1.38	Merck KGaA (2022)
32	PPD-T	1.44	ChemicalBook (2016)
33	PTFE	2.20	SpecialChem (2021)
34	PLA	1.24	Wagner and Lambert (2018)
35	PPS	1.35	SpecialChem (2021)
36	PVDF	1.78	SpecialChem (2021)

(b)

Risk factor 2 (RF2)			
Classes	***Classification***	***Mean density of polymers (g cm^{-3})***	***Score***
1	Moderate	$\rho<\rho s$ or $\rho=\rho s$	3
2	High	$\rho>\rho s$	5
Marine water density: $\rho s = 1.029$ g cm^{-3}			

categories (very low, low, moderate, high and very high) based on the data classification system corresponding to the score levels from 1 to 5 (Table 1c).

2.2.2. *Mean density of polymers (RF2)*

The decomposition of large marine plastic waste into microscale particles is inversely proportional to its density. When polymer density is less than that of marine water (1.029 g/cm^3) (Nayar et al., 2016), plastic waste floats on the sea surface, facilitating its degradation and even completely decomposition through biodegradation, Ultraviolet (UV) light, and weathering (Novotna et al., 2019). When the polymer density is greater than or equal to marine water, the suspended MPs in marine water have more potential to result in human exposure through the marine environment. Also, if high-density MPs settle on the seabed, they are inevitably contaminated by bacteria, algae, and heavy metals (Reve et al., 2018). As MPs settle, the surface area of MPs which is exposed to UV light and oxygen, is reduced. As a result, the degradation rate is dramatically reduced (Novotna et al., 2019). However, due to the movement of ocean currents and animal locomotion, the heavier MPs are also resuspended in marine water (Novotna et al., 2019). When MPs absorb heavy metals and toxic substances and become hetero-aggregated with microbial biofilms, the complex may cause greater harm to aquatic organisms and human health as they enter the food chain. In addition, toxic additives, metals, and oxides remaining in plastic waste will be re-released when broken down into micron-sized particles due to the ageing process (Rist et al., 2018). Table 2a and Fig. 4 show the mean density of selected polymers in comparison with marine water density. In the data classification system of RF2, if polymer density is below marine water density, it was scored 3; otherwise, it was assigned a higher score of 5 (Table 2b).

2.2.3. *Status of degradation in the marine environment (RF3)*

The decomposition of plastics in marine water is a particularly slow process. At present, it is widely known that even degradable plastics take at least 5 years to completely degrade in the marine environment (Ward and Reddy, 2020). Common popular polymers have a degradation time of 10–20 years or 500–1000 years, and most have lifespans of between 70 and 450 years (Ward and Reddy, 2020). However, these data are uncertain and based on estimates, and there is no real evidence to support its validation. Also, the physical and photolytic forces in oceans significantly retards the process of oxidative decomposition of plastics, especially in deep water, where plastics also accumulate negative buoyancy (Ward and Reddy, 2020). Due to their more prominent volume-to-area ratio, MPs are more likely to sorb pollutants in oceans

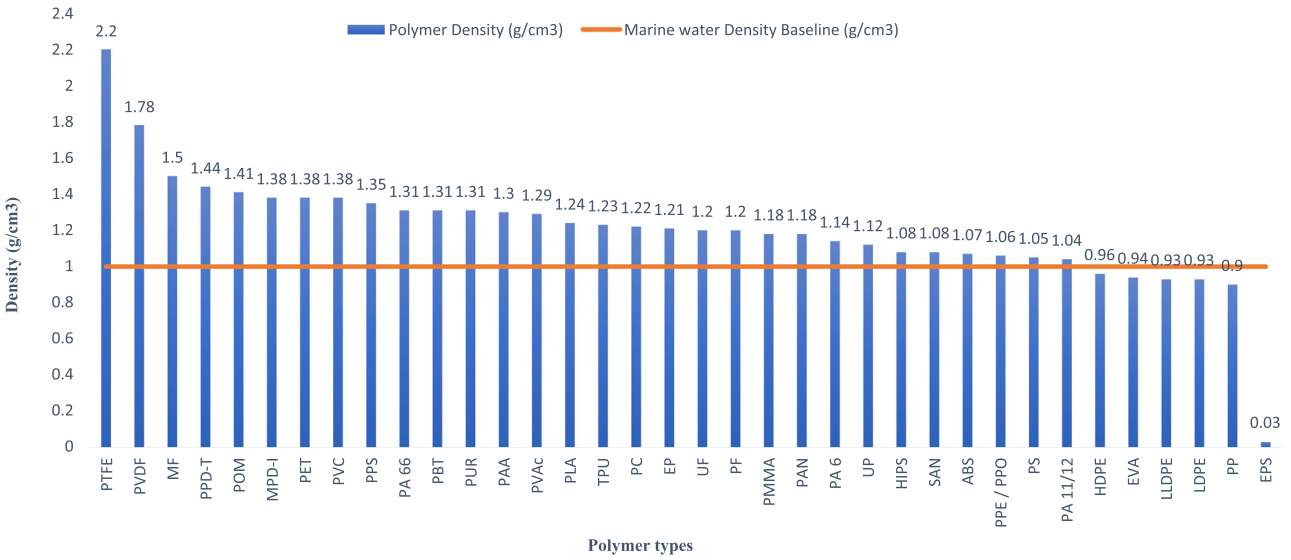

Fig. 4. The density of the selected polymer's presented in Table 2 and marine water density (baseline).

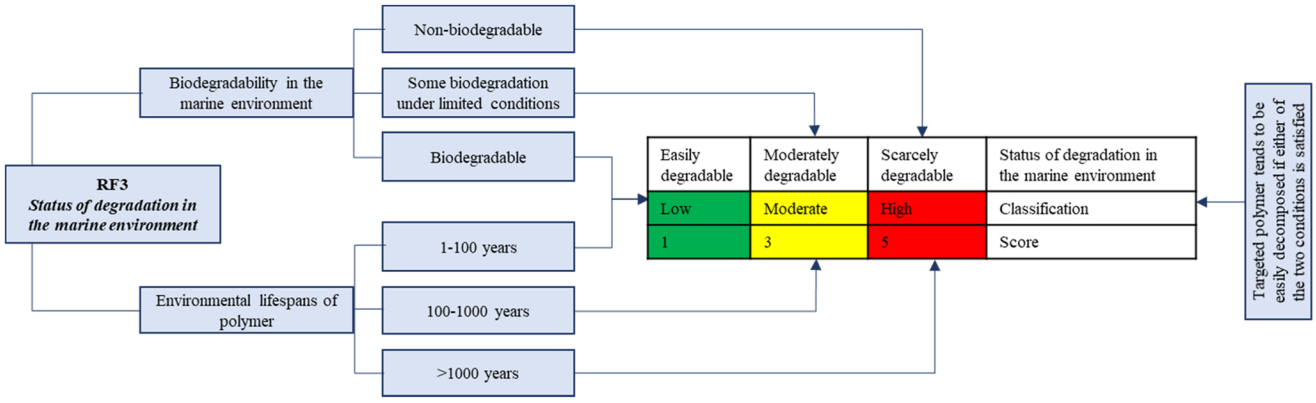

Fig. 5. Scoring strategy for degradability of plastics in the marine environment (risk factor (RF) 3).

and harbour microbial biofilms (Revel et al., 2018). This leads to extended dwell time in the marine environment, with a higher probability of human exposure to MPs by marine water, and potentially greater risk to human health. The earliest human use of plastic was in the early 1900 s, and the earliest plastic waste that can be detected is only 60 years old (Geyer, Jambeck and Law, 2017). There is no carbon detection technology to accurately measure the age of MPs, while chemical analysis (FTIR or Raman spectroscopy) has been used to quantify the level of weakening of plastics. As a result, it is not possible to extrapolate their longevity from this information in the marine environment (Laycock et al., 2017). Age estimation is complicated by the wide variation in weathering rates of plastic waste due to differences in polymer types, additive compositions, and environmental factors (Laycock et al., 2017). For instance, MP particles can move long distances either because they are in the rapid flow or simply because they have been in the ocean for a long time (Geyer, Jambeck and Law, 2017).

In this study, a new conception of the state of degradation in the marine environment was defined by combining data on environmental lifespan and biodegradability in the marine environment of the polymers based on information obtained in previous studies. Under the new interpretation (score strategy showed in Fig. 5, polymers are categorized as easily degradable (scored 1), moderately degradable (scored 3) and scarcely degradable (scored 5). Easily degradable means the polymer has an environmental lifespan of fewer than 100 years or is biodegradable. Likewise, moderately degradable signifies the polymer has an environmental lifespan between 100 and 1000 years or is partially biodegradable under certain conditions. Finally, scarcely degradable denotes the polymer has an environmental lifespan greater than 1000 years or is non-biodegradable. Thus, the targeted polymer tends to be viewed as scarcely degradable as long as either of the two conditions is satisfied. Table 3 and Fig. 6 show the status of degradation in the marine environment of different polymers.

2.2.4. *Particle size distribution of MPs (RF4)*

As plastic polymers typically have high molecular weights and lack natural analogues in marine environments (Lusher, Hollman and Mandoza-Hill, 2017), most conventional plastics are not easily biodegradable in marine or terrestrial environments. Plastic polymers are only physically degradable to some extent, for example, by physical forces such as waves or the abrasive action of sediment particles that fragment plastic waste into smaller pieces, but without altering the total mass of the plastic waste (Lusher, Hollman and Mandoza-Hill, 2017). Therefore plastic polymers typically only transform in size and distribution. In addition, when plastics are exposed to oxygen and UV radiation from sunlight, plastic polymers are oxidised, forming hydroperoxides which cause polymer chain breakage (Novotna et al., 2019).

The particle size of MPs is closely associated with tissue accumulation kinetics and distribution patterns (Rist et al., 2018). Particles of MPs smaller than 5 μm can be internalised by organisms through endocytosis by honeycomb cells, while particles of MPs larger than 0.5 μm may be

Table 3
Status of degradation in the marine environment of different polymers.

Number	Polymers	Environmental lifespans of common polymer/years[a]	Biodegradability in the marine environment	Status of degradation in the marine environment	Reference
1	LLDPE	450	Some biodegradation under limited conditions	Moderately degradable	Lusher et al. (2017)[b]
2	ABS	Forever	Non-biodegradable	Scarcely degradable	Lusher et al. (2017)
3	EPS	65	Non-biodegradable	Easily degradable	Lusher et al. (2017) [b]
4	EVA	450	Non-biodegradable	Moderately degradable	-[b]
5	HDPE	450	Some biodegradation under limited conditions	Moderately degradable	Lusher et al. (2017)[b]
6	HIPS	65	Non-biodegradable	Easily degradable	-[b]
7	LDPE	450	Some biodegradation under limited conditions	Moderately degradable	Ward and Reddy (2020); Lusher et al. (2017)
8	MF	Forever	Some biodegradation under limited conditions	Moderately degradable	El-Sayed, El-Baz and Othman (2006)
9	MPD-I	Forever	Non-biodegradable	Scarcely degradable	Zhang et al. (2017)
10	PA66	600	Non-biodegradable	Moderately degradable	Ward and Reddy (2020); Lusher et al. (2017)
11	PA6	600	Non-biodegradable	Moderately degradable	Ward and Reddy (2020); Lusher et al. (2017)
12	PA11/12	600	Non-biodegradable	Moderately degradable	Ward and Reddy (2020); Lusher et al. (2017)
13	PAA	450[c]	Non-biodegradable	Moderately degradable	Woods Hole Sea Grant Program. (2015); Polymer Science Learning Center, 2021;Ghafoori, Mehrvar and Chan (2014)
14	PAN	Forever	Non-biodegradable	Scarcely degradable	Lusher et al. (2017)
15	PBT	450	Non-biodegradable	Moderately degradable	-[b]
16	PC	Forever	Some biodegradation under limited conditions	Moderately degradable	Lusher et al. (2017)
17	PET	450	Non-biodegradable	Moderately degradable	Ward and Reddy (2020); Lusher et al. (2017)
18	PF	Forever	Non-biodegradable	Scarcely degradable	Chen et al. (2008)
19	PLA	0.5–1	Non-biodegradable	Easily degradable	Laycock et al. (2017); Lusher et al. (2017)
20	PMMA	Forever	Non-biodegradable	Scarcely degradable	Lusher et al. (2017)
21	POM	Forever	Non-biodegradable	Scarcely degradable	Lusher et al. (2017)
22	PP	200	Non-biodegradable	Moderately degradable	Ward and Reddy (2020); Lusher et al. (2017)
23	PPD-T	Forever	Non-biodegradable	Scarcely degradable	Lopez-Perez (2016)
24	PPE / PPO	Forever	Non-biodegradable	Scarcely degradable	Stack, O'Donoghue, and Birkinshaw (2003)
25	PPS	Forever	Non-biodegradable	Scarcely degradable	Li et al. (2020)
26	PS	50	Non-biodegradable	Easily degradable	Ward and Reddy (2020); Lusher et al. (2017)
27	PTFE	Forever	Non-biodegradable	Scarcely degradable	Zhao (2011); Lusher et al. (2017)
28	PUR	Forever	Non-biodegradable	Scarcely degradable	Lusher et al. (2017)
29	PVAc	78.65[d]	Non-biodegradable	Easily degradable	Calculated from Chai et al. (2009); Lusher et al. (2017)
30	PVC	Forever	Non-biodegradable	Scarcely degradable	Lusher et al. (2017)
31	PVDF	Forever	Non-biodegradable	Scarcely degradable	De Jesus Silva et al. (2020)
32	SAN	Forever	Non-biodegradable	Scarcely degradable	Jang and Wilkie (2005)
33	TPU	Forever	Non-biodegradable	Scarcely degradable	Puentes-Parodi et al. (2019)
34	UF	Forever	Non-biodegradable	Scarcely degradable	Arshad et al. (2016)
35	UP	Forever	Non-biodegradable	Scarcely degradable	Hashemi, Jamshidi and Aghdam (2018)
36	EP	Forever	Non-biodegradable	Scarcely degradable	Lusher et al. (2017)

Note
[a] When "forever" is given as the lifespan estimate, 10,000 years was used for the calculations.
[b] Based on homologous or chemically similar polymers.
[c] A lifespan of the main use products in the marine environment. For example, PAA is usually used in diapers (Woods Hole Sea Grant Program., 2015); Polymer Science Learning Center, 2021; Ghafoori, Mehrvar and Chan, 2014).
[d] Predicted environmental lifespan accordion to the study of Chai et al. (2009).

taken up through phagocytosis by macrophages in the small intestinal epithelium (Revel et al., 2018). Lusher, Hollman and Mandoza-Hill (2017) reported that MPs (0.1–150 μm, human: 0.2–150 μm) are transportable to the mammalian lymphatic system. On the one hand, MPs have physical toxicity, including intestinal obstruction or tissue abrasion by the progress of intestinal immune system activation and inflammation response (Rist et al., 2018). Other potential adverse effects of MPs include oxidative stress and inflammation, genotoxicity, apoptosis and necrosis, and even tissue damage, fibrosis and carcinogenesis (Lusher, Hollman and Mandoza-Hill, 2017). Given MPs have a large surface area to volume ratio and surface activity, they may induce pathogenic microorganisms to form biofilms (e.g. *Vibrio* spp.) on the surface of MPs (Revel et al., 2018). The particle size of the MPs decisively affects the degree of biological toxicity (Eerkes-Medrano et al., 2019).

The smaller the particle size of the MPs, the more physical and biological toxicity it may pose to human health. In this study, the mean particle size of MPs for specific polymers in marine environments was adopted as an essential impact factor of the risk level of MPs, based on the particle size data of MPs for individual polymer types recorded in previous studies (Table 4a and Fig. 7). Particle size distribution data of MPs is divided into five equal interval segments using mean and standard deviation (0.3 SD as the interval) to get an equal distribution for the data classification system of RF4 (Table 4b). Then, RF4 (physical and biological toxicity) are classified into 5 categories (very low, low, moderate, high and very high) based on the data classification system corresponding to the score levels from 1 to 5 (Table 4c).

2.2.5. *Hazard score based on monomer classification (RF5)*

Chemical toxicity is another important impact factor in assessing the risk of MPs to human health. Lithner et al. (2011) compared different polymers and classified their toxicity with a hazard ranking model. According to toxicity to humans, the hazards are divided into three categories viz. acute and chronic effects on aquatic organisms (carcinogenicity and mutagenicity), specific target organ toxicity, and acute oral toxicity; and polymers were ranked into five levels based on their

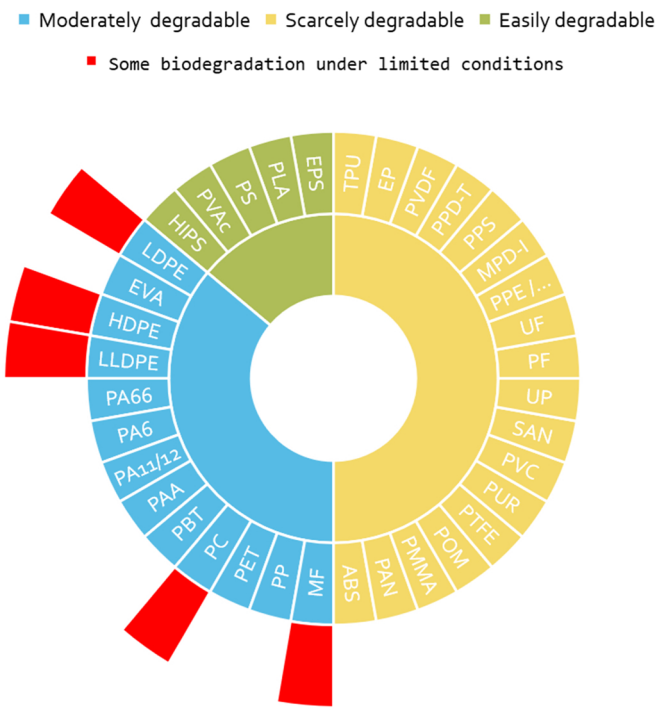

Fig. 6. The status of degradation of different polymers in the marine environment.

severity. According to the composition of the polymer, risk scores have been calculated in this study. The common variables in the model are the monomers used to produce different plastics compounds, including prepolymers, comonomers and plasticiser (Tebby et al., 2011). However, variables do not include polymerisation additives (such as initiators, catalysts, light stabilisers, flame retardants, chain transfer agents, suspending aids, surfactants and pigments), solvents, and monomer synthesis materials (Bergmann et al., 2015). Again, plastic polymer formulation is uncertain due to the involvement of different manufacturers (Bergmann et al., 2015).

Hazard classification criteria for substances used in polymer production are entirely based on EU CLP regulations which are further based on the United Nations Globally Harmonized System (GHS) (Lithner et al., 2011). The hazard levels V and IV correspond to the "phasing out" substance (V) and "risk-reducing" substance (IV) standards set by the Swedish Chemical Agency (Kass and Merten, 2018). Levels I to III come from assessing hazard categories in the GHS system (Kass and Merten, 2018). Each level of I-IV corresponds to an approximate risk score interval (a factor of 10 from 1 to 10,000) (Lithner et al., 2011) (Table 5). Choosing 10 as the multiplier between hazard categories provides a good distinction between five hazard levels (1−10,000) (Lithner et al., 2011). Therefore, the sum of hazard scores for the polymer is based on its monomer classifications (1−10,000). If multiple monomers are used in polymer production, the average weight fraction (wt%, usually converted from moles or percentages) of each monomer is multiplied by the hazard score of each monomer, and the combined sum is used to assess hazard levels of different polymers (Lithner et al., 2011) (Fig. A1).

$$H = \sum_{i=1}^{n}(P_{M_i} \times \sum_{s=1}^{m} h_s)\ldots \quad (1)$$

where, the left side of the equation is expressed as the hazard score of polymer (H). The terms of the right-hand side are the sum of monomers' (M_i) hazard level score (h_s) by percentage (P_{Mi}). The number of monomers used in the production of polymer is shown as n and i. Meanwhile, the number of hazard classes is shown as m and s.

2.3. The final score for the baseline model (BM) and other scenarios

The final score (S_{BM}) of the baseline model (BM) is based on the multiplication of the sum of probability score (RF1, RF2 and RF3) with the sum of potential impact score (RF4 and RF5) (Eq. 2). Scenario 1 considered all risk factors to be added (Eq. 3) to obtain the final score (S_1) instead of the probability-impact matrix. It is used to assess the influence of impact score (RF4 and RF5), toxicity (physical, biological and chemical toxicity). Scenario 2 was chosen to assess the influence of chemical toxicity factor (RF5, hazard score based on monomer classification) to final score (S_2) by adding physical and biological toxicity (RF4, particle size distribution of MPs) to the probability score instead of impact score (Eq. 4). Compared to the BM, scenario 3 only adds three probabilities together (Eq. 5) and eliminates the influence of impact factors to get the final score (S_3) to assess the influence of probability score (RF1, RF2, and RF3). Furthermore, MPs' degradation in the marine environment is hard to determine at present, as it may take 50–100 years or even more time from now for validation. Therefore, compared to BM, scenario 4 was used to evaluate the influence of errors caused by the degradability of polymers (RF3) through observing the final score (S_4), which eliminates the influence of RF3 in Eq. (6).

$$S_{BM} = (RF1 + RF2 + RF3) \times (RF4 + RF5)\ldots \quad (2)$$

$$S_1 = RF1 + RF2 + RF3 + RF4 + RF5\ldots \quad (3)$$

$$S_2 = (RF1 + RF2 + RF3 + RF4) \times RF5\ldots \quad (4)$$

$$S_3 = RF1 + RF2 + RF3\ldots \quad (5)$$

$$S_4 = (RF1 + RF2) \times (RF4 + RF5)\ldots \quad (6)$$

2.4. Scientific corroboration

To improve the validation of the baseline model outputs, it is critical to check with the monitoring data on MPs in marine environments from past studies. But full validation isn't possible in this model, and scientific corroboration is done by collating data from 144 published journal articles that presented results for MPs in the aquatic environment. By collating studies that reported MP monitoring results from different regions, it acted as a tentative qualitative corroboration for the model as it highlights common MPs recorded around the globe. Also, a word cloud could be used to quantitatively show the frequency of occasions of reported polymer's MPs.

3. Results and discussion

3.1. The baseline model (BM)

The breakup values of five risk factor scores (RF1, RF2, RF3, RF4 and RF5) of 36 polymers and the final scores (S_{BM}) are presented in Table 6. The highest-ranked 10 polymers are plotted on a bar chart (Fig. 8) according to their order from high to low as PUR (150), PVC (135), polyacrylonitrile (PAN) (120), acrylonitrile-butadiene-styrene terpolymer (ABS) (108), polymethyl methacrylate (PMMA) (108), styrene acrylonitrile copolymer (SAN) (99), thermoplastic polyurethanes (TPU) (88), unsaturated polyester (UP) (84), polyethylene terephthalate (PET) (78) and PS (77). Fig. 9 shows the cumulative bin distribution of the final score (S_{BM}) for 36 polymers.

3.2. Scenarios

A comparison of the final risk score of the baseline model (S_{BM}) with the final risk score of all scenarios (S1, S2, S3, S4) is presented in Table 7. Compared to the baseline model, the common polymers ranked in the top 10 of all scenarios (S1, S2, S3, S4) include PUR, PVC, and PMMA. In

Table 4
(a) Mean MPs particle size (μm) of the selected 36 polymers; (b) Data classification system of risk factor 4 (RF4) using mean and standard deviation of particle size distribution data of MPs; (c) Classification method of RF4.

(a)

Number	Abbreviation	Shape	Size/μm	Mean size/ μm	Reference
1	LDPE	Film	500–2000	1250	Zhao et al. (2018)
2	PP	Fragment	500–1000, 15–1660	794	Calculated fromZhao et al. (2018) andVianello et al. (2013)
3	HDPE	Fragment	500–1000	750	Zhao et al. (2018)
4	PET	Fragment	500–1000	750	Zhao et al. (2018)
5	PS	Fragment/ Sphere	500–1000, 42–529, 6–40, 10–500	295	Calculated fromECHA (2019):Zhao et al. (2018) andVianello et al. (2013)
6	PUR	Powder/ Sphere	7–30, 7–60	26	Calculated fromECHA (2019)
7	LLDPE	–	–	1250	Same as LDPE[a]
8	PVC	Fragment	500–1000, 60–163	431	Calculated fromZhao et al. (2018) andVianello et al. (2013)
9	SAN			295	Same as PS[a]
10	ABS	–	–	295	Same as PS[a]
11	PF	–	–	2665	Same as POM[a]
12	HIPS			295	Same as PS[a]
13	EPS	–	–	295	Same as PS[a]
14	EVA	–	–	1250	Same as LDPE[a]
15	PC	–	70–5000[b]	1300	Xu et al. (2018) ; Liu et al. (2019)[b]
16	PAN	–	18–950	484	Vianello et al. (2013)
17	UP	–	15–2413	1214	Vianello et al. (2013)[a]
18	POM	–	330–5000[b]	2665	Ivleva, Wiesheu and Niessner (2017)[b]
19	PMMA	Sphere, hemisphere/ Sphere/ Powder	5–50, 5–200, 6–40, 1–50, 2–12	37	Simulated mean calculated fromECHA (2019)
20	PA6	fibre		1233	Calculated fromZhao et al. (2018) and Vianello et al. (2013)
21	UF	–	–	2665	Same as POM[a]
22	PVAc	–	93	93	Vianello et al. (2013)[a]
23	EP	–	23–5000[b]	2512	Liu et al. (2019)[b]
24	PA66	fibre	500–3000	1233	Calculated from Zhao et al. (2018) and Vianello et al. (2013)
25	PBT	–	–	750	Same as PET[a]
26	PPE / PPO	–	0–5000	2500	-[c]
27	PAA	–	300–5000[b]	2650	Chen et al., (2020)[b]
28	MF	–	–	2665	Same as POM[a]
29	PA11/12	fibre		1233	Calculated from Zhao et al. (2018) and Vianello et al. (2013)
30	TPU	–	–	26	Same as PUR[a]
31	MPD-I	–	0–5000	2500	-[c]
32	PPD-T	–	0–5000	2500	-[c]
33	PTFE	–	3–15, 25–500, 45–5000	931	Calculated from ECHA (2019) and Zhao et al. (2020)
34	PLA	–	0–200[b]	100	Kazour et al. (2019)[b]
35	PPS	–	0–5000	2500	-[c]
36	PVDF	–	0–5000	2500	-[c]

(b)

Data classification system for risk factor 4 (RF4)	**Mean particle size of targeted polymer's MPs (μm)**
Mean	1248
Standard deviation (SD)	976
Segregation intervals for data classification	**Threshold values for scoring (μm)**
Max	2665
Mean + 0.45 × sd	1688
Mean + 0.15 × sd	1395
Mean - 0.15 × sd	1102
Mean - 0.45 × sd	809
Min	26

(c)

Risk factor 4 (RF4)				
Classes	***Classification***	***Mean particle size of targeted polymer's MPs (μm)***		***Score***
1	Very low	≤2665.00	<1688.45	1
2	Low	1688.45	<1395.37	2
3	Moderate	1395.37	<1102.29	3
4	High	1102.29	<809.22	4
5	Very high	809.22	≥26.00	5

Note:
[a] Based on homologous or chemically similar polymers.
[b] Microplastic found in this study includes that kind of polymer.
[c] In the absence of data, the average value of the prevailing size (1 μm-1 mm) of MPs was selected (2500 μm) (Bergmann et al., 2015; Ivleva et al., 2017).

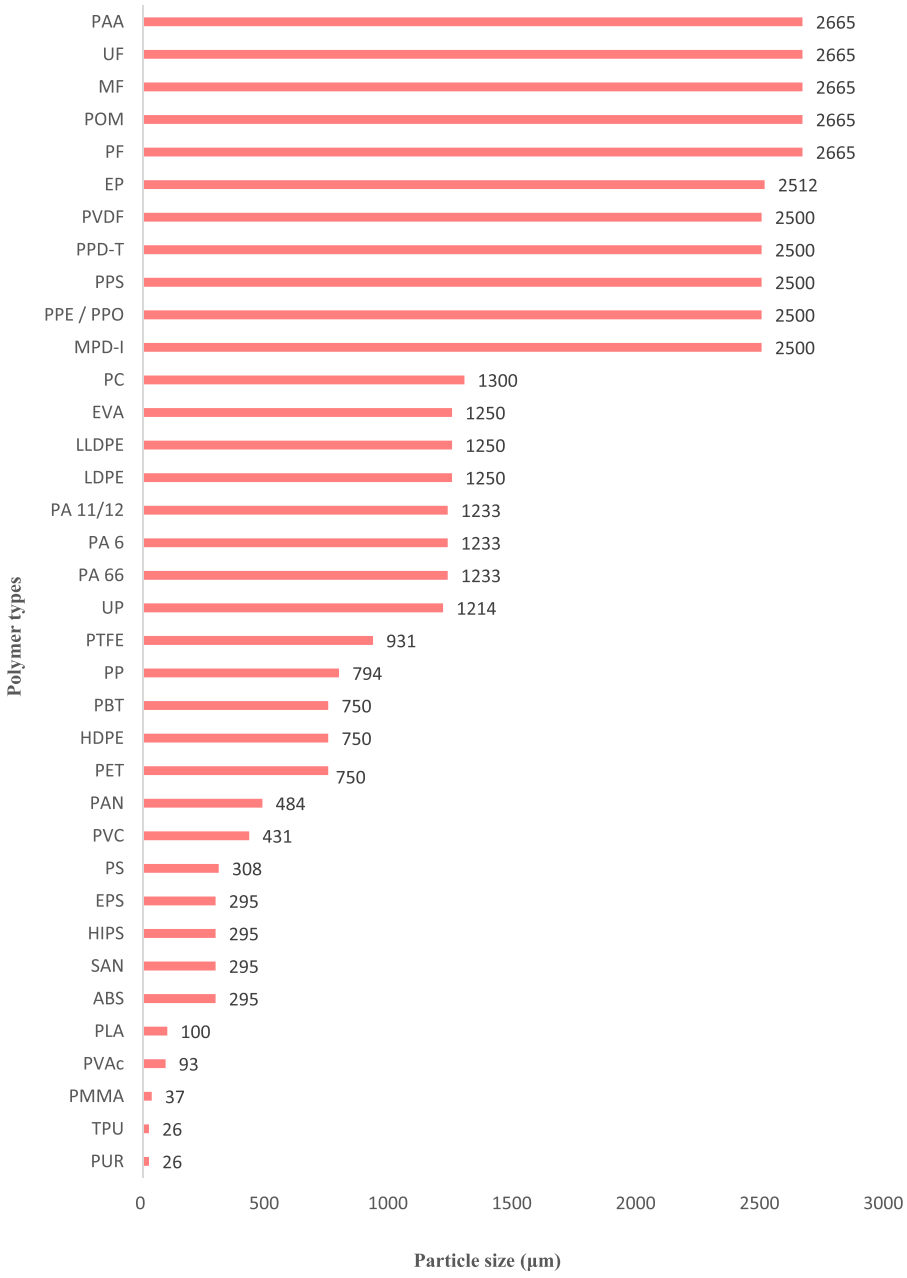

Fig. 7. Mean microplastics particle size (μm) of the selected polymers from Table 4a.

Table 5
Data classification system of risk factor 5 (RF5).

		Risk factor 5 (RF5)		
Classes	***Classification***	***Hazard score based on monomer classification* (Lithner, Larsson and Dave, 2011)**		***Score***
1	Very low	≤1	<10	1
2	Low	10	<100	2
3	Moderate	100	<1000	3
4	High	1000	<10000	4
5	Very high	10000	-	5

addition, the scientific corroboration in Table 7 shows that the most common polymers are found in past microplastic studies include PP (131), followed by PS (94), PET (61), PVC (49), LDPE (26), PUR (21), HDPE (19), PMMA (17), EPS (14), and PVAc (13). Scenario 1 shows approximately the same trends as the baseline model rankings, which means that the impact factors do not have a greater effect on the final

Table 6
Score breakup for five risk factors to form the final risk score for baseline model (S_{BM}).

Number	Abbreviation	Main Monomers (Lithner et al., 2011)	Global waste generations (million tonnes year^{-1})	RF1	Mean density (g cm^{-3})	RF2	Status of degradation in the marine environment	RF3	Mean size (μm)	RF4	Hazard score[a]	RF5	Risk score
1	PUR	Toluene di-isocyanate, Polypropylene oxide, Ethylene oxide, and Fluorocarbon ex HCF-134 / Propylene oxide, Sorbitol, diphenylmethane di-isocyanate, and C-pentane	16.00	5	1.31	5	Scarcely degradable	5	26	5	10,614	5	150
2	PVC	Vinyl chloride	15.00	5	1.38	5	Scarcely degradable	5	431	5	8518	4	135
3	PAN	Acrylonitrile	2.04	2	1.18	5	Scarcely degradable	5	484	5	10,581	5	120
4	ABS	Styrene, Acrylonitrile and 1,3-butadiene	4.49	2	1.07	5	Scarcely degradable	5	295	5	6552	4	108
4	PMMA	Methyl methacrylate	1.62	2	1.18	5	Scarcely degradable	5	37	5	1021	4	108
6	SAN	Styrene and Acrylonitrile	0.34	1	1.08	5	Scarcely degradable	5	295	5	2788	4	99
7	TPU	Poly(ethylene butylene adipate) (makes from Adipic acid, Ethylene glycol, Butanediol), Diphenylmethane di-isocyanate and Butanediol	0.19	1	1.23	5	Scarcely degradable	5	26	5	825	3	88
8	UP	1,2-propylene glycol, Maleic anhydride, Phthalic anhydride and Styrene	2.00	2	1.12	5	Scarcely degradable	5	1214	3	1266	4	84
9	PET	Ethylene glycol and Terephthalic acid (Dimethylterphtalate)	32.00	5	1.38	5	Moderately degradable	3	750	5	4	1	78
10	PS	Styrene	17.00	5	1.05	5	Easily degradable	1	308	5	30	2	77
10	HDPE	Ethylene	40.00	5	0.96	3	Moderately degradable	3	750	5	11	2	77
12	HIPS	Ethylene and 1,3-butadiene	4.35	2	1.08	5	Easily degradable	1	295	5	1628	4	72
13	PP	Propylene	55.00	5	0.9	3	Moderately degradable	3	794	5	1	1	66
14	PF	Phenol and Formaldehyde	5.00	2	1.2	5	Scarcely degradable	5	2665	1	1475	4	60
14	PC	Bisphenol A and Phosgene (Diphenyl carbonate)	2.15	2	1.22	5	Moderately degradable	3	1300	3	894	3	60
16	LDPE	Ethylene	31.92	5	0.93	3	Moderately degradable	3	1250	3	11	2	55
17	EP	Bisphenol A and Epichlorohydrin	0.80	1	1.21	5	Scarcely degradable	5	2512	1	5293	4	55
17	MPD-I	Isophthaloyl chloride and m-phenylenediamine	0.18	1	1.38	5	Scarcely degradable	5	2500	1	1187	4	55
17	PTFE	Tetrafluoroethylene	0.10	1	2.2	5	Scarcely degradable	5	931	4	0	1	55
20	PBT	Dimethyl terephthalate and 1,4-butanediol	0.47	1	1.31	5	Moderately degradable	3	750	5	0	1	54
21	POM	Formaldehyde / Trioxymethylene and Ethylene oxide (Dioxolane)	1.70	2	1.41	5	Scarcely degradable	5	2665	1	825	3	48
22	PA 66	Adipic acid and Hexamethylenediamine	0.59	1	1.31	5	Moderately degradable	3	1233	3	63	2	45
22	PA 6	ε-caprolactam	1.17	1	1.14	5	Moderately degradable	3	1233	3	50	2	45
24	LLDPE	Ethylene	25.08	5	0.93	3	Moderately degradable	3	1250	3	10	1	44
24	PPE / PPO	2,6-dimethylphenol	0.35	1	1.06	5	Scarcely degradable	5	2500	1	400	3	44
24	MF	Formaldehyde and Melamine	0.25	1	1.5	5	Moderately degradable	5	2665	1	882	3	44
24	PPS	1,4-dichlorobenzene and Sodium sulphide	0.05	1	1.35	5	Scarcely degradable	5	2500	1	897	3	44
24	UF	Formaldehyde and Urea	1.00	1	1.2	5	Scarcely degradable	5	2665	1	750	3	44
24	PPD-T	Terephthaloyl chloride and p-phenylenediamine	0.10	1	1.44	5	Scarcely degradable	5	2500	1	829	3	44
30	PVAc	Vinyl acetate	0.99	1	1.29	5		1	93	5	1	1	42

(*continued on next page*)

Table 6 (*continued*)

Number	Abbreviation	Main Monomers (Lithner et al., 2011)	Global waste generations (million tonnes year^{-1})	RF1	Mean density (g cm^{-3})	RF2	Status of degradation in the marine environment	RF3	Mean size (μm)	RF4	Hazard score[a]	RF5	Risk score
							Easily degradable						
30	PLA	Lactide	0.06	1	1.24	5	Easily degradable	1	100	5	0	1	42
30	EPS	Styrene and Pentane	2.72	2	0.03	3	Easily degradable	1	295	5	44	2	42
33	PAA	Acrylic acid	0.30	1	1.3	5	Moderately degradable	3	2665	1	230	3	36
33	PA 11/12	11-aminoundecanoic acid / Lauryl lactam	0.20	1	1.04	5	Moderately degradable	3	1233	3	0	1	36
35	EVA	Ethylene and Vinyl acetate	2.60	2	0.94	3	Moderately degradable	3	1250	3	9	1	32
36	PVDF	Vinylidene fluoride	0.02	1	1.78	5	Scarcely degradable	5	2500	1	0	1	22

Note:
[a] Based on monomer toxicity (Lithner, Larsson and Dave, 2011), ultimately due to the potential decay of polymers.

risk scores compared with probability factors. Scenario 2 highlights the potential impact of chemical toxicity on human health as risk factor RF5 is multiplied with the sum of other risk factors (RF1 +RF2 +Rf3 +R4). Therefore, compared to the baseline model, high-impact polystyrene (HIPS) and phenol formaldehyde resins (PF) have relatively higher risk scores. While PET and PS, with potential impacts mainly based on physical and biological toxicity (mean MP particle size), have low chemical toxicity potential (hazard score) and hence were ranked lower. This indicates that MP chemical toxicity has an impact on human health. Scenario 3 shows that if toxicity scores were not considered, the highest-scoring polymers in the baseline model retained on top in scenario 3. Risk rankings of PS, HDPE, PP, PC, HIPS exhibit a considerable variation from the baseline model, indicating that probability factors significantly affect the risk scores of polymers in scenario 3. MP degradation in the marine environment is not accounted for in Scenario 4; hence easily degradable polymers such as PS, HIPS, and HDPE got relatively higher scores than in other scenarios. In contrast, the risk scores corresponding to the scarcely degradable polymers, such as SAN, TPU, and UP, are reduced. Therefore, the degradability of polymers has a significant effect on the final risk score. The monitoring concentration of PP, PS, PET, PVC, LDPE in the marine environment is reflected in the last column of Table 7. However, this ranking is not in line with the baseline scenario because the first plastic polymers produced in the 1950 s would not have been fully degraded (Table 3), and maybe in the future, the polymers which are not easily degradable will take over the percentage share of moderately/easily degradable PP and PS (200 years and 50 years life-span, respectively) fragments in the marine environment.

3.3. Sensitivity analysis

A sensitivity analysis has been conducted, and the Spearman rank-order correlation coefficient values are presented in Fig. 10. The p-Values of all factors are < 0.0001, and all risk factors are positively correlated with the final risk scores as expected from equation Eq. (2). The most sensitive parameter of the S_{BM} is RF5 (coefficient +0.60) which is followed by RF4 (+0.54), RF1 (+0.52), RF3 (+0.32), and RF2 (+0.16). Therefore, the order of influence on the final risk score was monomer toxicity> particle size distribution> global waste generation of MPs> degradation of MPs in the ocean> Mean density of polymers (descending).

3.4. Polymers of concern in marine to human health

Based on the risk ranking of different polymers' MPs through marine water for human health (Fig. 8) in this study and comparing it with scientific corroboration (Table 7), the results are consistent. PUR is one of the most common plastic polymers used in daily life, with an annual global production of 27 million tons. Although PUR has a recovery rate of up to 40.74%, 16 million tons of PUR waste generation (Table 1 and Fig. 3) ends in the global environment every year and contributes significantly to the PUR MP concentration in marine water. Also, due to its higher density (1.31 g/cm^3) (Table 2 and Fig. 4) and resistance to degradation (Table 3 and Fig. 6) in the marine environment, it represents a strong potential exposure source for humans. On the one hand, the mean MP particle size of PUR is only 26 μm (Table 4 and Fig. 7), and it is the most hazardous polymer (scored 10614) based on monomer composition (extensive use of carcinogenic monomers and toxic additives in PUR production) (Fig. A1); therefore, PUR is the most potentially hazardous polymer considering physical, biological, and chemical toxicity for human health. On the other hand, PUR has been reported frequently (identified in 13 papers) (Table 7). Therefore, PUR, with the highest probability of human exposure via the aquatic environment and possible toxicity, is considered a critical MP polymer when considering potential human health effects.

In scientific corroboration, PVC (20 papers) and PMMA (8 papers) were detected in several studies (Table 7) and also have a high potential level of risk to human health for similar reasons as PUR. Meanwhile, PP (66 papers), PS (42 papers), PET (24 papers), LDPE (15 papers), and HDPE (9 papers), which have been reported extensively in numerous studies (Table 7), have a relatively low level of risk to human health because most of them (Table 6) show a relatively high degradation and low density resulting in a lower probability of exposure to the specific MPs, and a weaker potential toxicity impact. While EPS (8 papers) and PVAc (8 papers) were also widely identified (Table 7), they have low-risk levels (Table 6) due to their high degradability (low exposure probability) and low potential toxicity impact. In addition, despite their low waste generation, both of these are easily found in monitoring studies because they are bio-degradable polymers and are readily broken down to micro size in the marine environment (Table 6 and Table 7). However, since the global waste generation of PAN, ABS, SAN, TPU, and UP is less and resistant to breakdown, they accumulate in marine waters and are rarely identified in scientific studies (Table 7). However, their relatively high exposure probability (high density and scarcely degradable) and strong potential toxicity impact pose a high potential risk for human health (Table 6).

3.5. Results of scientific corroboration

Among all 196 experimental studies on MPs (both marine and freshwater, excluding studies on biomass, detailed in Tables A1 and A2) were screened, the main polymers that appeared most frequently in the identification of MP categories (Fig. 11) were PP (25%), PS (18%), PET

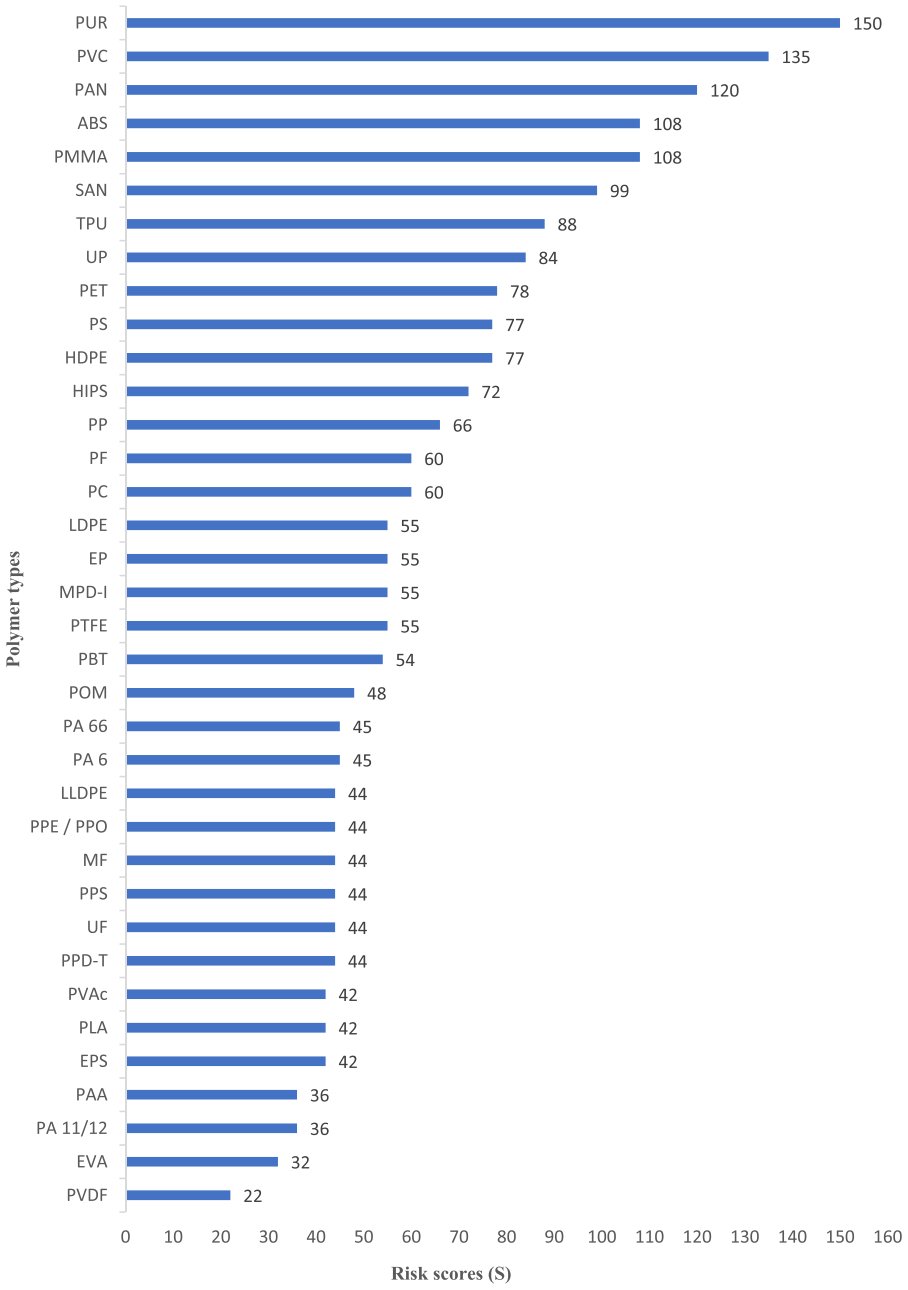

Fig. 8. Simulated risk ranking results of microplastics according to polymer types for baseline model (S_{BM}).

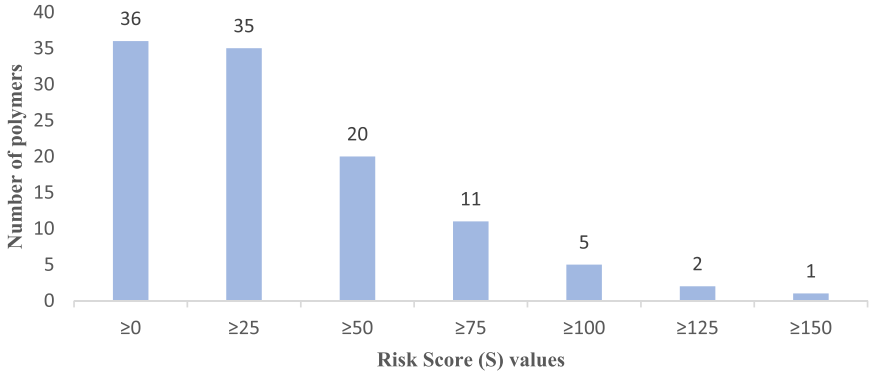

Fig. 9. Cumulative bin distribution of the final risk score for baseline model (S_{BM}).

Table 7
Comparison between risk ranking of 36 polymers in this current research (baseline model and different scenarios) and quantitative validation.

Number	Baseline model		Scenario 1		Scenario 2		Scenario 3		Scenario 4		Scientific corroboration	
	Polymers	Risk score	Polymers	Risk score	Polymers	Risk score	Polymer	Risk score	Polymers	Risk score	Polymers	Sum of appearance
1	PUR	150	PUR	25	PUR	100	PUR	15	PUR	100	PP	131
2	PVC	135	PVC	24	PAN	85	PVC	15	PVC	90	PS	94
3	PAN	120	PAN	22	PVC	80	PET	13	PAN	70	PET	61
4	ABS	108	ABS	21	ABS	68	PAN	12	PS	70	PVC	49
5	PMMA	108	PMMA	21	PMMA	68	ABS	12	ABS	63	LDPE	26
6	SAN	99	SAN	20	SAN	64	PMMA	12	PMMA	63	PUR	21
7	TPU	88	TPU	19	UP	60	UP	12	HIPS	63	HDPE	19
8	UP	84	UP	19	HIPS	52	PF	12	PET	60	PMMA	17
9	PET	78	PET	19	PF	52	POM	12	HDPE	56	EPS	14
10	PS	77	PS	18	TPU	48	SAN	11	SAN	54	PVAc	13
11	HDPE	77	HDPE	18	EP	48	TPU	11	UP	49	ABS	11
12	HIPS	72	HIPS	17	MPD-I	48	EP	11	TPU	48	EVA	10
13	PP	66	PP	17	PC	39	MPD-I	11	PP	48	PC	10
14	PF	60	PF	17	POM	39	PPE / PPO	11	PC	42	PAN	8
15	PC	60	PC	16	PPE / PPO	36	MF	11	LDPE	40	PTFE	6
16	LDPE	55	LDPE	16	MF	36	PPS	11	PBT	36	PA 66	5
17	EP	55	EP	16	PPS	36	UF	11	PVAc	36	EP	5
18	MPD-I	55	MPD-I	16	UF	36	PPD-T	11	PLA	36	LLDPE	4
19	PTFE	55	PTFE	16	PPD-T	36	PS	11	PF	35	SAN	4
20	PBT	54	POM	16	PS	32	HDPE	11	EPS	35	TPU	4
21	POM	48	PBT	15	HDPE	32	LDPE	11	LLDPE	32	PA6	3
22	PA 66	45	LLDPE	15	PAA	30	PP	11	EP	30	PAA	3
23	PA 6	45	PPE / PPO	15	LDPE	28	PTFE	11	MPD-I	30	POM	3
24	LLDPE	44	MF	15	PA 66	24	LLDPE	11	PTFE	30	PBT	2
25	PPE / PPO	44	PPS	15	PA 6	24	PVDF	11	PA 66	30	PLA	2
26	MF	44	UF	15	EPS	22	PC	10	PA 6	30	PA11/12	1
27	PPS	44	PPD-T	15	PET	18	PAA	9	POM	28	UP	1
28	UF	44	PA 66	14	PP	16	PA 66	9	PPE / PPO	24	MF	1
29	PPD-T	44	PA 6	14	PTFE	15	PA 6	9	MF	24	HIPS	1
30	PVAc	42	PVAc	13	PBT	14	PBT	9	PPS	24	PF	0
31	PLA	42	PLA	13	LLDPE	14	PA 11/12	9	UF	24	UF	0
32	EPS	42	EPS	13	PVAc	12	HIPS	8	PPD-T	24	PPE / PPO	0
33	PAA	36	PAA	13	PLA	12	EVA	8	PAA	24	MPD-I	0
34	PA 11/12	36	PA 11/12	13	PA 11/12	12	PVAc	7	PA 11/12	24	PPS	0
35	EVA	32	PVDF	13	PVDF	12	PLA	7	EVA	20	PPD-T	0
36	PVDF	22	EVA	12	EVA	11	EPS	6	PVDF	12	PVDF	0

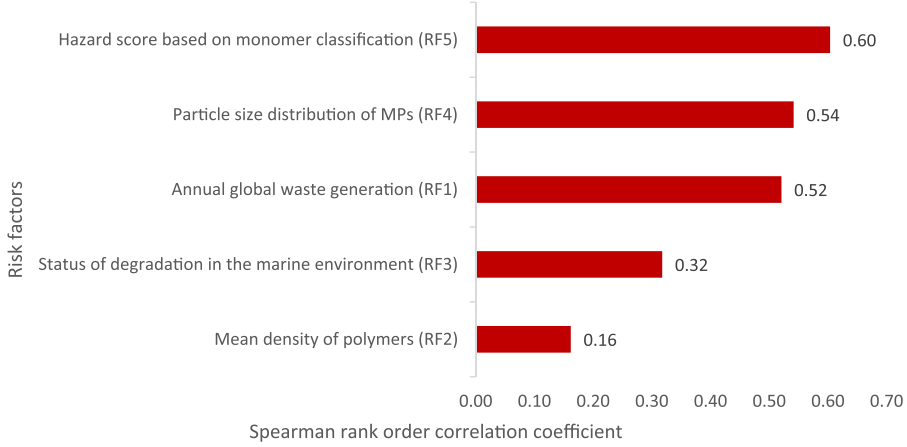

Fig. 10. Sensitivity analysis of five risk factors to final risk score using spearman rank order correlation coefficient.

(12%), PVC (9%), and LDPE (5%). These polymers are the most common sources of MP contamination in oceans, so it is necessary to evaluate their potential risk to human health, focusing on their risk scores in the risk ranking model.

In addition, the statistical output of previous experimental studies (Fig. 12) indicated that sediment and water samples share in almost equally half of all studies (Fig. 12a). Furthermore, nearly three-quarters of studies were concentrated in the marine environment, with 56% of marine studies using water samples in their investigations, while in freshwater, the value is up to 77% (Fig. 12b), which provides good data support for this model. In addition, studies on MPs are documented in several regions (Fig. 12c), including the Asia Pacific, Mediterranean

(a)

(b)

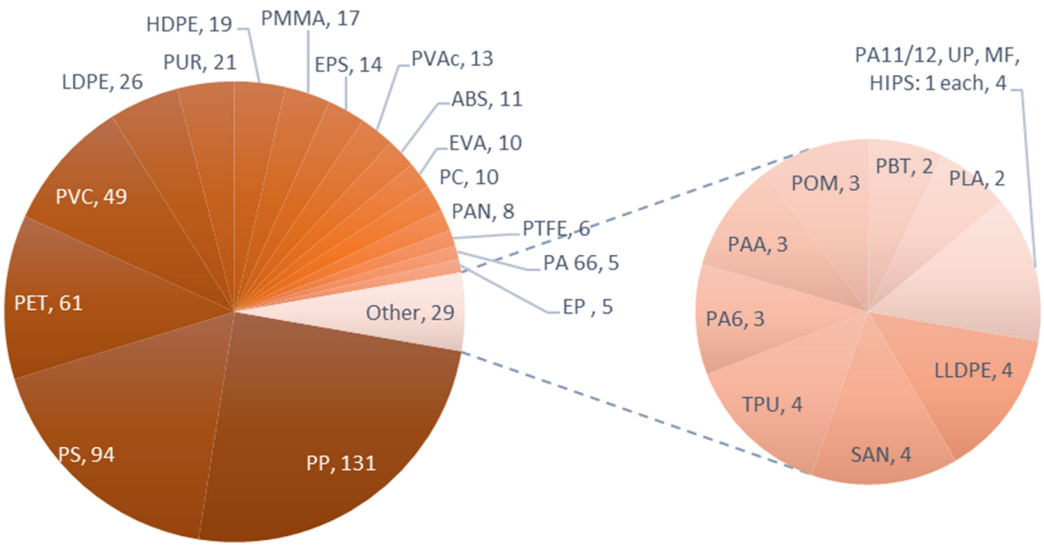

Fig. 11. The statistical output word cloud (a) and pie chart (b) of studies investigating levels of microplastics in the marine environment.

coast, and North America, where China and India have a large number of studies. However, there are still limited studies in Europe.

3.6. Recommendation

In the future, research should focus more on polymers, PUR, PVC, and PMMA, which are the most common and potentially harmful polymers. Regulations should be looked at to reduce their production and use while also restricting illegal dumping and promoting better ways for their recycling and disposal. The focus should be on the use and development of biodegradable plastics (EPS, polylactic acid (PLA), and PVAc) to replace them. Although PP, PS, PET, LDPE, and HDPE have a large production capacity and their risk values are low, these plastics require strict management on their use and recovery to avoid unnecessary contamination. In addition, ABS, SAN, TPU, and UP, though low in yield and easy to degrade, have high potential toxicity. So detailed toxicological experiments should be undertaken on these in future studies, and the production and use of these hazardous polymers should be carefully monitored.

Among the three probability factors, degradability requires further study to estimate more accurate information on the estimated biodegradability of these polymers in marine water. While for the two impact factors, chemical toxicity hazard values have been well ranked in the study by Lithner, Larsson and Dave (2011), however, physical and biological toxicity (mean MP particle size of polymers) remain in need of further data support and studies for a complete hazard ranking.

3.7. Limitations of impact (I) data

3.7.1. Data of mean MP particle size from marine MP researches

In this research, mean particle size data of MPs in marine water were collected from different geographical areas globally, including the Pacific, Atlantic, Indian, and Arctic Oceans. Although the shapes of MPs include fragments, films, fibres, and spheres, the size of MPs is often recorded as average diameter. So, it is assumed in this study that all MPs are spheres. All samples in those studies were taken from marine water using trawls or neuston nets and recovered by sieving or filtration (Adam et al., 2019). However, these processing methods may cause errors, mainly due to the limitation of sampling nets to capture smaller MP particles due to different cut-off sizes, which leads to an overestimation of particle size (Adam et al., 2019). However, such errors were not considered in this study.

3.7.2. Data of hazard score based on monomer classification

Although Lithner et al. (2011) created a unified model for hazard assessment of polymers, this model cannot reflect absolute differences between hazard categories and has not considered exposure routes and exposure levels. Furthermore, CLP does not mention characteristics of persistence, bio-accumulative toxicity, and endocrine disruption (Bergmann et al., 2015). Therefore, the risks of polymers with such

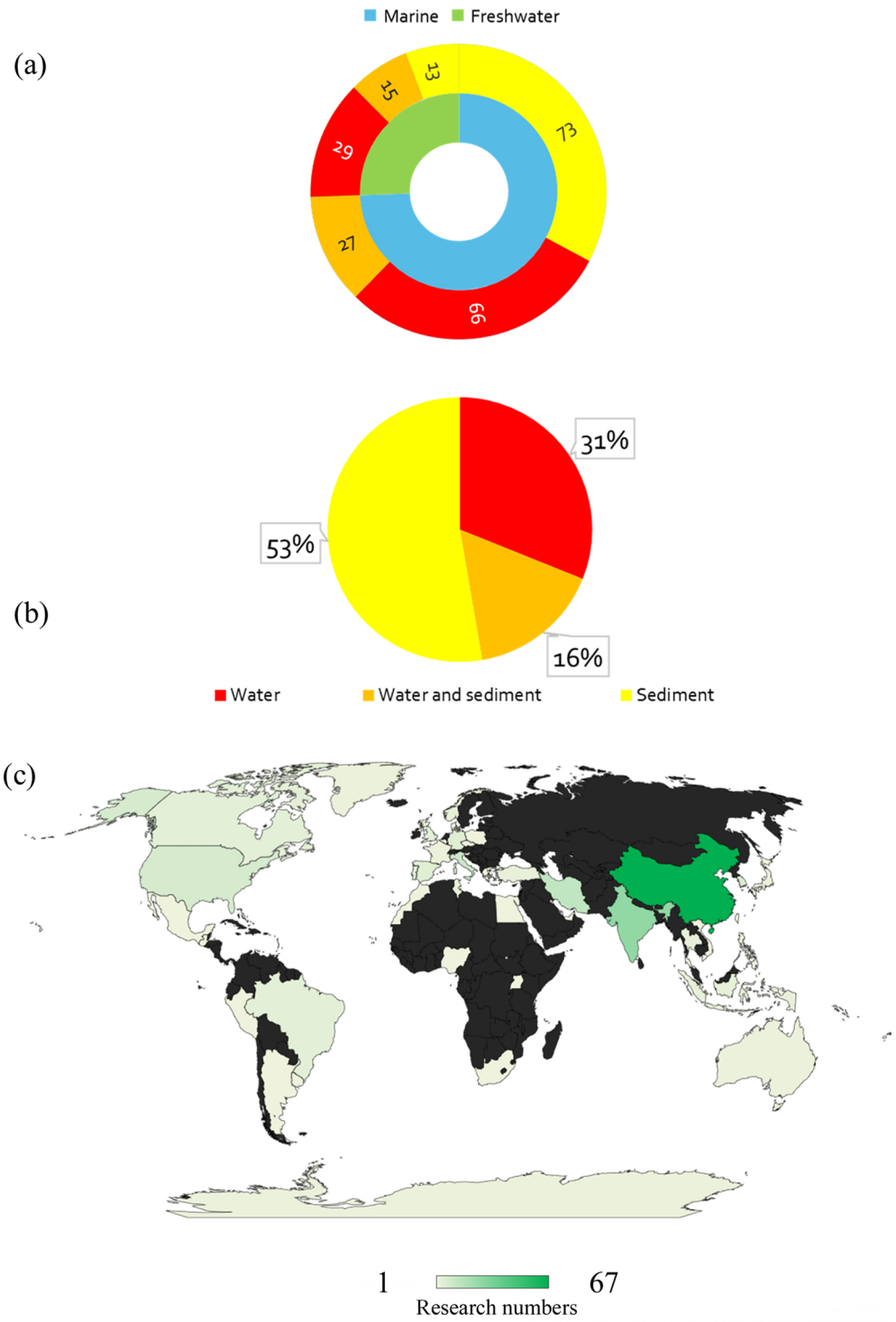

Fig. 12. Global scientific studies on levels of microplastics in the aquatic environment (total 196 studies): (a) Distribution of research in marine and freshwater; (b) Distribution of research in marine by sample types; (c) Distribution of research by regions (minimum 1 and maximum 67, black represents no data).

characteristics may have been underestimated. Furthermore, since information on concentrations (weight percentages) of various compounds required to manufacture polymers is usually commercially sensitive, the average values used in this method will influence the accuracy of RA (Lithner et al., 2011). Also, the hazard ranking only indicates that polymers are composed of hazardous substances; most of them have been transformed during polymerisation (Bergmann et al., 2015). During the product lifecycle from production, use and disposal into waste, hazardous substances or degradation products may be released, but it does not mean that polymers are inherently hazardous (Bergmann et al., 2015). However, it is temporarily impossible to estimate the impact of additives and residual monomers and solvents, as their concentration differs significantly among polymers and data are scarce (Kass and Merten, 2018). However, the model still lacks sufficient substance classification data to cover the extreme diversity of plastic polymers (chemical selection, formulation differences, and polymerisation methods), significantly affecting the model's accuracy (Kass and Merten, 2018).

3.8. Assumptions and future work

- Sediment, food chain, and air exposure routes are not considered in this model.
- Only 36 polymers were considered, while some rubber and novel polymers were not considered.
- Colour and shape, key indicators of microplastic morphology, are not considered risk factors.
- Using waste generation to estimate MP concentrations in marine water assumes that all waste ends up in the ocean and fails to account for quantities of waste that remain in terrestrial and air. Further research needs to be done to gain sufficient concentration data for specific polymer types of microplastics.
- Degradability may not be an accurate measure of the residence time of microplastics and requires more data to predict the lifespan of polymers in the marine environment.
- Data on the mean particle concentrations of specific polymer type microplastics are limited.
- Safety data for chemicals that are endocrine disruptors are not considered in the hazard assessment. However, many polymers are manufactured with monomers with endocrine-disrupting capabilities, which largely limits the ability to assess risk predictions for plastic polymers.

4. Conclusion

This study developed a new risk ranking methodology for MPs exposure from different polymers in marine water based on exposure probability factors (waste generation, density, and degradability) and potential toxicity (physical, biological and chemical) impact factors (particle size and hazard score based on monomer classification). PUR, PVC, PAN, ABS, PMMA, SAN, TPU, UP, PET, PS, and HDPE were found to be the most significant polymers with regards to the potential human health risk from food chain exposure routes influenced by marine waters. The most sensitive parameter of the final risk score of the baseline model is the potential chemical toxicity of polymers (RF5: coefficient +0.60) which is followed by the potential physical and biological toxicity of polymers (RF4: +0.54), annual global waste generation (RF1: +0.52), the status of degradation in the marine environment (RF3: +0.32), and the mean density of polymers (RF2: +0.16). Therefore, the order of influence on the final risk score was RF5 > RF4 > RF1 > RF3 > RF2 (descending). A complete risk assessment of top-ranked MPs based on polymer types may unify field experiment data collected worldwide into transparent analysis and evaluate the potential risk. Also, this risk ranking approach could support related organisations and governments to take necessary actions to monitor the most hazardous polymers or risk factors influencing the final risk and thus help minimise this potential risk.

CRediT authorship contribution statement

Zhihao Yuan: Conceptualization, Methodology, Formal analysis, Software, Data curation, Visualisation, Investigation, Writing – original draft. **Rajat Nag**: Conceptualization, Visualisation, Supervision, Writing – review & editing. **Enda Cummins**: Conceptualization, Resources, Supervision, Project administration, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors gratefully acknowledge the China Scholarship Council (CSC), China and University College Dublin (UCD), Ireland for supporting this study under CSC-UCD Scheme.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2022.128399.

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