DEVELOPMENT OF KNOWLEDGE ON PLASTIC PELLETS POLLUTION RESPONSE

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ABSTRACT

The majority of plastic objects are produced from a raw material called "plastic pellets" or “plastic nurdles”. Plastic pellets are mainly found in the form of granules (1 - 5 mm) but also exist in the form of flakes or powders. In addition to operational losses that can lead to environmental contamination (estimation of between 52,140 – 184,290 tonnes lost to the environment in the European Union in 2019), accidental and significant losses are possible during their transport on land, river and sea. For example, in June 2021, the sinking of the X-Press Pearl released 11,000 tonnes of plastic pellets into Sri Lankan waters. Unlike oils and chemicals for which good practices and guidelines are available to support decision-making in case of an accidental event, guidelines for the management of accidental spills of plastic pellets into the environment are still in development. In this context, Cedre conducted a review of existing knowledge on plastic pellet pollutions and performed analyses, experiments and equipment testing in its facilities to develop additional knowledge. The review demonstrated that pellets are mainly buoyant exhibiting similarities but also differences from oil in terms of behavior and fate and that several recovery techniques exist, some of which have already been tested during past incidents involving pellets. The review also highlighted several knowledge gaps regarding pellet composition, behavior but also concerning response techniques, confirming the need of further experimental studies. Our chemical analyses demonstrated that plastic pellets constitute a complex pollutant potentially containing a large diversity of chemicals depending on their application. In addition, the behavior assays conducted in Cedre facilities showed that external constraints (wind, current, swell) can rapidly disrupt the floating behavior of low-density polymers making it difficult to detect and observe the spilled pellets in water. The work is being continued by testing equipment (at sea and on the coastline) as

past incidents highlighted the challenge of pellets recovery and associated need of fit-for-purpose cleaning techniques.

INTRODUCTION

Since 1950, global production of plastic materials has followed an exponential trend, reaching 489 million tonnes (Mt) in 2022 (compared to 2 Mt in 1950) and using 4-6% of annual petroleum resources (OECD, 2023). The majority of plastic products are produced from a raw material called "plastic pellets" or “plastic nurdles”. Plastic pellets are mainly found in the form of granules (1 - 5 mm) but also exist in the form of flakes or powders.

Nowadays, plastic pellets are ubiquitous in coastal environments. This contamination can be linked to two main sources: (1) Operational losses along the supply chain; e.g. it is estimated that between 52,140 – 184,290 t of pellets are lost each year in the European Union (UE Commission, 2023); (2) Accidental events which can lead to a one-off but important spill of pellets in environments (e.g. road or rail accidents, incident on container ships including losses of containers). For instance, the sinking of the *X-Press Pearl* in 2021 caused the discharge of 11,000 t of pellets in Sri Lankan waters, while the container losses from the *MSC Susanna*, the *Trans Carrier*, and the *MSC Zoe* resulted in the discharge of 49.5 t in the port of Durban in 2017, 13.2 t in the North Sea, and 22.5 t in the North Sea, respectively (Cedre, 2023). The reduction in the emission of plastic pellets in the environment is now a global priority (*e.g.* UE Commission, 2023; OSPAR, 2022; IMO, 2022).

Plastic pellets found in the environment are highly diversified (e.g. polymer nature, chemical composition, colors, weathering state) (e.g. Karlsson et al., 2018; Jiang et al., 2022). However, their characteristics as pollutants (e.g. features, behavior, impacts) are poorly documented as well as recovery methods unlike oils and chemicals for which good practices and guidelines are available to support decision-making in case of an accident.

Based on existing knowledge, the first aim of this study was to highlight the main features of plastic pellets as pollutants and to identify the current gaps. Based on this state of the art and after adapting its facilities to allow testing on real pellets, Cedre conducted experimental assays in 2023 to improve the knowledge on plastic pellets in case of accidental pollution. Furthermore, we conducted assays at different scales (laboratory and pilot-scale) to obtain data on (1) the presence of compounds potentially hazardous (i.e. additives) in pellets, and their capacity to be released in seawater, (2) the behavior of different types of plastic pellets at sea.

METHODS

State-of-Art on plastic pellets

A state-of-art was conducted to identify the main features of plastic pellets as pollutants using the Cedre's documentation center "Alphonse Arzel" and the scientific database available on WebofScience and Google Scholar. In addition, reports from non-governmental organizations and authorities implied during accidental spills of plastic pellets were also identified and used to compile all relevant information on plastic pellets.

Chemical Analysis

Chemical analyses were performed on four types of plastic pellets: 3 batches of pellets of high-density polyethylene (HDPE) supplied by different industrial companies named HDPE-1 (length: 4.8 ± 0.3 mm; width: 2.4 ± 0.3 mm), HDPE-2 (length: 4.3 ± 0.2 mm; width: 2.1 ± 0.2 mm) and HDPE-3 (length: 4.7 ± 0.3 mm; width: 2.5 ± 0.5 mm), and Pellets of polystyrene (PS) supplied by an industrial company named “PS” (length: 3.9 ± 0.3 mm; width: 2.8 ± 0.3 mm). Photos of the pellets are in the Annex 1.

These chemical analyses aimed to (1) characterize the presence and concentration of a selection of compounds known to be found in plastic materials, (2) analyze the capacity of the different samples to release these compounds in seawater.

For the first objective, a liquid-solid extraction with methanol was performed to extract additives and compounds adsorbed on the surface of pellets. Triplicates of 1 gram each were placed in Buchi tubes, and 10 mL of pure methanol were added. The tubes were then covered with aluminum and placed on an orbital shaker (VWR-Ago02) for 24 hours at 200 rpm (rotations per minute). This step was carried out in the dark to prevent certain molecules from undergoing changes due to UV radiation. Samples were stored at -20°C until analyses.

For the second objective, 5 g of each sample were added in 1-liter amber glass bottles previously filled with 500 mL of natural seawater filtered through a 0.2 µm filter. The bottles were then agitated using an orbital shaker at 180 rpm for 36 hours in the dark. Subsequently, the samples were filtered through a 0.2 µm porosity glass fiber filter to remove the pellets. The leachates were stored at -20°C until chemical analyses.

In this first approach aiming at identifying general trends in the capacity of pellets to release their constitutive compounds, targeted-chemical analyses were conducted using a list of 56 compounds known to be present in plastic materials or added during manufacturing processes (PAHs, additives; e.g. Alassali et al., 2020; Rochman et al., 2013; Hermabessiere et al., 2017). All compounds are described in the Annex 2 and Annex 3, including 20 PAHs and 36 additives (divided into 6 groups according to their uses: “plasticizers”, “flame retardants”, “plasticizers and antioxidants”, “plasticizers and flame retardants”, “antioxidants”, and “UV stabilizers”). Quantification analyses were conducted by Gas chromatography–mass spectrometry (GC/MS) (Gardon et al., 2020).

Behavioral study

Pilot-scale tests ensure the acquisition of representative data on the behavior and weathering of pollutants following their release in the natural environment. The tests were conducted in Cedre's hydraulic channel (the Polludrome®), an experimental tool able to recreate natural phenomena such as the wind, current and swell in order to study the behavior of pollutants in conditions close to those found in the natural environment (internal length = 16.4 m; external length = 20.2 m; height = 1.4 m; width = 0.6 m).

Three batches of pellets supplied by different industrial companies, were studied during the tests: (1) HDPE pellets (HDPE-1 of the part “Chemical Analyses”, (2) PP (polypropylene) pellets (length: 4.4 ± 0.2 mm; width: 3.3 ± 0.2 mm), (3) PS pellets (PS of the part “Chemical Analyses”). During the tests, the different batches were subjected to three types of environmental constraints with three levels of intensity:

- Wind: (1) 2.5 knots, (2) 7 knots, and (3) 10 knots.
- Current: (1) 0.4 knots, (2) 0.7 knots, and (3) 0.9 knots.
- Swell: (1) Sea State 0 (calm sea without ripples and waves), (2) Sea State 1 (calm sea with ripples/waves with a maximum height of 10 cm), and (3) Sea State 2 (sea with waves from 10 to 50 cm).

During the tests, individual measurements were taken on four different pellets per batch (three measurement replicates per pellet) to assess the impact of different constraints and intensities on the speed (m/s) of the pellets on the water surface. In addition, complementary tests (n= 3 per pellets batch) were conducted on 10 g of pellets simultaneously placed in the Polludrome® to obtain qualitative information on the formation of a pellet slick or the presence of a heterogeneous distribution of pellets on the water surface.

RESULTS/DISCUSSION

State-of-Art

Main characteristics of pellets as pollutants

The literature analysis showed that plastic pellets as pollutants, are characterized by distinct physico-chemical properties from oils, despite some similarities, as pellets are solid, persistent (no rapid biodegradation; conventional polymers can resist for several decades), non-soluble, non-emulsifiable, non-dispersible, non-evaporating, non-viscous, and non-adherent.

Plastic pellets produced nowadays predominantly float (80% of the worldwide production = polyethylene + polypropylene; these two polymers having density lower than 1). Therefore, their behavior and fate are theoretically highly affected by currents, waves, tides but also by winds and rain, which could disseminate spilled pellets over large spatial scales. Pellets stranded or deposited in the environment can quickly be remobilized, displaced, or buried in soils if cleanup operations are not promptly implemented after the incident. Further experimental studies are needed to acquire numerical data and to better understand the real behavior of plastic pellets in aquatic environment to help modelers to predict trajectories of pellets following a spill.

Regarding environmental impacts of plastic pellets, they are still not well understood. To date, studies conducted after accidents (*MSC Susanna, Trans Carrier)* did not allow to evidence acute effects of spilled pellets on the studied animals in both cases (e.g. birds, fish). However, plastic pellets are persistent products contributing to global plastic pollution generating pressure on biodiversity and ecosystems. Experimental studies suggest potential effects at different levels including: (i) physical damages on habitats (*e.g.* effect on microbial activity, water retention, sediment temperature, smothering); (ii) accidental ingestion by animals (e.g., fish, birds) with potential physical disturbances leading to consequences at different levels of organization (e.g. cellular, tissue, individual, population); (iii) releases of smaller particles and leaching of chemicals in the environment or animals after ingestion; (iii) Transport of species including pathogenic or invasive species as pellets can be colonized by organisms (Cedre, 2023). Further studies are needed to better understand the real risk of plastic pellets during important spills in order to help stakeholders with robust risk assessment. In this context, there is an ethical and aesthetic obligation to clean-up as reasonably as possible, taking also into consideration the potential impact of clean-up (Foekema et al., 2021).

Main methods of recovery

Currently, to our knowledge, no response operations (aerial reconnaissance, deployment of containment and recovery equipment) directly at sea have been conducted during past accidents. Therefore, additional studies are needed to better prepare for future accidents.

By contrast, clean-up methods of plastic pellets on soil and coastline are better documented than operations at sea. To date, different methods have used to recover plastic pellets on the shoreline and separate them from the sand. Recovery methods include manual and mechanical recovery along with vacuuming. Separation methods include manual or mechanical cleaning along with sink-float separation.

The analysis of lessons learned from past accidents (e.g. *X-Press Pearl*, *Rena, MSC Susanna, Trancura, Trans Carrier*) demonstrated that shoreline clean-up of pellets can be particularly labour intensive, time consuming and tiresome and it appears impossible to recover all spilled pellets. More work is needed to improve existing clean-up methods or develop new ones.

Chemical Analysis

The quantity of targeted PAHs in the extracts (in ng/10 g of plastic pellets) of new pellets are similar among the four samples, without difference between HDPE and PS (values ranging from 100.5 ± 57.3 ng/10g of pellets to 314.1 ± 150.4 ng/10g of pellets; Figure 1). Overall, for the three HDPE samples, PAHs are largely dominated by 5 compounds (>80% of all PAHs): Fluorene, Fluoranthene, Pyrene, Phenanthrene and Dibenz[a,h]anthracene. However, specific differences were observed among the HDPE samples, for instance fluorene and phenanthrene were not detected in HDPE-1. These few differences are naturally linked to the different chemical feedstocks used for the pellet production. The analysis showed more important differences in PAHs composition according to the considered polymer. PAHs of the PS sample are dominated by 7 compounds (>90% of all PAHs) including PAHs detected in HDPE samples (Fluorene, Pyrene, Phenanthrene and Dibenz[a,h]anthracene; 65% of PAHs detected in the PS sample) and undetected in HDPE samples (Benzo[a]pyrene, Indeno[123-cd]pyrene and Benzo[ghi]perylene). These results suggest a low PAHs contamination in plastic pellets considering the targeted compounds by the analyses.

This hypothesis was checked by the analysis of leachates which highlighted the low ability to the tested pellets to leach rapidly their PAHs in seawater (ranging from 0 ng/L for the HDPE-3 to 14 ng/L for the HDPE-2). Considering all samples, two PAHs are mainly found in the leachates (>50% of all PAHs): Fluorene and Phenanthrene. In addition, in the HDPE-2 leachates, the PAHs composition was dominated by the Naphthalene (constituting 78% of the total PAHs composition) whereas this PAH was not detected in the extract samples. This fact could be explained by the different sensitivity of the methods used for extracts and leachates analyses.

The quantity of targeted additives in methanol extracts depends on the considered sample as highlighted by a 80-fold difference between the sample HDPE-1 (10 ng/10g of pellets) and HDPE-2 (828 ng/10g of pellets) (Figure 2). However, despite the variability in concentration, we found the same major additives in HDPE-2, HDPE-3 and PS: 4tOP (plasticizers and antioxidants), NP1OE (plasticizers and antioxidants), NP2OE (plasticizers and antioxidants) and ATBC (plasticizers and flame retardants). These 4 additives constitute more than 75% of the additives detected in extracts during the assays. In HDPE-1, only two additives were detected in the extracts (4tOP and BDE47 which is a flame retardant).

Thus, these results could suggest a low number of additives in the virgin pellets used for the study. This could be explained by the fact that the analyzed pellets were provided by initial producers and not converters which can modify the initial pellets by adding new additives to obtain new features (e.g. color, heat resistance) for the final plastic product. Therefore, the targeted analysis allowed the detection of few additives in virgin plastic pellets and leachates at low concentrations which could mean a low chemical risk. However, this hypothesis cannot be validated by our assays as we performed targeted chemical analysis although the compounds targeted are common in plastic materials. Indeed, more than 10,000 compounds are used in plastic industry, including more than 2,000 compounds considered as “of potential concern” (Wiesinger et al., 2021). Therefore, to confirm our hypothesis of a low number of additives leached by plastic pellets from initial producers, further investigations are needed using high-throughput nontargeted chemical analyses. In addition, toxicological studies will help to identify if pellets have the capacity to leach compounds at level inducing toxicity on different taxa, these studies allowing also to take into account potential “cocktail effects”.

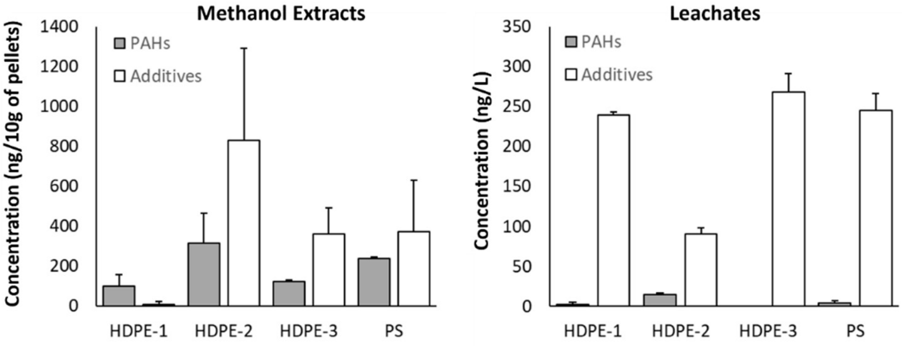

Figure 1. Concentration of additives and PAHs in methanol extracts (in ng/10 g of pellets) or leachates (in ng/L) of HDPE-1, HDPE-2, HDPE-3 and PS pellets.

Behavioral Analysis

Modelling of pellets trajectories following a spill is important to assist in the deployment of operational responses. To date, in case of pellet spills, models developed initially for oils and chemicals have been used but with uncertainty of the appropriate parametrization for pellets. Consequently, to help modelers to calibrate models for pellets, experiments are needed to obtain numerical data.

In the present study, we considered the three environmental constraints identified as important for the behavior of plastic pellets in aquatic environment: the wind, the swell, and the current. The results of the tests conducted in the Polludrome® are available in Table 1. Overall, the three environmental constraints had an impact on the behavior/movement of the pellets, the gradual increase of each parameter logically increase the horizontal speed of pellets.

Considering the horizontal movement of floating pellets, the gradual increase in the sub-surface current in the Polludrome© had the highest influence with average speeds of 0.4 ± 0.05, 0.5 ± 0.07 and 0.5 ± 0.05 m/s for HPDE, PP and PS, respectively, while the wind and swell allowed at the highest intensity level average speeds of 0.05 ± 0.02, 0.06 ± 0.03, 0.05 ± 0.02 m/s for HPDE, PP and PS.

Interestingly, assays with 10 g of each type of pellets showed formation of a slick at the seawater surface in static conditions while the presence of wind at all intensity levels caused pellet dissemination in the Polludrome©. This dissemination can be exacerbated by the variability in the morphological features of pellets from the same batch. Image analysis showed a length variability of 4-8% and a width variability of 7-20% according to the sample. These variabilities influence therefore the wind surface area of each pellet, gradually creating variations in speed between the different granules of the original slick, leading to the pellet dissemination in the Polludrome©. Regarding the currents and the swell, same observations were observed but in addition to an horizontal dissemination, we observed also a vertical dissemination along the water column (Polludrome© height: 80 cm). The floating balance was altered at the third intensity level of current for HDPE and PP pellets, causing the pellets to move into the water column. This balance was altered as early as the second intensity level for PS pellets. This difference from HDPE and PP pellets is explained by the higher density of polystyrene (~1.03 vs. 0.95 and 0.92 for polyethylene and polypropylene, respectively), facilitating movement into the water column and thus requiring less turbulent energy to break the flotation balance. Unlike the current, water surface agitation only altered the floating balance for PS pellets, and this occurred as early as the first intensity level. Once in the water column, we observed chaotic movement of pellets without any particular pattern, pellets moved over the entire water height (80 cm) without returning to their initial position at the surface. These experimental observations suggest that in the case of spills in open seas or in an environment with turbulence, aerial observation of plastic pellets on the water surface to assist the implementation of operational response (e.g deployment of confinement and recovery equipment at sea) would be difficult. This hypothesis should be tested in natural environment using pellet simulants.

These observations demonstrate that the buoyancy of 'floating' plastic pellets is not immutable but depends primarily on environmental constraints. All compiled information suggest that the current is the parameter with the highest influence on the pellet behavior, whether on horizontal velocity or vertical displacement of pellets in the water column. These numerical data and observation could be used by modelers to calibrate new tools adapted to better predict pellet trajectories in case of a spill. From an operational perspective, the observed dissemination of an original spill of pellets due to the environmental constraints in the Polludrome© could explain the high capacity of a “small amount” of pellets to easily contaminate several hundred kilometers of the coastline (Kystverket and Norwegian Coastal Administration, 2020).

Table 1. Speed of HDPE, PP, and PS pellets as a function of three environmental constraints (wind, current, swell). Results are expressed as mean ± standard deviation (n=4). Colored box indicates alteration of the pellet floating balance (*i.e.* transfer of plastic pellet in the water column).

Environmental factor		**HDPE**	**PP**	**PS**
Wind	2.5 knots	0.03 ± 0.01	0.04 ± 0.01	0.02 ± 0.01
	7 knots	0.06 ± 0.01	0.05 ± 0.01	0.03 ± 0.01
	10 knots	0.08 ± 0.01	0.09 ± 0.01	0.06 ± 0.01
Current	0.4 knots	0.03 ± 0.01	0.03 ± 0.01	0.04 ± 0.01
	0.7 knots	0.3 ± 0.01	0.2 ± 0.02	0.3 ± 0.01
	0.9 knots	0.4 ± 0.05	0.5 ± 0.07	0.5 ± 0.05
Swell	Sea State 0	0.01 ± 0.01	0.03 ± 0.01	0.02 ± 0.01
	Sea State 1	0.03 ± 0.01	0.04 ± 0.01	0.04 ± 0.01
	Sea State 2	0.1 ± 0.01	0.05 ± 0.01	/

CONCLUSIONS

The majority of plastic objects are produced from a raw material called "plastic pellets" or “plastic nurdles”. Accidental events can cause important spill of plastic pellets in aquatic environment, requiring rapid response operations to limit the spread of the contamination and the recovery of spilled pellets (e.g. at sea, along the coastline, on soils). Nevertheless, plastic pellets, as pollutants, are not well understood in terms of behavior, environmental risks, and the type of necessary response operations. The present study aimed to bring new data on important endpoints: (1) the chemical risk of plastic pellets, (2) the behavior at sea. The different assays showed plastic pellets contain a diversity of chemicals (including hazardous compounds) depending on the polymer type and the batch. Although, the tested pellets were able to leach their compounds in seawater, the level is low suggesting a limited risk regarding the targeted compounds by the analysis corresponding to common PAHs and additives found in plastic products. Regarding the acquisition of numerical data to help the calibration of models in order to improve the drift simulations of pellets following a spill, results showed that the three tested constraints (wind, current, swell) prevent a pellet slick formation leading to a rapid dissemination of pellets whether on the water surface or in the water column when the level of currents and swell are sufficiently high to break the floating equilibrium of plastic pellets. Consequently, these experimental results suggest that aerial observation of plastic pellets on the water surface of open seas would be difficult.

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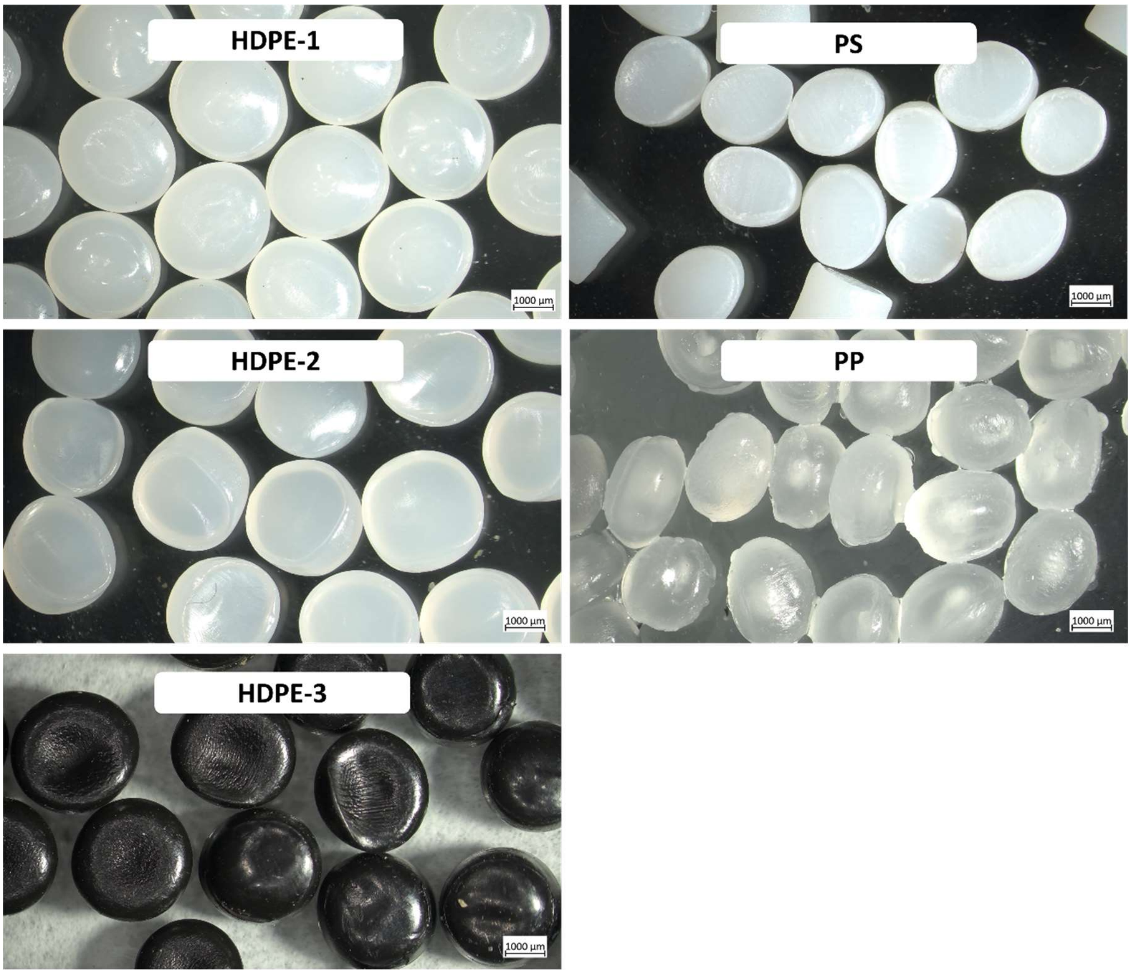
SUPPLEMENTARY INFORMATION

Annex 1 Photos of plastic pellets used in the study



Annex 2 List of PAHs targeted by the chemical analyses

PAHs	Abbreviations
naphthalene	N
1-methylnaphthalene	1-MN
2-methylnaphthalene	2-MN
benzothiophene	BTBPE
biphenyl	B
acenaphtylene	ANY
acenaphtene	ANA
fluorene	F
Dibenzothiophene	DBT
Phenanthrene	P
anthracene	A
fluoranthene	FL
pyrene	PY
2-methylfluoranthene	2-MFL
Benz[a]anthracene	BaA
chrysene	C
benzobfluoranthene	B(b)FL
benzokfluoranthene	B(k)FL
benzoepyrene	B(e)PY
benzoapyrene	B(a)PY
perylene	PE
indeno123cdpyrene	IN
dibenzoahanthracene	DBA
benzo(ghi)perylene	BPE

Annex 3 List of additives targeted by the chemical analyses

Additives	Abbreviations	Families
Tripropyl phosphate	TPrP	Flame retardant + plasticizers
Butylated hydroxy toluene	BHT	Antioxydant
4-tert-octylphenol	4-tOP	Plasticizer + antioxydant
4-octylphenol	4-OP	Plasticizer + antioxydant
Mix nonylphenols	NPs mix	Plasticizer + antioxidant
4-nonylphenol	4-NP	Plasticizer + antioxidant
Nonylphenol mono-ethoxylate	NP1OE	Plasticizer + antioxidant
Nonylphenol diethoxylate	NP2OE	Plasticizer + antioxidant
Tributyl Acetyl Citrate	ATBC	Flame retardant + plasticizer
Tris(1,3-Dichloro-2-propyl)phosphate	TDCPP	Flame retardant + plasticizer
Dibutyl phtalate	DBP	Plasticizer
Di-allyl phtalate	DAIP	Plasticizer
Diethyl phthalate	DEP	Plasticizer
Bis(2-ethylhexyl) adipate	DEHA	Plasticizer
Tri(2-ethylhexyl) phosphate	TEHPA	Plasticizer
Bis(2-ethylhexyl) phthalate	DEHP	Plasticizer
Dimethyl phtalate	DMP	Plasticizer
Di-n-hexylphtalate	DHP	Plasticizer
Benzyl butyl phtalate	BBP	Plasticizer
Dioctyl phtalate	DOA	Plasticizer
Diisoheptyl phtalate	DIHP	Plasticizer
Dicyclohexyl phtalate	DCHP	Plasticizer
Trinuvin 326	UV 326	UV Stabilizer
Trinuvin 328	UV 328	UV Stabilizer
Trinuvin 327	UV 327	UV Stabilizer
Uvinul 3008	Uvinul 3008	UV Stabilizer
2,4,4'-tribromodiphenyl ether	BDE 28	Flame retardant
Tricresyl phosphate	TCP	Flame retardant
Tricresyl phosphate	TCrP	Flame retardant
Tri-o-tolyl phosphate	TToP	Flame retardant
2,2',4,4',6-pentabromodiphenyl ether	BDE 100	Flame retardant
2,2',4,4'-tetrabromodiphenyl ether	BDE 47	Flame retardant
2,2',4,4',5-pentabromodiphenyl ether	BDE 99	Flame retardant
2,2',4,4',5,6'-hexabromodiphenyl ether	BDE 154	Flame retardant
2,2',4,4',5,5'-hexabromodiphenyl ether	BDE 153	Flame retardant
2,2',3,4',5,6'-heptabromodiphenyl ether	BDE 183	Flame retardant