PLASTIC PELLETS FOUND ON BEACHES ALL OVER THE WORLD CONTAIN TOXIC CHEMICALS

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International Pellet Watch

IPEN
for a toxics-free future

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IPEN is a network of over 600 non-governmental organizations working in more than 120 countries to reduce and eliminate the harm to human health and the environment from toxic chemicals. IPEN´s campaign on Toxic Chemicals in Plastics seeks to eliminate harm from chemicals in plastics when plastics are produced, used, recycled, and discarded.

ipen.org

International Pellet Watch is a nonprofit ecotoxicological research group whose mission is to monitor the occurrences of persistent organic pollutants (POPs), plastic waste, and plastic pellets, around the world. Based at Tokyo University of Agriculture and Technology, Laboratory of Organic Geochemistry in Tokyo, Japan, the group has been gathering data and educating the public about the hazards of plastic waste since 2005.

www.pelletwatch.org

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Front cover: Collecting pellet samples in Costa Rica. Photo: RAPAL

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ABSTRACT

Preproduction plastics in the size of lentils, known as plastic pellets, or nurdles, are used to make plastic products but are often lost during production, transportation, and storage. These pellets have been found on beaches all over the world since the 1970s. They can carry many different chemicals, both those that have been purposely added to the plastics and pollutants that attach (sorb) to the plastics in the environment. Some of these chemicals are especially concerning since they are known to have several negative effects on human health and the environment. In this study, plastic pellets from 23 different countries were analyzed for thirteen different types of polychlorinated biphenyls (PCBs) and ten benzotriazole UV stabilizers (BUVs). PCBs have been banned since the mid-1990s as part of the dirty dozen but are still frequently found in the environment. BUVs are frequently added to plastic products, but are known to leach out and to have endocrine-disrupting effects. All sampled PCBs and BUVs, including UV-328, were found in samples from all locations in the study. The concentrations were especially high in the samples from African countries, which illustrates how African countries often bear a heavy burden of plastic pollution, even though they are not major producers of neither chemicals nor plastics. The results from this study clearly show that beached plastics are not only bringing the physical pollution of plastic littering, but also chemical pollutants in the form of added and sorbed chemicals. Furthermore, it illustrates that plastics can play a very important role in the long-range transport of toxic chemicals.

KEY MESSAGES

- Plastics from all sampled locations contained all ten analyzed benzotriazole UV stabilizers, including UV-328
- Plastics from all sampled locations also contained all thirteen analyzed polychlorinated biphenyls
- The concentrations were especially high in African countries, even though they are not major producers of chemicals nor plastics
- The results show that with plastic pollution there is also chemical pollution
- The results also illustrate that plastics can play a very important role in the long-range transport of toxic chemicals

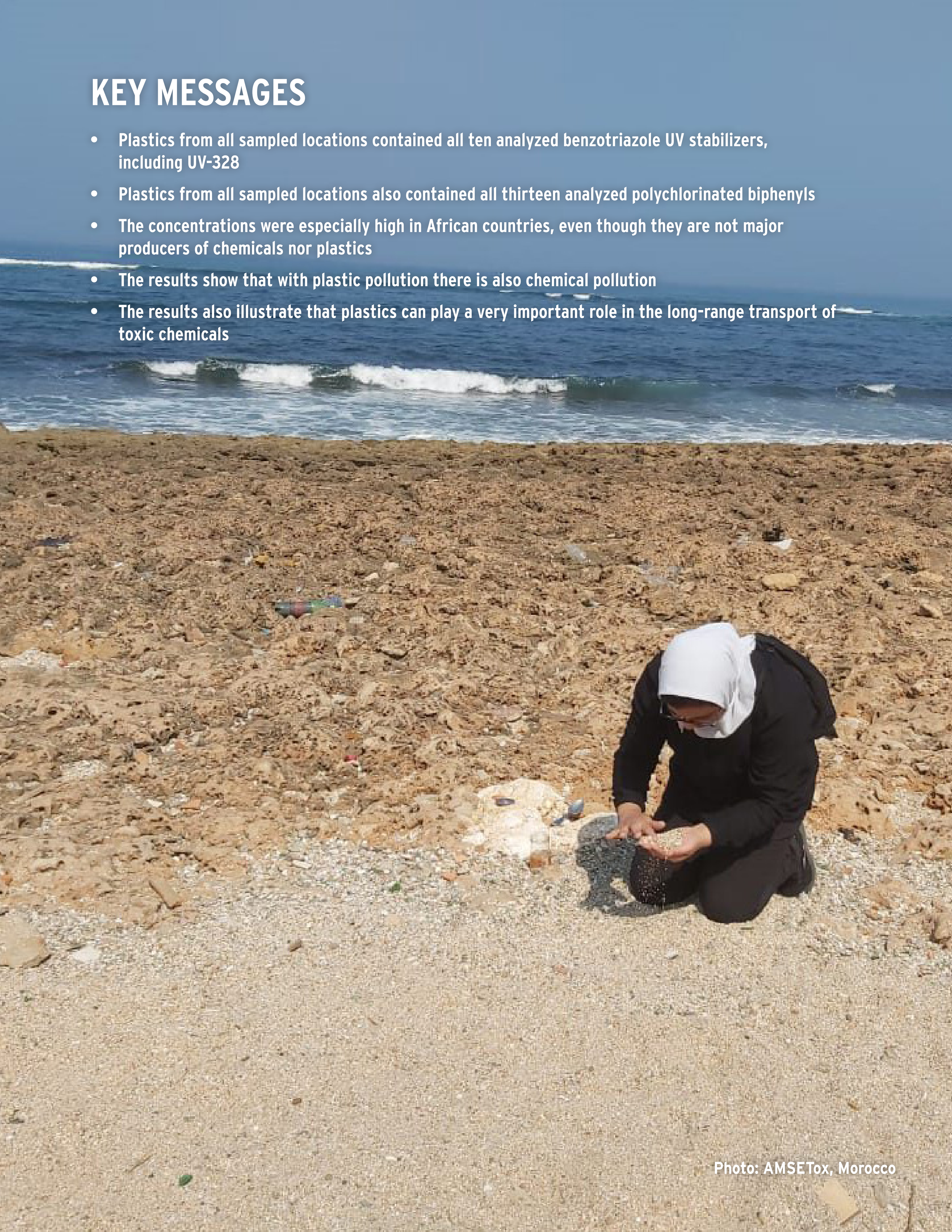
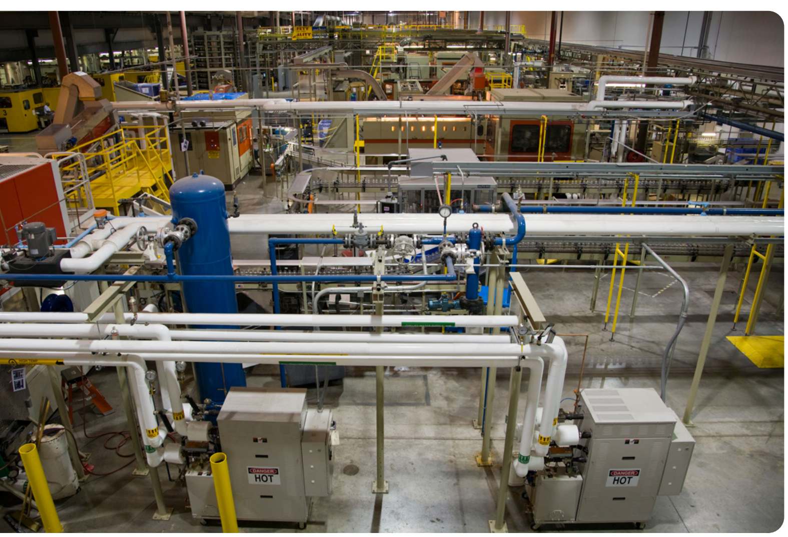


Photo: AMSETox, Morocco

BACKGROUND

The issue of plastic pellets that are discharged or spilled into the environment during production or transportation have been known for more than 50 years. Plastic pellets are small particles, 2-5 mm, of different colors. They can be made from different plastic polymers and are used to make plastic products. The pellets that are found on beaches are plastics that have been spilled into the environment, even before they became plastic products.

These pellets have been found on beaches and in open waters, all over the world, since the early 1970s (Carpenter *et al.* 1972). The researchers warned, already at that point, that the increased production of plastics, coupled with unsuitable waste management practices, would lead to an increase in plastic particles in the ocean. They also concluded that plastic particles could serve as a source of polychlorinated biphenyls in oceanic organisms, thereby flagging the potential for plastic particles to spread toxic chemicals.



Plastic pellets end up in the environment due to spills during production, transport, and storage. They can be spilled on land where rain and runoff can carry them to rivers and coastal areas, or they can be spilled directly into the water due to shipping accidents. Once in the ocean, floating pellets can travel far with the currents, and with them they bring a wide range of chemicals. These chemicals are either added to the pellets during production or consist of environmental contaminants such as PCBs that are sorbed (attached) to the pellets in the ocean.

CHEMICALS ADDED DURING PRODUCTION

The substances used in plastics include the building blocks, i.e., plastic polymers and monomers, as well as additives. Additives are various chemicals that are used to give the plastics specific colors and properties and to protect against UV radiation that causes the plastic to degrade. Typically, additives are not bound to the actual polymer, meaning that they can leach out from the plastics during the production, use, recycle, and disposal phases of plastics.

A recent systematic review found that over 10,000 chemicals are used in plastics (Wiesinger *et al.* 2021). Of those, 2,486 substances were identified to be of potential concern, but the results of the review suggest that less than half (47%) of those substances are subject to any risk management measures (Wiesinger *et al.* 2021).

The chemicals that travel with the plastics can leach into the environment and studies have confirmed the presence of chemicals typically associated with plastics in beach sand (Kwon *et al.* 2015, Kwon *et al.* 2020), in seabirds (e.g., Tanaka *et al.* 2019, Tanaka *et al.* 2020, Yamashita *et al.* 2021), and in several species of fish (e.g., Lu *et al.* 2016, Lu *et al.* 2019, Salvaggio *et al.* 2019, Peng *et al.* 2021).

Several of the chemicals that leach from plastics have been shown to have toxic effects (Zimmerman *et al.* 2021). Ultimately, this means that any kind of plastics can carry a large set of potentially harmful chemicals with them, of which many remain unregulated.

Plastic pellets are small plastic particles, approximately 2-5 mm, (about the same size as a lentil).

The pellets can also be called nurdles and are sometimes referred to as mermaid tears.

Photo: CGFED, Vietnam

UV stabilizers are amongst the most common plastic additives. According to Wiesinger and colleagues (2021), 762 “light stabilizers” are used in plastic products. One commonly used group of light stabilizers is benzotriazole ultraviolet stabilizers (BUVs) which are, aside from in plastics, also used in several other applications such as sunscreens, cosmetics, and coatings.

Like other plastic additives, BUVs are frequently found in the environment, for example in seafood (Kim *et al.* 2011), river sediment (Kameda *et al.* 2011), marine sediment (Apel *et al.* 2018) and wastewater (Lu *et al.* 2017). They are also found in beached plastics, i.e., plastic debris that has washed up on beaches (Rani *et al.* 2017; Tanaka *et al.* 2020). Moreover, some studies suggest that BUVs are persistent in the environment (Ruan *et al.* 2012). As BUVs typically are hydrophobic, thus having a higher affinity for lipids than water, they also have the potential to bioaccumulate in the lipids of organisms and studies on different trophic levels in the marine food chain confirm that BUVs are persistent and bioaccumulating (Nakata *et al.* 2009). Their hydrophobicity also gives them an affinity for sorbing to plastic litter in the environment, wherefore measured concentrations of BUVs in marine litter is likely to be a combination of additives and sorbed BUVs.

Plastic particles are especially frequent in seabirds, in fact, up until 2020, 180 species of seabirds had been reported to have ingested plastics (Kuhn and Franeker, 2020). This ingestion of plastic particles can in some cases lead to physical damage and death through obstruction of the gastrointestinal tract and subsequent starvation (Pierce *et al.* 2004, Roman *et al.* 2019b), as well as a higher incidence of cysts (Roman *et al.* 2019c). The ingested pellets can also provide an exposure route for chemicals used in the plastics into the birds and plastic additives, such as BUVs, are often found in seabirds (Tanaka *et al.* 2019, Tanaka *et al.* 2020b, Yamashita *et al.* 2021). Recent research shows that, although the birds can be exposed to BUVs from their feed, leaching from ingested plastic particles is also a significant exposure pathway (Tanaka *et al.* 2020b, Yamashita *et al.* 2021).

TABLE 1. SUMMARY OF INFORMATION REGARDING SAMPLED BUVs AS REPORTED IN THE EUROPEAN CHEMICALS AGENCY (ECHA) DATABASE

	Name in database	Used in	Toxicity	PBT	SVHC	POP
UV-P	2-(2H-benzotriazol-2-yl)-p-cresol	Coating products, sealants, plastic-based materials	Very toxic to aquatic life with long-lasting effects. May cause allergic skin reactions.	Under assessment	-	-
UV-PS	2-(2H-benzotriazol-2-yl)-4-tert-butylphenol	No public registered data	Unknown	-	-	-
UV-9	Oxybenzone	Cosmetics and personal care products, coating products, fillers, putties, modelling clay, finger paint	Very toxic to aquatic life with long-lasting effects	Under assessment	-	-
UV-234	2-(2H-benzotriazol-2-yl)-4,6-bis(1-methyl-1-phenylethyl) phenol	Coating products	Unknown	Under assessment	-	-
UV-320	2-benzotriazol-2-yl-4,6-di-tert-butylphenol	Plastics such as PVC and polyester	Harmful to aquatic life with long-lasting effects. May cause damage to organs through prolonged or repeated exposure, suspected of causing cancer.	Yes	Yes	-
UV-326	Bumetrizole	Coating products, adhesives, sealants, washing and cleaning products	Unknown	Under assessment	-	-
UV-327	2,4-di-tert-butyl-6-(5-chloro-benzotriazol-2-yl)phenol	Coatings, plastics, rubber and polyurethanes, cosmetics, food contact material, adhesives [1]	Harmful to aquatic life with long-lasting effects. Causes skin, eye, and respiratory irritation.	Under assessment	-	-
UV-328	2-(2H-benzotriazol-2-yl)-4,6-ditertpentylphenol	Coating products, air-care products, adhesives and sealants, lubricants and greases, polishes and waxes, and washing and cleaning products	May cause long-lasting harmful effects to aquatic life. May cause damage to organs through prolonged or repeated exposure.	Yes	Yes	Under assessment
UV-329	2-(2H-benzotriazol-2-yl)-4-(1,1,3,3-tetramethylbutyl) phenol	Air-care products, coating products, adhesives and sealants, lubricants and greases, polishes and waxes, and washing and cleaning products	Unknown	Under assessment	-	-
UV-350	2-(2H-benzotriazol-2-yl)-4-(tert-butyl)-6-(sec-butyl) phenol	Coatings, plastics, rubber and polyurethanes, cosmetics, food contact material, adhesives [1]	May cause long-lasting harmful effects to aquatic life. May cause damage to organs through prolonged or repeated exposure	Under assessment	Yes	-

PBT=Persistent, bioaccumulative, and toxic
SVHC= Substance of very high concern
POP= persistent organic pollutant
[1] Data on usage not reported in the listed database but detailed in ECHA 2020. Database: echa.europa.eu

BUVs have even been found in humans, for example in breast milk (Kim *et al.* 2019), human urine (Asimakopolous *et al.* 2013), and fat tissue (Wang *et al.* 2015), and several BUVs possess endocrine-disrupting potential (Sakuragi *et al.* 2021). Moreover, several BUVs are under assessment as being persistent, bioaccumulating, and toxic (PBTs) (ECHA database 2021), and UV-328 is under assessment for being a persistent organic pollutant (POP), to become listed under the Stockholm Convention. Some BUVs have been more studied than others, and information about the 10 BUVs included in this study is summarized in Table 1.

CHEMICAL POLLUTANTS SORBED TO THE PLASTICS

The chemicals that are not intentionally added to the pellets but that can sorb to the plastics in the environment include legacy POPs, such as PCBs (Yamashita *et al.* 2018). PCBs have been used in several different applications, including in transformers, heat exchangers, paper, plastics, lubricating oils, and paints (Erickson and Karley 2011). PCBs consist of 209 congeners, out of which about 130 (di- to deca-PCBs) are found in commercial mixtures. Between 1930 and 1990, more than 1.3 million tons of technical PCB mixtures were produced (Breivik *et al.* 2007). Due to the toxicity and environmental persistence of PCBs the production was stopped first in Japan in 1972, then in the US 1977, and in other countries by 1993 (Breivik *et al.* 2007), after being listed among the "dirty dozen", a group of chemicals that were categorized as POPs and banned or restricted under the Stockholm Convention (UNEP 2001).

Today, PCBs are still often found in the environment, despite being banned in most industrialized countries (Lu *et al.* 2021). Since 2005, International Pellet Watch (IPW) has used beached plastic pellets to analyze and monitor PCBs (http://www.pelletwatch.org/) (Takada and Yamashita 2016, IPW 2021). People from all over the world ship pellets to IPW for analysis. IPW then analyzes five subsamples and use the median value to categorize the concentration of PCB into different levels of pollution and create a database of values from around the world. The median has been found to correlate well with the concentrations in mussels as measured by Mussel Watch, a monitoring program established in the 1970s (Takada and Yamashita 2016).

Common sources of PCBs in the environment nowadays are the unintentional production in different industries such as paints (Anh *et al.* 2021), leaking from electrical transformers and contaminated sites, and the disposal and recycling of e-wastes. The latter has especially affected less industrialized Asian counties that, prior to 2017, received over 80% of the global electronic wastes (Lu *et al.* 2021). Moreover, since many buildings were built before the bans, PCBs often appear in indoor air, including indoor air in school buildings, which is concerning, as children are more sensitive to the toxic effects associated with PCBs (Marek *et al.* 2017). Additionally, PCBs exist as legacy pollutants in the environment, often accumulating in sediment where they can be re-suspended into the water column (Ogata, 2009).

PCBs are associated with several negative effects on the environment and human health, and in 2015, PCBs became classified as carcinogenic to humans (IARC, 2015). Moreover, even though they have been banned for many years, a study from 2018 showed that they still threaten the long-term viability of most of the killer-whale population (Desforges *et al.* 2018). They are also frequent in seabirds (Yamashita *et al.* 2018, Yamashita *et al.* 2021), and a global study of PCBs in seabirds found concentrations ranging from 1-60 000 ng/g lipid (Yamashita *et al.* 2018).

Although the effects of organisms ingesting microplastics with adsorbed PCBs, and how significant that pathway is compared to other pathways, are not fully elucidated, one study showed that PCBs can transfer to simulated clean gut systems in lugworms and cod, but the transfer was reported to be biphasic (Nor & Koelmanns, 2019), meaning that if the organisms had lower body burdens of PCBs it would transfer from the plastics to the organism, but if they had higher body burdens it would transfer to the plastics from the organism. Pellets with high concentrations of PCBs can thereby be assumed to spread PCBs to organisms/areas that are less polluted and studies on seabirds have shown that chemicals can transfer from ingested microplastics (Tanaka *et al.* 2020).

SOURCES AND TRANSPORT OF PLASTIC PELLETS

Plastic pellets end up in the environment due to continuous small-scale spills and due to larger spills following accidents. The continuous small-scale spills can amount to millions of pellets annually, from just one production site, and have been connected to a lack of preventive routines and limited accountability for the industry (Karlsson *et al.* 2018a).

Photo: Sören Funk, Unsplash.com

Larger spills, on the other hand, are often due to accidents during transport. These spills impact large coastal areas and clean-up attempts are often partly or largely volunteer-based. In 2012, outside of Hong Kong, 150 tons of plastic pellets were spilled into the ocean (Rochmann, 2013). For three months after that, volunteers were cleaning up beaches in the area. Still, six years later, big mounds of plastic pellets were being found on those same beaches (Gravier and Haut, 2020). Regardless of whether the pollution occurs from continuous smaller spills or occasional larger spills, the cleanup of the plastics, and its associated chemicals, will be challenging.

The polluter pays principle means that the entity that is responsible for the pollution should also remedy it. Plastic pellets enter the environment even before they have become plastic products and the responsibility for that pollution therefore belongs to either the producer or the subcontractors hired for transport and/or storage. There are also several international and regional policies and regulations that could be discussed in relation to plastic pellet pollution (for the EU, in part reviewed by the European Commission in 2013), (Stensgaard *et al.* 2017, and Karlsson *et al.* 2018a). The relevance of the policies depends partly on in which context the pellets are released (i.e., on land or at sea), partly on what they contain, and partly on whether they are considered as litter, industrial emissions, waste, or products.

Practically, the solutions to preventing the environmental pollution of plastic pellets are often quite straightforward. Suggested solutions include sweeping at the production and storage sites and the creation of barriers such as fences and/or filters. Still, continuous, and accidental spills are common and plastic pellets are frequently found in the environment.

Once in the environment, the floating pellets can get stuck on beaches close to the point of release (Karlsson *et al.* 2018a), and in seagrass (Carmen *et al.* 2021), but the pellets and their associated chemicals can also travel far from their point of release. Plastic pellets have been found all over the world (Fidra 2021, International Pellet Watch 2021), often on beaches that are not in any proximity to the plastic production plants.

Furthermore, a lot of floating plastics eventually sink. This can be either due to the material properties of the plastics themselves (i.e., they have a higher material density than seawater), or because the plastics get weathered and have things grow on them (Karlsson *et al.* 2018b), or because they get entangled with other debris.

AIM

The aim of this study was to investigate the presence and concentration of adsorbed PCBs and added and adsorbed BUVs in samples of beached pellets. The results will be used to highlight the concerns related to chemicals associated with plastic pellets polluting the environment and contribute to the development of recommendations on how to decrease the environmental pollution by plastic pellets and their associated chemical pollutants.

METHOD

Beached pellets were collected on beaches in 31 countries. The sampling was performed by IPEN partner organizations. The organizations looked for pellets on local sandy beaches along the high-tide line.

Approximately 100 pellets were collected per location. The samples were then wrapped in aluminum foil and sent to the VSCHT laboratory in the Czech Republic. There, all samples were sorted, and subsequently sent to the Laboratory of Organic Geochemistry of Tokyo University of Agriculture and Technology, Japan. Of the collected samples, 24 were analyzed for PCBs and 22 for BUVs (Table 2).

At the laboratory in Japan the pellets were, prior to analysis, sorted using a near-infrared spectrometer (Plascan-WTM OPT Research Inc., Tokyo, Japan) into polyethylene (PE), polypropylene (PP), and other polymers. Pigmented pellets were excluded from the analysis so that, in order to analyze pellets with a consistent weathering range, pellets that had reached a specific degree of yellowing could be chosen for analysis through comparing to internal laboratory standards (as described in Ogata *et al.* 2009).

Approximately 100 pellets were taken from each location. Those were then sorted according to the description above. Of the pellets that had the right level of yellowing and were confirmed to consist of polyethylene, five subsamples of five pellets each were taken per location for analysis. Extracts from the pellets were prepared using hexane. The extracts were subjected to acetylation, followed by fractionation by using silica gel column chromatography. PCBs (tetra- to nona-CB congeners) in apolar fraction were quantified using an ion-trap mass spectrometer fitted with a gas chromatograph (GC-MS). Thirteen congeners were quantified (CB# 66, 101, 110, 149, 118, 105, 153, 138, 128, 187, 180, 170, and 206). Details of the PCBs measurement method are available in Ogata *et al.* (2009).

Sample Collection: Plastic waste at the hide-tide line was sifted for plastic pellets. Roughly 100 pellets from each location were packaged, labeled, and sent for testing. Photos: Earth Thailand

TABLE 2. SAMPLE OVERVIEW

Country	Location	Sampled by	Latitude	Longitude	BUV	PCB
Argentina	Av. Félix U. Camet 1629 Mar del Plata, Buenos Aires, Argentina.	Bios Quilmes	S 37°58′ 54″	W 057°32′26″	x	x
Australia	Minim Cove	NTN	S 32°01′27″	E 115°46′15″	x	x
Bangladesh	Kalatoli Sea Beach	ESDO	N 21°14′02″	E 092°02′52″	x	x
Costa Rica	Playa Mantas	Rapal	N 09°42′22″	W 084°39′45″	x	x
Guinea	Benarés	Carbone Guinée	N 09°34′24″	W 13°36′29″	x	x
Jamaica	Fort Clarence	CARPIN	N 17°54′34″	W 076°53′00″	x	x
Kenya	Baobab beach	CJGEA	S 03°37′27″	E 39°52′28″	x	x
Malaysia	Penang straits	CAP	N 05°25′36″	E 100°19′35″	x	x
Mexico	Playas de Sánchez Magallanes en Cárdenas	Asociación Ecológica Santo Tomás	N 18°18′02″	W 093°50′38″	x	x
Morocco	Guy Ville beach	AMSETox	N 33°56′14″	W 006°56′49″	x	x
New Zealand / Aotearoa	Avon Heathcote estuary	Algalita	S 43°32′47″	E 172°42′50″	x	x
Nigeria	Finima beach	FOCONE	N 04°24′16″	E 007°08′03″	x	x
Philippines	Baseco beach	Ecowaste coalition	N 14°35′21″	E 120°57′16″		x
Poland	Poddąbie.	Buy responsibly foundation	N 54°37′33″	E 016°58′45″	x	x
Republic of Congo	Côte Sauvage	AED	S 04°49′18″	E 011°51′00″	x	x
Senegal	Plage De Hann Marinas	ADEC	N14°42′45″	W 017°25′50″	x	x
Sri Lanka	Colombo	CEJ	N 06°58′28″	E 079°52′09″	x	x
Tanzania	Coco beach	TABIO	S 06°46′28″	E 39°17′01″	x	x
Thailand	Bang Saen Beach Location	EARTH	N 13°17′54″	E 100°54′08″	x	x
Thailand	Mae Ram Phueng Beach	EARTH	N 12°35′32″	E 101°24′53″		x
Tunisia	Goulette Beach	AEEFG	N 36°59′42″	E 010°29′03″	x	x
Turkey	Uzunkum Plaiji	HEAL	N 41°10′14″	E 029°38′10″	x	x
USA	Cox Creek Texas	Wild at heart legal defence association	N 28°41′19″	W 096°31′43	x	x
Vietnam	Nguyen Tat Thanh beach	CGFED	N 16°5′29″	E 108°09′04″	x	x

Acetylated BUVs in polar fraction were measured by using a gas chromatograph equipped with a mass spectrometer (GC–MS; Agilent 7890A/5977). Details of the analysis method are available in Yamashita *et al.* (2021).

All BUVs were measured using a selected ion monitoring mode and quantified based on peak area. The concentrations of UV-326, UV-327, and UV-328 were corrected for the recoveries of the corresponding isotopically-labeled standards, whereas the other BUVs were not recovery-corrected.

Replicate aliquots were done of the pellet extracts to confirm the reproducibility with relative standard deviation (RSD) of analytical values less than 5% both for PCBs and BUVs. Recoveries were more than 80% for both classes of compounds. A procedural blank was run in each set of five samples. LOQ is defined as analytical values less than three times the corresponding blank. For Σ13 PCBs the LOQ was normally 0.3 ng/g. For BUVs the LOQ ranged from 0.5 ng/g-pellets for UV-320 to 13 ng/g for UV-328.

To compare the concentrations with previous samples median values were used. The median values of five subsamples have previously been found to correlate with concentrations in mussels (Ogata *et al.* 2009). Additionally, it excludes sporadic high concentrations (Yamashita *et al.* 2019), thereby making it more representative of the local pollution.

RESULTS

BENZOTRIAZOLE UV STABILIZERS

All analyzed BUVS were found in subsamples from all locations. All BUVs were also found in most of the subsamples (Figure 1). The highest concentrations were found in subsamples from Morocco, Jamaica, and Congo (Figure 3). Looking instead at median levels, the highest concentrations were in samples from Congo, Tanzania, and Senegal.

BUVs were measured in a total of 110 subsamples (5x22). Each subsample was analyzed for ten different BUVs. All ten analyzed BUVs were found in subsamples from all locations. UV-P was found in all subsamples. UV-326 to 329, UV-9, and UV-PS were all found in more than 90% of the subsamples. UV-350 and 320 were found in 80% and 70% of the subsamples respectively (Figure 1).

In terms of composition, UV-327, UV-326, and UV-9 were found in the highest concentrations. They also had the highest average and median values (Figure 2).

The highest total BUV concentrations were found in subsamples from Morocco, Jamaica, and Congo (Figure 3). Those three countries are also the countries with the highest averages. Looking instead at median levels of the total BUV concentrations, the highest concentrations were in samples from Congo, Tanzania, and Senegal.

The lowest measured total BUV concentrations for most countries was below 500 ng/g. Exceptions were Australia (528 ng/g), Senegal (520 ng/g), Tanzania (751 ng/g), and Congo (1258 ng/g).

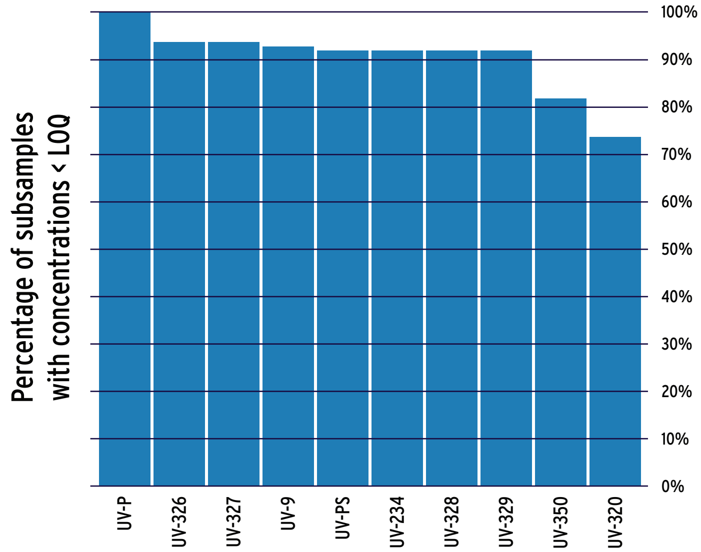


Figure 1. BUVs in pellet samples.

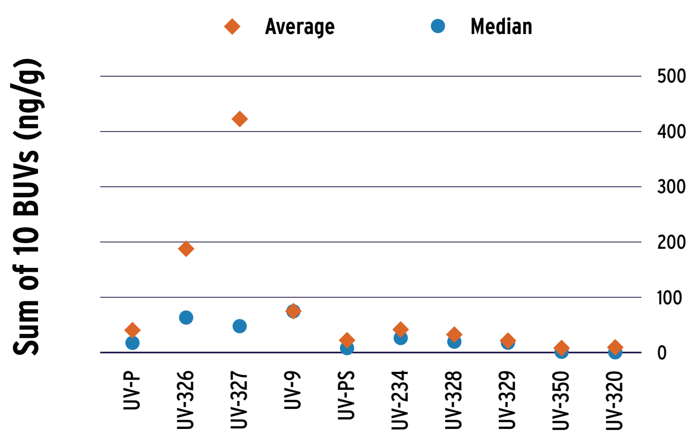


Figure 2. Averages and medians from all samples > LOQ

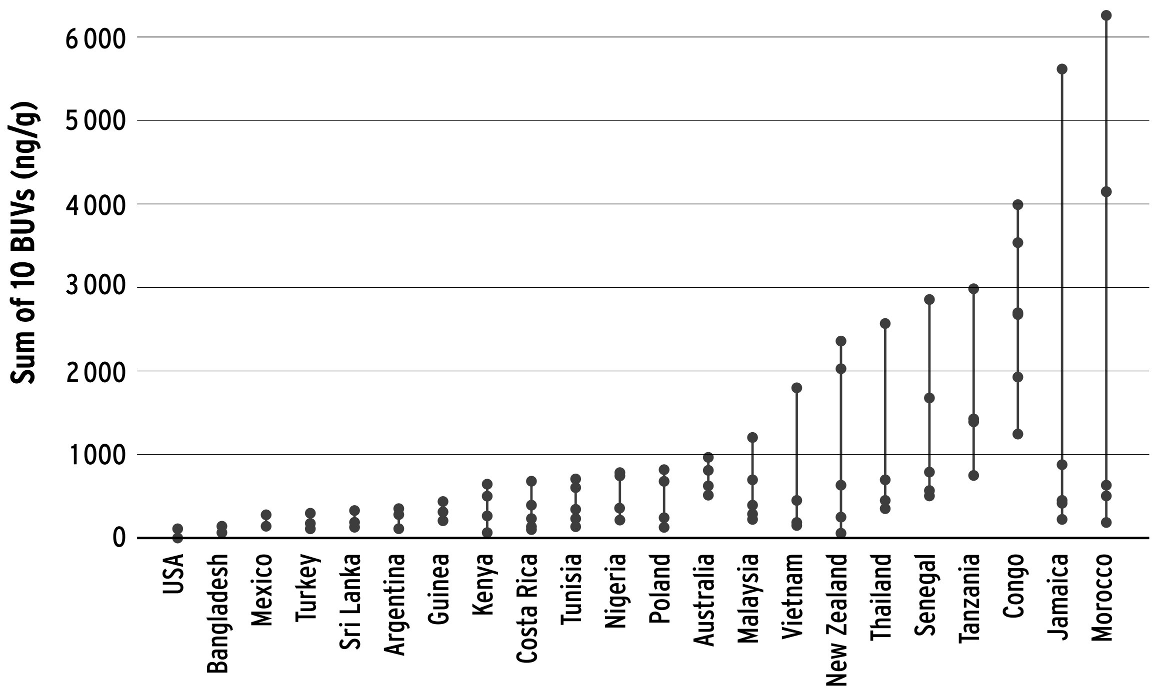


Figure 3. Concentrations of ten BUVs in subsamples of pellets from different countries.

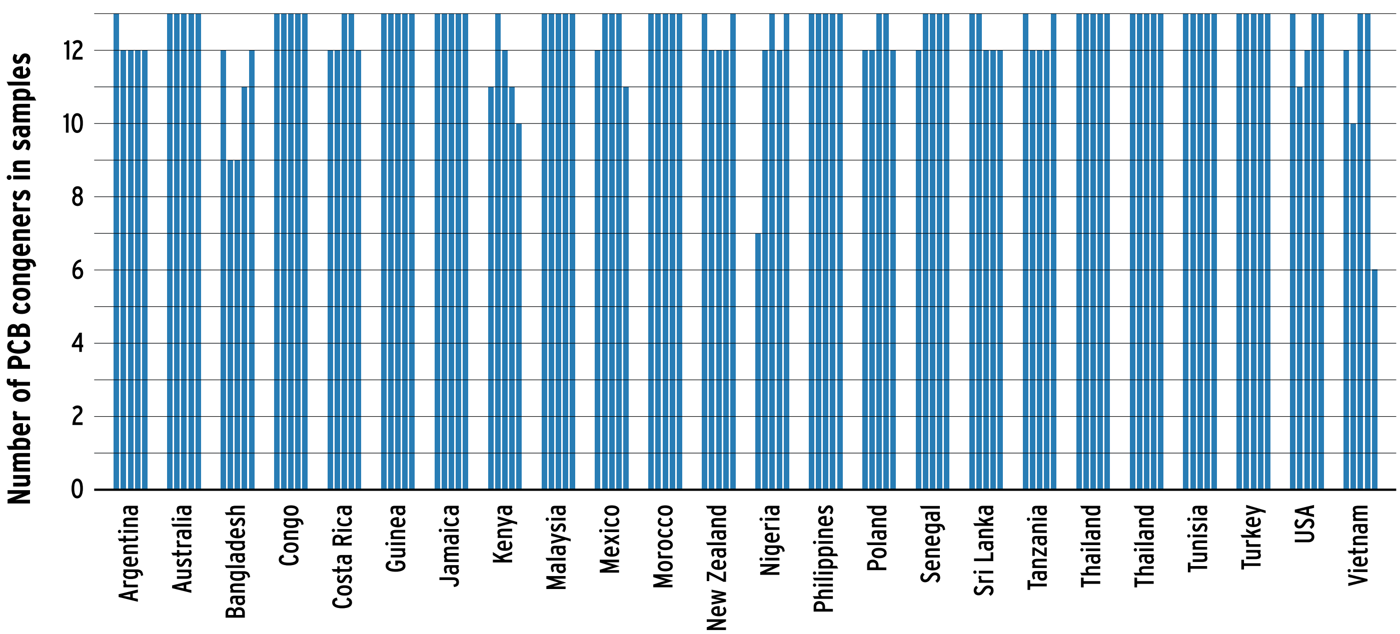


Figure 4. Number of detected PCB congeners in the samples (out of 13 total measured).

POLYCHLORINATED BIPHENYLS

All 120 subsamples had PCBs in them and all PCB congeners were present in samples from all locations (Figure 4). Samples from Senegal and Tunisia were extremely polluted (Figure 6). Samples from Tanzania, Congo and Morocco has concentrations ranging from moderately to highly polluted (Figure 6).

BUVs were measured in a total of 110 subsamples (5x22). Each subsample was analyzed for ten different BUVs.

PCBs were measured in a total of 120 subsamples (5x24). In each subsample thirteen different PCB congeners were measured. All subsamples had PCBs in them, and all measured PCB congeners were present in samples from all locations (Figure 4). 68% of all subsamples had detectable levels of all 13 PCB congeners. Regarding the PCBs composition (Appendix #), generally, penta-, hexa-, and hepta-CB congeners (i.e., CB110, 138, 180) were dominant. This pattern is similar to those observed in pellets from other locations in the world (e.g., Hosoda *et al.* 2014; Mizukawa *et al.* 2013; Yeo *et al.* 2015). The detailed composition varied among the locations. However, we could see a predominance of CB-138 and CB-180 in most of the samples and especially in samples with high PCBs concentrations, i.e., from Senegal, Tunisia, and Tanzania. In the pellets from these countries CB-138 accounted for more than 50% of the total for most of the samples.

Total PCB concentrations (sum of 13 congeners) in the analyzed pellet samples ranged from 1 to 4188 ng/g. In relation to the classifications used internationally by the International Pellet Watch, several subsamples and the medians of the pellets from Senegal and Tunisia would be considered extremely polluted (Figure 5).

The background level of PCBs pollution, which is derived from atmospheric transport and the long-range transport of PCBs when sorbed to floating plastics, is established at 10 ng/g. In this study, samples from three locations had concentrations corresponding to background levels. Those were found in Bangladesh (1-2 ng/g), Baobab beach, Kenya (3-9 ng/g), and Vietnam (1-10 ng/g) (Figure 6).

In Argentina (3-16 ng/g), Cox Creek Texas, USA (1-18 ng/g), Thailand (9-21 ng/g), Turkey (6-23 ng/g), Malaysia (8-35 ng/g), and Costa Rica (3-45 ng/g) the concentrations in the subsamples ranged from non-polluted to lightly polluted. In Thailand (9-68 ng/g) the concentrations ranged from non-polluted to moderately polluted.

PCBs Concentration (ng/g-pellet)

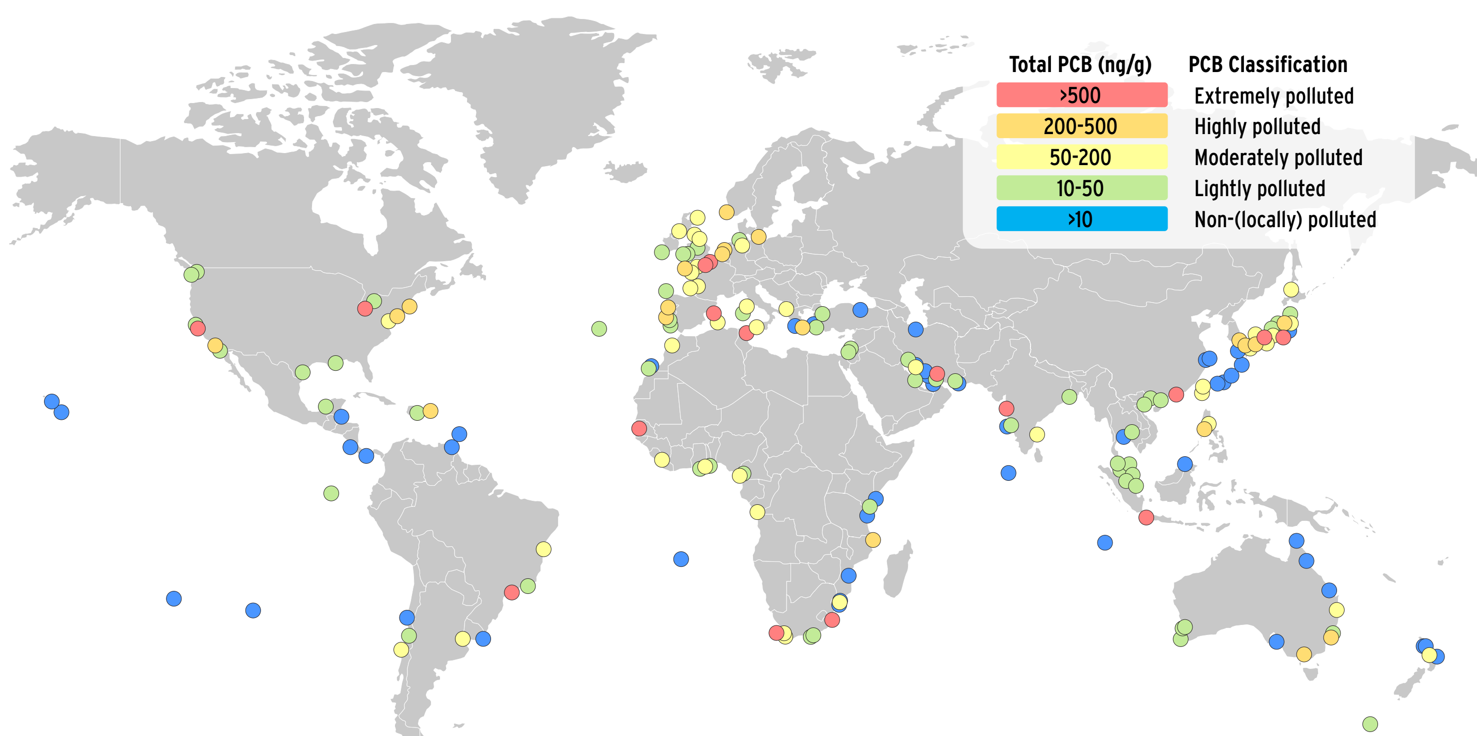
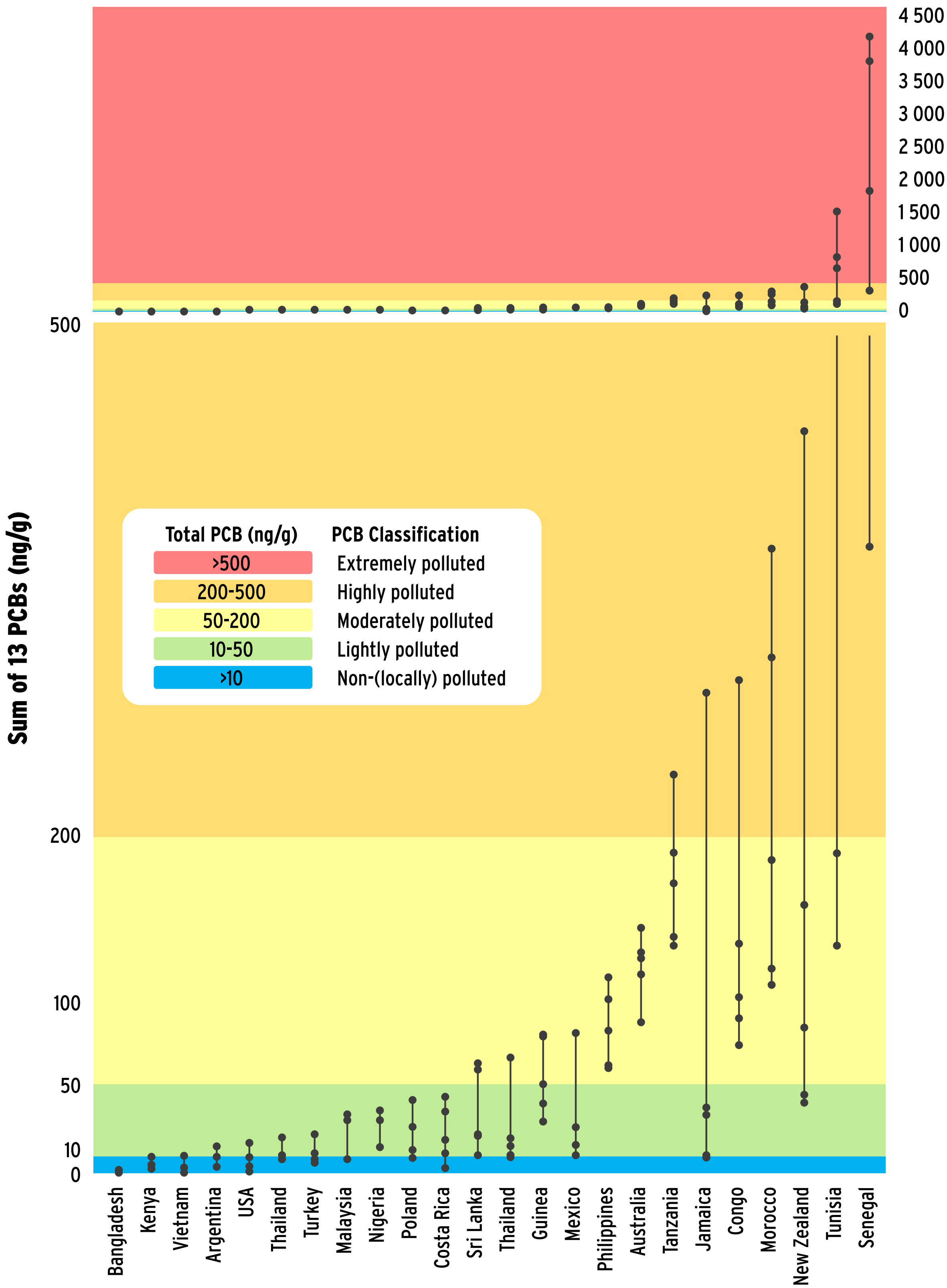


Figure 5. PCBs concentration (ng/g, median values) in plastic resin pellets around the world including samples from the current study.

Figure 6. Sum of 13 PCBs in pellets for all countries.
Bottom: Values below 500 ng/g. Top: Values scaled to include highest levels tested.



In Nigeria (15-37 ng/g) and Poland (10-43 ng/g) all subsamples had concentrations corresponding to lightly polluted.

In Sri Lanka (11-65 ng/g), Guinea (31-82 ng/g), and Mexico (11-83 ng) the concentrations ranged between lightly polluted and moderately polluted. In Jamaica (10-286 ng/g) and New Zeeland/Aotearoa (42-442 ng/g) concentrations ranged from lightly polluted to highly polluted.

Subsamples from the Philippines (62-116 ng/g) and Australia (90-146 ng/g) were moderately polluted. In Tanzania (135-238 ng/g), Congo (76-294 ng/g), and Morocco (112-372 ng/g) the concentrations ranged from moderately polluted to highly polluted and in Tunisia (135-1570 ng/g) it ranged from moderately polluted to extremely polluted. In Senegal (373-4188 ng/g) all subsamples were either highly or extremely polluted.

Figure 7. Beach in Congo sampled by AED Congo.

COMBINED LEVELS OF POLLUTANTS

Detailed results for the combined levels of pollutants

The concentrations of BUVs and PCBs in the plastics did not necessarily seem to co-occur with the level of contamination on the beaches. In Guinea, for example, the sampled beach had so much plastic pollution that it covered large parts of the actual beach (Figure 8), but the concentrations of BUVs and PCBs were quite low in comparison to some of the other samples (lightly to moderately polluted PCB concentrations).

The sampled beach in Congo, however, did not have as much visual pollution (Figure 7), but the sampled pellets had among the highest concentration of BUVs among the analyzed pellets (Figure 3).

Figure 8. Beach in Guinea sampled by Carbone Guinée.

DISCUSSION

In this study the presence of additives and sorbed POPs, more specifically BUVs and PCBs, were analyzed in pellets sampled on beaches from all over the world. All analyzed BUVs and PCBs were found in samples from all locations.

BENZOTRIAZOLE UV STABILIZERS

Ten different BUVs were analyzed in samples from 22 countries. Although all BUVs were found in samples from all locations, the concentrations for the BUVs were particularly high in Congo, Tanzania, Senegal, Morocco, and Jamaica.

The concentrations of individual BUVs in this study ranged from 1-5082 ng/g-pellet. In a previous study on beach debris, collected in South Korea, the concentrations of individual BUVs ranged from 0.3-81 700 ng/g. Beach debris collected in Hawaii had concentrations ranging between 0.2-1130 µg/g (200-1 130 000 ng/g). However, both of those studies looked at plastic products or fragments of plastic products.

THE FACT THAT ALL SAMPLED BUVs WERE DETECTED IN SAMPLES FROM ALL LOCATIONS, AND THAT THEY ARE ASSOCIATED WITH SEVERAL NEGATIVE EFFECTS ON HUMAN HEALTH AND THE ENVIRONMENT, ILLUSTRATES THAT THE PROBLEMS ASSOCIATED WITH UV-328 ARE SHARED WITH SIMILAR BUVs.

Regarding the composition of BUVs, the highest concentration in a subsample (5082 ng/g) and the highest average concentration (430 ng/g), were found for UV-327, which was present in 103/110 samples. UV-327 is listed as harmful to aquatic life, with long-lasting effects and is under assessment as persistent, bioaccumulating, and toxic (PBT) (ECHA 2021). Previous studies have found UV-327 in northern pike plasma (Lu *et al.* 2016) as well as in plastic debris (Rani *et al.* 2017, Tanaka *et al.* 2020a).

The highest median concentration (80 ng/g), on the other hand, was for UV-9, which was found in 102 out of 110 analyzed samples. UV-9 is listed as very toxic to aquatic life with long-lasting effects and is under assessment as a PBT (ECHA 2021). Studies have also shown that UV-9 can have estrogenic effects (Feng *et al.* 2020).

UV-P was found in all analyzed samples and is, just like UV-9, very toxic to aquatic life with long-lasting effects. It is also under assessment as a persistent, bioaccumulating and toxic chemical (ECHA 2021) and has been shown to induce estrogenic activity in in vitro and computational studies (Feng *et al.* 2020). It has previously been found in preen gland oil of seabirds (Yamashita *et al.* 2021). Studies have shown that it can activate the aryl hydrocarbon pathway in zebrafish (Fent *et al.* 2014) and in humans it shows an aryl hydrocarbon receptor ligand activity. The aryl hydrocarbon receptor pathway is a pathway that is often involved in how environmental pollutants affect animals. It can act as a trigger for several different types of effects such as hormonal imbalance, metabolic imbalance, carcinogenic activity, and developmental effects. UV-P also has a potential to bioaccumulate (Nagayoshi *et al.* 2015) and has shown androgen receptor antagonistic activities, and estrogen receptor agonistic activities (Sakuragi *et al.* 2021), meaning that they can disturb the hormonal balance through interacting with the receptors for estrogen and androgen in different ways.

The second highest median (67 ng/g) and average (193 ng/g) were found for UV 326. UV-326 was found in 103/110 samples and is under assessment as a PBT (ECHA). It has previously been found in liver samples from Arctic seals (Lu *et al.* 2019), preen gland oil of seabirds (Yamashita *et al.* 2019), and in plastic debris (Rani *et al.* 2017, Tanaka *et al.* 2020). UV-326 can, like UV-9, activate the aryl hydrocarbon pathway in zebrafish (Fent *et al.* 2014). And just like UV-9 it has shown aryl hydrocarbon receptor ligand activity in humans with the potential to accumulate (Nagayoshi *et al.* 2015).

Photo: Mare Nostrum, Romania

The third highest average concentration was for UV-328 which was found in 101/110 samples. UV-328 is under assessment as a POP and has previously been found in samples of plastic debris (Rani *et al.* 2017, Tanaka *et al.* 2020). UV-328 has also been detected in liver tissue of a Northern Fulmar in the Arctic (Lu *et al.* 2019), as well as the double crested cormorant (Lu *et al.* 2019 b), in preen gland oil of seabirds (Yamashita *et al.* 2019), blood plasma of bottle nose dolphins (Lu *et al.* 2016), and several species of fish (Lu *et al.* 2016, Lu *et al.* 2019). UV-328 acts as an estrogen receptor antagonist (Sakuragi *et al.* 2021). Upon metabolizing, the toxicity alters and has shown a more potent anti-androgenic activity (Zhuang *et al.* 2017). It can also cause an increased reactive oxygen species production in the algae *Chlamydomonas reinhardtii* (Giraudo *et al.* 2017). UV-328 has also been shown to induce transcription of ribosomal proteins in juvenile rainbow trout, as well as downregulate genes that are involved in immune responses (Giraudo *et al.* 2020).

UV-328 is a persistent chemical with a half-life of several months, and studies in soil have revealed a half-life of 79-223 days (Lai *et al.* 2014). It has a density higher than seawater, so if the chemical is released into surface waters it will sediment and although sediment can be transported over large distances, it is often slower than surface-water transport. However, since UV-328 is frequently released into the environment as an additive in low-density plastics, it can be transported far with surface-water transport, as evidenced by this study, where it is found in plastics on beaches all over the world.

Moreover, the fact that all sampled BUVs were detected in samples from all locations, and that they are associated with several negative effects on human health and the environment, illustrates that the problems associated with UV-328 are shared with similar BUVs. Often times, chemicals are assessed on a chemical-by-chemical basis, which is a slow process that can lead to one hazardous chemical being substituted by another equally hazardous chemical due to a lack of data for that specific chemical. By taking a class-based approach, chemicals that have similar chemical structures and properties are assessed together. Considering the many similarities between the different BUVs, and the fact that they have similar effects, a class-based approach towards BUVs would be preferrable to a chemical-by-chemical approach.

POLYCHLORINATED BIPHENYLS

The concentrations of BUVs and PCBs did not correlate. But, like BUVs, samples from 24 countries were tested for 13 PCB congeners and all congeners were found in all locations. The total concentrations varied from background levels for some locations to extremely high values when compared to previous global samplings (Hirai *et al.* 2011).

Most of the locations, 21 of 24 total, had samples with PCB concentrations that were higher than 10 ng/g-pellet. This means that there must be one or more local sources of PCBs around the locations, or that the pellets have adsorbed PCBs in one location and then brought the PCBs with them to the beach.

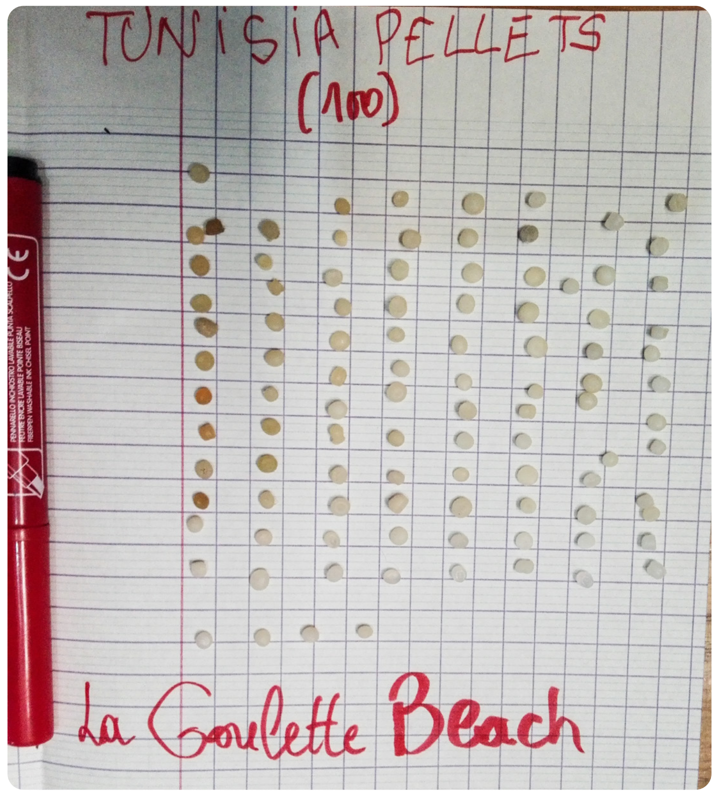
The extreme and high levels of PCBs found in several of the samples from Africa are comparable to levels found in US, Japan, and European cities in previous studies. These areas are known to have had rapid economic and industrial growth by the 1970s, causing a considerable amount of PCBs to be produced, used, and released into coastal areas (Karapanagioti *et al.* 2011; Mizukawa *et al.* 2013; Ogata *et al.* 2009; Yeo *et al.* 2015).

Global production of PCBs mainly occurred in the US, Europe, and Russia, and approximately 97 % of this was used in the Northern Hemisphere. Africa has never been a producer or major user of PCBs, so the concentrations found in the study are simply too high to be rationalized by historical usage of PCBs on the continent (Gioia *et al.* 2011). The sample from Senegal showed extremely high concentrations of PCBs. The samples were collected at Plage De Hann in Hann Bay, an area that is surrounded with industrial activities, located near the Mbeueuss landfill in Pikine city, 27 km from Dakar. In 2014, a study demonstrated that the e-waste scrap yard was the source of PCBs in pellets collected in Ghana (Hosoda *et al.* 2014), which could be a possible source also in this case, although we have no evidence of the sources for this specific location in our study.

Tunisia had the second highest concentrations of PCBs. The sample was taken in the Gulf of Tunis. The usage of PCBs in Tunisia is not well established. In 1986, the import into Tunisia of transformers or any equipment containing PCBs was banned. However, a large number of transformers containing PCBs are still used or presently stored in unsatisfactory conditions (Barhoumi *et al.* 2014). Therefore, there is a possibility of other local sources of PCBs.

In samples from Southeast Asia, we observed moderate concentrations in Baseco Beach, the Philippines, with 68 ng/g-pellet, categorized as moderately polluted. However, Bang Saen Beach in Thailand showed lower concentration with 16 ng/g-pellet, which can be categorized as lightly polluted. Compared to the global concentration (see Figure 5), the pollution status in Southeast Asia is comparable with previous samples that have been analyzed in the same region.

The samples from Jamaica and Mexico both have light concentrations with 27 ng/g-pellet and 16 ng/g-pellet respectively. Both these locations were located in remote areas with little to no industrial activities within the area and are at a considerable distance away from any substantial PCBs sources, which explains the low levels of PCB concentrations detected.



It should however be noted that the PCBs can have sorbed to the plastics locally or in another location prior to landing on the sampled beach. High concentrations of PCBs sorbed to the pellets have also been discussed to have a link to legacy PCBs in sediment, wherefore it is not necessarily linked to current releases of PCBs. Although the sorbtion and desorption of PCBs onto pellets is biphasic (meaning that PCBs both sorb and desorb from plastics) and strives towards an equilibrium, this process takes time (estimated to one year or more, according to Yamashita, 2018). It is therefore not possible to deduce whether the PCBs sorbed to the plastics locally or have been transported from an area with high concentrations of PCBs. Comparing median concentrations across the world the high concentrations still indicate the potential of local sources of PCBs.

It is also important to note that samples were collected from one or maximum two locations per country, meaning that it does not provide a high-resolution overview of the contaminants. It does however provide a snapshot, and the high and extreme cases in some of the samples from this study do give an indication of areas that potentially have high contamination levels of PCBs. Regulatory and remediating efforts in these areas might therefore be necessary.

It is also important to note that the samples analyzed in this study from Morocco, Congo, and Senegal had among the five highest concentrations of both BUVs and PCBs. Tanzania also had the fourth highest concentration of BUVs and Tunisia the second highest concentration of PCBs. All of these five countries are situated in Africa, where there is very limited production of chemicals and plastics. This study therefore further exemplifies how plastics move over national borders and highlights the importance of international collaborations to stop the spread of plastics and toxic chemicals in plastics.

CONCLUSIONS AND RECOMMENDATIONS

In this global study, all ten analyzed BUVs and all thirteen analyzed congeners of PCBs were found in pellets from all sampled locations. These results highlight that the preproduction plastic pellets, that are found on beaches all over the world, bring toxic chemicals with them.

The presence of several different BUVs, including UV-328, in the plastic pellets show that plastic pellet debris can increase the spread of these chemicals. Several BUVs are persistent, bioaccumulative, and toxic and have shown to act as endocrine disruptors in previous scientific studies. Taken together with the results in this study, it shows that multiple BUVs are ubiquitous contaminants that pose risks to human and environmental health. It is therefore important to take a class-based regulatory approach to prevent harm to the environment and human health from BUVs.

Measurements of PCBs, in this study, showed alarmingly high concentrations in several African countries, suggesting that there are regional sources of PCBs, possibly linked to e-waste sites. Consequently, particular attention must be paid to African countries facing problems related to legacy POPs.

Furthermore, it is important to note that the level of contamination on specific pellets is not a direct measure of the total level of contamination at that location, as some beaches are more littered than others. However, the presence of all analyzed BUVs and PCBs on all sampled beaches illustrate that it is not only the physical aspects of the litter that is polluting the beaches. These results also illustrate that even if a beach might visually have less waste plastic present, smaller amounts of visible contamination can still be accompanied with high levels of chemical pollution.

The challenges behind the situation as presented here are manifold. Plastic pellets are found all over the world and with them they carry additives and adsorbed environmental contaminants. In essence, this problematic situation can be traced back to a few key drivers.

Firstly, industries are rarely held responsible for plastic spills, even when there are regulatory frameworks that could prevent the spills if properly implemented. Secondly, banned chemicals are still ever-present in the environment due to their persistence and a lack of control mechanisms, particularly during recycling of e-wastes. And lastly, additives, such as UV stabilizers, associated with negative effects on environmental and human health, are still allowed in plastic products and frequently paired with very limited transparency.

To prevent further environmental contamination, it is therefore important that:

1. Overall production of plastics is decreased
2. Companies are held responsible to prevent and mitigate the release of plastic pellets,
3. Regulatory control mechanisms of banned chemicals are improved,
4. Additives associated with negative environmental and human health effects, such as BUVs, are substituted with safer alternatives.

Since plastic pollutants do not respect national borders, it is crucial that prevention and mitigation measures are created and implemented through international collaborations.

RECOMMENDATIONS

To tackle the problem of toxic chemical additives in plastics entering the environment, international agencies and policy makers should:

- Support the addition of UV-328 to Appendix A in the Stockholm Convention without exemptions;
- Accelerate the phase-out of 'groups' of toxic chemicals, rather than taking a substance-by-substance approach;
- Establish a right-to-know regulation that requires producers to publicly disclose substances and chemical additives used in products.
- Ensure that the polluter pays principle is enforced; and
- Ensure that companies involved in producing and handling pellets adopt strategies to avoid spills into the environment.

The plastics industry should:

- Establish routines to prevent the release of plastic pellets during production, transport, and storage;
- Stop adding toxic chemicals to plastics products;
- If some additives are essential for specific plastic products, the safety of those additives should be confirmed by a third-party organization; and
- List plastic ingredients, including additives, on labels and make the chemical content of plastics traceable throughout all its life and waste stages.

Overall, governments should work towards decreasing the production of non-essential plastics, including ending subsidies for fossil fuel extraction and plastic production facilities. Global agreements should prevent the release of plastics into the environment.

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