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MICROPLASTIC DISTRIBUTION AND DETECTION OF CHLORDECONE ON MICROPLASTICS IN MARINE SEDIMENTS IN GUADELOUPE: A PRELIMINARY STUDY§

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Abstract: Plastic pollution in the oceans is recognized as a worldwide problem. Since the 1950s, the production of plastics has been increasing and the first reports of microplastics (particles < 500 µm) in the marine environment began to appear in the 1970s. These particles represent a growing environmental problem due to their dispersion in seawater and marine organisms. Additionally, microparticles in general can adsorb pollutants that will then become bioavailable to organisms by being desorbed during digestion, which could be an important pathway for the contamination of organisms. In Guadeloupe and Martinique, an organochlorine pesticide called "chlordecone" was used from 1972 to 1993 in banana plantations and this very persistent pollutant contaminates soils, rivers, and coastal marine areas and accumulates in marine foodwebs. To examine these issues, we had two goals: 1) to assess the contamination of marine sediments by microplastics surrounding Guadeloupe; and 2) to determine the ability of microplastics to adsorb chlordecone, as has been demonstrated for other organochlorine pollutants. To do so, marine sediments were collected in triplicate from 12 sites in coral reef environments around the island. Microplastics from each sample were then enumerated by size, color and shape under a binocular microscope. The results indicate that microplastics are found in all the studied sites and that their distribution could be linked to marine currents or proximity to areas of significant human activities (port activities, agglomeration, etc.). Finally, our preliminary results indicated that chlordecone could be adsorbed onto microplastics, with a concentration ranging from 0.00036–0.00173 µg/µg of microfilter.

Key words: plastic fibers, pesticide, Caribbean, coastal pollution, coral reefs

Introduction

Since the 1950s, global plastic production has increased exponentially mainly due to being inexpensive, durable and a light weight material (Andrady 2011). However, only a portion of these plastics is recovered and recycled at the end of their life. In 2016, out of 27.1 million tons of plastics collected in Europe, only 31.1% were recycled (Plastic Europe 2018). Plastics then accumulate in landfills and gradually spread into the environment. Because they are not digested, plastic particles can obstruct the respiratory or digestive tracts and may lead to the death of marine mammals (Laist 1997), birds (Mallory 2008) and turtles (Bugoni et al. 2001). The first reports of microplastics in the marine environment appeared in the 1970s and 1980s (Carpenter and Smith 1972, Carpenter et al. 1972, Harper and Fowler 1987). These publications invited the scientific community to debate on the origin of these microplastics and their environmental and health consequences. Microplastics can be produced directly or resulting from physical, chemical or biological degradation. Various polymers are found in seawater, of which the most common are Polyethylene (PE), Polypropylene (PP), Polystyrene (PS), Polyethylene Terephthalate (PET), and Polyvinyl Chloride (PVC) (Brien 2007). Gregory and Andrady (2003) defined microplastics as plastic particles < 500 µm; < 1 nm, they are called "nano–plastics."

Plastics can adsorb several organic contaminants such as Polychlorinated Biphenyls (PCBs), Polycyclic Aromatic Hydrocarbons (PAHs) or Polybrominated Diphenyl Ethers (PBDEs), as well as heavy metals due to their hydrophobicity and generally high partition coefficient with organochlorine compounds (Teuten et al. 2007, 2009, Frias et al. 2010). These physical properties also allow plastics to carry endocrine disrupting substances (Gallo et al. 2018). Since their discovery, organochlorine compounds have been widely used and massively produced for many purposes such as to produce materials such as PVC and Teflon, as pesticides, or as compounds in insulators such as PCBs (Lange 2001). The abundance of these compounds has been responsible for many environmental issues, since most are toxic and have been distributed worldwide (Tanabe et al. 1996).

In the aquatic environment, organochlorine pesticides are generally present in low concentrations (µg/kg) or even in trace concentrations (ng/kg) but can nevertheless cause toxic effects in organisms due to bioconcentration and bio-

§*This article is based on a presentation given in November 2018 at the 71st annual Gulf and Caribbean Fisheries Institute conference in San Andrés, Columbia.*

magnification phenomena (Sonnenschein and Soto 1998). The adsorption of these molecules onto microplastics makes them much more bioavailable to aquatic fauna. The adsorbed concentrations can be several orders of magnitude higher than the concentration in seawater (Moore 2008). These pollutants can thus be desorbed when ingested by aquatic organisms (Devriese et al. 2017). Microplastics can also constitute a new pathway for contamination of marine food webs (Teuten et al. 2007).

Guadeloupe Island is particularly affected by an organochlorine pesticide called chlordecone. About 300 tons of this molecule were used in banana plantations in Guadeloupe and Martinique between 1972 and 1993 (Devault et al. 2016), which causes severe damage to the terrestrial and aquatic environment (Merlin 2015). Classified as POPs (Persistent Organic Pollutants) in the Annex A of the Stockholm Convention (i.e., to be eliminated), this pesticide is highly toxic to many organisms (MacPhee and Ruelle 1969, Roberts and Bendl 1982). Due to its structure, chlordecone is very persistent with an estimated half–life time of several decades (Cabidoche et al. 2009). Chlordecone is also biomagnified in the marine food–webs (Bouchon and Lemoine 2007, Dromard et al. 2018).

In recent years, many studies have focused on contaminant adsorption onto microplastics (Daugherty 2016, Devriese et al. 2017, Hüffer and Hofmann 2016, Koelmans et al. 2016, Teuten et al. 2007, 2009), but none have been conducted on tropical marine ecosystems and none have been focused on chlordecone. The present study aims to quantify the microplastics present in the marine sediments around Guadeloupe Island and their capacity to adsorb chlordecone.

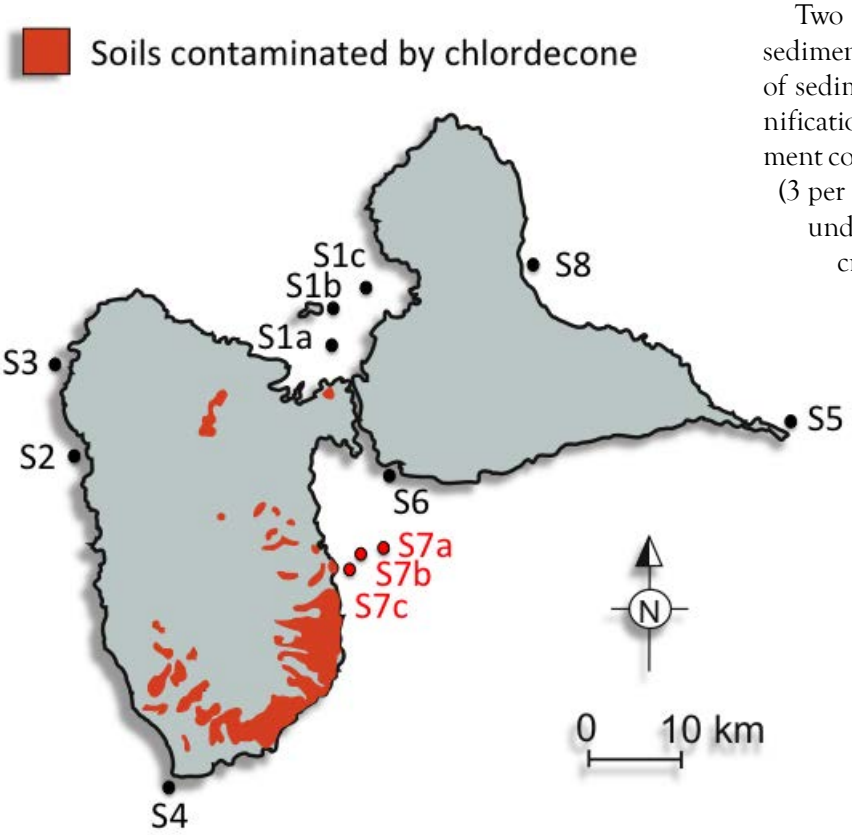

FIGURE 1. *Location of microplastic sampling sites around Guadeloupe Island. Sites with red label were used to study adsorbed chlordecone on microplastics. S1a: Lagoon of Grand Cul-de-sac Marin Bay (GCSM), S1b: Passe-à-Colas, S1c: External reef slope of GCSM, S2: Pointe Noire, S3: Deshaies, S4: Anse Dupuy, S5: Pointe des Châteaux, S6: Îlet Cochon, S7a: Caye à Dupont, S7b: Îlet Fortune, S7c: Lagoon of Petit-Bourg, S8: Moule.*

Materials and Methods

Sampling protocol

Triplicate marine sediments samples were collected in each of 12 sampling sites around Guadeloupe Island (Figure 1) based on their geographic location (north, south, east and west coasts), and different potential sources of exposure to chlordecone pollution. These sediment samples were collected in coral reefs between 3—5 m deep using glass containers to avoid contact with plastic material. For the same reason, the use of plastic gloves was avoided during all sampling and laboratory experiments. Sediments were transferred to the laboratory, placed in aluminum trays, and dried in an oven (60°C) until reaching a constant dry weight.

Microplastic enumeration

Two methods were used to extract microplastics from sediment. The first method consisted of direct observation of sediments under a binocular microscope (range of magnification: 16–25x), and was used to assess the level of sediment contamination around Guadeloupe. For each replicate (3 per site), 2 subsamples of 5g of sediment were examined under a binocular microscope in a glass Petri dish. Microplastics (fibers and fragments) were then sorted using fine forceps, counted, and enumerated (#/5g sediment). The identification of microplastics was done according to the size, the color and the shape of the particle as in Hidalgo–Ruz et al. (2012). Numbers of microplastics per site were compared using a Kruskal–Wallis (K–W) nonparametric ANOVA test, followed by a post–hoc comparison test (Multiple Comparisons Test after K–W, Sieger and Castellan 1988) if the K–W test was significant. All statistical analyses were conducted using the R software v3.5.1 (R Development Core Team 2018), package *pgirmess*, and using 0.05 as the critical significance level.

The second method was used to extract microplastics that were used to study the adsorption of chlordecone on the plastic particles. To do so, 3 sites were chosen due to their proximity to chlordecone–contaminated watersheds (sites S7a, S7b and S7c, Figure 1). To extract the microplastics from these samples, two 200 g sub–samples of sediment were added to 200 ml of a NaCl solu-

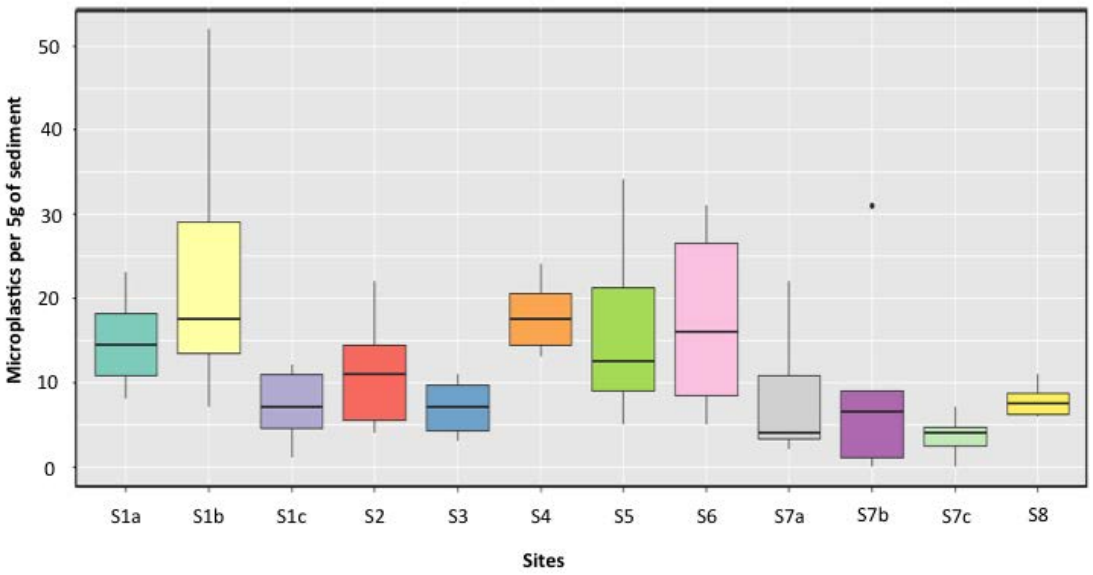

FIGURE 2. *Mean number of microplastics per 5 grams of sediment in each site. Location of the study sites is given in Figure 1. The boxplots depict the mean (horizontal bar), the second and third quartiles (shaded areas), the confidence interval (95%), and the dot is an outlier. S1a: Lagoon of Grand Cul-de-sac Marin Bay (GCSM), S1b: Passe-à-Colas, S1c: External reef slope of GCSM, S2: Pointe Noire, S3: Deshaies, S4: Anse Dupuy, S5: Pointe des Châteaux, S6: Îlet Cochon, S7a: Caye à Dupont, S7b: Îlet Fortune, S7c: Lagoon of Petit-Bourg, S8: Moule.*

tion with a salinity of 140, following the protocol of Martins and Sobral (2011). The supernatant was recovered and rinsed 3 times in 50 ml of the NaCl solution. This method was chosen to obtain a high amount of microplastics for the analysis. The supernatant, containing the microplastics recovered from sediments, was treated with a sodium hypochlorite solution (8%) to remove the organic suspended matter present. The supernatant was then filtered through 47 mm diameter Whatman GF/F glass microfiber filters (0.7 µm pore size) and rinsed with distilled water. However, since a deposit of material was still visible after filtration, they were thus filtered a second time on Whatman filters. Filters were then dried to constant weight in an oven (60°C).

Chlordecone extraction and analysis

Whatman glass microfiber filters containing microplastics were placed in a microwave in a pressure—resistant vessel containing 15 ml of a 50/50 mixture of dichloromethane and methanol to extract chlordecone potentially adsorbed onto microplastics. Due to the small amount of microplastics collected on filters, replicates of each site were pooled for the analysis. The extracts were recovered on glass—fiber filters concentrated under nitrogen flow and transferred into injection vials (3 rinses). A blank was used to control for any external contamination during the experiment. Chlordecone analysis was performed by liquid chromatography coupled with a tandem mass spectrometer (GC—MS—MS analysis, Agilent LC 1290 Infinity™) with internal calibration. Internal standard and measurement solution were prepared from a chlordecone solution marked with the carbon isotope ^{13}C at 5.549 µg/g^{1} and a native chlordecone solution at 5.668 µg/g^{1}. Acetonitrile and water were used as solvents for the chlordecone analysis and 2 ionic transitions were detected at m/z 506.9—426.7 and m/z 508.9—428.7. Acetonitrile blanks (spike) were introduced between each sample during injection.

Concentration of adsorbed chlordecone

The number of microplastics was estimated from the data obtained by counting of particles on these same samples (3 replicats per site, see "Microplastic enumeration" section). The mean weight of microplastics per filter (m_{MP}, µg/filter) was estimated according to the nature of the polymers (Callister 2007). The area values obtained by LC—MS—MS were used to calculate the quantity of chlordecone adsorbed (in µg) per µg of microplastic collected on the filters (M_{CLD}; µg/µg of microplastics).

Results

Contamination of marine sediments by microplastics

Microplastics, principally fibers, were found in the marine sediments of all the studied sites (Figure 2). Considering all replicates, the number of microplastics varied from 0—52 particles per 5 g of sediment. The maximum number of microplastics counted was observed along the drop—off of Passe—à—Colas channel (site S1b) and high values were also observed in the lagoon of the Grand Cul—de—Sac Marin (GCSM, site S1a), Anse Dupuy (site S4) and Îlet Cochon (S6). The sites containing the lowest amount of microplastics were Caye à Dupont (site S7a) and the lagoon of Petit—Bourg (site S7c).

The rank of the mean number of microplastics was significantly different among sites (K—W, $X^2_{11} = 29.8$, $p = 0.0017$). A post—hoc multiple comparison test indicated that differences ($p < 0.05$) in the rank of the mean number of microplastics by site were only observed between sites S7c and S4 and between sites S7c and S1b (i.e., sites with the lowest number of microplastics and those with the highest number of microplastics).

Concentration of adsorbed chlordecone

The high values of the dry weights of filters are due to the presence of mineral residues on filters. The concentra-

TABLE 1. *Mass (m) of chlordecone (in µg) per µg of microplastic collected on the 47 mm diameter Whatman glass filters (0.7µm pore size), calculated from LC-MS-MS data. CLD: chlordecone, MP—microplastics, M—concentraiton*

Sample	m_{CLD} (µg) per filter	m_{MP} (µg) per filter	M_{CLD} (µg/µg of MP)
Site S7a	0.00131	0.76000	0.00173
Site S7b	0.00030	0.84000	0.00036
Site S7c	0.00044	0.35000	0.00124

tion of chlordecone adsorbed (in µg) per µg of microplastic (M_{CLD}; µg/µg of microplastics) collected on the filters were 0.00036—0.00173 µg/µg of microfilter (Table 1).

Discussion

The results of the present study highlight the presence of microplastics in the sediments of all the studied sites. This result shows the omnipresence of microplastics along the coasts of Guadeloupe Island, as has been demonstrated in other regions including Florida (Graham and Thompson 2009) and in Europe (Claessens et al. 2011).

The location of sites S1a, S1b, and S1c can partially explain the different concentrations of microplastics found in marine sediment. Site S1a is located in the lagoon of the GCSM Bay and is protected from marine swell by a long reef barrier except during climatic events like hurricanes or in the case of strong northern swell. On the other hand, the proximity of Site S1a to the Rivière Salée, an inlet that separates the 2 islands of Guadeloupe (Grande Terre and Basse Terre) and that cross over the urbanized area of Pointe—à—Pitre, could explain its high plastic content. Indeed, a study on the currents of Guadeloupe indicated that the water in the Rivière Salée follows an alternating rhythm depending on the tide, but also mentioned that under usual conditions, waters from the Rivière Salée flow south to north (towards the GCSM Bay; Egis Eau 2012). In addition to currents, the tidal prism exchange of the lagoon waters is low, which would allow the accumulation of microplastics at the surface of the sediment in this part of the bay (Egis Eau 2012).

Site S1b, where sediments exhibited the highest concentrations of microplastics, is north of land and urban discharges, and is located along a channel and therefore more exposed to water movements. Furthermore, the high contamination at S1b by microplastics could be due to inputs from outside the island but also from an influence of the Riviere Salée. In this channel, water flows in both directions (north to south and south to north depending on the season, the tide, and the wind (Castaing et al. 1984, Pujos et al. 1992).

The high values of microplastics at sites S4 and S6 could be linked to local anthropogenic activities. Site S4 (Anse Dupuy) is located near a fishing harbor and a nautical work area with activities involving the use of synthetic resins, which could be found in the surrounding environment. Site S6 (Ilet Cochon) is the closest to the center of the island and the most important cities, which can be a source of microplastics discharge (e.g., use of PET water bottles) combined with poor waste management on the island. For these 2 sites, the contamination, therefore, appears to come directly from the island. In contrast, the low concentrations of microplastics found at Petit—Bourg (sites S7a and S7c) could be explained by their exposure to ocean swell and the predominant winds ("Alizées") that blow from east to west for most of the year that push water towards the coast. These conditions would therefore not be favorable for the retention of microplastics by the sediments. Overall, the concentration of microplastics differed by geographic location as exposure to marine swell or proximity to urbanized or harbor areas appears to play a role in the number of microplastics present in marine sediments. However, further studies are needed to confirm these potential links between human activities and the concentration of microplastics, including a more detailed study of the current patterns and the identification of the chemical nature of plastics would make it possible to trace their origin.

The present study documents the capacity of chlordecone to adsorb on microplastics, as it was detected at all 3 sites examined. Significant adsorption of chlordecone onto microplastics reflects its high affinity with plastic as noted with other organic pollutants. However, this adsorption depends on different parameters such as the polymer type, which could impact the adsorption affinity. Indeed, Cavani and Trifiro (1995) showed high adsorption rates of PAHs and PCBs on polystyrene. Daugherty (2016) showed that polypropylene sorbed more pollutants than polyethylene; however, the sorption forces were weaker and thus the pollutants could be more easily bioavailable to organisms. Bakir et al. (2012) found that PVC and High—Density Polyethylene (HDPE) could adsorb different concentration of Dichloro—diphenyltrichloroethane (DDTs) or Phenanthrenes (Phe). The type of contaminant also plays a role in the adsorption rate. Frias et al. (2010) studied adsorbed POPs on plastic samples and found a high concentration of adsorbed PCB relating to others categories of POPs. Organic pollutants also exhibit a competitive sorption rate. For example, Bakir et al. (2012) demonstrated that DDT competitively interfered with the sorption of Phe. Finally, the size and age of plastics are also important; older, more degradaed plastics have a higher adsorption capacity (Endo et al. 2005, Ogata

et al. 2009). In this study, it is not possible to quantify the differential capacity of chlordecone to be adsorbed onto different types of plastic because chlordecone was estimated on the entire sample (all categories of microplastics combined). An FT—IR analysis would be necessary to determine the characteristics of the plastics found in these environmental samples. Thus, the results of this study should be followed up with a more comprehensive examination of chlordecone.

Chlordecone adsorbed on microplastic particles suggests the possible existence of a new pathway of transfer of the molecule into marine trophic food—webs. However, while models of accumulation and biomagnification of chlordecone in the marine environment have been studied (Coat et al. 2011, Dromard et al. 2018), none of them have considered this potential pathway of contamination. The competitiveness of chlordecone compared to other pollutants, such as DDT or DDE found in the waters around Guadeloupe, is also not known. Adsorption tests on microplastics of pollutant mixtures in environmental concentrations would provide information on the behavior of chlordecone and allow a better estimation of its potential impact on marine organisms.

In conclusion, microplastics were found in all the sites studied around Guadeloupe. Inputs appears to come from both local sources, due to human activities on the island, but also from more distant sources (via oceanic swell, currents and tides). This preliminary study also assessed chlordecone contamination adsorbed on microplastics from marine sediments. Similar patterns of chlordecone absorption were found in coral colonies collected at the same 12 study sites during a previous study in Guadeloupe (Rochas 2018). Further studies are needed to focus on pollutants adsorbed on microplastics distributed in the water column or those present in marine organisms to get a more global overview of the contamination. In addition, a quantitative study to determine the nature of all pollutants adsorbed on microplastics would be necessary to estimate the potential synergistic effects of these molecules. A limited number of sites have been sampled to measure chlordecone adsorption, so our results may not be generalizable to the entire contaminated area. However, since the sites were chosen for their potential high level of chlordecone contamination, our study results likely reveal the maximum mean chlordecone values that can be found on microplastics of the sediments of Guadeloupe. Finally, a FT—IR analysis combined with tests on chlordecone adsorption could give important information about the nature of plastic, their adsorption rate, and allow a better estimation of the potential impact on marine wildlife.

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