



Elevated Levels of Anthropogenic Chemical Pollutants in Coastal Ecosystems of the Caribbean Netherlands:

A Case Study of Bonaire

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Contents

Contents	3
Summary	5
1 Introduction	7
1.1 Study Objectives	8
2 Methods	9
2.1 Study approach and area	9
2.2 Sample collection	10
2.2.1 Passive sampling	10
2.2.2 Sediment sampling	10
2.3 Chemical analysis and data processing	11
2.4 Status assessment criteria	12
3 Results and discussion	13
3.1 Passive sampler concentrations during the dry season	13
3.2 Sediment concentrations during the rainy season	14
3.2.1 Sediment characteristics	14
3.2.2 Polychlorinated biphenyls (PCBs)	15
3.2.3 Polycyclic aromatic hydrocarbons (PAHs)	16
3.2.4 Organochlorine pesticides (OCPs)	20
3.2.5 Brominated diphenyl ethers (BDEs)	20
3.2.6 Per- and polyfluoroalkyl substances (PFASs)	20
3.2.7 Metals	21
3.3 Dietary exposure pathways	24
3.3.1 Sea Turtle-Relevant Dietary Exposure Pathways	24
3.3.2 Application of sea turtle-specific limits within Risk Assessment	24
4 Summary and conclusion	27
5 Policy Recommendations	29
5.1 Establish a long-term coastal water quality monitoring program	29
5.2 Develop and implement local thresholds for sensitive species	29
5.3 Identify and regulate point source pollution	29
5.4 Enhance collaboration between local and national institutions	30
6 Quality Assurance	31
References	32
Justification	37
Annex Supplementary information (SI)	38
SI-1 Map and sample location of BOPEC of the study by De Zwart et al. (2013)	38
SI-1.1 Environmental conditions: total precipitation data	38
SI-1.2 Research locations and data	39

SI-2	Sampler sample results	40
SI-3	Sediment sample results	42
SI-4	Materials and Methods of Chemical Analysis.	46
	SI-4.1 Chemicals and standard solutions	46
	SI-4.2 Method	47

Summary

This study presents a baseline assessment of coastal contamination around Bonaire, combining sediment analysis, passive water sampling, and seagrass bioindicator data.

Sampling locations of this case study comprise potential point sources such as BOPEC, an oil storage and transfer facility; the Marina, a highly urbanized area; Lagun, a semi-enclosed bay located near a waste disposal site; and two reference dive sites (Captain Don's Reef and Red Slave) (Figure 1).

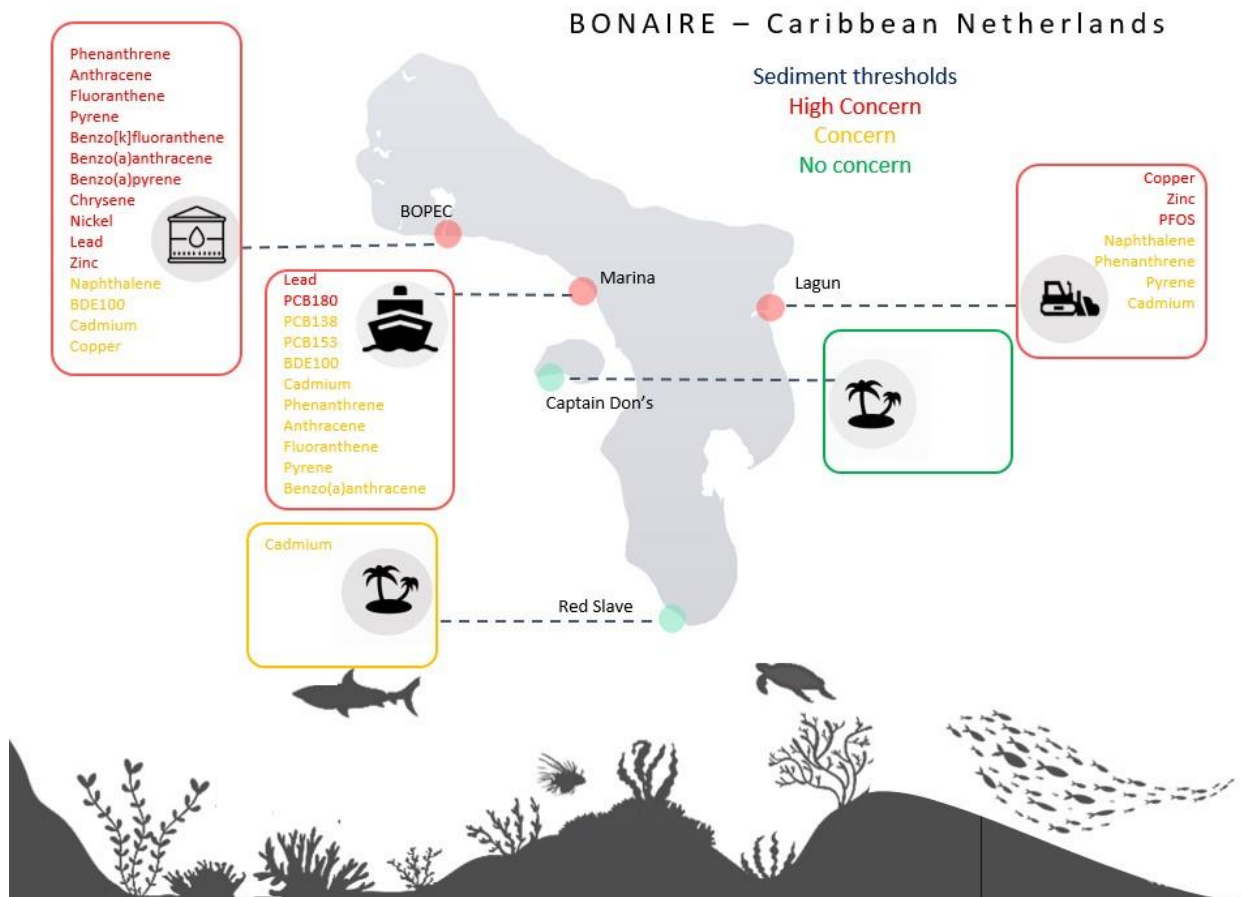


Figure 1 - Concentrations of chemicals in sediment samples collected monthly (November 2022–January 2023) in Bonaire, indicating exceedances of sediment quality thresholds (OSPAR, US EPA, RIVM, and the Canadian Environmental Protection Act, 1999). Only compounds with established threshold values are shown. Chemicals highlighted in orange exceed Background Assessment Concentrations (BACs), indicating concentrations above natural background levels in European waters. Chemicals shown in red exceed effect-based thresholds, above which adverse impacts on aquatic life are likely to occur. PCB = Polychlorinated Biphenyls; BDE = Brominated Diphenyl Ethers, PFOS=Perfluorooctanesulfonic acid.

Results confirmed widespread contamination with the highest levels detected at the three anthropogenically influenced sites (BOPEC, Lagun, and the Marina), in contrast, the reference sites, Red Slave and Captain Don's Reef, showed comparatively low or no contaminant levels, respectively (Figure 1).

- At **BOPEC**, polycyclic aromatic hydrocarbons (PAHs)—compounds commonly linked to petroleum emissions in marine sediments and known to be highly toxic to coral reefs—were consistently detected in marine sediments at concentrations exceeding established ecological thresholds from multiple organizations. These include the Effects Range-Low (ERL) set by the United States Environmental Protection Agency (US EPA) and adopted by the Oslo-Paris Convention (OSPAR) for the protection of the marine environment of the North-East Atlantic. Additionally, concentrations surpassed the Maximum Permissible Concentrations (MPC) set by the Dutch National Institute for Public Health and the Environment (RIVM) for marine sediments. Elevated levels of nickel, zinc, and lead were also recorded. These contaminants are most likely linked to oil spills, infrastructure leakage, or other legacy activities associated with the former oil storage facility.
- At **Lagun**, a foraging ground for green sea turtles (*Chelonia mydas*), the highest perfluorooctane sulfonate (PFOS), zinc, and copper concentrations were observed. PFOS concentrations exceeded RIVM secondary poisoning thresholds in marine sediments, posing potential risks to aquatic species and food web dynamics. Copper, a metal associated with the development of fibropapillomatosis, a disease characterized by tumor-like growths on the skin, within the mouth, or in the eyes of sea turtles, was also detected at this site. Notably, this disease has been most frequently reported at Lagun (Sea Turtle Conservation Bonaire, STCB, *personal communication*). Cadmium concentrations in seagrass leaves also exceeded known toxicity thresholds for green turtles and have previously been linked to adverse impacts on seagrasses in regions such as Australia. These contaminants are likely derived from surface runoff from the adjacent waste disposal site and landfill.
- At the **Marina** in Bonaire, sediment concentrations of polychlorinated biphenyls (PCBs) exceeded threshold values set under the Canadian Environmental Protection Act (1999) and applied by OSPAR during the wet season. PCBs—oily compounds formerly used in industrial applications such as electrical insulation, lubricants, flame retardants, and paint additives—are carcinogenic and known to cause endocrine disruption, neurotoxicity, and immunotoxicity in aquatic organisms. Lead levels in sediments also exceeded OSPAR environmental thresholds and are of particular concern due to their ability to impair coral reproduction and reduce symbiont density. During the dry season, both PCBs and polycyclic aromatic hydrocarbons were detected in passive samplers, indicating persistent contamination throughout the year. The Marina is situated in a densely urbanized area with intense boat traffic, factors that likely contribute to local pollutant inputs. In addition, PCBs may also originate from wastewater discharges in this highly developed zone. Given that the seagrass close to the Marina also serve as a foraging area for sea turtles, there is increased concern about ecological exposure. The bioaccumulative and biomagnifying properties of PCBs raise potential risks to marine wildlife and human health via trophic transfer. Further research is necessary to pinpoint contaminant sources and to better assess the ecological and toxicological risks posed by these pollutant groups.

Overall, contaminants identified in this study have been linked to adverse outcomes in corals (e.g., bleaching, reduced settlement), seagrasses (e.g., loss of shoot density), mangroves (e.g., impaired fertilization), and sea turtles (e.g., immunosuppression, fibropapillomatosis). Multiple hazardous chemicals at concentrations exceeding background levels indicate anthropogenic input. It raises concerns about the long-term resilience of ecosystems, particularly given the already declining condition of coral reefs and the increasing prevalence of coastal development.

In conclusion, this study highlights the urgent need for a structured, long-term coastal water quality monitoring program in the Caribbean Netherlands. In particular, biological sampling is crucial for a better understanding of exposure pathways and their effects across the food web. Without targeted action, continued pollution threatens to erode biodiversity, to jeopardize human health, and to undermine the ecosystem services that Bonaire's communities rely upon.

1 Introduction

Bonaire's coral reefs have experienced an estimated 80% decline in reef-building Scleractinian coral cover since the 1970s (de Bakker et al., 2019a). In recent years, this decline has been further exacerbated by increased frequency of coral bleaching events and outbreaks of coral disease, putting additional stress on already vulnerable reef ecosystems (Eckrich, 2024; Pepe et al., 2025). Research shows that reefs exposed to high levels of land-based runoff display reduced coral cover, increased vulnerability to disease and algal overgrowth, and lower coral diversity, which can lead to cascading effects on marine food webs and overall ecosystem stability (de Bakker et al., 2019b; Fabricius, 2005; Ku'ulei et al., 2015; Wenger et al., 2016).

Land-based emissions of anthropogenic chemicals with toxic properties can significantly degrade coastal water quality (Nalley et al., 2021). Maintaining concentrations of these contaminants below harmful levels is crucial for preserving the health of aquatic life and protecting communities that depend on these ecosystems (Nalley et al., 2021). Impacted waters may have profound ecological, economic, and social consequences (Hajji & Lucas, 2024; Wakwella et al., 2023).

To mitigate the risks of these impacts, the European Union has established regulatory frameworks that specify thresholds for chemical concentrations and ecological conditions, aiming to achieve a good water quality status. The Water Framework Directive (WFD), for example, mandates regular monitoring to ensure pollutant levels decrease and remain within safe limits for aquatic environments (Commission, 2008a). The Netherlands conducts extensive chemical monitoring under the Oslo-Paris Convention (OSPAR) and the Marine Strategy Framework Directive (MSFD) to protect marine ecosystems (Commission, 2008b).

Despite these efforts in the continental Netherlands, chemical pollution is yet a poorly understood and largely overlooked threat in the Caribbean Netherlands, comprising Bonaire, St. Eustatius, and Saba, where no structural, long-term coastal water quality monitoring is currently in place. Sustainable management of hazardous chemicals and waste is critical on small islands, where limited infrastructure increases vulnerability to pollution.

Addressing these challenges requires close collaboration among scientists, policymakers, and local communities. In 2021, Wageningen Marine Research (WMR) launched the Resilience and Restoration of Nature and Society project, which is funded by the Dutch Ministry of Agriculture, Fisheries, Food Security and Nature (LNVN). This project focuses on the islands of Bonaire, St. Eustatius, and Saba, special municipalities of the Netherlands. This project combines scientific research, policy development, and engagement with local and national stakeholders to strengthen ecosystem resilience and management capacity (Dogruer et al., 2021).

The project has laid the groundwork for identifying water quality indicators to support long-term monitoring, evaluate the effectiveness of mitigation measures, and guide restoration efforts. Monitoring includes, next to chemical pollutants, parameters such as temperature, salinity, turbidity, chlorophyll-a, inorganic nutrients, algal isotope signatures, and coral cover, extending into mesophotic reefs. The project also applies environmental DNA (eDNA), metabolomics, remote sensing, AI-assisted coral assessment tools, digital elevation models, and updated bathymetry to enhance the spatial understanding of processes influencing the marine ecosystems (Dogruer et al., 2021). Additionally, data is shared with a Deltares partner project to support hydrological and water quality modelling.

This report presents a preliminary assessment of chemical pollutants in sediments, a key exposure route for benthic habitat-forming species, such as corals and seagrasses, as well as species that rely on these benthic habitats, including sea turtles.

1.1 Study Objectives

This study aims to evaluate the presence, distribution, and ecological relevance of anthropogenic contaminants in Bonaire's coastal environment by integrating multiple sampling approaches. It provides first insights into the overarching question: Do chemical contaminants pose a risk to aquatic ecosystems in Bonaire?

Specific objectives are:

1. To determine the bioavailable fraction of contaminants in the water column during the dry season, using passive samplers (semi-permeable membrane devices) deployed in May and June 2022. This method captures the freely dissolved portion of contaminants in water, offering a proxy for chemical exposure levels experienced by aquatic organisms.
2. To assess contaminant concentrations in coastal sediments during the rainy season, when terrestrial runoff is expected to increase pollutant input. Sediment samples were collected monthly from November 2022 to January 2023 to identify concentrations that may exceed ecological thresholds.
3. To explore potential dietary exposure pathways by incorporating seagrass contaminant data from a previous study conducted in Lagun. This approach aims to assess potential ingestion risks for herbivorous marine species by comparing the seagrass contaminant results for Lagun with similar assessments from regions such as Moreton Bay, Australia.

Together, these objectives aim to provide a preliminary understanding of the exposure routes of contaminants and to inform future long-term monitoring and risk assessment efforts in Bonaire's coastal ecosystems.

2 Methods

2.1 Study approach and area

Passive samplers were selected as a proxy for chemical exposure levels experienced by aquatic organisms. Sediments were chosen because they act as both a sink and a source of chemical contaminants, representing a relevant exposure route for benthic species. Passive samplers were deployed at the three study sites, and sediment samples were collected at the same sites, as well as at two reference sites.

Sampling locations of this case study included potential point sources including BOPEC, an oil storage and transfer facility; the Marina, a highly urbanized harbour area; Lagun, a semi-enclosed bay located near a waste disposal site; and two reference dive sites in further proximity to point sources like Captain Don's Reef and Red Slave (Figure 2).

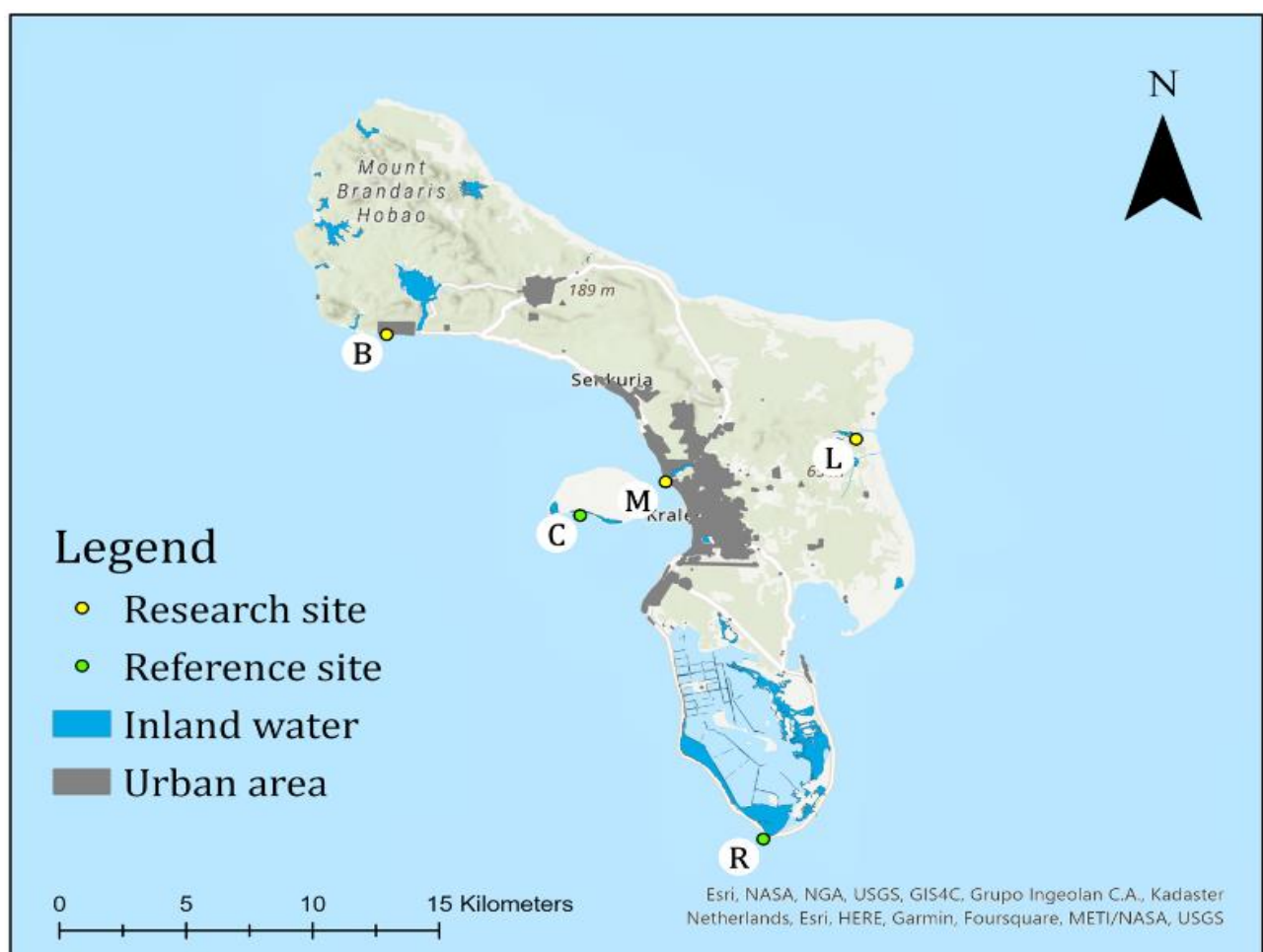


Figure 2 - Locations of the sample sites; BOPEC (B), Marina (M), Lagun (L) in yellow for passive sampling and sediment sampling as main research sites. Captain Don's Reef (C) and Red Slave (R) in green for sediment sampling as reference sites.

The reference sites are chosen due to their distance from human activity and, consequently, their distance from potential anthropogenic point sources of pollutants. Both reference sites were located near sites identified as having favourable reef health according to the Reef Health Index (RHI) model used for the Bonaire coral reef (Debrot et al., 2018). Captain Don's Reef is a dive site located on the coast of Klein Bonaire, a nature reserve with no permanent structures for human use. Captain Dons Reef is one of the diving locations farthest away from the urban areas of Bonaire, situated in a small bay. The second reference site is the dive site of

Red Slave, located at the island's southernmost tip. Only bordered by a small strip of undeveloped land and inland salt pans, human activity is relatively low. Furthermore, the predominant water current flows from south to north (van der Geest et al., 2020).

Of particular concern were substances known to exert toxic effects on marine life, including per- and polyfluoroalkyl substances (PFASs), polycyclic aromatic hydrocarbons (PAHs), organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), brominated diphenyl ethers (BDEs), and various metals. Many of these chemicals are banned or strictly regulated and are closely monitored in marine ecosystems worldwide due to their hazardous nature.

2.2 Sample collection

2.2.1 Passive sampling

The passive samplers are made from polydimethylsiloxane (PDMS) sheets. Before the passive samplers are placed at their research site in Bonaire, they are spiked by adding the performance reference compounds (PRCs) (containing a standard solution of PCB 1, 2, 3, 10, 14, 21, 30, 50, 55, 78, 104, 145 and 204) onto the sheet and placing them together in a glass jar containing deionized distilled water on a reciprocal shaker (Stuart SF1, Bibby Sterilin, Stone, UK) for six weeks to homogenise the concentration of PRCs in all passive samplers. Six passive sampler sheets are provided for each sample location, along with five blank sheets to which the PRC concentrations will be normalized.

The passive samplers at the BOPEC and Marina sites were deployed in May 2022 in a mesh cage at a depth of 10 m, in proximity to the coral reef and near the urban area. The passive sampler at Lagun was placed in shallow water at a depth of 0.5 m, close to the outflow from the landfill into Lagun, and was collected approximately six weeks later. Data on the sample location and dates are provided in SI-2.

2.2.2 Sediment sampling

Figure 2 shows the five sampling locations around Bonaire as the focus of investigation: the study sites BOPEC (B), Marina (M), and Lagun (L), and the reference locations Captain Don's Reef (C) and Red Slave (R).

At the BOPEC and Marina sites, sediment samples were collected every meter along a 10-meter transect at a depth of 10 meters, positioned near the former passive sampler locations and oriented perpendicular to the coastline. Sampling was conducted monthly from November 2022 to January 2023, resulting in three sampling events per site. At the two reference sites—Captain Don's Reef and Red Slave—sediment samples were also collected along 10-meter transects at a depth of 10 meters, perpendicular to the coast, but only once in January 2023. These 10 samples were taken strictly along the 1 m intervals, without targeting locations with visually higher organic carbon levels.

Due to the substantially shallower water at the outflow of Lagun compared to the other sites, a modified sampling approach was employed, as accessibility in this region was limited and there was a preference to minimize time spent in the water for safety reasons. Here, sediment samples were collected along a 5-meter transect, with 10 samples taken at 0.5-meter intervals, at a 2-meter distance from the former passive sampler location. As with the other sites, no distinction was made based on visually estimated organic matter content. Detailed information on sampling locations and dates is provided in SI-2.

2.3 Chemical analysis and data processing

Laboratory processing and mass spectrometry analysis were conducted at Wageningen Marine Research in the Netherlands from February to April 2023. A detailed description of the analytical methods is provided in the SI (SI-4). In brief, organic pollutants, including PAHs, PCBs, OCPs, PBDEs, and PFAS, were extracted using Soxhlet or solvent-based extraction techniques, followed by clean-up and quantification via GC-MS/MS and LC-MS/MS. All analyses were conducted in the chemical laboratory of WMR in IJmuiden, the Netherlands.

To support concentration normalization, the geochemical context and organic carbon content of the sediment samples were determined by the Royal Netherlands Institute for Sea Research (NIOZ) following the protocol of Verardo et al. (1990). Additionally, concentrations of trace metals (Cd, Cu, Hg, Ni, Pb, Zn) were analyzed by Triskelion B.V. using ICP-MS after digestion with aqua regia, following their internal protocols (WI-159 and WI-156).

All chemical concentrations in sediments were corrected for dry weight and normalized to 1% organic carbon (OC) to facilitate comparison with threshold values and sediment quality guidelines. The following equations were used:

Conversion from dry weight to wet weight:

$$C_{dw} = \frac{C_{ww}}{f_{dry}}$$

; C_{dw} is the concentration in dry weight (e.g., ng/g dw)

; C_{ww} is the concentration in wet weight (e.g., ng/g ww)

And f_{dry} is the fraction dry weight is (e.g., 80% dry matter = 0.80)

Organic carbon normalisation:

$$C_{oc} = \frac{C_{dw}}{f_{oc}}$$

; C_{dw} is the concentration in dry weight (e.g., ng/g dw)

; f_{oc} is the fraction of organic carbon (e.g., 0.2% organic carbon = 0.02)

The evaluation of passive samplers was previously described by Dogruer et al. (2025). Here, we used mathematical models to estimate the bioavailable fraction of contaminants in the water column. The samples were normalized to performance reference compounds and the field blanks to account for any possible background contamination.

2.4 Status assessment criteria

Assessment criteria according to OSPAR are included in the results [https://dome.ices.dk/OHAT/trDocuments/2023/help_ac_sediment_contaminants.html]. Concentrations significantly below the Background Assessment Concentration (BAC) are considered near the background. Effects Range Low (ERL), Environmental Assessment Criteria (EAC), and Federal Environmental Quality Guideline (FEQG) are three guideline threshold values used by OSPAR above which adverse effects are expected in marine organisms. The origin of the sources is indicated for the chemical group.

To evaluate the potential risk posed by hazardous chemical pollution to Bonaire's coastal ecosystems, sediment concentrations for PAHs and PFAS were compared to Maximum Permissible Concentrations (MPCs) derived from De Zwart et al. (2013) and Verbruggen (2012). This comparison was complemented by a previous risk assessment conducted on Bonaire's salinas, including Lake Goto, which is situated near the BOPEC study location. The evaluation, documented in the Follow-up Study on the Chemical Status of Lake Goto, Bonaire (De Zwart et al., 2013), provided valuable contextual data following a fire at the BOPEC oil storage facility. The threshold values used in our assessment were derived from these reports and normalized to 1% organic carbon (OC), utilizing the standard Dutch conversion from 10% organic matter (OM). Differences between the two limits presented in the reports are discussed in the RIVM report Janssen et al. (2004).

3 Results and discussion

3.1 Passive sampler concentrations during the dry season

Polydimethylsiloxane (PDMS), a silicone-based material housed in a protective cage, was used as a passive sampler to accumulate hydrophobic organic compounds from the aquatic environment over extended periods (Rusina et al., 2010). This approach enables time-integrated measurements of pollutant concentrations, reducing reliance on frequent grab sampling and better mimicking biological uptake (Dogruer et al., 2025).

Table 1 presents the concentrations of selected chemicals detected using PDMS passive samplers deployed at three coastal locations on Bonaire: BOPEC (an oil storage facility), the Marina (urbanised area with high boat traffic), and Lagun (adjacent to a waste processing site). These values were extrapolated from concentrations in the PDMS material (pg/g sampler) to estimated freely dissolved concentrations in water (pg/L), following the method described in Dogruer et al. (2025).

Table 1 - Estimated freely dissolved water concentrations (pg/L) derived from PDMS passive samplers., including polybrominated diphenyl ethers (BDEs), polychlorinated biphenyls (PCBs), and polycyclic aromatic hydrocarbons (PAHs)

Group	Compound (in pg/L)	BOPEC	Marina	Lagun
BDEs	BDE 100	1.3	<1.4	<0.61
	BDE 119	2.0	<1.4	<0.61
	BDE 75	2.8	<0.66	<0.38
PCBs	PCB 28	0.47	2.6	0.62
	PCB 31	<2.6	6.1	<3.3
	PCB 47	5.6	26	17
	PCB 49	<2.4	7.1	4.9
PAHs	Acenaphtene	<120	140	<90
	Chrysene	24	<3.9	<2.4
	Naphtalene	-	480	<510
	Pyrene	66	21	<10

The concentrations detected in the passive samplers represent the biologically available fraction of contaminants in the water column, those that are freely dissolved and readily available for uptake by aquatic organisms. However, comparing these concentrations directly to regulatory water quality thresholds requires caution, as most environmental standards are based on total water concentrations, including dissolved and particulate-bound components. Since passive samplers only reflect the dissolved fraction, measured values may underestimate the total environmental burden. On the other hand, they may better represent ecological risk, especially for hydrophobic compounds known to bioaccumulate via the waterborne pathway.

Among the BDEs, BDE-75, BDE-100, and BDE-119 were detected at BOPEC above detection limits, indicating a localized pollution source. These compounds may be leaching from equipment, building materials, or insulation treated with BDEs at the BOPEC facility as synthetic flame retardants.

The sum of detected PCBs reached 41.80 pg/L at the Marina, 22.52 pg/L at BOPEC, and 6.07 pg/L at Lagun. All of these values exceed the California Ocean Plan's 30-day average discharge limit of 19 pg/L (defined as the sum of Aroclors (Zeng et al. (2002)).

For comparison, a study from San Diego Bay (SDB)—one of the most urbanized coastal regions in the United States reported PCB concentrations ranging from non-detectable to 420 pg/L across multiple stations (Zeng

et al., 2002). While the levels detected in the current study during the dry season did not reach the upper concentrations reported in SDB, they nonetheless fall within the range of contamination concentrations observed in heavily urbanized areas, such as San Diego Bay.

PAHs were also detected at BOPEC and the Marina. Their presence during the dry season in Bonaire suggests the existence of chronic, year-round sources, such as ship exhaust, vehicular emissions, and potential industrial discharges. These findings challenge the assumption that pollution is introduced primarily during heavy rainfall, instead highlighting ongoing and unmanaged chemical inputs at all sites.

These findings point to the ongoing presence of legacy pollutants in the coastal zones of Bonaire, highlighting the persistent nature of PCBs and the need for continued monitoring and mitigation efforts.

Overall, this data demonstrates that passive samplers are valuable tools for obtaining initial pollution profiles, particularly in data-poor or remote environments. While no official threshold values currently exist for the bioavailable (dissolved) fraction of many contaminants in the water column, passive sampling offers critical insights for both known and emerging contaminants, such as UV filters and personal care products, as recently demonstrated in Lac Bay, and legacy compounds like PCBs and PAHs (from this study).

3.2 Sediment concentrations during the rainy season

3.2.1 Sediment characteristics

Dry weight analysis for each sample location and sampling month is provided in the SI-4. The other characteristics are measured per location, thereby combining the sediments from different months equally and homogenizing them into a single sample (Table 2).

Table 2 - Results of the sediment characteristics *C_{total}*= total carbon, *C_{org}*= organic carbon *N_{total}*= total nitrogen, *N_{org}*= organic nitrogen per study location, all expressed as percentages of the total dry weight.

Location	C _{total}	C _{org}	N _{total}	N _{org}	C _{org} /N _{total} ratio (n=2)
BOPEC	10.978	0.164	0.024	0.021	7.528
Marina	11.192	0.181	0.022	0.023	8.759
Lagun	6.574	0.502	0.049	0.053	11.060
Captain Dons	11.479	0.227	0.037	0.029	6.689
Red Slave	11.464	0.203	0.032	0.028	6.883

Sediment data indicate that the highest organic carbon content (*C_{org}*), total nitrogen content (*N_{total}*), and *C_{org}*/*N_{total}* ratio were detected in Lagun. Lagun is a large catchment area and shallow, semi-enclosed bay located on the island's east side. It is characterized by a limited presence of coral reefs and restricted water exchange. The area is surrounded by abundant mangrove vegetation and experiences episodic influxes of *Sargassum* spp., which may enhance the accumulation and preservation of organic matter (Van der Geest et al., 2024). These environmental conditions, including anoxia driven by *Sargassum* decay, likely reduce microbial degradation and promote the burial of organic-rich material (Chávez et al., 2020)). Lagun also shows the lowest total carbon (*C_{total}*) content among the sites, suggesting limited carbonate input and/or active dissolution of calcium carbonate (CaCO₃) in the sediment, a process enhanced under anoxic, low-pH conditions. This aligns with findings from studies in the Red Sea, which show that mangrove and seagrass-dominated sediments can act as net sites of carbonate dissolution due to ecosystem metabolism, especially in areas with low reef-derived carbonate input (Baldry et al., 2020).

In contrast, all other sites (Captain Dons, Red Slave, BOPEC, and Marina) are located on the island's west side and are characterized by coral reef-dominated environments, with calcareous sediment inputs. The two reference sites display lower *C_{org}*/*N_{total}* ratios along with higher nitrogen levels. In sediments, nitrogen is commonly associated with proteins and other labile organic compounds; thus, elevated nitrogen levels suggest

fresher, less decomposed organic inputs. Aligned with the selection as reference sites, results in lower C/N ratios at Captain Dons and Red Slave, indicative of more autochthonous (marine-derived) organic matter rather than more resistant terrestrial inputs (Lamb et al., 2006).

Consequently, the observed gradient (Captain Dons < Red Slave < BOPEC < Marina < Lagun) aligns with expected variation in organic matter sources and preservation conditions across the island’s distinct reef and non-reef habitats. The high carbonate input and calcification rates associated with the reef-dominated western sites further explain their higher inorganic carbon content, as supported by regional carbon budget assessments in coral reef ecosystems (Yan et al., 2024).

3.2.2 Polychlorinated biphenyls (PCBs)

PCBs are synthetic organic compounds that were once used in electrical equipment, industrial processes, and surface coatings. Despite their ban, PCBs persist in the environment due to their stability and resistance to degradation. These compounds can contaminate coral reef ecosystems through industrial discharges, land runoff, and atmospheric transport. PCBs accumulate in various organisms, including marine life such as filter feeders, fish, and marine mammals. Their environmental impact primarily stems from bioaccumulation in organisms and biomagnification throughout the food web, which affects air-breathing organisms at the apex of the food chain (Madgett et al., 2022). PCBs have been linked to detrimental impacts on reproduction, growth, metabolism, and survival. These effects encompass hepatotoxicity and immunotoxicity (Schaap et al., 2023; Wahlang et al., 2019; Xiao et al., 2021) . PCBs have been known to bioaccumulate in coral tissues; however, information on their biological effects is limited (T.-H. Chen et al., 2012; Ranjbar Jafarabadi et al., 2018).

A detailed summary of the results, including the sum of the PCBs, is provided in SI-2. At Marina, 11 of the 31 screened PCBs were present in the sediment samples collected in November 2022 (Table 3). PCB 180 was also detected in the sample from Marina collected in January 2023, with a concentration of 0.13 ng/g dw, which is above the BAC but below the EAC (see SI-2). Hence, PCB concentration in marine sediment decreases from high to low-participation periods (see SI-1.1). The sum of PCBs measured in Marina is 19.49 ng/g dw, for example, higher than the concentrations detected in Puerto Rico’s sediments, with a mean of 8 ng/g dw (%TOC ranging from 1 to 1.4) (Anthony S. Pait et al., 2007).

Table 3 - Results of the GC-MS/MS analysis of PCBs in sediment for the Marina site, expressed as ng/g dry weight, normalized to 1.0% organic carbon (OC) content. Only PCBs that were measured above the limit of detection (LOD) are shown. BAC= Background assessment criteria were developed within the Oslo and Paris Commission framework with scientific advice from the International Council for the Exploration of the Sea. Mean concentrations significantly below the BAC are said to be near background, and FEQGS were developed under the Canadian Environmental Protection Act, 1999 applied by OSPAR.

PCB	Marina-November (2022).	BAC	FEQGS
HCBd	0.007		
PCB 138	0.65	0.06	3.2
PCB 141	0.43		
PCB 149	1.44		
PCB 151	0.72		
PCB 153	2.16	0.076	16
PCB 170	1.44		
PCB 180	5.75	0.04	4.8
PCB 187	3.59		
PCB 194	2.16		
PCB 202	0.43		
PCB 206	0.72		

At the Marina in Kralendijk, the capital's port, various types of boats, ranging from small fishing vessels to luxury yachts, are docked, and they depart from here for the open sea. Furthermore, the urban areas surrounding the marina accommodate residential, industrial, and tourist-related activities associated with the detected chemicals. Consequently, the potential ecological impacts of PCB pollutants on coral reefs could manifest as a top-down disruption of the ecosystem's balance (van Dam et al., 2011). Furthermore, PCBs are carcinogenic and are known to induce endocrine disruption, neurotoxicity, and immunotoxicity in aquatic animals (Kandjo Ngoubeyou et al., 2022). Considering these locations are known for marine megafauna such as sea turtles to forage, a follow-up investigation, including biological sampling such as fish or sea turtle tissues, is highly warranted. The findings indicate risks to aquatic life, coral reef ecosystems, and potential human exposure in the area.

3.2.3 Polycyclic aromatic hydrocarbons (PAHs)

PAHs compounds from incomplete fossil fuel combustion, industrial processes, natural origins and are frequently present in petroleum products, vehicle emissions, and industrial effluents (De Zwart et al., 2013). They infiltrate coastal environments through oil spills, industrial releases, and atmospheric deposition, accumulating in sediments and the tissues of marine organisms, including corals (Sabourin et al., 2012). Existing research links PAH exposure to coral bleaching and diminished growth rates (Jafarabadi et al., 2021).

The sediment concentrations of 16 PAHs normalized to 1.0% organic carbon (OC) were analysed across multiple coastal sites in Bonaire. Figure 3 and Figure 4 present the PAH concentrations in comparison with established environmental thresholds: the Maximum Permissible Concentrations (MPCs) from RIVM, Effects Range Low (ERLs) from the US EPA applied by OSPAR, and the Background Assessment Concentrations (BACs) established under OSPAR. All concentration data are provided in the SI-3.

Out of the 16 PAHs analysed, only 12 had applicable environmental threshold values (MPC, ERL, or BAC). The two compounds, acenaphthylene and acenaphthene, were below the detection limit at all sites and excluded from further evaluation. Five PAHs (pyrene, fluoranthene, chrysene, benzo[a]anthracene, and phenanthrene) exceeded both the MPC and ERL thresholds during two consecutive sampling months (November and December 2022) at BOPEC, indicating these compounds as the most concerning. Two additional compounds, benzo[b]fluoranthene and benzo[a]pyrene, exceeded either the MPC or ERL thresholds in both November and December at BOPEC. Anthracene and benzo[k]fluoranthene exceeded MPC thresholds in December only, further supporting site-specific spikes in PAH levels.

Several PAHs, including naphthalene, fluorene, phenanthrene, pyrene, fluoranthene, and chrysene, exceeded the BACs established by OSPAR, primarily at BOPEC and occasionally at Lagun and Marina sites. These exceedances suggest the presence of diffuse contamination that may still fall below ecotoxicological thresholds but is notably above natural background levels. While no regulatory thresholds exist for benzo[g,h,i]perylene, dibenz[a,h]anthracene, and indeno[1,2,3-cd]pyrene, these were consistently detected at elevated levels at BOPEC.

BOPEC is situated on the island's northwest side, away from residential areas and near the Washington Slagbaai National Park. Adjacent to BOPEC is the outlet of Lake Goto, the largest saltwater (saliña) watershed on Bonaire and part of the National Park, into which the surrounding areas' run-off is collected before it flows into the sea. The elevated levels of PAHs are associated with the activities at the oil storage facility BOPEC.

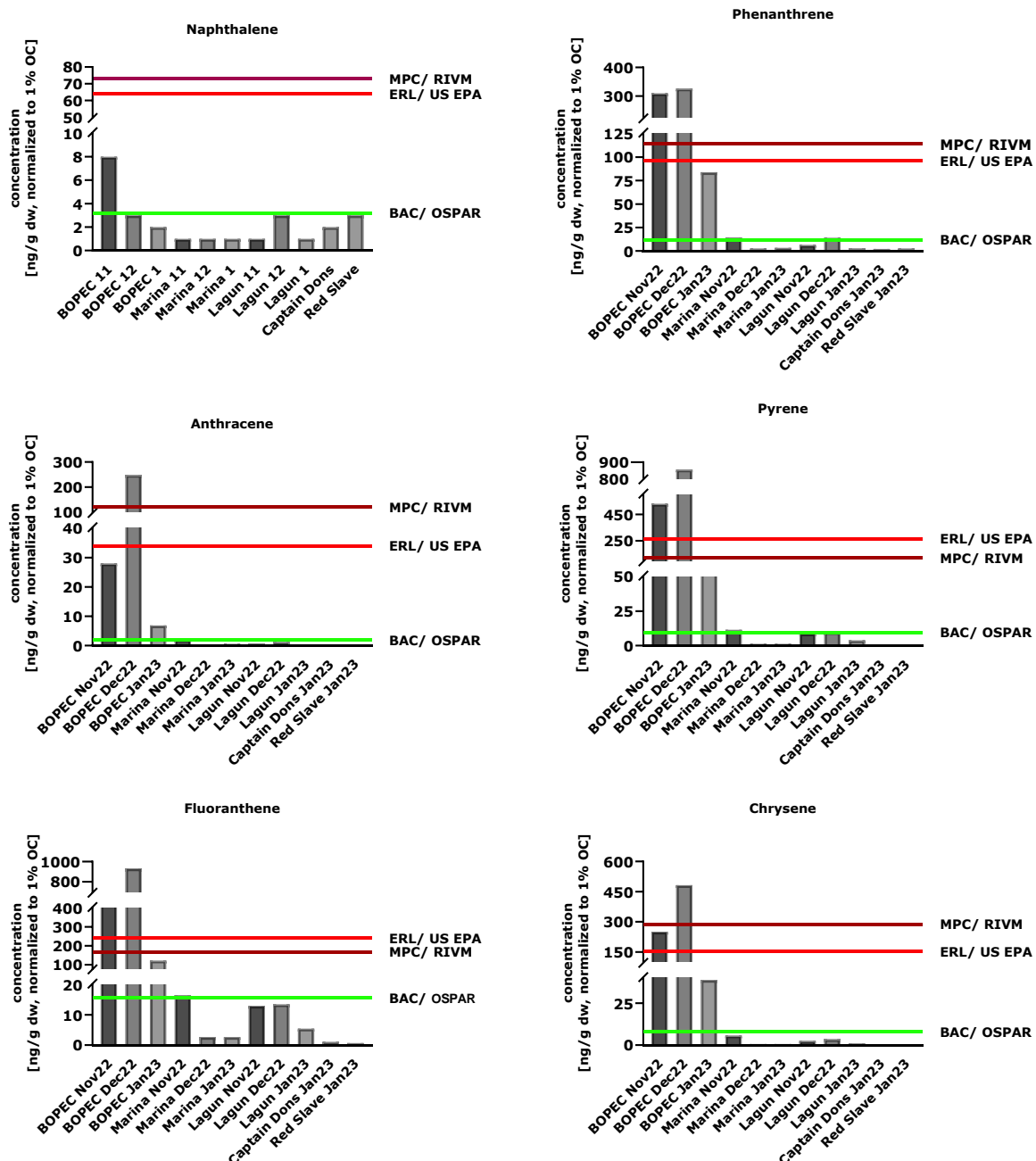


Figure 3 - Concentrations of polycyclic aromatic hydrocarbons (PAHs) in marine sediments from five sites and multiple sampling months, analyzed via GC-MS/MS and expressed in ng/g dry weight, normalized to 1.0% organic carbon (OC). Thresholds include Effects Range-Low (ERL) values from the US EPA, adopted by OSPAR (red), Background Assessment Concentrations (BACs) established by OSPAR in collaboration with ICES (green), and Maximum Permissible Concentrations (MPCs) for marine sediment derived by the Dutch RIVM (red). All thresholds were normalized to 1.0% OC for comparison.

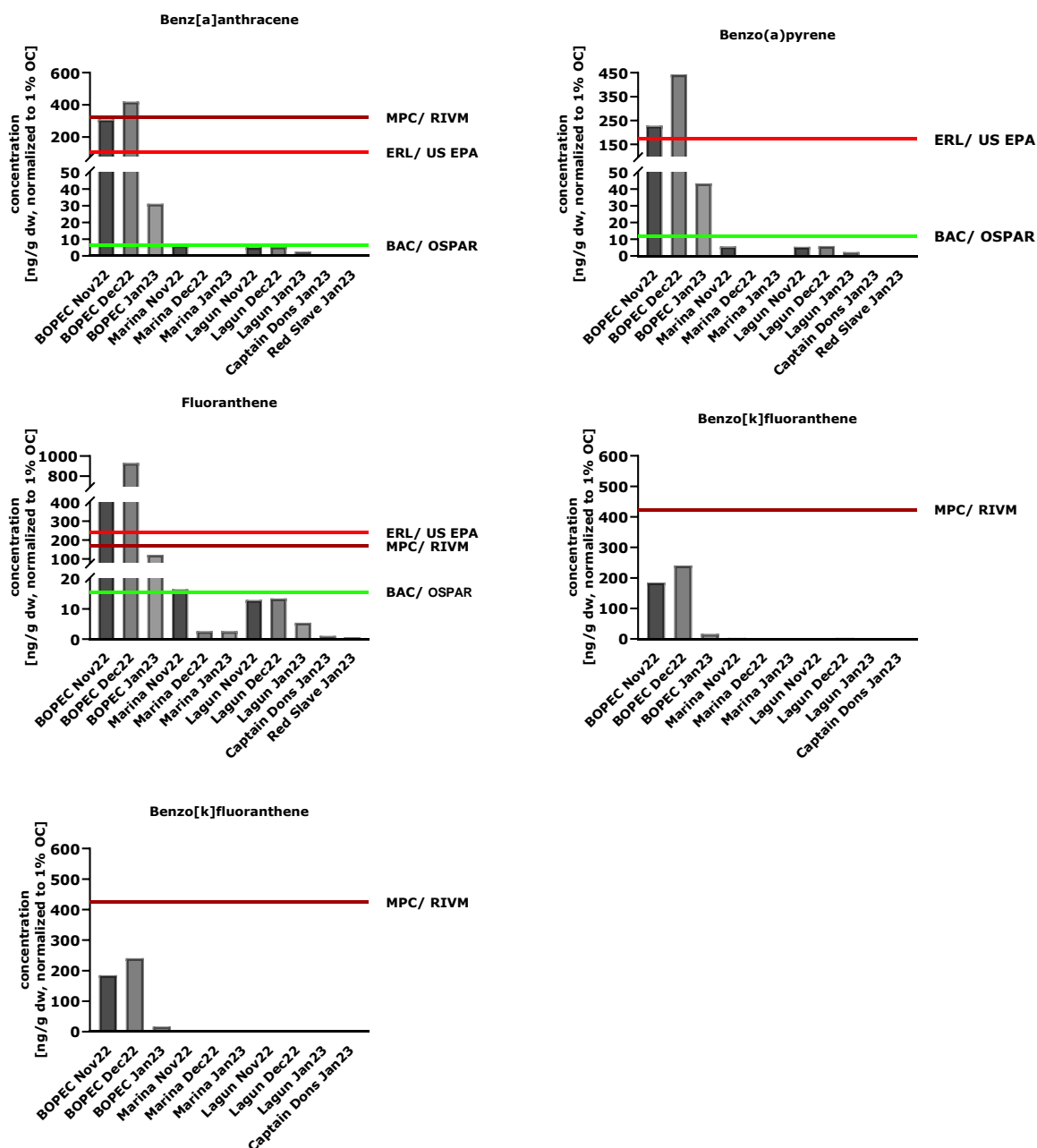


Figure 4 - Concentrations of polycyclic aromatic hydrocarbons (PAHs) in marine sediments from five sites and multiple sampling months were analyzed via GC-MS/MS and expressed in ng/g dry weight, normalized to 1.0% organic carbon (OC). Thresholds include Effects Range-Low (ERL) values from the US EPA, adopted by OSPAR (red), Background Assessment Concentrations (BACs) established by OSPAR in collaboration with ICES (green), and Maximum Permissible Concentrations (MPCs) for marine sediment derived by the Dutch RIVM (red). All thresholds were normalized to 1.0% OC for comparison.

In 2010, a petrochemical fire broke out at the BOPEC facility, releasing polycyclic aromatic hydrocarbons (PAHs) into the environment. To extinguish the fire, different per- and polyfluoroalkyl substances (PFASs) were used (De Zwart et al., 2013).

The data from the current study were compared with the historical data gathered after a fire at the BOPEC facility in 2012 (De Zwart, 2012, Location 9). Although these samples were collected in Lake Goto, a salinia inland water body with distinct physicochemical properties (e.g., salinity, hydrodynamics, and organic content) compared to our coastal marine sampling sites, they serve as a valuable reference. The 2012 and current samples were analysed by the same laboratory (formerly known as IMARES, now Wageningen Marine Research).

Figure 5 presents a time series comparison of all PAHs detected above the detection limit in both datasets. The results reveal that concentrations in BOPEC sediments (2022–2023) are multiple times higher than in Lake Goto in 2012, across nearly all PAH compounds assessed. These findings suggest either a continued or renewed input of PAHs in the area or the persistence of chronic contamination more than a decade after the incident. This comparison highlights the importance of repeated monitoring, especially after industrial accidents, to assess long-term environmental impacts more accurately.

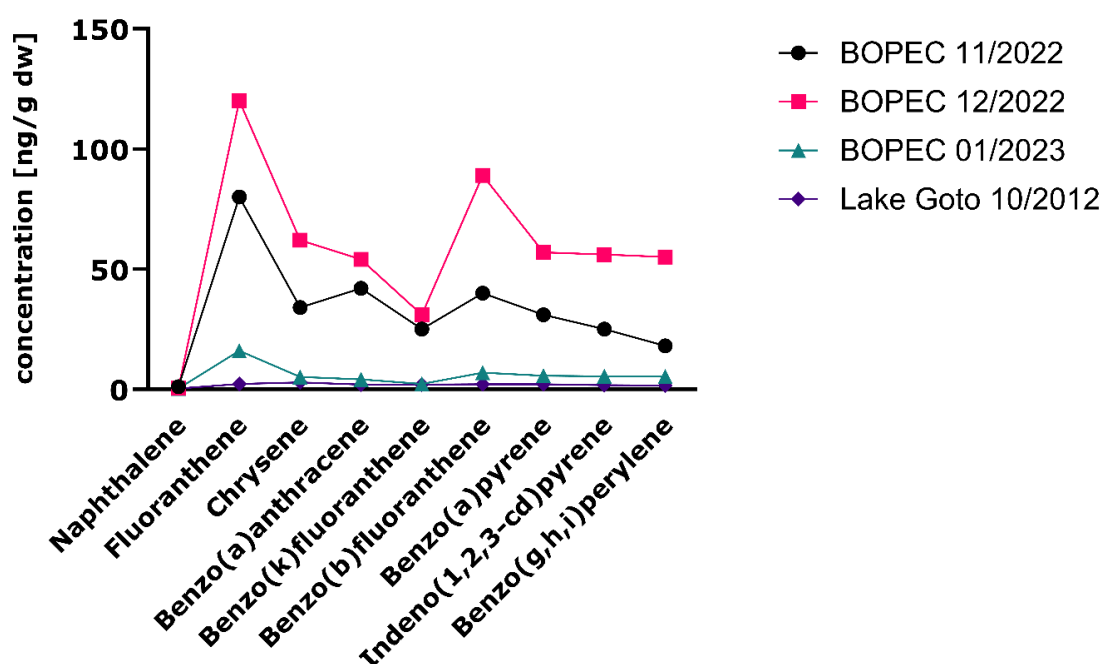


Figure 5 - Time series comparison of selected PAH concentrations (ng/g dry weight) in BOPEC sediment samples from November 2022 to January 2023, and historical data from Lake Goto (Location 9 in De Zwart, 2012, sampled in October 2012), analyzed by the same laboratory (formerly IMARES). Concentrations in BOPEC are consistently elevated, indicating persistent contamination and localized inputs.

PAHs are well-documented for their toxicity in marine ecosystems. Benzo[a]pyrene, for example, is known to induce oxidative stress in coral larvae (Farina et al., 2008), the most vulnerable stage in coral's life cycle. Furthermore, these compounds can have phototoxic effects on marine invertebrates (Bellas et al., 2008). They can disrupt reproduction and development in fish and crustaceans (Khan, 2013; Khan et al., 2023). Although vertebrates, such as fish, can metabolize PAHs, the metabolites formed can be potent carcinogens, posing a significant threat to exposed fish. Additionally, it is worth noting that coral reef environments tend to accumulate PAHs in biota more than in sediments, as previously demonstrated in Hainan and the Persian Gulf (Jafarabadi et al., 2021; Ranjbar Jafarabadi et al., 2018; Y. Li et al., 2021). These studies reveal higher PAH burdens in coral tissue than in adjacent sediments, suggesting that the biological impact at BOPEC may be even more severe than the sediment concentrations indicate alone. considering that existing research links PAH exposure to coral bleaching and reduced growth rates (Jafarabadi et al., 2021) and that thermal stress and bleaching are currently on the rise (Eckrich, 2024), further research is strongly warranted.

Overall, the stark contrast between BOPEC and background locations highlights strong spatial heterogeneity and emphasizes the need for targeted mitigation and long-term monitoring. The results raise concerns about benthic organisms and coral reef health and potential human exposure pathways in the region.

3.2.4 Organochlorine pesticides (OCPs)

OCPs are suspected of bioaccumulating in coral tissues and negatively impacting the algal symbiotic relationship (Kang et al., 2022; Markey et al., 2007). In the present study, the measured OCPs were not detected above the limit of detection (LOD) in the sediment samples.

3.2.5 Brominated diphenyl ethers (BDEs)

BDEs are flame retardants commonly used in consumer products, electronics, and textiles. These compounds can enter the marine environment by leaching from products and through the disposal of waste. In the present study, only one BDE congener, BDE-100, was quantified above the limit of detection (LOD). It was detected at the BOPEC site in January (0.061 ng/g dw) and at the Marina site in both December (0.067 ng/g dw) and January (0.074 ng/g dw) (Table 4). All other BDE congeners were below the LOD. A detailed summary of the results is provided in SI-2. The concentrations found in this study were relatively low, around 0.06 ng/g dw, compared to values reported in the literature. These concentrations are also near the Background Assessment Concentration (BAC), indicating a lower environmental risk. However, the chemicals were detected in the water column at BOPEC and Marina, implying an unmanaged input of these substances at those locations.

Table 4 - Results of the GC-MS/MS analysis of BDEs for the Lagun and Marina sites, expressed as ng/g dry weight, normalized to a 1.0% organic carbon (OC) content. Only BDEs that were measured above the limit of detection (LOD) are shown. BACs= Background assessment criteria were developed within the Oslo and Paris Commission framework with scientific advice from the International Council for the Exploration of the Sea. Mean concentrations significantly below the BAC are said to be near background and FEQGs were developed under the Canadian Environmental Protection Act, 1999.

Site	Sampling Date	BDE100 (ng/g 1%OC)	BAC (1% OC)	FEQG(1% OC)
BOPEC	January 2023	0.061	0.02	0.40
Marina	December 2022	0.067	0.02	0.40
Marina	January 2023	0.074	0.02	0.40

3.2.6 Per- and polyfluoroalkyl substances (PFASs)

PFAS, including PFOS, are synthetic chemicals extensively used in industrial processes and consumer products such as firefighting foams, non-stick coatings, and water-resistant textiles (Arvaniti & Stasinakis, 2015) . Due to their persistence, mobility, and bioaccumulative nature, PFASs have been detected in diverse environmental compartments, including marine sediments, surface waters, and biota (Jonker, 2024; Kissa, 2001; van den Heuvel-Greve & Foekema, 2024).

OSPAR does not currently provide threshold values for PFOS or its congeners in marine sediments. However, the Dutch National Institute for Public Health and the Environment (RIVM) has established environmental risk thresholds for PFOS in marine sediments, as outlined in De Zwart et al. (2013) . These include a Maximum Permissible Concentration for ecotoxicological effects (MPC eco) of 10 ng/g dry weight and a Maximum Permissible Concentration for secondary poisoning (MPC sp) of 3.2 ng/g dry weight, based on sediment with 10% organic matter. When normalized to 1% organic carbon (OC), the standard used in this study, these thresholds correspond to approximately 1.69 ng/g dry weight (dw) (MPC eco) and 0.54 ng/g dw (MPC sp).

Among all sampled locations, PFOS was detected only in sediment at Lagun, with a concentration of 6.92 ng/g dry weight (1% organic carbon, OC). Elevated levels of PFAS in solid surrounding waste incinerator have been reported elsewhere (Martin et al., 2023). At Lagun, the level exceeds RIVM-derived thresholds, indicating

potential ecotoxicological and food-web-related risks. Given that Lagun (December 2022) is a foraging habitat for green turtles (*Chelonia mydas*), this finding warrants further investigation, particularly into the accumulation of contaminants in local biota.

Emerging research indicates that PFOS may impact coral reef ecosystems by inducing oxidative stress, reducing photosynthetic activity, and disrupting lipid metabolism in symbiotic organisms (Bednarz et al., 2022). Recent studies have also raised concerns about the impact of PFOS on marine organisms, including sea turtles. For instance, research on green turtles (*Chelonia mydas*) in the North Pacific detected PFOS in their plasma, with concentrations correlating to human population densities, suggesting local sources of contamination. A risk assessment from this study indicated potential immunosuppressive effects in green turtles from areas with higher PFOS concentrations (Wood et al., 2021). Furthermore, PFOS exposure has been linked to developmental toxicity in hawksbill turtles (*Eretmochelys imbricata*), with maternal transfer of PFASs to eggs observed, which may potentially affect hatchling success and development (Wood et al., 2021). While direct studies on the impacts of PFOS specifically on green turtles are limited, the existing evidence indicates the need for further investigation into how PFOS and related compounds may affect the health and survival of these marine reptiles.

3.2.7 Metals

Metals are widely present in aquatic environments, originating from both natural sources (e.g., volcanic activity, forest fires) and anthropogenic activities, including mining, fuel combustion, industrial operations, and maritime infrastructure such as ports (Conrad et al., 2021). These elements can enter marine systems via land-based runoff during rainfall events, contributing to ecological degradation. While some metals, such as zinc, copper, and nickel, are essential micronutrients involved in physiological functions, others, including cadmium, mercury, and lead, are non-essential and toxic even at low concentrations, potentially impairing growth, immune function, and reproductive success in marine organisms (Tchounwou et al., 2012). Notably, many metals persist in sediments, bioaccumulate in tissues, and biomagnify (mercury) through aquatic food webs (Walker et al., 2005).

Metal concentrations in this study were normalized to 1.0% organic carbon (OC) content and compared against OSPAR BAC (Background Assessment Concentration) and ERL (Effects Range Low) thresholds. Figure 6 visualizes the results for BOPEC, Marina, Lagun, and reference sites Captain Don's Reef and Red Slave. OSPAR thresholds were adjusted from their original 2.5% OC reference.

Although normalization to aluminium (Al) is typically preferred in the OSPAR framework due to its lithogenic and conservative nature, this study adopted organic carbon (OC) normalization as an established alternative used in sediment geochemistry and marine pollution assessments (C.-F. Chen et al., 2024; Gao & Chen, 2012; Hosokawa et al., 2020; Li et al., 2021). While TOC-based normalization is not applied under OSPAR, it is considered scientifically valid. However, in volcanic terrains like Bonaire, iron (Fe) and titanium (Ti) might be more appropriate lithogenic normalizers. Incorporating these elements in future studies can enhance the accuracy of trace metal assessments and aid in distinguishing between natural geogenic levels and anthropogenic enrichment.

All mercury concentrations in sediment samples from Bonaire were below the limit of detection (LOD) and are therefore not shown. However, as mercury biomagnifies through the food web, low sediment levels do not necessarily indicate low concentrations in marine organisms. The only known study assessing mercury in biota from Bonaire dates back to 2013 and reported elevated mercury levels in mahi-mahi, wahoo, and blackfin tuna, exceeding the current OSPAR threshold for secondary poisoning in marine fish (20 µg/kg wet weight) (Davidson et al., 2013). This highlights the need to include biological samples in future monitoring efforts to better assess ecological and human health risks.

3.2.7.1 Cadmium

Cadmium was detected at all locations above BAC, except for Captain Don's Reef, our least impacted reference site (Figure 6). Although levels remained below the ERL, these thresholds are based on temperate species and may underestimate risk to tropical coral reef systems. Similar cadmium concentrations have been reported near urbanized coasts such as the South China Sea (300 ng/g), Bohai and Yellow Seas (200 ng/g), and the Suez Gulf (70–110 ng/g) (El Nemr et al., 2016; Li et al., 2021; Yang et al., 2021). Experimental studies suggest that cadmium can induce oxidative stress and tissue degradation in coral species (Mitchellmore et al., 2007).

3.2.7.2 Copper

Copper exceeded the ERL at Lagun in November and January, and exceeded the BAC at BOPEC in December and January (Figure 6). Lagun is situated near a landfill and a mangrove-seagrass ecosystem, serving as a foraging ground for green turtles (*Chelonia mydas*). These turtles exhibit high site fidelity and are ideal bioindicators of chemical pollution, often ingesting contaminated sediment during grazing. Elevated copper levels raise concerns given that fibropapillomatosis, a tumor-forming disease in turtles, has been reported at Lagun (SCTB, personal communication). Similar studies in Brazil have linked elevated copper exposure to fibropapillomatosis outbreaks in green turtles (da Silva et al., 2016).

Toxicity studies show that copper inhibits fertilization, larval settlement, and coral survival at concentrations as low as 22.6–101 µg/L (Nalley et al., 2021). A laboratory study on the mangrove species *Avicennia marina*, closely related to Bonaire's *Avicennia germinans*, found negative impacts at a concentration of 100,000 ng/g sediment (MacFarlane & Burchett, 2002). Additionally, coral larvae exposed to sediment containing 72,000 ng/g of copper showed inhibited settlement after just 36 hours (Negri et al., 2002). As all these studies indicate a lesser likelihood of acute toxicity, with values ranging from 13,037 to 25,752 ng/g at Lagun, chronic toxicity to mangroves and coral reefs cannot be excluded.

3.2.7.3 Zinc

Zinc concentrations exceeded the ERL at BOPEC in every month, and at Lagun in November and January. December values at Lagun were slightly below the ERL (13,037 ng/g vs. ERL 13,700 ng/g), but still exceeded the BAC (10,800 ng/g) (Figure 6).

Mean zinc concentrations were 116 times higher at BOPEC, 39 times higher at Lagun, and 8 times higher at Marina than at the reference sites. In *Acropora tenuis*, zinc impaired fertilization at just 10 µg/L after 5 hours (Reichelt-Brushett & Harrison, 2005), and larval metamorphosis was affected at sediment concentrations of 92,000 ng/g (Negri et al., 2002) a threshold exceeded in Lagun (up to 853,330 ng/g) and all months at BOPEC.

3.2.7.4 Nickel

Nickel exceeded the ERL at BOPEC in November and December (Figure 6). Nickel is a known trace metal in crude oil (Metwally et al., 1997). High sediment concentrations have been linked to chronic oil exposure in areas such as the Red Sea (Ali et al., 2011). Although BOPEC ceased operations in 2011, ongoing land-based runoff or leaching may still contribute to seasonal increases.

Nickel can reduce coral symbiont density and growth at concentrations of 3.5 µg/L, especially when combined with thermal stress (Biscéré et al., 2017, 2018). Uptake studies show significant nickel accumulation in coral tissue after only 7 days of sediment exposure (Gillmore et al., 2020).

3.2.7.5 Lead

Lead concentrations exceeded the ERL at BOPEC (December) and Marina (January), and the BAC at BOPEC in November (Figure 6). Levels ranged from 11,338 to 19,207 ng/g at BOPEC and from 9,884 to 20,744 ng/g at Marina, comparable to those in heavily industrialized marine systems, such as the South China Sea (4,180–58,700 ng/g; Zhu et al. (2010)). As lead has no biological function and is toxic at low levels, its presence is particularly concerning. Its accumulation may lead to disruptions in neurological and metabolic processes in reef species and other wildlife. Lead can reduce coral fertilization success and symbiont density (Nalley et al., 2021).

Overall, all BOPEC, Marina, and Lagun samples showed higher metal concentrations than the reference sites. December showed peak concentrations at BOPEC across all metals. Marina showed a slight downward trend, except for a peak in lead in January. These patterns and ecological vulnerabilities (e.g., coral reefs, seagrass beds, mangroves, and turtle foraging habitats) indicate significant environmental risks at impacted sites. The presence of multiple metals above guideline thresholds, often at levels comparable to those near industrial coastlines, underscores the urgent need for local monitoring, source control, and studies on ecological impact.

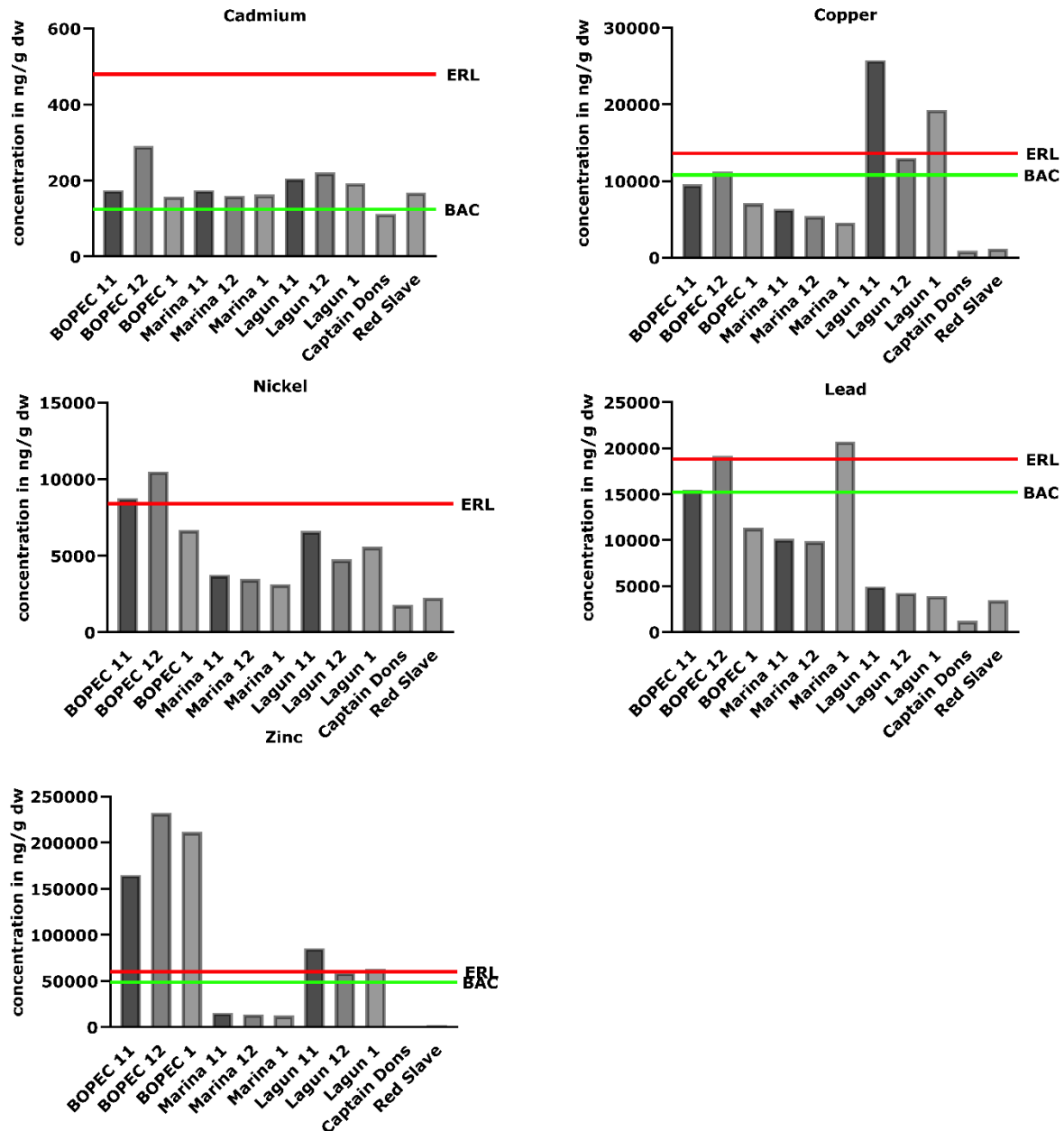


Figure 6 - Results of the ICP-MS analysis of metals for the five sites at different sample months, expressed in ng/g dry weight and normalized to a 1.0% organic carbon content. OSPAR ERL and BAC threshold values were provided and normalized from 2.5% to 1.0% OC.

3.3 Dietary exposure pathways

3.3.1 Sea Turtle-Relevant Dietary Exposure Pathways

Green sea turtles (*Chelonia mydas*), due to their herbivorous diet and close association with seagrass meadows and underlying sediments, are particularly susceptible to contaminant exposure (Dogruer et al., 2018; Dogruer et al., 2021; Gaus et al., 2012; Villa et al., 2017, 2019). Studies have demonstrated that green turtles can accumulate higher internal cadmium concentrations than many higher trophic-level marine species, such as dolphins, underscoring their role as sentinels of sediment-associated contamination (Fraga et al., 2018).

3.3.1.1 Benchmark Seagrass

To better understand species-specific exposure risks, Dogruer et al. (2021) developed and validated a mechanistic model simulating lifetime chronic cadmium exposure in green sea turtles inhabiting coastal Australian ecosystems. These data were used to establish quantitative relationships between internal cadmium burdens and blood-based biomarkers of toxicological effect. The model was based on a cohort study in which blood and cellular samples were collected from wild turtles and analyzed for their associations with environmental contaminant (Villa et al., 2019).

By applying the model in reverse, the authors estimated cadmium concentrations in dietary items, specifically seagrass, that would lead to internal concentrations just below the toxicity threshold. This led to deriving a maximum permissible concentration (MPC_{seagrass}) of 0.1 µg/g dry weight cadmium in seagrass. Recent elemental analysis of *Thalassia testudinum* leaf tissues collected from 3 sites in Lagun, Bonaire (March 2022), revealed mean cadmium concentrations of 3.27 µg/g dry weight (Ouwensloot, 2022). Notably, previous ecological studies have linked cadmium concentrations in this range to decreased rhizome density and reduced seagrass resilience (Fraser & Kendrick, 2017), indicating that this benchmark may serve a dual function by protecting both turtle health and habitat integrity.

3.3.1.2 Benchmark Sediment

To estimate corresponding cadmium concentrations in sediments, data from a long-term study conducted in North Queensland (Dogruer, 2022; Thomas et al., 2020) were used. This study reported cadmium concentrations in both seagrass and adjacent sediments. An estimated value was derived based on the observed ratios between seagrass and sediment cadmium concentrations (bioaccumulation factors) (Dogruer thesis). Sites with differing organic matter content were normalized to 1% total organic carbon (TOC). Based on measured TOC values in Bonaire sediments (approximately 0.074%), cadmium concentrations were adjusted, resulting in a TOC-normalized sediment threshold of approximately 81 ng/g dry weight at 1% TOC. Cadmium is associated with organic carbon in sediments but also binds to other geochemical fractions, including sulfides, carbonates, and oxides. Thus, while TOC normalization provides a practical comparison tool, a more comprehensive risk assessment should include additional sediment characteristics such as grain size, redox conditions, sulfide content, speciation, and lithogenic markers like aluminum (Al).

Overall, the lack of species-specific thresholds in tropical regions often results from logistical, ethical, and socio-political constraints; however, these gaps can be increasingly addressed through mechanistic modelling and targeted field data collection as well as by establishing species-specific bioassays.

3.3.2 Application of sea turtle-specific limits within risk assessment

3.3.2.1 Seagrass

In this study, we also used previously published data by Ouwensloot (2022) on trace metal concentrations in apical shoots of the seagrass species *Thalassia testudinum*, collected in Lagun, to assess potential ingestion risks for herbivorous marine species foraging on seagrass in Lagun. A description of the methodology used for analyzing trace metal concentrations in the apical shoots of *T. testudinum* can be found in Ouwensloot (2022). The risk-quotient (RQ) based risk assessment applies the sea turtle-specific seagrass cadmium limits of 100

ng/g dry weight as the predicted no-effect concentration (PNEC) and uses the measured concentration in seagrass (Cseagrass) within the following equation:

$$RQ = \frac{C_{seagrass}}{PNEC};$$

Cadmium concentrations in seagrass (*Thalassia testudinum*) were normalized against this threshold, such that a value above 1 indicates a level of concern, suggesting that ingestion of seagrass at those concentrations may result in adverse health effects in green turtles. Notably, the concentrations in Lagun surpass those reported in more urbanized and industrialized foraging habitats in Japan and Australia, where historically the highest cadmium levels have been recorded in green turtles (Pople et al., 1998).

Compared to these regions, cadmium concentrations in seagrass tissues from Bonaire's Lagun Bay are highly elevated, indicating that this Caribbean site also falls within the high-risk category and warrants further investigation and monitoring.

Table 5 - The risk-quotient(RQ)-based evaluation of the risk, as described in Dogruer et al. 2021. Seagrass concentrations are evaluated against the species-specific threshold for green turtles in Australia, the United States, Brazil, and Japan. Concentration in seagrass was normalised to the proposed threshold values. An RQ > 1 typically signals a potential risk and the need for further investigation or mitigation.

Location	Measured Concentration (ng/g dw)	PNEC (ng/g dw)	RQ
Japan	900	100	9.0
Australia	330	100	3.3
Brazil	140	100	1.4
United States	65	100	0.65
Bonaire (Lagun)	3270	100	32.7

3.3.2.2 Sediment

Using the proposed threshold of 81 ng/g dry weight in sediment concentrations, all study locations in Bonaire exceed the protective limit, with the lowest concentrations found at the site at Captain Don's, which is below the BAC limit by OSPAR (Figure 7). This is a notable finding, as Captain Don's is located off Klein Bonaire, an area known to support intensive green turtle nesting and foraging activity. The relatively low contaminant levels observed at this site may reflect more favourable environmental conditions for sea turtle health. Although sediment cadmium concentrations at the other sampling sites remain below the ERL, the data indicate that bioaccumulation in primary producers, such as seagrass, can lead to dietary exposures of concern for herbivorous species, like green turtles.

Overall, the results indicate a significant risk of chronic toxicological effects for green turtles in Lagun, particularly where both cadmium and copper levels are elevated. Continued monitoring of internal cadmium burdens in turtle tissues, investigation of trophic transfer dynamics, and broader assessments of sublethal and ecosystem-level impacts are highly needed to protect vulnerable turtle populations and the coastal habitats they depend on.

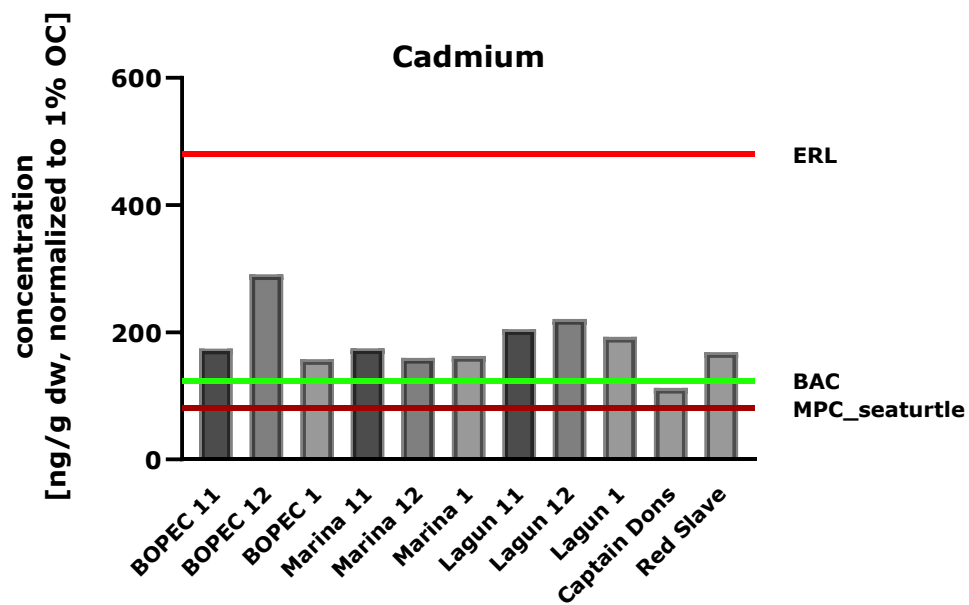


Figure 7 – Results for cadmium in sediment, including the extrapolated sea turtle-specific exposure limits from this study

4 Summary and conclusion

This study presents the preliminary findings of chemical assessment of coastal contamination around Bonaire, combining sediment analysis, passive water sampling, and seagrass bioindicator data. The findings provide compelling evidence of widespread contamination across these multiple matrices, particularly at three anthropogenically influenced sites: BOPEC, the Marina, and Lagun. Together, these locations exhibit elevated levels of persistent organic pollutants (POPs), toxic trace metals, and emerging contaminants, posing potential ecological risks to key marine habitats, including coral reefs, seagrass beds, mangroves, and areas where turtles forage. In contrast, reference sites (Captain Don's Reef and Red Slave) showed comparatively lower contaminant levels.

Detecting chemical concentrations in Bonaire's coastal sediments exceeding European Background Assessment Concentrations (BACs) is a noteworthy finding. BACs represent the expected baseline levels in minimally impacted environments and are used as a reference to distinguish between the natural background and anthropogenic inputs. Exceedance of these thresholds on a small island like Bonaire, often regarded as relatively pristine, suggests the presence of persistent local pollution sources, such as port activity, oil storage, vessel emissions, submarine groundwater discharge, or untreated waste water and surface runoff. It also raises concerns about the effectiveness of current waste management and regulatory oversight in safeguarding marine ecosystems. Furthermore, several compounds detected in this study not only exceeded BACs but also surpassed effect-based thresholds, such as the Effects Range-Low (ERL) values adopted by the US EPA and OSPAR or the Maximum Permissible Concentrations (MPC) established by the Dutch RIVM. These exceedances indicate that contaminant levels are high enough to pose ecological risks to aquatic life. While BAC exceedance alone does not confirm harm, the combination of background and effect-based threshold exceedances indicates the urgency for further investigation and potential mitigation.

Key outcomes:

- At BOPEC, polycyclic aromatic hydrocarbons (PAHs)—compounds often linked to petroleum emissions in marine sediments and known to be highly toxic to coral reefs—were consistently detected in marine sediments at concentrations exceeding established ecological thresholds from multiple organisations. These include the Effects Range-Low (ERL) set by the United States Environmental Protection Agency (US EPA) and adopted by the Oslo-Paris Convention (OSPAR) for the protection of the marine environment of the North-East Atlantic. Additionally, concentrations surpassed the Maximum Permissible Concentrations (MPC) set by the Dutch National Institute for Public Health and the Environment (RIVM) for marine sediments. Elevated levels of nickel, zinc, and lead were also recorded. These contaminants are most likely linked to oil spills, infrastructure leakage, or other legacy activities associated with the former oil storage facility.
- At Lagun, a foraging ground for green sea turtles (*Chelonia mydas*), the highest perfluorooctane sulfonate (PFOS), zinc, and copper concentrations were observed. PFOS concentrations exceeded RIVM secondary poisoning thresholds, posing potential risks to marine species and food web dynamics. Copper, a metal associated with the development of fibropapillomatosis, a disease characterized by tumor-like growths on the skin, within the mouth, or in the eyes of sea turtles, was also detected at this site. Notably, this disease has been most frequently reported at Lagun (Sea Turtle Conservation Bonaire, STCB, personal communication). Cadmium concentrations in seagrass leaves also exceeded known toxicity thresholds for green turtles and have previously been linked to adverse impacts on seagrasses in regions such as Australia. These contaminants are likely derived from surface runoff from the adjacent waste disposal site and landfill.
- At the Marina, polychlorinated biphenyls (PCBs) sediment concentrations exceeded the threshold developed under the Canadian Environmental Protection Act (1999) and applied by OSPAR during the wet season. PCBs, oily substances with various industrial uses like lubrication, electrical insulation, flame-retardants, and paint additives, are carcinogenic and are known to induce endocrine disruption,

neurotoxicity, and immunotoxicity in aquatic animals. Lead can reduce coral fertilization success and symbiont density. During the dry season, both PCBs and PAHs were detected in passive samplers, highlighting persistent contamination throughout the year. The Marina is situated in a densely urbanized area with intense boat traffic, factors that likely contribute to local pollutant inputs. In addition, PCBs may also originate from wastewater discharges in this highly developed zone. Given that the seagrass close to the Marina also serve as a foraging area for sea turtles, there is increased concern about ecological exposure. The bioaccumulative and biomagnifying properties of PCBs raise potential risks to marine wildlife and human health via trophic transfer. Further research is necessary to pinpoint contaminant sources and to better assess the ecological and toxicological risks posed by these pollutant groups.

The ecotoxicological implications are significant. Contaminants identified in this study have been linked to adverse outcomes in corals (e.g., bleaching, reduced settlement), seagrasses (e.g., loss of shoot density), mangroves (e.g., impaired fertilization), and sea turtles (e.g., immunosuppression, fibropapillomatosis). The presence of multiple stressors across habitats raises concerns about long-term ecological resilience, particularly given the already declining state of coral reefs and increasing anthropogenic pressure on Bonaire's coasts. In conclusion, this study highlights the importance of establishing a structured coastal water quality monitoring program in the Caribbean Netherlands. Without intervention, ongoing contamination may compromise biodiversity, human health, and the ecological services provided by Bonaire's marine ecosystems.

5 Policy Recommendations

The results of this study highlight the need for urgent and sustained action to address chemical pollution in Bonaire's coastal ecosystems. Although contamination levels in the Caribbean Netherlands are often assumed to be low due to the region's relatively undeveloped character, this report indicates that localised, chronic, and potentially harmful pollution is occurring. To prevent further ecological degradation and ensure alignment with environmental management standards applied in the Netherlands, the following recommendations are proposed:

5.1 Establish a long-term coastal water quality monitoring program

It is recommended to establish a permanent, government-supported monitoring programme to track pollutant concentrations in coastal waters, sediments, and biota. This programme is advised to:

- Adopt protocols in line with OSPAR, WFD, and MSFD standards.
- Include seasonal sampling, capturing both dry and wet season dynamics.
- Integrate passive sampling technologies (e.g., PDMS) and bioindicator species, such as algae, green turtles, marine invertebrates, and marine fish.
- Standardised data reporting and contributed to national and international environmental databases.

5.2 Develop and implement local thresholds for sensitive species

Most existing environmental quality standards are based on temperate ecosystems and may underestimate risks in tropical systems. There is an urgent need to:

- Develop species-specific toxicity thresholds for vulnerable marine fauna in the Caribbean Netherlands.
- Utilize mechanistic lifetime exposure modelling to derive Maximum Permissible Concentrations (MPCs), as demonstrated in the case of green turtles and cadmium.
- Prioritise assessments for organisms with ecological, economic, or conservation significance (e.g., coral, sea turtles, reef fish).

5.3 Identify and regulate point source pollution

The study identified BOPEC, the Marina, and Lagun as hotspots for contamination.

- Map and regulate point sources
- Regulate emission rates from point sources
- Implement containment, treatment, and remediation strategies to reduce further leaching of contaminants into the marine environment.
- Implement buffer zones, enhance stormwater infrastructure, and upgrade wastewater treatment facilities in coastal urban areas.

5.4 Enhance collaboration between local and national institutions

Effective pollution management in small island systems requires coordinated governance and management.

- Strengthen collaboration among Dutch ministries, local governments, knowledge institutes (e.g., RIVM, WMR, Deltares), local NGOs, and community stakeholders to build capacity and raise awareness.
- Include environmental pollution within broader marine spatial planning and resilience strategies.
- Launch targeted outreach campaigns on land-based sources of pollution and their effects on marine life and human health.

6 Quality Assurance

Wageningen Marine Research utilises an ISO 9001:2015 certified quality management system. The organisation has been certified since 27 February 2001. The certification was issued by DNV.

The Chemical and Benthos laboratory has an EN-ISO/IEC 17025:2017 accreditation for test laboratories with number L097. This accreditation has been granted by the Dutch Accreditation Council. As a result, the Chemical and Benthos laboratory has demonstrated its ability to provide valid results in a technically competent manner and to work in accordance with the ISO17025 standard. The scope (L097) of the accredited analytical methods can be found at the website of the Council for Accreditation (www.rva.nl).

The quality of the test methods is ensured in various ways. The accuracy of the analysis is regularly assessed by participation in proficiency tests. If the quality cannot be guaranteed, appropriate measures are taken.

In addition, a first-level control is performed for each series of measurements.

If desired, information regarding the performance characteristics of the analytical methods is available.

If the quality cannot be guaranteed, appropriate measures are taken.

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Justification


Report: C019/25

Project Number: 4318100362

The scientific quality of this report has been peer reviewed by a colleague scientist and a member of the Management Team of Wageningen Marine Research

Approved: Dr. ir. M.J.J. Kotterman
Researcher


Signature:

Signed by:

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Date: 16 April 2025

Approved: Dr. A.M. Mouissie
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Signature:

Signed by:

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Date: 16 April 2025

Annex Supplementary information (SI)

SI-1 Map and sample location near BOPEC of the study by De Zwart et al. (2013)



Figure 9 – Map of the sample location near BOPEC in 2010 conducted by De Zwart et al. (2013). Values at Location 9 was used for comparison in the present study.

SI-1.1 Environmental conditions: total precipitation data

Table 6 summarizes the monthly precipitation data recorded at the Flamingo Airport weather station on Bonaire, retrieved from Tutiempo Climate. The data shows the difference between a dry and rainy season in Bonaire.

Table 6 - Monthly total precipitation in millimetres

Month/Year	Total precipitation (mm)
May/2022	3
June/2022	75
November/2022	250
December/2022	83
January/2023	51

SI-1.2 Research locations and data

Table 7 - General information about the research locations and relevant data.

Site	Latitude	Longitude	Passive sampler Deployment date	Passive sampler Recollection date	Total passive sampler deployment days	Sediment sampling date	Depth	Type sediment	of
BOPEC	12.220295	-68.387336	14/05/2022	28/06/2022	45 days	08/11/2022	10m	Clear sand	coral
						06/12/2022			
						03/01/2023			
Marina	12.162792	-68.286162	14/05/2022	28/06/2022	45 days	08/11/2022	10m	Clear sand	coral
						06/12/2022			
						03/01/2023			
Lagun	12.179162	-68.217546	15/05/2022	05/07/2022	51 days	10/11/2023	0.5m	Greyish substance and fine brown sand	
						09/12/2023			
						05/01/2023			
Captain Don's reef	12.149976	-68.317461	-	-	-	02/01/2023	10m	Clear sand	coral
Red Slave	12.026339	-68.251753	-	-	-	12/01/2023	10m	Clear sand	coral

SI-2 Sampler sample results

Table 8 - Results of the GC-MS/MS analysis for all samplers, for the three sites in ng/l.

Compound	BOPEC	Marina	Lagun
PCB31	<0.0026	0.0061	<0.0033
PCB28	0.00047	0.0026	0.00062
PCB52	<0.0025	<0.0061	<0.0034
PCB49	<0.0024	0.0071	0.0049
PCB47	0.0056	0.026	0.017
PCB66	<0.0025	<0.0061	<0.0034
PCB101	<0.0052	<0.013	<0.007
PCB56	<0.0049	<0.012	<0.0067
PCB97	<0.0025	<0.0063	<0.0035
PCB87	<0.0025	<0.0063	<0.0035
PCB85	<0.0025	<0.0063	<0.0035
PCB110	<0.0042	<0.01	<0.0059
PCB151	<0.0027	<0.0066	<0.0036
PCB149	<0.0061	<0.015	<0.0084
PCB118	<0.0068	<0.017	<0.0094
PCB153	<0.0053	<0.013	<0.0073
PCB141	<0.0026	<0.0066	<0.0036
PCB105	<0.0025	<0.0063	<0.0035
PCB137	<0.0018	<0.0043	<0.0023
PCB138	<0.0053	<0.013	<0.0073
PCB187	<0.0037	<0.0093	<0.005
PCB202	<0.0039	<0.01	<0.0055
PCB128	<0.0026	<0.0066	<0.0036
PCB156	<0.0035	<0.0089	<0.0048
PCB180	<0.0027	<0.0069	<0.0038
PCB170	<0.0037	<0.0093	<0.005
PCB194	<0.0028	<0.0072	<0.0039
PCB206	<0.0062	<0.016	<0.0085

Table 9 - Results of the GC-MS/MS analysis for samplers for the three sites in ng/l.

Compound (ng/L)	BOPEC	Marina	Lagun
Naphthalene	nb	0.48	<0.51
Acenaphthylene	<0.27	<0.21	<0.2
Acenaphthene	<0.12	0.14	<0.09
Fluorene	<0.079	<0.064	<0.061
Phenanthrene	<0.17	<0.15	<0.14
Anthracene	<0.072	<0.069	<0.056
Fluoranthene	<0.086	<0.099	<0.068
Pyrene	0.066	0.021	<0.01
Benzo(a)anthracene	<0.0096	<0.019	<0.012
Chrysene	0.024	<0.0039	<0.0024
Benzo(b)fluoranthene	<0.0082	<0.019	<0.011
Benzo(k)fluoranthene	<0.0082	<0.019	<0.011
Benzo(a)pyrene	<0.0016	<0.0038	<0.0022
Dibenz(a,h)anthracene	<0.008	<0.019	<0.011
Indeno(1,2,3-cd)pyrene	<0.008	<0.019	<0.011
Benzo(g,h,i)perylene	<0.0081	<0.019	<0.011

Table 10 - Results of the GC-MS/MS analysis for all samplers, for the three sites in ng/l.

Compound	BOPEC	Marina	Lagun
b-HEPO	<0.031	<0.068	<0.039
HCb	<0.0013	<0.0029	<0.0016
HCBD	<0.0016	<0.0027	<0.0018
Heptachloor	<0.0012	<0.0031	<0.0016
Dicofol	<0.051	<0.045	<0.031

Table 11 - Results of the GC-MS/MS analysis for all samplers, for the three sites in ng/l.

Compound	BOPEC	Marina	Lagun
BDE28	<0.00039	<0.00062	<0.00036
BDE75	0.0028	<0.00066	<0.00038
BDE49	<0.00041	<0.00066	<0.00038
BDE71	<0.00041	<0.00066	<0.00038
BDE47	<0.00068	<0.0017	<0.00096
BDE66	<0.0034	<0.0076	<0.0042
BDE100	0.0013	<0.0014	<0.00061
BDE119	0.002	<0.0014	<0.00061
BDE99	<0.00058	<0.0014	<0.00061
BDE85	<0.00058	<0.0014	<0.00061
BDE154+BB153	<0.00077	<0.0019	<0.0011
BDE153	<0.00077	<0.0019	<0.0011
HBCD			
BDE138	<0.0017	<0.0038	<0.0022
BDE183	<0.0023	<0.0052	<0.003
BDE190	<0.00097	<0.0024	<0.0014

SI-3 Sediment sample results

Table 12 - Results of the GC-MS/MS analysis for all sediment samples, for the five sites and subsequent sample months in ng/g dry weight, normalised to 1.0% Organic Carbon (OC) content.

Compound	BOPEC- Nov	BOPEC- Dec	BOPEC- Jan	Marina- Nov	Marina- Dec	Marina- Jan	Lagun- Nov	Lagun- Dec	Lagun- Jan	Captain Don's	Red Slave
PCB31	<0,148	<0,156	<0,153	<0,144	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB28	<0,148	<0,156	<0,153	<0,144	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB52	<0,148	<0,156	<0,153	<0,144	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB49	<0,148	<0,156	<0,153	<0,144	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB47	<0,148	<0,156	<0,153	<0,144	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB66	<0,148	<0,156	<0,153	<0,144	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB101	<0,295	<0,311	<0,305	<0,288	<0,267	<0,268	<0,078	<0,165	<0,104	<0,219	<0,273
PCB56	<0,295	<0,311	<0,305	<0,288	<0,267	<0,268	<0,078	<0,165	<0,104	<0,219	<0,273
PCB97	<0,148	<0,156	<0,153	<0,144	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB87	<0,148	<0,156	<0,153	<0,144	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB85	<0,148	<0,156	<0,153	<0,144	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB110	<0,221	<0,233	<0,229	<0,216	<0,201	<0,201	<0,078	<0,132	<0,078	<0,164	<0,204
PCB151	<0,148	<0,156	<0,153	0,719	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB149	<0,295	<0,311	<0,305	1438	<0,267	<0,335	<0,104	<0,198	<0,104	<0,274	<0,273
PCB118	<0,369	<0,389	<0,382	<0,359	<0,334	<0,335	<0,13	<0,231	<0,13	<0,274	<0,341
PCB153	<0,295	<0,311	<0,305	2156	<0,267	<0,268	<0,078	<0,165	<0,104	<0,219	<0,273
PCB141	<0,148	<0,156	<0,153	0,431	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB105	<0,148	<0,156	<0,153	<0,144	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB137	<0,074	<0,078	<0,076	<0,072	<0,067	<0,067	<0,026	<0,066	<0,026	<0,055	<0,068
PCB138	<0,295	<0,311	<0,305	0,647	<0,267	<0,268	<0,078	<0,165	<0,104	<0,219	<0,273
PCB187	<0,148	<0,156	<0,153	3594	<0,134	<0,201	<0,052	<0,132	<0,078	<0,164	<0,204
PCB202	<0,148	<0,233	<0,229	0,431	<0,201	<0,201	<0,052	<0,132	<0,078	<0,164	<0,204
PCB128	<0,148	<0,156	<0,153	<0,144	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB156	<0,148	<0,156	<0,153	<0,216	<0,134	<0,201	<0,052	<0,132	<0,078	<0,164	<0,204
PCB180	<0,148	<0,156	<0,153	5,75	<0,134	0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB170	<0,148	<0,156	<0,153	1438	<0,134	<0,201	<0,052	<0,132	<0,078	<0,164	<0,204

PCB194	<0,148	<0,156	<0,153	2156	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136
PCB206	<0,295	<0,311	<0,305	0,719	<0,267	<0,268	<0,078	<0,165	<0,104	<0,219	<0,273

Table 13 - Results of the GC-MS/MS analysis for all sediment samples, for the five sites and subsequent sample months in ng/g dry weight, normalised to 1.0% Organic Carbon (OC) content. The sum (Σ) of the total PAHs, low molecular weight PAHs (LPAH; containing three or less rings) and high molecular weight PAHs (HPAH; containing more than three rings).

Compound	BOPEC- Nov	BOPEC- Dec	BOPEC- Jan	Marina- Nov	Marina- Dec	Marina- Jan	Lagun- Nov	Lagun- Dec	Lagun- Jan	Captain Don's	Red Slave
Naphthalene	8,12	3,11	2,29	1,44	0,67	1,34	1,30	3,29	0,78	2,19	2,73
Acenaphtylene	<0,044	<0,047	<0,046	<0,043	<0,04	<0,04	<0,016	<0,023	<0,016	<0,033	<0,041
Acenaphtene	<0,044	<0,047	<0,046	<0,043	<0,04	<0,04	<0,016	<0,023	<0,016	<0,033	<0,041
Fluoreen	21,40	14,00	5,34	0,72	0,20	0,27	0,26	0,30	0,21	0,16	0,41
Phenantrene	309,99	326,76	83,96	14,38	2,68	3,35	6,51	14,16	2,87	2,19	2,73
Anthracene	28,05	248,96	6,87	2,16	0,47	0,60	0,78	1,32	0,26	<0,033	0,41
Fluoranthene	590,47	933,59	122,12	16,53	2,68	2,68	13,02	13,50	5,47	1,09	0,68
Pyrene	531,42	855,79	91,59	11,50	1,34	1,34	8,86	9,88	3,91	0,44	0,41
Benzo(a)anthracene	309,99	420,12	31,29	6,47	0,33	0,54	5,21	5,60	2,61	0,16	<0,041
Chrysene	250,95	482,36	38,93	5,75	0,67	0,67	2,61	3,62	1,04	0,16	0,14
Benzo(b)fluoranthene	295,23	692,41	52,66	6,47	1,34	0,67	4,95	9,55	2,87	0,06	<0,041
Benzo(k)fluoranthene	184,52	241,18	16,79	3,59	0,47	0,54	3,13	4,28	1,30	0,22	<0,041
Benzo(a)pyrene	228,81	443,46	43,51	5,75	0,60	0,60	5,47	5,93	2,35	0,11	<0,041
Dibenz(a,h)anthracene	30,26	50,57	6,11	0,72	<0,04	<0,04	0,52	0,99	0,52	<0,033	<0,041
Indeno(1,2,3-cd)pyrene	184,52	435,68	41,22	3,59	0,67	0,67	4,17	4,94	2,09	<0,033	<0,041
Benzo(g,h,i)perylene	132,86	427,90	39,69	4,31	1,34	2,01	4,43	6,59	3,91	<0,033	0,68
Σ PAH	3106,59	5575,88	582,36	83,38	13,44	15,27	61,21	83,95	30,17	6,79	8,18
Σ LPAH	367,56	592,83	98,46	18,69	4,01	5,56	8,86	19,07	4,12	4,54	6,27
Σ HPAH	2739,02	4983,05	483,90	64,69	9,43	9,71	52,35	64,88	26,06	2,24	1,91

Table 14 - Results of the GC-MS/MS analysis for all sediment samples, for the five sites and subsequent sample months in ng/g dry weight, normalised to 1.0% Organic Carbon (OC) content.

Compound	BOPEC- Nov	BOPEC- Dec	BOPEC- Jan	Marina- Nov	Marina- Dec	Marina- Jan	Lagun- Nov	Lagun- Dec	Lagun- Jan	Captain Don's	Red Slave
BDE28	<0,015	<0,016	<0,015	<0,014	<0,013	<0,013	<0,005	<0,01	<0,005	<0,011	<0,014
BDE75	<0,037	<0,039	<0,038	<0,036	<0,033	<0,033	<0,013	<0,023	<0,013	<0,027	<0,034
BDE49	<0,037	<0,039	<0,038	<0,036	<0,033	<0,033	<0,013	<0,023	<0,013	<0,027	<0,034
BDE71	<0,037	<0,039	<0,038	<0,036	<0,033	<0,033	<0,01	<0,023	<0,013	<0,027	<0,034
BDE47	<0,037	<0,039	<0,038	<0,036	<0,033	<0,033	<0,01	<0,023	<0,013	<0,027	<0,034
BDE66	<0,015	<0,016	<0,015	<0,014	<0,013	<0,013	<0,005	<0,01	<0,005	<0,011	<0,014
BDE100	<0,052	<0,054	0,061	<0,058	0,06	0,067	<0,018	<0,033	<0,021	<0,044	<0,055
BDE119	<0,052	<0,054	<0,053	<0,058	<0,053	<0,054	<0,018	<0,033	<0,021	<0,044	<0,055
BDE99	<0,052	<0,054	<0,053	<0,058	<0,053	<0,054	<0,018	<0,033	<0,021	<0,044	<0,055
BDE85	<0,052	<0,054	<0,053	<0,058	<0,047	<0,054	<0,018	<0,033	<0,021	<0,044	<0,055
BDE154+BB153	<0,03	<0,031	<0,031	<0,029	<0,027	<0,027	<0,01	<0,02	<0,01	<0,022	<0,027
BDE153	<0,03	<0,031	<0,031	<0,029	<0,027	<0,027	<0,01	<0,02	<0,01	<0,022	<0,027
HBCD	<0,148	<0,156	<0,153	<0,216	<0,134	<0,201	<0,052	<0,099	<0,078	<0,164	<0,204
BDE138	<0,074	<0,078	<0,076	<0,072	<0,067	<0,067	<0,023	<0,033	<0,026	<0,055	<0,068
BDE183	<0,074	<0,078	<0,076	<0,072	<0,067	<0,067	<0,026	<0,066	<0,026	<0,055	<0,068
BDE190	<0,074	<0,078	<0,076	<0,072	<0,067	<0,067	<0,026	<0,066	<0,026	<0,055	<0,068
QCB	<0,052	<0,054	<0,053	<0,058	<0,053	<0,054	<0,018	<0,033	<0,021	<0,044	<0,055
HCb	<0,052	<0,054	<0,053	<0,058	<0,053	<0,054	<0,018	<0,033	<0,021	<0,044	<0,055
pp-DDE	<0,074	<0,078	<0,076	<0,144	<0,134	<0,134	<0,026	<0,066	<0,052	<0,109	<0,136
pp-DDD	<0,148	<0,156	<0,153	<0,144	<0,134	<0,134	<0,052	<0,099	<0,052	<0,109	<0,136

Table 15 - Results of the LC-MS analysis for all sediment samples, for the five sites and subsequent sample months in ng/g dry weight, normalised to 1.0% Organic Carbon (OC) content.

Compound	BOPEC- Nov	BOPEC- Dec	BOPEC- Jan	Marina- Nov	Marina- Dec	Marina- Jan	Lagun- Nov	Lagun- Dec	Lagun- Jan	Captain Don's	Red Slave
PFBA	<17,714	<17,894	<20,608	<14,376	<15,381	<15,403	<2,605	<8,234	<6,514	<14,775	<17,041
PFPeA	<0,738	<0,778	<0,763	<0,719	<0,669	<0,670	<0,130	<0,329	<0,261	<0,547	<0,682
PFHxA	<2,214	<2,334	<2,290	<1,438	<2,006	<2,009	<0,260	<0,988	<0,782	<1,642	<2,045
PFHpA	<0,738	<0,778	<0,763	<0,719	<0,669	<0,670	<0,130	<0,329	<0,261	<0,547	<0,682
PFOA	<0,738	<0,778	<0,763	<0,719	<0,669	<0,670	<0,130	<0,329	<0,261	<0,547	<0,682
PFNA	<2,214	<2,334	<2,290	<1,438	<2,006	<2,009	<0,260	<0,988	<0,782	<1,642	<2,045
PFDCa	<2,214	<2,334	<2,290	<1,438	<2,006	<2,009	<0,260	<0,988	<0,782	<1,642	<2,045
PFUnA	<0,738	<0,778	<0,763	<0,719	<0,669	<0,670	<0,130	<0,329	<0,261	<0,547	<0,682
PFDoA	<0,738	<0,778	<0,763	<0,719	<0,669	<0,670	<0,130	<0,329	<0,261	<0,547	<0,682
PFTTrA	<0,738	<0,778	<0,763	<0,719	<0,669	<0,670	<0,130	<0,329	<0,261	<0,547	<0,682
PFTeA	<2,214	<2,334	<2,290	<1,438	<2,006	<2,009	<0,260	<0,988	<0,782	<1,642	<2,045
PFBS	<1,476	<1,556	<2,290	<1,438	<1,337	<1,339	<0,260	<0,988	<0,782	<1,642	<2,045
PFHxS	<0,738	<0,778	<0,763	<0,719	<0,669	<0,670	<0,104	<0,329	<0,261	<0,547	<0,682
PFHpS	<4,428	<4,668	<5,343	<3,594	<4,012	<4,018	<0,781	<1,976	<1,824	<3,830	<4,772
PFOS	<0,738	<0,778	<0,763	<0,719	<0,669	<0,670	<0,130	6,916	<0,261	<0,547	<0,682
PFDS	<0,738	<0,778	<0,763	<0,719	<0,669	<0,670	<0,130	<0,329	<0,261	<0,547	<0,682

Table 16 - Results of the ICP-MS analysis for all sediment samples, for the five sites and subsequent sample months in ng/g dry weight, normalised to 1.0% Organic Carbon (OC) content.

Compound	BOPEC- Nov	BOPEC- Dec	BOPEC- Jan	Marina- Nov	Marina- Dec	Marina- Jan	Lagun- Nov	Lagun- Dec	Lagun- Jan	Captain Don's	Red Slave
Cd	174	291	157	174	159	162	204	221	192	112	168
Cu	9580	11234	7121	6352	5431	4588	25752	13037	19302	912	1190
Hg	<18.4	<19.5	<19.1	<18.0	<16.7	<16.7	<6.5	<8.2	<6.5	<13.7	<17.0
Ni	8756	10487	6672	3739	3476	3134	6614	4765	5590	1785	2262
Pb	15455	19206	11338	10124	9884	20744	4933	4279	3892	1233	3465
Zn	165057	232164	211745	15540	13625	12744	85330	59159	62959	1413	2091

SI-4 Materials and Methods of Chemical Analysis

SI-4.1 Chemicals and standard solutions

SI-4.1.1 Chemicals

Acetone, n-hexane, methanol, n-pentane, iso-octane (2,2,4-trimethylpentane) were obtained from LGC Standards (Wesel, Germany).

Sodium sulfate, copper, and hydrochloric acid were obtained from Sigma-Aldrich (Zwijndrecht, The Netherlands). Acetonitrile was obtained from Biosolve (Dieuze, France).

Methanol was obtained from Actua-All (Oss, The Netherlands). Diethyl ether was obtained from Riedel-de Haën (Seelze, Germany). ENVicarb, Silica gel 0.063-0.200 mm and Aluminium oxide 90 active basic were obtained from Merck (Darmstadt, Germany).

SI-4.1.2 Standard solutions

PCBs Polychlorinated biphenyls (PCBs; PCB 1, PCB 2, PCB 3, PCB 10, PCB 21, PCB 28, PCB 30 PCB 31, PCB 49, PCB 50, PCB 52, PCB 55, PCB 56, PCB 78, PCB 85, PCB 87, PCB 97, PCB 101, PCB 104, PCB 105, PCB 110, PCB 112, PCB 118, PCB 128, PCB 137, PCB 138, PCB 141, PCB 143, PCB 145, PCB 149, PCB 151, PCB 153, PCB 156, PCB 170, PCB 180, PCB 187, PCB 194, PCB 202, PCB 204, PCB 206, PCB 207, PCB 209).

PAHs Standard solutions for the polycyclic aromatic hydrocarbons (PAHs); naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, pyrene, fluoranthene, chrysene, benz[a]anthracene, benzo[k]fluoranthene, benzo[b]fluoranthene, benzo[a]pyrene, benzo[ghi]perylene, dibenz[a,h]anthracene, indeno[1,2,3-cd]pyrene) were obtained from Agilent (Palo Alto, CA, USA). Standard solutions for the deuterated PAHs (D8-naphthalene, D10-acenaphthylene, D10-acenaphthene, D10-fluorene, D10-phenanthrene, D10-pyrene, D10-fluoranthene, D12-chrysene, D12-benz[a]anthracene, D12-benzo[k]fluoranthene, D12-benzo[b]fluoranthene, D12-benzo[a]pyrene, D12-benzo[ghi]perylene, D12-dibenz[a,h]anthracene, D12-indeno[1,2,3-cd]pyrene), were obtained from LGC Standards (Wesel, Germany). Deuterated anthracene was obtained from Sigma-Aldrich (Zwijndrecht, The Netherlands).

OCPs Organochlorine pesticides (OCPs); hexachlorobutadiene (HCB), heptachlor, pentachlorobenzene (QCB), alpha-hexachlorocyclohexane (a-HCH), beta-hexachlorocyclohexane (b-HCH), gamma-hexachlorocyclohexane (g-HCH), hexachlorobenzene (HCB), octachlorostyrene (OCS), alpha-heptachlorepoxy (a-HEPO), beta-heptachlorepoxy (b-HEPO), dichlorodiphenyldichloroethane (pp-DDD), dichlorodiphenyldichloroethylene (pp-DDE), dichlorodiphenyltrichloroethane (pp-DDT) dicofol, dieldrin, endrin, mirex) were obtained from AccuStandard (New Haven, CT, USA).

BDEs Bromodiphenylethers (BDEs); hexabromocyclododecane, BDE 85, BDE 99, BDE 119, BDE 138, BDE 153, BDE 154, BDE 183, BDE 190).

PFASs Standard solution mixtures for the perfluorinated sulfonates (perfluorobutanesulfonate (PFBS), perfluorohexanesulfonate (PFHxS), perfluorooctanesulfonate (PFOS), and perfluorodecanesulfonate (PFDS)) and the carboxylic acids (perfluorobutanoic acid (PFBA), perfluoropentanoic acid (PFPA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDcA), perfluoroundecanoic acid (PFUnA), perfluorododecanoic acid (PFDcA), perfluorotridecanoic acid (PFTriA), and perfluorotetradecanoic acid (PFTeA)) as well as 13C3-PFBS, 13C3-PFHxS, 13C8-PFOS, 13C4-PFBA, 13C5-PFPA, 13C5-PFHxA, 13C4-PFHpA, 13C8-PFOA, 13C9-PFNA, 13C6-PFDcA, 13C7-PFUnA, 13C2-PFDcA and 13C2-PFTeA were obtained from Wellington Laboratories (Guelph, ON, Canada).

SI-4.2 Method

SI-4.2.1 Dry weight and organic carbon content

Dry weight

The bottom layer of a 100 mL beaker is filled with shell sand and weighed, and approximately 5 g of sediment sample is weighed and added for the moisture measurement. The sand and sediment are mixed with a glass stick, and the beaker with the sand/sediment mixture is transferred to an oven (WTB Binder, Tuttlingen, Germany) and heated to 105 °C for three hours to remove the moisture. Thereafter, the beakers are placed in a desiccator to cool down. The beaker, including contents, is weighed to obtain the dry weight and subtracted from the total weight before heating, which leads to the moisture contents (in percentage of the total weight). This is further calculated for the analyte concentrations by calculations 2.7 and 2.8.

Organic carbon content

The organic carbon content (OC) was analysed by the Royal Netherlands Institute for Sea Research (NIOZ) according to the protocol from Verardo et al. (1990). This is further calculated for the analyte concentrations by calculation 2.9. All OC values are provided in SI-1.

SI-4.2.2 Sample extraction and clean-up

Passive sampler extraction

The passive samplers were weighed, then transferred to a clean extraction thimble and a Soxhlet set-up was prepared. 600 mL of hexane/acetone (3:1) (v/v) mixture was added to the round bottom flask containing three clean boiling chips for the Soxhlet extraction. The extraction was performed for min. 12 hours overnight. The extract was concentrated with a rotavapor at 40 °C and 400 mbar (Hei-VAP advantage, Hei-VAP expert and Laborator 4003 control, Heidolph Instruments, Schwabach, Germany) to approximately 25 mL.

1 mL of extract was transferred to a pre-weighed tube and weighed again. 1 mL of PAH standard (0.084 ng/mL; D-10 anthracene and 0.074 µg/mL for the other deuterated PAHs) was added to the tube, and weighed. Hereafter, the sample was concentrated with the TurboVap (40 °C, 2-5 mbar) to 1 mL and transferred to a vial for further analysis. 5 mL of extract was transferred to a pre-weighed tube and weighed again. 1 mL of the standard was added (PCB 112/207; 0.01 and 0.04 µg/mL resp.) and weighed. Hereafter, the sample was concentrated with the TurboVap (40 °C, 2-5 mbar) to 1 mL and transferred to a vial for further analysis. 5 mL of extract was transferred to a pre-weighed tube and weighed again. 1 mL of PRC standard was added, and the tube was weighed, then 1 mL of BDE standard (BDE 58; 84 ng/mL) was added and weighed. The sample was concentrated with the TurboVap (40 °C, 2-5 mbar) to 1 mL and transferred to a vial for further analysis.

Sediment sample PFAS extraction and clean-up

PFASs in approximately 8 g sediment were weighed, then extracted and analysed based on the procedure described by Kwadijk et al. (2010). 350 µL internal standard solution (13C labelled PFAS at a concentration of 30 ng/mL in methanol) was added to 8 grams of sediment sample in a 50 mL polypropylene (PP) conical tube. 8 mL of acetonitrile was added to the conical tube and shaken thoroughly. The conical tube was placed in an ultrasonic bath (8510, Branson Ultrasonics Corporation, Danbury, CT, USA) and sonicated for 15 minutes. The sample was then placed in a reciprocal shaker (Stuart SF1, Bibby Sterilin, Stone, UK) on the highest setting for 30 minutes and then centrifuged for 8 minutes at 3135 rpm, after which the acetonitrile layer was transferred to a new 50 mL PP conical tube. The extract was concentrated to approximately 5 mL in the TurboVap (ZW700I, Zymark, Clackamas, ON, USA) at 65 °C and 2-5 mbar and 8 mL of acetonitrile was added. This extraction was repeated twice. Subsequently, 5 mL of hexane was added, and the sample was shaken for 10 min and then centrifuged for 15 minutes at 3135 rpm, after which the hexane layer was discarded. The acetonitrile layer was concentrated to approximately 5 mL and repeated twice, whereafter, the acetonitrile layer was concentrated to approximately 1 mL. The extract was stored in the freezer (-20 °C) overnight, after which the liquid layer was transferred to a new PP tube, and the frozen layer was discarded. This was concentrated in the TurboVap (65 °C, 2-5 mbar) until 1 mL after which 50 mg of

ENVicarb was added. The samples were mixed for 30 seconds using a vortex and subsequently centrifuged at 3000 rpm for 10 min. Extracts were transferred to a vial at 4 °C until analysis.

Sediment sample BDE, OCP, PCB and PAH extraction

Approximately 20 g of sediment sample was weighed and homogenised in a mortar. An excess of anhydrous sodium sulphate (approximately 30 g, dried overnight at 400 °C) was grinded with the sediment to dry the sample until it became a dry powdery mixture. The powder mixture was transferred to a clean Soxhlet extraction thimble, and a Soxhlet set-up was prepared.

600 mL of hexane/acetone (3:1) (v/v) mixture was added to the round bottom flask containing three clean boiling chips for the Soxhlet extraction. The extraction was performed for min. 16 hours overnight. The extract was concentrated with a rotavapor at 40 °C and 400 mbar (Hei-VAP advantage, Hei-VAP expert and Laborator 4003 control, Heidolph Instruments, Schwabach, Germany) to approximately 5 mL.

The extract was transferred quantitatively to a pre-weighed measuring cylinder and supplemented to 10 mL with pentane. One part of the sample of approximately 5 mL was transferred from the cylinder to a pre-weighed conical tube, and 1 mL of BDE standard (BDE 58 ≈84 µg/g) and 0.5 mL of PCB standard (PCB 112/207 ≈40 ng/mL) were added, weighed, and concentrated to approximately 2 mL in the TurboVap (40 °C, 2-5 mbar). 0.5 mL PAH standard (0.084 ng/mL for D-10 anthracene and 0.074 µg/mL for the other deuterated PAHs) was added to a different cylinder with another part of the sample of approximately 5 mL, which was then weighed again, quantitatively transferred to a conical tube, and concentrated to approximately 2 mL under constant nitrogen flow.

Sediment sample BDE, OCP and PCB clean-up

To activate the copper, copper powder was transferred to a flask, and 2M hydrochloric acid was added. The mixture was vortexed until the copper became shiny. The hydrochloric acid was discarded, and the copper powder was washed thrice with dH₂O, two times with acetone and three times with iso-octane. Lastly, iso-octane was added until the copper powder surface was completely covered. Before use, copper powder was transferred to a tube and completely dried under constant nitrogen flow.

Glass wool was placed in a column. 1.8 g anhydrous silica was weighed in a beaker, and pentane was added to obtain a silica slurry loaded on the column and washed with 1.5 mL of pentane. A spatula tip of the activated, dry copper powder was added to the silica-pentane layer. The extract for BDE, PCB and OCP analysis was then loaded on the column and the tube was rinsed with pentane to obtain all of the sample. The column was then eluted with 11 mL pentane and 25 mL 15 % diethyl ether in pentane (v/v) and the eluent was collected in a tube. 1 mL iso-octane was added, and the tube was concentrated in the TurboVap (40 °C, 2-5 mbar) to 3 mL, transferred to a smaller tube and further concentrated to 1 mL. The sample was transferred to a vial and stored at 4 °C until analysis.

Sediment sample PAH clean-up

Aluminium oxide was heated to 150 °C for three hours prior to the clean-up. Then, 5 mL of hexane, 5 g of aluminium oxide and 6 grams of silica were weighed and added to a column. A layer of activated copper powder was added.

Thereafter, the extract for PAH analysis was loaded on the column, rinsed with hexane, and eluted with 40 mL hexane and 100 mL 25% diethyl ether in hexane (v/v). The eluent was collected and concentrated in the rotavapor (40 °C, 250 mbar) to approximately 20 mL, then transferred to a tube and 0.5 mL of iso-octane was added. The sample was concentrated under constant nitrogen flow to approximately 0.5 mL. The sample was transferred to a vial and stored at 4 °C until analysis.

SI-4.2.3 Instrumental analysis

OCPs and PCBs

OCPs and PCBs were determined separately and quantified using a Shimadzu 2010 Plus GC coupled with a Shimadzu TQ8040 MS, AOC-20i Auto-Injector and an EI source. Separation was performed on an HT8 (30 m x 0.25 mm, 0.25 µm) column (Trajan Scientific and Medical, Ringwood, Australia). Dicofol was determined and quantified using a Shimadzu 2010 Plus GC coupled with a Shimadzu TQ8040 MS, AOC-20i Auto-Injector and using an NCI source. In Table 14 the used masses and collision energies are presented for the OCPs.

The OCPs and dicofol calibration curves were made with at least 6 calibration points ranging between app. 0.10 and 110 ng/g IS and the calibration curves for the PCBs were made with at least 6 points ranging between app. 0.30 and 290 ng/g IS. Separation was performed on a CPsil8 (50 m x 0.25 mm, 0.25 µm) column (Agilent Technologies, Palo Alto, CA, USA).

In Table 18 – Table 21 the used masses and collision energies are presented.

Table 17 -Used masses (m/z) and collision energies (CE) for the analysis of the dicofol using GC-MS.

Dicofol		
	target ion (quantification)	qualifier ion
Internal Standard		
D8-dicofol	258	260
Component		
Dicofol	250	252

Table 18 - Used masses (m/z) and collision energies (CE) for the analysis of the OCPs using GC-MS/MS.

OCPs				
Component	target ion (quantification)	CE target ion (v)	qualifier ion	CE qualifier ion (v)
Internal standard				
13C4 HCBD (Hexachlorobutadiene)	263.90>228.90	18	265.90>230.90	18
13C4 HCB (Hexachlorobenzene)	290.80>256.80	19	292.80>258.80	20
Component				
HCBD (Hexachlorobutadiene)	224.80>189.90	18	226.80>191.90	18
HCB (Hexachlorobenzene)	285.80>250.80	20	283.80>248.80	19
Heptachlor	271.80>236.90	18	273.85>238.85	18
Cis-heptachloro epoxide (b-HEPO)	352.80>262.90	20	350.85>260.85	18
Trans-heptachloro epoxide (a-HEPO)	288.85>253.00	10	290.90>254.85	10

Table 19 - Used masses (m/z) and collision energies (CE) for the analysis of the PCBs using GC-MS/MS.

PCBs				
	target ion (quantification)	CE target ion (v)	qualifier ion	CE qualifier ion (v)
Internal standard				
PCB112	324.00>254.00	25	326.00>256.00	25
PCB207	462.00>392.00	30	464.00>394.00	30
Component				
PCB31	256.00>186.00	25	258.00>186.00	25
PCB28	256.00>186.00	25	258.00>186.00	25
PCB52	290.00>220.00	25	292.00>222.00	25
PCB49	290.00>220.00	25	292.00>222.00	25
PCB47	290.00>220.00	25	292.00>222.0	25
PCB66	290.00>220.00	25	292.00>222.00	25
PCB101	324.00>254.00	25	326.00>256.00	25
PCB56	290.00>220.00	25	292.00>222.00	25
PCB97	324.00>254.00	25	326.00>256.00	25
PCB87	324.00>254.00	25	326.00>256.00	25
PCB85	324.00>254.00	25	326.00>256.00	25
PCB110	324.00>254.00	25	326.00>256.00	25
PCB151	360.00>290.00	25	362.00>292.00	25
PCB149	360.00>290.00	25	362.00>292.00	25
PCB143	360.00>290.00	25	362.00>292.00	25
PCB118	324.00>254.00	25	326.00>256.00	25
PCB153	360.00>290.00	25	362.00>292.00	25
PCB141	360.00>290.00	25	362.00>292.00	25
PCB105	324.00>254.00	25	326.00>256.00	25
PCB137	360.00>290.00	25	362.00>292.00	25
PCB138	360.00>290.00	25	362.00>292.00	25
PCB187	394.00>324.00	25	396.00>326.00	25
PCB202	428.00>358.00	25	430.00>360.00	25
PCB128	360.00>290.00	25	362.00>292.00	25
PCB156	360.00>290.00	25	362.00>292.00	25
PCB180	394.00>324.00	25	396.00>326.00	25
PCB170	394.00>324.00	25	396.00>326.00	25
PCB194	428.00>358.00	30	430.00>360.00	30
PCB206	462.00>392.00	30	464.00>394.00	30

Table 20 - Used masses (*m/z*) and collision energies (*CE*) for the analysis of the PRC-PCBs using GC-MS/MS.

PRC-PCBs				
Component	target ion (quantification)	CE target ion (v)	qualifier ion	CE qualifier ion (v)
Internal standard				
PCB 112	323.90>253.90	24	325.90>255.90	24
PCB 207	461.80>391.80	30	463.80>393.80	30
Component				
PCB 1	188.00>152.00	24	190.00>152.00	24
PCB 2	188.00>152.00	24	190.00>152.00	
PCB 3	188.00>152.00	24	190.00>152.00	
PCB 10	222.00>152.00	24	224.00>152.00	24
PCB 14	222.00>152.00	24	224.00>152.00	24
PCB 30	256.00>186.00	24	258.00>186.00	24
PCB 50	289.90>219.90	24	291.90>221.90	24
PCB 21	256.00>186.00	24	258.00>186.00	24
PCB 104	323.90>253.90	24	325.90>255.90	24
PCB 55	289.90>219.90	24	291.90>221.90	24
PCB 145	357.90>287.90	27	359.90>289.90	27
PCB 78	289.90>219.90	24	291.90>221.90	24
PCB 204	427.80>355.80	30	429.80>357.80	30

PAHs

Using an EI source, PAHs were determined and quantified using a Shimadzu Nexis 2030 GC coupled with a TQ8050 NX, AOC-20i Auto Injector. Calibration curves were made with at least 6 calibration points between 0.06 and 1500 ng/g IS. Separation was performed on a ZB-PAH-CT (30 m x 0.25, 0.20 µm) column (Phenomenex, Torrance, CA, USA). The separation was performed twice, once for naphthalene only and once for the other PAHs. In Table 18 used masses and collision energies are presented for the PAHs.

Table 21 - Used masses (*m/z*) and collision energies (*CE*) for the analysis of the PAHs using GC-MS/MS.

PAHs				
	target ion (quantification)	CE target ion (v)	qualifier ion	CE qualifier ion (v)
Internal standard				
D8-Naphtalene	136.00>134.00	10	136.00>108.00	20
D10-Acenaphtylene	160.10>159.10	10	161.10>160.10	10
D10- Acenaphtene	164.00>162.00	20	164.00>160.00	30
D10-Fluorene	176.00>174.00	20		
D10-Phenanthrene	188.00>160.00	20	187.00>159.00	30
D10-Anthracene	188.00>160.00	20	187.00>159.00	30
D10-Fluoranthene	212.00>210.00	20	212.00>208.00	30
D10-Pyrene	212.00>210.00	20	212.00>208.00	30

D12-Benzo(a)anthracene	240.00>238.00	10	240.00>236.00	30
D12-Chrysene	240.00>238.00	10	240.00>236.00	30
D12-Benzo(b)fluoranthene	264.00>260.00	45	264.00>236.00	30
D12-Benzo(k)fluoranthene	264.00>260.00	45	264.00>236.00	30
D12-Benzo(a)pyrene	264.00>260.00	45	264.00>236.00	30
D14-Dibenzo(ah)anthracene	292.00>288.00	35		
D12-Indeno(1,2,3-cd)pyrene	288.00>286.00	20	288.00>284.00	45
D12-Benzo(ghi)perylene	288.00>286.00	20	288.00>284.00	45
Component				
Naphtalene	128.10>102.00	20	128.10>126.10	20
Acenaphtylene	152.10>151.10	10	153.10>152.10	10
Acenaphtene	152.10>151.10	10	153.10>152.10	10
Fluorene	166.10>165.10	10	165.00>163.00	20
Phenanthrene	178.10>176.10	20	178.10>152.10	30
Anthracene	178.10>176.10	20	178.10>152.10	30
Fluoranthene	202.10>200.10	20	202.10>198.10	30
Pyrene	202.10>200.10	20	202.10>198.10	30
Benzo(a)anthracene	228.10>226.00	30	228.10>202.10	30
Chrysene	228.10>226.00	30	228.10>202.10	30
Benzo(b)fluoranthene	252.10>250.10	30	252.10>226.10	30
Benzo(k)fluoranthene	252.10>250.10	30	252.10>226.10	30
Benzo(a)pyrene	252.10>250.10	30	252.10>226.10	30
Dibenzo(ah)anthracene	276.10>274.10	30	276.00>275.00	20
Indeno(1,2,3-cd)pyrene	278.00>277.00	20	278.10>276.10	25
Benzo(ghi)perylene	276.10>274.10	30	276.00>275.00	20

BDEs

BDEs were determined and quantified using a Shimadzu 2010 GC (Shimadzu, Kyoto, Japan) coupled with a Shimadzu QP2010 Ultra MS and AOC-20s Auto-Injector, using a Negative Chemical Ionisation (NCI) source. Separation was performed on a CPsil8 (50 m x 0.25 mm, 0.25 µm) column (Agilent Technologies, Palo Alto, CA, USA). Calibration curves were made with at least 6 calibrations point ranging between app. 0.06 and 260 ng/g IS. In **Table 22** the used masses and collision energies are presented for the PBDEs.

Table 22 - Used masses (m/z) and collision energies (CE) for the analysis of the PBDEs using GC-MS.

PBDEs		
Component	Target ion (quantification)	Qualifier ion
Internal Standard		
PBDE 58	79	81
Component		
PBDEs	79	81

PFAS

PFASs were determined and quantified using an Agilent 1200 LC (Agilent Technologies, Palo Alto, CA, USA), coupled with a Micromass Quattro Micro MS (Waters, Milford, MA, USA), using an Electronic Ionization (EI) source. Separation was performed on a Fluorophase RP (100 x 2.1 mm, 5 µm) column (Agilent Technologies, Palo Alto, CA, USA). Calibration curves were made with at least 6 calibration points between app. 1 and 770 ng/g IS. In **Table 23** the used masses and collision energies are presented for the PBDEs.

Table 23 - Used masses (*m/z*) and collision energies (*CE*) for the analysis of the PFAS using LC-MS/MS.

PFAS				
	target ion (quantification)	CE target ion (v)	qualifier ion	CE qualifier ion (v)
Internal standard				
13C-PFHxS	402.7>103	30		
13C-PFOS	502.7>99	41		
13C-PFBA	216.7>217.8	8		
13C-PFHxA	314.7>269.8	9		
13C-PFOA	416.7>371.8	11		
13C-PFNA	467.7>422.8	11		
13C-PFDcA	514.7>469.8	13		
13C-PFUnA	564.7>519.8	15		
13C-PFDoA	614.7>569.8	13		
Component				
PFBS	299>99	25	299>80	25
PFHxS	398.7>99	30	398.7>80	30
PFHpS	448.7>99	35	448.7>80	35
PFOS	498.7>99	41	498.7>80	41
PFDS	598.7>99	50	598.7>80	50
PFBA	212.7>168.8	8		
PFPeA	262.7>218.8	9		
PFHxA	312.7>268.8	15	312.7>118.8	9
PFHpA	362.7>318.8	15	362.7>168.8	11
PFOA	412.7>368.8	11	412.7>168.8	15
PFNA	462.7>418.8	11	462.7>218.8	15
PFDcA	512.7>218.8	13	512.7>218.8	15
PFUnA	562.7>518.8	15	562.7>218.8	18
PFDoA	612.7>568.8	13	612.7>212.8	18
PFTTrA	662.7>618.8	15	662.7>218.8	18
PFTeA	712.7>668.8	15	712.7>218.8	18

Metals

Analysis of the metal contents of the sediment samples was performed by Triskelion (Utrecht, The Netherlands) according to their protocol WI-159 and WI-156, digesting the samples with aqua regia and analysing the samples using inductively coupled plasma-mass-spectrometry (ICP-MS). The analysed elements were cadmium (Cd), copper (Cu), lead (Pb), Zinc (Zn), nickel (Ni) and mercury (Hg). Further calculations for sediment samples are described at 2.7 Calculations. The calculations for the passive samplers are as performed in Rusina et al. (2010).

SI-4.2.4 Quality assurance

For every analysis run, a blank and a reference sample are extracted simultaneously with the sediment and passive sampler samples to verify that no contamination is present in the samples that would affect the results. Additionally, all the PAHs are adjusted to the blank, which is necessary because of their volatile nature and presence in the vicinity of the lab. All sample concentrations that were lower than the detection limit or five times the blank, as described in calculations 2.4 and 2.5, were regarded as below limit of detection (LOD) and shown with a less-than sign (<) and followed by the calculated LOD. All calibration curves had a $r^2 < 0.996$. Concentrations in the reference sample were within normal limits.

SI-4.2.5 Calculations

Concentrations of the studied samples are quantified with the following calculation.

First, the following calculation converts the blank from the GC/LC-MS data results in ng/g internal standard (IS) to ng absolute.

$$\text{Conc. blank (ng)} = \text{conc. blank} \left(\frac{\text{ng}}{\text{g}} \text{ IS} \right) * \text{weigh in IS (g)}. \quad (2.1)$$

Hereafter, the following calculation converts the analyte concentration from the GC/LC-MS data results in ng/g IS to ng absolute.

$$\text{Conc. analyte (ng)} = \text{conc. analyte} \left(\frac{\text{ng}}{\text{g}} \text{ IS} \right) * \text{weigh in IS (g)}. \quad (2.2)$$

Then, the blank correction is applied to the PAHs by subtracting the blank from equation (2.1) from the analyte from equation (2.2) by the following calculation.

$$\text{Conc. analyte corrected for the blank (ng)} = \text{conc. analyte (ng)} - \text{conc. blank (ng)}. \quad (2.3)$$

The final concentration for the analyte can be calculated from the corrected value of equation (2.3).

$$\text{conc. analyte} \left(\frac{\text{ng}}{\text{g}} \right) = \frac{\text{Conc. analyte corrected for the blank (ng)}}{\text{weigh in monster(g)}} \quad (2.4)$$

The limit of detection (LOD) is determined as the concentration of the lowest standard of the calibration curve by the following calculation.

$$\text{LOD} \left(\frac{\text{ng}}{\text{g}} \right) = \frac{\text{Conc. lowest standard} \left(\frac{\text{ng}}{\text{g}} \right) * \text{weigh in IS (g)}}{\text{weigh in monster(g)}} \quad (2.5)$$

When the concentration of the sample is above the calculated LOD but lower than five times the concentration of the blank, it is also regarded as below the limit of detection in the results by the following calculation.

$$\text{LOD} \left(\frac{\text{ng}}{\text{g}} \right) = \frac{5 * \text{conc. blank} \left(\frac{\text{ng}}{\text{g}} \text{ IS} \right) * \text{weigh in IS (g)}}{\text{weigh in monster(g)}} \quad (2.6)$$

The measured LC-MS/MS and GC-MS/MS results are corrected for the moisture content by the dry weight and the OC by the following three calculations. First, the percentage of wet weight is calculated.

$$\text{wet weight } (-) = \frac{\text{weight before drying (g)} - \text{weight after drying (g)}}{\text{weight before drying (g)}} \quad (2.7)$$

Then, the percentage of dry weight is calculated and converted to the dry weight.

$$\text{dry weight } (-) = 1 - \text{wet weight } (\%) \quad (2.8)$$

Finally, the results of the GC-MS/MS and LC-MS/MS, which were corrected for the dry weight, were normalised to the OC by the following calculation.

$$\text{Analyte conc. } \left(\frac{\text{ng}}{\text{g}} \right) = \text{measured conc. } \left(\frac{\text{ng}}{\text{g}} \right) \times \text{dry weight } (-) \times \text{OC } (-) \quad (2.9)$$

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