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Regional assessment of the historical trends of mercury in sediment cores from Wider Caribbean coastal environments

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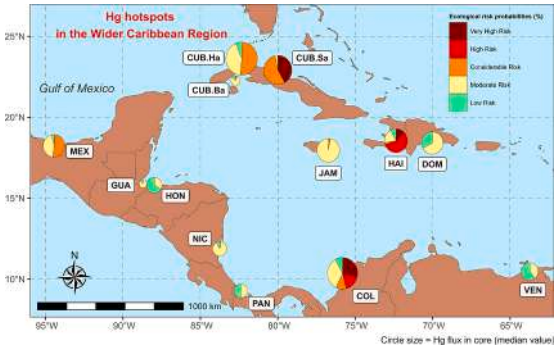
HIGHLIGHTS

- Hg sediment concentrations from 13 Wider Caribbean (WC) coastal sites are reported.
- Sediment Hg hotspots are identified in the WC region where Hg data are limited.
- Chloralkali plants and urban wastes are major Hg sources in the region.

GRAPHICAL ABSTRACT



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ABSTRACT

Spatial and temporal variations of mercury (Hg) concentrations, enrichment, and potential ecological risks were studied in a suite of lead-210 (^{210}Pb) dated sediment cores from 13 Wider Caribbean Region coastal environments. Broad variability of Hg concentrations (19–18761 ng g^{-1}) was observed, encompassing even background levels (38–100 ng g^{-1}). Most Hg concentration profiles exhibited a characteristic upward trend, reaching their peak values in the past two decades. Most of the sediment sections, showing from moderately to very severe Hg enrichment, were found in cores from Havana Bay and Sagua River Estuary (Cuba), Port-au-Prince Bay (Haiti), and Cartagena Bay (Colombia). These were also the most seriously contaminated sites, which can be considered regional Hg 'hotspots'. Both Havana Bay and Port-au-Prince Bay reportedly receive waste from large cities with populations exceeding 2 million inhabitants, and watersheds affected by high erosion rates. The records from the Sagua River Estuary and Cartagena Bay reflected historical Hg contamination associated with chloralkali plants, and these sites are of very high ecological risk. These results constitute a major contribution to the scarce regional data on contaminants in the Wider Caribbean Region and provide reference information to support the evaluation of the effectiveness of the Minamata Convention.

1. Introduction

Almost 40 % of the world's population lives within 100 km of the ocean (UN, 2017). Human settlements exert strong pressure on the coastal and marine environment, including habitat destruction and focused contamination resulting from industrial and domestic waste discharges that may deliver high loads of nutrients, organic pollutants, trace metals, and radionuclides. Among these contaminants, Hg is of utmost global concern as it is highly toxic and persistent in the environment and can be transferred through marine food webs, where it can pose health risks to the biota and even humans through seafood consumption (Anual et al., 2018).

Although Hg has natural sources, its presence in the environment is primarily from anthropogenic inputs, including atmospheric wet/dry fallout and direct releases to land and water (UNEP, 2013). Human activities have led to historic Hg-contaminated sites (so-called "legacy sites"), generally because of the lack or inadequacy of environmental regulations, pollutant-reduction technologies, and appropriated waste management practices. These activities mainly include Hg mining and quarrying, the chloralkali industry, coal-fired power plants, waste incineration, the production of chemical substances, and the use and disposal of products containing Hg, such as batteries and fluorescent lights (Futsaeter and Wilson, 2013). Once in the environment, Hg transport and fate are complex, influenced by chemical, physical, and biological factors, by which Hg can be transformed into the more toxic methylmercury (MeHg), which is readily taken up and retained by organisms and the trophic chain over time.

The Minamata Convention on Mercury (MCM), which came into effect on 15 August 2017, has the objective "to protect the human health and the environment from anthropogenic emissions and releases of mercury and mercury compounds". The MCM recognizes the relevance of monitoring environmental Hg concentrations to assess its long-term effectiveness since, as stated in the Convention's Article 22, the Conference of the Parties, based on the analysis of comparable monitoring data and available scientific information on the presence, movement, and trends of Hg levels in the environment and humans, will evaluate how these have been impacted by the measures taken under the Convention (UNEP, 2023).

Although most countries in the Caribbean Sea basin have ratified the Minamata Convention, limited knowledge of Hg levels in the main environmental compartments makes it impossible to assess trends in Hg contamination and, hence, the Convention's effectiveness. The lack of coastal ecosystem monitoring programs is still a challenge among countries in the Latin American and the Caribbean (LAC) region.

With the support of the International Atomic Energy Agency (IAEA), capacities were developed in LAC to use ^{210}Pb-dated sediment cores as environmental archives to reconstruct long-term contamination trends in coastal zones by using a standardized methodology (OIEA, 2021). Here we present the results of a comparative assessment of Hg contamination in 13 sediment cores collected in 11 countries (Colombia, Cuba, Haiti, Jamaica, Panama, Venezuela, Mexico, Nicaragua, Dominican Republic, Honduras, and Guatemala) from the Wider Caribbean Region (WCR) (Fig. 1), by researchers and laboratories of REMARCO (Research Network of Marine-Coastal Stressors in Latin America and the Caribbean; https://remarco.org/en/). The Hg concentrations from five cores included in this assessment have already been published elsewhere: Sagua River-Estuary (Díaz-Asencio et al., 2009); Havana Bay (Díaz-Asencio et al., 2011); Coatzacoalcos River (Ruiz-Fernández et al., 2012a); Gulf of Batabanó (Díaz-Asencio et al., 2016); and Cartagena Bay (Espinosa-Díaz et al., 2021). The objectives of the study were to i) systematically assess the spatial and temporal distribution of Hg in sediments from 13 geographically diverse coastal environments of LAC, ii) assess the enrichment and potential ecological risk of Hg in the cores, and iii) identify localized 'hotspots' of Hg in the region. Our working hypothesis was that the highest levels of Hg contamination are primarily caused by the proximity to large cities and legacy contamination from industrial sources. The IAEA-Technical Cooperation Programme Regional Project RLA7012, "Use of Nuclear Techniques to Address the Management Problems of Coastal Zones in the Caribbean Region" was implemented between 2007 and 2012 and included the systematic study of a broad range of coastal environments (Fig. 1), selected by the counterparts from the participant countries (Sanchez-Cabeza et al., 2019).

2. Materials and methods

2.1. Description of the study areas

2.1.1. Cartagena Bay (Colombia; core COL)

Cartagena Bay, located on the Caribbean coast of Colombia, has estuarine characteristics because its hydrodynamics are regulated by the Dique channel, an artificial stream that connects the Magdalena River, the main contributor of fluvial fluxes to the Caribbean (Restrepo and Kjerfve, 2000; Restrepo, 2008). Cartagena Bay has a surface area of approximately 84 km^2 and is connected to the Caribbean Sea by two straits, Boca Chica and Boca Grande. Legacy Hg contamination in Cartagena Bay is of interest due to pollution caused by an upstream chloralkali plant, which operated from 1967 to 1977 and used a Hg cathode for electrolysis in chlorine production, from which residues were ultimately discharged into the bay (Alonso et al., 2000; Cogua et al., 2012; Tosic et al., 2019; Espinosa-Díaz et al., 2021).

2.1.2. Gulf of Batabanó (Cuba; core CUB.Ba)

The Gulf of Batabanó is a large and shallow semi-enclosed water body in southern Cuba (20,850 km^2), fringed by mangrove forests and extensively covered by seagrass meadows, mainly *Thalassia testudinum*

(Piñeiro-Ramírez and Dumas-León, 2006) with patches of living coral reefs along the shelf. It is generally considered pristine (Alonso-Hernández et al., 2011). During the last decades, the Gulf of Batabanó has received high sediment loads (up to 1.35 million kg sediment year^{-1}) associated with coastal erosion promoted by deforestation for agricultural purposes (Schiettecatte et al., 2008). Siltation has also led to biodiversity loss, shifts in benthic communities, and reduced catches of commercially valuable species (Baisre, 2000, 2006, 2018; Baisre and Arboleya, 2006; Cerdeira-Estrada et al., 2008). In addition, the Gulf of Batabanó might be impacted by organic contamination due to agricultural wastes from Pinar del Río and the southern Havana province (Alonso-Hernández et al., 2011).

2.1.3. Havana Bay (Cuba; core CUB.Ha)

Havana Bay is an enclosed bay (5.2 km^2) adjacent to Havana City, with ~2.13 million inhabitants in 2016 (ONEI, 2017). Since the 19th century, population density, urbanization, and industrial, commercial, and harbor activities in Havana City have significantly increased (e.g., the city port receives ~50 % of the ships arriving in the country), causing severe damage to the natural resources. It receives untreated wastes from domestic and industrial sources (e.g., oil refinery and power plants) and high sediment loads resulting from soil erosion associated with agriculture and intensive forest exploitation in the catchment. Havana Bay is of environmental concern due to the presence of many contaminants, such as heavy metals (Díaz-Asencio et al., 2011), petroleum hydrocarbons, and organochlorine pesticides (Armenteros et al., 2009).

2.1.4. Sagua River Estuary (Cuba; core CUB.Sa)

The Sagua la Grande River Basin is in northcentral Cuba and has a catchment area of 2188 km^2, with numerous industrial parks and urban establishments (approx. 390,000 inhabitants). The Sagua River discharges into Santa Clara Bay, surrounded by mangrove forests with high marine biodiversity. A Hg-cell chloralkali plant has operated in Sagua la Grande City since the beginning of the 1980s and has caused widespread contamination of the surrounding environment (Gonzalez, 1991; Olivares-Rieumont et al., 2012) due to inefficient wastewater treatment. Previous studies in Sagua Bay, using ^{210}Pb-dated sediment cores, showed maximum Hg levels (2680 ± 130 ng g^{-1}) in 1990 (Díaz-Asencio et al., 2009; Alonso-Hernández et al., 2012). Hg levels in riverine fish muscles (1093 ± 319 ng g^{-1}) and coastal bivalves (596 ± 233 ng g^{-1}) indicated a direct impact from the chloralkali plant (Feng et al., 2019).

2.1.5. Haina River Estuary (Dominican Republic, core DOM)

Sewage and industrial waters from Santo Domingo City are transported to the coastal zone through the Haina River, which receives wastes, often without prior treatment, from Herrera and Haina, the two largest industrial zones in the country. In addition, one of the main ports of the Dominican Republic is located at its mouth. The high degree of contamination of the Haina River has caused an evident impact on the natural and landscape resources of the coastal area, the port, and its urban environment. In the mid-1990s, the first studies began to evaluate its pollution status, pointing to the town of Haina, located on the riverbank, as one of the ten most polluted places in the world (Kaul et al., 1999). Relatively high Hg concentrations in bivalves (290–7020 ng g^{-1}) and near-shore surface sediments (96–5650 ng g^{-1}) have been reported around the country (Sbriz et al., 1998).

2.1.6. Amatique Bay (Guatemala; core GUA)

Amatique Bay is a semi-enclosed bay on the Atlantic coast of Guatemala. It forms a complex ecosystem with coastal lagoons, swamps, and marshes (740 km^2). It is Guatemala's most important estuarine ecosystem due to its ecological and socio-economical value and great eco-touristic potential. The estuarine system is shallow, and three rivers and their corresponding basins (34 % of the country) drain towards the littoral zone. Agricultural activities (corn, beans, rice, and, to a lesser extent, banana, coconut, and pineapple crops) significantly impact woodlands, mangroves, and marshes. Loss of critical habitat, erosion, and sediment transport affecting water quality have been observed (Andrade et al., 2015). Contamination sources include agrochemicals and the lack of drainage and sewage treatment plants (Yáñez-Arancibia et al., 1999).

2.1.7. Port-au-Prince Bay (Haiti; core HAI)

The Port-au-Prince Bay has a coastline of about 45 km and an impact area of >500 km^2. It hosts the country's main industrial activities, including fuel storage, food processing industries, cement factories, loading ports, metal processing (aluminum and iron), and electricity generation. The bay receives effluents from the Froide and Gris Rivers, the latter carrying waste from distilleries, breweries, automobile

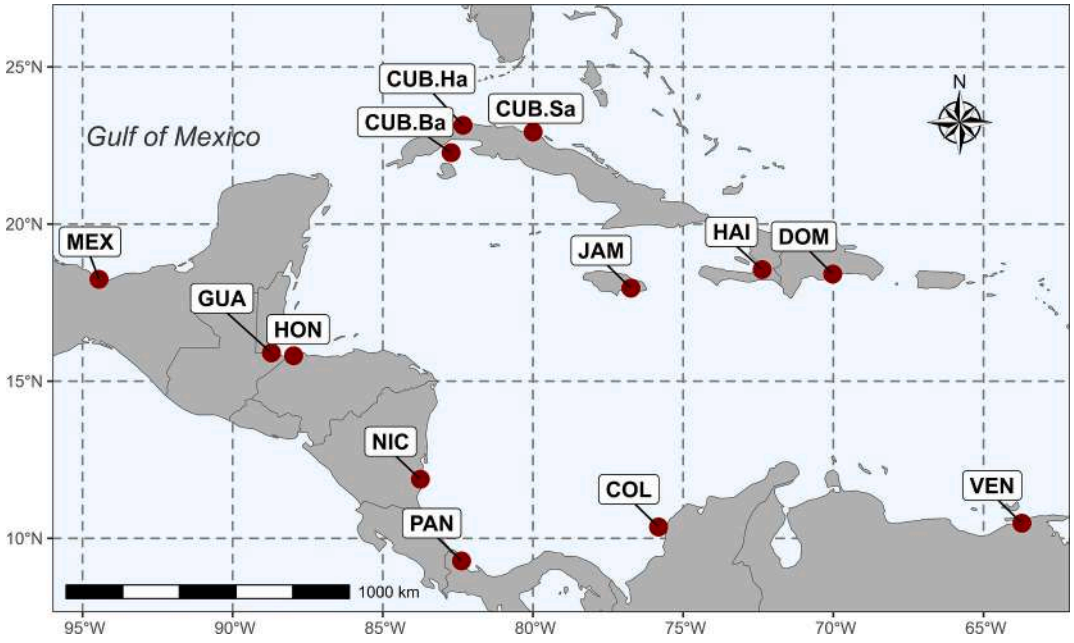

Fig. 1. Sampling locations in the Wider Caribbean Region (coordinates are in Table SI-1).

workshops, and a large agricultural area for bananas, sugar, and vegetables. The most significant activities impacting the coastal zone are acute deforestation, erosion, loss of vegetation cover, and pollution caused by liquid and solid wastes (UNEP, 1999).

More than 1,500 tons of solid waste are generated daily in the city (~2.5 million inhabitants), and 87 % of the domestic waste is directly discharged into the environment (Bras et al., 2009). Domestic wastewater shows high concentrations of ammonium and the presence of heavy metals (Emmanuel et al., 2009). Previous studies have reported chemical pollution in food and drinking water (Schwartzbord et al., 2013; Emmanuel and Emmanuel, 2021), heavy metals in manufacturing paint effluents (Angerville et al., 2005), and mobility and sorption mechanism of heavy metals in soil (Fifi et al., 2010, 2013), but there is no information reported on chemicals in sediments of this marine ecosystem.

2.1.8. Puerto Cortés Bay (Honduras; core HON)

Puerto Cortés Bay is a natural harbor with a 14 m maximum depth (INE, 2018). The primary sources of contamination are contaminated soils, eroded as a consequence of LUC for agriculture and livestock, domestic and industrial wastewater inputs through the Tulián and Medina Chamelecón rivers, manufacturing industry, agriculture, and commercial activities, including the contribution of oil residues generated by port activities. No Hg data have been reported for marine sediments.

2.1.9. Kingston Harbour (Jamaica; core JAM)

Kingston Harbour is located on Jamaica's southeastern coast and is the country's principal port. Kingston City has numerous industrial activities and hosts nearly one million people. Water quality has been badly degraded, and the surrounding land area has deteriorated, reducing its environmental use and tourism attractiveness. Kingston Bay's rich biodiversity is threatened by pollution and invasive species brought by shipping. Only about 25 % of the sewage receives limited treatment before release to the harbor. Uncollected solid wastes from the city are dumped into gullies and stormwater drains, ending in Kingston Bay. In addition, chemical releases into the Río Cobre and the use of agrochemicals, including nutrients, are also important sources of pollution in Kingston Bay. Although it is considered one of the most polluted bays in the Caribbean Sea (Webber and Webber, 1998; Galbraith-Smikle and Webber, 2017; Rose and Webber, 2019), few toxic metal data are reported for this ecosystem and, to the best of our knowledge, Hg contamination trends are unavailable for Kingston Bay.

2.1.10. Coatzacoalcos River Estuary (Mexico; core MEX)

The Coatzacoalcos River has Mexico's third highest water discharge (28,679 hm^3 $year^{-1}$). In the lower part of the basin is the industrial area of Minatitlan–Coatzacoalcos, one of Mexico's largest oil production centers, including the oldest refinery in the country. Trace metals and petroleum hydrocarbons contaminate the Coatzacoalcos River Estuary and wetlands surrounding the industrial area due to the discharges of untreated wastes and oil spills (Páez-Osuna et al., 1986; Vazquez et al., 1991; Rosales-Hoz and Carranza-Edwards, 1998). Increased trace element contamination since the early 1980s, related to higher inputs of eroded soils from the catchment due to extensive land use changes, coinciding with significant industrial and urban development (Ruiz-Fernández et al., 2012b). High levels of Hg in sediment and fish samples have also been observed (Ruelas-Inzunza et al., 2009; Buck et al., 2019). The mixed-use industrial zone of Minatitlán-Coatzcoalcos has been included among the global biological Hg hotspots, which are of heightened concern for human communities and the ecosystems they rely upon (Evers et al., 2014).

2.1.11. Bluefields Lagoon (Nicaragua; core NIC)

The Bluefields Lagoon (176 km^2) is a transition area between the Caribbean Sea and the drainage basins of the Escondido and Kukra rivers. The ecosystem is moderately contaminated by pesticides, polycyclic aromatic hydrocarbons, and some heavy metals (González et al., 1999; Ebanks Mongalo et al., 2015). Increased sedimentation rates is a major environmental problem in the bay (ranging from 5 (1970) to 42 (1998) mm $year^{-1}$) which was related to the socio-economic development in the area, and the occurrence of natural disasters (Dumailo, 2003).

2.1.12. Almirante Bay (Panama; core PAN)

The Almirante Bay is a semi-enclosed lagoon system in the Bocas del Toro Archipelago of Panama. The Changuinola River (~11 km northwest to the bay) runs adjacent to mainland banana plantations and transports sediments into the bay. Low concentrations of heavy metals, including Hg (highest value of 22.9 ± 8 ng g^{-1}), have been reported for surface sediments in the region (Berry et al., 2013).

2.1.13. Gulf of Cariaco (Venezuela; core VEN)

The Gulf of Cariaco is a deep-water body (975 km^2, maximum depth of 90 m) located south of the Araya Peninsula, Sucre State, which receives discharges of seventy-nine watercourses, the most important being the Manzanares and Carinicuao Rivers. Despite its biodiversity richness, marine fish biomass, such as the sardine *Sardinella aurita*, has gradually decreased over the last decade. The eastern coast, known as the "Saco del Golfo," has dense mangroves and seagrass meadows (*Thalassia testudinum*) of great ecological importance. Some trace elements determined in surface sediments were found in concentrations that can affect the biota (Fuentes et al., 2010).

2.2. Sampling and laboratory analysis

Sediment cores were collected at the 13 study sites using UWITEC gravity corer with a PVC liner (8.9 cm inner diameter, 1.0 m long) between 2008 and 2009. The cores were extruded and subsampled at 1-cm intervals; the samples were dried at ≤60 °C or freeze-dried and ground to powder in porcelain mortars. The radiometric analyses for ^{210}Pb dating were performed at Servicio Académico de Fechado (ICML-UNAM, Mexico), Centro de Estudios Ambientales de Cienfuegos (CEAC-Cuba) and the Radiometrics Laboratory (IAEA-EL, Monaco). Further sampling details and ^{210}Pb dating are reported in Ruiz-Fernández et al. (2020). ^{210}Pb depth profiles and ^{210}Pb-derived age models are available as Supplementary Information (Fig. SI-1 and Table SI-1).

Total Hg concentrations were determined with an Advanced Mercury Analyzer LECO AMA-254 (Hg detection limit of 0.01 ng) (Bolaños-Álvarez et al., 2016a) in sediment cores from Gulf of Batabanó, Havana Bay, and Sagua la Grande River-Estuary; and with a Direct Mercury Analyzer Milestone, DMA-80™ (Hg detection limit of 0.005 ng) in all other cores, in CEAC-Cuba (Bolaños-Álvarez et al., 2016b), CIEMAT-Spain, and CIRA/UNAN-Nicaragua. For Hg analyses, 20–50 mg aliquots of ground dry sediments were placed in quartz combustion containers, pre-cleaned in HNO_3 at 30 % for 24 h, and heated at 550 °C for 1 h. No metallic laboratory utensils were used. QA/QC procedures included the analysis of blanks and the assessment of precision through the analyses of replicates ($n = 2$) of all samples of sediments and of certified reference materials (PACS-1, BEST-1, SRM 1645, SRM 1646, IAEA 158, IAEA 433, IAEA 405) that were also used to assess the accuracy of the method. Recovery from reference materials ranged from 83 to 111 %, with a relative standard deviation (RSD) < 5 %.

Aluminum (Al) concentrations were determined by X-ray fluorescence (XRF) spectrometry (Spectro X lab 2000, Spectro Analytical Instruments) at IAEA-EL, Monaco. Sediment aliquots of ~4 g were placed into low-density polyethylene cells, with the bottom covered with Prolene® film, and analyzed under a helium atmosphere (Ruiz-Fernández et al., 2020). Replicate analysis ($n = 6$) of the reference materials IAEA-158, IAEA-405, and IAEA-433 showed strong consistency between certified and analytical values, with accuracy levels >90 % and relative standard deviations below 8 % (Ruiz-Fernández et al., 2020).

2.3. Data treatment

2.3.1. Risk assessment

The observed Hg concentrations in sediment were compared to the benchmarks Probable Effect Level (PEL; 700 ng g^{-1}) and Threshold Effect Level (TEL; 130 ng g^{-1}) (Long and MacDonald, 1998; Buchman, 2008), which allow identifying sediment rarely (<TEL), occasionally (>TEL < PEL), or frequently (>PEL) associated with adverse biological effects (Birch, 2011), such as changes in the benthic community structure (e.g., species richness or relative species abundance), histopathological disorders, hepatic lesions, abnormalities or high mortality rates (Long et al., 1995). These benchmarks are intended for screening purposes and help determine which chemicals present in the sediment might be toxic to the biota and identify the need for further site-specific investigation (e.g., biological and chemical testing) (Ruiz-Fernández et al., 2019).

In addition, the potential ecological risk factor for a substance (Er^i) (Hakanson, 1980) was used to assess the potential level of toxicity of the sediment (Eq. (1)):

$$Er^i = Tr(Hg)_{Hg} \times Cf(Hg) \quad (1)$$

where $Tr(Hg)$ is the toxic response factor for Hg (value = 40), $Cf(Hg)$ is the Contamination Factor, calculated as the ratio of the Hg concentration at each sediment section to the reference concentrations of each core. An $Er^i < 40$ indicates low ecological risk, $40 < Er^i < 80$ moderate, $80 < Er^i < 160$ considerable, $160 < Er^i < 320$ is high and $Er^i > 320$ is very high (Hakanson, 1980).

2.3.2. Background and baseline levels of Hg

Pre-anthropogenic or background levels of Hg (Table 1) for each core were established as the mean value of Hg sediment concentration in sections older than 100 years, which were only found in cores CUB.Ba, CUB.Ha, CUB.Sa, GUA, HAI, NIC, PAN, and VEN. For the other cores, a Hg baseline value (i.e., the content of an element at a given point in time) was established as the oldest core section (Table SI-1) corresponding to 1974 in COL, 1985 in DOM, 1947 in HON, 1944 in JAM, and 1956 in MEX.

2.3.3. Degree of contamination

The enrichment factor (EF, Eq. (2); Buat-Menard and Chesselet, 1979) was used to evaluate the degree of Hg contamination, i.e., the increment of Hg sediment concentrations above a reference (background or baseline) level. As Hg pre-anthropogenic values were unavailable for cores COL, DOM, HON, JAM, and MEX, in these cases, EF refers to the increment of Hg concentrations above a baseline Hg concentration. The EF is normalized by Al concentrations (Table SI-2). Al is commonly used as a proxy for the detrital input to the sediments (Loring and Rantala, 1992), and Al normalization is used to correct for dilution and to assess the enrichment of the trace elements by non-detrital components (Van der Weijden, 2002).

$$EF = \frac{\left(Hg/Al\right)_{section}}{\left(Hg/Al\right)_{reference}} \quad (2)$$

where $\left(Hg/Al\right)_{section}$ is the ratio of Hg to Al concentrations at each sediment section, and $\left(Hg/Al\right)_{reference}$ is the ratio of Hg to Al reference values (either pre-anthropogenic or baseline, Table 1). EF values were classified according to Birch (2003) : $EF \leq 2$ indicates no enrichment; $2 < EF \leq 3$ minor enrichment; $3 < EF \leq 5$ moderate enrichment; $5 < EF \leq 10$ moderately severe enrichment; $10 < EF \leq 25$ severe enrichment, $25 < EF \leq 50$ very severe enrichment, and $EF > 50$, extremely severe enrichment.

2.3.4. Hg fluxes

Hg fluxes are the product of the total Hg concentration (ng g^{-1}) and the ^{210}Pb-derived MAR (g cm^{-2} $year^{-1}$) (Table SI-2, Fig. SI-2). Hg flux ratios were determined as the ratio of the Hg flux in each core section to the reference Hg flux, calculated with the pre-anthropogenic or baseline concentration (Table SI-2).

2.4. Statistical analysis

Analysis of variance (ANOVA) and Tukey post-hoc test (95 % confidence) was used to evaluate the differences in Hg concentrations among cores. The statistical language R (R Core Team, 2023) in the RStudio (version 1.3.1056) environment (RStudio Team, 2023) was used for statistical analyses and graphs.

3. Results

The data supporting this study's findings are openly available in supplementary information.

3.1. Spatial and temporal variations of Hg concentrations

The Hg range of values obtained in sediment cores (19–18761 ng g^{-1}) presented a wide spatial (Fig. 2) and temporal variation (Fig. 3), including the Hg pre-anthropogenic and baseline values (38–100 ng g^{-1}) (Table 1). From the ANOVA results, COL core showed the broadest range of Hg concentrations (93–18761 ng g^{-1}) and the highest mean value (3233 ng g^{-1}). The average Hg concentrations (in descending order) for other cores were CUB.Ha (875 ng g^{-1}) ≈ CUB.Sa (757 ng g^{-1}) ≈ HAI (625 ng g^{-1}) > JAM (338 ng g^{-1}), PAN (184 ng g^{-1}) ≈ VEN (182 ng g^{-1}) ≈ MEX (176 ng g^{-1}) > NIC (91 ng g^{-1}) ≈ DOM (72 ng g^{-1}) ≈ HON (63 ng g^{-1}) ≈ GUA (51 ng g^{-1}) > CUB.Ba (28 ng g^{-1}).

In general, Hg concentrations increased from the bottom to the surface of most cores, except in DOM, COL, and VEN, where recent values were lowest, or in HON, where Hg values were almost

Table 1
Basic statistics and pre-anthropogenic mercury concentrations at 13 coastal sites in the Wider Caribbean region. Concentrations are expressed in ng g^{-1}. N is the number of observations.

Parameter	COL	CUB.Ba	CUB.Ha	CUB.Sa	DOM	GUA	HAI	HON	JAM	MEX	NIC	PAN	VEN
N	65	19	33	32	36	28	45	36	101	26	45	60	45
Min	93	19	370	33	48	38	100	60	273	87	53	67	82
Max	18761	45	1400	2684	120	59	1452	71	646	280	131	367	318
Median	1091	26	930	121	66	51	687	63	305	167	95	189	149
Mean	3233	28	875	757	72	51	625	63	338	176	91	184	182
SD	4772	7	343	918	18	6	359	2	68	68	15	67	78
Pre-anthropogenic levels	NA	22	427	33	NA	43	109	NA	NA	NA	64	202	221
Baseline levels	343	ND	ND	ND	61	ND	ND	63	273	90	ND	ND	ND
Baseline year	1974 ± 4				1985 ± 1			1947 ± 6	1944 ± 1	1956 ± 6			

NA: not available; ND: not determined.

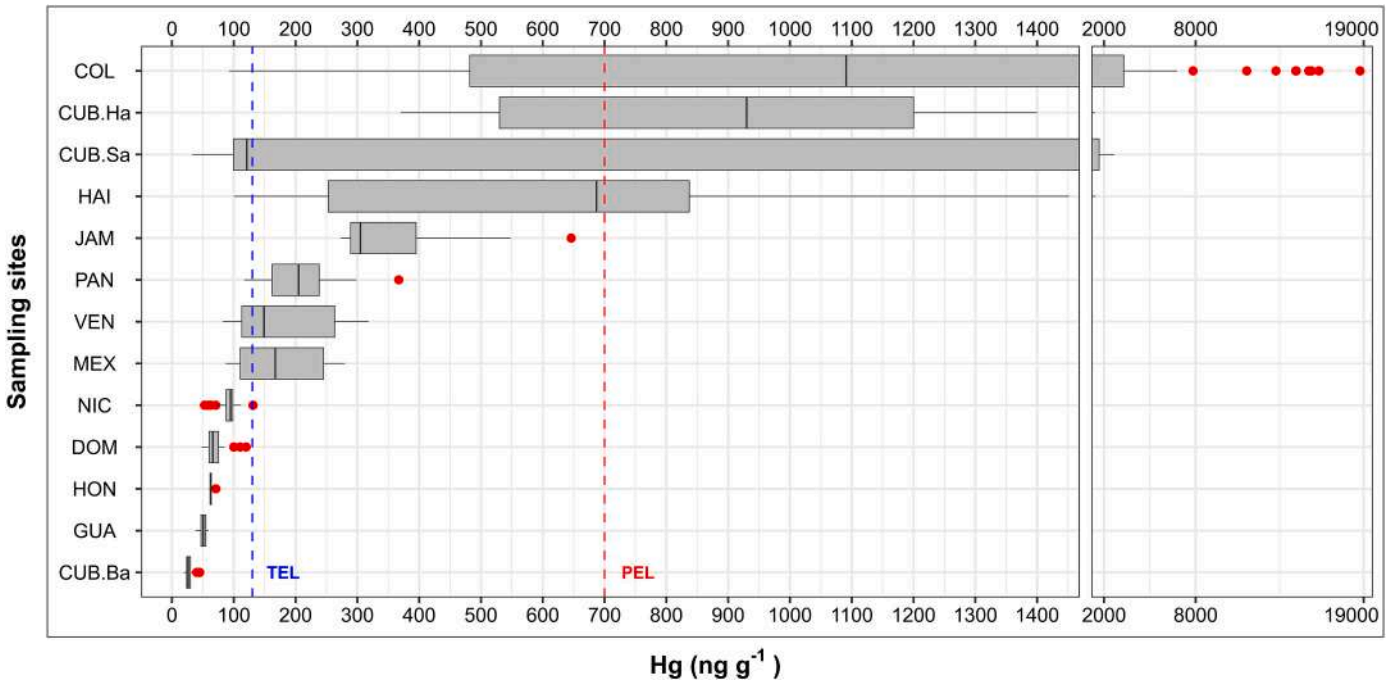

Fig. 2. Concentration ranges of mercury in sediment cores from 13 coastal sites in the Wider Caribbean Region. The blue dashed line indicates the threshold effect level (TEL = 130 ng g^{-1}), and the red dashed line indicates the probable effect level (PEL = 700 ng g^{-1}) (Buchman, 2008).

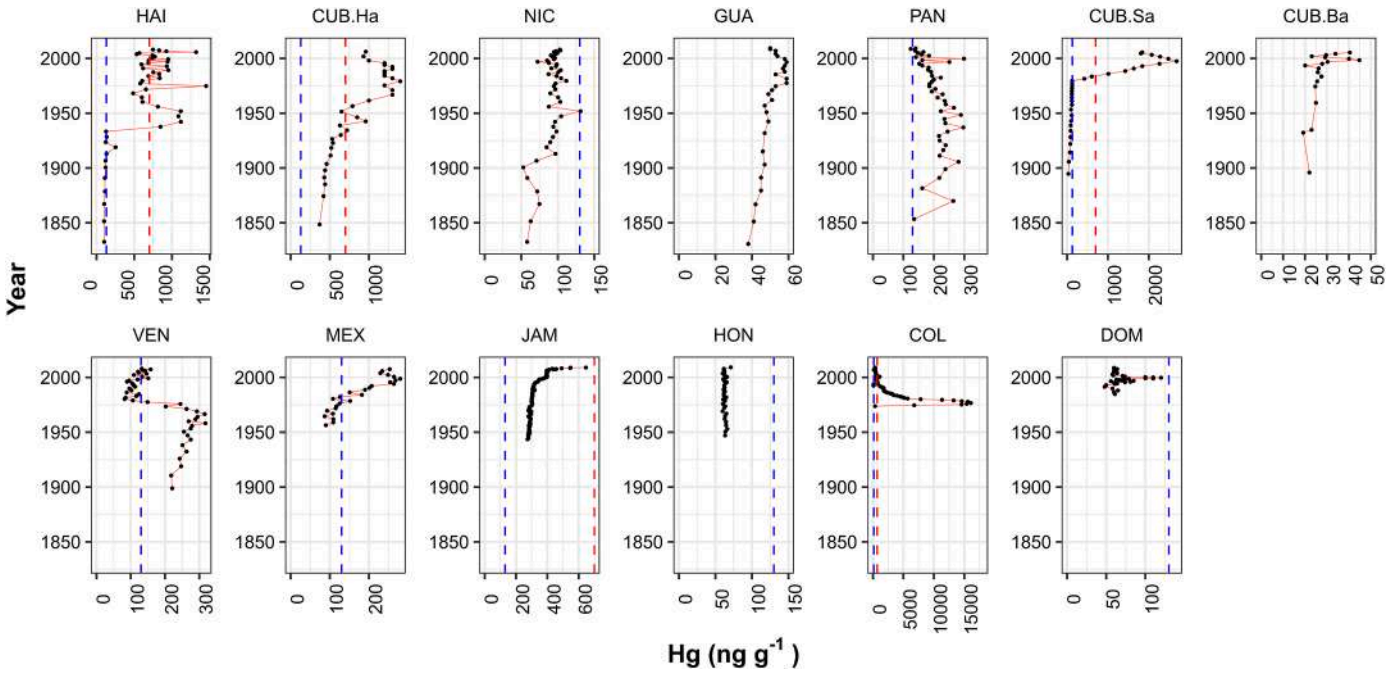

Fig. 3. Temporal profile of mercury concentration in sediment cores from coastal sites in the Wider Caribbean Region. The blue dashed line indicates the threshold effect level (TEL = 130 ng g^{-1}), and the red dashed line indicates the probable effect level (PEL = 700 ng g^{-1}) (Buchman, 2008).

homogeneous along the core (Fig. 3). The Hg concentration maxima, usually observed within the last 20 years, exceeded background values in cores CUB.Ha (3-fold), CUB.Sa (55-fold), and HAI (13-fold), or baseline values in core COL (82-fold). In contrast, Hg concentrations were comparable to baseline levels in DOM, HON, JAM, and MEX or to the background values in CUB.Ba, GUA, NIC, PAN, and VEN, suggesting a negligible or minor impact by Hg sources.

3.2. Contamination and risk assessment

For cores with pre-anthropogenic Hg concentrations, the EF (Table SI-2, Fig. 4) was ≤2.0 (negligible contamination) throughout the records of CUB.Ba, GUA, NIC, PAN, and VEN; and reached values as high as 96 in CUB.Sa, 10 in HAI, 3.2 in CUB.Ha, and 2.2 in CUB.Ba. Regarding the cores with Hg baseline values, the EF were <2.0 for HON and DOM and attained values up to 27 in COL, 4.3 in JAM, and 2.4 in MEX.

The mean EF value for CUB.Sa (26.5) was higher than 25 (i.e., very

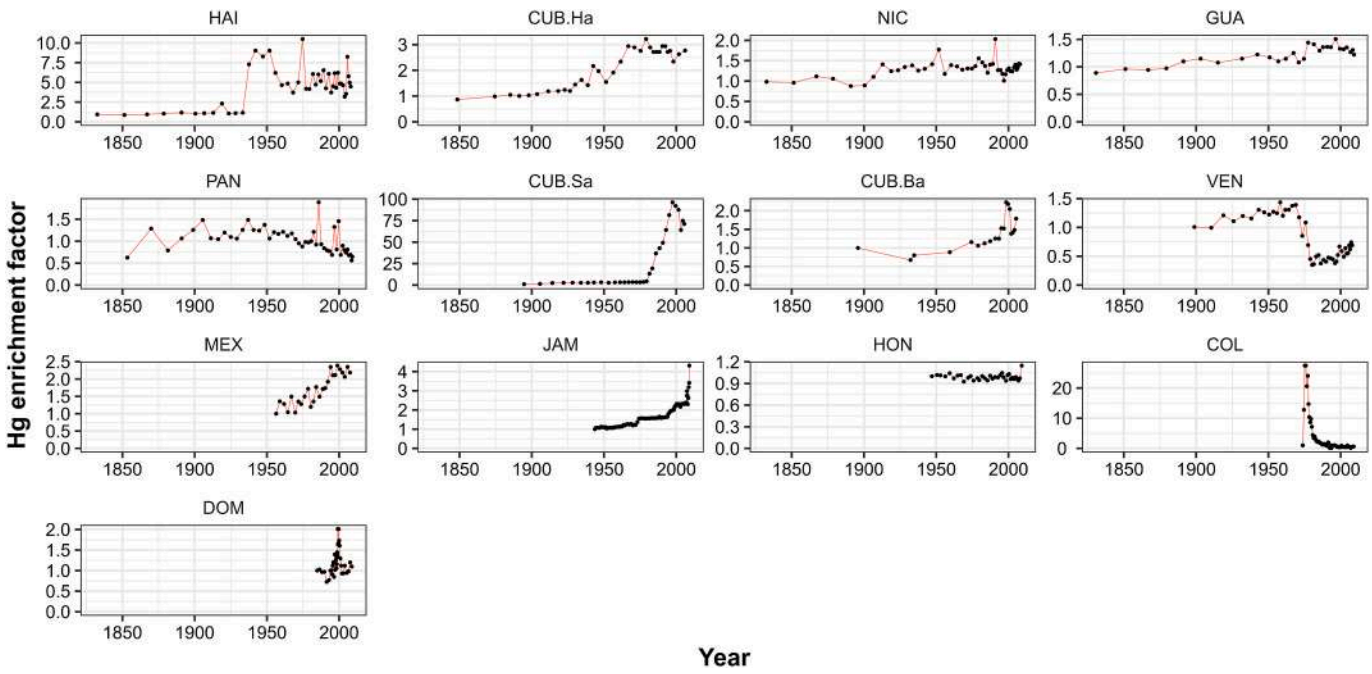

Fig. 4. Temporal evolution of the mercury enrichment factor in sediment cores from coastal sites in the Wider Caribbean Region.

severe enrichment) in the recent sections (15 cm), which indicates the presence of anthropogenic pollution sources. Although the EF mean values for COL (3.5) and HAI (4.4) exhibited moderate enrichment, some sediment sections showed higher EF values (up to 27.4 and 10.0, respectively). The mean EF for CUB.Ha (2.0) corresponded to sediment with "Minor enrichment", while the rest of the cores showed "No Enrichment" (EF < 2.0), indicating limited Hg input from non-natural sources (Fig. 4).

Most sediment samples from cores COL, CUB.Sa, CUB.Ha and HAI showed Hg values higher than TEL (130 ng g^{-1}), and >50 % (except for CUB.Sa, 30 %) were higher than PEL (700 ng g^{-1}) (Fig. 2), suggesting that these sediments are toxic. Hg concentrations in cores JAM, PAN, VEN, and MEX were between TEL and PEL values (suggesting that the sediments are toxic and occasional adverse effects are to be expected). In cores NIC, DOM, HON, GUA, and CUB.Ba, Hg concentrations were below TEL, suggesting that these sediments are non-toxic and adverse effects upon sediment-dwelling fauna would be expected only infrequently.

According to core mean Er^i values (Table SI-2), the ecological risk for the biological community was very high in CUB.Sa (918) and COL (377), high in HAI (229), considerable in CUB.Ha (82), moderate in HON (40), PAN (41), DOM (47), GUA (47), JAM (49), CUB.Ba (51), NIC (57), and MEX (78), and low in VEN (33).

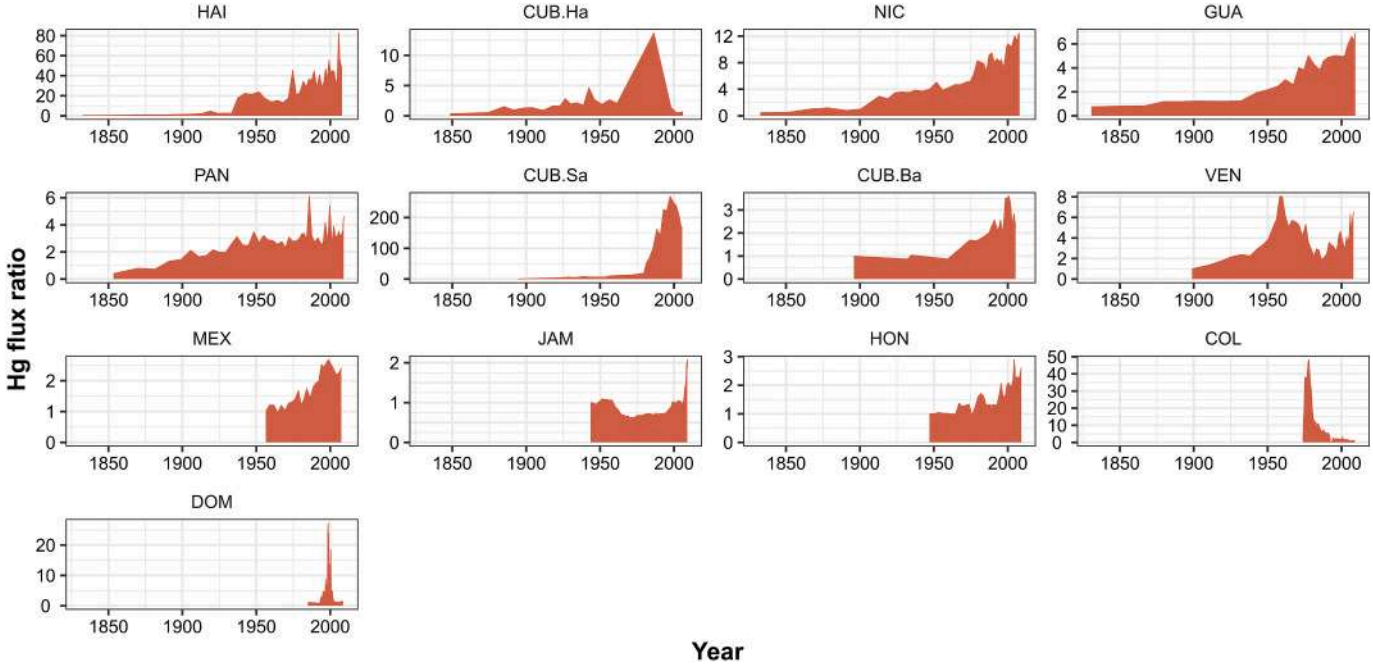

Fig. 5. Temporal evolution of the mercury flux ratio in sediment cores from coastal sites in the Wider Caribbean Region.

3.3. Hg flux ratio

Among the cores spanning >100 years, based on the Hg fluxes (Fig. SI-2), the Hg flux ratios (FR, Fig. 5) between 1900 and 1950 ranged between 2 and 5, except in core HAI, where a maximum of ~20 was observed. Over the second half of the 20th century, the Hg FR steadily increased towards the present, within the range of 2 to 6, in cores CUB. Ba, GUA, NIC, and PAN, whereas it abruptly fluctuated (ranging from 8 to 200) in CUB.Ha, CUB.Sa, HAI, and VEN. Regarding the youngest records, the Hg FR decreased towards the present in COL, remained almost constant in JAM, and increased steadily in HON, MEX, and DOM.

4. Discussion

Our results indicated that Hg concentrations, including the background values (38–100 ng g^{-1}, in sediments deposited >100 years ago), had wide spatial and temporal variability within the WCR. The high variability of background concentration is common; for instance, it ranged from 7.0 to 31 ng g^{-1} in coastal sediments from a reference zone free of industry in Coliumo Bay, Chile (Chandia et al., 2022), and it has been reported as 100 ng g^{-1} in Guanabara Bay, Brazil (Covelli et al., 2012), 130 ng g^{-1} in the Gulf of Trieste (Covelli et al., 2006), 70 ng g^{-1} in Quanzhou Bay, Southeast China (Yan et al., 2020), or 870 ng g^{-1} in Orbetello Lagoon, Italy (Romano et al., 2015). This variability of Hg background levels can be associated with the geological setting and the mineralogical composition of rocks and soils surrounding the study areas. For example, naturally occurring areas with high Hg levels are commonly associated with plate tectonic boundaries, volcanic activity, mineralized regions, or high crustal heat flow zones (Gustin et al., 2000). Also, high levels of Hg can be found in organic sedimentary materials, such as shales or soils enriched with Fe-Al-oxyhydroxides or pyrite (Fitzgerald and Lamborg, 2014).

The Hg concentrations recorded in most cores fell within the third and fourth quartiles (i.e., at the upper half of the Hg concentration distribution) of a database compiled from the literature for coastal and marine ecosystems around the world (Fig. 6). Concentrations in core COL (93–18761 ng g^{-1}) were among the highest reported values in the world. In contrast, Hg values in cores DOM (48–120 ng g^{-1}), GUA (38–59 ng g^{-1}), HON (60–71 ng g^{-1}), and NIC (53–131 ng g^{-1}) fell within the second quartile, and CUB.Ba within the first quartile (19–45 ng g^{-1}).

The highest post-1900 Hg concentrations observed in COL (93–18761 ng g^{-1}), CUB.Sa (33–2684 ng g^{-1}), CUB.Ha (370–1400 ng g^{-1}) and HAI (100–1452 ng g^{-1}) cores were within the ranges reported for sites seriously impacted by the wastes from chloralkali plants (Bloom et al., 2004; Bonsignore et al., 2015; Hatje et al., 2019), mining (Gehrke et al., 2011; Covelli et al., 2017), and other industrial and domestic activities in the coastal-marine environment around the world (Covelli et al., 2012; Aghadadashi et al., 2019; Caballero-Gallardo et al., 2020) (Fig. 6). The intermediate Hg concentrations in MEX (87–280 ng g^{-1}), PAN (67–367 ng g^{-1}), and VEN (82–318 ng g^{-1}) were similar to those reported for slightly contaminated coastal sites (Raygoza-Viera et al., 2014; Botello et al., 2018); whereas the lowest Hg concentrations found in NIC (53–131 ng g^{-1}), DOM (48–120 ng g^{-1}), HON (60–71 ng g^{-1}), GUA (38–59 ng g^{-1}) and CUB.Ba (19–45 ng g^{-1}) were similar to those found in marine and coastal areas with low anthropogenic influence (Sanders et al., 2006; Stupar et al., 2014; Li et al., 2016; Liu et al., 2019).

Most of the Hg profiles (Fig. 3) exhibited an increasing upward trend, with highest Hg flux ratios within the second half of the 20th century (Fig. 5), reaching peak values in the past two decades, as observed in cores from other sites in the world, such as coastal ponds in St. Thomas (US Virgin Islands; Benoit, 2018), the coastal sediments in New England (Fitzgerald et al., 2018), the coastal areas in the southeastern Gulf of Mexico (Ruiz-Fernández et al., 2019).

Both for the GUA and NIC cores (spanning >100 years) and the DOM and HON cores (younger records; Table 1), non-toxicity (Hg concentrations ≤ TEL) and negligible contamination (EF ≤2, either with respect to the pre-anthropogenic or the baseline values) were determined,

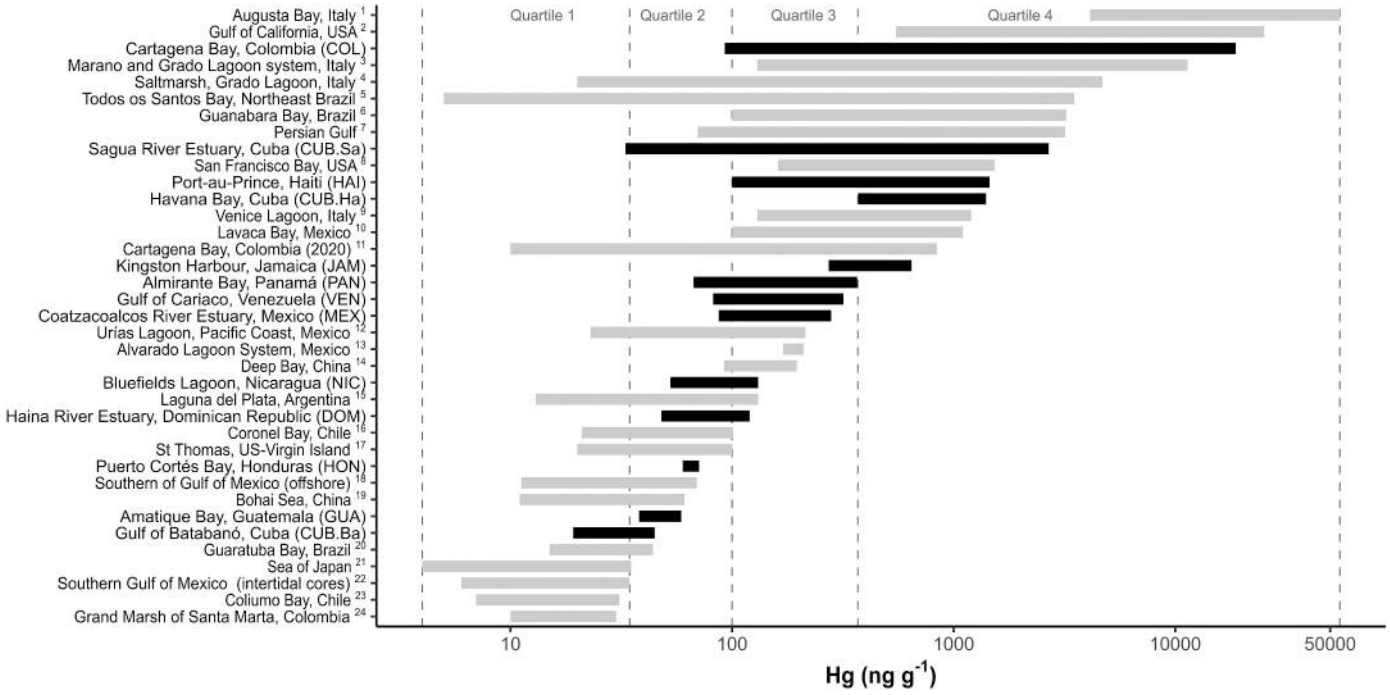

Fig. 6. Mercury concentrations in coastal and marine sediments in the Wider Caribbean Region (WCR) and worldwide. Bars denote Hg concentration ranges at this study sites within the WCR (black) and elsewhere (gray) reported in the literature: (Bonsignore et al., 2015)[1]; (Leal-Acosta et al., 2010)[2]; (Covelli et al., 2012)[3]; (Covelli et al., 2017)[4]; (Hatje et al., 2019)[5]; (Covelli et al., 2012)[6]; (Aghadadashi et al., 2019)[7]; (Gehrke et al., 2011)[8]; (Bloom et al., 2004)[9, 10]; (Caballero-Gallardo et al., 2020)[11, 24]; (Raygoza-Viera et al., 2014)[12]; (Botello et al., 2018)[13]; (Li et al., 2016)[14]; (Stupar et al., 2014)[15]; (Chandia et al., 2022)[16, 23]; (Benoit, 2018)[17]; (Ruiz-Fernández et al., 2019)[18, 22]; (Liu et al., 2019)[19]; (Sanders et al., 2006)[20]; (Song et al., 2015)[21].

suggesting that Hg concentrations were of little concern for these study sites. Contradictorily, Er^i (Table SI-2) indicated that Hg concentrations in these cores represented a moderate ecological risk, comparable to that of the PAN and VEN sediments that, although not contaminated (EF $\leq$ 2, Table SI-2), were found to be toxic (Hg concentrations between TEL and PEL). Studies elsewhere (e.g., Zhuang and Gao, 2014) have also reported contradictory results when using different risk assessment methods. Particularly, in the case of Er^i, the threshold for Hg low potential ecological risk (i.e., 40) is established by Tr(Hg), which means that the index will always indicate potential ecological risk even in the absence of contamination (Ma and Han, 2020). Considering that Hg concentrations in the sediment cores GUA, NIC, DOM, and HON were found to be non-toxic ($\leq$ TEL; Fig. 3), relatively low (Fig. 6), and not enriched (Fig. 4), it is unlikely that the sediments from these study areas represented moderate ecological risk, at least until the period of sampling.

Our results support the hypothesis that the proximity to large cities and industrial sources is a major factor controlling Hg contamination. Among the cores with the highest levels of contamination, the highest Hg concentrations and Hg flux ratios correspond to COL and CUB.Sa cores (Fig. 2). These sites were affected by the operation of chloralkali plants that used Hg as a cathode for electrochemical chlorine production (Díaz-Asencio et al., 2009; Espinosa-Díaz et al., 2021) during the decade they were in operation, released untreated waste into the environment (between 1967 and 1977 in Cartagena Bay, Colombia; between 1980 and 1990s in Sagua Estuary, Cuba). In both cases, the sedimentary records showed a substantial reduction in Hg concentrations after the plant's closure, which highlights the importance of taking proactive measures to protect the coastal environment to safeguard its natural integrity and ensure the long-term conservation and sustainable use of the coastal resources. Nonetheless, in case of sediment resuspension, either by natural (e.g., tidal currents, storms, or hurricanes) or anthropogenic (e.g., dredging, bottom trawling fishing, construction activities including flood defense works, port or wind farm construction) disturbance events, buried Hg-contaminated sediments can be dispersed, released and become bioavailable, posing a higher ecological risk to the biota (Eggleton and Thomas, 2004).

The primary sources of Hg in the CUB.Ha core were associated with the economic development of Havana City, particularly shipping activities tied to sugar and other product exports in the early last century. Between 1950 and 1990, population growth and land use changes triggered the erosion in the Havana Bay catchment. Subsequently, pollution decreased due to Cuba's economic decline and the implementation of environmental policies (Díaz-Asencio et al., 2011).

Increasing Hg concentrations in the HAI core, mainly between the years 1940 and 1950, were probably related to soil erosion promoted by accelerated forest loss, triggered by population growth and urban demands of charcoal (the most important energy source in Haiti), misguided forestry policies (such as the clearance of vast land extension to plant rubber trees, a valued commodity during the II World War), and unfortunate events, such as the deliberate, widespread falling of sacred mapou trees, led by the anti-Vodou campaigns driven by the church (Tarter, 2015; Dubois, 2016). Where forests are cleared and burned, elevated levels of Hg may be released into the environment (UNEP, 2008). Over the last decades, soil erosion caused by deforestation has intensified, and between 1958 and 1995, ~8 million m^3 of sediment ended up in Port-au-Prince Bay (UNEP, 2010). More recently, Hg pollution in Port-au-Prince Bay could be related to the discharge of untreated wastes from activities that potentially contribute to environmental Hg enrichment, such as charcoal and wood fuel, which constituted over 77 % of Haiti's primary energy supply (Lucky et al., 2014), which can be an important source of Hg through atmospheric pathways. Household wastewater can also be a significant source of Hg, as Port-au-Prince Bay (~ 3 million inhabitants) lacks a centralized sewage treatment system, and a substantial portion of the waste is inadequately disposed of and dispersed in the environment (Tull, 2017).

Our results showed that Hg contamination may still be a problem in most of the coastal areas studied, notably in Sagua Estuary and Havana Bay in Cuba, Kingston Bay in Jamaica, and Port-au-Prince Bay in Haiti, where the most recent Hg concentrations recorded at the time of sampling are indicative of toxic levels for the biota, that can harm human health through fish consumption since exposure to high levels of Hg has dangerous and sometimes irreversible effects on the nervous, digestive and immune systems, lungs and kidneys (WHO, 2017).

Given the extremely high Hg concentrations buried in sediment from Cartagena Bay, Sagua Estuary, Havana Bay, and Port-au-Prince Bay, their sources, and environmental fluxes, these ecosystems could be considered hot spots in the WCR, whose continuous monitoring is essential to guarantee the health of the population that depends on the consumption of marine organisms. Given the relevant role of coastal resources in the economy of WCR countries, our results strongly support the urgent need to comprehensively address Hg fluxes to the environment throughout its life cycle.

5. Conclusions

Our study showed a large spatial and temporal variability in Hg contamination across 13 sediment profiles from 11 Wider Caribbean Region (WCR) countries. The highest Hg concentrations found, which were comparable with those reported for severely impacted environments around the world, were linked to historical chloralkali plants operations (in Cartagena Bay and Sagua la Grande Estuary) and socioeconomic factors (Port-au-Prince Bay and Havana Bay), such as harbor activities, vegetation cover loss, and population growth. These four relevant ecosystems, identified as hotspots, are vulnerable to recurrent exposure to high concentrations of Hg as a result of sediment alterations caused by activities such as dredging or trawling. Hg contamination persists in most coastal areas studied, posing risks to local biota and human health through fish consumption.

Our study fills critical information gaps related to the spatial and temporal variability of Hg within the Wider Caribbean Region; using a standardized methodology, and provides comparable results and new knowledge that contributes significantly to the understanding of global Hg pollution. This work established a baseline for future studies on Hg contamination in the WCR, supporting the evaluation of the effectiveness of the Minamata Convention (as stated in the Convention's Article 22) that, based on the analysis of comparable monitoring data and scientific information on changes of Hg concentrations over time, would assess the impact of the measures taken under the Minamata Convention on mercury levels in the environment and humans. Future research should expand spatial and temporal coverage, including MeHg in sediments, water, and organisms in toxicologically relevant areas.

Undoubtedly, a prosperous economy in the WCR and a high quality of life depend on a healthy environment, which also provides the basis for the sustainable development of all human activities. Addressing environmental challenges in the WCR necessitates coordinated economic, social, and environmental policies and coherent governance frameworks, emphasizing pollution control, erosion management, water resources, and waste management for sustainable development.

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CRediT authorship contribution statement

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editing, Validation, Supervision, Investigation. **Juan Pablo Parra:** Investigation, Formal analysis. **Jesús Garay:** Validation, Supervision, Investigation. **Ramón Delanoy:** Investigation, Formal analysis. **Nicolás Solares:** Investigation, Formal analysis. **Katia Montenegro:** Writing – review & editing, Supervision, Investigation. **Alexis Peña:** Supervision, Formal analysis. **Fabiola López:** Writing – review & editing, Investigation, Formal analysis. **Ana Carolina Castillo-Navarro:** Validation, Formal analysis. **Miguel Gómez Batista:** Investigation, Formal analysis. **Alberto Quejido-Cabezas:** Supervision, Investigation, Formal analysis. **Marc Metian:** Supervision, Project administration, Investigation. **Libia Hascibe Pérez-Bernal:** Investigation, Formal analysis, Data curation. **Carlos M. Alonso-Hernández:** Writing – review & editing, Validation, Supervision, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

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

Data availability

The data used is in the supplementary material attached to this submission

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