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ECOTOXICOLOGY IN TROPICAL REGIONS

Chemical contamination assessment in mangrove-lined Caribbean coastal systems using the oyster *Crassostrea rhizophorae* as biomonitor species

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Abstract This paper aims to contribute to the use of mangrove cupped oyster, *Crassostrea rhizophorae,* as a biomonitor species for chemical contamination assessment in mangrove-lined Caribbean coastal systems. Sampling was carried out in eight localities (three in Nicaragua and five in Colombia) with different types and levels of contamination. Oysters were collected during the rainy and dry seasons of 2012–2013 and the tissue concentrations of metals, polycyclic aromatic hydrocarbons (PAHs), and persistent organic pollutants (POPs) were determined. Low tissue concentrations of metals (except Hg) and PAHs; moderate-to-high tissue concentrations of Hg, hexachlorocyclohexanes (HCHs), and dichlorodiphenyl-trichloroethanes (DDTs); detectable levels of chlorpyrifos, polychlorinated biphenyls (PCBs) (mainly CB28, CB118, CB138 and CB 153) and brominated diphenyl ethers 85 (BDE85); and negligible levels of musks were recorded in Nicaraguan oysters. A distinct profile of POPs was identified in Colombia, where the tissue concentrations of PCBs and synthetic musk fragrances were low to moderate, and Ag, As, Cd, Pb, and PAHs ranged from moderate to extremely high. Overall, the values recorded for HCHs, DDTs and PCBs in Nicaraguan mangrove cupped oysters greatly exceeded the reference values in tissues of *C. rhizophorae* from the Wider Caribbean Region, whereas only the levels of PCBs were occasionally surpassed in Colombia. Different contaminant profiles were distinguished between oysters from Nicaragua and Colombia in radar plots constructed using the main groups of contaminants (metals, PAHs, musks, PCBs, and organochlorine pesticides (OCPs)). Likewise, integrated pollution indices revealed differences in the levels of contaminants. Moreover, the profiles and levels in oyster tissues also varied with season. Thus, principal component analysis clearly discriminated Nicaraguan and Colombian localities and, especially in Colombia, seasonal trends in chemical contamination and differences amongst localities were evidenced. The geographical and environmental disparity of the studied scenarios may represent to a large extent the diversity of mangrove-lined Caribbean coastal systems and therefore the present results support the use of *C. rhizophorae* as suitable biomonitor species at Caribbean regional scale, where seasonal variability is a major factor controlling pollutant mobility and bioavailability.

Keywords *Crassotrea* · Mangrove cupped oyster · Bioaccumulation · Ecosystem health · Monitoring · Caribbean · Pollution · Pollution indices

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Introduction Mangroves are highly complex transitional coastal ecosystems that include 0.7% of world's tropical and subtropical forests and provide a unique ecosystem that represents complex food webs with a strong relationship with neighboring habitats (Bosire et al. 2008; Nagelkerken et al. 2008; Giri et al. 2011). In the last four decades, mangrove ecosystems worldwide have been severely impacted by a combination of natural and anthropogenic stressors (Lewis et al. 2011; Bayen 2012). In the Wider Caribbean Region (WCR), mangrove-lined coastal systems, an important natural resource for tourism, fisheries and storm protection, are vulnerable to land-based human activities (UNEP 1994).

In the WCR, mining, smelters, oil refineries, chemical industry, shipyards, untreated sewage sludge and diffuse contamination (e.g., metal piping and traffic) are the main anthopogenic sources of metal pollution; Cd, Hg, and Pb being the metallic contaminants of major concern (UNEP 2006). Burning of waste and vegetation and crude oil production and transportation result in elevated levels of polycyclic aromatic hydrocarbons (PAHs) at regional scale (UNEP 2002; Fernandez et al. 2007). The improper use and disposal of agrochemicals and industrial chemicals, and the release of by-products contribute to raise the environmentally relevant levels of persistent organic pollutants (POPs) such as diphenyl-trichloroethanes (DDTs) and other pesticides, polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) (UNEP 2006). Thus, persistent pesticides represent a major environmental concern in the WCR, where their use in agriculture has greatly increased during the last 30 years and the DDT residues have become ubiquitous (Rawlins et al. 1998; Fernandez et al. 2007). PCBs have been used extensively since the 1930s (e.g., in electrical transfomers) and correspondingly they have been detected in water, sediment, seafood and biota samples (Fernandez et al. 2007). Nevertheless, data and monitoring capacity for these POPs are limited at regional scale. Certainly, PCBs and PBDEs are contaminants distinctive of urban and industrialised areas; however, they have been found at increasing levels in developing countries as a result of poor waste management practices and improper burning (UNEP 2006). In view of their environmental relevance at regional scale, POPs ranked second in the priority rankings of Caribbean contaminant categories (GESAMP 2001). However, with the exception of some pesticides, the monitoring of most of them relies on occasional analyses for research purposes or for impact assessment, e.g., after accidental spills (UNEP 2004; GEF-REPCar 2011).

Pollution monitoring programmes have been carried out in the WCR coastal systems since the 1970s under the auspices of intergovernmental institutions (Siung-Chang 1997; UNEP 2006). Thus, the UNEP Caribbean Environment Programme (CEP) was established in 1981 in order to promote regional cooperation for protection and sustainable development of the Caribbean Sea. Moreover, several tools (e.g., the protocol for Pollution from Land-based Sources and Activities) were agreed in the Cartagena Convention in 1983 in order to classify water bodies, establish legally binding standards, identify major sources of pollutants and prevent pollution (Siung-Chang 1997). However, with notable exceptions (e.g., REDCAM in Colombia, Vivas-Aguas et al. 2014), monitoring activities at national scale are to our knowledge irregular and based on sporadic studies carried out by academics and research groups. A crucial issue in both national and regional monitoring programmes is that elsewhere, employed technology and assessment methods (e.g., Asia-Pacific Mussel Watch, USA Mussel-Watch and EU Marine Strategy Framework Directive; Goldberg 1975; Monirith et al. 2003; Kimbrough et al. 2008; Zampoukas et al. 2014) are not easily available or they are unaffordable and unsustainable; which often may be worsened by simple logistic hurdles (e.g., accessibility to sampling sites and sample transportation logistics). Other challenges include the paucity of baseline environmental data (e.g., concentrations of pollutants), the relatively poor baseline knowledge about contamination in mangrove-lined Caribbean coastal systems, and the limited human capacity and regional networking prospects (Rawlins et al. 1998; UNEP 2004, 2006). Moreover, ongoing monitoring programmes are aimed at recording the concentration of pesticides and other POPs, PAHs and metals in seawater, and sediments but only exceptionally in biota (Sericano et al. 1995; UNEP 2002, 2004, 2006; Rojas de Astudillo et al. 2005; Fernandez et al. 2007; Carvalho et al. 2009a; Vivas-Aguas et al. 2010, 2014; Castañeda-Chávez 2011; Alfonso et al. 2013; Kanhai et al. 2014, 2015). Indeed, monitoring programmes based on the Mussel-Watch approach (e.g., bivalves or other target organisms as biomonitors; Goldberg 1975; Sericano et al. 1995; Monirith et al. 2003; Kimbrough et al. 2008, 2009) are not systemically carried out, to our knowledge. In order to implement this approach in the WCR, research efforts must be addressed to solve logistic and technological questions and to select suitable biomonitor species.

Mangrove cupped oysters, *Crassostrea rhizophorae,* are potential biomonitors for pollution assessment in mangrove-lined coastal ecosystems because they (a) are filter-feeding sessile organisms inhabiting both clean and polluted sites attached either to mangrove roots or to coastal rocks; (b) bioaccumulate high concentrations of pollutants in their tissues (Wallner-Kersanach et al. 2000; Rainbow 2006; Van Lavieren et al. 2011; Torres et al. 2012; Kanhai et al. 2014); (c) respond to environmental insult (Nascimento et al. 1998; Silva et al. 2003; Rebelo et al. 2005; Zaccaron da Silva et al. 2005; Torres et al. 2012); (d) are widely distributed in tropical regions, e.g., from Caribbean to Southern Brazil; and (e) are easy to collect including a large range of size classes (Beeby 2001; Fox 2001; Basu et al. 2007; Valdez Domingos et al. 2007; Masson et al. 2010). Indeed, *C. rhizophorae* has already

been employed as biomonitor species in pollution monitoring studies aimed at deciphering spatial and temporal trends in chemical pollutants in tropical coastal zones in Brazil (Silva et al. 2003, 2006; Rebelo et al. 2005; Torres et al. 2012).

The present investigation aims at expanding the use of the mangrove cupped oyster, *C. rhizophorae*, as biomonitor species for pollution biomonitoring in mangrove-lined Caribbean coastal systems. For this purpose, a pilot field study was carried out in eight localities (three in Nicaragua and five in Colombia) with different types and levels of contamination, in two sampling campaigns during 2012–2013. Samples were collected in the rainy and dry seasons. The tissue concentration of metals, PAHs, and POPs in oysters was recorded as a measure of the nature and levels of bioavailable chemical contaminants and their seasonal variability in different representative scenarios of mangrove-lined Caribbean coastal systems including subtidal oyster reefs in coastal lagoons, intertidal prop roots of magrove trees, and intertidal rocky shores.

Material and methods

Sampling sites and sample collection

In Nicaragua, subtidal (<1 m depth) oyster reefs were studied in two localities (Fig. 1a): Bluefields (sampling sites: Punta Lora and Half Way Cay) and Pearl Lagoon (sampling site: Pigeon Cay). Punta Lora was considered as a prospective reference site (far away from urban settlements) whilst Half Way Cay and Pigeon Cay were selected as potentially polluted areas influenced by aquatic transport and urban discharges (GEF-REPCar 2011; Ebanks-Mongalo et al. 2013). In Colombia, intertidal prop roots of mangrove trees were studied in Cartagena Bay and Barbacoas Bay (Fig. 1b) and intertidal rocky shores in Santa Marta Bay (Fig. 1c). In Cartagena Bay, a mangle islet 200 m north of Isla Maparadita (0.5 Km offshore the Terminal of Cartagena Port) and Isla Brujas were selected as seemingly polluted sites, as shown in previous studies (Vivas-Aguas et al. 2010, 2014). Isla Brujas is an islet adjacent to the industrial zone of Mamonal (oil refineries, petrochemicals, and asphalt, cement and smelting plants) that also receives the direct impact of the Dique Channel. This channel was open five centuries ago to connect the Magdalena River with the Cartagena Bay for navigation and constitutes a major source of sediments and chemical pollution in the Cartagena Bay (Vivas-Aguas et al. 2014). In addition, Isla Barú in Barbacoas Bay was selected as a potential reference site; however, it also may be influenced by the Magdalena River to which it communicates by smaller channels via the Dique Channel since the 1950s (Gómez-Giraldo et al. 2009). In Santa Marta Bay, the Marina Santa Marta was selected as a sampling site subject to strong anthropogenic influence (Garcia et al. 2012), whereas the nearby Taganga Harbor was a priori considered as a reference site.

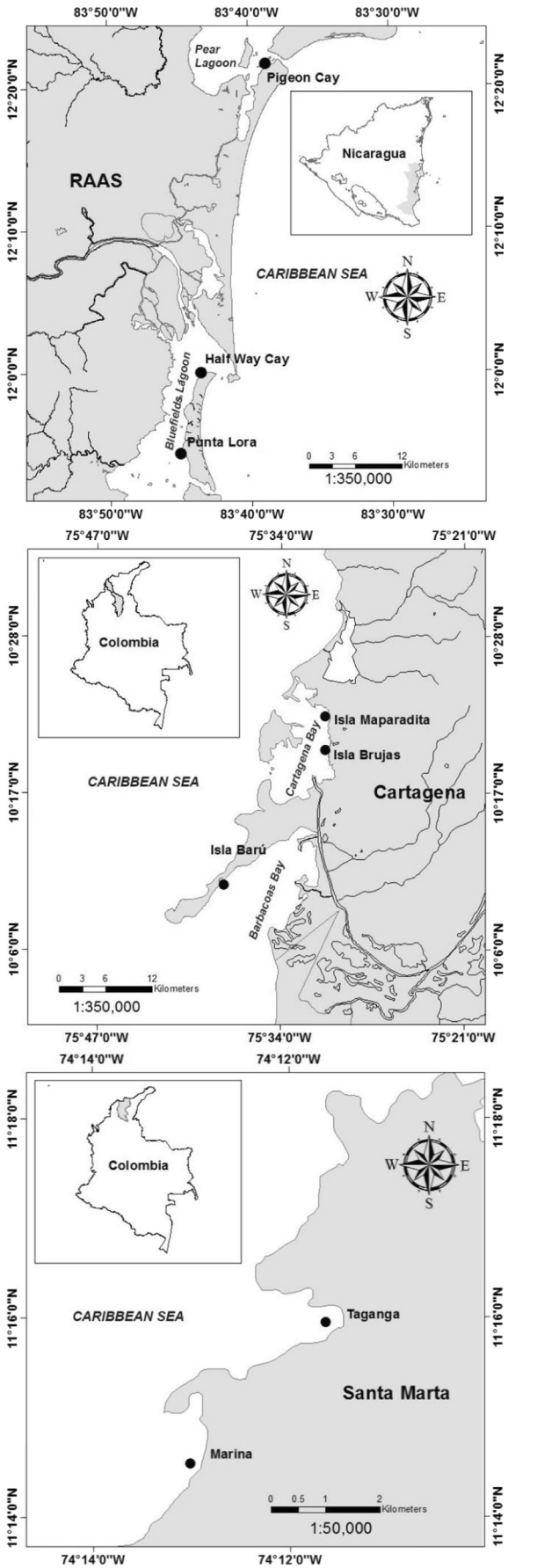

Fig. 1 Caribbean coastal maps from Nicaragua and Colombia, showing the localities where oysters *Crassostrea rhizophorae* were collected in 2012–2013. Nicaraguan coast: Pigeon Cay (12° 21′ 42.50″ N–83° 38′ 32.11″ W); Half Way Cay (11° 59′ 58.73″ N–83° 43′ 28.24″ W) and Punta Lora (11° 54′ 21.02″ N–83° 44′ 58.68″ W). Colombian coast: Isla Barú (10° 10′ 56.02″ N–75° 38′ 21.74″ W); Isla Brujas (10° 19′ 59.07″ N–75° 30′ 47.24″ W) and Isla Maparadita (10° 22′ 21.09″ N–75° 30′ 48.65″ W) in Cartagena Bay and Santa Marta Marina (11° 14′ 31.59″ N-74° 13′ 05.24″ W) and Taganga (11° 16′ 07.68″ N-74° 11′ 37.37″ W) in Santa Marta Bay

Sampling was carried out over 1 year (2012–2013) in the rainy season (October 2012) and in the dry season (March 2013). Seawater surface temperature was in the range of 30–32 °C during both seasons.

Up to 85 mangrove cupped oysters (*C. rhizophorae*) were collected per sampling site, of which 60 were used for biological effects assessment (Aguirre-Rubi et al. submitted) and the remaining 25 for the chemical analyses presented herein. Upon collection, oysters were placed in 15-L plastic boxes (two individuals/L) in seawater at ambient temperature and transported to the laboratory (for 3–6 h) before processing. Due to logistic problems, sampling could not be conducted in Pigeon Cay during the dry season.

Tissue concentrations of contaminants

Pools of magrove cupped oysters (25 individuals) were homogenised and freeze-dried before being stored (at −20 °C) and analyzed. For the analysis of metals (Ag, Al, As, Cd, Cr, Cu, Hg, Ni, Pb, Ti, V, and Zn), 100 mg of freeze-dried samples were digested in a microwave oven (Multi-wave 3000, Anton Paar, Austria) and the extracts were first filtered and then measured in an Inductively Coupled Plasma Mass Spectrometer (ICP-MS; NexION 300 Perkin Elmer, USA) (Bartolomé et al. 2010; Navarro et al. 2010). For the analysis of the organic contaminants, 0.3 g of freeze-dried samples and 0.3 g of Florisil dispersant were manually blended in a glass mortar and were transferred to a 10-mL glass syringe containing 0.6 g of deactivated silica and 4.0 g of activated silica. The target analytes were eluted with 25 mL of dichloromethane, and the eluate was evaporated to dryness using N_2 blowdown and reconstituted to a final volume of 140 L of n-hexane. All these extracts were analyzed by Gas Chromatography Mass Spectrometry (GC–MS) using an Agilent 7890A gas chromatograph coupled to an Agilent 7000 triple quadrupole mass spectrometer and an Agilent 7693 autosampler (Agilent Technologies). The MassHunter WorkStation Acquisition Software (Version B.05.02/Build 5.2.365.0, Agilent Technologies, 2008) was used for data acquisition and automatic integration and quantification of the results. This method was designed to measure up to 40 non-polar or slightly non-polar organic pollutants such as PAHs, PCBs, PBDEs, organochlorine pesticides (OCPs), organophosphorus pesticides and musk fragrances (Ziarrusta et al. 2015). In order to estimate the uncertainty of these measurements, we took into account the uncertainty of each method from their validation step and the experimental uncertainty of replicate samples whenever they were feasible. In the case of metal analysis, half of the samples were analyzed two times and, in the case of organic contaminants, replicates were only carried out in 20% of the samples. From those results, we have estimated an overall relative standard deviation of 10% for metals and 25% for organic contaminants.

Chemical pollution indices

The tissue concentrations of chemical contaminants in mangrove cupped oysters were compared with available environmental quality criteria for individual pollutants (Rimkus 1999; OSPAR Commission 2005, 2009, 2012; Kimbrough et al. 2008; Green et al. 2012): Background/Reference Concentrations (BRCs); Background Assessment Criteria (BACs), Highest Background Concentrations (HBCs), and Highest Low Concentrations (HLC). BRCs are intended to provide baseline or reference concentrations and to describe environmental conditions under which there is no anthropogenic influence on the concentrations of the target substances in the environment (Davies 2004; OSPAR 2005). BACs describe the threshold value for the background level, using data from reference sites (OSPAR Commission 2005, 2009, 2012, 2013). HBCs correspond to the upper limit to Class I in the Norwegian marine pollution monitoring approach, which recognises five classes from Class I, insignificantly polluted, to Class V, extremely polluted (Green et al. 2012). HLCs correspond to the upper limit of the Low Concentration Range in the NOAA's mussel watch pollution monitoring; this recognises three concentration ranges (low, medium, and high) for the tissue concentration of pollutants in oysters (Kimbrough et al. 2008).

The Chemical Pollution Index (CPI; Bellas et al. 2011, 2014; Beiras et al. 2012) was calculated for each site and season. For this purpose, Concentration Factors (CF) were calculated for each pollutant by dividing the tissue pollutant concentrations (C_{tiss}) by the corresponding environmental quality criterion (C_{eqc}): $CF = C_{tiss}/C_{eqc}$. Then, the following formula was applied: $CPI = \Sigma_i[log(CF_i)]$. Likewise, the Pollution Load Index (PLI; Tomlinson et al. 1980) was computed for each site and season by obtaining the *n*-root from the *n*-CFs that were obtained for all the pollutants, according to the following equation: $PLI = \sqrt[n]{CF1\ X\ CF2 \ldots\ldots CFn}$.

Statistical analyses

Statistical analyses were carried out with the aid of SPSS version 22 statistical package (IBM SPSS, Armonk, NY,

USA). The normality of data distribution (Shapiro-Wilk's test) and homogeneity of variance (Levene's test) were determined before proceeding with subsequent analyses. Significant differences between localities and seasons in pollutant tissue concentrations in oysters were analyzed by the *Z* score test ($p < 0.05$). Pearson's correlation between tissue concentrations of contaminants was carried out for Nicaraguan ($N = 5$) and Colombian ($N = 10$) oysters, separately; also between PLI and CPI for the complete set of studied localities. Principal component analysis (PCA) was performed (*N.* variables = 43) on the basis of leverage correlation after normalisation and standardisation of the data, using *The Unscramble v. 7.1* software (Camo, Norway).

Results

Nicaragua

Overall, metal tissue concentrations were not high (Table 1) and after applying the *Z* score test differences between seasons could only be established with a significance level of $p < 0.1$. Likewise, metal tissue concentrations were slightly higher in Half Way Cay than in Punta Lora, and statistical significance was established with $p < 0.1$, especially in the rainy season (Table 1). PAH tissue concentration was apparently higher in the dry season than in the rainy season (Table 2). In the rainy season, the highest values were recorded in Pigeon Cay and the lowest in Punta Lora, with Half Way Cay values in between (Table 2). In the dry season, tissue PAH concentration increased up to eightfold in Punta Lora but remained seemingly unchanged in Half Way Cay (Table 2). The most relevant PAHs found were acenaphthylene, acenaphthene, pyrene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene and dibenzo[ah]anthracene, especially in Pigeon Cay in the rainy season but also, to a lesser extent, in Half Way Cay and Punta Lora in the dry season (Table 2). The sum of the 16 USEPA PAHs (except napthalene) was much lower in Punta Lora during the rainy season than in any other Nicaraguan sample, which was accompanied by low values of $\sum_{HMW}$PAHs and $\sum_{LMW}$PAHs and a high $\sum_{LMW}$PAH/$\sum_{HMW}$PAHs ratio (Table 2). Unlike in the case of PAHs, POP tissue concentrations were higher in the rainy season than in the dry season (Table 3); nevertheless, significant differences after applying the *Z* score test could only be established at the level of $p < 0.1$ in most of the cases. HCHs and DDTs and their derivatives were recorded in oyster tissues in all the samples analyzed, irrespective of the locality and the season (Table 3). Interestingly, the highest tissue concentrations of pesticides (α-HCH, β-HCH, and DDT derivatives such as 4,4-DDE) were recorded in Punta Lora in the rainy season (Table 3). Musk fragrances were always below detection limits. The tissue concentrations for CB28, CB118, and CB153 were high and similar between localities and seasons, whilst for other individual PCBs (CB52, CB101, CB138, and CB180) tissue concentrations were higher in the dry season than in the rainy season, and especially high in Pigeon Cay (Table 3). Only one of the eight analyzed PBDEs (BDE85) was found in oyster tissues at measurable levels; this showed markedly high values in the rainy season in Pigeon Cay and Punta Lora (Table 3).

According to the Pearson's correlation analysis carried out (ESM 1), there existed association (a) amongst $_{LMW}$PAHs (other than acenaphthylene and pyrene) and some $_{HMW}$PAHs,

Table 1 Metal tissue concentration in *Crassostrea rhizophorae* (μg/g) from shallow subtidal oyster reefs of Nicaraguan mangrove lagoons

	LOD	Rainy season			Dry season	
		Pigeon Cay	Half Way Cay	Punta Lora	Half Way Cay	Punta Lora
Ag	0.0015	1.88^{a}	2.08^{a}	2.98^{b}	0.92^{a}	0.89^{a}
Al	15.3	469.67^{a}	665.83^{b}	225.42^{c}	408.77^{a}	488.05^{a}
As	0.01	6.81^{a}	4.23^{b}	2.96^{b}	4.39^{b}	4.59^{b}
Cd	0	1.39^{a}	2.50^{a}	1.78^{a}	1.78^{a}	3.59^{b}
Cr	0.22	0.79^{a}	1.04^{b}	0.36^{a}	0.54^{a}	0.39^{a}
Cu	0.11	557.13^{a}	541.19^{a}	220.02^{b}	487.66^{a}	357.22^{a}
Hg	0.004	0.16^{a}	0.18^{a}	0.12^{a}	0.08^{b}	0.13^{a}
Ni	0.54	1.00^{a}	1.21^{a}	0.54^{b}	0.75^{a}	1.38^{a}
Pb	0.015	0.25^{a}	0.33^{a}	0.21^{a}	0.26^{a}	0.39^{b}
Ti	0.94	18.32^{a}	24.17^{b}	11.15^{a}	11.27^{a}	12.68^{a}
V	0	1.43^{a}	2.07^{b}	0.73^{a}	1.07^{a}	1.44^{a}
Zn	2.36	1525.61^{a}	2544.21^{b}	936.76^{a}	1945.92^{a}	1490.22^{a}
∑Metals		2584.44^{a}	3789.04^{b}	1403.03^{c}	2863.41^{a}	2360.97^{a}

The superscript letters (a, b and c) indicate roughly significant differences between groups ($p < 0.1$)

LOD limit of detection

Table 2 PAH tissue concentration in *Crassostrea rhizophorae* (ng/g) from shallow subtidal oyster reefs of Nicaraguan mangrove lagoons

	LOD	Rainy season			Dry season	
		Pigeon Cay	Half Way Cay	Punta Lora	Half Way Cay	Punta Lora
Acy (1)	1	30[a]	30[a]	udl[b]	26[a]	31[a]
Ace (1)	1	52	29	29	38	38
Phe (1)	0.49	udl[a]	udl[a]	udl[a]	23[a]	83[b]
Ant	1	22[a]	5[b]	4[b]	10[b]	9[b]
Pyr (1)	1	29	udl	udl	udl	25
Benz[a]A (2)	1	24	6	5	13	12
Chr (2)	1	24[a]	5[b]	7[b]	12[b]	11[b]
B[b]F + B[k]F (2)	1	23	udl	3	10	10
B[a]P (2)	1	64[a]	50[a]	udl[b]	45[a]	55[a]
B[ghi]P	1	19[a]	20[a]	udl[b]	17[a]	22[a]
Ind (2)	2	26[a]	26[a]	udl[b]	23[a]	28[a]
D[ah]A (2)	2	29[a]	34[a]	udl[b]	30[a]	40[a]
∑PAHs (16; except Naph)		342[(a)]	204[(a)]	48[(b)]	248[(a)]	365[(a)]
$\sum_{HMW}PAHs^{\Sigma(2)}$ (carcinogenic)		190[(a)]	121[(a)]	15[(b)]	133[(a)]	156[(a)]
$\sum_{LMW}PAHs^{\Sigma(1)}$		111[(a)]	59[(a)]	29[(a)]	87[(a)]	177[(b)]
(%∑PAHs)		(32%)	(29%)	(60%)	(35%)	(48%)
Ind/B[ghi]P		1.37	1.30	n/a	1.35	1.27
$\sum_{LMW}PAHs/\sum_{HMW}PAHs$		0.58[(a)]	0.49[(a)]	1.93[(b)]	0.65[(a)]	1.13[(a)]
Ind/(Ind + B[ghi]P)		0.58	0.57	n/a	0.58	0.56

The superscript letters (a, b and c) indicate significant differences between groups ($p < 0.05$; $p < 0.1$, when shown between brackets). The following compounds were under detection limits in all the samples (superscript numbers refer to the sums and indices given in the table where they are integrated): Flu (LOD = 0.07)[(1)]; Flr (LOD = 0.34)[(1)]

LOD limit of detection, *udl* under detection limits, *n/a* not applicable

such as benzo[a]anthracene, chrysene, benzo[b]fluoranthene + benzo[k]fluo-ranthene, as well as As, chlorpyrifos and PCBs; (b) amongst benzo[a]pyrene, benzo[ghi]pyrene and indeno[1,2,3-cd]pyrene; and (c) between $_{HMW}$PAHs and HCHs (+) and $_{HMW}$PAHs and DDE (−).

The contaminant profile for each locality and season is illustrated in radar plots in Fig. 2a. For this purpose, the following variables were used to construct the radar plots using a spreadsheet: (a) sum of tissue concentrations of metals upon removal of outliers such as Al, Cu, Ti and Zn; (b) sum of 16 US EPA priority PAHs (USEPA 2001) except naphthalene; (c) sum of musks (HHCB + AHTM); (d) sum of 7 I.E. PCBs (OSPAR 2010); and (e) sum of OCPs (HCHs + DDTs). It can clearly be seen that (a) contaminant tissue concentrations were higher in the rainy season than in the dry season in all localities; (b) PCBs and OCPs were the most representative contaminants; and (c) OCPs seem to constitute a potential problem were elevated in Punta Lora during the rainy season.

Positive CPI values, always <1, were recorded in Pigeon Cay in the rainy season and in Half Way Cay and Punta Lora in the dry season (Fig. 3a). Overall, the main pollutants contributing to these CPI values were As, Cd, benzo[a]pyrene, HCHs, DDTs, and PCBs (ESM 2). PLI values were always <50, although PLI recorded in Pigeon Cay at the rainy season was 43.8 (Fig. 3c). Benzo[a]pyrene in the dry season and, generally, HCHs and DDTs were found to be the major contributors to PLI (ESM 3).

Colombia

With the exception of oysters from Taganga Bay, metal tissue concentrations were higher in the rainy season than in the dry season and not very dissimilar amongst localities, with the exception of the Zn tissue concentration (Table 4; *Z* score test, $p < 0.05$). In Taganga, the tissue concentrations of Al, As, Cr, Ni, Ti, and V were significantly higher in the dry season than in the rainy season (Table 4). PAH tissue concentrations were higher in the dry season than in the rainy season except in Isla Brujas where exactly the opposite was observed (Table 5). In the dry season, the most abundant PAH was phenanthrene in all the localities, although fluorene and pyrene were also relevant in Isla Brujas (Table 5). In the rainy season, in contrast, fluorene and anthracene were more concentrated in all the localities, and the levels of carcinogenic PAHs, especially chrysene, but also pyrene, benzo[a]anthracene, benzo[ghi]pyrene and indeno[1,2,3-cd]pyrene, were notably

Table 3 POP tissue concentration in *Crassostrea rhizophorae* (ng/g) from shallow subtidal oyster reefs of Nicaraguan mangrove lagoons

	LOD	Rainy season			Dry season	
		Pigeon Cay	Half Way Cay	Punta Lora	Half Way Cay	Punta Lora
α-HCH	50	$60^{(a)}$	$udl^{(a)}$	$102^{(b)}$	$udl^{(a)}$	$udl^{(a)}$
β-HCH	25	$89^{(a)}$	$90^{(a)}$	$102^{(a)}$	$87^{(a)}$	$116^{(b)}$
γ-HCH	2	$25^{(a)}$	$14^{(a)}$	$9^{(a)}$	$17^{(a)}$	$30^{(b)}$
δ-HCH	25	88^{a}	85^{a}	74^{b}	85^{a}	90^{a}
∑HCHs		262	189	287	189	236
4.4-DDE[3]	2	36^{a}	206^{a}	992^{b}	35^{a}	35^{a}
2.4-DDT[3]	25	$113^{(a)}$	$101^{(b)}$	$90^{(c)}$	$105^{(b)}$	$99^{(b)}$
∑DDTs[Σ (3)]		149^{a}	307^{a}	1082^{b}	140^{a}	134^{a}
4.4-DDT/4.4-DDE		0	0	0	0	0
2.4-DDD/4.4-DDE		0	0	0	0	0
(2.4-DDD + 4.4-DDE)/∑DDTs		$0.24^{(a)}$	$0.67^{(a)}$	$0.92^{(b)}$	$0.25^{(a)}$	$0.26^{(a)}$
∑OCPs		411^{a}	496^{a}	1369^{b}	329^{a}	370^{a}
Chlorpyrifos	2	$11^{(a)}$	$udl^{(b)}$	$udl^{(b)}$	$5^{(b)}$	$udl^{(b)}$
∑Musks[Σ (4)]		-	-	-	-	-
CB28[5]	1	$111^{(a)}$	$100^{(a)}$	$99^{(a)}$	$116^{(b)}$	$101^{(a)}$
CB52[5]	1	18^{a}	udl^{b}	udl^{b}	udl^{b}	udl^{b}
CB101[5]	1	19^{a}	udl^{b}	udl^{b}	udl^{b}	udl^{b}
CB118[5]	1	124	121	111	121	118
CB138[5]	1	22^{a}	udl^{b}	udl^{b}	udl^{b}	udl^{b}
CB153[5]	1	$47^{(a)}$	$29^{(b)}$	$28^{(b)}$	$36^{(b)}$	$35^{(b)}$
∑PCBs[Σ (5)] (PCB_7)		$341^{(a)}$	$250^{(b)}$	$238^{(b)}$	$273^{(b)}$	$254^{(b)}$
BDE85[6]	1	325^{a}	udl^{b}	89^{b}	udl^{b}	udl^{b}
∑PBDEs[Σ (6)]		325^{a}	udl^{b}	89^{b}	udl^{b}	udl^{b}
∑POPs		1088^{a}	746^{a}	1696^{b}	607^{a}	624^{a}

The superscript letters (a, b and c) indicate significant differences between groups ($p < 0.05$; $p < 0.1$, when shown between brackets). The following compounds were under detection limits in all the samples (superscript numbers refer to the sums and indices given in the table where they are integrated): 2.4-DDD (LOD = 5)[(3)]; 4.4-DDT (LOD = 50)[(3)]; 2.4-DDE (LOD = 5)[(3)]; HHCB (LOD = 8.8)[(4)]; AHTN (LOD = 10)[(4)]; CB180 (LOD = 1)[(5)]; BDE28 (LOD = 7.2)[(6)]; BDE47 (LOD = 1.3)[(6)]; BDE66 (LOD = 0.94)[(6)]; BDE99 (LOD = 6.6)[(6)]; BDE100 (LOD = 7.5)[(6)]; BDE153 (LOD = 50)[(6)]; BDE154 (LOD = 50)[(6)]

LOD limit of detection, *udl* under detection limits, *n/a* not applicable

elevated at Isla Brujas (Table 5). Likewise, POP tissue concentrations were noticeably higher in the dry season than in the rainy season, especially in Isla Brujas and Taganga (Table 5). The levels of HCHs, DDTs, chlorpyrifos, and PBDEs in oyster tissues were always below the detection limit (Table 6). The tissue concentrations of PCBs and, more markedly, of musk fragrances were higher in the dry season than in the rainy season in all the localities studied (Table 6). Besides, musk fragrances were below detection limits in Santa Marta Marina and Taganga Bay during the rainy season.

According to the Pearson's correlation analysis carried out (ESM 4), there was association (a) amongst different metals and of these with CB138; (b) amongst $_{HMW}$PAHs; and (c) between musk fragrances and of these with CB52.

Radar plots, constructed as above detailed, were used to illustrate the pollutant profile in Colombian oyster tissues (Fig. 2b). It can be observed that pollutant levels were higher in the dry season than in the rainy season. Isla Brujas was the locality most influenced by PAHs. Musk fragrances gained relevance in the dry season in all localities, especially in Taganga. PCBs and OCPs lacked any relevance in the pollutant profile of Colombian oyster tissues.

Positive CPI values (<1) were only recorded in Isla Brujas in the rainy season (Fig. 3b), with As, Cd, benzo[a]anthracene, chrysene, and benzo[ghi]pyrene as the main pollutants contributing to this CPI value (ESM 2). PLI values were always <50, although PLI recorded in Isla Brujas at the rainy season was 44.0, and PLI values recorded in the dry season in Isla Brujas and Isla Maparadita were, respectively, 19.9 and 11.1 (Fig. 3d). Cadmium, benzo[a]anthracene, chrysene, and benzo[ghi]pyrene were found to be the major contributors to PLI in Isla Brujas in the rainy season whilst in the

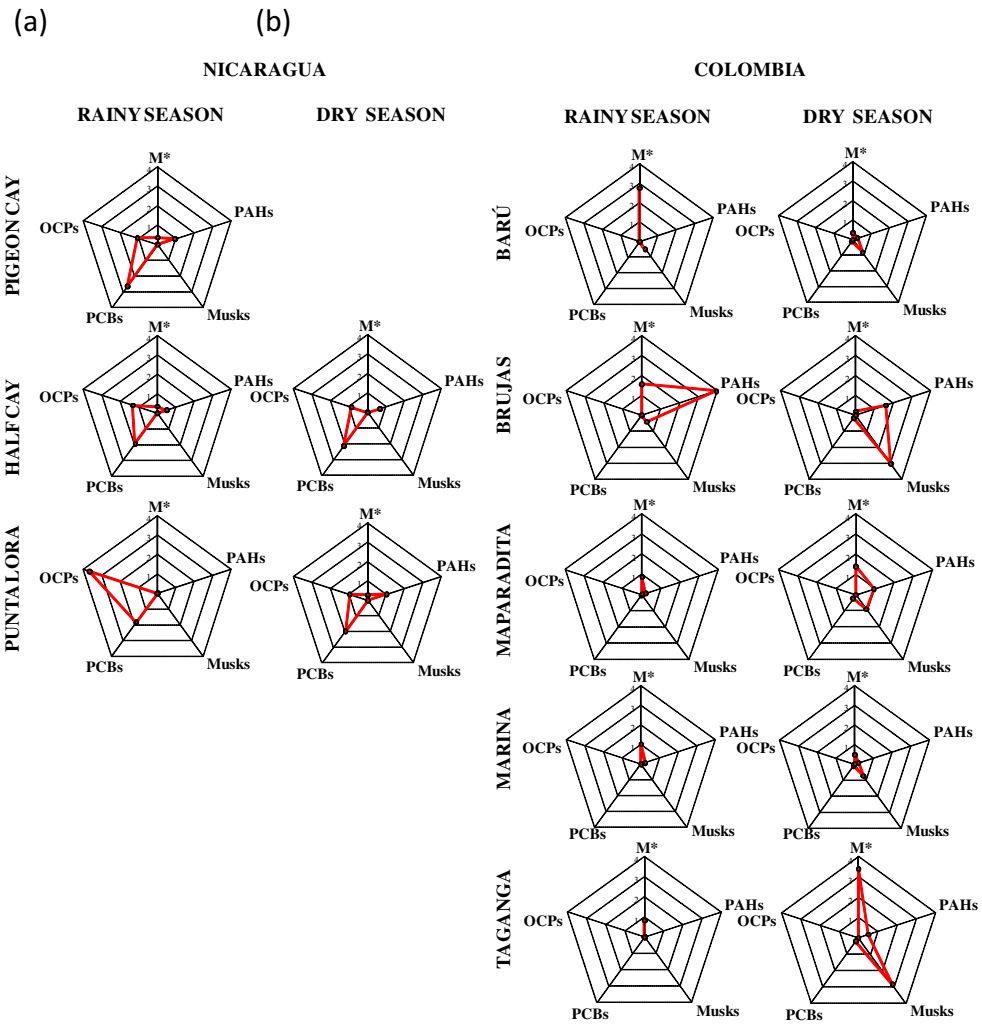

Fig. 2 Radar plots obtained using the total concentration of metals (other than Al, Cu, Ti and Zn: M*; μg/g dry-wt), the sum of the 16 USEPA priority PAHs (ng/g dry-wt), the sum of musks (ng/g dry-wt), the sum of the 7 I.E. PCBs (ng/g dry-wt), and the sum of OCPs (ng/g dry-wt), as variables to depict the tissue pollutant profile (red lines) for each locality and each season in oysters collected from Nicaragua and Colombia

dry season, the main contributors in both Isla Brujas and Isla Maparadita were HHCB, phenathrene, and Cd (ESM 3).

Comparative regional assessment

Principal Component Analysis of the tissue contaminant concentrations showed a 71% variance accounted for by three principal components (PC-1 43%; PC-2 17%; and PC-3 11%; Fig. 4). Communality was high (>74%) for all the items. PC-1 was interpreted in terms of rural vs. urban sources of contaminants, with high agricultural activities, aquatic transport, and rural wastewater disposal in the positive part of its axis (with low-to-moderate scores but high correlation loadings for Ace, Acy, B[a]P, B[b]F + B[k]F, D[ah]A, Ind, γ-HCH, δ-HCH, β-HCH, 4.4-DDE, 2,4-DDT, CB28, CB118, and CB153) and urban contamination in the negative part (with moderate scores and correlation loadings for Flr, AHTN, and HHCB). PC-2 was interpreted in terms of metal contamination, essentially because scores for the tissue concentrations of Al, As, Cr, Ni, Pb, Ti, V, and Zn were higher than the average (unlike for Cd and Ag), with high scores and high correlation loadings for Ni, Ti, and V, and moderate scores and correlation loadings for Al, As, Cr, Pb, V, and Zn. PC-3 was interpreted as seasonality. It was not strongly correlated with any contaminant tissue concentration and only low-to-moderate scores and correlation loadings were found for Hg, B[a]A, B[ghi]P, Chr, Ind, Chlorpyrifos, HHCB, AHTN, CB52, and CD138. However, the tissue concentrations of these chemicals exhibited the most marked seasonal variability, especially in Colombia, where the samples (localities) were clearly discriminated between the positive and the negative part of the PC-3 axis. Thus, at least three consistent clusters were outlined upon combining pairs of PCs (Fig. 4). In the PC-2 vs PC-1 biplot (Fig. 4a) as follows: (i) samples corresponding to Nicaraguan localities at both seasons were clustered lying very markedly towards the positive part of PC-1, characterised by moderate loading (values) of the oyster tissue concentrations of PAHs, OCPs, and PCBs (CB28, CB118 and CB153), (ii) samples from Santa Marta Marina and Taganga at both seasons were clustered at the centre of the top left quadrant around

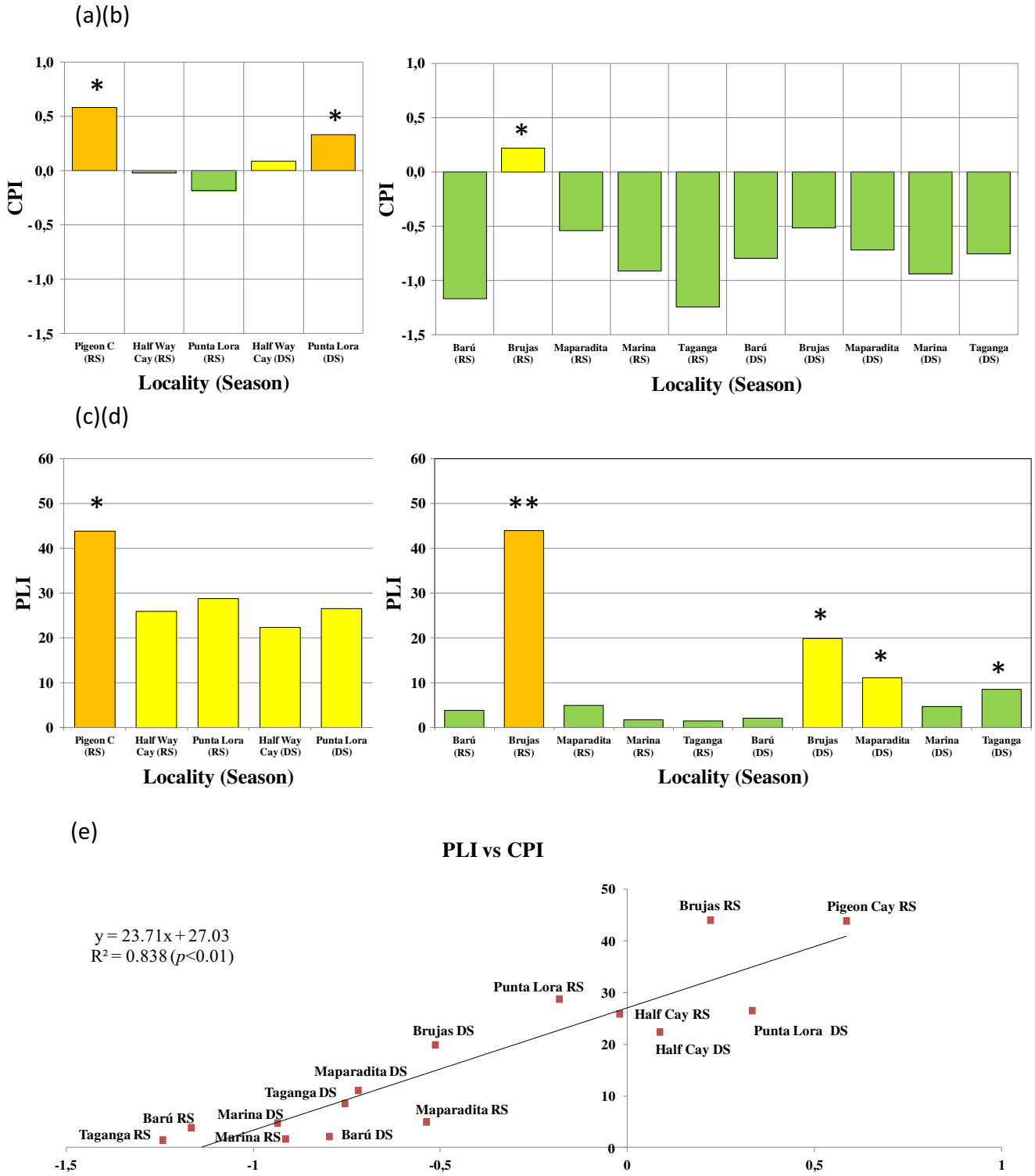

Fig. 3 **a**, **b** Chemical Pollution Index (CPI) in oysters from Nicaragua (**a**) and Colombia (**b**). CPI is categorised into three groups: "low pollution" when CPI ≤ 0, "moderate pollution" when 0 < CPI ≤ 1 and "high pollution" when CPI > 1 (according to Bellas et al. 2011, 2014). **c**, **d** Pollution Load Index (PLI) derived from the tissue concentrations of pollutants in oysters from Nicaragua (**c**) and Colombia (**d**) (Tomlinson et al. 1980). A PLI of 100 correspond to very contaminated sites and a PLI > 50 requires a potential risk and moderate contamination (Angulo 1996). **e** Linear regression of PLI against CFI. *RS* rainy season *DS* dry season

moderate-to-high concentrations of metals such as As, Cr, and Pb, and (iii) samples from Isla Barú during the dry season were clustered nearby moderate levels of musk fragrances. In addition, samples from Isla Brujas and Isla Maparadita at

Table 4 Metal tissue concentration in *Crassostrea rhizophorae* (μg/g) from intertidal roots/docks of Colombian mangrove swamps

	LOD	Rainy season					Dry season				
		Isla Barú	Isla Brujas	Isla Maparadita	Marina	Taganga	Isla Barú	Isla Brujas	Isla Maparadita	Marina	Taganga
Ag	0.0015	$1.08^{(a)}$	$1.52^{(b)}$	$0.89^{(a)}$	$0.21^{(a)}$	$1.24^{(a)}$	$0.48^{(a)}$	$0.41^{(a)}$	$1.47^{(c)}$	$0.06^{(d)}$	$0.28^{(a)}$
Al	15.3	351.01^{a}	317.84^{a}	260.03^{a}	609.10^{a}	522.14^{a}	190.17^{a}	90.45^{a}	58.35^{a}	302.04^{a}	949.40^{b}
As	0.01	8.17^{a}	4.97^{a}	4.67^{a}	13.06^{a}	7.82^{a}	6.87^{a}	6.48^{a}	7.04^{a}	10.60^{a}	33.03^{b}
Cd	0	28.03^{a}	16.60^{b}	6.95^{b}	0.92^{b}	1.08^{b}	2.54^{b}	3.43^{b}	15.88^{a}	0.76^{b}	1.14^{b}
Cr	0.22	0.79^{a}	1.40^{a}	4.71^{a}	3.21^{a}	4.44^{a}	2.11^{a}	0.28^{a}	0.23^{a}	1.22^{a}	6.39^{b}
Cu	0.11	62.42^{a}	184.73^{a}	82.11^{a}	206.10^{a}	105.18^{a}	66.45^{a}	38.72^{a}	54.89^{a}	465.80^{b}	113.04^{a}
Hg	0.004	$0.09^{(a)}$	$0.06^{(a)}$	$0.06^{(a)}$	$0.11^{(a)}$	$0.09^{(a)}$	$0.04^{(a)}$	$0.03^{(b)}$	$0.07^{(a)}$	$0.13^{(c)}$	$0.11^{(a)}$
Ni	0.54	1.07^{a}	1.09^{a}	0.66^{a}	1.27^{a}	1.70^{a}	0.41^{a}	0.49^{a}	0.43^{a}	0.64^{a}	2.34^{b}
Pb	0.015	$0.60^{a(a)}$	$0.54^{a(a)}$	$0.41^{a(a)}$	$0.69^{a(a)}$	$0.56^{a(a)}$	$0.37^{a(a)}$	$0.22^{a(b)}$	$0.15^{b(a)}$	$0.64^{a(a)}$	$0.79^{a(c)}$
Ti	0.94	10.97^{a}	8.72^{a}	5.99^{a}	32.72^{a}	30.74^{a}	5.74^{a}	3.47^{a}	2.33^{a}	21.07^{a}	72.64^{b}
V	0	1.25^{a}	1.23^{a}	1.25^{a}	1.82^{a}	2.29^{a}	0.76^{a}	0.63^{a}	0.38^{a}	1.14^{a}	4.16^{b}
Zn	2.36	495.46^{a}	1951.81^{a}	1401.70^{a}	2302.36^{a}	2093.90^{a}	1271.46^{a}	718.69^{a}	488.58^{a}	3541.70^{b}	1900.18^{a}
∑Metals		960.93	2490.53	1769.43	3171.57	2771.19	1547.40	863.30	629.80	4345.81^{b}	3083.50

The superscript letters (a, b and c) indicate significant differences between groups ($p < 0.05$; $p < 0.1$, when shown between brackets)

LOD limit of detection

Table 5 PAH tissue concentration in *Crassostrea rhizophorae* (ng/g) from intertidal roots/docks of Colombian mangrove swamps

	LOD	Rainy season					Dry season				
		Isla Barú	Isla Brujas	Isla Maparadita	Marina	Taganga	Isla Barú	Isla Brujas	Isla Maparadita	Marina	Taganga
Acy $^{(1)}$	1	udl	udl	udl	udl	udl	udl	udl	udl	udl	udl
Ace $^{(1)}$	1	udl	udl	udl	udl	udl	udl	udl	udl	udl	udl
Flu $^{(1)}$	0.07	0.8^{a}	16.1^{a}	udl^{a}	37.9^{b}	udl ^{a}	udl	udl	udl	udl	udl
Phe $^{(1)}$	0.49	$udl^{a(a)}$	$25.4^{a(a)}$	$udl^{a(a)}$	$udl^{a(a)}$	$udl^{a(a)}$	$117.4^{a(a)}$	$290.2^{a(b)}$	333.4^{b}	$106.2^{a(a)}$	$206.5^{a(a)}$
Ant	1	40.2 $^{a(a)}$	3.5 $^{a(b)}$	1.5 $^{a(b)}$	49.6^{b}	$42.1^{a(a)}$	udl $^{a(b)}$	udl $^{a(b)}$	udl $^{a(b)}$	udl $^{a(b)}$	udl $^{a(b)}$
Flr $^{(1)}$	0.34	udl^{a}	6.4^{a}	2.4^{a}	udl^{a}	udl^{a}	2.0^{a}	120.8^{b}	8.5^{a}	udl^{a}	udl^{a}
Pyr $^{(1)}$	1	udl^{a}	59.1^{a}	8.5^{a}	0.9^{a}	udl^{a}	udl^{a}	144.8^{b}	udl^{a}	udl^{a}	udl^{a}
Benz[a]A $^{(2)}$	1	udl^{a}	101.8^{b}	42.6^{a}	2.7^{a}	udl^{a}	udl^{a}	udl^{a}	udl^{a}	udl^{a}	udl^{a}
Chr $^{(2)}$	1	udl^{a}	837.1^{b}	45.2^{a}	22.0^{a}	udl^{a}	udl^{a}	udl^{a}	udl^{a}	udl^{a}	udl^{a}
B[ghi]P	1	udl	202.8	udl	udl	udl	udl	udl	udl	udl	udl
Ind $^{(2)}$	2	udl	47.3	udl	udl	udl	udl	udl	udl	udl	udl
∑PAHs (16; except Naph)		41.0^{a}	1299.5^{b}	100.3^{a}	113.1^{a}	42.1^{a}	119.4^{a}	555.8^{a}	341.9^{a}	106.2^{a}	206.5^{a}
$\sum_{HMW}$PAHs $^{\Sigma\,(2)}$ (carcinogenic)		n/a	986.3	87.8	24.7	n/a	n/a	n/a	n/a	n/a	n/a
$\sum_{LMW}$PAHs $^{\Sigma\,(1)}$ (%∑PAHs)		0.8 (2%)	81.6 (6%)	10.9 (11%)	38.8 (34%)	n/a	n/a	265.6 (48%)	8.5(2%)	n/a	n/a
Ind/B[ghi]P		n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
$\sum_{LMW}$PAHs/$\sum_{HMW}$PAHs		n/a	0.1	0.1	1.6	n/a	n/a	n/a	n/a	n/a	n/a
Ind/(Ind + B[ghi]P)		n/a	0.2	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a

The superscript letters (a, b and c) indicate significant differences between groups ($p < 0.05$; $p < 0.1$, when shown between brackets). The following compounds were under detection limits in all the samples (superscript numbers refer to the sums and indices given in the table where they are integrated): [b]F + B[k]F (LOD = 1)$^{(2)}$; B[a]P (LOD = 1)$^{(2)}$; D[ah]A (LOD = 2)$^{(2)}$

LOD limit of detection, *udl* under detection limits, *n/a* not applicable

Table 6 POP tissue concentration in *Crassostrea rhizophorae* (ng/g) from intertidal roots/docks of Colombian mangrove swamps

	LOD	Rainy season					Dry season				
		Isla Barú	Isla Brujas	Isla Maparadita	Marina	Taganga	Isla Barú	Isla Brujas	Isla Maparadita	Marina	Taganga
∑HCHs$^{\Sigma\,(3)}$		0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
∑DDTs $^{\Sigma\,(4)}$		0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
∑OCPs		0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
HHCB $^{(5)}$	8.8	11.1^{a}	9.6^{a}	0.4^{a}	udl^{a}	udl^{a}	19.6^{a}	71.0^{b}	20.9^{a}	17.5^{a}	67.1^{c}
AHTN $^{(5)}$	10	0.2$^{(a)}$	udl$^{(a)}$	udl$^{(a)}$	udl$^{(a)}$	udl$^{(a)}$	29.1$^{(a)}$	48.7$^{(b)}$	34.2$^{(a)}$	24.6$^{(a)}$	37.8$^{(a)}$
∑Musks $^{\Sigma\,(5)}$		11.3^{a}	9.6^{a}	0.4^{a}	0.0^{a}	0.0^{a}	48.6^{a}	119.6^{b}	55.0^{a}	42.1^{a}	104.8^{a}
CB28 $^{(6)}$	1	udl	3.7	5.7	0.6	udl	udl	udl	udl	udl	udl
CB52 $^{(6)}$	1	udl	udl	udl	udl	udl	14.4	21.0	27.6	13.5	24.1
CB138 $^{(6)}$	1	udl	udl	udl	udl	udl	udl	udl	udl	1.5	3.8
CB180$^{(6)}$	1	udl	udl	udl	udl	0.8	udl	udl	1.7	udl	udl
∑PCBs$^{\Sigma(6)}$ (PCB_7)		0.0$^{(a)}$	3.7$^{(a)}$	5.7$^{(a)}$	0.6$^{(a)}$	0.8$^{(a)}$	14.4$^{(a)}$	21.0$^{(a)}$	29.3$^{(b)}$	15.0$^{(a)}$	27.9$^{(a)}$
∑PBDEs$^{\Sigma\,(7)}$		0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
∑POPs		11.3$^{a(a)}$	13.3$^{a(a)}$	6.1$^{a(a)}$	0.6$^{a(a)}$	0.8$^{a(a)}$	63.0$^{a(a)}$	140.6$^{a(b)}$	84.4$^{a(a)}$	84.0$^{a(a)}$	158.3^{b}

The superscript letters (a, b and c) indicate significant differences between groups ($p < 0.05$; $p < 0.1$, when shown between brackets). The following compounds were under detection limits in all the samples (superscript numbers refer to the sums and indices given in the table where they are integrated): α-HCH (LOD = 50)$^{(3)}$; β-HCH (LOD = 25)$^{(3)}$; γ-HCH (LOD = 2)$^{(3)}$; δ-HCH (LOD = 25)$^{(3)}$; 4.4-DDE (LOD = 2)$^{(4)}$; 2.4-DDD (LOD = 5)$^{(4)}$; 2.4-DDT (LOD = 25)$^{(4)}$; 4.4-DDT (LOD = 50)$^{(4)}$; Chlorpyrifos (LOD = 2); CB101 (LOD = 1)$^{(6)}$; CB118 (LOD = 1)$^{(6)}$; CB153 (LOD = 1)$^{(6)}$; BDE28 (LOD = 7.2)$^{(7)}$; BDE47 (LOD = 1.3)$^{(7)}$; BDE66 (LOD = 0.94)$^{(7)}$; BDE85 (LOD = 1)$^{(7)}$; BDE99 (LOD = 6.6)$^{(7)}$; BDE100 (LOD = 7.5)$^{(7)}$; BDE153 (LOD = 50)$^{(7)}$; BDE154 (LOD = 50)$^{(7)}$

LOD limit of detection, *udl* under detection limits, *n/a* not applicable

the dry season were clearly discriminated from any other sample, being located towards the extreme bottom left quadrant (high levels of Flr and moderate-to-high of Cd and Phe). In the PC-3 vs PC-1 biplot (Fig. 4b), the Nicaraguan cluster remained as in the previous biplot but samples from Colombia appeared in two clusters: (i) rainy season samples in the top left quadrant, together with the highest loading values for Cd and Flu in Brujas and to a lesser extent in Isla Maparadita and Marina and (ii) dry season samples in the top left quadrant, together with the high loading values for AHTN and CB52, and moderate values for As, Phe, and HHCB. Finally, the PC-3 vs PC-2 biplot (Fig. 4c) discriminated all the samples both by location and season, although the loadings were in most of the cases low-to-moderate and only the 28% of the variance was accounted for. Samples from Santa Marta Marina and Taganga were clustered depending on the season, with moderate-to-high loadings for metals with less (e.g., Al, As, Cr, Ni, Pb, Ti, V, and Zn) and more (e.g., As and Hg) marked seasonal trends. Samples from Barú, Brujas, and Maparadita during the rainy season appeared in a cluster associated to Chr, B[a]A, and B[ghi]P and to a lesser extent to Cd, Ant, Flu, and Ind). Finally, samples from Isla Barú, Isla Brujas, and Isla Maparadita during the dry season and Pigeon Cay during the rainy season were distributed in the bottom left quadrant, without a clear chemical contamination profile as, except for AHTN and CB52, correlation loadings are low to moderate.

Discussion

Metals

In Nicaragua, metal tissue concentrations in mangrove cupped oysters were low, according to proposed criteria for oysters and mussels (Silva et al. 2003, Kimbrough et al. 2008, Alfonso et al. 2013, Solaun et al. 2013), with some exceptions. Mercury was found at moderate levels in the dry season and in Ponta Lora during the rainy season and at high levels in Half Way Cay and Pigeon Cay during the rainy season. This might be attributed to the presence of artisanal gold mining upstream the Escondido River (Dumailo 2003; MARENA 2010), as artisanal gold mining is known to be a major source of Hg pollution (Cordy et al. 2011). Cadmium was found at intermediate levels in Punta Lora during the dry season and Cu at intermediate levels in all the localities. Urban runoff and sewage are typical sources for these metals, especially when there is no sanitation system, as it is the case (Silva et al. 2001). In addition, antifouling paints, widely used because navigation is the main transportation means in the area, are a potential source for Cu (Wallner-Kersanach et al. 2000). Overall, metal tissue concentrations were higher in Half Way Cay than in Punta Lora during the rainy season, which is consistent because Half Way Cay is located at the sediment deposition zone of the lagoon, whilst Punta Lora is along the outflow of the secondary channel of the lagoon.

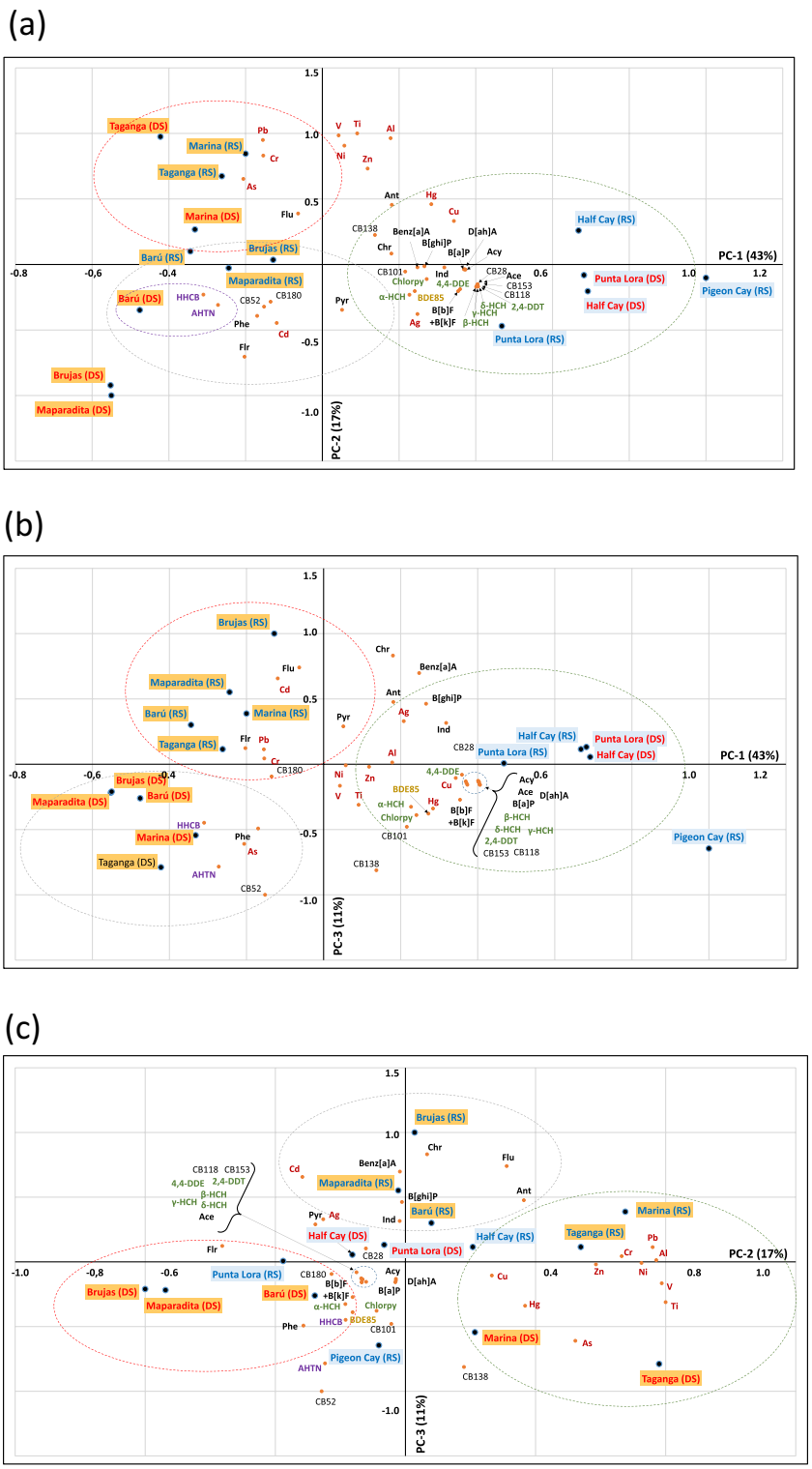

Fig. 4 Principal Component Analysis of the tissue pollutant distributions for oysters, *C. rhizophorae*, sampled in the Nicaraguan and Colombian Caribbean in the rainy and the dry season. Samples and variables are shown in biplots of combinations of the three principal components identified (**a** PC-2 vs PC-1; **b** PC-3 vs PC-1; **c** PC-3 vs PC-2): These three principal components explain 71% of the variance (PC-1 43%; PC-2 17%; and PC-3 11%). Two-three clusters can be distinguished in each biplot amongst the samples and are related to the tissue pollutant distributions in oysters. *RS* rainy season; *DS* dry season

In Colombia, the tissue concentration of As, Cd, Cr, Cu, and Zn in oysters was relatively high when compared with the concentrations reported in other Caribbean localities (Campos 1990; Silva et al. 2001; Cogua et al. 2012). In addition, the tissue concentration of Hg in Santa Marta Marina during the dry season and the tissue concentration of Pb in Isla Barú and Isla Brujas in the rainy season and in Santa Marta Marina and Taganga at both seasons were moderate (according to the criteria of Kimbrough et al. 2008). Extremely high Cd tissue concentrations were recorded in Isla Barú and Isla Brujas during the rainy season and in Isla Maparadita during the dry season, whilst moderate-to-high levels were found in Isla

Brujas during the dry season and in Isla Maparadita during the rainy season (according to NOAA criteria for *Crassostrea virginica;* Kimbrough et al. 2008). The values greatly exceeded those reported for *Crassostrea corteziensis* from coastal lagoons in Mexico as indicative of polluted areas (5.34 μg Cd/g dry-wt; Frias Espiricueta et al. 2009). The presence of high levels of Cd in Isla Brujas and Isla Maparadita could be associated to the industrial activities carried out in these localities, as well as to the influence of sediment loads received from the Dique Channel; however, the concentration of Pb, Cr, and Cd in seawater was determined to be below detection limits (Vivas-Aguas et al. 2014). Thus, the high levels of Cd recorded in the Isla Barú oysters' tissues in the rainy season might suggest that sediments of the Magdalena River could be an important source of metal contamination, because in the absence of detectable metals in seawater (Vivas-Aguas et al. 2014), these were recorded in oyster tissues (e.g., as a result of bioaccumulation). This controversy highlights the need to use biomonitors for metal contamination assessment rather than seawater analysis, as established since the pioneering proposal of Goldberg (1975).

In Santa Marta Marina oysters, the tissue concentration of As, Cu, and Zn can be considered moderate (Kimbrough et al. 2008), like the Ni tissue concentrations in Taganga oysters in the dry season. The main source of metals in this area is likely to be the port activities, since local metal industries and mining are absent (Vivas-Aguas et al. 2014). Thus, antifouling paints and vessel repair activities in the marina constitute the most likely sources of Cu and Zn in Santa Marta Marina (Wallner-Kersanach et al. 2000). The medium tissue concentration of As in Santa Marta Marina oysters and, more specifically, the high concentration of this metal recorded in Taganga oysters during the dry season are difficult to explain. According to geochemical data, As is a major potential contaminant in Colombia, as it is widespread throughout the country in the form of arsenopyrite associated with widely disseminated gold ores (Alonso et al. 2013). However, specific measurements carried out in sediments of the Ciénaga Grande de Santa Marta swamp revealed very low As concentrations (Perdomo et al. 1998). Nevertheless, Taganga cannot be considered a pristine reference site because the tissue levels of musk fragrances, As, and Cr were moderate to high (e.g., 6.39 μg Cr/g dry-wt in *C. rhyzophorae* from Taganga vs. 1–1.8 μg Cr /g dry-wt in *C. rhizophorae* from the nearby central Venezuelan coast; legal limit for shellfish consumption is 5 μg Cr/g dry-wt). The most probable source of these contaminants is the municipal wastewater discharges from Santa Marta that are delivered into Taganga Bay via a submarine outfall. On the other hand, Cd and Pb tissue concentration in oysters were low, in agreement with the low concentration of these metals detected in seawater during the study period (Vivas-Aguas et al. 2014).

Seasonality Except in Taganga, where the tissue concentrations of Al, As, Cr, Ni, Ti, and V were higher in the dry season than in the rainy season, both in Nicaragua and Colombia, the metal tissue concentrations were higher in the rainy season than in the dry season. In contrast, the concentrations of Cd, Cr, and Pb in seawater from Cartagena Bay and Barbacoas Bay were lower in the rainy season in 2012 than in the dry season in 2013 (Vivas-Aguas et al. 2014). It is conceivable that greater loads of suspended matter during the rainy season can contribute to enhance metal uptake and accumulation (Rebelo et al. 2003). Thus, Rebelo et al. (2003) found that the tissue concentration of Cd and Zn were the highest during the rainy season in *C. rhizophorae* from Sepetiba Bay (Brazil) and Alfonso et al. (2013) reported that the tissue concentrations of Al, Cd, and Ni were highest during the rainy season in *C. rhizophorae* from Buche and Mochima estuaries in Venezuela. Likewise, the highest tissue concentration of Pb and Zn in *Crassostrea gigas* and *C. cortezienzes* was recorded in the rainy season (Jara-Marini et al. 2008; Osuna-Martínez et al. 2011). However, contradictory patterns have been reported regarding seasonality of metal tissue concentrations in mangrove oysters. For instance, the Pb tissue concentration was higher in the dry season than in the rainy season in mangrove cupped oysters from Trinidad and Tobago (Kanhai et al. 2014) and in mangrove oysters *C. cortezienzes* from the Gulf of California (Páez-Osuna and Osuna-Martínez 2015). Conversely, Amado-Filho et al. (2008) did not find any significant difference in metal tissue concentrations between rainy and dry seasons in mangrove cupped oysters from Todos os Santos Bay (Brazil). In all these cases, seasonal trends seem to depend on the local variability in hydrodynamics of sediment particles, independently of the season (Rebelo et al. 2003). Thus, Páez-Osuna and Osuna-Martínez (2015) concluded that for the majority of the metals the tissue concentration in mangrove oysters from the Gulf of California, coastal lagoons was greater during the dry season than during the rainy season due to regionally relevant upwelling; whilst for Hg, high levels in the rainy season were associated with transport of materials from the watershed to the lagoon.

Polycyclic aromatic hydrocarbons

Tissue concentrations of PAHs reported in bivalves from Nicaragua within the framework of the International Mussel Watch are in the range of <100 ng/g for ∑PAHs (Sericano et al. 1995). In the present study, slightly higher concentrations were found for total PAHs except for Punta Lora in the rainy season. Nevertheless, the recorded tissue levels of total PAHs can be regarded as low (e.g., in *C. virginica,* values of

∑PAHs in the range of 47–828 ng/g are considered low; Kimbrough et al. 2008). In the case of Colombia, the values of ∑PAHs recorded in all the localities in the rainy season and in Isla Barú and Marina in the dry season were around or below the range of <100 ng/g (Sericano et al. 1995). As a striking exception, the oysters from Isla Brujas had much higher PAH concentrations (medium levels according to Kimbrough et al. 2008). Isla Brujas is located in the vicinity of an oil refinery and terminal nearby the Mamonal industrial complex and receives direct sediment input from the Dique Channel. It has been reported to present high concentrations of dissolved and dispersed hydrocarbons in seawater during the study period, especially in the rainy season (Vivas-Aguas et al. 2014). Similar PAH levels have been recorded in *C. rhizophorae* tissues in other WCR localities. Thus, total PAHs were in the range of 109–362 ng/g dry-wt in Trinidad and Tobago (Kanhai et al. 2015) and in the range of 66.8–240.7 ng/g dry-wt in Guadeloupe in the Lesser Antilles (Ramdine et al. 2012). Nevertheless, despite the low PAH levels, a potential risk cannot be disregarded (Neff et al. 2005) neither in Nicaragua nor in Colombia, as potentially carcinogenic PAHs (PAH_{HMW}; IARC 1987) were recorded in Nicaragua (except in Punta Lora in the rainy season) and in Isla Maparadita and Marina and, most outstandingly, in Isla Brujas in the rainy season. On the other hand, PAH tissue concentrations were considerably greater in the dry season than in the rainy season, especially in Punta Lora for phenanthrene and pyrene, and in all the Colombian localities for phenanthrene.

Regarding the likely origin of the PAHs, the diagnostic ratios Phe/Ant and Flr/Pyr (Neff et al. 2005) could not be calculated as some of the compounds were below detection limits. Instead, the ratio of $_{LMW}$PAHs to $_{HMW}$PAHs was used as chemical indicator of the petrogenic vs pyrolytic origin the PAHs (Baumard et al. 1998; Soclo et al. 2000). The ratio was >1 in Punta Lora and in Marina in the rainy season, indicating a predominant petrogenic origin whilst it was <0.65 in Half Way Cay and Pigeon Cay at both seasons and, most markedly, in Isla Brujas and Isla Maparadita in the rainy season. These results revealed a pyrolytic origin of the PAHs in these localities. In Punta Lora in the dry season, both petrogenic and pyrolytic sources of PAHs seem to be superimposed, as the value of $_{LMW}$PAHs/$_{HMW}$PAHs was close to 1. PAH isomer pair ratios are indicative of whether PAHs are derived from biomass, coal, and petroleum combustion (Yunker et al. 2002; Oros and Ross 2005). Thus, Ind/(Ind + B[ghi]P) isomer pair ratio (Yunker et al. 2002; Oros and Ross 2005) showed that in mangrove cupped oysters from Punta Lora in the dry season and in Pigeon Cay and Half Way Cay, the PAHs were derived primarily from biomass and coal combustion (Ind/(Ind + B[ghi]P) = 0.56–0.58). Meanwhile, this ratio indicated that petroleum combustion was the main source of PAHs in Isla Brujas in the rainy season (Ind/(Ind + B[ghi]P) = 0.20–0.50; Oros and Ross 2005). The likely source for these high PAH concentrations is the nearby oil refinery and asphalt plant (Mamonal industrial zone). The same approach was used to conclude that petroleum combustion was a major source of PAHs in San Francisco Estuary sediments and mussels and that only minor amounts of the PAHs in bivalves were derived from biomass (e.g., grasses, wood, and wood soot) and coal combustion (Pereira et al. 1999; Oros and Ross 2004, 2005).

Seasonality In Nicaragua, $_{LMW}$PAHs were dominant (60% of the total PAHs) in mangrove cupped oysters from Punta Lora in the rainy season whilst $_{LMW}$PAH% values were below 35% in Half Way Cay and Pigeon Cay, and values in Punta Lora in the dry season reached 49%. Hellou et al. (1993) and Gaspare et al. (2009) associated the dominance of $_{LMW}$PAHs to waterborne exposure, contrasting with dominance of $_{HMW}$PAHs for exposure through sediment and suspended matter. According to these criteria, Punta Lora oysters were exposed predominantly to waterborne PAH fractions in the rainy season and those from Half Way Cay and Pigeon Cay in Nicaragua in both seasons. Interestingly, Punta Lora in the dry season seemingly constituted an intermediate case, with oysters exposed to a mixture of water-borne and sediment or suspended matter associated compounds ($_{LMW}$PAHs = 48% of PAH_{total}). In Colombia, $_{LMW}$PAH% values were below 35% in all the localities in the rainy season (in Taganga it could not be calculated) and in Isla Maparadita in the dry season, indicating that oysters were exposed predominantly to waterborne PAH fractions, according to the criteria proposed by Hellou et al. (1993) and Gaspare et al. (2009). Conversely, in Isla Brujas in the dry season ($_{LMW}$PAHs = 48% of PAH_{total}) oysters were seemingly exposed to a mixture of waterborne and sediment or suspended matter associated PAH compounds.

Persistent organic pollutants

Organochlorine pesticides HCHs and DDTs are common pollutants in coastal areas and estuaries resulting mainly from agricultural practices and insect control (de Brito et al. 2002), which seems to be the case of Nicaraguan Caribbean mangroves. Bluefields Lagoon receives water load from the Escondido River basin, where corn, banana, sugarcane, oil palm, and coconut intensive agriculture has promoted the elevated application of pesticides for the last 50 years (GEF-REPCAR 2011; Ebanks-Mongalo et al. 2013). In agreement, dieldrin, DDTs, and terbufos were recently found in water, sediment, and mangrove cupped oysters from Bluefields at concentrations over environmental quality criteria (Ebanks-Mongalo et al. 2013). Likewise, high concentrations of HCHs and DDTs have been reported in water and sediments in other Nicaraguan Caribbean localities (GEF-REPCAR 2011). The use of DDTs is banned but public health institutions have permission to use DDT for sanitation purposes and

DDT derivatives are released to the environment, especially in the rainy season, to fight against mosquitoes (Bodin et al. 2011). Thus, the highest ∑DDT in *C. virginica* from Coastal Lagoons in the Gulf of Mexico was recorded in the rainy season, when the runoff transports DDT previously applied to nearby crop fields and urban areas (Castañeda-Chávez 2011). The levels of ∑DDTs values recorded herein are higher, especially in Punta Lora in the rainy season, than those reported in bivalves from Nicaragua within the framework of the International Mussel Watch (<10 ng/g dry-wt; Sericano et al. 1995). Likewise, they are much higher than in crab eggs (*C. granulate*) from impacted mangroves in Guanabara Bay at Brazil (98 ng/g dry-wt; de Souza et al. 2009). However, the values of ∑DDTs recorded are lower than those recorded in other areas; e.g., ∑DDTs of 156.2 ng/g dry-wt was reported in mangrove cupped oysters from the vicinity of Paranaguá City (Liebezeit et al. 2011) and ∑DDTs as high as 864 ng/g dry-wt was reported in freshwater snails from Hanoy City water canals (Nhan et al. 2001). The predominance of DDE amongst DDT and its derivatives indicates that exposure is chronic or at least long-lasting (Castañeda-Chávez 2011). In agreement, DDE was a major contributor to the DDT in oysters from Paranaguá City, as these were chronically affected by sewage discharge. Alike, DDE was the main POP compound found in tissues of oysters, *C. virginica* and *C. rhizophorae,* from Términos lagoon at the Gulf of Mexico (Carvalho et al. 2009a, 2009b) and in mussels, *Perna perna*, from Brazilian bays (Galvao et al. 2014). Therefore, the high levels of 4.4-DDE found in oysters from Half Way Cay and, most outstandingly, in those from Punta Lora, suggest that the source of DDTs is most likely long-lasting and uninterrupted. Likewise, significant levels of HCHs were recorded in Nicaraguan mangrove cupped oysters, especially in Punta Lora in the rainy season; these HCH levels were, however, of lower magnitude than the DDT levels. As pointed out by Nhan et al. (2001), who also found lower tissue concentrations of HCHs than of DDTs in freshwater snails from water canals in the region of Hanoi, there is no reason to believe that the amounts of HCHs used as pesticide have been lower than the employed amounts of DDT. However, the solubility in water of HCHs is much higher and their half lives much shorter than in the case of DDT, and therefore, the average environmental levels are lower in the long term. In any case, the values of ∑HCHs were similar to those reported in *C. virginica* and *C. rhizophorae* from Mexico (3–32 ng/g dry-wt; Carvalho et al. 2009a) and in bivalves from the WCR (12 ng γ-HCH/g dry-wt; Van Lavieren et al. 2011) but higher than those found in tissues of oysters from Senegal (1 ng/g dry-wt; Bodin et al. 2011) and in freshwater snails, *Angulyagra* sp. from water canals in the region of Hanoi (0.51 ng/g dry-wt; Nhan et al. 2001).

On the other hand, OCPs were below detection levels in the tissues of oysters collected from the Colombian localities in both seasons. In the early 2000s, DDT and its metabolites, heptachlor, aldrin, and HCHs were widespread pollutants in surface waters in Santa Marta Bay as the result of runoff from upstream plantations of coffee and banana, but in the recent decade, these chemicals have been below detection limits in tributary rivers as result of regulatory restrictions for the use of these OCPs (Vivas-Aguas et al. 2014). Similarly, a marked reduction in the OCP levels in seawater and sediments have been recorded in Cartagena Bay and Barbacoas Bay during the last decade (Vivas-Aguas et al. 2010). Thus, in the studied Colombian localities, OCP concentrations in surface waters were below detection limits in the 2012–2013 rainy and dry seasons (Vivas-Aguas et al. 2014), in agreement with our present records of OCP tissue levels in mangrove cupped oysters.

Chlorpyrifos Chlorpyrifos is a broad spectrum organophosphate insecticide widely used on food crops; however, few data are available about its concentration in marine biota. The range of chlorpyrifos in the tissues of marine bivalves along the US coast, including oysters (*C. rhyzophorae* and other species) and mussels, ranges from "non-detected" to 53 ng/g dry-wt, with a regional average of 0.78 ng/g dry-wt (Wade et al. 1998). In the present study, this organophosphate pesticide was only detected in Nicaraguan mangrove cupped oysters (in Half Way Cay in the dry season and Pigeon Cay in the rainy season) in the range of 5–11 ng/g dry-wt and not in the Colombian localities. In agreement, chlorpyrifos was recorded in sediments and mangrove cupped oysters from Bluefields in the 2011 rainy season (Ebanks-Mongalo et al. 2013) but not in surface waters of Cartagena Bay in the 2012–2013 period (Vivas-Aguas etal. 2014). Thus, in Nicaraguan Caribbean mangroves, high levels of chlorpyrifos seemed to be associated with currently ongoing agricultural practices. In Colombia, toxic levels of this organophosphate pesticide were detected in previous years (especially in 2011) in surface waters influenced by the Dique Channel input (intensive agriculture in the Magdalena River basin) and by agrochemical industry sited at the harbor area (Vivas-Aguas et al. 2010), but the levels were below detection limits in the 2012–2013 period (Vivas-Aguas et al. 2010). It seems that the environmental levels of this pesticide can be highly erratic, with transient peaks after recent application or discharges followed by low levels after a whilst. Indeed, chlorpyrifos has low solubility in water (thus, it is usually associated to suspended organic matter) and a relative short half-life (~7 days), and is volatile in water and metabolised by aquatic animals (Serrano et al. 1997). Consequently, the absence of a seasonal or geographical pattern can be associated to changes in the application and timing, so that detectable tissue concentrations might correspond to samples collected just after recent applications of the pesticide, as suggested by Wade et al. (1998). Nevertheless, this should not be overlooked because short

pulses of chlorpyrifos can exert long-term toxicity to marine invertebrates (CCME 2008).

Musk fragrances Synthetic musks are ubiquitous contaminants in the marine environment. Galaxolide (HHCB) and Tonalide (AHTN) are the major musk fragrances in sewage discharged into estuaries. The tissue concentration of musks recorded in Isla Barú, Isla Brujas, and Isla Maparadita in the rainy season are within the range of those previously reported for marine molluscs (e.g., <12 ng/g dry-wt for HHCB and <17 ng/g dry-wt for AHTN in North Sea mussels; Rudel et al. 2006). However, much higher concentrations were recorded in the dry season (interestingly, the top touristic season) in all the localities, in the range of 15–70 ng/g dry-wt for HHCB and 25–50 ng/g dry-wt for AHTN. Nevertheless, these values are still much below those recorded by for HHCB in mussels (1700 ng/g lipid = ~140 ng/g dry-wt; assuming 1% fat and 80% hydration) and clams (3000 ng/g lipid = ~240 ng/g dry-wt; assuming 1% fat and 80% hydration) from densely populated and industrialised zones of Canada (Gartemann et al. 1999). The high HHCB and AHTN concentrations in Taganga Bay and Isla Brujas during the dry season are most probably due to wastewater discharges from nearby sewage outfalls.

Polychlorinated biphenyls The levels of total PCBs recorded in Nicaraguan *C. rhizophorae* were lower than those recorded in crab eggs from impacted mangroves in Guanabara Bay in Brazil (570 ng/g dry-wt; de Souza et al. 2009) but higher than ∑PCBs reported in bivalves from Nicaragua within the framework of the International Mussel Watch (<10 ng/g dry-wt; Sericano et al. 1995) and for freshwater snails from the region of Hanoi (<60 ng/g dry-wt; Nhan et al. 2001). PCBs are essentially of industrial origin and therefore the tissue concentrations found in Nicaraguan mangrove cupped oysters are much lower than those found in harbors and industrial areas of industrialised countries (e.g., 2500 ng/g in mussels from Marseille harbor in France; Villeneuve et al. 1999). Nevertheless, remarkable tissue concentrations of CB28, CB118 and CB153 were recorded in all the Nicaraguan localities and seasons whilst CB52, CB101, CB138, and CB180 were found at lower concentrations. All these PCB congeners have been recognised as non- or poorly metabolised in molluscs and therefore they are bioaccumulated (Kannan et al. 1995; Nhan et al. 2001). Similarly, CB28 and CB153 were the two individual PCBs recorded at highest tissue concentrations in brown mussels (*Perna perna*) from south eastern Brazil bays (Galvao et al. 2014). Data on seasonality are not conclusive. In the present study, no seasonal trend was observed for CB28, CB118, and CB153 tissue concentrations, but CB52, CB101, CB138, and CB180 peaked up at the dry season. In contrast, *Crassostrea gasar* collected from the Saloum Delta (Senegal) had systematically higher PCB levels (by a factor of 2–3) during the rainy season compared to individuals sampled in the dry season (Bodin et al. 2011).

Polybrominated diphenyl ethers PBDEs are bioaccumulative halogenated compounds used as flame retardants in automobile, textile, and, mainly, electronics industries (Kimbrough et al. 2009) and are considered to be emerging environmental contaminants. BDE85 showed markedly high values in the rainy season in Pigeon Cay and Punta Lora, with values of total PBDEs in the range of 89–325 ng/g dry-wt. These values are much higher than previously reported in bivalves (Moon et al. 2007; Oros et al. 2007) and cannot be associated with a particular source at local scale.

Seasonality The tissue concentrations of HCHs, DDTs, chlorpyrifos, PCBs, and PBDEs in Nicaraguan mangrove cupped oysters were higher in the rainy season than in the dry season, especially for ∑OCPs in Punta Lora. In contrast, the levels of POPs in Colombian mangrove cupped oysters were higher during the dry season than during the rainy season, especially in Santa Marta Marina and Taganga. It seems, therefore, that the seasonal trend in the tissue concentration of POPs is not the same in Nicaraguan and Colombian oysters. In *C. gasar* from Senegal, high tissue concentrations of POPs in the rainy season were attributed to recent inputs from the surroundings through rainfall and river runoff (Bodin et al. 2011). Likewise, the highest POP tissue concentrations in *C. virginica* from Coastal Lagoons in the Gulf of Mexico were also recorded in the rainy season (Castañeda-Chávez 2011). It is conceivable that high POP tissue concentrations in the rainy season are the consequence of enhanced runoff that transports POPs from crop fields and urban areas to receiving waterways. This can be especially relevant in tropical countries where rainfall is intense during the rainy season and intensive agricultural practices lead to a high input of pesticides over watersheds (Daam and Van Den Brink 2010), like in the Nicaraguan mangroves, and less relevant in more industrial areas such as the Colombian mangrove-lined coastal systems studied herein.

Integrated chemical contamination assessment

Based on the determination of contaminant tissue levels in *C. rhizophorae*, the present study revealed that the chemical contamination profile was different between the two studied geographical areas, Nicaragua and Colombia; as shown by both radar plots (Fig. 2) and PCA (Fig. 4). Likewise, the levels of pollutants were different, as clearly reflected in the CPI and PLI values (Figs. 3a–d). These indices were significantly correlated to each other following a linear regression model (Fig. 3e), and therefore, they seem to be fully comparable integrative estimates of pollutants' levels. In addition, seasonality was a crucial factor affecting chemical contamination. Overall, low tissue concentrations of metals (except Hg) and

PAHs; moderate-to-high tissue concentrations of Hg, HCHs, and DDTs; detectable levels of chlorpyrifos, PCBs (mainly CB28, CB118, CB138 and CB 153), and BDE85 (in Pigeon Cay); and negligible levels of musks were recorded in Nicaraguan oysters. Conversely, a distinct profile of POPs was identified in Colombia, where the tissue concentrations of HCHs and DDTs were practically negligible, chlorpyrifos and PBDEs were below detection limits, and the tissue levels of PCBs were low (CB28 in the rainy season in Isla Brujas, Isla Maparadita, and Marina; and CB52 in all the localities in the dry season). In contrast, noticeable tissue concentrations of musk fragrances were recorded in all the localities in the dry season, and the levels of Ag, As, Cd, Pb, and PAHs in several localities and in particular seasons ranged from moderate to extremely high. In the WCR, mean values of 12 ng γ-HCH/g dry-wt, 4 ng PCBs/g dry-wt and 1 ng 4,4-DDE/g dry-wt have been reported in the tissues of *C. rhizophorae* (Van Lavieren et al. 2011). The values recorded in Nicaraguan mangrove cupped oysters largely exceeded these mean values reported for the WCR, whereas only the levels of PCBs were occasionally surpassed in Colombia. These profiles and levels of contaminants are useful to identify potential pollution problems and pollutant sources in the studied regions but they do not provide reliable indication of the deleterious effects that these pollutants may exert to biota and ecosystems, as recommended for pollution monitoring programmes (OSPAR 2013). For this reason, biological effects assessment was carried out in a parallel investigation aimed at relating oyster health condition to pollutant tissue levels (Aguirre-Rubí et al. submitted).

Moreover, it can be concluded that mangrove cupped oysters, *C. rhizophorae*, are suitable biomonitors for chemical contamination assessment in mangrove-lined coastal systems at regional scale in the Caribbean, including diverse habitats such as lagoons and swamps and over a wide geographical coastal region from Nicaragua to Colombia. Nevertheless, seasonality is crucial to properly design and conduct biomonitoring programmes. National and regional monitoring programmes such as IMA in Trinidad and Tobago (Siung-Chang 1997), GIWA in the WCR (UNEP 2006) and REDCAM in Colombia (Vivas-Aguas et al. 2014) would be greatly improved by including the tissue concentrations of chemical contaminants in mangrove cupped oysters in addition to the measurement of the levels of contaminants in water and sediments, as it is regularly done in other regions of the world such as Pacific Asia, Europe, Canada, and the USA (Monirith et al. 2003; Kimbrough et al. 2008; Zampoukas et al. 2014).

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