



Nutrients and trace elements of semi-arid dwarf and fully developed mangrove soils, northwestern Venezuela

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Abstract

Mangrove forests are characterized by a high carbon storage capacity. Anthropogenic activities and the climate change are threatening the stability of these ecosystems, specially those that are surviving on extreme environmental conditions, representing the most susceptible groups. This study aims to understand the geochemical differences of soils of mangrove forest with diverse degree of development of *Rhizophora mangle* and *Avicennia germinans*, located in two semi-arid regions from the Falcon state, Venezuela. The soil samples were collected in *Punta Caimán* region (dwarf mangroves of less than 2 meters height) and *Boca de caño* lagoon (fully developed mangroves of up to 8 meters height). The trace elements and macronutrients were determined in two sequential extraction, first with 1M *HCl* followed by an extraction with ultrapure grade *HNO₃*. Results showed that, in general, dwarf mangroves have limited capability to accumulate organic matter in the soil (average TOC of $0.6 \pm 0.2\%$) and that the sampled surface soils from the dwarf mangrove region had no differences with deeper material. On the other hand, the TOC of soils dominated by developed mangroves is $6 \pm 2\%$, representative of semi-arid conditions. Moreover, the soils dominated by dwarf mangroves are characterized high concentrations of trace elements related to the chalcophile group. The difference in soil chemistry between *Rhizophora mangle* and *Avicennia germinans* becomes significant when mangroves are fully developed. Finally, soils of dwarf mangrove forests presented a greater concentration of trace elements than soils of fully developed mangroves, which might be due to a different sediment source, to an anthropogenic influence or to the difference in organic matter.

Keywords Soil geochemistry · Mangroves · Nutrients · Trace elements

Introduction

Mangrove ecosystems are known for their net carbon sink characteristic, playing an important role in the global carbon cycle and in the preservation of wide number of species. Although they only occupy relative small area of the tropical and subtropical coasts, they are considered as blue carbon ecosystems due to their high carbon sink capacity (Alongi 2009; Perry and Taylor 2009; Shiau and Chiu 2020).

The development of a mangrove forest is influenced by several factors, *e.g.* solar radiation, temperature, pore-water

salinity, availability of oxygen, water and nutrients (Alongi 2009). The most important nutrients controlling the size, structure and production of mangrove forests are phosphorus and nitrogen (Reef et al. 2010); however, other elements classified as micronutrients (*e.g.* Zn, Cu, Co, Mn and Fe) are also relevant for the health of the ecosystem, although their concentration in the soil can be very low (Alongi 2017; Morel et al. 2006). The availability of these elements depends on their respective chemical species—or how they are present in the soil—which in turn depends on the physical chemical properties of the system (de Elias Onofre et al. 2007; Marchand et al. 2012). In regions of low water and nutrient availability (mainly N or P), and high pore-water salinity the growth of the mangrove plant is affected and it is common to find mangrove less than 2 m height, defined as dwarf mangroves, in such conditions (Feller et al. 2003; Medina et al. 2010).

The relative high rate of organic matter accumulation in mangrove soils and their high soil water content contribute to a predominance of anoxic conditions, allowing the

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activity of sulfate-reducing bacteria that precipitate sulfides of trace elements (Clark et al. 1998). Therefore, many mangrove soils are considered to store trace elements, associated to the chalcophile group, in an unavailable form to the mangrove (Clark et al. 1998; Marchand et al. 2006b). On the other hand, human activities, environmental changes, soil respiration or bioturbation are often altering the physical chemical conditions of the soils, allowing oxic conditions to prevail (Angeli et al. 2019; Araújo et al. 2012; Lacerda et al. 1993; Otero et al. 2017a). These alterations can dissolve the secondary sulphide minerals and release trace elements in a soluble phase, which, in some scenarios, can contaminate the mangrove ecosystem or be exported to the marine environment (de Elias Onofre et al. 2007; Marchand et al. 2012; Lacerda et al. 1993).

In this study, we have selected two different regions of mangrove soils, in which the surficial soils were previously characterized (Romero and Melendez 2013). These two regions present the same mangrove species (*Rhizophora mangle* and *Avicennia germinans*), with a defined zonation where *Rhizophora mangle* dominates the areas near the shore; present the same climate characteristics, a semiarid climate with sporadic heavy rain of short duration; and the attributes of the mangrove species were found to be the same as for mangroves of dry regions with strong seasonal characteristics (López et al. 2011). The development of the mangroves from the two studied regions is contrasting. Romero and Melendez (2013) suggest that the difference in the development is due to a phosphorus deficiency in the dwarf mangrove forest linked to removal of organic matter by action of tides and waves.

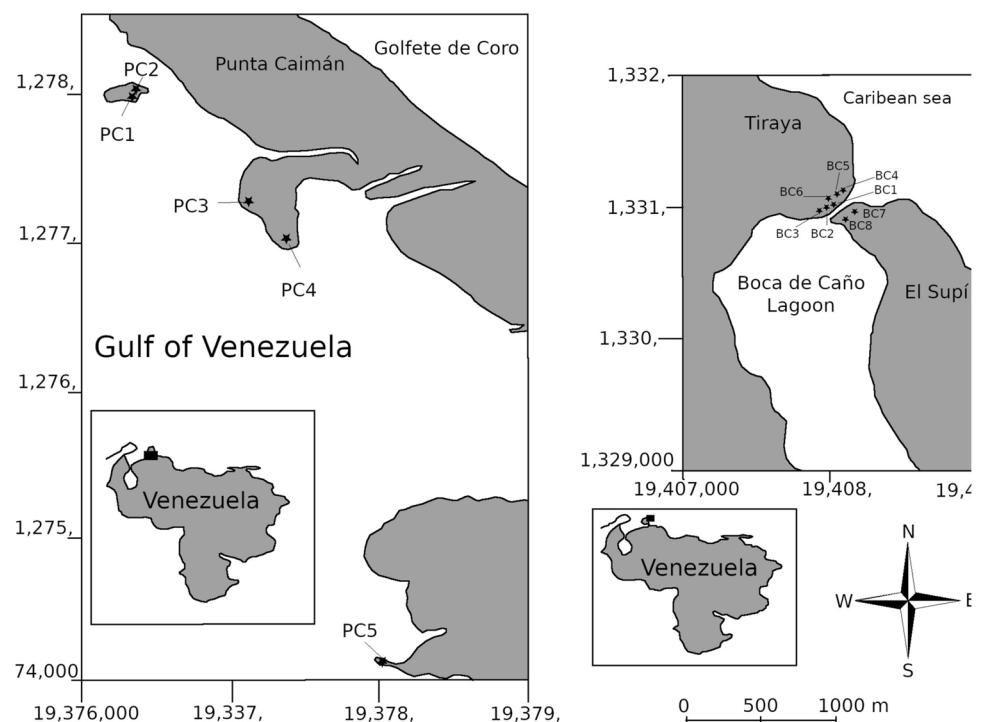
Nevertheless, the concentration of trace elements and the soils profiles were not addressed. The degree of development of the mangrove species will impact the biogeochemical processes occurring in the soil and, therefore, will have an effect on the concentration of chemical species. This study aims to determine which chemical species reflect this difference by analyzing the soil geochemistry at different depths.

Geological setting and methods

The study area is located in Falcon's basin, characterized by a transgression period during the Oligocene, the sedimentation took place in a marine environment; during the Miocene, the subsidence stopped and the environment of sedimentation changed to a continental type. The mineralogical composition of sediments from the Coro Gulf is influenced by the transported sediment by the *Mitare* river. This river flows through a large diversity of lithologies belonging to the sub-basin of west Falcon. In contrast, the geology of *Península de Paraguaná* consists of shale and limestone with high fossil contents, granite, sandstone, siltstone, metashales and ultramafic rocks (González et al. 1980).

In the region of *Punta Caimán* (PC), there is a small fishing community exploiting fish and, in minor quantity, oysters as main resources. This community depends on the stability of the mangrove ecosystem. In this region, the mangrove ecosystems are prone to a mixed semidiurnal tide: two high tides and two low tides of different heights and a lunar cycle of 24 h, 50 min and 28 seconds (Quintero and Terejova 2008).

Fig. 1 The location of the two study areas. Coordinates in meters, projection used is UTM



On the other hand, *Boca de Caño* (BC) is a wildlife refuge and fishing reserve, presenting relatively taller mangrove forests of *Rhizophora mangle* and *Avicennia germinans* than the previously mentioned region, about 6–8 m high, to the north of the lagoon, and relative short mangrove forests of the same species to the south, with less than 3 m height.

We defined five soil sampling points in *Punta Caimán* on February the sixth of 2010, and eight sampling points in the Wildlife refuge and fishing reserve of *Boca de Caño* Lagoon on July the third of 2010, both sites in Falcon state, in Venezuela (Fig. 1; Table 1). We collected samples at different depths (approximately 25 cm, 60 cm and 90 cm) in order to account for chemical changes in the soil profile. We measured the pH and the oxidation-reduction potential (ORP) of the bulk soil *in situ* by using a glass electrode and an oxidation reduction potential pocket probe (Hanna), respectively. We collected interstitial water samples in each sampling site and determined the salinity following the method explained in Wallace (1974), using the chloride concentration, determined by ionic chromatography (IC), HPLC WATERS.

We dried the soil samples at ambient temperature and sieved them using a stainless sieve with 2 mm of diameter. We collected a sub-sample by dividing it by quarters, we ground each sub-sample and homogenized them using an agate mortar. We carried out a sequential extraction by using 1 M *HCl*—associated to the adsorbed species, the amorphous oxide-hydroxides and carbonate minerals—and using ultrapure grade *HNO₃*—associated to crystalline phase of oxide-hydroxides, sulphides, organic matter and partial dissolution of clay minerals—(Araújo et al. 2012). We analyzed

the solutions using an inductively coupled plasma atomic emission spectrometer (ICP-AES) using a Perkin Elmer spectrometer, model Optime 3000, to determine the concentration of Fe, Al, Ni, Mn, Cu, Pb, Zn, Cr, Cd and Ca. Phosphorus was determined by implementing the modified Murphy and Riley photocolometric method for geological materials (Romero and Melendez 2013). Mercury was determined applying a direct analyzer Milestone DMA-80, while total carbon was measured using carbon analyzer LECO C-144; the percentage of organic carbon was determined implementing the Walkley and Black (1934) method; the total nitrogen was determined using the Jackson and Beltrán (1964) modified Kjeldahl method. Moreover, we conducted a mineralogical analysis using X-ray diffraction (XRD) crystalline powder method using a Bruker D8 advance.

We carried out multivariate statistics analyses after applying a normality test and adjusting the variables to a normal distribution. The multivariate analysis constituted of principal component analysis (PCA) and dendrograms for grouping samples for each study area. The number of groups of the dendrograms were defined based on the Elbow graphical method.

Results

In general, all soil cores presented a loamy texture with exception of sampling locations PC5 in *Punta Caimán* and small mangroves of *Boca de Caño* (BC7 and BC8), in which a sandy texture was dominant. Moreover, the zones between mangrove species in both regions was evident: the *Rhizophora mangle* dominated the areas near the shore, whereas *Avicennia germinans* grew farther from the shore. These zones followed a significant different salinity pattern observed in the pore-water (Table 1), with greater salinity observed in soils dominated by *Avicennia germinans*, with an average of 58 ± 7 ; and with a pore-water salinity of soils dominated by *Rhizophora mangle* with an average of 48 ± 5 (Romero and Melendez 2013).

The fully developed mangrove soils of the *Boca de Caño* region (BC) were of diverse characteristics: the sampling locations BC1, BC2 and BC3 (dominated by *Rhizophora mangle*) presented a contrasting thick organic-rich layer (more than 25 cm) with abundant clay texture at depth and oxides-hydroxides in small amount; the sampling locations BC4, BC5 and BC6 (soils dominated by *Avicennia germinans*) were characterized by a thin organic rich layer (less than 4 cm). The small mangroves from this region, sampling locations BC7 and BC8, characterized by 3 m height mangroves, did not present a significant organic-rich layer at the top layer. The mineralogical composition of mangrove soils in *Boca de Caño* region was characterized by quartz, plagioclase, calcite and clay minerals. On the other hand, the soils

Table 1 Site coordinates of *Punta Caimán* (PC) and *Boca de Caño* (BC) soil samples; Field Observations and pore-water salinity

Sample	Lat(mN)	Lon(19P mE)	Vegetation/height	Salinity (‰)
BC1	1,330,668	407.810	R. 8 m	40.9*
BC2	1,330,643	407.777	R. 8 m	46.2*
BC3	1,330,630	407.763	R. 8 m	47.1*
BC4	1,330,756	407.861	A. 8 m (submerged soil)	47.6*
BC5	1,330,736	407.824	A. 8 m	58.2*
BC6	1,330,702	407.773	A. 8 m	59.5*
BC7	1,330,568	407.854	R. 3 m	49.3*
BC8	1,330,562	407.835	A. 3 m	53.3*
PC1	1,277,617	376.256	Dwarf A. and R.	46.4
PC2	1,277,663	376.251	Dwarf A.	70.4
PC3	1,276,785	377.052	Dwarf R.	55.4
PC4	1,276,639	377.234	Dwarf and dry A.	55.4
PC5	1,273,733	377.979	Dwarf A.	61.0

*The Cl concentration was estimated by the ratio Na/Cl of 0.56

A.: *Avicennia germinans*. R.: *Rhizophora mangle*

The salinity of the seawater collected in *Boca de Caño* was 43.2 (‰)

of dwarf mangroves from *Punta Caimán* (PC), with exception of sampling location PC2, did not show a contrasting organic layer but a thin grayish layer (less than 5 cm). The mineralogical composition of soils from *Punta Caimán* was characterized by quartz and clay minerals, with significant amount of hematite. Halite was identified in all samples due to the high salinity of the pore-water. Moreover, thin

oxide-hydroxide layers were observed in all sampling locations where *Rhizophora mangle* was dominant.

Physical and chemical analyses

The ORP measurements, represented by the Eh values, are semi-quantitative given that a more representative value of the bulk soil ORP is obtained by using different probes with a defined distance between them. Therefore, the Eh values are not discussed into detail. The Eh vs. pH plot showed two distinguished groups, separately, mainly, by pH (Fig. 2). The more acidic samples corresponded to soils samples with high organic content, > 4 % (Fig. 3). The more basic soils corresponded to soils of dwarf mangroves with low organic content, < 2 %. However, one sample of *Punta Caimán*, PC2, located far from the shore in a small island, presented less pH (6.9) than the other soils dominated by dwarf mangroves (generally > 7.5), similar to the fully developed ones from *Boca de Caño*, with pH values of about 7 or less; this sample (PC2) had significantly greater organic carbon, 5.46 %, than other samples from *Punta Caimán*, less than 0.8 % (Fig. 3).

The soil profiles of developed mangrove soils from *Boca de Caño* showed a decrease in total organic carbon (TOC) content with depth, with average values of $6 \pm 2\%$ and $1.0 \pm 0.9\%$ at the top and bottom, respectively; soils related to dwarf mangrove forest from *Punta Caimán* region (with exception of sampling location PC2) did not present difference on organic content with depth (average of $0.6 \pm 0.2\%$, Fig. 3). Soils of developed mangroves displayed a decreasing

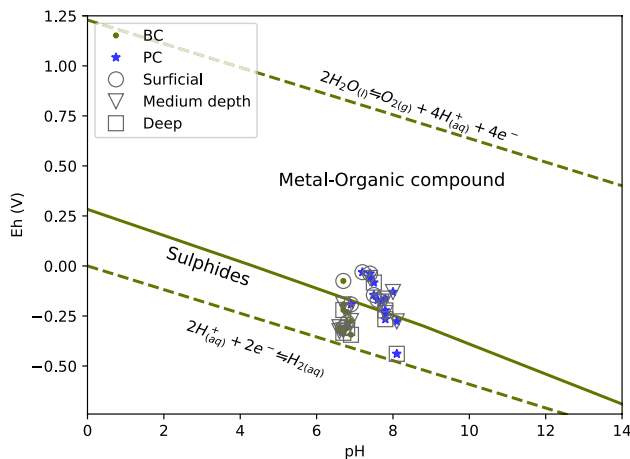
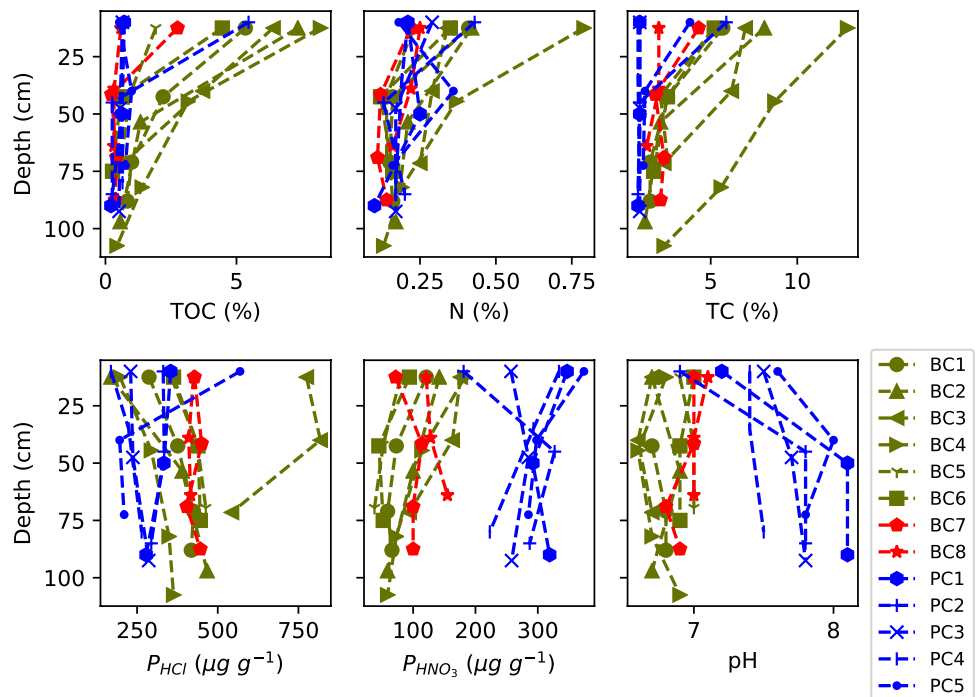


Fig. 2 Eh vs pH diagram for iron species in presence of sulphides. Calculations were done using the software “The Geochemist’s Workbench” version 11.0.5 and the dataset “thermos.com.v8.R6+.tdat” (Bethke and Yeakel 2016). Soil samples from *Punta Caimán* (PC) and *Boca de Caño* (BC) studied sites. Surficial, medium and deep samples correspond to < 25 cm, from 25 to 50 cm and > 50 cm, respectively

Fig. 3 Behavior of Carbon, nitrogen, P and pH in sampled soils from *Punta Caimán* (PC) and *Boca de Caño* (BC) with depth (cm). Green, red and blue colors stand for samplings sites of developed mangroves from BC, underdeveloped mangroves from BC and dwarf mangroves from PC, respectively



trend of nitrogen concentrations with depth, from $0.4 \pm 0.2\%$ at the top layer and $0.18 \pm 0.07\%$ in deeper layers; in *Punta Caimán*, the average of N concentration in all soil samples of dwarf mangroves was $0.20 \pm 0.07\%$, excluding the top of the sampling location PC2, which presented $0.43 \pm 0.05\%$ of N. The TC concentration in sampled soils followed a similar behavior with depth than the one observed for TOC; however, the top layer of the sampling location PC5 presented a significant increase of TC ($3.8 \pm 0.5\%$) as compared to

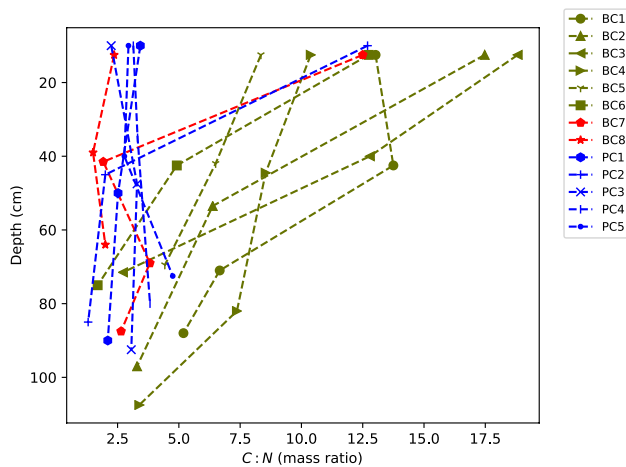
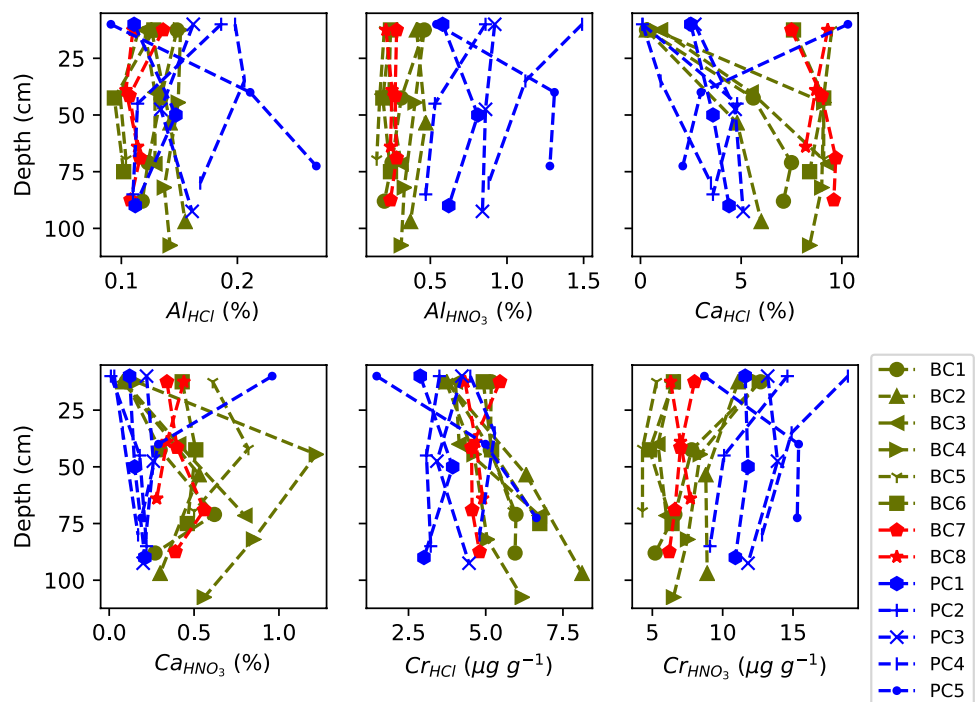


Fig. 4 Organic carbon to total nitrogen mass ratios in sampled soils from *Punta Caimán* (PC) and *Boca de Caño* (BC) with depth (cm). Green, red and blue colors stand for samplings sites of developed mangroves from BC, underdeveloped mangroves from BC and dwarf mangroves from PC, respectively

TOC values ($0.53 \pm 0.01\%$). In general, both regions showed an invariant P concentrations from the HCl extraction with depth. However, the P concentrations extracted using HNO_3 were different between both regions; the soils from *Boca de Caño* displayed less P_{HNO_3} than soils from *Punta Caimán*, average of $100 \pm 40ppm$ and $300 \pm 40ppm$, respectively, without including the top sample at point PC2, where P_{HNO_3} concentration was $180 \pm 10ppm$. The change with depth of C:N mass ratios in soils (Fig. 4) was similar to the observed for organic carbon (OC); the developed soils from *Boca de Caño* and the sampling location PC2 from *Punta Caimán* displayed a decrease of C:N ratios from about 10 at the top to 5 at deeper layers; however, the rest of the soils of dwarf mangroves showed C:N ratios less than 5. Furthermore, the fully developed mangroves (from samples BC1 to BC6) showed greater C:N ratios than the medium developed mangroves (samples BC7 and BC8), where the top layer of soils of *Rhizophora mangle* exhibit the highest C:N ratios (Fig. 5).

The average Al_{HCl} in soils from *Boca de Caño* and from *Punta Caimán* were $0.12 \pm 0.02\%$ and $0.16 \pm 0.05\%$, respectively, excluding the superficial sample from point PC5, which had a Al_{HCl} concentration of $0.091 \pm 0.002\%$ and showed increasing concentrations with depth until $0.268 \pm 0.002\%$. The aluminum concentrations from the HNO_3 extraction (Al_{HNO_3}) were greater in dwarf mangrove soils from *Punta Caimán*, with an average of $0.9 \pm 0.3\%$, compared to soils from *Boca de Caño*, with an average of $0.29 \pm 0.09\%$. In soils from *Boca de Caño* the Al_{HNO_3} concentrations did not change with depth; however, in the dwarf mangrove soils from *Punta Caimán* the concentrations

Fig. 5 Behavior of aluminum, calcium and chromium concentrations of HCl and HNO_3 extractions in soil samples from *Punta Caimán* (PC) and *Boca de Caño* (BC) with depth (cm). Green, red and blue colors stand for samplings sites of developed mangroves from BC, underdeveloped mangroves from BC and dwarf mangroves from PC, respectively



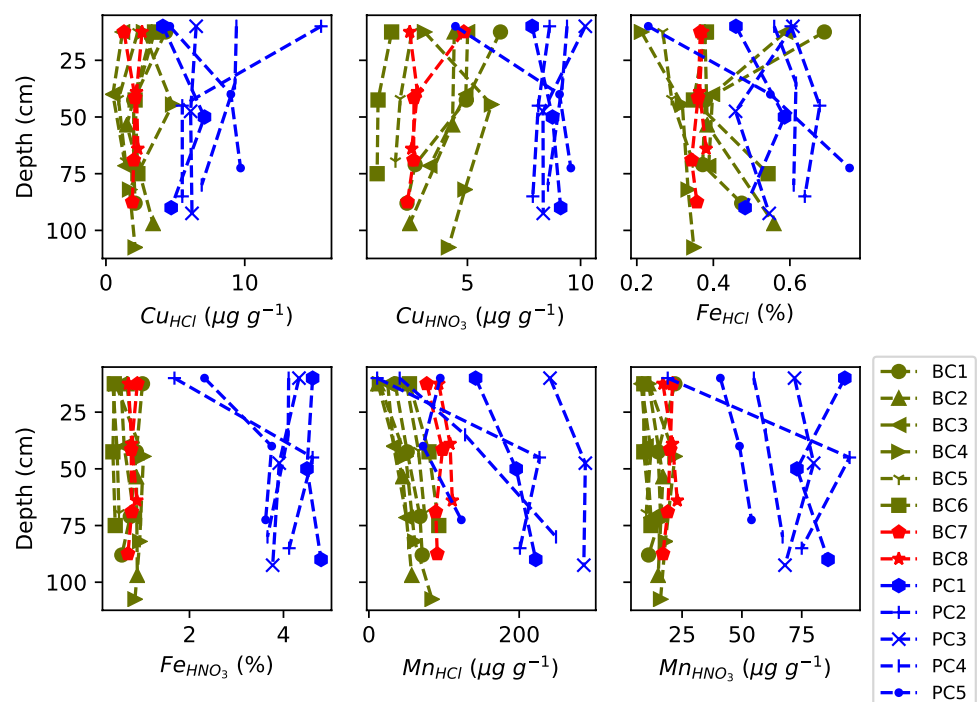
tended to decrease with depth, with exception of sampling location PC5, where Al_{HNO_3} increased with depth. The Ca_{HCl} concentrations in soils of developed mangroves from *Boca de Caño* were $< 1\%$ in the top layer and increased towards the bottom with an average of $8 \pm 2\%$. However, the soils from sampling location BC7 and BC8 (mangroves of 3 m height) did not show a change in Ca_{HCl} concentrations with depth (average of $8.9 \pm 0.8\%$). The Ca_{HNO_3} concentration in soils of dwarf mangroves from *Punta Caimán* had an average of $0.17 \pm 0.08\%$ (excluding the superficial sample of PC5) and did not change with depth; however, the Ca_{HNO_3} concentrations of sampling location PC5 tended to decrease with depth, from $0.96 \pm 0.03\%$ at the top to $0.19 \pm 0.03\%$ at the bottom. Furthermore, the soils of dwarf mangroves from *Boca de Caño* did not display a change in Ca_{HNO_3} concentrations with depth, average of $0.39 \pm 0.09\%$.

Cr concentrations in soils, on the other hand, did not vary with depth in the dwarf mangrove area from *Boca de Caño*, with $4.8 \pm 0.4 ppm$ and $7.0 \pm 0.7 ppm$ for Cr_{HCl} and Cr_{HNO_3} , respectively. The Cr_{HCl} concentrations in soils of developed mangroves from *Boca de Caño* increased with depth, from $< 5 ppm$ at the top to approximately 7 ppm at the bottom, a similar behavior was observed for sampling location PC5 from *Punta Caimán*. In general, the Cr_{HNO_3} concentrations in soils from *Punta Caimán* were greater than in soils from *Boca de Caño*. The Cu concentrations of both acid extractions were greater in soils from *Punta Caimán* than in those from *Boca de Caño*, with no significant trend observed with depth (Fig. 6). The Fe_{HCl} concentrations did not vary with depth in soil samples related to dwarf mangroves from both

studied regions, with exception of sampling location PC5, where an increasing trend with depth was detected, from $0.230 \pm 0.007\%$ at the top and $0.756 \pm 0.007\%$ at bottom. The Fe_{HCl} concentration related to fully developed mangroves from *Boca de Caño* presented a contrasting behavior with depth: the soils associated to *Rhizophora mangle* had greater Fe_{HCl} concentration at the top layer (average of $0.6 \pm 0.2\%$), the concentration tended to decrease towards a depth of 50 cm with about $0.4 \pm 0.2\%$ and tended to increase towards deeper regions; whereas the Fe_{HCl} concentration related to *Avicenia germinans* was rather constant, with exception of sampling location BC4, which is related to a submerged soil and the Fe_{HCl} concentration was found to be the lowest measured in this study ($0.212 \pm 0.007\%$). The behavior of Fe_{HNO_3} concentrations in soils with depth was found to be similar than the one observed for Mn_{HNO_3} . Moreover, the soils from *Punta Caimán* presented greater concentrations of Fe_{HNO_3} and Mn_{HNO_3} (average of $4.0 \pm 0.6\%$ and $70 \pm 20 ppm$, respectively) than the soils from *Boca de Caño* (average of $0.7 \pm 0.2\%$ and $15 \pm 5 ppm$, respectively). The Mn_{HCl} concentrations in soils from both regions showed a slight increased with depth.

The Ni_{HCl} and Zn_{HCl} concentrations in soils of developed mangroves from *Boca de Caño* did not change significantly with depth (Fig. 6), presented on average values of $5 \pm 1 ppm$ and $130 \pm 30 ppm$, respectively. On the other hand, the Pb_{HCl} concentrations were greater at the top layer of soils related to developed mangroves, with an average of $5 \pm 2 ppm$. The concentrations of Ni_{HCl} , Pb_{HCl} and Zn_{HCl} in soils of dwarf mangroves from *Punta Caimán* increased with depth, and

Fig. 6 Behavior of copper, iron and manganese concentrations of HCl and HNO_3 extractions in soil samples from *Punta Caimán* (PC) and *Boca de Caño* (BC) with depth (cm). Green, red and blue colors stand for samplings sites of developed mangroves from BC, underdeveloped mangroves from BC and dwarf mangroves from PC, respectively



were greater than the ones observed in soils from *Boca de Caño*, with averaged values of $8 \pm 2\text{ppm}$, $8 \pm 2\text{ppm}$ and $220 \pm 50\text{ppm}$, respectively. The concentrations of Ni_{HNO_3} , Pb_{HNO_3} and Zn_{HNO_3} in soils from *Punta Caimán* were greater than in soils from *Boca de Caño* (Fig. 7). The Cd_{HCl} and Hg concentrations of soils from *Boca de Caño* were on average $110 \pm 30\text{ppb}$ and $4 \pm 3\text{ppb}$, respectively; on the other hand, the measured Cd_{HCl} and Hg concentrations for the soils from *Punta Caimán* exhibited greater concentrations for both, with averages of $230 \pm 70\text{ppb}$ and $30 \pm 20\text{ppb}$, respectively (Fig. 8). The Cd_{HNO_3} concentrations for all samples were below detection limit. The Hg concentration in soils from *Punta Caimán* were in a similar range than the ones observed in other mangroves soils of Venezuela (Otero et al. 2017b).

Multivariate statistics

A principal component analysis (PCA) of the results (excluding Eh, pH and Hg values, because they were not measured for all samples) showed that there are two distinguished groups classified by their structural development (Fig. 9). The PC1 and PC2 accounted for 73.26 % of total variance. The PC1 described most of the variability, where trace elements associated to the chalcophile group are strongly related to the soils from the dwarf mangrove region. Whereas the Total organic carbon and nitrogen concentrations described the PC2, where only few samples are

grouped, specially the surficial samples with high TOC content. The small mangroves (of about 3 m height) from *Boca de Caño* (group 3 in Fig. 9) are grouped by their relative greater Ca and low TOC content. Furthermore, Cr_{HCl} concentration represents the only trace element fraction that is more related to the soils of *Boca de Caño* region.

The results obtained from the dendrograms suggested that mangrove soils from *Boca de Caño* Lagoon were classified into three groups (Fig. 10), the first group corresponded to superficial soil samples associated to fully developed *Rhizophora mangle* of approximately 8 meters height; the second group corresponded to soils of fully developed *Avicennia germinans* and underdeveloped mangroves, without significant differences between superficial and deeper samples; the last group is constituted by only one sample, which is the superficial sample of a submerged soil associated to fully developed *Avicennia germinans*. This sample showed no relationship to other soil samples, and it differentiated from other superficial soil samples of *Avicennia germinans* because of its greater concentration of organic carbon. On the other hand, in mangrove soils from the *Punta Caimán*, two groups were identified by multivariate analysis (Fig. 11). The first group comprised almost the totality of samples, without differences among depth or mangrove species. The second group corresponded to one superficial soil sample associated to *Avicennia germinans*, this soil sample was characterized by its sandy texture and by generally less concentrations of trace elements.

Fig. 7 Behavior of nickel, lead and zinc concentrations of HCl and HNO_3 extractions in soil samples from *Punta Caimán* (PC) and *Boca de Caño* (BC) with depth (cm). Green, red and blue colors stand for samplings sites of developed mangroves from BC, underdeveloped mangroves from BC and dwarf mangroves from PC, respectively

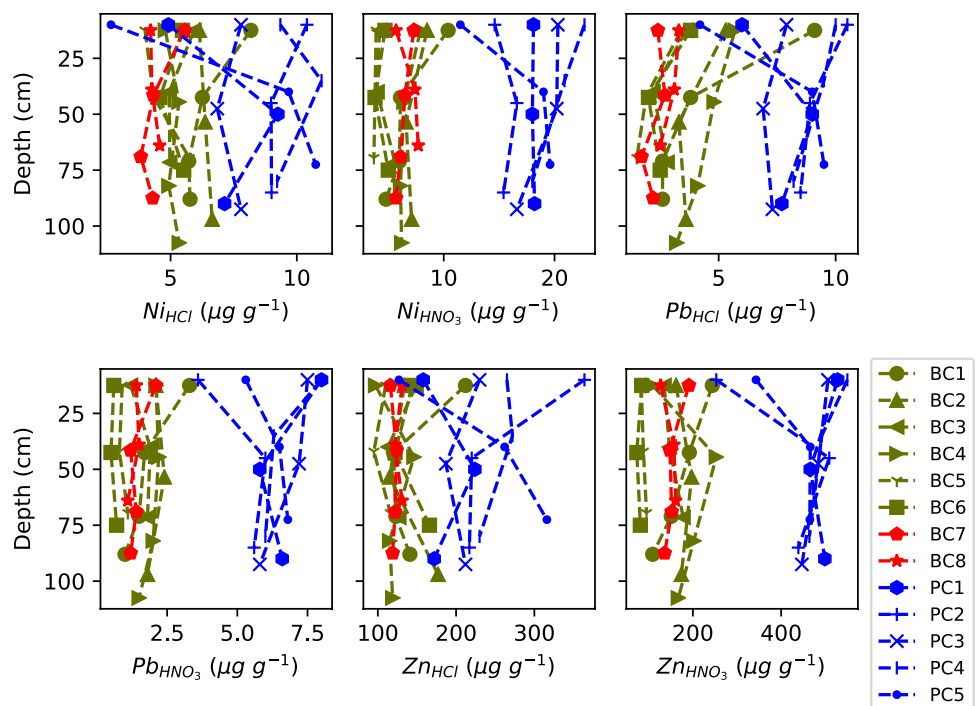


Fig. 8 Behavior of cadmium concentration of HCl extraction and total Hg concentration in soil samples from *Punta Caimán* (PC) and *Boca de Caño* (BC) with depth (cm). Green, red and blue colors stand for samplings sites of developed mangroves from BC, underdeveloped mangroves from BC and dwarf mangroves from PC, respectively

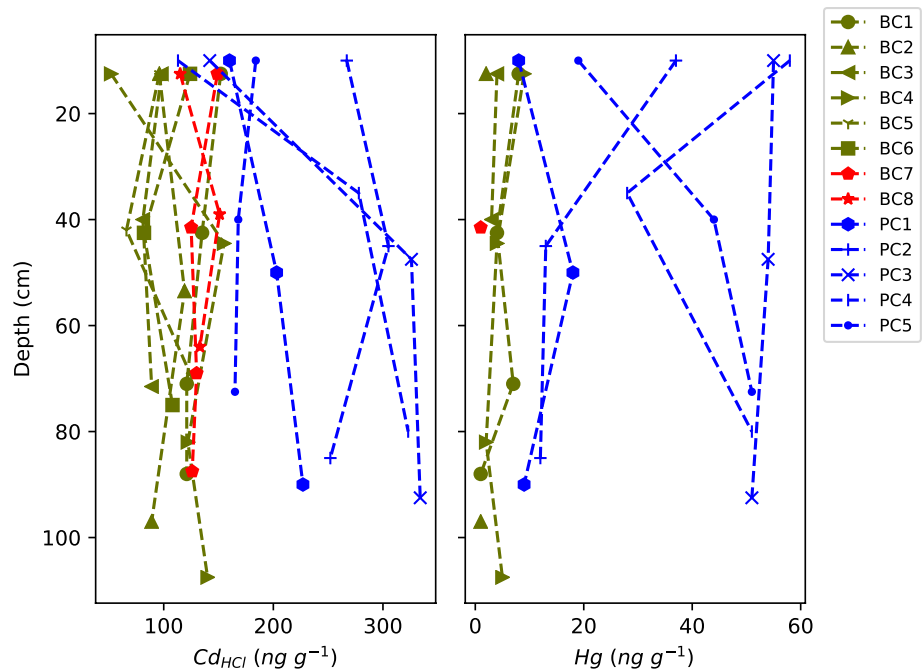
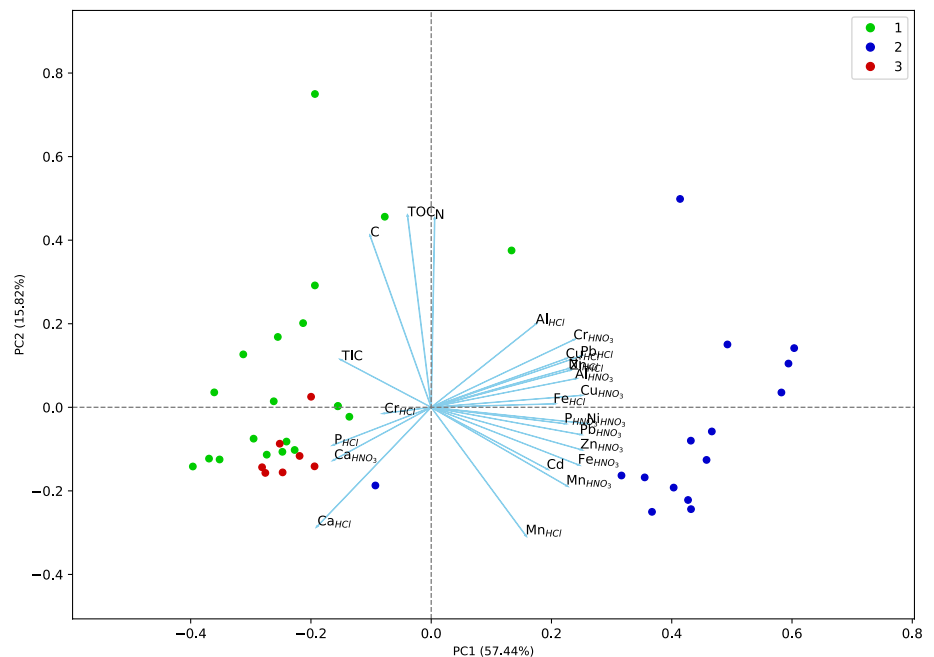


Fig. 9 Principal component analysis highlighting different regions with specific structural mangrove development: 1, fully developed mangroves of *Boca de Caño*; 2, dwarf mangroves of *Punta Caimán*; and 3, small mangroves of *Boca de Caño*



Discussion

The observed TOC values in this study are relatively similar to mangrove soils from other semi-arid areas (Alsamady et al. 2020; Araújo et al. 2012) but less than the average observed in mangroves soils from more humid areas located near the study region (Barreto 2008; Otero et al 2017b), which can be up to 20 %, and less than mangroves

soils from other studies in humid regions (Chiu and Chou 1991; Marchand et al. 2011).

There seems to be no limitation of P in the fully developed mangroves given that the P_{HNO_3} and the organic carbon concentrations show a good correlation (sampling locations BC1-6, Fig. 12). This suggests that the P is adsorbed into the organic matter. In general, P in the soils can be related to carbonate minerals, highlighted by the significant positive

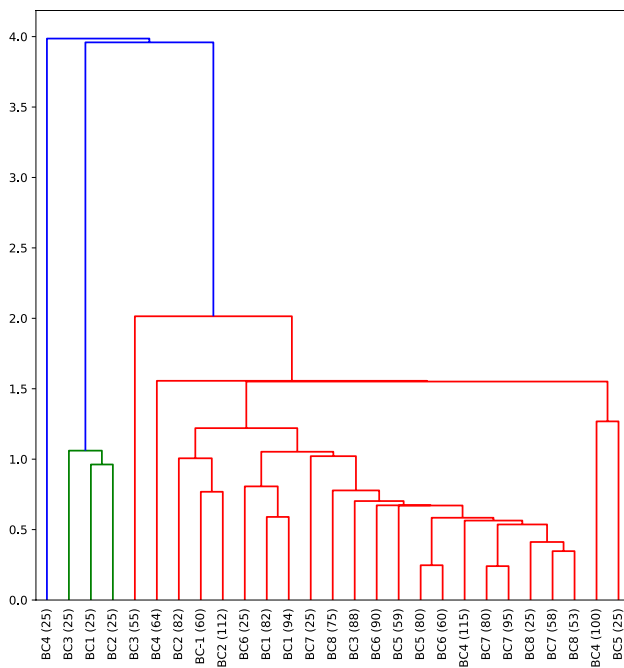


Fig. 10 Dendrogram grouping soil samples from *Boca de Caño* (BC). Sample depths inside parenthesis are in cm

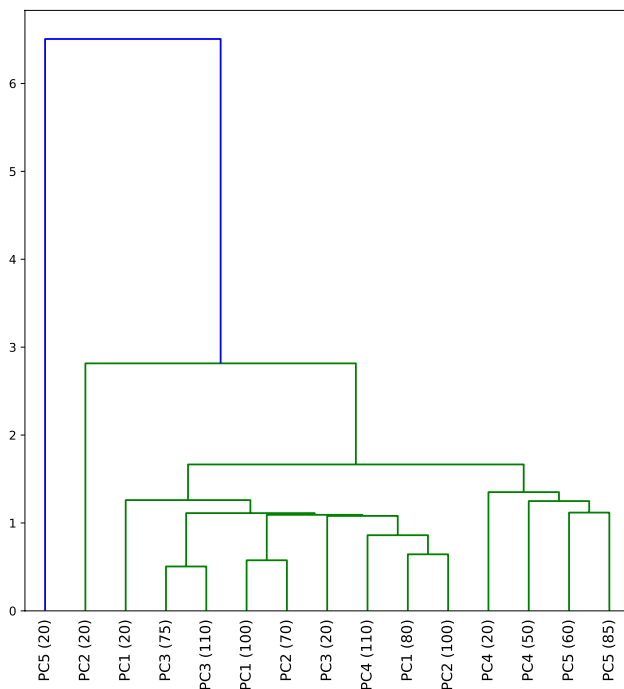


Fig. 11 Dendrogram grouping soil samples from *Punta Caimán* (PC). Sample depths inside parenthesis are in cm

correlation between P_{HCl} and Ca_{HCl} observed for all soil samples. Moreover, most of dwarf mangroves soils (excluding location PC2) show a positive correlation of P_{HNO_3} with

Fe_{HNO_3} , therefore, the P can be adsorbed into the crystalline oxides, more stable in the soil system and controlling the availability of P (Nóbrega et al. 2014).

The low C:N ratios in organic matter can serve as a good indicator to understand the stability of the soil organic matter, because microorganisms can easily process organic matter with low C:N, more amino-acids, than the high C:N ones, the latter is related to lignin or other more stable organic compounds (Manzoni et al. 2008, 2010). Moreover, C:N weight ratio of organic matter can indicate sources, *i.e.* low values are probably marine or *in situ* sources whereas high C:N values suggest terrestrial or refractory (degraded) organic matter (Bouillon et al. 2003). According to this, the C:N ratios of the sampled soils might represent a marine source or recent organic matter; the soils from *Punta Caimán* display less C:N values than the ones from *Boca de Caño*, which indicates that there is less lignin in the soil system.

The low accumulation of organic matter in most of the dwarf mangrove soils from *Punta Caimán* is mainly due to the action of waves and tides, which remove not only the particulate matter but also dissolved species—dissolved organic matter represented by fulvic and humic acids—that are more soluble in solutions of higher pH. For instance, the action of tides, which incorporate marine water to the soil, can dissolve more humic and fulvic acids than fresh water. This hypothesis is supported by the high organic carbon concentration observed in the soil sample of the dwarf *Avicennia germinans* (sampling location PC2) surrounded by *Rhizophora mangle* shrubs, which work as a barrier in the shore, protecting the detritus material and allowing the accumulation of organic matter far from the shore. In the *Boca de Caño* region, the soils related to fully developed *Rhizophora mangle* present, in general, a greater content of organic carbon than the ones dominated by *Avicennia germinans*, similar to other mangrove soils (Barreto et al. 2016; Marchand et al. 2011). The low soil organic carbon in deeper samples might be the result of less accumulation of organic matter at the time where the sediment was deposited or to the degradation of the organic matter through time.

The TOC and C:N values of soils are the most representative variables reflecting the difference in the degree of development of the mangrove forest. Both variables are affected by the effect of tides and removal of detritus material and their observed values might reflect these processes. Nevertheless, we suggest that both variables should be studied in other regions in order to corroborate if soils dominated by dwarf mangroves always display less TOC and C:N values than the ones dominated by fully developed mangroves. The pore-water salinity, although not significantly different, is slightly greater in soils dominated by dwarf mangroves, hence, it might also be one of the factors affecting the degree of development of the mangroves. However, the difference

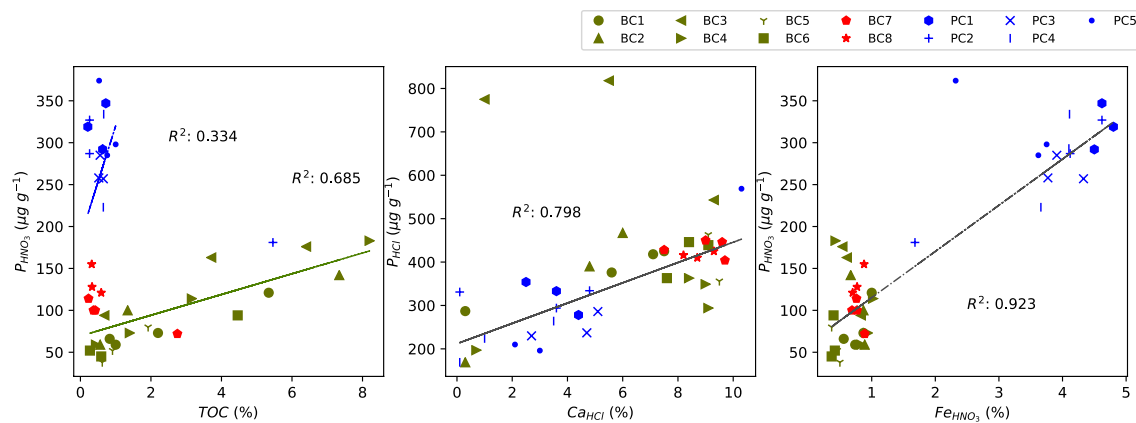


Fig. 12 Scatter-plots of P_{HNO_3} concentration against TOC content, P_{HCl} against Ca_{HCl} concentrations, and P_{HNO_3} against Fe_{HNO_3} concentrations in soil samples from *Punta Caimán* (PC) and *Boca de Caño*

(BC) with depth (cm). Green, red and blue colors stand for samplings sites of developed mangroves from BC, underdeveloped mangroves from BC and dwarf mangroves from PC, respectively

in pore-salinity in soils dominated by different mangrove species is significant and the observed pattern is the same for both regions.

Furthermore, the greater concentration of trace element found in soils dominated by dwarf mangroves compared to the ones dominated by fully developed mangroves might be the result of relatively low TOC concentration or to a significant difference in biogeochemical processes caused by differences in physical chemical conditions of the system. Taking into consideration that the difference in sediment source might also play a role. In this respect, Al might be one of the most significant trace elements analyzed in this study that might display the difference in source. However, more basic soils (dominated by dwarf mangroves) present greater Al concentrations, which is probably caused by increased dissolution of oxide-hydroxides of Al. The Al concentrations in both sites, are less than the total concentration measured *Golfete de Cuare*, an less arid region which is nearby (Otero et al. 2017b). The differences in Al concentration might be explained by the abundance of aluminosilicates in sediments and by secondary mineral precipitation and dissolution due to *in situ* alteration processes. Fe concentrations observed in *Punta Caimán* soils are greater than the concentrations found in mangrove regions located nearby (Otero et al. 2017a) and in other countries (Bastakoti et al. 2019, Marchand et al. 2006b). However, the concentration are similar to the ones measured in a semi-arid region located in the north-east coast of the state of Ceará, Brazil (Araújo et al. 2012). On the other hand, the Fe concentrations found in the region of *Boca de Caño* are similar to other humid soils (Bastakoti et al. 2019, Marchand et al. 2006a, Otero et al. 2017b). The soils from *Punta Caimán* region with low TOC content tend to display greater Fe_{HNO_3} concentrations. This effect might be caused by the precipitation of iron oxide-hydroxides in soils subject to tidal influence, where the marine water

intrusion increases the pH of the soil (Araújo et al. 2012). The low concentration of Fe_{HCl} observed in the submerged soil of *Avicenia germinans* (sampling location BC4), which presented pH of 6.8 ± 0.1 , might be the result of the vertical and horizontal mobilization of Fe in the reduced form, as Fe(+II), mainly due to limitation of oxygen in the soil (Banerjee et al. 2016; Kitaya et al. 2002). The submerged soil might allow more reducing conditions and limiting the diffusion of gases, affecting, consequently, the chemical properties of the soil.

The organic-rich soils from *Punta Caimán* and *Boca de Caño* regions tend to be more acidic and to have more reduced conditions, which can cause an increase in Fe mobilization along the soil profile, allowing sulphide mineral precipitation or reaching oxic areas where iron oxide-hydroxides could precipitate, *e.g.* due to bioturbation and root respiration (Araújo et al. 2012; Clark et al. 1998). This might be the reason of the less Fe_{HNO_3} concentration in the superficial sample of PC2 from *Punta Caimán* with respect to other samples from the same region. Although sulphate ions were not measured in this study, the high availability of sulphate ions in the marine water, primary source of water of these regions, may allow the precipitation of sulphide minerals under persistence reduced conditions (Sherman et al. 1998).

The Mn concentration of soils from *Boca de Caño* and *Punta Caimán* is greater than the observed in other mangrove soils of Venezuela (Otero et al. 2017b). The relative high concentration of Mn in the sampled soils are probably related to the carbonate phase (Otero et al. 2009), given the observed relationship between Mn_{HCl} and Ca_{HCl} (Fig. 13). The the sampling locations BC7, BC8 and PC5 presented coarser texture than other samples, with significant amounts of shell fragments. A coarser texture adsorbs less trace elements due to the low surface area compare to a clay or loam

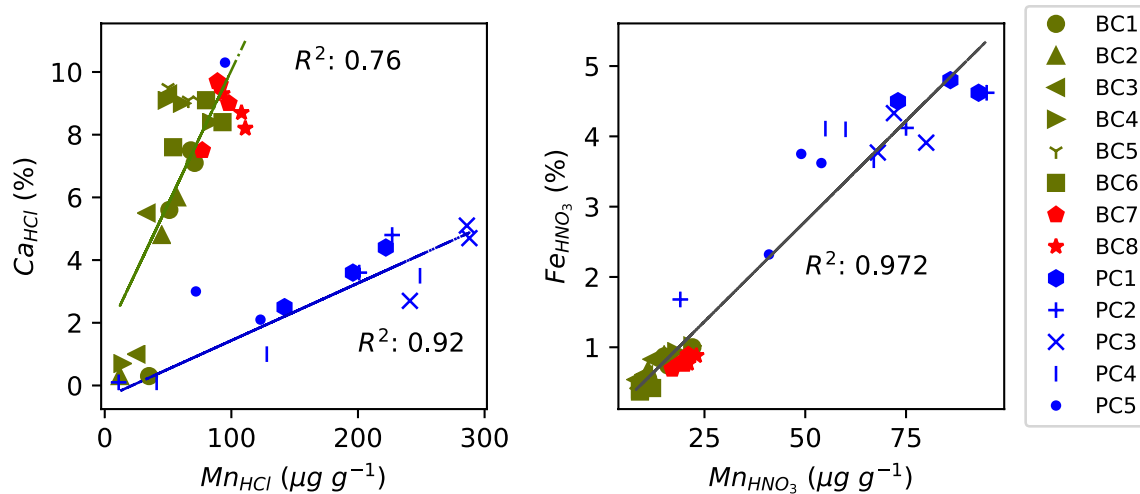


Fig. 13 Scatter-plots of Mn_{HCl} against Ca_{HCl} concentrations and Mn_{HNO_3} against Fe_{HNO_3} concentrations in soil samples from *Punta Caimán* (PC) and *Boca de Caño* (BC) with depth (cm). Green, red

and blue colors stand for samplings sites of developed mangroves from BC, underdeveloped mangroves from BC and dwarf mangroves from PC, respectively

textures. Moreover, the texture can also influence the diffusive limit of oxygen, hence, the redox characteristic of the soils (Chiu and Chou 1991; Clark et al. 1998). The positive correlation between Mn_{HNO_3} and Fe_{HNO_3} suggests the presence of crystallized oxides of iron, which can incorporate Mn into its structure (Otero et al. 2009). The greater concentration of Mn can be achieved by a reduction of Mn(IV) to Mn(II) and subsequent precipitation of authigenic carbonate minerals, which incorporate Mn into their structure. The less Mn concentrations in other mangrove soils might be related to a removal of Mn(II) by vertical or horizontal transport (Otero et al. 2009). The Cr_{HCl} concentration is correlated to Mn_{HCl} in soils of *Boca de Caño* and sampling location PC5 of *Punta Caimán* (Fig. 9), which might be associated to carbonate minerals. The Cr_{HNO_3} is probably related to crystalline phase of iron oxides, found in greater concentration in soils from *Punta Caimán*.

Comparison of trace element concentrations with other studies is rather difficult due to different methodologies applied during sample treatment. Therefore, most of the comparison are done by calculating the total concentration by combining both extractions ($HCl + HNO_3$). In this respect, the Cu concentration in soils from *Punta Caimán* is in the range to the ones measured in mangroves soils from French Guiana (Marchand et al. 2006b) and from Taiwan (Chiu and Chou 1991), but greater than the observed in different mangrove soils from Brazil (Angeli et al. 2019; Lacerda et al. 1993; Machado et al. 2014). In soils from *Boca de Caño*, however, the Cu concentration is in the less range compared to the previous mentioned studies. The Ni concentration in soils from *Punta Caimán* is relative greater than the one reported in other studies (Angeli et al. 2019; Chiu and Chou 1991; Machado et al. 2014; Otero et al. 2017b),

whilst the one measured in soils from *Boca de Caño* is less than the reported in those studies. The concentration of Zn (an essential trace element) in soils from *Punta Caimán* and from *Boca de Caño* are greater than the ones reported in other studies (Angeli et al. 2019; Chiu and Chou 1991; Machado et al. 2014; Otero et al. 2017b) but are similar to the ones reported in mangroves soils located in Sepetiba bay, Brazil (Lacerda et al. 1993). Environmental relevant trace elements concentrations, *i.e.* Cr, Cd, Pb and Hg, are similar to the ones observed in other mangrove soils (Alsamadany et al. 2020; Angeli et al. 2019; Chiu and Chou 1991; Kalaivanan et al. 2017; Lacerda et al. 1993; Machado et al. 2014; Otero et al. 2017b). These comparisons highlight the highly heterogeneity of the mangrove soils. Finally, there is no indication that mangrove soils from semi-arid regions or associated to dwarf mangroves will present greater trace element concentrations.

The adsorption of trace elements in soils is influenced by the soil properties, *i.e.* the texture, content of total organic carbon, Eh, pH and conductivity (Chiu and Chou 1991). Therefore, the availability and the geochemical behavior of trace elements is different in dwarf and fully developed mangrove soils, given that the latter present more organic matter. The accumulation of organic matter in the soil triggers other processes, *e.g.* cation exchange, acidity of soils, recycling of nutrients, accumulation of trace elements in forms not available to plants, and enhances the microbial activity (Alongi 2017; Clark et al. 1998; Morel et al. 2006).

Soil respiration by root and microbial activity may reduce soil organic carbon concentration and decreases pH, by the dissolution of produced CO_2 (Chambers et al. 2014), which speeds up the dissolution of minerals. The oxide-hydroxide layers observed in soils dominated by *Rhizophora mangle*

might be the result of a combination of soil respiration and bioturbation (Clark et al. 1998). Moreover, the soil organic matter and organic acids can potentially adsorb trace elements and nutrients, (Clarholm et al. 2015). The high concentration of organic matter and the persistent reduced condition allow to transforming trace elements in a form related to sulfides; however, in the dwarf mangrove soils from *Punta Caimán*, where the organic matter is less than in soils from *Boca de Caño*, a greater concentration of trace elements was found with respect to soils of developed mangroves from *Boca de Caño*. This relative high concentration of trace elements might indicate a difference in the source of the sediments, or resulted due to anthropogenic activities, given the proximity to an oil refinery (named Amuay) and the fact that there is a pipeline along this region, indicating previous human perturbation. Nonetheless, an anthropogenic influence is not possible to be confirmed because the trace element concentrations do not show a contrasting accumulation in the top soil layer with respect to the deeper samples.

The mangrove soils subject to tides will display variable oxidation-reduction potential and basic pH (due to intrusion of marine water), where some trace elements are more stable as oxide-hydroxides, which can increase trace element adsorption and precipitation, hence, decreasing the amount that could be incorporated into the food-chain if conditions remain stable (Chlopecka and Adriano 1997). For example, root respiration and organic acids produced near the rhizosphere can potentially dissolved amorphous oxides-hydroxides and incorporate trace elements into the food-chain (Krishnamurti et al. 1997).

A dwarf mangrove forest is a response of the plant to environmental conditions, water availability, pore-water salinity, the limitation of bioavailable nutrients or to the removal of organic matter. Despite that the soils of dwarf mangrove forests can have similar geochemical properties than the ones dominated by fully developed mangroves, the phenotypic response, which includes an increase of root production and organic detritus (Alongi 2015) controls the size of the mangrove. The degradation of the ecosystem by natural or anthropogenic influences can change the soil condition and stability, increasing the soil organic matter decomposition, hence, releasing carbon dioxide or methane to the atmosphere (Otero et al. 2017a). Moreover, a rapid change can limit the adaptation of the mangrove ecosystem (Alongi 2015). The action of tides and waves can increase the export loads of organic carbon to the marine ecosystem, specially in environments similar to the presented in this study.

The biogeochemical processes taking place in mangrove soils depend on physical, chemical, geological and biological properties (Alongi 2009), and the diversity of those conditions produces a highly heterogeneous soil. This heterogeneity should be consider in global applications and to

quantify the change in biogeochemical rates due to a changing climate. For instance, future models should consider the difference in organic carbon storage between dwarf and full developed mangroves. The mangrove forests located in coastal semi-arid regions should be focus of more attention because they are prone to decline if the adaptation of the mangrove is not fast enough (Alongi 2015). Furthermore, the mangrove forest may influence the marine biogeochemical cycles of nutrients and micronutrients given that it was identified that they might account for about 10 % of the terrestrial derived refractory DOC transported to the oceans (Dittmar et al. 2006). The carbon export to the marine environment, however, is not totally represented in global biogeochemical models and more information on rates is needed in order to validate future models. This work encourage to study in more detail these regions, where mangrove forests are more vulnerable.

Conclusions

The chemical composition and the geochemical characteristics of mangrove soils reflect the environmental (natural or anthropogenic) influences and allow the identification of susceptible ecosystems. The results obtained in this study showed that geochemical characteristics of soils associated to different mangrove species is notorious when the mangrove is fully developed. Moreover, although mangrove soils located in semi-arid regions present less organic carbon concentration than mangrove soils from humid regions, a wide range of organic matter content can be observed due to the effect of other significant processes, *e.g.* the effect of tides, marine water intrusion, and the texture of the soil, which also have an effect on the degree of development of the mangrove. The fully developed mangrove forest, *e.g.* in *Boca de Caño* region, are characterized by soils with neutral pH and anoxic conditions. These properties allow the accumulation of trace elements in the reduced form, most likely as sulphides. However, oxide-hydroxides could be formed due to bioturbation, allowing the migration of some trace elements and adsorption onto organic matter. Dwarf mangroves regions affected by the action of tides and waves are important exports of organic matter from the mangrove ecosystems into the marine environment. This effect may have a significant impact in future climate change, in which, an increase of the sea level will affect many mangrove ecosystems. Furthermore, the tidal influence allows the retention of trace elements adsorbed onto oxide-hydroxides and precipitation of authigenic carbonates that can incorporate Mn.

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Author contributions GRM conducted the analytical measurements and the analyses of the results. WM presented the initial idea and planned the field work. Both authors wrote and reviewed the manuscript.

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Declarations

Conflict of interest The authors declare no competing interests and non-financial competing interests.

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