



## An environmental magnetism approach to assess impacts of land-derived sediment disturbances on coral reef ecosystems (Cartagena, Colombia)



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### ABSTRACT

We used environmental magnetism methods to study recently deposited marine sediments from the estuarine ecosystems on the Caribbean coast of Colombia. Cartagena region has undergone an increasing sediment load during the last decades via sediment plumes from Magdalena River and its distributary man-made channel. Concentration dependent magnetic parameters show an increasing abundance of ferrimagnetic minerals on the uppermost sediments on sites located close to the continent (remanent magnetization  $SIRM = 5.4\text{--}9.5 \times 10^{-3} \text{Am}^2 \text{kg}^{-1}$ ) as well as faraway sites ( $SIRM = 0.5\text{--}1.7 \times 10^{-3} \text{Am}^2 \text{kg}^{-1}$  near Rosario Islands coral reef complex). The magnetic grain size and mineralogy along the cores are variable, showing the dominance of the magnetite-like minerals (remanent coercivity  $H_{cr} = 34.3\text{--}45.3 \text{mT}$ ), with a minor contribution of high-coercivity minerals ( $H_{cr} = 472\text{--}588 \text{mT}$ ). In addition, there is a moderate enrichment of elements Cu, Mo, and Zn (enrichment factor  $EF = 1.5\text{--}3.8$ ) that indicates the additional land-derived contribution on sediments. The environmental magnetism approach, which shows significant signals of magnetic minerals and trace elements, is a reliable tool to prove the presence of continental sediment supply in coral reef ecosystems.

### 1. Introduction

Natural systems such as coral reefs are complex and highly susceptible to environmental changes. Their behavior depends on the interaction of several independent forcing factors like climate variations and human activity affecting the environment. Fluvial sediment inputs in coral reefs have direct influence on coral health and diversity, affecting the distribution of coral communities (Hughes et al., 2003; Wolanski et al., 2005; Victor et al., 2006; Fabricius et al., 2013; McLaughlin et al., 2003). Sediment has been shown to be a major stressor to coral reefs globally. Storlazzi et al. (2015) showed that finer-grained and darker-colored sediments at higher suspended-sediment concentrations attenuate photosynthetically active radiation (PAR), significantly more than coarser, lighter-colored sediment at lower concentrations, and provide PAR attenuation coefficients for various grain sizes, colors, and suspended-sediment concentrations. Because fine-grained sediment particles settle-down slowly and are highly susceptible to resuspension, they remain in the water column longer, causing greater net impact by reducing light, essential for photosynthesis. Coral reef monitoring studies assessing the impacts of land-based sediment loads should concentrate on measuring fine grained lateritic and volcanic soils, as opposed to coarser grained siliceous and

carbonate sediments (Storlazzi et al., 2015). In the Caribbean, water transparency has steeply declined over the last 20 years at different locations from Guatemala to Honduras, and also at La Parguera in Puerto Rico (Jackson et al., 2014).

Environmental magnetism involves the use of magnetic techniques in situations in which the transport, deposition, or transformation of magnetic grains is influenced by environmental processes in the atmosphere, lithosphere, and hydrosphere. Environmental magnetism methods have been used in diverse fields, including land-use studies, limnology, oceanography, sedimentology, and soil science, among others (Verosub and Roberts, 1995). Magnetic proxies have the advantage of being determined with high sensitivity combined with fast laboratory processing; sample preparation is easy, laboratory instruments are relatively low cost, and most measurements are non-destructive (Chaparro et al., 2006; Gubbins and Herrero-Bervera, 2007). Magnetic measurements are being used as a rapid and useful tool to appraise heavy metal contamination in soils and sediments (Thompson and Oldfield, 1986; Evans and Heller, 2003). Thus, magnetic measurements provide new tools, in addition to traditional ones, to investigate the natural processes and anthropogenic influences in ecosystems such as soils (Petrovský et al., 2001; Blundell et al., 2009; Quijano et al., 2014), estuaries (Prajith et al., 2015; Chaparro et al.,

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2017a), lakes (Yang et al., 2007; Chaparro et al., 2017b), and rivers (Zhang et al., 2011; Krishnamoorthy et al., 2014).

The environmental magnetism method on marine sediment has not only been applied to investigate the recent influence of continental/anthropogenic sediments (e.g., Scoullou et al., 1979; Chan et al., 2001; Horng and Huh, 2011; Gwizdała et al., 2018), but also to investigate pollution records over decadal to centennial timescales (Locke and Bertine, 1986; Horng et al., 2009) and paleoclimate/paleoenvironmental records for longer periods (Bloemendal and deMenocal, 1989; Reinholdsson et al., 2013). Environmental magnetism studies on coral ecosystems are scarce around the world and absent in the Caribbean region. Among reported studies, Bothner et al. (2006) used sediment traps to evaluate the frequency, cause, and relative intensity of sediment mobility/resuspension along the fringing coral reef off of southern Molokai (Hawaii Island). Magnetic properties (e.g., isothermal and anhysteretic remanent magnetization, IRM and ARM, respectively, and ARM/IRM ratio) in those coastal sediments are a specific indicator of the land-derived component because the biogenic carbonate has non-magnetic properties. Land-derived particles carry their magnetic signature (volcanic rocks containing magnetite) potentially altered by weathering processes, as they are transported to and within the coastal ocean by streams, wind, and currents. Land-derived sediment, identified by the non-carbonate fraction and the presence of magnetite, is a ubiquitous component of sediment trap material, which indicates that fine-grained terrestrial sediment is constantly moving through reef ecosystems. Takesue et al. (2009) used geochemical tracers and magnetic properties (e.g., mass-specific magnetic susceptibility  $\chi$ , IRM, and S-ratio) to assess the influence of terrestrial runoff on a coral reef-fringed embayment in Kauai (Hawaiian island). They found that trace element ratios, magnetic domain size, and relative magnetite-hematite concentrations, should reflect sediment provenance. They also identified non-point source runoff from roads reported as the main contributor to elevated levels of Ni, Cu, Zn, and Pb in flood sediments, which may pose a greater threat to coral reef communities.

The Magdalena River, the main contributor of fluvial fluxes to the Caribbean Sea (Restrepo and Kjerfve, 2000; Restrepo et al., 2015), is probably the largest fluvial system worldwide, delivering continental sediments to coral reef ecosystems (Restrepo and Alvarado, 2011; Restrepo et al., 2016a). The coastal region of Cartagena, characterized by the Barbacoas and Cartagena bays, as well as by the presence of the largest coastal coral reef ecosystem in Colombia – the Rosario Island National Park – receives the Magdalena fluvial plumes via the Canal del Dique, a 114 km man-made channel (Fig. 1).

In the Cartagena region, previous studies (e.g., Moreno-Madriñán et al., 2015; Restrepo et al., 2016a) have analyzed the distribution of suspended sediment over coral reefs by using in-situ calibrated MODIS satellite images, demonstrating that corals have been strongly exposed to sediment plumes specially during the last decade. Nevertheless, some debate about the source of these sediments is still remaining, especially because the semi-open character of the Cartagena and Barbacoas bays allow the resuspension of bottom sediments to the near shore reefs (Restrepo et al., 2016a). In this area, other sources of sediments (e.g. eolian) are considered negligible versus fluvial sediments discharged by systems as the Magdalena River and associated Canal del Dique.

The proposed approach of environmental magnetism in coral reefs is an additional, reliable, and low-cost proxy to confirm the presence of land-derived sediments in coastal ecosystems, especially in a coastal region influenced by the continental fluxes of many rivers. The aim of this study is to determine: a) the magnetic properties of marine sediments for understanding the recent depositional processes in the Cartagena and Barbacoas bays; b) the presence of some trace elements, their enrichment and potential impact on different sites along the bays; c) the horizontal and in-depth parameter variation of bottom marine sediments; and d) the influence of continental sediment supply from the Magdalena River to the reef zones of the Rosario Islands, where the presence of ferrimagnetic and antiferromagnetic minerals may be

related to continental-source contribution.

## 2. Study area

The Cartagena Bay is located in northwest Colombia, on the Caribbean coast (Fig. 1). The Tierrabomba Island separates the bay from the open sea, leaving two sea inlets: Bocagrande in the north, and Bocachica in the south. The Bocachica inlet (15 m depth), the gap between Tierrabomba and Barú Peninsula, connects the bay with the major coral reef system in Colombia, the Rosario Island Coral Reef Natural Park, a 145 km<sup>2</sup> offshore coral reef (Restrepo et al., 2006).

Geologically, the Cartagena Bay area is located in the outer part of the Sinú fold belt, which is formed by Miocene to Pliocene marine sedimentary rocks deposited in turbiditic to coastal environments, like claystones, sandstones, gravels and calcareous sandstones, claystones and reef limestones (Duque-Caro, 1980). Locally, the hills and terraces in the bay (as La Popa Hill and Tierrabomba and Barú islands) are covered by a sequence of the Pleistocene Rotinet quartz-gravels over which is seated the La Popa Formation, a Pleistocene aged coral reef (Reyes et al., 2001). The observed rising of these coastal terraces at different levels (from ~20 to 3 m high) seems to be the response of active tectonics present along the bay which is expressed as faults and mud diapirism (Duque-Caro, 1980; Vernette, 1989).

The main freshwater source in the Cartagena Bay is the Canal del Dique, a 114 km long man-made distributary channel that connects the Magdalena River at Calamar with the Cartagena and Barbacoas bays (Fig. 1). The Canal del Dique has a great influence on the hydrology and sediment dynamics of these bays (Andrade et al., 2004; Restrepo et al., 2016a). The Canal has a high interannual river discharge variability, carrying most of its sediment load during short periods of time, being more pronounced during La Niña wet events. Between 1984 and 1998, the Canal transported 50% of its sediment load in seven extreme events (Restrepo et al., 2005). The highest sediment and water discharge rates occur during November, reaching  $31 \times 10^3$  t/d and 800 m<sup>3</sup>/s, respectively. Between 2000 and 2011, water and sediment fluxes have increased about 28% and 48% respectively, with interannual means of 397 m<sup>3</sup>/s of water discharge and  $5.9 \times 10^6$  t/y of sediment load (Restrepo and Correa, 2014; Restrepo et al., 2016a).

Sedimentation rates in the Cartagena and Barbacoas bays were obtained from the <sup>210</sup>Pb activity. Two cores were studied by Restrepo et al. (2016a) in order to assess sedimentation rates for the lower Magdalena River: one in the Pasacaballos delta, an outlet of El Dique into Cartagena Bay, and the other in the Barbacoas Bay, an area fed by a secondary delta system sourced by the Canal del Dique. No sedimentary discontinuities or changes in grain size were observed in the cores, which are absolutely dominated by silty sedimentation. For that reason, the interpretation of the <sup>210</sup>Pb results was based on the Constant Rate of Supply (CRS) model. The average accumulation rate for 10 samples in the core at the Barbacoas Bay is 0.75 cm per year and a similar value of 0.75 cm per year was estimated in the Pasacaballos Delta (25 samples). These fluvial-source sediments currently blanket the carbonate shelf and some are further transported reaching the more distal coral reefs of the Rosario Islands (Fig. 1). Along with the continental-bearing sediments, heavy metals, also detected, have been proved to come from the Magdalena basin and being flushed away to the bays and the Rosario Islands coral reef (Restrepo et al., 2016a).

The Rosario Islands coral reef, located in front of the Barbacoas Bay (Fig. 1), constitutes the major continental coral reef ecosystem in Colombia. In recent decades, the live coral cover has been reduced dramatically down to 22%, being replaced by the macroalgae cover (67%), which nowadays constitute the dominant feature in the reef (Restrepo and Alvarado, 2011). These authors discuss that the main causes of coral reef decline are the water quality in terms of turbidity, high sea surface temperature, and the use of dynamite and uncontrolled tourism in the Rosario Islands.

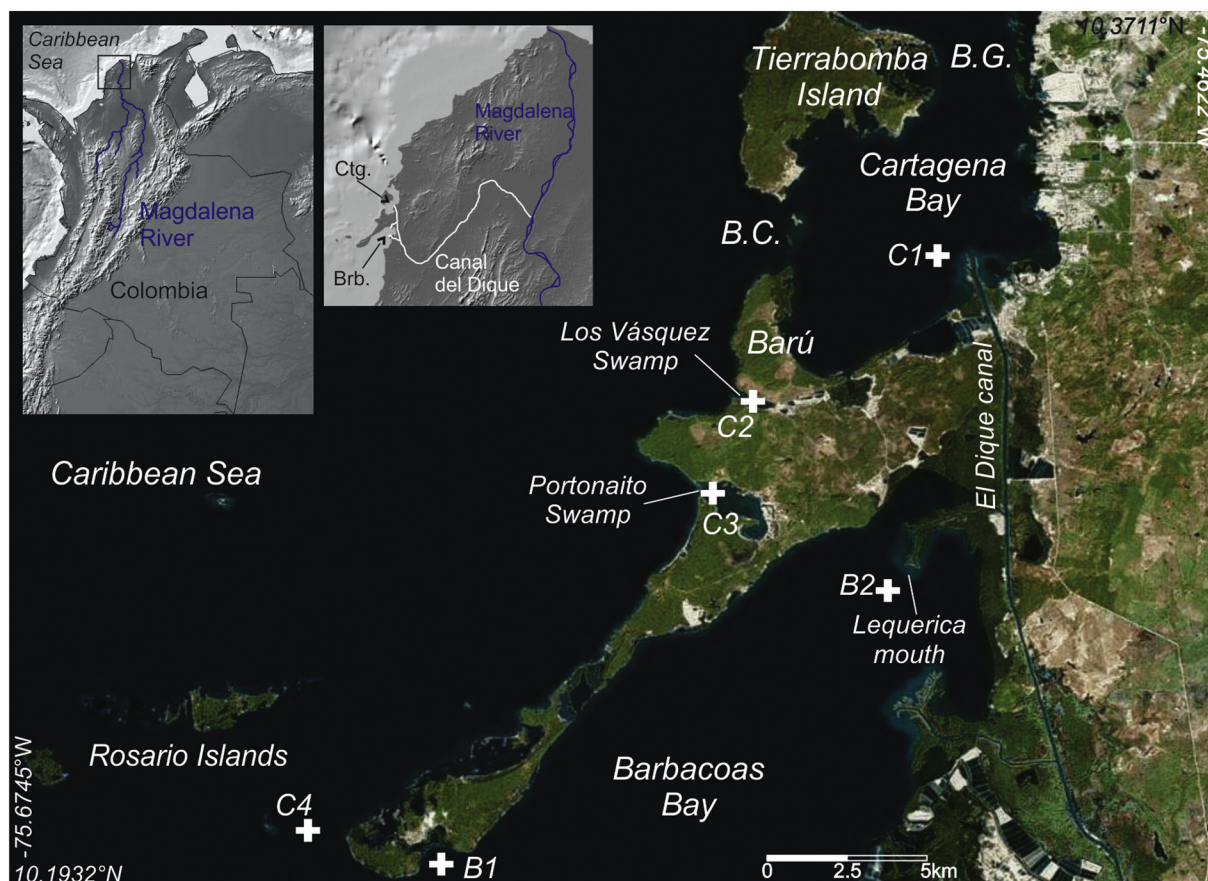


Fig. 1. Study area and sampling sites, Cartagena (Ctg.) and Barbacoas (Brb.) bays (Colombia). Sampling sites are: Canal del Dique (C1), Los Vásquez swamp (C2), Portonaito swamp (C3), Periquitos (C4), Punta Barú (B1) and Lequerica (B2).

### 3. Material and methods

#### 3.1. Sampling

In May 2014, six up to 30-cm-long sediment cores were collected in different places around the Cartagena and Barbacoas bays using a gravity piston corer. Sites were labelled as C1, C2, C3, C4, B1 and B2 and correspond to the following locations: Canal del Dique (Pasacaballos), Los Vásquez swamp, Portonaito (or Naito for short) swamp, Periquitos, Punta Barú and Lequerica, respectively (Fig. 1).

Marine-sediment cores are mostly made of clayey minerals. These cores were opened and sampled every centimeter in the Environmental Magnetism and Paleomagnetism Lab at EAFIT University (Colombia). These samples were put into glass containers and dried at 40 °C. In particular, samples from the Portonaito swamp, Periquitos and Punta Barú were sieved through a 600 µm mesh, due to the presence of abundant shell fragments.

A total of 119 samples were weighted and packed in 2.3 cm<sup>3</sup> plastic containers for magnetic analysis. Selected samples ( $n = 15$ ) from four cores were analyzed by inductively coupled plasma optical emission spectrometry (ICP-OES).

#### 3.2. Magnetic measurements

Magnetic measurements were carried out in the Laboratory of Magnetism and Paleomagnetism at the CIFICEN-IFAS (Argentina) and Environmental Magnetism and Paleomagnetism at EAFIT University (Colombia). Each sample was measured using different magnetic techniques: magnetic susceptibility and isothermal/anhyseretic remanent magnetization. Before running the measurements, each sample

was hardened with sodium silicate (diluted at 5% weight) to prevent particle movement in the process of acquisition of remanent magnetization.

Magnetic susceptibility ( $\kappa$ , volume) was measured with an MS2 susceptibility system from Bartington Instruments Ltd., linked to an MS2B dual frequency sensor (0.47 and 4.7 kHz). Replicate measurements were done for each sample on the higher sensitivity range ( $0.1 \times 10^{-5}$ SI), moreover  $\kappa$  values were corrected for drift through 5 measurement cycles (two air readings and three sample readings). Accuracy for measurement of  $\kappa$  is 1%. Parameter mass-specific magnetic susceptibility was calculated using this measurement and dry mass. Anhyseretic remanent magnetization was imparted to all samples with a device (pARM) coupled to an alternating field (AF) demagnetizer (Molspin Ltd.), by the superposition of increasing DC values of 10, 50 and 90 µT, with a peak AF of 100 mT. Remanent magnetization was measured with a spinner fluxgate magnetometer Minispin (Molspin Ltd.), which has a noise level  $< 2.5 \times 10^{-5}$  A/m and a sensitivity of  $10^{-4}$  A/m. These measurements were used to estimate the anhyseretic susceptibility  $\kappa_{ARM}$ , using linear regression with different DC values (7.96, 47.75 and 71.58 A/m). Also, parameters like ARM (ARM at DC = 71.58 A/m) and  $\chi_{ARM}$  both expressed on a basis of dry mass, the King's plot (King et al., 1982), and  $\kappa_{ARM}/\kappa$  were estimated.

Isothermal remanent magnetization measurements were made using an ASC Scientific pulse Magnetizer model IM-10-30. A total of 28 samples, chosen for detailed acquisition IRM measurements, were magnetized in 27 steps, under an increasing field, from ~1.7 mT to 2470 mT. The IRM was measured using the above-mentioned magnetometer. For all samples, the saturation of IRM (SIRM) was determined at a field of 2470 mT and expressed on a basis of dry mass. The remanent coercivity ( $H_{cr}$ ), S-ratio (S-ratio =  $-IRM_{300mT}/SIRM$ ) were

obtained applying a reverse field.

The experimental method proposed by Chaparro and Sinito (2004) was applied to discriminate magnetic mineralogy contribution: This method, based on the responses of different assemblages of magnetic minerals when they are subjected to a pulse-magnetizing dc field and a demagnetizing AF, separates the bulk backfield IRM curve into two individual magnetic phases experimentally. As indicated by Chaparro et al. (2005), the method was only carried out for backfield IRM measurements. In this study, separation was achieved using a high peak AF value ( $n = 102.5$  mT) as the filter. This AF value is chosen according to the markedly different response of low and high-coercivity magnetic materials. From measurements without an AF filter (backIRM(#i)) and with an AF filter (backIRM102.5(#i)), two magnetic phases (Phases 1 and 2) were obtained. Thus, the SIRM for each phase and its corresponding magnetic contribution (percent) to the total SIRM as well as the  $H_{cr}$  for each phase, were obtained.

### 3.3. Chemical analysis

The elements V, Cr, Co, Ni, Cu, Zn, Mo, Pb, and Fe were determined by ICP-OES using an Inductively Coupled Plasma Optical Emission Simultaneous Spectrometer, a Shimadzu 9000 (simultaneous high-resolution, norm EPA 200.7) at the Laboratory of Chemical Analysis (LANAQUI, CERZOS-CONICET-UNS).

The samples were dried in an electronic controlled stove under 40 °C for 24 h, and then sieved in order to eliminate major impurities. Then, they were digested in ultrapure grade nitric acid (US EPA SW-3052) using a MARS-5 microwave digestion system at 400 W, 800 psi and 200 °C for 15 min. The analysis was performed using the high-resolution ICP-OES Shimadzu 9000 with a torch injector (Glass Expansion, Australia) specially designed for high contents of diluted solids and salts.

Reported data correspond to an average value after measurements on two independent replicated samples; all of them within a standard deviation < 3.5%. Data were obtained using double methodology: external aqueous calibration and evaluation/correction of drifting and interferences using standard addition. All used standards are certified by the Chem-Lab, Zedelgem B-8210 and milli-Q water at 0.05 mS/cm and 18.2 MW/cm at 25 °C. Detection limits for each element are: 0.05 mg/kg (Co); 0.02 mg/kg (Cr); 0.04 mg/kg (Cu); 0.01 mg/kg (Fe); 0.05 mg/kg (Mo); 0.03 mg/kg (Ni); 0.04 mg/kg (Pb); 0.02 mg/kg (V); and 0.01 mg/kg (Zn).

The enrichment factor (EF) was calculated using the equation suggested by Szefer (1998),

$$EF_{Fe}^M = (M/Fe)_{\text{sample}} / (M/Fe)_{\text{background}} \quad (1)$$

where M is the given element and Fe is the reference element or normalizer; each ratio corresponds to the examined sample and the background reference. In this case, the upper continental crust (UCC, McLennan, 2001) was used as background value.

## 4. Results

### 4.1. Magnetic parameters

Magnetic parameters and related ratios were determined using measurements of magnetic susceptibility, isothermal and anhysteretic remanent magnetizations; hence parameters and inter-parametric ratios were used to characterize variations in magnetic properties in the analyzed samples. For all cores, concentration ( $\chi$ , ARM, and SIRM) and mineralogy ( $H_{cr}$ , S-ratio, SIRM/ $\chi$ ,  $\kappa_{ARM}/\kappa$  and ARM/SIRM) dependent magnetic parameters are detailed in the Supplementary data.

Mass-specific magnetic susceptibility values in Cartagena Bay (C1, C2, C3, and C4) vary from  $60.8 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$  to  $3.8 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$ , depending on the location and depth of the samples (Fig. 2). In site C1,

for example, there is a predominantly ferromagnetic (i.e. ferrimagnetic and antiferromagnetic) signal with values of  $\chi$  ranging from  $\sim 30.3$  to  $60.8 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$ , and ARM values ranging from 97.5 to  $161.2 \times 10^{-6} \text{ Am}^2 \text{ kg}^{-1}$ . These values indicate a change through time in the concentration of ferromagnetic minerals, with an increase of magnetic concentration upwards from recently deposited sediments, with maximum values in the first 4 cm. Values of SIRM increase up to two-fold, from  $5.4 \times 10^{-3} \text{ Am}^2 \text{ kg}^{-1}$  at the bottom to  $9.5 \times 10^{-3} \text{ Am}^2 \text{ kg}^{-1}$  at the uppermost sediments.

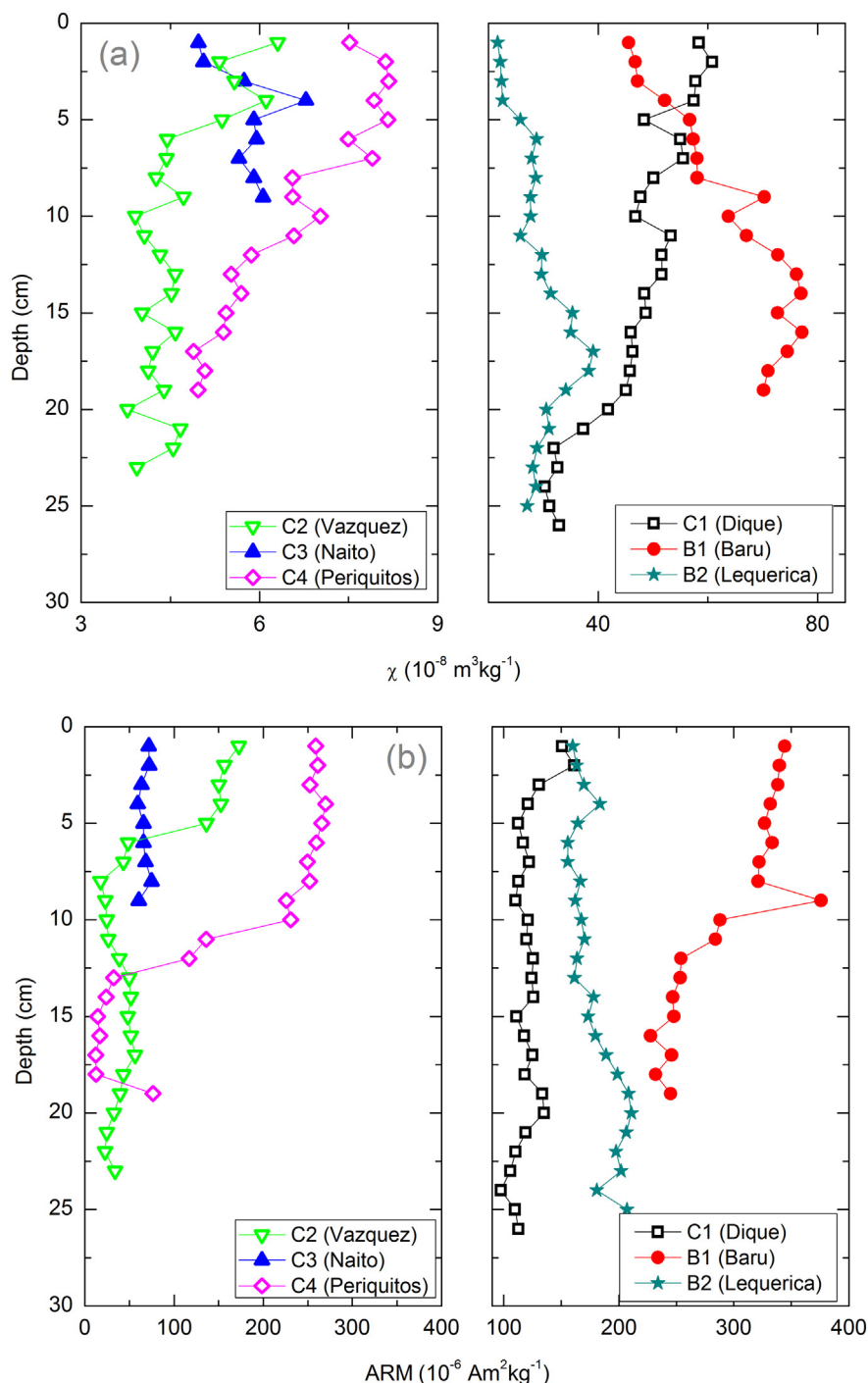
There is an abundance of paramagnetic minerals on sites C2, C3, and C4, although increases in  $\chi$  values in the uppermost sediments can be due to recent ferrimagnetic mineral input. Values of  $\chi$  in Los Vásquez swamp (core C2) vary from 3.8 to  $6.3 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$  (Fig. 2), and parameters ARM and SIRM show an increase in ferromagnetic minerals between 5 and 0 cm, with values of  $140.0 \times 10^{-6} \text{ Am}^2 \text{ kg}^{-1}$  and  $1.5 \times 10^{-3} \text{ Am}^2 \text{ kg}^{-1}$ , respectively. On core C3, in the depth interval between 8 and 6 cm, both ARM and SIRM show a minor increase in ferromagnetic mineral concentration, whereas  $\chi$  remains constant, indicating a dominance of paramagnetic minerals at this depth. On site C4 (Periquitos, in the Rosario Islands), there is an abundance of paramagnetic minerals, with  $\chi$  values between 4.9 and  $8.2 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$  (Fig. 2). Although mass-specific magnetic susceptibility values are low, the ARM and SIRM measurements show the presence of ferromagnetic minerals concentrated in the upper 10 cm of this core, with high ARM ( $270.0 \times 10^{-6} \text{ Am}^2 \text{ kg}^{-1}$ ) and SIRM values ( $1.7 \times 10^{-3} \text{ Am}^2 \text{ kg}^{-1}$ ).

Punta Barú site (B1), located on the Barbacoas Bay far from the Lequerica mouth, shows an important depth variation of magnetic minerals concentration (Fig. 2). This is visible for parameters as  $\chi$ , ARM, and SIRM. The  $\chi$  values vary between 45.5 and  $77.1 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$ . This parameter shows an increasing abundance of magnetic minerals with depth. Although the  $\chi$  curve shows a decrease in values upward to the uppermost sediments, ARM values show the opposite trend, ranging from  $227.7$  to  $375.9 \times 10^{-6} \text{ Am}^2 \text{ kg}^{-1}$ . A similar in-depth behavior could be seen for  $\chi$  parameter on Lequerica site (core B2), where the magnetic record is dominated by magnetic minerals, with  $\chi$  values ranging from 21.7 to  $39.1 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$ , and SIRM values from 3.8 to  $6.7 \times 10^{-3} \text{ Am}^2 \text{ kg}^{-1}$ . Magnetic minerals have maximum values of concentration dependent parameter at a depth interval of 12–20 cm, while  $\chi$ , ARM and SIRM parameters decrease upwards.

In all sampling sites,  $H_{cr}$  and S-ratio values show a relative mineralogical homogeneity, which is evidenced by low-coercivity magnetic phases of the magnetite-like mineral group on most of the sites; nevertheless, as it is discussed below (see Section 5.1), the contribution of high-coercivity magnetic minerals or hard phases is present in some sites.

Magnetite-like minerals are found on core C1, with high  $H_{cr}$  values from 41.6 to 49.5 mT; although in this core most of the samples show the presence of a ferrimagnetic phase, sections with  $H_{cr}$  values near 50 mT may have the presence of high-coercivity magnetic minerals, like hematite. This is evidenced in the S-ratio values that decrease down to 0.85 with the increase of  $H_{cr}$  values. Sites C2, C3, and C4 show  $H_{cr}$  values between 36.0 and 45.2 mT and also evidence phases of the magnetite-like mineral group; there is a relative quantity of high-coercivity magnetic minerals observed by the S-ratio values of about 0.78 (see Supplementary data).

Parameters  $H_{cr}$  and S-ratio in Barbacoas Bay show the dominance of a magnetite-like mineral group as well. In Punta Barú site (Core B1), there is evidence for the presence of low-coercivity magnetic phases ( $H_{cr}$  between 32.7 and 37.3 mT, and S-ratio between 0.84 and 0.90), hence dominated by ferrimagnetic over antiferromagnetic minerals. On the Lequerica site (core B2), the  $H_{cr}$  and S-ratio values vary from 40.6 to 49.7 mT and from 0.60 to 0.90, respectively. In the uppermost sediment section of this core, higher  $H_{cr}$  (lower S-ratio) values and therefore a relative increase of high-coercivity magnetic minerals are also observed.



**Fig. 2.** Concentration of magnetic dependent parameter versus depth for Cartagena (cores C1, C2, C3 and C4) and Barbacoas (cores B1 and B2) bays. (a) Mass-specific magnetic susceptibility  $\chi$ , and (b) anhysteretic remanent magnetization ARM.

On core C1 (Canal del Dique), magnetic grain size varies in the samples from 0.2 to 1  $\mu\text{m}$  according to the King's plot (King et al., 1982), and this difference is also evident in the related parameter  $\kappa_{\text{ARM}}/\kappa$  with a minor variation beginning at 18 cm depth (Fig. 3). This notwithstanding magnetic grain size is almost constant with depth in this core. On the other hand, magnetic grain sizes of cores C2, C3, and C4 (sites far from Canal del Dique) are smaller and characterized by the presence of finer ferrimagnetic particles on the uppermost sediments. On site C2, parameter  $\kappa_{\text{ARM}}/\kappa$  varies widely from 5.3 to 37.1 and ARM/SIRM ranges from 0.066 to 0.216. In Portonaito and Periquitos, both magnetic grain size dependent parameters show the presence of very fine ( $< 0.1 \mu\text{m}$ ) ferrimagnetic minerals in the uppermost section of the

cores. On site C4,  $\kappa_{\text{ARM}}/\kappa$  values vary also widely from 2.4 to 46.1 with an increase upwards.

In Barbacoas Bay, parameters  $\kappa_{\text{ARM}}/\kappa$  and ARM/SIRM show a magnetic grain size of 0.1  $\mu\text{m}$ , both in cores B1 and B2, according to the King's plot (King et al., 1982). On core B1,  $\kappa_{\text{ARM}}/\kappa$  values range from 3.5 to 8.2 and show a minor increase upwards ( $\kappa_{\text{ARM}}/\kappa = 7.2\text{--}8.2$ ), a condition that indicates a decrease in magnetic grain size. Nonetheless, on core B2, these parameters are almost constant with a minor increase for sediments in 0–4 cm, with  $\kappa_{\text{ARM}}/\kappa$  values of about 9.4 and ARM/SIRM values of 0.04 (Fig. 3 and Supplementary data).

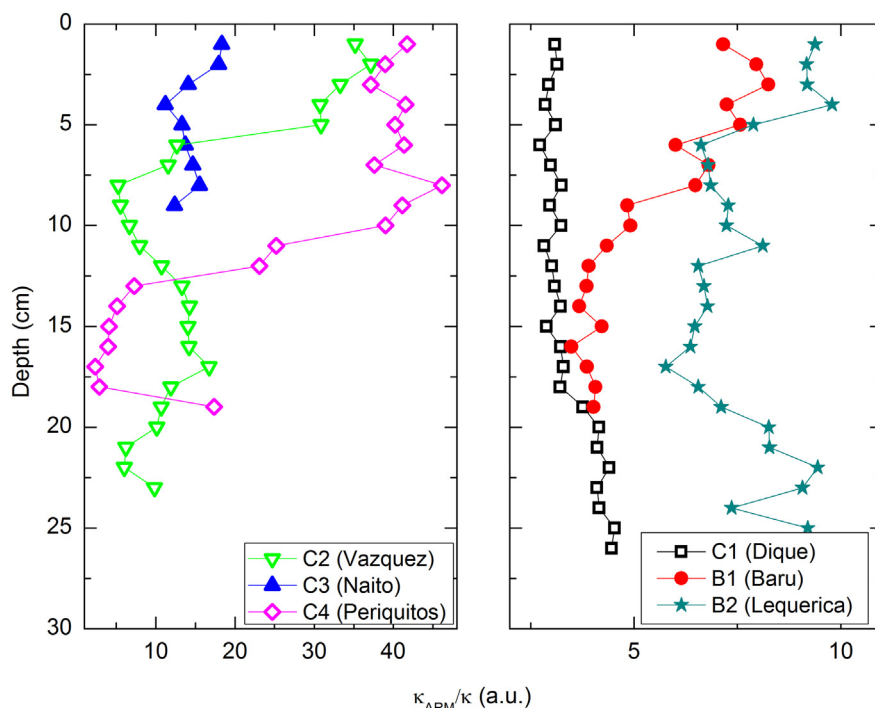


Fig. 3.  $\kappa_{ARM}/\kappa$  values versus depth for Cartagena (cores C1, C2, C3 and C4) and Barbacoas (cores B1 and B2) bays.

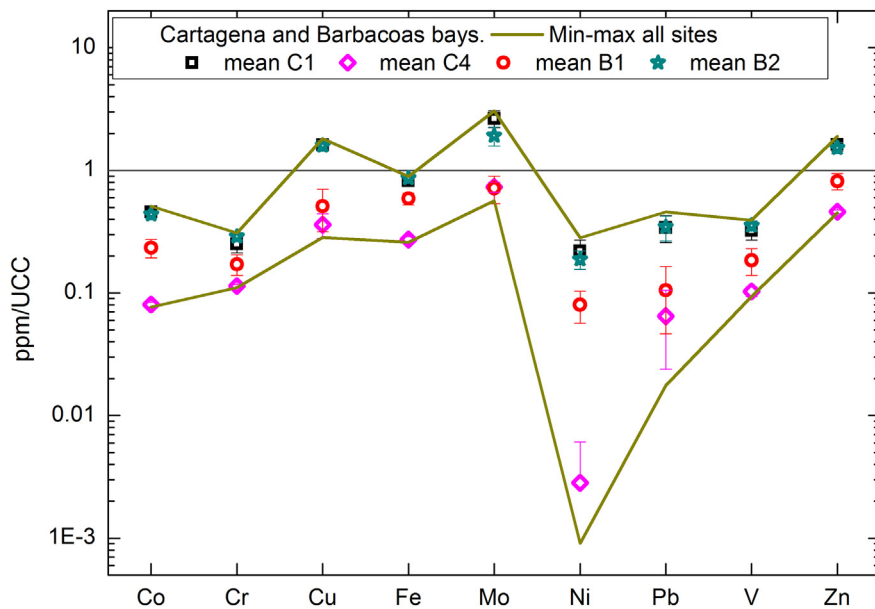


Fig. 4. Normalized values of selected elements for sites from Cartagena (Canal del Dique C1 and Periquitos C4) and Barbacoas (Lequerica B2 and Punta Barú B1) bays. Normalization was done using the upper continental crust (UCC, McLennan, 2001). Minimum and maximum values for all samples ( $n = 15$ ) and mean  $\pm$  s.d. values for each sediment core are shown.

#### 4.2. Trace elements

Four out of six sites were selected for chemical determination of Fe and trace elements Co, Cr, Cu, Mo, Ni, Pb V and Zn (Supplementary data). Such element concentrations were normalized to the upper continental crust (McLennan, 2001) in order to show the enrichments or depletion of chemical elements related to a reference pattern. The mean values for cores C1, C4, B1, and B2 are presented in the spider diagram (Fig. 4). Marine sediments of the study area present depleted chemical concentration in most elements, except in Cu, Fe, Mo, and Zn for sediment cores C1 and B2, values that are close to both channel mouths, i.e. Canal del Dique and Lequerica.

Elements behind UCC concentration are interpreted as weathered from minerals due to their chemical instability. On the other hand,

elements that present higher concentration than UCC should be due to additional pollution sources, such as mining, urban and agricultural activities. These concentration differences can be attributed to source-to-sink dispersal of continental/anthropogenic sediments in the Cartagena Bay area.

#### 5. Discussion

The main goal of this study is to compare the magnetic properties of marine sediments from various locations in the Cartagena and Barbacoas bays, and its relationship with the distribution of continental sediment supply coming from the Canal del Dique, represented by two sampling sites located near the mouths of the Canal del Dique at Cartagena (C1 core) and Barbacoas bays (B2 core), and additional

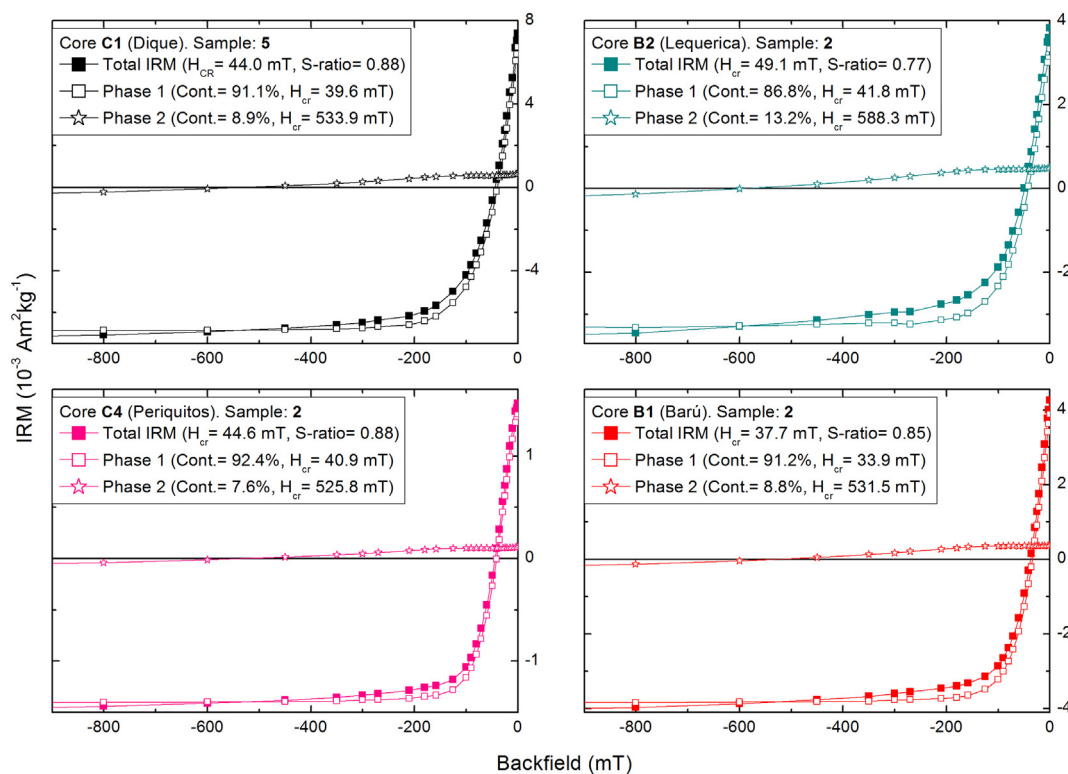


Fig. 5. Separation of IRM backfield measurements into two phases using the experimental method by Chaparro et al. (2005). Curves of Total back IRM (without AF filter), low-coercivity (Phase 1) and high-coercivity (Phase 2) phases, as well as their corresponding contribution to the SIRM and  $H_{cr}$  are detailed.

sampling sites located around the bays. There is also a special interest in evaluating the contribution of continental-bearing sediments from the Canal del Dique and Barbaecos channels to the Rosario Islands coral reefs.

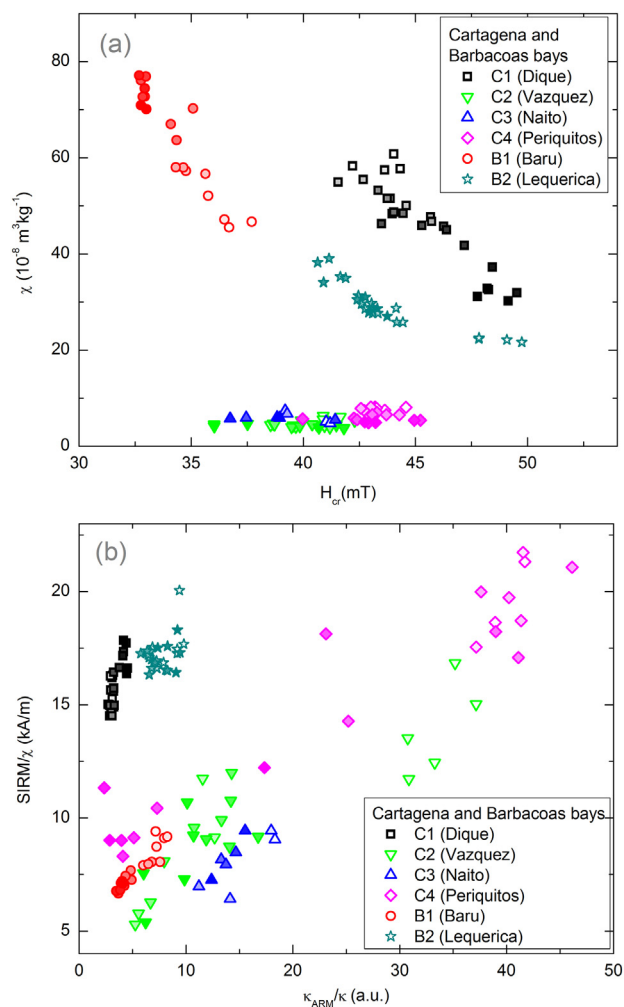
The geological setting around the Cartagena Bay and Barú Island is completely dominated by carbonate rocks formed on a carbonate platform from La Popa Fm (Reyes et al., 2001). The minerals present in the reef limestones belong to the carbonate and silicate groups, which are diamagnetic/paramagnetic minerals and give a weak magnetic signal compared to ferrimagnetic and antiferromagnetic minerals. At each site, it was assumed that the magnetic mineral components had a primarily continental/anthropogenic source, due to the absence of geological units with abundant strongly magnetic minerals in the rock formations nearby. Nevertheless, other possible pollution sources may have local influence. Although there is scarce or null information about neof ormation of ferromagnetic materials as end product of dynamite blasts, the intense use of explosives by fishermen within the coral reef ecosystems in the Rosario Islands since the 1970s (Werdning and Koster, 1977) had led to adverse pollution impact on this area. Even during the 1990s and beginning of the 21st century, fishing activities with dynamite were a common practice in the archipelago (Sánchez et al., 2006), which could have a local influence on pollution (Garzón-Ferreira and Díaz, 2003). Other pollution sources could come from the heavy traffic of boats and old commercial iron ships, in addition to sunken iron ships. According to Chan et al. (2001), magnetic enhancement on surficial layer of the seabed sediments in Hong Kong Harbor has a correlation with heavy metal contents, which implies shipping related contamination caused by navigation of ships, servicing, and scraping/painting while ships are at anchor. The occurrence of shipwrecks on the seabed may also constitute a source of contamination because salty water has a negative impact on the steel bode of wreck causing the increase of corrosion rate (Gwizdała et al., 2018). Although shipwrecks may pose a serious source of pollution, they are restricted to their vicinity of some coastal lagoons for building artificial reefs (Zarza-

González and Gómez, 2011). Eolian sediment transportation constitutes another possible source of magnetic minerals to the reef. Nevertheless, along the area, the local beaches would be the unique source of eolian sediments to be considered, which are mainly constituted by reef-related fragments and fluvial sediments. However, all these alternative pollution sources are considered negligible when compared to the power of fluvial systems (Canal del Dique) in the Cartagena and Barbaecos bays. The importance of these fluvial systems in the sediment distribution all along the Cartagena Bay is pointed-out by Restrepo et al. (2016b), showing that during ENSO cold phase, fluvial sediments from Canal del Dique are deposited across the entire bottom of the Cartagena bay.

### 5.1. Magnetic mineral composition of sediments

The magnetic mineralogy varies along the study area and shows a dominance of ferrimagnetic minerals such as magnetite, with mean  $H_{cr}$  values from 34.3 to 45.3 mT. Nevertheless, the contribution of high-coercivity minerals is evident from IRM magnetic separation analysis that shows, in some cases, the presence of high-coercivity phases (antiferromagnetic phases) that reach up to 13.5% of contribution to the SIRM (Fig. 5). Site C4 (Periquitos) located far from Canal del Dique (C1) show the presence of high-coercivity phases (i.e. hematite) with  $H_{cr}$  values of 471.8–525.8 mT and values of contribution to the SIRM lower than 11%. On Barbaecos Bay, the presence of high-coercivity phases with  $H_{cr}$  values of 531.5 mT and contribution to the SIRM lower than 9% is also observed in the site of Punta Barú (B1) located far from Lequerica site (B2).

The  $\chi$  and  $H_{cr}$  values are represented in Fig. 6a showing the concentration with respect to compositional variations. A similar magnetic composition is observed from mean  $H_{cr}$  values (Table 1) for sites close to the continent (C1 and B2) and the site close to the Rosario Islands (C4). Although core B1 shows lower  $H_{cr}$  values than core B2, high-coercivity magnetic phases are also present (Fig. 5). These sites, i.e.



**Fig. 6.** Biplots of (a) magnetic concentration dependent parameter  $\chi$  and mineral dependent parameter  $H_{cr}$ ; and (b) magnetic mineral dependent parameter  $SIRM/\chi$  and grain size dependent parameter  $\kappa_{ARM}/\kappa$  for Cartagena (cores C1, C2, C3, and C4) and Barbacoas (cores B1 and B2) bays. For each core, light colored symbols indicate uppermost sediment samples.

Canal del Dique (C1), Lequerica (B2), and Punta Barú (B1), present a common magnetic behavior as evidenced from statistical significant correlations between  $\chi$  and  $H_{cr}$  ( $R = -0.87$  to  $-0.93$ ;  $p < 0.01$ ), between  $\chi$  and  $\kappa_{ARM}/\kappa$  ( $R = -0.74$  to  $-0.96$ ;  $p < 0.01$ ), and between  $H_{cr}$  and  $\kappa_{ARM}/\kappa$  ( $R = 0.71$  to  $0.92$ ;  $p < 0.01$ ).

Although concentration dependent magnetic parameters of ferrimagnetic minerals differ in magnitude between C1 and C4, the compositional characteristics show that these two sites have a common magnetic mineralogy, for example, mean  $H_{cr}$  values are 45.3 and 43.2 mT, respectively (Table 1). The same occurs on cores C2 and C3, with mean  $H_{cr}$  values of 39.9 and 39.3 mT, respectively; indicating that these sites, which are located along the Cartagena Bay, share the same compositional characteristics. Nonetheless, core B1 shows quite a different concentration-composition relationship than the remaining sites, having lower mean  $H_{cr}$  values (34.3 mT) and opposing (to cores C1–4)  $\chi$  trend.

Parameters  $\kappa_{ARM}/\kappa$  and  $ARM/SIRM$  allowed the analysis of magnetic grain size variations (e.g.: Chaparro et al., 2017b; Lecomte et al., 2016). Based on statistical correlations between  $SIRM/\chi$  and  $\kappa_{ARM}/\kappa$ , as well as between  $SIRM/\chi$  and  $ARM/SIRM$  ( $R = 0.63$ – $0.99$ ;  $p < 0.01$ ), the parameter  $SIRM/\chi$  can also be used for magnetic grain variation on sites C2, C3, C4, and B1, (Fig. 6b). Coarser magnetic grains with mean  $\kappa_{ARM}/\kappa$  values of 3.4 (C1) and 7.7 (B2) are characteristic of fluvial

mouthing such as Canal del Dique and Lequerica. The King's plot indicates a magnetic particle size between 0.2 and 1  $\mu\text{m}$  on site C1, and  $< 0.1 \mu\text{m}$  on site B2. Although site B1 is located far from Barbacoas Bay delta (B2), it also shows coarser magnetic grains with mean  $\kappa_{ARM}/\kappa$  values of 5.4 (Table 1). Higher mean  $\kappa_{ARM}/\kappa$  (14.6–26.1) and  $ARM/SIRM$  (0.121–0.141, Table 1) values, and therefore finer magnetic particles, are present in sites C2, C3, and C4. Such finer particles are concentrated on the upper part of the cores (Fig. 3), where  $\kappa_{ARM}/\kappa$  and  $ARM/SIRM$  values increase in the first 5 cm (site C2) and 10 cm (site C4). This is observed as well in the concentration dependent magnetic parameter  $ARM$ , which is also sensitive to fine ferrimagnetic particles (Fig. 2).

## 5.2. Element enrichment in sediments

Calculated element enrichment factor for each trace element ranges between 0 and 3.8 (Supplementary data). The element enrichment factor has been widely used in environmental sciences to identify geogenic versus anthropogenic element sources. The EF values between 0.5 and 1.5 suggest that trace metal may come entirely from crustal contribution (e.g., weathering product), while values of  $EF > 1.5$  indicate that an important proportion of trace metals is delivered from non-crustal materials, e.g., non-point pollution sources (Zhang and Liu, 2002). For elements Cu, Mo and Zn, the EF values vary between 1.5 and 3.8 in Canal del Dique (C1), Periquitos (C4), and Lequerica (B2); and between 0.6 and 1.5 in Punta Barú (B1). The observed enrichment magnitudes close to the coast as well as on the reef platform indicate that coral ecosystems in the Cartagena region are influenced by additional continental and anthropogenic derived sediments.

According to the defined pollution categories for EF (Sutherland, 2000), the pollution contribution is minimal ( $< 2$ ) to moderate (2–5). On the other hand, the EFs are between 0 and 0.5 for elements Co, Cr, Ni, Pb and V for all sites. None of the analyzed samples overpass the Severe Effect Level (SEL), according to the Sediment Quality Guideline (SQG) for the reference pollutants Cr, Cu, Fe, Ni, Pb, Zn (McDonald et al., 2000). Nevertheless, cores Canal del Dique (C1) and Lequerica (B2) exceed the SQG Lowest Effect Level (LEL) for Cu, Fe, and Zn, and core Punta Barú (B1) exceed it for Cu and Fe. Although the obtained values indicate that the analyzed sediments are not heavily polluted to expect that sediment-dwelling organisms are currently affected by contamination, the Cartagena and Barbacoas bays sediments are not pollutant-free, especially in sites close to channel mouths. Similar to Restrepo et al. (2016a), this study also observed significant values of pollutant elements in sediments. For instance, Cu, Fe, and Zn exceed the LEL in several samples.

By using different magnetic properties on sampled sediments, our results indicate that Cartagena Bay is not the only source of pollutants. The Magdalena River seems to be loading polluted sediments that are later delivered to the Cartagena and Barbacoas bays, and eventually reach the Rosario Islands coral reefs.

## 5.3. Sources and distribution of sediments

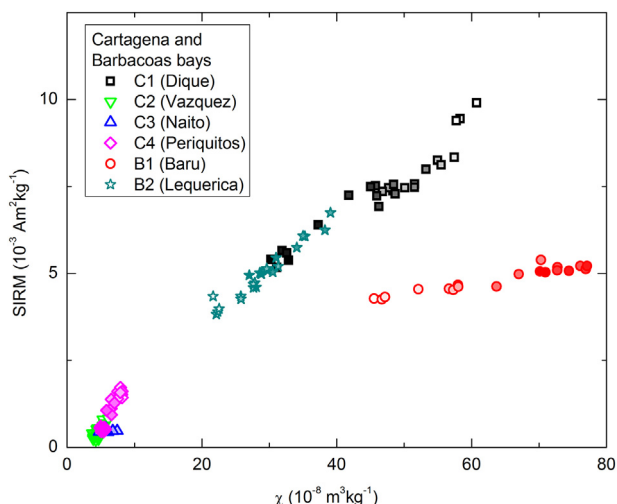
The relationship between concentration dependent magnetic parameters ( $\chi$  and  $SIRM$ ) is shown in Fig. 7. Sampling sites located close to Cartagena and Lequerica mouths (C1 and B2) show similar magnetic behavior, with significant statistical correlations between  $\chi$  and  $SIRM$  ( $R = 0.96$  and  $0.97$ ;  $p < 0.01$ ). Similarly, distal sampling sites (C4 and B1) also witness significant statistical correlations ( $R = 0.94$  and  $0.95$ ;  $p < 0.01$ ). In contrast, samples from core B1 (Punta Barú) that also have a significant statistical correlation between both magnetic parameters ( $R = 0.95$ ;  $p < 0.01$ ) display a different slope trend compared to the rest of the sediment samples (Fig. 7). This particular behavior could be explained by the action of specific processes and conditions, such as the presence of anthropogenic contaminants produced by human activities in Punta Barú.

The highest concentration of magnetic minerals are observed in the



**Table 1**  
Descriptive statistics of marine sediments from Cartagena and Barbacoas bays. Magnetic concentration and mineral dependent parameters.

Samples	$\chi$	ARM	SIRM	$H_{cr}$	S-ratio	SIRM/ $\chi$	$\kappa_{ARM}/\kappa$	ARM/SIRM
	$[10^{-8} m^3 kg^{-1}]$	$[10^{-6} A m^2 kg^{-1}]$	$[10^{-3} A m^2 kg^{-1}]$	[mT]	[a.u.]	[kA/m]	[a.u.]	[a.u.]
<b>Core C1 (Dique)</b>								
N	26	26	26	26	26	26	26	26
min	30.3	97.5	5.2	41.6	0.85	14.5	2.7	0.014
max	60.8	161.2	9.9	49.5	0.93	17.8	4.5	0.021
mean	46.6	121.1	7.3	45.3		15.9	3.4	0.017
s.d.	9.1	13.4	1.2	2.2		1.0	0.6	0.002
<b>Core C2 (Vazquez)</b>								
N	23	23	23	23	23	23	23	23
min	3.8	17.4	0.2	36.0	0.77	5.3	5.3	0.066
max	6.3	172.7	1.1	42.3	1.00	16.8	37.1	0.216
mean	4.6	62.7	0.5	39.9		9.8	15.4	0.121
s.d.	0.7	50.5	0.2	1.7		3.0	10.2	0.045
<b>Core C3 (Portonaito)</b>								
N	9	9	9	9	9	9	9	9
min	4.8	59.3	0.4	36.7	0.91	6.4	11.2	0.125
max	7.4	74.8	0.5	41.4	0.99	9.4	18.3	0.160
mean	5.9	66.8	0.5	39.3		8.1	14.6	0.141
s.d.	0.8	5.3	0.0	1.6		1.1	2.4	0.011
<b>Core C4 (Periquitos)</b>								
N	19	19	19	19	19	19	19	19
min	4.9	12.1	0.5	40.0	0.74	8.3	2.4	0.022
max	8.2	270.0	1.7	45.2	0.91	21.7	46.1	0.202
mean	6.6	156.1	1.1	43.2		15.6	26.1	0.123
s.d.	1.2	109.8	0.5	1.1		4.8	16.8	0.064
<b>Core B1 (Punta Barú)</b>								
N	19	19	19	19	19	19	19	19
min	45.5	227.7	4.3	32.7	0.84	6.7	3.5	0.044
max	77.1	375.9	5.4	37.7	0.90	9.4	8.2	0.080
mean	63.9	292.5	4.8	34.3		7.7	5.4	0.061
s.d.	10.8	46.9	0.4	1.6		0.9	1.7	0.013
<b>Core B2 (Lequerica)</b>								
N	25	25	25	25	25	25	25	25
min	21.7	155.6	3.8	40.6	0.60	16.3	5.8	0.028
max	39.1	210.9	6.7	49.7	0.90	20.0	9.8	0.046
mean	29.1	179.0	5.0	43.7		17.2	7.7	0.036
s.d.	4.6	18.3	0.7	2.4		0.7	1.2	0.005



**Fig. 7.** Concentration dependent magnetic parameters (SIRM and  $\chi$ ) for Cartagena (cores C1, C2, C3, and C4) and Barbacoas (cores B1 and B2) bays. For each core, light colored symbols indicate uppermost sediment samples and filled symbols correspond to lowermost samples.

channel mouths (Canal del Dique-C1 and Lequerica-B2), and Punta Barú (Core B1). The magnetic concentration dependent parameters  $\chi$  and SIRM are distinctively higher (mean values are

$29.1\text{--}63.9 \times 10^{-8} m^3 kg^{-1}$  and  $4.8\text{--}7.3 \times 10^{-3} Am^2 kg^{-1}$ , respectively) than values observed on the rest of the sites (mean values are below  $6.6 \times 10^{-8} m^3 kg^{-1}$  and  $1.1 \times 10^{-3} Am^2 kg^{-1}$ , respectively), as shown in Fig. 7. Core B1 in Barbacoas Bay shows a similar behavior to core B2 (site Lequerica) because it has magnetic concentration dependent parameters that show a decreasing trend upwards, with mean  $\chi$  values of  $63.9 \times 10^{-8} m^3 kg^{-1}$ , and decrease of  $\chi$  values to  $46.0 \times 10^{-8} m^3 kg^{-1}$  at the top (Fig. 8). Ferrimagnetic minerals in this site have lower coercivity values, reflecting distinctive characteristics.

Sampling sites C2, C3, and C4 are located farther from the Canal del Dique (C1) and show similar magnetic concentration parameters between them, characterized by lower concentration of ferrimagnetic minerals and a predominance of paramagnetic minerals. Although low values of magnetic concentration dependent parameters characterize C2, C3, and C4 cores, the presence of low and high-coercivity magnetic minerals with similar characteristics to the Canal del Dique is also observed in Periquitos (C4, Fig. 5).

All sediment cores taken from the Cartagena Bay reveal an increasing trend of ferrimagnetic mineral concentration upwards. Such behavior is evidenced in parameters as  $\chi$  (Fig. 8), ARM, and SIRM. Having higher concentrations on top of the sedimentary record may be explained via variation in the sedimentation rates, which were increased by 28% and 48% in the last decades (Restrepo and Correa, 2014). In addition, this increasing trend may also be explained by a higher input of continental sediments, which carry high concentrations of magnetic minerals due to the observed presence during the last decade of sediment plumes from Canal del Dique and Lequerica

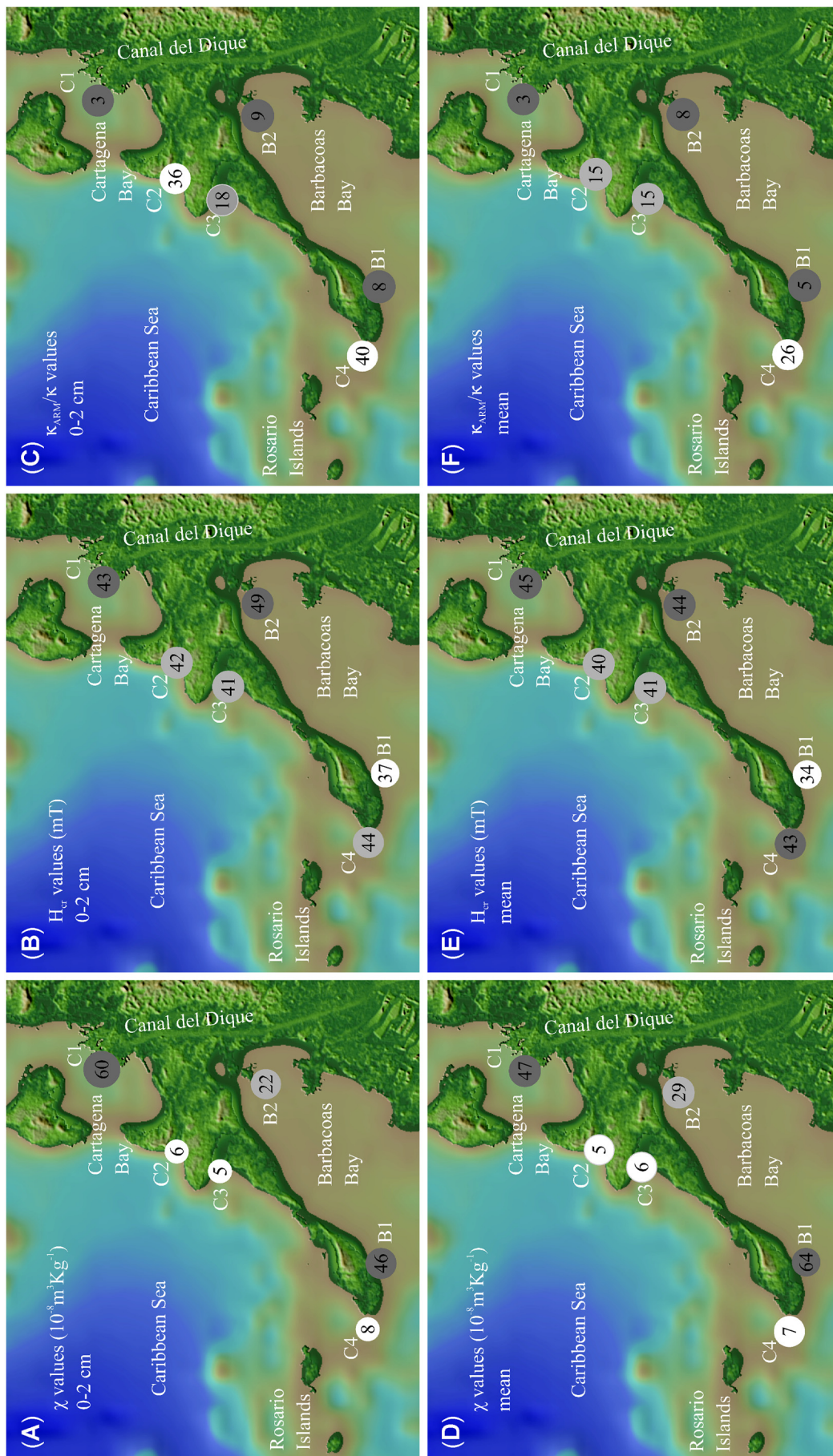


Fig. 8. Spatial and temporal variation of magnetic concentration and mineralogy dependent parameters  $\chi$  (higher values indicate higher magnetic concentration),  $H_{cr}$  and  $\kappa_{ARM}/\kappa$  (higher values indicate finer magnetic particles) in Cartagena and Barbaacoas bays. A-C) values of recently deposited sediments (0–2 cm). D-F) Whole core mean values.

channels (Restrepo et al., 2016a).

Core C4, sampled near the Rosario Islands, not only shows higher concentrations of ferrimagnetic minerals on the top of the core, but it also shows similar magnetic concentration parameters compared to other cores sampled in the Cartagena Bay (Fig. 7). This condition probably implies that the same sediments, which are delivered from Canal del Dique to the Cartagena Bay, are being transported all the way from the bay to the Rosario Island coral reefs and then being deposited over the calcareous shelf. In natural conditions, continental-bearing clastic sediments would be absent from environments like coral reefs. For instance, direct inputs of high loads of clastic sediments into coastal systems have catastrophic effects on the coastal sedimentary balance. An increasing input of sediments during the last decade has been recently witnessed in the Magdalena River and its distributary channel, the Canal del Dique, which has increased its sediment load by 48% between 2000 and 2011 (Restrepo et al., 2016a; Restrepo and Correa, 2014).

Parameters  $H_{cr}$ , contribution of magnetic phases, and S-ratio can be used to study the sediment provenance in this Caribbean area. Source-to-sink dispersal of continental/anthropogenic sediments in Cartagena Bay area can be traced from magnetic parameters, similar to other source-to-sink systems around the world (Hornig and Huh, 2011). Most of the sites show similar magnetic compositional characteristics (Fig. 6a and Table 1). This can be appreciated in Fig. 8, where ferrimagnetic minerals ( $H_{cr}$  values) in Periquitos (C4) are similar to Canal del Dique (C1) and Lequerica (B2).

There are grain size changes in time and space of magnetic particles for Cartagena and Barbacoas bays sediments. Magnetic grain size decrease (parameters  $\kappa_{ARM}/\kappa$  and ARM/SIRM increase) with distance from C1 to C4. Although smaller particles are deposited on more distal points or in closed systems as observed on C2, C3 and C4, their magnetic sizes show in-depth variations as well. In cores C2, B2 and C4, fine ferrimagnetic minerals are concentrated on the uppermost sediments (Fig. 3 and Fig. 8). Therefore, it is evident that the distribution of magnetic particles in this deltaic system has experienced changes with time showing different scenarios over the last decades.

## 6. Conclusions

The upward increase of ferrimagnetic mineral concentration in the sediment cores from Cartagena region reflect a marked sedimentation increase during the last decades, which is visible in parameters such as  $\chi$ , ARM, and SIRM. In Periquitos (core C4), far from Magdalena River delta, the arrival of finer ferrimagnetic minerals with a continental origin is confirmed from the magnetic mineralogy dependent parameters, such as  $H_{cr}$ , S-ratio,  $\kappa_{ARM}/\kappa$  and ARM/SIRM.

Magnetic particles deposited in the study area belong to magnetite minerals, with variable contributions of high-coercivity minerals on most locations. Magnetic mineralogical composition varies in the study area, while the  $H_{cr}$  values show a similar source of sediments for sampling points C1, C4, B1, and B2. Magnetic patterns obtained from core B1 (Punta Barú) seem to be different from the rest of the sampling sites. Here concentration parameters show a decreasing upwards behavior, similar to site B2 (Lequerica), a lower contribution of high-coercivity minerals, and magnetic grain sizes comparable to those observed in locations close to the coast.

The EF for elements Cu, Mo, and Zn indicates the additional continental and anthropogenic contribution on sediments close to the coast (sites C1 and B2) and site C4 located near the coral reefs (Rosario Islands National Park). However, the pollution contribution is minimal to moderate (EF = 2–5).

Magnetic grain size variability shows coarser particles deposited along the coastal area such as Canal del Dique and Lequerica mouths, and finer magnetic particles transported into distal zones. Besides such typical behavior of a deltaic system, the land-derived sediment has also experienced changes in magnetic size with time showing different

impacts over the last decades.

The integrated analysis of magnetic properties and trace elements presented here is an additional environmental proxy that corroborates the influx of land-derived sediment loads into the Rosario Islands coral reefs. Also, our results show the capabilities of applying environmental magnetism as a geoinicator to assess the presence and impact of continental fluxes on coastal ecosystems.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.marpolbul.2018.04.030>.

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