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RESEARCH ARTICLE

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Long-term trophic state responses of a large tropical coastal lagoon to land use changes and nutrient transport

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Abstract

The watershed of a large tropical coastal lagoon, historically hosting large sugarcane crops, which has been reduced during the last decades. It suggests that land use and land cover changes (LULCC) would lower the trophic state of the coastal lagoon due to less fertilization application and consequently reducing nutrient transfer into the lagoon. In this context, this study assessed the influence of LULCC, represented by the reduction of the sugarcane crops on the trophic state of a large tropical coastal lagoon. This influence was assessed by means of information about the long-term overall mass-balance budget, involving the estimation of the N and P loads from the sugarcane crops, other anthropogenic sources and natural processes; measurements of the river N and P fluxes into the lagoon; its seaward outflow and the long-term lacustrine nutrient retention. The results indicate that sugarcane crop reduction represented a decrease of 686 t of N and 51 t of P being emitted to the lagoon watershed. However, N and P loads from sugarcane crops were partially replaced by other anthropogenic activities, but even with the reduction of the nutrient fluxes, the long-term supereutrophic responses of the coastal lagoon was maintained. The sediment-related C/N and isotopic composition of the lagoon was sensitive to LULCC, indicating changes on the organic matter inputs into the lagoon. The trophic state was also influenced by the soil N and P accumulation and the long-term nutrient retention within the lagoon imposed by its restricted water connection to the sea.

KEYWORDS

agriculture, Atlantic Forest, isotopic composition, nitrogen, phosphorous, sugarcane plantation

1 | INTRODUCTION

Land use and land cover changes (LULCC) are highly diverse, leading to complex effects on aquatic ecosystems. The LULCC represented by deforestation, overgrazing and poor agricultural practices can induce an order of magnitude increase in erosion rates, affecting sedimentation and siltation of water bodies (Borelli et al., 2013; FAO, 2011; Li et al., 2007; Maranguit et al., 2017). In addition, differences in soil chemistry among diverse land uses and land covers may increase soil-bound and dissolved chemical loss to aquatic environments (Borelli et al., 2013; FAO, 2011; Fazhu et al., 2015; Groppo et al., 2015; Numata et al., 2007; Santos et al., 2013). Moreover, LULCC may also alter hydrology, as annual run-off is expected to increase as natural vegetation is replaced by other anthropogenic land covers

(Sterling et al., 2013). On the other hand, these inputs are suggesting that LULCC impacts on run-off vary from place to place (Li et al., 2009; Wang et al., 2012), as small effects on run-off were demonstrated when forests were altered to agricultural practices (Sajikumar & Remya, 2015).

Considering the negative LULCC effects on soil erosion, run-off and water chemistry, the literature has described that such changes have a direct impact on the water quality of aquatic environments, for example leading to eutrophication of inland and coastal waters (Huang et al., 2009; Mehdi et al., 2015; Nóbrega et al., 2018; Rodrigues et al., 2018; Yu et al., 2016). Lentic environments, such as coastal lagoons, are more vulnerable to eutrophication, in view of their physical features and the proximity of socio-economic activities, a primary source of nutrients, mainly nitrogen (N) and phosphorous (P) (Esteves et al., 2008; Kjerfve, 1994; Ménesguen & Lacroix, 2018). When natural vegetation is replaced by anthropogenic activities, such as urbanization, agriculture, animal husbandry and others, certain nutrients, such as N and P are increasingly mobilized across the landscape and may be ultimately exported to the coastal zone (Beusen et al., 2016; Paula-Filho et al., 2015; Pérez-Ruzafa et al., 2019).

Cultural eutrophication was widely recognized in studies from the beginning of 20th century, mostly in developed countries located in temperate regions of the northern hemisphere. Less information, in turn, was reported for lagoons from developing countries in the tropics, where LULCC has intensified and diversified during the last 50 years (FAO, 2007a; Lapola et al., 2014), and where eutrophication has been an important environmental and health issue (Azevedo et al., 2002; Campaneli & Molisani, 2019; Castro et al., 2016; Kosten et al., 2012; Le Moal et al., 2019). To date, many approaches have been applied to assess the spatiotemporal correlation between LULCC and the ecological status of aquatic environments (e.g., Geographic Information System-GIS, Life Cycle Assessment, geostatistical evaluation, stochastic modelling, nutrient budget and others), integrating views of the impacts beyond the typical ecological analysis of eutrophication (Bo & Qianqian, 2018; García-Ayllón, 2017; Ortiz-Reyes & Anex, 2018; Pérez-Ruzafa et al., 2019; Rodríguez-Gallego et al., 2017).

The LULCC in tropical regions such as in Brazil have exhibited the highest absolute rates of deforestation in the world, introducing low-productivity cattle pasture, while agriculture demands are continuously expanding (FAO, 2007b; Lapola et al., 2014). In one example, low-productivity pastures have been recently converted to sugarcane crops for ethanol production. From 1960 to 2007, sugarcane agriculture in Brazil increased from 1.4 million to 7.0 million ha, covering 2.5% of the country land area (FAO, 2007b; Rudorff et al., 2004). As example, sugarcane cover in a catchment in the State of São Paulo has increased from 7.0% to 26% between 1997 and 2007 which coincided with increased soil erosion and nitrogen pollution in adjacent aquatic environments (Martinelli & Filoso, 2008; Silva et al., 2007). In contrast, traditional sugarcane areas have reduced their production during the last decades, as water shortages to irrigation, increased soil erosion and nutrient deprivation, decreasing crop economic profits (Azevedo, 2004). It is expected that less sugarcane crops and, consequently, less fertilization may reduce nutrient soil input, also reducing the nutrient loss to surrounding aquatic environments, which could improve water quality in the long-term.

In this context, this study aimed to assess the influence of LULCC, represented by the reduction of the sugarcane crops, on the long-term trophic state of a large tropical coastal lagoon. This influence was assessed by means of mapping the LULCC and establishing the long-term trophic state of the lagoon and the mass-balance budget, involving the estimation of the relative N and P loads from the sugarcane crops, other anthropogenic sources and natural processes, measurements of the river N and P fluxes into the lagoon and its seaward outflow.

2 | MATERIAL AND METHODS

2.1 | Study area

The study area is located in the north of the State of Rio de Janeiro in Brazil. The annual sugarcane crop production in the region decreased from 7,091,450 to 1,200,000 tons from 1981 to 2017. The reduction of sugarcane crops and its possible effects were investigated at the Feia Lagoon, the second largest coastal lagoon in the Brazilian coast (Figure 1). It is located at the beach ridge and coastal plain geomorphologic unit and delimitated by the 'Serra do Mar' mountains. The lagoon was geologically formed during the Holocene after the last sea level change and the alterations of the medium-sized Paraiba do Sul River mouth alterations (Kjerfve, 1994; Martin et al., 1997). The lagoon drainage basin comprises 3,543 km^2, with a water surface area of 183 km^2 and depth varying between 0.3 to 2.2 m. Two small rivers feed the lagoon (Macabu and Ururai rivers) and the connection between the lagoon and the sea occurs by an artificial channel (Flechas channel) which is regulated by locks.

2.2 | Proposed method for assessing the effects of land use and land cover changes on the trophic state of the coastal lagoon

In order to assess the effects of the LULCC on the long-term trophic state of the large tropical coastal lagoon, we mapped the land transformation by using the GIS methodology and related the LULCC to the long-term trophic state of the lagoon and the N and P mass-balance budget for the entire aquatic system (Figure 2). The nutrient budget involved the estimation of the relative N and P loads from the sugarcane crops, other anthropogenic sources and natural processes, the measurement of the river N and P fluxes into the lagoon and its seaward outflow. Differences between N and P inputs and outputs provided an estimative of the nutrient retention within the lagoon and its watershed (Figure 2).

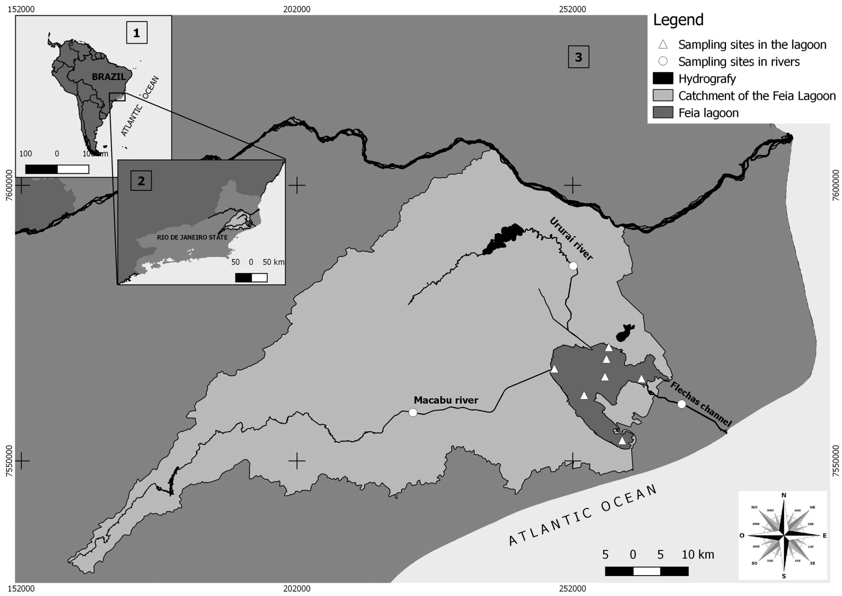

FIGURE 1 Map of the study area, including the sampling sites in the rivers and the seaward outlet channel (white circles) and in the lagoon (white triangle)

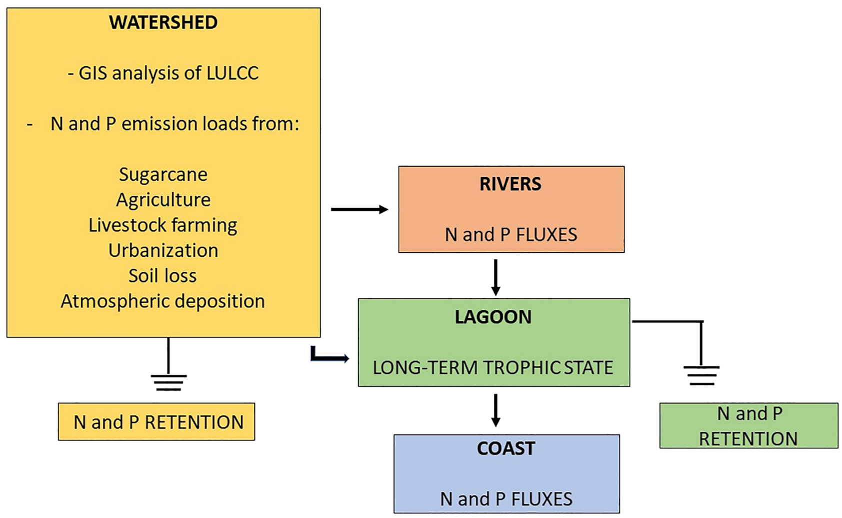

FIGURE 2 Flow diagram of the proposed method for assessing the effects of LULCC on the coastal lagoon eutrophication

2.3 | Mapping the land use and land cover changes

In the study area, the LULCC was assessed by the acquisition of 30 m-resolution LANDSAT 5 and LANDSAT 8 satellite images from 1984 and 2015 which represented, respectively, the maximum and minimum sugarcane producing period. Imaging processing and supervised classifications were performed using the ArcGIS10.1 software. ArcGIS 10.1-layer stacking was applied to convert composite bands (R6G5B4 bands) into a single layer for both the 1984 and 2015 images. A mask of the study area was created from the layer file using the *Extractbymask* command tool, followed by supervised image classification, collecting the polygon samples for each considered land use and cover classes. The Maximum Likelihood Classification method was applied, which ponders the distances between average class pixels by using statistical methods. The land use and cover classes were based on the level 2 of the Basic System of Land Use and Land Cover Classification set by IBGE-Instituto Brasileiro de Geografia e Estatística (2013), namely, forest, urban buildings, pasture, permanent and temporary crop fields, water bodies and vacant land. All classes were visually inspected in the field.

2.4 | Long-term nitrogen and phosphorous loads from the sugarcane crop, other anthropogenic activities and natural processes to the lagoon watershed

The temporal nitrogen (N) and phosphorous (P) loads from emission by the sugarcane crops and their relative importance on the set of natural and anthropogenic sources were evaluated by the Emission Factor Model (Lacerda et al., 2008; Nriagu & Pacyna, 1988; Paula-Filho et al., 2015) for the periods from 1978 and 1981 and from 2012 and 2015. This approach estimates N and P loads from natural processes such as soil leaching and atmospheric deposition and human activities, including urbanization, agriculture and animal husbandry. The data input includes the compilation of the range or mean values of parameters from the literature and from local conditions that may alter the reliability of the emission factors.

The N and P loads from atmospheric deposition are directly related to the basin area and the nutrient concentrations of the atmospheric deposition, adjusted by the retention rate of each element by the diversity of soils present in the basin, which were estimated according the Equation 1:

$$L^M_{atm} = \rho_{rw} \cdot A_{ws} \cdot (1-\alpha_{rs})/10^3 \quad (1)$$

where *LMatm* comprise atmospheric deposition loads (t year^{-1}), ρ*RW* is the deposition of each nutrient depending on rainwater concentrations, that is, the annual rainfall in the watershed (mg m^2 year^{-1}), *Aws* is the basin area (km^2) and α*rs* is the soil retention factor. Considering the incipient industrialization and urbanization of the study area, average N and P bulk atmospheric deposition for the Brazilian coast were applied, as follows: for N: 100 mg m^{-2} year^{-1} and P: 6.0 mg m^{-2} year^{-1}, considering the proposed range for N of 80–300 mg m^{-2} year^{-1} and for P of 4.0–10 mg m^{-2} year^{-1} (Mello & Almeida, 2004). These average values were weighted by the basin area and average annual rainfall for both periods (779 and 818 mm) (INMET–Instituto Nacional de Meteorologia, 2018) and corrected by the soil retention rates for each nutrient (N: 65% and P: 70%) (Golley et al., 1978; Malavolta & Dantas, 1980).

The N and P emissions from the physical and chemical denudation of diverse soil types of the watershed were calculated according to Equation 2:

$$L^M_s = \sum_{J=1}^{4} \rho sj.\, A_j.\, L_s.(1-\alpha_{rs}) \quad (2)$$

where $L^M{}_s$ represents each nutrient load from soil loss (t year^{-1}) as a function of ρ*sj* which is the concentration of each nutrient in the different soil types of the watershed, A_j is the area corresponding to each type of soil (km^2); L_s is the average soil loss of 410 t km^{-2} year^{-1} obtained from the proposed range of 60–760 t km^{-2} year^{-1} for tropical soils from gentle slopes and highly mechanized agriculture for Brazilian coastal areas (Goudie, 1987; Greenland & Lal, 1977); α*rs* is the soil retention factor for the different soils types (Latosol, Cambisol, Sand, Podzols, Alluvial/Regosol) in the watershed, and the average soil nutrient concentration from Brazilian coastal areas presenting low declivity and industrial development. The average N and P concentrations in soils (mg g^{-1}) are respectively: Alluvial soils: 900 and 500 mg g^{-1}; Sand: 500 and 100 mg g^{-1}; Latosols/Podzols; Planosols and Solonchack/Solonetz: 500 and 500 mg g^{-1} (Cantarella, 2007; Silva, 1996).

The N and P emission from domestic wastewater was assumed as non-treated prior to release into the watershed, which is noted in the study area. Domestic effluent loads are directly proportional to the average nutrient concentrations in wastewater (N: 52 mg L^{-1} and P: 15 mg L^{-1}) (Mota & Von Sperling, 2009); the watershed population (IBGE Cidades, 2014) and the amount of water consumed per capita (125 L inhab^{-1} day^{-1}) for rural inhabitants and 220 L inhab^{-1} day^{-1} for the urban population (IBGE-Instituto Brasileiro de Geografia e Estatística, 2013) which were calculated according to Equation 3:

$$L^M_{ww} = \sum_{i=1}^{2} \left(\frac{P_{ww}.\, P_u i.\, Q_u i.\, \beta.\, 365}{10^9} \right) + \sum_{i=1}^{2} \left(\frac{P_{ww}.\, P_r i.\, Q_r i.\, \beta.\, 365}{10^9} \right) \quad (3)$$

where $L^M{}_{WW}$ are the nutrient loads from untreated urban and rural domestic sewage in the watershed (kg yr^{-1}); P_{ww} are sewage concentrations (mg L^{-1}), P_ui and P_ri comprise the urban and rural populations, respectively, in each municipality within the lagoon watershed; Q_ui and Q_ri indicate the urban and rural water consumptions per capita, respectively, and β is the water/sewage return rate.

In order to calculate the nutrient loads emitted from urban run-off, an estimated low urbanization area of 0.69% to 0.89% for both inter-decadal periods were applied, as well as an average annual rainfall for each period (779 and 818 mm, respectively) and average N and P concentrations in urban run-off (N: 2.0 mg L^{-1} and P: 0.33 mg L^{-1}) (NCR-National Research Council, 2000), as presented in Equation 4.

$$L^M_{U_{rf}} = \left(\frac{p_{U_{rf}}.A_{Ui}}{10^6} \right) \quad (4)$$

where $L^M{}_{Urf}$ are the estimated nutrient loads resulting from urban run-off for the lagoon watershed; p_{Urf} are the concentrations obtained from urban run-off and annual rainfall and A_{Ui} is the urban area within the basin (km^2).

Emissions from solid waste disposal are given as a function of the watershed population, the average solid waste per capita production of 0.45 kg inhab^{-1} day^{-1} (IPEA–Instituto de Pesquisa Econômica e Aplicada, 2012), and the average N and P concentration in domestic fresh solid wastes (N: 8.9 g kg^{-1} and P: 5.6 g kg^{-1}) (Hjelmar et al., 2000). The estimates were corrected by N and P soil retention (Golley et al., 1978; Hadas et al., 2004). The Brazilian context in which improper waste disposal averages 42% of total solid waste produced (ABRELPE-Brazilian Association of Public Cleaning and Special Waste, 2011), as described in Equation 5, was applied for the basin:

$$L^M_{sw} = P_{sw}.\, Pi.\, G_{sw}.\, 365.\, (1-\alpha_{rs})/10^9 \quad (5)$$

where $L^M{}_{sw}$ are the nutrient loads from solid waste disposal within the basin; P_{sw} are the mean nutrient concentrations in the municipal solid waste; P_i is the population within the lagoon watershed; G_{sw}i is the per capita production of wastes; α_{rs} is the soil retention rate and δ_{sw} is the adequacy factor according to the type of the waste disposal.

Crop fertilizers are important parameters responsible for N and P emissions by agriculture (Embrapa, 2009; Filoso et al., 2006). Equation 6 was used to calculate the emission estimates from this source.

$$L_A^M = \sum_{j=1}^{5} \left(\frac{P_{tj} . A_{tij}}{10^3} \right) \tag{6}$$

where $L^M{}_A$ are the N and P loads from the sugarcane crops and the most common crops cultivated in the watershed (sugarcane, coconut, orange, coffee, corn, rice, bean, banana and cassava); P_{tj} are the N and P loads applied as fertilizer (kg ha^{-1}) (Embrapa, 2009; Filoso et al., 2006) and the nutrient loss percentage according to crop type (Golley et al., 1978; Silva et al., 2000) and A_{tij} is the cultivated area (ha $year^{-1}$) of each crop (IBGE Cidades, 2014).

The nutrient emissions from livestock farming were described in Equation 7:

$$L_{LF}^M = \sum_{j=1}^{4} p_{tj} . LF_{ij} . (1 - \alpha_{rs})/10^9 \tag{7}$$

where $L^M{}_{LF}$ are the loads originated from animal manure in the region (meat and dairy cattle, chicken, swine and horse), P_{tj} is the emission factor related to the annual amount of manure produced per animal in the watershed (*j*) (10, 2.5, 1.0 and 0.18 kg $animal^{-1}$ day^{-1}, for meat cattle, horses, swine, and chicken, respectively) and the N and P concentrations in raw manure (mg kg^{-1}) (Embrapa, 2004); LF_{ij} is the number of animals within the basin (*i*) (IBGE Cidades, 2014) and α_{rs} is the soil retention rate (Malavolta & Dantas, 1980; Silva, 1996).

The accuracy of the emission factor estimates was obtained comparing such results to the nutrient loads transported by the rivers to the lagoon, as well as, discussing the relevance of collected information and model uncertainties as a first-order estimative method.

2.5 | Measurements of the river N and P input, seaward output and overall mass-balance budget for the costal lagoon

The N and P concentrations and fluxes were determined for the main river tributaries flowing into the coastal lagoon and then to the sea by the channel outlet (Figure 1). The water sampling was coordinated by the Environmental Institute of the Rio de Janeiro (INEA) and comprised the years from 1978 to 1981 (14 samplings) and from 2012 to 2015 (15 samplings), respectively, the maximum and minimum sugarcane producing periods.

Water discharges were obtained from monitoring stations located on the river tributaries. Concerning the water discharges from the lagoon to the sea through the outlet channel, data on regularized flows from locks were obtained only for the 2012 and 2015 period. Lock operational routines are based on the 15 locks that regularize each water discharge of 14 m^3 s^{-1}. Personal communications concerning operational lock rules indicated that the channel locks have remained closed most of the time since their installation, regulating the lagoon volume according the water users, mainly for sugarcane crops.

Water samples were collected from both rivers in the same place of the discharge monitoring stations and in the outlet channel. In each sampling site, water was collected using a high-density PET bottle, stored in clean (2% detergent, 1.0 N HCl and pre-rinsed with local water) flasks and sent in an icebox to the INEA laboratory. At the laboratory, the unfiltered samples were simultaneously digested for total nitrogen (TN) and total phosphorous (TP) (persulphate method) determination (APHA-American Public Health Association, American Water Works Association–AWWA and Water Environment Federation–WEF, 2005). All analyses were performed in duplicate using analytical blanks, and precisions between replicates for all nutrients were less than 10%.

The instantaneous N and P fluxes were calculated by multiplying the nutrient concentrations and the river discharges of the tributaries. The annual fluxes were estimated using the average instantaneous load displayed in Equation 8 (Preston et al., 1989):

$$F = k \sum_{i=1}^{n} \frac{C_i Q_i}{n} \tag{8}$$

where *F* is the annual fluxes (t yr^{-1}), Ci comprises the N and P concentrations (mg L^{-1}), Q_i is the concomitant instantaneous flow (m^3 s^{-1}), *n* is the number of days presenting concentrations and flow data and K is the conversion factor used for the annual period. Seasonal fluxes were also calculated, grouping data for the dry (April to October) and rainy (November to March) seasons.

The N and P mass-balance budget was constructed from both 1978–1981 and 2012–2015 periods considering the fluxes from natural process and human activity emissions, the inputs from rivers into the lagoon and the lagoon's output to the sea. The difference between fluxes from the emission sources and the river inflow into the lagoon estimated the nutrient retention into the lagoon's watershed. Similarly, the balance between the river inputs into the lagoon and the outflow to the ocean estimated the N and P retention into the lagoon. However, this budget was calculated only for the 2012 and 2015 period, when the regulated water discharges of the seaward outlet channel were available.

2.6 | Long-term trophic state and limnology of the coastal lagoon

Simultaneously to the river monitoring, the limnological parameters were determined at seven sampling sites across the lagoon during the period from 1978 to 1981 ($n = 14$) and from 2012 to 2015 ($n = 13$) (Figure 1). This limnological survey was also coordinated by the Environmental Institute of the Rio de Janeiro (INEA). Such parameters included the water column and Secchi depths, water temperature, dissolved oxygen, pH, electrical conductivity, obtained by using specific probes at different depths (subsurface and bottom). Dissolved oxygen was determined by the titration Winkler method. Water samples were collected in the sub-surface and bottom of the water column using a Van Dorn bottle sampler, stored in clean (2% detergent, 1.0 N HCl and pre-rinsed with local water) flasks and sent in an icebox to the INEA laboratory for turbidity, chlorophyll a, total phosphorous, orthophosphate, nitrate, nitrite, ammonium, suspended particle and phytoplankton density analyses.

Duplicate water samples were filtered through a 0.45-μm membrane filters after collection and the filtrates were frozen until analysis. Filters were used to obtain the suspended particle concentrations by the gravimetric procedure. Filtrate water samples were analysed by spectrophotometric methods for ammonium (NH_4^+, indophenol blue method), nitrate and nitrite (NO_3^-, NO_2^-, Cd reduction method) and orthophosphate (PO_4^3, ascorbic acid molybdate method) (APHA-American Public Health Association, American Water Works Association–AWWA and Water Environment Federation–WEF, 2005). Non-filtered samples were simultaneously digested for total nitrogen (TN) and total phosphorous (TP) (persulphate method) determination by UV/Vis spectrophotometry (APHA-American Public Health Association, American Water Works Association–AWWA and Water Environment Federation–WEF, 2005). Water samples were also filtered through glass fibre filters to determined chlorophyll-*a* concentrations after 90% acetone extraction by the spectrophotometric method (APHA-American Public Health Association, American Water Works Association–AWWA and Water Environment Federation–WEF, 2005). For quantitative phytoplankton analyses, water samples were preserved in situ with 1% Lugol solution in opaque glass bottles. A sedimentation method was used for counting under an inverted microscope, according to the Utermöhl technique (400 × magnification). Phytoplankton were enumerated until at least 100 organisms (cell, colonies and filaments) per dominant species were recorded, at a precision of ± 20% within 95% confidence limits (Wetzel & Likens, 2000).

The long-term trophic state of the coastal lagoon was obtained using the index proposed by Lamparelli (2004), adapted from Carlson (1977), for shallow lentic environments. The index is composed by a set of equations (Equations 9 and 10):

$$\mathrm{TSI}\,(\mathrm{Chl}) = 10\,(6 - (0.92 - 0.34\,(\ln \mathrm{Chl}/\ln 2)) \tag{9}$$

$$\mathrm{TSI}\,(\mathrm{TP}) = 10\,(6 - (1.77 - 0.42\,(\ln \mathrm{TP}) - \ln 2)) \tag{10}$$

where Chl = Chlorophyll-*a* (μg L^{-1}); and TP = Total phosphorous (μg L^{-1}). The trophic limits defined were: Ultra-Oligotrophic: ≤ 47; Oligotrophic: 47 < TSI ≤ 52; Mesotrophic: 52 < TSI ≤ 59; Eutrophic: 59 < TSI ≤ 63; Supereutrophic: 63 < TSI ≤ 67; Hypereutrophic: >67. We calculated the TSI (Chl) and TSI (TP) for each sampling event, using the mean spatial concentration of each variable and then calculated the trophic state of the lagoon, based on the average between the TSI (Chl) and TSI (TP) values. The trophic state index for each sampling event was yearly averaged and the long-term trophic state was calculated as the average of annual TSI for both quadrennium.

The effects of LULCC on nutrient transport to the lagoon was analysed by one sediment core sampled in the southwest portion of the lagoon. The core was sliced at 2 cm intervals and at the laboratory, organic carbon (OC), $\delta^{13}C$, total nitrogen (TN) and $\delta^{15}N$ were determined for each interval. A mass of 10 mg was weighted in silver capsules, followed by acidifications through the addition of HCl (2 M) to remove carbonates (Brodie et al., 2011). Analysis was performed with an Elemental Analyser (Flash 2000) with interface CONFLO IV coupled to an isotope ratio mass spectrometer Delta V Advantage (Thermo Scientific, Germany). Quantification was performed using analytical curves from acetanilide for elemental composition standards (Elemental Microanalysis), with inter-replicate precision close to 97%. Accuracy was verified using the Standard Elemental Microanalysis/isotope—Low Organic Soil (1.52 ± 0.02% for C; 0.13 ± 0.02% for N; −27.5 ± 0.11% for $\delta^{13}C$; 6.70 ± 0.15% for $\delta^{15}N$), with above 98% recovery. The OC and TN content were expressed as percent (%) element. The detection limits of analytical methods were 0.05% and 0.02% for OC and TN, respectively. Carbon and nitrogen isotope ratios were expressed as ‰ relative to Pee Dee Belemnite (PDB) and atmospheric nitrogen, respectively, with analytical precision of 0.1‰.

In order to determine whether changes in the limnological parameters, including trophic state, were statistically significant between the inter-decadal period, the rainy and dry season and in the surface and bottom water column of the lagoon, an unequal variance t-test for independent groups was applied. Results were considered significant at a probability level of less than 5%. All analyses were carried out using the GraphPad Prism 5.0 (GraphPad software, USA).

3 | RESULTS

3.1 | Land use and land cover changes of the lagoon watershed

The Figure 3 indicates the LULCC that occurred at the Feia Lagoon watershed for 1984 and 2015, respectively. During the interdecadal period, sugarcane crop areas decreased by 32% and were replaced by permanent crops, vacant lands, urbanization, pasture and forests, which increased their areas, respectively, by 276%, 103%, 28%, 6.7% and 1.9%. However, considering the absolute area, pastures were the main land use and cover that replaced sugarcane crops covering most of the watershed, followed by forests, temporary crops and sugarcane, vacant lands, urbanization and permanent crops.

3.2 | Long-term relative importance of sugarcane crop, other anthropogenic activities and natural processes on the N and P loads to the lagoon watershed

Comparing 1984 to 2015, the sugarcane crops reduced their relative nutrient loads to all other sources from 54% to 46% for N and from 14% to 11% for P. For the entire period, N loads from the sugarcane crops to the lagoon watershed decreased from 2753 to 2067 t $year^{-1}$ while P reduced from 203 to 152 t $year^{-1}$ (Table 1). The anthropogenic activities were the main N and P sources acting throughout the watershed, but slightly reduced their total emission during the sampling period, mainly due to the observed sugarcane crop reduction.

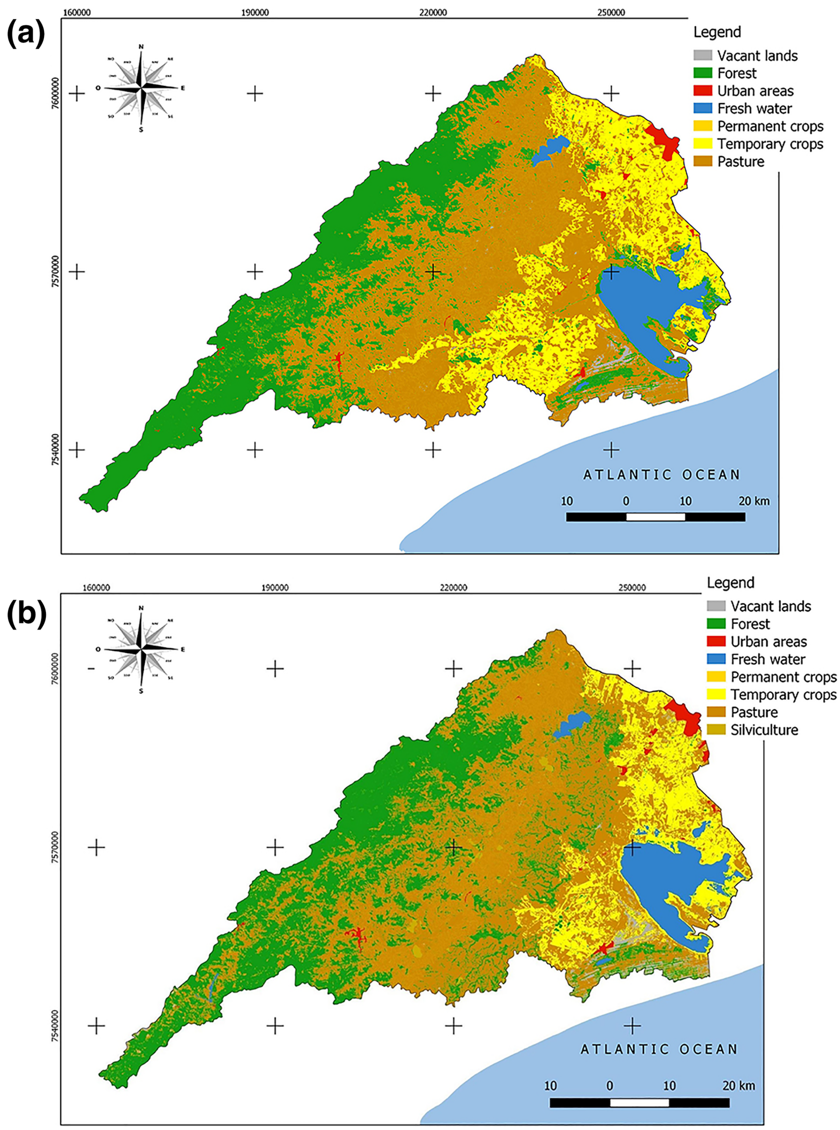

FIGURE 3 Land use and land cover changes of the lagoon watershed for 1984 (a) and 2015 (b)

TABLE 1 Temporal N and P loads (t $year^{-1}$) and relative importance (%) of sugarcane crop, natural processes and other anthropogenic emissions to the coastal lagoon watershed

	N		P	
Sources	1978–1981	2012–2015	1978–1981	2012–2015
Total emission	5085	4471	1484	1404
Sugarcane crop	2753 (54%)	2067 (46%)	203 (14%)	152 (11%)
Soil denudation	655	655	10	10
Atmospheric deposition	52	56	4.0	4.0
Total—Natural processes	717 (14%)	711 (16%)	14 (<1%)	14 (<1%)
Temporary and permanent agriculture	2814	2081	226	164
Livestock farming	968	855	990	861
Domestic waste water	582	818	254	365
Municipal solid wastes	0.3	0.3	0.1	0.1
Urban run-off	4	6	1	1
Total—Anthropogenic sources	4368 (86%)	3760 (84%)	1471 (99%)	1391 (99%)

Among anthropogenic activities, agriculture and livestock farming were the main N and P sources to the watershed, respectively. Considering agriculture crops, sugarcane was the main source, even after decreased crop cultivation, followed by coconut and mango crops. Among pastures, cattle for meat production were the main watershed herd, with increased production during the sampling period (39%), but other herds presented an expressive reduction. Urbanization represented by untreated domestic sewage emissions has a relatively N and P contribution to the watershed, presenting increasing values from 1984 to 2015, when the watershed population increased from 163,979 to 224,102 inhabitants. Among natural processes, soil denudation, mainly from Podzols, Latosols and Gleisols, which comprise the largest areas, were the main natural sources of N and P to the watershed.

3.3 | Determination of river N and P inputs, seaward outflow and mass-balance budget for the coastal lagoon

During the first quadrennium (1978–1981), the water discharge of tributaries into the lagoon ranged from 3.1 to 34 $m^3 s^{-1}$. The nitrogen concentrations varied from 50 to 450 $\mu g L^{-1}$ while phosphorous varied from 10 to 660 $\mu g L^{-1}$. For the 2012 and 2015 quadrennium, discharges varied from 1.5 to 124 $m^3 s^{-1}$. The nitrogen concentrations of tributaries varied from 20 to 1,230 $\mu g L^{-1}$ and phosphorous levels varied from 10 to 350 $\mu g L^{-1}$. Monitoring information for the seaward outlet of the lagoon was available only for 2012–2015 period, while the locks were closed and the seaward outflow from the lagoon was restricted during 62% of the monitoring period. When the locks were opened, water discharge to the ocean varied from 27 to 154 $m^3 s^{-1}$. Nitrogen concentrations in the outlet channel were higher (70–3,-300 $\mu g L^{-1}$) than in the contributing rivers, while values were similar for P (20–350 $\mu g L^{-1}$). Average fluxes indicated that rivers contributed with 173 t $year^{-1}$ for N and 84 t $year^{-1}$ for P to the lagoon during 1978 and 1981, and with 265 t $year^{-1}$ for N and 115 t $year^{-1}$ of P during 2012 and 2015. Higher nutrient fluxes to the lagoon during the second quadrennium are attributed to the higher water discharges from the rivers during this period. From 2012 to 2015 period, the seaward N and P fluxes from the controlled channel were measured at 161 t $year^{-1}$ and 34 t $year^{-1}$, respectively (Figure 4).

The mass-balance budget was calculated including the N and P loads from natural and anthropogenic emissions, the river inflow into the lagoon and then to the sea by the lagoon's outlet channel (Figure 4). Comparing the N and P fluxes from the emission sources and the river inputs into the lagoon, 94% of N and 91% of P loads emitted from the sugarcane crops, other anthropogenic activities and natural processes were retained across the watershed soils. When comparing the river inputs into the lagoon and the seaward outflow from the outlet channel, the budget indicated that 60% of N and 30% of P inputs int the lagoon by rivers remained within the lagoon. From the total nutrient loads from the natural and anthropogenic sources across the watershed, only 3.6% and 2.4% of N and P, respectively, were exported to the sea. The retention status observed for the 2012–2015 period might be assumed as also occurring for the 1978–1981 period, considering that channel locks have remained closed most of the time since their installation, regulating the lagoon volume according the water users. Thus, the lagoon would always have retained fluvial N and P fluxes.

3.4 | Long-term trophic state and limnology of the coastal lagoon

The inter-decadal limnological measurements described the trophic state responses to the long-term land use and land cover changes and nutrient transport across the watershed (Table 2). The lagoon's limnology between both quadrennia showed differences for water temperature ($p < 0.0001$) and electrical conductivity ($p < 0.01$), which were lower during the 2012 and 2015 period; and pH ($p < 0.05$), total phosphorous ($p < 0.01$), nitrate ($p < 0.05$) and nitrite ($p < 0.01$), which were higher during this period. The other variables displayed similar conditions between both periods.

When evaluating the data per season in both decadal sampling periods, few limnological parameters displayed significant differences between the dry and rainy seasons within the decadal, with PO_4^{3-} and Secchi depths higher during the dry season and temperature, pH and chlorophyll *a* and phytoplankton density higher in the rainy season (Table 2). This seasonal analysis suggests a reduced variability of the lagoon's limnology, which was also confirmed when a comparison between the surface and the bottom of the water column was carried out. Only water temperature was statistically higher at the surface compared to the bottom water column, while all other limnological parameters were similar at the surface compared to bottom waters.

For the trophic state calculation, the average total phosphorous concentrations were 93.7 ± 65.9 $\mu g L^{-1}$ and 113 ± 113 $\mu g L^{-1}$ for the first and second quadrennium, respectively; while for chlorophyl, the mean values were 12.9 ± 11.9 $\mu g L^{-1}$ and 14.3 ± 11.8 $\mu g L^{-1}$,

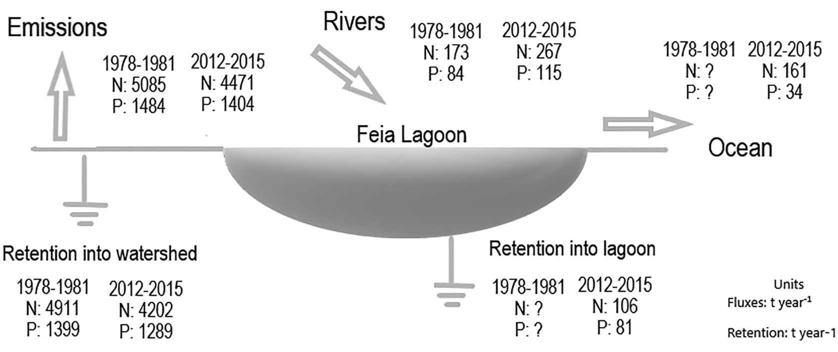

FIGURE 4 Overall N and P mass-balance budget for the coastal lagoon (t $year^{-1}$), including the natural and anthropogenic emission loads, river inputs and the output to the ocean. The sinkhole symbol represents the N and P retention into the watershed and lagoon

TABLE 2 Mean ± standard deviation and range of limnological parameters and trophic state of the large tropical coastal lagoon between the sampling periods and seasons (symbol represent that means are significantly different at $p < 0.05$)

			1978–1981		2012–2015	
	1978–1981	**2012–2015**	***Dry***	***Wet***	***Dry***	***Wet***
Water column depth (m)	1.26 ± 0.56	1.46 ± 0.27	1.19 ± 0.4	1.32 ± 0.7	1.45 ± 0.3	1.47 ± 0.2
	0.23–2.7	0.60–2.0	0.80–2.0	0.23–2.7	0.60–1.8	0.80–2.0
Secchi depth (m)	0.38 ± 0.37	0.42 ± 0.38	0.46 ± 0.4†	0.23 ± .0.1†	0.47 ± 0.4	0.39 ± 0.3
	0.05–2.00	1.70	0.1–2.0	0.15–0.4	0.1–1.7	0.1–1.1
Water temperature (°C)	25.0 ± 2.9†	21.0 ± 2.5†	24 ± 2.4†	29 ± 1.6†	20 ± 1.8†	24 ± 2.3†
	20.1–32.0	17.9–26.7	20–32	26–31	17–25	19–27
Dissolved oxygen (mg L^{-1})	7.17 ± 1.88	6.86 ± 2.08	7.3 ± 1.2	6.8 ± 2.6	7.3 ± 1.8	6.1 ± 2.1
	1.4–9.0	1.8–10	3.6–9.0	1.4–9.0	1.9–10	2.4–8.4
Conductivity (μS cm^{-1})	172 ± 130†	105 ± 63.2†	152 ± 77	118 ± 51	108 ± 74	99 ± 37
	30–627	23–432	52–356	30–183	45–432	46–173
pH	6.39 ± 0.77†	6.77 ± 0.65†	6.5 ± 0.7	5.9 ± 0.8	6.6 ± 0.7†	7.0 ± 0.5†
	4.7–7.6	4.3–7.9	4.7–7.2	4.7–7.3	4.3–7.9	6.2–7.6
Turbidity (TNU)	68.8 ± 57.3	56.7 ± 62.0	67 ± 57	88 ± 36	52 ± 57	63 ± 68
	11–239	17–314	11–239	41–142	1.7–225	5.0–314
Suspended particle (mg L^{-1})	68.1 ± 59.8	48.8 ± 49.6	67 ± 61	87 ± 37	46 ± 51	57 ± 57
	6.0–240	0.70–241	6.0–240	37–141	2.0–191	9.0–241
Nitrate (μg L^{-1})	77.3 ± 89†	111 ± 75†	45 ± 34†	210 ± 117†	119 ± 73	77 ± 71
	10–134	10–320	10–156	50–437	10–320	20–230
Nitrite (μg L^{-1})	7.40 ± 7.10†	16.8 ± 15.5†	6.0 ± 6.0†	9.0 ± 9.0	20 ± 18†	8.0 ± 1.0
	1.0–40	10–80	1.0–26	2.0–36	5.0–80	7.0–10
Ammonium (μg L^{-1})	87.3 ± 90.4	121 ± 136	80 ± 90	120 ± 110	130 ± 160	100 ± 40
	10–400	10–770	10–400	10–328	10–770	60–170
Total P (μg L^{-1})	93.7 ± 65.9†	113 ± 113†	85 ± 69	96 ± 56	115 ± 120	122 ± 101
	10–290	20–610	10–290	10–206	20–610	20–501
Orthophosphate (μg L^{-1})	30.2 ± 26.8	25.5 ± 18.4	36 ± 28†	12 ± 4.0†	31 ± 23 $^{\psi}$	22 ± 10 $^{\psi}$
	10–100	10–110	10–100	10–20	10–110	10–40
Chlorophyll *a* (μg L^{-1})	12.9 ± 11.9	14.3 ± 11.8	19 ± 9.2†	33 ± 22†	17 ± 13	11 ± 10
	0.70–64	0.1–50	1.0–39	8.9–64	1.0–50	0.5–35
Phytoplankton Density (10^6 organisms L^{-1})	5.12 ± 5.51	3.12 ± 3.44	3.5 ± 2.6†	14 ± 9.5†	3.6 ± 0.37	2.5 ± 0.32
	0.43–25	0.087–17	0.43–9.8	0.29–25	0.26–17	0.18–11
Trophic state index	64 ± 3.0	65 ± 2.3	63 ± 3.0	65 ± 4.0	65 ± 3.0	64 ± 2.0
	58–70	61–70	58–67	62–68	61–70	61–65
Trophic state	Supereutrophic	Supereutrophic	Supereutrophic	Supereutrophic	Supereutrophic	Supereutrophic
	Mesotrophic–Hypereutrophic	Eutrophic–Hypereutrophic	Mesotrophic–Hypereutrophic	Eutrophic–Hypereutrophic	Eutrophic–Hypereutrophic	Eutrophic–Supereutrophic

respectively. Such variables presented a large concentration range during the long-term monitoring of the lagoon limnology (Table 2). On average, the trophic state index calculated for both decadal periods were 64 and 65, respectively, classifying the lagoon as supereutrophic for both the higher and lower sugarcane production periods, respectively. However, analysing each sampling event, the lagoon's trophic state varied from mesotrophic to hypereutrophic during the 1978–1981 period and from eutrophic to hypereutrophic for the 2012–2015 period (Table 2).

The long-term C/N ratio and $\delta^{13}C$ and $\delta^{15}N$ composition were registered in the lagoon's sediments (Figure 5). The temporal profile revealed the past composition (assumed in the sediment core layer 16–80 cm) with a C/N ratio from 12.9 to 31.2, while $\delta^{13}C$ varied from −25.4‰ to −28.7‰ and $\delta^{15}N$ from 5.6 to 2.2‰. In more recent

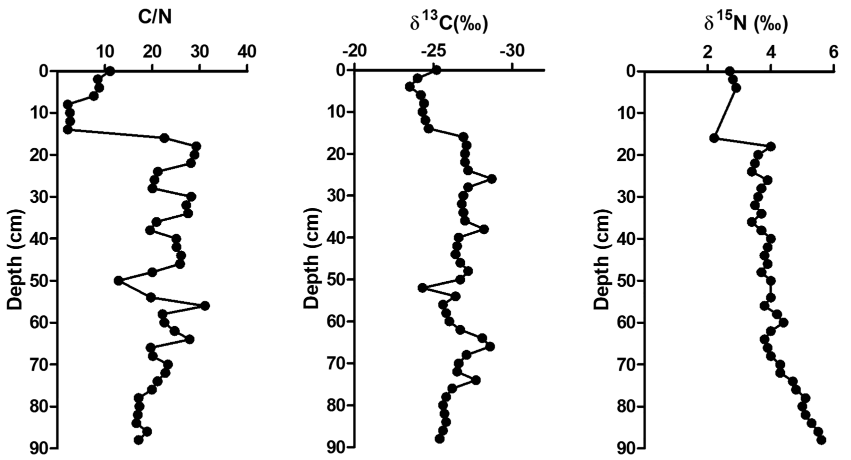

FIGURE 5 Elemental and isotopic C and N composition of the lagoon sediment

study period (assumed in the sediment core layer 8.0–14 cm), C/N ratio was strongly reduced to about 2.2–2.7, while $\delta^{13}C$ ranged from -24.3 to -24.7‰ and $\delta^{15}N$ from 2.0‰. The grain size distribution of the surface up to 14 cm was dominated by fine sandy fraction with a mean concentration of OC and NT was 0.18% to 0.02%, respectively. After this depth, there is an increase OC and NT (average, 5.04% and 0.27%, respectively). These signal changes in the hydrodynamics and the depositional environment of the lagoon.

4 | DISCUSSION

The reduction of the sugarcane crop and replacement by permanent crops, vacant lands, urbanization, pasture and forest did not follow the most reported LULCC in Brazil, where crops have been inserted in abandoned or low-productivity cattle pastures, and forests have made way for crops and pasture (FAO, 2007a; Lapola et al., 2014). In the study area, the sugarcane crop area decreased by approximately 32%, and such LULCC represented a reduction of 686 and 51 t, respectively, of N and P being emitted to the Feia Lagoon watershed.

In many areas in Brazil, sugarcane as annual crops require about 3.13 million tons of fertilizers, with application rates of 80–100 kg ha^{-1} $year^{-1}$ (Martinelli & Filoso, 2008). However, N uptake efficiency by sugarcane is low, with only 20–40% of the applied N being assimilated by plant tissues (Basanta et al., 2003). In addition, N fertilizers can be lost as NH_3 to the atmosphere by volatilization or during nitrification from soils or plant senescence, but may return to watersheds as dry (NH_3) or wet (NH_4^+) depositions (Holland et al., 1999). Compared to other crops, N loads from fertilizer in sugarcane crops are relatively low, but lead to N soil excess. Moreover, sugarcane also influences soil removal, as changes from natural forest vegetation to intensive sugarcane crops increase soil loss rates from 0.03 to 12.6 t ha^{-1} $year^{-1}$ (Junior et al., 2019). As a consequence, the intensification of N and P fertilization and soil loss in sugarcane crop fields increased nutrient exports to rivers (Filoso et al., 2006) that undergo different pathways, including run-off and leaching to groundwater depending on soil characteristics (Basanta et al., 2003; Oliveira et al., 2000, 2002).

During the sugarcane peak production, this activity was the main N and P emission source contributing to the supereutrophic state of the lagoon. However, over the study years, sugarcane crops were replaced by other anthropogenic activities, leading to slightly reduced N and P emissions (13 and 5.3%, respectively), mainly those related to the livestock farming, agriculture and less for urbanization (untreated domestic sewage, solid waste disposal and urban run-off), which is usually reported as the main nutrient source for many regional coastal lagoons, causing long-term eutrophic conditions (Campaneli & Molisani, 2019).

The emission factor accuracy was verified by comparing the calculated loads to the measured N and P flows from the river tributaries to the Feia Lagoon. The N and P loads calculated by the Emission Factor Model (N: 4,471 and P: 1,404 t $year^{-1}$) were higher than the river fluxes (N: 266 and P: 12 t $year^{-1}$). The differences were attributed to (1) the upstream position of the gauging stations in the tributary rivers that underestimate the N and P fluxes; (2) sampling limitation to measure large river flows during episodic rainfall events which transport most of the nutrients; (3) unknow nutrient fluxes retained by two reservoirs located at both contributing rivers; (4) nutrient retention across the coastal plain and decrease of downstream export by rivers (Molisani et al., 2021; Reddy et al., 1999). Concerning the input data for the Emission Factor Model, the reduced availability of some regional data for calculations contributed to the estimate's uncertainties. The smaller uncertainties are associated to the atmospheric deposition, domestic wastewater and urban run-off, for which more regional data is available in the literature. Higher uncertainties are attributed to municipal solid waste, due to the diversity of residues and related N and P concentrations. Emission factors calculated for soil denudation, agriculture and animal husbandry have large uncertain due to the ample soil distribution and nutrient retention capacity. Depending on the element and the emission category, the calculated N and P loads may vary from 4 to 23-fold, within the range proposed by the literature (Lacerda et al., 2008; Molisani et al., 2013; Nriagu & Pacyna, 1988; Paula-Filho et al., 2015).

The dominance of anthropogenic N and P emissions over natural emissions has been reported for many coastal ecosystems, and increases in nutrient inputs are an important cause of the coastal eutrophication in a global scale (Claussen et al., 2009; Le Moal et al., 2019). The long-term maintenance of the supereutrophic state of the coastal lagoon can also be explained by the mass-balance budget, which indicates that the waterbody retained the N and P inputs, but that such retention loads represented a small fraction of the total nutrient emissions from natural and anthropogenic loads in the watershed.

Consequently, most of the N and P emissions were retained in soils that act as a buffer and temporary sink that slow down chemical transference to coastal zones (Reddy et al., 1999; Sébilo et al., 2013; Withers & Jarvie, 2008). The nutrient budget also indicated reduced N and P exportation from the lagoon to the coast and, consequently, high material retention within the lagoon based on its intermediate water residence times, estimated at 66 days (Campaneli & Molisani, 2019). For some coastal lagoons, eutrophication is linked to restricted exchanges with the sea and high residence times, where lagoons become more sensitive to nutrient enrichment and eutrophication (Mudge et al., 2008; Tett et al., 2003).

In the present study, the LULCC represented a small reduction of nutrient fluxes to the lagoon and thus contributing to maintenance of its supereutrophic state. The long-term prevalence of eutrophic conditions in other coastal lagoons has been observed in areas undergoing the continuous expansion of agriculture, urbanization and other economic activities (Campaneli & Molisani, 2019; Contreras-Espinosa et al., 1996; Rivera-Guzmán et al., 2014). Homeostatic mechanisms are based on control by trophic web chains, including high turnover in species composition and spatio-temporal variability of environmental conditions (Pérez-Ruzafa et al., 2019). Physical processes also maintain the limnological conditions of ecosystems, where higher lagoon water volume is reported relative to a small catchment area, which overcomes the drastic effects of nutrient input (Kalff, 2003), as expected for the Feia Lagoon. However, even in 'stable' coastal lagoons, primary production may be sensitive to small water quality alterations (e.g., turbidity, water transparency and nutrient enrichment from anthropogenic activities) and may suddenly develop dystrophic crisis (Koch, 2001; Pérez-Ruzafa et al., 2019; Rivera-Guzmán et al., 2014). For example, a coastal lagoon in Spain maintained water transparency and low nutrient and chlorophyll *a* level for two decades, despite the recent order of magnitude increase in nitrate concentration and LULCC. However, the lagoon suddenly displayed altered limnological conditions, increasing nutrient and chlorophyll contents in the water column and loss of transparency, diagnosed as an abrupt dystrophic crisis, which suddenly disappeared, leading to a rapid lagoon recovery (Pérez-Ruzafa et al., 2019).

An ecosystem may present gradual changes in its environmental conditions, resulting in small ecological effects until a threshold is reached and drastic changes are installed (Scheffer & Carpenter, 2003). Seasonal and spatial ecosystem heterogeneity increases resilience and reduce the influence of key environmental factors from which drastic shifts of ecosystem features are observed (van Nes & Scheffer, 2005). The lagoon in fact presented considerable spatial and short-term temporal variation in its limnological parameters, for example, the increasing nitrate concentrations, a wide range of dissolved oxygen (1.4 to 10 mg L^{-1}) and trophic state changes ranging from mesotrophic to hypertrophic. On the other hand, lagoon homogeneity was observed between the surface and bottom water column, with no chemical stratification. No stratification conditions are related to the shallowness of the lagoon, maximizing the effects of winds and storm events, as described for many other coastal lagoons (Kjerfve, 1994). Thus, the spatial and temporal heterogeneity of this lacustrine environment may contribute to its resilience to eutrophication, as stated by Scheffer and Carpenter (2003) and van Nes and Scheffer (2005). On the other hand, water column and inter-decadal N and P emission homogeneity, in association to the long water residence times of this restricted coastal lagoon and long-term accumulation of organic matter and nutrients, may contribute to decreased lagoon resilience, explaining its supereutrophic conditions. At one level, there is constant change, as the shift in the limnological conditions over time, at the same time that the whole intricate system is in balance.

Although, the water-related eutrophication index provides insights on the trophic state, sediment-related indicators on the aquatic lentic environments are mostly sensitive to changes in LULCC (Rodríguez-Gallego et al., 2017). The C/N ratio and isotopic composition from the lagoon sediments was similar to the results from an inland lagoon formed by a river tributary of the Feia Lagoon (Silva & Rezende, 2002). This study showed a ^{210}Pb-dated sediment core which covered a time period from 1848 to 1998 (Silva & Rezende, 2002). The older C/N ratio ranged from around 25 to 30 and suggested larger contribution of the allochthonous materials from the watershed, mainly vascular plants with C/N ratio higher than 20 (Cloern et al., 2002; Meyers, 2003). The δ^{13}C values found represent a mixture between C3 and C4 vegetation and together with C/N ratio, we observe the changes initiated between 1950 and 1960 attributed to the increasing deforestation of the watershed for sugarcane expansion, as described by Silva and Rezende (2002). From 1960, the C/N ratio decreases may be attributed to sugarcane crop intensification, in view of the fact that the sugarcane soil C/N ratio of 16 was lower than that of organic matter sources (grass = 49 and sugarcane = 61). For instance, soil organic matter δ^{13}C decreased from −25‰ to −20‰ as the sugarcane plantation aged from 12 to 50 years (Martinelli et al., 2002). In addition, the C/N ratio reduction at the Feia Lagoon may also reflect the increasing importance of autochthone organic matter sources, including phytoplankton biomass with high protein and low cellulose content, with a C/N ratio ranging from 4 to 10 (Meyers, 2003). The inflexion point at a depth of 6 cm can already reflect the replacement of part of the sugarcane crops mainly by pasture lands, agriculture and forest. Thus, the increasing C/N might represent an anew organic matter input from grass (C/N = 49) and vascular plants (>20) into the lagoon (Cloern et al., 2002; Martinelli et al., 2002; Meyers, 2003). Therefore, the inflexion point at a depth of 14 cm represents a change in the relationship between the surface run-off of soils with C4 organic matter content and the hydrodynamics of the lagoon, characterized by the dominance of the clay fraction for fine sand.

5 | CONCLUSION

This study concluded that LULCC, exemplified by the sugarcane crop partially replaced by permanent crops, livestock farming, forest and urbanization, slightly reduced the N and P loads to the coastal lagoon. However, the small reduction in the nutrient loads did not alter the long-term supereutrophic responses of the coastal lagoon, mainly to the anthropogenic N and P loads. Such ecological responses were also controlled by the soil accumulation of most natural and anthropogenic N and P emission loads, and the long-term nutrient retention within the lagoon, imposed by the its restricted connection to the sea, being the lagoon sediments considered a nutrient source to the water column, especially for total phosphorus. The sediment-related C/N and isotopic composition of the lagoon was sensitive to LULCC, indicating changes on the organic matter and nutrient inputs into the lagoon.

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CONFLICT OF INTEREST

The authors of the submitted manuscript declare no potential conflict of interest.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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