



Effects of recent urbanization on carbon and nitrogen burial rates of sedimentary records in a tropical coastal lagoon (Brazil)

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ABSTRACT

Land use and land cover changes (LULCC) are a global environmental issue that has impacted biogeochemical cycles worldwide. Sedimentary records can demonstrate the effects of LULCC on aquatic ecosystems, where the recent urbanization has been linked to changes in carbon and nitrogen burial. In this study, we reconstructed long-term LULCC and sedimentary records of carbon (C), nitrogen (N), phosphorus (P), and sediment burial rates in a eutrophic tropical coastal lagoon affected by recent urban expansion. Based on analyses of 30 years of satellite imagery and sedimentary records from 1932 to 2013, we revealed that urban expansion over low-productivity agricultural-pasture areas increased siltation and C, N, P concentrations and fluxes in the coastal lagoon. Large temporal variability of such parameters revealed not only the effects of LULCC on the lagoon's burial rates, but also the influence of artificial sand barrier openings, which connect the studied lagoon to the sea, reducing C, N, P, and particle deposition in the sediment. Our results support multi-proxy methods to assess the relationships between recent urbanization, rising C, N, and P burial rates, and the eutrophication process. We highlight that artificial sandbar openings, the current eutrophication management strategy for coastal lagoons, are ineffective in reducing the eutrophication state, even in the recent scenario of decreasing C, N, and P burial rates.

1. Introduction

Land use and land cover change (LULCC) has affected over 32% of global land area from 1960 to 2019, a scenario four times greater than previously estimated (Winkler et al., 2021). While LULCC is mainly attributed to deforestation and agricultural expansion in the tropics, in temperate regions it is associated with reforestation or afforestation, cropland intensification, and urbanization (Song et al., 2018). Tropical regions present the highest absolute deforestation rates in the world,

where tropical forests are converted to low-productivity cattle pasture, while agricultural demands continue to rise alongside urbanization.

As a result, LULCC may alter watershed hydrology, soil erosion, material transport, and the water quality of aquatic ecosystems. Kayitesi et al. (2022) examined the hydro-morphological consequences of LULCC in tropical regions, finding that it increased streamflow, surface flow, water and sediment yields, erosion rates, and channel migration while reducing evapotranspiration and groundwater recharge. Furthermore, natural vegetation replacement by urbanization, agriculture, and animal

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husbandry increases material transfer within watersheds, including organic matter, as a C source, and other nutrients mobilization. For example, in some urban and agricultural watersheds, anthropogenic sources may account for up to 82–90% of total nitrogen (N) loads emitted, outweighing natural emission processes such as soil loss and atmospheric deposition (De Paula Filho et al., 2015, 2019; Campanelli et al., 2021; Arueira et al., 2022).

C burial in depositional ecosystems, such as lakes and lagoons, is recognized as a key component of the terrestrial C cycle. However, data are scarce on the magnitude and temporal variability of C burial rates in areas where natural forests surround such depositional systems, as well as how such rates change over time in response to LULCC (Anderson et al., 2013; Mendonça et al., 2017; Amora-Nogueira et al., 2022). According to Amora-Nogueira et al. (2022), lake sediments influenced by tropical forests comprise a significant worldwide organic C sink, estimated at 7.4 Tg.C.yr⁻¹. Since 1900, global lake C burial rates have grown by approximately 72 Tg yr⁻¹, offsetting 20% of yearly CO₂ freshwater emissions (Anderson et al., 2020). Aside from the importance of lakes and lagoons in global C budgets, identifying shifting sources of organic C and N in sedimentary profiles is critical for understanding the consequences of LULCC in biogeochemical processes and negative effects, such as eutrophication and algal blooming events.

Tracking historical LULCC and its consequences on coastal ecosystems is especially important because coastal zones suffered the greatest urbanization expansion between the 1970s and 2000s in comparison to any other region in the world (Seto et al., 2011; Neumann et al., 2015; Rentschler et al., 2023). Human-induced pressure is anticipated to have greater impacts on coastal lagoons, because they serve as sinks for continental runoff, which is primarily influenced by human activities, and have the potential for long-term material accumulation and negative effects on water quality, deposition rates, and organism distribution (Kjerfve, 1994; Esteves et al., 2008; Rivera-Guzmán et al., 2014; Pérez-Ruzafa et al., 2019; Amora-Nogueira et al., 2023).

Long-term satellite imaging combined with geochemical traces in sedimentary records may provide pivotal information to track changes in C and N transport in these ecosystems (Meyers and Teranes, 2001; Davies-Vollum et al., 2019; Amora-Nogueira et al., 2023). LULCC is commonly assessed using satellite or airborne spatial imagery, which has become increasingly available over the last decades due to advancements in remote sensing technology and several satellite launches (Wulder et al., 2012; Yu et al., 2014; Xiao et al., 2019). This technique provides spatially explicit data on physical properties such as shape, texture, and color, which are used to classify different types of land use (Wang et al., 2022). Additionally, with the capability of satellites to revisit the same area, it is possible to compare land use types over different periods and quantify LULCC. As a result, remote sensing has been extensively used to track LULCC, understand their drivers, and assess their effects on several environmental processes, especially regarding aquatic ecosystems and watersheds (Li et al., 2008; Magalhães et al., 2022). For instance, remote sensing has been used to link urban expansion to increased runoff and extreme flooding (Lei and Zhu, 2018; Seidl et al., 2020), nutrient emissions intensification and eutrophication of lagoons (Rodríguez-Gallego et al., 2017; Obubu et al., 2022), and siltation caused by sediment transport (Ruiz-Luna and Berlanga-Robles, 2003).

We present a case study of the effects of LULCC represented by the recent urbanization and the long-term C and N changes registered on the sedimentary records of a coastal lagoon. The recent urbanization of this watershed started in the 1980s and intensified in the 2000s, induced by offshore oil exploration in the Campos Basin on the Brazilian coast which accounted for more than 80% of national oil production, being today the second most productive exploration marine oil basin (ANP, 2023). Most of the studied impacts from offshore oil exploration are usually focused on the marine environment and relate to drilling, oil spills, pollution, navigation, and marine fisheries, among others (Venegas-Li et al., 2019; Andrews et al., 2021). However, the

implementation of the offshore oil productive chain leads to onshore urbanization and inland environmental changes within the enterprise zone of influence (Esteves et al., 2008; Molisani et al., 2013; Lins-de-Barros, 2017; Beraldi et al., 2019; Andrews et al., 2021; Campanelli et al., 2021; García and Dias, 2023).

Thus, in a few decades, urbanization has replaced forest cover and other land uses, and as mentioned by the literature, the lagoon's sedimentary records can offer data about the variability and magnitude of natural and human land covers in C and N burial rates. Furthermore, the long-term dynamic in nutrients, chlorophyll *a*, and process of eutrophication has been characterized for this coastal lagoon, which can provide evidence for linking LULCC to C and N sources, burial rates, and eutrophication. The purpose of this study is to reconstruct long-term LULCC, C, N, P, and sediment burial rates in a tropical lagoon watershed under the effect of recent urbanization, which is a significant driver of LULCC on a global scale, particularly in developing nations.

2. Material and methods

2.1. Study area

The study region serves as the primary operational base for the Campos Basin offshore oil productive chain in Macaé, on the South-eastern Brazilian coast. The population in the municipality has expanded from 40,000 in 1940 to more than 260,000 in 2024 (IBGE, 2024). Other surrounding cities have also seen their populations grow over the last 40 years, primarily due to the oil and gas chain, in an unplanned urbanization process. This resulted in a myriad of inland environmental impacts, such as irregular sewage disposal, chemical contamination, eutrophication, siltation, and biodiversity loss (Molisani et al., 2013; Lins-de-Barros, 2017; Farias et al., 2019; Beraldi et al., 2019).

Among these impacted ecosystems, the Imboassica lagoon watershed stands out as an example where LULCC has led to biogeochemical changes (Palma-Silva et al., 2002; Bozelli et al., 2009; Beraldi et al., 2019). The Imboassica lagoon, like other coastal lagoons was formed by the natural impoundment of a small contributing river during the last Holocene transgression episode, when both river and marine sediment inputs built a sand barrier between the river and the sea (Kjerfve, 1994). The lagoon is an orthogonally-oriented choked brackish water lagoon (Kjerfve, 1994; Melo et al., 2007), with an average depth of 1.43 ± 0.51 m, a limnetic surface of approximately 2.71 km² and a water volume of 3.55 × 10⁶ m³ (Melo et al., 2007). Its watershed (Fig. 1) is 59.99 km², with altitudes ranging from -9m to 169m asl. Freshwater inputs from first to third-order rivers average 1.21 m³ s⁻¹. The watershed's geology is mostly represented by the Paleoproterozoic "Região dos Lagos" complex, which is composed of biotite gneisses and orthogneisses. Fine to coarse alluvial sand Cenozoic deposits and holocene/pleistocenian marine and aeolian sandy deposits adjacent to the coastline are also found on the watershed (Almeida and Silva, 2012). According to the Köppen-Geiger climate classification, the area has a tropical climate with a dry winter (Kotteck et al., 2006). Annual rainfall averages 1202 mm varying from 31 to 196 mm between the driest (April–October) and wetter (November–March) months. Previous studies indicated that the Imboassica watershed LULC was mainly attributed to pasture lands, forests, and urbanization (Barreto, 2009). The watershed is home to approximately 10,000 people and numerous oil exploration operation facilities are placed in the area. Urbanization has expanded over the past few decades, leading to increased raw sewage emissions into the lagoon. This has caused a rise in the lagoon's trophic state and a shift from a clear-water, Charophyte-dominated mesotrophic state to a turbid-water, phytoplankton-dominated hypertrophic state, with more frequent cyanobacterial blooms (Farias et al., 2019; Palma-Silva et al., 2002).

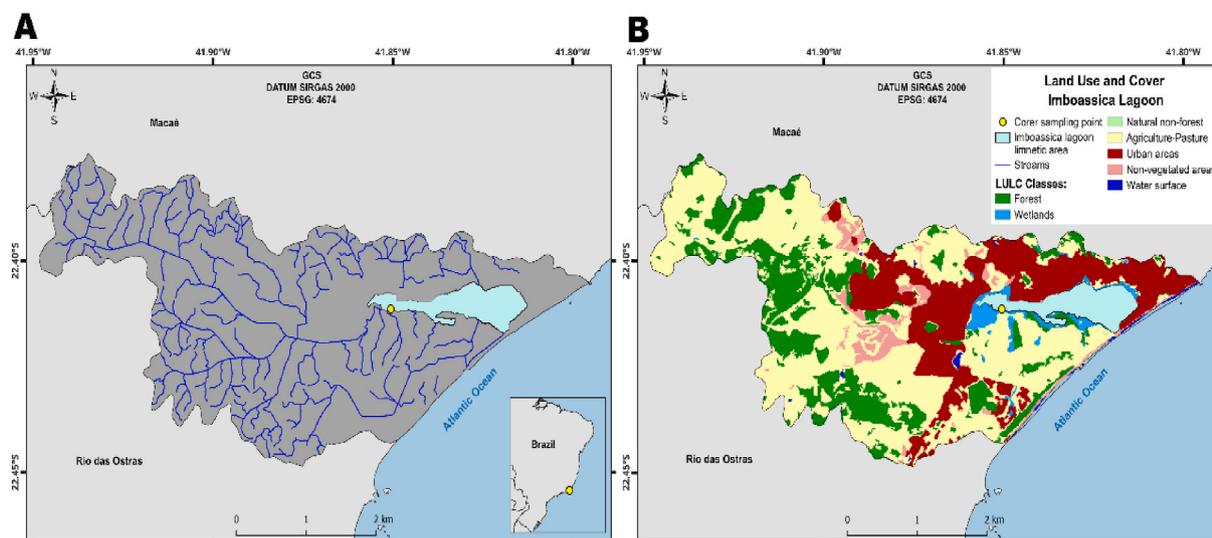


Fig. 1. Map of the study area, including the lagoon's watershed delimitation (dark gray area), its hydrography (dark blue), the Imboassica lagoon's limnetic surface (light blue), the sediment core sampling location (yellow circle) (A), and the watershed's land use and cover classification for the year of 2021 (B).

2.2. Land use and land cover analysis

To identify LULCC in the Imboassica watershed, we analyzed raster images made available by the MapBiomias Project. This project is a multidisciplinary initiative that monitors annual land use changes across the Brazilian territory using high-resolution, temporally comprehensive data (Souza et al., 2020). The methodology employed by MapBiomias involves analyzing Landsat images with a 30m resolution on the Google Earth Engine platform. The LULC raster products generated by MapBiomias are annual mosaics, created from multiple raster images captured during periods of expected low cloud coverage. For the Atlantic Biome, this period spans from April to September. The original Landsat images are first processed by cloud masking and removal. Then, spectral indexes, reflectance bands, and spectral mixture modeling-derived variables are used for land use and cover classification using a *Random Forest* algorithm available in Google Earth Engine, after 100 iterations. Training samples are drawn from areas where land use has not changed over the 38 years of data collection. Further details on image acquisition, processing, and land cover classification are available on the project's website (MapBiomias, 2022).

For our analysis, we used Level 3 raster images from MapBiomias Collection 7.0 and reclassified the themes into seven main categories of interest: Natural Forest, Wetlands, Other Natural Non-Forest Areas, Agriculture-Pasture, Urban Areas, Other Non-Vegetated Areas, and Water Surface. The MapBiomias Collection 7.0 Level 2 classification had a global accuracy of 84.4% (MapBiomias, 2024). We analyzed images acquired annually from 1985 to 2015 and quantified the area of each LULC class yearly after polygonization. All spatial analyses were conducted under the Albers Equal Area Conic Projection, as recommended for area calculation in the Brazilian territory (IBGE, 2019). All geospatial analyses were performed using the software QGIS v. 3.28.8.

2.3. Sediment core collection and laboratory analyses

In 2015, one sediment core was collected from the western portion of the Imboassica Lagoon (22° 24' 41" S; 41° 51' 02" W) (Fig. 1). The sediment core was carefully extruded and sectioned at 1 cm intervals, and each sample was homogenized and placed in previously sterilized containers. In the laboratory, the apparent density ($\text{g} \cdot \text{cm}^{-3}$) was estimated by taking the volume of each sediment layer and a constant dry weight. The granulometry of the dried samples was determined using a laser diffraction particle analyzer (Shimadzu model SALD3101).

Elemental (%C and %N) and stable isotope compositions ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) were determined in sediments using an Isotopic Mass Spectrometer Thermo Quest-Finnigan Delta (Finnigan-MAT), attached to an Elemental Flash 2000 analyzer (Thermo Scientific). Pee Dee Belemnite carbonate and atmospheric nitrogen were used as standard values and analytical precision was $\pm 0.1\%$ for $\delta^{13}\text{C}$ and $\pm 0.2\%$ for $\delta^{15}\text{N}$. The C and N quantification was performed using analytical curves from Buffalo River standards (SRM 2704, National Institute Standards), with an inter-replicate precision of 95%. Accuracy was verified using the Standard OAS/isotope—Low Organic Soil (elemental microanalysis), with a 94% recovery. Total phosphorus (TP) concentrations were determined by spectrophotometry using the molybdenum blue complex method in a Varian 50 Bio UV-visible spectrophotometer model (Golterman et al., 1978) after 16 h of extraction in 1N HCl, subsequent filtering in 8 μm mesh filters, and 10-fold dilution of the extracted solution (Aspila et al., 1976).

The sediment's chronology was determined through the sediment accumulation rates by measuring the natural radioisotope ^{210}Pb . ^{210}Pb has a half-life of 22.3 years and yields accumulation rates for approximately the past 100 years (Smoak and Patchineelam, 1999). Sediments were oven-dried (50 °C), disaggregated, and homogenized before analyses and their water content was determined as the weight loss after drying. Sediment sub-samples were sealed and stored for at least 21 days to allow the establishment of secular equilibrium between ^{226}Ra and its granddaughters. Samples were counted for at least 24 h to determine ^{226}Ra and ^{210}Pb activities by gamma-ray spectrometry, using a Canberra HPGe (High Purity Germanium Detector) spectrometer with an efficiency of 40%, housed in a lead shield and coupled to a multichannel analyzer installed at the Laboratório de Radioisótopos Aplicados ao Meio Ambiente of Fluminense Federal University. The ^{214}Pb and ^{214}Bi , ^{226}Ra activities were determined by a weighted average of two ^{214}Pb energies (295.2 and 351.9 keV) and a ^{214}Bi gamma peak at 609.3 keV, while ^{210}Pb activities were determined by the direct measurement of 46.5 keV gamma peak.

Sediment accumulation rate (SAR) was calculated for each sample according to equation (1) and expressed in $\text{g} \cdot \text{cm}^{-2} \cdot \text{yr}^{-1}$:

$$\text{SAR}_{(x)} = d_{(x)} \cdot \omega_{(x)} \quad (1)$$

Where, $d_{(x)}$ represents the density ($\text{g} \cdot \text{cm}^{-3}$) of sediment sample x , $\omega_{(x)}$ is the sedimentation rate ($\text{cm} \cdot \text{yr}^{-1}$) of sediment sample x , given by the distance between samples x and $x-1$ divided by x and $x-1$ samples respective dating.

Burials of TOC, TN, and TP were calculated according to equation (2) and expressed in $\text{mg}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$:

$$T = \omega \cdot \rho_s \cdot (1 - \emptyset) \cdot C \quad (2)$$

where: ω is the sedimentation rate ($\text{cm}\cdot\text{yr}^{-1}$); ρ_s is the mean density ($\text{g}\cdot\text{cm}^{-3}$); \emptyset is the porosity and C is the concentrations of each element in each sediment core layer ($\text{mg}\cdot\text{g}^{-1}$) (Bernier, 1980).

2.4. Statistical analysis

The LULC and sediment profile data was grouped in three periods, according to each sample's respective age, following the recent history of the watershed land use and cover. Group I encompassed a period before the oil economy establishment in the region (1932–1970). Group II comprised the period of establishment of the oil and gas industry in the watershed (1970–1996). Group III (1997–2013) represented the expansion of the oil economy in Brazil and the consequent urban expansion in the area. Thus, each year's sample was considered a replicate within each group. For LULC analysis, we only compared Group II (1985–1996) and Group III (1997–2015) because satellite data was only available for the period after 1985.

We applied univariate analyses of variance (One-Way ANOVA) to test for differences between group means. We tested statistical assumptions of normality and homoscedasticity by applying Shapiro-Wilk's normality test and Levene's homoscedasticity test, respectively. When data did not meet assumptions even after log transformation, we applied the Kruskal-Wallis test. We also tested the correlation between variables using Pearson's linear correlation. In addition, we applied Principal Component Analysis (PCA) to group each year in a multidimensional space in the function of the sediment data, after eliminating autocorrelated variables. All statistical analyses were performed at a confidence interval of 95% in R Studio software v. 4.3.0.

3. Results

3.1. Long-term LULCC in the lagoon's watershed

Satellite images indicate that the urban area within this watershed increased 5.6 times, accounting for 3.7% of the watershed area in 1985 and 20.7% in 2015 (Table 1; Fig. 2; SM1 on supplementary material). Natural forests covered 11% of the Imboassica watershed in 1985 and increased by 4.3% between 1985 and 2015. Urbanization expanded primarily towards pastoral and agricultural areas, which comprised 76.8% of the watershed in 1985 and lost 29.7% of its former area by 2015. During the studied period, wetlands, which primarily include cattail (*Typha domingensis* Pers.) mats in our study, expanded in area by 123%, while the lagoon's water surface decreased by 12%. There were also slight increases in other non-vegetated regions (2.9-fold) and natural non-forested areas (1.6-fold) in the watershed, but they accounted for a small share of the basin (less than 2.6%).

Urban areas in the lagoon's watershed were significantly smaller in

Table 1
LULC classes' area in km^2 (sd) for 1985–1996 (Group II) and 1997–2015 (Group III) (n = number of samples).

LULCC/Group	1985–1996/Group II (n = 12)			1997–2015/Group III (n = 19)		
	Mean	Min	Max	Mean	Min	Max
Urban areas	3.1 (0.44)	2.2	3.6	7.6 (3.1)	3.6	13
Agriculture-pasture	46 (0.32)	46	47	41 (4.1)	32	46
Natural forest	5.7 (0.44)	5.3	6.7	6.7 (1.0)	5.8	9.3
Wetland	1.2 (0.11)	1.1	1.5	1.4 (0.1)	1.3	1.6
Natural non-forest	0.04 (0.01)	0.03	0.06	0.05 (0)	0.04	0.06
Non-vegetated areas	0.50 (0.02)	0.43	0.5	0.6 (0.27)	0.43	1.5
Water surface	3.4 (0.07)	3.2	3.4	3.1 (0.13)	2.9	3.4

Group II, i.e. the 1985–1996 period (mean = 3.1 km^2 ; sd = 0.44; One-Way ANOVA; df = 1; F value = 43.7; $p < 0.05$) in comparison with Group III, the period between 1997 and 2015 (mean = 7.6 km^2 ; sd = 3.1) as in Table 1. This effectively indicates urban areas' expansion through time. Meanwhile, agricultural-pasture lands reduced significantly (Kruskal-Wallis test; $X^2_{(1)} = 21.375$; $p < 0.05$) from 46.4 km^2 (sd = 0.32) in Group II to 41 km^2 (sd = 4.13) in Group III during the same period.

We observed a significantly negative correlation between agriculture-pasture lands and urban areas (Adj. $R^2 = 0.97$; $p < 0.05$; $F = 1108$; df = 29; Fig. 3A). Urban area increase was constant between 1985 and 2015 in the watershed, and our data suggest that urbanization spread toward pasture lands conversion (SM2). We also observed a decline in water surface area significantly correlated to wetlands area increase (Adj. $R^2 = 0.55$; $p < 0.05$; $F = 38.41$; df = 29; Fig. 3B). This likely represents a siltation process related to urban expansion in the Imboassica watershed. Water surface area was significantly smaller for Group III (mean = 3.1 km^2 ; sd = 0.07; One Way ANOVA; df = 1; F value = 45.17; $p < 0.05$) in comparison with Group II (mean = 3.4 km^2 ; sd = 0.07). The opposite was found for wetland areas. Group III wetland area was significantly greater (mean = 1.2 km^2 ; sd = 0.11; Kruskal-Wallis; $X^2_{(1)} = 12.45$; $p < 0.05$) than observed for Group II (mean = 1.4; sd = 0.1).

3.2. Long-term changes in sedimentary records

The vertical sediment profile sampled in 2015 encompassed a period spanning over eight decades, from 1932 to 2013. The average sedimentation rate was 0.44 cm/year , with predominantly fine sediments, mostly represented by silt, corresponding to $67.69\% \pm 11.16\%$ of the total sediment sample mass. Sandy clasts corresponded, on average, to $9.64\% \pm 15.57\%$ of sample mass, while clay sediments corresponded to $22.66\% \pm 9.53\%$, although two events of pronounced granulometric changes were observed in the sediment profile (Fig. 4). The Sediment Accumulation Rate (SAR) varied from $0.04 \text{ g cm}^{-2}\cdot\text{yr}^{-1}$ to $0.21 \text{ g cm}^{-2}\cdot\text{yr}^{-1}$ among sediment layers, with an average SAR of $0.12 \text{ g cm}^{-2}\cdot\text{yr}^{-1} \pm 0.04$. SAR exhibited an increasing trend starting from the 1990s, followed by a reduction trend between 2006 and 2013.

SAR was similar between Group I (1932–1970) and Group II (1973–1996), with means equal to 0.12 ± 0.04 and 0.09 ± 0.02 , respectively. On the other hand, SAR increased significantly in Group III (1997–2013), when urbanization increased in the watershed, with an average of $0.16 \pm 0.03 \text{ g cm}^{-2}\cdot\text{yr}^{-1}$. These differences were supported by analysis of variance (One-Way ANOVA; df = 2; F value = 10.0; $p < 0.05$), followed by Dunn's post-hoc test, which indicated significant differences between groups I and III and II and III, but not between groups I and II. Although we observed a clear increase in the median clay content in the lagoon's sediment from 1932 to 2013 (from 19.2% to 32.3%), we found no significant differences in clay content between groups. We observed a similar opposite pattern for sand content. Although no significant differences were observed in sand content between groups, we found consistently decreasing medians between groups I (12.4%), II (1.2%), and III (0.1%). Silt median values presented a hump-shaped behavior, increasing from Group I (67.8%) to Group II (76.5%) and decreasing in Group III (62.3%). Some sediment layers showed drastic changes in grain size likely due to water mass movements, such as the higher sandy content and lower fine particles in the 1954 layer. This is corroborated by the negative correlation observed between sand and clay contents (linear model; df = 34; Adj. $R^2 = -0.58$; $p < 0.05$; SM3).

The average TOC concentration (Fig. 5) increased significantly from Group I (mean = $25.5 \text{ mg g}^{-1} \pm 10.4$) to Group II (mean = $43.9 \text{ mg g}^{-1} \pm 10.7$) but thereafter declined significantly in Group III ($20 \text{ mg g}^{-1} \pm 8.2$; One-Way ANOVA, df = 2, F = 16.06; $p < 0.05$; SM4). TN and TP concentrations increased consistently from Group I to Group III, with significant increases between groups II and III for TN (One-Way ANOVA,

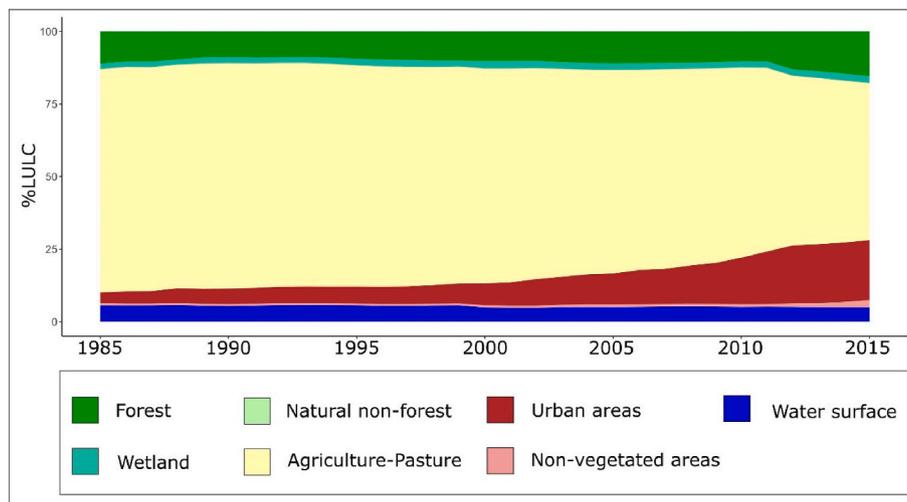


Fig. 2. Land use and land cover (%) of the Imboassica lagoon's watershed between 1985 and 2015.

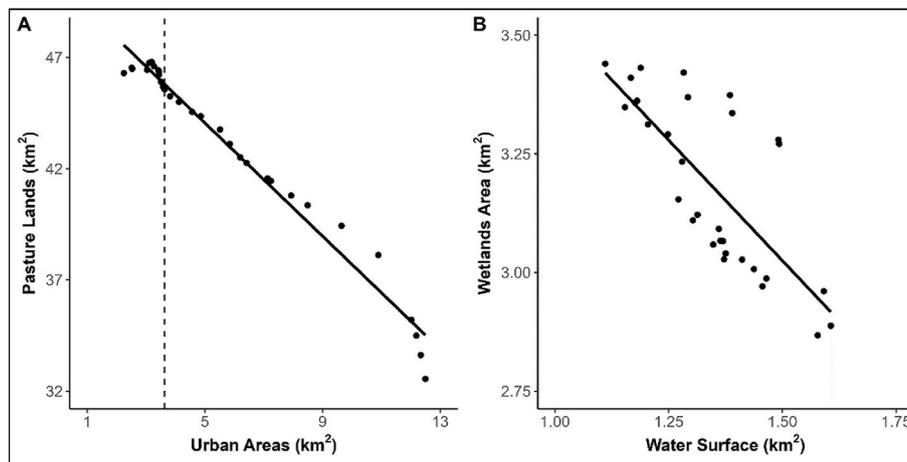


Fig. 3. Linear regression between annual pastoral and urban areas (A); and between wetlands and water surface areas (B). The vertical dashed line in A indicates the year 1997 when the Brazilian state broke the monopoly for oil exploration in Brazil.

df = 2, $F = 19.56$; $p < 0.05$) and TP (One-Way ANOVA, df = 2, $F = 35.84$; $p < 0.05$). TOC burial was similar between groups, with no significant differences between groups found. On the other hand, temporal increases were observed for TN fluxes, which increased significantly from Group II (mean = $1.1 \text{ g m}^{-2} \cdot \text{yr}^{-1} \pm 0.46$) to Group III (mean = $2.2 \text{ g m}^{-2} \cdot \text{yr}^{-1} \pm 0.85$; One-Way ANOVA; df = 2, F value = 19.56, $p < 0.05$). The same was observed for TP fluxes, which increased significantly from Group II (mean = $61.4 \text{ mg m}^{-2} \cdot \text{yr}^{-1} \pm 25.2$) to Group III (mean = $267.8 \text{ mg m}^{-2} \cdot \text{yr}^{-1} \pm 133.8$; One-Way ANOVA; df = 2; F value = 35.84; $p < 0.05$). For these variables, there were no significant differences between groups I and II.

The average C:N decreased significantly from 39.8 ± 6.5 in Group I to 17.5 ± 7 in Group III (One-Way ANOVA, df = 2, F value = 36.7; $p < 0.05$), with significant differences only observed between Group III and the other groups. $\delta^{13}\text{C}$ readings ranged from -29.0 to -24.6‰ and showed a significant increase from the late 1990s, followed by a decrease in the second half of the 2000s decade. Analysis of variance found significant differences in $\delta^{13}\text{C}$ isotopic values (Kruskal-Wallis test; $X_{(2)} = 12.95$, $p < 0.05$) between Group III (mean = $-25.8 \pm 1.2\text{‰}$) and Groups I (mean = $-27.5 \pm 0.3\text{‰}$) and II (mean = $-27.8 \pm 0.2\text{‰}$). $\delta^{15}\text{N}$ isotopic values in the sediment profile ranged from 2.0 to 4.4‰, with a pronounced peak in 1943 and a consistent increase from 2005 up to the end of the record. We found significant differences in $\delta^{15}\text{N}$ isotopic signature only between Group I and III (One-Way ANOVA; df = 2; F

value = 4.3; $p < 0.05$; followed by Dunn's post hoc test).

A Principal Component Analysis (PCA) grouped recent samples (1997–2013) in function of greater $\delta^{13}\text{C}$, $\delta^{15}\text{N}$ and TP fluxes and low C:N (Fig. 6). Meanwhile, Group II (1970–1996) and Group I (1932–1970) were associated with increased TOC concentrations and fluxes, as well as greater C:N, although Group I was less associated to TOC fluxes than Group II. The first Principal Component of the PCA accounted for 48.4% of the data variability, whereas the second Principal Component explained 22.5% of it.

4. Discussion

Our results show long-term LULCC associated with forest loss converted in pastoral areas and recent urbanization that led to a great change in nutrient dynamics and deposition in the Imboassica lagoon. This concurs with global trends of LULCC in tropical regions, where natural vegetation had been previously converted to low-productivity agriculture-pasture fields and has been recently converted to urban areas (Lapola et al., 2014; Achard et al., 2014).

Simultaneously with urbanization expansion in the watershed, the Imboassica lagoon's water surface has decreased by 12% in 30 years, leading to marginal wetlands expansion by 123%, where monospecific cattail mats have been consistently spreading. Concomitant urban expansion and increases in the SAR and clay content registered in the

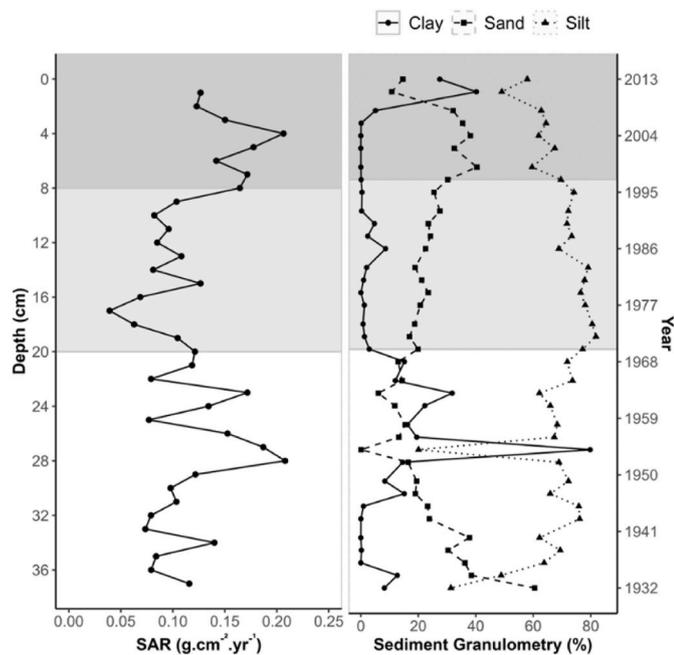


Fig. 4. Sediment accumulation rate (SAR) and granulometric profiles dated by ^{210}Pb in the sediment core of the Imboassica lagoon. Black circles with continuous lines represent clay content; black squares with dashed lines represent sandy clasts; and black triangles with dotted lines represent silt sediments.

sediment core suggest that the Imboassica lagoon is under a LULCC-related process of siltation, indicated by water-surface reduction and cattail wetlands expansion. LULCC affects tropical watersheds by increasing streamflow, runoff, sediment production, and erosion rates (Kayitesi et al., 2022). A review of sediment transport in agricultural and urban watersheds found that urbanization expansion leads to an increase in median annual sediment yield by at least three times in comparison to background levels (Russell et al., 2017). Widespread impervious surface cover increases runoff, particularly during brief periods of heavy rainfall, and affects erodible downstream areas (Ferreira et al., 2020). In coastal watersheds, sediments are leached towards coastal ecosystems, such as coastal lagoons, where continental-coastal sediment exportation is modulated and these systems consequently become sediment sinks and are threatened by siltation processes (Kjerfve and Magill, 1989; Avramidis et al., 2008; Duck and Da Silva, 2012). Although agricultural lands yield between 88% and 62% less sediments than urbanized lands (Russell et al., 2017), they exacerbate soil erosion because natural vegetation suppression and tilling practices expose bare soils and contribute to enhancing load transmissions (Stenfert Kroese et al., 2020).

The SAR in the Imboassica lagoon was lower than in other anthropized tropical coastal lagoons (Cerdeira et al., 2016; Amora-Nogueira et al., 2023). Nonetheless, early interventions on land cover before 1932 converted natural forests to pastoral lands and are believed to have increased sediment mobilization, transport, and deposition into the lagoon. Early deforestation and river channeling in watersheds have been linked to increases in sediment loading into other coastal lagoons resulting in early eutrophication and environmental change (Freitas et al., 2002; Dinis et al., 2006; Katsuki et al., 2019; Ruiz-Fernández et al., 2020).

Large runoff and sediment transport related to LULCC can also lead to increased nutrient burdens across watersheds (Old et al., 2006; MacKenzie et al., 2022). TOC, TN, and TP burial increased in the sediment core following urbanization in the studied site. The PCA plot distinguished the 1997–2013 period from previous periods since the intensification of urbanization was linked to TN, TP burial, and SAR. The

TOC, TN, and TP levels in Imboassica lagoon were comparable to moderately contaminated tropical coastal lagoons (Amora-Nogueira et al., 2023), but lower than to other contaminated lagoons (Cerdeira et al., 2016).

The long-term dynamics of nutrients, chlorophyll, and the eutrophication process of the Imboassica lagoon have been monitored since 1992 (Farias et al., 2019). The lagoon's limnology was described in terms of distinct phases. Before urbanization, the lagoon was in a stable state of clear water with submerged Charophytes occupying most of the benthic compartment, although inorganic turbidity could be observed after rainfall events (Palma-Silva et al., 2004; Farias et al., 2019). Between 1992 and 2016, there was a significant rise in mean TN (0.63 ± 0.19 to $3.0 \pm 0.98 \text{ mg.L}^{-1}$), TP (0.04 ± 0.02 to $0.18 \pm 0.18 \text{ mg.L}^{-1}$), and chlorophyll a (9.4 ± 7.4 to $122 \pm 39 \mu\text{g.L}^{-1}$) in the pelagic compartment of the Imboassica lagoon. During this period, especially after May/June 1999, the trophic state increased from mesotrophic to hypertrophic, and turbid waters dominated by phytoplankton, with marginal *Typha domingensis* wetlands covering 15% of the lagoon area (Palma-Silva et al., 2002; Farias et al., 2019). Dredging such banks to minimize eutrophication revealed increases in N and P concentrations in the water column, suggesting that dredging and bank removal would worsen the eutrophication process of the lagoon (Marotta et al., 2009).

A similar pattern was observed in a coastal lagoon (Spain) over the previous 22 years, in the context of eutrophication caused by an increase in nutrient inputs from human activities (Pérez-Ruzafa et al., 2019). During 22 years of monitoring, nutrient concentrations in the water column increased, but the lagoon remained homeostatic, with clear water, low chlorophyll levels, and the presence of submerged macrophytes. However, an abrupt increase in nutrient concentrations caused a shift to higher phytoplanktonic primary productivity, lowered water transparency, and the loss of submerged macrophytes. Despite nutrient inputs, the lagoon's resilience was supported by top-down regulation and a rapid turnover in species composition, related to the limited connectivity between the coastal lagoon to the sea. This trophic evolution sequence was similar to that observed in the Imboassica lagoon and other ecosystems alike (Amora-Nogueira et al., 2023; Campanelli et al., 2021). Nonetheless, while the Spanish lagoon was resilient to eutrophication due to a limited connection to the sea and top-down biological control (Pérez-Ruzafa et al., 2019), the Imboassica lagoon presents sporadic and usually human-induced sandbar opening events that connect it to the sea. This could be related to the lower resilience to eutrophication of the Imboassica lagoon.

Identifying shifting sources of organic C and N in sedimentary profiles is critical for understanding the consequences of LULCC on the eutrophication of the lagoon. The decrease in C:N from 34 (1932–1970) to 15 (1997–2013) revealed that as urbanization expanded, organic matter input from allochthonous vegetation was replaced by autochthonous phytoplankton-derived organic matter, an indicative of sewage-driven eutrophication. The C:N ratio has been widely used to track sources of sediment organic matter (Müller and Mathesius, 1999; Kausshal and Binford, 1999; Meyers, 2003), because terrestrial plant-derived organic matter presents higher C:N (≥ 20) due to relatively low protein (N) content and abundant cellulose (C), while phytoplanktonic-derived organic matter presents low C:N ($\sim 4\text{--}12$) because of high protein contents and absence of cellulose (Meyers and Ishiwatari, 1993; Meyers, 1994; Filley et al., 2001; Das et al., 2008). The C:N ratios suggest that after 1995, the sediment organic matter origin shifted to phytoplanktonic-derived, which must be linked to raw sewage disposal, nutrient over-enrichment, and henceforth higher trophic levels (Palma-Silva et al., 2002; Farias et al., 2019).

Moreover, the effects of increased urbanization and raw sewage emissions on the lagoon's phytoplanktonic productivity were also registered by the $\delta^{13}\text{C}$ measurements. Because the lighter ^{12}C isotope is more prone to mobility from the dissolved inorganic carbon (DIC) pool, as phytoplanktonic productivity grows, the ecosystem DIC bulk becomes enriched with ^{13}C isotopes, which are then recorded on the sediment

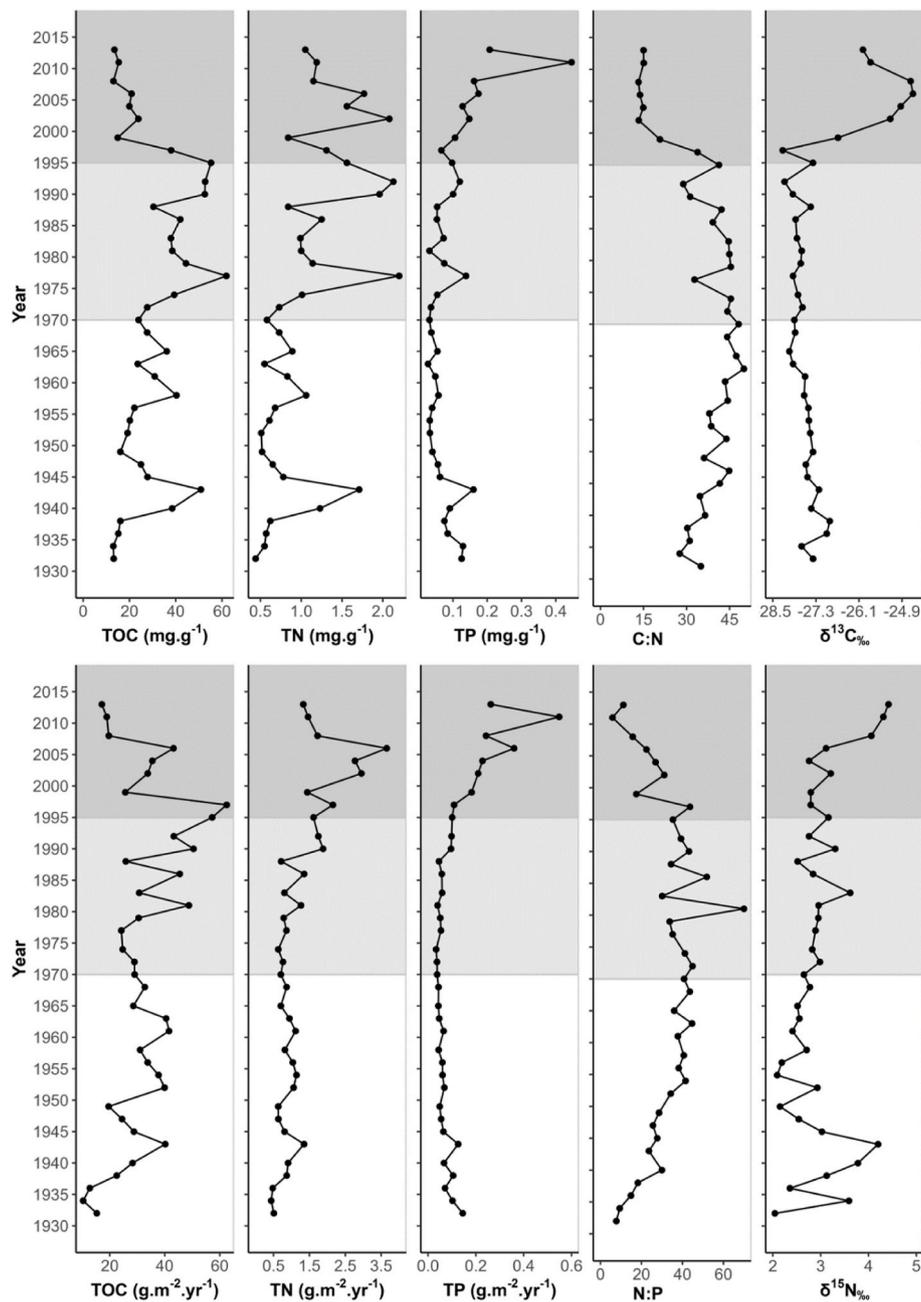


Fig. 5. Long-term C, N, P concentrations, fluxes, and isotopic composition in the sediment profile from the Imboassica Lagoon.

after precipitation (Schelske and Hodeli, 1991). Nutrient enrichment in aquatic environments increases ecosystem productivity and DIC fixation rates, resulting in higher $\delta^{13}\text{C}$ isotopic values (Schelske and Hodell, 1995). The Imboassica lagoon's $\delta^{13}\text{C}$ isotopic enrichment from a range between -27.0 and -28.5‰ (1932–1995) to -24.9‰ after 1995 registers an increase in phytoplanktonic primary productivity.

Although $\delta^{13}\text{C}$ may also indicate a shift in organic matter origin, during the whole study period, the long-term $\delta^{13}\text{C}$ isotope values ranged from -28.22‰ to -24.6‰ , which are consistent with both C3 terrestrial plants and lacustrine algal-derived organic matter (Meiers and Ishiwatari, 1993). Despite the presence of pasture grasslands throughout the watershed, historical measurements of C4-derived organic matter from grass range from -11.0‰ to -15.0‰ (Roeske and O'Leary, 1984; Meiers, 1994) and are significantly higher than the $\delta^{13}\text{C}$ observed in the Imboassica lagoon. On the other hand, the *Typha domingensis*, an emergent C3 macrophyte that develops dense monospecific mats in the

Imboassica lagoon's margins, presents an average $\delta^{13}\text{C}$ signature of -29.5‰ (Santos Fonseca et al., 2015) and could be contributing to the bulk organic matter, particularly in marginal areas. The limnetic particulate organic carbon (POC) in a comparable lagoon close to our study site is formed by the organic matter decomposition of littoral emergent macrophytes, which when settled becomes part of the sediment (Marinho et al., 2010).

The recent increase in $\delta^{15}\text{N}$ in the sediment core could be related to raw home sewage discharges from the lagoon's expanding population. The highly urbanized Rodrigo de Freitas lagoon showed an upward $\delta^{15}\text{N}$ trend over time, suggesting an increase in sanitary sewage releases (Vezone et al., 2021). Sediments with $\delta^{15}\text{N}$ enrichment ranging from 3.0 to 9.0‰ were connected to higher untreated sewage discharges and higher human population density (Vizzini and Mazzola, 2006; Cerda et al., 2016). Thus, the increasing urbanization in the Imboassica lagoon, for example, towards rural regions of the watershed, encourages the use

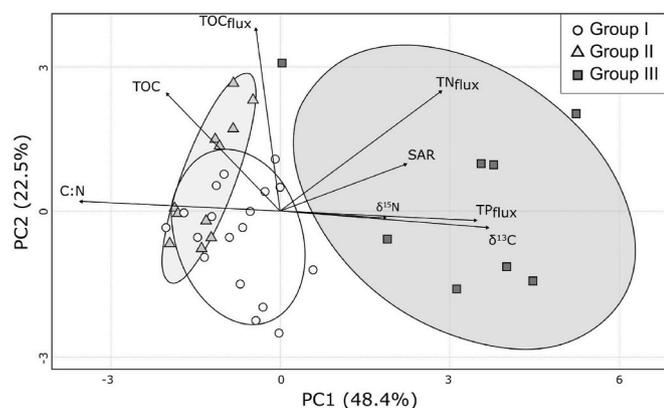


Fig. 6. PCA plot representing the grouping of the different characteristics of the three groups in the sediment core.

of technologies to limit the impact of raw sewage in the eutrophication of the lagoon (Gautam et al., 2021; Perumpully et al., 2023). Similarly to $\delta^{13}\text{C}$, $\delta^{15}\text{N}$ can be used to monitor ecosystem eutrophication. This is related to elevated $\delta^{15}\text{N}$ isotope values due to phytoplanktonic and bacterial ^{14}N utilization preferences (Altabet and Francois, 1994).

Average nutrient concentration and burial rates increased across the three analyzed period groups, although significant temporal variability, such as rapid decreases in SAR, TOC, TN, and TP were observed. This could be linked to sandbar breaching events, temporally connecting the lagoon and the Atlantic Ocean. Sandbar breaching events may occur naturally during high precipitation periods. However, human interventions have intensified the frequency of sandbar openings to increase marine fish and shrimp larvae recruitment and improve fisheries stocks and production in lagoonal systems (Trnski, 2002; Able et al., 2017). Sandbar breaching is also a cultural practice for the fisherman in the studied area, as mentioned by August de Saint-Hilaire, a French naturalist who visited the region during an expedition in the 19th century and described a sandbar opening event in the Imboassica lagoon and related it to fishing purposes (Saint-Hilaire, 1833). Other interests related to artificial sandbar openings include flood control and water quality improvement, particularly in developing territories, such as the studied site. Unplanned urbanization and irregular sewage emissions lead to periodic clogging of households within lagoonal flooding areas and water quality impairment, evoking drastic management practices to tackle flooding events and impede human losses (Suzuki et al., 1998; Sahu et al., 2014; Prestrelo and Monteiro-Neto, 2016; Davies-Vollum et al., 2019). Between 1992 and 2017, the lagoon's sandbar was artificially opened 22 times, primarily to decrease the water volume and control flooding risks (Farias et al., 2019).

Even with increased sewage intake and a higher average of the measured sedimentary parameters from 1997 to 2013, the lagoon had a decline in several parameters due to the increased frequency of sand barrier opening during the urbanization peak. Other regional lagoons are also subject to this intervention since short-lived flushing of lagoon water reduces TOC, TN, and TP burial; but soon after the lagoon closes, such nutrient burial increases and sometimes exceeds earlier levels. Thus, long-term increases in TOC and TN burial continued even with transitory reducing peaks associated with the lagoon opening, indicating that such intervention did not represent the projected alleviation of eutrophic conditions in this lagoon, as sewage is still disposed of in the lagoon (Amora-Nogueira et al., 2023). Suzuki et al. (1998) monitored several physical, chemical, and biological water quality variables in a sewage-driven eutrophic coastal lagoon, and the relationship to ephemeral sand barrier breaks revealed an enrichment of dissolved inorganic nutrients during the first two to eight days, followed by a period when seawater's physical and chemical characteristics prevailed. The lagoon reverted to pre-opening water conditions within 10–20 days,

with significant levels of eutrophication, chlorophyll *a*, TN, and TP.

Our multi-proxy approach showed significant relationships between LULCC and biogeochemical processes during the last decades in the Imboassica lagoon's watershed. The satellite imagery quantified the LULCC with a comprehensive spatial and temporal resolution. The elemental and isotopic proxies were accurate (approximately 95% and 94%, respectively), validating the estimated burial rates. The predicted C burial rates range between 20 and 60 $\text{g m}^{-2}\text{.yr}^{-1}$, comparable to natural and human-altered lake organic C burial rates globally (36.7–114 $\text{g m}^{-2}\text{.yr}^{-1}$) (Amora-Nogueira et al., 2022). The burial rates of C, N, and P were likewise similar to another coastal lagoon located in the same morphological unit of the Serra do Mar mountains (TOC: 12–136 $\text{g m}^{-2}\text{.yr}^{-1}$; TN: 1.24–17 $\text{g m}^{-2}\text{.yr}^{-1}$; TP: 0.08–1.03 $\text{g m}^{-2}\text{.yr}^{-1}$) (Amora-Nogueira et al., 2023).

5. Conclusion

Satellite images and sedimentary records collected over the last decades have supported the reconstruction of the watershed's LULCC and biogeochemical changes in the coastal lagoon. The LULCC caused by deforestation, low-productivity agricultural-pasture areas, and recent urbanization increased sediment and nutrient accumulation in the lagoon, resulting in the intensification of siltation and eutrophication processes. This situation can also be potentialized by the loss of lagoon area and an increase in wetlands, both of which serve as siltation indicators during urbanization expansion. Although average SAR, TOC, TN, and TP concentrations and burial increased from 1932 to 2013, temporal variability indicated the influence of raw sewage emissions from urbanization. It also revealed that a temporary artificial connection of the lagoon to the sea could have different responses, reducing nutrient and particle deposition. Thus, an integrated management and mitigation approach should be implemented at the local and regional levels, with a focus on reforestation, particularly of riparian vegetation and other permanent protection areas; sewage treatment; variable connections to the sea; and lagoonal floodplain conservation to prevent siltation and eutrophication.

CRediT authorship contribution statement

Roberto Nascimento de Farias: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Theo Dias Arueira:** Writing – original draft, Visualization, Methodology, Investigation, Formal analysis. **Arthur de Barros Bauer:** Writing – original draft, Visualization, Methodology, Formal analysis. **Carlos Eduardo de Rezende:** Writing – review & editing, Validation, Resources, Methodology, Funding acquisition. **Marcelo Gomes de Almeida:** Methodology, Conceptualization. **Carla Regina Alves Carvalho:** Methodology, Funding acquisition. **Marcella Vidal:** Methodology. **Guilherme Sardenberg Barreto:** Writing – review & editing, Conceptualization. **Joao Augusto A. Novaes:** Writing – review & editing, Methodology, Formal analysis. **Marcos Paulo Figueiredo de Barros:** Supervision, Resources, Project administration, Funding acquisition, Conceptualization. **Mauricio Mussi Molisani:** Writing – review & editing, Supervision, Resources, Methodology, Funding acquisition, Formal analysis, Conceptualization. **Francisco de Assis Esteves:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envres.2024.120161>.

Data availability

Data will be made available on request.

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