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Abstract

The Jacuí Delta drains into Lake Guaíba which is the main source of water for more than two million people in South Brazil and has suffered from pollution with heavy metals and phosphorus. The objective of the current research is to demonstrate how the use of sediment source tracing techniques, in combination with sediment flux monitoring, can improve understanding of the sediment source contributions to one of the largest lakes in South America. The sediment flux monitoring results were based on data obtained from 12-years of records of water flow and suspended sediment concentrations. The sediment source fingerprinting approach was based on the use of geochemical tracers. Based on the results of the source tracing study, the respective contributions of the tributaries to the sediment in Lake Guaíba were estimated to be as follows: the Jacuí River (median of 54% - interquartile range (IQR) 34-71%), the Caí River (12%, IQR 7-16%), the Sinos River (5%, IQR 1-20%), and the Gravataí River (16%, IQR 10-30%). These results are similar to those derived from the sediment flux monitoring, namely: the Jacuí River (70%), the Caí River (19%), the Gravataí River (4%), and the Sinos River (7%). These results demonstrate that the sediment source fingerprinting approach combined with sediment flux monitoring can provide a useful means of estimating the respective sediment contributions from individual tributaries in large and complex delta systems and may provide a powerful tool to guide water resource management.

Keywords: sediment source fingerprinting, sediment tracing, water flow and sediment monitoring, elemental geochemistry, source to sink.

Highlights

- The results provided by fingerprinting and sediment flux monitoring were very similar.
- Zn, Cr, Mn and Ba were able to correctly classify 95% of the tributary sediment samples.
- The Jacuí River is the main sediment source to Lake Guaíba (54%).
- The Jacuí River has the lowest specific sediment yield (SSY) (20 Mg km^{-2} $year^{-1}$).

The Sinos and Caí Rivers have SSYs 2.2 and 4.3 times higher than the Jacuí River.

1. Introduction

Most large centers of population have developed close to sources of water. However, as they grow and utilize this natural resource, large loadings of contaminants, such as those associated with domestic and industrial sewage, are frequently generated and released into waterbodies. As a consequence, it is common to observe contamination of water bodies with heavy metals that are harmful to human and environmental health (Maggioni dos Santos et al., 2021). Moreover, the increasing occurrence of algal blooms, due to excessive inputs of phosphorus, have attracted worldwide attention, because they seriously affect aquatic life, local landscapes, tourism, and drinking water supplies (Duquesne et al., 2021; Mu et al., 2021).

Lake Guaíba is a freshwater lake which is surrounded by the city of Porto Alegre (Fig. 1) (which is the capital of the State of Rio Grande do Sul) and other large cities, such as Canoas and Guaíba. This lake is the main water supply for more than two million people. It a covers a surface area of 482 km^2 and is fed by water coming from four main rivers: the Jacuí River, the Sinos River, the Caí River, and the Gravataí River. Those rivers converge and flow into the Jacuí Delta, forming a transitional environment (from fluvial to lacustrine), and this water then flows through the lake until it reaches the Patos Lagoon (de Andrade et al., 2019). In the last several decades, episodes of water quality problems in Lake Guaíba have become increasingly frequent, mainly due to eutrophication (Andrade and Giroldo, 2014; Ribeiro et al., 2012).

The Jacuí River is the main tributary of Lake Guaíba, as it drains an area that covers about 84% of the total catchment area of the lake. The catchment of the Jacuí River is characterized by many sources of diffuse pollution linked to agricultural activities. The impact associated with industrial activities and urbanization is limited. The Lower Jacuí catchment is mainly occupied by rice fields, with a direct connectivity of water and sediment between the floodplain and the river channel. The Upper Jacuí catchment is more heterogeneous and is characterized by the cultivation of row crops, such as soybean and corn under no-tillage in plateau areas with deeper soils, and tobacco grown in steeper areas with shallower soils. Intensive livestock production of pigs and poultry is found across the entire region. The lack of land use planning and the limited implementation of soil conservation measures result in a large contribution of sediment from the cropland areas (Tiecher et al., 2017, 2015). In addition, the intensive use of fertilizers and pesticides has resulted in the contamination of river sediment with phosphorus (Bender et al., 2018; Reichert et al., 2019; Tiecher et al., 2019; Zafar et al., 2017, 2016), various agrochemicals (de Castro Lima et al., 2020; Fernandes et al., 2019) and veterinary drugs (Camotti Bastos et al., 2018).

The Gravataí, Sinos and Caí catchments are much smaller than that of the Jacuí River, and correspond to approximately 2.3, 4.3 and 5.9% of the area draining to Lake Guaíba, respectively. These catchments are impacted by both agricultural and industrial activities. The Caí, Gravataí, and dos Sinos Rivers are well known for their pollution, especially by heavy metals, as they flow through industrial areas in a dense urban region and suffer from the associated deleterious environmental impacts (de Andrade et al., 2018a).

Recent studies have shown that the sediment of Lake Guaíba is contaminated with high levels of heavy metals (de Andrade et al., 2019) and phosphorus (de Andrade et al., 2018b) due to the pollution originating from the different tributaries. This may increase water treatment costs and significantly decrease the quality of the water supplied to a large metropolitan region (McDonald et al., 2016). As most of the pollution found in Lake Guaíba likely originates from diffuse sources within its tributary catchments and no point source is known to directly release contaminants into the lake, it is necessary to better understand the contribution of sediment from each tributary, in order to propose solutions to limit the sources of contamination. Therefore, there is a need for quantitative information on the sources of the sediment delivered to the lake. However, to the best of our knowledge, the amounts of sediment contributed by each tributary to Lake Guaíba have not to date been quantified.

In order to improve our understanding of water and sediment dynamics in catchments, a suite of different monitoring techniques and numerical models have been developed around the world. The monitoring of water flow and sediment discharge through time will reflect the impacts of the intrinsic characteristics of the site, as well the effects of climate and land use change on the river regime (Minella et al., 2017). The sediment yield, which is generally obtained through water flow and sediment monitoring, integrates all the processes operating and the characteristics of each tributary (Lemma et al., 2020; Zou et al., 2020).

Another way to obtain an estimate of the contribution of a given tributary (or sediment source) is by using methods based on the fact that the physical and chemical characteristics of the sediments are related to the characteristics of the main tributaries. For example, if a given source of sediment (tributary) is enriched in some chemical element compared to others, this chemical element can be used as a tracer of that source. However, the use of a limited set of chemical elements generates mathematical uncertainty in the determination of sources (Collins & Walling, 2002), especially when studying multiple sediment sources. In this sense, Yu & Oldfield (1989) have demonstrated that the quantitative resolution of multiple sources of sediments can be achieved by using composite sets of chemical elements as tracers, or fingerprints – what gave the fingerprinting technique its name. In this sense, Minella et al. (2008) demonstrated the potential value of combining sediment source tracing techniques with traditional water flow and sediment monitoring approaches, when documenting the impact of improved land management on the suspended sediment load of a catchment. Statistical and computer modelling techniques, such as the sediment fingerprinting approach, provide a powerful suite of methods for quantifying the contribution of each potential source delivering sediment into water courses (Collins et al., 2020). This technique has been increasingly used in Brazil during the last decade, mainly in the southernmost region of the country (Minella et al., 2014; Ramon et al., 2020; Rodrigues et al., 2018; Tiecher et al., 2016; Valente et al., 2020). However, this method is now attracting interest in other parts of the country (Batista et al., 2018; Bispo et al., 2020; Franz et al., 2014; Lima et al., 2020).

The sediment fingerprinting approach primarily relies on the identification of potential sediment sources in the field (Collins et al., 2017), and the subsequent analysis of the chemical fingerprints of both the potential sources and the transported sediment (Walling et al., 2013). Fingerprint properties to be used as tracers must show a statistical difference between the potential sources (Koiter et al., 2013). The pre-selection of potential tracers to be included in this approach can depend on availability of laboratory infrastructure or rely on a targeted approach relying on pedological or lithological knowledge (Batista et al., 2018). Following both approaches, the tracer properties must remain conservative during the continuum of erosion, transport, deposition and remobilisation processes that may occur during sediment transfer across the catchment (Owens et al., 2016). In addition, for the statistical approach, tracers are generally selected after passing a non-parametric test to identify those properties that differ between potential sources, which are then entered into a discriminant function analysis in order to select the minimum set of tracer properties that provides the maximum discrimination between the potential sources (Collins et al., 2020). Subsequently, a modelling exercise is carried out to determine the percentage contribution of each sediment source (Collins et al., 2020).

Therefore, the objective of this research was to estimate the contribution of sediment from the four main tributaries to the Jacuí Delta, which drains into Lake Guaíba, between 2000-2011. Our hypothesis is that the use of sediment source tracing techniques in combination with more traditional monitoring techniques, can improve our understanding of the sediment sources associated with one of the largest freshwater lakes in South America. For this purpose, we used data publicly available from long-term monitoring of discharge (Q) and suspended sediment concentration (SSC) combined with the shorter-term monitoring of chemical element concentrations in bed sediment (2000-2011).

2. Materials and methods

2.1. Study site

The study reported was undertaken in the Jacuí River Delta, which drains into Lake Guaíba, located in the State of Rio Grande do Sul (RS), Brazil (Fig. 1). The climate of Rio Grande do Sul is of the temperate subtropical type, classified as Humid Mesothermal with an annual rainfall between 1300-1500 mm in the southern half of the state and 1500-1800 mm in the northern half (Dubreuil et al., 2018). The four seasons are well defined, and rainfall is well distributed throughout the year. Annual average temperatures vary between 15 and 18°C (ranging from -10°C to 40°C).

The total drainage area of Lake Guaíba is 85,139 km², of which 482.2 km² represents the lake itself and 28.1 km² the Jacuí delta (SEMA, 2020). The remaining area corresponds to small streams that drain directly into the lake (2,463 km^2 – 2.9%). Lake Guaíba has four main tributaries: the Gravataí River, the Caí River, the Sinos River, and the Jacuí River (Table 1). Together, these four tributaries account for 96.5% of the catchment area of the lake. This area contains 61% of population of the Rio Grande do Sul State population (6 million inhabitants), with a large concentration in the metropolitan region of Porto Alegre, the capital. The main characteristics of these four tributaries of Lake Guaíba are shown in Table 1.

The drainage area of Lake Guaíba embraces a great diversity of geology (Fig.2 and 3), soil type (Fig. 3) and climate characteristics which control the main land uses. In the northern part, the highest areas of the State are found, while to the west the Depressão Central is associated with the lowest altitudes. Finally, to the south, the Planalto Sul-Riograndense (Escudo Sul-Rio Grandense) consists of a vast plateau (Fig. 4). The existing native vegetation comprises Mixed Ombrophilous Forest, Seasonal Forest and the Pampa Biome. However, much of this vegetation has been altered, with only residual native vegetation patches found on the steep slopes of the valleys, especially in the Jacuí River basin, and more specifically in the Taquari-Antas River basin. A summary of the main land uses in each tributary provided in Table 1, is based on the Mapbiomas database for the year 2018 (Souza et al., 2020). The proportion of the catchment surface covered with soils derived from basalt and rhyolite (volcanic rocks) is higher in Jacuí, Caí and Sinos river catchments (ranging from 31-51%) compared to the Gravataí catchment (only 3%) (Figs. 2 and 5). In contrast, the proportion of the basin covered with soils derived from sedimentary rocks is the highest in the Gravataí catchment (83%) (Figs. 2 and 5).

2.2. Tributary sediment contributions estimated using flow and sediment monitoring data

2.2.1. Flow and sediment monitoring data acquisition

Flow and sediment monitoring data were obtained through the monitoring network coordinated by the Brazilian National Water Agency (*Agência Nacional de Águas – ANA*) and operated by the Geological Survey of Brazil (*Companhia de Pesquisa de Recursos Minerais - CPRM*), which is responsible for collecting field observation data (http://www.snirh.gov.br/hidroweb/serieshistoricas). Initially, a review of ANA monitoring data for the four tributaries was undertaken to establish the availability of data on flow and suspended sediment concentration. However, the entire Gravataí catchment (1,977 km^2), currently lacks any monitoring station with data available, and therefore it was not possible to estimate its sediment discharge to Lake Guaíba using flow and sediment monitoring data. The Caí and Sinos catchments are equipped with monitoring stations, with data available for locations close to their confluences with Lake Guaíba, and these were used to estimate their respective suspended sediment fluxes. Conversely, we decided to apply regionalization to estimate sediment yield in the Gravataí tributary. For this purpose, we used the monitoring data from the Sinos tributary, due to the similar characteristics of these catchments.

The Jacuí catchment is the largest tributary of Lake Guaíba and contains several monitoring stations. As none of these are located close to its mouth, a sediment yield regionalization study was conducted using the existing monitoring stations in its sub-catchments, namely: the Alto Jacuí, Baixo Jacuí, Pardo, Vacacaí and Taquari-Antas sub-catchments. For the Pardo and Baixo Jacuí sub-catchments, there were no data available and the sediment yield values for these sub-catchments were estimated using the results of the regionalization based on the aforementioned sub-catchments. Table 2 shows the origin of the data used in the regionalization of the specific sediment yield and to estimate the total sediment input into Lake Guaíba. Fig. 1 shows the location of each tributary and the monitoring stations used in the current analysis.

2.2.2. Estimation of the annual suspended-sediment yields (SY) and specific-sediment yields (SSY) of the Lake Guaíba tributaries

Discharge (Q) and suspended sediment concentration (SSC) data were obtained from the Brazilian Water Agency (ANA) website HIDROWEB (https://www.snirh.gov.br/hidroweb/apresentacao) which provides access to the hydrological and water quality data from the national monitoring network. In Brazil, suspended sediment concentration data are collected randomly four times a year during several years (Horowitz et al., 2015) using a standard isokinetic sediment sampler as described by Edwards and Glysson (1999). Unfortunately, the random sampling schemes adopted by ANA in Brazil rarely include measurements during floods, which are the periods of greatest sediment transport and highest sediment concentrations (Merten et al., 2006). Therefore, if such data are used to estimate the annual suspended sediment yield from a catchment, it is likely to be underestimated. Fig. 7 shows the flow frequency analysis with the number of sediment samples for the SSC determination for the Lake Guaíba tributaries used to estimate the annual suspended-sediment yields. Values of daily SSC for the Caí, Sinos, and Jacuí Rivers were estimated using the sediment-rating curve procedure as proposed and described in detail by Horowitz et al. (2001) and Horowitz (2010, 2003). The SSC was carried out by filtration and evaporation methods (Shreve & Downs, 2005). Filtration was performed with a 934 AH glass fiber filter membrane (Guy, 1969), which has the capacity to retain particles larger than 1.5 µm. Using the evaporation method, the samples were placed in an oven at 105°C +/- 5°C until weight constancy.

Briefly, values of SSC for the available samples and associated values of flow discharge (Q) for each site were log-transformed and a simple sediment-rating curve was established for each site (Fig. 8). These rating curves and their associated regression equations were used in combination with the records of daily discharge to synthesize time series of daily SSC and estimates of daily sediment load were calculated, according to equation (1):

$q_{ss} = Q \times SSC \times 0.0864$ (Eq. 1)

Where: q_{ss} represents daily sediment load (Mg day^{-1}), Q is the daily mean water discharge (m^3 s^{-1}), SSC is the daily suspended sediment concentration (mg L^{-1}), and 0.0864 is a constant used when converting q_{ss} into Mg day^{-1}. The annual sediment yield (SY) was calculated by summing the daily q_{ss} values and the specific sediment yield (SSY) was estimated by dividing the SY (Mg year^{-1}) by the catchment area (km^2).

It is important to note that the rating curves used in our study are based on all available sediment concentration data for the sampling stations (from 1978-1986 to 2017-2019) and not just the data for the study period (2000-2011). We are aware that this may be a limitation of the approach used, as it is impossible to assume that conditions will have been stationary over the total sampling period, due to land use change, for example. However, we decided to use this longer series of data to increase the number of samples used to derive the rating curves. Even using all the period available, the coefficients of determination are low (Fig. 8) indicating that the fit is quite poor and that there is considerable uncertainty associated with the predicted sediment concentration values.

The sediment yield from each tributary to Lake Guaíba was determined as the mean annual suspended sediment yield for the period 2000-2011, which was the same period as that corresponding to the period of collection of sediment samples used to analyze the concentration of chemical elements for the sediment tracing study using the fingerprinting method (see 2.3.1).

In the absence of monitoring data for some of the Jacuí River sub-catchments [Baixo Jacuí, Pardo, Forqueta and Baixo Taquari-Antas (Table 2 and Fig. 1)], SSY was estimated using the regionalized SSY, taking account of physiographic similarities between sub-catchments (Fig. 1, 2, 3 and 4). Thus, for example, the SSY of the Forqueta sub-catchment (Table 2 and Fig. 1) was estimated based on the area-weighted average SSY of the Carreiro and Guaporé sub-catchments, since they share very similar physiographic characteristics. The same approach was considered for the Gravataí River using the monitoring data from Sinos River.

2.3. Estimation of tributary sediment contributions using the fingerprinting approach

2.3.1. Sediment sampling and analysis

In this study, fine bed sediment was used as a surrogate for suspended sediment when establishing its geochemical composition, as suggested by Horowitz and Elrick (2017). Bed sediment sampling was carried out between 2000 and 2011 at seven sites by the Municipal Department of Water and Sewage (DMAE) of Porto Alegre, RS – Brazil, the environmental agency responsible for monitoring the water and sediment quality of Lake Guaíba. The four main river tributaries draining into Lake Guaíba, namely, the Jacuí River, the Gravataí River, the Caí River, and the Sinos River were considered to be the primary potential sediment sources and samples of bed sediment were collected close to their outlets into the lake. Information regarding the sampling locations and the number of sediment samples collected for each tributary is presented in Fig. 6.

Within the Jacuí River delta, three sites were sampled to characterize the deposited sediment found at that location (Fig. 1). These included the Ilha da Pintada Channel that receives water from the Jacuí River only (used as a control site), the Navegantes Channel that receives water from all four rivers, but which only receives part of the Jacuí River flow, and Lake Guaíba representing a position that integrates all the water and sediment supplied to the lake by the four main tributaries. Information regarding the location and number of sediment samples collected at each sampling site within the Jacuí River delta is also presented in Fig. 6.

At all seven sampling sites, bed sediment was sampled using a Petersen dredge. Once collected, the material was oven-dried (50°C) and sieved to < 0.177 mm (Mesh Tayler nº 80). The < 0.177 mm fraction of the sediment was assumed to represent suspended sediment deposited at that location. The particle size was defined by the DMAE agents to remove coarse sand particles, which are considered not to transport chemical elements and other pollutants. After that, we used the range test to avoid using tracer chemical element in our study that may be affected by enrichment or impoverishment. The concentrations of eleven metals in this material were analyzed by atomic absorption spectrophotometry after pseudo-total digestion (EPA 3051a for Al, Fe, Mn, Ba, Zn, Cu, Pb, Cr, Ni, Cd, and EPA 7473 for Hg (EPA, 2007)). Bed sediment sampling was undertaken once or twice a year between 2000 and 2011 (except for 2002 and 2003 due to budget constraints), with one sampling in spring/summer and another in autumn/winter (Fig. 6), without a fixed calendar, resulting in the collection of a total of 129 samples.

2.3.2. Tracer selection and sediment source apportionment

The selection of tracer properties was performed following three steps: (i) a range test; (ii) the Kruskal–Wallis *H* test (KW-H test); and (iii) a linear discriminant function analysis (LDA). The range test was based on the interquartile range (IQR) as proposed by Batista et al. (2018). When the median ± IQR (25^{th} and 75^{th} percentiles) of the concentration of chemical elements in bed sediments lied outside the range of the concentration of the chemical elements in the bed sediment sampled in the sources (the four main tributaries), the chemical element was considered not conservative and was excluded from further analysis. The KW-H test was used to test the null hypothesis ($p<0.01$) that the sources belonged to the same population (Collins et al., 1997). The KW-H test demonstrated which properties exhibited statistically significant differences between the four potential sediment sources and could be considered as potential tracers).

Subsequently, a multivariate LDA was undertaken to determine the minimum number of variables or sediment properties that maximizes the discrimination between the sources. LDA was performed only with the variables that passed the KW *H*-test. LDA is based on the Wilks' lambda (Λ^*) value from the analysis of variance, where the criterion used by the statistical model is the minimization of Λ^*. A Λ^* value of 1 is found when all the group means are the same whilst a low Λ^* value means that the variability within the groups is low compared to the overall variability. The LDA was performed in the forward mode, with 0.05 used as the maximum or minimum significance of F required to include or remove a property.

A mass balance mixing model was used to estimate the source contributions by minimizing the sum of squared residuals (SSR). The mixing model was solved using a Monte Carlo simulation with 2,500 iterations (Batista et al. 2018) (Eq. 2). Optimization constraints were set to ensure that source contributions were non-negative and that they summed to 1.

$$SSR = \sum_{i=1}^{n}\left(\left(\mathrm{C}_i - \left(\sum_{s=1}^{m}\mathrm{P_s S_{si}}\right)\right)/\mathrm{C}_i\right)^2 \qquad \text{(Eq. 2)}$$

where *n* is the number of parameters in the model chosen by the three-step selection process; C_i is the bed sediment sample property (i); m is the number of sources; P_s is the contribution of source (s); S_{si} is the mean of parameter (i) in source (s).

Analyses were undertaken using R software (R Development Core Team, 2017). The modelled source contributions are represented by the median and interquartile range (IQR, Q_{25} and Q_{75}) of predicted values from the Monte Carlo simulations for each individual sample. The source contribution for each sediment sampling site was obtained from the average of the median and IQR values obtained for each individual sample.

3. Results and discussion

3.1. The SY and SSY inventory for Lake Guaíba

The SY data indicate that during the period 2000-2011 the Jacuí River was the main source of sediment (16.9 x 10^6 Mg) to Lake Guaíba (70% - Table 3). However, the SSY of the Jacuí River (20 Mg km^{-2} $year^{-1}$) is much lower than those estimated for the Caí River (85 Mg km^{-2} $year^{-1}$) and the Sinos River (44 Mg km^{-2} $year^{-1}$).

Although the Caí and Sinos Rivers drain catchments with similar surface areas (5,004 km^2 and 3,667 km^2), the Caí River (85 Mg km^{-2} $year^{-1}$) contributes almost twice as much sediment as the Sinos River (44 Mg km^{-2} $year^{-1}$) (Table 3). The higher SSY in these two catchments can be explained by the complex dynamics occurring in these drainage basins. The Caí catchment is characterized by steep slopes (Fig. 4) and shallow soils such as Leptosols and Regosols (Fig. 3), and the fragile environment is strongly impacted by agricultural activities.

The Sinos catchment has the second smallest drainage area (3,667 km^2) but has the second highest concentration of inhabitants per km^2 (333). The number of industries located along the river flood plains increases in a downstream direction, causing great pressure on water resources (Nascimento et al., 2015). In addition, with exception of the less urbanized areas in the headwaters, the catchment is devoid of dense vegetation cover. In the lower parts of the catchment, where there is a higher population density, there is sparse vegetation cover and the presence of paddy fields (flooded rice) in the riverbanks (Bianchi et al., 2019; Pedde et al., 2015).

The extensive plains used mainly for agricultural activities found in the lower Taquari-Antas and Baixo Jacuí Rivers in the Jacuí catchment result in a greater potential for sediment to be deposited before it reaches Lake Guaíba (Fig. 1 and 4). Therefore, it is possible that material remains stored in the channel within these plains and that part of this material does not reach the Jacuí Delta. This catchment has a low population density compared to the other investigated zones, with around 38 inhabitants km^{-2}. Therefore, even though this catchment provides the greatest contribution of sediment to Lake Guaíba and has the highest SY, it is characterized by the lowest SSY among the main tributaries. Therefore, the largest sediment input into Lake Guaíba supplied by this river reflects the size of the Jacuí catchment and its higher flow contribution – it supplies 86.3% of the water that reaches Lake Guaíba (Table 1). Despite the low temporal coverage of discharge and suspended sediment monitoring records of the lower part of Jacuí, the sediment contribution data obtained with the fingerprinting technique demonstrate that the sediment contribution of Jacuí may be even lower (54% - see further ahead).

Although it is not possible to calculate the contribution of sediment from the Gravataí catchment due to the lack of flow and sediment monitoring data, it is characterized by the highest population density of the four main tributaries of Lake Guaíba (656 inhabitants km^{-2}). However, when the sediment contribution was calculated, based on the SSY (Mg km^{-2} $year^{-1}$) of the Sinos River, a value of 1.0 x 10^6 Mg was obtained, which corresponds to 4% of the sediment that reaches Lake Guaíba. Land use and cover in this catchment are dominated by two distinct activities, with agriculture dominating in the upper parts with steep slopes and widespread urban-industrial use being found in the lower reaches of the river (Table 1). Accordingly, as in the Sinos River, water is widely used for irrigation of rice fields, which demands a large amount of water, causing great pressure on the water courses in this area.

3.2. The sediment fingerprinting approach

High pass rate in the conservativeness test was found. Ten of the 11 chemical elements analysed (Al, Fe, Mn, Ba, Zn, Cu, Pb, Cr, Ni, Cd and Hg) passed the interquartile range (IQR) test; only Hg was found not to be conservative (Table 4). Notwithstanding, it is well known that the conservative behavior all depends on the type of range test that is conducted as it may be more or less restrictive regarding those properties that are considered to remain conservative. We know that this is a crucial hypothesis of the sediment fingerprinting method. Unfortunately, there are few alternative methods to take this process into account. When high local anthropogenic pollution levels are found (which is not the case in the current research, otherwise it would have been outlined by the range test), alternative analysis techniques targeting the residual metal fraction were shown to provide an alternative, which may be recommended in study areas characterized by high local anthropogenic pollution levels (Dabrin et al., 2021).

From the ten elements that passed the range test, nine passed the KW-H test ($p<0.01$) and therefore all except Cd were considered as potential tracers (Table 4). These nine tracers were then entered into a discriminant function analysis. Four tracers (Zn, Cr, Mn, and Ba) were selected as the minimal set of tracers that most successfully accounted for the differences between the tributaries of Lake Guaíba (Table 5).

The Wilks' lambda parameter was only 0.015 (Table 5). This means that the total variance due to errors in the source discrimination was 1.5%. Accordingly, the optimal set of four tracers (Ba, Cr, Mn, and Zn) explained approximately 98.5% of the differences observed in sediment collected in the main tributaries of Lake Guaíba. Using these four tracers only, it was possible to correctly classify 94.6% of the source samples, and the tributary groups of samples were significantly separated by an average squared Mahalanobis distance of 41.6 (Table 5 and Fig. 9). This discrimination is considered sufficient and although including more chemical tracers could result in a higher classification of samples in their groups of origin, we chose not to include them to avoid information redundancy and increased complexity of the model.

The differences in chemical element concentrations found in sediment transported by the four main tributaries of Lake Guaíba can be explained by the difference in their respective geologies/soil types (Mn) and by the main human activities occurring in each catchment (Cr and Zn). In the State of Rio Grande do Sul, soils derived from basalt and rhyolite are naturally richer in Cr and Zn than soils derived from sedimentary rocks (Althaus et al., 2018). Therefore, when analyzing the main lithologies found in the catchments, it would be expected that the levels of Cr and Zn in the bed sediment collected in the Jacuí, Caí and Sinos rivers would be similar (Fig. 5). However, it is possible to observe that among these three catchments, the bed sediment of the Jacuí River has the lowest concentrations of Cr and Zn (Fig. 11). The higher content of these metals in sediments of Caí and Sinos rivers can be explained by the historical occurrence of tannery (leather and footwear industry) activities in these subcatchments (de Andrade et al., 2019). The low content of Zn and Cr clearly indicates that the sediment sampled on the Ilha da Pintada Channel has great similarity to the bed sediment sampled at the mouth of the Jacuí River (Fig. 10e).Based on the natural content of geochemical elements in the soils of each catchment, low Zn concentrations were expected in sediment transported by the Gravataí River where sandstone derived soils predominate (Fig. 5). However, the sediment sampled in this river showed the highest Zn content (Fig. 11). This is likely due to the fact that the river flows through the metropolitan region between the cities of Gravataí, Porto Alegre, and Canoas, where a large amount of untreated sewage is discharged into the river (de Andrade et al., 2019, 2018b).

In contrast to Cr and Zn, the contents of Mn found in the sediment followed the respective natural Mn concentrations associated with the dominant lithologies (Althaus et al., 2018) found in each catchment. Accordingly, the higher the proportion of basalt and rhyolite outcrops in the catchment (and the lower the sedimentary rock outcrops), the higher the concentration of Mn in the sediment (Fig. 11).

In the Ilha da Pintada Channel, 83% (IQR of 67-93%) of the sediment contribution was estimated to originate from the Jacuí River (Fig. 12). This was expected, because the Jacuí River is the main water source at this sampling location. Accordingly, the remaining 17% of the sediment transported past this site is expected to originate from the other three tributaries (i.e. the Sinos, Gravataí, and Caí Rivers). This also indicates that there is likely to be substantial circulation of the suspended sediment in the Jacuí delta as observed by Nicolodi et al. (2013), which may be affected both by the variation of the river flow and by the dominant wind direction.

The Navegantes Channel showed respective sediment contributions from the rivers of Jacuí (0.1%, IQR 0.1-4.2%), Caí (20%, IQR 8-29%), Sinos (17%, IQR 5-38%), and Gravataí (55%, IQR 40-68%) (Fig. 12). This unbalanced mixture is likely explained by the location of this sampling point close to the mouth of the Gravataí River (Fig. 12). In addition, there is a limited circulation of material originating from the Jacuí River and a greater circulation of water from the other tributaries at this location (Nicolodi et al., 2013), causing a dilution effect (Fig. 12). Finally, at the sampling point where the sediment was expected to reflect all the potential sediment sources contributing sediment to Lake Guaíba, their respective contributions were estimated as follows: Jacuí River (54%, IQR 34-71%), Caí River (12%, IQR 7-16%), Sinos River (5%, IQR 1-20%), and Gravataí River (16%, IQR 10-30%).

Contrary to the estimate that 86% of the waters entering Lake Guaíba come from the Jacuí River (Table 1), the results of the current sediment fingerprinting study suggests that only half of the sediment (54%, IQR 34-71%) accumulating in the lake originates from this river (Fig. 13), which is characterized by the lowest SSY among the investigated tributaries (Table 3). For dos Sinos and Caí rivers, where both sediment fingerprinting and flow and sediment monitoring were available, and where most of the catchment area was covered by the discharge and sediment monitoring network (approximately 86% – Table 2), the respective sediment contributions estimated using both approaches were similar (Fig. 13). Although the results of the two approaches were also similar for the Jacuí River, this comparison should be avoided or made with caution, as only half of the catchment area was covered by the discharge and suspended sediment monitoring network (Table 2) and most of the lower part of the catchment area where sedimentary rocks predominate was not monitored.

3.3. Limitations and future research perspectives

Studies carried out in large and complex river basins such as the Lake Guaíba basin require multiple lines of evidence to better understand their suspended sediment dynamics. The current research attempted to provide such information by using two different but complementary techniques. Although the results of a decade of monitoring and sample collection (2000-2011) using the two different techniques resulted in similar estimates of the sediment contributions from the tributaries, the inter-annual variation of the relative contributions of tributaries did not show any clear correlation between the two approaches (data not shown). This may be due to the different sampling strategies on which each of the approaches relied. Although the sediment rating curve may provide a good basis for describing the sediment regime of watercourses (Efthimiou, 2019), the monitoring of suspended sediment concentrations remained very limited in our study. Only four field measurements of suspended sediment concentrations were conducted per year according to a random schedule, which may not cover the high magnitude rainfall-discharge events (Fig. 7 and 8), which are responsible for transporting most of the sediment load associated with a given year (Horowitz, 2008). Moreover, it is expected that estimates of annual sediment load provided by the rating curve approach do not reflect actual inter-annual variation in sediment loads, since the same lumped rating curve is used for every year and the annual estimates are largely a reflection of the water discharge record for each year and take no account of likely variations in concentration between different events and years. Therefore, the rating curve approach is more appropriate for estimating the mean annual sediment load and not annual sediment loads.

Another important limitation and source of uncertainty in our study is that only about 52.3% of the total area of the Lake Guaíba catchment was covered with discharge and sediment concentration monitoring (Table 2). The only tributaries that were covered with a good monitoring of water discharge and sediment concentration were the Caí and Sinos rivers (with networks covering ca. 86% of the total area). The entire Gravataí River catchment was not monitored, and a significant section of the Jacuí River flowing across sedimentary rocks was not monitored at all (Fig. 1).

In addition, the sediment sampling strategy used in the source fingerprinting approach involved collecting one or a maximum of two sediment samples per year at each sampling site. This may be sufficient to discriminate the sediment that comes from each tributary (Fig. 9 and 10), but it may fail to capture the seasonal variation in the sediment source contributions that reach the Jacuí Delta. Therefore, future studies should consider a more intensive sediment sampling programme in both space and time and particularly the collection of more samples throughout the year, including during flood periods, in order to improve sediment yield predictions using the rating-curves, and also to capture the inter-event variation of sediment sources using the fingerprinting approach.

Future studies should also involve more detailed sampling in the lower portions of Lake Guaíba and include the Dilúvio Stream as a potential source of sediment (Fig. 12). Although the Dilúvio Stream is located below the Jacuí Delta and it has a much lower water discharge than the four tributaries evaluated in the present study, it is an urban river highly impacted by heavy pollution with metals and phosphorus resulting from anthropogenic activities (Maggioni dos Santos et al., 2021). The Dilúvio Stream catchment has been polluted by several sources, including unplanned land development, discharge of untreated sewage and dense road traffic. Furthermore, this river is devoid of riparian forest along most of its course, and in the lowest 12 km section, the stream has been channeled, which increases the contribution of surface runoff from the catchment and the associated input of road-derived contaminants (Maggioni dos Santos et al., 2021). Accordingly, although the sediment contribution of the Dilúvio Stream to Lake Guaíba is likely to be low, it may supply a large number of pollutants that should be explicitly considered in future research.

In addition to denser sampling of sediment in the lower portions of Lake Guaíba to assess the current tributary contributions of suspended sediment, future studies could also collect and analyse sediment cores from the Lake to reconstruct the degradation of this aquatic systems during the Anthropocene. For this purpose, dating of sediment cores could be based on the identification of natural excess lead-210 ($^{210}Pb_{ex}$) and artificial radionuclides (e.g. caesium-137 (^{137}Cs)). Moreover, environmental DNA (eDNA) analyses of sediment could also help to reconstruct the changes in land use/farming practices across the catchment during the last several decades (Evrard et al., 2019; Foucher et al., 2020). The eDNA signatures have been shown to be conservative during soil erosion and sediment transfer in river systems and will be protected from further degradation after the deposition of the sediment in water bodies (Capo et al., 2021; Foster et al., 2020). The combined use of physical-chemical tracers such as those employed in the current research and novel biological indicators such as eDNA in sediment archives associated with sediment sinks could provide additional lines of evidence regarding sediment source contributions and their changes through time to guide the future management of soil and water resources such as those found in Lake Guaíba and its drainage basin.

4. Conclusions

Lake Guaíba, one of the largest lakes in South America, drains a complex hydrographic system where a large amount of sediment accumulates leading to environmental problems. To the best of our knowledge, the current research provides the first quantitative estimate of the respective contributions of the four main tributaries to the sediment accumulated in Lake Guaíba. To this end, two different approaches, based on sediment flux monitoring and on the analysis of geochemical tracers using a sediment fingerprinting approach were used. The estimates of the sediment contributions from the tributaries were similar for both approaches. The Jacuí River provided the main source of sediment that flows into Lake Guaíba, contributing between 54 and 70% of the sediment input to Lake Guaíba. Although the catchment of the Jacuí River accounts for about 87% of this hydrographic region, it has the lowest specific sediment yield among the investigated tributaries. Although the Caí (5,004 km^2) and Sinos (3,667 km^2) catchments are of similar size, their respective contributions to the sediment input to Lake Guaíba were different, with an average of 12 to 18% for the River Caí, which was twice to three times greater than the contribution of the Sinos River (between 5 and 7%). Finally, for the Gravataí River, the estimated sediment contribution ranged from 4% (when relying on sediment flux monitoring using dos Sinos River historical data) to 16% (when relying on fingerprinting). These results reflect the more intensive land use of a region characterized by steeper slopes in the Caí River catchment leading to a much higher sediment yield compared to other sub-catchments. These results show that the sediment fingerprinting approach could provide a useful tool for estimating the sediment contributions from contrasting tributaries in complex and extensive hydrographic systems. Combining the two approaches can provide a powerful tool to guide the decisions to be made by water resource managers. In the future, sediment cores could be collected in Lake Guaíba to attempt to reconstruct the degradation of this aquatic system during the Anthropocene based on sediment dating with natural ($^{210}Pb_{ex}$) and artificial radionuclides (e.g. ^{137}Cs) and the analysis of next-generation source tracers such as environmental DNA (eDNA) to reconstruct the impact of past land use/farming practices in this region.

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Figures

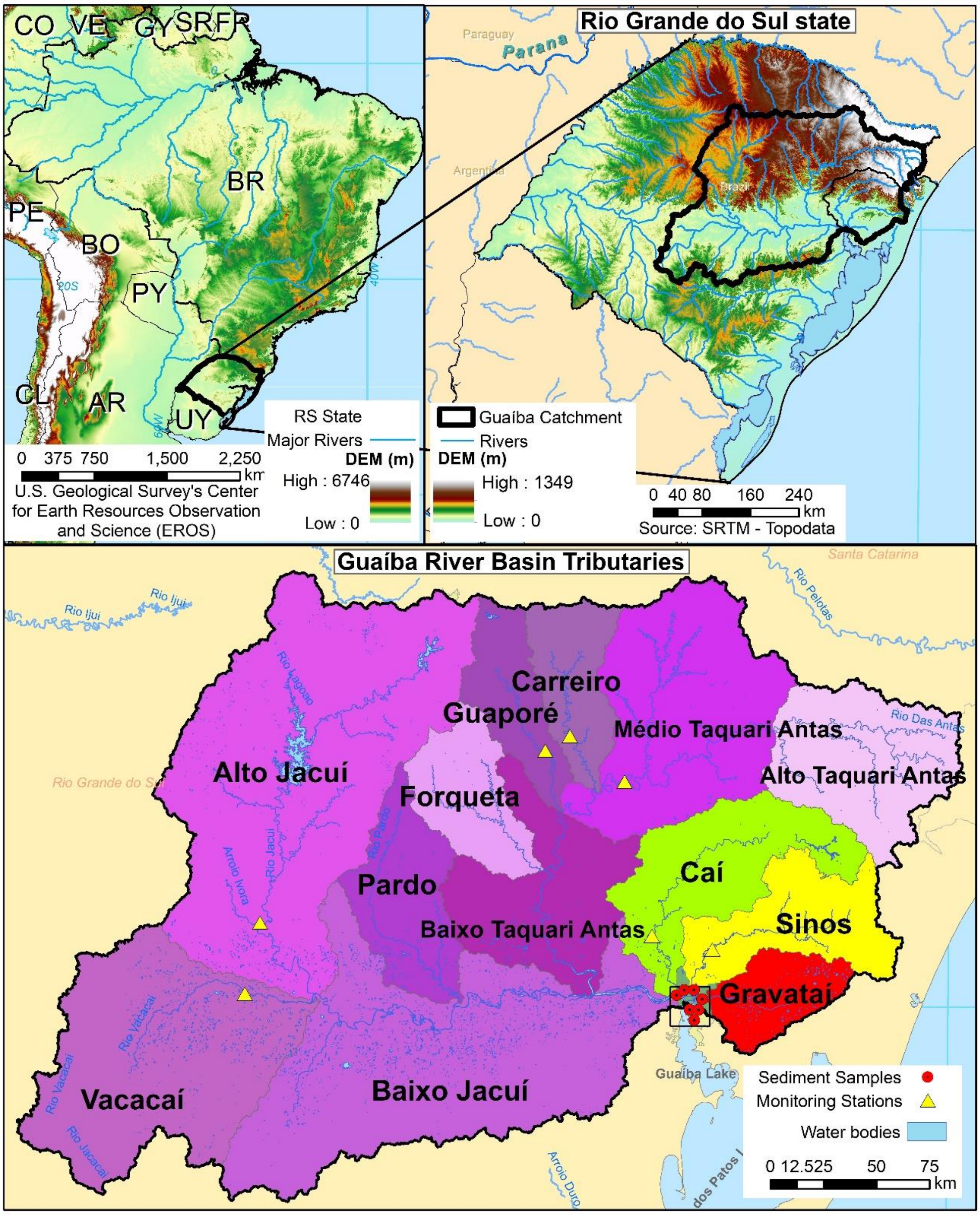

Fig. 1.

Location of Lake Guaíba and its basin in South America, Digital Elevation Model (DEM) of the region and delineation of the four main tributary catchments (Jacuí River, Caí River, dos Sinos River, and Gravataí River).

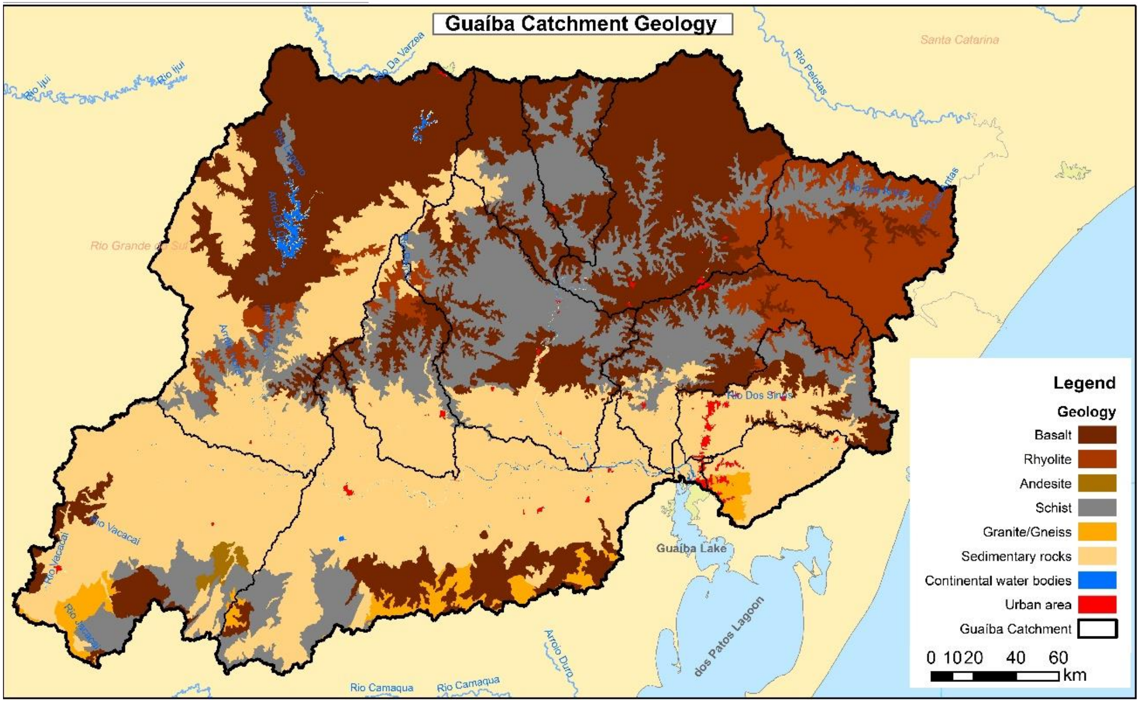

Fig. 2.
Geology of the main four tributary catchments draining into Lake Guaíba (Jacuí River, Caí River, dos Sinos River, and Gravataí River).

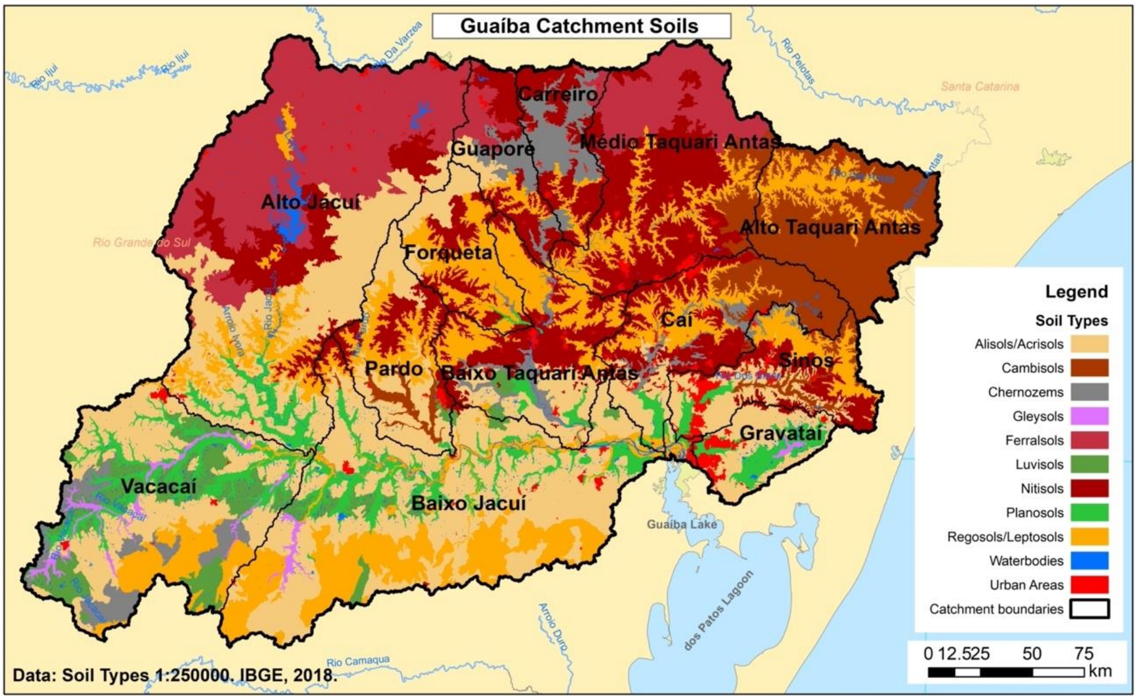

Fig. 3.
Distribution of soil types in the Lake Guaíba basin.

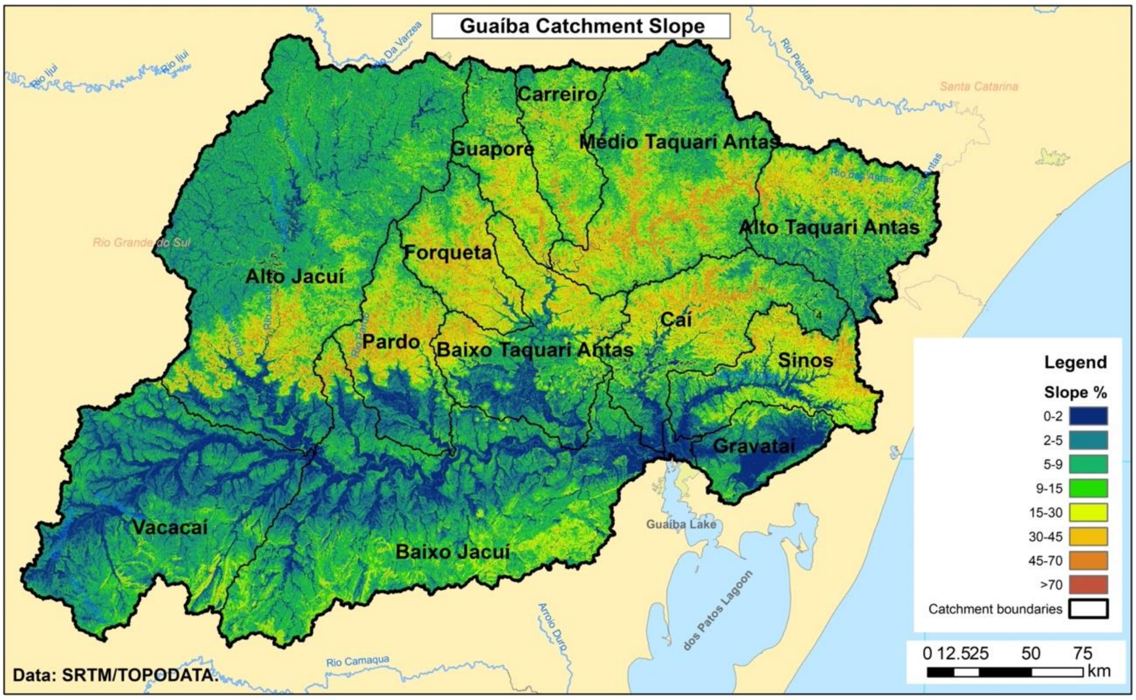

Fig. 4.
Slope distribution in the Lake Guaíba basin.

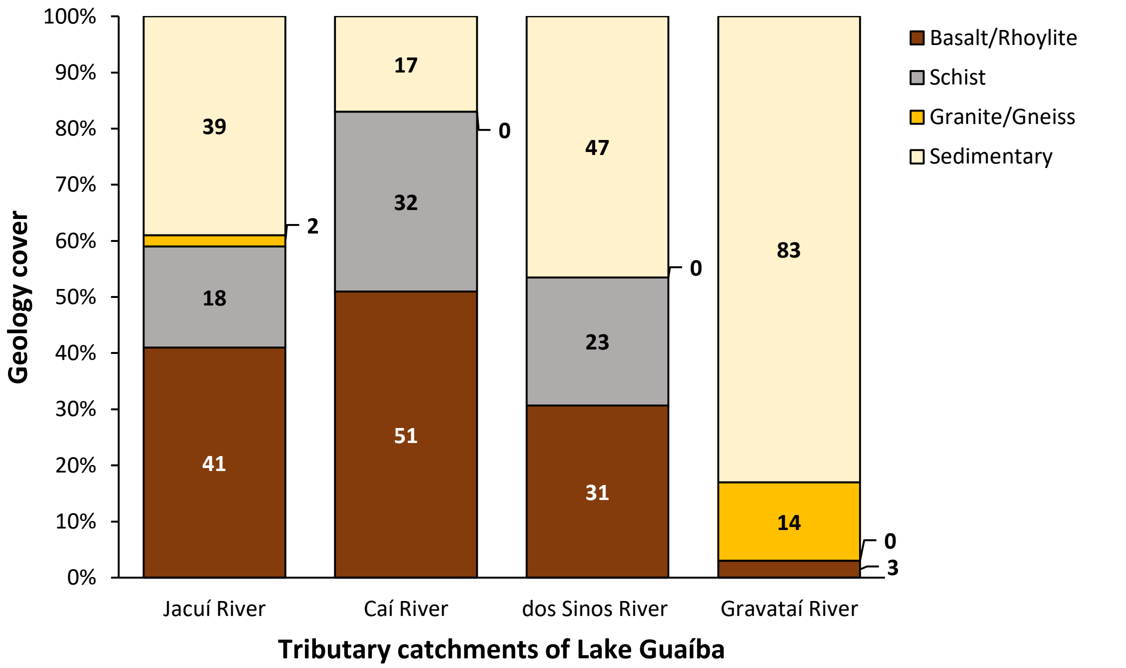

Fig. 5.
Proportion of the main types of lithology in the four main tributary catchments draining into Lake Guaíba (Jacuí River, Caí River, dos Sinos River, and Gravataí River).

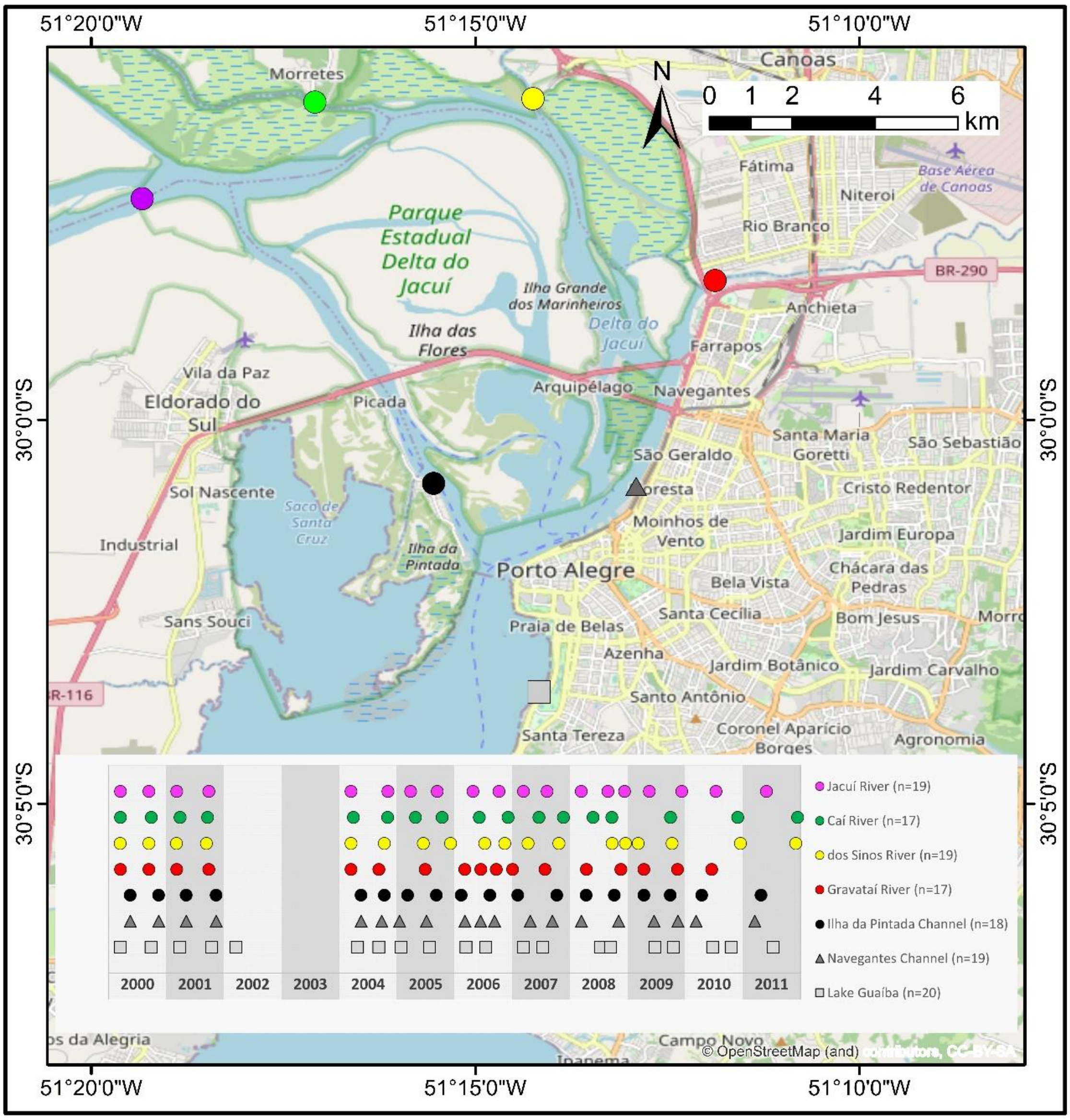

Fig. 6.

Spatial and temporal distribution of the sediment sampling in the four main tributaries of Lake Guaíba and in the Jacuí Delta.

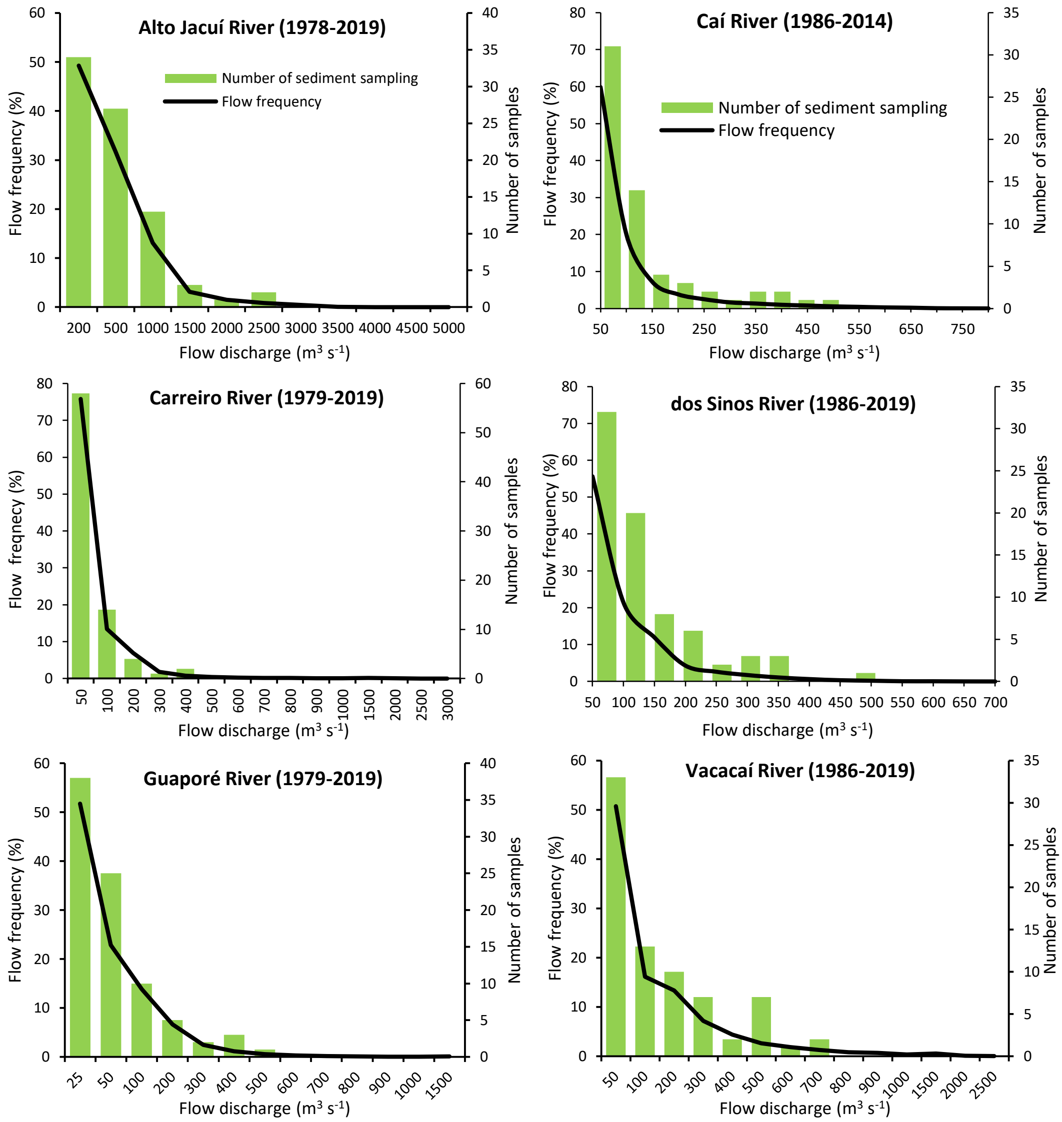

Fig. 7.
Flow frequency analysis with the number of sediment samples used for the suspended sediment concentration analysis in the tributaries draining into Lake Guaíba.

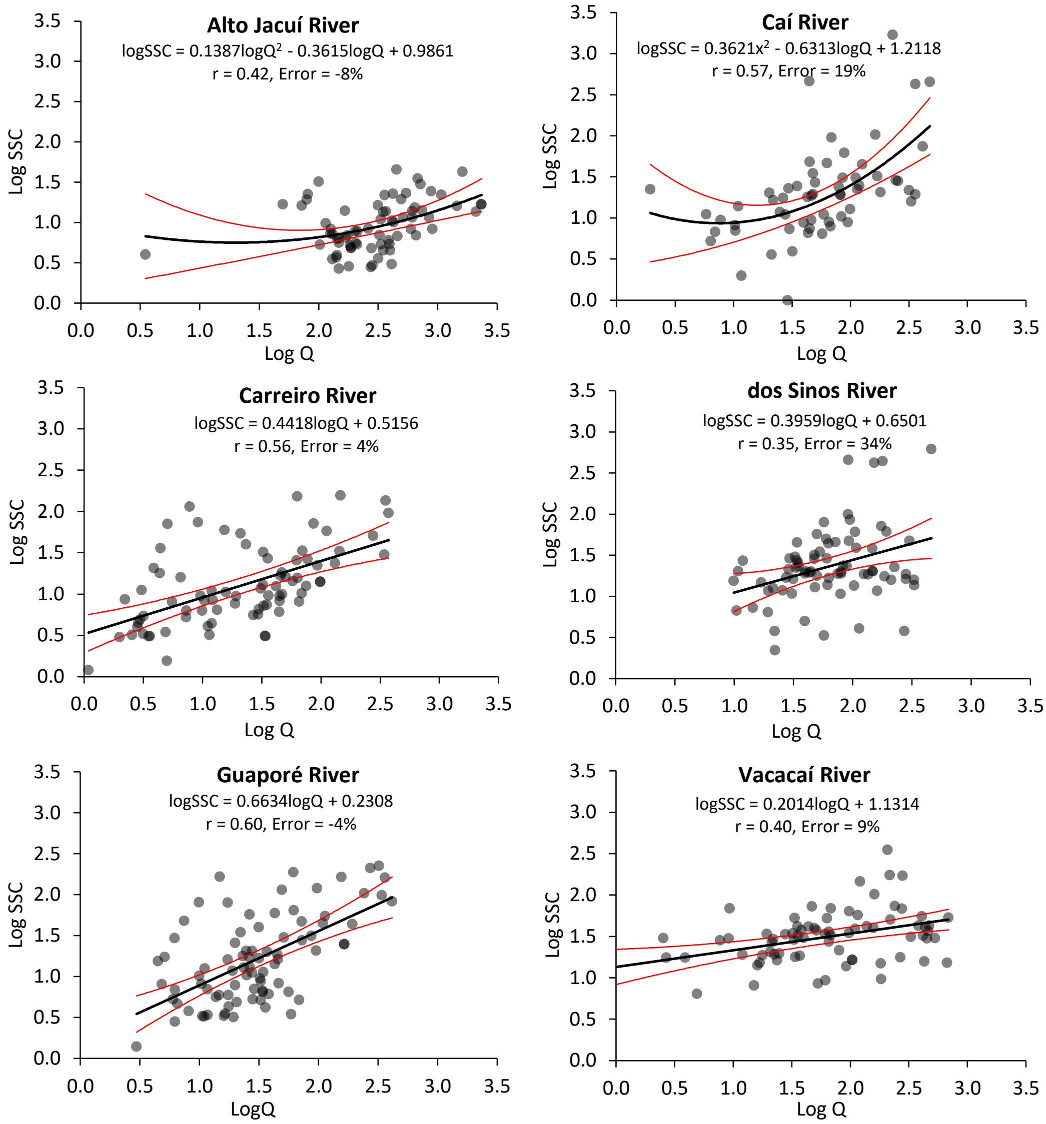

Fig. 8.

Rating curve and their associated regression equations of suspended sediment concentration (SCC, mg L^{-1}) and flow discharge (Q, $m^3 s^{-1}$) for different tributaries of Lake Guaíba. Black line is the equation and the red lines indicate the 95% confidence interval.

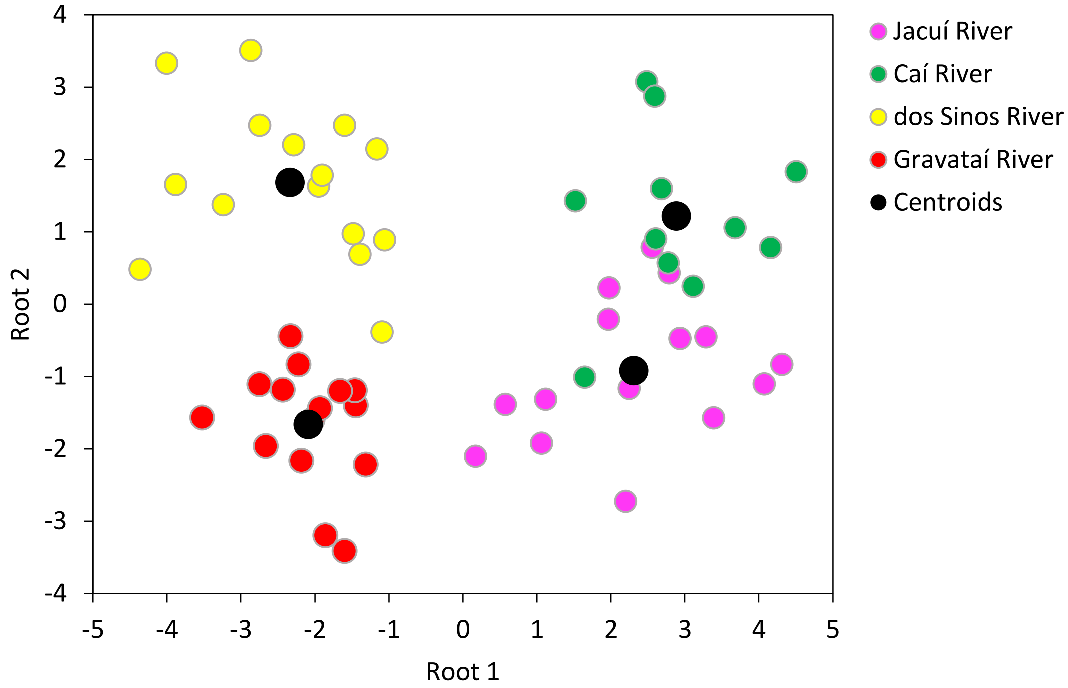

Fig. 9.
Two-dimensional scatterplots of the first and second discriminant functions provided by the stepwise discriminant function analysis (DFA).

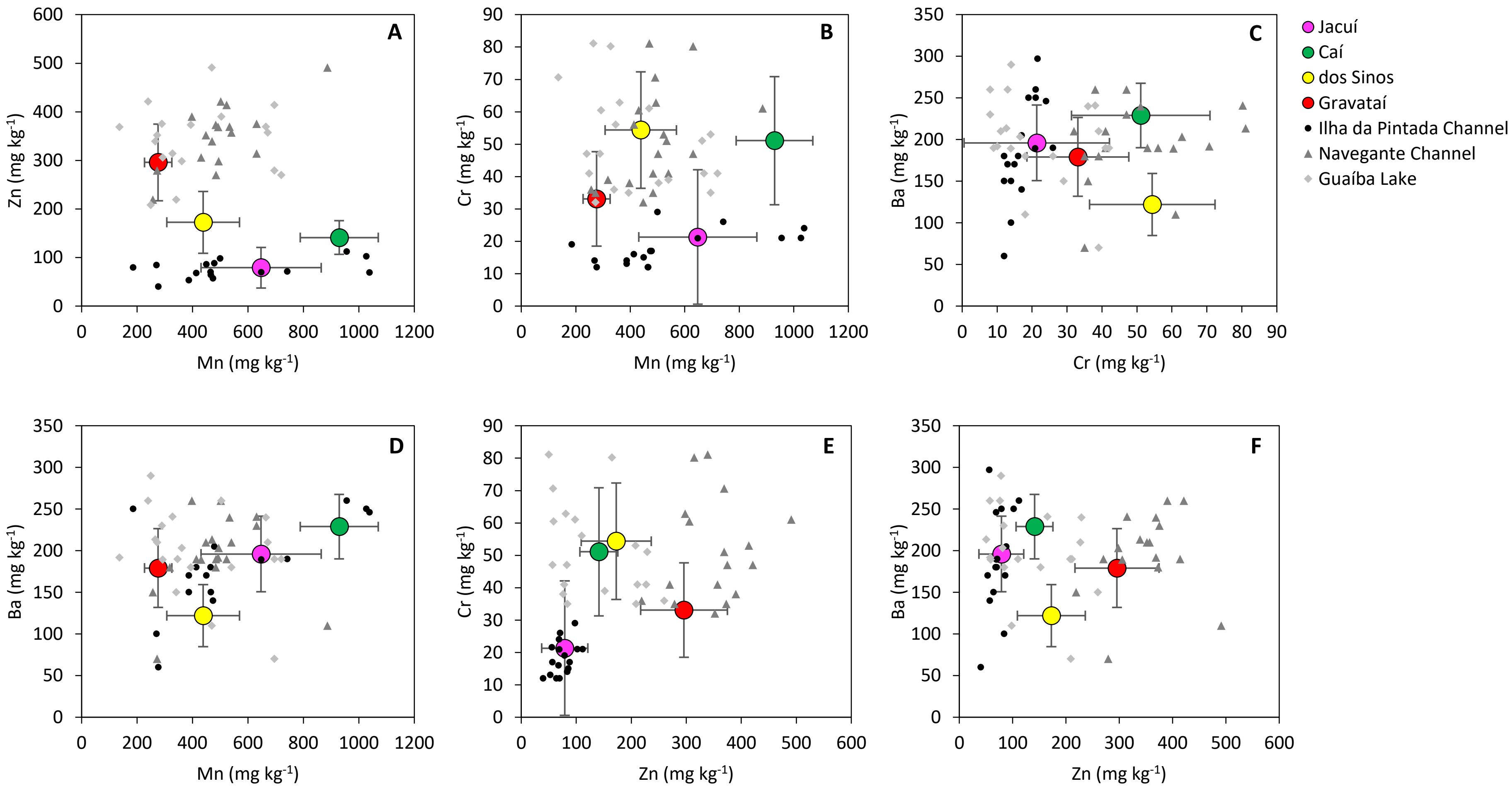

Fig. 10.
Relationships between the concentration of chemical elements selected as chemical tracers of the source of sediments in Jacuí Delta. Whiskers indicate the standard deviation of the concentration of each chemical element in each tributary draining into Lake Guaíba.

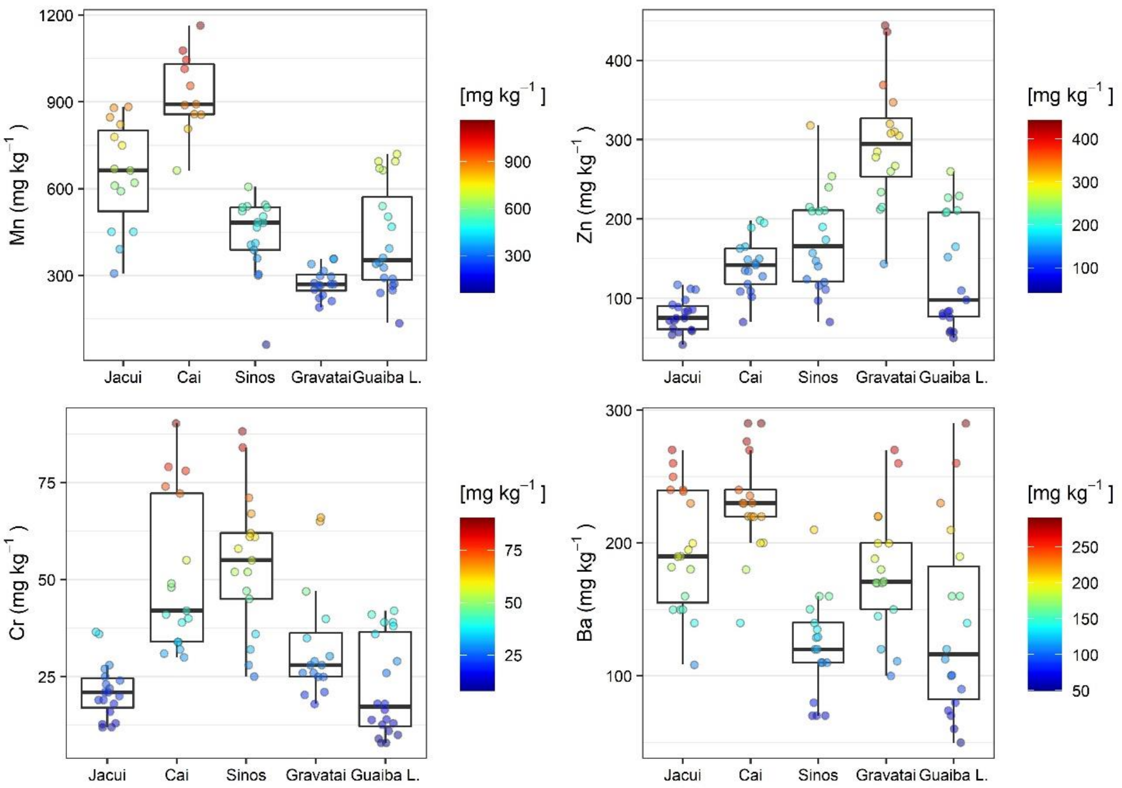

Fig. 11.

Concentrations in Mn, Zn, Cr and Ba analysed in sediment collected from 2000 to 2011 in the four main tributaries and in Lake Guaíba.

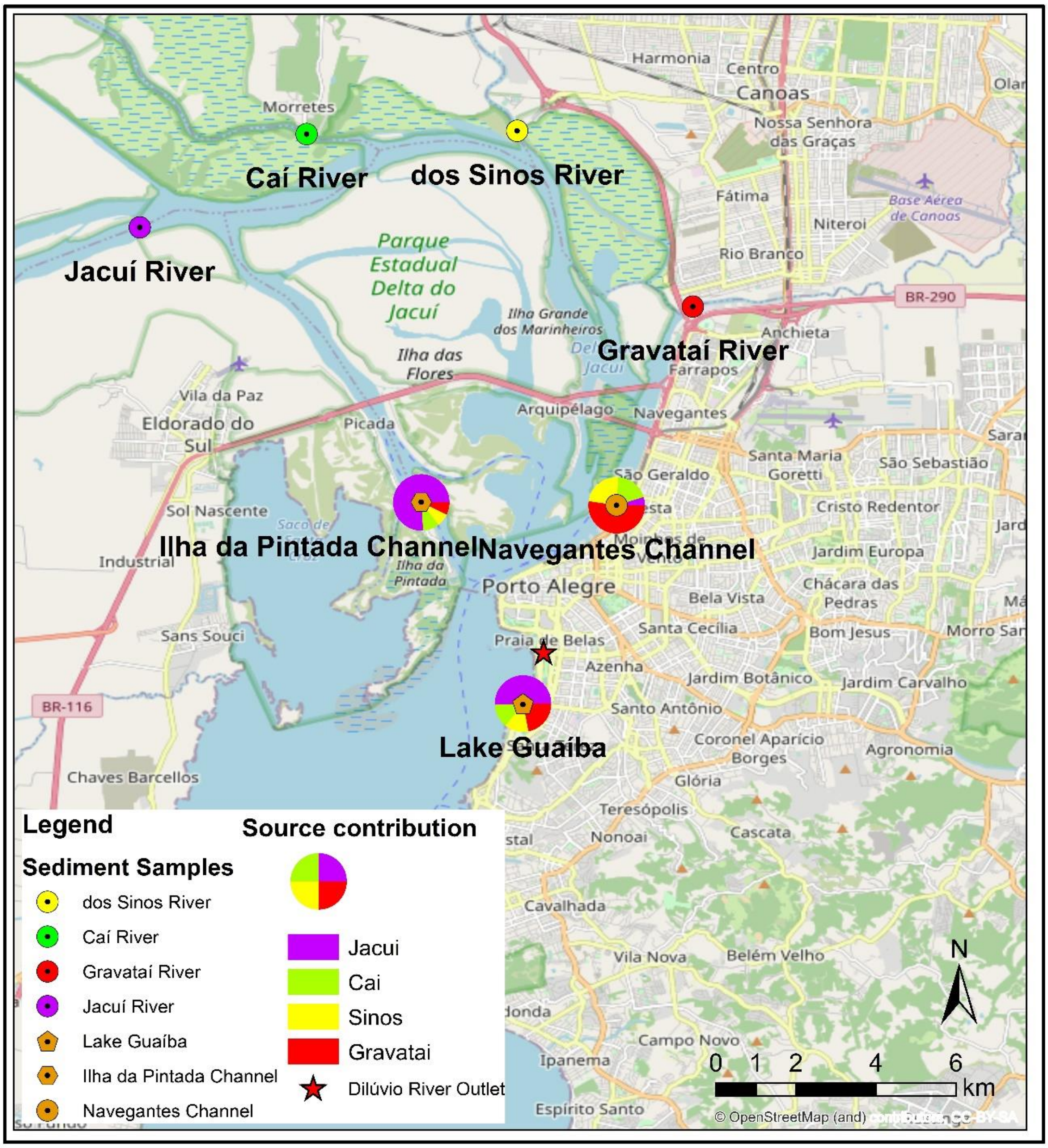

Fig. 12.

Sediment sampling sites used to characterize the sediment output from the four main tributaries of Lake Guaíba (Jacuí, Caí, dos Sinos and Gravataí Rivers), and summary of tributary contributions to sediment at three different sediment sampling sites in the Lake Guaíba delta (Ilha da Pintada Channel, Navegantes Channel, and Lake Guaíba). Results correspond to the average calculated for the entire period between 2000 to 2011.

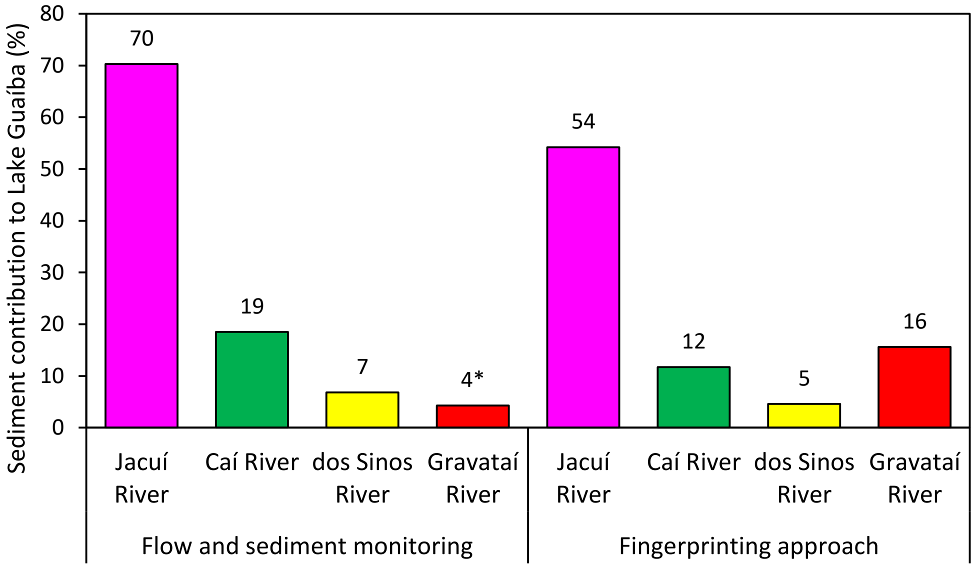

Fig. 13.
Comparison of tributary contributions to sediment of Lake Guaíba based on flow/sediment monitoring and sediment fingerprinting approaches for the period comprised between 2000 to 2011. *As there were no data available to estimate the contribution of sediment from the Gravataí River using the river flow and sediment monitoring approach, the contribution was estimated using the monitoring data available from the dos Sinos River.

Tables

Table 1.

Characteristics of the main tributaries of Lake Guaíba.

Tributary[1]	**Gravataí River**	**Sinos River**	**Caí River**	**Jacuí River**	**Sum of four tributaries**
Area[1] (km^2)	1,977	3,667	5,004	71,518	82,166
Mean annual rainfall[1] (mm)	1,700	1,350	1,150	1,400	-
Average altitude[1] (m)	400	600	1,000	730	-
Contribution to water discharge into Lake Guaíba [2] (%)	1.6	4.7	5.3	86.3	97.9
Proportion of the area of Lake Guaíba catchment (%)	2.3	4.3	5.9	84.0	96.5
Main soil types [3]	Acrisols Gleysols Planosols	Acrisols Planosols	Acrisols Nitisols Plinthosols	Acrisols Ferralsos Planosols	-
Land use[4]					
Cropland (%)	32	12	16	44	-
Grassland/pasture (%)	34	21	20	22	-
Forest (%)	22	59	61	32	-
Water bodies (%)	1.7	0.2	0.6	1.3	-
Urban (%)	9.4	7.5	2.5	0.9	-

[1] Fundação Estadual de Proteção Ambiental Henrique Luis Roessler – FEPAM. 30-year historical average.

[2] Plano da Bacia Hidrográfica do Lago Guaíba. The remaining 2.1% of water discharge originates from the small streams flowing directly into Lake Guaíba from its immediate vicinity.

[3] Soil map EMATER/DIT. Department of Soil Science of the Universidade Federal do Rio Grande do Sul.

[4] Souza et al. (2020).

Table 2.
Data sources and strategy for processing discharge and sediment concentration data for calculating the sediment inputs from the main tributaries of Lake Guaíba.

Lake Guaíba tributaries	Jacuí River catchment	Taquari-Antas sub-catchment	Proportion of the total area (%)	Total area (km^2)	Area covered by hydrological monitoring (km^2)	Area covered by hydrological monitoring (%)	Data source
Gravataí River			2.3	1,977	0	0.0	Not available
Caí River			5.9	5,004	4,360	5.1	ANA monitoring gauge station nº 87270000
Sinos River			4.3	3,667	3,130	3.7	ANA monitoring gauge station nº 87382000
Jacuí River			84.0	**71,518**	37,011	43.5	
	Alto Jacuí		20.2	17,230	13,998	16.4	ANA monitoring gauge station nº 85400000
	Vacacaí		11.8	10,010	6,754	7.9	ANA monitoring gauge station nº 85600000
	Pardo		4.3	3,623	0	0.0	Estimated[1]
	Baixo Jacuí		16.7	14,225	0	0.0	Estimated[1]
	Taquari-Antas		31.0	**26,400**	16,259	19.1	
		Carreiro	3.0	2,563	1,817	2.1	ANA monitoring gauge station - 86500000
		Guaporé	24.1	20,489	2,043	2.4	ANA monitoring gauge station - 86560000
		Alto e Médio Taquari-Antas	15.7	13,383	12,399	14.6	CERAM - Antas River bridge
		Baixo Taquari-Antas	6.0	5,125	0	0.0	Estimated[2]
		Forqueta	3.3	2,840	0	0.0	Estimated[2]
Total area of the four main tributaries			96.5	**82,166**	44,501	52.3	
Jacuí Delta			0.03	**28**			
Guaíba Lake			0.6	**482**			
Other small streams			2.9	**2,463**			
Total			100.00	**85,139**			

[1] Estimated by the authors using monitoring data from the Vacacaí and Alto Jacuí sub-catchment.
[2] Estimated by the authors using monitoring data from the Carreiro, Guaporé, Alto e Médio Taquari-Antas sub-catchment.

Table 3.
Contribution of suspended sediment load from each tributary to Lake Guaíba between 2000 and 2011 as estimated by the flow and sediment monitoring.

Guaíba Lake tributaries	**Area**	**Suspended sediment load for the period between 2000-2011****		
	km^2	10^6 Mg	%	Mg km^{-2} year^{-1}
Jacuí River	71,518	16.9	70	20
Caí River	5,004	5.1	19	85
Sinos River	3,667	1.9	7	44
Gravataí River*	1,977	1.0	4	44
Total		27.56	100	-

* Estimated based on the suspended sediment load from the Sinos River.
** Although the contribution values add up to 100%, this estimate was based on a discharge and suspended sediment monitoring coverage of only 52.3% of the total catchment area of Lake Guaíba (see Table 2). Furthermore, it only takes into account the four main tributaries and does not consider the contribution of small streams flowing directly into the Lake Guaíba (2.9% of the catchment area of Lake Guaíba - Table 2).

Table 4.
Metal concentrations in sediment samples collected in the four main tributaries of Lake Guaíba and in three sites across the Jacuí Delta, and outputs of the Kruskal-Wallis test and interquartile range test.

Tracer property	Lake Guaíba tributary				Kruskal-Wallis test		Jacuí Delta			Interquartile range test
	Jacuí River	Caí River	Sinos River	Gravataí River	H-value	p-value	Ilha da Pintada Channel	Navegantes Channel	Lake Guaíba	
Al (g kg^{-1})	33.87	47.68	30.36	45.87	16.6	0.0008	31.49	53.86	44.51	Passed
Fe (g kg^{-1})	36.11	52.52	30.58	28.57	25.3	<0.0001	32.22	39.98	34.43	Passed
Mn (mg kg^{-1})	647.66	929.19	438.56	276.40	44.7	<0.0001	539.26	484.62	423.59	Passed
Ba (mg kg^{-1})	196.03	228.96	121.95	179.13	31.4	<0.0001	187.48	195.18	138.71	Passed
Zn (mg kg^{-1})	78.96	141.13	172.53	295.85	49.4	<0.0001	74.49	349.16	131.29	Passed
Cu (mg kg^{-1})	51.98	65.48	43.22	64.33	12.2	0.0069	39.19	109.02	41.01	Passed
Pb (mg kg^{-1})	20.66	29.93	19.74	50.01	25.4	0.0069	24.75	63.89	26.14	Passed
Cr (mg kg^{-1})	21.33	51.08	54.37	33.09	38.5	0.0069	18.02	50.91	22.10	Passed
Ni (mg kg^{-1})	22.63	42.18	26.15	22.76	23.5	0.0069	20.75	36.75	18.89	Passed
Cd (mg kg^{-1})	0.25	0.29	0.20	0.22	3.5	0.3206	0.23	0.25	0.21	Passed
Hg (mg kg^{-1})	0.05	0.08	0.16	0.16	47.8	0.0069	0.06	0.42	0.12	Failed

Table 5.
Output of the linear discriminant function analysis.

DFA parameters	
Wilks' lambda	0.015
Variance due to differences between sources (%)	98.5
Selected tracers	Zn
	Cr
	Mn
	Ba
River sediment samples correctly classified (%)	
Jacuí River	88.9
Caí River	87.5
Sinos River	92.3
Gravataí River	100.0
Total	92.3
Squared Mahalanobis distances	
Jacuí vs. Caí	5.7
Jacuí vs. Sinos	39.1
Jacuí vs. Gravataí	69.3
Caí vs. Sinos	36.8
Caí vs. Gravataí	79.2
Sinos vs. Gravataí	19.4
p-levels	
Jacuí vs. Caí	0.0017
Jacuí vs. Sinos	<0.0001
Jacuí vs. Gravataí	<0.0001
Caí vs. Sinos	<0.0001
Caí vs. Gravataí	<0.0001
Sinos vs. Gravataí	<0.0001