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Modeling flow-related phosphorus inputs to tropical semiarid reservoirs

Maria de Jesus Delmiro Rocha, Iran Eduardo Lima Neto *

Department of Hydraulic and Environmental Engineering, Federal University of Ceará, Block 713 – 1st Floor – Center of Technology, Fortaleza, Ceará, Brazil

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ABSTRACT

Hydrological data and total phosphorus (TP) concentration at reservoirs' outlet were combined in a transient complete-mix model to obtain mean input loads and inlet concentration-flow relationships. This approach was designed to investigate the issue of phosphorus pollution in semiarid regions with intermittent rivers. The methodology was applied for twenty reservoirs in the State of Ceará, Brazilian semiarid. The modeled TP loads correlated well ($R^2 = 0.74$) with reference loads estimated from environmental inventories, with only 10% of underestimated results. The average input loads per unit area of the catchments ranged from about 4 to 40 kg km^{-2} yr^{-1}, which were considerably lower than the national average of about 500 kg km^{-2} yr^{-1}. This was attributed to lower precipitation indexes, intermittent river regime and a high-density reservoir network, peculiar of the Brazilian semiarid. Meanwhile, the input load per unit area of a small and highly populated urban catchment, with higher precipitation indexes and deficient sanitation was substantially higher (2626 kg km^{-2} yr^{-1}). Moreover, the fitted TP concentration-flow relationships directly reflected different TP input sources: strong u-shaped behavior marked the curves of highly non-point source dominated catchments, whereas a dilution pattern prevailed in those with significant point source inputs. The model validation with measured riverine TP concentration reached a NSE of 0.63. However, peak values in TP concentration during low flow rates sensitively affected the fitting of the models. In spite of non-point source dominance in the catchments, some relationships presented a slight signal of this use type. The variation range of the fitting parameters in comparison with other studies, as well the expected behavior of the curves in light of land use characteristics, strongly support the methodology applied in this study. The proposed approach will potentially help address the TP issue in tropical semiarid regions. Furthermore, the paper presents a simple way to deal with the challenging lack of monitored data in such environments.

1. Introduction

Phosphorus is a critical nutrient with regards to primary productivity in water bodies (Ansari and Gill, 2014; Le Moal et al., 2019). Identifying the main contribution sources of total phosphorus (TP) load is a first step to understand relevant P inputs to rivers and reservoirs, and guides water quality management strategies (Sharpley, 2016; Lun et al., 2018). However, the relation between catchment characteristics and TP concentration remains poorly understood in regions such as the Brazilian semiarid, where many reservoirs are failing to meet national water quality standards (Pacheco and Lima Neto, 2017; Lima et al., 2018; Mesquita et al., 2020; Lira et al., 2020; Wiegand et al., 2021). An additional challenge comes from the high-density reservoir network (one per about 5 km^2) and mostly intermittent rivers (Lima Neto et al., 2011; Campos et al., 2016; Mamede et al., 2012, 2018).

A catchment-based approach is required to understand the delivery pathways of TP sources entering rivers, lakes and reservoirs and to further address P-related water quality problems (Chen et al., 2015b; Krasa et al., 2019). Natural and anthropogenic TP point sources (PS) and non-point sources (NPS) have fundamental differences, distinct transfer pathways and contrasting relationships with flow rates (Bowes et al., 2008). Nutrient wash-off in runoff, erosion processes and river bed remobilization can increase TP concentration in water bodies located in NPS dominated catchments (Jarvie et al., 2010; Moura et al., 2020). Otherwise, high TP concentrations during low flow rate indicate PS dominated catchments (Jarvie et al., 2013; Sharpley et al., 2013). There are also more complicated catchments that contribute significantly with the two types of sources (Greene et al., 2011).

The absence or scarcity of water quality data, however, imposes serious difficulties for TP load estimation (Johnes, 2007; Chen et al., 2015a). Hence, nutrient balance in a catchment becomes an available option (Han et al., 2018). Physical-based and empirical models are

* Corresponding author.
E-mail address: iran@deha.ufc.br (I.E. Lima Neto).

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widely applied for TP load estimation and simulation with good accuracy (Mockler et al., 2017). However, physical-based approaches are of difficult implementation in data-scarce regions (Bowes et al., 2009), and then empirical models may be an alternative with acceptable precision (Rattan et al., 2017). The Export Coefficient Modeling (ECM) approach is an empirical model widely applied for P load estimation in the State of Ceará, Brazil (Ceará, 2020b) and other regions world-wide (Johnes, 1996; Ding et al., 2010; Matias and Johnes, 2012; Delkash and Al-faraj, 2014; Greene et al., 2015; Tsakiris and Alexakis, 2015). The combination of the Loss Coefficient Method (LCM) with the Nutrient Losses Empirical Model (NLEM) is another approach based on export coefficients to estimate TP load (Zhang et al., 2020). Meantime, it should be noted that indirect methodologies such as calibration processes in a coupled reservoir-catchment approach remain little explored as a viable alternative.

Vollenweider (1968) started pioneer studies on TP modeling in lakes by proposing a mass-balance model applied to well-mixed lakes, allowing transient analysis when multi-year TP concentration and flow data are available. The model's predictive ability was tested over the past decades and additional improvements were performed in recent studies (Chapra and Dolan, 2012; Shimoda and Arhonditsis, 2015; Chapra et al., 2016; Katsev, 2017; Araújo et al., 2019; Lira et al., 2020). Additionally, for the Brazilian semiarid, the occurrence of a diurnal mixing cycle with weak stratification patterns turns plausible a complete-mix hypothesis (Lima Neto, 2019). Also, for data-scarce regions, this parsimonious model stands out as a feasible solution.

Estimating the catchment production of P load in regions such as the Brazilian semiarid can contribute to understand the patterns of P input to water supply reservoirs (Chaves et al., 2013; Wu et al., 2016; Paula Filho et al., 2019). Subsequently, the development of concentration-flow relationships. This approach is of particular interest in regions with intermittent rivers (von Schiller et al., 2017; Chaves et al., 2019), where reservoir related issues are of great concern for water quality management (Lopes et al., 2014; Lacerda et al., 2018; Wiegand et al., 2021).

The nutrient-flow relationship is described by a power-law function that can be applied to a dataset comprised of paired TP concentration and inflow measurements (Bowes et al., 2008, 2009). This methodology has been successfully applied to perennials rivers throughout the globe (Bowes et al., 2009b, 2010, 2014, 2010; Bieroza and Heathwaite, 2015). The coefficients of the flow-nutrient model contribute to the understanding of dominant P inputs (Bowes et al., 2008; Chen et al., 2015a), as the model stands on fundamental hydrological differences between PS and NPS dominated catchments. The application of this model covering paired mean TP concentration and discharge at reservoir inlet is still a new approach to be verified for semiarid environments.

Thus, the objective of this study was to evaluate and discuss the main external TP input sources to reservoirs from Brazilian semiarid catchments and develop concentration-flow relationships for their intermittent rivers. The specific goals are: (i) to provide a methodology to estimate and evaluate average TP load and concentration at reservoirs' inlet through a mass-balance approach, (ii) to extend the application of TP-discharge relations to regions of intermittent rivers, (iii) to improve the understanding of flow conditions on TP concentration at reservoirs' inlet, and (iv) to provide a tool to address the TP issue in water bodies in data scarce regions.

2. Material and methods

2.1. Study area

As depicted in Fig. 1, the study includes twenty reservoirs located in the State of Ceará, Brazilian semiarid, monitored by the Water Resources Management Company of the State of Ceará (COGERH). Their capacities range from 0.3 to 6700 hm^3 and their catchments present different TP sources. The smallest reservoir, called Santo Anastácio, is located in the capital of the State, closer to the coastal zone. The main water uses of these reservoirs are human supply, aquaculture and irrigation. According to COGERH, fish farming is practiced in 40% of these reservoirs (CEARÁ, 2020b). Regarding the ecological protection of these ecosystems, specifically fauna and flora, the adverse climatic conditions imposed upon them, the priority use given to human supply and the incipient regulatory measures in this field, make them self-managed. In extreme drought periods, many reach their lowest volume or a complete dryness, destroying almost all life in the ecosystem. When they are very polluted, eutrophication triggers an intense proliferation of aquatic macrophytes. The reservoirs with a relevant practice of aquaculture are those that receive more attention in mitigating the potential impacts of volume and water quality changes, mostly due to economic losses. Thus, these management issues regarding life in the ecosystems still need thorough attention.

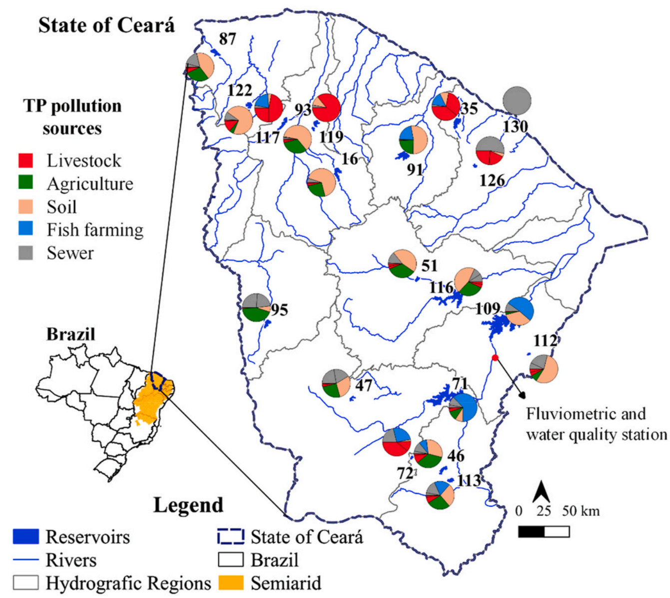

Fig. 1. Study sites location in the State of Ceará – Brazil highlighting the different TP sources of pollution of the catchments. Catchment and reservoir details presented in Table 1.

Table 1 summarizes important data of the catchments and reservoirs. The total catchment area of these reservoirs reaches 89,296 km^2, which represents about 60% of the state's area. These catchments are characterized by mild slopes varying normally from 0 to 15°. Only the catchments of the reservoirs Arrebita e Angicos present steeper slopes of 30 and 75°, respectively. The soils are not deep as they are located in the crystalline geological formation and the most representative soil types are the red-yellow argisol, litolic neosol and chromic luvisol (Jacomine et al., 1973; Araújo and Medeiros, 2013). These conditions favor the inexistence of perennial rivers and practically no base flow. The precipitation indexes vary from about 600 $mm.yr^{-1}$, in the Flor do Campo reservoir catchment, to about 1400 $mm.yr^{-1}$, in the Santo Anastácio reservoir catchment. Details of the rainfall patterns are available in the supplementary material. As typical of semiarid regions, there are two marked periods: the rainy season and the dry season. The rivers are intermittent with flows primarily occurring during the rainy season. In a special situation, the Santo Anastácio reservoir receives an almost constant flow of untreated sewage through an urban drainage channel, even during the dry season (Fraga et al., 2020). More details of the physical characteristics of the catchments and their land uses can be found in CEARÁ (2020b).

High-quality agricultural land use with irrigated crop production can be found in the Jaguaribe river basin, which covers about 40% of the catchments of the studied reservoirs. However, subsistence agriculture and animal husbandry, such as cattle, pig and poultry farming, is

Table 1
Physical characteristics of the reservoirs, catchments and the period of available water quality data.

ID	Reservoir	Catchment area (km^2)[a]	(Inhab km^{-2})[b]	Reservoir capacitiy (hm^3)	Average depth (m)	Catchment average precipitation (mm yr^{-1})	Data period	Environmental inventory date[b]
16	Edson Queiroz	1778	24.1	254	15.5	687	2008–2020	2011
35	Sítios Novos	446	854	126	11.1	899	2008–2020	2008
46	Rosário	337	171	47.2	15.3	1036	2010–2020	2011
47	Rivaldo de Carvalho	318	220	20.1	9.0	657	2008–2020	2011
51	Quixeramobim	7005	10.3	7.9	10.6	705	2009–2020	2011
71	Orós	24,538	5.4	1940	26.3	728	2008–2020	2011
72	Olho D'água	72.0	485	19	13.0	948	2008–2020	2008
87	Itaúna	781	67.4	72.6	8.2	1176	2011–2020	2011
91	General Sampaio	1582	3.9	322	18.0	729	2008–2020	2011
93	Forquilha	191	91.9	50.1	9.1	726	2008–2020	2008
95	Flor do campo	663	71.4	105	5.4	615	2010–2020	2011
109	Castanhão	44,800	1.8	6700	38.2	836	2008–2020	2011
112	Canafístula	329	62.5	13.1	6.5	830	2008–2020	2011
113	Cachoeira	143	356	34.3	14.7	983	2008–2020	2011
116	Banabuiú	14,200	6.3	1601	20.5	671	2010–2020	2011
117	Ayres de Sousa	1102	209	96.8	19.1	1125	2010–2020	2009
119	Arrebita	79.0	255	18.5	9.0	801	2008–2020	2011
122	Angicos	286	122	56.1	12.2	1119	2008–2020	2011
126	Acarape do Meio	210	524	29.6	19.7	1292	2008–2020	2008
130	Santo Anastácio[c]	4.0	7204	0.3	5.0	1042	2013–2019	–

[a] CEARÁ (2020a).
[b] CEARÁ (2020b).
[c] Araújo et al. (2019); Mesquita et al. (2020).

extensively practiced by the dispersed rural population. In spite of being essentially rural, there are some cities in the catchments with population densities ranging from 1.8 to 7204 inhab.km^{-2}, as shown in Table 1. Note that the highest population density refers to a neighborhood of the city of Fortaleza, where the Santo Anastácio reservoir is located, the only with an urban catchment.

2.2. Reservoir modeling

2.2.1. Total phosphorus input load modeling

Eq. (1) presents the transient complete-mix model proposed by Vollenweider (1968) (Chapra, 2008). The model requires parameters related with water balance, morphometry and TP dynamics. Due to the elevated interannual and seasonal variations in the morphometric parameters of semiarid reservoirs, this model is particularly suitable. The required data to perform the modeling was provided by COGERH (Ceará, 2020a). For Santo Anastácio reservoir, the data was obtained from Araújo et al. (2019) and Mesquita et al. (2020).

$$TP_r(t) = TP_{r,0}.\exp(-(\frac{Q_s}{V}+k).t) + [W/(Q_s + kV)].[1-\exp(-(\frac{Q_s}{V}+k)\,t)] \quad (1)$$

in which $TP_r(t)$: TP concentration at reservoir outlet in a given time (kg m^{-3}); $TP_{r,0}$: Initial TP concentration (kg m^{-3}); t: Elapsed time (yr); V: Average reservoir volume for a specified period (m^3); W: TP load (kg yr^{-1}); Q_s: Released water (m^3 yr^{-1}); k: TP decay coefficient (yr^{-1}).

The application of Eq. (1) consisted of taking paired TP measurements made by COGERH during the rainy period. These measurements are generally collected four times a year, two of them during the rainy period and analyzed according to APHA (2005). They are usually measured in January or February and, then in May or June, encompassing the beginning and the end of the wet period. These measurements were classified according to date as $TP_{r,o}$ and TP_r (t) and the elapsed time was calculated. The volume (V), released flow (Q_s) and decay coefficient (k) are the required additional parameters to estimate the TP load. It is expected that, once the elapsed time between the measurements encompasses almost the entire rainy season, the estimations significantly account the load entering the water body through inflow. Ultimately, particularly for the Santo Anastácio reservoir, once there are available TP concentration and inflow data measured directly at the reservoir's inlet, the TP load was estimated from the product of these two variables.

2.2.2. Retention time (RT) and phosphorus decay coefficient (k)

In line with the assumed complete-mix hydraulic behavior, the real residence time was replaced by the theoretical hydraulic residence time (V/Q). V is volume and Q the inflow, both averages of the measurement elapsed time (Salas and Martino, 1991; Castellano et al., 2010). This assumption is considered consistent once Brazilian semiarid reservoirs present a very weak thermal stratification which breaks-up on a daily cycle (Lima Neto, 2019). Furthermore, for relatively well-mixed lakes, simpler approaches to obtain this parameter have demonstrated good accuracy (Pilotti et al., 2014).

The phosphorus decay coefficient (k) was calculated by applying Eq. (2), as function of the RT. This empirical correlation was adjusted and validated by Toné and Lima Neto (2020) for several Brazilian semiarid reservoirs and also validated by Lima (2016) and Araújo et al. (2019) for the Acarape do Meio and Santo Anastácio reservoirs, respectively. It is noteworthy that similar empirical relations were developed for temperate and tropical lakes by Vollenweider (1976) and Salas and Martino (1991), respectively. However, this particular correlation (Eq. (2)) intended to account the effect of higher water temperatures of the Brazilian water bodies (~30 °C) in comparison with temperate (~10 °C) and tropical lakes (~20 °C). This increased water temperature results in increased consumption rate by algae and decreased water viscosity, which favors faster TP sedimentation rates, as proposed by Castagnino (1982) and Toné and Lima Neto (2020).

$$k = 4/RT^{1/2} \quad (2)$$

2.2.3. Total phosphorus measurements and water balance data

The reservoirs were selected according to the availability of TP concentration and TP load information. Excepting for the Santo Anastácio reservoir, which measurements of TP_r and W were taken from Araújo et al. (2019) and Mesquita et al. (2020), the remaining 19 reservoirs integrate a continuous monitoring program by COGERH. Their catchments have an environmental inventory also prepared by COGERH with TP load data estimated by the ECM methodology (Ceará, 2020b). It accounts for the main TP source contributions in an annual timestep. The details regarding TP load calculation methods are available in CEARÁ (2020b).

The last required parameters were: reservoir's volume (V), reservoir inflow (Q), evaporation (E), and released flow (*Qs*). The volume V was obtained through daily water level measurements and level-volume curves provided by CEARÁ (2020a). The values of Q and *Qs* were obtained from COGERH on a monthly basis and then converted to a daily average. Finally, the evaporated volume E was calculated from the product of the average surface area of the reservoir by the evaporated height, available from the climatological station of each reservoir (Brasil, 2020). The reservoir surface area was also obtained from level-area curves provided by CEARÁ (2020a).

2.2.4. National standards of water-column TP concentration for water quality protection

The issues related with water quality, limiting nutrient concentration and classification of water bodies are regulated by the Brazilian National Environment Council (Brasil, 2005). The level of water quality in order to ensure the appropriate water uses is deliberated through the classification of the water bodies. A reservoir may be classified as Special Class and Classes I, II and III. The Special Class defines water bodies with a high water quality level destined for the preservation of aquatic environments and the natural balance of aquatic communities. Class I ensures safety for human primary contact, human consumption after conventional treatment and irrigation. Lastly, Classes II and III describe less noble usage and the human supply is allowed after advanced treatment. Each class poses limiting TP concentration. For lentic environments, the limits for classes I, II and III are 20, 30 and 50 μg L^{-1}, respectively. For lotic environments, the limits are 100 μg L^{-1} for classes I and II, and 150 μg L^{-1} for class III. The studied reservoirs and their tributaries are framed as Class II. Then, the measured and modeled TP concentrations were evaluated in accordance with these standards.

2.3. Riverine TP concentration

The TP concentration at the reservoirs' inlet (TP_{in}) was calculated through the ratio between the modeled loads (W) and the inflows (Q) for each period. Then, a dataset of paired TP_{in} and Q was constructed for each reservoir. Whenever available, measured riverine TP concentrations were also considered. It is noteworthy that the methodology applied in this work intends primarily to estimate the flow-related TP load during the rainy season (i.e., external load), not accounting for the internal TP contributions in the overall TP balance. Note that the high inflow rates of the wet period favor the assumed well-mixed behavior in the reservoirs and weaken the already subtle stratification patterns, potentially reducing the risk of internal load (Lima Neto, 2019; Rocha et al., 2020). The literature reports that the external load is more significant during the rainy season, especially in semiarid regions (Song et al., 2017; Cavalcante et al., 2018; Barbosa et al., 2019), while internal loading is more impactful in the dry period (Coppens et al., 2016). Moreover, since the studied catchments are mostly rural with high NPS dominance, one expect significant nutrient wash-off from external sources (Jiang et al., 2019). In addition, during the wet period, the water level is higher and wind velocity smaller (Jalil et al., 2019), reducing the effect of wind shear in deeper water layers and minimizing P resuspension from bottom sediments (Bai et al., 2020).

2.3.1. Flow-concentration model development

The flow-concentration approach is mainly useful in data-scarce regions for integration of water resources management and/or further calibration of more complex models. Once the relationship is satisfactorily calibrated, the empirically-fitted model is a feasible tool to analyze the catchment land use contribution to the water quality and predict TP concentration (Chen et al., 2015a). The TP_{in} concentration can be expressed as presented in Eq. (3), accounting for the contribution of PS pollution (first term) and NPS pollution (second term), as proposed by Bowes et al. (2008).

$$TP_{in} = A.Q^{B-1} + C.Q^{D-1} \tag{3}$$

The four fitting parameters A, B, C and D in Eq. (3) were calibrated using the non-linear least squares regression procedure PROC NLIN of the SAS statistical analysis package (SAS, 2018). The A and C coefficients have unit of load while B and D are dimensionless. Two constraints were applied to provide reliable solutions. First, it was assumed $0 \leq B \leq 1$, which implies that the TP concentration from PS-derived TP load will decrease with increasing river discharge (Q). Second, $D > 1$, as NPS-derived TP load and concentration will both increase with increasing Q (Bowes et al., 2008). Furthermore, parameters A and B were both constrained as greater than zero. Particular constraints such as a total PS dominance ($C = D = 0$) or NPS dominance ($A = B = 0$) were not considered as the studied catchments present significant proportions of the two main sources (CEARÁ, 2020b). The Nash–Sutcliffe Efficiency (NSE) was calculated to each fitted model as a performance indicator (Moriasi et al., 2007).

In order to analyze TP patterns depending on the inflow and catchment influence, the data was not grouped by seasonal periods. The intermittent regime of the semiarid rivers was another relevant aspect to avoid this grouped analysis. Furthermore, the discharge at which the estimated PS and NPS TP inputs are equal ($Q = Q_e$) was calculated using the coupled A, B, C, and D parameters according to Eq. (4). When $Q < Q_e$, PS inputs dominate the TP load as compared to NPS. Conversely, NPS inputs dominate the TP load when $Q > Q_e$.

$$Q_e = \left(\frac{A}{C}\right)^{\frac{1}{(D-B)}} \tag{4}$$

2.4. Application and validation of the flow-concentration model for the castanhão reservoir

The Castanhão reservoir, the largest in the State of Ceará, has fluviometric and water quality stations managed by COGERH located near its main entrance (see Fig. 1). Thus, particularly for this reservoir, a comparison between the daily inflow from the water balance and the fluviometric station was possible, for which the indicators NSE and R^2 were calculated. Furthermore, the flow-concentration model for the TP_{in} concentration was applied for the entire study period using the inflow data from the fluviometric station. Then, the modeled concentrations were compared with a subset of the measured values in the water quality station and evaluated by the same indicators (NSE and R^2).

2.5. Uncertainty analysis and validation

The fundamental data used to develop the nutrient-flow relationships were the TP load. However, the load estimation process discussed arise uncertainties mainly related with water quality and quantity data (Hollaway et al., 2018). Then, an uncertainty analysis along with the validation of the estimated loads were performed. The validation was possible through the evaluation of the R^2 and the bias between the mean modeled TP load and the reference load calculated by COGERH. Furthermore, as several TP loads were estimated throughout the studied period, the coefficient of variation was calculated for each catchment and an analysis of this statistics was carried out in light of available literature. Ultimately, reported mean error values for the ECM methodology was gathered to perform a comparison of the range of variation of the modeled TP load with the range of variation of the reference TP load.

3. Results and discussion

3.1. Catchment hydrology and water quality characterization

The volume variation, inflow regime, sampled TP_r and estimated TP_{in} concentration for the studied period are presented, respectively, in Fig. 2a–d, along with the limits of TP concentration according to national standards. The interannual variability of the volume is shown in Fig. 2a. One can see a wide range of volume variation among reservoirs, with maximum reductions spanning from about 50% to 5% of the maximum observed volumes. For instance, the volume of the Castanhão reduced from 5192 to 289 hm^3, while the volume of the Canafístula from 10.6 to 0.06 hm^3. The small reservoirs usually present an annual cycle due to their frequent "emptying process", while the larger ones sustain water for several years (Lopes et al., 2014; Lacerda et al., 2018). The volume of the Santo Anastácio is approximately constant (0.3 hm^3),

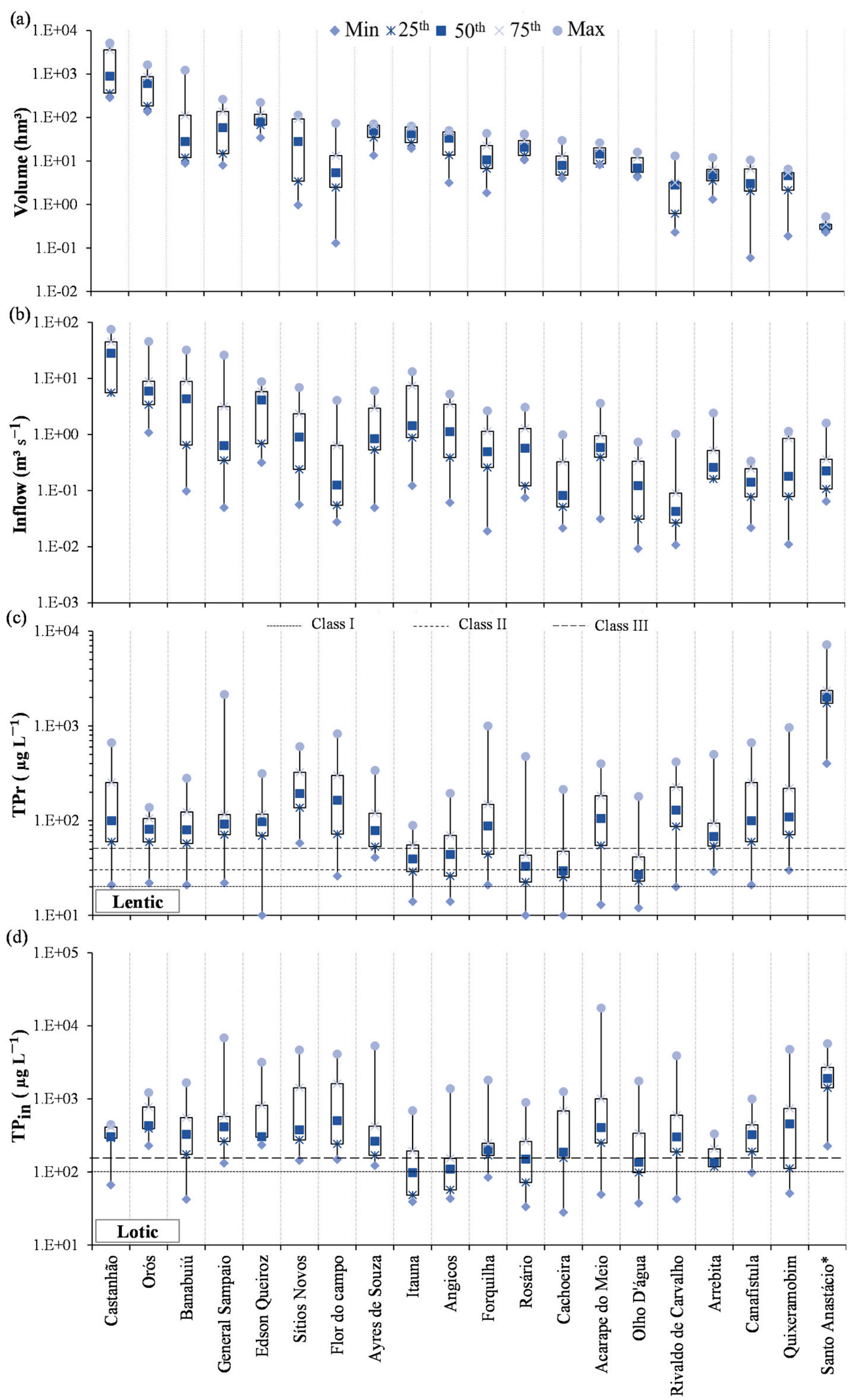

Fig. 2. Variation range of (a) volume, (b) Inflow, (c) measured TP_r (d) estimated TP_{in} along with the limits of TP concentration according to national standards. * Measurements from Araújo et al. (2019) and Mesquita et al. (2020).

since it receives continuously sewage from a drainage channel. In the wet season, though, the volume increases to about 0.5 hm^3 due to urban runoff (Fraga et al., 2020).

Fig. 2b shows a marked variability in the mean inflow to the reservoirs during the wet seasons of the study period. The reservoir Olho D'água presented the lowest inflow, from 0.01 to 0.7 $m^3 s^{-1}$, while the highest values were observed in the Castanhão reservoir, from 2.1 to 74.7 $m^3 s^{-1}$. The seasonal and interannual variabilities of volume and inflow are highly influenced by changes in precipitation levels, particularly in the State of Ceará (Broad et al., 2007). While a decline in the mean inflow is expected during dry periods in most regions (Zhang et al., 2015), due to the semiarid condition of the studied area, the inflow is completely interrupted during the dry season. Additionally, the high-density reservoir network strongly contributes to this inflow interruption in downstream reservoirs (Mamede et al., 2012, 2018). Conversely, the inflow to Santo Anastácio reservoir had a lower variation of 0.1–1.6 $m^3 s^{-1}$, even in the dry season.

The measured TP concentrations in the reservoirs (TP_r) are shown in Fig. 2c. The ranges were: 10–400 μg L^{-1} (minimum values), 27–2011 μg L^{-1} (median values) and 89–7181 μg L^{-1} (maximum values). The Orós and General Sampaio reservoirs presented the lowest (about 6 times) and highest (about 100 times) variations, respectively. The water quality deterioration (TP_r = 2145 μg L^{-1}) observed in the General Sampaio reservoir was associated with large NPS pollution due to the heavy precipitations in 2009, mainly considering its proximity to the city of General Sampaio (Chaves et al., 2013). Contrastingly, the values of TP_r in the Santo Anastácio reservoir ranged from 225 to 7181 μg L^{-1}, as a result of massive untreated sewage input from its drainage channel.

With regards to the accordance with Brazilian standards for water quality, most reservoirs had their concentrations over the limiting value of 30 μg L^{-1} for their classification (see Fig. 2c). The reservoirs which better performed were Rosário, Cachoeira and Olho D'água, with 46%, 56% and 52% of the TP_r measurements under this limit. The implications of exceeding these limits rely mainly on triggering eutrophication processes, for which TP is a critical nutrient (Le Moal et al., 2019), and intensifying the water quality degradation. The recovery of the water quality related to this nutrient has been a natural process with no measures taken by environmental regulatory agencies on record. TP_r is strongly affected by changes in hydrological parameters (Zhang et al., 2020). For the studied period, the lower the volume, the higher the concentration in the water column for all reservoirs, which is an expected behavior for this nutrient (Wu et al., 2016). Furthermore, the input TP load variability is another explaining factor for TP_r variation through time.

The different variations in the water quality of the reservoirs may have several causes related to the catchment and the reservoir itself, such as: seasonal patterns and variations in the flow regime (Rattan et al., 2017), different land uses (Lun et al., 2018), variable water retention time (Chaves et al., 2013) and significant water level variation (Sharpley et al., 2013). Previous studies in lakes and artificial reservoirs have found TP_r concentrations ranging from about 1 to 3270 μg L^{-1} (Matias and Johnes., 2012; Rattan et al., 2017; Han et al., 2018; Li et al., 2020), which are of the same order as those observed in the present study.

Ultimately, the TP concentration modeled at the reservoirs' inlet as an average of its tributaries (TP_{in}) is presented in Fig. 2d, along with the limiting concentrations. An overall description shows that the lowest value was 28 μg L^{-1} (Cachoeira) and the largest was 17,515 μg L^{-1} (Acarape do Meio), 2.56-fold higher than the second highest value of 6839 μg L^{-1} for the General Sampaio inlet waters. The maximum TP_{in} concentrations were considerably high during low flow period (0.001–0.2 $m^3 s^{-1}$). The 75th percentile, though, reached more usual values ranging from 154 to 2707 μg L^{-1}, while the flow varied from 0.09 to 45 $m^3 s^{-1}$. The median concentrations ranged from 98 to 1897 μg L^{-1}. As for the limiting concentrations, the TP_{in} of the reservoirs Santo Anastácio, Edson Queiroz and Orós resulted in all estimated values over the established limit for both classes. Angicos, Itaúna and Rosário, though, better met the standard, with 64%, 62% and 50% of the estimated values under the limit concentration for Class II, respectively. For the remaining, only the modeled values between the minimum and the 25th percentile fell below the reference concentration of 100 μg L^{-1}.

The estimated TP_{in} concentration values were very far from those reported by Chen et al. (2015a) (10–200 μg L^{-1}), Bowes et al. (2009) (400 μg L^{-1} in average), Bowes et al. (2015) (85–447 μg L^{-1}), Greene et al. (2011) (500 μg L^{-1} in average), and Mouri et al. (2011) (540 μg L^{-1} in average). In contrast, they were closer to those observed by Rattan et al. (2017) (55–1740 μg L^{-1}), Bowes et al. (2014) (20–2000 μg L^{-1}), Bieroza and Heathwaite (2015) (10–2000 μg L^{-1}), Bowes et al. (2010) (~0–4000 μg L^{-1}) and Bowes et al. (2008) (~0–8000 μg L^{-1}). Remarkably, TP_{in} values superior than 16,000 μg L^{-1} were found during low flow periods of a Mediterranean intermittent river with an arid catchment highly dominated by PS pollution (Perrin et al., 2018). These hydrologic regimes and climate condition are quite similar to the ones of the studied area as well as the high TP_{in} concentrations.

The major inflows of the reservoirs come from intermittent rivers, except for Santo Anastácio, whose inflow come from an urban drainage channel. For the rural reservoirs, higher TP_{in} values were obtained in the alternation of wet and dry phases, a low flow period denominated hydrological contraction (von Schiller et al., 2017), before the river complete dryness. This aspect might be associated with a lack of dilution of TP from point sources during low flow rates (Bowes et al., 2009). Contrastingly, the bioavailability of P compounds in the sediments of intermittent rivers on drying potentially contributes to eventual peak concentration during high flow events (von Schiller et al., 2017). In addition, aspects such as the enlargement of P delivery from land to water bodies through a high hydrologic connectivity (Chen et al., 2015b; Rattan et al., 2017), high concentrations of suspended algal material (Bowes et al., 2009), catchment urbanization Mouri et al. (2011) (Rattan et al., 2017); and the low water quality of the reservoir during the modeling period can contribute to high TP_{in} estimations.

3.2. Modeled TP load

The estimation of the TP load produced in the catchments was achieved by the application of Eq. (1). The variability presented in Fig. 3 results from changes in TP_r and water balance parameters for each modeled interval. The reservoirs with the largest variations in the estimated TP load were also those with the largest variability in their inflows, such as the General Sampaio and Acarape do Meio reservoirs (see Fig. 2b). The reference TP load values estimated by COGERH through the ECM approach is also shown in Fig. 3.

The estimated annual TP loads ranged from 0.03 t yr^{-1}, for the catchment of the Rivaldo de Carvalho reservoir, to 1164 t yr^{-1} for the Castanhão catchment. Considering only the Orós and Castanhão catchments, the median value was 214 t yr^{-1}, while including all 19 reservoirs managed by COGERH, the median drops to 7.2 t yr^{-1}, which turns these first two catchments the largest TP producers. It is interesting to stress, however, that even being much smaller, the Santo Anastácio reservoir receives annually an average TP load of about 16.6 t yr^{-1} (Araújo et al., 2019), which is twice higher than the median value for the above-mentioned reservoirs.

As can be seen in Fig. 3, the estimated TP loads were slightly under the reference values for 25% of the catchments, and more significantly underestimated for only 10% of them, represented by the catchments of the reservoirs Orós and Angicos. For 35% of the catchments, the reference load and the median estimated load were the closest, while, for the remaining, the reference value was somewhere between the range of modeled loads. The temporally spaced measured data might have contributed to the underestimation of the TP load. Low sampling frequency during the rainy season tends to underrepresent peak storm or high-flow events and the intense phosphorus dynamics right after (Johnes, 2007; Bowes et al., 2009). Besides, particularly for the larger

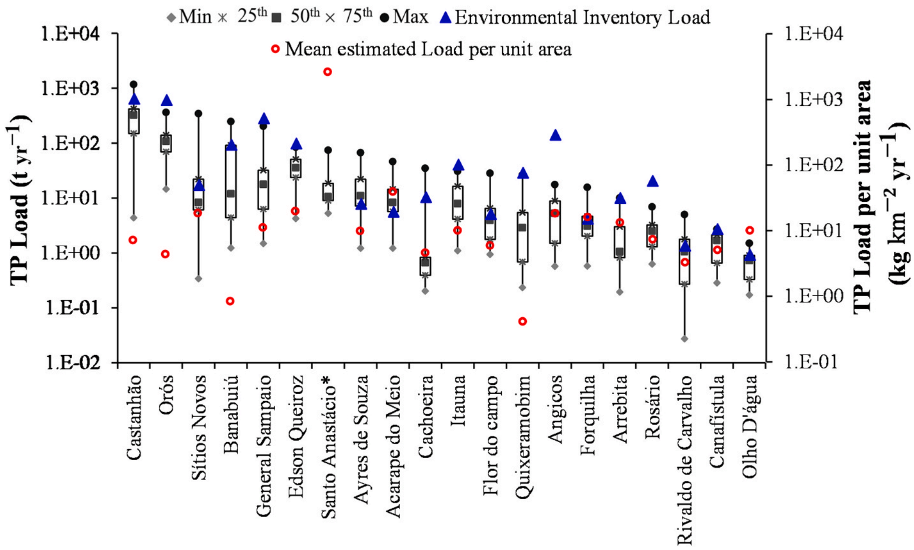

Fig. 3. Estimated TP load by mass balance and reference TP load by the ECM modelling approach. * Estimated TP load from measured data of TP_{in} and inflow (Araújo et al., 2019; Mesquita et al., 2020).

catchments, the high density of reservoirs also plays an important role in regulating upstream P retention (Hu et al., 2020). Bowes et al. (2009) evaluated the impact of sampling frequency influence on annual TP load estimation. Their estimation with monthly measurements produced a mean deviation of −21 to 35% from the one carried with weekly measurements. Therefore, improved load estimates are produced from more frequent sampling, which highlights the challenge of dealing with low data availability. In the meantime, as water quality or quantity data are not carried out frequently, modeling with the available data remains the possible option in data-scarce regions, such as the Brazilian semiarid.

The reconstruction of TP load through different modeling approaches was widely performed in the literature. Rattan et al. (2017) obtained values of 117 t yr^{-1} in Canada. Chen et al., (2015a) found TP loads around 357 t yr^{-1} in China. Zhang et al. (2020) obtained TP loads reaching peak values over 2700 t yr^{-1} in China. Mockler et al. (2017) assessed annual TP emissions for 16 major river catchments with values up to 3000 t yr^{-1}. Lastly, Han et al. (2018) evaluated the great Three Gorges Reservoir basin and estimated TP load of about 43,977 t yr^{-1}. In comparison, the studied catchments do not present a so intense TP load production (see Fig. 3). A plausible explanation may be the existence of a dense reservoir network, which potentially contribute to P retention, as observed for the sediment production (Lima Neto et al., 2011; Mamede et al., 2018).

The TP load production from NPS pollution is expected to be higher in larger catchments (Marques et al., 2019). However, the activities' intensity developed in the basin plays a more important role. Rural areas where predominate livestock and agriculture produce about four times more TP load than urban areas (Li et al., 2020). The catchments of Castanhão and Orós concentrate the most relevant irrigated croplands of the study area and the first one shows the highest amount of TP production (656 t yr^{-1}) (CEARÁ, 2020b). The TP load from fertilizers is another concern to be considered for the catchment of Orós, the second largest TP producer (624 t yr^{-1}). Agriculture and soil contribute with 80% of NPS pollution in this catchment. Indeed, cultivated lands can be a major secondary pollutant to surface waters (Li et al., 2020).

With respect to the among-catchment variation of the input load, specific anthropogenic uses of each catchment play an important role in this variability (Hong et al., 2012; Chen et al., 2015b; Hu et al., 2020), while the intervariation of TP load may be better explained by its relation with seasonal variations in rivers discharge (Han et al., 2018). Stream discharge fundamentally influences annual TP load along with the river's own nutrient retention capability (Torres et al., 2007). Also, hydrologic events (Jeznach et al., 2017), such as peak storms (Chen et al., 2012), mainly in NPS dominated. Besides, legacy P remobilization during slow flow paths may be taken into consideration as a relevant aspect (Sharpley et al., 2013).

The TP loads per unit area were also presented in Fig. 3 to support a comparison among catchments. Despite the high TP loads estimated for Orós and Castanhão catchments, their values per unit area were relatively low: 4.4 and 7.2 kg km^{-2} yr^{-1}, respectively. On the other hand, the catchment of Acarape do Meio presented the highest value, 39.3 kg km^{-2} yr^{-1}, possibly as consequence of its relatively small area and potentially high river connectivity. This catchment presents the highest precipitation index, which also favors higher TP runoff. Contrastingly, Lun et al. (2018) found a net soil P budget for Brazilian croplands of about 500 kg $km^{-2}.yr^{-1}$. Although the loads in the studied catchments come from more uses than croplands, the values are very low compared to this national average. Lower precipitation indexes and the intermittent regime of the rivers, potentially reducing TP runoff, contribute to these low estimates. Additionally, the TP retention in the numerous small upstream reservoirs within the catchment impacts this result (Lima Neto et al., 2011; Mamede et al., 2012, 2018). Contrastingly, the Santo Anastácio reservoir presented a TP load per unit area of 2626 kg km^{-2} yr^{-1} (Araújo et al., 2019), significantly higher than the above-mentioned national average. This is a combined result of the high precipitation indexes in a small urban catchment that additionally disposes untreated sewage into the reservoir.

3.3. *Assessment of TP point and non-point sources to the catchments*

To describe the proportions of TP load from different sources in each catchment and further contrast with the behavior of the nutrient-flow relationships, Fig. 4 shows the percentages of contributions from agriculture, soil, fish farming, livestock and sewer sources according to the environmental inventory of each catchment (Ceará, 2020b). Note that for the Santo Anastácio reservoir, it was assumed the sewer discharge as the main TP source (Pacheco and Lima Neto, 2017). It is noteworthy that some simplified hypothesis adopted when the environmental inventories were elaborated might bring uncertainties. They might be primarily related with empirical parameters such as loss coefficients from soil and pasture, as well as TP load per capita. This uncertainty analysis, however, is later addressed.

The NPS loads refer to animal husbandry, agriculture and soil erosion, while PS loads refer to fish farming and sewer production. The

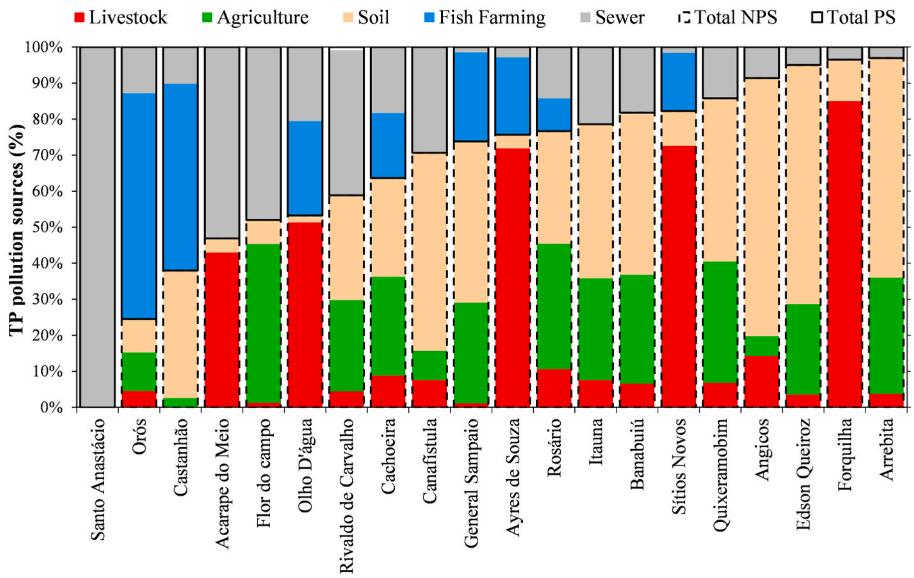

Fig. 4. Total phosphorus point source and non-point source load characterization for the studied catchments in the State of Ceará.

total NPS load exceeded PS loads in seventeen catchments, except the Castanhão, Orós and Acarape do Meio reservoirs. This accounts for a NPS dominance in 86% of the catchments. Forquilha and Arrebita reservoirs presented the highest NPS contributions (97% and 97%, respectively), while Orós and Castanhão the highest PS contributions (76% and 62%, respectively). The representative sewage contribution for the Orós reservoir is justified by the location of a large city (>100,000 inhabitants) in its surroundings. Its low coverage of sanitation facilities largely leads to untreated sewage inputs (Barbosa et al., 2012; Ceará, 2020b). For the Castanhão reservoir, Paula Filho et al. (2019) have estimated the P contribution of 28% of the catchment as 340 t yr^{-1}. Furthermore, in the rural areas of the State of Ceará, septic tank systems are the main method for disposal of human wastes. Lastly, although not monitored, the approximation for sewer contribution as the most significant TP source for Santo Anastácio is consistent with the characteristics of its catchment as well as previous studies (Araújo et al., 2019).

Overall, sewer contributed the most as PS load in 77% of the catchments, while fish farming accounted significantly for the PS load produced in the Orós (64%) and other reservoirs as shown in Fig. 4. Regarding the representativeness of NPS, livestock dominated in 25% of the catchments, while soil contribution prevailed in 55% as the most significant non-point source. Agriculture, then, predominated in the remaining catchments. The predominance of NPS over PS in the study area highlights the importance of animal husbandry and agriculture for the local population. Furthermore, the among-catchment heterogeneity of TP sources is largely explained by differentiated anthropogenically-sourced load, agricultural intensity, cattle density and soil type (Ceará, 2020b).

Regarding the soil influence on NPS contribution, soil erodibility plays an important role in the mobilization of pollutants (Li et al., 2020). Poorly drained soils can transfer the most NPS contribution (Greene et al., 2011), particularly in semiarid regions (Paula Filho et al., 2019). This process might be driven by rainfall through runoff events (Greene et al., 2011) as well as the soil type (Jacomine et al., 1973). The rainy season in Brazilian semiarid, well-defined from January to May, accounts for more than 80% of annual precipitation and 90% of annual erosivity (Araújo and Medeiros, 2013). In addition, riparian ecosystems are important to control NPS pollution from soil and sediments (Guo et al., 2014), providing a TP load reduction of up to 30% (Zinabu et al., 2017). In the studied area, these ecosystems are severely degraded, particularly for some water bodies such as Orós (Chaves et al., 2019). Ultimately, soil erodibility can also make available legacy P, although quantifying the real contribution of this TP input is very difficult (Sharpley et al., 2013).

Together, the aforementioned aspects related with high TP concentrations (see Fig. 2c), the estimated TP loads (see Fig. 3) and their main sources (see Fig. 4), arise the problem of how to reduce this nutrient source. This is particularly important considering human supply as the main use of water for these reservoirs. Among the non-point sources of pollution, the soil contribution is one of the most representative and difficult to address. It depends mainly on the condition and characteristics of the local vegetation, the degree of exposure to processes such as erosion and desertification, the land use by the diffuse rural population and the action of natural phenomenon. Thus, focusing on this source of pollution is more impractical, at least in a mid-term perspective. With regard to agriculture, undefined standards or widespread fertilizer application might increase the availability of TP to be carried out in runoff events, which can be addressed mainly through regulation by environmental agencies. Ultimately, the sewer pollution is the pressing P source through which is possible and necessary to take reduction measures. This approach, however, requires mostly a governmental effort to expand sanitation facilities, which currently cover only 41% of the urban areas of the state (Ceará, 2020c). These measures may, in the medium and long term, help decrease the transfer of P from the catchment to water bodies and possibly contribute to the improvement of water quality.

3.4. Adjusted nutrient-flow relationships

The fitted models encompassing average inflow and TP concentration at reservoir's inlet are presented in Fig. 5, while their modeling parameters are shown in Table 2. Three main nutrient-flow patterns should be highlighted: an inverse curve, when PS dominance occurs, representing a dilution pattern; a linear or exponential curve, for NPS dominance where a combined increase in flow and concentration is more likely; and, a u-shaped curve for PS and NPS codominance (Greene et al., 2011). These curves describe the existing relations with four parameters (A, B, C and D).

Analyzing the adjusted models along with the catchment sources of contribution of TP load reveal that the relationships presented a consistent shape form for the majority of the studied sites, with only few particularities. The adjusted curve for the reservoirs Castanhão, Forquilha and Sítios Novos presented clear u-shaped forms. Consistently,

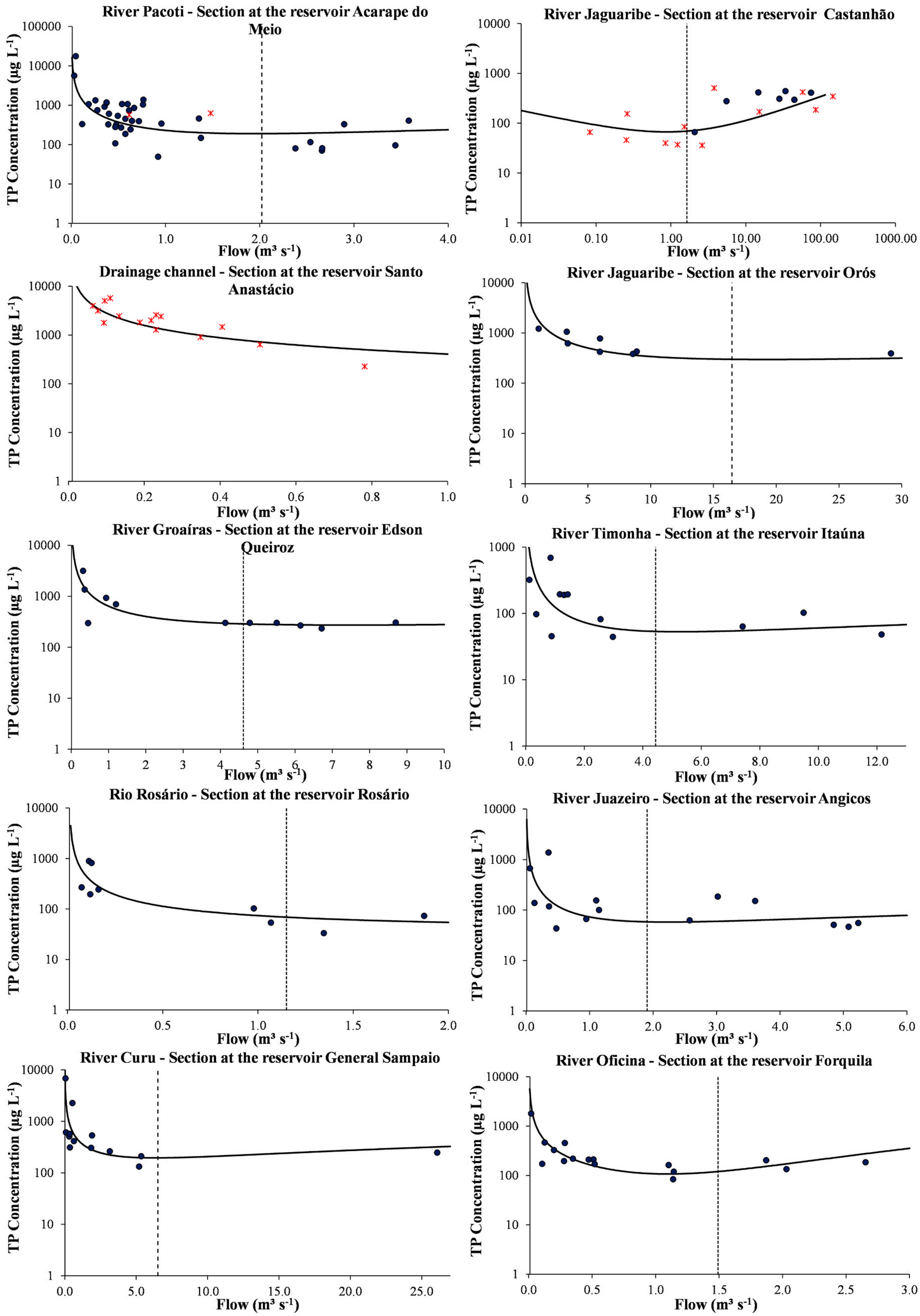

Fig. 5. Fitted models between mean TP_{in} and average inflow for the twenty studied reservoirs: dot (modeled concentration), asterisk (measured concentration), dashed line (changing flow) and continuous line (adjusted model).

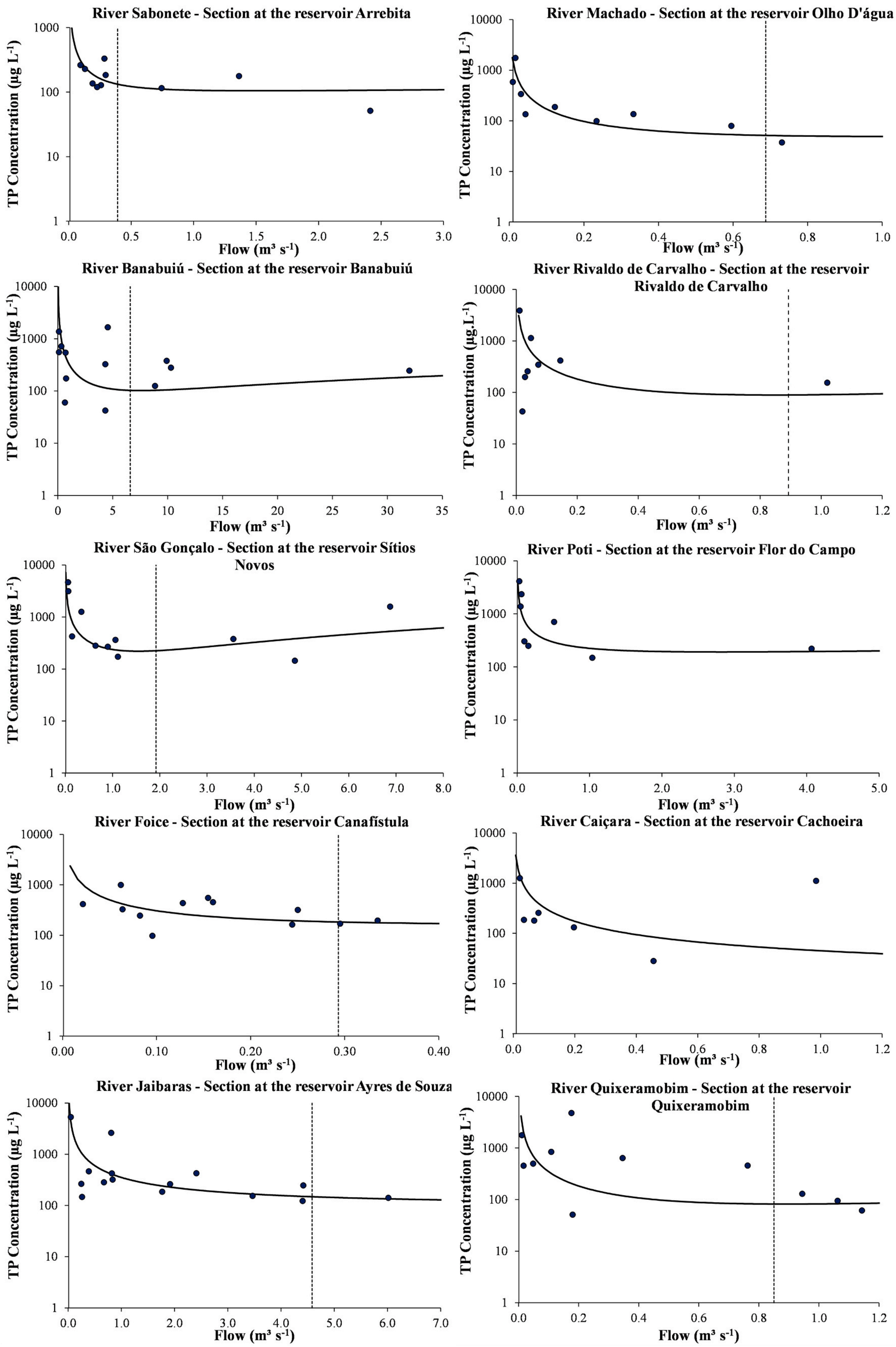

Fig. 5. (*continued*).

Table 2
Parameters of the TP_{in}-inflow adjusted models (A, B, C and D) along with the model performance indicators (NSE and R^2) and the inflow that equalizes point and non-point loads (Q_e).

Catchment's reservoir	Main river	Model parameters				Q_e ($m^3.s^{-1}$)	NSE	R^2
		A	B	C	D			
Ayres de Souza	Jaibaras	291.4	0.10	61.0	1.13	4.6	0.73	0.75
Forquilha	Riacho Oficina	85.0	0.13	23.0	3.40	1.5	0.60	0.94
Edson Queiroz	Groaíras	569.0	0.10	70.0	1.47	4.6	0.58	0.62
General Sampaio	Curu	390.0	0.26	23.0	1.77	6.5	0.56	0.61
Rivaldo de Carvalho	Rivaldo de Carvalho	40.3	0.10	50.0	2.00	0.9	0.56	0.60
Flor do campo	Poti	143.4	0.31	80.0	1.40	1.7	0.44	0.78
Santo Anastácio*	Drainage channel	406.9	0.16	0.0	0.00	–	0.41	0.53
Orós	Jaguaribe	1858	0.10	21.0	1.70	16.4	0.37	0.66
Sítios Novos	São Gonçalo	184.0	0.24	53.0	2.15	1.9	0.37	0.84
Rosário	Rosário	39.3	0.02	34.3	1.00	1.2	0.32	0.37
Castanhão	Jaguaribe	40.6	0.68	26.5	1.55	1.6	0.32	0.46
Acarape do Meio	Pacoti	183.0	0.07	47.0	2.00	2.0	0.29	0.52
Olho D'água	Machado	18.0	0.05	31.0	1.50	0.7	0.27	0.38
Arrebita	Sabonete	27.0	0.04	80.0	1.20	0.4	0.16	0.25
Cachoeira	Caiçara	20.0	0.01	35.0	2.00	0.7	0.10	0.21
Angicos	Juazeiro	55.0	0.02	18.0	1.75	1.9	0.07	0.15
Quixeramobim	Quixeramobim	67.0	0.17	47.5	1.90	1.2	−0.28	0.02
Canafístula	Foice	30.4	0.10	137.7	1.33	0.3	−0.43	0.06
Itaúna	Timonha	115.7	0.02	9.0	1.73	4.4	−0.91	0.10
Banabuiú	Banabuiú	320.0	0.03	12.0	1.77	6.6	−2.89	0.17

the percentage of TP load from NPS contribution is 97% and 82% for Forquilha and Sítios Novos, respectively. In contrast, Castanhão, with 38% of non-point source dominance, also presented a strong pattern of NPS dominated catchments. This can be attributed to eventual inaccuracies in the load estimation process in its environmental inventory, mainly due to the difficulty to account accurately non-point sources of pollution in a large catchment (44,800 km^2). Still, as the model for this reservoir present several TP_{in} measurements, the result is assumed to be reliable. None of the studied sites presented a single linear or exponential curve, except the Santo Anastácio reservoir, which presented only a dilution pattern. This is in line with the high contribution from point sources as well as the predominantly urban usage of the catchment.

Many sites presented a slightly u-shaped pattern with a decrease in TP_{in} as flow increases followed by an increase in concentration as flow increases. This aspect reveals a PS dominance for low flow rates and a NPS dominance for high flow rates, in line with which is expected since the catchments present contributions from point and non-point sources in significant proportions. At these sites, the discharge at which the estimated PS and NPS TP inputs are equal (Q_e) marks the inversion of the curve. Furthermore, it is important to highlight that the highest TP concentrations occurred during flows below the Q_e value in 90% of the catchments.

A high variability in the TP_{in} was observed in the fitted models of the Banabuiú and Quixeramobim reservoirs, which impacted in the adjustment of the model. This scatter in the estimated values may suggest large hysteresis patterns during some storm events (Bowes et al., 2014). For the Banabuiú, high TP_{in} derived from estimated TP loads with markedly different values in the pair $TP_{r,0}$-$TP_r(t)$, with elevated $TP_r(t)$ measurements. Sporadic peaks in TP_{in} concentration during low flow rates suggests the existence of a pollution source neither continuously constant nor diffuse, such as a random pollution event (Bowes et al., 2008). Cattle access points and local effluent thrown into rivers, for instance, might work as sporadic PS contributions (Jarvie et al., 2010). This aspect also elucidates the contrasting dilution pattern observed in these rural catchments.

The fitted model for the Acarape do Meio reservoir (with loads coefficients A = 183, B = 0.07, C = 47, and D = 2) derived from the largest TP_r dataset and also presented measured TP_{in} data by Lima et al. (2018). Prevails in the model low flow rates and a dilution pattern. The catchment present significant PS pollution near the tributaries, such as sewage treatment stations from which three discharge their effluents directly into the main river of the catchment (Lima et al., 2018). Its A coefficient, which accounts for PS contribution, is accordingly higher than 60% of the others.

With regards to the A and C fitting parameters shown in Table 2, they describe for each model the relevance of PS and NPS contributions, respectively. The A coefficients varied from 18 to 1858 with a median of 100.4, while the C coefficients varied from 0 to 137, with a median of 34.7. The adjusted model for the Orós presented the largest A coefficient (A = 1858) and, accordingly, its catchment presented the second highest PS contribution. The D coefficient, the flow-dependent one, describes the changing rate of the load with flow and remains greater than one to account for NPS contributions. D ≈ 1.0, however, suggests the poor ability of the model to predict only the mean TP concentration. Physical phenomena such as P remobilization into the river bed potentially lead to superior D values (Bowes et al., 2008). The best fit for two models in this work, however, presented D = 1. This difficulty to detect input signals from NPS contribution was also reported by Bowes et al. (2008). Regarding the B coefficient, the values always greater than zero indicate a PS load removal from water such as losses to river sediment through sorption process. This is particular intensified in slow water velocity, low flow, and when the sediment P sorption capacity is not saturated (Sharpley et al., 2013; Jarvie et al., 2013). Consumption and deposition are other intensifying processes of TP removal from the water (Ansari and Gill, 2014; Gargallo et al., 2017; Braga et al., 2019). Thus, the low flow levels during the studied period may have largely contributed to B parameters less than 1.

The majority of the fitted models were realistic with respect to the data, with modeled TP_{in} concentration versus average daily discharge showing satisfactory Nash–Sutcliffe coefficients. NSE coefficients over than 0.20 are considered satisfactory for NPS dominated catchments as highlighted in previous applications (Greene et al., 2011; Chen et al., 2015a). The fitted model for Ayres de Souza presented the best NSE (0.73). For those models with a poor performance metric (Canafístula and Itaúna reservoirs), their NSE was sensitively affected by isolated TP_{in} peak value. The NSE for Canafístula would change from −0.43 to 0.15 without this peak value. Similar analysis applies for the Itaúna reservoir. Ultimately, the indicators for the models of the reservoirs Quixeramobim and Banabuiú reservoirs have presented a low performance mainly due to the variability of the estimated TP_{in} in spite of the good overall pattern of the curve.

The model developed for the Castanhão reservoir (with loads coefficients A = 40.6, B = 0.68, C = 26.5, and D = 1.55) was applied to

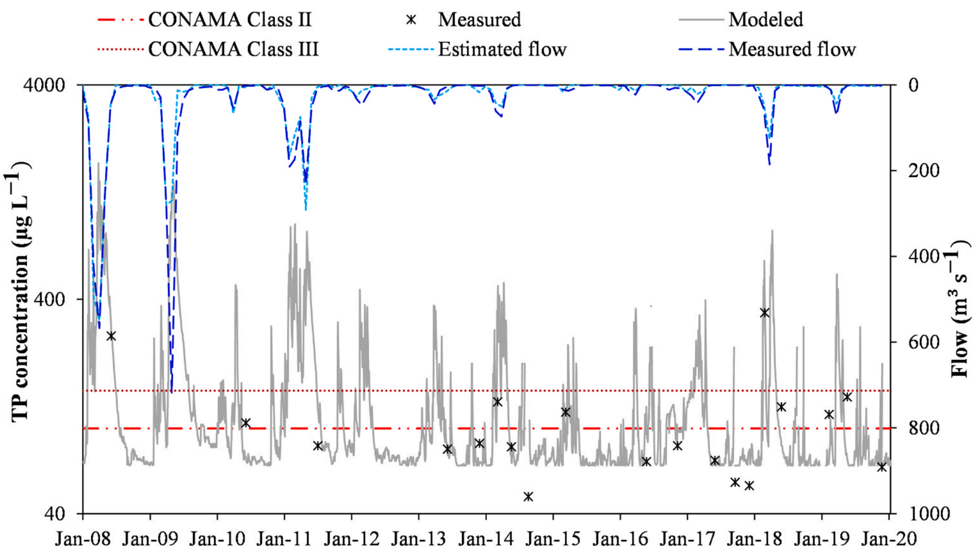

Fig. 6. Modeled TP_{in} concentration for the inlet of the Castanhão reservoir by the application of the adjusted flow-concentration curve.

estimate TP_{in} from measured flow data. The available measured riverine TP concentration was used to evaluate the model's performance. Fig. 6 presents the results of this modeling along with the national standards for water quality. Additionally, the measured and estimated flow are shown. The comparison between modeled and measured TP concentration resulted in a Nash coefficient of 0.63 and a R^2 of 0.70. As can be observed, the validation of the model included both low and high flow periods. However, some unusual low concentrations were not reached by the model, which may be due to the unrepresentativeness of this low concentration pattern in the entire dataset (<10%).

During high flow events higher concentrations are obtained as a result of the u-shape pattern of the model to reflect strong NPS contribution from wash-off. In the meantime, as low flow rates prevailed during the studied period, the dilution pattern of the model (for $Q < Q_e$) stood out and low concentrations were observed. Even under these conditions, only 53% of the estimated values were under the limit of Class II. Ultimately, the comparison between measured and estimated flow resulted in a NSE of 0.81 and a R^2 of 0.82, which highlight the quality of the flow data obtained from the water balance. This result reinforces the application of the flow estimated from water balance as a workaround to the inexistence of fluviometric stations at the inlet of several studied reservoirs.

3.5. Evaluation and comparison of the proposed models

The proposal and analysis of TP concentration-inflow relationships were carried out previously by several authors (Bowes et al., 2008, 2009b; 2010, 2014,1015; Greene et al., 2011; Bieroza and Heathwaite, 2015; Chen et al., 2015a). The detailed description of the parameters presented by the first four researches was, then, compared with the ones obtained in the present study. It is noteworthy that, even though the relationships reported by Bowes et al. (2009b) were developed with soluble reactive phosphorus concentration, once the fraction represented more than 65% of the TP load (Bowes et al., 2009b), their coefficients are likely to represent an order of magnitude reliable to perform the comparison. The compared studies were performed in perennial rivers and the general statistic, minimum, median and maximum values of the parameters are presented in Table 3. Bowes et al. (2008, 2009b, 2010) highlight the relevant influence of human occupation and PS contribution in their studied catchments, while Bowes et al. (2014) presented relationships with both TP load dominance.

The highest A coefficient shown in Table 3 was 11,776, about six times higher than the value of 1858 obtained in this study, suggesting a stronger influence of PS pollution. With regards to the A coefficient, the values reported by Bowes et al. (2014) were similar to those obtained in the presented study. As aforementioned, the similarities rely mainly in the NPS contribution to the catchments. The C coefficient, which mostly characterizes NPS contribution, reached the minimum values in this study in comparison with the others. In median terms, it was closer to the results obtained by Bowes et al. (2014). The minimum C value was fitted for the catchment of the Santo Anastácio reservoir. Accordingly, high C coefficients are not expected in urban catchments. Some particularities, such as B ≥ 1 and D ≤ 1 observed by Bowes et al. (2008), were not observed in the present study. The D and B coefficients were those more similar in all the studies. Particularly for the Santo Anastácio, the parameters related with the NPS pollution were nill. The model stated this way, accounting mainly PS contributions, reached the best performance. Additionally, this behavior is widely supported by the studies that highlighted sewer as the main TP input source for this water body (Pacheco and Lima Neto, 2017).

3.6. Uncertainty analysis

The three main approaches that integrate this research present some degree of related uncertainties that were evaluated to ensure the effectiveness of the methodology. Those three refer to the use of the reference

Table 3
Comparison among the parameters of the TP concentration-discharge relationships.

Statistics	A coefficient			B coefficient			C coefficient			D coefficient		
	Min	Med	Max	Min	Med	Max	Min	Med	Max	Min	Med	Max
This study	18.0	237	1858	0.01	0.2	0.7	0.0	45.2	138	0.00	1.7	3.4
Bowes et al. (2008)	0.01	316	11,776	0.0	0.0	1.1	0.9	84.6	490	0.97	1.5	4.0
Bowes et al. (2009b)	193	368	464	0.0	0.0	0.0	11.8	29.9	47.7	1.1	1.4	1.7
Bowes et al. (2010)	10.5	1765	6613	0.0	0.2	1.0	10.6	20.9	41.6	1.0	1.4	2.0
Bowes et al. (2014)	0.00	71.0	4806	0.0	0.0	0.0	9.0	61.0	585	1.0	1.4	1.8

load estimated by COGERH to validate the modeled loads, the TP load modeling process itself and associated errors and the uncertainties with the TP_{in} concentration-flow relationships. With regards to the reference load, the concerning aspects with the ECM methodology are the adequacy of the coefficients to each catchment as well as the precision in determining total population, land use and soil type. Literature reports a range of mean error in the application of this methodology varying from 4.3% to 31% (Johnes, 1996; Ding et al., 2010; Matias and Johnes, 2012; Delkash and Al-faraj, 2014; Tsakiris and Alexakis, 2015). Then, a comparison was carried out between the reference TP load with a maximum range of variation of 31% and the modeled load with its related range of variation.

For the catchments whose reference load was underestimated in modeling, increasing or decreasing the reference load by a percentage of error did not significantly enhanced the already obtained deviation. This analysis suggests that the reference load and its variability due to some inherent percentage of error is largely covered by the greater variability of the estimated loads. Furthermore, the variability in the input data to the modeled load might have a large influence on this deviation, as previously discussed. The correlation analysis between the mean modeled load and the reference load, though, reached a $R^2 = 0.74$. This result was considered satisfactory to the quality of the modeling process (Moriasi et al., 2007).

The coefficient of variation for the modeled loads ranged from 0.6 to 2.4 with mean of 1.1. Kulasova et al. (2012) reported a range 0.05–0.2, significantly lower than that obtained in this study. Uncertainties associated with discharge measurement, sample collection and laboratory analysis significantly contribute to this result. Sample collection, only, adds an uncertainty of 109% in the worst scenario for TP concentration and data aggregation, such as average measures, adds 13% of uncertainty (Harmel et al., 2006, 2009). Also, the range of variation in the estimation process of NPS loads vary from 30% to 110% (Harmel et al., 2006). The sampling interval between TP measurements might also imply uncertainties to the developed concentration-discharge relationships and their applicability. Quarterly average concentration-flow relations might reach from 10% to 20% uncertainty in load estimation (Dolan et al., 1981). Moreover, for the top 5% of flows, the uncertainties associated with the applied traditional power law fit might achieve even higher percentages (Hollaway et al., 2018).

3.7. Model implications and limitations

The methodology introduced in this research coupled catchment and reservoir. Designed from the modeling of average TP concentration at reservoirs' inlet from reservoirs inside characteristics, nutrient-flow relationships were adjusted. This methodology is replicable to semiarid regions with intermittent rivers where strategic water supply reservoirs are of greatest concern. It has the advantage of enhancing the understanding of the most relevant TP sources to a water body, evaluate the patterns of TP concentration coming from the catchment and predict the influence of inflow rates in water quality. Its main purpose is help addressing the TP issue in regions highly affected with lack of measured data in rivers and mostly monitored data in reservoirs. This methodology also provides means to evaluate seasonal TP concentrations only by classifying the levels of observed flow, with the advantage of simplicity to easily estimate averaged riverine TP concentration. Furthermore, it might be directly applicable in other regions with similar characteristics.

The development of relationships between TP concentration and river discharge to address PS or NPS influence of external P sources is widely reported. However, it was mostly applied to perennial rivers, a contrasting reality with the one of the studied area, with intermittent rivers and reservoirs for human supply. Thus, the approach adopted in the current paper tries to address the characteristics of the inlet waters of reservoirs as these water bodies are of utmost importance for semiarid regions. Additionally, nutrient-flow relationships can be used to address and understand the impacts of the catchment in the water quality of the reservoir.

Existing limitations related with the TP load estimation occur due to the utilization of simple relationships for the retention time RT and TP sedimentation rate k, as well as the reference TP load to guide the estimation process. However, the data limitation on the studied region strongly reduces the available alternatives to carry out a data comparison or validation. As previously presented, inaccuracies related with the sampling interval, analysis and data quality manipulation might impact the load estimation process and the modeled outputs. Another limiting aspect is related with the under-representation of high flow events during the studied period with only five years of precipitation indexes above average. This way, low flow events were more representative. This might have impacted the total TP load estimated from non-point sources. Moreover, the models indicated that, despite the studied catchments being rural, point sources were more influential than non-point sources. This may be due to aspects such as the proximity of the cities to the water bodies or because the streams remained with low flow conditions. Note that a dilution pattern prevails during low flow rates.

4. Conclusions

This research aimed to investigate the flow-related TP input load to semiarid reservoirs and obtain phosphorus-discharge relationships pairing average inflow and TP concentration at reservoir's inlet (TP_{in}). This was reached through the application of a simple modeling approach to predict TP_{in} from hydrological data and water quality measurements inside the reservoir. This model was particularly designed to support the issue of phosphorus pollution in semiarid data-scarce regions of intermittent rivers where artificial water supply reservoirs prevail as the most important water body.

The TP load produced in the catchments and delivered to the reservoirs was estimated throughout the application of a mass balance model. The modeled loads were considered satisfactory with only 10% of underestimated results in comparison to the reference TP load and a correlation between modeled and reference load with $R^2 = 0.74$. The estimated TP input loads per unit area of the rural catchments ranged from about 4 to 40 kg km^{-2} yr^{-1}, which were significantly lower than the national average of about 500 kg km^{-2} yr^{-1}. This was attributed to lower precipitation indexes and the intermittent regime of the rivers, which potentially reduce TP runoff. Additionally, the high-density reservoir network promotes TP retention in the thousands of small reservoirs distributed over the catchments. On the other hand, the average TP input load per unit area of an urban catchment was significantly higher (2626 kg km^{-2} yr^{-1}), which was a result of its relatively small area, higher precipitation indexes and low sanitation coverage.

The existing among-catchment variation of TP flux was mainly due to differentiated activities' intensity carried out in each basin. In general, a predominance of NPS over PS was observed in sixteen of the twenty catchments. The main non-point source of TP load were animal manure, agricultural production and natural soil erosion, whereas point source loads went from fish farming and sewer. With regards to TP_{in} estimation, remarkably high values were obtained, especially during low flow periods. This aspect suggests that PS pollution was more significant during the period, even when the catchments markedly presented rainfall related pollution dominance.

The TP-discharge models were realistic, with modeled TP_{in} concentration versus average daily discharge showing acceptable Nash–Sutcliffe coefficients. For the Castanhão reservoir, the concentrations resulting from the application of the model compared with measured riverine concentration achieved an NSE of 0.63. The predominance of NPS over PS of phosphorus supply in a great majority of the catchments was markedly reflected in three TP-discharge relations, with a strong u-shaped form behavior. For the others, only a slight NPS signal was observed prevailing a dilution pattern. Additionally, peak values in TP_{in} concentration sensitively affected the fitting of the models.

Regarding to the parameters of the model, the comparison between the ones obtained in this study with those from others researches highlighted the quality of the adjusted curves. Similar characteristics related to the range of the values of the four fitting parameters A, B, C and D were observed. The empirically-fitted models, once calibrated to the local data, have presented themselves as a feasible tool to analyze the phosphorus supply from the catchment to the reservoirs. It is expected that the simple methodology presented in this paper will help understand the TP issue in arid/semiarid regions and assist integrated water quality management actions in strategic water supply reservoirs.

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.jenvman.2021.113123.

Author contribution

Maria de Jesus Delmiro Rocha: Conceptualization; Data curation; Formal analysis; Investigation; Methodology; Validation; Writing – original draft. Iran Eduardo Lima Neto: Conceptualization; Formal analysis; Funding acquisition; Project administration; Supervision; Writing – review & editing.

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