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Hydrochemistry applied to assess the chemical weathering and soil removal rates in the Sorocaba River basin, São Paulo State

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ABSTRACT: Chemical weathering and soil removal rates are responsible for the Earth's landscape, composition of surface and groundwater, producing the soils and buffering the composition of the atmosphere. This study aimed to assess the chemical weathering and soil removal rates in the Sorocaba River basin, São Paulo State, Brazil, allowing answering the questions about the dynamics of fluvial transport of dissolved and suspended solids, the chemical weathering processes and associated atmospheric/soil CO_2 consumption, and the relationship between chemical weathering and soil erosion rates. The annual specific flux of total suspended solids and total dissolved solids were 49.59 and 60.97 t/km^2/yr. The chemical weathering process dominant in the Sorocaba River basin was the monosiallitization (RE = 2.4), with an associated atmospheric/soil CO_2 consumption of 2.3×10^5 mol/km^2/yr. The chemical weathering and soil removal rates were 7.2 and 29.8 m/Myr, respectively, indicating a soil thickness reduction. Finally, the soil removal rate in the Sorocaba River basin is almost 3-fold higher than the Cenozoic soil removal rates, being this difference related to the current land use which increased the soil removal processes.

KEYWORDS: Fluvial geochemistry; disturbed watershed; water-rock interactions; rainwater and anthropogenic influences.

INTRODUCTION

Chemical weathering is typically a destructive process, which allows the development of new minerals from the weathering of primary minerals. In addition, water-rock interactions are responsible for the Earth's landscape, composition of surface and groundwater, producing the soils and buffering the composition of the atmosphere, being this process one of the main mechanisms of atmospheric CO_2 removal and consequent deposition of carbonates Ca^{2+} and Mg^{2+} in oceans, playing an important role in moderating terrestrial climate (Gaillardet *et al.* 1999, Millot *et al.* 2002). Residual products are subject to other processes of the supergene cycle, such as erosion, transport, and sedimentation, which ultimately lead to continental denudation, with consequent flattening on the relief (Teixeira *et al.* 2000).

Pioneering studies to investigate the nature and composition of the dissolved and suspended load transported by rivers were performed in the 1960-70s (Barth 1961, Johnson *et al.* 1968, Gibbs 1970, Tardy 1971, Martin and Meybeck 1979). Since then, many studies have been carried out to assess chemical weathering and soil erosion rates using mass-balance models adjusted to atmospheric and anthropogenic (mainly originating from domestic sewage and industrial and agricultural activities) contributions, once the total river fluxes integrate the contributions of these different sources (Probst 1986, 1992, Meybeck 1987, Lasaga *et al.* 1994, White and Blum 1995, Boeglin and Probst 1996, 1998, Boeglin *et al.* 1997, Gaillardet *et al.* 1999, Semhi *et al.* 2000, Millot *et al.* 2002, Meybeck *et al.* 2003, Walling and Fang 2003, Riebe *et al.* 2004, Chakrapani 2005, Weijden and Pacheco 2006, Louvat *et al.* 2008, Gurumurthy *et al.* 2012, Laraque *et al.* 2013, Li *et al.* 2014). The interest in assessing the chemical weathering and soil removal rates in watersheds under different geological and climatic setting also occurred in Brazil (Stallard and Edmond 1981, 1983, 1987, Moreira-Nordemann 1980, 1984, Mortatti *et al.* 1997, 2008, Gaillardet *et al.* 1997, Bortoletto Junior *et al.* 2002, Conceição and Bonotto 2003, 2004, Mortatti and Probst 2003, Bonotto *et al.* 2007, Sardinha *et al.* 2010, Fernandes *et al.* 2012, 2016a, Conceição *et al.* 2015, Couto Júnior *et al.* 2019, Spatti Júnior *et al.* 2019).

The state of São Paulo established 21 units of Water Resources Management (UGRHI), according to Law No. 7,663, published in December 30th, 1991 (São Paulo 1991). The Sorocaba River basin belongs to UGRHI-10 (Médio Tiête — Sorocaba), presents well-defined climatic seasonality (tropical climate) and a diverse geological and geomorphological context. Successive cycles of development and diversification of human activities have occurred since its occupation in the seventeenth century. Nowadays, this watershed covers 18 municipalities (1,212,376 inhabitants), an important industrial park, with over 1,850 enterprises and

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large agricultural areas (IBGE 2010). Approximately 65% of the demands for public supply in the Sorocaba River basin are supplied by Itupararanga Reservoir (IPT 2006). Despite its importance, few studies have been conducted in the Sorocaba River basin related to the rainwater chemical composition and annual atmospheric deposition (Conceição *et al.* 2011, 2013), the chemical weathering rates in the Upper Sorocaba River basin (Sardinha *et al.* 2010, Fernandes *et al.* 2016a), the water quality of the Itupararanga Reservoir (Pedrazzi *et al.* 2013, 2014), and the origin and flux of trace elements and isotopic composition of particulate organic matter in suspended sediment (Fernandes *et al.* 2012, 2016b).

Thus, this study aims to assess the chemical weathering and soil removal rates in the Sorocaba River basin, allowing answering the following questions:

- What are the dynamics of fluvial transport of dissolved and suspended solids?;
- What are the chemical weathering processes and associated atmospheric/soil CO_2 consumption?;
- What is the relationship between the chemical weathering and soil removal rates?

STUDY AREA

The Sorocaba River basin is located in the southeastern portion of São Paulo State, Brazil, between latitudes 23 and 24ºS and longitudes 47 and 48ºW, and occupies an area of 5,269 km^2. Considered the most important tributary of the left bank of Tietê River, Sorocaba River travels 227 km before flowing into Tiete River, in Laranjal Paulista municipality (IPT 2006). This watershed is inserted into two main geomorphological units: Atlantic Plateau and Paulista Peripheral Depression (Ross 1996 — Fig. 1). The Atlantic Plateau presents metamorphic rocks belonging to the São Roque Group and Embu Complex, with associated granitic rocks (Godoy *et al.* 1996). The relief is comprised of hills shapes with convex tops and deep valleys with altitudes that range between 800 and 1,000 m a.s.l. and slope above 20% (Ross 1996, Perrota *et al.* 2005). In the Paulista Peripheral Depression outcrop the sedimentary rocks belonging to the Parana Sedimentary Basin (Paleozoic-Mesozoic), i.e., Itararé Group (diamictic, sandstones, mudstones, and rhythmites), Guatá Group (siltstones and sandstones), and Passa Dois Group (siltstones, mudstones, and shales) (Conceição and Bonotto 2004, IPT 2006). The relief presents hills with tabular and large convex tops, prevailing altitudes between 600 and 700 m a.s.l. and slopes ranging from 5 to 10% (Ross 1996, Perrota *et al.* 2005).

The predominant soils in the study area are Red Argisol (49%), Red Latosol (38%), and Red-Yellow Latosol (9%), according to the Brazilian soil classification (EMBRAPA 2013, Oliveira *et al.* 1999), corresponding to Ultisols and Oxisols in the USDA nomenclature (USDA 1999), respectively. Forests, fields, and Savanna characterized the original vegetation. Currently, with the agricultural occupation and the urbanization processes, land use is characterized by the predominance of the

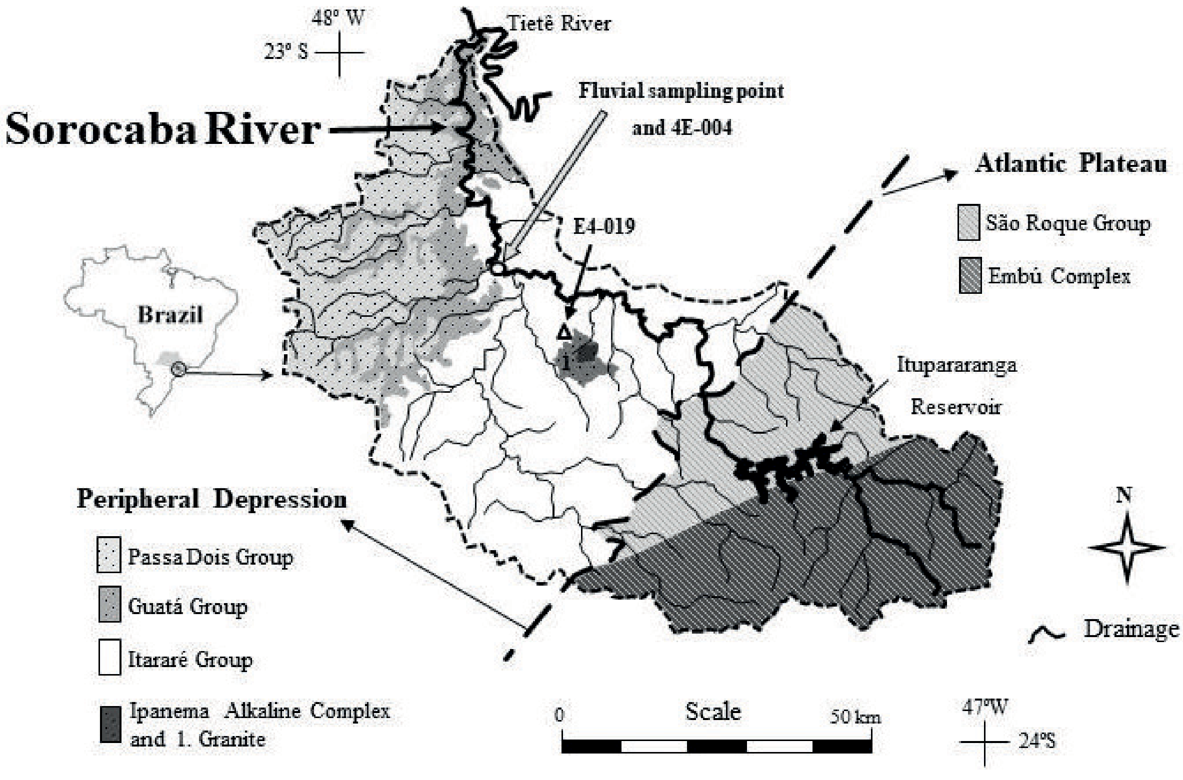

Figure 1. Geological map of Sorocaba River basin with location of the fluvial sampling point at the Tatuí municipality, and the pluviometric and fluviometric stations (E4-019 and 4E-004, respectively).

pastures and fields (77%), followed by areas with agricultural crops (14%), reforestation areas (3%), original vegetation cover (2%), and urban areas (4%) (IPT 2006).

The climate is Cwa type, according to the Köppen classification (Köppen 1948), characterized by the predominance of rainfall in summer and dryness in winter, with an average annual temperature of 18 to 22ºC (IPT 2006). Figure 2A shows the monthly averages of rainfall and discharge in the Sorocaba River basin from 1979 to 2008, calculated from the monthly historical data of the Pluviometric station E4-019 (23º20'S, 47º41'W) and the Fluviometric station 4E-004 (23º19'S, 47º46'W) (DAEE 2010), respectively. During this period, the average annual rainfall was 1,276 mm, where January and August were the months with the highest and lowest rainfall values (236 and 35 mm, respectively). In the same historical period, the average annual discharge was 53.8 m^3/s, with the highest monthly average in February (98.3 m^3/s) and lowest in August (33.7 m^3/s). Figure 2B shows a significant positive linear correlation between the average monthly values of rainfall and discharge of these 30 years.

MATERIALS AND METHODS

Sampling and analytical methods

The river sampling point was established approximately 500 m upstream from the confluence of the Sorocaba and Tatuí rivers, in the municipality of Tatuí (Lat. 23º19'09"S, Long. 47º46'44"W), as can be seen in Figure 1, covering an area of 3,942 km^2, i.e., 74.8% of the total area of the Sorocaba River basin, with a total population of 1,061,023 inhabitants (IBGE 2010) and the urban sewage treatment percentage estimated at 17.5% (IPT 2006). This sampling point was chosen due to there being a fluviometric station installed (limnimetric ruler and an automatic limnigraph), managed by DAEE/CTH, with daily discharge data since 1940. These data were used to validate the discharge measurements performed during the sampling period.

Twelve fluvial water sample collections were carried out at the Sorocaba River, covering one complete hydrological cycle (Jun/2009 to Jun/2010). Sorocaba River waters (1,000 mL) were collected in each sampling at 1.5 m deep, using a single-stage punctual sampler. The samples were separated into two 500 mL aliquots, one crude and the other preserved with 0.1 mL of concentrated H_2SO_4. Both aliquots were stored in identified polyethylene bottles and kept at 4ºC until laboratory processing.

Discharge (Q), hydrogenionic potential (pH), electrical conductivity (EC), and temperature (T) were characterized in the field using direct reading equipment. The discharge was represented by the product of the wet river channel cross-section area (m^2), obtained by bathymetry, and the average velocity of the water flow in this section (m/s) quantified using a Digital Micromolinete Global Water FP 101. The pH values were determined using a DM2 Digimed portable pHmeter, with a relative accuracy of 0.01% and calibrated with standard solutions DM-S1B (pH 4.01) and DM-S1A (pH 6.86). In addition, EC and T were quantified using the Digimed DM3 sensor, with a resolution of 0.01 mS/cm, relative accuracy of 0.05% and automatic temperature compensation, previously calibrated with conductivity standard solutions DM S6A (1,412 mS/cm and DM S6B (146.9 mS/cm).

Crude fluvial water samples were filtered through cellulose membrane filters (0.45 mm), previously dried and weighed. These filtered samples were analyzed by ion chromatography Dionex ICS-90 equipped with analytical columns IonPac® CS12A 4x250 mm and IonPac® AS14A 4x250 mm, for the quantification of dissolved ions (Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, SO_4^{2-}, PO_4^{3-}, and NO_3^-), with a detection limit of 0.001 mg/L (Dionex Corporation 2004) and quantification limit of 0.01 mg/L (Ribani *et al.* 2004). The HCO_3^- was represented by the alkalinity content and was quantified by the Gran method (Edmond 1970). The preserved fluvial water samples were filtered through a glass fiber membrane filter (0.3–0.6 mm) and used in the quantification of dissolved Si^{4+} concentration by optical emission spectrometry with inductively coupled argon plasma, ICP-OES Optima 3000 DV, with a detection limit of 0.02 mg/L, and the result was expressed in terms of SiO_2. The total dissolved solids (TDS) correspond to the sum of dissolved cations, anions and silica. The total suspended solids (TSS) was quantified by gravimetry (APHA 1999), considering the retained material in the cellulose membrane filter after drying in a stove at

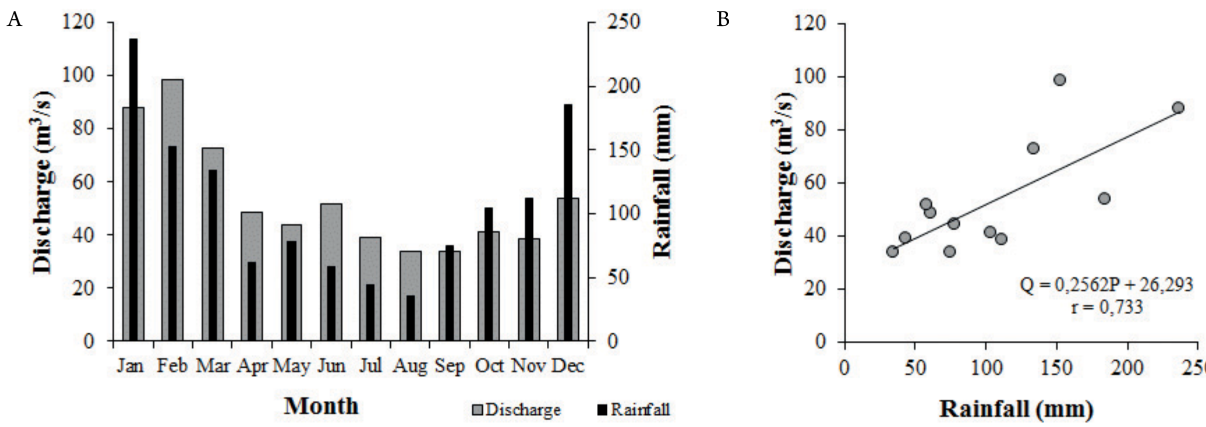

Figure 2. (A) Monthly average rainfall and discharge for a 30-year period (1979–2008) in the Sorocaba River basin, and (B) relationship between the monthly average rainfall and discharge for the same period.

60ºC to constant weight. The analysis of the river water samples was performed at Stable Isotope Laboratory (dissolved ions, HCO_3^- and TSS) and Analytical Chemistry Laboratory (dissolved silica), both located at CENA/USP.

Theoretical background

The fluvial fluxes (F_W, in t/km²/yr) of dissolved chemical species, TDS and TSS related to chemical weathering and soil removal processes, were calculated using a mass balance model expressed in Equation 1 (White and Blum 1995), considering negligible the fluxes from the biomass change and derived from the ionic exchange sites in clay minerals.

$$F_W = F_{river} - F_{rainfall} - F_{anthropogenic} \quad (1)$$

In which:
F_{river} = the measured river flux (t/km²/yr);
$F_{rainfall}$ = the atmospheric inputs (t/km² /yr);
$F_{anthropogenic}$ = the anthropogenic influences (t/km²/yr).

The R_E index can be used to determine the predominant process of chemical weathering of rocks in a drainage basin. Initially proposed by Tardy (1971), this index is equivalent to the molecular ratio $(SiO_2)/(Al_2O_3)$ of secondary minerals neoformation within the soil profile. Boeglin and Probst (1998) modified the R_E index, being expressed by the molar ratio of chemical dissolved species in the surface waters (Eq. 2).

$$R_E = \frac{3K + 3Na + 2Ca + 1.25Mg - SiO_2}{0.5K + 0.5Na + Ca + 0.75Mg} \quad (2)$$

The atmospheric/soil CO_2 consumption during chemical weathering processes (F_{CO2} – in mol/km²/yr) was estimated by the sum of corrected fluxes of Na^+, K^+, Ca^{2+}, and Mg^{2+} ($F_{(ion)\,sil}$ - in mol/km²/yr), according to Equation 3 (Gaillardet *et al.* 1999, Gurumurthy *et al.* 2012).

$$F_{CO2} = F_{Na\,sil} + F_{K\,sil} + 2\,F_{Ca\,sil} + 2\,F_{Mg\,sil} \quad (3)$$

The chemical weathering of rocks (IQ – in t/km²/yr) can be estimated through the sum of the corrected annual fluvial flux of Na^+, K^+, Ca^{2+}, Mg^{2+}, and SiO_2 ($F_W(ion)$ - in t/km²/yr), i.e., after correction of atmospheric inputs and anthropogenic contributions, according to Equation 5 (Probst 1992). The ratio among the IQ and the average density of rocks for the watershed represent the chemical weathering rate (Wq - in m/Myr), as expressed in Equation 5.

$$IQ = F_W(Na^+) + F_W(K^+) + F_W(Ca^{2+}) + F_W(Mg^{2+}) + F_W(SiO_2) \quad (4)$$

$$W_q = \frac{IQ}{\rho} \quad (5)$$

The soil removal rates (W_m in m/Myr) can be calculated through Equation 5; however, the use of corrected TSS annual flux (t/km²/yr) and the average soil density (g/cm³) is necessary instead of IQ and average density of rocks, respectively (Mortatti *et al.* 1997, Boeglin and Probst 1998).

RESULTS

Table 1 shows the results of Q, pH, EC, T and the concentrations of dissolved ions and SiO_2, TDS, and TSS, with their respective discharge weighted average for the study period.

The discharge showed seasonal variation in consonance with the historical data of the monthly average (Fig. 2A), with the highest value obtained in Jan/2010 (230.40 m³/s) and the lowest in Jun/2009 (28.77 m³/s). Despite the similar seasonality,

Table 1. Physical and chemical parameters for the Sorocaba River surface waters.

Parameter	Unit	Sampling date												C_{WAV}
		Jun/09	Jul/09	Aug/09	Sep/09	Nov/09	Dec/09	Jan/10	Feb/10	Mar/10	Apr/10	May/10	Jun/10	
Q	m³/s	28.77	32.60	81.35	67.01	110.03	228.58	230.49	118.64	98.56	71.79	48.85	31.64	95.69
pH		6.8	6.9	6.9	6.9	6.7	6.5	6.6	6.8	6.8	6.9	6.8	6.9	6.7
EC	mS/cm	136.9	141.8	108.6	110.9	82.2	70.9	73.7	98.1	89.5	99.0	115.0	128.0	104.6
T	ºC	16.7	17.0	16.5	19.5	26.3	26.2	27.5	25.0	26.8	25.5	22.0	20.3	22.4
SiO_2	mg/L	34.00	28.00	13.72	13.66	14.00	9.91	9.53	11.70	11.58	12.07	16.00	28.00	13.05
Ca^{2+}	mg/L	17.92	16.90	9.20	10.40	9.00	5.69	6.80	10.40	11.70	13.41	13.08	15.00	9.43
Mg^{2+}	mg/L	1.32	1.02	0.90	0.97	0.86	0.80	0.87	0.92	1.00	1.10	1.10	1.40	0.93
Na^+	mg/L	19.15	15.92	10.59	11.41	6.38	5.64	5.61	7.22	8.03	8.65	10.94	13.82	8.03
K^+	mg/L	2.26	1.90	1.70	1.83	1.65	1.47	1.70	1.75	1.90	2.00	2.10	2.30	1.75
HCO_3^-	mg/L	51.31	45.05	36.03	38.59	35.21	25.77	30.26	38.00	36.20	37.99	39.86	43.41	34.25
Cl^-	mg/L	18.90	15.20	7.86	8.62	4.26	3.63	4.40	5.48	6.28	7.50	8.64	10.75	6.22
SO_4^{2-}	mg/L	7.74	6.56	4.47	4.82	4.08	2.71	2.37	2.72	4.40	5.28	5.99	9.25	3.88
NO_3^-	mg/L	4.82	3.10	2.06	2.29	1.53	0.66	0.87	1.45	3.40	4.00	5.38	5.61	2.02
PO_4^{3-}	mg/L	0.13	0.16	0.07	0.04	0.07	0.02	0.03	0.05	0.16	0.16	0.19	0.21	0.07
TDS	mg/L	157.55	133.81	86.61	92.63	77.05	56.29	62.45	79.69	84.64	92.16	103.28	129.74	79.64
TSS	mg/L	19.50	31.00	70.33	50.83	105.33	74.00	66.33	25.83	66.67	41.67	18.33	11.83	59.56

C_{WAV}: weighted average element/compound concentration for the study period; Q: discharge; EC: electrical conductivity; T: temperature; TDS: total dissolved solids; TSS: total suspended solids.

the average discharge for the study period (95.69 m^3/s) was 1.8 times higher than the historical annual average for the period of 1979–2008 (53.8 m^3/s). This is justified by the fact that the rainfall in the study period (2,101 mm) was higher than the historical average (1,276 mm), with a direct impact on the discharge values. During the historical period, a similar occurrence was observed only in 1983, with an annual rainfall of 2,054.0 mm and average discharge of 143.49 m^3/s.

The Sorocaba River waters presented a pH close to neutral, ranging from 6.5 to 6.9. The EC showed a significant seasonal variation (annual average of 104.6 mS/cm), with values below 74 mS/cm in the months of highest rainfall and discharge, and values above 135 mS/cm in Jun and Jul/2009. During the dry season (May to October), EC values were higher than the expected limit for natural waters, i.e., 100 mS/cm (Hermes and Silva 2004). The T followed the seasonal variation, with the lower values in winter (16.5ºC in Aug/2009) and higher in summer (27.5ºC in Jan/2010).

The concentration of [TSS] was directly related to the discharge (Fig. 3A). According to Probst (1986), for most world rivers the model obtained for the relationship between [TSS] and Q ($[TSS] = a.Q^b$) presents positive b exponent with values between 1 and 2, indicating that the increase in [TSS] is a function of the discharge increase. This exponent in the model established for the Sorocaba River was 0.7039, indicating that the [TSS] was also influenced by rainfall. This influence is highlighted in the November and December 2009, when the fluvial water sampling was performed after two days of significant precipitation, with accumulated volumes of 45.8 and 25.9 mm, respectively.

On the other hand, the relationship between [TDS] and discharge was inverse and significant (Fig. 3B), which characterizes the dilution process with increasing discharge. Among the dissolved chemical species that composes the TDS, evaluable on a molar basis of C_{WAV}, the anionic predominance of HCO_3^- (33.1%) was verified, followed by Cl^-, SO_4^{2-}, NO_3^-, and PO_4^{3-}, while for the cations the greatest participation was Na^+, with 20.6%, followed by Ca^{2+}, Mg^{2+}, and K^+, respectively, and the SiO_2 represented 12.8% of the TDS. The relationship "sum of cation *vs.* sum of anion" (Probst 1992), in meq/L, indicated a deficit of anionic charge in the charge balance (Fig. 3C). It can be attributed to the presence of dissolved organic anions not counted in this study, such as dissolved organic carbon (Probst *et al.* 1992, Boeglin and Probst 1996, Laraque *et al.* 2013).

DISCUSSION

Dynamics of fluvial transport in the Sorocaba River basin

The fluvial fluxes integrate the contributions of the chemical weathering and soil removal processes that occur in natural watersheds. However, nowadays it is also necessary to consider the atmospheric inputs and anthropogenic influences in the fluvial dynamics (Stallard and Edmond 1981, Mortatti *et al.* 1997, Semhi *et al.* 2000, Bortoletto Junior *et al.* 2002, Conceição and Bonotto 2004, Weijden and Pacheco 2006, Mortatti *et al.* 2008, Conceição *et al.* 2010, Hissler *et al.* 2015, 2016).

The F_{river} of dissolved chemical species, TDS and TSS were quantified in the specific transport form, the result of the product between C_{WAV} and average discharge of the study period weighted by surface of study area, according to the stochastic methodology proposed by Probst (1992). $F_{rainfall}$ was represented by the specific input of solute, obtained from the total precipitation in the study period (2,101 mm) and the average concentration of dissolved chemical species obtained by Fernandes (2012).

The $F_{anthropogenic}$ for dissolved chemical species, TDS and TSS were obtained using secondary data, despite the uncertainties associated with these data regarding the reality of the studied basin. In relation to dissolved load, it was considering the *per capita* values of the dissolved chemical species present in untreated domestic effluents discharged directly in the river (g/hab/day) established by Mortatti *et al.* (2008, 2012) for the Médio Tietê basin (SiO_2 = 0.84, Ca^{2+} = 7.50, Mg^{2+} = 1.3, Na^+ = 13.1, K^+ = 2.6, HCO_3^- = 42.0, Cl^- = 7.1, and SO_4^{2-} = 12.5), and the total population upstream of the sampling point (1,061,023 inhabitants). The anthropogenic contribution of SiO_2 was considered negligible, such as reported in other studies (Mortatti *et al.* 2008, 2012). On the other hand, the $F_{anthropogenic}$ associated to suspended sediment load was represented by the *per capita* TSS load contained in untreated urban sewage (0.022 kg/hab/day), obtained from average production of untreated urban sewage (100 L/hab/day) and respective TSS average concentration (220 mg/L), both global references data published by Tchobanoglous and Burton (1991), the total population upstream of the sampling point and the respective percentage of urban sewage treatment (17.5%) (IBGE 2010).

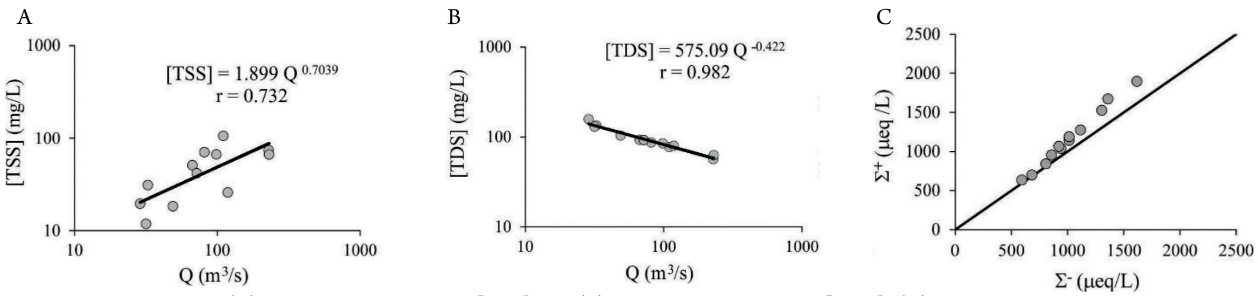

Figure 3. Relationships (A) between discharge and [TSS] and (B) between discharge and [TDS], (C) and charge balance in the Sorocaba River in the study period, with S^+ and S^- corresponding to total dissolved cations and anions, respectively.

The fluxes of cations, anions, silica, TDS, and TSS in the Sorocaba River basin are shown in Table 2. The total fluvial flux of TDS was 33% higher than that observed to TSS flux. Among the dissolved chemical species, the HCO_3^- presented the highest fluvial flux, corresponding to 43% of TDS, followed by SiO_2 (16.4%), Ca^{2+} (11.8%), Na^+ (10.1%), and Cl^- (7.8%), while the fluvial flux presented by SO_4^{2-}, NO_3^-, K^+, Mg^{2+} and PO_4^{3-} were lower than 5 t/km²/yr and together represented the remaining 10.9% of TDS. The atmospheric inputs account for 17.3% of the total specific flux of TDS in the Sorocaba River. Regarding the anthropogenic inputs, there was a higher contribution to the dissolved load (ca. 14% of the fluvial TDS) than to the suspended solids load (ca. 4% of the fluvial TSS).

Assuming that the suspended load represents approximately 90% of the total sediment river flux (Walling and Fang 2003), the specific flux of the total suspended solids exported by the Sorocaba River was estimated at 45.59 t/km²/yr. After correction of the anthropogenic contributions, the specific flux related to the soil removal (F_w) was 43.81 t/km²/yr. According to the classification proposed by Meybeck *et al.* (2003) for the world's rivers, from very low to extremely high, the soil removal in the Sorocaba River basin was considered as medium-specific sediment flux (range from18.25 to 73 t/km²/yr).

Chemical weathering processes and atmospheric/soil CO_2 consumption

The weathering process is characterized according to the classification proposed by Pedró (1966), where $R_E \approx 0$ characterizes the total hydrolysis process called allitization, with only aluminum and iron fixed as insoluble hydroxides; when $R_E \approx 2$, the process is called partial hydrolysis with monosiallitization, occurring the kaolinite formation; and to $R_E \approx 4$ the predominant process is the partial hydrolysis with bisiallitization and is related to the formation of mineral 2:1, such as montmorillonite.

The predominant process of chemical weathering of rocks in the Sorocaba River basin, was determined using the R_E index (Eq. 2) and corresponded to 2.4, value that characterizes the predominance of partial hydrolysis with a tendency to monosiallitization, i.e., to the kaolinite stability domain, similar to that observed in the Amazonas River basin (Mortatti and Probst 2003). However, in two watersheds (Tietê and Piracicaba river basins) located in the same region of the Sorocaba River, a different situation was verified, i.e., a tendency to the bisiallitization domain, probably due to extensive agricultural areas with a high degree of soil tillage, fact that may influence the remobilization of major ions instead of silica (Bortoletto Junior 2004).

According to Conceição and Bonotto (2004) and Fernandes *et al.* (2016a), the main minerals found in the igneous and metamorphic rocks of the Sorocaba River basin are biotite ($K(Mg,Fe)_3(Si_3Al)O_{10}(OH)_2$), muscovite ($KAl_2(Si_3Al)O_{10}(OH)_2$), sillimanite ($Al_2SiO_5$), quartz ($SiO_2$), microcline ($KAlSi_3O_8$), oligoclase ($(Na,Ca)(Si,Al)_4O_8$), and hornblende ($Ca_2Na(Mg,Fe)_4(Al,Fe,Ti)AlSi_8AlO_{22}(OH)_2$). For sedimentary rocks, quartz, albite (NaAlSi3O8), microcline, kaolinite ($Al_2Si_2O_5(OH)$), and illite ($K_{0.9}Al_2Si_4O_{10}(OH)_2$) were highlighted. Theoretically, the weathering reactions involving the mineral rock of the Sorocaba River basin indicate that the Na^+ has its origin in the hydrolysis of albite, hornblende and plagioclase; K^+ ions are derived from the hydrolysis of muscovite, microcline, biotite and illite; the Ca^{2+} can be attributed to the hydrolysis of hornblende and plagioclase; and Mg^{2+} can be released by the hydrolysis of hornblende and biotite. In addition, the Sorocaba River basin does not contain volumetrically significant $Cl^–$, $NO_3^–$, $PO_4^{3–}$ or $SO_4^{2–}$ bearing minerals. Therefore, only small inputs of these anions are expected in the rivers due to water-rock interactions. Quartz and kaolinite are not weathered and remain in the soil profile, as well as the supergene minerals, i.e., kaolinite, goethite (FeOOH), and rutile (TiO_2).

The atmospheric/soil CO_2 consumption during the chemical weathering processes in the Sorocaba River basin was obtained using Equation 3 and corresponded to 2.3×10^5 mol/km²/yr. This value was lower than that observed in the Tietê River basin (3.8×10^5 mol/km²/yr, Bortoletto Junior 2004). However, it was higher than other Brazilian watersheds, such as the Amazonas Basin (0.3×10^5 mol/km²/yr, Mortatti and Probst 2003) and Jamari and Jiparana basins (0.8×10^5 and 1.4×10^5 mol/km²/yr, respectively, Mortatti *et al.* 1992) in northern region, or in the Paraná Basin (0.9×10^5 mol/km²/yr, Gaillardet *et al.* 1999) and Piracicaba Basin (1.4×10^5 mol/km²/yr, Bortoletto Junior 2004), both in the Southeastern Brazilian region.

Chemical weathering and soil removal rates

The *IQ* value in the Sorocaba River basin, obtained using Equation 4 and the data of Table 2, corresponded to a flux of 19.1 t/km²/yr, representing 31.4% of TDS flux at the river. The Amazonas and Tietê River basins showed higher fluxes (*IQ*) than that observed for the Sorocaba River

Table 2. The annual flux (t/km²/yr) of total suspended solids (TSS), total dissolved solids (TDS), dissolved silica, cations and anions in the Sorocaba River basin.

Species	TSS	TDS	SiO_2	Ca^{2+}	Mg^{2+}	Na^+	K^+	HCO_3^-	Cl^-	SO_4^{2-}	NO_3^-	PO_4^{3-}
F_{river}	45.59	60.97	9.99	7.22	0.71	6.15	1.34	26.22	4.76	2.97	1.54	0.06
$F_{rainfall}$*	---	10.57	---	2.99	0.12	0.40	0.28	3.60	0.71	1.24	1.18	0.05
$F_{antropogenic}$	1.78	8.54	0.08	0.74	0.13	1.29	0.26	4.13	0.70	1.23	---	---
F_w	43.81	41.85	9.91	3.49	0.47	4.46	0.80	18.49	3.35	0.50	0.36	0.01

*Data reported in Fernandes (2012).

basin, with 32.2 e 41.4 t/km^2/yr, respectively (Mortatti and Probst 2003, Bortoletto Junior 2004). The chemical weathering rate (*Wq*) for the Sorocaba River basin was calculated using Equation 5 and the regional value of the mean density of rocks (2.65 g/cm^3 — Brasil 1983) and corresponded to 7.2 m/Myr. This rate was 22 and 54% higher than those obtained for the Tietê (5.9 m / Myr) and Piracicaba (4.7 m / Myr) river basins, respectively (Bortoletto Junior 2004).

The soil removal rate in the Sorocaba River basin was 29.8 m/Myr, considering that the average soil density is 1.47 g/cm^3 (Fernandes *et al.* 2012). This rate was lower than that observed in the Amazonas River basin (123 m/Myr, Mortatti and Probst 2003) and higher than that in the Jamari and Jiparaná river basins (6.5 m/Myr in both basins, Mortatti *et al.* 1992). The Tietê and Piracicaba river basins, located in the same geographical region as the Sorocaba River, presented higher rates when compared to those obtained in this study, i.e., 42.6 and 37.0 m/Myr, respectively (Bortoletto Junior 2004). Considering the chemical weathering and soil removal rates in the Sorocaba River basin (7.2 and 29.8 m/Myr, respectively), in the present climatic setting, there is a soil thickness reduction.

The cooling/denudation crustal rates quantified using apatite fission track (AFT), apatite (U-Th)/He (AHe) and *in situ* cosmogenic ^{10}Be could be used to compare the present soil removal rates obtained by a fluvial mass-balance with the Cenozoic soil removal rates. Values of past denudation obtained in southeast Brazil ranging from 8.8 to 15.7 m/Myr, using *in situ* cosmogenic ^{10}Be (Cherem *et al.* 2012). Hackspacher *et al.* (2004) used AFT ages to indicate a cooling/denudation rate of 11 m/Myr at the boundary between the Paraná Sedimentary Basin and the basement rocks. The soil removal rate in the Sorocaba River basin (29.8 m/Myr) is almost 3-fold higher than the estimates of Cenozoic denudation reported Cherem *et al.* (2012) and Hackspacher *et al.* (2004). This difference can be explained by the present land use in the Sorocaba River basin, where the replacement of the original vegetation by agricultural and livestock activities increased the erosion processes and, consequently, the present denudation rates, even though the study region has remained roughly in the same latitude during the drift to west of South America, since the time of the separation of continents and the basalt eruptions of the Serra Geral Formation.

Couto Júnior *et al.* (2019) evaluated three different scenarios from land use changes and how they have affected soil loss in a watershed located in the PPD, using the USLE model. Similar to Sorocaba River basin, the main types of soils occurring in the studied area were Ultisol and Oxisol. The authors verified a similar soil removal rate, it was almost 3-fold higher than the long-term denudation rates suggested by the literature for the Peripheral Depression, and reinforced that the increase in the denudation rate is mainly related to land use/land cover changes than to the soil type present in the studied area.

CONCLUSION

This study aimed to evaluate the chemical weathering of rocks and soil removal processes that occur in the Sorocaba River basin and allowed a better understanding of the dynamics of fluvial transport of dissolved and suspended solids, of the chemical weathering processes and the atmospheric/soil CO_2 consumption and of the relationship between chemical weathering and soil removal rates. The TSS concentration was directly related to the discharge and influenced by rainfall, with higher concentrations recorded after rainfall events. However, the TDS concentration showed dilution behavior in a wet period. The annual specific flux of TDS was 60.97 t/km^2/yr, but after the atmospheric inputs and anthropogenic contributions (ca. 17 and 14%, respectively) this value was corrected to 41.85 t/km^2/yr and represents the fluvial flux related to the chemical weathering of rocks. The total annual specific flux of TSS was 45.59 t/km^2/yr, with a small portion derived from the anthropogenic contributions (ca. 4%). The chemical weathering process showed a tendency to monosiallitization ($R_E = 2.4$), with an atmospheric/soil CO_2 consumption rate of 2.3×10^5 mol/km^2/yr. The chemical weathering and soil removal rates were 7.2 and 29.8 m/Myr, respectively, indicating a soil thickness reduction. The present soil removal rate in the Sorocaba River basin was almost 3-fold higher than the Cenozoic soil removal rates, reinforcing that the human-landscape systems are complex and affect the natural denudation rates, and, consequently, the present landscape evolution in the State of São Paulo.

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