Relevance of tyre wear particles to the total content of microplastics transported by runoff in a high-imperviousness and intense vehicle traffic urban area.

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Abstract

Microplastics (MPs) are an emerging pollutant and a worldwide issue. A wide variety of MPs and tyre wear particles (TWPs) are entering and spreading in the environment. TWPs can reach waterbodies through runoff, where main contributing particulate matter comes from impervious areas. In this paper, TWPs and other types of MPs that were transported with the runoff of a high populated-impervious urban area were characterised. Briefly, MPs were sampled from sediments in a stormwater detention reservoir (SDR) used for flood control of a catchment area of ~ 36 km^2, of which 73% was impervious. The sampled SDR is located in São Paulo, the most populated city in South America. TWPs were the most common type of MPs in this SDR, accounting for 53 % of the total MPs; followed by fragments (30 %), fibres (9 %), films (4 %) and pellets (4 %). In particular, MPs in the size range 0.1 mm-0.5 mm were mostly TWPs. Such a profile of MPs in the SDR is unlike what is reported in environmental compartments elsewhere. TWPs were found at levels of 2,160 units/(kg sediment·km^2 of impervious area) and 87.8 units/(kg sediment·km street length); MP and TWP loadings are introduced here for the first time. The annual flux of MPs and TWPs were $7.8x10^{11}$ and $4.1x10^{11}$ units/(km^2·year), respectively, and TWP emissions varied from 43.3 to 205.5 kg/day. SDRs can be sites to intercept MP pollution in urban areas. This study suggests that future research on MP monitoring in urban areas and design should consider both imperviousness and street length as important factors to normalize TWP contribution to urban pollution.

Keywords: Microplastic pollution; High urbanization; Imperviousness; Environmental management.

1. Introduction

Microplastics (MPs) are defined as plastic particles ranging from 1 µm to 5 mm (Frias and Nash, 2019). They commonly originate from products such as textile, plastic waste, pigment particles or some personal care products (Skaf et al., 2020). The MPs have different sizes, shapes, colours, compositions and density, and the well-established high stability of polymer constituents of MPs, increases and prolongs their negative impact in the environment (Anbumani and Kakkar, 2018; Ma et al., 2020). Furthermore, MPs may transport toxic organic chemicals, heavy metals and microorganisms that could affect aquatic organisms, and some have the potential for bioaccumulation and biomagnification (Ma et al., 2019; Kumar et al., 2021).

MPs tend to be at high levels in areas with large population worldwide (Bronwe et al., 2011; Vaughan et al., 2017; Jiang et al., 2019, Nematollahi et al., 2022), due to the generation of high amounts of litter with non-appropriate waste disposal and high number of vehicles (Li et al., 2020; Koutnik et al., 2021). The escalation in population density, associated with urbanisation, is associated with an increase in impervious areas (Ramezani et al., 2021; Kawakubo et al., 2019; Wu et al., 2020), which subsequently affects the response of an area after a rainfall events, typically increasing runoff in the area (Miller et al., 2014; Huang et al., 2008; Jacobson, 2011). Runoff from storms washes out impervious surfaces in densely populated areas, carrying the previous build-up of plastics and its degradation products into water bodies (Triebskorn et al., 2019; Grbić et al., 2020; Lange et al., 2021; Smyth et al., 2021).

Brazil is considered one of the most urbanised medium-income countries in the world (Lima and Rueda, 2018). São Paulo, with 12,396,372 inhabitants (Brasil, 2022), is the largest city in Brazil, second largest city in Latin America and the sixth most populous city in the world. São Paulo has an urbanisation rate of 99.1 % (Collaço et al., 2019). When urbanisation arises from a poorly controlled occupation process, as in São Paulo, impervious surfaces reach high levels (above 50 %) (Martins et al., 2018), resulting in high volume runoffs and frequent floods (Lima et al., 2018; Simas and Rodrigues, 2020). Flood events are intensified by climate conditions, and these promote transport of MPs.

Stormwater detention reservoirs (SDRs) have been used as an engineered solution to minimise impacts of flooding (Santos and Mazivieiro, 2016), by equalising the runoff volume during rainfall events (Szelag et al., 2019), especially in South-Eastern Brazilian states. After a rainfall event, the accumulated rainwater is pumped out of SDRs back to the river, and this leaves different solid materials remaining at the bottom of the reservoir. SDRs differ across previous studies (Lin et al., 2021; Niu et al., 2022; Koutnik et al., 2022) because these SDRs relate to different impervious surface and operational conditions. SDRs were indicated by the team as potential hotspots for MPs, which are found in large number and variability in such sites (Moruzzi et al., 2020).

While fibres are reported by different monitoring studies to be the dominant MPs in urban areas (Nematollahi et al 2022), tyre wear particles (TWPs) have the potential to become one of the largest type of MPs[1]. In addition, TWPs have been found in the environment and can be a risk to ecosystems and human health (Wik and Dave, 2009; Halle et al., 2021). This paper primarily assesses the relevance of TWPs to the total content of MPs in an urban area with high imperviousness and vehicle traffic. This is an urban characteristic that can be relevant to the understanding of different pathways taken by MP pollution to the environment that has not yet been addressed in previous studies. For this purpose, samples of sediments transported by urban runoff were collected from the bottom of the Jardim Arize SDR, municipally of São Paulo, and MPs, including TWPs, were characterised.

[1] Here we adopt the general definition of MP proposed by Verschoor (2015), which accounts MP as all man-made macromolecular material, including synthetic rubber.

2. Material and Methods

2.1. Study area description

The city of São Paulo created the Flood Control Programme in the Aricanduva River basin in the 1990s as a way of reducing and/or minimizing the impacts of floods over vulnerable population during intense rainfall events in the region. This involved implementation of flood storage reservoirs on or adjacent (in-line or off-line types) to the Aricanduva River and its tributaries. The Aricanduva River basin has high impervious surface rates and a long history of flooding (Simas and Rodrigues, 2020). In addition, the traffic in the area reaches one the highest loading of vehicles in São Paulo city with values as high as 24,700 vehicles/day in the main arterial road only (CET, 2019).

The Jardim Arize SDR (23°33'42.10" S and 46°30'35.92" W), built in 2002 (Canholi, 2014), is located in the Aricanduva River basin. This SDR drains an area of 36.437 km^2 (Fig. 1), with 390,614 inhabitants and an average of demographic density and Human Development Index (HDI) of 10,723 inhabitants/km^2 and 0.79, respectively (São Paulo, 2017). The Jardim Arize SDR is an engineered structure, which operates as an off-line SDR with total capacity of 160,000 m^3 (17,750 m^2 and average depth of 9 m). It was designed to reduce the Aricanduva River flow from 94 m^3/s to 75 m^3/s, for a rainfall of 10 years of return period (TR) (Canholi, 2014). Stormwater comes from a side weir, positioned in the middle length of the SDR. The weir is set at a level that watercourse can accommodate flow at normal period. During a rainy period, any additional flow, established from the maximum level for critical stormwaters, passes over the weir into the SDR. The flow then spread quickly over the SDR area, and stays until the end of the critical rainfall period, where detained water is pumped out back to the watercourse. The sediment brought by the flow remains at the SDR bottom and is scratched and put aside for removal. Lorries can then remove the sediment to a controlled landfill as final destination. The cycle is then repeated for new critical stormwaters and the frequency of cleaning depends upon the amount of sediment carried by the runoff. Figure 2 shows the schematic sequential operation for the Jardim Arize SDR. The climate of the study area is classified as humid tropical, with wet summer (October to March) and dry winter (April to September) (Simas et al., 2017).

2.2. Land use description, hydrological settings and SDR sediments

The land-use mapping of the catchment area was performed as outlined by Lupinacci et al. (2017, 2022) and Couto Júnior et al. (2019), consisting of a visual interpretation of satellite images acquired from Sentinel 2A. The following land-use classes were mapped: (i) water bodies, (ii) permeable areas (reforested areas, exposed soil and unoccupied areas), and (iii) impervious areas (residential/commercial and industrial areas), which are areas of intensive use and include buildings and road systems. The total street length was also calculated from the land use mapping.

The region of São Paulo city has the average annual rainfall between 1300 mm and 1500 mm (Lima et al, 2018). The average monthly and annual rainfall in the study area was quantified using data from the E3-035 rainfall station (23º39'04" S and 46º37'2" W) from 1936 to 2019 (DAEE, 2020). The rainfall intensity for different TR (in years) was obtained using Equation 1 (DAEE, 2018), for $10 \leq d \leq 1440$ min.

$$I = 32.77(d+20)^{-0.8780} + 16.10(d+30)^{-0.9306}\left[(-0.4692 - 0.8474Ln(Ln\left(\frac{TR}{TR-1}\right)\right] \quad (1)$$

where I is rainfall intensity (mm/min) and d is rainfall duration (min).

Rainfall intensities, responsible for the runoff and washing out of MPs and TWPs, for the sampling period were assessed. For this, data from two rainfall stations, namely Interlagos

(23º43'28” S and 46º40'39” W) and Henry Borden (23º42'11” S and 46º40'26” W) were evaluated from January to August 2019, according to the dataset available at Water and Energy Department’s database (DAEE, 2020). Finally, data related to the composition of residues, collected monthly from the studied SDR, in tonnes (AMLURB, 2022), were used to quantify the amount of accumulated sediment.

2.3. Sampling and microplastics separation

Sediment samples were collected from Jardim Arize SDR (23°33'42.10" S and 46°30'35.92" W) (Fig. 3a). The reservoir operates intermittently, according to the runoff flow, and samples were taken when the Jardim Arize SDR was without water (in August 2019). Two superficial sediment samples (2 kg) were collected at the bottom of SDR (Fig. 3b), equidistant 50 m from the weir, corresponding to the 2nd and 3rd quartile for length, at the central position of SDR’s width, covering the entire depth (~ 50 cm). A preliminary analysis did not find difference ($p<0.05$) in the profile of MPs across sampling positions. Also, due to the operational characteristics in the Jardim Arize SDR, every sediment sample collected results from the transport of sediment from various rainfall events. Hence, the content of the SDR is itself a composite sample (Fig. 2). The samples were then stored in sealed glass containers and transported to the laboratory.

The sediment samples were dried at room temperature and from each sample, three representative fractions (30 g each) were collected, totalising six samples where MPs and TWPs were quantified. Each sample was added to a glass beaker with 300 mL of a solution containing water and $ZnCl_2$ (> 98 %, from Sigma-Aldrich, Brazil) dissolved at a density of 1.6 g/cm^3. The mixture was manually homogenized, transferred to the Imhoff cone and after 24 h of sedimentation, the supernatant water was filtered through a 0.45 μm cellulose nitrate membranes filter with 47 mm of diameter (Sartorius, Brazil). A blank sample consisting of the same amount of free-plastic soil was treated in parallel to monitor potential contamination by new microplastics from the laboratory environment during the procedure.

2.4. Quantification and characterization of microplastics

After use, the membrane filters were placed in closed glass petri dishes for drying (around 23 °C for at least 24 h) and subsequent quantification and characterization of all MPs and TWPs, followed the procedures outlined by Moruzzi et al. (2020). The TWPs, among the MPs found in the sediment samples, were identified using visual criteria, as performed by Leads and Weinstein (2019). TWPs are black particles, elongated/cylindrical in shape, may be partially covered with other road impurities, and have a rough surface and rubber consistency, which remains even when handled with tweezers. MPs and TWPs were visually identified and counted using a stereo microscope (Zeiss Discovery V12 SteREO, Germany) with an integrated camera (AxioCam ER 5s, Germany). The particles were classified into categories according to their type (fibres, films, fragments, pellets and tyres) and size (0.1 mm to 0.5 mm, 0.5 mm to 1.0 mm and 1.0 mm to 5.0 mm). Particles lower than 0.1 mm were not considered because of limitations of the light microscope used. MPs in the samples were counted as the number of MPs per kilogram of sediment (units/kg). Preliminary tests were set to confirm the main TWP characteristics, and colour was found to be a remarkable trait for visual inspection. Random samples of black particles were then taken and TWPs were confirmed by ATR FT-IR (Varian 640-IR, Netherlands) with spectra from 400 – 4000 cm^{-1}, comparing the spectra obtained with an online database (Bio-Rad Sadtler, Brazil). Additionally, the morphology and composition (presence of sulfur) of TWPs was characterized using Scanning Electrical Microscopy with Energy Dispersive X-Ray Spectroscopy (SEM-EDS) (JEOL - JSM-6010 LA, USA), which confirmed the presence of sulfur in the specimens.

2.5. Annual flux and emission factor

The annual flux of MPs (F_{MPs}) and TWPs (F_{TWPs}) were determined using Equation 2, considering the total loading of sediments to SDRs for the catchment area.

$$F_{MPs}\ or\ F_{TWPs} = \frac{C.S}{A}.10^3 \quad (2)$$

where F_{MPs} and F_{TWPs} stand for annual flux of MPs and TWPs [units/(km^2.year)], respectively; S is the total mass of sediments (t/year); C is either the MPs or TWPs (units/kg); A is the catchment area (km^2), being the ratio C/A the loading of MPs or TWPs [units/(kg.km^2)].

The TWP emission factor within the catchment area (T_{TWP} in kg/day) was estimated through the Equation 3 (Järlskog et al., 2020). The F_{tw} for the type of vehicle was based on Kole et al. (2017) and NV obtained from CET (2019), based on 13 peak hours for the study area. The peak hours represent the time interval between 7 am and 8 pm, where most of the vehicle traffic concentrates. For the L_C, only the arterial roads, which receive the highest volume of vehicle traffic, were considered.

$$T_{TWP} = F_{tw}.NV.L_c.10^3 \quad (3)$$

where F_{tw} is the emission factor of tyre wear particles [kg/(vehicle.km)]; NV is the annual average of daily traffic (vehicles/day); and L_C is the street length of arterial roads within the catchment area (km).

3. Results

3.1. Land use mapping

The land-use mapping of the catchment area is presented in Figure 4. The residential/commercial areas (52.67 %) are the main land-use in the study area, followed by industrial areas (20.37 %) and reforest (15.32 %) (Table S1). The impervious area was 26.60 km² in 2019, which represented ~ 73 % of the total study area. In 2019, the total street length of the study area was 640 km, with a street density of 24 km/km². The arterial roads sum 65.9 km and cross the catchment area from East to West mostly.

3.2. Rainfall characteristic of the catchment area

The annual rainfall average was 1,437.1 mm, with the annual rainfall values ranging from 887 mm (1963) to 2,228 (1983) in the study area over 80 years (Fig. S.1a). The monthly average rainfall was 119.9 ± 68.9 mm, with January (237.6 ± 91.1 mm) being the rainiest month and August (37.8 ± 33.0 mm) the driest (Fig. S.1b). Figure S.1c, derived from Equation 1, indicates rainfall intensities expected for different durations and TRs. For TR between 2 and 10 years, rainfall intensities up to 63 mm/h are expected, with duration of 60 min, while ~ 94 mm/h are estimated for TR of 10 years and duration of 30 min. Lower intensity rainfalls, with intensities < 54 mm/h, are more likely to occur with TR values (TR < 5 years). The maximum registered rainfall intensity during the sampling period was 56 mm/h, corresponding to TR of 6 years (Table S.2), with 84 % chance to be equalled or surpassed within 10 years.

3.3. SDR's sediments

The monthly average mass of sediments removed from the SDRs in São Paulo is shown in Figure S.2, where the data was compiled from AMLURB (AMLURB, 2022). The quantity of sediments removed from the SDR was 15,900 ± 976 tonnes/month on average, ranging from 14,280 ± 1,680 tonnes (in January, average 2013-2020) to 18,421 ± 6,150 tonnes (in December, average 2013-2020). The sediments are collected by the municipal cleaning service and deposited in controlled landfills engineered with necessary civil structures to prevent soil and groundwater contamination.

3.4. MP quantification and characterisation

The total amount of MPs (unit/kg), including TWPs, found in the Jardim Arize SDR is presented in Table 1. Figure 5 shows the distribution of MPs by type and size. MPs in the sediment samples from the Jardim Arize SDR were 109,089 units/kg, with TWPs being the most common MPs (57,461 units/kg), followed by fragments (32,456 units/kg), fibres (10,022 units/kg), films (4,622 units/kg) and pellets (4,528 units/kg) in that order. Figure 6 shows representative examples of different MPs. Optical microscopy images (Fig. 7a and 7b) and SEM micrographs (Fig. 7c) illustrate typical rough surfaces of TWPs. In addition, the characterization with FTIR confirmed that the particles identified were tyre fragments (Fig. 7d). The MPs detected on the Jardim Arize SDR were mainly 0.1 to 0.5 mm (~ 90 %), followed by MPs ranging from 0.5 mm to 1.0 mm (~ 6 %) and 1.0 mm to 5.0 mm (~ 4 %), being 0.1 mm the lowest size that the methodology used in this work allowed to study. TWPs prevailed with 57 % of the MPs size ranging from 0.1 mm to 0.5 mm.

3.5. *Annual flux and emission factor*

Based on the average of 190,800 t of sediment/year deposited on the SDRs (Fig. S.2), compiled from the database presented by AMLURB (2022), the annual flux of MPs and TWPs were $7.8\text{x}10^{11}$ and $4.1\text{x}10^{11}$ units/(km^2.year), respectively. For the T_{TWP}, Brazilian F_{tw} values for mass of tyre debris per vehicle and kilometre were used for calculation, according to Kole et al. (2017): 0.132 g/(vehicle.km) for automobiles, 0.007 g/(vehicle.km) for motorcycles, 1.068 g/(vehicle.km) for trucks and 0.204 g/(vehicle.km) for buses. The *NV* was considered in the range of 5,200 – 24,700 vehicles/day (CET, 2019). The percentage of each vehicle category was 79.10 % for cars, 15.74 % for motorcycles, 1.37 % for trucks and 2.99 % for buses. The L_C was 65.91 km within 640 km. Finally, the T_{TWP} was estimated from 43.3 to 205.5 kg/day depending on the daily vehicle traffic.

4. Discussion

4.1 Rainfall characterisation and SDR sediments for the sampling period

Figure S.2 indicates that the average amount of sediments removed monthly from the SDRs in São Paulo city from 2013 to 2020 had low variation with time. As such, sediments that accumulated at the bottom of São Paulo SDRs were approximately removed at a constant rate, regardless of the season, following operational and logistic arrangements. While this can be considered a disadvantage for a paired relation analysis between rainfall and sediment transport, it provides a more homogeneous sample for the assessment of the long-term accumulation of MPs and allows for a more consistent evaluation of representative samples.

4.2 Comparison of total amount of microplastics in different sites in Brazil and elsewhere

A summary of the total amount of MPs identified in sediments from sites close to urban areas (in Brazil and elsewhere) is presented in Table 2. Previous studies indicate great variation in the number of MPs, depending on the sampling point. For instance, MPs measured by Zheng et al. (2020) in China was ≤ 20 units/kg, whilst values ranging from ≤ 80 to ≤ 3,763 units/kg were found in streams and river from New Zealand (Dikareva and Simon, 2019) and Germany (Klein et al., 2015). On the other hand, ponds and lakes may present values as high as ~ 28,000

(Liu et al., 2019) or 128,000 units/kg (Ballent et al., 2016). The reason for such variation comes not only from social and environmental aspects (Ballent et al., 2016; Zhang et al., 2020), but also specific characteristics of the water bodies and reservoirs sampled. The volume of water bodies and reservoir characteristics are expected to bring down the total amount of MPs brought by runoff. Rivers, lakes and ponds may have multiple inlet contribution (tributaries) as well as having multipurpose uses (e.g. drinking water source, energy generation, etc).

A previous study showed MPs in a Brazilian SDR of ~ 57,500 units/kg (Moruzzi et al., 2020), whereas the present work has found ~ 109,000 units/kg, possibly because the catchment area and land use differ from one another. Although the levels of MPs found were very different than this study, the high total content of MPs on the SDR confirms that stormwater reservoirs are hotspots of MPs and should be more investigated and used to intercept MP pollution. The presence of MPs in multipurpose reservoirs not designed specifically for flood control but for other uses - such as water supply, irrigation and power generation - has also been reported (Niu et al., 2022; Di and Wang, 2018; Lin et al., 2021). Furthermore, unlike most of SDRs deployed in the municipally of São Paulo, these reservoirs are not impervious tanks, so the particles can infiltrate into soil. Therefore, SDRs are adequate for the assessment of the abundance and characterization of MPs transported by runoff in impervious catchment areas, as they are designed to contain flood and built with an impermeable bottom (Santos and Mazivieiro, 2016), thus acting as a barrier for these particles (Moruzzi et al., 2020).

4.3. Different sizes and types of MPs

Except for pellets, which were exclusively found in the size ranging between 0.1 and 0.5 mm (where 0.1 mm was the smallest MPs analysed), the distribution of MPs in this lowest size range constitutes 90 % of the total MPs. This is in accordance with previous investigations in sediment samples reported by Moruzzi et al. (2020), Lin et al. (2021), Zheng et al. (2020)

and Nematollahi et al. (2022). In addition, TWPs prevailed in the size ranging from 0.1 to 0.5 mm, as they are more easily transported from pathways by high-frequency surface runoff, while larger particles (> 0.5 mm) require higher flows and can also be trapped in the environment close to the pathways where they are generated (Järlskog et al., 2021; Klöckner et al., 2020).

It is noticeable that a great number of MPs come from TWPs. TWPs were the main contributors to MPs in the studied SDR. Relevance of TWPs in the total content of MPs has been described by few studies in Australia (Ziajahromi et al., 2020), Sweden (Järlskog et al., 2021) and USA (Leads and Weinstein, 2019; Werbowski et al., 2021). TWPs are generated by the friction between the tyre and the road surface (Knight et al., 2020) and are made from natural and synthetic rubber (polyisoprene or styrene-butadiene rubber) (Järlskog et al., 2020). The magnitude of TWP release is related to the number of vehicles (Wik e Dave, 2009) and depends upon the tyre and road characteristics (composition and conservation conditions), imperviousness of the catchment area and environmental factors, such as weather conditions and driving style (Zhang et al., 2020; Yan et al., 2021). Street length can play an important role on emission levels of TWPs alongside the traffic load, which is why normalized pollution levels are herein proposed. Finally, atmospheric movement and deposition may also affect the distribution of MPs and TWPs (Koutnik et al., 2022) (Sun et al., 2022; Li et al., 2022; Koutnik et al., 2021). However, it is likely that wet regions are more prone to be influenced by TWPs from emissions from traffic, where runoff may modulate MP transport during rainy period, as shown by de Carvalho et al. (2022).

Fragments were the second most abundant MP type (30 %) found in the Jardim Arize SDR. Large amounts of MP fragments (from 50 to 70 %) have also been reported by several authors (Moruzzi et al., 2020; Ballent et al.,2016; Dirakeva and Simon, 2019; Niu et al., 2022). Given that TWPs can also be classed as fragment, some discrepancies in the proportions of the type of MPs were expected. Since fragments originate from the degradation of solid waste and other larger plastics (Horton et al., 2017; Akdougan and Guven, 2019), their presence in greater amounts in densely populated places, where there is also greater use of plastic products, is expected (Browne et al., 2011; Jiang et al., 2019).

Fibres accounted for 9 % of the total MPs observed in this study, which is consistent with the value reported by Moruzzi et al. (2020) in another Brazilian SDR, in the municipally of Poa. Their abundance increased with size (0.5 mm to 1.0 mm, 35 %), becoming predominant in the range of 1.0 mm to 5.0 mm (55 %). Fibres have been the dominant MPs in other urban areas, such as in Three Gorges Reservoir sediments, China (Di and Wang, 2018), in Lake Bolsena and Chiusi sediments, Italy (Fischer et al., 2016), in Jiaozhou Bay sediments, China (Zheng et al., 2020), in Danjiangkou Reservoir sediments, China (Lin et al., 2021), in Ahvaz soil samples, Iran (Nematollahi et al., 2022) and in Tibet Plateau sediment, China (Jiang et al. (2019). The sources of fibres are mainly textile products, such as clothing and carpets (Bronwe et al., 2011), mainly due to the discharge of effluents from washing machines (Dris et al., 2018). Therefore, higher percentages of fibres are expected in combined systems or separated sewage systems. However, since the beginning of the 20th century, Brazil has had separated systems, i.e. stormwater and sewage are collected by different pipes. This may show that SDRs in urban areas are capturing different pollution aspects from other types of reservoirs, soils, lakes, rivers and bays.

Films, accounting for 4 % of total MPs (Fig. 5a), were more prominent in the larger defined size ranges of MPs (representing 18 % in the range of 0.5 mm – 1.0 mm and 17 % within 1.0 – 5.0 mm). Films are secondary MPs from plastic waste, with a characteristic thin layer morphology, generated from the fragmentation of packaging materials and plastic containers (Di and Wang, 2018), and are also expected in greater abundance in places with high population density and use of plastic products.

Finally, pellets were among the lowest concentrations on MPs measured in this work (4 %) and is in agreement with the literature (Rodrigues et al., 2018; Lin et al., 2021; Di and Wang, 2018; Jiang et al., 2019; Moruzzi et al., 2020). However, is not in keeping with the well-established greater importance that pellets/beads acquired in the mass media communication, and their subsequent restructured use in personal care products in some countries. Indeed, pellets primarily come from cosmetics industry and hygiene products (Di and Wang, 2018; Jiang et al., 2019).

4.4. Influence of imperviousness, street length and vehicle traffic on the content of MPs and TWPs

MP pollution, like other type of pollution, tends to be high in urban areas (Vaughan et al., 2017), and there is a clear variation in the characteristics of such pollution between rural and urban areas (Di and Wang, 2018). In addition to the input and movement of contaminants (Grbić et al., 2020), the structure of the urban areas and vehicle traffic influence the quantity and transport of MPs in the area (Järlskog et al., 2021). Urbanization is also related to the increase of impervious surfaces (Rameziani et al., 2021; Wu et al., 2020) and total street length (Peponis et al., 2007), both being factors with high potential to influence the transport of MPs, including the total content of TWPs.

The Aricanduva watershed has a high-impervious surface, high demographic density, and also 640 km of street length. These characteristics are similar to other parts of São Paulo for low-income occupation (Sobrinho and Tsutiya, 1999), but may be very different from wealthier areas that have better infrastructure and lower demographic density. Both, imperviousness and street length are related to tyre wear. They both may help to understand TWP pathway from similar urban conditions in Brazil or elsewhere. In this study, the total amounts of MPs and TWPs were normalized by the total impervious area (km^2) and the total street length (km), resulting in 4,101 units/(kg·km^2) and 170.5 units/(kg·km) for MPs, and 2,160 units/(kg·km^2) and 87.8 units/(kg·km) for TWPs. Such normalized data has not been presented in previous studies and this work constitutes the first recommendation for MPs in urban environments.

In addition, the abundance of MPs and TWPs in the Jardim Arize SDR ranging from 0.1 mm to 0.5 mm may be explained by the high residential imperviousness area and frequency of moderate to low rainfall intensities (low TR), with small particles being easily transported by low overland flows. Previous studies have also shown the importance of runoff for the transport and occurrence of MPs in areas with high impervious surface (Hong et al., 2016; Strobach et al., 2019; Triebskorn et al., 2019; Grbić et al., 2020; Wu et al., 2020; Lange et al., 2021; Rameziani et al., 2021; Smyth et al., 2021). High-impervious areas such as Jardim Arise increase the effect of runoff on MPs and TWPs transport, and also facilitates the interpretation of the MPs pollution at SDRs.

The annual flux of MPs and TWPs were 7.8×10^{11} and 4.1×10^{11} units/(km^2.year), respectively, for 190,800 t of sediment/year deposited in SDRs (AMLURB, 2022). Although from different sources, the annual flux obtained has the same order of magnitude as those presented by Dris et al. (2018). The lack of data on this topic is still a challenge and the heterogeneity of sources and types of MPs makes comparisons difficulty. However, the results presented here may assist future researchers in assessing MP pollution. Finally, the T_{TWP} values estimated in this study (43.3 to 205.5 kg/day), based on local data, are much higher than the 0.81 kg/year reported by Kole et al. (2017), who considered the TWP generation data, global vehicles and population. Apparently, in densely populated areas, such as the city of São Paulo, there is a significant increase of the TWP emission due to imperviousness and street length, associated with the vehicle traffic. Future research should address the effect of the type of road on the emission of TWP.

5. Conclusions

The study area located in the municipally of São Paulo covered 36 km^2, 73 % of which had impervious surface and 640 km of street length. TWPs were the main fraction (53 %) of MPs in SDR sediments from street runoff, and their abundance can be related to the characteristics of the contributing area such as high imperviousness and street density. Most of the TWPs counted were within the lowest range examined (0.1 mm to 0.5 mm), and this can be attributed to their easy transportation by surface runoff as a result of the frequent rainfall events. TWPs at levels of 2,160 units/(kg·km^2) of impervious area and 87.8 units/(kg·km) of street, and TWP emissions ranging from 43.3 to 205.5 kg/day, demonstrate the significance of this kind of pollution in urban areas and are, for the first time, measured as loading by unit of area or street length. The annual flux was $7.8x10^{11}$ units/(km^2.year) for MPs among which $4.1x10^{11}$ units/(km^2.year) were for TWPs only. It is expected that MP and TWP pollution similar to what has been found in the Jardim Arize SDR will be present in other urban areas with similar land-use, around the world. Furthermore, we propose that further investigations consider the imperviousness, street length and vehicle traffic as important features for runoff transport of TWPs in urban areas and recommend SDRs as a strategy to collect and reduce MP pollution in the environment.

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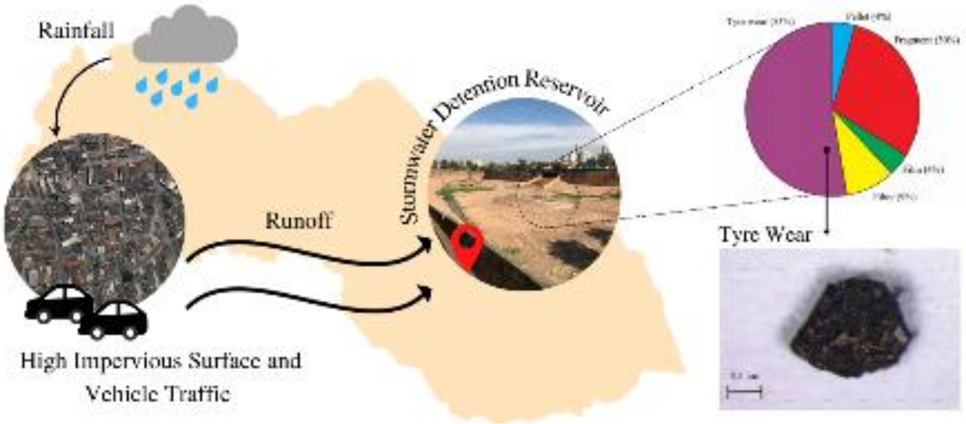
Graphical Abstract and Figures



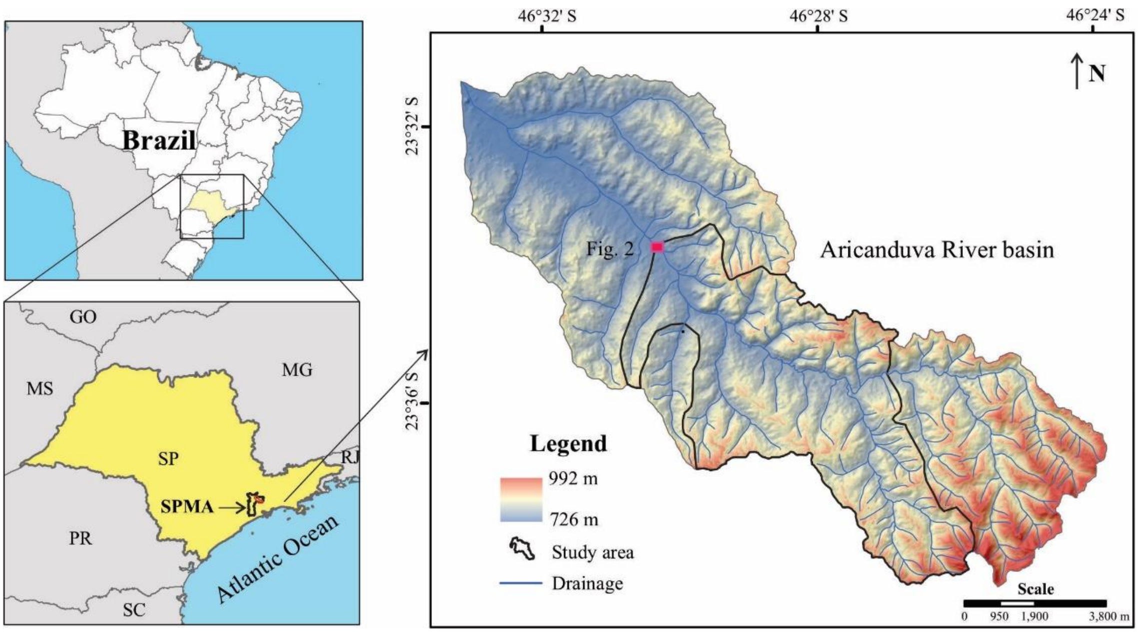


Figure 1 – Maps of the São Paulo Metropolitan Area (SPMA) (left) with digital elevation model (USGS/SRTM3”) using 30 m of resolution for Aricanduva River basin (right). The Aricanduva River basin contributes to runoff into SDR.

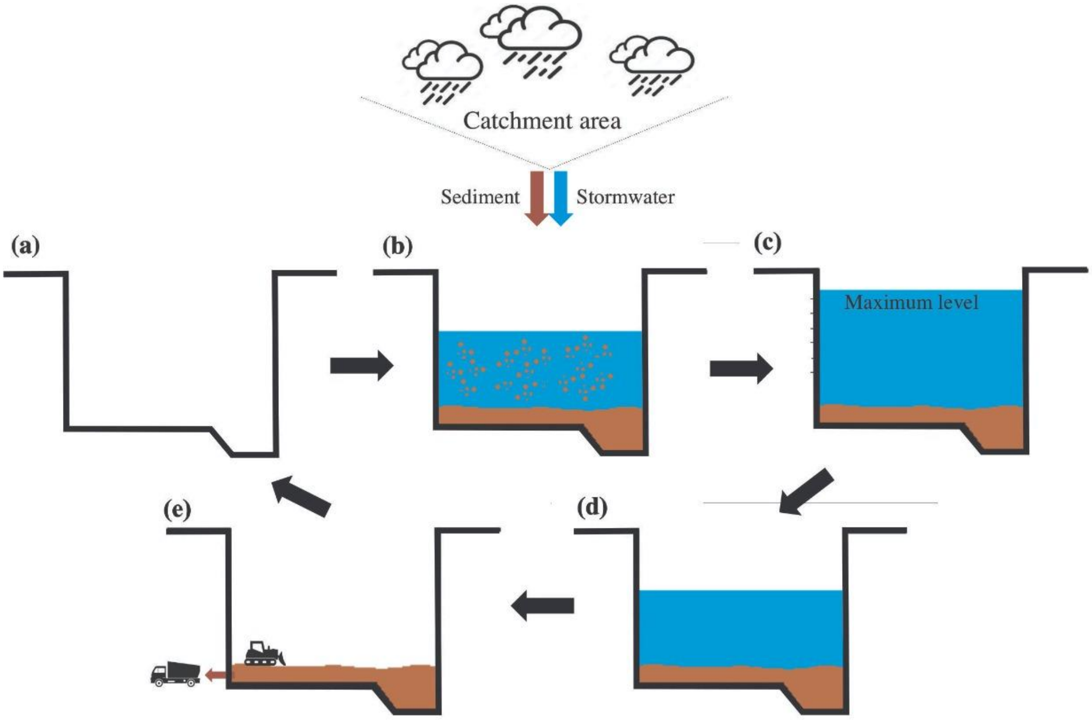


Figure 2 – Schematic sequential operation for the off-line Jardim Arize SDR. Empty SDR (a) receives runoff from side weir (b) up to the stablished maximum level (c) from where water is pumped out back to river (d), and remaining sediment is scratched and put aside, where the sediment is removed to a controlled landfill. The frequency of cycles depends upon the rainfall and the amount of sediment carried by runoff.

Figure 3 – Jardim Arize SDR marked with continuous red line, with the image from Google Earth Pro - 06/28/2020 (a). Overview of the Jardim Arize SDR, with the sediment scratched and put aside for removal (red circle) (b). Detail of the circled area (c).

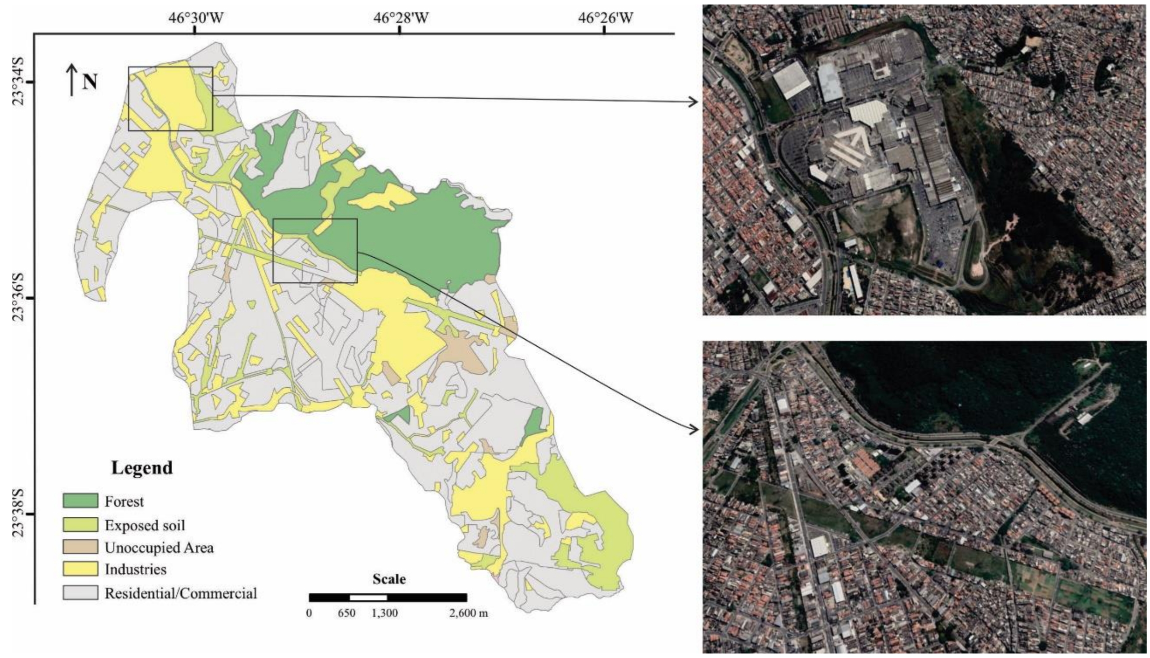


Figure 4 - Land use mapping of the catchment area, with details extracted from Google Earth Pro - 28/06/2020.

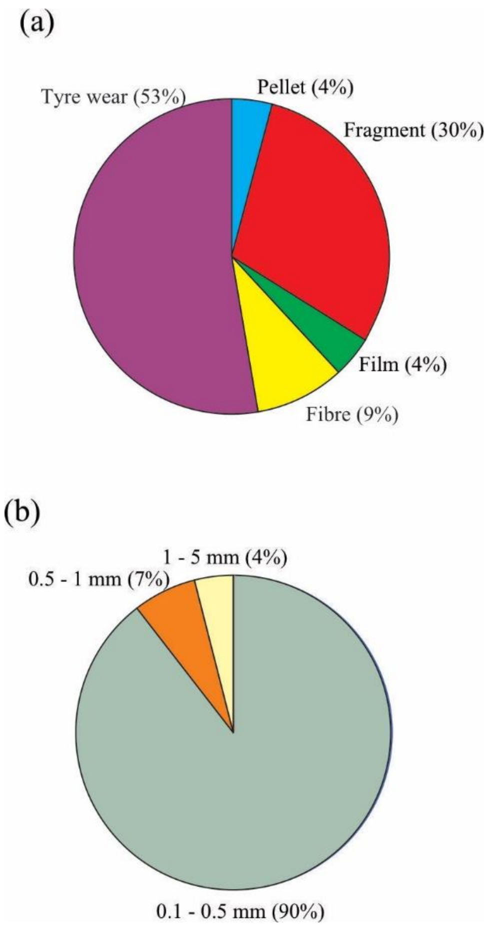


Figure 5 – Distribution of MPs, considering type (a) and size (b), in sediments collected in the Jardim Arize SDR.

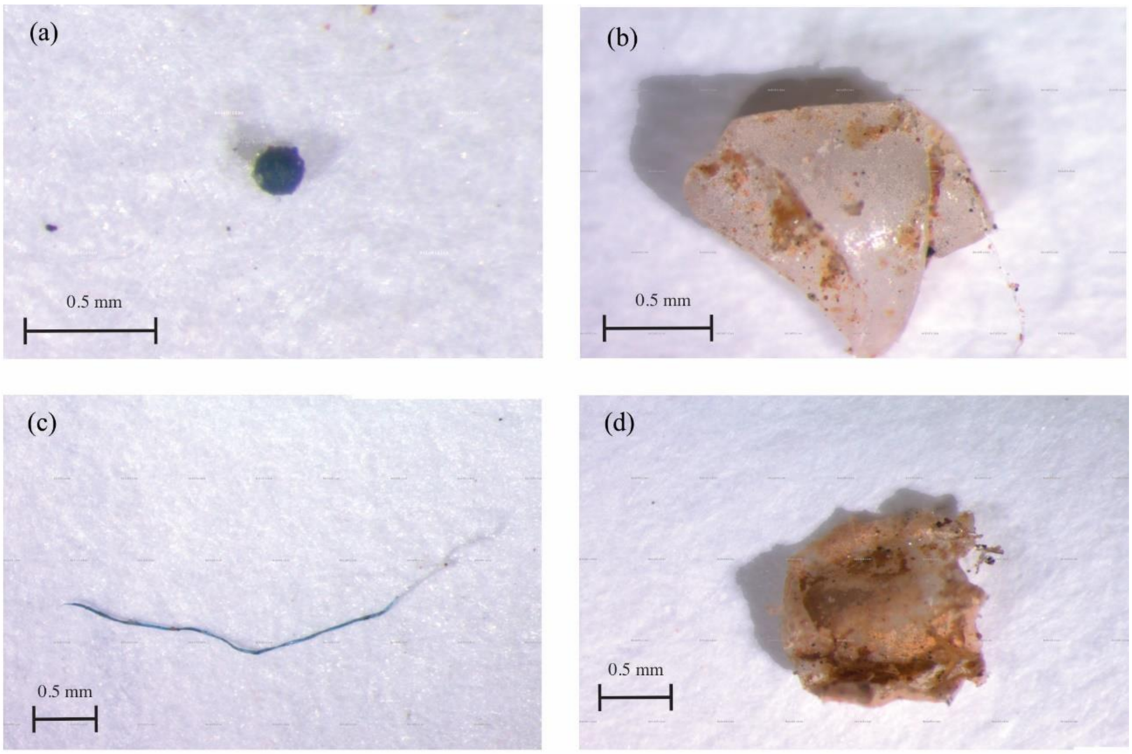


Figure 6 – Examples of different types and shape of MPs in sediments collected in the Jardim Arize SDR: pellet (a), fragments (b), fibre (c) and film (d).

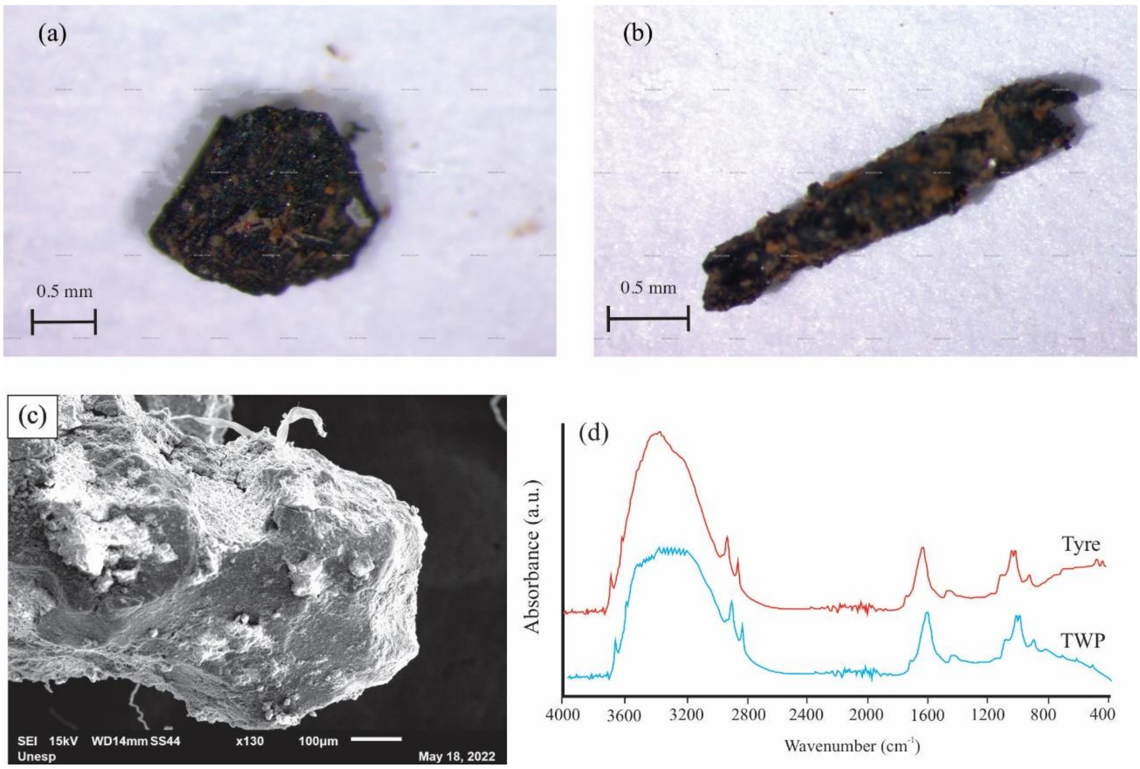


Figure 7 – Examples of MPs identified as TWPs in the Jardim Arize SDR. Optical microscopy images (a and b) and SEM images showing the morphology of TWPs (c). ATR FTIR spectra comparing tyre standard with TWPs (d).

Table 1. Characteristics of MPs found in sediments collected in the Jardim Arize SDR classified by type and size (units/kg). The values in brackets correspond to relative standard variations from three sediment samples, where each sample was 2 kg taken from the surface of the bottom of the SDR.

Type\Size	**0.1–0.5 mm**	**0.5–1 mm**	**1.0–5.0 mm**	**Total (units/kg)**
Tyre wear	55,294 (±46%)	1800 (±33%)	367 (±71%)	57,461 (±45%)
Fragment	30,089 (±41%)	1550 (±39%)	817 (±90%)	32,456 (±41%)
Fibre	5122 (±71%)	2506 (±55%)	2394 (±60%)	10,022 (±47%)
Film	2622 (±47%)	1250 (±78%)	750 (±64%)	4622 (±52%)
Pellet	4528 (±38%)	–	–	4528 (±38%)
Total	**97,656 (± 38%)**	**7106 (± 33%)**	**4328 (± 65%)**	**109,089 (± 38%)**

Table 2. MPs (units/kg) in sediments sampled in urban areas found in previous investigations and this study.

Location	**Country**	**MPS (units/kg)**	**Reference**
Jiaozhou Bay	China	≤27	Zheng et al. (2020)
Streams in Auckland	New Zeland	≤80	Dikareva and Simon (2019)
Bloukrans River	South Africa	≤160	Nel, Dalu and Wasserman (2018)
Tibet Plateau	China	≤195	Jiang et al. (2019)
Lake Bolsena and Chiusi	Italy	≤266	Fischer et al. (2016)
Three Gorges Reservoir	China	≤300	Di and Wang (2018)
Edgbaston Pool	UK	≤300	Vaughan et al. (2017)
Beijiang River	China	≤544	Wang et al. (2017)
Antuã River	Portugal	≤1265	Rodrigues et al. (2018)
SCMs in Los Angeles	EUA	≤2784	Koutnik et al. (2022)
Lake Poyang	China	≤3153	Liu et al. (2019a)
Danjiangkou Reservoir	China	≤3237	Lin et al. (2021)
Rhine e Main River	Germany	≤3763	Klein et al. (2015)
Jiayan Reservoir	China	≤15,700	Niu et al. (2022)
Lake Ontario	Canada	≤28,000	Ballent et al. (2016)
Poá SDR	Brazil	≤57,542	Moruzzi et al. (2020)
Stormwater Pound	Denmark	≤127,986	Liu et al. (2019)
Jardim Arize SDR	Brazil	≤109,089	Present study

Supplementary Material

Table S.1 – Surface area by land use classes in the study area.

Land use	**Area (km^2)**	**Area (%)**
Water bodies	0.06	0.15
Reforest	5.58	15.32
Exposed soil	3.47	9.52
Unoccupied area	0.71	1.94
Residential/commercial	19.18	52.67
Industrial	7.42	20.37
Total	**36.43**	**100.00**

Table S.2 - Rainfall intensity (mm/hour) for a return period (TR) between 2 and 10 years and duration of 10, 20, 30 and 60 min for São Paulo.

Duration (min)	**TR (years)**			
	2	**5**	**6**	**10**
10	94.3	124.3	129.6	144.1
20	73.1	97.4	101.8	113.5
30	60.0	80.5	84.2	94.1
60	39.6	53.7	56.2	63.0

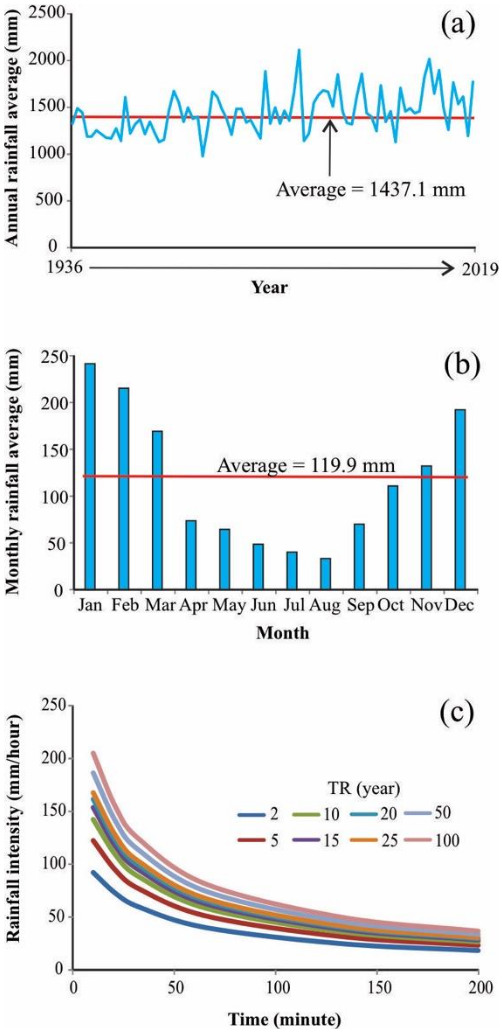


Figure S.1 – Annual (a) and monthly (b) rainfall average in the period between 1936 and 2019 (DAEE, 2020). Rainfall intensity with different return period (TR) (c).

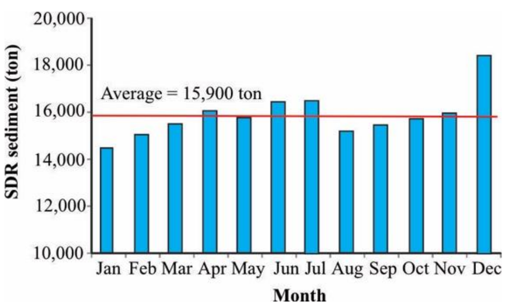


Figure S.2 – Monthly average of sediments removed from SDRs in São Paulo, between 2013 and 2020 (AMLURB, 2022).