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Rainfall and PM Removal on Tree Leaves: A Study of Santiago, Chile's Native Species

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Abstract: Green infrastructure, such as street trees, can help improve air quality, but the role of rainfall in cleaning total particulate matter (TPM) from tree leaves is not well understood, especially in cities like Santiago, Chile. This study measured TPM deposited on leaves and its elemental composition of two native tree species, *Quillaja saponaria* and *Schinus molle*, by five independent rainfall episodes. The results showed significant differences in how each tree species responded to rainfall. The total amount of TPM finally removed or retained at the leaf level in the five rainfall events studied was 4.72 and 8.43 mg/g_{ldw} for *Q. saponaria* and *S. molle*, respectively. *Q. saponaria* decreased TPM levels after rainfall, while *S. molle* exhibited mixed responses, increasing or decreasing TPM accumulation on leaves after different intensities of rainfalls. Elemental analysis revealed metals such as lithium and nickel—potentially linked to electric vehicle batteries—and tin and antimony—potentially linked to industrial processes. Rainfall benefited air quality by removing heavy metals from the atmosphere and aiding plant recovery from TPM accumulation. However, further research is needed on metal speciation in TPM and its foliar uptake by plants. This study provides some insights into the complex interactions between trees leaves, TPM deposition, and rainfall.



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Keywords: urban air pollution; street trees; *Quillaja saponaria*; *Schinus molle*; elemental analysis of TPM

1. Introduction

Urban air pollution poses a significant health problem, with an estimated 8.8 million premature deaths per year attributed to exposure to ambient air pollution, reducing life expectancy by 2.9 to 3.5 years on average [1].

Santiago, the capital of Chile, is a modern city and one of South America's most dynamic urban centres. Santiago is located 543 m above sea level with more than 6 million inhabitants, which concentrates much of the cultural economic, industrial and commercial activity of the country. Santiago boasts a well-developed infrastructure with old buildings and modern skyscrapers and public transportation, including an extensive metro system [2]. In Santiago, air pollution is exacerbated by both unfavourable weather patterns and geographical conditions. Major emission sources, such as transportation, industrial activities, and residential heating, contribute to poor air quality, increasing the population's exposure to air pollutants [3–6]. Transportation is responsible for 61.1% of the city's air

pollution, while residential firewood use contributes 37.1% [7]. The air quality in Santiago is particularly poor during the autumn and winter due to the city's geographical location, which is surrounded by mountains (between 2000 and 5000 m.a.s.l.) that trap pollutants and limit their dispersion [8–11]. As a result, there are high levels of particulate matter (PM_{2.5} and PM₁₀), which pose a severe health risk [8,12]. Compared to other capital cities in the region, the air quality in Chile is still higher. According to the last World Air Quality Report, the PM_{2.5} average annual concentration in Santiago was 21.3 μgm^{-3} , Lima 19.7 μgm^{-3} , Bogotá 13.4 μgm^{-3} , and Buenos Aires 9.6 μgm^{-3} [13].

Several studies have linked PM exposure to increased morbidity and mortality from respiratory and cardiovascular diseases [14,15]. Additionally, research conducted in Chile has shown a strong correlation between PM_{2.5} and respiratory issues, such as asthma and pneumonia [16–18].

In response to these health challenges, green infrastructure (GI) has emerged globally as a potential approach to urban planning to alleviate human exposure to air pollution [19–21]. Green infrastructure incorporates nature-based solutions to provide various environmental, social, and economic benefits known as ecosystem services [22,23]. Green infrastructure, such as, street trees, hedges/shrubs, green walls and roofs, wetlands, and parks, among other green spaces, can address diverse environmental and urban challenges if they are strategically incorporated into urban settings [24–27]. For example, GI can help with climate regulation, air pollution control, and improving both mental and physical health [28–36]. Moreover, several studies have demonstrated that, in particular, street trees can reduce PM in urban areas, highlighting the potential of this GI type to improve air quality [25,37–40].

The mechanisms by which GI can reduce human exposure to air pollution or how they can ameliorate air pollution are complex [19,41–43]. Multiple factors are responsible for the favourable action of GI on air quality, including street layouts, meteorological conditions, and vegetation characteristics [43]. Large and coarse particulate matter are deposited on leaves, and fine, ultra-fine particles, and gases are absorbed through the stomata of the leaves, resulting in removing air pollutants from the atmosphere [44–47]. Local weather parameters such as wind and rain can remobilise the PM deposited on leaf surfaces [48–50]. Studies have examined the effect of rainfall in washing off PM from leaves (Table 1). These studies revealed species differences in the rate at which PM is washed off, primarily due to the variations in leaf surface [51–53]. The PM retention capacity on leaf surfaces, however, does not always correlate with the stability of leaf surface PM during rainfall [51,53].

Table 1. Summary of the relevant review papers from 2017 onwards discussing the effect of rainfall to remove pollutants deposited on leaves.

Key Findings	Method Used	Pollutant Studied	Reference
Precipitation effectively removes PM from plant leaves. Changes in rainfall characteristics, such as intensity and duration, influence the efficiency of PM scavenging, especially for water-soluble ions.	Artificial rainfall system	water-soluble ions (SO ₄ ⁻² , Ca ⁺² , Na ⁺ , NO ⁻³ , Cl ⁻ , Mg ⁺² , K ⁺ , F ⁻ , and NH ₄)	Cong et al., 2022 [54]
Rainfall intensity and duration affect the particulate removal process. The maximum particle removal amount occurs at 30 mm/h precipitation. The most easily removal particle size ranges from 10 to 100 μm .	Artificial rain simulator	Particle size range of 10–100 μm	Zhou et al., 2020 [55]

Table 1. Cont.

Key Findings	Method Used	Pollutant Studied	Reference
Wash-off masses of large particles increased by rainfall intensity. Large fractions were washed off preferentially in early rainfall.	Artificial rainfall experiment	Three fractions of PM large particles (10–100 μm), coarse particles (3–10 μm), and fine particles (0.2–3 μm)	Xu et al., 2019 [56]
There was a significant impact of rainfall in washing the PM (PM ₁ , PM _{2.5} , and PM ₁₀) off the leaves. There was a differential impact of rainfall in remobilising PM on different species of plants. A high rainfall intensity had a significant positive impact on PM wash-off from the leaves.	Artificial rain simulator	Three fractions of PM: PM ₁₀ , PM _{2.5} and PM ₁	Weerakkody et al., 2018 [57]
Plants with high PM accumulation show greater PM wash-off and retention. Cumulative PM wash-off increases exponentially with rainfall amount, while wash-off duration shortens with increased rainfall intensity. PM retention varies based on rainfall intensity and plant species.	Artificial rainfall experiment	Three fractions of PM	Xu et al., 2017 [58]
Accumulation of PM on foliage was analysed over days and over a week. Changes in PM deposition on the leaves were mostly affected by rain and to a lesser extent by wind, but the extent of the effect was species-specific. Precipitation affects also PM retained in waxes.	Natural rainfall experiment	PM ₁₀ and PM _{2.5}	Popek et al., 2019 [59]
Rain and wind can remove PM of all sizes, PM > 100 μm is most easily removed. Leaf size had a greater effect on PM retention than the presence of rough microstructures such as hair and grooves. Small leaves are more conducive to PM retention than large leaves.	Natural rainfall experiment	Four sizes (PM > 100, PM _{10–100} , PM _{2.5–10} , and PM _{2.5})	Xu L. et al., 2021 [51]
Only 6 out of the 65 papers considered studied PM wash-off from leaves; as shown in these studies, smooth leaves may have a higher PM wash-off level than rough leaves.	Review	PM	Xu X. et al., 2020 [60]

This fieldwork study aims to provide new insights into the effect of rainfall on the removal of total particulate matter (TPM) from leaves by quantifying the amount and elemental composition of TPM removed during the austral winter. The research focuses on two evergreen native tree species (*Quillaja saponaria* and *Schinus molle*) across five independent rainfall episodes that followed a prolonged long drought of 14 consecutive years in Santiago [61].

2. Materials and Methods

2.1. Site Description

Santiago has a cool, semi-arid climate (BSk) with Mediterranean (Csb) patterns, according to the Köppen climate classification [62]. The sampling site was located in an urban area corresponding to Eloísa Díaz Campus, Universidad de Chile in Santiago (33°25′15.4″ S; 70°39′10.5″ W). The Campus is home to various exotic, native, and evergreen species. The selected species grow under similar environmental conditions on the campus, including exposure to pollutant sources, climatic conditions, solar radiation, and irrigation. The campus is situated north of Santiago, and three major roads pass through the area, which serve as key connections between the city centre and different public services [63,64]. Inside the campus, no firewood is used, and there are no other identifiable sources of air pollution nearby [64].

The elemental composition of PM in Santiago, Chile, from 2011 to 2018, indicates that, for the fine fraction, the mass contributions of crustal elements (Si, Al, Ca, and Fe) have remained stable over the last eight years. Similarly, the mass contributions of elements associated with anthropogenic sources, such as Pb, Br, and Cl (primarily from traffic and wood burning), have also shown stability during this period. For the coarse fraction, three distinct groups of elements were identified: Zn and Cu, and these elements are predominantly associated with brake wear, and K, Si, Mn, and S and Fe, V, Ca, Sr, and Ti, and these groups likely have a mixed origin, potentially influenced by soil resuspension, industrial emissions, and vehicular activity [65].

Geological Characterisation of the Study Area

Santiago is located in a basin filled by quaternary alluvial sediments mostly derived from the chemical and mechanical erosion of the Cenozoic intermediate and basic volcano-sedimentary formations of the Andes. The classification of the Santiago Basin's soils can be grouped as follows: (i) Rock: predominantly found in the surrounding mountain ranges and isolated hills; (ii) Fluvial deposits: soils formed by river activity, ranging from coarse materials like sandy gravels to silts; (iii) Alluvial deposits: resulting from the transport of materials by water, such as gravels in a sandy-clay matrix, interspersed with layers of sand, silt, and clay; (iv) Fine-grained soil deposits: composed of silts and clays with interbedded layers of volcanic ash, gravels, and sands; and (v) Volcanic ash deposits: fine fragments of volcanic rock less than 2 mm in diameter. These are part of sandy loam or clay loam soils. Additionally, copper, molybdenum, silver, and zinc deposits are known, grouped into two mining districts located east and northeast of the city. Lime, gypsum, sand, and gravel are the most important non-metallic mineral resources of the area [66,67].

2.2. Tree Species Selection

Two species were selected for study, *Schinus molle* and *Quillaja saponaria*, and both are recognised as native species according to the Chilean legislation [68] (Figure 1). These trees are located less than 100 m from each other inside the campus to minimise microclimate variations that could affect the results.

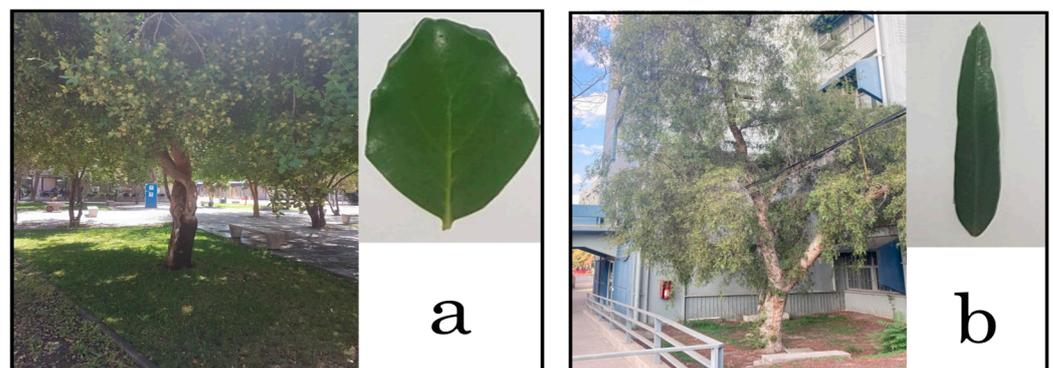


Figure 1. Selected tree species and leaves from *Quillaja saponaria* (a) and *Schinus molle* (b).

Schinus molle (Spanish name Pimiento) is a species introduced to Chile by the Incas many hundred years ago, common name Pepper or Molle (Figure 1b). It is currently from 18.4746° to 33.4569° latitude S. It is an evergreen tree with compound, alternate leaves 15 to 30 cm long, 15 to 41 opposite, lanceolate linear leaflets, 0.85 to 5 cm long. It can grow up to 25 m in height and has rapid growth and longevity [69]. It is widely used as an urban tree because of its resistance to pollution, easy and inexpensive propagation, and minimal need for watering, and it tolerates high temperatures and is highly resistant to drought [70,71].

Quillaja saponaria (Spanish name Quillay) [68,69,71]. It can be found from 29.9533° to 38.5400° latitude S. Is an evergreen tree that can reach up to 15 m in height and has single and alternate leaves. It is a medium-growing species with a longevity of approximately 100 to 150 years (Figure 1a).

2.3. Sample Collection and Rainfall Events

Healthy, adult, well-developed leaves, free from pests and visible damage or drought symptoms, were sampled from each species. Leaves were randomly selected prioritising branches well exposed to the environment at a height of 1.7–2 m above ground level, according to the protocol for the exposure of the population to urban pollution [72]. To obtain a better representation of the tree species, the branches were selected from different parts of the tree in the azimuthal direction, meaning that random branches were selected at the same height from north, south, east, and west of the tree. This ensured representation of weather exposure and tree canopy. The leaves per species were collected one hour before the onset of the first rain in each episode and again one hour after the end of the final rain (Table 2).

Table 2. Duration (days) and mm of rainfall during the austral winter, June–July 2022.

Rainfall Episodes	1st	2nd	3rd	4th	5th
Duration (days)	4	4	2	3	3
Start	2 June	22 June	30 June	8 July	13 July
Final	6 June	27 June	2 July	11 July	16 July
Rainfall level (mm) ¹	26.4	12.4	17.6	37.2	17.0

¹ Data from the National Meteorological Office [61].

The leaf area index (LAI) of the tree species was measured through fieldwork photographs using HEMIV4 (Delta T devices Inc., Cambridge, UK). Photos were taken with a 180° capture angle using a fisheye lens. The images were analysed using the Gap Light Analyzer software v2.0. The LAI for *Q. saponaria* was 0.33 m²m⁻² and *S. molle* was 0.35 m²m⁻². Both species exhibited similar canopy coverage; however, the number of leaves collected for each species depends on the size of the leaf. For *Q. saponaria*, 50 to 70 leaves were selected per rainfall episode, with an average leaf area of 5.47 ± 1.09 cm², as reported in prior studies on this species in Santiago, Chile [73]. In contrast, for *S. molle*, 100 to 120 leaflets were collected per episode, with each leaflet having an average area of 1.96 ± 0.47 cm², consistent with values reported in other investigations [74,75]. This sampling approach ensured a fair and representative comparison between the two species, accounting for differences in individual leaf size while maintaining consistency in the canopy-level analysis.

Information on the five rainfall episodes studied during 2022 was provided by the National Meteorological Office [61], which supplied the millimetres (mm) of rain per minute that fell during each rainfall (Table 2). In this study, rainfall episodes refer to periods of two or more days of intermittent rainfall, where precipitation occurs sporadically rather than continuously. The total rainfall for each episode is measured cumulatively in millimetres, representing the aggregate precipitation over the course of the episode. During the austral winter of 2022 in Santiago, nine rainfall episodes occurred, of which five were analysed in this study, corresponding to 68.7% of the total precipitation of that season. No rainwater was collected during this study, as the primary objective was to investigate TPM removal due to rainfall on tree leaves rather than to analyse the chemical composition of the rainwater itself.

The wind condition in Santiago was obtained from the closest meteorological station to the university campus (Quinta Normal, latitude: −33.44404, longitude: 70.68303), as reported by the National Meteorological Office (Figure 2). This provides a broader context

for wind conditions influencing PM resuspension and deposition, even though precise micro-scale variability near the sampled trees could not be captured.

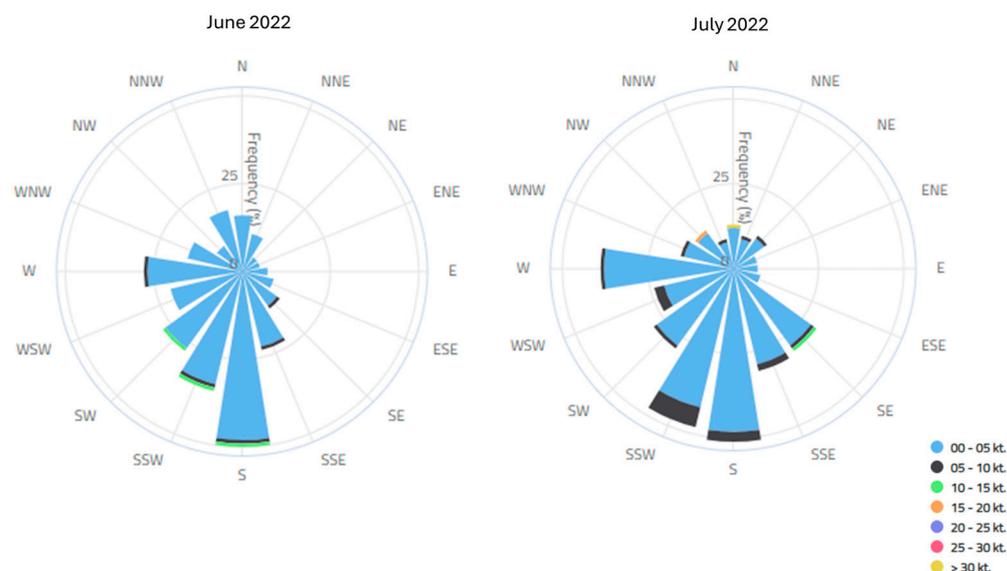


Figure 2. Wind rise for Santiago during the study period (June–July 2022), illustrating the predominant wind directions and intensities (knots). Data sourced from the National Meteorological Office’s Quinta Normal Station (latitude S 33.44404, longitude W70.68303) [76].

2.4. Quantitative Analysis of the Total Particulate Matter on Leaves

The gravimetric method was used to quantify the total particulate matter (TPM) (particles with a diameter < 45–50 μm), deposited on leaves before and after rainfall episodes [77–79]. The selected leaves for each species were carefully cut, isolated, and then transported to the laboratory located on campus.

In the laboratory, each leaf was washed with water and ethanol p.a. (99.9%), while a soft brush was used to gently clean the abaxial and adaxial surfaces, ensuring complete removal of PM from all leaf folds [78]. The solution was filtered through a pre-weighed borosilicate filter (Pallflex Products Corp, Putnam, CT, USA, TX40HI20-WW, 47 mm diameter, Marlborough, MA, USA). The filters were weighted before and after filtration to determine the TPM mass sample using Equation (1). An analytical balance of 0.01 mg precision, installed inside an anaerobic chamber with controlled humidity conditions, was used to weight the filters [80]. Following the filtration process, the washed leaves were dried at 70° for 72 h to measure their dry mass.

$$TPM \text{ deposited on leaves} = \frac{M_1 - M_0}{g_{ldw}} \text{ (mg } g_{ldw}^{-1}) \quad (1)$$

where M_0 is the pre-weighting filter (without deposited TPM), M_1 is the post-weighting filter (with TPM), and g_{ldw} is the gram of dry leaf [81,82].

2.5. Elemental Composition of Total Particulate Matter

Acid digestion of filters with TPM was performed at the Environmental Chemistry Laboratory of the Institute of Public Health (ISP, Santiago, Chile). Each filter was carefully inserted into a 50 mL test tube, and 6 mL of HNO_3 and 18 mL of HCl were added. Then, the tubes were sonicated for 20 min at 25 °C and brought to 115 °C for 1 h in a hot block, Environmental Express. The resulting solution was filtered with folded cellulose paper and placed in a 50 mL volumetric flask with deionised water. The elementary determination of the TPM composition was performed using ICP-MS (Agilent Technologies, 7700 series, Kita

Hachioji, Tokyo, Japan). Two certified standards were used: metals in soil (SQC001-30G) and CRM207-225G-Loamy Sand 3 (Sigma-Aldrich, Laramie, WY, USA). The analysis was done separately before and after the rainfall.

2.6. Quantitative Analysis of Total Particulate Matter at the Species Level (Individual)

The models proposed by Dobbs [83] were used to determine leaf biomass; this means transforming the experimental results at the leaf level to the species (individual) level (Table 3). This model was developed to estimate tree leaf biomass (B_{lf}) for ten Chilean species in Santiago [83]. The diameter at breast height values (BHD) and tree height (H_t) were experimentally measured [84]. In this way, the TPM retained or removed by individual trees was extrapolated using the following Equation (2):

$$TPM_{(\text{deposited on tree})} = B_{lf} \times TPM_{(\text{deposited on leaves})} \times 10^{-6} \quad (2)$$

where $TPM_{(\text{deposited on tree})}$ is the total particulate matter retained or removed by the tree (kg), B_{lf} is total estimated dry leaf biomass of the tree (g), calculated using the Dobbs model, and $TPM_{(\text{deposited on leaves})}$ is particulate matter retained per gram of dry leaf biomass (mg g_{ldw}^{-1}), and 10^{-6} the conversion factor from milligrams to kilograms.

Table 3. The model for calculating biomass (B_{lf}) uses the diameter at breast height (BHD) in cm and the height (H_t) of the individual in metres.

Species	Parameters	Biomass (B_{lf}) ¹
<i>Q. saponaria</i>	BHD = 43 cm	$0.0028BHD^{26821}$
<i>S. molle</i>	BHD = 60 cm; $H_t = 16$ m	$0.1577 + [0.004BHD^2(H_t)]$

¹ Equation from Dobbs (2004) [83].

2.7. Statistical Analysis

For descriptive analysis, boxplots were employed using R software (version 4.3.2) and two statistical tests were carried out. The Shapiro-Wilk test was used to verify the normality of the samples, and the Dixon test was used to identify possible outliers.

As for the analysis of the particles deposited on each tree leaf, a paired-samples *t*-test was performed to detect significant differences between the analysed species (*S. molle* and *Q. saponaria*).

During the analysis of the effect of rainfall on elemental concentrations in TPM, a non-normal distribution was observed using the Shapiro–Wilk test, which was further confirmed by the detection of outliers using the Dixon test. Given the limited size of the data set, a non-parametric analysis was chosen. The Wilcoxon test was applied to assess possible differences between tree species. Finally, a cluster analysis was carried out to identify possible groups of elements that could indicate emission sources.

3. Results

3.1. Total Particulate Matter Retention Capacity and Washed off Effect on Leaves

Considering the five rainfall episodes, *Q. saponaria* showed a difference in TPM concentration deposited on leaves before and after rainfall, which was not observed in the case of *S. molle* (Figure 3). Before the rain, *Q. saponaria* retained an average of 0.49 mg/g_{ldw} of TPM on their leaves, lower than the retention observed in *S. molle*, which retained 3.05 mg/g_{ldw} , an approximately six times higher value. After the rainfall events, it was found that 12% of TPM was washed off (reduced) from *Q. saponaria* leaves compared to pre-rain conditions. In contrast, *S. molle* slightly increased the TPM post-rainfall, rising to 3.2 mg/g_{ldw} (Figure S1 in the Supplementary Materials).

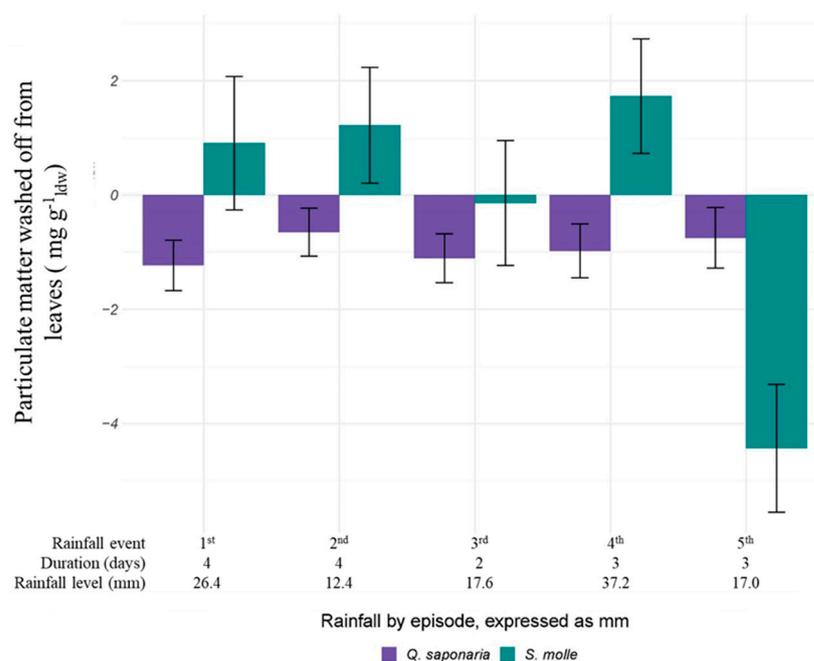


Figure 3. Total particulate matter retained (positive values) or removed (negative values) from the leaves by each rainfall episode, expressed as $\text{mg}/\text{g}_{\text{ldw}}$ by *Quillaja saponaria* (purple bars) and *Schinus molle* (green bars).

After each rainfall event, rain washed off TPM from the *Q. saponaria* leaves, with values ranging between 0.65 ± 0.42 and $1.23 \pm 0.44 \text{ mg}/\text{g}_{\text{ldw}}$ and totalled $4.72 \pm 0.46 \text{ mg}/\text{g}_{\text{ldw}}$ across the five rainfall episodes (Table 4). The quantity of TPM in *Q. saponaria* leaves remained relatively constant over time after precipitation, showing consistent behaviour compared to *S. molle*. The results for *S. molle* differed showing different concentrations of TPM in different rainfall events. Specifically, during the first (26.4 mm), second (12.4 mm), and fourth (37.2 mm) rain events, the TPM increased between 0.91 ± 1.17 and $1.73 \pm 1.00 \text{ mg}/\text{g}_{\text{ldw}}$. In contrast, during the third (17.6 mm) and fifth (17 mm) rain events, the removed TPM was $4.57 \pm 1.11 \text{ mg}/\text{g}_{\text{ldw}}$ (Table 4, Figure 3).

Table 4. Total particulate matter (in mg) deposited (by g_{ldw}) before and after each rainfall for both studied tree leaf species.

Rainfall Episodes	<i>Q. saponaria</i> Before ($\text{mg g}_{\text{ldw}}^{-1}$)	<i>Q. saponaria</i> After ($\text{mg g}_{\text{ldw}}^{-1}$)	<i>S. molle</i> Before ($\text{mg g}_{\text{ldw}}^{-1}$)	<i>S. molle</i> After ($\text{mg g}_{\text{ldw}}^{-1}$)
1st	6.79 ± 0.32	5.56 ± 0.30	13.18 ± 0.91	14.09 ± 0.73
2nd	6.93 ± 0.32	7.29 ± 0.27	8.40 ± 0.62	9.62 ± 0.80
3rd	7.11 ± 0.32	6.00 ± 0.28	13.96 ± 0.66	13.82 ± 0.87
4th	7.64 ± 0.30	6.66 ± 0.36	12.18 ± 0.65	13.91 ± 0.76
5th	7.13 ± 0.39	6.38 ± 0.35	16.51 ± 0.84	12.08 ± 0.74

3.2. Total Particulate Matter Retention Capacity and Washed off Effect at the Species Level (Individual)

Applying Dobbs models [83] (Table 3) and experimental values of breast height diameter (BHD) and the height (Ht) of the tree species determined by Tagle [84], the values obtained at the leaf levels were transformed to tree species levels (the whole tree). In this way, rainfall washed off $0.9 \pm 0.03 \text{ kg}$ of TPM from *Q. saponaria* tree species in total after the five rainfall episodes. However, for *S. molle*, rainfall washed off $1.05 \pm 0.25 \text{ kg}$ of TPM during the third (17.6 mm) and fifth (17.0 mm) rain episodes and accumulated on leaves $0.9 \pm 0.24 \text{ kg}$ of TPM in the first (26.4 mm), second (12.4 mm), and fourth (37.2 mm) rain episodes (Figure S2 in the Supplementary Materials). In total, $1.94 \pm 0.25 \text{ kg}$ was removed

per tree species (Figure 4). This approach provides an estimation of the total contribution of *S. molle* tree species to TPM removal during rainfall episodes.

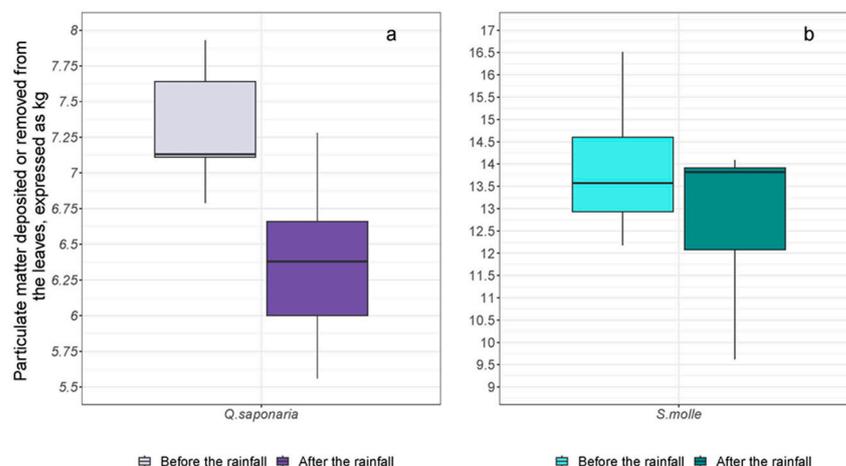


Figure 4. Amount of particulate matter deposited/removed, expressed as kg/individual, from the species *Quillaja saponaria* (a) and *Schinus molle* (b) before and after the rainfall episodes.

The statistical analysis showed that *Q. saponaria* had significant differences in TPM on its leaves (and species) before and after the five rainfall episodes, while *S. molle* did not (Table 5). After the rainfall episodes, there was a continuous decrease in TPM concentration for *Q. Saponaria* leaves, as indicated by the negative values of the *t*-test analysis (−8.714). In contrast, the *t*-test analysis for *S. molle* was close to zero (−0.127), indicating minimal variation between the leaf level and tree species level.

Table 5. *t*-test results comparing particles retained or removed in each tree species at the leaf level (mg/g_{ldw}) and individual level (mg/kg). Statistical parameters showing the central tendency, *t*-test value (statistic) dispersion (Df), and possible normality/atypicality of the distributions of the variables (*p*-value).

Variable	Unit	Group 1	Group 2	N1	N2	Statistic	Df	<i>p</i>
<i>Q. saponaria</i>	mg/kg	Before the rainfall	After the rainfall	5	5	−8.714	4	1
<i>Q. saponaria</i>	mg/g _{ldw}	Before the rainfall	After the rainfall	5	5	−8.714	4	1
<i>S. molle</i>	mg/kg	Before the rainfall	After the rainfall	5	5	−0.127	4	0.905
<i>S. molle</i>	mg/g _{ldw}	Before the rainfall	After the rainfall	5	5	−0.127	4	0.905

¹ Significance level = *p*-value < 0.05.

After five rainfall episodes, it was observed that *S. molle* tended to retain TPM on its leaves, while *Q. saponaria* showed a tendency to always wash off the TPM on its leaves. The amount of TPM on *Q. saponaria* leaves remained relatively stable over time after precipitation, showing stability in its behaviour compared to *S. molle* (Figure S2 in the Supplementary Materials).

3.3. Elemental Concentrations in Total Particulate Matter Deposited on Leaves

Sixteen elements were found in the TPM deposited on the leaves of *Q. saponaria* and *S. molle*: Ag, As, Be, Cr, Cu, Fe, Li, Mn, Mo, Ni, Pb, Sb, Se, Sn, V, and Zn. It is worth noting that, while Be and Ag were detected, their concentrations were not consistently above the quantification limits and, thus, were not always included in the analysis. Both tree species differed in terms of concentrations of elements before and after rainfall (Table S1 in the Supplementary Materials contains the elements concentrations before and after rainfall episodes). The highest concentrations were recorded in *S. molle* in the range from 1 to 10⁷ mg/kg compared to the range from 1 to 3 × 10⁴ in *Q. saponaria* (Figures 5 and 6).

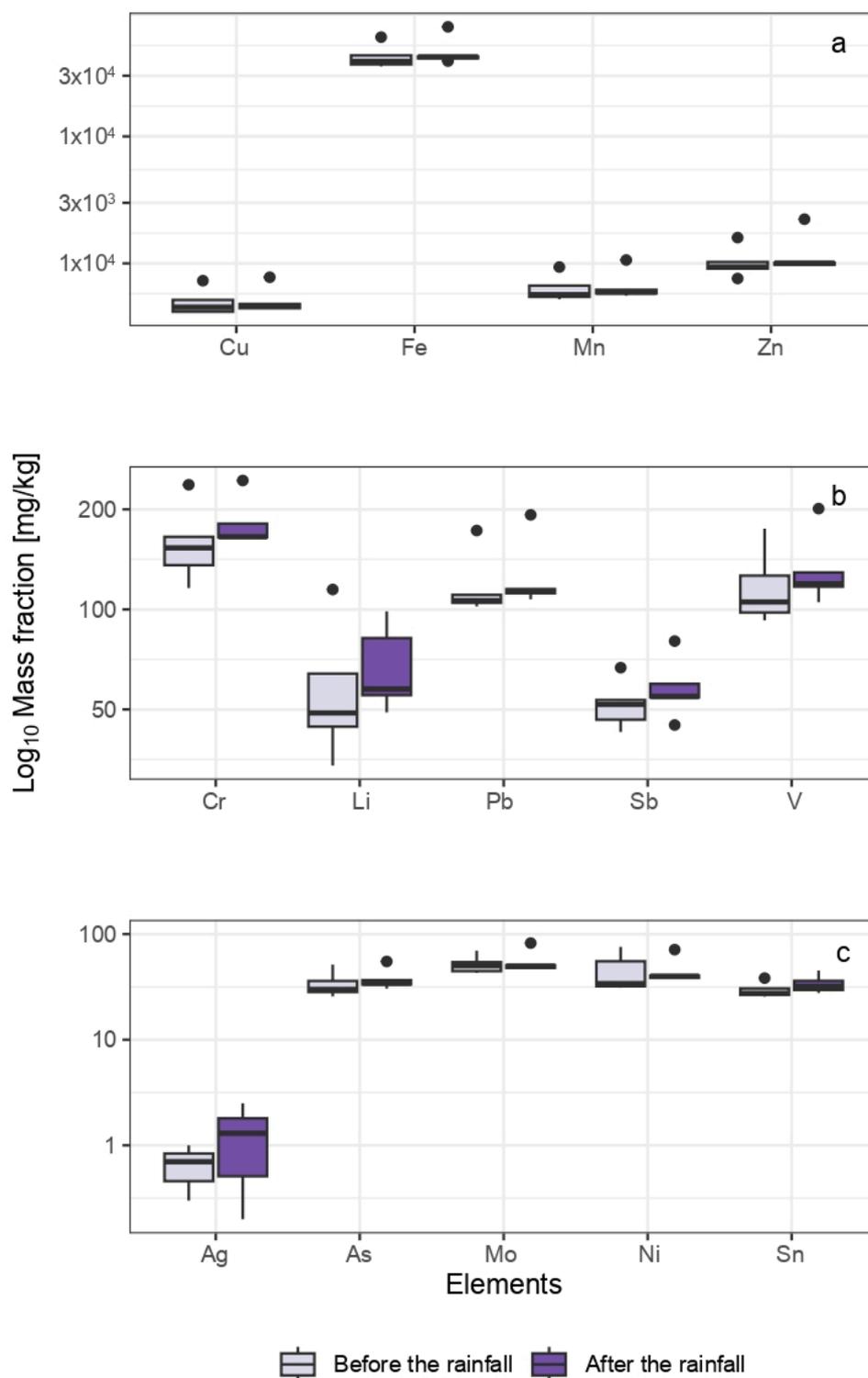


Figure 5. Metal concentrations on *Quillaja saponaria* (mg/kg) before and after the five rainfall episodes. (a) Elements with the highest concentrations; (b) elements with moderate concentrations; (c) elements with the lowest concentrations.

After the five rainfall events, the deposition of the elements onto the leaves of *Q. saponaria* was likely facilitated by rainfall, leading to increased concentrations of most elements, such as Cr, Cu, Fe, Mn, Mo, Pb, Sb, Se, Sn, V, and Zn on the leaves of this species. The opposite trend was observed in *S. molle*, as its elemental composition decreased after rainfall. In addition, for a few elements, such as As, Mo, Ni, and Sn, the concentrations remained almost the same before and after the rain for both tree species (Figures 5 and 6).

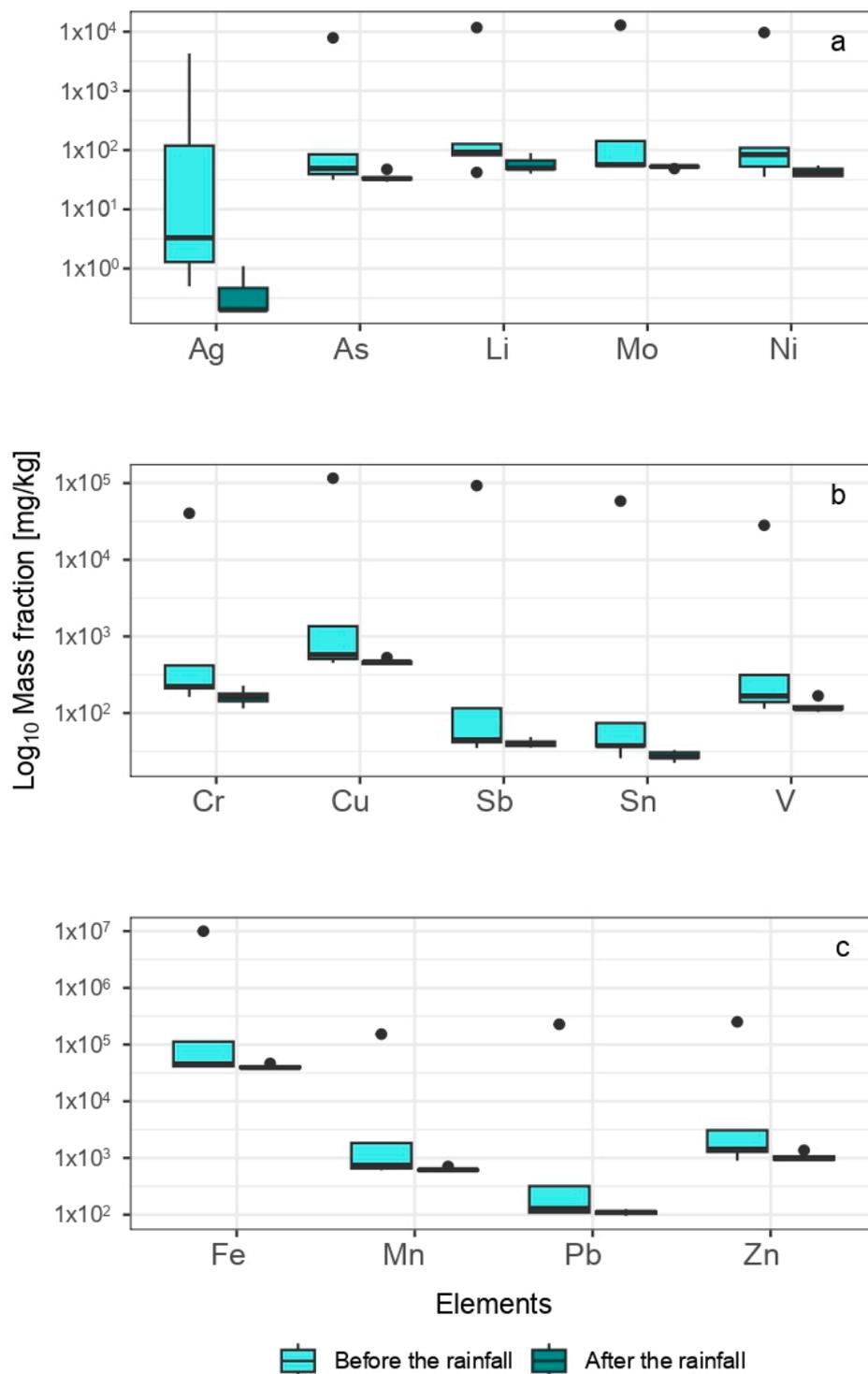


Figure 6. Metal concentrations on *Schinus molle* (mg/kg) before and after the five rainfall episodes. (a) Elements with the highest concentrations; (b) elements with moderate concentrations; (c) elements with the lowest concentrations.

Each rainfall episode affects TPM accumulation in different ways. In each rainfall episode, most of the elements were washed off (negative values) from *Q. saponaria* leaves. Moreover, in the first episode (26.4 mm), the elemental concentrations of Fe, Mn, Mo, Ni, Pb, Se, and Zn were the lowest compared to the other rain episodes. In some other rainfall episodes, however, elements were retained on the leaves (positive values), such as As, Cr, Sb, Sn, and V in the first event (26.4 mm); Li and Ni in the second episode (12.4 mm); and Cu and Mo in the fourth episode (37.2 mm) (Figure S3 in the Supplementary Materials).

Different elemental concentrations were observed in the rainfall episodes for *S. molle*. Elements such as Cu and Mo consistently rose in concentration during the first three rainfall episodes, with an average rainfall of 18.9 mm. However, during the fourth rainfall episode, which had a rainfall of 37.2 mm, the concentrations of these elements decreased by more than half. The elements quantified in each rainfall episode were As, Ag, Cr, Cu, Fe, Li, Mn, Mo, Ni, Pb, Se, Sb, Sn, V, and Zn; all showed increased concentrations on the leaves in the fifth rainfall episode (17.0 mm). Elements that decreased in concentrations were Cr, Ni, and Se in the second episode (12.4 mm) and As, Mn, Ni, Pb, Sn, and Zn in the fourth rainfall episode (37.2 mm) (Figure S4 in the Supplementary Materials).

Based on the Shapiro–Wilk test, it was observed that the elemental concentrations and TPM obtained before and after the rainfall on each tree species did not behave normally. Additionally, some data fell outside of this behaviour, which should be considered due to the scarcity of data. The Dixon test showed that eliminating anomalous data must not be ruled out. The collected data did not follow a normal distribution. However, some peculiarities suggested differences between groups observed before and after rain events in certain variables, although this was inconclusive. To explore this further, the Wilcoxon test was used to combine the observations made before and after the rain (Table 6). Subsequently, a cluster analysis was performed to find a group of elements pointing to some sources of emission (Figures 7 and 8).

Before rainfall, two possible sources of association were identified in the TPM accumulated on the leaves of *Q. saponaria*: one formed by Li and Ni and the other by Sb and Sn. For the other associations, the specific source of As, Cr, V, and Zn and Cu, Fe, Mn, Mo, and Pb was less evident. A group that brings together the elements As, Cr, V, and Zn was also observed before the rainfall for both trees *Q. saponaria* and *S. molle* (Figures 7 and 8).

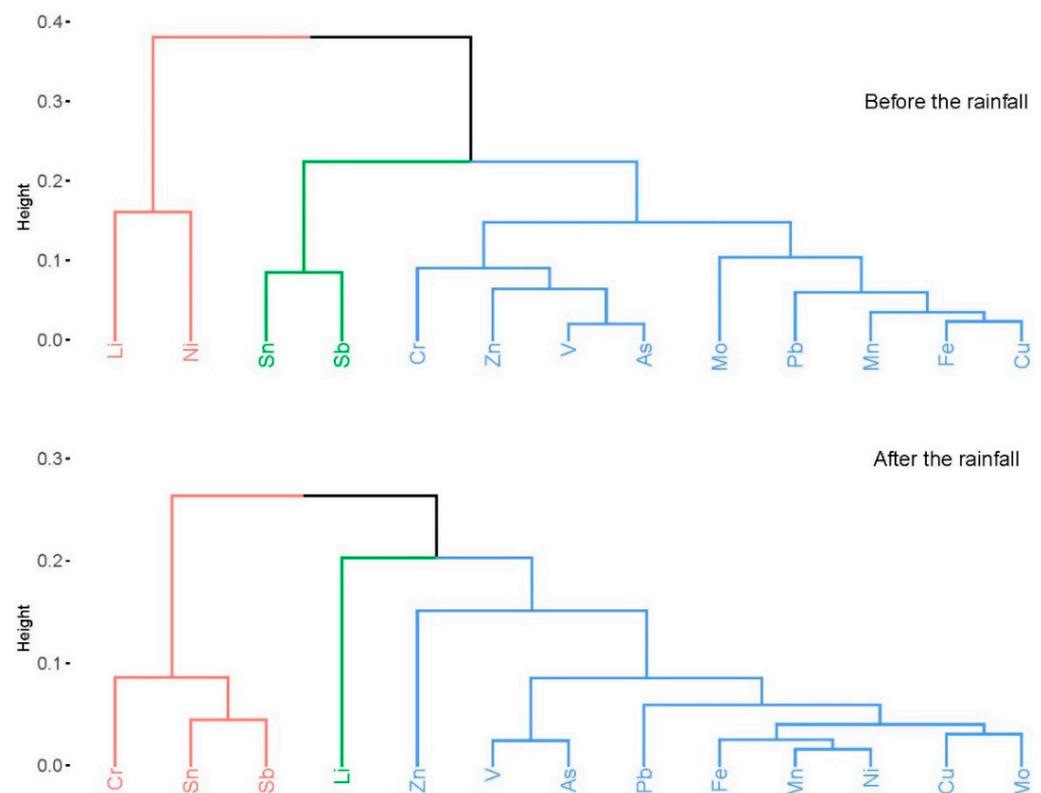


Figure 7. Dendrogram corresponding to the elemental concentration of the total particulate matter deposited/removed from the leaves of *Quillaja saponaria* before and after the total period of rainfall.

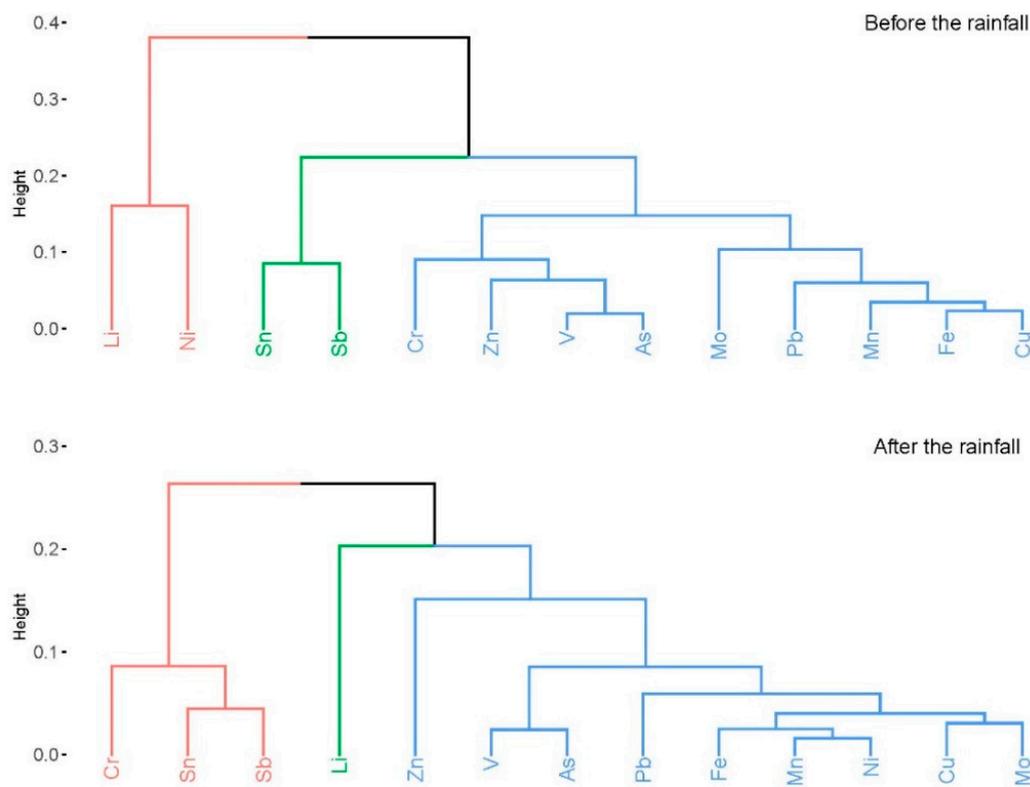


Figure 8. Dendrogram corresponding to the elemental concentration of the total particulate matter deposited/removed from the leaves of *Schinus molle* before and after the total period of rainfall.

Table 6. Statistical parameters of the Wilcoxon test and Dixon test for the relationship between TPM on tree species (*Quillaja saponaria* and *Schinus molle*) and rainfall episodes at the level of the leaves (mg/g_{ldw}) and the level of the individual (kg/individual).

Variable	Media	Median	SD ¹	Wilcoxon’s Q Test		Dixon’s Q Test	
				W.obs	p-Value	Q.obs	p-Value
mm_rainfall	22.12	17.6	9.83	0.91	0.46	0.44	0.44
<i>Q. saponaria</i> (mg/g _{ldw})	0.46	0.47	0.05	0.98	0.97	0.21	0.82
<i>S. molle</i> (mg/g _{ldw})	2.94	3.11	0.54	0.92	0.39	0.21	0.81
<i>Q. saponaria</i> (kg/individual)	6.85	6.95	0.73	0.98	0.97	0.21	0.82
<i>S. molle</i> (kg/individual)	12.78	13.5	2.35	0.92	0.39	0.21	0.81

¹ Standard deviation; Significance level = p-value < 0.05. High W values close to 1 and high p-values; the variables could follow a normal distribution. For the Dixon test, high values in the Q statistic and high p-values indicate an absence of outliers in the data.

After rainfall, only the association of Cr, Sn, and Sb remained distinguishable. In the case of *S. molle* before the rainfall, the first component corresponds to the association of Pb, Sb, and Sn elements and the second to the association of Li and Ni elements. After rainfall, when some elements are removed and others retained, no clear distinction of source was possible (Figures 7 and 8).

4. Discussion

The finding of this study aligns with Xu et al.’s [58] investigation, which highlighted the impact of rainfall on the removal of PM from leaf surfaces. In line with the findings of Xu et al. [58], this study also highlights that the number of days of TPM accumulation and the maximum capacity of the plants should be considered. TPM wash-off is strongly influenced by both the environmental growth conditions and the specific characteristics of the plant. Regarding the environmental growth conditions, the two tree species studied in

this research were located less than 100 m apart within the university campus, minimising microclimate variations. Both species were exposed to similar environmental conditions, including pollutant levels and water availability, which helped control external factors. However, one species, *S. molle*, was located near a wall, which may have influenced the amount of TPM deposited on its leaves, the amount of rainfall reaching them, and wind patterns. This, nonetheless, accurately reflected the real-world conditions that different tree species encounter in urban environments. Popek et al. [59] found that changes in PM deposition on the leaves are mostly affected by rain and, to a lesser extent, by wind, but the extent of the effect was species-specific. Collecting leaves from various parts of the tree canopy in an azimuthal pattern ensured a comprehensive representation of weather exposure and canopy structure. This approach facilitated more reliable comparisons while accounting for the variables that affect TPM removal.

Zhou et al. [85] found that coarse particles removal by rainfall was highest across different rainfall intensities. Interestingly, at a higher rainfall intensity (45 mm/h), the mass of PM removed from leaf surfaces was lower than at a lower intensity (30 mm/h). Their study also revealed that the response to rainfall events varies significantly among species, which aligns with the findings of this study. In this study, two rainfall episodes of similar magnitude, the third (17.6 mm) and fifth (17 mm), removed 4.57 mg/g_{ldw} of TPM from *S. molle* leaves, while the other episodes, averaging 25.3 mm, removed 3.86 mg/g_{ldw} of TPM. It is important, however, to approach these comparisons with caution. Zhou et al. [70] conducted their experiments under artificial rainfall conditions, whereas this study was conducted under natural conditions with fewer rainfall episodes and varying intensities. This difference in study conditions may impact how PM wash-off behaves. Wang et al. [86] conducted a field study on an evergreen tree species (*L. lucidum*) in China between spring and autumn, observing a relationship between the amount of PM washed off and the amount of water fallen. Their study demonstrated that higher precipitation levels were associated with increased removal of PM from leaf surface, differing from our results. This variation could be attributed to differences in rainfall patterns, specifically, high-intensity, short-duration rainfalls compared to multiple low-intensity rainfalls spread over days. While Wang et al. [86] and this study highlight the cleaning effect of rainfall, the quantity of PM removed varies depending on specific rainfall patterns, leaf traits, and environmental conditions. Popek et al. [59] found that the accumulation of PM changes with short-term (daily changes) and longer time periods (weekly changes) of rainfall. Further field research is needed to explore how different rainfall intensities and frequencies impact PM wash-off and foliar uptake, improving our understanding of these dynamics across various environmental contexts.

How PM is accumulated or washed off by leaf is still ongoing discussion [44,60]. The ability of leaves to accumulated PM varies among tree species and leaf traits, such as leaf morphologies [44–47]. A combination of different leaf traits is a key factor for enhancing PM retention. For example, it is known that rough leaves, with complex shapes, high stomata density, and longer persistence with leaf blade total developed, have been associated with the highest PM deposition values [43,44]. Since the deposition PM occurs mainly through inertial deposition [85], it is expected that leaves with more complex traits will have higher PM accumulation [42]. The leaves of *Q. saponaria* are simple, alternate, smooth, and ovate, traits that likely facilitate easier TPM removal after rainfall episodes. In contrast, *S. molle* leaves are opposite, lanceolate, and have characteristic grooves along the central vein. These leaf differences may account for the more complex wash-off effect observed in *S. molle*, where TPM is sometimes retained even after rainfall. The grooves in *S. molle* leaves could trap particles, limiting their removal and resulting in inconsistent TPM reduction across rainfall episodes. This suggests that leaf morphology plays a crucial role in determining how effectively PM is washed off during rainfall [42]. Other factors, such

as rainfall intensity, rain variations during the episode, and the physiology of the trees, could also influence the PM accumulation. Understanding how leaf traits influence and interact with rainfall episodes in PM accumulation will enhance our comprehension of the PM removal process by rain.

Another finding reveals distinct species responses to rainfall. The total amount of TPM finally removed at the leaf-level in the five rainfall episodes studied was 4.72 and 8.43 mg/g_{ldw} for *Q. saponaria* and *S. molle*, respectively. In the case of *Q. saponaria*, the leaves were washed consistently during all rainfall events, regardless of the amount of water fallen. The five rainfall episodes removed 0.9 ± 0.03 kg/tree species and eventually left the possibility of continuing to accumulate TPM without the danger of clogging the stomata of the leaves [59]. *S. molle* had a different behaviour, and in some episodes, TPM increased, and they retained more TPM on leaves contrary to the expected outcome that TPM is washed off by rain. To the authors' knowledge, this phenomenon has not been documented previously. A possible explanation may be the species' location in the urban environment and its specific characteristics, as previously discussed. *S. molle* is situated near a wall, which could influence TPM resuspension and wind turbulence, thereby affecting the TPM content on leaves. This observation highlights the need for further studies in natural environments to better capture real-world conditions and the daily variability of rainfall events. As Xu et al. [60] recommended, future research should focus on studying the wash-off of leaf-retained PM in greater detail. Moreover, providing more insights into TPM reduction based on leaf-retained TPM wash-off mass would be invaluable for future landscape planning and design.

Before the rainfall events, an association between Li and Ni elements was observed in the elemental concentrations of TTPM washed by the rainfall on the leaves of *Q. saponaria* and *S. molle*. This finding contributes to new information about the composition of the atmospheric TPM in Santiago, Chile, as Li was never reported before in the air of Santiago, not in PM₁₀, or PM_{2.5} [87,88] not even in TPM in the first study of Air Quality in 1976 using Instrumental Neutron Activation Analysis (INAA) [89]. No information about recycling system of Li-ion batteries was found in Santiago. However, Li and Ni are critical metals present in e-waste, typically in lithium-ion batteries and electric vehicles [90]. Lithium-ion batteries are currently used as energy-storage technology and contain Li_x-Ni_y-Oz in different proportions in the city. They are widely used in transportation, including hybrid and electric vehicles (EVs) [91]. It has been observed that EVs influence the content of Ni in PM [92]. While electric vehicles powered by lithium-ion batteries do not produce direct emissions during operation, their production, damage during use, and recycling at the end of their life cycle can contribute to environmental pollution. Studies have shown that thermal runaway events or improper recycling processes can release PM containing potentially toxic elements like Li and Ni, which may impact water and soil quality [93,94]. The detection of Li in this study highlights the need to further investigate the connection between urban PM and the growing adoption of lithium-ion battery technology. Additionally, public transportation in Santiago has incorporated nearly 2000 electric buses [95] since 2017, underscoring the importance of understanding the environmental implications of such technologies [93,94] use, e-waste, and disposal. This finding could prompt further investigations into the study of the elemental composition of PM to track changes in the vehicle market [91]. For example, the presence of Ba, Pb, Cr, and Fe-rich particles [96] on the leaf surface could indicate the popularity of the use of cars with combustion engines, while the presence of Ni-rich and rare earth elements (REEs)-rich particles could be linked to the use of EVs. It is important to keep monitoring the elemental composition of PM to address these emerging trends. It could be a good alternative enforcing policies that encourage the planting of particular species capable of capturing these elements.

The association of Sb and Sn elements in *Q. saponaria* and Pb in *S. molle* was also observed. These three elements are usually emitted from the recovery process of lead-containing products and polymetallic alloys [97]. In addition, a subgroup consisting of Cu, Fe, Mn, and Mo was also identified in both species before rainfall. These elements have been previously reported in the atmospheric PM of Santiago, Chile, coming from crustal matter, motor vehicles, combustion and copper smelting [87,88]. Some heavy metals, such as Cu, Cr, Fe, Mn, Ni, and Zn found in this study could be found in micronutrients necessary for living organisms, including plants, but they may induce noxious effects at higher levels [98]. Barium (not found in this work), Cu, Fe, Mn, Mo, Sb, and Zn have been interpreted in specific settings to be indicative of brake wear and can serve as indicators of traffic-related re-suspension [99,100], Zn and Cu, particularly in the coarse fraction, have been associated with brake wear since 2011 in Santiago [65]. Lead could also be present in the atmosphere coming from burning unleaded gasoline in vehicles and industries and leaded gasoline in piston-engine aircrafts [101], as well as legacy leaded gasoline through particle resuspension [102]. In Chile, vehicles without leaded gasoline were only introduced in 1992, and formal sales of leaded gasoline were phased out by mid-2003 [95].

This research not only provides new insights into the effect of rainfall on TPM wash-off but also updates and expands the existing knowledge on the elemental composition of airborne particles in Santiago. However, it is crucial to consider the local climatic context, particularly the prolonged 14-year drought in Santiago. These conditions likely enhance the resuspension of soil and dust, which can significantly influence the observed elemental composition of PM. Further targeted studies are needed to comprehensively understand the dynamics of airborne elemental compositions in cities, particularly under unique climatic conditions.

Rainfall could have a role in improving air quality by facilitating the removal of heavy elements from both the atmosphere and plant leaves. According to Popek et al. [59], rainfall is necessary to allow plants to capture PM again, as rain has the “wash-off” effect. Furthermore, studies have indicated that atmospheric metals, such as lead, can be retained by plant leaf structures like trichomes and cuticular waxes [103], which contributes to air quality improvement. However, the process of foliar uptake of heavy metals remains insufficiently understood and required further investigation [98].

This research provides more information about the effect of rainfall on TPM removal on leaves, particularly under conditions of low-intensity, intermittent rain that persists over two or more days. The results of this study can have important implications for urban planning and policy, especially when choosing plant species for urban green spaces. Understanding the impact of rainfall on PM removal and washing off would be the next level for computational models. This tool could help researchers, urban planners, and stakeholders understand the complex interactions between deposition, resuspension, and wash-off processes. It would also help in understanding how meteorological conditions and different leaf traits influence these processes and provide more accurate results about the effect of trees on air quality. One example of such a tool is the current Vegetation Impact Dynamic Assessment (VIDA) model, which intends to facilitate the exploration of dynamic interactions among these processes [104]. However, the challenges remain in the lack of availability of measurements of PM on leaves and high-resolution air concentration data to validate this kind of model.

Limitations and Recommendations for Future Studies

The limited number of rainfall episodes studied after a prolonged drought impacts the reliability of this research; a small sample size reduces the study’s statistical power, making it more challenging to detect significant effects or trends. Consequently, these results may not be generalisable to broader contexts, restricting the ability to draw wide-ranging conclusions or apply findings to other regions. However, this fieldwork study under

natural conditions in a Mediterranean city in South America is among the first to examine rainfall's wash-off effect outside a controlled experiments indoor, offering a more realistic view of how rainfall influences TPM removal in situ. Analysing only a few rainfall episodes might not accurately capture seasonal changes or long-term trends, which could impact the conclusions of the study. Therefore, it is important to conduct further fieldwork studies that include long-period rainfall analysis in different geographical zones to comprehend the impact of rainfall throughout different seasons, including changes in leaf seasons.

5. Conclusions

The results of this study demonstrate the wash-off effect on TPM deposited on leaves. The research indicates that there is no connection between the amount of TPM accumulated and washed off from tree leaves and the amount of rainfall. However, the study showed differences in how *Quillaja saponaria* and *Schinus molle* respond to rainfall, particularly in terms of the total PM that is either removed from or remains on their leaves. Based on these findings, the following conclusions can be made:

- Two rainfall episodes, with an average rainfall of 17.3 mm, resulted in significant loss of TPM from *S. molle* leaves, while *Q. saponaria* were minimally affected by the amount of rainfall. Further research is needed to examine how different intensities and durations of rainfall affect the retention and wash-off of PM from the leaves of urban tree species.
- Elemental analysis of TPM washed-off on the leaves reveals associations with certain elements like Li and Ni, potentially linked to lithium-ion batteries from electric vehicles, as well as Pb, Sb, and Sn associated with industrial processes and still leaded gasoline in Santiago, Chile.
- Rainfall might positively affect air quality by removing heavy elements from the atmosphere and leaves, aiding in the recovery of plants' ability to capture more PM and reduce eventual phytotoxic effects due to heavy elements accumulated on their surface. However, there is a lack of data on the effect of metal speciation in TPM on its foliar uptake by plants, highlighting a need for further research.

The results presented in this study mark a pioneering effort in Chilean research that highlighted the complex relationship between trees, particulate matter deposition, and rainfall, a phenomenon that has received limited attention to date. The study also highlights gaps in understanding the role of metal speciation in TPM foliar deposition, emphasising the need for further investigation to comprehensively address the impacts of pollution on vegetation health and ecosystem dynamics.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/atmos16010051/s1>, Table S1: Elements found in the total particulate matter analysed, Figure S1: Amount of total particulate matter deposited and removed on *Quillaja saponaria*'s leaves (a) and *Schinus molle*'s leaves (b) before and after the rainfall episodes (mg/g_{ldw}). Figure S2: Total particulate matter retained (positive values) or removed (negative values) from the species by each rainfall episodes (kg/individual) for *Quillaja saponaria* (purple bars) and *Schinus molle* (green bars). Figure S3: Elemental concentration, expressed as log mg/kg, retained (positive values) or removed (negative values) from the leaves of *Quillaja saponaria* in every rainfall episode. Figure S4: Elemental concentration, expressed as log mg/kg, retained (positive values) or removed (negative values) from the leaves of *Schinus molle* in every rainfall episode.

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