






## Article

# Spatial Distribution and Chemical Composition of Road Dust in Two High-Altitude Latin American Cities <sup>†</sup>

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- <sup>†</sup> This paper is dedicated to the memory of Professor Beatriz H. Aristizábal.



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**Abstract:** Road dust (RD) resuspension is one of the main sources of particulate matter in cities with adverse impacts on air quality, health, and climate. Studies on the variability of the deposited PM<sub>10</sub> fraction of RD (RD<sub>10</sub>) have been limited in Latin America, whereby our understanding of the central factors that control this pollutant remains incomplete. In this study, forty-one RD<sub>10</sub> samples were collected in two Andean cities (Bogotá and Manizales) and analyzed for ions, minerals, and trace elements. RD<sub>10</sub> levels varied between 1.8–45.7 mg/m<sup>2</sup>, with an average of 11.8 mg/m<sup>2</sup>, in Bogotá and between 0.8–26.7 mg/m<sup>2</sup>, with an average of 5.7 mg/m<sup>2</sup>, in Manizales. Minerals were the most abundant species in both cities, with a fraction significantly larger in Manizales (38%) than Bogotá (9%). The difference could be explained mainly by the complex topography and the composition of soil derived from volcanic ash in Manizales. The volcanic activity was also associated with SO<sub>4</sub><sup>2−</sup> and Cl<sup>−</sup>. Enrichment factors and principal component analysis were conducted to explore potential factors associated to sources of RD<sub>10</sub>. Elements such as Cu, Pb, Cr, Ni, V, Sb, and Mo were mainly associated with exhaust and non-exhaust traffic emissions.

**Keywords:** PM<sub>10</sub>; dust resuspension; sediment load; non-exhaust emissions; chemical profile; enrichment factors; Colombia

## 1. Introduction

Particulate matter (PM) emitted by road transport can be released both from the exhaust pipe, as a result of incomplete fuel combustion, and from non-exhaust processes [1]. Exhaust particles have shown a trend of decreasing emissions due to stringent tailpipe standards, better fuel quality, and migration towards zero- or low-emission vehicles in

recent decades [2]. However, detailed information concerning non-exhaust PM emissions is still relatively limited in developing countries, particularly in Latin America, where there is not enough knowledge regarding physicochemical characteristics of PM, its spatial distribution, and associated adverse health effects, among other variables.

Non-exhaust emissions have been identified as one of the most significant sources of air pollution in urban environments due to their contribution of coarse, fine, and ultrafine particles [1,3–6]. These emissions are constituted by direct releases of particles due to abrasion processes, including tire, brake, clutch, and road wear [7–9]. They also include the resuspension of road dust, namely PM emitted from natural and anthropogenic sources, deposited on the road surface, and resuspended as a result of vehicular turbulence and the action of the wind [2].

Road dust is composed mainly of crustal dust and soil material, but also of particles derived from several anthropogenic sources, such as traffic, industrial emissions, quarries, and construction activities, among others [6,10,11]. Therefore, the chemical composition of road dust is variable and is associated with the characteristics of the emission sources. This includes abundant crust elements, such as Al, Fe, Mg, Na, P, and Ti, among others, but also hazardous components such as Pb, V, Cd, Cu, Sb, and polycyclic aromatic hydrocarbons (PAHs). The latter ones generate adverse effects on human health, especially on the cardiorespiratory system [12]. Road dust loadings, as well as their chemical speciation, vary geographically depending on parameters such as meteorology, geology of the region, and even the type of pavement sampled [13,14].

Some studies have investigated the association of road dust particles and mortality from cardiorespiratory diseases and other causes. Researchers have found that an incremental increase of particles related to road dust resuspension can increase mortality risk by 4.0%, higher than the observed for vehicle exhaust [15]. Other studies have reported a significant daily mortality because of an increase of  $10 \mu\text{g}/\text{m}^3$  of particles related to road dust in Stockholm [16]. Lastly, chemical species associated with road dust and mineral resuspension, such as  $\text{SiO}_2$ , Ca, Fe, and Ti, have shown deleterious health effects [17,18]. Consequently, studying the loadings and the chemical composition of road dust, as well as its spatial variability, is crucial to identify the origin of the PM, to formulate mitigation measures and to identify potential adverse effects on the population.

Studies on loadings and chemical characterization of particles below  $10 \mu\text{m}$  from road dust ( $\text{PM}_{10}$  but termed  $\text{RD}_{10}$  in this study) have been carried out in several cities. For example, Alves et al. [7] used a portable resuspension chamber to collect road dust from five main roads in Oporto and an urban tunnel in Braga, Portugal. Researchers reported dust loadings of  $0.48 \pm 0.39 \text{ mg PM}_{10}/\text{m}^2$  for asphalt paved roads. Furthermore, they found that crustal and anthropogenic elements, associated with tire and brake wear, dominated the inorganic fraction. Vlasov et al. [19] analyzed potentially toxic elements (PTEs) in road dust in Moscow, finding that the main pollutants of the  $\text{PM}_{10}$  fraction included Sb, Zn, W, Sn, Bi, Cd, Cu, Pb, and Mo. They concluded that particles were most contaminated in the central part of the city due to the large number of cars and traffic congestions. Zhang et al. [20] analyzed a total of 64 dust samples collected from five urban roads and four parks in Tianjin, China to determine the size distribution and elemental composition of the  $\text{PM}_{10}$  fraction. The researchers found that crustal elements accounted for 30.14% of the  $\text{PM}_{10}$  fractions and the most abundant trace elements were Zn, Mn, and Cu (range, 277 to 874 mg/kg). Pant et al. [21] obtained road dust samples collected at two sites in Birmingham, UK and one site in New Delhi, India. They found that dust loadings were found to be much higher for New Delhi ( $72.9 \pm 24.3 \text{ mg}/\text{m}^2$ ) compared to Birmingham (range,  $9.34 \pm 5.56$  to  $12.1 \pm 9.3 \text{ mg}/\text{m}^2$ ). In addition, Cu was found to be a factor significantly associated with oxidative potential in the  $\text{PM}_{10}$  fraction.

Although some emission inventories have recognized resuspended road dust as one of the most significant sources of PM in urban centers [22], research on  $\text{RD}_{10}$  conducted in Latin American countries has been scarce. Pachón et al. [23] studied influencing factors of road dust in Bogotá, finding that meteorology, land use, traffic characteristics, and road

conditions determine  $RD_{10}$  levels in the city. Ramírez et al. [24] characterized inorganic compounds (water-soluble ions, major and trace elements, organic and elemental carbon) and estimated source contributions to the  $PM_{10}$  fraction of road dust sampled in Bogotá, the capital of Colombia. Crustal elements were the most abundant species, accounting for 49–62% of the  $PM_{10}$  fraction, followed by OC (13–29%), water-soluble ions (1.4–3.8%), EC (0.8–1.9%), and trace elements (0.2–0.5%). On the other hand, Ramírez et al. [25] evaluated road dust in Barranquilla, a major industrial city in the Caribbean region, finding that the major elements, including Al, Ca, Fe, K, Mg, Na, and S, were the most abundant species, accounting for  $23\% \pm 18\%$  of the mass of thoracic particles.

The objective of this paper was to show an analysis of the spatial distribution and chemical composition of road dust in two high-altitude Latin American cities. This study was conducted in two cities in Colombia (Bogotá and Manizales), located in the Andean mountains at 2000 m above sea level (m.a.s.l.). The cities were selected for presenting different demographic conditions, transportation patterns, industrial dynamics, and pollution levels. The aims of the research were (i) to determine the  $RD_{10}$  load in each city according to land use, (ii) to evaluate the spatial distribution and the chemical composition of  $RD_{10}$  in Bogotá and Manizales, and (iii) to explore potential factors associated to sources of  $RD_{10}$  for the two cities mentioned.

## 2. Materials and Methods

### 2.1. Sampling Locations

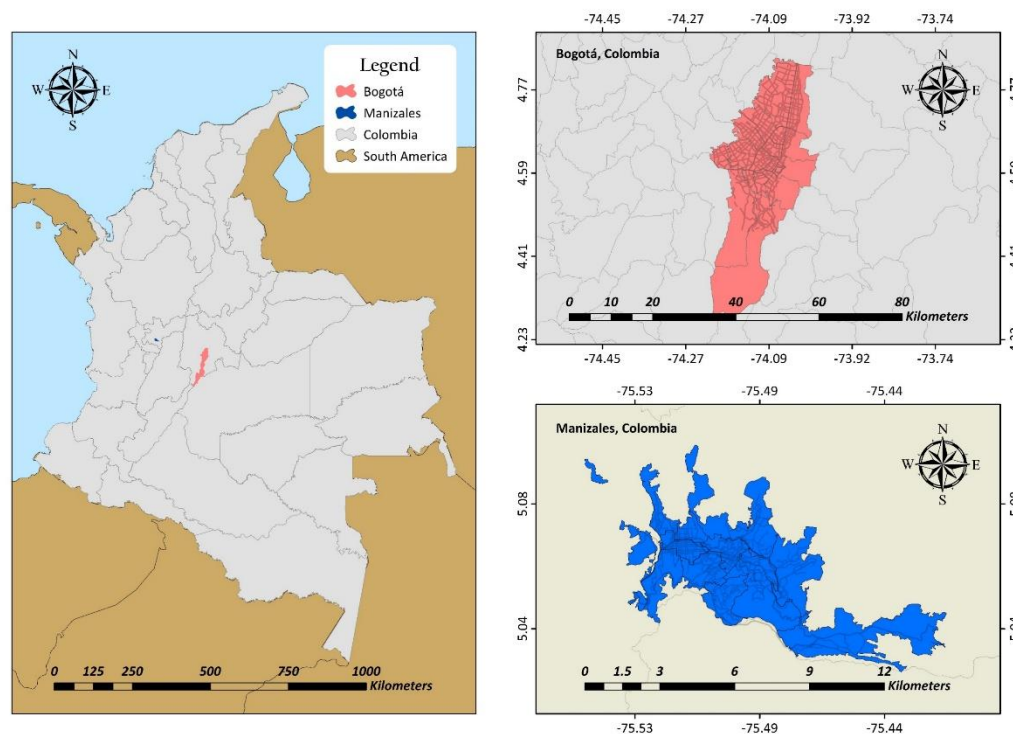
Bogotá is the capital of Colombia and the largest city in the country (population of 8.1 million), located in the South America Andes Cordillera at an altitude of 2640 m.a.s.l. (Figure 1). The urban region has an approximate area of 420 km<sup>2</sup> and a high population density, ~17,700 inhabitants per km<sup>2</sup> [24]. Meteorological conditions in Bogotá are characterized by having an average temperature of 15 °C and average annual precipitation of 844 mm. Winds from NE–E and SE–E predominate, with average speeds of 1.5–3.5 m/s. The trade winds reach the city between June and August, favoring pollutant dispersion. The city has a high motorization rate, 313 vehicles per 1000 inhabitants in the year of 2018 [26]. The city's vehicle fleet in 2018 was composed of 2.4 million vehicles, in which 47% were passenger cars, 25% pick-up trucks, 22% motorcycles, 3% heavy-duty cargo fleet, 2% taxis, and 1.9% collective transport buses [27]. Private vehicles and motorcycles use mainly gasoline, taxis use gasoline and vehicular natural gas (NG), buses use diesel and NG, and heavy vehicles use predominantly diesel as fuel.

Bogotá has a mix of industries, such as manufacturing, chemical, plastics, food and beverages, metallurgy, exploitation of quarries, and mining (nonmetallic) areas, among others. This sector has an intensive use of fossil fuels for goods production and transportation. The industries are located both within the urban area and at nearby municipalities, generating land-use and environmental conflicts [28]. Frequently, PM exceeds national air quality standards in the southern and western areas of the city, given the emission sources, and local and meteorological conditions [24].

Manizales is a medium-sized Andean city (population of 434,403 inhabitants) [29], located in the central-west of Colombia on the western slope of the Cordillera Central at an altitude of 2150 m.a.s.l. [30] (Figure 1). The urban region has an approximate area of 54 km<sup>2</sup> [29] and high population density (~7504 inhabitants per km<sup>2</sup> in the urban area). Meteorological conditions in Manizales are characterized by a high annual precipitation (1670 mm) with a high spatial variability. The diurnal temperature profile ranges between 12–24 °C, high relative humidity between 69–86%, and low wind speeds ( $\leq 2$  m/s). These meteorological conditions enhance minimum dispersion of pollutants in the urban area [31,32]. Local atmospheric chemistry is influenced by proximity to the Nevado del Ruiz volcano (located approximately 28 km from the southeast of Manizales and 140 km from the northwest of Bogotá), one of the most active in Latin America, registering significant activity since 2010 with daily SO<sub>2</sub> and ash emission episodes [33]. Valley–mountain

wind circulation patterns in the city are characterized as ascending by day and descending by night [30,31], with possible transport of volcanic emissions during downslope wind.

Manizales has a high motorization rate, 455.2 vehicles per 1000 inhabitants in the year of 2018 [31,34]. The city's vehicle fleet in 2017 was composed of 169,142 vehicles, in which 48.3% were passenger cars, 47.1% motorcycles, 1.4% taxis, 1.5% collective transport buses, and 1.7% pick-up trucks. Private vehicles and motorcycles use gasoline, taxis use gasoline and NG, and buses and heavy vehicles use diesel as fuel. Manizales has a diverse industrial sector of food and beverages, chemical, plastics, metallurgy, foundry, and minerals, among others. These industries use natural gas, coal, and diesel as fuels [32].



**Figure 1.** Location of the study cities—Bogotá and Manizales, Colombia.

## 2.2. Road Dust Samplings

A total of 41 samples were obtained from the two cities, 20 samplings were carried out in Bogotá and 21 in Manizales. Road dust sampling was conducted in dry weather and at least 48 h after a precipitation event. This condition guarantees that particles are mobilized and not retained by the surface humidity [35]. Field campaigns were carried out between December 2017 and March 2018 in Bogotá, and between July and September 2019 in Manizales (Table 1). As a result of the closeness with the equator (latitudes between 4.5 and 5.1 degrees north for Bogotá and Manizales, respectively), the seasonal variations of temperature and solar radiation between these two cities in question are small, and even negligible if the length of the day is considered. Of all samples, 23 were collected in areas of the cities where commercial activities predominate (average conditions typical of each city)—7 in Bogotá and 16 in Manizales; nine samples were taken in sites of interest such as industrial areas—6 in Bogotá and 3 in Manizales; nine samples were taken in sites of interest with little activity, such as residential areas—7 in Bogotá and 2 in Manizales.

**Table 1.** Average loading of RD<sub>10</sub> at each location.

ID Point	Length	Latitude	Sampling Date	Classification	RD <sub>10</sub> (mg/m <sup>2</sup> )
Bogotá					
1	74°03'59.07" W	4°45'34.48" N	19 December 2017	Commercial Land Use	3.26
2	74°01'28.05" W	4°45'21.12" N	19 December 2017	Residential Land Use	7.04
3	74°01'56.41" W	4°43'12.28" N	20 December 2017	Residential Land Use	14.31
4	74°02'50.84" W	4°41'14.78" N	20 December 2017	Residential Land Use	18.92
5	74°03'58.63" W	4°38'22.69" N	22 December 2017	Commercial Land Use	7.06
6	74°04'14.20" W	4°39'26.73" N	27 January 2018	Commercial Land Use	10.33
7	74°05'38.62" W	4°41'18.01" N	26 January 2018	Commercial Land Use	4.60
8	74°07'25.07" W	4°43'27.55" N	26 March 2018	Commercial Land Use	45.75
9	74°09'11.51" W	4°40'31.39" N	26 January 2018	Industrial Land Use	1.82
10	74°07'25.32" W	4°39'07.38" N	26 January 2018	Industrial Land Use	2.41
11	74°07'11.76" W	4°38'07.65" N	24 January 2018	Industrial Land Use	7.08
12	74°05'52.42" W	4°37'33.33" N	24 January 2018	Industrial Land Use	8.35
13	74°04'36.13" W	4°36'29.91" N	22 December 2017	Commercial Land Use	12.07
14	74°06'01.31" W	4°36'11.21" N	21 December 2017	Residential Land Use	9.40
15	74°08'13.84" W	4°35'48.87" N	23 January 2018	Industrial Land Use	23.14
16	74°10'06.66" W	4°37'47.06" N	3 March 2018	Residential Land Use	21.15
17	74°10'38.49" W	4°35'51.84" N	27 January 2018	Industrial Land Use	6.05
18	74°07'33.38" W	4°34'09.98" N	25 January 2018	Commercial Land Use	10.65
19	74°05'14.33" W	4°34'11.51" N	25 January 2018	Residential Land Use	16.71
20	74°07'02.07" W	4°31'00.78" N	27 March 2018	Residential Land Use	6.04
Manizales					
1	75°27'05.00" W	5°01'40.58" N	2 September 2019	Industrial Land Use	26.75
2	75°27'04.86" W	5°01'58.84" N	20 August 2019	Industrial Land Use	11.72
3	75°27'41.15" W	5°02'10.10" N	2 September 2019	Industrial Land Use	0.77
4	75°27'56.27" W	5°01'55.02" N	31 July 2019	Mixed Land Use	8.54
5	75°27'59.94" W	5°02'02.87" N	30 August 2019	Residential Land Use	2.52
6	75°28'52.54" W	5°02'01.90" N	28 August 2019	Mixed Land Use	1.74
7	75°29'01.39" W	5°02'49.34" N	9 August 2019	Mixed Land Use	1.96
8	75°28'54.88" W	5°03'05.36" N	8 August 2019	Mixed Land Use	7.19
9	75°29'24.04" W	5°03'07.42" N	28 August 2019	Residential Land Use	2.22
10	75°29'12.30" W	5°03'27.14" N	9 August 2019	Mixed Land Use	2.72
11	75°29'38.04" W	5°03'35.71" N	30 August 2019	Mixed Land Use	3.17
12	75°30'06.37" W	5°03'50.11" N	20 August 2019	Mixed Land Use	2.73
13	75°29'55.54" W	5°04'12.86" N	22 August 2019	Mixed Land Use	3.57
14	75°30'20.88" W	5°03'57.53" N	6 August 2019	Mixed Land Use	6.46
15	75°30'38.84" W	5°04'04.84" N	22 August 2019	Mixed Land Use	1.43
16	75°30'55.37" W	5°04'04.19" N	1 August 2019	Mixed Land Use	7.14
17	75°30'54.04" W	5°04'13.44" N	28 August 2019	Mixed Land Use	4.16
18	75°31'00.55" W	5°04'08.76" N	6 August 2019	Mixed Land Use	5.92
19	75°30'20.88" W	5°04'07.18" N	8 August 2019	Mixed Land Use	6.52
20	75°31'29.75" W	5°04'17.44" N	1 August 2019	Mixed Land Use	7.53
21	75°31'51.10" W	5°03'21.28" N	6 August 2019	Mixed Land Use	5.03



A dry dust sampler designed at the Spanish National Research Council (CSIC) was used in the field campaigns. A description of the instrument is available elsewhere [8,35]. Briefly, the instrument consists of a PVC deposition chamber followed by a stainless steel elutriation filter designed to allow passage to only PM<sub>10</sub>-grade material [23]. Road sediments from the pavement of the active traffic lanes were sampled by suction (in a total area of 3.0 m<sup>2</sup> divided into three sections of 1.0 m<sup>2</sup> each), using a vacuum pump (a 30 min sampling was performed at each section with an air flow of 25 ± 2.5 L/min) powered by a field generator (located at some distance downwind with respect to the sampling area). These sediments were immediately resuspended in the deposition chamber. The particles that were small and/or light enough to be carried by the air current continued their journey through the system and entered the stainless steel elutriation filter. Finally, the particles able to penetrate this barrier (PM<sub>10</sub>) were collected on 47 mm filters made of quartz fiber (in Bogotá) and glass fiber (in Manizales). PM<sub>10</sub> losses by sedimentation in the deposition chamber have been shown to be negligible (on average only 0.6% and 0.1% (in volume) of samples sieved at 250 µm and 63 µm, respectively) [35]. The surroundings to the sampling point were observed for safety, traffic characteristics, road condition, and land-use types [23,36].

Filters were baked at 500 °C for 12 h before sampling and then conditioned for 48 h at a constant temperature and relative humidity (20 ± 2 °C and RH 50% ± 5%). The weight of each filter was determined with a microbalance every 24 h. The mass fraction of road dust per m<sup>2</sup> (RD<sub>10</sub>) was calculated at each location by averaging the mass collected at the three sampling points.

### 2.3. Chemical Analysis

After sampling, filters were kept refrigerated in the laboratory in a sealed bag protected from light until analysis. The road dust mass fraction per m<sup>2</sup> (RD<sub>10</sub> load) at each site was calculated by difference in filter weights before and after sample collection, and averaging the mass collected after weighing three times (mg/m<sup>2</sup>). After the sample weights were established, the filters were sent for destructive analysis at the “Laboratorio de Análisis Ambiental de la Universidad Nacional de Costa Rica” (Heredia, Costa Rica). Each filter was divided into small parts for the analysis of ions (3.5 cm<sup>2</sup> approximately for ion chromatography, IC), minerals, and trace elements (6.25 cm<sup>2</sup> approximately for inductively coupled plasma mass spectrometry, ICP-MS).

For mineral and trace elements, a portion of the impacted filter was acid-digested using 10 mL of a mixture of HNO<sub>3</sub> (5.55% v/v) and HCl (16.75% v/v) with deionized water. Microwave heating for 15 min was used by temperature ramps that reached 200 °C. After achieving 200 °C, the temperature was maintained for another 15 min. Then, this solution was filtered through a quantitative filter and diluted with deionized water in a 25 mL volumetric flask. A 15 mL aliquot was transferred to the sampler tubes of the spectroscope. The digested samples were injected to the ICP-MS by a peristaltic pump to the nebulizer system, where it was transformed into an aerosol due to the action of argon gas. The ions that originated in the argon plasma were injected into the mass spectrometer, mass/charge ratio was used for the identification.

Another portion of the filter was leached into 30 mL of deionized water in an ultrasonic bath for 45 min for the extraction of water-soluble ions. The residual liquid was then filtered through a membrane filter and diluted with deionized water in a 50 mL volumetric flask. A portion of the liquid was taken then for further analysis by IC. Sulphate, nitrate, and chloride ions were analyzed [37]. The blanks were submitted to the same chain of custody and treatment as the samples. The Supplementary Materials Table S1 shows the calculations made for the determination of charges for each chemical species.

## 2.4. Enrichment Factors

Enrichment factors (EFs) were calculated for the fraction of road dust using Equation (1). EFs have generally been used to determine the degree of enrichment from anthropogenic sources in road dust [24].

$$EF_{\text{Element}} = \frac{(\text{Element } RD_{10} / \text{Reference } RD_{10})}{(\text{Element Crust} / \text{Reference Crust})} \quad (1)$$

where element is the concentration of the element under consideration and reference is the concentration of the chosen referenced element. Subscripts indicate to which source the concentration refers, sample ( $RD_{10}$ ) or the earth's crust (Crust). EF values were calculated based on the upper continental crust composition (see Supplementary Materials Table S2) using aluminum as the reference element [38]. Aluminum was selected because it is one of the most abundant elements in the earth's crust, and it has been the most used normalizing element in the geochemical literature [38,39]. The enrichment values of elements from anthropogenic sources are classified according to the following ranges: minimal ( $EF = 1-2$ ), moderate ( $EF = 2-5$ ), significant ( $EF = 5-20$ ), very high ( $EF = 20-40$ ), and extremely high ( $EF > 40$ ) [40].

## 2.5. Principal Component Analysis (PCA)

A PCA was carried out to explore qualitative contributions to the chemical composition of  $RD_{10}$  from different factors associated with sources such as brake wear, tire wear, fugitive dust, industrial emissions, and natural emissions, among others. The factors identified by PCA can be associated with emissions sources, but they do not represent the actual sources. The application of a more refined receptor model was not possible due to the lower number of samples. PCA was performed for the dataset recorded in Bogotá and Manizales, and a varimax rotation (with Kaiser normalization) was applied to facilitate the interpretation of the results. In general, this method of rotation maximizes the sum of the variance of the squared loadings, where 'loadings' means correlations between variables and factors. This usually results in high factor loadings for a smaller number of variables and low factor loadings for the rest. The remaining components all have eigenvalues of more than one. In simple terms, the result is that a small number of important variables are highlighted, which makes it easier to interpret the principal component analysis [41].

## 3. Results and Discussion

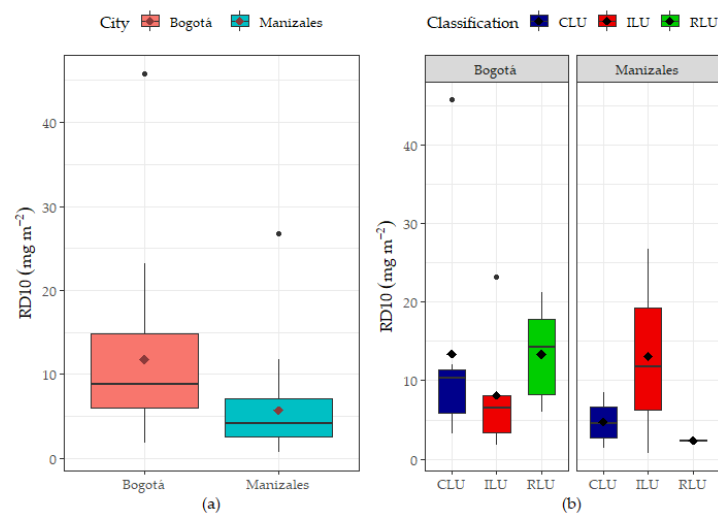
### 3.1. $RD_{10}$ Levels

The  $RD_{10}$  loadings observed in Bogotá (average  $11.8 \text{ mg/m}^2$ , interquartile range between  $6.0-16.1 \text{ mg/m}^2$ ) and Manizales (average  $5.7 \text{ mg/m}^2$ , interquartile range between  $2.4-7.2 \text{ mg/m}^2$ ) were comparable to those reported for European cities such as Barcelona ( $3.0-80.0 \text{ mg/m}^2$ ), Birmingham ( $3.8-42.7 \text{ mg/m}^2$ ), the Andalusia region ( $2.0-22.0 \text{ mg/m}^2$ ), and Turin ( $0.8-42.7 \text{ mg/m}^2$ ), but they were higher than Girona ( $1.3-7.1 \text{ mg/m}^2$ ), Porto ( $0.1-0.9 \text{ mg/m}^2$ ), Paris ( $0.7-2.2 \text{ mg/m}^2$ ), and Zürich ( $0.2-1.3 \text{ mg/m}^2$ ). However,  $RD_{10}$  values in Bogotá and Manizales were lower than New Delhi in India ( $44.0-106.0 \text{ mg/m}^2$ ) (Table 2).

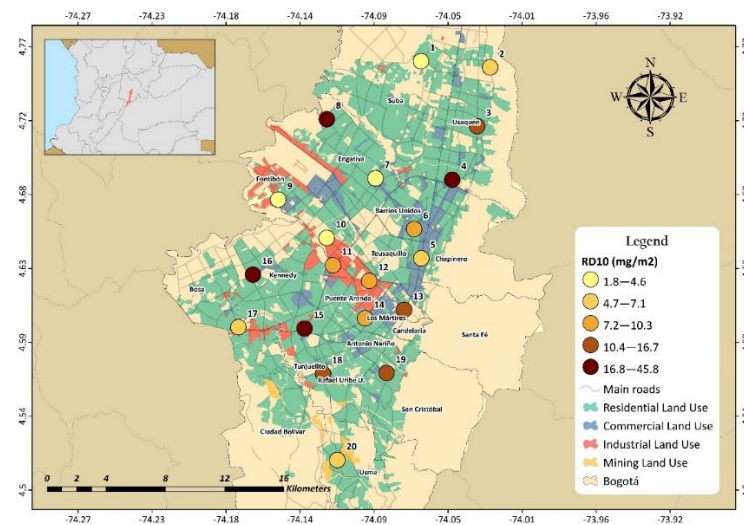
$RD_{10}$  loadings were higher and more variable in Bogotá than in Manizales (Figure 2, panel a). The three highest loadings in Bogotá ( $45.8$ ,  $23.1$ , and  $21.1 \text{ mg/m}^2$  at ID points 8, 15, and 16, respectively, Figure 3) were sites located close to demolition/construction works in the southwest of the city, the same area where ambient  $PM_{10}$  is frequently exceeded. In Manizales, the three highest loadings ( $26.7$ ,  $11.7$ , and  $8.5 \text{ mg/m}^2$  at ID points 1, 2, and 4, respectively, Figure 4) were found in sites with industrial activity and poor road pavement conditions in the eastern areas.

**Table 2.** Comparison of road dust loads from this study and others reported for different regions.

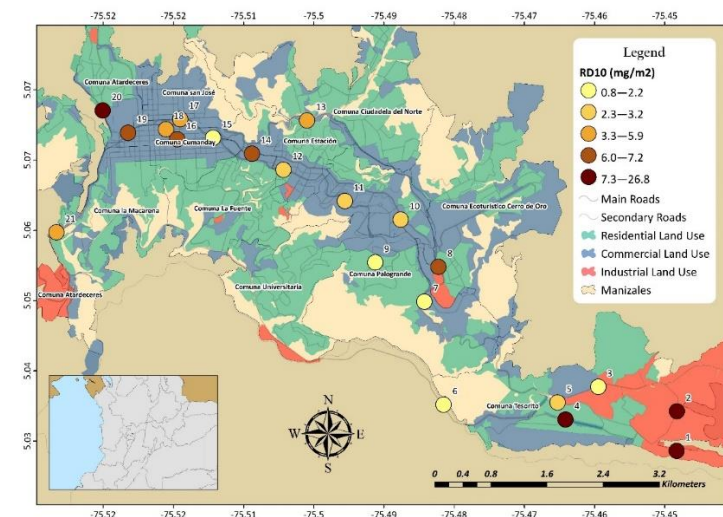
Source	City	Site Features	RD <sub>10</sub> (mg/m <sup>2</sup> )	Season
[35]	Barcelona, Spain	City Center—Urban Zone	3.0–23.0	Summer
		Ring roads with heavy traffic	24.0–80.0	
		Urban tunnel	13.0	
		Demolition/construction area	2.5–471.0	
[8]	Barcelona, Spain	Urban Zone	3.7–23.1	Spring/ Summer
	Girona, Spain	Urban Zone	1.3–7.1	
		Demolition/construction area	48.7	
	Zürich, Switzerland	Urban Zone	0.2–1.3	
[42]	Andalucía, Spain	Urban Zone	2.0–22.0	Summer
[21]	Birmingham, UK	High traffic highway	3.8–21.8	Dry season
		High traffic tunnel	3.0–36.1	
	New Delhi, India	High traffic highway	44.0–106.0	
[17]	Paris, France	Urban Zone	0.7–2.2	Spring
		Cobblestone pavement	10.3	
[7]	Porto, Portugal	Paved road	0.1–0.9	Summer
		Cobblestone pavement	50.0	
[43]	Barcelona, Spain	Urban Zone	1.1–3.4	Summer
	Turin, Italy	Urban Zone	0.8–8.8	Summer and winter
		Roads in proximity to unpaved parks	12.3	
		Demolition/construction area	11.5–42.7	
Current study	Bogotá, Colombia	Urban Zone	1.8–45.7	Dry season
	Manizales, Colombia	Urban Zone	0.8–26.7	

**Figure 2.** Road dust loads boxplots (a) for Bogotá and Manizales and (b) grouped by land use for Bogotá and Manizales—CLU: commercial land use; ILU—industrial land use; RLU—residential land use.





**Figure 3.** Spatial distribution of road dust loads in Bogotá.



**Figure 4.** Spatial distribution of road dust loads in Manizales.

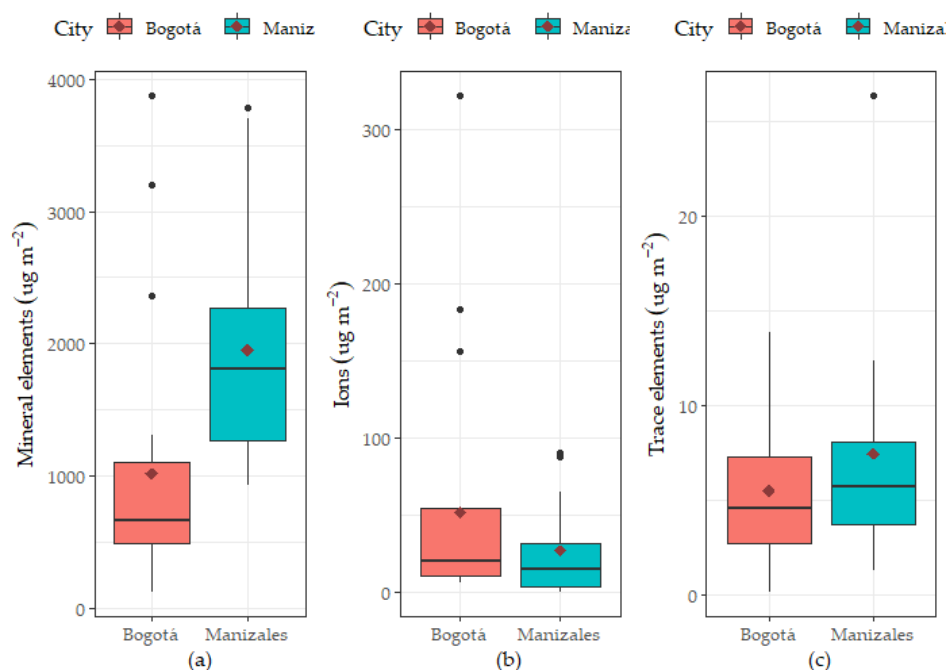
Differences in RD<sub>10</sub> levels in Bogotá and Manizales can be partially explained by urban planning. As it can be identified from Figures 3 and 4, Manizales displays a central axis of commercial activity surrounded by residential areas. The industrial sector is located mainly towards the southeastern areas of the city. In contrast, industrial facilities in Bogotá are spread in the central and southwestern areas of the city, in conjunction with residential and commercial areas. These differences in land-use features determine, in part, levels of RD<sub>10</sub>. The higher values in Bogotá were not only attributed to industrial zones but to areas with a mix of industries, heavy traffic, commercial facilities, and poor road condition, in addition to building and road construction.

Dust loadings in residential areas in Bogotá were larger than those observed in commercial or industrial zones. In contrast, RD<sub>10</sub> levels were the highest under industrial land use in Manizales. Residential areas showed the lowest values (Figure 2, panel b). As it was described, urban development in Bogotá has resulted in zones where industrial and commercial sectors coexist with residential use. One of such zones is the southwest of the city (Figure 3). This area has heavy traffic, a higher fraction of unpaved roads and poor pavement conditions, quarrying/mining activity, and the presence of eroded soils. In Manizales, areas categorized under industrial land use also show heavy vehicular traffic and poor pavement quality, which explains the highest RD<sub>10</sub> values. In residential areas with predominance of light vehicle circulation, dust loadings were the lowest (average

2.37 mg/m<sup>2</sup>). Lastly, in areas with mixed land use (institutional, commercial, and services), the average RD<sub>10</sub> was 4.58 mg/m<sup>2</sup> (Figure 2, panel b and Figure 4).

### 3.2. Chemical Profile

Mineral elements such as Al, Ca, Fe, K, Mg, Mn, Na, and Ti represented 9.0% the total RD<sub>10</sub> mass in Bogotá, with an average load of 1020 µg/m<sup>2</sup>; meanwhile, mineral contribution in Manizales comprised 37.8% of the RD<sub>10</sub> mass (1952 µg/m<sup>2</sup>). Mineral compounds are abundant in the thoracic fraction of road dust [25] and can influence the mass percentage distribution. The mineral fraction was significantly larger in Manizales than Bogotá (Figure 5a). However, other studies have found greater contributions of the mineral component in RD<sub>10</sub> in Colombia, 62% ± 26% in Bogotá and 23% ± 18% in Barranquilla [24,25]. In cities such as Barcelona, New Delhi, and Fushun, the contribution was found to be in the range of 25–35% [21,35,44]. The contribution percentages of the main elements to total mass might be underestimated due to the fact that elements such as metalloids (Si), oxides (Al<sub>2</sub>O<sub>3</sub>, CO<sub>3</sub><sup>2−</sup>, SiO<sub>2</sub>), and organic matter were not analyzed.



**Figure 5.** Total load of (a) mineral elements, (b) ions, and (c) trace elements determined for Bogotá and Manizales.

The fraction of water-soluble ions (SO<sub>4</sub><sup>2−</sup>, NO<sub>3</sub><sup>−</sup> and Cl<sup>−</sup>) represented less than 1.0% of the total mass in Bogotá (average load 51.52 µg/m<sup>2</sup>) and Manizales (average load 26.64 µg/m<sup>2</sup>) (Figure 5b). Previously, a larger ions contribution of 3.8% ± 1.1% was observed in Bogotá [24], similar to Barcelona, Spain (2.4%) [35], but higher than Fushun, China (0.5%) [44].

Trace metals (Ag, As, Be, Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb, Sb, Se, and V), with an average load of 5.54 µg/m<sup>2</sup> and 7.48 µg/m<sup>2</sup>, represented 0.05% and 0.13% of the total mass captured from RD<sub>10</sub> in Bogotá and Manizales, respectively (Figure 5c). In Bogotá, Barcelona, and Fushun, trace metals represented 0.5%, 0.36%, and 0.66%, respectively [24,35,44]. The Supplementary Materials Table S3 shows results of each of the chemical species analyzed.

In Bogotá, 89.4% of the mineral load was composed of four crustal metals: Al (33.6%), Ca (28.0%), Fe (19.5%), and Na (8.3%). The origin of these metals can be associated to: (i) poor road condition and unpaved roads; (ii) low or null vegetation cover [45]; (iii) road and building construction [17,35]; and (iv) quarries and mining areas [46] (Figure 6a). In Manizales, five crustal elements comprised 92.8% of the mineral components: Na (34.3%),

Al (20.5%), K (14.1%), Ca (14.0%), and Fe (9.9%). The sources of these elements are mainly related to the resuspension of fugitive dust (earth's crust material of volcanic geological formation, Figure 6b). In effect, previous studies in the Manizales area have highlighted the presence of high contents of Si, Al, and Fe, as well as compounds such as plagioclase feldspar (Ca-Na) and volcanic glass, among others, in the chemical composition of the soil derived from volcanic ash [47]. Likewise, Trejos et al. [36] found traces of volcanic ash, such as hydrated complexes of Al, Si, and K; heavy metals like As, Hg, Cd, and Pb; and specific compounds, such as ammonia salt and plagioclase feldspar (Ca-Na), in soil dust from Manizales. This volcanic influence may explain the higher loads of Al, K, and Na in Manizales in comparison to Bogotá (Figure 7).

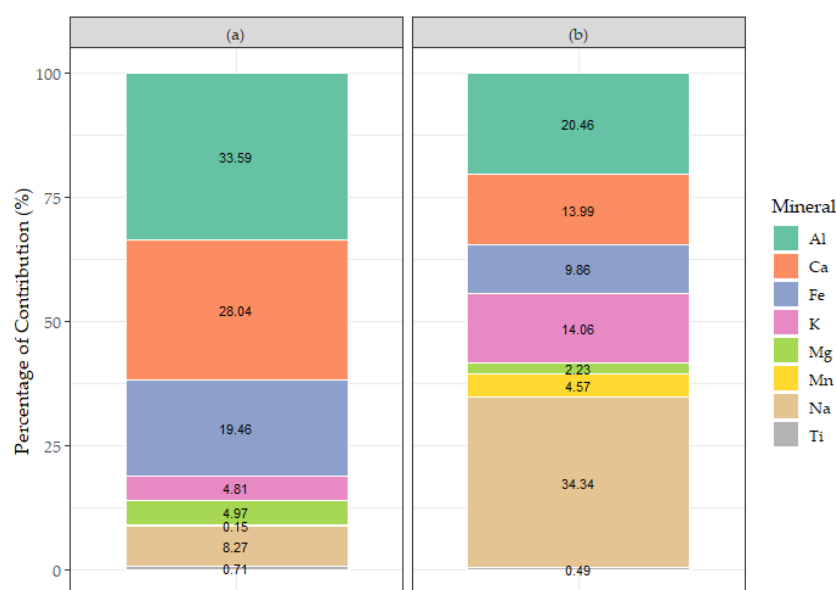


Figure 6. Average composition of main elements for: (a) Bogotá and (b) Manizales.

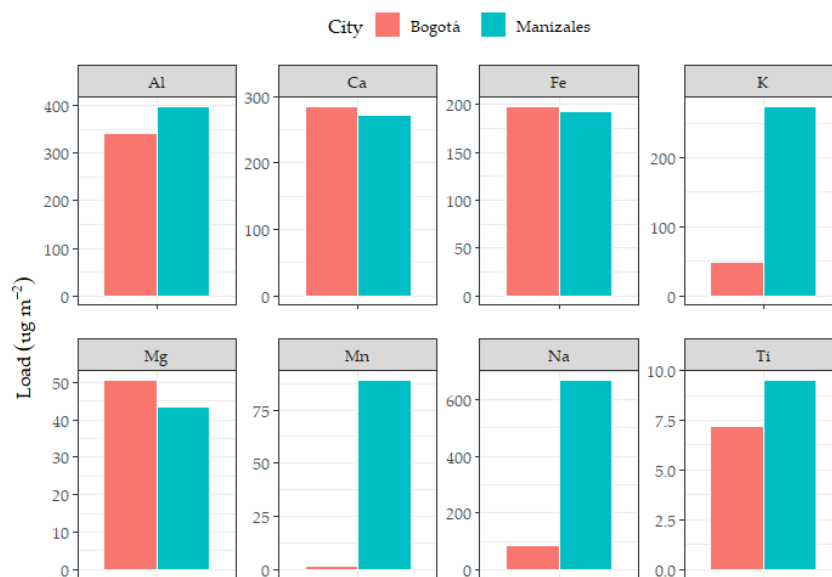
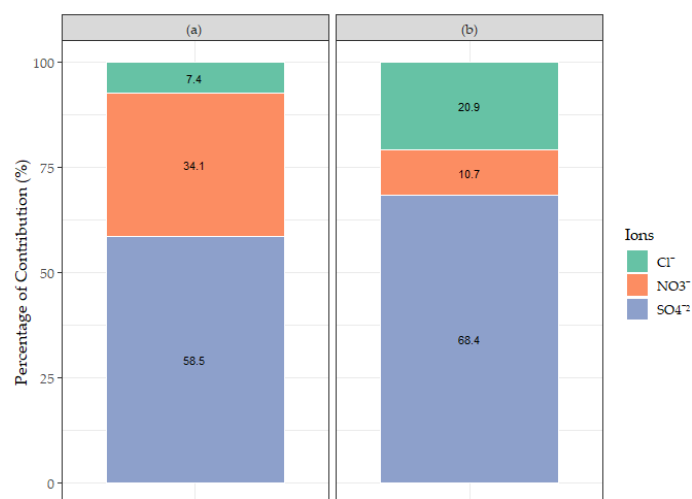


Figure 7. Individual load of main elements obtained in Bogotá and Manizales.

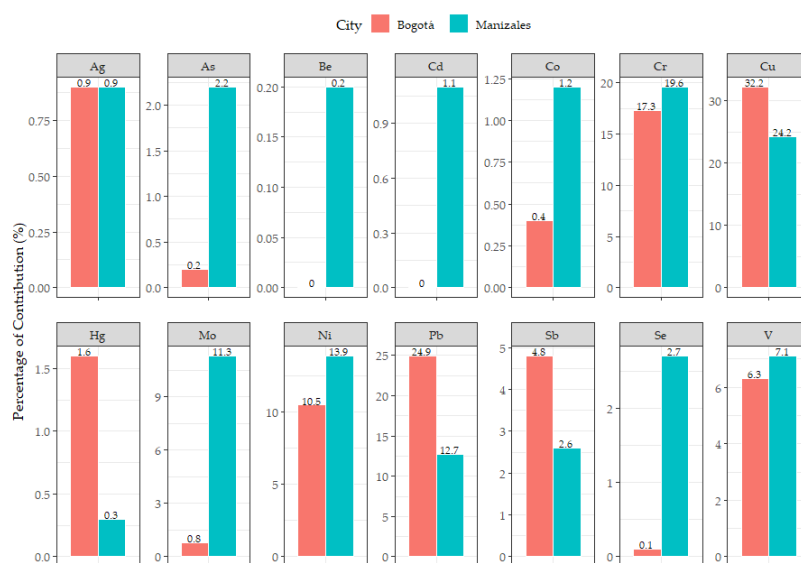
Despite their low fraction in mass, sulphate and nitrate were the most abundant ions in soil dust in Bogotá and Manizales (Figure 8). The precursors of these compounds are associated with pavement wear [46], vehicle exhaust emissions, and formation of

secondary inorganic aerosols [48,49].  $\text{Cl}^-$  showed a contribution of 7.4% in Bogotá and 20.9% in Manizales. The latter is mainly associated with the composition of soils derived from volcanic ash, where  $\text{Cl}^-$ ,  $\text{HCO}_3^-$ , and  $\text{SO}_4^{2-}$  predominate among soluble anions [47] and the deposition of secondary particles formed from precursor gases, such as HCl [24], emitted from the Nevado del Ruiz volcano.



**Figure 8.** Water-soluble ion distribution for (a) Bogotá and (b) Manizales.

Trace elements, such as Cu, Pb, Cr, Ni, V, and Sb, comprised 96.0% of the mass attributed to this component in Bogotá (Figure 9). These metals are commonly associated with vehicle exhaust emissions, especially the use of antioxidants, additives, and lubricants (Cu and Pb), mechanical abrasion (Cu), and tire/brake wear (Cu, Pb, Ni, V, and Sb) [10,17,19]. In the case of Manizales, the contents of Cu and Pb were lower, but there was a significant mass of Mo (11%) in comparison to Bogotá (Figure 9). Molybdenum is mainly associated with emissions from the steel industry [50,51] and brake wear [42]. Both situations are relevant in Manizales, with a relatively large industrial sector and a complex topography that requires a strong use of brakes in steep streets with slopes that could be greater than 22% [52].



**Figure 9.** Average composition of traces for Bogotá and Manizales.

### 3.3. Source Exploration by EFs

Enrichment factors were estimated with the aim to explore the anthropogenic influence of some crustal elements (Ca, Fe, K, and Mn) and trace metals (As, Co, Cr, Cu, Mo, Ni, Pb, Sb, Se, and V). Aluminum was used as a reference element, given its abundance in the upper continental crust. In Bogotá, EFs showed extremely high values ( $EF > 40$ ) for Mo and Sb; very high ( $20 < EF < 40$ ) for Cu and Pb; high ( $5 < EF < 20$ ) for Ni, Se, and Cr; and moderate ( $EF < 5$ ) for Ca, Fe, V, and As. In Manizales, EFs were extremely high ( $EF > 40$ ) for Mo, Se, Sb, and Mn; very high ( $20 < EF < 40$ ) for Cu and As; high ( $5 < EF < 20$ ) for Ni, Cr, and Pb; and moderate ( $1 < EF < 5$ ) for K, Ca, V, Co, and Fe (Figure 10).



Figure 10. Enrichment factors in Bogotá and Manizales.

The presence of Sb, Ba, Cu, Fe, and Mn in road dust samples can be associated with brake wear [53]. Antimony was, in particular, the element that recorded the highest levels of enrichment in Bogotá ( $EF = 271$ ), and it was among the highest in Manizales ( $EF = 130$ ). Despite the relatively low number of vehicles (169,142), the complex topography and steep roads in Manizales imply the heavy use of brakes [36]. In Bogotá, the greater vehicular volume (approximately 2 million vehicles) and traffic congestion explain the frequent use of brakes.

The EF of Sb was higher than those reported in high-traffic cities such as Moscow, Russia ( $EF = 44$ ) [19] and those registered in some roads in Barranquilla ( $EF$  close to 100) [25], but it was similar, and even lower, than those observed in high traffic roads in Viana do Castelo, Portugal ( $100 < EF < 1000$ ) [13]. Ramírez et al. [24] reported a Sb enrichment factor close to 140 for commercial-use areas in Bogotá, which was lower than those found in this study. In accordance with the chemical composition of the brake pads, a Cu/Sb ratio  $> 7$  has been proposed for brake wear emissions in road dust [21,35]. An average Cu/Sb ratio of 7.6 for Bogotá and 9.2 for Manizales was observed, confirming the relevant contribution of brake wear to road dust.

Molybdenum registered an enrichment factor eight times higher in Manizales ( $EF = 1213$ ) than Bogotá ( $EF = 151$ ). The obtained EF values of Mb in cities such as Moscow ( $EF = 7$ ) [19] and Toronto ( $EF = 0.9$ ) [51] were much lower than the values observed in Colombia. Molybdenum has been associated with various industrial emissions, mainly those related to incinerators and with the mechanical/metallurgical industry [19]. Likewise, Mo is associated with brake pad abrasion [51]. A high correlation of Mo with Co (0.96), Hg

(0.73), Pb (0.74), Se (0.61), and V (0.94) in Manizales suggests a common source of origin, related with the industrial activity in the city (see Supplementary Materials Figure S1). In addition, the high correlation of Mo with elements found in volcanic ash, such as Cd (1.0), Pb (0.74), V (0.94), and Fe (0.84), suggests a potential contribution of volcanic emissions to the observed enrichment levels [36].

Selenium showed the second-highest EF value in Manizales ( $EF = 576$ ), a level that was higher than those reported in Bogotá [24] and Viana do Castelo [13]. The enrichment of Se is commonly associated with anthropogenic emissions, including combustion (coal, oil, wood, and biomass), non-ferrous melting, manufacturing, and use of agricultural products. The correlations of Se with As (0.63), Cd (0.62), Mo (0.61), and V (0.63) (Figure S1) suggest a contribution from industrial activity and possible influence of the Nevado del Ruiz volcano [24,36,54].

Cu, Pb, Ni, and Cr presented high and very high EFs ( $5 < EF < 40$ ), widely associated with mechanical abrasion and tire/brake wear [19,24,51]. In Manizales, As also showed a high EF, possibly associated with coal combustion [55] and volcanic emissions (moderate correlation of As with  $SO_4^{-2}$  (0.62) and Se (0.63)).

### 3.4. Factors Associated to Sources in PCA Analysis

In the PCA analysis, 92.2% of the total variance could be explained by five factors in Bogotá and six factors in Manizales (Table 3). Factors that explain at least 10.0% of the data variability were considered. In Bogotá, PC1 explained 42.3% of the variance and showed high correlations with elements associated with dust resuspension, pavement wear, road traffic, and the use of antioxidants, additives, and vehicle lubricants (V, Al, Fe, Ti, As, K, Pb, Co, Mn, Ca, and Cr) [10,17]. PC2, explaining 25.9% of the variance, showed high correlations with Se, Na,  $NO_3$ , and Mg and a moderate relation with  $SO_4$  and Ca. PC2 is associated with construction/demolition activities [35,46]. Lastly, PC3 (16.7% variance explained) is influenced by non-exhaust trace elements, such as Sb, Cu, Mo, and Ni [19].

In Manizales, the first component (PC1) explained 37.8% of the total variance and had high correlations with Co, V, Mo, Cd, Fe, Ca, Pb, Mg, and Ti, suggesting an influence of tire and brake wear, as well as combustion processes [42,50,51]. PC2, a component that explained 18.7% of the total variance, was mainly influenced by  $SO_4$ , As, and K with a lower influence of Sb, Mg, Ti, Al, and Cr. This factor may be associated with the road abrasion and volcanic ash emissions [24,36]. Lastly, PC3 (10.1% of the variance) was correlated with Na, K, and Al, suggesting a contribution related to the resuspension of fugitive dust and volcanic ash [36,47].

**Table 3.** Rotated component of PCA for road dust in Bogotá ( $n = 20$ ) and Manizales ( $n = 14$ ). Only significant values ( $>0.32$ ) are presented. Highest values ( $>0.7$ ) are shown in red.

Rotated Component Matrix <sup>a</sup> —RD <sub>10</sub> Bogotá					Rotated Component Matrix <sup>b</sup> —RD <sub>10</sub> Manizales						
Element	PC1	PC2	PC3	PC4	Element	PC1	PC2	PC3	PC4	PC5	PC6
V	0.97				Co	0.98					
Al	0.96				V	0.97					
Fe	0.96				Mo	0.96					
Ti	0.92				Cd	0.96					
As	0.85	0.35			Fe	0.91					
K	0.82	0.52			Ca	0.83					
Pb	0.78		0.4		Pb	0.79				0.49	
Co	0.78	0.47			Mg	0.75	0.54				
Mn	0.77	0.57			Ti	0.71	0.54				
Ca	0.75	0.61			SO <sub>4</sub>		0.90				
Cr	0.71	0.38	0.52		As	0.35	0.79				
Se	0.34	0.90			K		0.77	0.55			
Na	0.33	0.88			Na	0.49		0.72		0.38	



Table 3. Cont.

Rotated Component Matrix <sup>a</sup> —RD <sub>10</sub> Bogotá					Rotated Component Matrix <sup>b</sup> —RD <sub>10</sub> Manizales						
Element	PC1	PC2	PC3	PC4	Element	PC1	PC2	PC3	PC4	PC5	PC6
NO <sub>3</sub>		<b>0.88</b>			Ni				<b>0.86</b>		
Mg	0.54	<b>0.77</b>			Mn					<b>0.97</b>	
Sb			<b>0.86</b>		NO <sub>3</sub>						<b>0.90</b>
Cu	0.35	0.36	<b>0.83</b>		Cu	0.43	0.39		0.53		0.52
Mo		0.45	<b>0.80</b>		Sb		0.61				0.46
Ni			<b>0.72</b>	0.42	Al	0.58	0.52	0.5			
Cl				<b>0.92</b>	Se	0.56	0.37		0.4	−0.38	
SO <sub>4</sub>	0.49	0.63		0.55	Cr		0.52		−0.66		
Cd					Cl			−0.84			
Eigenvalues	8.9	5.4	3.5	1.5	Eigenvalues	8.3	4.1	2.2	2.1	1.9	1.7
Variance (%)	42.3	25.9	16.7	7.2	Variance (%)	37.8	18.7	10.1	9.3	8.5	7.7
Cum. (%)	42.3	68.3	85.0	92.2	Cum. (%)	37.8	56.5	66.6	76	84.5	92.2

Extraction method: principal component analysis. Rotation method: varimax with Kaiser normalization. <sup>a</sup> Rotation converged in 6 iterations. <sup>b</sup> Rotation converged in 14 iterations.

#### 4. Conclusions

Road dust loadings and chemical composition were investigated in two Andean cities with different sizes, urban planning, and emission sources. RD<sub>10</sub> levels were larger in Bogotá (average of 11.8 mg/m<sup>2</sup>) than in Manizales (average of 5.7 mg/m<sup>2</sup>) due to a higher vehicular volume and a large mix of land use features. Residential areas in Manizales showed the lowest RD<sub>10</sub> values (average of 2.4 mg/m<sup>2</sup>), whereas industrial zones contributed the largest dust loadings (average of 13.1 mg/m<sup>2</sup>). Residential areas mixed with industrial and commercial activity showed significant levels of road dust in Bogotá (average of 13.4 mg/m<sup>2</sup>). Construction and demolition activities were identified as relevant emitters of road dust (dust loadings between 45.8–21.1 mg/m<sup>2</sup>).

Crustal elements (Al, Ca, Fe, K, and Na) were the most abundant elements in road dust in both cities. The complex topography in Manizales and the closeness to the Nevado del Ruiz volcano explain the high levels of crustal and trace metals in road dust, especially Mo, Se, Sb, and Mn. The volcanic activity was also associated with SO<sub>4</sub><sup>2−</sup> and Cl<sup>−</sup>. In Bogotá, Cu, Pb, Cr, Ni, V, Sb, and Mo were associated with combustion and non-exhaust (brake, tire, and road wear) emissions. Finally, the study of the influential factors and chemical composition of road dust, as well as its spatial variability, is crucial for public agencies and research centers to identify the origins of PM, formulate mitigation measures, and identify possible adverse effects on the population.

**Supplementary Materials:** The following are available online at <https://www.mdpi.com/article/10.3390/atmos12091109/s1>, Figure S1: Correlation matrix of chemically analyzed elements for road dust in the cities of (a) Bogotá and (b) Manizales, Table S1: Calculation methodology for determining loads in RD<sub>10</sub>, Table S2: Concentrations in upper continental crust, Table S3: Results of chemical species analyzed for each city.

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