Ecological Indicators 126 (2021) 107667

Contents lists available at ScienceDirect

Ecological Indicators

journal homepage: www.elsevier.com/locate/ecolind

Pollution monitoring in two urban areas of Cuba by using *Tillandsia recurvata* (L.) L. and top soil samples: Spatial distribution and sources

Yasser Morera-Gómez [a,d,*], Carlos Manuel Alonso-Hernández [a], Alejandro Armas-Camejo [a], Orlando Viera-Ribot [a], Mayra C. Morales [b], Daniellys Alejo [c], David Elustondo [d], Esther Lasheras [d], Jesús Miguel Santamaría [d]

[a] Centro de Estudios Ambientales de Cienfuegos (CEAC), AP 5. Ciudad Nuclear, CP 59350 Cienfuegos, Cuba
[b] Department of Chemistry Engineering, Universidad Central de Las Villas, Carretera a Camajuaní Km 5.5, Santa Clara, Cuba
[c] Department of Chemistry, Universidad Central de Las Villas, Carretera a Camajuaní Km 5.5, Santa Clara, Cuba
[d] Instituto de Biodiversidad y Medioambiente (BIOMA), Universidad de Navarra, C/Irunlarrea s/n, 31008 Navarra, Spain

ARTICLE INFO

Keywords:
Tillandsia recurvata
Top soils
Major and trace elements
Cluster analysis
Urban pollution
Cuba

ABSTRACT

This work provides a comprehensive report on the chemical composition of 47 major and trace elements in *Tillandsia recurvata* (L.) L. and top soil samples from the cities of Cienfuegos and Santa Clara in Cuba. The main aims were to provide new information on the urban pollution degree in Caribbean urban regions where the availability of data of urban health indicators are very limited and to identify the main pollution sources.

The abundance of the analyzed elements at both type of samples were different at each urban regions suggesting the influence of various sources. Top soils were slightly contaminated with Zn, V, Ba, Pb, Ni, Cr, Cu, Co and Hg and seriously contaminated with Ni and Cr in Santa Clara. These and other elements such as Se, S, P, Cd, Mo and Ca where highly enriched in *T. recurvata* indicating a significant impact of anthropogenic sources in the air quality of both urban areas. Cluster analysis helped us associate most of the elements with an anthropogenic origin with three main pollution sources: road traffic, industrial emissions and oil combustion. The spatial variability was particularly useful to identify some of these sources including the emissions from diesel and fuel oil combustion in power stations, biomass burning and metallurgic industries. The results also showed that V and Ni were strongly associated to the oil combustion and that V/Ni ratio indices in both indicators can be used to trace this type of sources.

The results presented in this study confirmed the conclusion that both *T. recurvata* and top soils can be used as feasible indicators of the health of Caribbean urban ecosystems and the distribution of the main pollution sources that are affecting them.

1. Introduction

Urban ecosystems are extraordinarily vulnerable to anthropogenic activities. The accelerated increase in the atmospheric concentrations of various pollutants in urban areas is affecting the health of their ecosystems and the well-being of their inhabitants (Markert et al., 2011; McDonald et al., 2018; Pope and Dockery, 2006). Therefore, the establishment and implementation of indicators of changes resulting from environmental pollution in urban areas has become a very effective tool that can support managers in decisions related to public health policy and environmental protection (Piazzetta et al., 2019).

Some of the most commonly used indicators in urban environments are biomonitors, top soils, street and road dust, atmospheric deposition and atmospheric particulate matter (Abbasi et al., 2018; Bermudez et al., 2009; Fernandez-Olmo et al., 2014; Modabberi et al., 2018; Querol et al., 2007). The first three indicators are economically feasible, applicable to large areas and fast alternatives for the analysis of the origin, distribution and effects of urban pollution in comparison with the last two, which usually require longer monitoring periods and a greater consumption of resources.

Biomonitoring is a simple and relatively inexpensive tool for spatio-temporal surveys of environmental quality because it provides the integrated effect of all the environmental factors, including air pollution and weather conditions (Wannaz and Pignata, 2006). The most popular

* Corresponding author at: Centro de Estudios Ambientales de Cienfuegos (CEAC), AP 5. Ciudad Nuclear, CP 59350 Cienfuegos, Cuba.
E-mail address: yasser@ceac.cu (Y. Morera-Gómez).

https://doi.org/10.1016/j.ecolind.2021.107667
Received 8 May 2020; Received in revised form 4 March 2021; Accepted 27 March 2021
Available online 8 April 2021

types of air quality biomonitors are epiphytic plants, such as lichens and mosses (Boltersdorf et al., 2014; Boltersdorf and Werner, 2014; Chaparro et al., 2013; Izquieta-Rojano et al., 2016). It has been demonstrated that epiphytic plants reflect total atmospheric deposition, concentrations and patterns of trace elements (Boltersdorf et al., 2014; Boquete et al., 2015; Malm et al., 1998; Schröder et al., 2013).

Although lichens are frequently used in temperate regions, *Tillandsia* species are more appropriate to monitor atmospheric pollution in tropical areas because they are adapted to hot and dry air conditions showing high tolerance to hydric stress (Cortés, 2004; Malm et al., 1998). The roots of these organisms only act as a means of anchoring to the substrate, so their assimilation function is carried out by foliar trichomes that allow the absorption of water, nutrients and dust directly from the air and atmospheric deposition (Boltersdorf et al., 2014; Montero Alvarez et al., 2017). Therefore, they have a strong independence from the soil. This characteristic makes their elemental composition and physiological responses a reliable indicator of the environment where they are living.

Some species of the genus *Tillandsia*, such as *Tillandsia recurvata* (L.) L., tend to occur naturally in polluted and urbanized sites (Piazzetta et al., 2019). This epiphytic Bromeliaceae is common to grow on different types of trees and also on power lines. They have a reduced stem and non-functional roots and adult individuals tend to be spherical, ca. 10–12 cm in diameter (Zambrano Garcia et al., 2009). Several studies have proved that *T. recurvata* is a highly competitive plant, able to survive in urban environments and in areas with multiple active stresses, thereby showing its potential to be used as a biomonitor of air pollution in urban areas (Castaneda Miranda et al., 2016; Mejía-Echeverry et al., 2018; Piazzetta et al., 2019). *T. recurvata* has some physiological resistance to high levels of SO_2 which may partially explain their abundance in polluted urban environments (Zambrano Garcia et al., 2009; and references therein).

Top soils and street dust have also been employed as indicators of the health of urban ecosystems (Bourliva et al., 2017; Hu et al., 2018; Modabberi et al., 2018). They have proved to be reservoirs of harmful substances, in particular potentially toxic elements emitted by

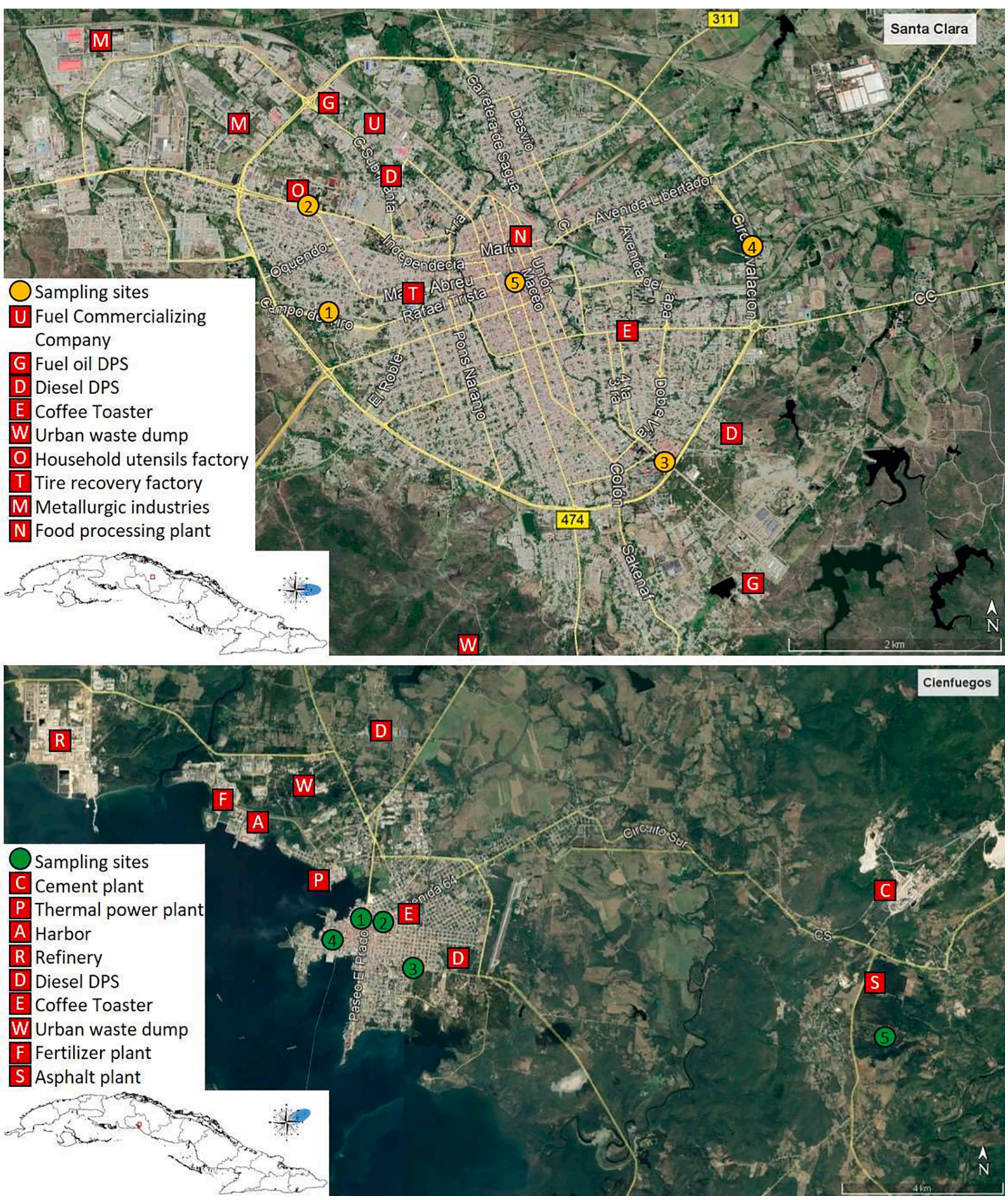

Fig. 1. Sampling sites and potential pollution sources in the areas of study. Decentralized Power Stations (DPS).

industries, hospitals, solid waste deposits, wastewaters, construction activities, painting, road traffic and pavement alteration, among other urban sources. As a consequence, resuspension of top soils and road dust is one of the major sources of airborne particles in urban environments, including the finest particles that have been proved to cause adverse health and environmental effects (e.g. Bourliva et al., 2017; Ferreira-Baptista and De Miguel, 2005).

Many governments worldwide have invested considerable resources and efforts to monitor the health of urban ecosystems employing these and others indicators in order to design and implement policies devoted to reducing the impact of urban pollution. Unfortunately, the availability of data of indicators in Caribbean urban areas is very limited and dispersed geographically.

This study presents a comprehensive report on the chemical composition of 47 major and trace elements analyzed in *T. recurvata* and urban top soil samples at two different cities in Cuba, providing new information on their urban pollution degree. The main goals were to assess the urban pollution employing two different indicators (epiphytic plants and top soil) and identify the main pollution sources based on specific atmospheric tracers.

2. Methods

2.1. Monitoring areas

Samples were collected in two cities located in the central part of Cuba (Fig. 1). Cienfuegos is a coastal city with a population of ~ 160,000 inhabitants and an important tourist and industrial activity. A significant number of potential contamination sources are present within and around this urbanized area (Morera-Gómez et al., 2020). These include a large oil refinery, a fertilizer plant, a thermal power plant fed mainly with S-rich Cuban crude oil (Turtós Carbonell et al., 2007), a cement plant mainly fueled with petroleum-coke and an asphalt plant. In addition, there is a remarkable shipping activity, with a harbor containing large pier and shipbreaking yards, coal storage and packing, scrap storage and classification sites, very intense transportation activities including ferrous scrap trucks and busy ports used for transportation.

Santa Clara, on the other hand, is an inland city with a population of ~ 240,000 inhabitants located 60 km NE from Cienfuegos. The main pollution sources in this region are the Fuel Commercializing Company of Villa Clara, a household utensils factory, metallurgical and metal-mechanic industries including iron and steel smelters and a cycle factory, a tire recovery factory, a soft drink factory, food processing plants, industry of dietary products, among others (Alejo et al., 2011, 2010).

Apart from these specific industrial sectors, there are other important sources of pollution common to both regions: an outdated vehicle fleet, mostly diesel and with very old cars in a bad state of maintenance; several Decentralized Power Stations (DPS), which often operate with diesel or fuel oil (Herrera et al., 2013); suburban waste dumps, where at certain times of the year spontaneous or incidental incineration occurs; steam boilers in schools, university and hospitals mostly using diesel or fuel oil; incinerators mainly located in hospitals and coffee roasting plants. Several villages and agricultural areas and some resorts are also located in these areas. The main pollution sources among those mentioned above are represented in Fig. 1.

Both regions have a tropical climate typical of the Caribbean, characterized by two distinct seasons: dry (approximately from May to October) and wet (November to April). The average annual rainfall is ~ 1360 mm in Cienfuegos and ~ 1070 mm in Santa Clara, mostly produced during the wet period (~80%) (Alejo et al., 2011; Morera-Gómez et al., 2020). The yearly prevalent wind directions in these areas are ENE and E (Fig. 1), respectively.

2.2. Samples collection and preparation

Passive biomonitoring, based on collection of existing plants was used in the study. In this regard, *T. recurvata* was the only species found in both urban environments, confirming its ability to survive in areas with multiple active stresses. Samples of *T. recurvata* were collected in 5 and 4 sites of Santa Clara and Cienfuegos, respectively (Fig. 1). Four sampling campaigns were carried out between 2015 and 2016 taking samples in both dry and wet seasons: in Santa Clara, samples were collected in November 2015 and March, June and September 2016; while in Cienfuegos sampling campaigns took place in June and December 2015, and June and September 2016. The monitoring sites at both cities were located in roads or highways with high level of vehicular traffic (sites 1 and 3 in Cienfuegos and 2, 3 and 4 in Santa Clara) and in parks surrounded by busy roads (sites 2 and 4 in Cienfuegos and 1 and 5 in Santa Clara). At each sampling site, composite samples, made up of 3 to 6 plants from the same or nearby trees were collected guaranteeing enough plant mass (>150 g) prior to the analysis. Samples were taken at a height of 2 m or higher in order to minimize resuspension of soils and particles from roads. Additionally, three composite samples of *T. recurvata* were collected in the Botanic Garden of Cienfuegos (site 5 in Fig. 1) in September 2016 for comparison with the studied sites. Top soils samples (0–5 cm of roadside or parks soils) were collected in the same sampling places at the same time except for the control sample. All samples were stored and labeled in self-sealed polyethylene bags and immediately taken to the laboratory for preparation.

Unwashed samples of *T. recurvata* and top soils were oven-dried at 45C to constant weight, grounded and passed through a 250 μm sieve. The fraction < 250 μm was stored in polyethylene bags until further analysis.

2.3. Chemical analyses

Concentrations of 46 major and trace elements (B, Be, Na, Mg, Al, P, S, K, Ca, V, Ti, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Rb, Sr, Cd, Sn, Sb, Cs, Ba, La, Ce, Pr, Nd, Sm, Eu, Gd, Dy, Ho, Er, Tm, Yb, Lu, Tl, Pb, Th, U, Mo and W) were determined using inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500a) according to the methodology described in González-Miqueo et al. (2010). Briefly, 0.25 g (dry weight) of epiphytic plant and top soils samples were digested in a closed microwave digestion system (CEM Co., Mars X press) using 9 mL of concentrated nitric acid and 12 mL *aqua regia*, respectively. A multi-element solution (Li, Sc, Y, In, Bi) was added to each sample and used as internal standards for further analysis by ICP-MS. Mercury concentrations were determined by atomic absorption spectrometry using a Mercury Analyzer (MA-2000 Series, Nippon) on 50 mg of sample. Soil samples from the 4th sampling campaign were not measured by ICP-MS due to technical issues.

For the quality control of the analytical procedure, two plant materials were used in order to ensure the quality of *T. recurvata* analyses. A pine needle sample from the 15th ICP Forests ring test (Sample 1-Pine needles) and the interlaboratory reference material M2-*Pleurozium Schreberi* (Steinnes et al., 1997). For top soils, the certified reference materials *Soil 0217-CM-73007* and *0217-CM-7003 (Silty Clay Loam)* were used. In both cases, these QC samples were measured every ten samples. In addition, one blank (only reagents) and one duplicated sample were also measured every ten samples. Elemental recovery from reference materials were in the range 76–101% with a relative standard deviation (RSD) below 7% for epiphytic plants and between 74 and 115% with RSD < 9% for top soils. Here, the heavier lanthanoids (Dy to Lu) showed lower recoveries in the range 47–63% (RSD < 6%). The RSD from duplicated samples were generally below 10% for all the elements analyzed in both type of samples. All measured concentrations were above the detection limit evaluated as 3 times the standard deviation in procedural blank samples.

2.4. Enrichment factors

Enrichment factors (EFs) were calculated in order to evaluate the degree of anthropogenic influence on the element concentrations in *T. recurvata* tissue. EF were calculated as follows:

$$EF_X = (C_X/C_{Al})_{T.recurvata}/(C_X/C_{Al})_{UCC}$$

where $(C_X/C_{Al})_{T.recurvata}$ represents the concentration ratio of an elements of interest (*X*) with respect to aluminum (Al) in the plants tissue and $(C_X/C_{Al})_{UCC}$ is the average concentration ratio of X to Al in the upper continental crust (UCC) (Rudnick and Gao, 2014). For their evaluation, EF < 2 means deficiency to minimal enrichment (concentrations are predominantly due to crustal sources), $2 <$ EF < 10 indicates a moderate enrichment (influence of both crustal and non-crustal sources) and EF $\geq$ 10 a high enrichment (non–crustal sources) (Dehghani et al., 2017; Pellegrini et al., 2014). The composition of soil samples wasńt used for normalization because the results showed that many elements in the urban soils analyzed here presented higher concentrations than in the UCC and Cuban background soils (Alfaro et al., 2018, 2015), indicating a significant degree of pollution (see Section 3.1)

2.5. Statistical analysis

Data processing was initially based on descriptive statistics (average, min–max range and standard deviation). The non-parametric Mann-Whitney *U* test was used to evaluate the seasonality (dry and wet periods) of the element concentrations at each city and the non-parametric Kruskal-Wallis test, followed by multiple comparisons post hoc, was performed to compare the content of elements between both studied cities and the control site in Cienfuegos, and also to evaluate the spatial variability at each city.

In order to discriminate the groups of chemical elements as tracers of distinct sources, an explorative hierarchical cluster analysis (HCA), using the Ward's method as linkage function and Pearson correlation as the metric distance, was performed. Before HCA, the variables were standardized by means of z-scores normalization (with a mean of 0 and a standard deviation of 1). Some pollutants were excluded from this analysis taking into account the following assumptions: average EF < 2 or no significant correlation with any other pollutant, because they tended to form factors with single pollutants (Zambrano Garcia et al., 2009). In the case of soils samples only pollutant with concentration ratios to UCC or Cuban background soils > 1 or with a significant

Table 1
Summary of the chemical composition (dry weight) of top soils samples.

		Cienfuegos						Santa Clara						TV[b]	IV[b]
	Unit	mean	min	max	SD	R_{UCC}[a]	R_{CBS}[a]	mean	min	max	SD	R_{UCC}	R_{CBS}		
Ca	%	6.81	3.89	9.02	1.90	2.7	3.3	3.61	1.66	8.19	2.10	1.4	1.7		
Al	%	3.51	2.57	4.40	0.61	0.4	0.5	4.16	2.90	6.01	1.03	0.5	0.5		
Fe	%	3.01	2.35	3.66	0.48	0.8	0.5	5.35	3.95	9.10	1.42	1.4	0.8		
Mg	%	0.59	0.41	0.93	0.18	0.4	0.6	2.39	1.17	5.28	1.06	1.6	2.4		
K	%	0.34	0.25	0.45	0.06	0.1	0.7	0.27	0.14	0.46	0.10	0.1	0.5		
S	%	0.16	0.12	0.19	0.03	2.5	19.8	0.14	0.09	0.23	0.03	2.3	17.8		
Ti	%	0.14	0.01	0.19	0.06	0.4	0.3	0.22	0.13	0.38	0.08	0.6	0.5		
P	%	0.12	0.05	0.21	0.06	1.9	1.6	0.09	0.04	0.28	0.07	1.4	1.2		
Mn	%	0.08	0.04	0.12	0.02	1.0	0.4	0.12	0.09	0.18	0.03	1.5	0.6		
Na	%	0.03	0.02	0.05	0.01	0.01	0.1	0.05	0.04	0.07	0.01	0.02	0.2		
Zn	$\mu g\ g^{-1}$	160.3	62.85	272.7	76.62	2.4	1.8	109.7	46.51	292.9	64.02	1.6	1.2	140	720
Sr	$\mu g\ g^{-1}$	135.8	94.64	182.4	30.48	0.4		107.4	58.28	198.9	39.87	0.3			
Ba	$\mu g\ g^{-1}$	132.0	77.71	181.0	36.12	0.2	1.0	198.1	49.18	460.1	121.9	0.3	1.5	160	625
V	$\mu g\ g^{-1}$	120.4	80.77	161.8	23.74	1.2	0.9	121.3	72.50	168.3	34.79	1.3	1.0	42	250
Pb	$\mu g\ g^{-1}$	119.9	21.69	339.9	108.6	7.1	3.5	67.89	10.70	316.1	84.40	4.0	2.0	85	530
Cr	$\mu g\ g^{-1}$	88.28	53.05	128.9	29.25	1.0	0.2	609.7	131.4	1834.2	566.1	6.6	1.3	100	380
Ni	$\mu g\ g^{-1}$	55.00	29.61	94.66	23.69	1.2	0.2	534.4	93.20	1488.5	461.9	11.4	1.8	35	210
Cu	$\mu g\ g^{-1}$	53.60	25.77	82.26	21.20	1.9	0.6	92.18	21.56	375.9	109.5	3.3	1.1	36	190
Rb	$\mu g\ g^{-1}$	47.85	39.32	58.09	6.69	0.6		36.26	20.64	58.76	12.12	0.4			
Ce	$\mu g\ g^{-1}$	22.20	19.48	24.39	1.41	0.4	0.9	15.34	9.61	21.13	3.54	0.2	0.6		
Sn	$\mu g\ g^{-1}$	13.75	1.94	30.13	10.66	6.5		6.60	0.99	56.65	14.05	3.1			900
Co	$\mu g\ g^{-1}$	11.20	8.57	14.28	2.12	0.6	0.4	54.58	22.94	145.1	37.34	3.2	1.7	9	240
La	$\mu g\ g^{-1}$	11.17	10.09	12.13	0.75	0.4	0.7	6.69	4.35	9.73	1.95	0.2	0.4		
Nd	$\mu g\ g^{-1}$	11.10	10.04	12.13	0.66	0.4	0.6	8.34	5.86	10.77	1.26	0.3	0.5		
As	$\mu g\ g^{-1}$	5.49	4.34	7.34	0.97	1.1	0.5	2.12	0.68	4.66	1.22	0.4	0.2	29	55
Pr	$\mu g\ g^{-1}$	2.81	2.59	3.11	0.16	0.4	0.6	1.97	1.40	2.64	0.35	0.3	0.4		
Sm	$\mu g\ g^{-1}$	2.37	2.05	2.63	0.19	0.5	0.5	2.05	1.36	2.64	0.34	0.4	0.5		
Gd	$\mu g\ g^{-1}$	2.23	1.93	2.46	0.19	0.6	14.9	2.19	1.35	3.06	0.48	0.5	14.6		
Sb	$\mu g\ g^{-1}$	2.12	0.49	5.74	1.46	5.3	0.3	1.32	0.58	3.48	0.80	3.3	0.2	3	15
Dy	$\mu g\ g^{-1}$	1.90	1.67	2.17	0.17	0.5	1.4	2.11	1.26	3.16	0.58	0.5	1.6		
Th	$\mu g\ g^{-1}$	1.52	1.35	1.79	0.14	0.1		0.94	0.44	2.14	0.49	0.1			
Mo	$\mu g\ g^{-1}$	1.41	0.25	2.33	0.82	1.3	2.8	0.80	0.34	1.30	0.29	0.7	1.6	3	200
Er	$\mu g\ g^{-1}$	1.11	0.98	1.27	0.10	0.5	0.5	1.27	0.73	1.89	0.36	0.6	0.5		
Yb	$\mu g\ g^{-1}$	1.04	0.93	1.20	0.09	0.5	0.6	1.18	0.70	1.69	0.30	0.6	0.6		
U	$\mu g\ g^{-1}$	0.84	0.65	1.32	0.18	0.3		0.37	0.20	0.60	0.13	0.1			
Cs	$\mu g\ g^{-1}$	0.80	0.60	1.12	0.18	0.2		0.57	0.15	1.12	0.33	0.1			
Eu	$\mu g\ g^{-1}$	0.68	0.61	0.77	0.05	0.7	22.8	0.64	0.38	0.93	0.15	0.6	21.2		
Cd	$\mu g\ g^{-1}$	0.63	0.21	1.00	0.25	7.0	0.5	0.47	0.22	1.51	0.35	5.2	0.4	0.8	12
Hg	$\mu g\ g^{-1}$	0.48	0.04	1.85	0.68	9.5	4.8	0.13	0.04	0.51	0.12	2.5	1.3	0.3	10
Be	$\mu g\ g^{-1}$	0.40	0.35	0.46	0.04	0.2		0.36	0.24	0.60	0.11	0.2		1.1	30
Ho	$\mu g\ g^{-1}$	0.39	0.34	0.44	0.03	0.5	0.7	0.43	0.26	0.65	0.12	0.5	0.8		
Tm	$\mu g\ g^{-1}$	0.17	0.15	0.19	0.01	0.6	8.5	0.19	0.11	0.28	0.05	0.6	9.5		
Tl	$\mu g\ g^{-1}$	0.17	0.13	0.28	0.04	0.2		0.12	0.06	0.17	0.04	0.1		1.0	15
Lu	$\mu g\ g^{-1}$	0.16	0.14	0.18	0.01	0.5	0.2	0.18	0.10	0.24	0.04	0.6	0.2		

[a] R_{UCC} and R_{CBS}: concentration ratio to UCC (Rudnick and Gao, 2014) and Cuban soils background values (CBS; Alfaro et al., 2018, 2015), respectively.
[b] TV and IV: Target value and Intervention value according to Swartjes (1999), respectively.

contamination degree according to Swartjes, (1999) were included. Spearman's correlation was also performed to support the above statistical analysis.

Plots and statistical analyses were carried out using the software Origin (version 9.0, Origin Lab Corporation, Northampton, MA, USA) and IBM SPSS (version 25, IBM Corp., New York, USA).

3. Results and discussion

3.1. Top soils

3.1.1. Chemical composition of top soil samples

Table 1 summarizes the concentration (dry weight) of major and trace elements determined in top soils in both cities. The concentration ratios with respect to UCC (Rudnick and Gao, 2014) and Cuban background soils values (Alfaro et al., 2018, 2015) are also included for comparison purpose. Major elements were ordered according to the following abundance: Ca > Al > Fe > Mg > K > S > Ti > P > Mn > Na in Cienfuegos and Fe > Al > Ca > Mg > K > Ti > S > Mn > P > Na in Santa Clara. Calcium was the most abundant element in Cienfuegos, being considerably higher than in Santa Clara, where Fe and Al presented higher abundances. Concentration of Mg in Santa Clara and Ca, S and P in both cities, were higher than in UCC and Cuban background soils (e.g. concentration ratio > 1, Table 1). The elements Fe and Mn also showed concentrations higher in Santa Clara than in UCC but lower than in Cuban soils. The rest of major elements showed similar or lower average concentration than UCC and Cuban soils. The higher abundance of Ca observed in Cienfuegos (6.81 ± 1.90%) can be explained because this city is located in a karst area and the influence of the limestone dust is exacerbated by the cement production during all the year in an area where the predominant winds facilitate the arrival of this kind of material to the urban region (see Fig. 1). The higher values of S concentrations in our urban areas (0.16 ± 0.03% in Cienfuegos and 0.14 ± 0.03% in Santa Clara), on the other hand, could be related to the combustion of heavy oils, often employed in Cuba (Núñez Caraballo et al., 2019; Turtós Carbonell et al., 2007).

The abundance of trace elements were very different at both urban regions: in Cienfuegos it was dominated by elements in the sequence Zn > Sr > Ba > V > Pb > Cr > Ni > Cu, whereas in Santa Clara the dominant abundance was Cr > Ni > V > Ba > Zn > Sr > Cu > Pb. Anthropogenic tracers such as Zn, Pb and Hg presented average concentrations higher than both, UCC and Cuban soils (concentration ratio > 1, Table 1) at both cities, suggesting an important degree of pollution. Concentrations of Cu, Sb, Sn and Cd in the soil samples were also higher than in UCC at both cities, but similar or lower than those found in Cuban soils. Concentrations of most of these elements were generally higher in Cienfuegos. However, significantly higher concentrations of Cr (610 ± 566 μg g^{-1}), Ni (534 ± 431 μg g^{-1}) and Co (54.58 ± 37.34 μg g^{-1}) were observed in Santa Clara. No seasonality for any of the determined elements was found in both regions.

Díaz Rizo et al. (2013) reported concentrations of Ni, Cu, Zn and Pb in urban soils from Cienfuegos consistent with our results and found a high degree of contamination for the last three elements. These authors also reported concentrations of Co, Ni, Cu, Zn, Pb and Fe in urban soils from industrial areas, parks and school grounds in La Habana (Diaz Rizo et al., 2011a) within the range of variation observed in most of the elements analyzed in this study. Our results were also comparable with the concentrations of Co, Ni, Cu, Cr, Pb and Fe in urban agricultural soils collected around a steel-smelter plant in La Habana, but significantly lower concentrations were found for Zn (Díaz Rizo et al., 2015). In general, concentrations of Cr, Co and Ni in Santa Clara were significantly higher than those observed in previous studies conducted in Cuba. Nevertheless, Díaz Rizo et al. (2011b) reported similar levels of Cr and Ni in soils from the city of Moa (Holguín province), located near the biggest Ni mines of Cuba.

3.1.2. Top soils contamination degree

As mentioned above, top soils correspond to the uppermost layers of urban soils (0–5 cm) collected in areas of parks very crowded by people and tourists as well as on roadsides roads, where this material can be resuspended easily. Therefore, the importance for human health and the environment becomes more relevant. Due to the lack of official Cuban guidelines for suitable concentrations of heavy metals in soils, we compared our results with the soil quality standards set by the Dutch Authorities (Swartjes, 1999), that establish a Target value (TV) and an Intervention value (IV) and allow soils to be classified as *clean*, *slightly contaminated* and *seriously contaminated* (See Table 1). According to these standards, soils from Cienfuegos can be classified as *slightly contaminated* with Zn, V, Pb, Ni, Cu, Co and Hg; while those form Santa Clara can be classified as *slightly contaminated* with Ba, V, Cu and Co and *seriously contaminated* with Ni and Cr.

3.1.3. Cluster analysis

Urban soils, specifically roadside and park soils, are generally formed by landfills with very different geological origins, which cause great heterogeneity in this type of samples. However, top soils are particularly affected by contributions from all kinds of urban activities and the atmospheric deposition and can help understand where the emissions of many pollutants are originated (Bourliva et al., 2017; Modabberi et al., 2018).

The Cluster analysis performed with the data obtained from the top soils samples in Cienfuegos allowed to identify three main groups of elements (Fig. 2a). Cluster I and II are respectively formed by S-Ca and V-Ni, elements that can be associated to the cement plant, the

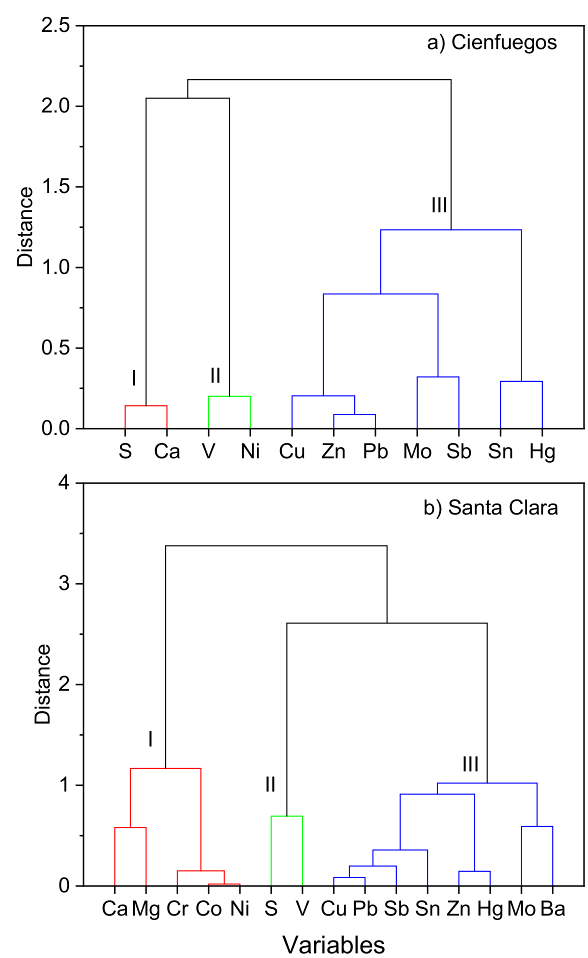

Fig. 2. Hierarchical cluster analysis on chemical elements in top soil samples from Cienfuegos (a) and Santa Clara (b).

surrounding limestone quarries and the combustion of heavy oil and petroleum coke in the cement kiln (Moreno et al., 2010; Turtós Carbonell et al., 2007). Cluster III includes elements as Cu, Zn, Pb, Mo, Sb, Sn and Hg often associated to the road traffic and industrial emissions (Adamiec et al., 2016; Alves et al., 2018; Christoforidis and Stamatis, 2009; Taiwo et al., 2014).

In Santa Clara, three main groups of elements were also distinguished (Fig. 2b). Cluster I grouped Ca, Mg, Cr, Co and Ni. Most of these elements were significantly correlated between them (Cr, Ni and Co; $r > 0.9$, $p < 0.01$; Table S4) and presented a high degree of contamination, suggesting a clear anthropogenic origin. The last elements are considered tracers of anthropogenic sources such as metallurgical industries (Moreno et al., 2010; Pandolfi et al., 2011). In Cluster II are associated V and S, typical tracers of heavy oil and petroleum coke combustion (Moreno et al., 2010), while in Cluster III we observed elements related to the road traffic and industrial emissions (Cu, Pb, Sb, Sn, Zn, Hg, Mo and Ba) as in Cienfuegos.

3.2. *Tillandsia recurvata*

3.2.1. Chemical composition of *T. recurvata* tissues

Table 2 summarizes the concentration of major and trace elements in *T. recurvata* in the cities studied and in the control site. The sequence of abundance for major elements was slightly different in the three sites: Ca > Fe > K > Al > S > Mg > Na > P > Ti in Cienfuegos, Ca > K > Mg > Fe > Al > S > Na > P > Ti in Santa Clara and Ca > K > Al > Fe > S > Mg > Na > P > Ti in the control site. Calcium was the most abundant element at the tree sites and the highest average values observed in Cienfuegos (1.75 ± 0.66%), doubling the value at the control site (0.84 ± 0.16%), could be indicating the high exposure to limestone dust and the cement industry relating emissions as it has been noticed in recent studies in this urban region (Morera-Gómez et al., 2019, 2018). Average concentrations of all major elements at both cities were higher or similar than the values reported for the control site. All the studied elements except Mg showed the highest values in Cienfuegos.

The abundance of trace elements were significantly distinct at the three sites and the elements were ordered according to the following sequence: Zn ~ Mn > Ba > Pb ~ Sr > V > Cu > Cr in Cienfuegos, Mn > Zn > Ni > Cr > Sr > Pb ~ Ba ~ Cu in Santa Clara and Sr > Mn > Zn > Cu ~ Ba > B > Rb > V in the control site. The average concentration of most trace elements was higher in the urban regions than in the control site and, similar to the major elements, highest values were generally observed in Cienfuegos. Only Cr (31.59 ± 23.43 $\mu g\ g^{-1}$), Ni (38.85 ± 29.89 $\mu g\ g^{-1}$) and Co (2.93 ± 2.35 $\mu g\ g^{-1}$) presented higher averages in Santa Clara, in line with what was observed in the soil samples. These high values might be the result of the metallurgical factories located in

Table 2
Summary of the chemical composition (dry weight) of *Tillandsia recurvata* (L.) L. tissues.

		Cienfuegos				Santa Clara				Control				KWA*
	Unit	mean	min	max	SD	mean	min	max	SD	mean	min	max	SD	Sig.
Ca	%	1.75a**	0.82	3.45	0.66	1.14b	0.72	2.10	0.36	0.84b	0.66	0.95	0.16	*0.002*
Fe	%	0.48a	0.29	1.03	0.19	0.34	0.13	1.01	0.20	0.19b	0.16	0.21	0.02	*0.008*
K	%	0.41	0.24	0.78	0.13	0.47	0.24	0.78	0.17	0.47	0.40	0.52	0.06	
Al	%	0.39a	0.26	0.75	0.12	0.23b	0.11	0.46	0.10	0.31b	0.16	0.45	0.14	*0.001*
S	%	0.27	0.15	0.42	0.07	0.22	0.15	0.34	0.05	0.19	0.13	0.23	0.05	*0.046*
Mg	%	0.24a	0.13	0.35	0.06	0.37b	0.21	0.80	0.14	0.19a	0.15	0.23	0.04	*0.001*
Na	%	0.19	0.10	0.95	0.21	0.15	0.09	0.32	0.05	0.15	0.11	0.19	0.06	
P	%	0.08	0.04	0.12	0.02	0.09	0.04	0.14	0.02	0.08	0.07	0.09	0.01	
Ti	%	0.02a	0.01	0.03	0.01	0.01b	0.00	0.02	0.00	0.01b	0.01	0.01	0.00	*0.000*
Zn	$\mu g\ g^{-1}$	101.0a	47.34	174.5	41.84	51.20b	24.01	110.4	22.51	22.17b	16.85	26.24	4.82	*0.000*
Mn	$\mu g\ g^{-1}$	100.3a	52.29	168.5	30.80	77.95	44.70	175.2	33.93	50.49b	37.68	61.88	12.16	*0.008*
Ba	$\mu g\ g^{-1}$	44.44a	19.93	87.12	20.77	19.75b	6.90	43.74	9.08	18.59	15.36	22.82	3.83	*0.000*
Pb	$\mu g\ g^{-1}$	34.97a	13.87	73.68	14.57	19.92b	4.57	54.85	13.53	3.60b	1.87	5.21	1.67	*0.001*
Sr	$\mu g\ g^{-1}$	34.66a	18.57	54.46	13.77	22.03b	10.19	37.32	7.15	62.31a	30.30	85.69	28.69	*0.002*
V	$\mu g\ g^{-1}$	30.39a	17.72	50.49	9.81	12.13b	5.16	33.74	7.57	7.55b	6.81	7.98	0.64	*0.000*
Cu	$\mu g\ g^{-1}$	24.75	13.96	43.18	10.18	19.13	6.51	40.33	10.92	19.07	11.37	24.81	6.93	
Cr	$\mu g\ g^{-1}$	15.66	10.25	29.15	5.06	31.59b	9.99	104.1	23.43	6.17a	5.44	6.56	0.63	*0.002*
Ni	$\mu g\ g^{-1}$	12.43a	8.52	19.86	3.79	38.85b	11.19	125.9	29.89	4.34a	4.17	4.44	0.15	*0.000*
B	$\mu g\ g^{-1}$	10.28	8.17	13.01	1.80	15.39	6.52	29.37	7.33	15.62	12.09	17.69	3.08	
Rb	$\mu g\ g^{-1}$	5.62a	2.06	16.55	4.21	6.66	2.59	15.02	3.22	12.68b	11.18	15.62	2.55	*0.016*
Ce	$\mu g\ g^{-1}$	3.44a	1.83	5.53	1.23	1.29b	0.46	2.81	0.65	0.98b	0.77	1.11	0.18	*0.000*
Co	$\mu g\ g^{-1}$	1.72	0.92	3.03	0.57	2.93a	1.02	10.36	2.35	0.75b	0.69	0.78	0.06	*0.009*
La	$\mu g\ g^{-1}$	1.68a	0.98	2.85	0.58	0.68b	0.27	1.48	0.33	0.51b	0.41	0.58	0.09	*0.000*
Nd	$\mu g\ g^{-1}$	1.28a	0.68	2.65	0.50	0.53b	0.21	1.30	0.27	0.48b	0.38	0.53	0.08	*0.000*
As	$\mu g\ g^{-1}$	1.18a	0.65	3.24	0.67	0.39b	0.15	0.99	0.19	0.66	0.30	1.02	0.36	*0.000*
Sn	$\mu g\ g^{-1}$	0.97a	0.10	2.83	0.73	0.50	0.08	1.37	0.33	0.04b	0.02	0.06	0.03	*0.006*
Mo	$\mu g\ g^{-1}$	0.91a	0.59	1.51	0.25	0.47b	0.25	0.88	0.15	0.41b	0.26	0.52	0.13	*0.000*
Pr	$\mu g\ g^{-1}$	0.35a	0.19	0.67	0.13	0.14b	0.06	0.34	0.07	0.13b	0.10	0.14	0.02	*0.000*
Sm	$\mu g\ g^{-1}$	0.27a	0.14	0.55	0.11	0.11b	0.04	0.28	0.06	0.11	0.08	0.12	0.02	*0.000*
Gd	$\mu g\ g^{-1}$	0.26a	0.14	0.53	0.10	0.11b	0.04	0.29	0.06	0.10b	0.08	0.11	0.02	*0.000*
Se	$\mu g\ g^{-1}$	0.26	0.16	0.48	0.09	0.22	0.11	0.41	0.09	0.19	0.13	0.25	0.06	
Cd	$\mu g\ g^{-1}$	0.24a	0.11	0.41	0.08	0.19	0.06	0.49	0.10	0.07b	0.05	0.08	0.02	*0.005*
Sb	$\mu g\ g^{-1}$	0.24a	0.15	0.38	0.07	0.19	0.11	0.35	0.06	0.07b	0.05	0.08	0.01	*0.003*
Dy	$\mu g\ g^{-1}$	0.22a	0.11	0.45	0.09	0.10b	0.04	0.26	0.05	0.08b	0.06	0.09	0.01	*0.000*
Hg	$\mu g\ g^{-1}$	0.19a	0.08	0.37	0.08	0.10b	0.03	0.18	0.05	0.11	0.11	0.13	0.01	*0.002*
Er	$\mu g\ g^{-1}$	0.12a	0.06	0.26	0.05	0.06b	0.03	0.16	0.03	0.04b	0.04	0.05	0.01	*0.000*
Yb	$\mu g\ g^{-1}$	0.11a	0.06	0.24	0.05	0.06b	0.02	0.15	0.03	0.04b	0.03	0.04	0.00	*0.000*
Cs	$\mu g\ g^{-1}$	0.10a	0.07	0.17	0.03	0.07b	0.03	0.13	0.03	0.19a	0.12	0.22	0.05	*0.002*
W	$\mu g\ g^{-1}$	0.09a	0.03	0.24	0.07	0.10a	0.02	0.32	0.09	0.03b	0.02	0.03	0.00	*0.028*
Eu	$\mu g\ g^{-1}$	0.08a	0.04	0.15	0.03	0.03b	0.01	0.09	0.02	0.03	0.02	0.03	0.00	*0.000*
Tl	$\mu g\ g^{-1}$	0.05a	0.02	0.18	0.04	0.03b	0.01	0.06	0.01	0.04	0.03	0.05	0.01	*0.015*
Tm	$\mu g\ g^{-1}$	0.02a	0.01	0.04	0.01	0.01b	0.00	0.04	0.01	0.01	0.01	0.01	0.00	*0.013*

* Kruskal-Wallis ANOVA. Only Sig < 0.05, that means significant differences between sites, are displayed.
**Different letters means significant differences at the 0.05 level.

the northwestern part of the city. As for soil samples, no seasonal behavior was observed for most of elements in *T. recurvata* at both cities. Only Hg contents in Cienfuegos showed higher values during the dry period. Morera-Gómez et al. (2019) also failed to find seasonality for all these elements in atmospheric bulk depositions in Cienfuegos, suggesting that their concentrations were mostly affected by fluctuations in the emissions rates rather than by the weather conditions.

There are no previous reports on chemical composition of *T. recurvata* in Cuba, and as far as the authors know, neither in other Caribbean islands. Nevertheless, Montero Alvarez et al. (2006) and Estévez Alvarez et al. (2011) reported similar concentrations of V, Co, Ni, Cu, Zn, Sr, Cd and Pb in lichen samples in La Habana. That study also found lower concentrations of Na, Mg, Al, Ca, Cr and Mn than those observed in *T. recurvata* in the present study. Similarly, concentration values observed in the sites studied here are in the range reported in *T. recurvata* in the Mezquital Valley in Mexico (Zambrano Garcia et al., 2009), a region characterized by an intensive agriculture and numerous industrial facilities, including several cement plants. In general, concentrations reported here are within the range found for *T. recurvata* in urban areas in Central and South America (Castaneda Miranda et al., 2016; Cortés, 2004; Mejía-Echeverry et al., 2018; Piazzetta et al., 2019; Sanchez-Chardi, 2016).

3.2.2. Enrichment factors

The EFs for each element at each monitored city is displayed in Fig. 3. The elements S, Hg, Se, Cd, Pb, Zn, P, Cu, Mo, Ca, B and Sb were highly enriched at both cities (EF > 10). In addition, Ni and Cr were highly enriched in Santa Clara, confirming they were mainly originated from non-crustal sources. Sn, V, Ni (only in Cienfuegos), As, K, Cr (only in Cienfuegos), Mg, Mn, Fe, Sr and Co showed a moderate enrichment in both urban regions, whereas the rest of elements presented a minimal enrichment. Although most of the moderately and highly enriched elements showed higher concentrations in Cienfuegos, their enrichment factors were lower than those found in Santa Clara. This situation could be due to the influence of the resuspended limestone dust associated to the cement production activity under the prevailing easterly winds. The different degree of enrichments observed in *T. recurvata* in Cienfuegos is in agreement with those reported for atmospheric particulate matter in this region, including the urban aerosols and atmospheric bulk deposition (Morera-Gómez et al., 2020, 2019, 2018).

3.2.3. Cluster analysis

The cluster analysis in Cienfuegos allowed the association of the elements in three main groups (Fig. 4a). Cluster I includes Ca, a tracer of the activities related to the cement industry and activities in the limestone quarries (Abril et al., 2014; Zambrano Garcia et al., 2009) and S, Pb, Sb, Zn, Cd and Mo, elements usually associated to industrial emissions (Taiwo et al., 2014). Cluster II, on the other hand, groups V and Ni, which are specific tracers of combustion heavy oils (Moreno et al., 2010). Finally, Cluster III includes Cu and Sn. Both Cu and Sn are typical tracers of tire and brake wear emissions and Sn is also emitted from vehicle exhaust (Amato et al., 2016; Grigoratos and Martini, 2015; Johansson et al., 2009; Piazzetta et al., 2019). All these elements were associated with the same sources in recent studies on atmospheric particulate matter carried out in Cienfuegos (Morera-Gómez et al., 2019, 2018), thus confirming the feasibility of this epiphytic plant as a biomonitor of air pollution.

In Santa Clara, elements were divided in three clusters (Fig. 4b). Cluster I, constituted by B, Ca, Se, S, Cu, Hg and V, includes elements associated to the road traffic (e.g. Cu) and the resuspension of soil and road dust or construction activities (e.g. Ca) (Jeong et al., 2019). Along with them, other typical tracers that appear in this cluster are V, S and Se, elements related to the combustion of heavy oil (Mehdi et al., 2013; Moreno et al., 2010). Cluster II separated elements such as Cr, Co and Ni (closely correlated and highly enriched with respect to what was observed in Cienfuegos), Zn, Pb, Mo, Cd and Sn. Thus, Cr, Co and Ni are

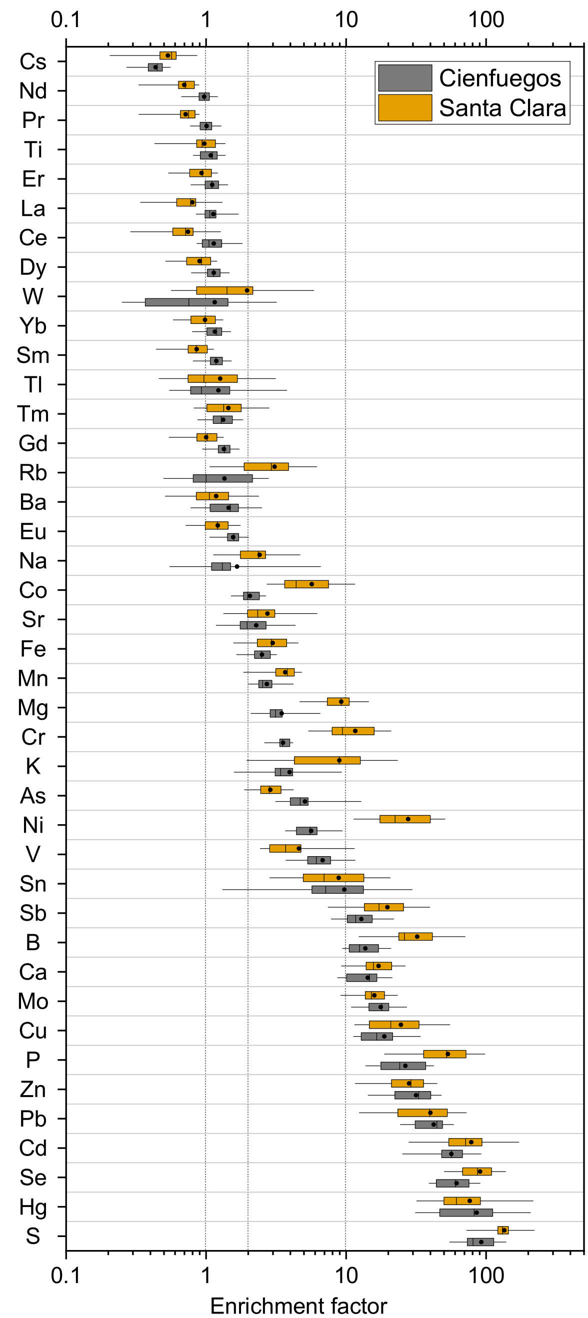

Fig. 3. Boxplot for the element enrichment factors in *Tillandsia recurvata* (L.) L. samples collected in the city of Cienfuegos and Santa Clara. Whisker represent the min–max range.

strongly correlated both in top soils and plant tissue (r > 0.9, p < 0.01; Tables S3 and 4), which supports the conclusion that they could be related to emissions from the metallurgical industries. Cluster III is formed by K, Sb and P. Potassium is highly but negatively correlated with most mineral elements (r > 0.6, p < 0.01; Table S3), indicating a non-mineral origin. Concentrations of K in air are often associated to biomass burning and agricultural sources (Viana et al., 2013; Zambrano Garcia et al., 2009). Similarly, P can also originate from the use of fertilizers and combustion sources, such as fires for agriculture purposes (Giampaoli et al., 2016). Besides fossil fuel combustion, non-ferrous metals derived from refining process and traffic emissions, waste incineration and incineration of sewage sludge are the major anthropogenic sources of Sb (Grigoratos and Martini, 2015; Tian et al., 2011).

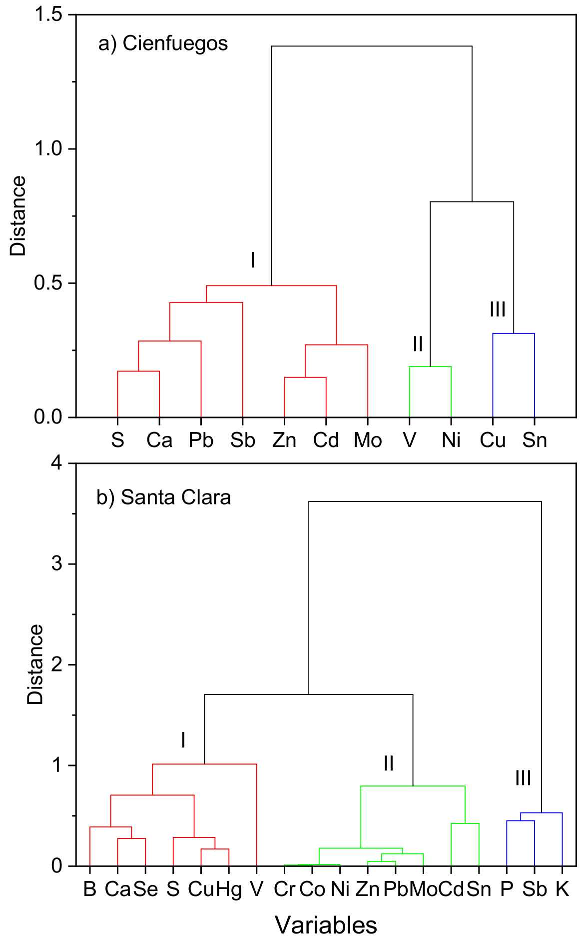

Fig. 4. Hierarchical cluster analysis on chemical elements in *Tillandsia recurvata* (L.) L. samples from Cienfuegos (a) and Santa Clara (b).

3.3. Pollution sources

Several pollutants in top soils and *T. recurvata* samples, showed a significant spatial variability in both cities (Kruskal-Wallis ANOVA at 0.05 level) regardless of the type of soil in which they develop. Moreover, most of them were significantly enriched (Fig. 3) or showed an important degree of contamination (Section 3.1). The spatial variations of those elements was roughly consistent in both type of samples in Cienfuegos (Fig. 5), suggesting that their presence in top soils and *T. recurvata* was mainly caused by the same sources. Elements typically associated to road traffic such as Cu, Zn, Sb and Pb showed the highest concentrations in the sampling site 1, located in the main avenue entering the city and very close to traffic lights. The highest concentrations of S and Ca was also observed in this site for both type of samples, while the lowest ones were found in site 4, one of the most touristic parks in the city. Mo and Sb presented a variability similar to that found for S in *T. recurvata*, with concentrations decreasing from site 1 to 4, while the highest values in top soils were detected in sites 1 and 4. These results suggest that oil combustion is the most likely source of these elements. Although there were no significant differences in the case of the epiphytic plants, there was a tendency to observe the highest Hg contents in site 4, where soils samples also reached maximum concentrations. The origin of Hg in Cienfuegos ambient it is not entirely clear (Morera-Gómez et al., 2020, 2018), but the extremely high values observed in soil samples from site 4 may indicate a point source pollution that should be further investigated. Concentrations of K also showed a different dynamic in *Tillandsia* and top soil samples since *Tillandsia* showed high values in site 2 and low in site 4, whereas for soil samples the highest values were found in sites 3 and 2 and the lowest in site 1. Site 2 is located in a park close to a coffee roaster, which could increase the presence of these elements in the air due to the burning of biomass in this factory. Similarly, Ni also presented a different dynamic, with higher concentrations in roadside soils (site 1 and 3) and lower concentrations in soils from parks (site 2 and 4), while no differences were observed in plant samples. This dynamic may be associated to the wear and tear of very old car parts.

In Santa Clara (Fig. 6), Cr, Co and Ni presented a very similar spatial variation in *T. recurvata,* with their highest concentrations in site 2, which is located just in front of the household utensils factory where electroplating processes with Ni and Cr are performed (Fig. 1). Moreover, other metallurgical plants, mainly iron and steel smelters and a cycle factory, surround this site, being able to provide these elements. Cr, Co and Ni concentrations were also high in site 1, which is located near site 2 and very close to the tire recovery facility. High concentrations of these elements were also found in top soils in site 2 but, in contrast to *Tillandsia*, remarkable contents were also observed in site 4, located close to a bus stop in the main highway of the city, which could be related to the wear and tear of car parts. Concentrations of Ba and Pb presented consistent distributions in both type of samples with their largest values in sites 2 and 5, except for Pb in *T. recurvata* that also

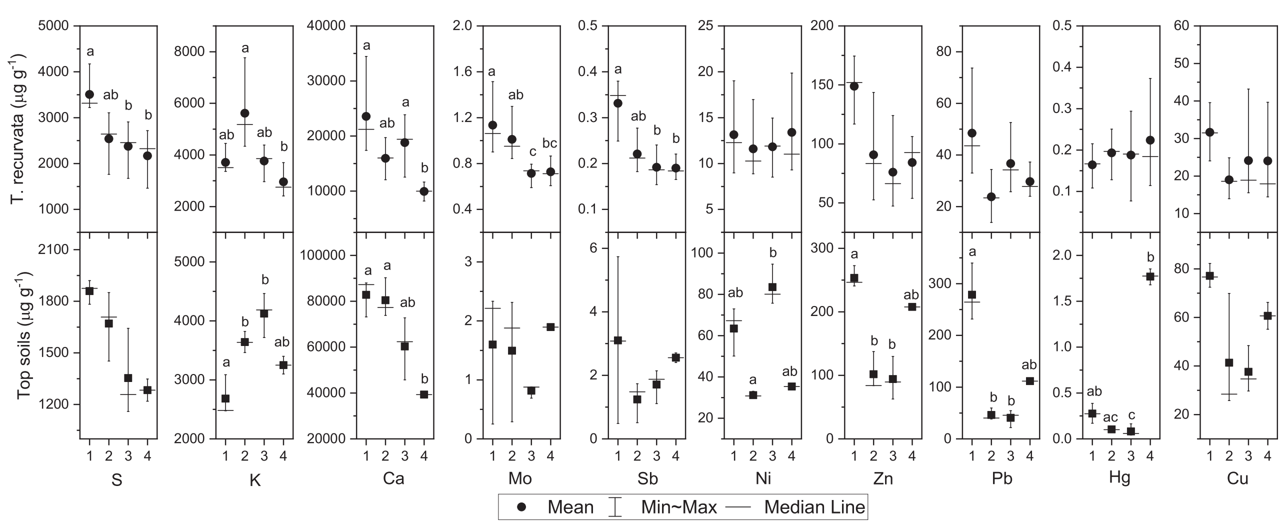

Fig. 5. Interval plot for the concentration of selected elements in *Tillandsia recurvata* (L.) L. and top soil samples collected in Cienfuegos as function of the sampling site. Different letters means significant differences at the 0.05 level (Kruskal-Wallis ANOVA).

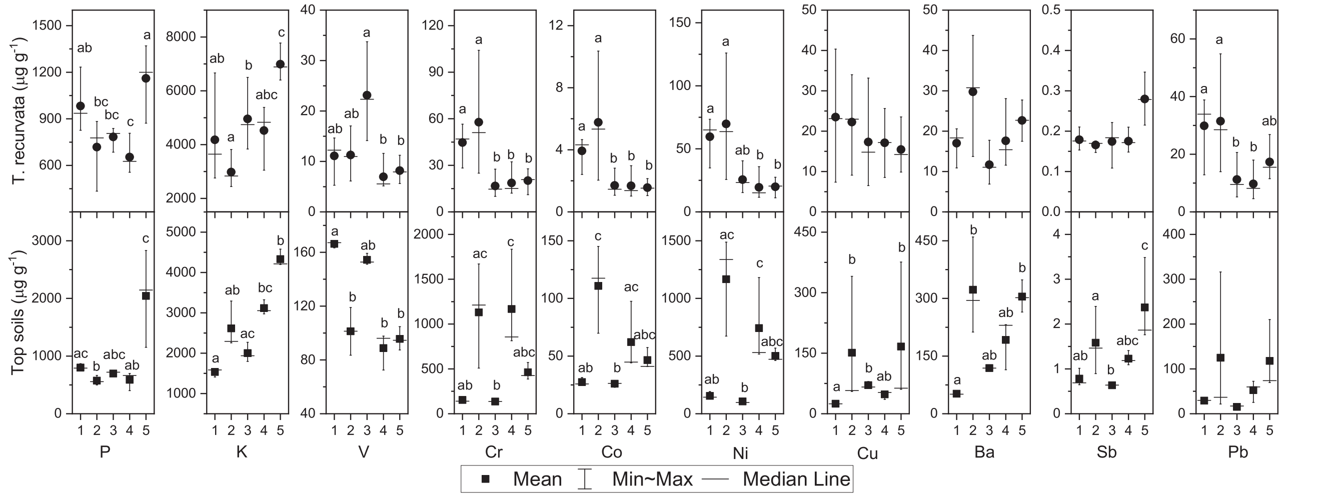

Fig. 6. Interval plot for the concentration of selected elements in *Tillandsia recurvata* (L.) L. and top soil samples collected in Santa Clara as function of the sampling sites. Different letters means significant differences at the 0.05 level (Kruskal-Wallis ANOVA).

presented high contents in site 1. A similar spatial distribution was found for Cu and Sb in soils, with the highest values in sites 2 and 5. However, things were different in *Tillandsia* as no significant spatial variation was found for Cu, and Sb was more consistent with K and P. These last two elements presented similar spatial variations for both type of samples. Thus, both top soils and *T. recurvata* showed the highest contents in site 5, located in a park in the city center downwind of a nearby coffee roaster (~600 m) and very close to food processing plants (~300 m) located at north, that burns wood as fuel, which could facilitate the arrival of emissions related to the biomass burning. This park has usually a high density of birds so we should not rule out the effect of their droppings on the concentrations of K and P both in *T. recurvata* and top soils. However, other elements that should also be affected by this cause such as N (not published data), Ca or Mg did not show so high concentrations in this site. Lastly, V distribution was remarkably different to the rest of elements, with the highest concentrations in site 3 for *T. recurvata* and in sites 1 and 3 for soils. The site 3 is located very close to a small diesel electric generator located at a hospital and close to fuel oil and diesel DPS (Fig. 1). The immediate vicinity of this site is characterized by the presence of several hospitals and the school of medicine, which are equipped with steam boilers that use similar fuels.

3.3.1. *The V/Ni ratio variability*

Based on the above results, we specifically examined the relationships between V and Ni in order to gain insight into their main sources. V and Ni are the most abundant metals present in crude oil so, much of their atmospheric emissions come mainly from oil-fired power plants, petrochemical complexes, and to a lesser extent, shipping transport and other industries that use residual oils and petroleum cokes (Moreno et al., 2010). In Cuba, most of the power plants are fueled with sulfur-rich Cuban crude oil (Turtós Carbonell et al., 2007) and all the cities have several DPS operating with diesel or fuel oil (Herrera et al., 2013). As a result, about 96% of the power generation comes from fossil fuels, and 26% of this is produced by DPS (Herrera et al., 2013). In addition, cement plants generally employ petroleum coke as fuel. Therefore, the combined use of these two elements can be a useful tool to trace the emissions from these kinds of sources.

The relationship between the different concentrations of V and Ni in *T. recurvata* is displayed in Fig. 7a. The V/Ni signature shows very different patterns in both urban regions. In Cienfuegos, all sampling sites follow a single tendency with a strong positive correlation between these two heavy metals ($R^2 = 0.66$, $p < 0.001$), suggesting the presence of a unique major source. The V/Ni ratios in these sites averaged 2.5 ± 0.6 and showed seasonality with lower values in the wet season (2.3 ± 0.3)

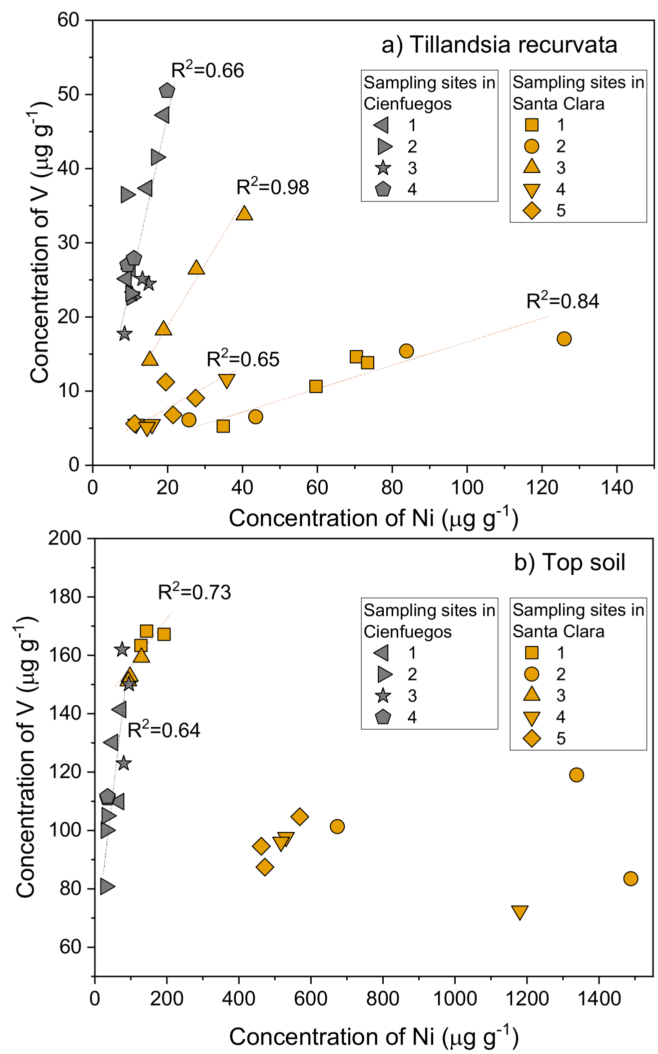

Fig. 7. Relationship between the concentrations of V and Ni in *Tillandsia recurvata* (L.) L. (a) and top soil samples (b) collected in Cienfuegos and Santa Clara. In the case of Cienfuegos, linear fit in both type of samples correspond to samples from the 4 urban sampling sites. In the case of Santa Clara, linear fits correspond to different patterns associated to different sampling sites. See more details in the text.

than in the dry (3.0 ± 0.8) season. These average values were lower than the 4.4 ± 0.2 observed in emissions from the power plant located in this city (Morera-Gómez et al., 2018) and the 7.5 ± 0.2 found in the petroleum coke used in the cement plant located ENE to the city (Morera-Gómez et al., 2019). However, they were consistent with values around 3, typically reported for shipping emissions (Pandolfi et al., 2011; Viana et al., 2014). Morera-Gómez et al. (2018) also found important V and Ni contributions in aerosols from Cienfuegos associated to road traffic emissions (V/Ni ratio of 2.1), since cars in Cuba are very old and mainly fueled with diesel. Other potential emission sources were the DPS and steam boilers; however, the V/Ni signature in diesel and fuel oil, the most used fuels in these sources, was not analyzed in this study.

In Santa Clara, in contrast, three different patterns associated to different sampling sites were distinguished. Samples collected at site 3, very close to the DPS, showed concentrations of V and Ni strongly correlated ($R^2 = 0.98$, $p < 0.05$), with an average V/Ni of 0.92 ± 0.06. Herrera et al. (2013) demonstrated that the site 3 is highly affected by emissions coming from the diesel and fuel oil DPS located NE and SE, respectively. The values observed in samples from sites 1 and 2, also strongly correlated ($R^2 = 0.84$, $p < 0.01$), showed a high Ni-enrichment with the lowest V/Ni ratio (0.15 ± 0.02). These samples were influenced by emissions related to metallurgical industries close to both sites (Fig. 1). Ultimately, samples from sites 4 and 5 exhibited lower concentrations (V/Ni = 0.33 ± 0.02), although significantly correlated as well ($R^2 = 0.65$, $p < 0.05$), with a tendency in-between the observed for the previous sites. A result that suggest a mixed influence from both type of sources, fossil fuel combustion and metallurgical industries. The lower concentrations found in these sites can be also due to the fact they are located from the center to the east of the city, where they are more likely to receive the contribution from rural areas under the prevailing wind direction (Fig. 1).

Top soil samples from Cienfuegos (Fig. 7b) mostly followed a single tendency ($R^2 = 0.64$, $p < 0.01$) with V/Ni averaging 2.4 ± 0.7, very similar to the observed in *T. recurvata,* but with no seasonal variation. Interestingly, higher concentrations of both elements were measured in roadside samples (123–162 $\mu g\ g^{-1}$ of V and 50–95 $\mu g\ g^{-1}$ of Ni, V/Ni = 1.9 ± 0.4), whereas lower concentrations were found in samples from parks (80–111 $\mu g\ g^{-1}$ of V and 30–35 $\mu g\ g^{-1}$ of Ni, V/N = 3.1 ± 0.2) . Result in roadside soil samples was consistent with the V/Ni ratio of 2.1 obtained in aerosols from Cienfuegos associated to road traffic emissions (Morera-Gómez et al., 2018). In the same way, V and Ni in soils of Santa Clara were strongly correlated ($R^2 = 0.73$, $p < 0.05$) in sites 1 and 3, with a V/Ni = 1.3 ± 0.3, but no significant correlation was observed in site 4 and 5 with a lower V/Ni ratio (0.19 ± 0.01). Finally, site 2 showed the most Ni-enriched samples as was observed in *Tillandsia* samples with no correlation.

These results confirmed that V and Ni are strongly associated to the oil combustion and can be used to trace this type of sources. Moreover, the extremely higher concentrations of Ni in Santa Clara appear to be controlled by emissions from metallurgical industries, specially the household utensils factory.

4. Conclusions

This study provides the first report on the chemical composition of *T. recurvata* in Cuba and one of the most comprehensive reports, in terms of number of analyzed elements, in the Caribbean region.

The chemical composition and its variability in both, *T. recurvata* and top soils, suggested the influence of distinct sources type and different levels of contamination in the urban environments of Cienfuegos and Santa Clara. Top soils presented a significant degree of contamination with Zn, V, Ba, Pb, Ni, Cr, Cu, Co and Hg, being Ni and Cr pollution especially important in Santa Clara. These and other elements such as Se, S, P, Cd, Mo and Ca where highly enriched in *T. recurvata,* indicating the impact of anthropogenic sources in the air quality of both urban areas.

The main pollution sources were related to the anthropogenic activities such as: road traffic, oil combustion and different types of industries. The spatial variability of certain pollutants and the V/Ni indices in both type of samples probed to be particularly useful to identify some of these urban pollution sources. Thus, emissions derived from combustion processes in Decentralized Power Stations (DPS) and biomass burning in coffee roasters plants were identified as common sources in both cities. In addition, tracers derived from metallurgical industry were identified in Santa Clara. The obtained results were similar in both indicators, showing a strong consistency between the degree of contamination and the enrichment observed for several elements in the studied areas. Thus, the content and spatial variability of pollutants in both, *T. recurvata* and the upper fraction of soils appear to be good indicators of changes resulting from the urban environmental pollution.

The evidences presented here support, therefore, the implementation of environmental quality monitoring

programs based on these indicators that can help decision makers in the design of efficient urban pollution mitigation plans.

CRediT authorship contribution statement

Yasser Morera-Gómez: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data curation, Writing - original draft, Writing - review & editing, Visualization. **Carlos Manuel Alonso-Hernández:** Conceptualization, Supervision, Project administration. **Alejandro Armas-Camejo:** Writing - review & editing. **Orlando Viera-Ribot:** Investigation. **Mayra C. Morales:** Investigation, Writing - original draft. **Daniellys Alejo:** Investigation, Writing - original draft. **David Elustondo:** Conceptualization, Resources, Writing - original draft, Writing - review & editing, Supervision. **Esther Lasheras:** Investigation, Validation, Resources. **Jesús Miguel Santamaría:** Conceptualization, Resources, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This research received funding from "la Caixa" Banking Foundation. This study was also supported by the International Atomic Energy Agency (IAEA) through the IAEA TC Project CUB/7/008 "Strengthening the National System for Analysis of the Risks and Vulnerability of Cuba's Coastal Zone Through the Application of Nuclear and Isotopic Techniques" and National Program PNUOLU/4 –1/2 No./2014 of the National Nuclear Agency (Cuba).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ecolind.2021.107667.

References

Abbasi, S., Keshavarzi, B., Moore, F., Mahmoudi, M.R., 2018. Fractionation, source identification and risk assessment of potentially toxic elements in street dust of the most important center for petrochemical products, Asaluyeh County, Iran. Environ. Earth Sci. 77, 673. https://doi.org/10.1007/s12665-018-7854-z.

Abril, G.A., Wannaz, E.D., Mateos, A.C., Pignata, M.L., 2014. Biomonitoring of airborne particulate matter emitted from a cement plant and comparison with dispersion modelling results. Atmos. Environ. 82, 154–163. https://doi.org/10.1016/j.atmosenv.2013.10.020.

Adamiec, E., Jarosz-Krzemińska, E., Wieszała, R., 2016. Heavy metals from non-exhaust vehicle emissions in urban and motorway road dusts. Environ. Monit. Assess. 188, 369. https://doi.org/10.1007/s10661-016-5377-1.

Alejo, D., Morales, M.C., Nuñez, V., Bencs, L., Van Grieken, R., Van Espen, P., 2011. Monitoring of tropospheric ozone in the ambient air with passive samplers. Microchem. J. 99 (2), 383–387. https://doi.org/10.1016/j.microc.2011.06.010.

Alejo, D., Morales Perez M. C., Alfonso G., Rosa Dominguez E., Herrera I., Nunez V., 2010. Diagnosis of the air quality in a zone affected by combustion gases sources. Chem. Eng. Trans. 21, 199–204. https://doi.org/10.3303/CET1021034.

Alfaro, Mirelys Rodríguez, Montero, Alfredo, Ugarte, Olegario Muñiz, do Nascimento, Clístenes Williams Araújo, de Aguiar Accioly, Adriana Maria, Biondi, Caroline Miranda, da Silva, Ygor Jacques Agra Bezerra, 2015. Background concentrations and reference values for heavy metals in soils of Cuba. Environ. Monit. Assess. 187 (1) https://doi.org/10.1007/s10661-014-4198-3.

Alfaro, Mirelys Rodríguez, Nascimento, Clístenes Williams Araújo do, Biondi, Caroline Miranda, Silva, Ygor Jacques Agra Bezerra da, Silva, Yuri Jacques Agra Bezerra da, Accioly, Adriana Maria de Aguiar, Montero, Alfredo, Ugarte, Olegario Muñiz, Estevez, Juan, 2018. Rare-earth-element geochemistry in soils developed in different geological settings of Cuba. CATENA 162, 317–324. https://doi.org/10.1016/j.catena.2017.10.031.

Alves, C.A., Evtyugina, M., Vicente, A.M.P., Vicente, E.D., Nunes, T.V., Silva, P.M.A., Duarte, M.A.C., Pio, C.A., Amato, F., Querol, X., 2018. Chemical profiling of PM10 from urban road dust. Sci. Total Environ. 634, 41–51. https://doi.org/10.1016/j.scitotenv.2018.03.338.

Amato, F., Favez, O., Pandolfi, M., Alastuey, A., Querol, X., Moukhtar, S., Bruge, B., Verlhac, S., Orza, J.A.G., Bonnaire, N., Le Priol, T., Petit, J.-F., Sciare, J., 2016. Traffic induced particle resuspension in Paris: Emission factors and source contributions. Atmos. Environ. 129, 114–124. https://doi.org/10.1016/j.atmosenv.2016.01.022.

Bermudez, Gonzalo M.A., Rodriguez, Judith H., Pignata, María L., 2009. Comparison of the air pollution biomonitoring ability of three Tillandsia species and the lichen Ramalina celastri in Argentina. Environ. Res. 109 (1), 6–14. https://doi.org/10.1016/j.envres.2008.08.014.

Boltersdorf, S.H., Pesch, R., Werner, W., 2014. Comparative use of lichens, mosses and tree bark to evaluate nitrogen deposition in Germany. Environ. Pollut. 189, 43–53. https://doi.org/10.1016/j.envpol.2014.02.017.

Boltersdorf, Stefanie H., Werner, Willy, 2014. Lichens as a useful mapping tool?—an approach to assess atmospheric N loads in Germany by total N content and stable isotope signature. Environ. Monit. Assess. 186 (8), 4767–4778. https://doi.org/10.1007/s10661-014-3736-3.

Boquete, M.T., Fernandez, J.A., Carballeira, A., Aboal, J.R., 2015. Relationship between trace metal concentrations in the terrestrial moss Pseudoscleropodium purum and in bulk deposition. Environ. Pollut. 201, 1–9. https://doi.org/10.1016/j.envpol.2015.02.028.

Bourliva, Anna, Christophoridis, Christophoros, Papadopoulou, Lambrini, Giouri, Katerina, Papadopoulos, Argyrios, Mitsika, Elena, Fytianos, Konstantinos, 2017. Characterization, heavy metal content and health risk assessment of urban road dusts from the historic center of the city of Thessaloniki, Greece. Environ. Geochem. Health 39 (3), 611–634. https://doi.org/10.1007/s10653-016-9836-y.

Castaneda Miranda, A.G., Chaparro, Marcos A. E., Chaparro, Mauro A. E., Boehnel, H.N., 2016. Magnetic properties of Tillandsia recurvata L. and its use for biomonitoring a Mexican metropolitan area. Ecol. Indic. 60, 125–136. https://doi.org/10.1016/j.ecolind.2015.06.025.

Chaparro, Marcos A. E., Lavornia, J.M., Chaparro, Mauro A. E., Sinito, A.M., 2013. Biomonitors of urban air pollution: Magnetic studies and SEM observations of corticolous foliose and microfoliose lichens and their suitability for magnetic monitoring. Environ. Pollut. 172, 61–69. https://doi.org/10.1016/j.envpol.2012.08.006.

Christoforidis, Achilleas, Stamatis, Nikolaos, 2009. Heavy metal contamination in street dust and roadside soil along the major national road in Kavala's region, Greece. Geoderma 151 (3-4), 257–263. https://doi.org/10.1016/j.geoderma.2009.04.016.

Cortés, E., 2004. Investigation of air pollution in Chile using biomonitors. J. Radioanal. Nucl. Chem. 262 (1), 169–276.

Dehghani, S., Moore, F., Keshavarzi, B., Hale, B.A., 2017. Health risk implications of potentially toxic metals in street dust and surface soil of Tehran, Iran. Ecotoxicol. Environ. Saf. 136, 92–103. https://doi.org/10.1016/j.ecoenv.2016.10.037.

Díaz Rizo, O., Castillo, F.E., López, J.O.A., Merlo, M.H., 2011a. Assessment of Heavy Metal Pollution in Urban Soils of Havana City, Cuba. Bull. Environ. Contam. Toxicol. 87, 414–419. https://doi.org/10.1007/s00128-011-0378-9.

Díaz Rizo, O., Cazorla, L.L., Céspedes, D.G., Rodríguez, K.D. A., Leyva, O.T., Rieumont, S. O., López, Y.T.B., 2015. Assessment of heavy metal content in urban agricultural soils from the surrounding of steel-smelter plant using X-ray fluorescence. Nucleus 57, 38–43.

Díaz Rizo, O., Coto Hernández, I., Arado López, J.O., Díaz Arado, O., López Pino, N., D′Alessandro Rodríguez, K., 2011b. Chromium, Cobalt and Nickel Contents in Urban Soils of Moa, Northeastern Cuba. Bull. Environ. Contam. Toxicol. 86, 189–193. https://doi.org/10.1007/s00128-010-0173-z.

Díaz Rizo, O., Quintana Miranda, E., Hernández, A., M, C., Cartas Águila, H., ĎAlessandro Rodríguez, K., López Pino, N., López, A., O, J., 2013. Heavy metal and natural radionuclide levels in urban soils from Cienfuegos city, Cuba. Nucleus 54, 17–22.

Estévez Alvarez, J., Montero Alvarez, A., López Sánchez, D., Pupo González, I., Hernández Torres, D., Pérez Arriba, O., Iglesias Brito, H., Wolterbeek, B., 2011. Biomonitoring of the atmospheric pollution in Havana during 2004–2005 survey. Nucleus 50, 18–23.

Fernandez-Olmo, I., Puente, M., Montecalvo, L., Irabien, A., 2014. Source contribution to the bulk atmospheric deposition of minor and trace elements in a Northern Spanish coastal urban area. Atmospheric Res. 145, 80–91. https://doi.org/10.1016/j.atmosres.2014.04.002.

Ferreira-Baptista, L., De Miguel, E., 2005. Geochemistry and risk assessment of street dust in Luanda, Angola: a tropical urban environment. Atmos. Environ. 39 (25), 4501–4512. https://doi.org/10.1016/j.atmosenv.2005.03.026.

Giampaoli, P., Wannaz, E.D., Tavares, A.R., Domingos, M., 2016. Suitability of Tillandsia usneoides and Aechmea fasciata for biomonitoring toxic elements under tropical seasonal climate. Chemosphere 149, 14–23. https://doi.org/10.1016/j.chemosphere.2016.01.080.

González-Miqueo, L., Elustondo, D., Lasheras, E., Bermejo, R., Santamaría, J.M., 2010. Heavy metal and nitrogen monitoring using moss and topsoil samples in a pyrenean forest catchment. Water. Air. Soil Pollut. 210 (1-4), 335–346. https://doi.org/10.1007/s11270-009-0256-9.

Grigoratos, Theodoros, Martini, Giorgio, 2015. Brake wear particle emissions: a review. Environ. Sci. Pollut. Res. 22 (4), 2491–2504. https://doi.org/10.1007/s11356-014-3696-8.

Herrera, I., De Ruyck, J., Ocaña, V.S., Rubio, M., Martínez, R.M., Núñez, V., 2013. Environmental impact of decentralized power generation in Santa Clara City, Cuba: an integrated assessment based on technological and human health risk indicators. Appl. Energy 109, 24–35. https://doi.org/10.1016/j.apenergy.2013.03.085.

Hu, W., Wang, H., Dong, L., Huang, B., Borggaard, O.K., Hansen, H.C.B., He, Y., Holm, P. E., 2018. Source identification of heavy metals in peri-urban agricultural soils of southeast China: an integrated approach. Environ. Pollut. 237, 650–661. https://doi.org/10.1016/j.envpol.2018.02.070.

Izquieta-Rojano, S., Elustondo, D., Ederra, A., Lasheras, E., Santamaria, C., Santamaria, J. M., 2016. Pleurochaete squarrosa (Brid.) Lindb. as an alternative moss species for biomonitoring surveys of heavy metal, nitrogen deposition and delta N-15 signatures in a Mediterranean area. Ecol. Indic. 60, 1221–1228. https://doi.org/10.1016/j.ecolind.2015.09.023.

Jeong, C.-H., Wang, J.M., Hilker, N., Debosz, J., Sofowote, U., Su, Y., Noble, M., Healy, R. M., Munoz, T., Dabek-Zlotorzynska, E., Celo, V., White, L., Audette, C., Herod, D., Evans, G.J., 2019. Temporal and spatial variability of traffic-related PM2.5 sources: comparison of exhaust and non-exhaust emissions. Atmos. Environ. 198, 55–69. https://doi.org/10.1016/j.atmosenv.2018.10.038.

Johansson, Christer, Norman, Michael, Burman, Lars, 2009. Road traffic emission factors for heavy metals. Atmos. Environ. 43 (31), 4681–4688. https://doi.org/10.1016/j.atmosenv.2008.10.024.

Malm, Olaf, de Freitas Fonseca, Marlon, Hissnauer Miguel, Paula, Rodrigues Bastos, Wanderley, Neves Pinto, Fernando, 1998. Use of epiphyte plants as biomonitors to map atmospheric mercury in a gold trade center city, Amazon, Brazil. Sci. Total Environ. 213 (1-3), 57–64. https://doi.org/10.1016/S0048-9697(98)00074-6.

Markert, Bernd, Wuenschmann, Simone, Fraenzle, Stefan, Graciana Figueiredo, Ana Maria, Ribeiro, Andreza P., Wang, Meie, 2011. Bioindication of atmospheric trace metals – with special references to megacities. Environ. Pollut. 159 (8-9), 1991–1995. https://doi.org/10.1016/j.envpol.2011.02.009.

McDonald, Brian C., de Gouw, Joost A., Gilman, Jessica B., Jathar, Shantanu H., Akherati, Ali, Cappa, Christopher D., Jimenez, Jose L., Lee-Taylor, Julia, Hayes, Patrick L., McKeen, Stuart A., Cui, Yu Yan, Kim, Si-Wan, Gentner, Drew R., Isaacman-VanWertz, Gabriel, Goldstein, Allen H., Harley, Robert A., Frost, Gregory J., Roberts, James M., Ryerson, Thomas B., Trainer, Michael, 2018. Volatile chemical products emerging as largest petrochemical source of urban organic emissions. Science 359 (6377), 760–764. https://doi.org/10.1126/science:aaq0524.

Mehdi, Y., Hornick, J.-L., Istasse, L., Dufrasne, I., 2013. Selenium in the environment, metabolism and involvement in body functions. Molecules 18, 3292–3311. https://doi.org/10.3390/molecules18033292.

Mejía-Echeverry, D., Chaparro, Marcos A. E., Duque-Trujillo, J.F., Chaparro, Mauro A. E., Castaneda Miranda, A.G., 2018. Magnetic Biomonitoring as a Tool for Assessment of Air Pollution Patterns in a Tropical Valley Using Tillandsia sp. Atmosphere 9, 283. https://doi.org/10.3390/atmos9070283.

Modabberi, S., Tashakor, M., Soltani, N.S., Hursthouse, A.S., 2018. Potentially toxic elements in urban soils: source apportionment and contamination assessment. Environ. Monit. Assess. 190, 715. https://doi.org/10.1007/s10661-018-7066-8.

Alvarez, Alfredo Montero, Estévez Alvarez, Juan R., do Nascimento, Clístenes Williams Araújo, González, Iván Pupo, Rizo, Oscar Díaz, Carzola, Lázaro Lima, Torres, Roberto Ayllón, Pascual, Jorge Gómez, 2017. Lead isotope ratios in lichen samples evaluated by ICP-ToF-MS to assess possible atmospheric pollution sources in Havana, Cuba. Environ. Monit. Assess. 189 (1) https://doi.org/10.1007/s10661-016-5739-8.

Montero Alvarez, A., Estévez Alvarez, J.R., Iglesias Brito, H., Pérez Arriba, O., López Sánchez, D., Wolterbeek, H.T., 2006. Lichen based biomonitoring of air quality in Havana City west side. J. Radioanal. Nucl. Chem. 270, 63–67. https://doi.org/10.1007/s10967-006-0310-7.

Moreno, Teresa, Querol, Xavier, Alastuey, Andrés, de la Rosa, Jesús, Sánchez de la Campa, Ana María, Minguillón, MariCruz, Pandolfi, Marco, González-Castanedo, Yolanda, Monfort, Eliseo, Gibbons, Wes, 2010. Variations in vanadium, nickel and lanthanoid element concentrations in urban air. Sci. Total Environ. 408 (20), 4569–4579. https://doi.org/10.1016/j.scitotenv.2010.06.016.

Morera-Gómez, Yasser, Alonso-Hernández, Carlos Manuel, Santamaría, Jesús Miguel, Elustondo, David, Lasheras, Esther, Widory, David, 2020. Levels, spatial distribution, risk assessment, and sources of environmental contamination vectored by road dust in Cienfuegos (Cuba) revealed by chemical and C and N stable isotope compositions. Environ. Sci. Pollut. Res. 27 (2), 2184–2196. https://doi.org/10.1007/s11356-019-06783-7.

Morera-Gómez, Y., Elustondo, D., Lasheras, E., Alonso-Hernández, C.M., Santamaría, J. M., 2018. Chemical characterization of PM10 samples collected simultaneously at a rural and an urban site in the Caribbean coast: local and long-range source

apportionment. Atmos. Environ. 192, 182–192. https://doi.org/10.1016/j.atmosenv.2018.08.058.
Morera-Gómez, Y., Santamaría, J.M., Elustondo, D., Lasheras, E., Alonso-Hernández, C. M., 2019. Determination and source apportionment of major and trace elements in atmospheric bulk deposition in a Caribbean rural area. Atmos. Environ. 202, 93–104. https://doi.org/10.1016/j.atmosenv.2019.01.019.
Núñez Caraballo, V., Rodríguez Rojas, R., Gómez Camacho, L., Herrera Moya, I., Morales Pérez, M.C., 2019. Influence on air quality of sulfur dioxide emissions from fixed agricultural sources in Villa Clara (in Spanish). Cent. Agríc. 46, 86–95.
Pandolfi, Marco, Gonzalez-Castanedo, Yolanda, Alastuey, Andrés, de la Rosa, Jesus D., Mantilla, Enrique, de la Campa, A. Sanchez, Querol, Xavier, Pey, Jorge, Amato, Fulvio, Moreno, Teresa, 2011. Source apportionment of PM10 and PM2.5 at multiple sites in the strait of Gibraltar by PMF: impact of shipping emissions. Environ. Sci. Pollut. Res. 18 (2), 260–269. https://doi.org/10.1007/s11356-010-0373-4.
Pellegrini, Elisa, Lorenzini, Giacomo, Loppi, Stefano, Nali, Cristina, 2014. Evaluation of the suitability of Tillandsia usneoides (L.) L. as biomonitor of airborne elements in an urban area of Italy, Mediterranean basin. Atmospheric Pollut. Res. 5 (2), 226–235. https://doi.org/10.5094/APR.2014.028.
Piazzetta, Karime Dawidziak, Ramsdorf, Wanessa Algart, Maranho, Leila Teresinha, 2019. Use of airplant Tillandsia recurvata L., Bromeliaceae, as biomonitor of urban air pollution. Aerobiologia 35 (1), 125–137. https://doi.org/10.1007/s10453-018-9545-3.
Pope, C. Arden, Dockery, Douglas W., 2006. Health effects of fine particulate air pollution: Lines that connect. J. Air Waste Manag. Assoc. 56 (6), 709–742.
Querol, X., Viana, M., Alastuey, A., Amato, F., Moreno, T., Castillo, S., Pey, J., de la Rosa, J., Sánchez de la Campa, A., Artíñano, B., Salvador, P., García Dos Santos, S., Fernández-Patier, R., Moreno-Grau, S., Negral, L., Minguillón, M.C., Monfort, E., Gil, J.I., Inza, A., Ortega, L.A., Santamaría, J.M., Zabalza, J., 2007. Source origin of trace elements in PM from regional background, urban and industrial sites of Spain. Atmos. Environ. 41 (34), 7219–7231. https://doi.org/10.1016/j.atmosenv.2007.05.022.
Rudnick, R.L., Gao, S., 2014. Composition of the Continental Crust. In: Holland, H.D., Turekian, K.K. (Eds.), Treatise on Geochemistry (Second Edition). Elsevier, Oxford, pp. 1–51. https://doi.org/10.1016/B978-0-08-095975-7.00301-6.
Sanchez-Chardi, A., 2016. Biomonitoring potential of five sympatric Tillandsia species for evaluating urban metal pollution (Cd, Hg and Pb). Atmos. Environ. 131, 352–359. https://doi.org/10.1016/j.atmosenv.2016.02.013.
Schröder, Winfried, Pesch, Roland, Hertel, Anne, Schonrock, Simon, Harmens, Harry, Mills, Gina, Ilyin, Ilia, 2013. Correlation between atmospheric deposition of Cd, Hg and Pb and their concentrations in mosses specified for ecological land classes covering Europe. Atmospheric Pollut. Res. 4 (3), 267–274. https://doi.org/10.5094/APR.2013.029.
Steinnes, Eiliv, Rühling, Åke, Lippo, Harri, Mäkinen, Ahti, 1997. Reference materials for large-scale metal deposition surveys. Accreditation Qual. Assur. 2 (5), 243–249. https://doi.org/10.1007/s007690050141.
Swartjes, F.A., 1999. Risk-based assessment of soil and groundwater quality in the netherlands: standards and remediation urgency. Risk Anal. 19, 1235–1249. https://doi.org/10.1023/A:1007003332488.
Taiwo, A.M., Harrison, R.M., Shi, Z., 2014. A review of receptor modelling of industrially emitted particulate matter. Atmos. Environ. 97, 109–120. https://doi.org/10.1016/j.atmosenv.2014.07.051.
Tian, H.Z., Zhao, D., He, M.C., Wang, Y., Cheng, K., 2011. Temporal and spatial distribution of atmospheric antimony emission inventories from coal combustion in China. Environ. Pollut. 159 (6), 1613–1619. https://doi.org/10.1016/j.envpol.2011.02.048.
Turtós Carbonell, L., Meneses Ruiz, E., Sánchez Gácita, M., Rivero Oliva, J., Díaz Rivero, N., 2007. Assessment of the impacts on health due to the emissions of Cuban power plants that use fossil fuel oils with high content of sulfur. Estimation of external costs. Atmos. Environ. 41 (10), 2202–2213. https://doi.org/10.1016/j.atmosenv.2006.10.062.
Viana, M., Hammingh, P., Colette, A., Querol, X., Degraeuwe, B., de Vlieger, I., van Aardenne, J., 2014. Impact of maritime transport emissions on coastal air quality in Europe. Atmos. Environ. 90, 96–105. https://doi.org/10.1016/j.atmosenv.2014.03.046.
Viana, M., Reche, C., Amato, F., Alastuey, A., Querol, X., Moreno, T., Lucarelli, F., Nava, S., Calzolai, G., Chiari, M., Rico, M., 2013. Evidence of biomass burning aerosols in the Barcelona urban environment during winter time. Atmos. Environ. 72, 81–88. https://doi.org/10.1016/j.atmosenv.2013.02.031.
Wannaz, Eduardo D., Pignata, María L., 2006. Calibration of four species of Tillandsia as air pollution biomonitors. J. Atmospheric Chem. 53 (3), 185–209. https://doi.org/10.1007/s10874-005-9006-6.
Zambrano Garcia, A., Medina Coyotzin, C., Rojas Amaro, A., Lopez Veneroni, D., Chang Martinez, L., Sosa Iglesias, G., 2009. Distribution and sources of bioaccumulative air pollutants at Mezquital Valley, Mexico, as reflected by the atmospheric plant Tillandsia recurvata L. Atmospheric Chem. Phys. 9, 6479–6494.