Distribution Maps of the Different Levels of Elemental Concentrations Accumulated by the Lichen in the Northeast of Algeria

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Abstract
An evaluation of environmental pollution in the region of Bordj Bou Arreridj (BBA), Algeria according to metallic trace elements has been carried out, to determine the levels of the 10 elements accumulated in lichens and the different sources found in the region. A total of 192 samples of *Xanthoria parietina* lichen were collected over an area of 3920.42 km². Sampling sites include urban sites, rural sites, green parks, sites near high traffic streets and industrial enterprises. The lichen samples were analyzed by FAAS for the ten elements and their concentrations were mapped. Concentrations of Pb, Cd, Sb and Zn were higher at urban sites and increased with proximity to highways and industrial areas. These results suggest that the composition of lichen elements is strongly affected by road traffic. While the sources of the elements Co, Ni, Fe, Mn and Cr probably come from dust from quarrying and contaminated soil deposits in particular, to the north and west of the region. This mapping of metal pollution can establish the first biological monitoring network in the study area.
Keywords: Biomonitoring of lichens, Metallic elements, Pollution sources, Distribution maps, BBA.
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1. Introduction

Emissions such as the elements have long been recognized as potential threats to the environment and human health (Azimi *et al.* 2005). In this problem, the metallic trace elements (MTE) that are mentioned in the literature constitute the most important category of atmospheric pollutants. (Pacyna & Pacyna 2001; Gombert *et al.* 2005; Gouzy & Ducos 2008; Rauch & Pacyna 2009; Aslan *et al.*2011; Sen & Peucker-Ehrenbrink 2012; Agnan *et al.* 2013).

Air pollution monitoring is essential in order to control air quality and to be able to identify and reduce anthropogenic sources (Garrec & Van Haluwyn 2002). Road traffic and dust deposits are two of the most serious sources of air pollution (Wu *et al.* 2020). In order to follow this pollution with the measurement of MTE in the atmosphere, physical and chemical methods as well as biological methods are available (De Temmerman *et al.* 2004). Physico-chemical measurements are objective and precise methods, but the use of this technical equipment is not widespread for economic reasons, compared to biological methods (Conti & Cecchetti 2001; Wolterbeek 2002; Tian *et al.* 2015).

Plants are known to be very suitable organisms for biological monitoring of air quality and they can be very effective in detecting environmental changes (Markert *et al.* 1997; Mulgrew & Williams 2000).

The use of lichens as bio-monitors in air pollution biological monitoring studies evolved from long-standing observations of the relationships between air pollution and the presence of lichen species (Wolterbeek *et al.* 2003; Rzepka & Cuny 2008; Loppi 2019). This use offers the means to carry out preliminary estimates of the propagation of this pollution, the location and identification of pollution sources, and the method of mapping pollution levels in a given area (Sloof & Wolterbeek 1993; Loppi *et al.* 2004). Spatio-temporal maps of atmospheric pollution levels are carried out to monitor MTE near sources of road and industrial emissions (Cuny 2012). The analysis of pollutants in atmospheric deposits is facilitated by the bioaccumulation method, due to the absence of a root system; lichens take up and accumulate concentrations of MTE in their thalli from wet and dry deposition (Shukla *et al.* 2014; Sett & Kundu 2016).

The most common epiphytic lichen, *Xanthoria parietina*, is the species most used in biological monitoring studies (Nash 2008), because of its ability to accumulate high amounts of MTE in polluted areas (Nimis *et al.* 2001; Scerbo *et al.* 2002 ; Dörter *et al.* 2020). This species was chosen as the target organism of the present study in the passive biological surveillance for the accumulation of MTE in the region of Bordj Bou Arreridj (BBA) in northeast Algeria. The study was carried out with the aim of establishing distribution maps of MTE concentrations within urban, industrial and rural sites at BBA and identify the sources of air pollution emissions in the region and their impact.

2. Materials and methods

2.1 Study area

The sampling was carried out in the region of BBA located in the North East of Algeria, which has a total area of 3920.42 km². Sampling strategies were put in place taking into account topography, urbanization and industry. Was carried out in the region of more than 658,968 inhabitants, is centered in the east and the center of the region, where high levels of air pollutants occur mainly emitted by road traffic and industry. In contrast, many mountainous sites are sparsely populated, devoid of local sources of atmospheric pollution, but the harvest has an environmental impact.

The region is determine by a semi-arid continental climate, more humid on the north side in mountainous sites with higher elevations and drier on the south side.

2.2. Sample processing and analysis

Lichen samples were collecting from 34 sites near main roads, public gardens, rural sites, urban sites and industrial areas (Figure 1). Lichen samples were taken naturally in situ from tree trunks. Lichen thalli are collected from at least (3 to 6) trees at a height of more than 1 m above the ground to avoid any contamination that could influence the composition of the sample (Nimis *et al.* 2002).

The collected samples are kept in paper bags and are referenced by all the necessary indications of site and sampling, then are transported to the laboratory in a clean environment for processing and mineralization. *Xanthoria parietina* thalli were digesting in the laboratory and under aseptic conditions (Picardie 2004). The most widely used MTE solubilization method is mineralization by acid attack. It is carried out in a firm and hot environment at 150°C to avoid loss of volatile elements. All steps are performed in Teflon containers to prevent moisture absorption.

Lichen samples were washed with distilled water to remove plant debris using plastic tweezers and a ceramic knife, after rinsing, the thalli were put in filter papers and oven dried for 72 hours. The dried thalli were ground into a fine powder so as to promote the dissolution of the elements to be analyzed. The powder obtained after grinding is calcined in a muffle furnace, the temperature of which is gradually increased to 500 °C from 02 until 04 hours, using quartz capsules. Then the fine powder is placed in an acid solution ; about 0.5 g of dry plant material with 5 ml of hydrofluoric acid (HF 40%) and 1.5 ml of perchloric acid ($ClHO_4$ 70%). The solution is homogenized before heating to 160°C, until the solution has completely evaporated. After almost total evaporation, a second dissolution is carried out by adding 0.5 ml of nitric acid (65% HNO_3) and 5 ml of distilled water. The solution is left to stand for 30 min in the refrigerator at a temperature of 4 °C, then put back into solution on a hot plate at 60°C for one hour. The resulting mixture is transferred to a 50 ml flask for filtration adjusting the volume with distilled water.

Then these concentrations of the trace metal elements Cd, Cu, Cr, Co, Fe, Mn, Ni, Pb, Sb and Zn are determined by flame atomic absorption spectrophotometry (FAAS).

2.3. Data analysis

The concentrations of elements accumulated by *X. parietina* at the levels of urban sites, sites near roads, rural sites and industrial areas of the BBA region were mapping from the coordinates of the sampling points by the ArcMap 10.5 software. The maps were using to represent the surface distribution of metal pollution in the study area. The results of the analyzed lichens were evaluated using Statistica 10 software.

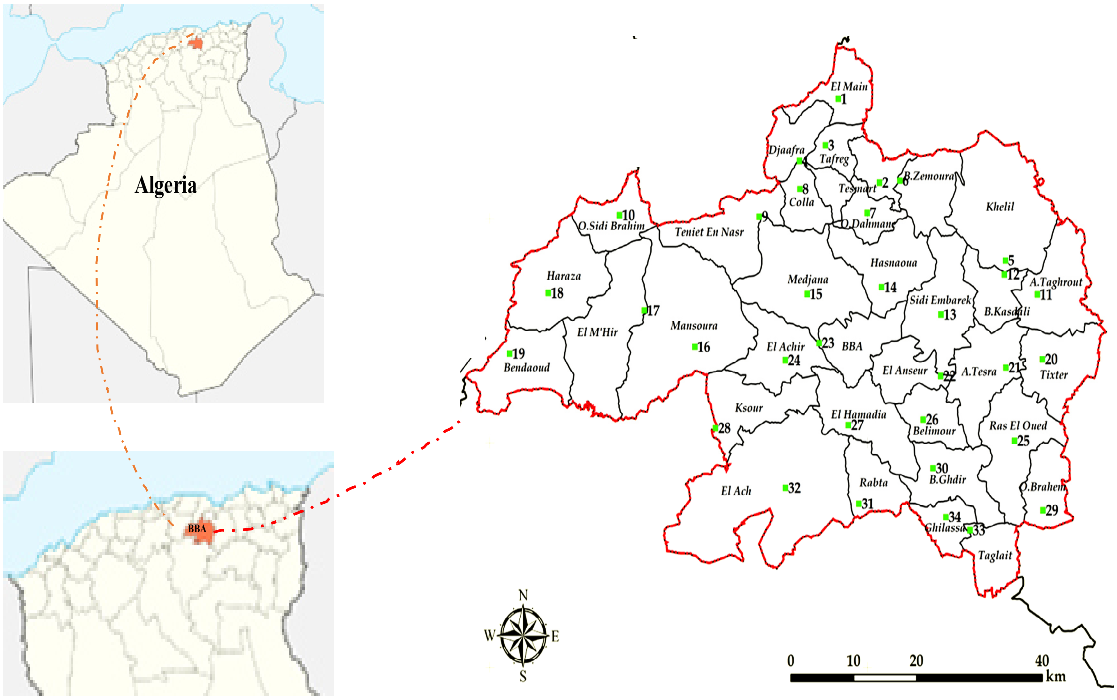

Figure 1: sites sampled in the BBA region North-Est of Algeria

3. Results and discussion

3.1. Variation in MTE concentrations

A principal component statistical analysis is performe on all the sites to determine the variability of the concentrations of MTE estimated in the thalli of *X. parietina* and the relationship of these concentrations with the sites studied. The mean concentrations of ten elements in the sites sampled at the level of the BBA region varied between 3.25 mg/kg and 43184.38 mg/kg depending on the location of the site (Table 1).

It is noted that all the concentrations of MTE estimated in the thalli exceed the certified standard, except the Pb concentrations are lower than the certified standard or the minimum values have been recorded in rural sites. While, Iron has the highest average concentration in the entire study region (43184.88±16373.66 mg/kg).

The coefficient of variation (CV) of MTE concentrations in the thalli measured is varied between 18.2% and 62%, high CV values were observed for Ni, Mn, Pb, Cu and Fe. A significant variability noted by the concentration of Ni (62%) with an average of (64.96±40.32), while Sb and Cd are the least abundant in the thalli and also present an absence of variability 19.10% and 18.2% respectively. The average variation of the ten elements analyzed is 35.17%.

Table 1. Mean of the concentrations of the elements (mg/kg); median; standard deviations; max and min values found in *X. parietina* in the BBA region are expressed as the mean coefficient of variation (CV) and the certified values and standard deviations (CRM 482) by the FAAS (Quevauviller *et al.,* 1996).

Element	*Mean±SD*	*Median*	*Min-Max*	*CV(%)*	*Certified values±SD*
Cd	3.25±0.62	3	2-5	18.2	0.56±0.02
Cr	313.44±97.27	327.89	127.9-472.1	31.1	4.12±0.15
Cu	49.81±19.28	51	23-87	38.8	7.03±0.19
Co	556.48±152.59	586.09	254.3-814.8	27.4	0.32±0.03
Fe	43184.38±16373.66	42950	7000-73000	37.9	804±16
Mg	671.88±312.36	550	300-1300	46.5	33±0.5
Ni	64.96±40.32	65.77	3.3-133.4	62	2.47±0.07
Pb	98.44±39.54	96	6-206	40.1	40.9±1.4
Sb	186.75±35.73	185.5	93.00-253.00	19.1	0.35±0.09
Zn	476.63±145.91	438.08	323.90-983.52	3.6	100.6±2.2

3.2. MTE distribution maps

A cluster analysis was using, in order to map the levels of metallic pollution by the atmosphere in the urban area of BBA. This analysis revealed two groups of elements (Figure 2). In each small group, one element was chosen to represent that group. The distribution maps of concentrations measured in the lichen thalli *X. parietin*a are represented in the ten metallic trace elements (Cd, Cu, Cr, Co, Fe, Mn, Ni, Pb, Sb, and Zn).

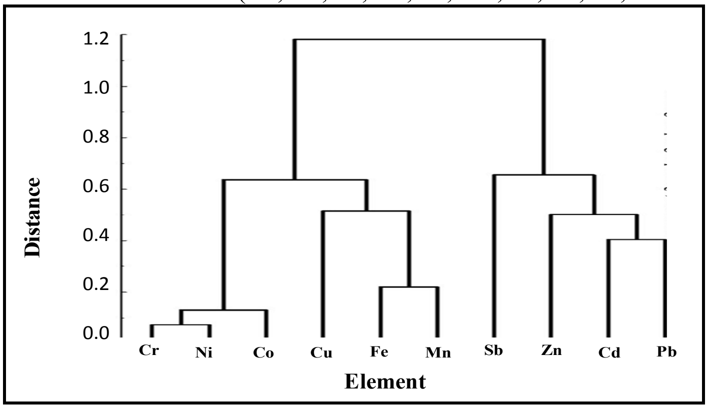

Figure 2: Classification of MTE in *X. parietina* thalli

3.2.1. Concentration of Lead (Pb)

Anthropogenic lead emissions have long been dominated by road traffic, due to the presence of Pb in gasoline (Loppi *et al.* 2004; Li *et al.* 2012 ; Dörter *et al.* 2020), but this contribution of gasoline to atmospheric Pb is decreasing due to the increased use of unleaded gasoline (Yenisoy-Karakas & Tuncel 2004). However, other industrial activities that can be major sources of this toxic element in the environment (Agnan *et al.* 2013). In the study area, the distribution of the highest Pb values are recorded in urban sites that are close to highwayssuch as Ain Taghrout (206 mg/kg), Medjana (173 mg/kg), Tixter (169 mg/kg) and Hasnaoua (136 mg/kg) sites. While, minimum concentrations are observed in rural sites away from main roads (Figure 3a). The high Pb values in the study area are mainly related to traffic, due to the use of many older cars.

3.2.2. Concentration of Cadmium (Cd)

Cd concentrations are around 3 mg/kg in most of the sites located to the south and west to the north of the BBA region. The maximum concentrations of this element are observed near major roads and industrial activities (Figure 3b). Cd is an element, used in vehicle components industry (Ouali-Alami *et al.* 2014), which confirms that the contamination in urban sites at BBA by this element is of anthropogenic origin from various vehicular and industrial emissions.

3.2.3. Concentration of Antimony (Sb)

In the literature, authors attribute the high levels of Sb mainly to road traffic, waste incineration and the production of accumulators, rubber additives and in paints (Azimi *et al.* 2005; Tian *et al.* 2015). This contamination by Sb extends to rural stations where the maximum concentration is recorded in Ouled-Dahmane (253 mg/kg), probably coming from sources located outside the borders of the study area, as reported in France by Agnan et al. (2013). It is noted that the results of the distribution map of the concentrations of Sb at BBA are similar to those of the results of the distribution maps of Pb and Cd (Figure 3c). These high levels of Sb in urban and industrial sites, so they seem to be related to road traffic emissions (Yenisoy-Karakas & Tuncel 2004).

3.2.4. Concentration of Zinc (Zn)

These high values are due to road traffic as has been suggested in the literature (Scerbo *et al.* 2002; Conti 2008; Kinalioglu *et al.* 2010), but also this element is widely used in the manufacture of metals, production of cement, steel, plastic and paints (Lin *et al.* 2002; Demiray *et al.* 2012). Another source of Zn-related pollution that can be a threat in the region is agricultural activity, through the use of fertilizers and pesticides that produce relatively high zinc concentrations (Mulgrew & Williams 2000; Pignata *et al.* 2002). The highest concentrations of Zn are observed at urban sites close to main roads, and around industrial sites (Figure 3d). There is no metallurgical industry in BBA, the production of cement and plastics can be a point source that emits pollutants in significant quantities into the atmosphere. Moderate concentrations have been found in agricultural sites far from pollution sources, these concentrations may be due to the use of fertilizers.

3.2.5. Concentration of Iron (Fe)

Iron is one of the main components of soil minerals, thus, this element is used as building materials (Nimis *et al.* 2001; Aslan *et al.* 2011; Demiray *et al.* 2012; Parzych *et al.* 2016; Francovà *et al.* 2017). The high rate of Fe in our sites is linked to geographical factors since Iron is a soil element which undergoes significant bioaccumulation in the thalli. The highest values were also distributed in the North and West of the region, these values gradually decrease towards the South and East where the lowest value was recorded (Figure 3e). The second explanation for the high rate in the thalli is due to the accumulation of dust from anthropological activities (cement industry, quarrying, brick making and gravel processing) which are frequently found in the BBA region (Branquinho *et al.* 1997; Branquinho *et al.* 2008; Aslan *et al.* 2011). The Fe values analyzed in the thalli of *X. parietina* are varied between 7000 mg/kg and 73000 mg/kg with a median of 42950 mg/kg.

3.2.6. Concentration of Manganese (Mn)

Manganese in the air is of natural origin, it is an important element derived from the soil. Thus, this element is used in the production of batteries and alloys (Markert *et al.* 1997; Yenisoy-Karakas & Tuncel 2004; Dron *et al.* 2016; Parzych *et al.* 2016). The highest Mn concentrations were distributed north and west of BBA (Figure 3f). Thus, this element is used in the production of batteries and alloys (Azimi *et al.* 2005). This may be the reason for the high concentrations of Mn found in the region especially to the north and west of BBA. The Mn values obtained from this analysis are between (300 mg/kg-1300 mg/kg) with a median of 550 mg/kg.

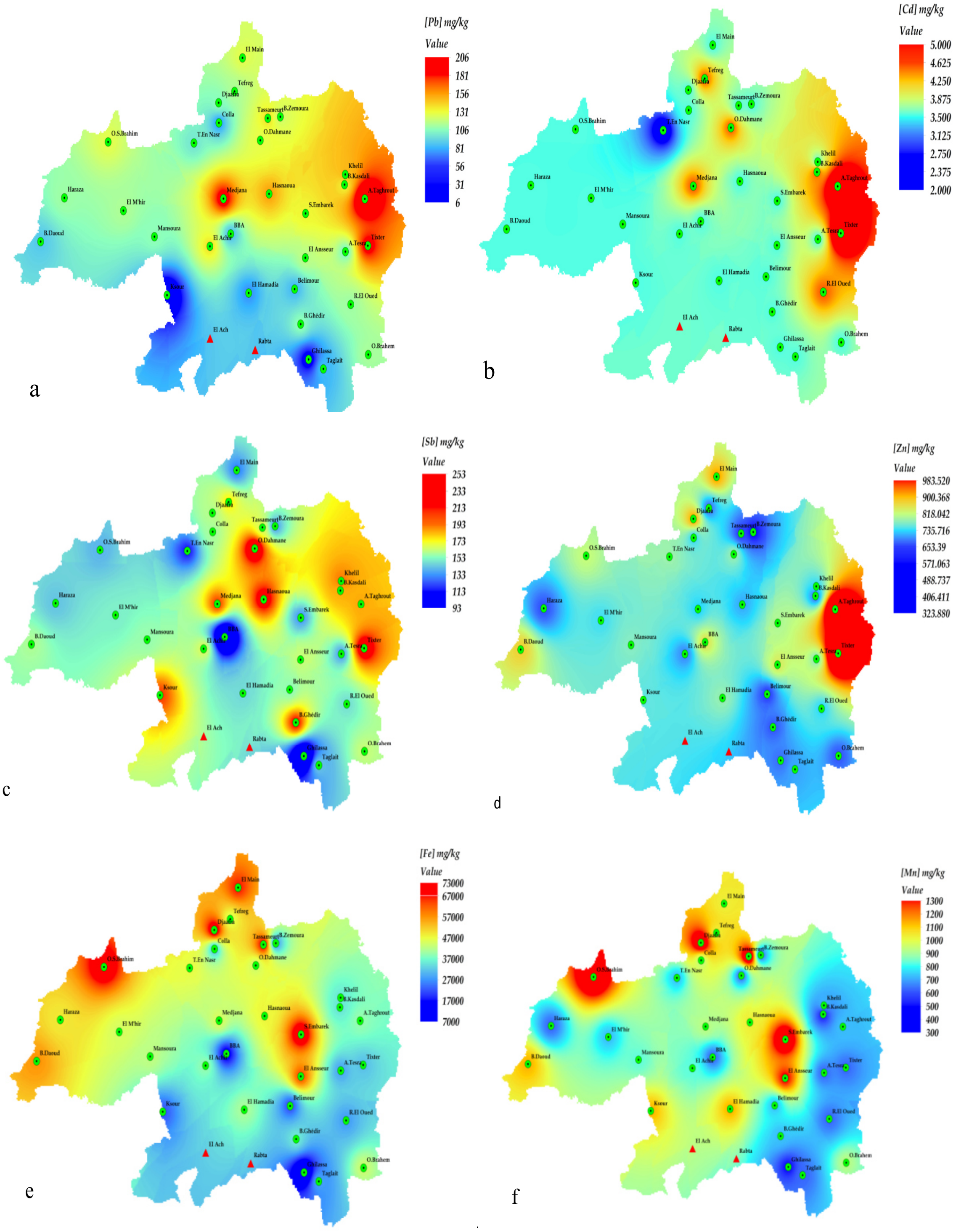

Figure 3: Distribution map of element concentrations (a) lead, (b) cadmium, (c) antimony, (d) zinc, (e) iron, (f) manganese (g) copper (h) cobalt, (i) chromium and (j) nickel (mg/kg) in the lichen *X. parietina* in the BBA region north-est of Algeria

3.2.7. Concentration of Copper (Cu)

The distribution of Cu concentrations in the region is similar to those of Fe and Mn, the low values of which are located to the south and east of the BBA region. While the highest values are distributed to the north and west, particularly in agricultural sites (Figure 3g). Several studies carried out in the world show that Cu is an element that comes not only from road traffic and the metallurgical industry but also from agricultural activities (Pignata *et al.* 2002; Tian *et al.* 2015; Parzych *et al.* 2016). This suggests that the use of fertilizers may be a source of Cu contamination in agricultural sites in the study area. We can suggest another source of Mn in the region, in particular, in the sites of El M'hir, Mansoura, El Ansseur, El Achir and Ouled Sidi-Brahim, which includes rail transport.

3.2.8. Concentration of Cobalt (Co)

The results of the Cobalt distribution map show that the high concentrations are distributed west of the BBA region (Figure 3h). Dust from quarrying and manufacturing of building materials may be the reason in the region studied (Dörter *et al.* 2020). The Co values are varied between 254.30 mg/kg and 814.80 mg/kg, the median value is 586.09 mg/kg.

3.2.9. Concentration of Chromium (Cr)

In the study region, the highest accumulation of Cr is observed in the western parts, particularly at agricultural sites (Figure 3i). The high level of concentrations of this element in agricultural sites can be partly explained by the contribution to its presence in the soil (Scerbo *et al.* 2002). The range of chromium concentrations measured in the thalli of *Xanthoria parietina* being between 127.90 mg/kg and 472.11 mg/kg with a median value of 327.89 mg/kg.

3.2.10. Concentration of Nickel (Ni)

In the study region, the highest Ni concentrations were found in the West (Figure 3j). The high rate of this element may be due to pollution transported from neighboring regions. These concentrations decrease towards the eastern stations where the lowest levels were observe at industrial and urban sites. Ni contents in lichen thalli vary between 3.72 mg/kg and 133.40 mg/kg with a median value of 65.77 mg/kg.

The distributions of the analyzed Cr, Co and Ni elements represented on the maps (Figs. 3h-3i & 3j) are similar. Nevertheless, there is one possibility that would explain the high levels of these three elements at sites in the west of the region that are characterized by agricultural and grazing activities: probably the element composition of western BBA lichens is strongly affected by dust deposition from polluted soil (Liu *et al*. 2016). In addition, topography and meteorological factors may also have an important role in the high accumulation of these elements (Szczepaniak & Biziuk 2003; Gür & Yaprak 2011; Dron *et al.* 2021).

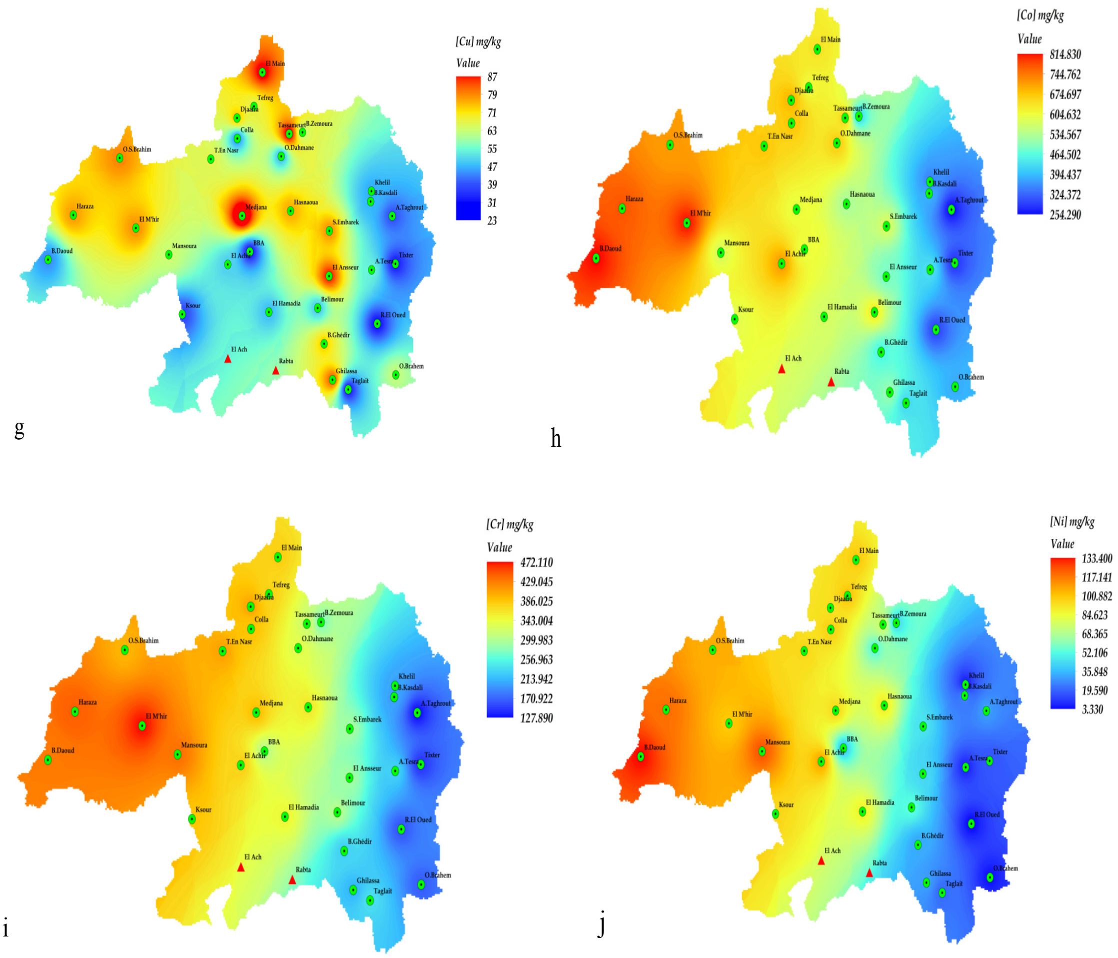

Figure 3. (Continued)

4. Conclusion

Element distribution maps were drawn to visualize the spatial distribution of accumulated MTE concentrations in *X. parietina* thalli in the sampling region at BBA, as indicative of different local sources. Road traffic emissions, dust from quarries and building materials, small industries, as well as highly urbanized sites are possible sources of metallic trace elements in the BBA region.

Traffic is a main source of Pb, Cd, Zn and Sb, the abundance of the concentrations of these elements in the studied region decreases from urban sites (high road traffic) towards agricultural sites (low road traffic). Another cause that can explain the high concentrations of Pb, Cd, and Sb in the urban stations of the region, would be due to industrial activities.

The highest contributions of elements such as Fe, Co, Mn, Cr, Cu and Zn, were observing in the northern and western parts of BBA, especially at sites relatively close to agricultural areas and quarries. For Ni and Cr are affected by the soil, but there are obviously other unknown sources around the sites located in the west of the region. The maps made in this study could be a valuable tool for decision-making processes aimed at reducing metal pollution in the study area.

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