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# Geochemical characterization and spatial distribution of heavy metals from urban dust in Chetumal, Mexico

# Caracterización geoquímica y distribución espacial de metales pesados del polvo urbano en Chetumal, México

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## Abstract

The first diagnose of heavy metal (Cd, Cr, Cu, Fe, Mn, Ni and Pb) concentrations present in Chetumal, Mexico and its spatial distribution was carried out by analyzing 86 samples of urban dust through atomic absorption spectrophotometry. The assessment of the extent of pollution was undertaken by the use of the Mexican Residential Soil Guideline Values, the calculation of the contamination factor and the pollution load index. The results showed concentrations of heavy metals below the Mexican guidelines in the city, except for chromium and lead in a few samples. However, using the contamination factor the concentrations for chromium, lead and copper are exceeded in some samples. The map of pollution load index shows the areas requiring immediate attention from the decision makers.
**Keywords:** Contamination factor, spectrophotometry, pollution load index, geostatistics, indicator Kriging.

## Resumen

El primer diagnóstico de concentraciones de metales pesados (Cd, Cr, Cu, Fe, Mn, Ni y Pb) presentes en Chetumal, México y su distribución espacial se realizó analizando 86 muestras de polvo urbano usando espectrofotometría de absorción atómica. La evaluación del alcance de la contaminación se realizó usando los Valores de la Norma Mexicana para Suelo Residencial (NMSR), el cálculo del factor de contaminación y el índice de carga contaminante. Los resultados mostraron concentraciones de metales pesados por debajo de las normas mexicanas, excepto para el cromo y plomo en unas pocas muestras. Sin embargo, usando los índices de contaminación los límites para cromo, plomo y cobre se exceden en varias muestras. El mapa del índice de carga contaminante muestra las áreas que requieren atención inmediata por parte de los tomadores de decisiones.
**Descriptores:** Factor de contaminación, espectrofotometría, índice de carga contaminante, geoestadística, indicador Kriging.

## Introduction

In recent years, concerns about pollution levels of heavy metals in urban areas have risen, due to their relationship with cases of cancer, i.e. pulmonary, bronchial, among others, which are considered some of the main death causes in the U.S (Siegel *et al*., 2012) and the world (WHO, 2013).

The variety of sampled substrates used for heavy metal (HM) pollution assessments can include soil, as a measure of historical and former contamination; living organisms such as tree leaves, earthworms, lichens, etc. used as bio-monitors; suspended particular matters ($PM_{10}$ and $PM_{2.5}$) and urban dust as a measure of recent contamination (Bautista *et al*., 2011a; Aguilera *et al*., 2018; Delgado *et al*., 2019).

Urban dust is a heterogeneous mixture composed by combustion-related emissions, brake abrasion and tire wear particles (Adachi and Tainosho, 2004; Aguilera *et al*., 2018); weathering of paint, industrial emissions (Al-Khashman, 2004 and Morales *et al*., 2014); and other elements that get suspended and transported by air drafts to settle and mix with soil (Sánchez *et al*., 2015 and Cortés *et al*., 2015). These can then become harmful to humans due to its ease of being inhaled, ingested, or absorbed through skin (Donaldson *et al*., 2001; Cakmak *et al*., 2014 and Yann *et al*., 2014).

Chetumal has no record of former study related, therefore this study has aimed to obtain the first diagnosis of heavy metal contamination (Cd, Cr, Cu, Fe, Mn, Ni and Pb) in urban dust, assessment of the quantity of dust existent through the use of Mexican Residential Soil Guideline Values (MRSGV), calculation of contamination factors (CF) per metal and the pollution load index (PLI) for the metals mentioned in the latter part.

## Methods and materials

Chetumal city is located southeast of Mexico, as the capital of Quintana Roo, it has 151,243 inhabitants (INEGI, 2010) and a number of motor vehicles close to 111,000 (INEGI, 2014). The state is mainly focused on touristic activities and very scarce industry (INEGI, 2016). The soil has generally been classified as Leptosol over limestone (Pacheco and Alonzo, 2003 and Bautista *et al*., 2011b).

Sampling in Chetumal was performed in May 2013 during the dry season, when particle mobility was considered greater. A systematic, homogenously distributed sampling was carried out with the idea of incorporating the greatest variation, as well as to be able to make a spatial analysis using geostatistical tools (Webster and Oliver, 1990; Aguilera *et al*., 2018; Delgado *et al*., 2019). Sampling covered homogeneously the urban area; 86 urban dust samples were collected. Each site was located the closest to the street intersection; logging in street names, observations, and coordinates using a GPS *Garmin GPSmap 60C*.

Sampling consisted on delimiting a 1 $m^2$ surface at each site using a cotton string, then samples were swept and collected from the pavement (Wei and Yang, 2010; Aguilar-Reyes *et al*., 2011 and Bautista *et al*., 2011a; Aguilera *et al*., 2018). Samples were weighed, tagged and packed.

The samples were dried in the shade for two weeks, then ground and sieved through a 2 mm mesh and weighed (Aguilar-Reyes *et al*., 2011). The extraction was performed digesting 0.5 g of sample and 5 mL of $HNO_3$ (CONC) in a MARS Xpress microwave CEM (USEPA, 2007a). For the heavy metal content, the atomic absorption spectrophotometry (AAS) technique (USEPA, 2007b) using a PERKIN-ELMER Analyst 700 spectrophotometer. Calibration curves were made with a standard IV multi-element Merck Millipore solution and $HNO_3$ 2% v/v. Dilutions being 100, 50, 30, 20, 15, 10 and 5 mg $L^{-1}$. The calculated limit of detection was 0.0021 mg $L^{-1}$.

Contamination indexes were calculated and used as a tool for contamination site and source location. The CF is the ratio between the heavy metal concentration and the background concentration in the area of interest (Tomlinson *et al*., 1980). Values below 1 indicate insignificant contamination *in-situ*, values from 1 to 3 show moderate contamination; from 4 to 6 considerable and >6 very high (Ihl *et al*., 2015).

The CF is calculated as follows (Tomlinson *et al*., 1980):

$$CF = \frac{Cmsample}{CmBackground} \quad (1)$$

Where *Cm Sample* represents the element concentration found in the sample and *Cm Background* is the natural metal content in the area of study. *Cm Background* values used were the lowest concentrations found in this study corresponding to sample *P1*, taking the following values: 23.8 mg $kg^{-1}$ Pb; 21.4 mg $kg^{-1}$ Cu; 13.3 mg $kg^{-1}$ Cr; 2.5 mg $kg^{-1}$ Cd; 21.5 mg $kg^{-1}$ Ni; 76.7 mg $kg^{-1}$Mn and 4240 mg $kg^{-1}$ Fe. The PLI was calculated to merge the CF per metal of each sampled site (Chandrasekaran *et al*., 2015). The PLI is defined as "the $n^{th}$ root of the product of the CF" (Bhuiyan *et al*., 2010):

$$PLI = \sqrt[n]{CF1 * CF2 * CF3 * ... * CFn} \quad (2)$$

A PLI equal to 0 indicates excellent quality, a value of 1 the presence of baseline concentrations of heavy metals, and above 1 means progressive degeneration of the quality (Tomlinson *et al*., 1980).

## Results and discussion

The highest dust quantities were found in the central area of the city. The weighed dust in samples averaged 270±100 g $m^{-2}$, going from 80 up to 820 g $m^{-2}$. Published papers reporting dust quantities in Mexico are scarce, thus no comparison with other studies is possible. Regarding the exposition, chronic exposition to urban dust is as harmful as it is the exposition to very high heavy metal concentrations in dust (Cakmak *et al*., 2014).

In this regard, this study is one of the first reporting concentrations of heavy metal per area unit, among these: Pb (67.9±99.7 mg $m^{-2}$), Cr (16.5±16.9 mg $m^{-2}$), and Fe (3306±2889 mg $m^{-2}$) and low quantities of Cu (28.1±61.5 mg $m^{-2}$) and Mn (31.5±21.5 mg $m^{-2}$) (Table 1). Particularly, Cr and Pb presented the highest per area unit in four samples (*P7, P8, P12* and *P13*) located on the southeast part of Chetumal. As expected, the zone with governmental buildings (administrative downtown) contains the greatest quantity of dust, due to the presence of high-density traffic roads heading downtown. It is worth mentioning that the calculated means show a high variability, this may be due to the different HM sources.

The concentrations of Cr, Cu, Pb and Fe are recognized as being of anthropic origin, that is to say contaminants, because they presented high values of standard deviation and are typically of non-Gaussian distribution. In contrast, heavy metal concentrations of natural origin, as Cd and Mn, have small standard deviations and a Gaussian distribution (Aguilera *et al*., 2018). In the case of Ni, some samples with high concentrations were found, which reveals contamination in some places (Table 1).

Table 1. Descriptive statistics of concentration, amount, CF of heavy metals in urban dust. Variance (Var), standard deviation (SD), minimum (Min), maximum (Max), skewness (Sk), Kurtosis (Kt), contamination factor (CF) and Mexican legislation residential MRSGV (NOM)

| Heavy metals | | Mean | Median | Mode | Var | SD | Min | Max | Sk | Kt |
|---|---|---|---|---|---|---|---|---|---|---|
| Cd | mg $kg^{-1}$ | 3.8 | 3.6 | 3.5 | 0.7 | 0.9 | 1 | 9 | 2.6 | 14.8 |
| | mg $m^{-2}$ | 1 | 11.5 | - | 0.4 | 0.6 | 0.3 | 3 | 3.4 | 15.9 |
| | CF | 1 | 1 | 1 | 0.3 | 0.5 | 0 | 3 | 0.9 | 0.4 |
| | NOM | 37 | - | - | - | - | - | - | - | - |
| Cr | mg $kg^{-1}$ | 65 | 52.2 | - | 7199 | 84.9 | 8 | 728 | 6.4 | 46.8 |
| | mg $m^{-2}$ | 16.5 | 8.4 | - | 286 | 16.9 | 2.5 | 117 | 1.6 | 3.8 |
| | CF | 5 | 4 | 3 | 41 | 6.4 | 1 | 55 | 6.4 | 46.6 |
| | NOM | 280 | - | - | - | - | - | - | - | - |
| Cu | mg $kg^{-1}$ | 96 | 61.7 | 107 | 26223 | 161.9 | 7.4 | 1440 | 7 | 57.1 |
| | mg $m^{-2}$ | 28 | 14.3 | - | 3782 | 61. 5 | 1.37 | 539 | 7.1 | 57.6 |
| | CF | 5 | 3 | 3 | 57 | 7.5 | 0 | 67 | 7 | 56.8 |
| Fe | mg $kg^{-1}$ | 11758 | 10360 | - | 3.4E+07 | 5834.5 | 130 | 27750 | 0.9 | 0.6 |
| | mg $m^{-2}$ | 3307 | 37.7 | - | 8346321 | 2889 | 21 | 19820 | 4.2 | 19.3 |
| | CF | 3 | 2 | 2 | 2 | 1.4 | 0 | 7 | 0.9 | 0.5 |
| Mn | mg $kg^{-1}$ | 114.7 | 113.2 | 125.2 | 1466 | 38.3 | 0.75 | 229 | 0.2 | 0.9 |
| | mg $m^{-2}$ | 31.5 | 24.6 | - | 462 | 21.5 | 0.12 | 134 | 2 | 5.6 |
| | CF | 2 | 1 | 1 | 0.3 | 0.6 | 0 | 3 | 0.4 | -0.4 |
| Ni | mg $kg^{-1}$ | 37 | 35.3 | 33.9 | 91 | 9.5 | 19 | 106 | 4.6 | 32.2 |
| | mg $m^{-2}$ | 10 | 2427.6 | - | 29 | 5.4 | 2.9 | 34 | 2.8 | 11.9 |
| | CF | 2 | 2 | 2 | 0.2 | 0.5 | 1 | 5 | 2.6 | 21.9 |
| | NOM | 1600 | - | - | - | - | - | - | - | - |
| Pb | mg $kg^{-1}$ | 257 | 158.6 | - | 157418 | 396.8 | 23.8 | 3396 | 6.2 | 47 |
| | mg $m^{-2}$ | 68 | 0.8 | - | 9940 | 99.7 | 5.8 | 636 | 1.5 | 2.5 |
| | CF | 11 | 7 | 5 | 279 | 16.7 | 1 | 143 | 6.3 | 86 |
| | NOM | 400 | - | - | - | - | - | - | - | - |

Notably, the average values of HM found in Chetumal do not exceed the MRSGV established by the NOM 147 for residential soils (Table 1); however, for samples P23 and P54, the limits of Pb (400 mg $kg^{-1}$) were exceeded. These same samples show very high concentrations of total Cr, although the Mexican regulation only takes into account Cr (VI) concentrations in the MRSGV.

The Mexican regulations need to broaden the number of elements given in soil guideline values. Moreover, it is needed to establish in the legislation guideline values suitable for the heavy metal assessment in urban dust considering background values.

Cd and Mn were the heavy metals with the lowest contamination factor values, maps show areas with values close to 3 (Figure 1). Fe and Ni reached values greater than 6 of the contamination factor, these are considered high (Figure 2).

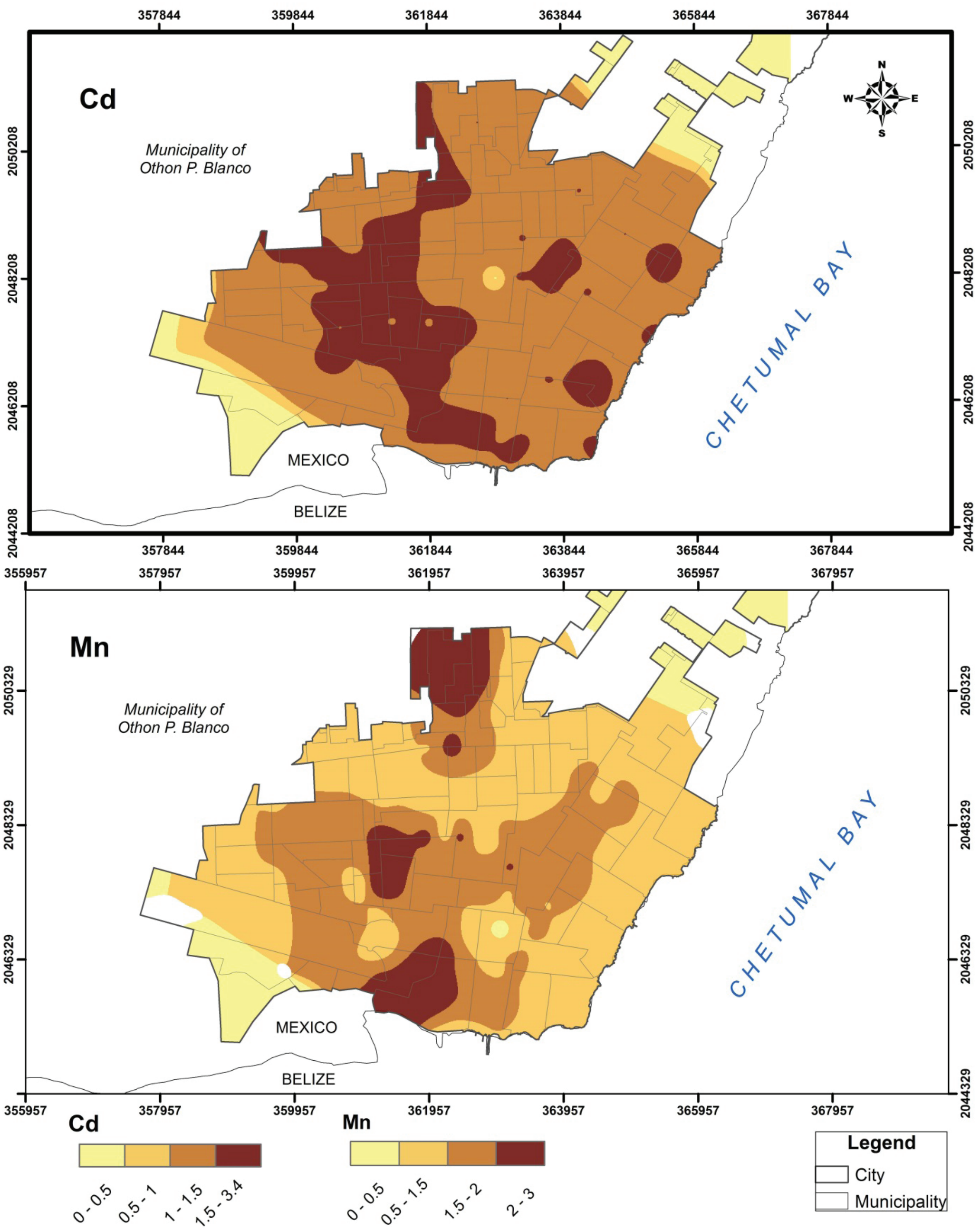


Figure 1. Maps of contamination factor of Cadmium and Manganese

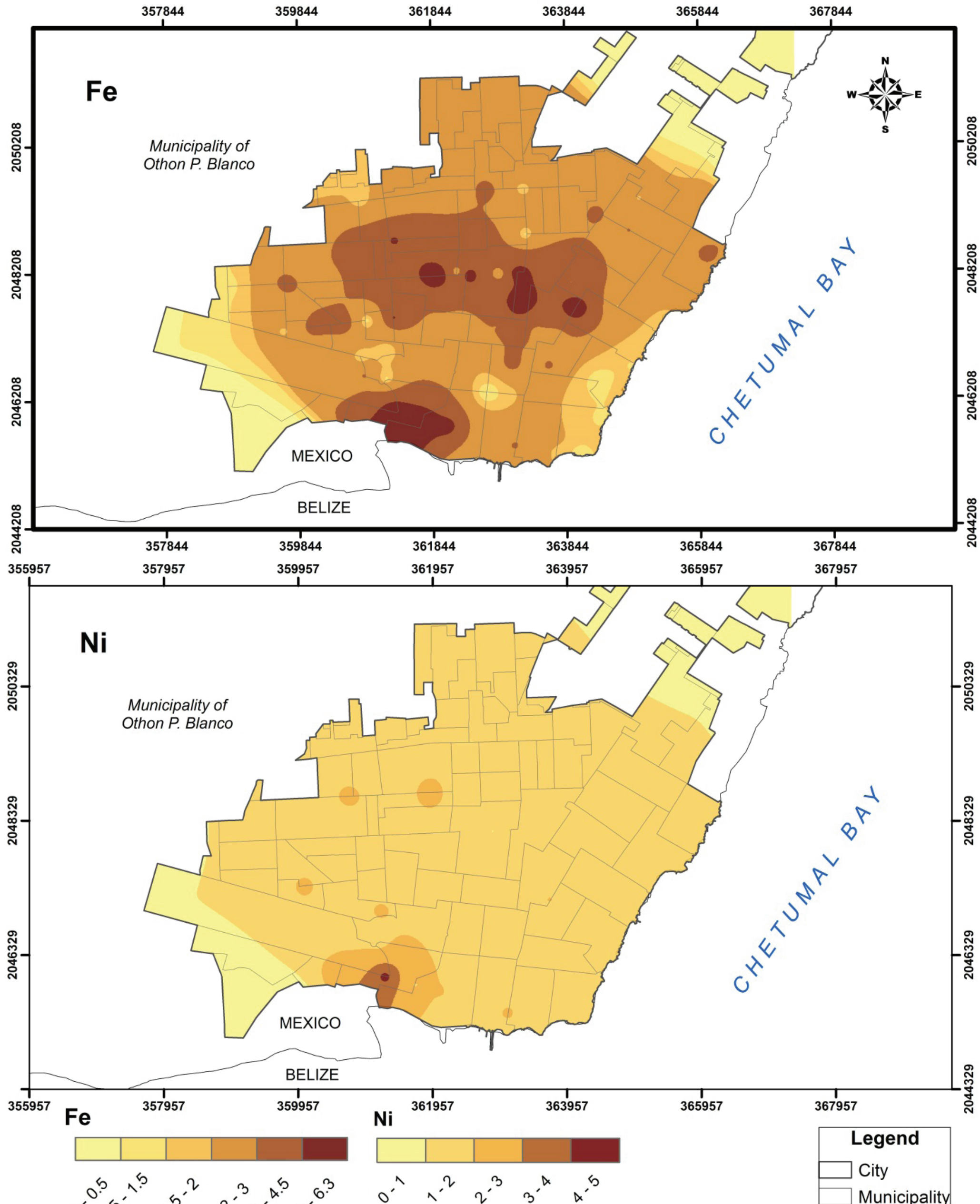


Figure 2. Maps of contamination factor of Iron and Nickel

The analysis of contamination factor in metals such as Cr, Cu and Pb showed very high contamination levels with values up to 55, 67 and 143, respectively (Figures 3 and 4). For the case of Cu, 11 samples showed very high contamination, whereas Cr showed values of very high contamination in nine samples with the highest value in sample (*P23*). Finally, for Pb 51% (44 samples) showed CF>6, hence the city is considered as highly contaminated by Pb. For the case of Fe, only one sample (*P82*) showed a CF=7.

The average calculated PLI for the city was 3±0.9, showing that the overall sampled sites contain two-fold the background concentration of heavy metals of the area. Sample *P54* expresses a value equal to 6.5 on the southeast of the city; this is the sample with the highest values in Cr and Pb (Figure 3). The probabilistic PLI map

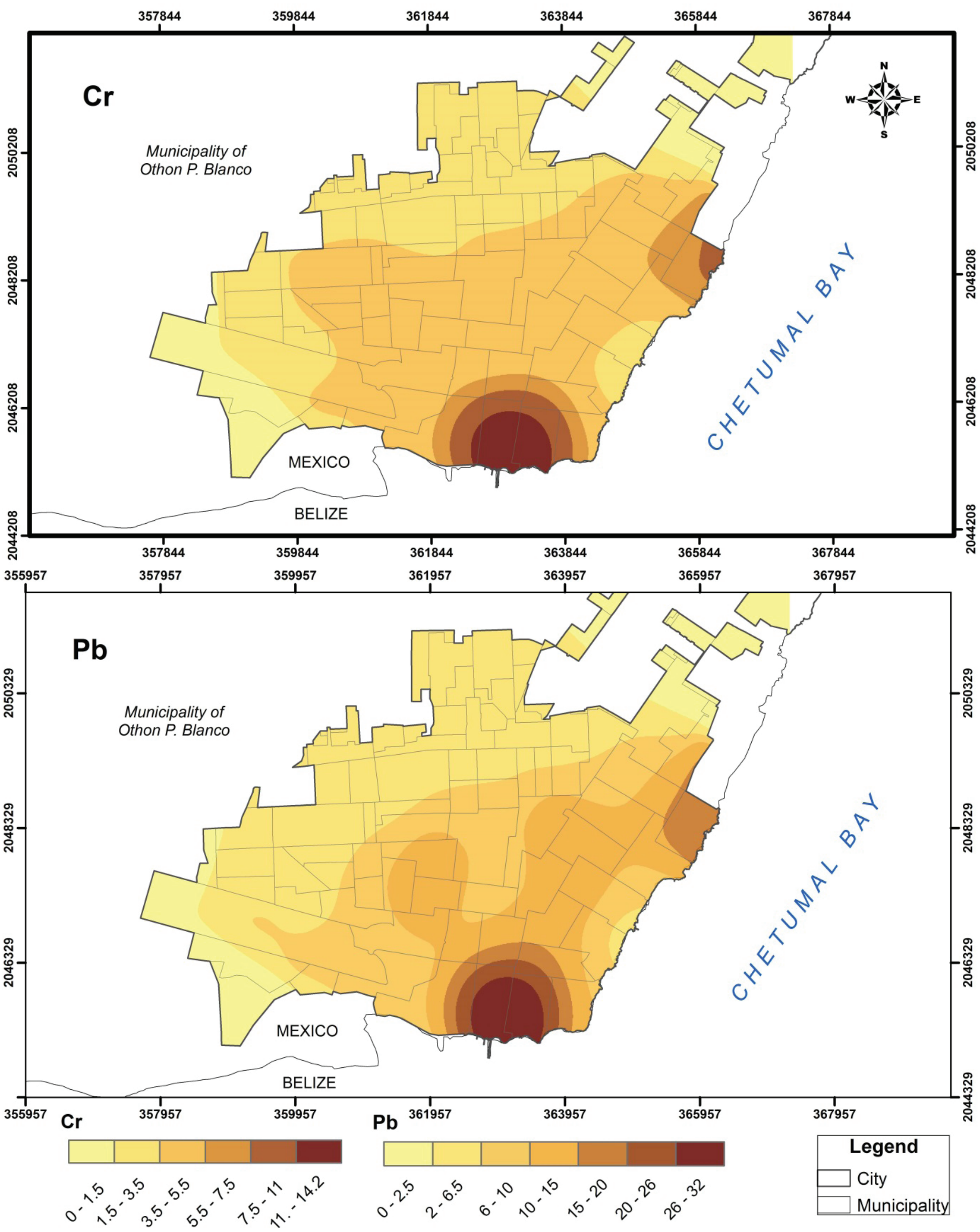


Figure 3. Maps of contamination factor of Chromium and Lead

indicates that values greater than 0.8 are areas that exceed a value of 3 and are those that require immediate attention (Figure 4). If calculating the PLI can provide an idea of the pollution extent in an area (Tomlinson *et al*., 1980), it can also demonstrate the zones that require urgent attention. In this study it is shown that in the port terminal the higher PLI values were reached, so it is suggested that part of the contamination of the boats is in the urban dust, as was also reported by Cortés *et al*. (2015) for the case of the city of Ensenada. In conclusion to having found PLI values up to 3, it is very likely to find human health consequences in middle and long-term.

As mentioned by Padoan *et al* (2017) urban dust may be originated from either natural or anthropogenic activities. The former referring to natural weathering

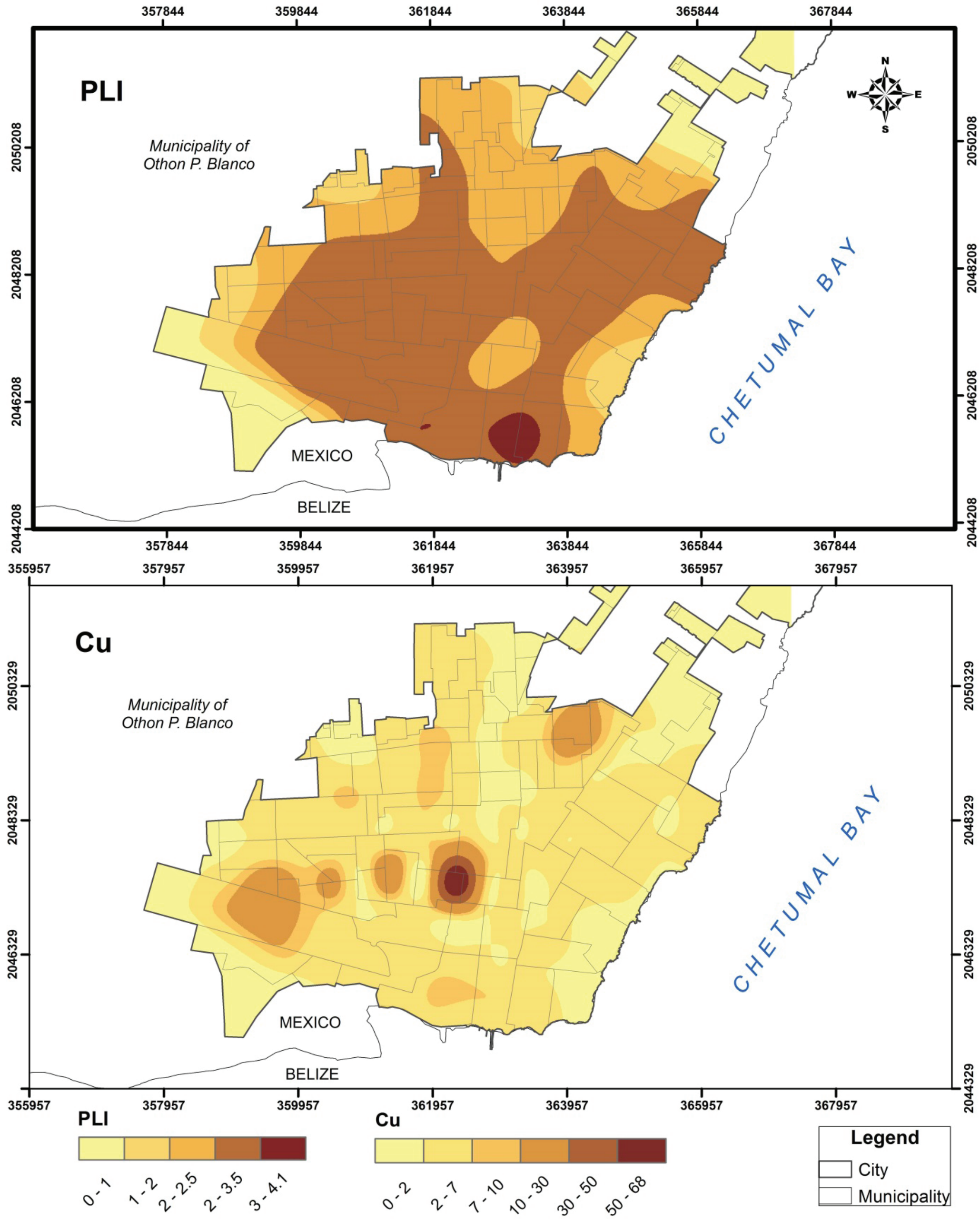


Figure 4. Maps of pollution load index and contamination factor of Cupper

and the latter to wear and tear, caused by vehicle use, or inner combustion vehicles (Adachi and Tainosho, 2004).

Elements such as Fe may have been introduced by cargo trucks coming from rural areas rich in Vertisols and Luvisols (Kabata, 2011). Morever, for the cases of Pb and Cr, an often seen cause may be the weathering of leaded yellow road paint, which was observed in some samples. Some concentrations in Chetumal are comparable to those seen in streets of Plymouth, UK by Turner and Lewis (2018). Additionally, the deposition of atmospheric particles produced by car exhausts may also contribute to these concentrations. Finally, the origins of Cu have been tracked to brake dust (Adachi and Tainosho, 2004).

Since the zone with more abundant urban dust was located in the center of the city, two hypothesis can be proposed: the dust production is greater in this area or the high-density traffic in this area creates air drafts that accumulate the urban dust in only one site. This is debatable with studies of the air dynamics. Finally, for prevention it is recommended for the decision makers to plan strategically the urban transport routes, as well as the establishment of an urban dust-quality monitoring plan that will include some government guideline values. For mitigation, the implementation of buffer areas rich in clays might aid in the sequestration of cations contained in urban dust.

In agreement with Cortes *et al.* (2016), if we consider that "*environmental pollution is the presence in the environment of any agent in places, forms and concentrations that can be harmful to the health, safety or well-being of the population*", we can say that in this study we identified harmful agents, concentrations, amounts and polluted places, however, in future studies will be necessary the identification and evaluation of chemical forms of heavy metals, using techniques of sequential extraction and toxicity in the population (Bautista, 1999; Covelo *et al*., 2007).

## Conclusions

Based on the residential soil guideline values proposed by the Mexican government, it can be concluded that there are signs of isolated sources of contamination in urban dust only; however, this conclusion is not completely representative given the lack of guideline values for urban dust. Also, considering the CF calculated, Chetumal has contamination problems with copper, chromium and lead in the south is the most polluted area. Preventive actions to counteract the air draft dynamics are strongly advised to avoid the increase in HM concentrations and mitigation actions are also advised to reduce the already high HM concentrations.

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