Ecotoxicological Risk Assessment Of Heavy Metals From Leachate Of The Wild Landfill Of El Hajeb City (Morocco)

Abderrahmane Gamar, Zakaria Khiya, Touria Zair, Mohammed El Kabriti, Abdelaziz Bouhlal, Fatima El Hilali

Abstract: The presence of heavy metals in landfill leachates poses a risk to the environment, especially in the unpaved dumps where their dissipation in groundwater might occur. This study is focused on the assessment of heavy metal load in landfill leachate of El Hajeb (Morocco) and their effects on the environment in general. To do this, raw leachate samples were collected monthly from basins available at the heart of the landfill and analyzed using the atomic absorption spectrophotometer (AAS) to quantify the heavy metals during the period which extends of May, 2015 to February, 2017. The results showed the presence of Pb (2.68 mg/L), Co (0.92 mg/L), Hg (0.009 mg/L), Cr (5.14 mg/L), As (1.95 mg/L), Cd (0.41mg/L) and Mn (61.52 mg/L) in significant amounts. What makes these extremely toxic effluents and thus presented a potential danger for the health of the riparian population and the adjacent groundwater. The determination of heavy metals content will thus make it possible to orient the choice of the leachate treatment process to answer the strictest standards of rejection.

Index Terms: Leachate; Heavy metals; Discharge; Contamination; El Hajeb (Morocco).

1. Introduction

Progress in urban life has generated a worrying accumulation of household waste that must eventually be landfilled [1]. The major and inevitable consequence of such a landfilling is the production of a leachate that can give rise to serious ecological problems. Indeed, where the site is not equipped with a containment system, leachates rich in organic and inorganic materials, but also heavy metals can contaminate underground and surface aquatic systems. Heavy metals once released into the environment can remain in waterways for decades or even centuries, with concentrations that are high enough to pose a health risk. The solubility and mobility of metals may increase in the presence of natural and synthetic complexing ligands and humic substances. Reduction/oxidation and adsorption/desorption reactions are the major mechanisms that dictate the mobilization of arsenic into the environment. The inorganic forms of arsenic are arsenate [As (V)] and arsenite [As (III)], which are the most important redox states of arsenic in nature and the most common toxic forms [2].The solubility of the As and Cr metals can also increase because of the reducing condition of the leachate which changes the ionic state of the metals (i.e., As (V) → As (III) and Cr (VI) → Cr (III)) [3].

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Manganese exists in the aquatic environment in two main forms: Mn (II) and Mn (IV). Movement between these two forms occurs via oxidation and reduction reactions that may be abiotic or Micro-biologically mediated. Mn (II) dominates at lower pH. Although aluminum production waste are not classified as hazardous. According to Petrovic and Thomas [4], when the buried aluminum comes into contact with alkaline water pH = 9 that enters from a variety of sources, such as alkaline incinerator ash, it results from it undesirable chemical reactions which can occur at any moments. Several methods are used to clean up the environment from these kinds of contaminants, but most of them are costly and difficult to get optimum results. Some heavy metals are at the origin of serious problems of public health. Indeed, according to Environment Canada and Health Canada [5], arsenic and its compounds are considered a priority and carcinogenic pollutant, while cobalt and its components are classified by the European Union as carcinogenic pollutants. The mercury can circulate between the large compartments of the environment (water/air/soil) by migrating into its ionic (and more rarely metallic) forms and being transported by living organisms (bioturbation). Mercury (Hg) is a global pollutant that affects fish, wildlife, and human health [6]. The recent work [7] revealed that the leachates of this discharge are in a young, acidogenic and reducing state (low pH values about 6.65, a high organic loading of COD of the order of 23597.5 mgO2/L and an dissolved oxygen quantity scarcely exceeding 1 mgO2/L. This tends to increase the solubility, adsorption and modification of the ionic state of heavy metals. Environmental risk assessment of heavy metals and other chemicals often uses the Risk Quotient (RQ) method to characterize risk quantitatively. For each ecological receptor, a RQ is a ratio of exposure to effect and calculated by dividing the estimated or actual environmental concentration by the appropriate toxicity end-point. The RQ can be used by risk analysts and decision makers to assess whether the value exceeds any predetermined threshold levels of concern for the purpose of making comparisons of environmental risk among and identify the need for considering a regulatory action. Consequently, the ecological relevance of estimated risk quotient is one of the main items in recent ecotoxicological research. The RQ (risk quotient) is calculated, if a value less than 1, then there is an acceptable risk. However, if the RQ is greater than 1, there is an unacceptable level of risk and measures to reduce contamination should be taken with urgency. The aim of this study was to assess quantitatively the potential hazards of some heavy metals (Pb, Cu, Cd, Cr, As, Co, Ni, Cr, Al, Mn and Hg) in leachates from the wild dump of El Hajeb city (Morocco) and to provide, in the case of diffusion of these leachates to adjacent groundwater, their eco-toxicological risks through measuring toxicity end-point and estimating RQs in relationship with level of concern. This will thus constitute a contribution to the program of continuous monitoring of groundwater sources around the study area.

2. Materials and methods

2.1. Presentation of the study site

The city of El Hajeb is in 30 km in the South of Meknes, the nearest town, and in 60 km in the southwest of Fes and its total population is of 36.491 inhabitants. Its climate is temperate. The average pluviometry is rather important, approximately 650 mm a year. The altitude from the sea level is 1054 m. Lithology of the region, reveals that it is a carbonated, fractured, karstified environment and favorable to the development of a high permeability (from $3.7.10^{-3}$ to $5.4.10^{-2}$ cm/s). The subsoil of the study area contains, at a depth of 5 m, the underground water. The limestone covers this water table. It is characterized by intensive agricultural development where all the land is cultivated. The majority of crops oblige farmers to resort to irrigation. The municipal dump, which serves this city, is located to the west and about 12 km from this town (Fig. 1). Its geographical coordinates in degrees are: latitude 33°39'55.7 "North; Longitude 5°27'29.1 "West. The average amount of waste produced is estimated at 11,000 tons per year, for a daily ratio of about 1 kg/j/capita. The main constituents of the municipal waste dump of the town of El Hajeb is household waste with a 90% share (Table 1). Their examination showed that they contain a high level of organic matter of about 78 % (Table 2).

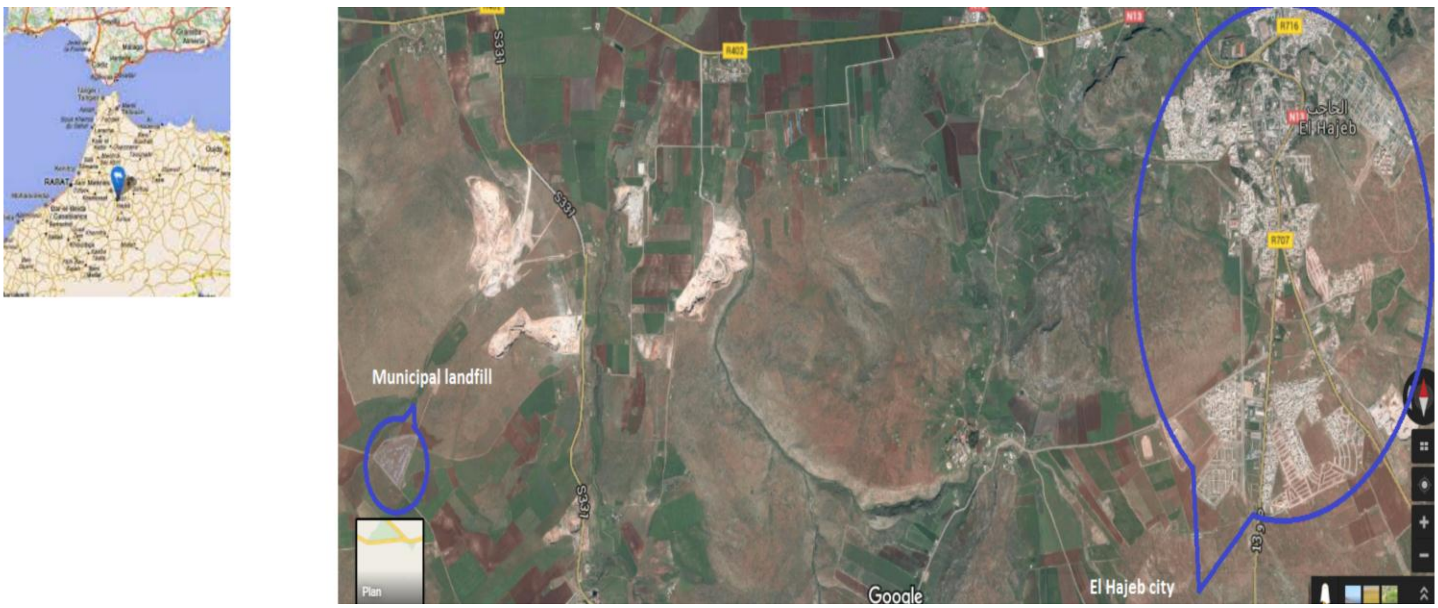

Fig. 1 *The wild landfill of El Hajeb city (Morocco).*

TABLE 1 *LANDFILL WASTE AND THEIR PERCENTAGES*

Waste type	Rate (%)
Slaughterhouse waste	1.55
Green waste	1.1
Waste from markets	2.2
Industrial waste	0.55
Hospitals waste	3.1
Household waste	91.5

TABLE 2 *CONSTITUENTS OF HOUSEHOLD GARBAGE*

Types of household garbage	Quantity (%)
Organic mater	78.1
Paper and Cardboard	5.8
Glass-debris from ceramics	0.8
Fabrics	4.1
plastics	6.3
Metals	0.3
Wood	3.1
Various	1.5

2.2. Sampling and analysis

2.2.1. Sampling mode

The leachate samples were collected from the municipal dump in El Hajeb city at monthly intervals during the period from May 2015 to April 2017 and were stored in 5-L polyethylene sterilized and opaque bottles pre-washed and conserved with nitric acid. All contained samples were kept in a dark ice cooling tank at 4 ° C to prevent chemical absorption and ionic interference before analyzes of heavy metals and transported to the lab. A total of 336 samples were collected.

2.2.2. Analysis method

We have dosed, into the leachate, the following heavy metals: Ni, Hg, Co, Cr, Zn, Pb, Mn, Al, Cd, Cu and As which are considered to be of greatest concern. The concentrations of heavy metals were determined using the Inductive Coupled Plasma Technique (Optima 2000DV, Perkin-Elmer, USA). Arsenic (As) was analyzed using Atomic Absorption technique with hydride- generation system (Shimadzu AA-6300, Japan). Hg analysis was performed using an AAS in cold steam.

2.3. Statistical and comparative analysis

Descriptive statistical analysis presented below was performed using the IBM SPSS Amos 21.0 software. The results of heavy metals analysis were presented in the form of mean ± SE. The level of statistical significance was estimated at the significance threshold Alpha = 0.05 (Shapiro-Wilk statistical test). At this threshold, most statistical results are satisfactory and significant. Environmental risk comparative analysis was expressed using risk quotient (RQ) calculated as the ratio between the determined concentration and that of the available standard. Seven standards concerning the landfill leachates and the direct and indirect rejections in underground and surface waters have been selected to calculate the RQ: Moroccan norms [8], European Union legislations [9], Quebec regulations [10], World Health Organization guidelines [11], U.S Government Environmental Protection Agency guidelines [12] and Food and Agricultural Organization standards [13].

3. RESULTS AND DISCUSSION

3.1. Trace metals

In this study, the order of heavy metals concentration were Mn > Zn> Ni> Cr >Al> Pb> Cu> As> Co > Cd > Hg from highest concentration to lowest concentration (Table 3).

TABLE 3

RESULTS AND DESCRIPTIVE STATISTICAL ANALYSIS OF THE HEAVY METAL CONTENTS OF THE LEACHATES FROM THE DISCHARGE

Heavy metals	Leachate samples (n = 336)								
	Unit	Mean ± SE	Min	Max	CV	CS	CK	*p-Value*	MN
Cd	$mg.L^{-1}$	0.41 ± 0.008	0.2000	0,6100	0.39	-0.12	-1.57	< .0001	0.2
As	$mg.L^{-1}$	1.97 ± 0.06	0.0000	2.7900	0.54	-1.06	-0.72	< .0001	0.05
Cr	$mg.L^{-1}$	5.14 ± 0.22	0.6900	11.1400	0.79	0.39	-1.38	< .0001	0.5
Co	$mg.L^{-1}$	0.92 ± 0.03	0.0200	1.7300	0.67	-0.22	-1.12	< .0001	0.1
Cu	$mg.L^{-1}$	2.52 ± 0.04	1.3100	3.4800	0.32	-0.40	-1.22	< .0001	3
Ni	$mg.L^{-1}$	6.36 ± 0.13	3.0200	9.5700	0.38	-0.06	-1.42	< .0001	5
Pb	$mg.L^{-1}$	2,68 ± 0.06	1.3600	4.3700	0.45	0.27	-1.57	< .0001	1
Zn	$mg.L^{-1}$	6.96 ± 0.13	3.8300	10.4100	0.35	0.15	-1.32	< .0001	5
Hg	$mg.L^{-1}$	0.009 ± 0.0001	0.0070	0.0120	0.21	-0.27	-1.43	< .0001	5e-4
Al	$mg.L^{-1}$	5.05 ± 0.15	1.2300	8.4100	0.56	-0.15	-1.60	< .0001	10
Mn	$mg.L^{-1}$	61.52 ± 0.073	31.8200	86.7100	0.34	-0.21	-1.51	< .0001	1

Min: Minimal value; Max: Maximal value; Mean: Average value; SE: Standard Error; CV: Coefficient of Variation; CS: Coefficient of Skewness; CK: Coefficient of Kurtosis MN: Moroccan Norms [8].

Pb, As and Cr

The statistical evolution of the measured values for all sampling points is demonstrated by the Tukey-box representation described in figure 1-A. The limits of Pb stipulated by the norm were exceeded for all the samples (Table 3), indeed the concentration of Pb in the leachate studied ranged from 1.36 to 4.37 mg/L and exceeded the national regulations, set at 1 mg/L. The average recorded of the order of 2.68 ± 0.06 mg/L was very close to the calculated median value (Fig. 2-A), indicated elimination the disposal of Pb batteries, chemicals for photograph processing, Pb-based paints and pipes. For As, given the reducing state of the leachate under study, the As (III) was assumed to be the dominant specie. The Table 3 indicated an average concentration in As of 1.97 ± 0.06 mg/L almost 40 times higher than the norm set at 0.05 mg/L. Precisely 331 samples (98% of samples) exceeded this limit. The median calculated at 2.5 mg/L greatly surpassed the mean (Fig. 2-B). High values were may come from dyeing, paints, insecticides and rat poison. Arsenic is used as an essential component of animal feed, herbicides and pesticides, lead batteries, and can also come from hospital waste. Concerning the Cr, based on the chemistry of chrome speciation, the Cr (III) was assumed to be the dominant specie. It showed concentrations varied between 0.69 and 11.14 mg/L with a range of 10.45 mg/L (Fig. 2-C). Precisely 25% of the samples exceeded at least 16 times the limits set by the norm set at 0.5 mg/L (Fig. 2-C) and the average of 5.14 ± 0.22 mg/L in Cr overstepped 10 times this same norm (Table 3). Trace concentration of Cr can be associated with dumped materials containing Cr such as stainless steel, paint pigments, and wood preservatives.

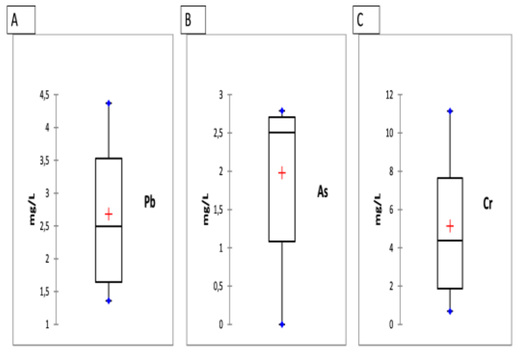

Fig. 2 *Diagrams in Tukey boxes of Pb (A), As (B) and Cr (C) concentrations*

Cd, Ni and Zn

The Cd limits set by the national standard at 0.2 mg/L were outreached by 52% of the sampling (Fig. 3-A). The found average, of the order of 0.41 ± 0.008 mg/L, exceeded twice the norm (Tableau 3). The presence of Cd is attributed to 37% of plastics (mainly PVC) where it is used as an additive and/or sometimes as a colorant and similar wastes such as paints, pigments, plastics, battery effluents, scrap metal, old slum boxes. Another heavy metal, the Ni was examined. The course of the values measured, were showed in Fig. 3-B. The Ni concentration ranged from 3.02 to 9.51mg/L with an average of 6.36 ± 0.13 mg/L, concentration almost equal to its calculated median concentration. The limits of Ni, set by the norm, set at 5 mg/L, were beyond existing by about 75% of the sample that is precisely 248 samples (Fig. 2-B). It may come from special wastes such as batteries, paint pigments, stabilizers or rubbers. In average, the Zn content was 6.96 ± 0.13 mg/L; such value slightly exceeded the norm set at 5 mg/L and the median calculated at 6.80 mg/L. The limits of Zn, established by the norm, were surpassed by more 75% of the number of sampling precisely 256 samples (Fig. 3-C). Zinc is also among the most leachable elements in fresh waste. It can come of batteries and other electric appliances, paints and pigments, alloys and solders, biocides (herbicides, pesticides, conservative) and chemical fertilizers.

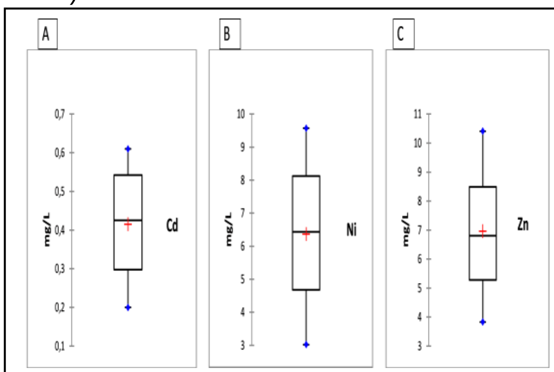

Fig. 3 *Diagrams in Tukey boxes of Cd (A), Ni (B) and Zn (C) concentrations*

Cu, Co and Hg

Concentration levels of Cu fluctuated between 1.31 and 3.48 mg/L (Table 3). Although about 25% of the samples showed concentrations higher than the national standard fixed at 3 mg/L, overall the average value of 2.52 ±0.04 mg/L in Cu did not overshoot this regulation. The average detected, slightly lower than the median calculated at 2.66 mg/L (Fig. 4-A), indicated the presence of fresh waste containing printing inks or paints. Co contents varied from 0.02 to 1.73 mg/L (Table 3) with an average of 0.92 ± 0.02 mg/L, such value was on the one hand considerably higher than the norm 0.1 mg/L, set by the regulation (Table 3) and on the other hand slightly lower than the median calculated at 0.97 mg/L, with a range of 1.71 mg/L and an interquartile range of 0.67 mg/L. About 95% of the samples, exceeded the limit cited (Fig. 4-B). These high concentrations of Co can be attributed to the rejections of paints, varnishes, agricultural fertilizers and animal feed additives. Another heavy metal controlled was Hg. The statistical course of the measured values for all sampling points is illustrated in figure 4-C. This metal showed the smallest range of the order of 0.005 mg/L and interquartile range of 0.0028 mg/L, given by its Tukey box diagram. This explained the very low dispersion of the results. The average concentration in Hg was 0.009 ± 0.0001 mg/L. The limits of Hg stipulated by the norm at 0.0005 mg/L were surpassed 18 times by this average. The concentration found in Hg indicated that batteries, lamps, fluorescent tubes ..., measuring instruments in laboratories (thermometers, barometers, densimeters ...), dental amalgams, cinnabar (pigment known since antiquity), plastics, paper and wax were probably present in the landfill.

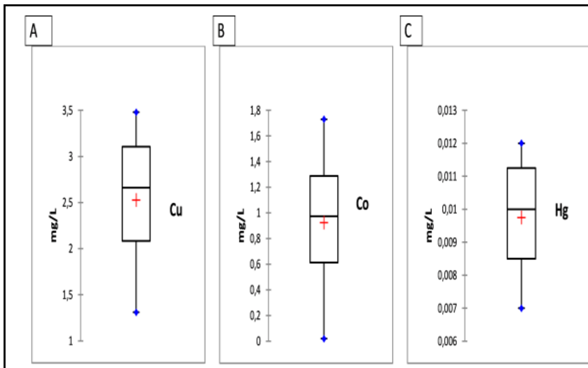

Fig. 4 *Diagrams in Tukey boxes of Cu (A), Co (B) and Hg (C) concentrations*

Mn and Al

The Mn concentrations recorded in the study leachate varied between 31.82 and 86.71 mg/L, the average found 61.52 ± 0.073 mg/L exceeded the norm set at 1 mg/L. All samples (336 samples) outstripped this limit, whereas the calculated median of 63.79 mg/L was slightly above the mean (Fig. 5-A). These high values reflected the high amount of dissolved manganese in anaerobic environment (leachate under study) in which the particulate manganese oxides have been reduced. The major anthropogenic sources of manganese group in landfill come from the manganese compounds used in dry-cell batteries and as fertilizers, as a livestock supplement in animal feed, for seed treatment of small grains such as wheat and as a preservative for fresh flowers and fruits.

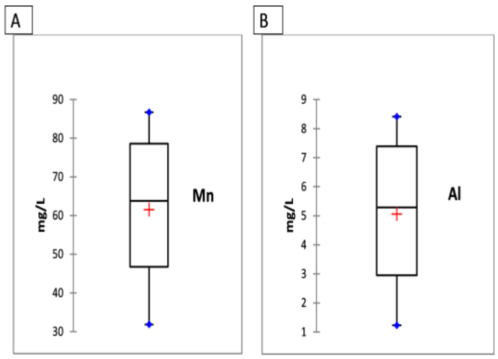

Fig. 5 *Diagrams in Tukey boxes of Mn (A) and Al (B) concentrations*

The last heavy metal examined was Al (Table 1). Al contents varied between 1,23 and 8.41 mg/L, although Aluminum does not pose a risk to human health, the mean value 5.05 ± 0.15 mg/L of Al during the study period did not exceeded the regulations set at 10 mg/L. All the samples (336 samples) did not exceed the limit mentioned, while the calculated median of 5.29 mg/L slightly exceeded the mean (Fig. 5-B). The major sources of aluminum are domestic waste.

3.2. Characterization of the eco-toxicological risk of heavy metals: risk quotient (RQ)

For the present study most of the standards indicated values of RQ strongly > 1 for all metals (Fig. 6), with the exception of Hg, whose norms for its existence are very limited, this reflected, according to these standards, alarmingly, the major risk of contaminating that represented the majority of the metals of the discharge towards the environment and precisely on the human health in the event of leachate dissipation to the groundwater main source of drinking water and irrigation, therefore, measures to reduce contamination should be taken urgently. Indeed several compounds of lead, cadmium, cobalt and inorganic arsenical compounds have a carcinogenic potential, recognized for humans [6]. While Al, in addition to its probable link with Alzheimer's disease, it can cause undesirable chemical reactions and destruction of the soil tightness of the discharge. On the other hand, the presence of Cu at high concentrations can lead to intoxication [14]. Whereas the Ni can cause gastrointestinal disorders and Cr tends to accumulate in spleen, intestines, kidneys, liver and testes [15]. Furthermore high concentrations of some zinc compounds can cause the stomach cramps, nausea, vomiting and anemia. Similarly for the Mn, ingestion of high doses causes burns in the digestive tract and the central nervous system is most likely to be severely affected by this excess manganese [16]. The mercury can have fatal effects on the ecosystem. It is neurotoxic, toxic, eco-toxic and plays a role of endocrine disruptor. It is bio-available and bio-cumulative. Two groups are generally more sensitive to the effects of mercury; fetuses are particularly sensitive to the impact of mercury on development, while exposure to methylmercury in utero may result from the mother's consumption of the contaminated water [17]. Nevertheless, Moroccan norms (Fig. 6-A) for Al and Cu, WHO guidelines (Fig. 6-B) for Pb and EU regulation (Fig. 6-E) for Hg all indicated RQ <1, thus suggesting an acceptable risk to the environment and human health in the event of contamination. It remains to be noted that the EU standards (Fig. 6-E) did not report any information on the Zn and Mn.

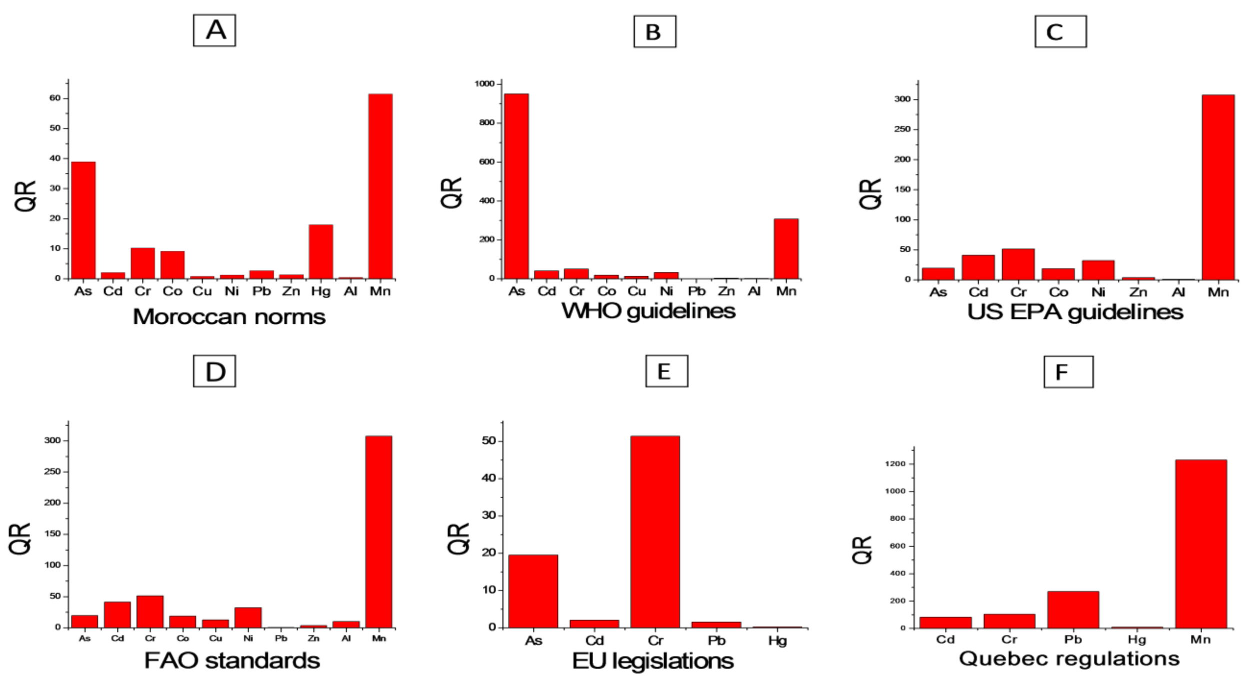

Fig. 6 *Risk quotients calculated according to Moroccan norms (A), to WHO guidelines (B), to US EPA guidelines (C), to FAO standards (D), to EU legislations (E) and to Quebec regulations (F).*

4. CONCLUSION

This study allowed assessing the degree of contamination of the leachate from the landfill of El Hajeb city by heavy metals. Indeed it was found that the majority of these heavy metals exceeded the limits allowed by most of the standards and the risk quotients have proved to be greater than 1. Heavy metals are not degradable and their continued accumulation poses a serious risk to human health by the possibility of their diffusion through the soil strongly permeable to subterranean waters excessively used for the production of drinking water, the watering livestock and the irrigation in the region. This expected contamination was associated with household, agricultural and hospital waste. These results put the accent on the necessity to better solids waste management, including the separation of waste and regulations on their eliminations. On the other hand, metals whose contents do not exceed the accepted limits could also constitute a danger to human health. Indeed these metals by bioaccumulation and Biomagnification could be found in toxic doses at certain level in the trophic chain. Classic examples of the effect of bio-concentrated toxic substances are the painful and fatal disease of Itai-Itai, caused by chronic cadmium poisoning, and Minamata disease, caused by chronic mercury poisoning. Besides, any program for the continuous monitoring of groundwater sources around the study area will contribute massively to the control of any contamination that may occur.

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