Natural content of heavy metals on cattle regions soils of Mayabeque and Artemisa province in Cuba

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In order to determine the pseudo-total concentrations of heavy metals (HM) on Cuban cattle soils with established grasslands from Mayabeque and Artemisa provinces, and to propose reference values of quality, samples of eleven representative soils were collected. They were taken at two depths and the pseudo-total and phyto-availability contents of the HM were assessed. After a cluster analysis, the soils were distributed into four groups according to the similarities in the contents of Cu, Zn, Cd, Pb, Ni and Cr. A great variation of the natural values of HM in the different types of soils was demonstrated. Many of the values found are over those commonly reported in the international literature, especially those included in the third group, corresponding to ferralitic regions.

Key words: *degradation, trace elements, geochemical fund, values of reference*.

In Cuba, soils pollution is one of the main environmental problems declared in the National Environmental Strategy 2007-2010 (CITMA 2007). This situation is associated to the synergic action of multiple stressing factors: loss of biological diversity, erosion, salinization, reduction of nutrients, climatic change, among others.

The causes of the grasslands degradation in cattle regions are multiple and interrelated (Brown 2003 and Padilla *et al*. 2003). In Cuba, grasslands cover at about 1.2 millions of hectares and have degraded progressively in the last years. The area of improved pastures decreased from 50 %, in the 80's, to 18-19 % at present (Anon 2000 and 2004). If urgent actions are not taken at present to stop the soils degradation, there is the risk of desertification. The 14 % of the Cuban territory (1 580 996 ha), distributed in 24 edaphoclimatic sub-zones, is affected by this process. Those soils addressed to cattle rearing are also included in these sub-zones (Fuentes *et al*. 1997 and Vega *et al*. 2011).

The heavy metals (HM) in natural stage are present in the soils, even when no antropic perturbation in the environment has occurred. Their concentration increases may occur due to natural processes or induced activities (Guilherme *et al*., 2005, Pedroso *et al*. 2005 and Amaral Sobrinho *et al*. 2009). Assessing the soil pollution and adopting reference values for HM levels are important to keep the agricultural and ecological functions of the soils.

Up to the present, in Cuba, HM reference values supporting the pollution level of the soils have not been defined yet. If considering that the natural levels of HM vary with the edaphic cover, it would be appropriate to compare the proposed reference values of quality in respect to the legislations of other countries.

During the last 25 years, in Mayabeque and Artemisa provinces, researches have been conducted using different technologies in grasslands, several crops and sugar cane. Through an integral methodology (Febles *et al*. 2008), these studies have allowed a system of agro-environmental indicators and the establishment of limiting values of sustainability in places of reference (Febles *et al*. 2009 and Febles *et al*. 2011). However, the studies related with the soils pollution due to HM in these strategic agroecosystems are still insufficient. In this sense, studies conducted by Muñiz *et al*. (2000, 2001 and 2006) highlight.

Considering these antecedents, the objective of this study was to determine the pseudo-total contents of HM on the main soils of Mayabeque and Artemisa provinces under natural conditions. It is also pretended to propose reference values of quality on the main types of soils, so risks sceneries in other cattle areas of the countries may be provided.

Materials and Methods

The research was conducted in representative locations of Mayabeque and Artemisa provinces, described by Febles *et al*. (2008a) and Febles *et al*. (2008b). The soil samples were collected at a depth of 0 - 20 cm (samples A) and of 20 - 40 cm (samples B), with uniform space distribution and in environments with functional biological diversity. In this area, the vegetation is practically virgen or under the natural regeneration of the tree component, with species such as marabu (*Dichrotachys glomerata*), aroma (*Acacia farmesiana*) and palm trees (*Roystonea regia*). These, acting as a fallow induced in the last two decades, propitiated certain resilience (Astier-Calderón *et al*. 2002) or morphoedaphological stability in the processes (Pablos *et al*. 2007 and Febles and Vega 2010).

In these environments, poorly disturbed or cuasi virgen, the soils may be used in quality of patterns or reference profiles, typical of each grouping (Febles *et al*. 2010).

The soils samples were broken into pieces, ground and shifted at 210 µm. They were submitted to an extract of aqua regia ($3HNO_3$+1HCl) to count the pseudototal contents (does not destroy the silicates' structures) (ISO 1995). The phyto-available fraction of the metals in the soils samples was extracted with acetic acid (Sahuquillo *et al*. 1999). The HM analyses were conducted with five analytic repetitions per sample. The mean values of the repetitions per each type of soil were considered for the statistical assessments.

The elements concentrations in the resulting extracts of the soil were counted with an spectrometer of plasma emission (ICP-OES), mark Pekín Elmer, model ÓPTIMA 3000, with LD (mg kg^{-1}) of 0.508 and LQ (mg kg^{-1}) of 1.69 for zinc; LD (mg kg^{-1}) of 0.026 and LQ (mg kg^{-1}) of 0.097 for cadmium; LD (mg kg^{-1}) of 0.020 and LQ (mg kg^{-1}) of 0.067 for lead; LD (mg kg^{-1}) of 0.01 and LQ (mg kg^{-1}) of 0.05 for iron; LD (mg kg^{-1}) of 0.01 and LQ (mg kg^{-1}) of 0.020 for manganese and LD (mg kg 1) of 0.03.

In order to validate the determination of the pseudototal content of zinc, cadmium, iron, manganese, lead and chromium on the soil, the reference certified material NIST SRM 2709a (San Joaquin Soil, with concentrations of: zinc of 103 ± 28 mg kg^{-1}; cadmium, 0.371 ± 0.002; lead 17.3 ± 0,1; manganese of 529 ± 18 mg kg^{-1}; iron of 3.36 ± 0.07 %) was used.

The analyses of organic carbon, pH, interchangeable cations, potential acidity, calculation of the basis, cationic interchange capacity and the granulometric analyses were conducted according to EMBRAPA (1997).

For identifying the similarity structure between the different soils based on the pseudototal values (Cu, Zn, Ni, Cr, Cd and Pb) of the samples, at depths of 0-20 cm and of 20-40 cm, were submitted to the cluster method Simple Link, from the Euclidian distance (SAS, 2010). The cutting point of the dendrogram was carried out according to the criteria of Milligan and Cooper (1985), who consider a cutting point of 1.25 times or standard deviations of the differences between the linking distances.

The data were submitted to a correlation analysis of Spearman (SAS 2010) to assess the intercorrelation between the variables studied. Later, the correlation matrix was organized in a color matrix at scale of equal intervals.

Results and Discussion

The natural concentration of HM on the soils depends, mainly, on the origin material, the formation processes and on the composition and proportion of the components in their solid phase. The relation of the soil with the material of origin is evidenced when the soil is formed *in situ*, being it less expressive on those originated on sediments. The soils originated from basic rocks have naturally higher contents of these elements, compared to those formed on granites, gneis, areniscas and limolites (Oliveira 1996, Facchinelli *et al*. 2001, Fadigas *et al*. 2006 and Utermann *et al*. 2006). Apart from the nature of the material of origin, other factors such as percentage and composition of the clay fraction, content of organic matter and physico-chemical conditions may influence on the richness of HM on the soil (Barona and Romero 1996, Oliveira 1996 and Fadigas *et al*. 2010).

According to studies of Campos *et al*. (2004) and Jaimez *et al*. (2004), the soils of higher geographical distribution in the areas of the old Havana provinces (table 1) are those of the Brown Sialitic group (24. 2 %), followed by those of the Ferralitic (22.1%) and, in third place, those of the Ferralic group (13.2 %).

The firsts (Brown Sialitic) are mainly associated with rocks of the terrigenous-carbonated type and carbonataded-terrigenous (margas, marlys limestone) and, in lower degree, with basic ignea and ultra basic rocks. Its distribution coincides, in great extent, with high waved or slightly waved plains, where the soils have been formed due to tropical meteorization of sialitic type. In them, their evolution has been modulated by different manifestations of erosion (present and geological) and by an edaphic process called self-restrain (Hernández 1973).

In general, they are characterized by being carbonated soils, or saturated by alkaline-terreous, predominating the calcium in the interchange. They have cationic interchange capacity (CIC), generally superior to 30 $cmol^{(+)}$. kg^{-1} (determined in the clay fraction). The content of free iron compared with the total one (Fe_F/Fe_T) is lower than 40 %. The organic matter content, generally, is middle (2.0-4.0 %). The pH, under natural conditions, is generally neutral to slightly alkaline.

In second place appear the soils of the Ferralitic group (22.1%) are distributed, mainly, in the plainest areas (carsic pediplain) of the Meridional Carsic Plain Havana-Matanzas and in areas of higher stability of the Havana-Matanzas Heights and Bejucal-Madruga-Coliseo (high plains).

Differently from those brown, the geological environments influencing the most on its nature and properties are the followings: rainfall over

1 200 mm and annual mean temperatures superior to 20 °C (Roose 1981 and Collinet 1988) that, except quartz, affect the rest of the primary minerals, washing the alkaline basis and alkaline-terreous. This brings about the accumulation of oxides and hydroxides of iron and aluminum, predominating the clayish minerals type 1:1, like etahaloisita, caolinita, goethita, hematita and traces of gibbsita (Obregón 1979, Sarazin *et al.* 1982, Kampf and Schwertmann 1982, Schwertmann and Murad 1983, Vizier 1983 and Boulet *et al*. 1985). This dynamics favors and induces the ferratilization processes (Barranco and Díaz 1988 and Herrera 1996) and is one of the main causes justifying its wide distribution in the country (Ascanio 1980 and Ascanio *et al*. 1983).

Lithologically, these soils are based on limestone or any other carbonated materials with different degree of impureness (Gonou 1997 and Febles 2007), with mean ratios of SiO_2/Al_2O_3 and SiO_2/R_2O_3 in clay inferior to two, low CIC and slightly acid pH (Delgado 1987, Izquierdo *et al*. 1990, Aleva 1994 and Vargas 2009). They can be dated with fluctuating age between 120 000 and 450 000 years, according to the dissolution coefficient of the Cuban limestone (Iturralde-Vinent 1972) and to their impureness content (Camacho and Ruíz 1980).

This close genetic association with the Ferralitic soils (paragenesis), the Ferralic group is distributed in the present provinces of Mayabeque and Artemisa. This group has similar characteristics to those of the previous, but dealing with saturate soils, with natural pH values, superior to 6.5 (neutral) and CIC > 20 $cmol^{(+)}$. kg^{-1} (determined in clay).

Its origin is associated with the Ferralitic soils and, like them; they are derived soils from members with content in hard limestone. However, sometimes they have less caved limestone (slightly superior clay). This determines higher cohesivity and size of the aggregates, and an evolution process called "incomplete ferralitization" (Instituto de Suelos 1999).

The three described groups are the edaphic cover of highest agricultural importance in the territory, due to their agro-productive attributes (from medium productive up to highly productive) and to their geographical distribution.

Other soils of agricultural importance in both provinces are those of the Sialitic Humic grouping (7.6 %), followed by those of the Hydromorphic group (6.9 %) and the Fersialitic (2.8%). The Alluvial soils (at present Fluvisol: 2.2 %) and the Allytic (0.9 %) are restricted in the southwest of the provinces. Those of the vertical or Vertisol type (0.2%), only distributed to the east and northeast of the area, and those of the Ferritic

Table 1. Links between the main type of soils and the stony underlying materials in Mayabeque and Artemisa provinces, in Cuba.

Type (Paneque *et al*. 1991)	Lithology	Surface (km^2)	Area (%)	World Reference base (2006)	New classification version (Hernández *et al*. 1999)
Brown Sialitic Brown with carbonates Brown without carbonates	Soft limestone (marly limestone)	1561.30	24.20	Cambisol Calcic	Brown sialitic group. Brown Type
Red Ferralitic Hydrated red Ferralitic Typical Red Ferralitic humified; Yellowish ferralitic Gley Ferralitic	Hard limestone, generally caved	1429.30	22.10	Ferralsol Rhodic Ferralsol Rhodic-Endostagnic Ferralsol Rhodic-Humic Acrisol Chromic-Ferric Gleysol Ferric	Typical red ferralitic Humic red ferralitic Yellowish ferralitic, typical lixiviated Yellowish Lixiviated, humic and gley
Ferralic Red Ferralic, humic	Hard limestone, generally little caved	854.40	13.20	Cambisol Ferralic-Rhodic-Humic	Red ferralic, typical
Humic Sialitic Humic Carbonated	Margas and calcareous lutites	489.90	7.60	Feozem Chernic	Humic Calcimorphico, typical
Fersialitic Brown fersialitic Soft redish brown	Variated (limestone, serpentinites)	183.70	2.80	Cambisol Chromic-Humic	Brown fersialitic soft brown redish
Alluvial	Alluvial deposits	141.80	2.20	Fluvisol Haplic	Fluvisol
Ferritic Dark Typical humified	Basic and ultrabasic rocks	1.40	0.02	Ferralsol Rhodic-Ferric Humic	Dark red ferritic, humic
Total		4661.80	72.20		

group occupy only 1.4 km^2 (0.02% of the total), limited to serpentinite blocks around Madruga town.

The studied soils belonging to Mayabeque and Artemisa provinces, form four groups (figure 1), corresponding with the criteria proposed by Milligan and Cooper (1985). These soils express the pollution dangers of the edaphic cover in the cattle establishments.

The following types of soil were included in group 1: Brown with Carbonate (on margose limestone), Reddish Brown Fersialitic (on fragmented hard limestone), Humified Typical Red Ferralitic (on limestone), Yellowish Ferralitic (on fragmented hard limestone), Yellowish Gley Ferralitic (on limestone eluvia), Carbonatic Humic (on marly limestone) and Fluvisol (on sediments). This group is characterized by intermediate metal values in respect to the rest (table 2) and has the highest geographical part of both provinces.

Even when the middle contents of HM in this group are no the highest, they are higher than those values considered as normal in other countries (table 2) (Kabata-Pendias and Pendias 1984, CETESB 2005, Fadigas *et al.* 2006 and Ballesta *et al.* 2010).

The group 2 is formed by samples of Brown soils without carbonate, sustained on serpentinite (samples 2A and 2B). These samples are different from those of the group 1 because they have higher values of Ni and Cr compared to the rest of the soils of this group. However, the concentrations of Cu, Zn, Cd and Pb are similar. Nevertheless, the values of Ni and Cr of the samples of group 2 were not high enough to be included in group 4 (table 2).

In the groups 3 samples of the Eutric Typical Ferralic soil (marly limestone) and hydrated Red Ferralitic soil (slightly unsaturated ferralitic cover) were included. The

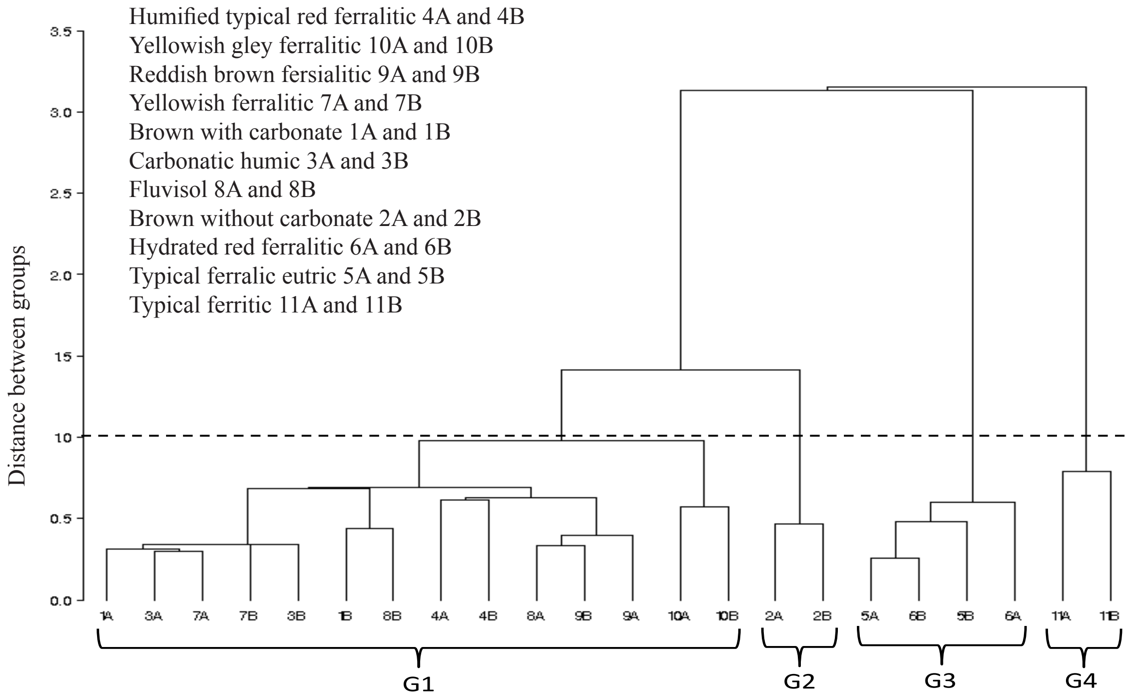


Figure 1. Suggestion up to the cutting point of the dendrogram, according to the simple linking method

Table 2. Mean values and standard deviation of HM content for the four groups formed with the cluster analysis.

Groups	Ni, mg kg^{-1}	Cr, mg kg^{-1}	Cu, mg kg^{-1}	Zn, mg kg^{-1}	Pb, mg kg^{-1}	Cd, mg kg^{-1}
G1	68.0 ±43	78.0 ±37	63.0 ±28	114.0 ±48	44.0 ±5.2	3.1 ±1.1
G2	676.0 ±79	1259.0 ±87	68.0 ±6.6	121.0 ±205	36.0 ±208	2.0 ±0.1
G3	292.0 ±12	290.0 ±32	138.0 ±8.5	462.0 ±11	67.0 ±2.2	10.9 ±0.8
G4	4663.0 ±474	2944.0 ±359	48.0 ±104	83.0 ±6.8	37.0 ±1.0	2.2 ±0.2
Cetesb (2005)	13.0	40.0	35.0	60.0	17.0	0.5
Fadigas *et al*. (2006)	5-35	19-65	2-119	6-79	3-40	0.3-1.5
Ballesta *et al* (2010)	42.6	113.4	27.0	86.5	44.2	4.4
Kabata-Pendias and Pendias (1984)	0.2-660	1.4-1384	1.0-323	3.5-622	1.5-189	0.01-2.53

main characteristic of this group is the highest values of Cu, Zn, Cd and Pb in respect to the other soils. The contents of Cd and Zn are over the normal values under natural conditions.

Similar values have been reported in certain environments in German soils (Utermann *et al*. 2006) in forest vegetation, with maximum proportions of Zn, varying between 534 and 3 628 mg kg^{-1}, and of Cd, ranging between 15.9 and 3.0 mg kg^{-1} for the soils formed from alluvial sediments and calcareous rocks, respectively. The contributions of the carbonated rocks, and especially of those of the serpentinite type in the high contents of HM are demonstrated.

The group 4 is hardly formed by superficial and sub-superficial samples of the Typical Ferritic soils (on serpentinite). This group has, as main characteristic, high contents of Cr and Ni.

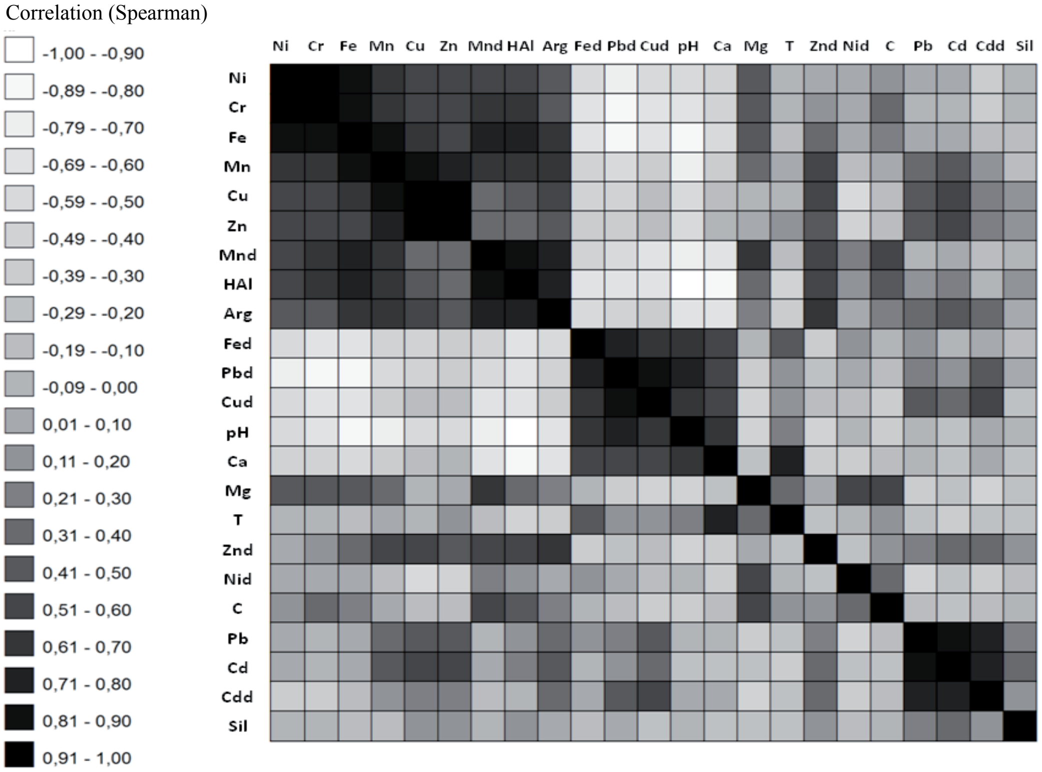
Generally, the ultra basic rocks like serpentinites, are materials with high proportions of Cr and Ni (Facchinelli *et al*. 2001). The soils originated on this substrate tend to have higher content of these elements. Facchinelli *et al*. (2001) applied an analysis of principal components in data obtained in Italy and confirmed a high relation between the values of Cr, Ni and Co and the soils originated of serpentinitic rocks. The Ferritic soils, characterized by a more advanced of meteorization between the Cuban soils, show the highest values of oxides in the clay fraction (Hernandez *et al*. 1999). The Fe and Mn oxides propitiate high adsorption of HM in the soils fractions (Amaral Sobrinho *et al*. 2009).

The high values of Ni and Cr, observed in group 4, are quite over those reported in the international literature, in soils uder natural conditions. In studies conducted in thirteen countries of the European Union (Utermann *et al*. 2006) very high contents were only reported in four agricultural regions on calcareous rocks of the Slovakian Republic. In this area, maximum values of Ni and Cr, of 2 066 and 6 096 mg kg^{-1} respectively, were found. Meanwhile, the 90 percentile values for agricultural areas and on calcareous rocks were of 60.7 and 122.1 mg kg^{-1}, in respect to the forest areas on carbonated materials. The contents of Ni and Cr were of 242 and 665 mg kg^{-1}, respectively.

In Cuba, previous studies indicated anomalies in the contents of Ni and Cr in some soils. That was proved by Baró (2011), who reported higher values than those obtained in this study.

The data showed two structures of high correlation, as they can be interpreted as rulling factors of the soil in the performance of these variables. These structures may be identified in the figure 2 due to the color contrasts. Those colors tending to black express positive high correlation and high negative those tending to white.

The first structure comprises the variables pseudototal



Significant correlation at 1% equal to ±0.54.
The letter d signify availability of the element

Figure 2. Correlation matrix of Spearman in 22 samples of the superficial and subsuperficial covers in representative cattle soils of Mayabeque and Artemisa provinces.

HM (Ni, Cr, Fe, Mn, Cu and Zn), Mn available (Mnd), H + Al, arcilla, Fe available (Fed), Pb available (Pbd), Cu available (Cud), pH and Ca (figure 2). The fact of these elements being closely related with variables associated to the pedologic formation such as H + Al, arcilla, pH and Ca, indicates a natural occurrence of the pseudototal values of these HM on the soil. This structure corroborates the results of Fadigas *et al*. (2006 and 2010) for the most representative soils of Brazil.

The second structure is reflected by the variables pseudototal Pb and Cd and the available Cd (Cdd). The isolation of the variables Pb and Cd in this structure may indicate an anthropic or natural pollution. It promotes a maintaining rupture of these elements proportions in respect to the rest. The geological formations favorable to the concentration of some HM, in punctual conditions, are examples that may justify this phenomenon.

This structure seems to be associated to a pedo-geological event, as the high concentrations of pseudototal Cd and available Cd are related with the soils of group 3. This is limited for the type typical Ferralic eutric (marly limestone) and hydrated Red Ferralitic (Ferralitic cover in unsaturated medium).

Studies of Patraa *et al*. (2008), when comparing the affected areas due to anthropic pollution with similar not-polluted environments, reveal the influence of the high values of Cd and Pb on the chemical composition of milk. In the cattle rearing establishments of the provinces under study, located in soils of group 3 (ferralitic regions), the risks possibilities in respect to the HM transference, mainly of Cd, throughout the grasslands, the water supplied to the animals and milk and meat should be considered. This danger has been corroborated in classic studies conducted by Voisin (1963) in other environments.

Great variation of the HM natural values on the main types of soils of the cattle regions studied is presented, especially the concentrations of Cd, Cu, Ni, Pb and Zn that are over the threshold values reported in the specialized international literature. This manifests the danger these elements represents in the feeding chain.

The HM content values obtained in the soils of Mayabeque and Artemisa provinces may be interpreted as a first approaching to design management strategies and mitigation addressed to risks sceneries in cattle establishments in Cuba.

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