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ADSORPTION OF CD, CR, NI, AND PB IN SATURATED SOILS AT MAXIMUM ADSORPTION CAPACITY OF THESE HEAVY METALS

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ABSTRACT

Environment pollution and particularly of the soil compartment has been set as a major socio-environmental problem, mainly when it concerns the accumulation of certain chemical elements in the soil. Identifying the adsorption mechanisms, if the process tends to be more specific or electrostatic, or even if the behavior of one metal when other has been previously adsorbed already may contribute to the development of techniques capable to minimize the environmental problems caused by waste rich in heavy metals on the ground previously deposited. Selected soils for this study belong to the predominant classes of the State of Minas Gerais, which have received dosages equivalent to the maximum adsorption capacity (MAC). Soil that are incubated with doses equivalent to MAC of the heavy metals Cd, Cr, Ni, and Pb and that later received larger dosages of those same metals, presented similar behavior when adsorption is analyzed in increasingly different added dosages. There was no correlation between adsorbed values of heavy metals Cd, Cr, Ni, and Pb in soils incubated with the MAC of those metals, with the classes of soils. Heavy metals Cr and Pb were those that adsorbed the largest amounts of heavy metals previously incubated in studied soils, and also those that were adsorbed more strongly in the same situation.

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INTRODUCTION

Environment pollution has been set as a major socio-environmental problem, mainly when it concerns the accumulation of certain chemical elements in the soil. Metals accumulate in soils due to natural and anthropic processes such as, for example, atmospheric deposition deriving from industrial activities, deposition of sludge, domestic waste or by-products, use of fertilizers and pesticides. High concentrations of heavy metals in the soil may affect ecosystems' productivity, biodiversity, and sustainability, becoming risk for human and animal health (Sun *et al*., 2001). Reactions with heavy metals in soils are ruled by principles that rule ions interactions with solid surfaces. Thus, the adsorption, desorption, and the consequent bioavailability of the heavy metals are controlled by the soils features, such as pH, redox potential, clay content, organic matter, presence of Al, Fe, and Mn oxides and calcium carbonate (McLean and Bledsoe, 1992; Sparks, 1995; Rieuwerts *et al*. 1998; Krishnamurti *et al*., 1999). Essays have been developed aiming at understanding the processes that determine metals competition and adsorption, mostly adding them jointly to the soil, which results in the establishing the adsorption sequences that help predicting the behavior of those metals and, consequently, their behavior in multi elements systems. Several studies have shown the affinities sequences of metals for the available adsorption sites in different soils, residues, and other materials (Gomes *et al*., 1997; Fontes *et al*., 2000; Gomes *et al*., 2001; Fontes and Gomes, 2003). There is a more common adsorption sequence for soils, as follows: Crop>Cu>Zn>Ni>Cd. Literature points to position changes between Cr and Pb, and Ni and Cd (Matos *et al*., 1996; Gomes *et al*., 2001; Fontes & Gomes, 2003; Pierangeli *et al*., 2004; Basílio *et al*., 2005; Usman, 2008; Kamala-Kannan, 2008). Importance has also been given, beyond chemical characteristics, to soil physical features in relation to adsorptive behavior of heavy metals. Moreira (2004) states that soils with greater adsorption capacities of metals were those with clay and very clayish textures, while less amounts of adsorbed metals referred to soils with sandy texture. Similar data was obtained by Suzuki et. al. (2006), which noticed that metals are adsorbed in the soil, mostly in their clayish fractions and soils with silt-sandy texture may not efficiently adsorb heavy metals when put in contact with domestic waste deposits, leading to underground waters contamination in regions close to those deposits, which may lead to compromising the quality of water bodies. It was evaluated, in this study, the behavior of a pre-adsorbed heavy metal through addition of other heavy metals in different concentrations equivalent to its maximum adsorption capacity for each surveyed soil.

METODOLOGY

Selected soils for the essay belong to the predominant classes of the State of Minas Gerais, which are: Oxisols, Alfisols, Inceptisols, Entisols, and Ultisols. It was sought to work with soils, within classes, horizons A and B that could present contrasting characteristics in chemical and physical terms. The main differences between soils chosen to develop the current study were: amount of clay, organic matter content, and pH values of the soil samples. The collecting points are inserted in areas without anthropic influence or change, and samples collect with stainless tools and conditioned in sealed and identified plastic bags. Prior to analyses, samples were shade dried, screened in 2 mm mesh, homogenized and identified. After this procedure, 500 g. samples from these soils received doses equivalent to their maximum adsorption capacities (MAC) of Cd, Cr, Ni, and Pb and they were kept incubated in field capacity for 30 days. In samples of 2 g of soil, screened in 2 mm mesh opening, derived from incubated soils, 20 mL of 0.01 mol L-1 solution of NaNO3 (1:10 ratio) were added having initial concentrations of Cd, Cr, Ni, and Pb in triplicate. Metals were individually added in each sample through Ni (NO3)2, Cd (NO3)2, Pb (NO3)2 e Cr (NO3)3 solutions. These amounts were equivalent to 0; 0.5; 1; and 1.5 fold to the maximum adsorption capacity that had been previously determined for each studied metal and soil. The soil-solution set was conditioned in polyethylene tubes with capacity of 50 mL and after 24 consecutive hours shaking in horizontal shaker at 120 g. After shaking, suspensions underwent centrifugation at 2,5 g for 5 minutes, the supernatant filtered in quantitative paper filter and collected for determination of metal contents by flame atomic adsorption spectrophotometry. The amount of adsorbed metal was calculated by the difference between the added amount of metal and the remaining quantity in the balance solution. The results were presented and discussed using the multivariate statistical analysis grouping method in the STATISTICA version 7.0 (Statsoft, 2004) software.

RESULTS AND DISCUSSION

The results of the readings of metals previously incubated (Table 1), due to later addition of other metals in the system, indicates the capacity of the second metal to displace the first, therefore, it may be inferred that the concur for the same sites or over the adsorption mechanisms of each of the metals in the study. In this case, an adsorption increase of the second by means of the non-accrual of the first in the solution would indicate that the adsorption sites for the metals would be different. In this study, the added metals provided equivalence of applied doses or even higher than the doses used for incubation of the first metal. Studied soils pH suffered greater reductions with the addition of Cr and Pb, and less with addition of Cd and Ni. It is known that metals adsorption is a process much influenced by the soil pH (Rieuwerts *et al*., 1998), while pH decrease favor the formation of free metallic cations, and using simple bonding for obtaining soils groupings with similar behavior. The hierarchic analysis of grouping (Sneath & Sokai, 1973) was undertaken by calculating the Euclidian distance between samples for the set of 19 soil samples with individual doses of other three heavy metals, and by using the simple bonding for obtaining the soil grouping with similar behavior. The analysis result was presented as dendrogram graphic that helped indentifying groupings of the samples. In general, the division and the grouping of the soil samples presented a very important result that was the ordering of these samples according to the adsorbed quantities of the metal that was later added. Soils that initially received similar quantities of the second metal have grouped, showing that they behaved similarly when analyzing the quantities of this adsorbed metal in the different dosages (Figures 1, 2, 3 and 4).

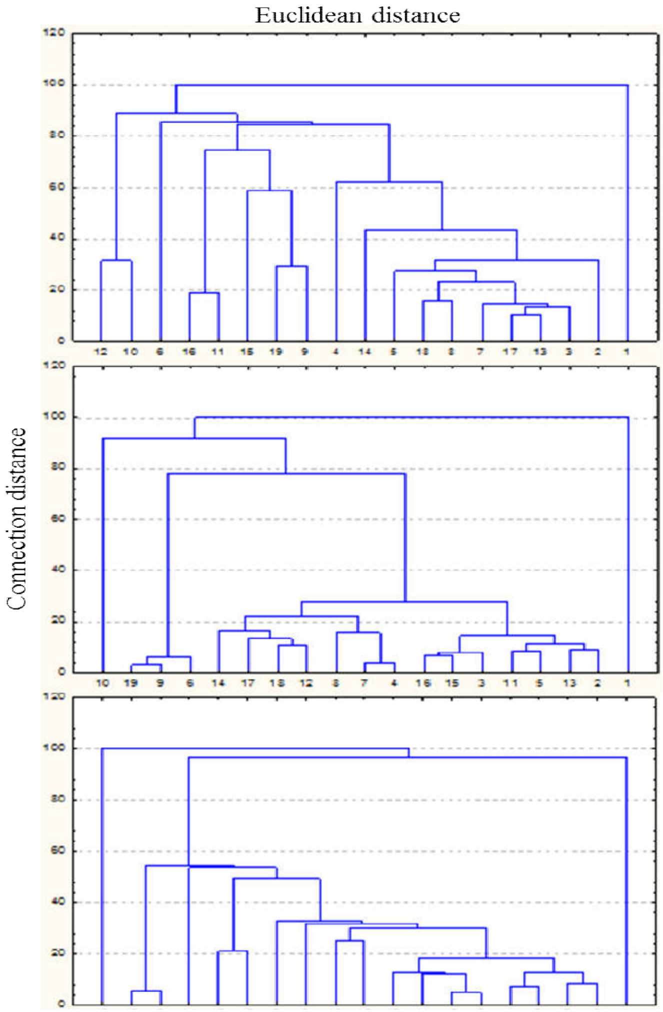

Figure 1. Hierarchical cluster analysis with adsorption of Cr, Pb e Ni respectively in soils incubated of Cd MAC.

Soils incubated with Cd: It was observed in soils previously incubated with doses equivalent to MAC that Pb and Cr were the most adsorbed metals, followed by Ni in of Cd, while the sequence Pb>Cr>Ni was the most common for the majority of studied soils (Figure 4). This result shows the equivalence mechanisms of adsorption for Pb and Cr as counterpoint to the mechanism through which Ni is adsorbed or if these metals are adsorbed through the same mechanism, the force with which Cr and Pb are adsorbed is higher than that of Ni. Generally, the greater added quantities led to greater metals adsorption. When there was predominance of Cr adsorption in relation to Pb, it occurred primarily in the latosoils. This behavior may be related to the fact of latosoils offer more specific adsorption sites, while the Cr is more strongly adsorbed than Pb in these conditions. Literatures indicate that trivalent metals, such as Cr, should be preferably adsorbed over all bivalent metal (McBride, 1994). Cd was desorbed as consequence of later addition of increasing individual doses of Cr, Pb, and Ni (Figure 4). Generally, greater doses added of the second metal caused greater Cd desorption. Pb, Cr, and Ni, in this order, were metals that causes the greatest Cd desorption in the studied soils.

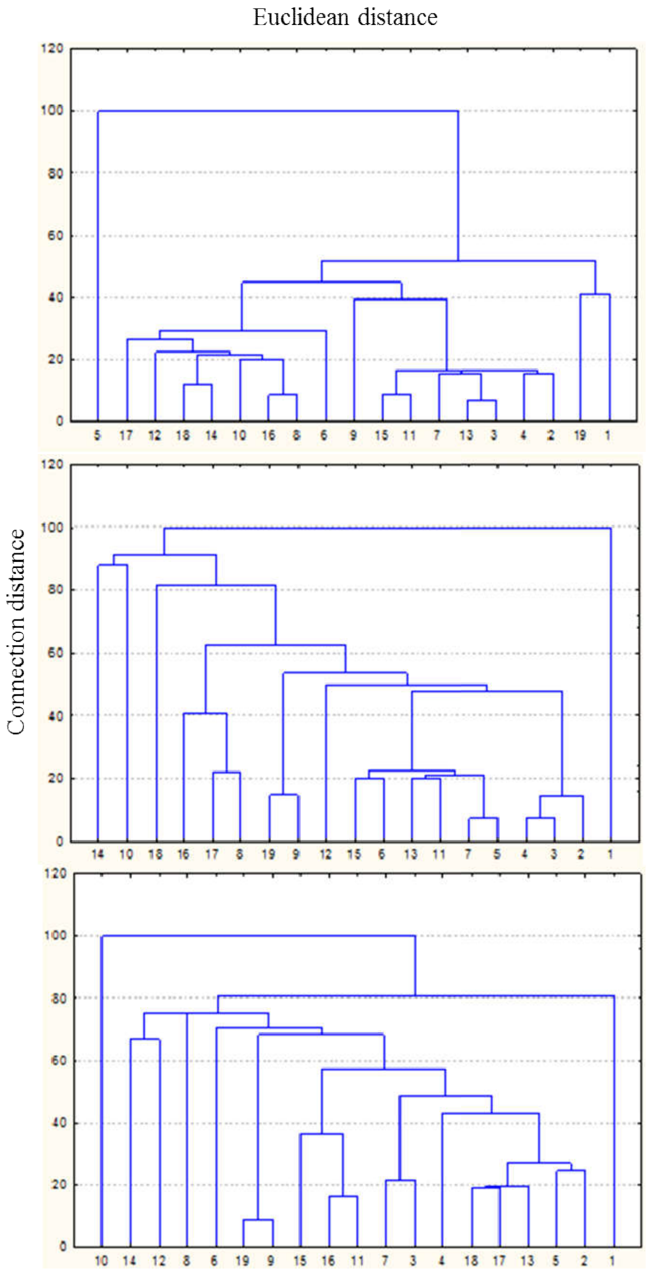

Figure 2. Hierarchical cluster analysis with adsorption of Cd, Ni and Cr respectively in soils incubated of Pb MAC

In samples 7, 9, 10, 11, 13, 15 16, and 19, in average, 80% of previously incubated Cd remained adsorbed after adding 1.5 fold the equivalent amount of maximum adsorption capacity of Pb. This behavior may indicate that Pb is been adsorbed by other sites that are not the same already occupied by Cd. The observed correlations between quantity of adsorbed metal in soils incubated with Cd and the physic-chemical features of these soils indicate that there was great similarity between adsorptive behaviors of soils incubated with Cd and that had received increased and individual doses of Cr, Pb, and Ni. The adsorption of Pb and Cr did not only correlate with saturation by Aluminum. A positive correlation between organic matter content in the soil was observed, attribute that confers to soils great affinity for cations (Hodgson, 1963), with adsorptive behavior of Cr, Pb, and Ni in previously incubated soils with maximum adsorption capacity for Cd. Concerning classes of soils used here it was noticed that Cr, Pb, and Ni adsorption in previously Cd incubated Ultisols and Alfisols (Table 6) resulted in adsorption sequence Cr>Pb>Ni. The observed behavior for Oxisols indicates a greater adsorption of Cr, followed by Pb and Ni (Cr>Pb>Ni), been the latter, generally, the same adsorption sequence of heavy metals used for the studied Inceptsols and Entisols. This behavior did not differ between soils or between their horizons.

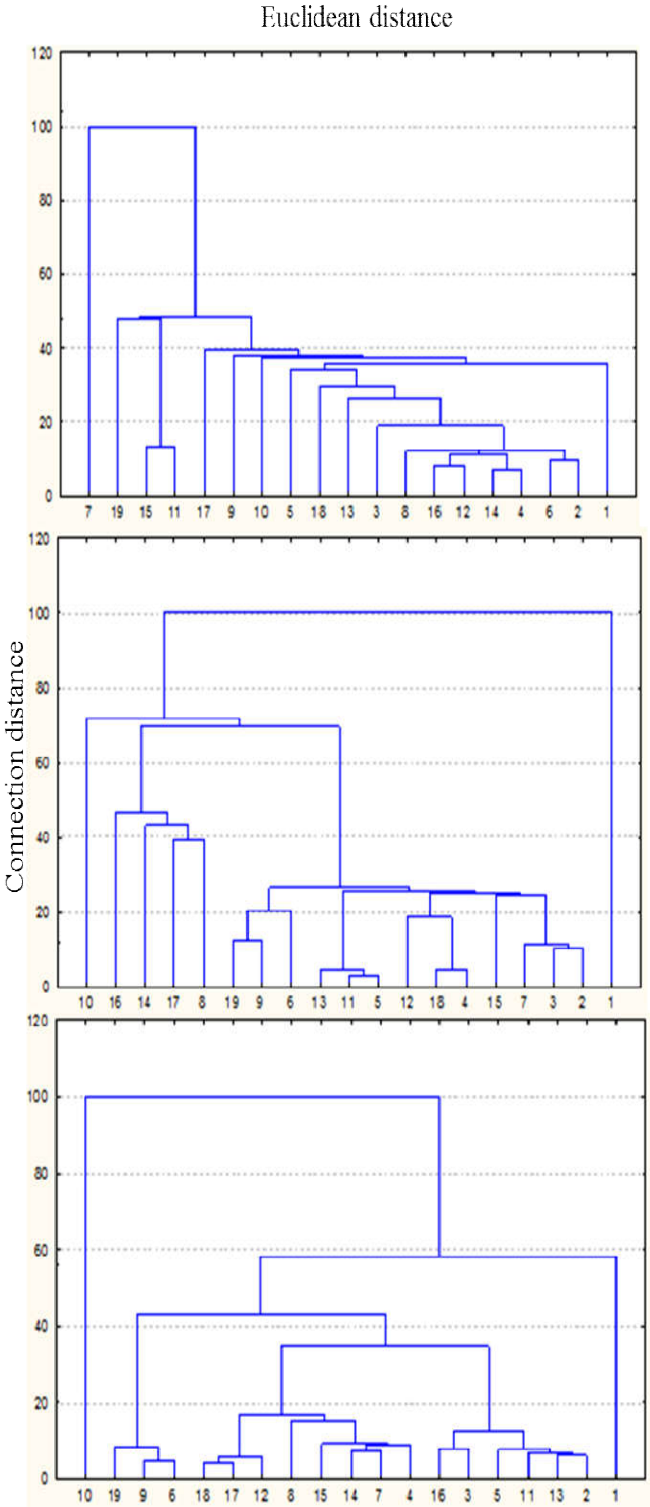

Figure 3. Hierarchical cluster analysis with adsorption of Cd, Ni and Cr respectively in soils incubated of Cr MAC

Soils incubated with Cr: The heavy metal Pb, in most of studied soils, was the one that was kept most adsorbed compared to Cd and to Ni in Cr incubated soils. In soils in which it is noticed adsorption preference for Pb over Cadmium, some authors observed that the preference for Pb can be attributed due to the smaller size of its hydrated ionic radius (Pb = 0,401nm e Cd = 0,426nm) (Gao *et al.*, 1997; Pardo, 2000; Phillips, 1999), in addition to Pb great affinity for functional groups of organic matter as phenolic and carboxylic groups. The fact of Pb is more electro negative than Cd (2.10 and 1.69 for Pb and Cd, respectively) and having a pK (constant of balance) (7.78 for Pb and 11.70 for Cd) makes Cd a metal with electrostatic adsorptive features in reactions involving complexation

Table 1. Maximum adsorption capacities (mmol kg-1) of Cd, Ni, Cr, and Pb used in previous incubation of soils

				ALFISSOLS					
Horizon A					Horizon B				
SOIL	Cd	Ni	Cr	Pb	SOIL	Cd	Ni	Cr	Pb
1	140.75	133.30	116.26	156.42	11	17.21	26.36	18.26	22.66
2	8.96	15.35	12.82	19.86	12	5.02	12.48	4.51	7.99
3	8.38	15.82	10.35	15.88	13	12.43	24.14	10.19	19.24
				OXISOLS					
4	7.13	15.82	16.76	10.72	14	4.95	7.58	6.62	9.62
5	20.48	21.12	10.46	21.36	15	16.57	29.19	29.97	16.69
6	8.95	42.68	65.70	49.56	16	6.04	11.48	17.80	16.32
7	10.43	15.70	11.55	10.52	17	3.22	9.94	9.86	10.65
				INCEPTISSOLS					
8	4.84	8.43	9.60	12.88	18	5.38	15.93	8.80	9.29
9	22.07	44.51	40.69	46.78	19	36.18	53.35	34.75	47.65
				ENTISSOLS					
10	2.89	5.56	4.30	3.24					

(1) Alfisol (2) Ultisol, (3) Inceptisol, (4-7) Oxisol, (8-9) Inceptsol, (10) Entisol, (11) Alfisol, (12) Ultisol, (13) Alfisol, (14-17) Oxisol, (18-19) Inceptisol. Classification System of Soil Taxonomy (2006).

by inner sphere (Huheey, 1983; McBride, 1994). Greater amounts of added heavy metals in solution provided greater adsorptions of these metals in the different applied doses. The fact that Ni be more adsorbed than Cd in previously incubated soils with Cr, may be explained by the fact of Ni be adsorbed by non-specific or electrostatic mechanism.

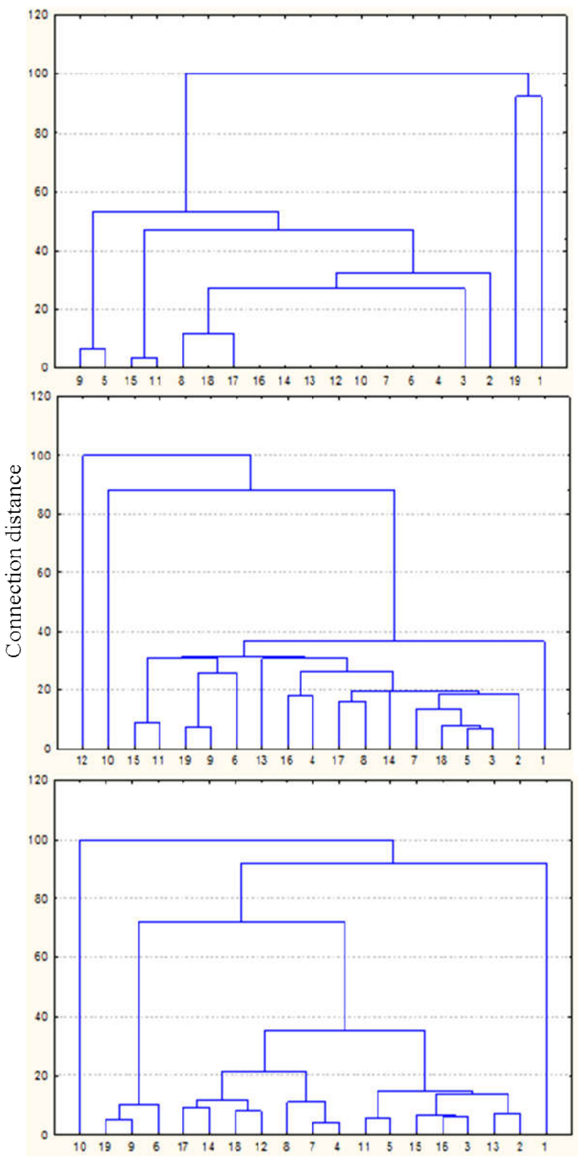

Figure 4. Hierarchical cluster analysis with adsorption of Cd, Cr and Pb respectively in soils incubated of Ni MAC

In higher incubation doses of Cr (116,26 mol kg^{-1}), Cd was not adsorbed as observed for the other used soils, which indicated that this metal may be more easily leached if it contacts a soil that has high Cr adsorbed values in its matrix, setting up thus a major environmental problem. Pb is the metal able of more desorption in previously incubated Cr. Soils incubated with Chrome Maximum Adsorption Capacity, the most observed adsorption sequence of heavy metals in studied soil was (Pb>Cd>Ni).

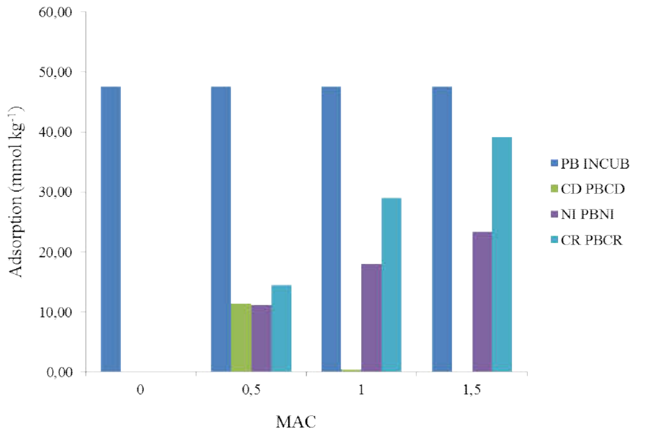

Figure 5. Cd, Ni, and Cr adsorption in previously incubated soils with doses equivalent to Maximum Adsorption Capacity (MAC) of Pb

Soils incubated with Ni: The heavy metal Pb, in most of studied soils, was the one that was kept most adsorbed compared to Cd and to Cr. In soils incubated with Ni, there was less Cd adsorption in comparison to Cr and Pb. This behavior indicates difficulty for Cd to remain adsorbed in soils that already are in sites occupied by Ni. With Cd been specifically adsorbed, but with less strength than Cr and Pb, this behavior should have been expected since some authors indicate Ni as more strongly adsorbed regarding Cd. Pb was the heavy metal that desorbed Ni the most off the exchange complex. In soils incubated with Nickel Maximum Adsorption Capacity, for the same horizon, the most observed adsorption sequence of heavy metals in studied soil was (Pb>Cr>Cd).

Soils incubated with Pb: Generally, there was predominance of higher adsorbed values of Cr, Ni, and Cd, respectively, in soils incubated with Pb maximum adsorption capacity. In the case of soil 1 (Figure X), it was not noticed Cadmium adsorption in the later additions of the corresponding to 1 and 1.5 fold of Cadmium MAC, which may indicate that it has the same adsorption mechanism as Pb, but with less strength; thus, Cd is one of the metals that causes most concern to the environment (high mobility in the soil). Nickel was adsorbed in the three applied doses; this behavior indicates that Ni is adsorbed by a mechanism that is different to Pb. On the other hand, Chrome, for which it was also noticed accentuated adsorption by the same mechanism as that of Pb. Generally, Cr accounted for the greatest values of desorbed Pb. In soils incubated with Maximum Adsorption Capacity for Lead, for the same horizon, the most adsorbed metal was Chrome. For the studied Argisoils, the second most adsorbed metal was Nickel, followed by Cadmium; on the other hand, Cadmium was the second most adsorbed metal followed by Nickel for the latosoils, cambisoils, and neosoils.

CONCLUSION

- The most important features for adsorbing metals by these essays, already having an adsorbed metal, were the organic matter content and the CTC;
- The heavy metals Cr and Pb were the ones that adsorbed the largest amounts of previously incubated metals in studied soils, and also the most strongly adsorbed ones.

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