IMPACT OF ACIDIFICATION ON POLLUTANTS FATE AND SOIL FILTRATION FUNCTION

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Abstract

The objective of this paper was to investigate the effects of simulated acid load on the fate of inorganic pollutants (Cd, Pb), soil sorption potential, soil filtration function. We made use of a short-term acidification pot experiment with grown plant of spring barley cultivated at 4 different soil types (Fluvisol, Cambisol, Stagnosol, Podzol). The potential of soil filtration was evaluated according to the Eq.: [Soil filtration function]=[Potential of soil sorbents]+[Potential of total content of inorganic pollutants]. Potential of soil sorbents (PSS) is defined by qualitative (pH, organic matter quality - $A^{400}/_{600}$) and quantitative factors (carbon content-Cox, humus layer thickness-H) according to the Eq.:[PSS]=F(pH)+F($A^{465}/_{665}$)+F(Cox)*F(H). Acid load significantly influenced soil sorption potential and thus affected increase in Cd and Pb mobility what was reflected in their transfer into the plants. Results of soil filtration function showed significant change of filtration function in Cambisol.

Key words: *acidification, soil reaction, Cd, Pb, soil filtration function*

Introduction

Ecosystem services are defined as the beneficial flows arising from natural capital stocks and fulfilling human needs. Dominati (Dominati *et al.*, 2010) defined three groups of ecosystem services: provisioning, regulating and cultural ecosystem services. Soils provide regulating services which enable humans to live in a healthy environment as well as to grow a healthy crop. Soil regulating services include among other filtering of risk elements. Commonly, soil filtration function means ability to keep pollutants and prevent them from leaching and contamination of groundwater or from entrance to food chain. Sorption processes play an important role in soil fixation of potential risk elements. The toxicity of inorganic risk elements varies from one element to another, decreasing in order of: Hg > Cd > Ni > Pb > Cr (Hansen *et al.*, 2001). Some of them are essential in small concentrations. Hg, Pb and Cd are non-essential and belong to the most toxic among inorganic risk elements. Cadmium and leads are highly toxic to plants, animals and humans. Many authors (Yong *et al.*, 1992, Hansen *et al.*, 2001, Bolan *et al.*, 1999, Makov-

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níková, Barančíková, 2012, Lacarce *et al*., 2012) estimated the influence of selected soil parameters on mobile and potentially available risk element content. The soil reaction, the content and quality of soil organic matter, the content and quality of clay fraction and iron and manganese oxides belong to the main factors which influence the mobility of the potential risk elements in soil (Zeien and Brűmmer, 1989, Blume and Brűmmer, 1991, Boruvka and Drabek, 2004).

Soil pH value is the most important parameter, influencing elements mobility in soil. This phenomenon is related to soil sorbents charges depending on the value of soil reaction. Cadmium activity is strongly affected by acid soils whereas in alkaline soils Cd is rather immobile. Cd reveals a tendency to be removed from solution in the form of hydroxides or to precipitate on the clay surface with increasing pH (Yong *et al*., 1992, Naidu *et al.*, 1996). The soil organic matter and sesquioxides may largely control Cd mobility, and in alkaline soil, the precipitation of Cd compounds likely accounts for Cd equilibrium. Lead is reported as the less mobile element among the other heavy metals. This statement is supported by the relatively low Pb concentration in natural soil solution (Donisa *et al*., 2003, Lacarce *et al*., 2012).

Acidification is one of the most significant soil degradation processes, which may affects the soil filtration function through its effect on the physical, chemical and biological characteristics of soils. According to Adriano (2001), the pH can be viewed as the master variable of all the driving factors because it can affect the surface charge and subsequent adsorption of solutes by variable charge soil components, such as layer silicate clays, organic matter, and oxides of Fe and Al. Prognoses of the negative ecological consequences of acidification are often underestimated. It is important to clarify how the acidification affects soil filtration function. The aim of this study was to investigate the effects of simulated acid load on the fate of inorganic risk elements (Cd, Pb), soil sorption potential and thus soil filtration function (immobilization potential of Cd and Pb).

Material and methods

We made use of a short-term pot experiment with grown plant of spring barley cultivated at different soil types Fluvisol, Cambisol, Stagnosol, Podzol (tab. 1. 2) with simulated acid load. The soils were classified according to the World Reference Base (2006). Soil samples were collected from humic horizons where 5 soil samples were mixed from 1 plot of 10x10 m. Homogenised soil samples of 4 kg were placed in plastic flow containers with 4 dm^3 volumes. We seeded 100 grains of spring barley into two lines. After the plant emergence on the 11^{th} day, we reduced the number of plants to 20 pieces per container (2 lines of 10 plants in every container).

Two treatments were applied, treatment A: watering with acid solution of pH 2.5 (mixture of distilled water, sulphuric and nitric acid, 3:1 ratio, added into the distilled water) and treatment B: watering with distilled water of pH 5.4. A volume of 200 ml was applied three-times per a week in four repetitions. In recalculation three months of pouring with the solution of pH 2.5 equals pouring with solution of pH 4.4 during 19 years (calculated according annual weighed precipitation mean in Slovak territory). Soil samples were air dried, homogenized, sieved on 2 mm and analysed. By selective sequential extraction procedure (Zeien and Brümmer, 1989) the first and the second fractions of Cd and Pb were determined (1.fraction - mobile, 2. Fraction - exchangeable).

Cd, Pb content was analysed by electrochemical AAS – Varian Spectra A 300/400 Zeeman. Soil organic carbon (as Cox) was determined by wet combustion (Nikitin, 1972), quality of organic matter was determined by Kononova method (1966). Humus quality was assessed by the ratio of absorbances at the wavelengths of 400 and 600 nm ($A^{465}/_{665}$), Soil texture and content of particles < 0,01 mm was determined by pipette method, and pH in $CaCl_2$ potentiometrically (Kolektív, 2011).

Table 1 *Initial soil properties*

	pH in $CaCl_2$	Cox in %	$A^{465}/_{665}$	Soil texture (<0.01 mm) in %
Fluvisol	6.77	1.46	5.40	35.39
Stagnosol	4.69	3.69	5.30	39.46
Cambisol	4.55	1.60	5.10	54.24
Podzol	4.43	6.20	5.72	20.93

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Table 2 *Total content, mobile and exchangeable content of Cd and Pb - initial values in soils*

	Total content in $mg.kg^{-1}$		Mobile content in $mg.kg^{-1}$		Exchangeable content in $mg.kg^{-1}$	
	Cd	Pb	Cd	Pb	Cd	Pb
Fluvisol	2.520	195.00	0.266	0.292	1.518	15.200
Stagnosol	0.260	21.00	0.085	0.535	0.046	1.422
Cambisol	0.380	38.85	0.091	2.562	0.017	1.962
Podzol	0.650	47.00	0.182	1.258	0.076	2.499

For the evaluation of soil filtration function following equation was used (Makovníková *et al.*, 2007): [Soil filtration function] = [Potential of soil sorbents] + [Potential of total content of inorganic pollutants] (Eq. 1). Potential of soil sorbents (PSS) includes: pH value (pH), soil organic carbon (Cox), quality of organic matter (optical parameters $A^{400}/_{600}$), and humus layer thickness (H). Potential of soil sorbents was calculated according to equation (Makovníková *et al.*, 2007): [PSS]=F(pH) + F($A^{465}/_{665}$) + F(Cox)*F(H) (Eq. 2).

Score values of soil properties are in Table 3a,b, categorization of soil sorbents is in Table 4, categorization of soil contamination potential in Table 5, categorization of soil filtration function in Table 6. The level of Cd, Pb concentration were evaluated according to the Slovak Soil Law (2004). The score evaluation was determined separately for each element because the Cd affinity to soil sorbents is different than Pb affinity,

Table 3a *Score values of soil properties –Cd*

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pH in $CaCl_2$	score	Cox (%)	score	$A^{465}/_{665}$	score	depth	score
< 5.5	4.0	<1.0	1.00	> 6.01	1.0	< 20 cm	2.0
5.51 – 6.00	3.2	1.00-1.50	0.80	5.51 – 6.00	0.8	21 – 30 cm	1.0
6.01 – 6.50	2.4	1.51-2.00	0.60	5.01 – 5.50	0.6	> 30 cm	0.5
6.51 – 7.00	1.6	2.01-2.50	0.40	4.51 – 5.00	0.4		
> 7.0	0.40	>2.50	0.10	<4.5	0.1		

Table 3b *Score values of soil properties –Pb*

pH in $CaCl_2$	score	Cox (%)	score	$A^{465}/_{665}$	score	depth	score
< 4.05	2.0	<1.0	1.00	> 6.00	2.0	< 20 cm	2.0
4.04 – 4.50	1.6	1.00-1.50	0.80	5.51 – 6.00	1.6	21 – 30 cm	1.0
4.51 – 5.50	1.2	1.51-2.00	0.60	5.01 – 5.50	1.2	> 30 cm	0.5
5.51 – 6.00	0.8	2.01-2.50	0.40	4.51 – 5.00	0.8		
>6.00	0.20	>2.50	0.10	<4.5	0.2		

Table 4 *Categorization of soil sorbents*

Range of rating score	Soil category
> 4.00	very low
3.01 – 4.00	low
2.01 – 3.00	medium
1.01 – 2.00	high
< 1.00	very high

Table 5 *Categorization of soil contamination potential*

Range of rating score	Soil category
0	very low (lower than 80% of limit value)
1	low (equal 80% lower than 120%)
2	medium (equal 120% lower than 200%)
3	high (equal or higher than 200%)

Table 6 *Categorization of soil filtration function*

Range of rating score	Soil category
> 6.9	very low
5.1 – 6.9	low
4.1 – 5.0	medium
3.1 – 4.0	high
< 3.0	very high

Results and discussion

The results show (fig.1, 2, 3) that simulated acid load induced higher activity of buffer systems in the soils and significant changes of the soil chemical properties. Simulated acid load resulted in significant changes in the values of soil reaction. The greatest reduction of pH value in $CaCl_2$ from 4.43 to 4.00 (decrease about -62 847.10^{-9} [H^+]) was recorded on Podzol. Changes in total heavy metal content were not significant (fig. 4). Soil organic carbon (Cox) content increased in treatment A in all soil types. In comparison of treatment A and treatment B, acid load caused higher increase of Cox in treatment B in Cambisol and Podzol, and distilled water watering caused higher increase of Cox in Fluvisol and Stagnosol. Simultaneously with Cox changes, acid load induced changes in organic matter quality (fig. 3).

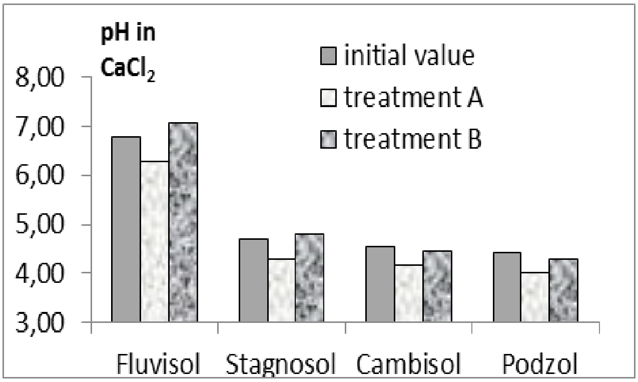

Figure 1
Changes in pH in $CaCl_2$

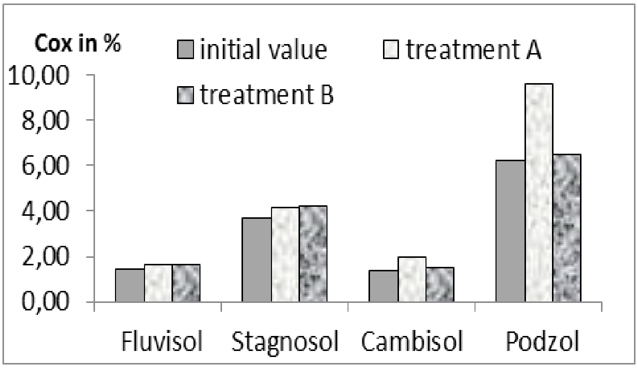

Figure 2
Changes in Cox in %

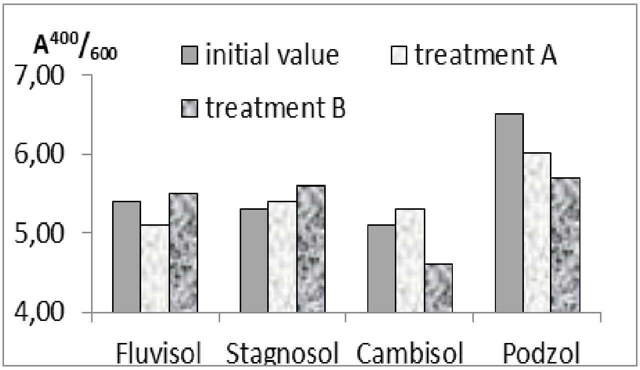

Figure 3
Changes in $A^{465}/_{665}$

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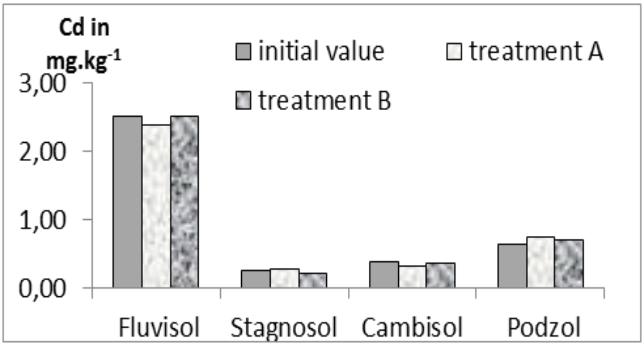

Figure 4
Changes in Cd total content changes

Although total analyses give information about heavy metals content in soils, mostly it is not sufficient criterion for risk estimation and it should be supplemented with the analyses specifying soil contaminant elements form and mobility. Mobile and exchangeable fractions of Cd and Pb (Zeien and Brűmmer, 1989) were analysed (Tab. 7).

Table 7. *Mobile and exchangeable content of Cd and Pb – treatment A and B*

	Mobile content in $mg.kg^{-1}$		Exchangeable content in $mg.kg^{-1}$		Mobile content in $mg.kg^{-1}$		Exchangeable content in $mg.kg^{-1}$	
	Cd	Pb	Cd	Pb	Cd	Pb	Cd	Pb
	Treatment A				Treatment B			
Fluvisol	0.375	0.383	1.635	16.200	0.283	0.299	1.541	16.040
Stagnosol	0.086	0.815	0.043	1.405	0.084	0.598	0.046	1.386
Cambisol	0.102	3.473	0.015	2.125	0.082	2.486	0.012	1.863
Podzol	0.236	1.535	0.065	3.245	0.098	1.063	0.061	2.481

We registered increase of Cd in the mobile fraction (Table 8) (in % of initial value) evoked by acidification in the order: Fluvisol (40.9%) > Podzol (29.6%) > Stagnosol (1.2%) > Cambisol (1.0%) and increase of Pb in mobile fraction in the order: Stagnosol (52.0%) > Cambisol (34.0%) > Fluvisol (31%) > Podzol (29%) (tab.7). Such changes - increase of mobile fraction - are determined by decrease of $pH/CaCl_2$ below critical value of 6.5 for Cd and 4.4 for Pb together with absence of carbonate buffer system in investigated soils. I is recommended that soil pH should be maintained at pH 6.5 or greater in land receiving biosolids containing Cd (Adriano, 2001). Cd mobility increase is in agreement with the results reported by Lianzhen (Lianzhen Li et al., 2014) in anthropogenic contaminated soils, where Cd is weakly bound with soil sorbents. Soil sample of Fluvisol is from location anthropogenically contaminated. Bolan *et al.* (1999) observed that approximately 50% of the pH-induced increase in surface negative charge in variable charge soils was occupied by Cd. In comparison of treatment A and treatment B, acid load caused higher increase of mobile Cd and Pb content in Fluvisol, Cambisol and Podzol which was reflected in increased Cd and Pb transfer into the plants (fig.5, 6). The changes in the exchangeable fractions of Cd and Pb were not so expressive than in mobile fraction (except Fluvisol). We assume that the part of Cd bound in this fraction was transferred into the mobile fraction. Acid load significantly influenced soil sorption potential and thus affected increase in Cd and Pb mobility.

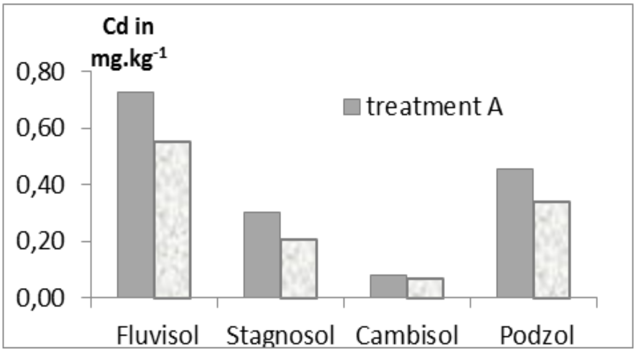

Figure 5
Cd content in plant

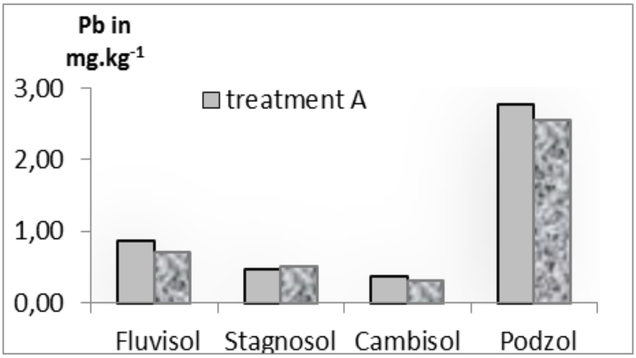

Figure 6
Pb content in plant

The rating system with different importance of parameters was used for evaluation of the individual sorbents as well as for the categories of filtration function. The importance of parameters that determine potential sorption is different therefore the parameters are weighted according to results of factor analysis (Makovníková *et al*., 2007, Makovníková, Barančíková, 2009). Higher value of soil reaction, Cox, depth of humus horizon and lower value of $A^{400}/_{600}$increase soil filtration function. Therefore the score values of soil properties influenced soil potential sorption are opposite to contamination score values, that means the highest score values of soil parameters represent the lowest influence to soil potential sorption. The high pollutants content was evaluated by the high point value and present high risk. High soil sorption potential results by low point value decreases transport risk of inorganic pollutants in ecosystem. The soil sorption potential was calculated according to Eq.2 and evaluation of soil filtration function was made according to Eq.1. The Table 8 shows the changes of the soil sorption potential as well as the soil filtration function caused by the acidification.

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Table 8 *Evaluation of soil filtration function for Cd*

Soil type	Potential of soil sorbents		K potential		Filtration function	
	Rating score	Category	Rating score	Category	Rating score	Category
Initial value						
Fluvisol	3.00	medium	3	high	6.00	low
Stagnosol	4.70	very low	0	very low	4.70	medium
Cambisol	5.00	very low	0	very low	5.00	medium
Podzol	4.90	very low	2	medium	4.90	low
Treatment A						
Fluvisol	3.60	**low**	3	high	6.60	low
Stagnosol	4.70	very low	0	very low	4.70	medium
Cambisol	5.20	very low	0	very low	5.20	**low**
Podzol	5.20	very low	2	medium	7.20	very low
Treatment B						
Fluvisol	1.60	medium	3	high	4.60	*medium*
Stagnosol	4.90	very low	0	very low	4.90	medium
Cambisol	5.20	very low	0	very low	5.20	**low**
Podzol	4.90	very low	2	medium	6.90	very low

Table 9 *Evaluation of soil filtration function for Pb*

Soil type	Potential of soil sorbents		K potential		Filtration function	
	Rating score	Category	Rating score	Category	Rating score	Category
Initial value						
Fluvisol	2.20	medium	3	high	5.20	low
Stagnosol	2.50	medium	0	very low	2.50	very high
Cambisol	3.00	medium	0	very low	3.00	very high
Podzol	3.80	low	2	medium	5.80	low
Treatment A						
Fluvisol	2.00	*high*	3	high	5.00	*medium*
Stagnosol	2.90	medium	0	very low	2.90	very high
Cambisol	3.80	**low**	0	very low	3.80	**high**
Podzol	4.10	**very low**	2	medium	6.10	low
Treatment B						
Fluvisol	2.00	*high*	3	high	5.00	***medium***
Stagnosol	2.90	medium	0	very low	2.90	very high
Cambisol	3.20	**low**	0	very low	3.20	**high**
Podzol	3.30	low	2	medium	5.30	low

Acidification also affects soil sorbents in dependence of individual soil type characteristics, and induces changes in soil quality. The distribution of Cd fractions in soil is different from Pb fractions (Yong *et al*., 1992, Zeien and Brűmmer, 1989, Makovníková, 2001), therefore was the influence of acid load more significant in the case of Cd than Pb. As to Cd, a considerable fraction of the total content is represented by potential mobile pools, while high proportions of Pb are bound in soil organic matter and occluded on poorly and well crystalline Fe-oxides that corresponds to the results of Waller and Pickering (1993) and Bolan *et al*.(2003). In agreement with this fact, acid load significantly influences Cd mobility. There are many factors and processes that determine the soil sorption potential. That is a reason why also our results concerning to PSS varies among the different soil types. Results of soil sorption potential showed that acid load caused negative change of filtration function in one soil type (Fluvisol) in the event of Cd and in two soil types in the event Pb (Cambisol, Podzol). The comprehensive assessment of soil filtration function showed that acid load caused negative change of filtration function in one soil type (Cambisol) in the event of Cd as well as Pb. Results of soil organic matter quality (fig. 3) showed that acid load caused deterioration of organic matter quality. According to the some theories (e.g. Kononova, 1966), humic acids as aromatic compounds have a lignin origin. Lignin significantly contributes to the soil humic acid composition (Adani, *et al*., 2007). Decrease of pH value and increase $A^{465/}{}_{665}$ shift Cambisol from category of medium into low category of soil filtration function for Cd and from category of very high into high category of soil filtration function for Pb. We used in the evaluation of soil filtration function total content of risk elements, because these date are available in existing databases in Slovakia (Čurlík, Šefčík, 1999, Makovníková *et al*., 2007, Kobza *et al*, 2009). The categories with low and very low filtration function for Cd cover 16.2% of all agricultural soils of Slovakia which is less than for Pb (31.4 %) (Makovníková and Barančíková, 2009) .

Conclusion

This work evaluates the impact of acidification on the soil filtration function for Cd and Pb. Results showed that acid load caused negative changes in filtration function of Cambisol in the case of Cd and Pb. Despite the fact, that there were not determined negative changes in soil filtration function of Fluvisol, Stagnosol and Podzol, their initial filtration capacity were low which was reflected in increased Cd and Pb transfer into the plants. We assume that determination and assessment of the mobile and exchangeable risk element fractions are more precise and can provide a more realistic view on the fate of risk elements in soil as well as on the evaluation soil filtration function. The use of available fraction (mobile and exchangeable) in evaluation soil filtration function is difficult since these data are not included in the existing database.

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IMPACT DE L'ACIDIFICATION SUR LES POLLUANTS DESTIN ET DE LA FONCTION DE FILTRATION DU SOL

Résumé

L'objectif de cet article consiste à étudier les effets de l'impact d'une charge acide simulée sur la distribution des polluants inorganiques (Cd, Pb), sur le potentiel de sorption du sol, ainsi que sur la fonction de filtration de sol. Une expérience de végétation (orge de printemps) simulant des précipitations acides a été effectuée sur 4 différents types de sol (Fluvisol, Cambisol, Stagnosol, podzol). Le potentiel de la fonction de filtration de sol a été évalué selon l'équation : [Fonction de filtration de sol] = [potentiel de sorption du sol - PSS] + [potentiel de la teneur totale en polluants inorganiques]. Le potentiel de sorption du sol est défini comme un ensemble de paramètres qualitatifs (pH, qualité de matière organique - $A^{465}/_{665}$) et de facteurs quantitatifs (teneur en matière organique - Corg, profondeur de l'horizon d'humus - H) selon l'équation: [PSS] = F(pH) + F(Q46) + F(Corg) * F(H). La charge acide a eu un impact significatif sur le potentiel de sorption du sol ce qui s'est manifesté par une mobilité élevée de Cd et Pb et par leur transfert ultérieur dans les plantes. Les résultats ont montré une modification négative de la fonction de filtration du sol Cambisol.
Mots-clés: *l'acidification, la réaction du sol, Cd, Pb, la fonction de filtration du sol*

L'IMPATTO DELL'ACIDIFICAZIONE SUL DESTINO INQUINANTI IN FUNZIONE DEL GRADO DI FILTRAZIONE NEL SUOLO

Riassunto

L'obiettivo di questo articolo è di indagare gli effetti dell'induzione di un aumento del grado di acidità del suolo sul destino di contaminanti inorganici (Cd, Pb), sul potenziale adsorbente e sulla funzione di filtraggio del suolo. Abbiamo applicato una sperimentazione in vaso a breve termine, con accrescimento di orzo primaverile coltivato in quattro differenti tipi di suolo (Fluvisol, Cambisol, Stagnosol, Podzol). Il potenziale adsorbente è stato valutato sulla base di Eg.:[funzione di filtro del suolo]=[potenziale adsorbente del suolo-PSS]+[Potenziale di contenuto totale di contaminanti inorganici]. Il potenziale adsorbente del suolo è definito da parametri qualitativi (valore di pH, qualità della materia organica - $A^{465}/_{665}$) e fattori quantitativi (contenuto di materia organica-Corg, spessore degli strati di humus-H) secondo l'Eq.:[PSS]=F(pH)+F($Q^4{}_6$)+F(Corg)*F(H). Il grado di acidità significativo ha influenzato il potenziale adsorbente del suolo and di conseguenza ha comportato un aumento della mobilità di Cd e Pb che si è riscontrato nel loro trasferimento nelle piante. I risultati della funzione filtro del suolo hanno mostrato un cambiamento significativo nei Cambisol (da classe media a classe bassa di funzione filtro del suolo).
Parole chiave: *acidificazione, reazione del suolo, Cd, Pb, funzione filtro del suolo*