

Different forms of chromium in soils with and without contamination: content and vertical distribution

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Abstract. This study aimed to investigate the concentration levels, vertical distribution, and association of various forms of chromium (Cr) with reactive phase carriers in Dystric Cambisols in natural soil-forming environments and in remediated dumpsite areas. Soil samples were collected from two sites and analyzed using a combination of advanced analytical methods. The mean Cr content in the uncontaminated soils was similar to global and regional background levels. In these soils, Cr was mainly bound in stable soil compounds and accumulated in the middle and lower parts of the soil profile. The contents and distributions of total and potentially environmentally available Cr forms depended largely on the clay content and on soil compounds enriched by Al and Fe. The distribution of the water-soluble Cr form was largely dependent on the content of organic, Ca-, and Mn-containing soil compounds. Compared to the uncontaminated soil, significant increases in the content and accumulation of all studied Cr forms were observed in remediated dumpsite soil. This was accompanied by an increase in the proportion of extractable Cr forms and stronger associations of Cr with clay, organic, and Fe- and Mn-rich soil phases. These soil phases act as geochemical microbarriers, effectively stabilizing Cr ions in contaminated soils.

Key words: Dystric Cambisols, chromium, soil contamination, municipal waste, organic and mineral soil phases.

Различные формы хрома в загрязнённых и незагрязнённых почвах: содержание и вертикальное распределение

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Аннотация. Изучено содержание и вертикальное распределение различных форм хрома (Cr) во взаимосвязи с реакционно-активными фазами бурозёмов, сформированных в естественных условиях и на территории рекультивированной свалки бытового мусора. Отобранные на этих двух участках образцы почв проанализированы с использованием комбинации современных аналитических методов. Содержание Cr в незагрязнённых почвах оказалось сходным с уровнями мирового и регионального фоновых значений. В незагрязнённых почвах Cr преимущественно входил в состав стабильных почвенных соединений и накапливался в средней и нижней частях почвенного профиля. Концентрация и распределение общих и потенциально-доступных форм Cr в основном определялись содержанием глины и соединений, обогащённых Al и Fe. Распределение водорастворимой формы Cr зависело от наличия органических, Са- и Мп-содержащих соединений. По сравнению с незагрязнённой почвой, в почве рекультивированной свалки отмечено значительное увеличение всех изученных форм Cr и более высокий уровень статистически значимой положительной связи уровня концентрации потенциально-доступных форм Cr с глинистыми, органическими и обогащёнными Fe и Мп почвенными фазами. Отмеченные почвенные фазы представляют собой геохимические микробарьеры, способные эффективно стабилизировать ионы Cr в загрязнённых почвах.

Ключевые слова: бурозёмы, хром, загрязнение почв, муниципальные отходы, органические и минеральные почвенные фазы.

Introduction

Geochemical surveys of the diffuse input of toxic and potentially toxic elements into dumpsite soils are increasing due to growing environmental risk awareness. Increased waste generation has not always been commensurate with waste management capacity. One consequence of improper waste management is trace element contamination of soils underlying and surrounding waste dumpsites (Agbeshie et al. 2020). The presence of high concentrations of trace elements in soils is a major concern because these elements do not undergo degradation (chemical, physical, or microbial) and tend to accumulate in organic and mineral soil phases, thus being retained in the soil for long periods (Timofeeva et al. 2021).

Among trace elements, chromium (Cr) is one of the most common elements accumulating in dumpsite soils due to its widespread use in various industries (metallurgical, refractory, and chemical), resulting in large quantities of Cr-containing compounds in industrial, residential, and municipal waste (Kotas, Stasicka 2000; Bavaresco et al. 2017). The Cr content in municipal and industrial wastes from various sources can be as high as 10200 mg kg⁻¹, with an average of 1993 mg kg⁻¹ (Kabata-Pendias 2010). Chromium is also a potentially toxic trace element that, at concentrations above threshold levels, is phytotoxic to plants and toxic to humans (Anjum et al. 2017; Babula et al. 2008; Kotas, Stasicka 2000).

Numerous studies have shown that the Cr content in natural soils is largely dependent on the mineralogical composition of the parent material (Bavaresco et al. 2017; Kabata-Pendias 2010; Kotas, Stasicka 2000). Chromium contents have been examined in various soil types, demonstrating that natural baseline total Cr content varies greatly (from 2 to 1100 mg kg⁻¹) (Kabata-Pendias 2010). Soils in urban and industrial regions worldwide contain high Cr levels, posing a risk to the local natural environment (Kabata-Pendias, Szeke 2021). Based on criteria for contaminated land, Cr concentrations in soil above 100 mg kg⁻¹ indicate contamination. Recent research suggests that a Cr content of 100 mg kg⁻¹ in soil is a threshold above which plant antioxidant potential is suppressed and plant growth is reduced (Levizou et al. 2019).

The phytoavailability, toxicity, mobility, accumulation, and distribution of Cr in soils, and hence its environmental contamination potential, are influenced by soil physical, chemical, and mineralogical properties. Soil-specific studies have shown that Cr ions are mainly associated with soil organic matter, Fe- and Mn-rich compounds (mostly oxides), and clay minerals (Bavaresco et al. 2017; Kabata-Pendias, Szeke 2021; Kotas, Stasicka 2000). The relationship of Cr with different soil phases and compounds varies depending on soil acidity, Cr oxidation state, and environmental conditions (Kabata-Pendias, Szeke 2021; Levizou et al. 2019). Nonetheless, the main trends of Cr behavior in soils remain unclear because of location-specific environments and different pedogenic processes that affect both the association of Cr ions with reactive phase-carriers and the natural Cr content, as well as the dynamics of soil system sorption activity for Cr under anthropogenic influence.

In this context, we conducted research focused on the concentration, vertical distribution, accumulation, and association of Cr ions with reactive phase-carriers in soils (Dystric Cambisols) formed both without (natural soil-forming environment) and with direct anthropogenic impact (contamination with municipal waste).

The principal aims of this study were (i) to quantify the total Cr content and the concentrations of potentially environmentally available and water-soluble forms of Cr; (ii) to identify the main factors controlling the vertical distribution and accumulation of Cr

ions in soils from a conditionally pollution-free natural environment and in soils contaminated with municipal waste.

Materials and Methods

The uncontaminated site was located in a coniferous broad-leaved forest massif, considered a natural area (43°37'23" N, 131°51'49" E). The contaminated site was located on a remediated municipal waste dumpsite (43°35'47" N, 131°37'24" E). Remediation of the dumpsite consisted of removing the contaminated upper part of the soil profiles, followed by allowing natural self-overgrowing with native plant associations over 16 years.

The soil used in this study is typical for the southern part of the Russian Far East. It is classified as a Dystric Cambisol according to the World Reference Base for Soil Resources (2006). The Dystric Cambisol consists of four horizons (A, Bw1, Bw2, BC). The soil location favors regular saturation from atmospheric precipitation and surface runoff. The soils from both study sites are characterized by significant weathering processes that increase the accumulation of clay in the middle and lower parts of the soil profiles.

Soils were collected from four soil profiles, sampling the main genetic horizons. After carefully removing roots and stones, the soil samples were ground to a powder.

Soil clay content was analyzed using the recommendations of Pansu and Gautheyrou (2006). Soil organic carbon (SOC) content was determined on dried (105 °C) samples using a TOC-L analyzer (Shimadzu) with a CSN configuration and an SSM-5000A solid sample measurement module.

Total Cr, SiO₂, Al₂O₃, Fe₂O₃, MnO, and CaO contents were determined via energy dispersive X-ray fluorescence spectrometry (EDX) using a Shimadzu EDX-800HS-P instrument as previously described (Timofeeva et al. 2021). To measure the concentration of potentially environmentally available Cr-containing compounds (PEAC), soil samples were completely dissolved in HF and HNO₃ based on Pansu and Gautheyrou (2006). Chromium in soil water-soluble components (WSC) was extracted at a 1:10 soil-to-water ratio. Chromium concentrations in the suspensions were analyzed by atomic absorption spectrometry (Atomic Absorption Spectrophotometer AA-7000, Shimadzu) as previously described in detail (Timofeeva et al. 2021).

Each chemical analysis was performed with three parallel replicates. Analysis of variance was used to determine significant differences and to generate correlation matrices for the concentrations of different Cr forms and selected soil properties. The significance level (P) did not exceed 0.05.

Chromium contamination levels in soils from the contaminated site were evaluated using the contamination factor (CF), calculated from the total Cr content as the ratio of the element's concentration in contaminated soils to its concentration in uncontaminated pristine soils, based on Antoniadis et al. (2017).

Results

The total Cr content in the studied soils was compared with previously determined mean background Cr contents for surface soils worldwide (59.5 mg kg⁻¹), as reported by Kabata-Pendias (2010). Additionally, we compared our results with the regional mean background Cr content in soils of the Primorsky Krai (66.0 mg kg⁻¹) (Golov 2004). The mean concentrations and standard deviations of Cr in the studied Dystric Cambisols are given in the table.

The total Cr content in uncontaminated soils was lower than or similar to the world-soil background value and the regional mean background content throughout the studied

Table. Chromium content and contamination factor of Cr in the Dystric Cambisols.

Таблица. Содержание хрома и коэффициент загрязнения в буроземах.

Form	Horizons (depth, cm)			
	A	Bw1	Bw2	BC
	mg kg ⁻¹			
Uncontaminated soils				
Total	54.82 ± 2.29	61.93 ± 2.70	64.05 ± 2.66	64.42 ± 2.93
PEAC*	19.19 ± 0.94	24.80 ± 1.16	26.59 ± 1.26	18.03 ± 0.51
WSC**	1.06 ± 0.14	1.59 ± 0.16	1.61 ± 0.17	0.67 ± 0.06
Contaminated soils				
Total	187.17 ± 10.03	342.55 ± 16.81	406.93 ± 17.60	161.92 ± 7.09
PEAC	152.39 ± 6.99	267.66 ± 11.32	355.00 ± 14.60	113.24 ± 5.50
WSC	14.54 ± 0.59	19.08 ± 0.85	18.92 ± 0.77	5.65 ± 0.16
CF	3.41	5.53	6.35	2.51

* PEAC – concentrations of potentially environmentally available Cr form; ** WSC – concentrations of water-soluble Cr form; values are means (n = 6), with standard deviations.

soil profiles. Analysis of the vertical distribution of total Cr content revealed an increase with depth. Pronounced peaks of Cr were found in the Bw2 and BC horizons, where the total Cr content was equivalent to the global and regional background values. The concentrations of Cr ions after acid and water extractions, expressed as percentages of the total content, indicate that the PEAC and WSC forms accounted for 36.13% and 2.01% (median values), respectively (table). The results of Cr fractionation showed that the PEAC and WSC forms were dominant in the middle part of the soil profiles (40.04% and 41.51% of the total Cr content in the Bw1 and Bw2 horizons) and lower (27.99%) in the BC horizon. In general, the vertical distribution of Cr in the studied uncontaminated soils reflects the cycling of elements in the soil-plant system of the natural environment.

Significant correlations were observed between Cr (all forms) and soil compounds enriched by Ca (r_{Cr-Ca} from 0.59 to 0.95) (figure).

The total Cr content and concentration of the PEAC form were mainly associated with clays and with soil compounds enriched by Al and Fe ($r_{Clay-Cr}$ 0.68 and 0.87; r_{Al-Cr} 0.83 and 0.72; r_{Fe-Cr} 0.59 and 0.77, respectively). In contrast, the relationship between these components and the concentration of the Cr WSC form was negative (figure). This result suggests that Cr ions associated with clay and Al- and Fe-enriched soil compounds represent forms of Cr that are less available for mobilization. The combination of SOC and soil compounds enriched by Ca and Mn best explained the variability of the WSC Cr form (r_{SOC-Cr} 0.81; r_{Ca-Cr} 0.95; r_{Mn-Cr} 0.78). Additionally, positive correlations were observed between the Cr PEAC form and soil Mn-containing compounds (r_{Mn-Cr} 0.83). This suggests the formation of complexes of Cr ions mainly with Ca and Mn colloids.

The values in Table 1 indicate that the Cr concentrations varied considerably between the two study sites. The total Cr content in soils previously contaminated with municipal waste was elevated relative to the uncontaminated soils, the world-soil background value, and the regional mean background value, by factors ranging from 2.43 to 6.87. This indicates enrichment of Cr throughout the entire soil profile. The calculated contamination factor (CF) values indicated that Cr is a major contributor to soil contamination (table). The total contents and contamination factors of Cr were highest in the middle soil horizons and decreased with increasing depth. Our results suggest that past intensive input of Cr

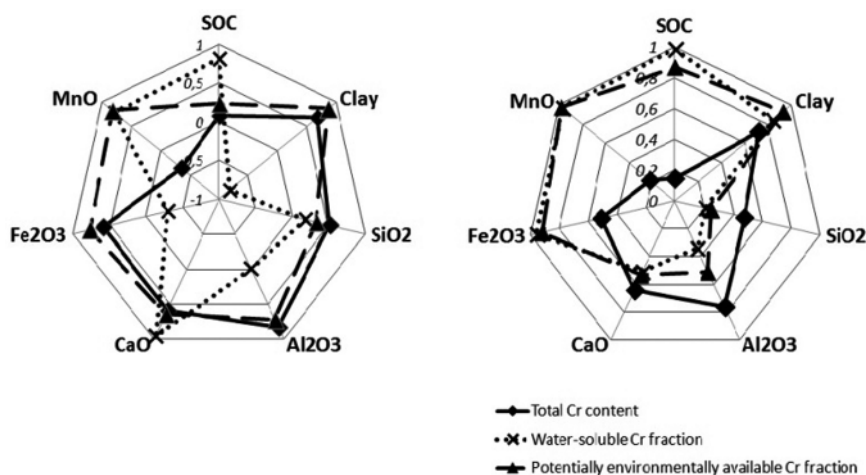


Figure. Correlation coefficients between the Cr concentration and chemical and physical properties of soils.

Рисунок. Коэффициенты корреляции между содержанием Cr и химическими и физическими свойствами почв.

from waste has resulted in the formation of a geochemical anomaly, and Cr poses a high ecological risk to the surrounding ecosystems in the studied contaminated area.

Our work demonstrates increased Cr mobilization after water and acid extractions in the soils of the former municipal waste dumpsite compared to uncontaminated soils. In terms of percentage values, the mean PEAC and WSC Cr form concentrations were 80.68% and 5.44%, respectively. The vertical distribution of absolute concentrations of these Cr forms confirmed the distribution of total Cr content. However, the percentage levels show enrichment of the WSC form of Cr in the upper horizons (7.76% of total Cr content). The percentage levels of the PEAC Cr form had a relatively distinct peak in the A and Bw₂ horizons. The enrichment of the surface horizon with water- and acid-soluble Cr-containing compounds resulted from contamination with municipal waste associated with the corrosion of Cr-containing materials and the migration of decomposition components in the soils.

The vertical distribution of total and PEAC forms of Cr indicated a strong affinity for clay and soil compounds enriched with Ca, Al, and Fe (r Clay-Cr 0.72 and 0.93; r Ca-Cr 0.64 and 0.53; r Al-Cr 0.76 and 0.51; r Fe-Cr 0.51 and 0.92, respectively). A similar relationship between Cr and soil compounds was observed in soils from uncontaminated areas (figure). Compounds enriched by Mn and Fe ions, and SOC, were the soil factors that best explained the distribution and accumulation of the WSC and PEAC forms of Cr (r SOC, Fe, and Mn - Cr from 0.87 to 0.99). Our results show an increased affinity of SOC and soil Fe- and Mn-rich compounds for retaining Cr ions in soils previously contaminated with municipal waste compared to uncontaminated soils.

Discussion

In natural soils, Cr is a lithophile element, and maximum Cr contents are generally found in the lower soil horizons, which contain weathered parent materials (Kabata-Pendias, Szteke 2021). Furthermore, the significant positive correlation between Cr and Al and Fe oxides confirms the high affinity of Cr for the parent material in different soil types (Wang et al. 2009). We also observed an increase in Cr content in the lower part of

the uncontaminated soil profiles (BC horizon). Our data indicate that in uncontaminated soils, Cr associated with clay and Al- and Fe-containing soil compounds largely represents Cr ions embedded in the crystal lattice of these compounds, where Cr is less available for remobilization. The enrichment of the studied contaminated soils with Cr throughout all horizons is direct evidence of past contamination with municipal waste. This is supported by the increased amounts of Cr in both industrial and municipal wastes and by the wide use of Cr in the production of metal alloys, electroplating, leather tanning, and wood preservation (Bavaresco et al. 2017; Kabata-Pendias 2010).

Our work demonstrates that in uncontaminated soils, SOC and Ca- and Mn-containing soil compounds regulated the concentrations of the Cr form most available to the surrounding environment (plants, groundwater). The capacity of SOC and Mn oxides and hydroxides for Cr retention is well-known. However, the combination of different mechanisms (such as surface adsorption, co-precipitation, substitution, and complexation with organic ligands) controlling the interactions of Cr with SOC and Mn-containing soil compounds depends on specific soil properties, especially the redox status (Sparks 2003; Trebien et al. 2011). A study of Cr behavior in soils with alternate wetting and drying periods indicated that co-precipitation and substitution of Cr with Mn were predominant due to the association of Cr with freshly precipitated Mn oxides (Essington 2003). Despite the presence of contrasting redox cycles, the association of Cr with Mn in the soils from both study fields was related to both surface precipitation of Cr and the formation of outer-sphere complexes, as well as to the adsorption of Cr and the formation of strong chemical bonds between Cr-containing compounds and Mn. Fendorf and Zasoski (1992) suggested that surface complexes of Cr ions on Mn oxides and hydroxides are formed before electron transfer during Mn reduction by organic compounds. In general, increased organic matter content enhances the soil's ability to absorb Cr (Bavaresco et al. 2017).

The affinity of Cr for SOC and Mn-containing soil compounds has a significant ecological aspect (Timofeeva et al. 2021). Manganese oxides and hydroxides have high redox activities and can oxidize significant amounts of Cr(III), which has relatively low toxicity and mobility, to Cr(VI), which is highly toxic, soluble, and mobile, during their reduction (Leita et al. 2009). Trebien et al. (2011) found that the amount of Cr (III) oxidation depended on the concentration of easily reducible Mn oxides and that soluble organic compounds in soil decreased Cr (VI) formation due to Cr (III) complexation. Literature data on the stability of Cr-organic matter complexes are contrasting. Studies in soils with different organic matter contents have shown that organic compounds promote the reductive dissolution of easily reducible Mn oxides and form stable, low-redox-activity complexes with Cr (III) (Bavaresco et al. 2017). However, Cr (III) bound to both humic acids and fulvic acids is easily mobilized (Kabata-Pendias 2010). The close relation of the water-soluble Cr forms to SOC in the soils from both study sites points to the formation of unstable complexes, most likely due to electrostatic interaction of Cr with negatively charged functional groups of the organic matter, from which Cr may be easily replaced by other cations such as Ca.

The vertical distribution of Cr in contaminated soils indicates accumulation of this element in the middle part of the soil profiles. This also has an ecological aspect, because accumulation of Cr in the Bw1 and Bw2 horizons helps prevent element transport to deeper soil layers and subsequent input into groundwater. The enrichment of contaminated soils by potentially environmentally available and water-soluble forms of Cr, and the increase in their association with clay, SOC, and Mn- and Fe-containing compounds, can most likely be explained by the presence of Cr originating from municipal waste in compounds

that were available for transformation and complexation with clay, organic, and Fe- and Mn-rich soil phases.

Conclusion

The levels of Cr content and its vertical distribution in the uncontaminated soils depend on natural environmental conditions, especially the composition of the mineral parent material. The Cr values were lower than or similar to the world-soil background value and the regional mean background content. The concentrations of total and potentially environmentally available forms of Cr were strongly positively related to the clay content and to different soil compounds enriched by Al and Fe. The results of this study indicate that only small portions of Cr ions are available to plants in uncontaminated soils. The distribution of the water-soluble Cr form was largely dependent on the mobilization and distribution of SOC and Ca- and Mn-containing soil compounds in soils from the uncontaminated area.

Soil contamination with municipal waste is a factor that increases the content, accumulation, and mobility of Cr. Despite remediation over 16 years, the studied contaminated soils continued to have elevated Cr concentrations compared to the world-soil background value and the regional mean background content. Our results indicate that clay, organic, and Fe- and Mn-rich soil phases are the main reactive phases that influence Cr precipitation and complex formation. Chromium originating from the decomposition of municipal waste was primarily transformed by these soil phases into a potentially environmentally available form (which is less available for mobilization compared to water-soluble forms).

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