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Chemical fractionations and mobility of heavy metals in soils of eastern Uttar Pradesh

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ABSTRACT

In the current investigation, an attempt has been made to assess the various chemical forms and mobility factor (MF) for lead (Pb), cadmium (Cd), chromium (Cr) and nickel (Ni) in four soils of eastern Uttar Pradesh. For this purpose, two different surface soil samples were collected from each of Entisol, Inceptisol, Vertisol and Alfisol soil orders during 2019–20. The modified Tessier sequential extraction procedure was applied to determine the chemical pools of Pb, Cd, Cr and Ni in each soil. Results indicated that total metal content follows the order of Pb>Cr>Ni>Cd across the orders, whereas mobility factor of micronutrients could be arranged as: Cd>Pb>Ni>Cr. The bioavailable metal fraction of all micronutrients, i.e. exchangeable pool (F1) was maximum in Inceptisol. The highest carbonate bound (F2), Fe–Mn oxides bound (F3), organically bound (F4) and residual (F5) fraction were recorded in Vertisol, Alfisol, Inceptisol and Alfisol, respectively for all heavy metals. The mean percentage value of Pb and Ni fractions were in the order of F5>F3>F4>F2>F1, whereas Cd whose chemical fractions follows the order of F2>F3>F4>F5>F1. The Pb was highly mobile in Vertisol, Ni in Inceptisol, while Cd and Cr in Entisol. Therefore, availability and mobility of heavy metals in the soil environment depends mainly on their association with various chemical fractions of the soil which relies on the mineralogical origin of the metals.

Keywords: Chemical fractionation, Heavy metals, Mobility factor, Soil orders

In recent years, heavy metal contamination in soils has gained worldwide attention because of its high toxicity, non-biodegradability and long-term accumulative behavior (Fajardo *et al.* 2019). Heavy metals not only change the soil fertility, but also disturb the microbial community and lead to biodiversity loss (Kasemodel *et al.* 2019). In general, high levels of metal pollution in soil are recorded due to long-term application of industrial effluents, sewage, polluted river water and municipal solid waste (Golui *et al.* 2020). Metals are transferred from polluted soils to human food chain via crops grown there on. Metals are present in various physico-chemical associations in soils such as: i) water soluble; ii) exchangeable; iii) occluded or co-precipitated with oxides, carbonates and phosphates, or other secondary minerals; iv) linked to organic substances; and v) ions in the crystalline lattices of primary minerals (Ryan *et al.* 2002). Water-soluble and exchangeable fractions are considered readily mobile and available to plants, while metals incorporated into the structure of primary and secondary minerals appear relatively inert. The carbonate bound fraction or metal occluded in Fe–Mn oxides or complexed with organic matter (OM) is considered as relatively active fractions depending on the chemical environment of soil. However, a small fraction of metal is present in bioavailable form (Massas *et al.* 2013). The 'sequential extraction' a multi-step extraction makes it possible to differentiate the mobile and residual fractions and to characterize the different labile fractions (Kouchou *et al.* 2020).

Most of the studies evaluated total metal content to monitor heavy metals load in soils (Gowd *et al.* 2010). Though total metal content is used to assess the long-term soil enrichment and to estimate the origin of the metals, moreover it provides negligible information on the potential bioavailability of the metals. The metal fractions in soil have a strong relationship with bioavailability of metal (Massas *et al.* 2013). In this backdrop, the present investigation was carried out with the objectives of assessing the distribution of lead (Pb), cadmium (Cd), chromium (Cr) and nickel (Ni) in different fractions in four major soil orders in eastern Uttar Pradesh and evaluating their mobility factor.

MATERIALS AND METHODS

Location and collection of soil samples: The GPS based surface soil samples (0–15 cm) were collected from four major taxonomic orders of eastern Uttar Pradesh, i.e. Entisol,

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Inceptisol, Vertisol and Alfisol. For each soil orders, two soil samples were collected from two separate locations. Entisol was collected from lower bank of the river Ganga, one from Shikhar block of Mirzapur (25º13' N and 82º87' E) which was designated as Entisol-I and another from Pindra block of Varanasi (25º31' N and 83º02' E) which was designated as Entisol-II. Inceptisol was sampled from agricultural land of Shikhar block, Mirzapur (25º15' N and 82º87' E) and Agricultural Research Farm at Institute of Agricultural Sciences, Banaras Hindu University, Varanasi (25º25' N and 82º99' E) and labeled as Inceptisol-I and Inceptisol-II, respectively. Agricultural land of Nagar city (25º10' N and 82º50' E) and Shikhar (25º80' N and 82º47' E) block of Mirzapur was chosen for the collection of Vertisol and tagged as Vertisol-I and Vertisol-II, respectively. The Alfisol-I was collected from agricultural land of Chakia block of Chandauli (25º01' N and 83º21' E) and Alfisol-II was collected from Agricultural Research Farm at Rajiv Gandhi South Campus, Banaras Hindu University, Mirzapur (25º06' N and 82º60' E).

Processing and analysis of soil samples: Soil samples were collected and analyzed during 2019–20 in the Department of Soil Science and Agricultural Chemistry, Institute of Agricultural Sciences, Banaras Hindu University, Varanasi. Soil samples were air-dried at room temperature followed by grounding in wooden mortar and pestle and passed through a 2 mm sieve and homogenized properly and further grounded to pass through 0.5 mm sieve for chemical fractions analysis. The modified Tessier sequential extraction procedure was applied to determine the chemical fractionation of Pb, Cd, Cr and Ni in each soil order by atomic absorption spectroscopy (AAS) (Tessier *et al.* 1979). Fraction 1 (F1), fraction 2 (F2), fraction 3 (F3), fraction 4 (F4) and fraction 5 (F5) were estimated and designated as: exchangeable, carbonate bound, Fe–Mn oxides bound, organically bound and residual pools of the micronutrients in soil, respectively. The total trace elements content in the soil samples were determined by the summation of all five fractions (F1–F5). The mobility factor (MF) of Pb, Cd, Cr and Ni was calculated based on the content of trace elements in the mobile fractions to the sum of all fractions (Gusiatin and Kulikowska 2015).

$$\text{Mobility factor (MP)} = \frac{F1 + F2}{F1 + F2 + F3 + F4 + F5} \times 100$$

The F1–F5 is the trace elements content in each fraction (mg/kg) according to the modified Tessier sequential extraction procedure. This equation shows the potential mobility of trace elements in the soil.

The data were statistically analyzed with one-way analysis of variance (ANOVA) using SPSS version 16.0 software. Duncan's multiple range test (DMRT) was performed to test the significance of the difference between the treatments at $P \leq 0.05$.

RESULTS AND DISCUSSION

Lead fractions and mobility factor: The F1, F2, F3, F4 and F5 fractions of Pb in soils ranged from 1.83–6.49, 7.23–18.6, 25.2–52.8, 16.8–26.9 and 34.7–56.9 mg/kg, respectively (Table 1). The maximum F1 fraction was recorded in Inceptisol-II, F2 in Vertisol-I, F3 in Alfisol-I, F4 in Inceptisol-II and F5 in Alfisol-II. This result was supported by the observations made by Yan *et al.* (2017). The mean percentage value of Pb pools of total Pb were in the order of F5 (37.9%)>F3 (30.1%)>F4 (17.7%)>F2 (10.6%)>F1 (3.60%). The occurrence of Pb was mostly concentrated in the oxide bound fraction of soil or may be incorporated in the lattice structure in the soil (Asmoay *et al.* 2019). Thus, the highest concentration of total Pb was in Alfisol-I and Vertisol-II (141 mg/kg), whereas the minimum in Entisol-I (89.4 mg/kg). The mobility of Pb in various soil orders represent: Alfisol-II>Alfisol-I>Entisol-I>Entisol-II>Vertisol-II>Inceptisol-II>Inceptisol-I>Vertisol-I (Table 1). The large capacity of soil to adsorb Pb is often attributed to the presence of Fe–Mn oxides and OM (Golui *et al.* 2020). Therefore, Pb exhibit lowest mobility in Alfisol (major association with Fe–Mn oxides).

Cadmium fractions and mobility factor: Soil Cd has mainly associated with the carbonate bound fraction across the soil orders (36.6% of the total extracted Cd), while smaller values in the exchangeable (11.5%) fractions. Also, the other researchers showed that Cd was mostly associated with the carbonate and Fe–Mn oxides bound fractions and very little to the organically bound and residual fractions (Duplay *et al.* 2014).The maximum F1 (0.32 mg/kg), F2 (0.94 mg/kg), F3 (0.66 mg/kg), F4 (0.39 mg/kg) and F5 (0.34 mg/kg) Cd pools was recorded, respectively in Inceptisol-II, Vertisol-I, Alfisol-I, Inceptisol-II and Alfisol-I (Table 1). In case of total Cd content, Vertisol-II (2.39 mg/kg) had the maximum value, whereas the lowest in Entisol-I (1.68 mg/kg). The findings of the present investigation were lined with the findings of Jalali and Hemati (2013). The higher percentage of Cd in the exchangeable and carbonate bound fractions indicated the high solubility and mobility of Cd in soils (Beygi and Jalali 2019). Therefore, Entisol-II noted the maximum MF 54.0% for Cd, however Alfisol-II recorded the lowest 38.8% (Table 1). The mobility factor of Cd across the soil orders appeared in the order of Entisol>Vertisol>Inceptisol>Alfisol.

Chromium fractions and mobility factor: The concentration of each chemical form of Cr in soil is shown in Table 2. The highest exchangeable Cr pool (F1) was recorded in Inceptisol-II (1.08 mg/kg) and the lowest in Entisol-I (0.41 mg/kg). Moreover, the highest F2, F3, F4 and F5 fractions were documented in Vertisol-I (6.14 mg/kg), Alfisol-I (10.8 mg/kg), Inceptisol-II (14.7 mg/kg) and Alfisol-I (68.1 mg/kg), respectively. In summary, on an average, the maximum proportion of the extracted Cr of total Cr was associated with the F5 (67.6%) followed by F4 (15.4%), F3 (10.3%), F2 (5.65%) and F1 (1.00%) pool. A significantly high proportion of Cr was associated with organically bound and residual fraction, which is in agreement with other workers (Hseu *et al.* 2018; Zhang *et al.* 2020). In case of total Cr content, Alfisol-I (91.8

Table 1 Distribution of lead and cadmium (mg/kg) in different soil fractions and mobility factor

Soil type	Lead							Cadmium						
	F1[#] (mg/kg)	F2 (mg/kg)	F3 (mg/kg)	F4 (mg/kg)	F5 (mg/kg)	Total (mg/kg)	MF (%)	F1 (mg/kg)	F2 (mg/kg)	F3 (mg/kg)	F4 (mg/kg)	F5 (mg/kg)	Total (mg/kg)	MF (%)
Entisol-I	1.83c (2.05)*	10.9cde (12.1)	25.2d (28.2)	16.8c (18.8)	34.7c (38.8)	89.4c	14.2c	0.21bcd (12.5)	0.69b (41.1)	0.34b (20.2)	0.21c (12.5)	0.23c (13.7)	1.68e	53.4a
Entisol-II	2.49bc (2.57)	12.0cd (12.4)	27.1cd (28.0)	17.4c (18.0)	37.8bc (39.1)	96.8c	15.0bc	0.24abcd (13.4)	0.73ab (40.8)	0.35b (19.6)	0.22bc (12.3)	0.25bc (14.0)	1.79de	54.0a
Inceptisol-I	5.91a (4.91)	13.8bc (11.5)	32.4cd (26.9)	24.9a (20.6)	43.5abc (36.1)	120b	16.4ab	0.28ab (13.1)	0.77ab (36.0)	0.51ab (23.8)	0.32ab (15.0)	0.26bc (12.1)	2.14bc	48.9a
Inceptisol-II	6.49a (5.01)	14.7bc (11.3)	36.4bcd (28.1)	26.9a (20.7)	45.1abc (34.8)	130ab	16.4ab	0.32a (13.9)	0.78ab (33.9)	0.53ab (23.0)	0.39a (17.0)	0.28abc (12.2)	2.30ab	47.8a
Vertisol-I	5.64a (4.25)	18.6a (14.0)	37.6bcd (28.3)	22.5abc (17.0)	48.5abc (36.5)	133ab	18.3a	0.26abc (11.1)	0.94a (40.2)	0.56a (23.9)	0.30abc (12.8)	0.28abc (12.0)	2.34ab	51.1a
Vertisol-II	5.26a (3.74)	16.7ab (11.9)	41.7abc (29.6)	24.8ab (17.6)	52.3ab (37.2)	141a	15.7bc	0.28ab (11.7)	0.92a (38.5)	0.57a (23.8)	0.32ab (13.4)	0.30ab (12.6)	2.39a	50.0a
Alfisol-I	4.40ab (3.11)	9.41de (6.65)	52.8a (37.3)	19.4bc (13.7)	55.4a (39.2)	141a	9.80d	0.18cd (8.53)	0.65b (30.8)	0.66a (31.3)	0.28abc (13.3)	0.34a (16.1)	2.11bc	39.3b
Alfisol-II	4.32ab (3.15)	7.23e (5.27)	47.5ab (34.6)	21.3abc (15.5)	56.9a (41.4)	137ab	8.45d	0.15d (7.58)	0.62b (31.3)	0.64a (32.3)	0.26bc (13.1)	0.31ab (15.7)	1.98cd	38.8b
Mean	4.54 (3.60)	12.9 (10.6)	37.6 (30.1)	21.7 (17.7)	46.8 (37.9)	124	14.3	0.24 (11.5)	0.76 (36.6)	0.52 (24.8)	0.29 (13.7)	0.28 (13.5)	2.09	47.9
SEm (±)	0.65	1.24	4.46	1.91	4.48	6.11	0.65	0.03	0.07	0.06	0.03	0.02	0.08	2.09
CD (P≤0.05)	1.94	3.72	13.4	5.71	13.4	18.3	1.95	0.09	0.20	0.19	0.10	0.06	0.23	6.28

*Data in parentheses indicates proportion of each fraction in total Pb and Cd present in soil, respectively

[#]F1, Exchangeable fraction; F2, carbonate bound fraction; F3, Fe–Mn oxides bound fraction; F4, organically bound fraction and F5, residual fraction, and MF, mobility factor.

Table 2 Distribution of chromium and nickel (mg/kg) in different soil fractions and mobility factor

Soil type	Chromium							Nickel						
	F1[#] (mg/kg)	F2 (mg/kg)	F3 (mg/kg)	F4 (mg/kg)	F5 (mg/kg)	Total (mg/kg)	MF (%)	F1[#] (mg/kg)	F2 (mg/kg)	F3 (mg/kg)	F4 (mg/kg)	F5 (mg/kg)	Total (mg/kg)	MF (%)
Entisol-I	0.41d (0.89)*	3.38cd (7.35)	5.66b (12.3)	7.44e (16.2)	29.1c (63.3)	46.0c	8.31a	0.47cd (1.75)*	1.60cde (5.95)	5.51c (20.5)	2.37d (8.81)	17.0d (63.0)	26.9b	7.94abc
Entisol-II	0.46d (0.87)	3.55cd (6.74)	5.80b (11.0)	8.18de (15.5)	34.7c (65.8)	52.7bc	7.68a	0.66bcd (2.25)	1.79bcd (6.10)	5.93c (20.2)	2.63d (8.96)	18.3cd (62.5)	29.3b	8.59ab
Inceptisol-I	0.98ab (1.42)	4.09bc (5.92)	6.10b (8.83)	14.1a (20.4)	43.9bc (63.5)	69.1ab	7.42a	1.11ab (2.95)	2.22abc (5.89)	7.59ab (20.1)	4.93ab (13.1)	21.8bcd (57.9)	37.7a	9.07ab
Inceptisol-II	1.08a (1.42)	4.82b (6.32)	6.29b (8.24)	14.7a (19.2)	49.5abc (64.8)	76.3a	7.81a	1.24a (3.07)	2.76a (6.83)	7.72ab (19.1)	5.59a (13.8)	23.1bcd (57.2)	40.4a	10.2a
Vertisol-I	0.87abc (0.99)	6.14a (6.99)	8.72a (9.92)	12.2abc (13.9)	59.9ab (68.2)	87.9a	8.04a	0.89abc (2.14)	2.41ab (5.79)	8.70a (20.9)	4.66b (11.2)	25.0abc (60.0)	41.7a	8.15abc
Vertisol-II	0.89abc (1.00)	5.92a (6.64)	8.96a (10.1)	13.1ab (14.7)	60.3ab (67.6)	89.1a	7.70a	0.13d (0.44)	1.35de (4.59)	6.72bc (22.8)	2.18d (7.41)	19.0cd (64.7)	29.4b	5.21abc
Alfisol-I	0.59cd (0.64)	2.23e (2.43)	10.8a (11.8)	10.0cde (10.9)	68.1a (74.2)	91.8a	3.11b	0.39cd (0.85)	1.03e (2.24)	9.19a (20.0)	3.65c (7.92)	31.8a (69.0)	46.1a	3.20c
Alfisol-II	0.66bcd (0.74)	2.54de (2.84)	9.52a (10.7)	10.8bcd (12.0)	65.9a (73.7)	89.4a	3.62b	0.44cd (1.05)	1.15de (2.74)	8.97a (21.3)	3.78c (8.99)	27.7ab (65.9)	42.0a	3.92bc
Mean	0.74 (1.00)	4.08 (5.65)	7.73 (10.3)	11.3 (15.4)	51.4 (67.6)	75.3	6.71	0.67 (1.81)	1.79 (5.01)	7.54 (20.6)	3.72 (10.0)	23.0 (62.5)	36.7	7.03
SEm (±)	0.10	0.32	0.75	0.86	6.58	6.97	0.35	0.17	0.22	0.51	0.28	2.35	2.60	1.55
CD (P≤0.05)	0.31	0.97	2.26	2.67	19.7	20.9	1.04	0.50	0.66	1.54	0.85	7.04	7.80	4.64

*Data in parentheses indicates proportion of each fraction in total Cr and Ni present in soil, respectively

[#]F1, Exchangeable fraction; F2, carbonate bound fraction; F3, Fe–Mn oxides bound fraction; F4, organically bound fraction and F5, residual fraction, and MF, mobility factor.

mg/kg) recorded the maximum value, whereas the lowest in Entisol-I (46.0 mg/kg). Kouchou (2020) also found a similar association of Cr. Chromium was highly mobile in Entisol-I, as it observed the highest MF 8.31% (Table 2) and the mobility factor of Cr follows the order of Entisol>Vertisol>Inceptisol>Alfisol. Chromium is mostly associated with the low molecular weight organic acids and low adsorption capacity of Cr in sand particles increased its mobility (Borah *et al.*2018).

Nickel fractions and mobility factor: The Inceptisol-II recorded the maximum F1 (1.24 mg/kg), F2 (2.76 mg/kg) and F4 (5.59 mg/kg) Ni fraction among the soil orders (Table 2). However, Alfisol-I documented the highest F3 (9.19 mg/kg) and F5 (31.8 mg/kg) Ni pool. The highest percentage of Ni out of total Ni was associated with the residual fraction (62.5%) across the soil orders. About 20.6% of total Ni was associated with Fe–Mn oxides bound, while only 1.81% of total Ni occurred with exchangeable fraction. The Fe–Mn oxide has the ability to absorb metals present in soil solution and to fix them in its lattice layer because of its large surface area (Golui *et al.* 2020).The order of association between total Ni content and different soil orders could be arranged as: Alfisol>>Inceptisol>Vertisol>Entisol. The highest amount of total Ni, i.e. 46.1 mg/kg was recorded in Alfisol-I and the lowest in Entisol-I (26.9 mg/kg). The total Ni content of studied soils is similar to world average Ni contents (8.9–57.7 mg/kg) in non-contaminated soils (Alloway 2013). In case of mobility factor, the maximum 10.2% was noted in Inceptisol-II and the minimum 3.20% in Alfisol-I (Table 2). The mobility factor of Ni in different soils follows the order: Inceptisol>Entisol>Vertisol>Alfisol. In soils, Fe–Mn oxides are the most important metal holders thus, Ni mobility is significantly controlled by its distribution in this fraction (Tashakor *et al.* 2014). Therefore, Alfisol recorded the lowest MF for Ni.

Evaluation of various chemical forms of Pb, Cd, Cr and Ni in four major taxonomic orders of eastern Uttar Pradesh revealed that Cd content was highest in carbonate bound fraction whereas other trace elements in residual pool irrespective of soil orders. The total trace elements content follows the order of Pb>Cr>Ni>Cd across the soil orders whereas the mobility factor of micronutrients could be arranged as: Mn>Cu>Zn>Fe. The bioavailable, i.e. exchangeable pool of Pb, Cd, Cr and Ni was maximum in Inceptisol. The mean percentage value of Pb, and Ni fractions were in the order of F5>F3>F4>F2>F1, whereas Cd whose chemical fractions follows the order of F2>F3>F4>F5>F1. The Pbwas highly mobile in Vertisol, Ni in Inceptisol, while Cd and Cr in Entisol.

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